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Comment on acp-2021-238

Anonymous Referee #2

Referee comment on "Evaluation of SO_2, SO_4^{2-} and an updated SO_2 dry deposition parameterization in UKESM1" by Catherine Hardacre et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-238-RC1, 2021

[General comments]

Hardacre et al. report the impact of revising a dry deposition parameterization for SO2 in the earth system model UKESM1. The model behavior before and after revising the dry deposition parameterization is evaluated using ground-level observations of SO2 and sulfate aerosols in USA and Europe, dry deposition fluxes of SO2 estimated in USA and satellite measurements of SO2 vertical column densities over USA, Europe and South to Northeast Asia, including their temporal trends over the period from 1987 to 2014. Major changes in the dry deposition parameterization are the consideration of leaf wetness after rainfall and more than 100-fold decrease of surface resistance from 148 s/m to 1 s/m on the water surface. The authors state that the relative humidity dependence of leaf and soil wetness is also a new addition to the revised algorithm. Other changes, which the authors state are of minor importance, are the lowering of reference height for the aerodynamic resistance calculation from 50 m to 10 m and the use of an alternative stability correction formula again for the aerodynamic resistance calculation. Although I argue that the revised treatment of wet surface/canopy resistance for SO2 employed in this study is not necessarily very new (e.g., Pleim and Ran, 2011, Figure 2), the description of models in the literature often lacks clarity in the details of what they are doing in the dry deposition parameterizations. There are indeed ongoing efforts in the air quality modeling community to come up with measures for evaluating the behavior of sub-components of dry deposition parameterizations (e.g., Hayden et al., 2021; Galmarini et al., 2021).

The results of model runs before and after the changes in the dry deposition parameterization are thoroughly evaluated against observations. This is a strength of the paper. On the other hand, the authors do not delve into changes in the atmospheric budgets (or lifetimes) of SO2 and sulfate aerosols, which obscures the meaning of the present study for the aerosol-climate modeling community. Obviously, the motivation of the study comes from significant high biases found in the ground-level concentrations and vertical column densities of SO2 simulated by UKESM1. The new parameterization has alleviated this problem; however, it has degraded the model performance in the simulation of ground-level aerosol sulfate concentrations in eastern USA and Europe, biased low before and even lower after the implementation of the new parameterization. In addition, the modeled SO2 dry deposition fluxes in USA are biased high before and even higher after the implementation of the new parameterization. This may be a case where the status quo of compensation errors is broken in the representation of atmospheric sulfur cycles; in the discussions section, the authors list a number of

challenges that may need to be scrutinized in their forthcoming model versions.

The authors could make the paper more informative to the community if they would provide the global or regional (over SO2 source regions like eastern USA and Europe) budgets of SO2 and sulfate aerosols in the model and how they change before and after the implementation of the new parameterization. In addition, there is no demonstration of changes in the dry deposition velocities themselves. In my opinion, Section 4 and Figure 10 should be preceded by discussions and figures of how the new additions related to leaf and soil wetness affect the calculated dry deposition velocities across the globe and perhaps in different seasons. I recommend the authors to deal with the following aspects before the manuscript is accepted for publication.

[Specific comments]

- 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).
- 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).
- 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).
- 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 SO2 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 SO2 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 SO2 and low biases in the ground-level aerosol sulfate concentrations in USA and Europe).

- The reactive uptake of SO2 on dust aerosols can notably reduce the SO2 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 SO2 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?

[Technical suggestions]

L158-159: "Gas- and aqueous-phase oxidation of …"

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

L126: "ares" -> "areas"

L136: "to be developed" -> "being developed"

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.

L201: Garland and Branson (1977) reported the dry deposition of SO2 to pine forest. Please correct me if I am wrong, but I cannot find the surface resistance of SO2 on the water surface in this study.

Figure 2 caption: "(a, c)" -> "(a, d)", "(b, d)" -> "(b, e)" and "(c, e)" -> "(c, f)"

L376, 485 & 488: "peninsular" -> "peninsula"

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

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

L486: “9” -> “8”

L503: “this aspect of THE change”

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

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

L690: “would BE beneficial”

[References]

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