

# Reply to referee # 1

October 14, 2020

Dear referee,

thank you very much for your comments and suggestions 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 Major concerns

As mentioned above the study is strongly linked to the work by Winterstein et al. (2019). Unfortunately, both studies have been conducted with different model versions. Moreover, the reference simulation for the MLO, REF QFLX, has been performed using a third model version / set-up. I have a hard time understanding why the authors did not simply apply the same model version as in Winterstein et al. (2019)? The authors want to make us believe that the model modifications do not have a significant impact on the outcome and try to circumvent this issue by showing differences of differences, which by the way does not increase readability, but how can you be sure that the climate background state has no impact on the modelled response to 2x (5x) methane (CH<sub>4</sub>)?

We would like to clarify that the slow feedbacks can only be assessed as the shown difference of differences even if exactly the same model version was used. They are defined as the SST-driven contribution to the overall response and can therefore only be assessed as the difference of the overall response (as simulated in the MLO simulations) and the rapid adjustments (as simulated in the fSST simulations). We feel that we have not stated this clearly enough in the previous manuscript. We will state this in the introduction (see remark to line 6) and at the beginning of Sect. 3.3.1 (line 179 ff, see below).

The MLO simulations were performed at a later time than the fSST simulations. The submodel MLO-CEAN was not yet implemented in its full functionality in the model version used for the fSST simulations and backporting was not reasonable due to other changes in the model. Therefore, and also considering the computational cost of the simulations, we decided to run the MLO simulations with the most advanced cleanly defined model version available at that time.

Yes, we find differences in the simulated reference states of REF MLO and REF fSST (as shown for e.g. ozone (O<sub>3</sub>) in Fig. S19), but these differences are small enough that they do not affect the conclusions about the differences between the full response and the rapid adjustments, i.e. the slow feedbacks.

**Old, l. 179 ff** Winterstein et al. (2019) analysed the quasi-instantaneous impact of doubled and fivefold CH<sub>4</sub> mixing ratios on the chemical composition of the atmosphere. In this section we investigate how tropospheric warming and associated climate feedbacks (see Sect. 3.2) modify these rapid adjustment patterns. For this purpose the difference patterns of the mixed layer ocean (MLO) sensitivity simulations are compared to those of the fSST simulations.

**New, l. 179 ff** Winterstein et al. (2019) analysed the quasi-instantaneous impact of doubled and fivefold CH<sub>4</sub> mixing ratios on the chemical composition of the atmosphere. In this section we investigate the respective slow feedbacks that are assessed as the difference between the full response (as simulated in the MLO simulations) and the rapid adjustments (as simulated in the fSST simulations) and therefore visualized as differences of the differences.

As already mentioned above the presentation of the results is strongly linked to the paper by Winterstein et al. (2019). Without knowing that paper, I find it often very difficult to follow the argumentation. For example, the paper discusses on increase in SWV due to enhanced atmospheric methane, but Fig. 6 displays negative changes in SWV as it present the difference SWV response in the MLO and fSST runs. This way of presenting the results is not very intuitive as the reader first has to look into the supplement to find the SWV response to enhanced CH<sub>4</sub> in the MLO runs and then has to think about differences between the fSST and the MLO set-up. For the sake of readability and clarity I suggest to re-structure the paper as follows: First present the results of the MLO runs and move several of the figures provided in the supplement to the main paper, and then discuss the differences to Winterstein et al. (2019), maybe only for one case (2x or 5x), if the paper turns out to become too long.

We understand that it is difficult to follow the presentation of the previous structure without knowing Winterstein et al. (2019). Therefore, we will make the following changes.

- We will add a short summary of the most important findings and conclusions of Winterstein et al. (2019) in the introduction (see remark to line 6). This implies that we do not need to refer to Winterstein et al. (2019) in the results section too often. We will review the results section with regard to this.
- We will show panel plots of the overall response (MLO) and slow feedbacks (difference between MLO and fSST) for temperature, hydroxyl radical (OH), water vapour (H<sub>2</sub>O), and O<sub>3</sub>. This should make it easier to interpret the slow response in comparison to the full response.
- We will also include a short description of the full response where we think it is necessary, e.g. for O<sub>3</sub>. However, as the slow feedbacks represent only small modifications of the rapid adjustments, we think that it is not necessary to discuss the full response separated from the slow feedbacks and, that it would largely repeat the study of Winterstein et al. (2019).

The argumentation is often very qualitative, but not quantitative. A good example is the discussion of SWV changes and their attribution to changes in CPT and CH<sub>4</sub> oxidations. The CPT changes could be transferred into a change in H<sub>2</sub>O entry values, and from the model simulations it should be easy to calculate SWV production from CH<sub>4</sub> oxidation. With that, the importance of both effects could be quantified. This is only one example, but there are several places where some more quantification would be desirable.

We agree that more quantification would be desirable. Therefore, we went through the manuscript and have the following suggestions for improvement, first regarding the discussion about the H<sub>2</sub>O response:

- We had already included the relative change of H<sub>2</sub>O entry values in l. 273, but we will also include the absolute values in ppm as these might be more relevant. We have estimated the H<sub>2</sub>O entry value as the tropical (10°S–10°N) mean H<sub>2</sub>O mixing ratio at 70 hPa following Revell et al. (2016).
- The quantification of stratospheric water vapour (SWV) production from CH<sub>4</sub> oxidation is not so straightforward. It has often been assumed that two H<sub>2</sub>O molecules result from one oxidized CH<sub>4</sub> molecule, but Frank et al. (2018) showed that the yield deviates from two molecules and further varies with height. Tracing the chemical pathways to determine the actual yield of H<sub>2</sub>O is not so trivial and requires a comprehensive tagging mechanism (see also Frank et al., 2018). Another possibility is to estimate H<sub>2</sub>O from CH<sub>4</sub> oxidation as  $H_2O_{CH_4} = H_2O - H_2O_{entry}$ . We have already done this qualitatively when we compared the change of H<sub>2</sub>O entry mixing ratio that is slightly higher in the MLO runs with the response of H<sub>2</sub>O in the middle and upper stratosphere that is lower in the MLO runs. We will calculate it explicitly with the formula above and include a Figure in the supplement.

**Old, l. 271 ff** The SWV mixing ratio at a given location and time can be approximated as the sum of these two terms (Austin et al., 2007; Revell et al., 2016). We calculate the amount of tropospheric H<sub>2</sub>O entering the stratosphere as the tropical (10°N–10°S) mean H<sub>2</sub>O mixing ratio at 70 hPa following Revell et al. (2016). The H<sub>2</sub>O entry mixing ratio increases by about 10 % (40 %) in the CH<sub>4</sub> doubling (fivefolding) experiments (both MLO and fSST). The relative increases are insignificantly higher in both MLO experiments compared to the respective fSST experiment. Furthermore, the zonal mean tropical cold point temperature (CPT) increases in all sensitivity simulations (see Fig. S9). The magnitude and the latitude dependence of the CPT changes are very similar for both doubling and both fivefolding experiments, although slightly larger for the MLO experiments in line with the changes of the H<sub>2</sub>O entry mixing ratio. Changes of the amount of tropospheric H<sub>2</sub>O entering the stratosphere can therefore not explain the differences in the SWV response between MLO and fSST in the middle and upper stratosphere. The increases of the H<sub>2</sub>O entry mixing ratio and the CPT are both slightly stronger in the MLO experiments and would therefore suggest a stronger increase of SWV in the MLO experiments. On the contrary, the increases of SWV are weaker in the middle and upper stratosphere in the MLO experiments compared to fSST. The contribution of the CH<sub>4</sub> oxidation on SWV can explain these weaker increases of SWV in the MLO experiments. The strengthening of the CH<sub>4</sub> oxidation in the stratosphere is weaker in the MLO experiments resulting likewise in a weaker increase of SWV produced by CH<sub>4</sub> oxidation.

**New, l. 271 ff** The SWV mixing ratio at a given location and time can be approximated as the sum of these two terms following Austin et al. (2007); Revell et al. (2016) as

$$H_2O = H_2O_{\text{entry}} + H_2O_{\text{CH}_4}.$$

We calculate the amount of tropospheric H<sub>2</sub>O entering the stratosphere as the tropical (10°S–10°N) mean H<sub>2</sub>O mixing ratio at 70 hPa following Revell et al. (2016). The H<sub>2</sub>O entry mixing ratio increases by 9.08 % (0.14 ppm) in S2 fSST, 9.77 % (0.17 ppm) in S2 MLO, 38.53 % (0.57 ppm) in S5 MLO, and 38.86 % (0.68 ppm) in S5 MLO. Furthermore, the zonal mean tropical CPT increases in all sensitivity simulations (see Fig. S9). Though differences exist between the reference CPT in MLO und fSST, the magnitude and latitudinal structure of the CPT changes are very similar for both doubling and both fivefolding experiments. They are also a bit larger for the MLO experiments (again consistent for the S2 and S5 case), in line with the response of the H<sub>2</sub>O entry mixing ratios. Changes of the amount of tropospheric H<sub>2</sub>O entering the stratosphere can therefore not explain the weaker increase of SWV in the MLO experiments compared to fSST in the middle and upper stratosphere.

To illustrate the effect of CH<sub>4</sub> oxidation on the SWV response, Fig. S8 shows the response of H<sub>2</sub>O from CH<sub>4</sub> oxidation estimated using Eq. 2. As discussed in the previous paragraph, the strengthening of the CH<sub>4</sub> oxidation in the stratosphere is weaker in the MLO experiments. This results in a weaker increase of SWV produced by CH<sub>4</sub> oxidation in the middle and upper stratosphere (see Fig. S8 c) d)) and can explain the difference of SWV response between MLO and fSST as shown in Fig. 6.

In addition, we will also include the following points:

- We will add the tropospheric CH<sub>4</sub> lifetime when only the temperature dependent reaction rate coefficient responds to the forcing (see remark to line 205–214).
- To quantify the composition changes in the tropical lower stratosphere we will give average values of CH<sub>4</sub> and O<sub>3</sub> changes in boxes in this region.

For CH<sub>4</sub>:

**Old, l. 238 ff** Another aspect to note in Fig. 5 is the more than 5×CH<sub>4</sub> increase in the lowermost tropical stratosphere for S5 MLO. This feature indicates enhanced tropical upwelling, which leads to larger CH<sub>4</sub> mixing ratios in the tropical lower stratosphere. This feature is more pronounced in S5 MLO than in S5 fSST, in line with the more pronounced changes of tropical upwelling in the MLO set-up as discussed in Sect. 3.2.

**New, l. 238 ff** Another aspect to note in Fig. 5 is the more than 2× or 5×CH<sub>4</sub> increase in the lowermost tropical stratosphere. This feature indicates enhanced tropical upwelling, which leads to larger CH<sub>4</sub> mixing

ratios in the tropical lower stratosphere. It is more pronounced in the MLO than in the fSST experiments, in line with the more pronounced changes of tropical upwelling in the MLO set-up as discussed in Sect. 3.2. The average deviation from  $2\times$  or  $5\times\text{CH}_4$  for a region in the tropical lower stratosphere ( $30^\circ\text{S}$ – $30^\circ\text{N}$ , 70–20 hPa) is 0.16 % for S2 fSST, 0.37 % for S2 MLO, 0.23 % for S5 fSST, and 1.31 % for S5 MLO.

For  $\text{O}_3$ :

**Old, l. 298** A dominant feature is the stronger decrease of  $\text{O}_3$  in the lowermost tropical stratosphere in S5 MLO compared to S5 fSST of up to 18 percentage points (p.p.). This difference also exists between the S2 simulations, albeit weaker (4 p.p.).

**New, l. 298** A dominant feature is the stronger decrease of  $\text{O}_3$  in the lowermost tropical stratosphere in S5 MLO compared to S5 fSST of up to 18.39 p.p.. The average difference between S5 MLO and S5 fSST for a region in the tropical lower stratosphere ( $30^\circ\text{S}$ – $30^\circ\text{N}$ , 100–20 hPa) is 6.33 p.p.. This difference also exists between the S2 simulations, albeit weaker (with a maximum difference of 4.68 p.p. and an average difference of 1.67 p.p.).

## 2 Specific comments

The title is very general, almost the same meaning as Winterstein et al. (2019).

We will change the title to *Slow Feedbacks Resulting from Strongly Enhanced Atmospheric Methane Concentrations in a Chemistry-Climate Model with Mixed Layer Ocean* to emphasize that this study focuses on the slow SST-driven feedbacks.

L6 and introduction: It would be nice to see a short definition/description of instantaneous and slow responses / feedbacks. Maybe it would be helpful to add a schematic to the paper, which shows the considered processes and clearly separates fast and slow effects.

While we think that the key parameters of the conceptual radiative forcing, radiative feedback, and climate sensitivity framework adopted here, have all been mentioned and defined in the original manuscript, we admit that the referee’s proposal of a compact presentation in the introduction is certainly worthwhile. To account for the referee’s request, we have reorganized and somewhat extended the introduction, starting at l.42. However, since our manuscript already contains a lot of Figures, we tend to not include an additional schematic.

**Old, l. 42** However, these studies did not focus on the climate impact of  $\text{CH}_4$ . Other recent studies assessing climate feedbacks and climate sensitivity of  $\text{CH}_4$  did not include radiative contributions from chemical feedbacks in their analysis (Modak et al., 2018; Smith et al., 2018; Richardson et al., 2019).

Winterstein et al. (2019) assessed chemical feedback processes and their radiative impact (RI) in sensitivity simulations forced by 2-fold ( $2\times$ ) and 5-fold ( $5\times$ ) present-day (year 2010)  $\text{CH}_4$  mixing ratios. As their simulation set-up prescribed sea surface temperatures (SSTs) and sea ice concentrations (SICs) and thus suppressed surface temperature changes, the parameter changes in their simulations have the character of rapid adjustments (e.g., Forster et al., 2016; Smith et al., 2018). In the effective radiative forcing (ERF) framework, rapid adjustments of radiatively active species are counted as part of the forcing and are to be distinguished from slow climate feedbacks that are coupled to surface temperature changes (Sherwood et al., 2015). Climate sensitivity parameters, reflecting the degree of surface temperature change per unit forcing, have been found to be less dependent on the forcing agent with this definition compared to previous definitions of radiative forcing (RF) (e.g., Shine et al., 2003; Hansen et al., 2005; Richardson et al., 2019).

As a follow-up on Winterstein et al. (2019), we assess the respective SST-driven climate feedbacks, their effect on the quasi-instantaneous response of the chemical composition, and consequently resulting radiative feedbacks. Consistent with Winterstein et al. (2019), we perform sensitivity simulations with  $2\times$  and  $5\times$  present-day  $\text{CH}_4$  mixing ratios with the ECHAM/MESSy Atmospheric Chemistry (EMAC) chemistry-climate model (CCM) (Jöckel et al., 2016), but this time coupled to a MLO model instead of prescribing SSTs and SICs. To our knowledge, this is the first study assessing the response to strong increases of  $\text{CH}_4$  mixing ratios in a fully coupled CCM, meaning that the interactive model system includes atmospheric dynamics, atmospheric chemistry, and ocean thermodynamics.

**New, I. 42** However, these studies did not focus on the climate impact of  $\text{CH}_4$ . In climate feedback and sensitivity studies it has become standard to distinguish between rapid adjustments of the system (that develop in direct reaction to the forcing, independently from sea surface temperature changes) and feedbacks driven by slowly evolving temperature changes at the Earth’s surface (e.g., Colman and McAvaney, 2011; Geoffroy et al., 2014; Smith et al., 2020). Under this concept, the rapid radiative adjustments are counted as an integral part of the radiative forcing, yielding the so-called effective radiative forcing (Shine et al., 2003; Hansen et al., 2005). The concept has been found to be physically more meaningful than other radiative forcing frameworks, as the climate sensitivity parameter, i.e., the global mean surface temperature change per unit radiative forcing, is becoming less dependent on the forcing agent (Hansen et al., 2005; Sherwood et al., 2015; Richardson et al., 2019). However, recent studies of climate feedbacks and sensitivity to a  $\text{CH}_4$  forcing adopting the effective radiative forcing concept did not account for the radiative contribution from chemical feedbacks in their analysis (Modak et al., 2018; Smith et al., 2018; Richardson et al., 2019).

Winterstein et al. (2019) assessed chemical feedback processes and their RI in simulations forced by 2-fold ( $2\times$ ) and 5-fold ( $5\times$ ) present-day (year 2010)  $\text{CH}_4$  mixing ratios. As their simulation set-up used prescribed sea surface temperatures (SSTs) and sea ice concentrations (SICs) and thus suppressed surface temperature changes, the parameter changes in their simulations match the rapid adjustment and effective radiative forcing concept (e.g., Forster et al., 2016; Smith et al., 2018). Rapid radiative adjustments to stratospheric ozone and water vapor changes were found to make a considerable contribution to the  $\text{CH}_4$  effective radiative forcing, in line with previous respective findings (e.g., Shindell et al., 2005, 2009; Stevenson et al., 2013). SWV mixing ratios were found to increase steadily with height under increased  $\text{CH}_4$  in the quasi-instantaneous response as analysed by Winterstein et al. (2019). Rapid adjustments of the chemical composition of the stratosphere lead to increases of OH favoring the depletion of  $\text{CH}_4$ , which is an important in situ source of SWV. The increased SWV mixing ratios cool the stratosphere, thereby affecting  $\text{O}_3$ . In the troposphere, the enhanced  $\text{CH}_4$  burden leads to a strong reduction of its most important sink partner, OH, thereby affecting the  $\text{CH}_4$  lifetime. Winterstein et al. (2019) found a near-linear prolongation of the tropospheric  $\text{CH}_4$  lifetime with increasing scaling factor of  $\text{CH}_4$  for the two conducted experiments ( $2\times$  and  $5\times\text{CH}_4$ ).

As a follow-up on Winterstein et al. (2019), we assess the respective slow SST-driven response of the chemical composition and resulting radiative feedbacks. Consistent with Winterstein et al. (2019), we perform sensitivity simulations with  $2\times$  and  $5\times$  present-day  $\text{CH}_4$  mixing ratios with the EMAC CCM (Jöckel et al., 2016), but this time coupled to a MLO model instead of prescribing SSTs and SICs. For radiative forcing strengths as discussed here, equilibrium climate sensitivity simulations using a thermodynamic mixed layer ocean as lower boundary condition have been shown to represent the surface temperature response yielded in (much more resource demanding) model setups involving a dynamic deep ocean sufficiently well (e.g., Danabasoglu and Gent, 2009; Dunne et al., 2020; Li et al., 2013). The slow feedbacks are assessed as the difference between the full response (as simulated in the MLO simulations) and the rapid adjustments (as simulated in the simulations with prescribed SSTs and SICs). To our knowledge, this is the first study assessing the response to strong increases of  $\text{CH}_4$  mixing ratios in a fully coupled CCM, meaning that the interactive model system includes atmospheric dynamics, atmospheric chemistry, and ocean thermodynamics.

L90/91: I am bit confused by the description of the applied  $\text{CH}_4$  boundary condition. I thought that  $\text{CH}_4$  is relaxed towards to observational data set, and that this data set is simply multiplied by 2 (5) for the sensitivity runs. Why the “equilibrium  $\text{CH}_4$  fields of the respective fSST simulations”? What is the

difference / advantage?

We apply the nudging of the CH<sub>4</sub> mixing ratio to the observational data set only at the lower boundary. The atmospheric CH<sub>4</sub> mixing ratios are free to adjust to this forcing. In the stratosphere, for example, the increase of CH<sub>4</sub> mixing ratio deviates from the increase factors of 2 and 5, respectively. As the equilibrium fields of CH<sub>4</sub> mixing ratio from the fSST experiments are already close to the respective equilibrium of the MLO simulations, the initialization with these fields shortens the spin-up. We will reformulate the sentence to state this point more clearly.

**Old** The MLO simulations have been initialized with the equilibrium CH<sub>4</sub> fields of the respective fSST simulations, thus the initial CH<sub>4</sub> fields of S2 MLO and S5 MLO were implicitly scaled by two and five, respectively.

**New** The MLO simulations have been initialized with the equilibrium CH<sub>4</sub> fields of the respective fSST simulations. As the latter are already close to the respective equilibrium CH<sub>4</sub> fields of the MLO simulations, the initialization with these fields shortens the spin-up.

L91 onwards: What is the advantage / difference between the relaxation approach and simply prescribing the CH<sub>4</sub> concentration at the surface? What relaxation timescale is used? With the long lifetime of CH<sub>4</sub> there should not be a large difference?

Indeed, it is in principle the same as the relaxation time (10800 s) is short in comparison with the CH<sub>4</sub> lifetime and transport times. We will add the nudging coefficient to the manuscript.

**Old** Alike the fSST simulations, the CH<sub>4</sub> lower boundary mixing ratios of the MLO simulations are prescribed by Newtonian relaxation (i.e. nudging).

**New** Alike the fSST simulations, the CH<sub>4</sub> lower boundary mixing ratios of the MLO simulations are prescribed by Newtonian relaxation (i.e. nudging) with a nudging coefficient of 10800 s.

REF QFLX: This simulation should be the same as REF fSST, shouldn't it? Does REF MLO also include the gravity wave set-up as described in Appendix B? If not, do you expect any impact?

In principle, REF QFLX should be the same as REF fSST, but the simulations were performed with different model versions. ALL MLO simulations use the same gravity wave set-up as the fSST simulations for consistency. The different gravity wave set-up does mainly influence the middle atmosphere. We therefore presume that the influence on the ground is so small that the heat flux correction is not affected.

**Old** In the REF QFLX simulation the setting of the non-orographic gravity wave drag parameterization (GWAVE, Baumgaertner et al., 2013) was different than in the other simulations, ...

**New** In the REF QFLX simulation the setting of the non-orographic gravity wave drag parameterization (GWAVE, Baumgaertner et al., 2013) was different than in all the other simulations (fSST and MLO), ...

L127/128: What is the reason for the negative bias and observed total column CH<sub>4</sub>? This is a good example where an explanation seems to be given in Winterstein et al. (2019), but is unfortunately not

summarized in the present study.

Thank you for this note. We will add a short explanation to the text.

**Old, l. 127 ff** Consistent with REF fSST (see Winterstein et al., 2019), there is a negative bias between the REF MLO and the observed total CH<sub>4</sub> columns of less than 4 % (not shown). Given that relative comparisons between sensitivity simulations and the reference are the main target of our analysis, REF MLO represents CH<sub>4</sub> conditions of the year 2010 sufficiently realistic for our purpose.

**New, l. 127 ff** Consistent with REF fSST (see Winterstein et al., 2019), there is a negative bias between the REF MLO and the observed total CH<sub>4</sub> columns of less than 4 % (not shown). Note that not all the observations originate precisely from the year 2010. The global annual mean CH<sub>4</sub> surface mixing ratios have, for example, risen by about 0.024 ppm from 2010 to 2014 ([www.esrl.noaa.gov/gmd/ccgg/trends\\_ch4/](http://www.esrl.noaa.gov/gmd/ccgg/trends_ch4/)), the year of the study by Klappenbach et al. (2015). In addition, the CH<sub>4</sub> lifetime could be slightly underestimated. The CH<sub>4</sub> lifetime in EMAC lies in the middle to lower range in comparisons with other CCMs (Jöckel et al., 2006; Voulgarakis et al., 2013). However, given that relative comparisons between sensitivity simulations and the reference are the main target of our analysis, REF MLO represents CH<sub>4</sub> conditions of the year 2010 sufficiently realistic.

L136/137, Fig. S1: I have also worked with the ECHAM5 MLO, and I am a bit concerned about the difference pattern shown in Fig. S1, namely the temperature difference around 60S, especially over the eastern hemisphere. In my simulation the MLO was in much better agreement with the reference SST climatology. Any thoughts about this?

We have derived the flux correction at the surface that stabilizes the MLO reference run from the surface fluxes of the fixed SST reference run. If you did likewise in your coupling exercise, one possibility could be that your basic model (with fixed SSTs) has had an ideally balanced top of the atmosphere radiation balance, with optimally low correction fluxes. (In our case the original global radiation balance was -1.14 Watt per square meter ( $\text{W m}^{-2}$ ) for REF fSST.) As the largest temperature deviations occur near the ice edge, another possibility could be that you provided a multiple iteration of the correction fluxes in these regions to ensure optimal reproduction of the ice edge location in the reference run with MLO. Did you?

MLO: A more general question to the MLO: The MLO does not consider heat exchange with the deep ocean, but all forcing goes into the MLO. Up to which forcing strength is the usage of an MLO justified?

We feel that there is robust, long standing evidence for a sufficient reproduction of climate sensitivity parameters simulated by deep ocean coupled AOGCMs by MLO coupled AOGCMs in case of forcing strengths at least up to carbon dioxide (CO<sub>2</sub>) doubling (Danabasoglu and Gent, 2009; Dunne et al., 2020). This evidence has been explicitly confirmed for the ECHAM5 climate model (Li et al., 2013), which is the atmospheric model basic to the chemistry-climate model setup used in our paper. Problems may arise for larger forcings (4xCO<sub>2</sub> and higher) with strong ocean mixed layer warming, which is not transferred to the deep layers, but as our forcings are much smaller than for CO<sub>2</sub> doubling, that should not be an issue here.

We will add a clarifying sentence to the paragraph introducing the MLO setup (l. 58, see also remark to line 6):

**Old, l. 58** ... of prescribing SSTs and SICs.

**New, l. 58** ... of prescribing SSTs and SICs. For radiative forcing strengths as discussed here, equilibrium climate sensitivity simulations using a thermodynamic mixed layer ocean as lower boundary condition have been shown to represent the surface temperature response yielded in (much more resource demanding) model setups involving a dynamic deep ocean sufficiently well (e.g., Danabasoglu and Gent, 2009; Dunne et al., 2020; Li et al., 2013).

L173 onwards: If Bony et al discussed a similar feature for CO<sub>2</sub>, then why not adding a short (speculative) discussion for CH<sub>4</sub>?

Our previous formulation was a bit vague. What we wanted to indicate is the following: Bony et al. (2013) found differences between the fast and the slow (temperature driven) response of the tropospheric tropical circulation in CO<sub>2</sub> increase experiments. We will state that more clearly. However, we still think that a detailed discussion of the processes leading to these differences is beyond the scope of this paper. As proposed by the second referee, Peer Nowack, we will add an outlook on tropospheric circulation changes in CH<sub>4</sub> increase simulations as this is surely an interesting research question by itself.

**Old, l. 173 ff** A similar feature has been noticed and discussed in CO<sub>2</sub> increase simulations, too (e.g. Bony et al., 2013). However, ...

**New, l. 173 ff** Differences between the fast and the slow response of the tropospheric tropical circulation have been noticed and discussed in CO<sub>2</sub> increase simulations, too (e.g. Bony et al., 2013). However, ...

L205-214: It would be nice to see some more quantification of the temperature effect on the CH<sub>4</sub> lifetime!

Fig. 3 shows the total effect on the CH<sub>4</sub> lifetime that results from changes of CH<sub>4</sub>, OH and the temperature dependent reaction rate coefficient. A possible quantification of the temperature effect on CH<sub>4</sub> lifetime would be the comparison with the CH<sub>4</sub> lifetime calculated using only a changed reaction rate coefficient corresponding to temperatures of 2× and 5× CH<sub>4</sub>. However, also the abundance of OH is influenced by temperature changes as we show in this study. Therefore, changing only the reaction rate coefficient would not represent the whole temperature/climate effect on the CH<sub>4</sub> lifetime. Nevertheless, we will include the isolated effect of the temperature dependent reaction rate on the CH<sub>4</sub> lifetime in Fig. 3.

**Old, l. 205 ff** Additionally, the tropospheric warming in the MLO sensitivity simulations results in a faster CH<sub>4</sub> oxidation as its reaction rate increases with temperature.

**New, l. 205 ff** Additionally, the tropospheric warming in the MLO sensitivity simulations results in a faster CH<sub>4</sub> oxidation as its reaction rate increases with temperature. The isolated effect of the temperature dependent reaction rate is indicated by the blue squares in Fig. 3. They show the CH<sub>4</sub> lifetime corresponding to REF MLO, except for the reaction rate coefficient that was calculated with temperatures corresponding to 2× and 5× CH<sub>4</sub>.

L232: Why is the tropospheric CH<sub>4</sub> response marginally larger? Tropospheric is largely controlled by boundary condition? Remaining effect from CH<sub>4</sub> oxidation?

Yes, we think you are absolutely right. We will reorganize the paragraph to state this more clearly.



**Old, l. 232 ff** Winterstein et al. (2019) investigated whether the increase of atmospheric CH<sub>4</sub> follows the doubling or fivefolding for fSST conditions linearly. Tropospheric CH<sub>4</sub> is largely controlled by the nudging at the lower boundary through mixing and responds linearly to the increase. However, the CH<sub>4</sub> increase between 50 and 1 hPa has found to be smaller than a strictly linear relation would predict. This indicates enhanced chemical CH<sub>4</sub> depletion in the stratosphere due to changes in the chemical composition. Fig. ?? shows the relative difference between the annual zonal mean CH<sub>4</sub> of S2 MLO (S5 MLO) and 2× (5×) the zonal mean CH<sub>4</sub> of REF MLO. The doubling or fivefolding of the reference CH<sub>4</sub> serves to emphasize regions where the increase factor of the CH<sub>4</sub> mixing ratio deviates from 2 or 5, respectively. The response of tropospheric CH<sub>4</sub> is marginally larger than a linear increase in both MLO experiments. This is in line with the response of tropospheric CH<sub>4</sub> in the fSST simulations. As for the fSST simulations, the CH<sub>4</sub> increase in the extratropical stratosphere is weaker than a linear increase in both MLO sensitivity simulations. The non-linearity is less pronounced in the two MLO sensitivity experiments compared to the respective fSST experiments (compare with Fig. 3 in Winterstein et al., 2019) suggesting that the chemical depletion of CH<sub>4</sub> is enhanced in the MLO experiments as well, however, less strongly than in the fSST experiments.

**New, l. 232 ff** Fig. 5 shows the relative differences between the annual zonal mean CH<sub>4</sub> of S2 MLO (S5 MLO) and 2× (5×) the zonal mean CH<sub>4</sub> of REF MLO. The doubling or fivefolding of the reference CH<sub>4</sub> serves to emphasize regions where the increase factor of the CH<sub>4</sub> mixing ratio deviates from 2 or 5, respectively. The response of tropospheric CH<sub>4</sub> is marginally larger than a linear increase in both MLO experiments. This is in line with the response of tropospheric CH<sub>4</sub> in the fSST simulations. Tropospheric CH<sub>4</sub> is largely controlled by the nudging at the lower boundary through mixing and is, therefore, prevented to adjust to the lifetime increase as discussed above. The slightly positive values in Fig. 5 indicate a small residual of this effect. As for the fSST simulations, the CH<sub>4</sub> increase between 50 and 1 hPa is smaller than the factors of 2 or 5, respectively. This effect is less pronounced in the two MLO sensitivity experiments compared to the respective fSST experiments (compare with Fig. 3 in Winterstein et al., 2019) suggesting that the chemical depletion of CH<sub>4</sub> is enhanced in the MLO experiments as well, however, less strongly than in the fSST experiments.

L246 onwards: Again the argumentation in this section stays mainly qualitative (“weaker increases of OH are presumably connected...”). Although the arguments sound reasonable, it should be possible to keep track of chemical production / loss budgets in a CCM

Unfortunately, it is not trivial to keep track of the chemical production and loss budgets of OH in a comprehensive chemical mechanism such as MECCA. It is theoretically possible, but would require a complex tagging mechanism as presented by, e.g., Gromov et al. (2010). In the present simulations we did not use this mechanism as it is computationally expensive and can, therefore, not be applied to global simulations that cover multiple decades. For simple mechanisms, as for example the CH<sub>4</sub> sink reactions, keeping track of the budget is straightforward.

L293: Which one is the limiting OH precursor? Water vapor or ozone? I would imagine that depends on the atmospheric region?

As already replied to the previous remark, it is not easy to determine the production and loss budgets of OH from our simulation results. Determining the more important OH precursor is also not straightforward and would require additional calculations. Nicely et al. (2020), for example, assessed the contribution of various drivers to the CH<sub>4</sub> lifetime long-term trend (as proxy for OH) with a machine learning algorithm.

Here, we can only speculate if H<sub>2</sub>O or O<sub>3</sub> is the limiting precursor for stratospheric OH. Our reasoning here is that, as the increase in OH is smaller in the MLO runs, while the entry of tropospheric H<sub>2</sub>O is stronger, the limiting precursor is presumably O<sub>3</sub>.

L297: Please add a short summary of the explanation for the O<sub>3</sub> response given in Winterstein et al. (2019).

We will add a short summary of the explanation for the O<sub>3</sub> response.

**Old, l. 296 ff** Winterstein et al. (2019) gave a detailed explanation of the processes leading to the resulting O<sub>3</sub> pattern that is also valid for the MLO simulations.

**New, l. 296 ff** Winterstein et al. (2019) gave a detailed explanation of the processes leading to the resulting O<sub>3</sub> pattern that is also valid for the MLO simulations. As the O<sub>3</sub> catalytic depletion cycles are less efficient at lower temperatures radiative cooling in the stratosphere results in increased O<sub>3</sub> mixing ratios in the middle stratosphere (between 50 and 5 hPa). Additionally, increased abundances of H<sub>2</sub>O favor the depletion of excited oxygen (O(<sup>1</sup>D)), likewise reducing the sink of O<sub>3</sub> and favoring increases of the O<sub>3</sub> abundance. Reduced O<sub>3</sub> mixing ratios in the lowermost tropical stratosphere indicate enhanced tropical upwelling of O<sub>3</sub> poor air from the troposphere into the stratosphere. Above 2 hPa, increases of OH lead to enhanced depletion of O<sub>3</sub> resulting in reduced O<sub>3</sub> mixing ratios.

Fig. S9: Would be nice to see the difference in CPT for the reference simulations, fSST and MLO, as well.

We will include the difference of cold point temperature of REF MLO and REF fSST in Fig. S9. In addition, we will make the following change to the manuscript (see also answer to major concern 3)).

**Old, l. 276 ff** The magnitude and the latitude dependence of the CPT changes are very similar for both doubling and both fivefolding experiments, although slightly larger for the MLO experiments in line with the changes of the H<sub>2</sub>O entry mixing ratio.

**New, l. 276 ff** Though differences exist between the reference CPT in MLO und fSST, the magnitude and latitudinal structure of the CPT changes are very similar for both doubling and both fivefolding experiments. They are also a bit larger for the MLO experiments (again consistent for the S2 and S5 case), in line with the response of the H<sub>2</sub>O entry mixing ratios.

### 3 Technical corrections

**Page 7, line 189, Equation (1):** is there a bug in the listed units? E.g., units for reaction rate coefficient? [cm<sup>3</sup> mol<sup>-1</sup> s<sup>-1</sup>]? Otherwise the lifetime is not in [s].

**Page 7, line 190:** [kg], to be consistent with the other units.

**Pages 14-15, lines 324-325:** It is not necessary to additionally mention numbers listed in a table here.

Thank you for these suggestions and corrections. We fully agree and changed the manuscript accordingly. The unit of the concentration in Eq. (1) is [cm<sup>-3</sup>].