Reply to reviewer #1

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We thank reviewer #1 for her/his comments and the 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*.

1 General comments

This manuscript is a thorough evaluation of the EMAC v2.55 sulfur simulations and makes a useful contribution by (i) closing a model-internal sulfur budget (ii) documenting how the model compares with satellite data in 2019 to evaluate how it responds to volcanic emissions and (iii) evaluating against long-term measurements, 2010–2019

The paper is well organized and generally clear. However, it is unnecessarily long, and the presentation of results is at times too detailed, making it difficult to extract the main messages and scientific significance. The description of the model setup partly repeats work published elsewhere, and it is not entirely clear what is new compared to earlier model versions (e.g., Jöckel et al., 2016).

Jöckel et al. (2016) describes our model setup and contribution to CCMI-1. Here, we analyse data from our contribution to CCMI-2022. This information is written in Section 3.1, lines 70-71 (original manuscript). However, since this is indeed an importatn information, we added to the abstract as well:

In this study, we present, for the first time, a comprehensive examination of atmospheric SO_2 *simulated by the EMAC model, here operated under the Chemistry-Climate Model Initiative (CCM-2022) protocol*

For the interactive gas–particle chemistry, more detail would be beneficial, as the current description is incomplete for interpreting SO lifetime and deposition. I.e. the statement that "the simulation did not involve an interactive aerosol submodel" needs clarification. Does this mean that interactions with ammonia are excluded? If so, this should be explicitly stated, as ammonia strongly influences sulfur oxidation pathways, cloud pH, and the partitioning and deposition of sulfur.

This was indeed unclear. The interactions with ammonia are included in the liquid phase chemistry scheme. 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.*

2 Specific comments

- For the emissions it is important to notice that earlier global inventory for China have underestimated the reductions after around 2010. I am not sure if CMIP6 emissions have taken this into account? In Line 35 it is written that the emissions in China remain high, which is true, but the authors should also mention the substantial reductions in recent years.
  We added a reference analysing the recent reduction of SO_2 emissions over China.

- Why is the comparison made with EDGAR 5 instead of the more recent EDGAR 6.1 (2024)? Using EDGAR 6.1 would enable comparison over the full 2000–2019 period. It would strengthen the analysis to run short sensitivity tests (1–2 simulations) using alternative inventories (e.g., EDGAR 6.1) to quantify whether biases in CMIP6 emissions explain the overestimation. If such tests are beyond the study's scope, this limitation should at least be discussed.
  In our revised manuscript we added the more recent EDGAR 8.0 inventory to cover the full period. However, the interpretation of our model results and our conclusions do not change. Additional simulations with alternative emission inventories are indeed beyond scope, because the focus here is the evaluation of the CCMI-2022 model results (see also reply to General comments).

- Why are you not comparing to the more recent EDGAR6.1 (from 2024) instead of EDGAR5? Then you would be able to compare it with the whole 2000-2019 period. It would have been useful to run the model with EDGAR and not only compare the emissions inventory to evaluate more directly if it biases in the CMIP6 emissions that cause the overestimation. Not necessarily a full rerun of the model but quantify the bias by re-running 1–2 short sensitivity test with different emissions. Though I understand if this is beyond the capacity of this work.
  In our revised manuscript we added the more recent EDGAR 8.0 inventory to cover the full period. However, the interpretation of our model results and our conclusions do not change. Additional simulations with alternative emission inventories are indeed beyond scope, because the focus here is the evaluation of the CCMI-2022 model results (see also reply to General comments).

- Sulfur data from Africa (INDAAF; https://indaaf.obs-mip.fr) and from Canada (CAPMoN) could have been included to provide a more complete global picture. In the U.S., CASTNET is responsible for air and aerosol data, whereas wet deposition data originate from the National Atmospheric Deposition Program (NADP). It appears NADP data were used,

but this is not stated explicitly. NADP also includes many more sites than those used here.

In principle it would have been possible to include other regions in our analysis as well. However, in other regions, compared to the three regions USA, Europe, and East Asia/China, the network coverage is usually lower and time series of observations have gaps in the considered time range. For instance, according to our inspection, the INDAAF data contain only 4 stations with SO_2 measurements, but none covering the persiod 2000 - 2019, which was the criterion for our selection (see our Appendix A3 Ground-based measurements).

Moreover, we intentionally focused on the three regions since they are, on the one hand, the largest emitters in the industrial and energy sectors, and on the other hand, are observed by rather dense observational networks providing sufficiently long time series for evaluation.

For the USA we used the CASTnet data from the URL as indicated in our Appendix A3.1 USA. This data indeed comprises the wet deposition data from NADP/NTN. We added this information to the revised manuscript:
Dry deposition fluxes in this data set are derived with the Total Deposition Science Committee (TDep) Measurement Model Fusion method using the measured air concentrations from CASTnet and simualted CMAQ deposition velocities and fluxes. The wet deposition fluxes in this data set are estimted with the TDep Measurement Model Fusion method using the concentrations in precipitation and precipitation amounts measured by the National Atmospheric Deposition Program / National Trends Network (NADP/NTN).
Again, we selected only stations at which the measurements cover the two decades 2000 - 2019.

- The model response to volcanic emissions is evaluated only against TROPOMI satellite data. You write that "it is difficult to ascertain whether the differences originate near the surface or higher up in the atmosphere." It would strengthen the analysis to use in-situ data from 2019, which are available for that year and would allow a more direct comparison.
  Indeed, we tried this direct comparion, however the results were not conclusive. In-situ data are only aivailable near-surface, where the volcanic SO_2 signal is weak or entirely absent, because the volcanic plume is transported and dilutet at higher altitudes.

- It is not written how trends were calculated, i.e. linear regression, Sen's Slope . Mann Kendall. This should be added to the methods.
  Trends have been calculated with linear regression. This information has been added to the revised manuscript, Appendix B, and in the text.
  For trend comparisons, it would be appropriate to refer to regional studies covering similar periods. Some suggestions:
  We thank the referee for pointing out the additional literature.

  - In Europe recent work done by EMEP: https://doi.org/10.4209/aaqr.230237. Seems like you have somewhat larger trend for SO2 (0.05 ug/m3/y) compared to EMEP (0.034 ug/m3/y). The difference may stem from site selection, trend method, or emissions used.

Unfortunately we cannot retrace the 0.034 $\mu g m^{-3} yr^{-1}$. Aas et al. (2024) report a SO_2 decline of -0.067 $\mu g m^{-3} yr^{-1}$ (see their supplementary Table S3) for the years 2000-2019, which is slightly larger than our derived trend. We added this information to the revised manuscript:

This trend, calculated by linear regression, is comparable to the trend of -0.067 $\mu g m^{-3}$ derived by Aas et al. (2024, see thier Table S3 for the perios 2000 – 2019), which has been derived with a different method and based on a slightly different number of stations.

– Several studies in North America. Eg.: NADP data: https://doi.org/10.1016/j.atmosenv.2023.119783, CASTNet: https://doi.org/10.5194/acp-22-12749-2022 and from Canada by CAPMoN: https://doi.org/10.5194/acp-22-14631-2022

The comparison of our results with those of Conrad-Rooney et al. (2023) is limited, nevertheless we added to the revised text:

Conrad-Rooney et al. (2023) analysed the wet-deposition of SO_4^{-2} for urban and suburban sites separately, whereas we did not distinguish different site classes and analysed the total sulfur deposition. Thus, the results are not directly comparable. Nevertheless, Conrad-Rooney et al. (2023, see their Figures 2D and 5D) estimated for the year 2000 a wet deposition rate (urban stations) of approx. 20 $kg(SO_4^{-2}) ha^{-1}$, equivalent to 6.7 $kg(S) ha^{-1} yr^{-1}$. For 2018, they derived a wet deposition rate of 5 (rural) to 7.5 (urban) $kg(SO_4^{-2}) ha^{-1} yr^{-1}$, equivalent to 1.7 to 2.5 $kg(S) ha^{-1} yr^{-1}$, respectively. These numbers agree, given the limitations of this comparison, with our results for the eastern USA (see Figure 8d, blue area).

W.r.t. to Benish et al. (2022) we added:

Benish et al. (2022) derived for the USA a decrease of the total sulfur depostion from 5.3 $kg(S) ha^{-1}$ in 2002 to 1.8 $kg(S) ha^{-1}$ in 2017 (see their Figure S14 (b)). These total fluxes are lower than the wet deposition fluxes derived by Conrad-Rooney et al. (2023) and than our results, but the derived trends are consistent.

– Japan: https://doi.org/10.1016/j.envpol.2021.117842

We added:

In addition, Yamaga et al. (2021) found "no clear increase or decrease trends in the S deposition amounts throughout the 15-year period" (2003 - 2017) at 8 stations in Japan. This is also in line with our results (Figure 12, top right panel) due to the large standard deviations and because the overal decline is largely driven by emission reductions over China.

– China: https://doi.org/10.3390/su17198815

We added:

... This is an average decline of 0.53 $kg(S)\, ha^{-1} yr^{-1}$ between 2014 and 2019 based on station data in South China and Japan. Xi et al. (2025) reported a China nationwide average decline rate (2013 – 2023) of -0.244

$\mathrm{kg(S)\,ha^{-1}yr^{-1}}$ *and furhter showed that the trends in South and Central China are larger (negative) than this average (see their Figure 2). Thus, our results can be considerd to be consistent.*

3 Technical corrections/spelling errors

- Line 70: "caclulated" to calculated

  Done.

- Line 80-81: "sun-synchrinously" to sun-

  Typo is corrected.

- Line 89: "isopren to isoprene.

  Done.

- Line 146: "histrotical" to historical.

  Done.

- Line 149: "soley" to solely.

  Done.

- Line 230: "comparatative" to comparative.

  Done.

- Line 260: "correpsonding" to corresponding.

  Done.

- Line 294: "drived" to derived

  Done.

- Line 328: "depostion" to deposition

  Done.

- Line 364: "heve" to have.

  Done.

- Line 543; "cylce" to cycle

  Done.

- Line 604: "retrieveal" to retrieval.

  Done.

– Line 658: “resepctively“ to respectively.

Done.

– Line 723: “anlyses” to analyses

Done.

– Line 724: “caclualted” to calculated

Done.

– Line 746 “undr” to under. Multiple instances of “acces” and “reslts” to be replaced with access and results.

Done.

– Table 2 caption: “aersol” to aerosol.

Done.

– Figure caption 12. EMEP should be replaced with EANET

Corrected. Thanks for spotting this error.

– Standardize Tg(S)/yr vs Tg(S)/a

We adapted all units to the exponential form as requested by the journal guidelines, i.e. $\mathrm{Tg(S)\,yr^{-1}}$ and similar.

– There is a mix of units used for air concentration and fluxes (e.g., μg m^3 vs mmol m^2 and kgS/ha). These should be standardized throughout the paper for clarity and comparability.

Our intention was to use the units as reported with the corresponding measurements. Yet, we see the issue. Thus, in the revised manuscript we converted the deposition flux units $\mathrm{mmol/m^2}$ into $\mathrm{kg(S)\,ha^{-1}}$ in Section 6.3 to be consistent with Sections 6.1 and 6.2.

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