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Interactive comment on "Modelling black carbon absorption of solar radiation: combining external and internal mixing assumptions" *by* Gabriele Curci et al.

Anonymous Referee #1

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General comments:

This work presents an interesting modelling study on the impact of mixing assumptions in the optical properties and radiative impact of aerosols in the atmosphere. The focus is on the absorption of Black Carbon (BC) and the impact of assuming external or internal mixtures. The authors use results from the regional models contributing to the third phase of the Air Quality Model Evaluation International Initiative (AQMEII), and through the use of an off-line tool to compute absorption properties (FlexAOD), model results are compared with AERONET sunphotometer retrievals.

The weaknesses of the study are: (1) the inconsistency of using prescribed microphys-

ical and optical properties for the aerosols different to the ones used by the models. The mass simulated by the set of models from AQMEII are strongly dependent on the microphysical properties of the aerosols assumed by each model (i.e., density, size distribution). The authors should justify that the harmonization applied in the microphysical properties of the aerosols has a second order impact on the understanding of the uncertainty associated with the mixing states assumed in coated BC particles. (2) A second point that should clarified in the manuscript is the treatment of the Brown Carbon (BrC). If mineral dust is excluded, BrC is the second most absorbing aerosol in the atmosphere, and the treatment in models should be specified. Some organic aerosols have absorbing properties depending on the emission source, others may experience a browning effect through its aging. Some specific discussion on how models deal with that is required to understand the distinction done between bright organic aerosol and BrC. This has a significant impact on the results and conclusions of the work. (3) The underestimation of the aerosol mass concentration at surface level and in the column observed in the AQMEII models introduces an important uncertainty on the results discussed in the manuscript. A lack of a comprehensive evaluation of the chemical composition of the aerosols is lying in the fundaments of the work. Thus, some discussion on that should be introduced in the revised manuscript and make it clear in the conclusions.

The paper is generally well-written. I recommend it to publish in ACP after the previous weakness/questions and following specific and technical comments are addressed.

Specific comments:

- Page 4 Line 9: What is the criterion followed to select the AERONET stations? Are only urban stations selected?

- Page 5 Line 4: Is not the 1.2 thershold for AAE too small for BrC? Lack and Cappa (2010) showed that AAE values, ranging from $1-1.6$, can be observed for internally mixed OC/BC particles and suggested that aerosols with AAE exceeding 1.6 should

be classified as BrC.

- Page 5 Line 6: What is the difference between your approach and Wang et al. (2016)? Would this affect the results of the work?

- Page 5 Line 22: What are the chemical boundary conditions used in the models?

- Page 6 Line 4: A justification of the different results over Europe of model ES1 and over US of model DK1 compared with the others would be useful. Differences are quite significant.

- Page 6 Line 8: There are observational networks in EU and USA that measure chemical composition of particles (i.e., EBAS, EMEP, IMPROVE). Why are not used here to quantify the errors on BC, organic mass, sulfate and dust concentrations? The work presented in the manuscript would benefit from a clear quantificaction of the errors of the models on the chemical composition of the aerosols, at least at surface level.

- Page 6 Line 12: Zhang et al. (2017) showed that some aircraft experimental data suggest that BrC constitute a significant part of absorbing carbonaceous aerosols, especially at high altitudes (> 5km). Some model disagreement with observations seems related to the treatment of BrC. The assumptions used in the present work related to BrC should be described in more detail.

- Page 6 Line 15: It would be useful to understand the reasons of the differences of ES1 and TR1 models predicting the secondary inorganic species. If emissions are the same ones for all the models, the differences should not be so large among models.

- Page 6 Line 21: Here again, there is a need to clarify how BrC is treated in the models. There is some evidence that some BrC comes from secondary organic aerosols (Laskins et al., 2015).

- Page 6 Line 31: It would be useful to include here the description of how BC absorption enhancement and BC core mass fraction are calculated. Right now, the definitions are in the caption of Figure 2.

- Page 7 Line 7: It seems that EU models reproduce the AOD for the wrong reasons considering the significant bias in surface PM2.5. This points to the need of an evaluation of the chemical composition of this PM2.5. What are the implications to the results obtained from the work? It can be expected than a different aerosol vertical distribution will considerably modify the findings of the work.

- Page 7 Line 27: Are the optical properties of the shell species computed as an homogeneous internal mixing?

- Page 8 Line 4: From the results, it turns out that the methodology to combine size distributions is a critical point. This should be remarked in the conclusion of the manuscript as an open issue that deserves further analysis.

- Page 9 Line 18: Would the use of the original models size distribution reduce this bias instead of using a uniform assigned distributions? The assumption of a bulk aerosol implies a loss of detail from the original model results.

- Page 9 Line 26: It would be interesting to present the statistics of the models for this final subset of data. It is expected that the bias on PM2.5 and AOD will be reduced and the confidence with the findings may be stronger.

- Page 11 Line 5: The treatment of BrC in the simulations is not clear. From Table 4, the use of an imaginary refractive index of 0.021 following Highwood et al. (2009) for POM is already quite absorbing. Tang et al. (2016) and Kirchstetter et al. (2004) suggest an imaginary value of 0.03 for BrC from biomass burning. It seems that the assumptions selected for the optical properties of POM contributes to a more absorbing aerosol than the other way around.

In Table 4, it seems there is a typo in the imaginary refractive index of POM, which is set to 0.21. Highwood et al. (2009) suggest a value of 0.021 at 550 nm for the organic aerosol.

- Page 11 Line 14: Results of combining external and internal mixtures (PIM cases)

perform quite similar to CSBC. It would be worth mention it here.

- Page 11 Line 15: A general discussion comparing the EU and US results would be interesting as closing paragraph.

- Page 11 Line 26: This last sentence should be included in the abstract to clarify that the models are compared under BC dominant conditions.

- Page 11 Line 30: What are the recommendations for the modellers to improve the absorption of they systems? There is still significant variability among models, but some insights from the results indicate internal coating or mixtures of external and internal coating are recommended approaches. This can be highlighted in the conclusions. A comment on the impact seen between CSBC and CSBCV is an important result of the work. Only the method used to derive a resulting distribution of an internal mixture is still a significant source of uncertainty.

- Page 23 Table 3: Are the AOD values presented here computed with FlexAOD? Why are there such differences compared with Table S1, where US3 reports an AOD at 440 nm of 0.05?

- Page 23 Table 4: There is a typo in the imaginary part of the refractive index of POM. The refractive index of POM is more representative of a pure BrC rather than a mixture of white organic aerosol and BrC. Nakayama et al. (2012) or Liu et al. (2013) suggest a refractive index of 1.486 - i 2.5e-5 at 550 nm for secondary organic aerosols. Concerning the growth factor, is only the growth factor at 90% considered? The growth factor starts to be non-negligible at 75% relative humidity. Some description on role of the growth factor on the resulting refractive index of an internal mixture would be relevant for the work. Why are BC and POM considered hydrophobic?

- Page 24 Table 6: Some clarification on the numerical failures of the optical calculations is needed.

Technical corrections:

- Page 2 Line 15: I suggest to move this last sentence to the last paragraph of the introduction.

- Page 3 Line 22: Replace "external or internal" by "external and internal".

- Page 5 Line 23: Delete the second point after the reference (Flemming et al., 2015).

- Page 7 Line 16: Check syntax of the references "(source of data Highwood (2009) and Hess et al. (1998))".

- Page 7 Line 29: Delete the point after the word "mixing".

- Page 8 Line 11: Finalize the sentence with "and particle volume are:".

- Page 9 Line 15: Include a closing point at the end of the paragraph.

- Page 22 Table 2: Complete the table with the "Aerosol model description" for the University of Aarhus WRF-DEHM model.

- Figure 2, 3, 5 and 8: The quality or resolution of the figures is low for final publication.

References:

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