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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of the modal aerosol scheme GLOMAP-mode on aerosol forcing in the Hadley Centre Global Environmental Model" *by* N. Bellouin et al.

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We thank Steve Ghan and the anonymous referee for their positive comments and constructive criticisms, which prompted improvements to the manuscript. In particular, aerosol direct forcing is now presented for each component individually, which facilitates the analysis of differences between the two aerosol modules, as predicted by the reviewers.

In the following, we respond to each of the reviewer comments, which are written in bold.

1 Responses to comments by Reviewer 1

This is a well-written description of a comparison between estimates of aerosol radiative forcing by two very different representations of the aerosol. All conclusions are supported by analysis.

Only one comment will require considerable effort: distinguishing direct forcing by each aerosol component. It is not essential, but would strengthen the message significantly.

Section 7 of the revised manuscript now discusses a new figure, reproduced below as Figure 1, which shows the direct forcing for sulphate, fossil-fuel black carbon, fossil-fuel organic carbon, and biomass-burning aerosols. Those component direct forcings support the analysis of differences in total direct forcing already included in the paper: sulphate direct forcing is weaker in CLASSIC than in GLOMAP-mode (-0.28 and -0.67 Wm^{-2}, respectively), while fossil-fuel black carbon direct forcing is stronger in CLASSIC than in GLOMAP-mode (+0.19 and +0.12 Wm^{-2}, respectively). Those differences can be readily traced back to differences in aerosol residence times. In addition, the more absorbing nature of GLOMAP-mode biomass-burning aerosols yields a direct forcing of 0 Wm^{-2} for that component, weaker than the -0.07 Wm^{-2} obtained in CLASSIC.

1. Line 113. Ghan et al., 2001 is not a sectional model, so remove this citation. It is a modal model (same as Easter et al., 2004), so you can cite it when discussing modal models if you wish.

Thank you, Ghan et al. (2001) is now a reference for modal aerosol modelling.

2. Line 125. Add reference to Liu, X., et al., 2012: Toward a minimal representation of aerosols in climate models: Description and evaluation in the Community Atmosphere Model CAM5. Geosci. Model Dev., 5, 709–739, doi:10.5194/gmd-5-

709-2012.

This reference is very relevant, and added to the manuscript.

3. Figure 1 should make it clear that number concentration is predicted for each mode.

"Mass and number" has been added to each of the 5 boxes describing the GLOMAP-mode aerosol modes, and the caption has been made clearer by writing "CLASSIC represents the mass of the external mixture..." and "GLOMAP-MODE simulates the mass and number of five aerosol modes..."

4. Line 358. Why does the parameterisation of wet deposition yield smaller sulphate wet deposition rates in GLOMAP-mode? Why isn't wet deposition treated the same in both models? Is this a result of the feedback of sulfate on precipitation?

Wet deposition is not treated in the same way because the representation of sulphate aerosol in CLASSIC includes a mode representing aerosols dissolved into cloud droplets (the "dissolved mode"). Dissolution pathways are either through instantaneous in-cloud nucleation of cloud droplets onto accumulation-mode particles, or diffusion of interstitial Aitken-mode particles into cloud droplets. Wet-deposited mass is taken naturally from the dissolved mode, in proportion of precipitated water. GLOMAP-mode does not include a dissolved mode, and must instead compute the amount of wet-deposited mass from precipitation diagnostics. The fact that wet deposition rates are in the end smaller in GLOMAP-mode than CLASSIC stems from the choice of parameters in the two schemes, and may change in the future if the parameterisations are calibrated differently.

The last paragraph of section 3 has been clarified by rewriting the sentence as: "Sulphate resides in the atmosphere two days longer in GLOMAP-mode than in CLASSIC because parameterisations of wet deposition differ between the two schemes, as dis-

cussed in the last paragraph of section 2 above, and the parameterisation used by GLOMAP-mode ends up yielding smaller deposition rates of sulphate aerosols."

5. Line 364. According to Table 2, the difference in BC lifetime is 10, not 12 days.

The discrepancy is now corrected: numbers in the Table are the correct ones.

6. Line 375. But the jet mode could influence the particle surface area, which influence the competition for water and hence the activation of accumulation mode particles. Compare the surface area of the modes and decide.

In this study, as described in Section 6, the empirical relationship of Jones et al. (2001) between cloud condensation nuclei and cloud droplet number is used. For CLASSIC, cloud condensation nuclei are equal to the aerosol number in the accumulation and dissolved modes: activation is not explicitly resolved. Therefore, in the CLASSIC representation, jet-mode sea-salt, and all other aerosol components, influences the indirect effect through its number only.

The manuscript has been clarified by rewriting the sentence as: "The jet mode is therefore optically inefficient for the direct effect, and provides little aerosol number for the parameterisation of the first indirect effect used in this study, described in section 6."

7. Lines 395-399. It does not appear that dust AOD has been removed from the AOD shown in Figure 2, and shouldn't be if the simulated AOD is to be compared with the assimilated distribution. So delete this sentence?

Mineral dust AOD has not been removed: rather, a common distribution is shared by all simulations in order to focus on actual CLASSIC/GLOMAP-mode differences. The manuscript has been clarified by rewriting the sentence as: "Although mineral dust differs between the HadGEM2 and HadGEM3 simulations for the technical reasons stated in Section 3, those differences are not relevant to this study and would divert attention from actual differences between CLASSIC and GLOMAP-mode. Therefore,

mineral dust AOD is hereafter taken from the HadGEM3 simulation only."

8. Line 524. For which measure of mode radius are the optical properties determined? Surface mode radius?

The modal radius is for the size distribution of the number. This is now clarified in the manuscript.

9. Figure 4. Is the SSA the column mean (AOD-weighted) or the value at a particular level?

The SSA is computed here as the ratio between column-integrated scattering and extinction AOD. This is now clarified in the figure caption.

10. Lines 734-756 and Figure 7. It should not be difficult to distinguish between the direct forcing by each aerosol component (sulphate, black carbon, organic carbon). Just call your radiation scheme with each component zeroed separately. This provides valuable information about the cause of the difference in the total direct forcing, and just requires one more set of runs (1850 and 2000 emissions) for each model. An example of this is Ghan et al. J. Climate, October 2012.

and

11. Lines 865-868. The longer residence time of BC in CLASSIC would also raise the direct forcing. It would be helpful to have separate estimates of forcing by BC and sulfate.

This has now been done: see response to your first comment, above. Thank you for the suggestion.

12. Lines 911-912. The correct reference is Abdul-Razzak and Ghan (2000), which is designed for modal schemes.

Thank you, the correct reference is now used.

2 Responses to comments by Reviewer 2

The paper from Bellouin et al. presents a clean and well documented comparison between two aerosol model schemes in almost the same host model. This is very valuable to understand differences between aerosol models in principal. The presentation, the graphics, the tables are in good shape. However, I have some questions, recommendations and objections, which the authors should take up to make this paper complete. In the present form I find it a little bit too optimistic with respect to the quality of the GLOMAP model. It should be published in ACP after revisions as requested below.

The statements regarding the advantages of using GLOMAP-mode over CLASSIC have been toned down, as described below.

Abstract : P21438, Line 10 : Missing that only AOD is compared.

The abstract now reads "HadGEM simulations of the aerosol optical depth..."

Line 12: why are the residence times different?

The abstract now reads "Because of differences in wet deposition rates, GLOMAP-mode sulphate..."

Line 13: Under estimate can also be due to organic Aerosol or nitrate. I do not see a thorough enough evaluation of absorption in this paper.

The statement needs to be taken in the context of longer residence time of fossil-fuel black carbon in CLASSIC. Evaluating absorption is difficult, especially in the Arctic

where high-quality AERONET sites providing inversion of single-scattering albedo are lacking. For this reason, we think the statement "likely overestimates absorption in remote regions" is cautious and fair.

Line 22: Why is the cloud susceptibility different? Make this more clear... I thought the meteorological model was the same. I think the definition of cloud susceptibility is partly independent of the aerosol background. Clouds can have different susceptibility for the same background aerosol.

This is a good point, as "cloud susceptibility" is defined in the paper as the change in cloud albedo due to a change in CDNC, which is indeed independent of the aerosol. The manuscript then goes on using the same term when referring to the change in CDNC in response a change in CCN, which does depend on the aerosol module. This is unhelpful, and "cloud susceptibility" is changed to "ability to change cloud droplet number concentrations" throughout the paper, where appropriate.

Line 28: I do not agree that the results really show that CLASSIC lacks sophistication or realismn.

and

Page 21439, line 3: Again, I don't see that "those findings" "improves the fidelity in aerosol forcing". It might well be, but that would require more arguments to conclude so.

Note that we used the verb "suggest" in both instances. In the absence of strong observational constraints on radiative forcing, it is difficult to judge objectively which of the two schemes does the better job, especially for aerosol-cloud interactions. However, it is reasonable to expect that GLOMAP-mode, being able to represent the processes that modify aerosol mass without changing aerosol number, is a step in the right direction.

P 21441, line 18: I wonder if one could tune the CLASSIC scheme in such a way, that it outperforms GLOMAP with respect to AOD. I think this should be discussed. AOD is a limited constraint for a multi-component, multi-size model.

AOD is a convenient metric as it is routinely observed, but we agree that it only provides constraints that are limited to direct effects. In addition, GLOMAP-mode was not expected to dramatically outperform CLASSIC when simulating the AOD: the ability to support internal mixtures, or simulate aerosol mass and number independently, is not a key driver here. However, the situation is very different for aerosol-cloud interactions, as described in the paper.

The introduction now includes the statement: "GLOMAP-mode is expected to better represent the physical mechanisms involved in aerosol-cloud interactions, while maintaining the quality of the simulation of aerosol optical depths and direct effects."

P 21442, line 13: Maybe insert right here that you do not activate nitrate in CLASSIC.

Agreed, in the list of aerosol species provided by CLASSIC, ammonium nitrate is now followed by "(which is not activated in this study)".

P21443, l 24: if SOA is diagnostic in CLASSIC, what is then done exactly for biomass burning aerosols?

The CLASSIC diagnostic SOA only represents secondary aerosols from biogenic emissions, this is now clarified in the manuscript. Freshly-emitted biomass-burning represents primary aerosols only, and condensation of VOCs is simply represented by an increase in mass upon ageing.

P21443, l 28: what is a "dissolved mode"?

The manuscript now reads "Because CLASSIC uses so-called dissolved modes to independently represent the aerosols that are dissolved in cloud droplets (Jones et al.,

2001), CLASSIC wet deposition..."

P21445, l 25: ....but dust differences will propagate to AOD. Not clear in the manuscript how dust is impacting the AOD evaluation. Also Why is the lifetime of dust different in the two aerosol schemes?

The way differences in mineral dust AOD are accounted for when comparing against AERONET and MACC is now clarified, see response to Reviewer 1's 7th comment, above.

Dust residence time is different in the two simulations because the size distributions of emitted particles are different. This is now briefly mentioned in the manuscript.

P21447 , l 15: you mean inter annual variability?

Yes, this is now clarified in the manuscript.

L20: the ratio in dust burden is 3.5, the AOD ratio is 2.2. Why is there this difference?

Because mineral dust mass is distributed differently across size bins, which explains the differences in residence time, as stated above, and also affects the optical properties of that aerosol.

L24: dust is removed from CLASSIC and GLOMAP ? and then for evaluation against MACC and Aeronet added again? Unclear. It should be left in for all comparisons. Or did you remove dust from MACC?

Please see the response to Reviewer 1's comment number 7, above.

P21449, l14: It would be interesting to know also the mean bias at Aeronet sites, not just the RMSE.

The paper now gives the bias and absolute bias for CLASSIC and GLOMAP-mode, averaged across all AERONET sites. Bias is -0.058 for CLASSIC and -0.039 for GLOMAP-mode, indicating the tendency of both schemes to underestimate the total aerosol optical depth. Absolute bias is 0.074 for CLASSIC and 0.065 for GLOMAP-mode. Note that GLOMAP-mode is the better model, in agreement with results for RMSE and correlation.

P21452, l12: I do not find this absorption evaluation convincing. I disagree that the conclusion on the superior quality of GLOMAP in the last phrase can be drawn here.

We acknowledge that the evaluation of absorption is less extensive than that of total aerosol optical depth, but this is partly justified by the decreased coverage and increased uncertainty of ground-based retrievals of aerosol single-scattering albedo compared to optical depth. The last statement of section 5.1 was toned down to: "GLOMAP-mode represents the contrast between South American and African aerosol absorption better than CLASSIC."

P21454, l5: While cloud phase production might falsely produce too many particles, DMS is not producing new particles through nucleation in CLASSIC ! Compensation of errors? Please discuss and quantify better.

The CLASSIC aerosol scheme does not simulate particle number (a fixed size distribution is assumed), and thus does not simulate nucleation. As shown in Fig. 5, there is no compensation of errors, and CLASSIC overpredicts the magnitude of the CCN response in comparison to GLOMAP-mode. GLOMAP-mode fully accounts for aerosol microphysical processes, and thus provides a more realistic representation of the fate of the sulphur from the DMS perturbation. New CCN in GLOMAP-mode are formed via nucleation and condensational growth with the extra sulphuric acid from the DMS perturbation. As noted in the text however, the majority of SO_2 in the marine boundary layer is oxidized in the aqueous-phase, and is not available for nucleation nor conden-

sational growth.

Section 6.1 of the manuscript is revised as follows: “The CLASSIC scheme, which does not simulate aerosol number and assumes a fixed modal distribution for sulphate aerosol, is only able to respond to an increase in sulphur by directly increasing the CCN number.” Also, to better document the response in GLOMAP-mode: “The GLOMAP-mode response is more realistic, as aqueous-phase oxidation only grows existing CCN, and does not create new CCN (Woodhouse et al., 2012). Note however that GLOMAP-mode does create a limited number of new CCN via nucleation and condensational growth resulting from the extra sulphur.”

P21456, l 25; mitigated???? mixed arguments, eventually expand sentence.

The sentence has been rewritten as “Differences in global-averaged direct forcing between CLASSIC and GLOMAP-mode are partly reduced by the positive forcing due to black carbon at high latitudes in CLASSIC, and the positive forcing exerted in cloudy sky by the more absorbing biomass-burning aerosol in GLOMAP-mode.”

P21458, l21: I don’t see how the comparison “highlights problems of CLASSIC to represent growth”? Which problems? I don’t think much is shown in the respective paragraphs. Is the size distribution wrong in CLASSIC wrt data?

The statement has been slightly rewritten to make it clearer: “The comparisons also highlight how simpler mass-based schemes have problems representing growth processes which should increase aerosol mass without increasing aerosol number, such as aqueous sulphate production.”

L25: unless you evaluate the number concentrations and cloud susceptibility in climate models one can not draw this general conclusion on the failings of mass based schemes. The mass based model could have been a priori correct wrt number concentrations on average.

We find the statement to be a reasonable summary of the results described in Section 6.1. Processes where aerosol mass increases but aerosol number does not change have been observed and their mechanisms are well understood. If an aerosol model is not able to represent those processes, then it lacks the sophistication needed to study some Earth System feedbacks, such as the sensitivity to changes in dimethyl-sulphide emissions. The development of the more complex modal schemes has been motivated in part by those failings.

P21458, l19: I suspect that also the black carbon forcing is more positive in the classic model. Please explore and document bc and sulphate forcing.

Section 7 of the revised manuscript now discusses a new figure, reproduced below as Figure 1, which shows the direct forcing for sulphate, fossil-fuel black carbon, fossil-fuel organic carbon, and biomass-burning aerosols. As suspected by the reviewer, black carbon forcing is more positive in CLASSIC than GLOMAP-mode, at +0.19 and +0.12 Wm^{-2}, respectively.

P21460, l 5: How do you know that the wet deposition of sulfate is improved? You would need to evaluate against data.. It is not so clear how different the wet deposition really is in the two aerosol models.

We agree that the statement implies that wet deposition of sulphate in GLOMAP-mode needs improvements, which has not been proven in the manuscript. The statement is removed, replaced by “The addition of nitrate aerosols to the GLOMAP-mode scheme will also increase anthropogenic AOD.”

L28: “clear improvements”?? Maybe better “are better founded on physical principles of aerosol dynamics”

Agreed, the statement is rewritten as suggested.

Caption of Figure 1: All-sky shortwave direct radiative forcing at the top of the atmo-

sphere, in Wm^{-2}, for (top to bottom) sulphate, fossil-fuel black carbon, fossil-fuel organic carbon, and biomass-burning aerosols, as simulated by the CLASSIC (left) and GLOMAP-mode (right) aerosol schemes.

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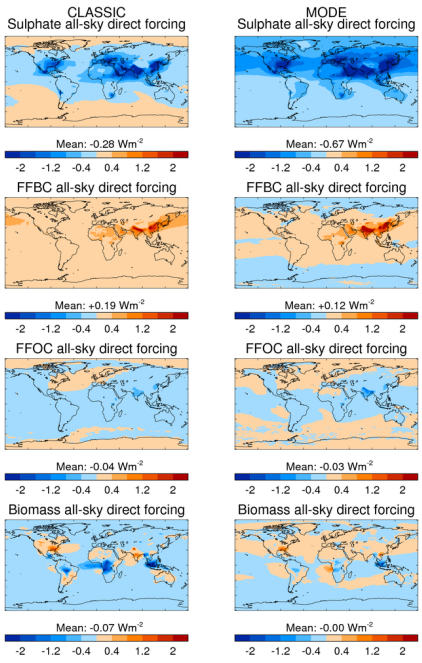

Fig. 1. See text for caption.