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Interactive comment on "Coupling aerosol optics to the chemical transport model MATCH (v5.5.0) and aerosol dynamics module SALSA (v1)" *by* E. Andersson and M. Kahnert

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Below you will find our answers (in black) together with the original comment (in blue). The references to the pages and line numbers are to the supplemented document (which also contains this answer).

The authors describe the implementation of a new aerosol optics model into the atmospheric chemistry transport model MATCH. They compare simulation results with this new optics model to results with the previously implemented one. In order to assess the significance of the differences in simulated optical properties for estimates of the aerosol radiative forcing and for data assimilation, they discuss simulations with

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the new optics model in combination with the former bulk aerosol treatment in MATCH and in combination with the more recent aerosol dynamics model SALSA (as part of MATCH). The differences between the simulations with and without aerosol dynamics serve as a reference to evaluate the significance of the impact of the different optics models.

From my point of view, the topic falls well within the scope of GMD, the idea of this study is sound, and it is well motivated. I think the aerosol modeling community will benefit from this study once the following comments to the authors are adequately addressed.

Thank you for taking your time reviewing our paper. Your comments have been most valuable to improve our paper and we specially thank you for being thorough and constructive in your comments. Below you will find your comments in blue together with our answers in black for each comment in major, minor, specific and typo section. Following the comments and answers, a document highlighting the differences between the original manuscript and the revised manuscript is added. To which we also refer in the answers.

1 Major comments

We will answer the first three comments together, since they are interrelated, and since the reviewer presented these comments as three different alternatives to improve the manuscript.

1. In order to assess the significance of the differences between the simulations, I think it is necessary to provide some measure of uncertainty or variability in the simulated aerosol optical properties and radiative effects, depending on the simulated aerosol (component) mass mixing ratios. From my point of view, an analysis of the involved nonlinearities that the authors mention in the Conclusions cannot

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be completely deferred to a later study, as they also impact the assessment of the significance of differences between simulations with aerosol (optics) models of different complexity. If, as the authors state, the "test cases [may not be] in any way representative for typical aerosol and black carbon loads", then the study seems to me like a purely academic exercise.

2. Additionally (or maybe alternatively, as this would also provide a backdrop against which to gauge the significance of the discussed simulation differences) the study would benefit from comparisons with observations, in order to put the aerosol optics and dynamics impacts into a context of simulation-observation differences. AERONET and various satellite data sets come to mind here, and lidar observations could probably also be used.

3. A third option that might help make the conclusions a bit more robust would be to run another simulation with the old optics model in combination with SALSA and check if the MT-SALSA differences (using the old optics model) and the EXT-CGS differences (using SALSA) are similar to those discussed in the manuscript.

We first want to respond to the reviewer's concern about the representativeness of our study (comment 1). Although we picked only four locations and two points in time, we *did* choose these cases in order to cover as different situations as possible (two points over land, one north and one south of the Alps; two points over water, one in the north, one in the south; one summer and one winter day). The main goal we pursue with our approach is not only to quantify the differences between the two optics models, but also to show where those differences come from. We start our analysis by considering how morphological differences in the model particles cause differences in single-scattering optical properties. Then we investigate how those differences impact the radiative fluxes throughout the atmosphere, and how that, in turn, impacts the TOA radiative forcing. We want to help the reader appreciate that subtle microphysical properties impact radiometric properties of particles, particle populations, and macroscopic media. This

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chain of physical processes would remain obscure in a statistical analysis; it can only be revealed in a detailed analysis of selected cases. We are aware that this may be mind-boggling and demanding for the reader, and that a simple statistical analysis would be much simpler to follow. But we also think that our approach is quite valuable, because we do not just tell the reader what kind of errors one may introduce by using very simple optics models, but we show why this is so.

We have deleted the misleading part in Sect. 3.1 regarding the representativeness of the selected cases.

Now we turn to the reviewer's main concern. As we understand it, the reviewer wants more facts to strengthen our main conclusion, namely, that the choice of optics model can have a significant impact on the calculated radiometric properties. The reviewer makes three alternative suggestions, 1. Provide a measure of uncertainty or variability (also mentions analysing the non-linearities of the optics model); 2. Perform comparison with observations; or 3. Perform additional computations with the old optics model implemented into SALSA.

We performed additional computations and analyses that combine some ideas of 1. and 3. More specifically, we implemented, as the reviewer suggested, the old external-mixture/homogeneous-sphere optics model into SALSA and repeated the computations. Further, we computed mean errors for different optical properties over the entire geographic region and averaged over a whole month. The results are presented in a new table (Table 4 in the revised version), and discussed in a dedicated section (Sec. 3.1 in the revised version). The statistical analysis confirms the essential conclusions of this study. We believe that this addresses the reviewer's main concern by supplementing our study with more quantitative information on the differences between the optics models. We also make it clear in the revised version that the analysis of averaged optical properties mainly serves to draw some general conclusions, while the case-studies mainly serve didactic purposes to explain *why* different optics models can give

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different predictions for radiative fluxes.

We did not analyse the non-linearities of the optics model; even a crude analysis would probably take several months of work. However, we are currently applying for funding for a dedicated project that will deal with just this problem. Also, we do not wish to add a comparison with observations to this study. This would completely change the character of this paper. As chemical transport models have numerous sources of error and biases, a meaningful comparison with measurements comparison would require a comprehensive analysis of all model uncertainties. This would completely shift the focus of this paper away from the optics model toward various other error sources in MATCH and SALSA, which are not the subject of this study.

4. Please check the publications by Jacobson (2000), Matsui et al. (2013), and Klingmüller et al. (2014), and refer to them if and where appropriate.

   The paper by Jacobson (2000) is based on the use of the core-shell model, which underestimates the absorption cross section (which was, however, not known until quite recently). We added a reference to this paper in the discussion of our optics model in Sect. 2.3.2 (p. 13, l. 346). We have also added the references Matsui et al. (2013) (p. 15 l.407), Andersson et al. (2015) (p.4 l. 104, p. 7 l. 216, p.15 l. 407) and Klingmüller et al. (2014) (p. 4 l. 97) as well as Kokkola et al. (2008) (p.4 l. 104).

5. Please give some details on the model setup, e.g., model domain, horizontal and vertical resolution, etc.

   This information has now been added to Sect. 2.2 (p. 5 l. 139-145).

6. As far as I understand, the effects of including more detailed assumptions on internal particle structure are only studied for BC-containing particles, i.e., not for dust-containing particles, for instance. Please clarify this throughout the manuscript.

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The paper does have a dedicated methodology section (Section 2). In subsection 2.3.2 we give a very detailed, itemized, and unambiguous description of the optics model, which we believe leaves very little room for misunderstandings. Therefore, we would prefer not to repeat the model description in other parts of the paper, for the sake of conciseness.

7. Section 3.1.1 requires improvement.

   - Please expand on the results shown in Table 3:
     - What do you mean by "the same behaviour"?
     - Are the reasons for this "same behaviour" also the same?
     - What about Northern Italy in winter?

     We have re-written the whole result section as well as the mentioned paragraph in a, hopefully, clearer way, This paragraph can now be found at p. 22 l. 605-612.
   - What is a "dominant feature" (p. 10754, l. 12) if the "differences [...] are almost negligible" (p. 10755, ll. 4 and 5)?

     We re-wrote this sentence as follows: "Again, differences in ΔF_{net} at TOA are mainly caused by corresponding differences in the upwelling diffuse radiative flux ΔF_{u}." See p. 22 l. 618-619.
   - The paragraph on Fig. 8 confuses me:
     - How do you arrive at the conclusion that the MT-SALSA difference is "not as prominent" over the Mediterranean as over Northern Italy, given that in the next paragraph you state that this difference is small at all locations except the Mediterranean?

       This was indeed confusing, because in the first paragraph we referred to absolute differences, but in the following we mean relative differences (but we did not say this clearly). We have now made this distinction

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throughout the text. In addition to this comment and the next item of this comment, we have changed the paragraph about black carbon forcing in the context of comparing MT and Salsa, completely. Figure 8 is now also referred to as figure 10 according to specific comment 26. See p. 23.

- The word "difference" appears very often in this paragraph and refers to different contexts. This makes the discussion hard to follow.
  We agree that this paragraph was confusing and little helpful. The main point is that the two models are similar, because the optical properties do not differ appreciable in this case. We emphasised this fact in the revised version, and deleted the rest of the text.
- Please expand on the "multiple scattering effects".
  Reference to multiple scattering effects has been removed — see preceding item.

- Please expand on the differences between summer and winter. How can a larger difference between the MT and SALSA simulations be tied to a larger difference in the BC mixing ratios if the summer ΔF_{net} difference is greater over the Mediterranean, where the BC difference is smaller, than over Northern Italy?

  When taking a closer look at the concentration differences (not shown), then it becomes clear that the sensitivity of the forcing rate to concentration differences is rather low. Thus, when we have differences by a factor of 2–10, this has a noticeable effect on the forcing rate. But the tiny differences we see in summer between the two models over Northern Italy and the Mediterranean has no clear effect. We have re-written and expanded the discussion of the table accordingly. See p. 23 l. 659-671.

8. In many cases the language of the manuscript does not seem precise enough to me. For (some) details, see the Minor comments and Specific comments below

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We did our best to improve the preciseness of our language (see our response to the minor comments).

2 Minor comments

1. What are the "known and important effects from using aerosol dynamics"? (Quoted from the Abstract, but also appears in the Conclusions in similar form.)

   We rephrased the abstract and conclusion as well as added references to make our statement more elaborated and robust. We also realise that the term "aerosol dynamics" is a broad and non-specific term, therefore we have exchanged this term to the more proper "aerosol microphysics". This will help us address the vague formulations that the referee pointed out.

   The statement refers to the fact that the microphysical processes involving aerosol particles (aerosol dynamics) strongly affects their size distributions and mixing state, as well as their direct and indirect radiative forcing. This has been investigated by several studies, among others Matsui et al. (2013) that looks at how ageing processes affects radiative processes together with different mixing states and Kokkola et al. (2008) as well as Andersson et al. (2015) describes and evaluates an aerosol microphysical module in a chemical transport model.

2. In order to avoid confusion concerning different types of atmospheric or climate models as much as possible I would recommend to follow the terminology outlined in Dameris and Jöckel (2013)

   Thank you for noting the terminology of model types. We have now used the term "Chemistry-Climate" model instead of "Earth system" model throughout the text. The changes occur on p. 1 l. 4, p. 2 l. 30, 34 and 50, and p. 53 l. 836.

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3. I would suggest to replace most, if not all, occurrences of "aerosols" by an appropriate choice of "aerosol", "aerosol particles", "aerosol populations", "aerosol components" or similar, as the term "aerosol" technically does not only refer to the particulate phase, although it is often used this way in spoken language.

   We have searched through the whole document and replaced most occurrences of "aerosols" with either "aerosol particles", "particles" or "aerosol components".

4. Furthermore, there is often a distinction between "aerosol" and "black carbon", although the latter is of course a component of the former.

   Black carbon is, of course, part of the aerosol phase. In the text we do talk about both aerosol particles and black carbon particles. In the introduction and methodology sections we introduce and describe the importance of model aerosol particles mass concentrations, size distribution and mixing state on the remote sensing and climate related properties. We continue with distinguishing the black carbon particles, since those are the particles we focus on improving with the new aerosol optics model.

   Later on we continue with a distinction between "aerosol particles" and "black carbon particles" when we in the model have "removed" all the aerosol components or only the black carbon components. This to study "aerosol radiative forcing" and "black carbon radiative forcing". We believe that it is this latter distinction we might have caused some confusion to the reader. However, due to the reformulation of "aerosols" (from one of the above comments) we believe the issue will become clearer. We also reformulated appropriately by adding "all aerosol components" instead of just "aerosol components" to p. 19 l. 528, p. 20 l. 551, 554-555 and 555-556, where we start to present the results for the aerosol radiative effects.

5. Please be specific as to what is compared, evaluated, mixed, etc.: is it the model, the simulation output, the size bins, the aerosol components, etc. For instance, "comparing the differences in the optics models to other sources of error" (p.

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10739, l. 9) should be replaced by something like "comparing the differences between simulations with the two optics models to errors from other sources".

Thank you for being thorough with our text. We have now changed the text to "comparing the differences in the optics model output to other sources of error", see p. 4 l. 100-101.

6. I would prefer if you referred to table contents rather than to table rows (e.g., p. 10743, ll. 9 and 10; p. 10759, l. 21)

   The first example on p. 10743 an ll. 9 in the discussion paper does refer to table contents and not to rows and columns. However, the second example does refer to rows, which we have changed in the revised edition to "in June (summer) for all four geographical locations and in December (winter) for the locations Poland and North Sea", see p. 32 and l. 785-786.

7. Please avoid generalizations. For instance, what is "required" (p. 10737, l. 19) or "over-simplified" (e.g., p. 10760, l. 4) certainly depends on the application.

   We have looked into the occurrences of the more generalising words and we have changed the following; on p. 2 l. 53. Further, we have removed all instances of "over-simplified".

8. Please define technical terms at first use, e.g. external vs. internal mixture (defined later), backscattering coefficient (defined later), effective radius (not defined at all), etc.

   Thank you for pointing this out. We have now restructured and rephrased the text slightly in order to avoid confusion with newly introduced terms. The effective radius is now defined with its corresponding equations together with its introduction at page 21 and line number 590. The external mixture assumption is now explained on p. 3 l. 68 where it is first mentioned. The internal mixing is now defined on p. 3 l. 81, and for the optical properties, we have added an appendix

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(Appendix A, B and C) describing how these parameters are derived in the model set-ups.

9. Please keep in mind that an interested reader should in principle be able to reproduce your simulations. For instance, this requires complete information about the size distributions assumed for emitted particles.

   Thank you for pointing this out, we have now added appropriate references and extra information. Regarding the different chemical transport versions, i.e. with and without aerosol microphysics, they are fully described in Andersson et al. (2013) and Andersson et al. (2007), respectively. Regarding the size distribution assumed for the emitted particles, they are written in Table 1 which now also includes the appropriate reference, Andersson et al. (2007). For the MATCH set-up with aerosol microphysics, the emission size distribution is directly refereed to with a specific citation, table 4 and figure 6 in Andersson et al. (2013), on p. 7 and l. 193-194.

10. I would appreciate if table and figure captions contained more, and more specific information.

    We realise that too little information was put in the caption of many of the figures and some of the tables. Therefore, we have now added more specific information regarding table 3, 4 and 5 as well as figure 6, 7, 9, 10 and 11. More information regarding the time of the chosen events as well as the content of the table and plots has been added.

11. SALSA bins are not only distinguished by size, but also by composition. The term "size bin" therefore seems inappropriate, or at least incomplete.

    Regarding the terminology we could have used the term "size composition bin", but we have chosen to follow the terminology from Andersson et al. (2015). See also the answer to minor comment nr. 13.

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12. I find it difficult to understand Table 2 without some further expansion of Sect. 2.2.2. Some of the questions that come to mind are:

    - Why are the smallest particles assumed to remain dry?
    - Could you give an example for externally mixed PNHx particles?
    - Why does PNOx appear only in internally mixed particles in one single size bin?

    In order to make Table 2 more understandable, we have extended the section about the aerosol microphysics module SALSA, section 2.2.2, page 6-7. The specific questions are addressed below:

    - Table 2, it is stated that the smallest size bin of particles that remains dry, i.e. that are not included in cloud droplet activation is size bin 3 with a radius of 9-25nm. As stated in McFiggans et al. (2006), particles with radius r < 40nm are not involved in cloud droplet activation. Our next size bin in MATCH-Salsa is 25-49nm and therefore includes non-dry particles.
    - An example for externally mixed PNHx would be pure ammonium sulphate, $(NH_4)_2SO_4$.
    - PNOx only appears in single size bin because the development of PNOx-chemistry in MATCH-SALSA is still in a very early stage. As noted in the specific comments, a more elaborated description of PNOx is desired; we have added a comment to section 2.2.2, p. 7 l. 190-195.

13. Furthermore, it should be mentioned in Sect. 2.2.2 that SALSA explicitly tracks particle number mixing ratios.

    What MATCH-Salsa tracks is particle number concentrations, i.e. number of particles per volume of air per size interval. This has been added in the section 2.2.2 on p. 6 l. 186-187.

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14. The term "bin" should be used consistently. In Sect. 2.3.1, for instance, three different words are used: "class", "bin", and "mode".

    We understand that different terminology might confuse the reader. We have revised the text and consistently use the term "bin" now, which is consistent with the literature, e.g. Andersson et al. (2015) and Kokkola et al. (2008). However, we still do use the term "mode" when appropriate, i.e., when referring to a modal model, such as one using a log-normal size distribution. The changes occurred at: p. 6 l. 174, p. 8 l. 231 and 232, p. 9 l. 269, and p. 14 l. 371.

15. Sects. 2.3.1 and 2.3.2: It could be stated more clearly that the external mixture assumption is only used in conjunction with the MT aerosol module and the mixing state-resolved optics model is used both with the MT scheme and with SALSA.

    In our new revised edition of the manuscript we have done as the reviewer suggested in major comment nr. 3, i.e., we added a fourth model set-up where we include the MATCH-SALSA CTM version together with the old EXT optics model (Salsa-EXT). This should be clear by the list on page 15 in the revised edition of the manuscript.

16. Please specify what is meant by the "total flux" (e.g., p. 10752, l. 13).

    We have now changed the formulation "total flux" to what we actually defined in section 3.1, the "net radiative flux" on page 20 l. 563 and p. 21 l. 1.

17. Vague formulations like "slightly", "somewhat", "quite" should be avoided as much as possible. Quantitative information is preferred.

    Thank you for pointing this out, we have now removed vague formulations in the text.

18. Sect. 3.1.2: If possible, please check (the inter-model differences in) ΔF_u as a function of wavelength to support your conclusion. Otherwise, please explain

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why the IR AOD effect should be "dominant" over the g effect. Furthermore, this seems to contradict what you state in the introduction to Sect. 3.1 (p. 10751, l. 16): "At other wavelengths (not shown) the optical properties behave similarly."

First, we removed the rather imprecise statement in the introduction. Secondly, we have added an appendix (Appendix E) with all the simulated wavelengths for the in depth analysed model set-ups (MT-EXT,MT-CGS and Salsa-CGS), which will support our statement in the section, where we discuss the possible impact on the black carbon radiative fluxes from different wavelengths and the two optics models (see p. 24 l. 706-708).

3 Specific comments

1. Correct citation format (possibly "citep" instead of "citet"): p. 10738, l. 18, p. 10740, l. 25, p. 10744, ll. 2 and 3, p. 10748, l. 7.

   Thank you for noticing these errors, they are now corrected for with the right type of citation format. See p. 3 l. 79, p. 5 l. 140, p. 9, l. 245 and 251, as well as p. 14 l. 377 in the revised edition of the manuscript.

2. p. 10739, l. 7: Please remove the word "rates".

   The word "rates" has now been removed, see page 4 line number 99.

3. p. 10739, l. 15: Neither of the optics models is actually evaluated here, at least not in the sense that its output is compared to observations.

   We follow the reviewer's advice and use a different formulation since "evaluation" is commonly used in the context of observations. Instead of "evaluation", we now use "comparison".

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4. p. 10740, l. 5: Probably "fraction" is supposed to mean "component" here, otherwise please specify the "whole" of which a fraction is discussed here.

   What is referred to as "The SIA fraction" is the secondary inorganic aerosol fraction of aerosol particles. This has now been made clearer in the text, see p. 5 l. 153.

5. p. 10741, l. 15: Please summarize briefly how the emissions inventory was generated.

   We have added some more detailed information on how we unified the EMEP gridded emissions with the non-gridded BC and OC emissions given by Kupiainen and Klimont in order to obtain gridded OC and BC emissions. See p. 5 l. 163 - p. 6 l. 173.

6. Sect. 2.2.1 vs. Table 1: Are PPM (incl. BC and OC) assigned to three (Table) or four (text) size bins?

   Other PPM, BC and OC are only assigned to three of the four available size bins. In the text we have stated that there are four available size bins, and in table 1 we list what is in each of the four size bins.

7. Sect. 2.2.1: The last two sentences should be moved up, where the corresponding modules are mentioned.

   Thank you, this has now been edited for. The changes can be found on p. 5 l. 155-157.

8. p. 10742, l. 7: Please summarize briefly what the simplified PNOx description is.

   We have added a brief clarification, stating that PNO_x in the current version is simply computed by the mass-transport model, and the mass is assigned to the SALSA-size bin No. 15. See page 7 and l. 190-192.

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9. Section 2.3.1: Please add a reference to Table 1, and add information on the hydrophilic/hydrophobic assumption for "other PPM".

   We have added a reference to table 1, Andersson et al. (2007), but we are not sure what the reviewer means when asking for more "information on the hydrophilic/hydrophobic assumption for "other PPM". However, we realised that we need to clarify what "other PPM" means. This has been done in the section 2.3.1 together with the description of the gridded EMEP emissions. See page 6 l. 172-173.

10. p. 10741, l. 15: Please state briefly how the Gerber (1985) parametrization is applied.

    We added the information that the parametrisation computes the wet radius as a function of dry radius, relative humidity, and temperature. See p. 9 l. 255-256.

11. p. 10744, l. 14: Please state briefly how the optical properties are interpolated onto intermediate water volume fractions.

    They are interpolated linearly, which we have added to the text as well, see page 9, l. 263.

12. Section 2.3: Please state clearly that optical properties are computed from the (effective) refractive indices.

    This is only true for homogeneous internal mixtures. Section 2.3 makes some rather general observations on optics modelling. We have looked through the text and wrote "effective refractive index" wherever appropriate, but we retained "refractive index" whenever no effective-medium assumptions are involved.

13. Sect. 2.3.1, 3rd paragraph: I find this confusing. If I understand it correctly, you explain how to get from mass mixing ratios, which you simulate, to particle number mixing ratios (but not number densities). In case this is correct, please

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restructure the paragraph accordingly, and add information on how to convert particle mass to particle volume. Furthermore, as single scattering albedo and asymmetry parameter are properties of individual particles (in contrast to aerosol optical depth and backscattering coefficient), please clarify that the particle number concentration/size distribution is required here for the averaging, rather than for the computation of the per-particle properties. Finally, the terms "mean radii", and "variances" are used incorrectly here. Please either give mean radii and variances, or use the appropriate terms: "geometric mean radii" and "geometric standard deviations"

We have added two sections in an appendix (Appendix A and B) that explain in detail how to convert mass mixing ratio to number *density* (not number mixing ratio!), and how to compute ensemble-averaged optical properties. Further, we explain how to compute radiometric properties of the medium, such as optical depth and backscattering coefficient, from the ensemble-averaged optical properties of the particles. The whole averaging procedure is a bit tricky when working with truncated size modes, as we do in the old optics model. For this reason, we did not want to include all this in the text, as it would be a bit of a detour away from the more essential issues. However, the appendix now provides all the equations necessary for the interested reader who wants to understand the details of the size-averaging process. We hope that this clarifies the reviewer's questions. We would also like to point out that single-scattering albedo and asymmetry parameter are not *exclusively* properties of individual particles, as the reviewer said. They can also be properties of a small *ensemble* of particles. To compute ensemble-averaged single-scattering albedos, asymmetry parameters, etc. the number density/size distribution is required just as much as for computing AOD and other radiometric properties of the medium. This should be clear now when reading the appendix. Finally, we have followed the reviewers advise and now use the terms "geometric mean radius" and "geometric mean standard deviation". See the whole paragraph starting on page 9 l. 267 and finishing on

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page 10 l. 281.

14. p. 10746, l. 2: Please specify how the averaging over particle orientations is achieved (e.g., analytically over all orientations, or using a sampling technique, etc.)

    We have added a sentence specifying that the orientation-averaging has been done analytically, and we added a reference to the paper by Khlebtsov (1992), who first derived the analytic expressions for non-axisymmetric particles. See page 10 and line number 310.

15. p. 10746, l. 15: Please specify "size" (i.e., measured as what).

    We have clarified this text by rewriting the sentence to "... were computed in the range of 100-500 nm (volume-equivalent radius)" on page 7 line number 198-201 in the revised edition of the manuscript.

16. p. 10747, l. 24: Please specify the core-shell partitioning parameter, and state whether this was taken from the given reference, or whether it was found in the same way. Furthermore, please comment on whether the parameter is/should be size- and/or composition-dependent.

    The parameter was taken from the given reference. It is independent of size, volume-fraction, and optical parameter, but it does depend on wavelength. We have provided this information in the revised text (see page 13 and 14, l. 357-365 blue marked text) and added a table (table 3 on page 14) with the core-shell partitioning parameter as a function of wavelength.

17. Section 2.3.2, item 2: Parts of the third paragraph should be moved to item 3, or to a separate one, as item 2 deals with the treatment of internally mixed BC. Furthermore, it is again unclear here how "other PPM" is treated. Please also state whether BC is treated the same way both in the core and the (gray) shell.

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To make this section more clear, we made a fourth item to the enumerated list containing the treatment of all internally mixed particles, except black carbon. See page 14 line number 379-380. We also added information that "other PPM" is interpreted as dust particles, see page 6 l. 172-173 and page 7 l. 211-212.

18. Section 2.3.2: As the optics model is the essential novelty of this study, I think the 28 wavelength bands, 37 discrete BC volume fractions, etc. should be given explicitly in this publication, maybe in an Appendix or in a Supplement. Please also state briefly how the interpolation is performed and add some mathematical formulas to the description of the size-averaging procedure (in the main text), so that it is easier to understand.

    The 28 wavelength bands and the refractive indices of each aerosol component are given in an extra table in appendix D, page 37. The 37 discrete BC volume fractions are now given in the text, page 14 line number 383. The interpolation is performed linearly, and the size-averaging is now explained in the new appendix.

19. Section 2.4: As stated above, I think this is not an "evaluation" in a strict sense. Furthermore, the second paragraph and the first sentence of the third paragraph actually belong to the Results section rather than to the description of the method. Please also comment on how clouds might affect your results, either here, or in Sect. 3.1.

    We replace "Evaluation" by "Methodology for comparing". We also moved the figure showing AOD over the model domain and the discussion of the figure to the results-section. We have only considered clear-sky situations. As we did not perform any calculations involving clouds, we refrain from speculating on the possible effects these and other complications would have on our results.

20. Sect. 3.1: Please mention that Fs and Fd are counted positive downward, and Fu is counted positive upward.

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We have added to the definition on page 20 and line number 534-537 that F_s and F_d are accounted for with a forward direction downwards and F_u upwards.

21. Sect. 3.1.1: This section could be more concise. Some sentences could be removed completely (this also applies to Sects. 3.1.2 and 3.2). Other parts should be more precise, or even expanded (cf. Major comment on this section). Please clarify (at least) the following points:

    - The magnitude of ΔF_u would also increase with decreasing altitude in case the AOD per layer was constant.
    - Aerosol extinction does not result in the generation of diffuse flux. Actually, the opposite is the case: the processes that generate the diffuse flux are responsible for the extinction.
    - The difference in "convertible flux" (last sentence of first paragraph) actually increases with altitude; it is its magnitude that decreases.
    - When you discuss "mass" you actually refer to "mass mixing ratios". (This appears in similar form in Sect. 3.3 ("mass densities").)
    - "Number density"" is actually what you call in in Sect. 2.3.1. What you refer to here is more likely "number mixing ratio".
    - Although the assumption is unavoidable in the optics model in connection with the MT module, aerosol number mixing ratio does not necessarily increase with the mass mixing ratio in reality. (This appears again in Sect. 3.3, 2nd paragraph.)

    - Yes, this is, of course true. We meant to say that the *sharp* increase in the magnitude of ΔF_s is caused by the *sharp* increase by the AOD per layer. This is now clarified in the revised text, p. 20 l. 556-558.
    - We disagree. The process that generates diffuse flux is scattering. Extinction is just the generic term for the combined effect of scattering and

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absorption. We reformulated the text to preclude misinterpretations; it now reads "extinction in the form of scattering results in the generation of diffuse flux", see page 20 line number 558.

- Yes, we changed the text accordingly.
- Yes, we are referring to mass mixing rations, since we use the figure with vertical profiles of aerosol particle distribution with the unit ppb(m). In section 3.3 we now use "mass mixing ratio" instead of "mass densities", page 30 line number 764.
- No, by "number densities", we mean particle number concentration (see answer to specific comment 13).
- Correct. We have rephrased our statement on page 21 l. 598-599.

22. p. 10757, l. 8: Please specify which two locations

    Thank you for pointing this out, we have now changed the sentence to "The black carbon forcing in Fig. 9 (Northern Italy) and (Mediterranean) 8 display different behaviours in radiative fluxes, comparing the EXT (blue) and CGS (red) model results.", see page 25 line number 714-716.

23. Sect. 3.2: As this still deals with the radiative forcing I would suggest to move it into Sect. 3.1.

    We have now moved section 3.2 to section 3.1.3.

24. Table 1: Please replace "wind blown" by "dust" or similar.

    We have changed "Wind blown" to "Dust" in Table 1.

25. Table 2: I suggest to sort the table by bin size rather than by composition.

    Table 2 is sorted according to the number of size bins, which has different size ranges, mixing states and composition. In order to change the table and sort it by

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the size alone would be rather confusing, since most size ranges appears more than one time. with different mixing states and compositions. We would therefore like to keep Table 2 as it is.

26. Figures:

- I suggest to increase the font size (everything except the titles in Fig. 11).
- As you mainly discuss "TOA" effects, it might be advisable to reduce the number of vertically resolved plots in Figs. 5-8 to those that are actually required to understand the discussion.
- It looks as if the plotted data in Figs. 5-8 was somehow averaged/interpolated in/onto 1 km bins/levels. Please comment on this in the text.
- Why do the lowermost plots in Figs. 7 and 8 show differences between calculations with and without BC instead of just the BC values?
- Please comment on the increase in BC ΔF_u with altitude as simulated with SALSA in the lowest kilometre in Fig. 8.
- Maybe replace "Bulk" by "MT" in Figs. 9 and 10.
- As sulfate and nitrate are not discussed in the text, they can as well be left out of Fig. 10.
- Following the structure of the discussion, I recommend to swap Figs. 7/8 and 9/10.

- The font sizes have now been increased for all the figures (except the titles). The font weight has also been changed to "bold" to make it more readable.
- We believe that all the vertically resolved figures (Fig. 5-8) are helpful for understanding our analysis, so we would prefer to not erase any of them.

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- Fig. 5-8 show the radiative fluxes together with the corresponding optical properties aerosol optical depth, single scattering albedo and the asymmetry parameter. The radiative transfer model we have used operates on these fixed levels. The optical properties have therefore been interpolated onto these levels. This is now mentioned in the text, see page 17 line number 466.
- The lowermost plots in Figs. 7 and 8 *do* show just the BC-values, but it is the differences between the solid and dashed lines one should focus on, because this is what we need in order to understand the forcing of BC. This was different for the total aerosol forcing in Figs. 5 and 6: to understand the total forcing of aerosols, we need to look at just the optical properties of aerosols. (In the absence of aerosols, the optical properties of aerosols are, trivially, zero!)
- We have thought about this, but did not find a satisfactory explanation for the increase in ΔF_u with altitude over the lowest kilometre in Fig. 8.
- We have now replaced the occurrences "Bulk" with "MT" for consistency.
- Sulphate is however mentioned in the text, whereas nitrate is not. Therefore we agree with the reviewer to remove nitrate form the figure showing the aerosol mass mixing ratios.
- We now follow the order of the discussion and have placed the figures of the effective radius and the profiles of aerosol mass mixing ratios before the black carbon forcing discussion.

4 Typos

- p. 10736, l. 19: effect → affect

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- p. 10739, l. 14 and l. 15: model → models
- p. 10740, l. 7: hydrophillic → hydrophilic
- p. 10745, l. 16: was → is
- p. 10745, l. 21: calculation → calculations
- p. 10751, l. 11: are → is
- p. 10751, l. 17: 6→5
- p. 10752, l. 4: Algier → Algiers
- p. 10752, l. 17: at higher altitudes → [delete]
- p. 10752, l. 24: then → than
- p. 10757, l. 1: downdwelling → downwelling
- p. 10757, l. 7: a → [delete]
- p. 10760, l. 3: differences → difference
- p. 10761, l. 4: an → and
- p. 10762, l. 4: extend → extent
- p. 10762, l. 9: acknowledge → acknowledges
- p. 10764, l. 4: Nousainen → Nousiainen
- Table 3: Polen → Poland

We have gone through the typos and corrected them accordingly.

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Please also note the supplement to this comment:
http://www.geosci-model-dev-discuss.net/8/C4298/2016/gmdd-8-C4298-2016-supplement.pdf

Interactive comment on Geosci. Model Dev. Discuss., 8, 10735, 2015.