Reply to Reviewer #1

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We thank Reviewer #1 for their comments and their evaluation of our paper. Below, we repeat each comment (in blue) and address it (in black). Changes of the manuscript are written in italics.

1 General comments

1.1 Ocean model coupling

A mixed layer ocean model is coupled in the REF-SSTvar simulations. How representative are the SSTs in these runs compared to the prescribed SSTs? This would have impacts on air-sea interactions and thus affecting atmospheric chemistry and composition in addition to the GSAT. The biases in reference cases may propagate into the perturbed cases, which would affect your assessment in the climate response. Do you think the biases could be canceled out between the reference run and perturbed run?

You are right that we did not discuss possible differences between both reference simulations. The study by Stecher et al. (2021) used the same setup of the MLOCEAN submodel. As their study was one of the first to use this configuration, they provide a more detailed evaluation of differences between the reference simulations with prescribed sea surface temperatures (SSTs) and with MLO. In addition, Appendix C of the PhD thesis of the first author (Stecher, 2024) compares the reference simulation used in this study (REF-SSTfix and REF-SSTvar). We added a short text with both references in line 149. Concerning the distribution of chemical species, the zonal mean difference of methane (CH_4) mixing ratios between REF-SSTfix and REF-SSTvar is below 0.5% in the troposphere. Ozone (O_3) mixing ratios are up to 2% smaller in the southern troposphere in the MLO reference, which can be linked to a higher abundance of water vapour (H_2O) and thereby a stronger chemical sink. O_3 mixing ratios are up to 5% larger in the tropical tropopause region in the MLO reference, which is linked to a slightly larger (less than 1 hPa) tropopause pressure in the MLO simulation (see Stecher, 2024, Appendix C).

Add in line 149:

Appendix C of Stecher (2024) provides a comparison of the two reference simulations. Overall, the simulation REF-SSTvar reproduces the simulation REF-SSTfix well. The largest differences are in the Southern Hemisphere (SH) polar region, where the MLO model tends to underestimate the sea ice area of the prescribed climatology, which has been noted for a similar application of the MLOCEAN submodel as well (Stecher et al., 2021).

1.2 Methane radiative feedback

The simulations between reference cases and perturbed cases suggest larger sensitivity of the methane lifetime towards climate change compared to previous studies and the authors attribute such difference to the different methane representations in the models, e.g., emission driven vs prescribed lower boundaries. But the authors also point out in the manuscript, the model-specific parameterizations, mechanisms, etc would lead to the model-dependent results. For example, using different radiative transfer models give very different radiative feedback as shown in the manuscript. How to better quantify the sensitivity to different model setup or model schemes? How to improve the model confidence in assessing methane radiative feedback?

We think that it is important to differentiate between the sensitivity of firstly, the response of OH and CH_4 mixing ratios, and secondly, of the quantification of the corresponding radiative effects here. In our study, the radiative effect corresponding to the same CH_4 change depends strongly on the radiative transfer scheme, with the PSrad scheme providing the more reliable estimates. The response of OH and CH_4 could also depend on the radiation scheme driving the model, but our simulations results cannot answer this question. As we mention in the outlook, such a comparison is planned in the future.

The model spread of previously published estimates of the radiative feedback of CH_4 is caused by the spread of the OH response, as the CH_4 radiative effects are derived using formulas, e.g. by Etminan et al. (2016), (Heinze et al., 2019; Thornhill et al., 2021). The OH response is influenced by many factors and it would indeed by valuable to identify reasons for CCM differences in future studies.

In general, we think that a multi-model comparison of the CH_4 feedback from chemistry-climate simulations driven by CH_4 emission fluxes would be helpful. These estimates could be compared to previous multi-model mean estimates from simulations with CH_4 prescribed at the lower boundary to assess the influence of CH_4 emission fluxes. We modified the paragraph starting in line 601 also taking into account comment 7 and 8 by referee 2.

Modify line 601 (see also comment 7 and 8 by referee 2):

Modified:

The sensitivities of the CH_4 lifetime per unit change of global surface air temperature (GSAT) are -6.7 % K^{-1} for 1.35×carbon dioxide (CO_2) and -7.6 % K^{-1} for 2.75×CH_4, which is larger compared to previous CCM results using prescribed CH_4 mixing ratios at the lower boundary (Voulgarakis et al., 2013; Thornhill et al., 2021; Stecher et al., 2021). The results of the comparable CH_4 increase experiment with prescribed CH_4 surface mixing ratios (Stecher et al., 2021) provides a clear indication that the lifetime change per temperature change is larger in the CH_4 emission driven set-up. A comparable CO_2 increase simulation using EMAC with prescribed CH_4 surface mixing ratios is not available, but the comparison to the results of other CCMs (Voulgarakis et al., 2013; Thornhill et al., 2021) indicates the same effect (see Sect. 3.1). Estimates of the CH_4 lifetime change per temperature change from other chemistry-climate models (CCMs) driven by prescribed CH_4 emission fluxes would be helpful to verify the influence of CH_4 emission fluxes in comparison to prescribing CH_4 at the lower boundary. Additionally, the multi-model differences of the CH_4 lifetime change per unit change of GSAT are large (Voulgarakis et al., 2013; Thornhill et al., 2021) and it would be valuable to identify reasons behind CCM differences in future studies.

1.3 Ozone

There are a lot of discussions in the manuscript on the impacts on tropospheric and stratospheric ozone. What about impacts on surface O3, which is more relevant to the health effects.

Tropospheric and stratospheric O_3 changes are important to understand the corresponding radiative effects and health effects are not the focus of the paper. Nevertheless, we added the response of surface ozone in the supplement and mention it shortly in the main manuscript.

Include in line 453:

The spatial distribution of the climate response of surface O_3 is likewise similar for the CO_2 and the CH_4 perturbation (see Fig. S4).

2 Specific comments

Page 9, Section 3.1, line 265-267, How sensitive of OH levels to lightning NOx? Do changes in lightning NOx play a role here?

Thank you for pointing this out. As we show later in the manuscript, lightning NO_x emissions increase by about 0.3 Tg(N) a^{-1} in the climate response, which is expected to lead to enhanced OH production. To estimate the effect on the CH_4 lifetime, we use the multi-model mean sensitivity of the CH_4 lifetime towards lightning NO_x emission change of -4.8% (Tg(N) $a^{-1})^{-1}$ from Thornhill et al. (2021), which suggest a shortening of the CH_4 lifetime due to the lightning NO_x emission change by 1.4%. The CH_4 lifetime shortens by 7.4% in total. We will adapt the text as follows:

Modify line 265:

Previous:

The OH response is largely driven by the increase of tropospheric humidity associated with higher temperatures.

Adapted:

Firstly, emissions from lightning NO_x increase by about 0.3 Tg(N) a^{-1} in the climate response (see Tab. 3), which leads to enhanced production of OH. To estimate the effect on the CH_4 lifetime, we use the multi-model mean sensitivity of the CH_4 lifetime towards lightning NO_x emission change of -4.8% (Tg(N) $a^{-1})^{-1}$ from Thornhill et al. (2021), which suggest a shortening of the CH_4 lifetime due to lightning NO_x emissions by 1.4%. Additionally, the increase of the tropospheric humidity associated with higher temperatures leads to enhanced production of OH.

Page 13, line 375-380: How do you treat N2O in the model? Does your model read N2O emissions or prescribe N2O at lower boundaries?

Nitrous oxide (N_2O) mixing ratios are prescribed at the lower boundary. We added this information to the methods section.

Add in line 142 (see also reply to referee 2):

The mixing ratios of CO_2, N_2O and ozone depleting substances (ODS) are prescribed at the lower boundary using monthly mean values of the year 2010 (Meinshausen et al., 2011; Carpenter et al., 2018). For the radiation, a CFC-11 equivalent is calculated lumping additional radiatively active ODS via radiative efficiencies following the approach by Meinshausen et al.

(2017). For the short-lived halocarbons $CHCl_2Br$, $CHClBr_2$ *and* CH_2ClBr, *as well as* CH_2Br_2 *and* $CHBr_3$ *surface emissions are prescribed from Warwick et al. (2006) and Liang et al. (2010), respectively.*

Figure S2 shows the difference in the specific humidity. Is specific humidity in your model also affected by chemistry? In other words, is water vapor a prognostic chemical tracer explicitly involved in the chemical reactions?

Yes, H_2O is a prognostic chemical tracer in the chemistry module MECCA. Its chemical feedback modifies the prognostic specific humidity, and vice versa.

Add in line 126:

The chemical feedback on H_2O *modifies the prognostic specific humidity, and vice versa.*

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