Dear Dr. Gerasopoulos,

Please find attached the response to the reviewer comments. Moreover, I highlighted in red font color the main changes in the revised text. I did not include very obvious and small changes, typos and so on, in this “red version”, but they are done as mentioned in the responses.

Kind regards,

Antti Arola

We would like to first express our thanks to the reviewer for his/her constructive comments. The responses to these are below after the reviewer points that are in bold.

The manuscript "Measurement-based direct radiative effect by brown carbon over Indo-Gangetic Plain" by Arola et al. presents a novel approach to determining the total aerosol, black carbon (BC), and brown carbon (BrC) direct radiative effect (DRE) using AERONET inversion data products obtained from four AERONET sites in the Indo-Gangetic Plain region. In addition to providing valuable information regarding the sign and magnitude of the DREs, the manuscript details a unique analysis on the seasonality of the DRE for each aerosol type. Overall, the manuscript presents interesting results to a solid analysis and would make a nice contribution to ACP. However, before publication, I believe the following points should be addressed:

-Some statistics on the relevant parameters are needed. Error bars representing one standard deviation on the monthly averages should be included on all the panels in Fig. 3. How variability in these parameters impact monthly differences in the DREs should be assessed in order to confirm the seasonal cycle.

In the revised version, we included a new box-plot figure (thus including also the variability, through 25% and 75% percentiles, in addition to median and mean) of monthly imaginary indices at RNIR wavelengths and the difference between 440nm and RNIR. The former essentially determines our BC and the latter BrC concentrations. This had to be a separate and new plot, since the Figure 3 would not have remained clear and readable enough with several error bars. The variability in these imaginary indices contains the major information regarding also the amount of variability in our retrieved fractions of absorbing components and thus in our DRE calculations. For this reason, we presented only the variability in the parameters of the upper panels of Figure 3 (in the previous version), as the clear link between the upper and other panels is stressed.

We did not, however, conduct new simulations to assess whether the variability in BC and BrC fractions could affect the seasonal cycle that we have obtained. If the DRE calculations were made fully consistently, this would have required to start from the actual retrievals of BC and BrC fractions, with assumed variability in the input information (imaginary indices). Alternatively, we made a rough assessment by looking at the variability of BC and BrC fractions at the months of largest warming and cooling (in the Figure 3 of the revised version). This figure confirms that there is essentially no overlap in BC fractions between these months and thus suggests that the variability overall (BC and BrC) is not nevertheless large enough to substantially mask our clear seasonality of BrC DRE. This is briefly stressed in the revised version.

-As suggested by Lack and Cappa (2010) and Kim et al. (2015) there may be a spectral dependence of kBC, especially those particles containing coatings. Some analysis or discussion on how a spectrally-dependent kBC would impact the calculated BrC volume fractions (and thus the BrC DREs) should be included in the text. Alternatively, if the assumption that considering only pure, uncoated BC is indeed valid for this region and/or this method, this should be stated and supported with references.

Lack and Cappa 2010 did not discuss spectral variability of kBC at all. And in Kim et al. 2015 (if we now talk about the same paper), the dependence was very minor indeed: from 0.67 at 467nm to 0.63 at 660nm. We are assuming a mixing scenario that is similar to Lack and Cappa, that is internal mixing; however, we are assuming a homogeneous internal mixture, whereas Lack and Cappa assumed a BC core with a BrC coating. We are using a homogeneous internal mixture, because that is what is used in the AERONET retrieval.

-It is my understanding the BrC refractive index (and its spectral dependence) are not well established. A reference and reason as to why the selected BrC refractive index was used would be helpful, even if this is already stated in the referenced Schuster et al. (2015a) paper.

It is certainly true that there exists a wide range of published values of kBrC. We use Kirchstetter et al. 2004 for our baseline refractive index of BrC, because it provides reasonable maximum and median fractions for BrC and also reasonable BrC/BC ratios. This has been discussed in Schuster et al. 2015a and is now also briefly mentioned, as a justification, in our revised version.

-The results section (Section 3, starting on page 21591, line 5) was a bit confusing; as is, the discussion jumps back and forth between Fig. 4 and 5 too much. I suggest restructuring it so that the discussion and description of the more generalized relationship between BrC and BC volume fractions and the differences in k (i.e., Fig. 5) precedes the discussion of the time series in Fig. 4. Also the lines in the bottom panel of Fig. 5 are confusing. I understand each point represents the monthly average BC and BrC volume fractions starting with January at the star but it's not clear it's really showing anything valuable. I recommend only plotting April and November (the middle of the two interesting periods) or removing them all together and referring to Fig. 3.

Thanks for pointing out the need for clarified discussion between these two Figures (Figure 5 and 6 of the revised version). First, however, we want to emphasize the definite need to keep the Figure 5, which brings information that cannot be conveyed from the Figure 3. However, we modified the figure and hopefully it is now easier to interpret. It includes now only two months, as suggested by the reviewer. Moreover, we excluded a separate upper panel of 440nm case and included the difference in imaginary index at 440nm as isolines in the lower RNIR plot. Therefore, the current figure includes only one panel and contains much more information in a one glance. In addition, we tried to make the link between these two figures (Figures 4 and 5) more obvious to the reader.

-Two of the most interesting and important findings of this work are 1) the seasonality in BrC radiative forcing and 2) the fact assuming spectrally-invariant BrC can result in offsets upwards near a factor of 2 in BrC DRE. However, the significance of these results seems to get a bit lost in the comparison to the total aerosol DRE which appears to have no relationship seasonally or otherwise to BrC DRE. Further, because this comparison occurs at the very end of the discussion, it leaves the reader with a bit of "so what?" feeling. I suggest re-framing the big picture and focusing more on the potential errors introduced in the estimated positive forcing associated with carbonaceous aerosols when BrC DRE is neglected. This can be done by including a panel showing the monthly contribution of BrC DRE to total carbonaceous aerosol DRE (BrC +BC) as a ratio or percentage, either in addition to or in replacement of the BC DRE in Fig. 6 and adding some corresponding text in the discussion. This would show that, while BrC

might be a minor contributor to the total aerosol effect in this region, it does contribute with some significance to uncertainties in the positive RF (upwards near 10% for some months based on Fig. 6) associated with aerosol absorption.

In the revised version, we slightly modified the "big picture" as suggested with more discussion regarding the BrC impact in total carbonaceous effect as well. Overall we included results of different DREs (Total, BC, BrC) in a Table 3 of the revised version, which is useful to estimate quantitative importance of BrC from different point of view.

Other specific comments:

Title: I'm not sure if this should be considered "measurement-based" since the analysis relies on parameters obtained indirectly (retrieved) from radiance and direct-sun measurements. I would suggest removing "measurement-based" and starting the title either as "Retrieved direct-radiative: : :" or just "Direct-radiative: : :"

The title has been modified.

Page 21586, line 25 and Page 21587, lines 7-8: So are Level 1.5 or Level 2.0 size distributions used? Clarify.

Yes, we agree that these statements became confusing. For size distributions we ended up using only Level 2 data. This has been now clarified.

Page 21589, lines 3-8: For those instances in which dust is placed in the fine mode, how is BrC distinguished from dust for k440? Do any of these instances occur in March-May? If yes, how does this impact the results shown for the pre-monsoon dust season (March-May) when BrC is noted to have a high BrC volume fraction?

Page 21589, lines 28-29: See above. If goethite and hematite were not distinguished from BrC in the fine mode, could the enhancement in BrC volume fraction be because it is really a BrC + fine-mode dust estimate?

This is to answer both two comments above. The algorithm of Schuster et al. 2015a places dust in the fine mode, when AERONET indicates a very strong spectral dependence of the imaginary index (as indicated by the branch A of their Figure 6). In these cases, it is assumed that there is zero BrC in the fine mode, so there is no risk of overestimating the contribution of BrC to the fine mode in these cases, while underestimation is possible. However, these cases did not occur in our results, i.e. we did not have dust in fine mode in any month. An opposite problem could then happen though: in reality, there should have been fine mode dust, while the algorithm placed only BrC to account for the spectral dependence of imaginary index. These cases, of course, would impact our estimations. However, there is no way to estimate the probability of these cases, when the evidence generally about the dust in fine mode is missing as well.

Page 21593, lines 20-25: What are the values compared to the non-BrC DREs? A table of these values would be useful.

There is a new table hopefully giving enough interesting details regarding the DRE of BrC in relation to the radiative effect of other components.

Minor editorial comments:

Page 21584, line 4: Remove "those" in second sentence.

Done.

Page 21585, line 1: Need an "and" before (2).

Done.

Page 21586, line 7: AERONET does both a direct-sun and sky radiance measurements at 440, 670, 870, and 1020nm. Add "also" between "are" and "used" to make this clearer.

Done.

Page 21592, line 24: Remove "best."

Done.

Page 21594, line 8: Add "an" between "using" and "identical."

Done.

We would like to first express our thanks to the reviewer for his/her constructive comments. The responses to these are below after the reviewer points that are in bold.

General Comments: This is a well-written and informative paper regarding aerosol Brown Carbon (absorbing Organic Carbon) and computed aerosol forcing effects in a very important aerosol source region (the IGP region of India and Pakistan). I believe this paper clearly meets the standards of ACP and makes a useful contribution to the literature, and should be published with only suggested minor revisions (see below).

Specific Comments:

Page 21586, lines 5-6: The AERONET UV filters (340 and 380 nm) have a full width at half maximum (FWHM) of 2 nm as compared to 10 nm for all other channels.

This is now corrected.

Page 21586, line 17: The AOD is larger than 0.5 at 440 nm (include the wavelength)

Wavelength is now included.

Page 21587, lines 18-19: The IGP is bounded to the north by the Himalayan foothills and to the south by lower altitude mountains.

Sentence modified as suggested.

Page 21587, lines 25-27: It would be useful to know if these monthly mean AOD values in Figure 2 are computed from the all of the Level 2 direct sun measurements (most robust method to compute mean climatology of AOD) or from only those direct sun measurements associated with almucantar retrievals.

In our case, mean AODs were based on almucantar retrievals only (thus taken from inversion product, not from direct sun, which would have been more appropriate). This is now clarified.

Page 21587, lines 27-29: Perhaps you could provide the percentage of total retrievals that were L2 retrievals of the entire data set utilized in this study.

These percentages are now included in the text.

Page 21589, lines 8-9: Please clarify, are the number of imaginary indices the number of almucantar retrievals?

This is correct and is now clarified in the text.

Page 21589, line 15: Is the total volume mentioned here for the fine mode only or both

fine and coarse modes?

It was for both fine and coarse mode. This has been now clarified.

Page 21591, lines 1-2: Please briefly describe how you extended the four wavelengths of the albedo data from AERONET retrieval input data (440, 675, 870 and 1020 nm) to the total SW spectrum.

This has been now explained in the revised version.

Page 21595, lines 6-8: Maybe you could mention the advantage here of your approach in the ability to separate dust from BrC absorption.

Now this advantage is briefly mentioned.

Page 21595, Conclusions: You might consider mentioning how critical the BrC absorption is for UV radiation and remote sensing from satellite in the UV wavelengths (i.e. OMI, etc.). Alternatively this could be added to the Introduction if you feel it is not too disruptive of the flow of the paper.

This has been now briefly mentioned in the conclusions.