

Dear referee#3,

we thank you very much for your in-depth review of our manuscript egosphere-2024-324. Please find our replies to your comments below. Your original comments are repeated in italics, our replies in normal font, and text passages which we included in the text are in bold.

*General comments: The manuscript offers a comprehensive assessment of transport emissions' impacts on ozone and the hydroxyl radical (OH) across different transport sectors using the EMAC Chemistry-Climate model's simulations. The study uses an innovative approach to quantify contributions to OH for the years 2015 and projections for 2050, under various SSPs. The analysis extends to ozone radiative forcing and methane lifetime reductions, highlighting the manuscript's value to environmental policy and planning stakeholders. The manuscript is well written and provides an extensive analysis of the impacts arising from various emission scenarios. However, there are several areas where further development could enhance the study's robustness. After addressing the suggestions outlined below, the manuscript should be considered for publication due to its valuable contribution to the field.*

Reply: Thanks a lot for your overall positive comments on our manuscript. We revised the manuscript based on your suggestions and the comments from the other referees. Please find our detailed comments and changes below.

*Model Evaluation: The manuscript would greatly benefit from a dedicated validation section. Such a section should detail the model's proficiency in simulating the chemical environment and meteorology for the base year of 2015. This could include comparisons of model outputs with observed data or results from prior studies to establish the model's skills*

Reply: Thanks a lot for your suggestion. Referee#1 raised a similar point. Therefore, we added a short model evaluation with additional details in the Supplement.

Generally, our model configuration has only minor updates (e.g. more recent model version, changed emissions) compared to the configuration in Jöckel et al. (2016) including a detailed evaluation. We compared some key species with the results of the *RC1SD-base-10a* model simulation as described by Jöckel et al. (2016). Overall, our analysis shows that the magnitude of the differences between the results of the two simulations is as expected given the different emission inventories. Therefore, we conclude that the detailed evaluation presented by Jöckel et al. (2016) holds also for the simulations analysed in the present manuscript. In addition, we compared upper tropospheric/lower stratospheric ozone simulation results with the SWOOSH data-set. This comparison confirms the known positive bias of ozone in the troposphere. Moreover, we want to stress that our goal was not to use the 'best' available emissions inventory for present day, but to use the CMIP6 emissions inventory in their original form, since model results based on these emissions inventories have been used in many

studies.

The added subsection reads:

The EMAC model has been extensively evaluated in the past. Jöckel et al. (2016) present a detailed evaluation of various atmospheric variables, including tropospheric and stratospheric ozone. From these evaluations we know that EMAC has a positive bias of tropospheric ozone and a negative bias of carbon monoxide. Estimates of the methane lifetime simulated by EMAC are typically at the lower end of the range of values estimated by other models. However, multi-model inter-comparisons show that the biases compared to observational data of EMAC are within the range of those of comparable models (Naik et al., 2013; Stevenson et al., 2013; Voulgarakis et al., 2013; Young et al., 2013).

Given these extensive previous evaluation efforts, we reduce the evaluation of our model results to a minimum. In a first step we compare the ozone mixing ratios of the results from our PD simulation with the results of the *RC1SD-base-10a* simulation discussed by Jöckel et al. (2016). The set-up of both simulations are very similar, despite changes of the emission inventories, small updates and bug-fixes in the model infrastructure, and the fact that we simulate more recent years. Ozone is larger by 2–4 nmol mol<sup>-1</sup> in PD compared to *RC1SD-base-10a* in the extra-tropical lower and middle troposphere. In the extra-tropical free troposphere the difference between the two simulation is slightly larger, reaching up to 8 nmol mol<sup>-1</sup>. In the tropical troposphere the difference range between -2–2 nmol mol<sup>-1</sup>. Overall, the change is lower than 8 % with the largest increase in the Southern Hemisphere, dominated by the variability of the polar vortex. Figures of the comparison of ozone and of further trace gases are provided in the Supplement (see Supplement Sect. S10). From this analysis we conclude that the extensive evaluation presented by Jöckel et al. (2016) remains valid.

In addition, we compared the simulated ozone mixing ratios in the upper troposphere and lower stratosphere (UTLS) with Satellite measurements published as the Stratospheric Water and OzOne Satellite Homogenized dataset (SWOOSH) by Davis et al. (2016). The SWOOSH data are a homogenized, gridded, monthly-mean data set for ozone and water vapour based on several satellite data. For the considered period the data set is based on Aura MLS. We used the SWOOSH data in version 2.6 with a horizontal resolution of 2.5° and 31 vertical levels. Horizontally, the SWOOSH data are interpolated onto the slightly coarser EMAC grid, vertically the data are interpolated onto the much coarser SWOOSH grid similar as by Pletzer and Grewe (2024). The monthly-mean SWOOSH data are compared with monthly-mean data from the model, meaning that satellite data and model data are not co-located in space and time. Averaging Kernels

of the Satellite are not considered, accordingly the satellite data can only be used for a qualitative evaluation. The evaluation is performed for the years 2013–2017.

Figure A1 shows the difference between the ozone mixing ratios of the PD simulation and the SWOOSH data. Overall, the inter-comparison confirms the known bias of simulated ozone, as discussed above, also in the upper troposphere. We would like to stress that the results can only be used for qualitative evaluation (i.e. confirming the ozone bias), as neither averaging kernels are used, nor are the data spatially and temporally co-located. Moreover, the number of considered years are very limited and we found that the magnitude and location of the peak of the upper tropospheric ozone bias strongly depends on the approach used for vertical remapping due to the limited vertical resolution of SWOOSH. For a detailed quantitative evaluation of UTLS ozone we refer to previous inter-comparisons for example with the IAGOS (in-situ measurements on board passenger aircraft) measurements presented by Jöckel et al. (2016); Pletzer et al. (2022); Cohen et al. (2024).

*Model Description: The description of the EMAC model setup, including its chemical mechanisms, is thorough. Nonetheless, the manuscript would benefit from additional details on the model’s parameterizations, particularly those influencing ozone and other chemical species. This should encompass radiation, deposition, and boundary layer schemes, as well as the land surface model used. A clarification on whether the simulations incorporate direct radiation feedback would be pertinent.*

Reply: Thanks for this suggestion. As stated in the manuscript the model set-up is very similar to the set-up of (Jöckel et al., 2016). Therefore, we wanted to keep repetitions to a minimum, but of course the information in the manuscript should reflect the most important details. However, we added some more details on the mentioned processes. In Sect. 2 we described the ‘QCTM’ mode of EMAC which is applied in all simulations. In this operation mode the same (prescribed) climatologies from previous simulations are used for all radiatively active trace gases. With this approach we achieve identical model dynamics in all simulations. This approach is very important to be able to quantify even small perturbations (see Deckert et al., 2011). These small perturbations might not be detectable in a statistically robust way or require very long integration times.

To make this more clear in the manuscript we added some more details on the description of the QCTM mode.

In the description of the RF calculation: **It is important to note that neither the radiative fluxes from  $O_3$  nor the ozone contributions (e.g.  $O_3^{\text{SHP}}$ ) feed back onto the dynamics. Instead, prescribed cli-**

matologies are used for the forcing of the dynamics (as described in Sect. 2.4).

In the description of the QCTM mode: In this mode, mixing ratios of the radiatively active trace gases are prescribed for the radiation calculations. **This means that in each simulation the same radiative forcings by the prescribed mixing ratios are considered.**

As stated in the manuscript the model set-up is very similar to the set-ups described and evaluated by Jöckel et al. (2016) including also detailed description of the considered processes (see also Jöckel et al., 2010). We expanded our short description with details on the dry deposition scheme, calculation of photolysis rates, the radiation scheme, the boundary layer and the land-surface model. The added points are:

- Heterogeneous reactions in the stratosphere (submodel MSBM, Jöckel et al., 2010) as well as aqueous phase chemistry and scavenging (submodel SCAV, Tost et al., 2006) are included. **Photolysis rates are calculated using JVAL (Sander et al., 2014).**
- **Dry deposition is considered via the submodel DDEP (described as DRYDEP by Kerkweg et al., 2006). It is based on the big-leaf approach by Wesely and Hicks (2000).**
- **The radiation is largely based on the original radiation scheme from ECHAM5 (Roeckner et al., 2003), but restructured and expanded with additional features such as multiple diagnostic calls as described by Dietmüller et al. (2016).**
- **The land surface model and the boundary layer implementation are modularized versions (see also Jöckel et al., 2016) of the original implementations of ECHAM5 described in detail by (Roeckner et al., 2003).**

*Introduction and Methods: Structural Suggestions: Consider relocating parts of the limitations and uncertainties discussion, currently in line 610, to the introduction or methodology sections. This would help set the reader's expectations early in the manuscript.*

Reply: Thanks for the suggestion. We would like to keep the structure as it is, because we expanded/restructured the discussion a lot based on the reviews/community comments. Nevertheless, we did add some information about the limitations in the introduction. The changed paragraph reads:

We performed simulations for 2015 and for the three considered SSPs. **Each simulation covers five years and simulates the same present-day meteorology. Accordingly, the influence of climate change on atmospheric**

**composition is not considered, but the effect on the results are discussed in detail.**

*Climate Change Impact: Incorporating a discussion on the potential changes and impacts of climate change on atmospheric chemistry and transport patterns is recommended. This includes a thorough consideration of radiation feedbacks and their prospective effects on future climate change scenarios. For example, changes in surface and atmospheric temperatures can profoundly influence ozone chemistry; as temperatures increase, so do evaporation rates, which lead to a higher concentration of water vapor in the atmosphere, potentially affecting OH and ozone levels. Additionally, changes in cloud cover can alter photolysis rates, thereby impacting ozone formation and destruction*

Reply: Thanks for the suggestions. The community comment from Owen Cooper raised similar concerns. We restructured the discussion and expanded our discussion on the effect of climate change on ozone. We want to stress that the effects of climate change are not considered in these simulations and we can't assess the effects of radiation feedbacks from our model results by design. This is due to the use of the 'QCTM' mode. For further studies it is very important to also include the effects of climate change on atmospheric chemistry. This would, however, need a different model set-up with much more expansive and much longer time-slice and transient simulations due to the large signal-to-noise ratio.

The changed discussion reads: **Effects of climate change**

The signals of emissions from specific regions or specific emission sources (such as e.g. aviation) are small. To quantify these signals, we apply the QCTM mode (see Sect. 2) in which chemistry and dynamics are decoupled. Accordingly, the dynamics (and therefore the climatic state) is identical in every model simulation. This approach of applying present day dynamics for future emission scenarios is commonly used when the effects of certain emission changes or sources on the atmospheric chemistry are investigated on the global and regional scale (e.g., Eyring et al., 2007; Hoor et al., 2009; Hodnebrog et al., 2012; Righi et al., 2015; Matthias et al., 2016).

Due to this approach, however, **our model simulations do not consider changes of meteorology and climate between 2015 and 2050. Accordingly**, emissions which are based on meteorological conditions (e.g. biogenic emissions, lightning-NO<sub>x</sub>) are identical in all simulations. With climate change, these emissions are likely to increase (von Schneidemesser et al., 2015). This increase could alter the contributions of the anthropogenic emissions, for instance increased biogenic VOC emissions may affect the ozone production efficiency, while increased lightning-NO<sub>x</sub> in the upper troposphere may compete with NO<sub>x</sub> emissions from the aviation sector.

**Moreover, increased biogenic emissions and changed atmospheric conditions (e.g. increased temperature and its effects on kinetics) likely lead to an increase of ozone near highly polluted regions (known as**

'climate-penalty', Zanis et al., 2022). In addition, climate change likely leads to an decrease of ozone in remote regions due to the increase of water vapour (known as 'climate-benefit', Zanis et al., 2022). In addition, during periods of droughts and heat-waves, reduced ozone deposition to vegetation could increase ground-level ozone (Lin et al., 2020).

Altogether, this could affect also the contributions of the traffic emissions. A reduced life-time of ozone, especially over the oceans, would likely lead to a reduction of ozone attributable to shipping emissions. Also long-range transport, especially the source-receptor relationships, might be affected by changes of the ozone lifetime. At the same time, the increase of ozone in polluted regions in a changing climate could affect ozone contributions especially from land transport emissions.

Koffi et al. (2010) considered the effects of climate change on the ozone effects of transport emissions applying a 5 % emissions reduction. Globally, they report a small decrease of the ozone changes caused by transport emissions due to climate change, but with strongly varying regional patterns. **The effect of climate change on ozone contributions (i.e. applying a tagging approach) needs to be analysed in follow-up studies.**

*Land Surface Model Considerations: It is important to discuss the implications of land surface model choices within the simulations. Soil moisture variability, alterations in land use, and vegetation cover driven by climate scenarios play an important role in the soil's chemical processes and the land's overall energy budget. For instance, soil moisture dependent on the chosen climate scenario affects soil chemistry, influencing how land surface models simulate these processes. Similarly, changes in land use and vegetation cover have the potential to modify the absorption, reflection, and emission of radiant energy at the land surface. Moreover, the type of vegetation and temperature changes can affect the deposition of chemical species and their uptake by plants.*

Reply: We agree that these aspects also affect ozone. As stated in the reply above effects of climate, land use etc. are not considered in the present study. We added a short note about these effects in the overall discussion. This part reads:

**Similar to changes of the climate, changes in land-use also affect ozone (e.g. Wu et al., 2012; Wang et al., 2020). The effects of land-use change on ozone are due to various processes, such as changes of biogenic emissions, effects on dry deposition, and changes of temperature (e.g., by effects on radiation and evapo-transpiration). Land-use change, and the corresponding effects, also heavily depend on the considered scenario (Popp et al., 2017). These effects are not considered to isolate the effects of the emissions changes only.**

*Sensitivity Analysis: Could a sensitivity analysis be performed to evaluate how various model assumptions, such as chemical reaction rates and deposition processes in a changing climate, might affect the outcomes? This would contribute to understanding the study's conclusions' robustness.*

Reply: Thanks for the suggestions. As stated above our model set-up currently does not consider the effects of climate change by design. Therefore, the current model set-up is not suitable to assess questions such as "chemical reaction rates and deposition processes in a changing climate, might affect the outcomes?". This would require a different model set-up which is out of scope of the present study. However, as stated above, this would be the next step for follow up studies.

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