Reply to reviewer #2

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We thank reviewer #2 for her/his comments and the detailed evaluation of our paper. Below, we repeat each comment (in blue) and address it (in black). Changes of text in the manuscript are written in *italics*.

- This manuscript comprehensively evaluates sulfur dioxide in the EMAC chemistry-climate model. This is done by first verifying that the sulfur cycle is closed, meaning mass is neither lost nor gained. The SO2 depositions and vertical column densities are then evaluated with a set of ground-based and satellite observations. The structure of the manuscript is generally very well laid out and the methods are explained thoroughly and well executed. However, there are some areas where more concrete explanations are needed.

  Thank you very much for this encouraging summary.

- P4, L90: The authors mention that the aerosol is not interactive in the simulations used for this study and refer to the aerosol radiative effects and heterogeneous chemistry. Is sulfate aerosol represented in some form in these simulations? If so, please state for clarity. If not, what does this imply for the sulfur cycle?

  This was indeed unlclear. In the revised manuscript we replaced the misleading sentcence by:

  SO_2 *in the gas phase is oxidized by the hydroxyl-radical* OH *or direclty photolysed. The major sink, however, is by transition into the aqueous phase, mainly cloud water, and further oxidation in the liquid phase. Since gaseous* SO_2 *is not released on evaporation of cloud and rain droplets, the sulfur contents is in these cases transferred into a so-called residual (res) pseudo-aerosol tracer* $SO4_{res,cs}$ *with characteristics of a coarse mode soluble (cs) aerosol. This tracer is treated as aerosol tracer, e.g. by the sedimentaion submodel (SEDI), by the dry deposition submodel (DDEP), and by the wet scavenging submodel (SCAV). The details of the gas phase and liquid phase chemistry are documented as supplemantary material.*

- P16, Figure 5: Looking at this figure, the SO2 burden appears to decline exponentially in the TROPOMI retrieval, whereas all model simulations depict a more gradual, almost linear decay. The authors elaborate that the differences might stem from repetitive injections of SO2 into the atmosphere. However, the different changes in the decline rates suggest a difference in removal processes as well.

Honestly speaking, we do not see this in Fig. 5. If there is a significant difference between the shape of declines at all, it is more the other way around: the model simulated declines are more exponentially shaped, whereas the TROPOMI retrieval based estimate is more linear in shape. Moreover, we do not have any indication that the sink processes represented in the model are not first order exponential, i.e. describable by an SO_2 lifetime, which is determined mainly by washout, dry deposition, and oxidation with OH.

- Fig 7, 9, 11: The near surface SO2 concentrations in EMAC seem to be more spread out, is there an underestimation of initial removal close to the source or could it be related to how SO2 is emitted in the model or could the distribution be "flatter" due to e.g. numerical diffusion?

  This is indeed a very good observation of our results. Indeed, presumably the rather coarse model resolution is the cause. Whereas SO_2 in reality is in large parts emitted from point sources, in the model these emissions are instantaneously distributed over the entire grid-box. This is a well-known and common limitation of large-scale models for atmospheric chemistry.

- Minor comments:

  - L6: close $\longrightarrow$ closed

    Done.

  - L6-8: This sentence is a bit long and confusing

    We split the sentence into two and reformulated: *First, the tropospheric sulfur budget simulated by EMAC is verified to be closed. This closure means that all sulfur sources and sinks are balanced and no artificial gain or loss occurs over time due to numerical or conceptual errors.*

  - L60: Jöckel et al. in parentheses

    Done.

  - L89: isopren $\longrightarrow$ isoprene

    Done.

  - Table 4: Biomasse $\longrightarrow$ Biomass

    Done.

  - L204: citet $\longrightarrow$ cited

    Done.

  - L230: comparatative $\longrightarrow$ comparative

    Done.

  - L365: heve $\longrightarrow$ have

    Done.