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Geoscientific Model Development Discussions

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

Anonymous Referee #1

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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 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.

1 Major comments

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 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.

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.

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

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.

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?
   - What is a "dominant feature" (p. 10754, l. 12) if the "differences [...] are almost negligible" (p. 10755, ll. 4 and 5)?
   - 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?
     - The word "difference" appears very often in this paragraph and refers to different contexts. This makes the discussion hard to follow.
     - Please expand on the "multiple scattering effects".
   - 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?

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.

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.)

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 & Jöckel (2013).

3. I would suggest to replace most, if not all, occurences 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.

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

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

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).

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.

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.

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.

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

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

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 PNH_x particles?
    - Why does PNO_x appear only in internally mixed particles in one single size bin?

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

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

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.

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

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

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 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.”

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.

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

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.

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.

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

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

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

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

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

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

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

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

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 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”.

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.)

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

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.

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.

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.

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.

20. Sect. 3.1: Please mention that F_s and F_d are counted positive downward, and F_u is counted positive upward.

21. Sect. 3.1.1: This section could be more consise. 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_s 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 n_i 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.)

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

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

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

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

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 kilometer 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.

4 Typos

- p. 10736, l. 19: effect → affect
- 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

References

Dameris, M. & Jöckel, P. (2013). Numerical Modeling of Climate-Chemistry Connections: Recent Developments and Future Challenges. *Atmosphere*, 4(2), 132–156.

Jacobson, M. Z. (2000). A physically-based treatment of elemental carbon optics: Implications for global direct forcing of aerosols. *Geophysical Research Letters*, 27(2), 217–220.

Klingmüller, K., Steil, B., Brühl, C., Tost, H., & Lelieveld, J. (2014). Sensitivity of aerosol radiative effects to different mixing assumptions in the AEROPT 1.0 submodel of the EMAC atmospheric-chemistry–climate model. *Geoscientific Model Development*, 7(5), 2503–2516.

Matsui, H., Koike, M., Kondo, Y., Moteki, N., Fast, J. D., & Zaveri, R. A. (2013). Development and validation of a black carbon mixing state resolved three-dimensional model: Aging processes and radiative impact. *Journal of Geophysical Research: Atmospheres*, 118(5), 2304–2326.

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