Reply to Reviewer #2

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We thank Reviewer #2 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 Specific comments

1. In Section 2.1, you state that chlorine and bromine halogen chemistry is included and that oxidation of methane by chlorine is considered. Can you include some clarification on whether the chlorine sink is only relevant in the stratosphere and/or whether the methane sink in the marine boundary layer through tropospheric halogen chemistry is included?

   The methane sink through the reaction with chlorine is accounted for in the whole atmosphere. However, the reaction with chlorine accounts for only about 0.23% of the total tropospheric methane (CH_4) loss in our reference simulations (see Table S1, which we added to the supplement). We added this information in the Methods section. We also added the following information about emissions of the short-lived halogens $CHCl_2Br$, $CHClBr_2$, CH_2ClBr, CH_2Br_2 and $CHBr_3$, as these are expected to affect the chlorine sink.

   – **Add in line 123:**

      Oxidation by the hydroxyl radical (OH) dominates the tropospheric CH_4 sink, so that the reaction with chlorine (Cl) accounts for only 0.23% of the total chemical tropospheric CH_4 loss (see Tab. S1 in the supplement). Therefore, we focus on CH_4 lifetime changes with respect to oxidation by OH.

   – **Add in line 142 (see also reply to referee 1):**

      The mixing ratios of carbon dioxide (CO_2), nitrous oxide (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.

2. In Section 2.1, what criteria were used to determine whether "a quasi-equilibrium is reached"?

We added the following information.

Add in line 168 (see also community comment by Zosia Staniaszek) :

Time series of the global mean surface CH_4, the total atmospheric masses of CH_4 and ozone (O_3), the TOA radiation balance, and GSAT (for the MLO simulations) were monitored to decide whether an equilibrium is reached. In addition, we assessed the spin-up of the mass of CH_4 of the simulation $ERFCH_4$ in more detail. A curve fit was applied to the spin-up period to derive the atmospheric mass of CH_4 in equilibrium. The mass of CH_4 follows the exponential function of the form $a - b \cdot exp(-t/c)$ closely. The mass of CH_4 in the last year of the spin-up, simulation year 90, is about 0.5% smaller than the derived equilibrium estimate (parameter a), and therefore spun-up sufficiently well (see Fig. S14). The derived perturbation lifetime (parameter c) is 21.6 years. We note that the perturbation lifetime is larger than that of the CH_4 emission reduction experiment by Staniaszek et al. (2022). As the perturbation lifetime increases with increasing CH_4 burden (Holmes, 2018), this can be expected. In addition, model differences and the magnitude of the emission change might play a role.

3. Line 240: You state that the radiative effects of ozone and water vapour are calculated separately for the troposphere and stratosphere – can you include what definition you use for the tropopause?

The climatological tropopause ($tp_{clim}= 300$ hPa $- 215$ hPa $\cdot \cos^2(\phi)$) is used. We added the missing information.

Add in line 240:

To define the region in which the stratospheric temperature adjustment is applied, as well as to separate tropospheric and stratospheric radiative effects of O_3 and the specific humidity, the climatological tropopause tp_{clim} is used consistently with the CH_4 lifetime calculation (see Sect 2.1).

4. Section 3.1, line 262: You mention that the lifetime with respect to OH oxidation is reduced when the model is allowed to respond to the CO2 perturbation. Although stratospheric oxidation is a more minor sink for methane than tropospheric oxidation by OH, I wondered whether you could also diagnose the stratospheric lifetime

We calculated the stratospheric CH_4 lifetime using Eq. 2, analogously to the tropospheric lifetime, but taking into account all grid boxes above the tropopause. The results of the CH_4 loss, i.e. $\frac{1}{\tau}$, are shown in Tab. S1 in the supplement.

We mentioned the following points in the manuscript regarding the stratospheric CH_4 loss:

Add in line 256:

The stratospheric CH_4 loss by reaction with OH, Cl, and excited oxygen ($O(^1D)$) is reduced by about 2% (see Tab. S1 in the supplement).

Add in line 267:

... and the stratospheric CH_4 loss does not change (see Tab. S1 in the supplement), ...

Add in line 400:

In the stratosphere, CH_4 loss by OH is enhanced due to an increase in stratospheric HO_x, whereas CH_4 loss by Cl is

reduced (see Tab. S1 in the supplement). Overall, chemical stratospheric CH_4 loss is increased by 17.5%. However, the increase of CH_4 mixing ratios is larger than the increase factor of surface emissions of 2.75 in the whole stratosphere as well.

5. Section 3.1, line 269: Given the lack of significant differences in the oxidants, you hypothesize that the change in methane concentrations in the lower stratosphere are due to transport, i.e., because of reduced tropospheric concentrations and a more efficient Brewer Dobson circulation. Do you have any mass flux diagnostics that can support that statement?

We added a Figure showing the response of the residual streamfunction to the supplement and referred to it in the text as follows. Just to be clear, we think that enhanced tropical upwelling alone would lead to an increase of the CH_4 mixing ratio in the tropical lower stratosphere as air masses with larger CH_4 mixing ratio are transported from the troposphere into the stratosphere more efficiently.

Add in line 270: *Tropical upwelling is enhanced in the climate response (see Fig. S13 in the supplement).*

Add in line 447:: *(i.e. strengthening of the Brewer-Dobson circulation, see Fig. S13 in the supplement)*

6. Section 3.1: It is interesting that the sensitivity of the methane lifetime to climate change in EMAC appears to be stronger than in Voulgarakis et al. and Thornhill et al. I'm not convinced that it is due to methane being more fully interactive here, and it would make an interesting follow-up study to try to unpick the reasons behind these model differences.

We agree that it would be valuable to identify reasons for the multi-model differences in future studies. We added this in the outlook, see our answer to comment 7.

7. Section 3.1, lines 303-305: You state that ", the explicit treatment of the CH4 feedback in our set-up allows for a subsequent feedback of OH and correspondingly for a self-feedback on the CH4 lifetime, which can explain the enhanced sensitivity of the CH4 lifetime towards climate change." Have you verified in this setup that if methane was driven by concentration-based boundary conditions that the sensitivity of methane lifetime to temperature would be more comparable to that in other models?

We agree that the comparison with a CO_2 increase experiment with EMAC with concentration-based boundary for CH_4 would be desirable. Unfortunately, we do not know of a suitable simulation for which the CH_4 lifetime change per temperature change was calculated. For the experiments of Dietmüller et al. (2014) the CH_4 lifetime change is not available anymore.

However, for the CH_4 increase experiment, the results of Stecher et al. (2021) provide a fair comparison with concentration-based boundary for CH_4. There is a clear indication that the lifetime change per temperature change is larger in the CH_4 emission driven set-up. We modified the statement in line 601.

Modify line 601 (see also comment 7 and comment 1.2 by referee 1):
Previous:
Consequently, the sensitivities of the CH_4 lifetime per unit change of global surface air temperature (GSAT), -6.7 % K^{-1}

for $1.35\times CO_2$ and -7.6 % K^{-1} for $2.75\times CH_4$, are larger in the present study compared to previous CCM results using *prescribed CH_4 mixing ratios at the lower boundary (Voulgarakis et al., 2013; Thornhill et al., 2021a; Stecher et al.,* *2021).*

Modified:

The sensitivities of the CH_4 lifetime per unit change of GSAT are -6.7 % K^{-1} for $1.35\times CO_2$ and -7.6 % K^{-1} for *$2.75\times CH_4$, 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., 2021a; 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., 2021a) 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., 2021a) and it would be valuable to identify reasons behind CCM differences* *in future studies.*

8. Section 3.1, lines 306-310: Here, you argue that the model response in EMAC is more consistent with f=1 than estimates of f = [1.2, 1.4]. I wonder how representative the range of 1.2-1.4 is for the EMAC model. Do you know what the feedback factor from EMAC is in concentration-driven simulations or even from your ch4 flux perturbation simulation?

We calculated the feedback factor f from the present CH_4 emission increase simulation, which suggests f=1.55 (see also reply to community comment by Zosia Staniaszek). This is at the larger end of previously published estimates. The feedback factor f increases with increasing CH_4 burden (Holmes, 2018), so that f=1.55 is most likely not representative for smaller CH_4 perturbations with the EMAC model. Applying f=1.55 implies an even larger reduction of CH_4 mixing ratio to 1.61 ppmv.

We applied the formula here to show that for the CH_4 emission driven simulation, the CH_4-OH feedback is already included in the OH and the CH_4 lifetime responses. Therefore, using the formula with f>1 applies the CH_4-OH feedback twice, which results in a larger CH_4 reduction than simulated by the model. We rephrased the paragraph and extended the range of f.

Modify in line 306:

Previous:

If the CH_4 mixing ratio can not adapt to changes in its lifetime, the corresponding CH_4 equilibrium mixing ratio can be *estimated using Eq. 1, which indicates a global mean CH_4 equilibrium mixing ratio in the range of 1.63 to 1.66 parts* *per million volume (ppmv) for f = [1.2, 1.4] for the present changes of the CH_4 lifetime. Thus, Eq. 1 suggests a larger* *reduction than simulated by the model, which adjusts to a global mean CH_4 equilibrium mixing ratio of 1.69 ppmv*

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(see Tab. 2). However, if the feedback factor is not applied (f=1), Eq. 1 gives 1.68 ppmv, which is in close agreement with the simulated response of CH_4 mixing ratios and supports the assumption that the sensitivity of OH and the CH_4 lifetime towards climate change is larger, if the feedback of CH_4 is explicitly simulated as thereby the CH_4-OH feedback is implicitly included in the simulated response.

Modified:

If the CH_4 mixing ratio can not adapt to changes in its lifetime, the corresponding CH_4 equilibrium mixing ratio can be estimated using Eq. 1. The feedback factor f in the equation accounts for the CH_4-OH feedback. In our CH_4 emission driven simulation the CH_4-OH feedback is implicitly included in the simulated response of OH and the CH_4 lifetime, so that using Eq. 1 with $f > 1$ applies the CH_4-OH feedback twice in this case. Eq. 1 indicates a global mean CH_4 equilibrium mixing ratio in the range of 1.61 to 1.66 ppmv for $f = [1.19, 1.55]$ for the present changes of the CH_4 lifetime. Thus, it suggests a larger reduction than simulated by the model, which adjusts to a global mean CH_4 equilibrium mixing ratio of 1.69 ppmv (see Tab. 2). However, if the feedback factor is not applied (f=1), Eq. 1 gives 1.68 ppmv, which is in close agreement with the simulated response of CH_4 mixing ratios.

9. Section 3.3: Here you state that Table 4 shows the total SARF, ERF, ΔGSAT and the associated climate sensitivity parameters λ, as well as individual radiative effects corresponding to the composition changes of CH4, O3 and stratospheric H2O. You then go on to compare SARF and ERF for the co2 and ch4 perturbation simulations. In the case of the CH4 perturbation simulation, the ERF is a factor of 3 larger than the SARF. I think it's important to state upfront that the SARF here is only capturing the direct radiative effect of CH4 alone, whereas the ERF captures the radiative effects from the ch4-driven chemical adjustments (e.g., ozone, stratospheric water vapour).

We added a clarification at the beginning of Sect. 3.3. and in the caption of Table 4.

Add in line 487 and in caption of Table 4:

ERF includes physical and chemical adjustments, whereas SARF represents the radiative effect of the CO_2 or CH_4 composition change and the corresponding stratospheric temperature adjustment only.

10. Section 3.3: Here, the radiative effect of stratospheric water vapour from the methane flux perturbation experiment seems to be nearly comparable in magnitude with that from ozone. This doesn't appear to be consistent with the relative contributions from water vapour and ozone in the present-day forcing from methane. Can you comment further on its radiative effect?

We assume that you are referring to the studies by Thornhill et al. (2021b); O'Connor et al. (2022), which quantify the contribution of O_3 and SWV to the pre-industrial to present-day CH_4 radiative forcing. The relative contribution of O_3 is about 16-27% and 13-21%, and of SWV about 0.5% and 2-7% in the studies by Thornhill et al. (2021b) and O'Connor et al. (2022), respectively. So the ratio $\frac{RF_{SWV}}{RF_{O3}}$ is 0.02 for Thornhill et al. (2021b), and 0.10 (Table 3, Single forcing method) and 0.54 (Table 2, Elimination method) for O'Connor et al. (2022). In our study, the relative contribution of O_3 is about 47% and of SWV 30%. For both, O_3 and SWV, the relative contributions are overestimated as the direct CH_4 radiative forcing is underestimated, which we discuss in the manuscript in detail. $\frac{RF_{SWV}}{RF_{O3}} = 0.63$ in our study, which

is larger than in previous studies, but somewhat comparable in magnitude to the results of the Elimination method by O'Connor et al. (2022).

In general, it is difficult to decide whether model/methodological differences or the different time period/magnitude of the perturbation causes the difference. In addition, the differences in the radiative forcing could be caused by the sensitivity of the water vapour response, or by the sensitivity of the water vapour radiative forcing. For example, it makes a difference if the associated stratospheric temperature adjustment is included. We are unsure if the corresponding stratospheric temperature adjustment is included in the estimate provided by Thornhill et al. (2021b). Thus, we decided to not discuss SWV radiative forcing differences in the text as model, methodological and forcing differences can play a role here. A more targeted multi-model/multi-method initiative could help to attribute and to better understand differences.

2 Technical comments

Thank you for the suggested improvements for wording. We have adopted the suggestions. For the following comments, we deviated slightly from the suggestion.

- Comment 22: We reformulated the paragraph and hope that it is easier to understand now.

- Comment 36: The sentence reads now. *Biogenic emissions of* NO_x *and* C_5H_8 *increase in the full response as well*.

- Comment 48: We changed the occurrence in line 688, but not in line 687 as in line 687 the expression *climate feedback of CH$_4$* refers to the change in CH_4 mixing ratios (*feedback on CH$_4$ mixing ratios*), but also to the corresponding radiative feedback (*of CH$_4$*).

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