Interactive comment on "Size-selected black carbon mass distributions and mixing state in polluted and clean environments of northern India" *by* Tomi Raatikainen et al.

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We would like to thank Referee #1 for the constructive comments. The comments below are shown with italicized font and our replies with the upright font. The changes to the manuscript and Supplementary Material are specified in the supplement.

I think this paper needs to be more conclusive (too many hypothesis at the moment) before it can be accepted. The writing itself I feel a bit lengthy. The major points in particular:

-the BC sources are not clear, some back trajectory analysis will be helpful. This unclearness goes through all of the texts when discussing if the source is local or

transported etc. The discussions on diurnal variation are also weak because of lack of source analysis. With clear source analysis, these discussions should be tidier.

We have done back trajectory analysis, but unfortunately we did not see clear correlation between our rBC observations and trajectory parameters such as average direction or altitude. Therefore, the trajectory analysis was left out from the manuscript. We have now shortened the discussion about sources, because we don't have any additional information about these.

-the main limitation is most of the information is derived from DMA360nm, which could only represent a fraction of total BC. The most populated total particle size may not represent the most populated rBC size, therefore combing the rBC size at other DMA sizes will be also useful. I would suggest to analyse and compare the rBC information at all DMA sizes together.

rBC mass and number size distributions (described by the log-normal distribution parameters) are derived using the available DMA sizes ($>$ 100 nm). Size-resolved results such as the rBC core mean diameters are represented by the bin mean values (Fig. 5) and time series from one DMA size (360 nm). Only one DMA size is selected, because the parameters are so similar that selecting another DMA size would lead us to the same conclusions. Better explanation for selecting only the 360 nm DMA size is given in the updated manuscript.

-the chamber temperature introduces some instrument bias, I guess the SP2 laser power was affected by this? how have you corrected this regarding the reduced detection efficiency when reduced laser power?

We have added laser power diagnostic to the Supplementary Material. It seems that laser power has decreased in Gual Pahari so that the detection efficiency is lower for rBC cores when particle size is smaller than 220 nm. A correction would have required detailed quantification of the detection efficiency, which is not possible anymore. We

have therefore limited our calculations to sizes larger than 200 nm, where the effect of the detection limit seem to be small, and used the much larger 360 nm mobility size bin in most calculations.

-the BC mixing state as derived from DMPS-SP2 is not clear, did you calculate as rBC size divided by mobility size? I don't think page 7 has explained what you have done sufficiently. This is really important but this only appears in supplement. The uncertainty of this method is largely from the particle morphology, however it is hard to tell without particle mass measurement (though you measured rBC mass but this is not the total BC-containing particle mass). I think the main texts need to address this uncertainty aided by more thorough analysis at different DMA sizes.

The text in page 7 was indeed unclear regarding the calculations, but it has now been revised. We will also clarify that both rBC and mobility diameters are measured quantities and therefore accurate within the typical measurement uncertainties. We no longer assume that the particles would be spherical, but just use the measured parameters. The reasons for focusing on the single DMA size are given above and in the updated manuscript.

-The bimodal mode of rBC under cleaner environment looks interesting which needs more detailed analysis, such as how much fraction of the smaller mode, how will this fraction be related to the sources. Some very relevant references may be helpful to aid this observation (doi:10.5194/acp-14-10061-2014; doi:10.5194/acp-12-1681-2012).

The fraction of the larger mode is given in the text (0.5-0.8) and the rest are from the smaller mode (0.2-0.5). The two references are familiar to us, but their source analysis is based on different instruments and numerical methods (positive matrix factorization of high-resolution aerosol mass spectrometer data in doi:10.5194/acp-14-10061-2014 and cluster analysis of aerosol time-of-flight mass spectrometer data in doi:10.5194/acp-12-1681-2012). These methods cannot be applied to the current

data, because the different DMA sizes are strongly correlated (see Fig. 5). We have done source analysis using trajectory data, but specific sources could not be identified. Therefore, the source analysis was left out of the manuscript.

Specific:

Fig.3 I would like to see a full set of rBC core size distribution for all of the DMA sizes, also the project standard deviation.

All DMA sizes do not provide valid information and the relevant DMA sizes are correlated, so that the same conclusions could be made based on any of these. However, the Supplementary Material has been updated with the rBC core size distributions for the usable DMA size range and also the standard deviations are shown. Because standard deviations make the figures less readable, these are shown only in the Supplementary Material.

Fig.4 There is a significant fraction of tail on the rBC mass distribution. This seems to be two modes of BC distribution, maybe we could do a lognormal fitting on one mode and then the remaining is the other mode. and why is that?

Bimodal log-normal fits have been added to Fig. 4, but only the smaller mode is fully resolved due to the SP2 sizing limits. Fractions of the modes in the SP2 sizing range are now reported. We don't have good explanation for the origin of the large particles.

Fig. 5 what do the small markers stand for? What is the point for the fitting?

Small markers indicate the data that is not used in the fitting. It is now clarified these data points are ignored (due to instrument detection limits or poor counting statistics). The fits are convenient parameterizations for the rBC mixing state parameters, which can be used in other studies.

Page 8-10, I found the whole section is a bit too lengthy but not really discussing your own results.

This section has been made significantly shorter and more focused on our results (especially LEO). Especially the text about mass fractal dimension parameter has been removed completely.

I found problematic for the fitting in Fig. 7. Because you are measuring the total particle mobility size but only the rBC mass content.

This figure has been removed (see the previous comment).

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-435/acp-2016-435-AC1-supplement.pdf