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Discussions

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Reply on RC1

Catherine Hardacre et al.

Author comment on "Evaluation of SO_2, SO_4^{2-} and an updated SO_2 dry deposition parameterization in the United Kingdom Earth System Model" by Catherine Hardacre et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-238-AC1, 2021

Reply to Reviewer 1

We thank the reviewer for their careful reading of the manuscript and their suggestions for improvement. The Reviewer's comments and our responses can be found below.

[Specific comments]

1. The crux of this study should be the impact of changing the dry deposition parameterization on the simulated tropospheric sulfur cycles. In order to serve for this purpose, the authors should beef up the contents of the manuscript in the model description and the discussion of dry deposition velocities. In section 2.2.1, the authors should provide a clear description of mathematical formulae employed for prescribing the leaf cuticle and soil resistances to SO2 uptake as a function of relative humidity and references for the basis of the employed formulae. Section 4 should begin with the discussion of simulated dry deposition velocities themselves (rather than the dry deposition fluxes) before and after the implementation of the new parameterization (see, for example, Ganzeveld et al., 1998).

=> We have now included an Appendix (Appendix A) to provide a more detailed description of the changes to the SO2 dry deposition parameterization. The reader is directed to this from Section 2.2.1. This appendix includes a table illustrating how cuticular restistance (Rcut) and soil resistance (Rsoil) in are represented differently in UKESM1 and UKESM1-SO2. We include the mathematical formulae for calculating Rcut as a function of humidity and clearly state the references used for these formulae. In keeping the technical details of the SO2 dry deposition modifications separate from the main text we aim to keep the manuscript more streamlined for the reader.

=> As suggested by the reviewer we have now included an analysis of the SO2 dry deposition velocities in UKESM1 and UKESM1-SO2. In section 4.1 we have included a new figure showing the annual mean, DJF mean and JJA mean for SO2 dry deposition velocities for UKESM1, and difference plots showing the impact of the modifications to the SO2 dry deposition paramterization on the deposition velocities in UKESM1-SO2. These results are then summarized in Section 4.1, paragragh 1.

=> In response the comment in paragraph 1 of the reviewer's General Comments, we clarify here that we do not mean to suggest that we have developed a new treatment of surface canopy wetness. Rather that this is an update for the SO2 dry deposition parameterization in UKESM1. We have reworded the text in Section 2.1.1 to clarify that we have adapted the findings from observational studies and applied them to UKESM1.

2. It is not clear whether other aerosol-climate and earth system models share some of the model biases reported in this study, which I hope the authors will touch on when revising the manuscript. The authors allude to inaccuracies in CMIP6 emissions as one of the error sources. It leads me to wonder if other models participating in CMIP6 exhibit the same problem as identified in this study. In addition, the authors need to elaborate the point of argument by Pope and Chipperfield (2021) regarding "total SO2 emissions in CMIP6 are moderately larger than the HTAP-OMI and EDGAR data sets" (L576-579).

=> With the exception of Aas et al., (2019) we are not currently aware of a study that has evaluated SO2/SO4(2-) across the CMIP6 ensemble. Aas et al. (2019) compare modelled and observed trends in SO2 and SO4 over the recent historical period, finding that the models do capture these trends over most regions. This is also the case for UKESM1 and UKESM1-SO2. We have added a snetence in Section 3.1, Para. 1 to highlight the agreement between out findings and those of Aas et al., (2019). However, it is not clear from Aas et al. (2019) how the models are are biased relative to SO2 or SO4(2-) concentrations. We note that in a communication from Stephen Smith in the open discussion of this manuscript that the CMIP6 emissions over the Western USA are too high, which we have highlighted in our revised manuscript, along with an elaboration on the statement regarding the differences between the CMIP6 dataset and the HTAP-OMI and EDGAR data sets (see Discussion, Para. 3). Further, Aas et al., (2019) also suggest that there are larger uncertainties in the emissions and representivity of the SO2 emissions over East Asia compared with Europe and North America, although the emission datasets used in that study were different from the CMIP6 emissions used in this study. We aim to evalaute sulphur species in the wider CMIP6 ensemble in a future study and this may shed some light on any systematic bias in the CMIP6 SO2 emissions dataset.

3. The analysis of the present model results will become much stronger if the authors can dive deeper into the metrics of model behavior related to the budget of atmospheric sulfur compounds and its changes with the revision of the dry deposition parameterization. This will allow us to grasp the broader context of this study. For example, the authors could calculate the regional lower-tropospheric budgets of SO2 and sulfate following Chin and Jacob (1996, Figs. 2-3) or re-iterate the global budget of SO2 discussed in Mulcahy et al. (2020, Tables 4-5) with possible extension of the comparison with yet other models. The point is that knowing the proportions of SO2 lost via dry and wet deposition and via oxidative conversion to sulfate provides a more in-depth measure of UKESM1's performance in its sulfur cycle. As it stands in the present version of the manuscript, this aspect is discussed only qualitatively. Another useful metric would be the SO2 lifetime and its seasonal variations estimated from regional SO2 vertical column densities and emission intensities (e.g., Lee et al., 2011, Fig. 2; Buchard et al., 2014, Fig. 3).

=> We have now calculated the SO2 budget using a 2 year AMIP simulation covering the period 1981–1983 inclusive. The SO2 budget for UKESM1 is in good agreement with that shown in Mulcahy et al. (2020). The budget for UKESM1-SO2 clearly shows the impact of the dry deposition modifications on the SO2 burden, SO2 lifetime and the deposition and oxidation processes. The SO2 budget is presented in Appendix C, with the main findings summarized in Section 4.1, para. 2 and discussed in the Discussion, para. 5 and 7.

4. The authors state that emissions from the energy and industrial sectors are all emitted into the first model layer (line 156), which seems to have been indicated by Mulcahy et al. (2020) as one potential weakness for the handling of this process in UKESM1. The injection of SO2 emissions from large stacks across several vertical layers above the lowest model layer (in lieu of plume-rise modeling) has improved the agreement of ground-level SO2 concentrations simulated by GEOS-5/GOCART model with observations in USA,

whereas the SO_2 vertical column densities did not change significantly (Buchard et al., 2014). The authors should refer to this finding when discussing the model evaluation against observed ground-level SO_2 concentrations. Perhaps it is too much to ask a new set of model runs for testing this emission treatment problem within the present study, but I am inclined to an idea that it can alleviate many problems identified in this study (high biases in the ground-level concentrations and dry deposition fluxes of SO_2 and low biases in the ground-level aerosol sulfate concentrations in USA and Europe).

=> The reviewer is correct in that varying the height at which SO_2 emitted in to the atmosphere, as opposed to emitting all SO_2 at the surface, can reduce model bias in surface SO_2 concentrations. This was demonstrated by Mulcahy et al. (2020) in their comparison between UKESM1 and HadGEM-GC3.1, although model bias in atmospheric SO_4 concentrations was not reduced in this study. In emitting all SO_2 at the surface in UKESM1 we maintain consistency with other Met Office model configurations, however, implementing a vayring emission height is a key development target for the model. We thank the reviewer for pointing us towards the study by Buchard et al. (2014) and we have cited this paper in the Discussion, para. 4.

5. The reactive uptake of SO_2 on dust aerosols can notably reduce the SO_2 concentrations and has a very large impact over China (e.g., Dentener et al., 1996; Liao et al., 2003, Bauer and Koch, 2005). It doesn’t appear that UKESM1 accounts for this process, hence another possible contributor to the model SO_2 bias especially over China.
The authors should justify the change of reference height from 50 m to 10 m for the computation of aerodynamic resistance, by explaining whether it comes with changes in the configuration of vertical layer thickness of the model. The reference height should be in general approximately half the thickness of the lowest model layer (e.g., Ganzeveld and Lelieveld, 1995, Section 3.2); if the lowest model layer thickness is substantially greater than 20 m (say, 40 m or greater), it calls for a strong rationale for using the reference height at 10 m. The authors should clarify the point of argument by Holtslag and De Bruin (1988) if UKESM1’s lowest model layer thickness is much greater than 20 m. Toyota et al. (2016, Section 2.2) gave a rationale in favor of Ganzeveld and Lelieveld (1995) for the choice of reference height from the mathematical formulation of aerodynamic resistance. Toyota et al. (2016) also noted that stability corrections applied for the computation of aerodynamic resistance are often inconsistent between dry deposition and host meteorological modules employed in the same model system. Does the use of the Holtslag and De Bruin (1988) function instead of the Dyer (1974) function reduce or eliminate this problem of inconsistency with meteorological flux calculation (i.e., u^* and L) within UKESM1?

=> The reviewer is correct in that UKESM1 does not account for the reactive uptake of dust on aerosols and we thank the reviewer for highlighting this potential source of bias in the model. Although we do not have specific plans to include this process in the model, it could be a target for future development.

=> We updated to Holtslag and De Bruin (1988) because it is considered a slightly more up-to-date set of equations for describing the fluxes in the boundary layer. In addition, by changing the the reference height from 50 m to 10 m we are making it more more consistent with the height of the lowest model level, which is 20 m in UKESM1. Unfortunately using Holtslag and De Bruin (1988) instead of Dyer does not increase consistency between meteorological stability functions and those used in UKESM1/UKCA. We are interested in updating the meteorological fluxes to also use Holtslag and De Bruin (1988) but this was not possible within this study. We agree with the reviewer that ideally there would consistency across the various flux formulations and we will aim study this, though changing the flux formulation across the meteorological subroutines is a major task.

[Technical suggestions]

1. L158-159: “Gas- and aqueous-phase oxidation of …”

=> Corrected as suggested

2. L113: Would you classify the gravitational settling as part of the wet deposition processes?

=> The approach for dry deposition of aerosol in GLOMAP-mode within UKCA (where UKCA is the chemistry and aerosol model in UKESM) is the same as that described in Section 2.2.2 of Mann et al.(2010) with a dry deposition velocity (Vd) for each aerosol mode given as the combination of a gravitational settling velocity (Vgrav) and one-over the sum of the aerodynamic and surface resistances (Ra and Rs) i.e. Vd = Vgrav + (1 / Ra + Rs).

(Mann et al., Description and evaluation of GLOMAP-mode: a modal global aerosol microphysics model for the UKCA composition-climate model, Geosci. Model Dev., 3, 519–551, https://doi.org/10.5194/gmd-3-519-2010, 2010.)

3. L126: “ares” -> “areas”

=> Thank you for highlighting theis error. Corrected.

4. L136: “to be developed” -> “being developed”

=> Sentence modified to "UKESM1 is the latest generation Earth System (ES) model developed in the UK." UKESM1 is the first generation of the model to be developed and was released in Jan 2020. As such it is not quite correct to say that this model version is still being developed.

5. L190: The soil pH is not taken care of in the model even after the revision to the parameterization, right? Please clarify. The authors may also want to cite Ganzeveld et al. (1998), which dealt with changes in the soil pH in their global tropospheric sulfur chemistry-transport model.

=> The reviewer is correct, soil pH is not accounted for in UKESM's SO_2 dry deposition parameterization.

6. L201: Garland and Branson (1977) reported the dry deposition of SO_2 to pine forest. Please correct me if I am wrong, but I cannot find the surface resistance of SO_2 on the water surface in this study.

=> Thank for highlighting this error. We have now used the correct citation i.e., Garland, J. A., The dry deposition of sulphur dioxide to land and water surfaces, Proc. R. Soc. Lond. A. 354, 245-268 (1977)

7. Figure 2 caption: “(a, c)” -> “(a, d)”, “(b, d)” -> “(b, e)” and “(c, e)” -> “(c, f)”

=> Thank you for highlighting theis error.Corrected.

8. L376, 485 & 488: “peninsular” -> “peninsula”

=> Thank you for highlighting theis error. Corrected.

9. Table 6 caption: Please come up with a better phrase for what “zonally averaged (median) time series” mean.

=> We have re-worded this caption to clearly describe what the data represents.

10. Figure 9: Change the figure title for “South East Asia” to “South to North East Asia”.

=> *******

11. L486: “9” -> “8”

=> Thank you for highlighting theis error. Corrected.

12. L503: “this aspect of THE change”

=> Thank you for highlighting theis error. Corrected.

13. L538: “NMB = 0.25” -> “NMB = -0.25” and “NMB = 0.43” -> “NMB = -0.43”

=> Thank you for highlighting theis error. Corrected.

14. L579: The authors need to be more specific about the data merging between OMI and HTAP.

=> We have now included a reference to the relevant citation Liu et al., (2018) in Discussion, para. 3 as we didn't create the data set, just explore it. If the reviewer or reader are interested the details of how the data set was derived, we suggest they follow the reference.

15. L690: “would BE beneficial”

=> Thank you for highlighting theis error. Corrected.