Reply to referee # 2

October 14, 2020

Dear Peer Johannes Nowack,

thank you very much for the positive comments on our manuscript. In the following we reply to your comments point-by-point. The indicated pages of the answers relate to the discussion paper.

1 Thoughts on the wider context

This work only considers the effects of increased methane in isolation, which is useful to separate its effect from those of other climate forcing agents. However, given the dependency of methane on, e.g., OH, I would expect that simultaneous CO2 forcing found in the real world could strongly interact with this picture, possibly even in a non-linear fashion. I assume that the reduction in OH driven by methane increases, for example, would be largely offset by increases in tropospheric OH under additional CO2 forcing? I am not asking that the study is revised in this sense, but the potential of such interactions should be mentioned somewhere, unless the authors can make strong arguments against this idea. A simple way to achieve this would be to add another clarifying sentence to the paragraph l. 204-214, where you discuss the importance of water vapour and ozone changes, which will also be driven by CO2 forcing and the associated tropospheric warming, thus impacting OH.

Thank you for making this point. We fully agree and will rephrase the paragraph as follows.

Old, l. 212 ... century. However, the tropospheric warming in the RCP8.5 scenario is stronger because it includes the effects of all greenhouse gass (GHGs) and not only the effect of methane (CH_4). This can explain the larger offset of the CH_4 lifetime response reported by Voulgarakis et al. (2013).

New, l. 212 ... century. However, the tropospheric warming in the RCP8.5 scenario is stronger because it includes the effects of all GHGs, as opposed to the isolated effect of CH_4 in our experiments. Additional warming induced by other GHGs, in particular carbon dioxide (CO_2), would drive water vapour (H_2O) and ozone (O_3) increases as well. Therefore, the reduction in hydroxyl radical (OH) driven by CH_4 increases in our experiments is expected to be more strongly offset under a simultaneously active CO_2 forcing.

Did the authors look at changes in the tropospheric circulation at all (cf. Chiodo Polvani 2016, Nowack et al. 2017)? I don't think any study has explored the specifics of the response to methane forcing, with its coupled effects on ozone and stratospheric water vapour before. I am NOT referring to the difference be tween the fixed SSTs and MLO runs here (Figure 2), as this might indeed beyond the scope of this work. If the model set-up allows (fairly short simulations and constrained ocean response), a short section on some central aspects of the tropospheric circulation response could further increase the impact of this paper. Otherwise, maybe suggest this point for future work with fully coupled ocean models. I could also imagine that the (lack of) tropospheric circulation changes might affect the stratospheric circulation response, e.g. through wave forcing and propagation, which might be worth commenting on

The tropospheric circulation in response to CH_4 forcing with and without interactive chemistry would be a very interesting research question, indeed. However, we think that it would open up a new subject area. Considering that this paper is already quite long, we think that a discussion about tropospheric circulation changes is beyond the scope of the present paper and we prefer to leave this point for future work. Moreover, in a future study we plan to use a CH_4 emission flux boundary condition, as opposed to the prescribed CH_4 surface mixing ratios here, so that tropospheric CH_4 can adjust to changes in its sinks. We will include a suggestion of the topic for this study in the conclusions section.

Old, l. 460 The contribution of sea surface temperature (SST)-driven climate feedbacks to the total CH_4 induced O_3 response shows remarkable similarities to the O_3 response to climate feedbacks in CO_2-forced climate change simulations (Dietmüller et al., 2014; Nowack et al., 2018; Chiodo and Polvani, 2019). The consistency between the O_3 feedbacks resulting from these different forcing agents encourages the separation of the O_3 response patterns into rapid adjustments and climate feedbacks in future studies. Rapid adjustments are specific to the forcing, whereas climate feedbacks are driven by surface temperature changes and are therefore expected to be less dependent on the forcing agent (Sherwood et al., 2015).

New, l. 460 The contribution of SST-driven climate feedbacks to the total CH_4 induced O_3 response shows remarkable similarities to the O_3 response to climate feedbacks in CO_2-forced climate change simulations (Dietmüller et al., 2014; Nowack et al., 2018; Chiodo and Polvani, 2019). The consistency between the O_3 feedbacks resulting from these different forcing agents encourages the separation of the O_3 response patterns into rapid adjustments and climate feedbacks in future studies. Rapid adjustments are specific to the forcing, whereas climate feedbacks are driven by surface temperature changes and are therefore expected to be less dependent on the forcing agent (Sherwood et al., 2015). However, the overall response of O_3 (rapid adjustments and slow feedbacks) is quite different under CH_4 forcing compared to CO_2 forcing owing to chemically induced feedbacks under CH_4 forcing. Chiodo and Polvani (2017); Nowack et al. (2017) suggested that feedbacks from interactive O_3 under CO_2 forcing have the potential to significantly alter the tropospheric circulation. As the overall O_3 response is different under CH_4 forcing, also modified feedbacks on the tropospheric circulation are expected. Those are planned to be assessed using a simulation set-up with a CH_4 emission flux boundary condition to simulate feedbacks of tropospheric CH_4 to changes in its chemical sinks.

2 Minor comments

l. 6-8: it might be the passive use of verbs that makes this paragraph slightly hard to read, or also the reference to the Winterstein et al. (2019) study. After all, all you seem to say is that: "Strong increases in CH4 reduce hydroxyl radical concentrations in the troposphere, thereby extending CH4 lifetime. We find that slow climate feedbacks counteract/dampen this effect (through increases in tropospheric water vapour and ozone(?); maybe mention the mechanism).

Thank you for this suggestion. We will modify the text as follows.

Old, l. 6 We find that the slow climate feedbacks counteract the reduction of the hydroxyl radical in the troposphere, which is caused by the strongly enhanced CH_4 mixing ratios. Thereby also the resulting prolongation of the tropospheric CH_4 lifetime is weakened compared to the quasi-instantaneous response considered previously.

New, l. 6 Strong increases of CH_4 lead to a reduction of the hydroxyl radical in the troposphere, thereby extending the CH_4 lifetime. Slow climate feedbacks counteract this reduction of OH through increases in tropospheric H_2O and O_3, thereby dampening the extension of CH_4 lifetime in comparison with the quasi-instantaneous response.

l. 11-13: Maybe more explicitly say as well that the middle-upper stratospheric changes cannot be explained by changes in cold point temperature

Thank you for this hint. We will change the text as follows.

Old, l. 11 In the middle and upper stratosphere, the increase of stratospheric water vapour is reduced with respect to the quasi-instantaneous response. Weaker increases of the hydroxyl radical cause the chemical depletion of CH_4 to be less strongly enhanced and thus the in situ source of stratospheric water vapour as well.

New, l. 11 In the middle and upper stratosphere, the increase of stratospheric water vapour is reduced with respect to the quasi-instantaneous response. We find that this difference cannot be explained by the response of the cold point and the associated H_2O entry values, but by a weaker strengthening of the in situ source of H_2O through CH_4 oxidation.

l. 25: would rephrase "influenced". After all water vapour concentrations are also influenced anthropogenically, only is the effect indirect.

Yes, this is indeed not correct. We will replace it by "directly emitted by human activity".

l. 58-60: I am fairly sure that some of the NASA-GISS simulations by Drew Shindell might have had similar model set-ups but probably looked at other research questions?

Thank you for this note. You are right, the work of Shindell et al. (2005, 2009) and Stevenson et al. (2013) should be mentioned here. We generally extended the introduction and also included these citations (see also reply to referee 1).
In addition, we will include the citation of Shindell et al. (2009) and Stevenson et al. (2013) when referring to Fig. 8.17 of the IPCC report: e.g., Fig. 8.17 in IPCC, 2013 derived from Shindell et al., 2009; Stevenson et al., 2013.

l. 85: Why not attempt a sensitivity analysis of the entire transient data following Gregory et al. GRL (2004) as well? Is the signal too small for the slope to be derived robustly? Gregory et al. A new method for diagnosing radiative forcing and climate sensitivity, Geophysical Research Letters (2004)

The signal is indeed too small for the slope to be derived robustly. We have actually tried this method and included Fig. 1 exemplary for the 5xCH4 case in this reply. For the 2xCH4 case, the signal to noise ratio is even worse.
One solution to reduce the uncertainty would be to calculate an ensemble of spin-up phases as proposed by, e.g., Ponater et al. (2012). This would be, however, computationally expensive. Therefore, we used the "fixed SST" method to quantify effective radiative forcing (ERF) as recommended by Forster et al. (2016).
We will include a short sentence in line 329, where we discuss the climate sensitivity.

Old, l. 329 Under the reasonable assumption that the total radiative impacts (RIs) from the fSST experiments represent the corresponding ERFs with chemical rapid adjustments included (Winterstein et al., 2019), we calculate the climate sensitivity parameters λ as 0.61 $\pm$ 0.17 K W^{-1} m^2 and 0.72 $\pm$ 0.07 K W^{-1} m^2, respectively.

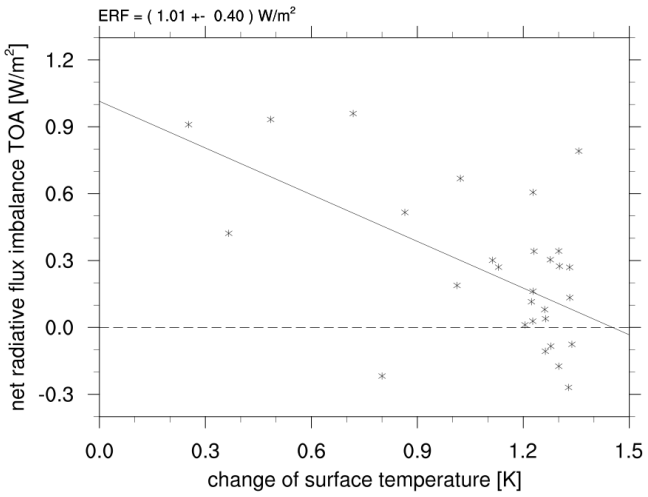

Figure 1: Regression of surface temperature response against net radiative flux perturbation at the TOA for S5 MLO following Gregory et al. (2004).

New, l. 329 The forcing strengths of 2× and 5×CH_4 turn out too small to robustly quantify the corresponding climate sensitivity parameters λ with a sensitivity analysis of the entire transient data following Gregory et al. (2004). Therefore, we calculate λ, under the reasonable assumption that the total RIs from the fSST experiments represent the corresponding ERFs with chemical rapid adjustments included (Winterstein et al., 2019), as 0.61 ± 0.17 K W^{-1} m^2 and 0.72 ± 0.07 K W^{-1} m^2, respectively.

l. 99: I suppose methane is not an emission flux then? Would be good to clarify to avoid misunderstandings.

Yes, that's right. The CH_4 mixing ratios are prescribed at the lower boundary.

We will add a clarifying sentence.

Old, l. 92 Alike the fSST simulations, the CH_4 lower boundary mixing ratios of the mixed layer ocean (MLO) simulations are prescribed by Newtonian relaxation (i.e. nudging).

New, l. 92 Alike the fSST simulations, the CH_4 lower boundary mixing ratios of the MLO simulations are prescribed by Newtonian relaxation (i.e. nudging). Thus, no CH_4 emission flux boundary was used, but pseudo surface fluxes were calculated by the MESSy submodel TNUDGE (Kerkweg et al., 2006) to reach the prescribed CH_4 lower boundary mixing ratios.

In addition, we will reformulate the following sentence.

Old, l. 99 All other prescribed boundary conditions, such as emission fluxes, in the sensitivity simulations are identical to the respective reference simulations and represent conditions of the year 2010 in general.

New, l. 99 Apart from CH_4, all other boundary conditions and emission fluxes used in the sensitivity simulations are identical to the reference simulations and represent conditions of the year 2010 in general.

l. 139: One way of quantifying the importance of the climatological surface temperature differences would be to compare the global mean surface temperatures. I assume those differences should be smaller but possibly more relevant. Given that the MLO simulations are also free-running, could those effects

also just represent some form of internal variability, which, if I understand correctly could still affect the sea ice distribution through atmospheric variability and its effect on SSTs? Higher latitudes can show similarly large variability for fully coupled ocean models. Similar arguments could apply to the NH (cf. l. 143). Looking at Fig. S1, I would think that the overall difference is positive, but the visual effect overemphasizes those changes in SH high latitudes which make up quite a small area. For climate sensitivity aspects, I would actually be more interested in the differences in tropical low-cloud regions which appear to stand out?

You are right, the differences are smaller on the global scale than at higher latitudes. The highest differences occur near the sea ice edge, which poses the largest challenge to being reproduced by a thermodynamic ocean/ice model. While avoiding to let this section become too long, we have tried to improve the balance in the discussion of regional and global differences.

Old, l. 136 The reduction of sea ice concentration (SIC) results in up to 1.5 K higher SSTs in the Southern Ocean in REF MLO compared to the prescribed climatology (see Fig. S1). Zonal mean air temperatures in the Southern Hemisphere (SH) extra-tropical troposphere are likewise up to 1 K higher in REF MLO compared to REF QFLX on annual average (not shown). As the contribution of Antarctic sea ice melting to global surface albedo feedback and climate response is comparatively small, a substantial underestimation of the climate sensitivity from this effect is not to be expected.

In the Northern Hemisphere (NH), the monthly climatology of sea ice area is generally well reproduced (see Fig. S2). However, in boreal winter and spring REF MLO overestimates the prescribed climatology of sea ice area with a maximum deviation of 1.33×10^9 km^2 in April. The larger SICs result in about 0.5 K lower SSTs on annual average in REF MLO in the Greenland Sea and in the Barents Sea (see Fig. S1), where the increase of SIC is located (not shown). In the Hudson Bay and in the Labrador Sea, on the other hand, the sea ice cover is reduced in REF MLO resulting in about 1 K higher SSTs in REF MLO compared to the prescribed climatology (see Fig. S1). The deviation from the prescribed climatology is strongest in this region in boreal summer. In summary, REF MLO simulates sufficiently realistic oceanic conditions for our purpose.

New, l. 136 The reduction of SIC results in up to 1.5 K higher SSTs in the Southern Ocean in REF MLO compared to the prescribed climatology (see Fig. S1). In the NH, the annual cycle of the sea ice area is generally well reproduced (see Fig. S2), except for a slight overestimation of the sea ice area in REF MLO resulting in about 0.5 K lower annual mean SSTs in the Greenland Sea and in the Barents Sea (see Fig. S1). However, the sign of the global and annual mean surface temperature difference between REF MLO and REF fSST is determined by the positive REF MLO bias related to the Antarctic sea ice reduction. The global mean difference is 0.28 K, much less than the regional maxima near the ice edges, and with a small contribution of about 0.10 K from the tropical belt. It is unlikely that this will lead to substantial biases in the estimation of global mean surface temperature response and climate sensitivity in the intended equilibrium climate change simulations.

l. 174/175: "would be beyond the scope"?

Yes, thank you for this suggestion. We will reformulate the sentence.

l.180: It would indeed be useful to see the overall response, the rapid adjustment response and the difference due to slow feedbacks as subplots next to each other.

We understand that our previous presentation was difficult to follow, especially when not knowing the study of Winterstein et al. (2019). We decided to show 2x2 panel plots of the full response (MLO) and the slow feedbacks (difference between MLO and fSST) for S2 and S5 for temperature, OH, H_2O, and O_3. This should simplify the interpretation of the slow response. However, we decided to not show the rapid adjustments (fSST) again as this would duplicate the work of Winterstein et al. (2019). As the slow feedbacks impose only small modifications, the patterns of the full response and the rapid adjustments are qualitatively very similar and it should be possible to follow the presentation.

l. 193: the tropopause is defined how?

Here, we used a climatological tropopause calculated as:
$\mathrm{tp_{clim}} = 300\ \mathrm{hPa} - 215\ \mathrm{hPa} \cdot \cos^2(\phi)$
The used troposphere definition is recommended by Lawrence et al. (2001), when calculating the CH_4 lifetime. We will add the following sentence to the text.

Old, l. 192 B is the region, for which the lifetime should be calculated, e.g. all grid boxes below the tropopause for the mean tropospheric lifetime.

New, l. 192 B is the region, for which the lifetime should be calculated, e.g. all grid boxes below the tropopause for the mean tropospheric lifetime. For the CH_4 lifetime calculation a climatological tropopause, defined as $\mathrm{tp_{clim}} = 300\ \mathrm{hPa} - 215\ \mathrm{hPa} \cdot \cos^2(\phi)$, with ϕ being the latitude in degree north, is used as recommended by Lawrence et al. (2001).

l. 228: revise sentence

In response to a comment by referee 1, we have restructured the whole paragraph (see answer to referee 1 to reply to line 232).

l. 258: you mean 'stratospheric abundance'

Actually, we referred to the abundance of H_2O in the troposphere and the stratosphere here. The abundance of tropospheric H_2O is indirectly influenced by CH_4 through the CH_4-induced tropospheric warming. However, we admit that the first sentence was not very meaningful and we decided to restructure the paragraph.

Old, l. 258 H_2O is a precursor of OH and its abundance is also influenced by CH_4 mixing ratios. Winterstein et al. (2019) reported a steady increase of H_2O with height for the CH_4 doubling and fivefolding experiments with prescribed SSTs and SICs. Figure 6 shows the difference of the H_2O response between the MLO and the fSST simulations (see Fig. 5 in Winterstein et al., 2019 and Fig. S8 for the respective response patterns of H_2O in the fSST and the MLO simulations, respectively). As the saturation vapour pressure increases with temperature, the warming of the troposphere in the MLO simulations consistently leads to a stronger increase of the tropospheric H_2O mixing ratio in comparison with the respective fSST simulation. The maximum difference between MLO and fSST can be found in the upper tropical troposphere and extratropical lowermost stratosphere and reaches 11 percentage points (p.p.) (35 p.p.) for the 2× (5×) CH_4 experiments.

New, l. 258 Winterstein et al. (2019) reported a steady increase of stratospheric water vapour (SWV) with height for the fSST experiments as an outcome of the enhanced CH_4 depletion as discussed in the previous

paragraph, whereas tropospheric H_2O remained largely unaffected. The warming of the troposphere in the MLO simulations consistently leads to an increase of the H_2O mixing ratios also in the troposphere as evident from Fig. 6. The maximum difference in tropospheric H_2O response between MLO and fSST can be found in the upper tropical troposphere and extratropical lowermost stratosphere and reaches 11 p.p. (35 p.p.) for the 2× (5×) CH_4 experiments.

Figure 6: another case where it would be useful to see the overall response as well instead of just the difference to the rapid adjustment response. Same for Figure 7. 2x2 panels.

Yes, we agree. As stated in the answer to the previous remark to line 180, we will show 2x2 panel plots of the full response (MLO) and the slow feedbacks (difference between MLO and fSST) for S2 and S5 for temperature, OH, H_2O, and O_3.

l. 334: the efficacy of ERF methane of close to 1 appears surprising to me – see e.g. the 145 Hansen et al. Efficacy of climate forcings, Journal of Geophysical Research (2005).

Looking at Table 1 of Hansen et al. (2005) we find efficacy values between 1.05 and 1.08 under the effective radiative forcing framework (with 1.5xCO2, equivalent to a forcing of 2.38 Wm-2 as a reference). This may seem at odds with the most recent work of Richardson et al. (2019), who suggest a CH_4 efficacy value well below 1. However, in their work the reference is 2xCO2 (equivalent to about 4 Wm-2, while the 3xCH4 simulation runs with 1.2 Wm-2 only). This is a dangerous comparison as the climate sensitivity parameter tends to depend on the strength of the forcing. Compare, e.g., with Hansen et al. (2005)'s 1.25xCO2 and 2xCO2 runs, and it becomes obvious that 3xCH4 vs. 1.25xCO2 would probably make a more fair comparison. Many recent studies also show, how delicate the climate sensitivity parameter of CO_2 can depend on the forcing strength.
We think, however, that the main difference between previous work and our study is the inclusion of ozone and water vapor contributions to the methane forcing. Thus, in a chemistry-climate model, the "effective climate sensitivity of methane" will probably contain components from pure CH_4, pure O_3, and pure stratospheric H_2O. Hence, the finding of an efficacy close to 1 in our framework is indeed a surprise that deserves further investigation.

l. 368: how is this calculation of the effect on stratospheric temperatures done precisely? Could you provide more detail about the calculations? Are they expected to be robust in different regimes of the atmosphere, e.g. in the lowermost stratosphere vs the tropical upper stratosphere? What is "addst" in equation (2)?

We feel that from our previous formulation it was not clear that the stratospheric adjusted temperature response is the one shown in Fig. 9 and Fig. S11. We will formulate this clearer and use the abbreviation ΔT_{adj} already, when introducing the calculation of stratospheric adjusted temperatures. "addst" is the EMAC internal abbreviation for the adjusted stratospheric temperatures. We agree that the naming is not very intuitive and will replace it by "adj".
The calculation of the adjusted temperatures response is regime-independent. However, it is not meaningful if the radiatively induced temperature adjustment initiates dynamic processes whose effects on the temperature field are stronger than the radiatively induced changes. This would be the case in the troposphere. However, as the stratosphere is highly stable the radiatively induced temperature response dominates. This is the case in the lower as well as in the upper stratosphere.

Old, l. 354 Following Winterstein et al. (2019) we calculate the stratospheric adjusted temperature response to changes in CH_4, tropospheric and stratospheric H_2O, and tropospheric and stratospheric O_3, as well as

their individual contributions for S2 MLO and S5 MLO (see Fig. S11 for simulation S2 MLO and Fig. 9 for simulation S5 MLO).
The difference of the adjusted stratospheric temperature response between S5 MLO and S5 fSST is shown in Fig. 10 (for S2 see Fig. S12).

New, l. 354 Following Winterstein et al. (2019) we calculate the stratospheric adjusted temperature response ΔT_{adj} to changes in CH_4, tropospheric and stratospheric H_2O, and tropospheric and stratospheric O_3, as well as their individual contributions, for S2 MLO and S5 MLO (see Fig. S11 for simulation S2 MLO and Fig. 9 for simulation S5 MLO). ΔT_{adj} represents the temperature response induced by composition changes of radiatively active gases (Stuber et al., 2001).
The difference of ΔT_{adj} between S5 MLO and S5 fSST is shown in Fig. 10 (for S2 see Fig. S12).

Old, l. 368 By calculating the difference between the total temperature response in the regular simulations and the sum of the individual contributions of CH_4, H_2O and O_3 to the adjusted stratospheric temperatures, we attempt to identify the dynamical effect ($\Delta\tilde{T}_{dyn.}$) in the stratospheric temperature response as

$$\Delta\tilde{T}_{dyn.} = \Delta T(\text{SX-REF}) - \Delta T_{addst}(\text{SX*-REF*})$$

with X being either 2 or 5. A similar approach was, for example, used by Rosier and Shine (2000) and Schnadt et al. (2002) to distinguish between the radiative impact of trace gases and dynamical contributions to the total temperature response.

New, l. 368 By calculating the difference between the total temperature response in the regular simulations ΔT and the sum of the individual contributions of CH_4, H_2O and O_3 to the adjusted stratospheric temperatures (ΔT_{adj}^{total}, see Fig. 9 a) and Fig. S11 a)), we attempt to identify the dynamical effect ($\Delta\tilde{T}_{dyn.}$) in the stratospheric temperature response as

$$\Delta\tilde{T}_{dyn.} = \Delta T(\text{SX-REF}) - \Delta T_{adj}^{total}(\text{SX*-REF*})$$

with X being either 2 or 5. A similar approach was, for example, used by Rosier and Shine (2000) and Schnadt et al. (2002) to distinguish between the radiative impact of trace gases and dynamical contributions to the total temperature response.