Reply to the reviewers' comments to ChAP 1.0: A stationary tropospheric sulphur cycle for Earth system models of intermediate complexity

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We are grateful for the reviewers for the constructive comments which led to the much improved presentation of our results. In addition, we are grateful to suggestions for future work – all they open important routes for improving our scheme.

The most important changes in the manuscript are as follows:

- The second referee's suggestions are discussed as limitations of the contemporary version of the scheme and noted as routes to improve it.
- In addition, an issue of distinguishing heavy and light rain in wet deposition calculations is noted and discussed.
- Some typos (in particular those found by the first referee) are corrected.

Below, the point-to-point replies to the comments and suggestions are listed. Original comments and suggestions are typed in italic.

Review 1

This reviewer made only one editorial comment, *"Numbering in Figs. 5-7 do not correspond to the figure captions – there are no designations d, e"* .
This was a misprint: items 'b' and 'c' in panel titles were erroneously duplicated. Upon revision, this misprint is corrected.

Review 2

Comments

- *I may have missed it but I didn't find the information about what time step was used in the simulations shown in the manuscript: if it is not present,*

could you please add it?
The stationary approximation embedded into ChAP removes the necessity to specify the time step, and time stepping is completely determined by the monthly mean forcing data. The corresponding note is added to Sect. 3 of the manuscript.

- *In the discussion of the limitations (Section 6), I think two major assumptions should be at least mentioned: that of the fixed lifetime of SO_2/SO_4 as well as the vertical length scale. While the values chosen appear sensible and in line with results from more complex models, the fact that the spatio-temporal variations of these parameters is not accounted for could have an impact on the results of ChAP1.0. The vertical length scale for example probably varies a lot between day and night (in clear-sky conditions), while the lifetime of SO_4 is heavily impacted by its main sink, wet deposition, and in turn by the occurrence of precipitation. It is possible that the tuning stage compensated partly for not taking these into account (and the other hypothesis outlined in Section 6).*
Apart from the apparently simplistic formulations of the conversion and deposition rates (see Sect. "Suggestions"), the first assumption was not used in the tuning procedure. Contrary to the previously available scheme for the tropospheric sulphur cycle designed for EMICs (Bauer et al., 2008), our scheme does not employ an assumption of fixed lifetimes for both $\mathrm{SO_2}$ and $\mathrm{SO_4}$. In ChAP, both lifetimes are determined by the conversion and deposition coefficients which depend on climate and on burden of the compounds coming from the earlier steps of chemical chains. We note to non-systematic variations of both lifetimes between different simulated time slices. The respective statement is added to Conclusions.
However, our scheme does employ an explicit assumption of fixed vertical scales for $\mathrm{SO_2}$ and $\mathrm{SO_4}$. We agree that it should be properly discussed as a limitation of the contemporary ChAP code. The respective discussion in Sect. 6 is extended upon revision. In addition, this limitation is explicitly listed in Conclusions, where it was missed in the previous version of the manuscript.

- *The tuning procedure (Section 4, line 240): where does the observed SO_4 burden per unit come from?*
Yes, this issue was missed in the previous version of the paper. In the revised version, it is stated explicitly in Sect. 4 that we used the CMIP5 sulphate burdens per unit area in place of $B_{\mathrm{SO_4,o}}$ in Eq. (16).

Suggestions

- *For* $\mathrm{SO_2}$ *lifetime, the authors may think of using the very simple parameterization from Huneeus et al. (2007), as a function of latitude only: (from Remy et al. : 2019): "The conversion rate (per second) can be* *written as*

$$C_0 = \frac{\exp\left[-\frac{\delta t}{(C_1 - C_2 \cos\theta)}\right]}{\delta t},$$

(16), where δt is the time step, θ is the angular latitude, and C_1 and C_2 are e-folding times in days representing the lifetime at the pole and the Equator set to 8 and 5 days, respectively, for operational cycles up to 43R1."
Yes, it would de a valuable option to parametrise an impact of the OH abundance on oxidation rate of SO_2. The respective note and a corresponding reference is added to Sect. 6 of the manuscript.

- *For dry deposition: to use different values over ocean and land (and possibly, ice/snow). That would be quite simple to implement and test and could give a bit more variability to the model.*
  We agree that it is sensible to prescribe dry deposition rate coefficients as a function of land surface type with a distinction between the open ocean, snow/ice, and land without ice and snow. This possibility is omitted on purpose in the present manuscript. The reasoning behind this choice is due to i) the neglect of the oceanic sources of sulphur which directly hampers tuning of $k_{\mathrm{SO_2,dry}}$ and $k_{\mathrm{SO_4,dry}}$ over the ocean, and ii) an attempt to demonstrate the ability of the present, simplistic version of ChAP to reproduce large-scale properties of the sulphur compounds distribution in the atmosphere. Nonetheless, we opt to try this option in future. The corresponding discussion is added to Sect. 6.

- *For wet deposition, to distinguish between solid and liquid precipitations, i.e. to split $k_{\mathrm{SO_4,wet}}$ in $k_{\mathrm{SO_4,wetrain}}$ and $k_{\mathrm{SO_4,snow}}$, and then compute $k_{\mathrm{SO4,wetrain,0}}$ and p_0 specifically for both rain and snow. Wet deposition by snow is generally much less intense than by rain, so this again could make a difference.*
  We agree that contemporary implemented formulation (Eq. (15)) does not distinguish between different precipitation types: light rain, heavy rain, and snow. Light and heavy rains show principally different efficiencies for removing hygroscopic aerosols from the atmosphere. Snow is an inefficient aerosol remover as well. The work to implement a distinction between different precipitation types in our scheme is under way and is expected to be implemented into the next version of ChAP.