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Dear Editor,

we have modified the original manuscript of our paper in accordance with the replies to the referees, as requested. In particular, there are some changes with respect to the figures, which have now different headlines (as the designation of the simulations has been changed). One figure originally part of the supplement is now in the main paper (as Figure 8), while one new figure has been included in the supplement (all according to the referees' recommendation).

A manuscript version with all text changes specifically marked has also been uploaded.

Thanks and best regards
Franziska Winterstein and co-authors

Implication of ~~extreme~~ strongly increased atmospheric methane concentrations for chemistry-climate connections

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Abstract. Methane (CH₄) is the second most important directly emitted greenhouse gas, ~~which atmospheric concentration the~~ atmospheric concentration of which is influenced by human activities. In this study, numerical simulations with ~~a chemistry-climate model~~ the CCM EMAC are performed aiming to assess possible consequences of significantly enhanced CH₄ concentrations in the Earth's atmosphere for the climate.

5 We analyze experiments with 2xCH₄ and 5xCH₄ present day (2010) mixing ratio and its quasi-instantaneous chemical impact on the atmosphere. The massive increase in CH₄ strongly influences the tropospheric chemistry by reducing the hydroxyl radical (OH) abundance and thereby extending the CH₄ lifetime as well as the residence time of other chemical ~~po~~llutantssubstances. The region above the tropopause is impacted by a substantial rise in stratospheric water vapor (SWV). The stratospheric ozone (O₃) column increases overall, but SWV induced stratospheric cooling also leads to an enhanced ozone
10 depletion in the Antarctic lower stratosphere. Regional patterns of ozone change are affected by modification of stratospheric dynamics, i.e. increased tropical up-welling and stronger meridional transport towards the polar regions. We calculate the net radiative impact (RI) of the 2xCH₄ experiment to be 0.69 W/m² and for the 5xCH₄ experiment to be 1.79 W/m². A substantial part of the RI is contributed by chemically induced O₃ and SWV changes, in line with previous radiative forcing estimates.

To our knowledge this is the first numerical study using a CCM with respect to two/fivefold CH₄ concentrations and it is
15 therefore an overdue analysis as it emphasizes the impact of possible strong future CH₄ emissions on atmospheric chemistry and its feedback on climate.

1 Introduction

Methane (CH₄) is a potent greenhouse gas (GHG), subject to strong anthropogenic emissions that contribute substantially to global warming. It is not just by itself a radiatively active gas but is chemically active as well, strongly influencing the
20 chemical composition of the atmosphere. Beyond that its sources are prone to temperature and it is generally expected that climate change (i.e. surface warming) will lead to enhanced CH₄ emissions, accelerating the temperature rise. For instance additional CH₄ emissions are expected from wetlands due to climate-driven changes (Gedney et al., 2004; Zhang et al., 2017; Ma et al., 2017). Moreover, a large quantity of CH₄ is stored as methane hydrate not only in permafrost soil but also in the sea floor. Permafrost soil stores about a hundredfold of the current CH₄ burden in the atmosphere and oceanic methane hydrates

store even a thousandfold (IPCC, 2013). Current estimates indicate that GHG emissions from thawing permafrost soils could represent a major terrestrial biogeochemical feedback to climate change over the coming decades (Comyn-Platt et al., 2018).

At the same time, it is under debate whether a possible strong release of CH₄ from thawing permafrost in the Arctic region could potentially force an abrupt climate change (as discussed by O'Connor et al., 2010). At present, the release of methane hydrate from reservoirs is highly uncertain as well as the magnitude of future natural and anthropogenic emissions of methane. Increasing surface temperatures cause enhanced CH₄ emissions from thawing permafrost soils to the atmosphere, but the amount is currently poorly constrained (Hayes et al., 2014; Schaefer et al., 2014; Koven et al., 2015; Schuur et al., 2015). For instance, Dean et al. (2018) stated that there is basically no significant increase of Arctic methane emissions at the moment, though it may increase towards the end of the ~~21st~~21st century.

Nevertheless, permafrost thaw could potentially release trapped CH₄ and transform frozen soil to wetland areas, which would then add to Arctic CH₄ emissions. Moreover, ongoing heating of the Arctic sea surface temperature (SST) will also enhance future CH₄ production in the ocean, and a reduction of sea ice concentration (SIC) may increase the direct transfer of CH₄ from the ocean to the atmosphere. In particular, enhanced SST can increase the production of CH₄ as permafrost underlying the continental shelf begins to thaw (Miller et al., 2018). How a changing climate will impact future CH₄ emissions remains a topic of debate in atmospheric science, since emissions from the most climate sensitive CH₄ sources, i.e. wetlands, are difficult to quantify precisely.

Although there remain important knowledge gaps about the magnitude of CH₄ emissions, it is important to improve our understanding of how strongly future CH₄ emissions may impact our atmosphere and the environment. About 90% of the emitted CH₄ is removed in the troposphere. A change in tropospheric CH₄ concentration affects the oxidizing capacity of the atmosphere, modifies ozone in the troposphere and influences the CH₄ lifetime itself (e.g. Saunio et al., 2016; Frank, 2018; Holmes, 2018). Additionally, it also ~~influences~~affects the stratosphere. For example, enhanced CH₄ emissions will lead to increased abundance of stratospheric water vapor (SWV) and ~~by due to~~ this strongly influence stratospheric ozone (O₃) (Stenke and Grewe, 2005; Revell et al., 2016).

To assess the direct and indirect effects of ~~severely~~strongly enhanced CH₄ emissions on atmospheric composition and Earth climate, numerical model studies are able to support investigations such as to identify potential signatures impacting climate change. So far only a limited amount of numerical studies are available concerning the impact of very strong CH₄ emissions. Exemplary, the effect of twofold CH₄ was investigated in a 1D radiative-convective climate model by Owens et al. (1982) and by MacKay and Khalil (1991). Shang et al. (2015) used a Chemical Transport model (CTM) but doubled CH₄ emission over China only. Other CTM studies have focused on recent changes and fluctuations of the atmospheric CH₄ concentration (e.g. Dalsøren et al., 2016) or have tried to explain CH₄ trends, which is a challenge because of important uncertainties in the global CH₄ budget, i.e. the balance of surface sources and atmospheric and surface sinks (Saunio et al., 2016). Furthermore, CTMs are limited in assessing climate change related issues, because they do not include the feedback between chemistry and dynamics. Smith et al. (2018) investigated the fast radiative feedbacks (adjustments) in a model-intercomparison using simulations with 3×CH₄ simulation without considering the chemical feedback effects. This investigation includes only physical components of the atmosphere, like direct cloud, water vapor and temperature adjustments, and the total radiative adjustment ~~result~~

resulted in a value near zero. This example even more motivates an assessment of simulations that include chemically driven atmospheric adjustments to increases of CH₄.

To our knowledge studies using data derived from chemistry-climate model (CCM) simulations including extreme CH₄ emissions (i.e. beyond current and near-future amounts) are not available so far. A CCM is an atmospheric global circulation model that is interactively coupled to a detailed chemistry module. In contrast to CTMs, in CCMs the simulated concentrations of the radiatively active gases are used for the calculations of net heating rates. Changes in the abundance of these gases due to chemistry and advection influence heating rates and, consequently, variables describing atmospheric dynamics. This creates a dynamical-chemical coupling in which the chemistry influences the dynamics and vice versa. Since CH₄ influences other trace gases due to its oxidation products as well as the removal of hydroxyl radical (OH), a comprehensive chemistry module is necessary. In simulations with doubled carbon dioxide (CO₂), in contrast, the feedback on climate and chemistry is induced only by its radiative impact. Apart from accounting for the direct radiative impact of CH₄ the use of a CCM is strongly desired, since the atmospheric CH₄ chemical feedback is a key process for understanding the variations in atmospheric CH₄ and its effects on other chemical constituents of the atmosphere (Holmes, 2018).

The present work is the first study investigating atmospheric effects due to extreme CH₄ emissions with such a CCM. Idealized simulations of significantly enhanced CH₄ concentrations are performed, i.e. twofold (2xCH₄) and fivefold (5xCH₄) enhanced CH₄ concentrations compared to present day condition, allowing to assess possible future consequences for atmospheric composition considering chemical feedback processes. In a first step, ~~which is described in this paper,~~ we conducted CCM simulations without interactive ocean coupling, i.e. the surface conditions regarding SST and SIC are prescribed (suppressed surface temperature feedback). Equivalent to the work of Smith et al. (2018), the results can be interpreted as rapid adjustments to a sudden CH₄ enhancement before the ocean reacts to the perturbation, which would occur on a by far larger time scale.

In this study we will use the ECHAM/MESSy Atmospheric Chemistry (EMAC) CCM (Jöckel et al., 2016) assessing the range of atmospheric responses by abrupt increases of CH₄ concentrations. A short description of EMAC is given in Section 2 as well as an explanation of the simulation strategy. In Section 3 the reference simulation representing near-present day condition is briefly evaluated with observations (Subsect. 3.1) and a discussion of the impact of twofold and fivefold increased CH₄ concentrations in respective scenario simulations is presented in Subsect. 3.2. In the final Section 4 we draw some conclusions from our investigation and give a brief outline for follow-up investigations.

2 Description of the model and simulation strategy

We use the EMAC model in the version 2.52 (Jöckel et al., 2010) and operate it at a resolution of T42L90MA corresponding to a quadratic Gaussian grid of approx. 2.8° x 2.8° in latitude and longitude with 90 levels up to 0.01 hPa. More details on the Modular Earth Submodel System (MESSy) can be found in Jöckel et al. (2016).

We conducted one reference simulation (REF) and two sensitivity simulations (~~S1 and S2~~ and S5) as 20-year time slice simulations representing in general year 2010 conditions. Monthly SST and SIC are thereby repeatedly prescribed, representing

a climatological annual cycle of the years 2000–2009 based on global analyses of Rayner et al. (2003). An at least 10-year long spin-up ~~simulation period~~ preceding each simulation (likewise time slice) ensures quasi steady state conditions, but has been neglected in the evaluation. The spin-up started with initial conditions using a restart file representing the year 2010 of a reference simulation with so called specified dynamics (SD) of the Earth System Chemistry integrated Modelling (ESCiMo) project (Jöckel et al., 2016). To reduce the length of the spin up of the sensitivity simulations we started those using twofold and fivefold CH₄ mixing ratios compared to the initialization of the reference, respectively.

The lower boundary condition of CH₄ in the reference simulation, i.e. the CH₄ surface mixing ratio, is prescribed by Newtonian relaxation (i.e. nudged) following a zonal mean estimate from the National Oceanic and Atmospheric Administration/Earth System Research Laboratory (NOAA/ESRL) based on observations of CH₄ surface mixing ratios. These observations are provided by the Advanced Global Atmospheric Gases Experiment (AGAGE; <http://agage.eas.gatech.edu>) and National Oceanic and Atmospheric Administration/Earth System Research Laboratory (NOAA/ESRL; <http://www.esrl.noaa.gov>). The mean surface mixing ratio is about 1.8 parts per million volume (ppmv). The two sensitivity simulations (~~S1 and S2~~ and S5) are carried out nudged at the surface to a twofold CH₄ surface mixing ratio (compared to the present day reference) ~~being of~~ about 3.6 ppmv and a fivefold CH₄ surface mixing ratio ~~being of about~~ 9.0 ppmv, respectively. Although this does not correspond to an equivalent increase in the surface fluxes, it scales the surface mixing ratio directly to the intended value, as has been done in similar studies (see Kirner et al., 2015; Forster et al., 2016; Smith et al., 2018). To put the chosen scaling factors in perspective, a surface mixing ratio of 3.6 ppmv (similar to the doubling above) will be reached according to the Representative Concentration Pathways (RCP) 8.5 scenario towards the end of the ~~21st~~ 21st century (Riahi et al., 2007). The RCP 8.5 is the so called baseline climate change scenario, which does not employ any climate mitigation target (Riahi et al., 2011). Other prescribed conditions (SST, SIC, CO₂ etc.) of the sensitivity simulations ~~S1 and S2~~ and S5 are identical to REF. Particularly, ~~all other GHG concentrations and online simulated emissions, as well as online simulated trace gas emissions,~~ represent 2010 conditions.

In the following, changes and feedbacks are assessed by comparing the reference simulation REF with ~~S1 and S2~~ and S5, with focus on changes in the simulated chemically and radiatively active trace gases. To quantify the associated radiative impact (RI), the EMAC option for multiple radiation calls is ~~employed~~ used in submodel RAD (Dietmüller et al., 2016) in a separate additional simulation, which allows to estimate individual components of the total radiative impact. This simulation is run for one year (plus one year spin-up) and uses climatological 20 year means of the species of interest ~~;~~ (namely CH₄, O₃ and SWV) from the corresponding reference or sensitivity simulation (REF, S2 and S5). The results ~~corresponding to one of the previously introduced experiments are indicated~~ are indicated hereafter by the associated simulations name and ~~a asterix~~ an asterisk (i.e. REF*, ~~S1* and S2*~~ and S5*). RAD performs multiple radiation calls with different input within one time step. Only the first call is used for providing the radiative heating feedback to the basemodel, while the other calls produce “perturbed” radiative fluxes and stratospheric temperature changes that are used diagnostically for calculating a stratospheric temperature adjusted RI (Stuber et al., 2001; Dietmüller et al., 2016). In our set-up the first call receives the reference mixing ratios of the chemical species while the other calls receive climatological means derived from the sensitivity simulations, replacing either all component species combined or each of the three species individually.

3 Discussion of results

3.1 Evaluation of the reference simulation

The ~~reference simulation REF is set up to represent conditions comparable to the~~ set-up of the reference simulation represents near-present atmospheric conditions ~~in~~ of 2010. To ensure that this simulation is sufficiently realistic, the simulation results
5 of CH₄ mixing ratio in the troposphere and the stratosphere are compared to data derived from atmospheric observations indicated below. These observations are independent to the data sets used for the lower boundary condition, to ensure an objective evaluation.

For a detailed assessment of the performance of EMAC in general and how EMAC compares to observations (e.g. regarding temperature and ozone) we refer to Jöckel et al. (2016). This publication also includes an evaluation of transient simulations
10 regarding CH₄ in the Upper Troposphere and Lower Stratosphere (UTLS), using measurements of the Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container (CARIBIC) project, ~~which indicates a good.~~ That evaluation indicated a good CH₄ representation with relative differences of less than 5%.

In general, observed surface mixing ratios of CH₄ indicate a north-south gradient with larger CH₄ mixing ratios in the North-
15 ern Hemisphere (NH) mostly due to large wetland regions and anthropogenic sources on the northern continents. This north-south gradient is by design (nudging to ~~observation-based~~ zonal mean surface mixing ratios based on observations) apparent in ~~the simulation data as well our performed simulation.~~ The simulated CH₄ gradient of the ~~model REF simulation~~ is compared to observations during from a ship cruise of the research vessel *Polarstern* (~~Klappenbach et al., 2015, see Supplement Fig. S1~~)
20 (Klappenbach et al., 2015, see Supplement Fig. S1). The simulation results reproduce the observed north-south gradient of the ship cruise qualitatively well, although an offset of about 0.055 ppmv exists. Note that the observations ~~in~~ on the *Polarstern* were conducted in 2014, while the simulation represents 2010 conditions. Global CH₄ surface mixing ratios have risen between 2010 and 2014 by about 0.030 ppmv, which explains ~~a large part~~ some of the offset.

Additionally, the average vertical CH₄ profile of the REF simulation is evaluated using balloon borne measurements from
25 Röckmann et al. (2011). The CH₄ mixing ratio in the troposphere is approximately constant as a result of well-mixed tropospheric conditions. Above the tropopause the mixing ratio of CH₄ decreases with altitude. This vertical gradient apparent in the balloon borne observations is reasonably reproduced in REF (see Supplement Fig. S2).

Furthermore, a general comparison of ~~zonally averaged~~ the zonal mean of CH₄ mixing ratio from REF above the tropopause
~~of REF~~ is done with observations from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) instrument mounted on the ENVISAT satellite (Fischer et al., 2008). The zonally averaged CH₄ MIPAS climatology (2002–2012) ~~in~~ from
Plieninger (2017) corresponds qualitatively and quantitatively to our simulation results in Figure 1a.

30 ~~Annual zonal mean of absolute CH₄ mixing ratios (in parts per billion volume (ppbv)) of reference (REF) (a), S1 (b) and S2 (c). Note the different color scales.~~

~~Our~~ The REF simulation represents the observed CH₄ mixing ratios in the stratosphere and mesosphere and also shows the apparent double bulge in the upper stratosphere with slightly higher values in the NH.

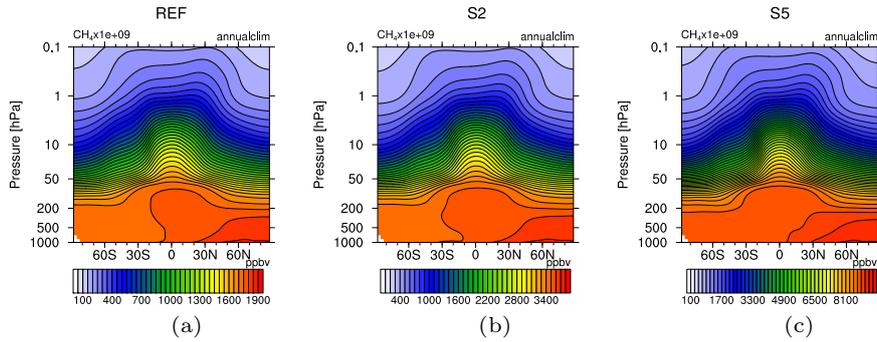


Figure 1. Annual zonal mean of absolute CH₄ mixing ratios (in ppbv) of reference (REF) (a), S2 (b) and S5 (c). Note the different color scales.

Overall, the agreement of the reference simulation results with observations ~~turned out to be sufficient~~ is suitable for the purpose of our intended study. In the next section we compare the reference simulation with the two sensitivity simulations, to present the general impact of strongly enhanced CH₄ concentrations.

3.2 Impact of twofold and fivefold increased CH₄ concentrations

5 In this sub-section we investigate the impact of twofold and fivefold surface CH₄ mixing ratio on the chemical composition of the atmosphere and analyze the new chemical equilibrium after a sufficient spin up.

Since SST and SIC are prescribed, ~~any possible~~ the larger part of the feedback on tropospheric temperature is largely suppressed in the present simulations. Therefore, in this study we are only considering rapidly evolving chemical feedback effects including respective radiative adjustments and temperature adjustments in the stratosphere.

10 The oxidation capacity of the atmosphere is often measured in terms of the CH₄ lifetime (Karlsdóttir and Isaksen, 2000; Dentener et al., 2003; Naik et al., 2013; Voulgarakis et al., 2013). In this study we calculate the tropospheric CH₄ lifetime according to Jöckel et al. (2006) as

$$\tau_{CH_4} = \frac{\sum_{b \in B} M_{CH_4}}{\sum_{b \in B} k_{CH_4+OH}(T) \cdot c_{air}(T, p, q) \cdot OH \cdot M_{CH_4}}, \quad (1)$$

with M_{CH_4} being the mass of methane in kg , T the temperature, p the pressure and q the specific humidity, all depending on time and the specific box $b \in B$, with B being the set of all considered grid boxes, e.g. all boxes which lie below the tropopause. $k_{CH_4+OH}(T)$ is the reaction coefficient of the reaction $CH_4 + OH \rightarrow$ products in [$cm^3 s^{-1}$]. $c_{air}(T, p, q)$ is the concentration of air in ~~molecules (molec.)~~ cm^{-3} [$molec. cm^{-3}$] and OH the mole fraction of OH in one mole of the chemical tracer per one mole of air ($mol mol^{-1}$)_{dry air}.

Our calculations yield a nearly linear increase of tropospheric CH₄ lifetime with respect to the CH₄ scaling in the sensitivity simulations (see Fig. 2). It is known that the tropospheric CH₄ lifetime is anti-correlated with OH concentration (Montzka et al., 2011). The strongly enhanced CH₄ mixing ratios reduce the atmospheric OH mixing ratio, which leads to a longer

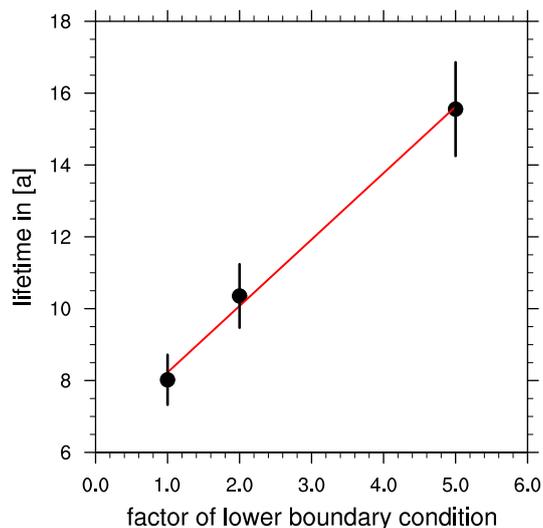


Figure 2. CH₄ lifetime calculated according to equation 1 versus the corresponding scaling factor applied to the reference lower boundary condition in the respective simulation: REF (1.0), [S1-S2](#) (2.0), [S2-S5](#) (5.0).

(tropospheric) CH₄ lifetime. It is not sure that the quasi-linear behavior will hold for even larger or smaller scaling factors, since the chemistry determining the OH abundance is highly non-linear. There are also not enough data points (sensitivity simulations) for a definite proposition on strictly linear dependence. Nevertheless, we assume that these results give evidence that in the troposphere CH₄ and OH for the given range are almost linearly anti-correlated. The simulations clearly show that increasing CH₄ emissions increase the residence time of CH₄ in the atmosphere and therefore its global warming potential (GWP).

Next, we investigate the impact of CH₄ concentration increases on changes in CH₄ depletion, thus analyzing possible non-linearities in the chemical cycles. For this [purpose](#) we compare both sensitivity simulation results with the reference CH₄ mixing ratio multiplied by 2 and 5, respectively, as is shown in Fig. 3. This approach makes it possible to see where CH₄ is impacted by non-linear processes, [hence i.e.](#), where the twofolding (fivefolding) at the surface does not lead to an equal increase in the upper layers in the steady state.

In both sensitivity simulations, the troposphere is largely controlled by the nudging at the lower boundary due to turbulent mixing. In this area the differences of the sensitivity simulations and the scaled reference simulation are near-zero (though slightly positive). Larger CH₄ mixing ratios reduce its most important sink reactant in the troposphere, namely OH, which leads to a reduction of the CH₄ depletion compared to the reference.

The n-fold methane concentrations at the surface do not generate a [likewise-corresponding](#) n-fold methane concentration throughout the upper stratosphere. In the sensitivity simulation with twofold (fivefold) CH₄, lower CH₄ values of about 5% (10%) are found between 50 and 1 hPa compared to the corresponding n-folded reference. Identically prescribed SST in all three model simulations determines to a large extent the forcing of atmospheric dynamics and also constrains to a large part the

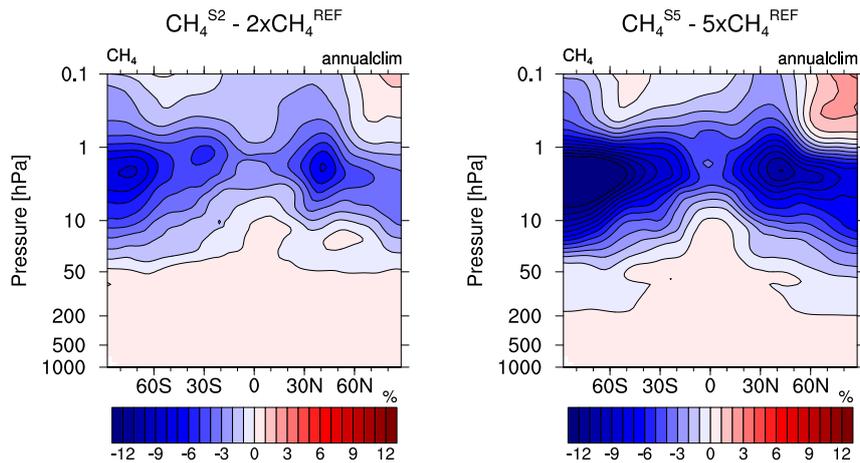


Figure 3. Difference between the annual zonal mean CH_4 of the simulations S1-S2 relative to the twofold and S2-S5 relative to the fivefold annual zonal mean of the reference in (%).

stratosphere (see Garny, 2010). Therefore, modified atmospheric circulation patterns are unlikely the cause of these changes in stratospheric CH_4 mixing ratios.

An explanation for the relatively strong relative depletion in CH_4 in the upper stratosphere could be the change of the reaction rates for the CH_4 decomposition via OH and chlorine (Cl), which are both temperature dependent. However, since the stratosphere cools in the sensitivity simulations (as will be discussed below) this also cannot explain the simulated reduced CH_4 content in the stratosphere. Nevertheless, the deviation of a linear signal in stratospheric CH_4 mixing ratio gives evidence that more CH_4 in the upper stratosphere is destroyed due to secondary feedbacks caused by changes in the chemical composition of the stratosphere (particular O_3 , SWV and OH) and will be discussed in the paragraphs below.

The OH concentration in the atmosphere is determined by its precursors, which are water vapour (H_2O) and O_3 , the photolysis rate of O_3 , as well as by its sinks, which are mostly CH_4 and carbon monoxide (CO). The decline of OH in the troposphere by 20 – 30% in the S1-S2 simulation compared to REF (see Fig. 4) is caused, as stated above, by the increased sink via CH_4 . In the stratosphere, however, OH increases by about 30% in the twofold CH_4 case (S1-S2) and by 60 – 80% in the fivefold CH_4 case (S2-S5 , see Fig. 4). The mixing ratio of OH increases especially in the upper stratosphere at higher latitudes and fits to the decline in CH_4 in the same regions (see Fig. 3). However, increases in CH_4 mixing ratios influence the abundance of OH precursors, namely H_2O and O_3 via direct and secondary chemical effects and thereby feedback on the production of the CH_4 associated sink OH.

Oxidation of CH_4 in the stratosphere produces additional H_2O and is therefore an important source for SWV (Hein et al., 2001; Rohs et al., 2006; Frank et al., 2018). The enhanced CH_4 mixing ratios in the stratosphere cause a steady increase of SWV with height in both sensitivity simulations as indicated by Fig. 5. In the twofold CH_4 case (S1-S2) the amount of H_2O is

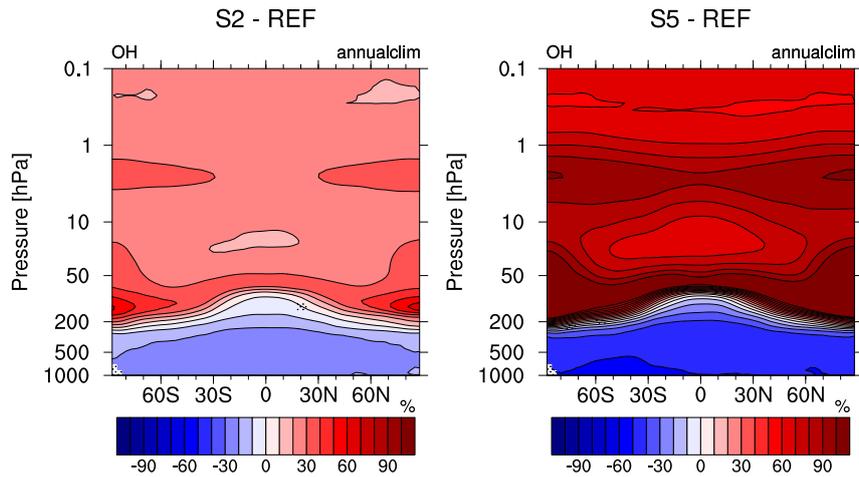


Figure 4. Comparison of the relative changes (%) in annual zonal mean OH mixing ratio of the sensitivity simulations [S1 and S2](#) and [S5](#) (two- and fivefold CH₄, respectively) compared to the reference REF. Non-stippled areas are significant on a 95% confidence level according to a two sided Welch's test.

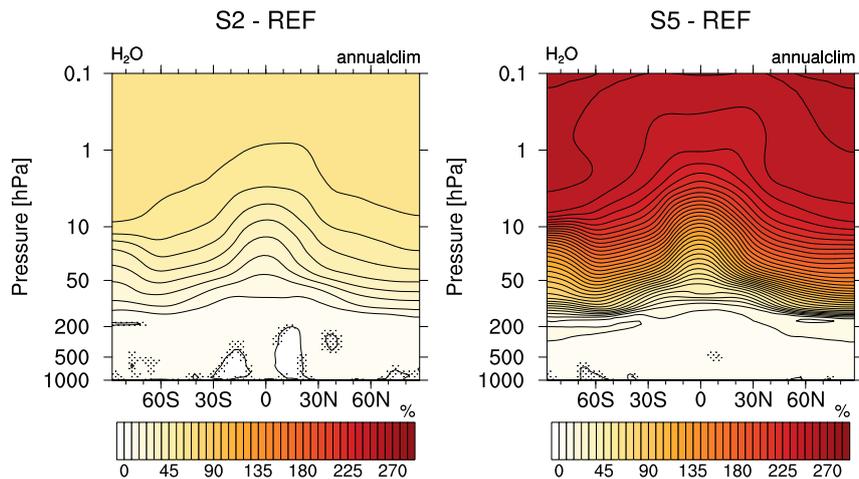


Figure 5. Comparison of the relative changes (%) in annual zonal mean H₂O mixing ratio of the sensitivity simulations [S1 and S2](#) and [S5](#) (two- and fivefold CH₄, respectively) compared to the reference REF. Non-stippled areas are significant on a 95% confidence level according to a two sided Welch's test.

enhanced by up to 50% in the middle and higher stratosphere, in the fivefold experiment ([S2S5](#)) the SWV increases by more than 250%. [The additional H₂O leads to increasing OH in the upper stratosphere and lower mesosphere \(Fig. 4\).](#)

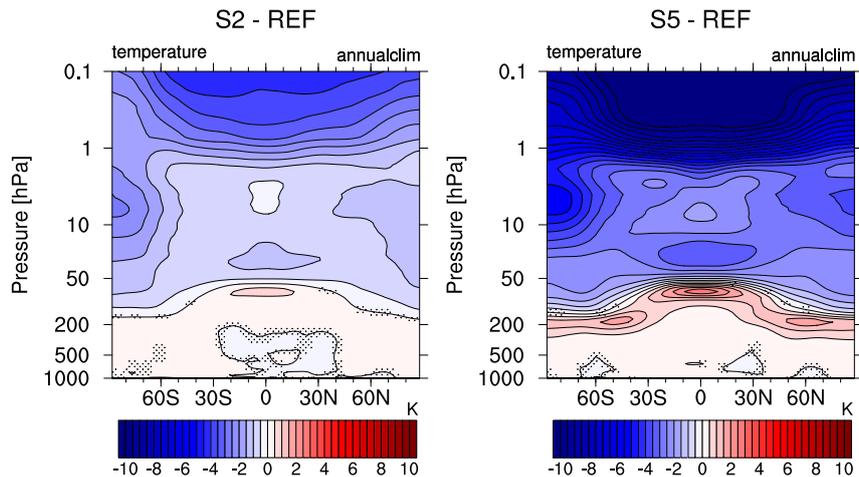


Figure 6. Comparison of the absolute changes (K) in annual zonal mean temperature of the sensitivity simulations [S1](#) and [S2](#) and [S5](#) (two- and fivefold CH_4 , respectively) compared to the reference REF. Non-stippled areas are significant on a 95% confidence level according to a two sided Welch's test.

The chemical changes indicated above influence the atmospheric temperature. However, since the SST is prescribed, the temperature response is largely suppressed in the troposphere. Confirmation [gives is given in](#) Fig. 6, where only a small change in tropospheric temperature is detected in both sensitivity simulations ([S1](#) & [S2](#) & [S5](#)). The stratosphere, however, shows larger changes in temperature. It can adjust to the perturbation since its temperature is mostly controlled by local radiative heating from trace gases and changing dynamics.

Around the tropopause there is a slight warming in the twofold CH_4 case ([S1S2](#)), which is reaching values of up to +3 K in the fivefold case ([S2S5](#), see Fig. 6). Elsewhere in the stratosphere, however, the higher abundance of CH_4 induces a stratospheric cooling in our simulations. The twofold CH_4 mixing ratios in [S1S2](#) lead to a stratospheric cooling of about ~~-1~~ ~~-2~~ ~~-1~~ to ~~-2~~ K and a mesospheric cooling of up to -5 K. The results of the simulation with fivefold CH_4 mixing ratios indicate a cooling of about -3 K in the stratosphere and more than -10 K in the mesosphere.

As will be ~~shown below~~ [discussed in detail later in this section](#), these temperature changes are ~~mainly~~ induced by the radiative ~~impact of CH_4 cooling from increasing CH_4 and H_2O in the stratosphere and mesosphere~~ ~~and by the changes in atmospheric chemistry due to enhanced chemically reactive CH_4~~ , but in particular by the ~~chemically induced O_3 decrease and its associated radiative effect~~. However, this is clearly a matter of two-way interaction, as the cooling also impacts on chemical reaction rates affecting OH and O_3 (see below).

~~Furthermore, the extreme~~ [As evident from Fig. 7, the strongly enhanced](#) CH_4 concentrations prescribed in our sensitivity simulations have an impact the vertical profile of O_3 . Concentrations of O_3 increase between 50 and 5 hPa (see Fig. 7) and

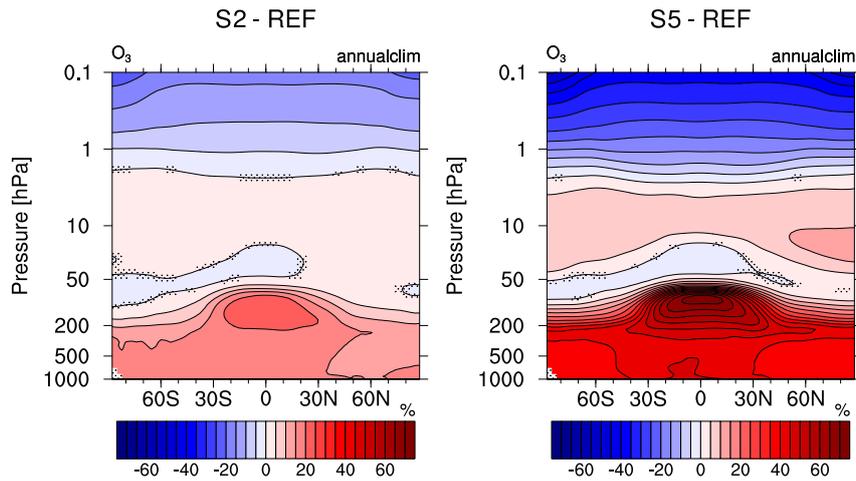


Figure 7. Comparison of the relative changes (%) in annual zonal mean O_3 mixing ratio of the sensitivity simulations ~~S1 and S2~~ and S5 (two- and fivefold CH_4 , respectively) compared to the reference REF. Non-stippled areas are significant on a 95% confidence level according to a two sided Welch's test.

decrease above. There is also a decrease between 50 hPa and 20 hPa in the tropics and between 100 hPa and 50 hPa at the southern pole.

The pattern of ozone reduction in the lowermost tropical stratosphere is typical for an enhanced tropical up-welling, which transports ozone depleted air from the upper troposphere to the lower stratosphere (Deckert and Dameris, 2008; Dietmüller et al., 2014). Although the main factor for such a strengthening, namely the SST, is prescribed, the increase in the GHG CH_4 alone can also lead to an enhanced tropical up-welling (Garny et al., 2011). A similar pattern in stratospheric ozone changes due to CH_4 increases (i.e. increase between 2000 and 2040–2049 according to the Intergovernmental Panel on Climate Change (IPCC) A1B greenhouse gas scenario) has been shown by Kirner et al. (2015). Nonetheless, we expect the impact on the tropospheric up-welling to intensify further in simulations, where the SSTs ~~can~~ are allowed to adjust to the CH_4 ~~induced~~ climate change radiative forcing.

In the middle stratosphere the O_3 production/depletion is influenced by the increased SWV and the corresponding stratospheric cooling, which restrains the reaction rates of O_3 depleting catalytic cycles (Portmann and Solomon, 2007; Braesicke et al., 2013). Furthermore, ~~Excited oxygen ($O(^1D)$)~~ $O(^1D)$ is depleted by increased abundances of H_2O , which reduce the sink of O_3 and eventually lead to increased O_3 abundances. Above 2 hPa, increases in OH facilitate O_3 destruction in the upper stratosphere and mesosphere (Kirner et al., 2015). Beyond that, there are inter-hemispheric differences in O_3 mixing ratios in the polar regions (increased O_3 at the polar middle stratosphere at about 30 hPa) that are an indication for a strengthening of the meridional transport towards the poles in the extreme case of $5 \times CH_4$ (see Supplement Fig. S4). In southern winter (Supplement Fig. S4 upper right) this transport is suppressed by the polar vortex and is forming a corona of increased O_3 mixing ratios

outside the polar vortex. The stratospheric cooling also leads to enhanced forming of polar stratospheric clouds (PSCs) during the southern hemispheric winter and therefore to enhanced O₃ depletion in the southern lower stratosphere (see Supplement Fig. S3 c and S4 c), (Dameris, 2010), as discussed in, e.g., Dameris (2010). The severity of the depletion does barely increase from S1 to S2 to S5, which may be explained by a saturation effect reached with respect of additional PSCs. The same effect can be noticed in the total O₃ column. Overall the total column of O₃ (see Supplement Fig. S5) increases due to the rise of CH₄ in the atmosphere, except in the Southern Hemisphere (SH) polar region, where S2 and S5 show about the same depletion in total O₃ column.

Overall, the twofold CH₄ mixing ratio with respect to 2010 is found to induce a radiative impact (RI) of about 0.69 Watt per square meter (W/m²) in the twofold case (S1S2) and a RI of about 1.79 W/m² in the fivefold case (S2S5) (see Table 1). The RI is calculated by the difference of the sum of long-wave and short-wave radiation at the top of the atmosphere between the reference and the respective sensitivity simulation. As the simulations are performed with prescribed SST, this net RI has the character of an effective radiative forcing (ERF), (Forster et al., 2016) as pointed out by Forster et al. (2016). It includes the RI of CH₄ itself, as well as rapid adjustments from physical and chemical processes. The chemical processes specifically include changes in SWV and O₃ and have been quantified by estimating their individual RIs (including stratospheric temperature adjustments) with the EMAC submodel RAD (Dietmüller et al., 2016) in a separate simulation resulting in estimates corresponding to the reference simulation REF* and the sensitivity simulations S1* and S2* (see and S5* (see Section 2 for a detailed explanation on the of this simulation).

The twofolding and fivefolding CH₄ enhancements in the sensitivity simulations S1 and S2 and S5 correspond to a net increase of surface CH₄ of 1800 ppbv and 7200 ppbv, respectively. For example, the increase of the surface CH₄ mixing ratio from 1750-722 ppbv (pre-industrial) to 1803 ppbv (2011) has led to a ERF of 0.48 W/m² ± 0.1 W/m² (IPCC, 2013, Chap. 8, Tab. 8.2) (IPCC, 2013, Chap. 8, Tab. 8.2), which corresponds to a net increase of surface CH₄ of about 1100 ppbv. The net increase of twofold CH₄ mixing ratios (S1S2) is larger than the increase since pre-industrial times. Thus, at first glance, the net RI calculated in this study seems consistent with the value in the IPCC AR5 and previous estimates from other models for a tripled CH₄ concentration (assumed +3534 ppbv) (Forster et al., 2016; Smith et al., 2018), which are for example for HadGEM2 1 W/m² and for CESM1 1.4 W/m².

However, those previous estimates do not account for contributions from O₃ and stratospheric H₂O changes (see also Smith et al. (2018)) (see also Smith et al., 2018). Only by looking at the direct RI simulations of individual species a clearer picture emerges (see Table 1). The individual RIs of the chemical variations is once more reasonable compared with similar calculations of the IPCC (Chapter 8, Figure 8.17) (Chap. 8, Fig. 8.17, derived from Shindell et al., 2009; Stevenson et al., 2013). We detect comparably low values for the RI of CH₄, a feature which has been reported before for the ECHAM5 radiation module (Lohmann et al., 2010). Part (but only part) of the underestimation can be attributed to a near zero shortwave absorption contribution that is known from radiation schemes used in other climate models (Etminan et al., 2016; Smith et al., 2018). As a residuum of the sum of chemically induced RI (Table 1) and the ERF, we estimate a pure physical RI of 0.03 W/m² (S1*S2*, 2xCH₄) and -0.03 W/m² (S2*S5*, 5xCH₄), respectively, i.e., small contributions that compare very well to the results of Smith et al. (2018).

Table 1. An estimation of separate RI in $[W/m^2]$ of the changes in the chemical species CH_4 , SWV and O_3 . Values are calculated using the RAD submodel (Dietmüller et al., 2016) in a separate simulation using 20 years climatologies of the individual species. Solely values of the totals are directly calculated from the presented simulations RI, [S1 and S2](#) and [S5](#).

Simulation	CH_4	SWV	O_3	chemical RI	physical RI ¹	total RI
S1* S2* (2x CH_4)	0.23	0.15	0.27	0.66	0.03	0.69
S2* S5* (5x CH_4)	0.51	0.55	0.76	1.82	-0.03	1.79

¹ total RI – chem. RI.

The separation of the individual RI in the main contributing species allows to further identify the individual contribution to temperature adjustments in the stratosphere (see [Supplement Fig. S5 and S6](#) [Fig. 8](#)). The stratosphere cools [by about \$-1\$ to \$-2\$ K](#) mostly due to SWV (up to -1.4 K). This cooling is amplified by the increased CH_4 and tropospheric O_3 , but reduced by [the radiative heating of](#) increased stratospheric O_3 in the [area-altitude domain](#) between roughly 20 hPa and 1 hPa. This [induces a dipole pattern in results in a quadrupole structure of](#) the total temperature change [pattern](#) (warming in the [lower-stratosphere](#) [troposphere with maximum around the tropopause level](#), cooling in the [lower/middle stratosphere](#), [warming-less cooling](#) in the upper stratosphere and [again stronger](#) cooling in the mesosphere, see [Fig. 8 and Supplement Fig. S5 and S6](#)). Only a minor contribution to the stratospheric temperature change is [contributed-made](#) by tropospheric H_2O , as to be expected. The difference between the pure RI of all trace gas changes (Figs. [S5a](#), [S6a](#) [8 a and Supplement Fig. S6 a](#)) and the net stratospheric temperature change (Fig. 6) is small, indicating a dominating role of radiative effects [over induced dynamical changes](#) in forcing the temperature response in [S1 and S2](#) and [S5](#).

4 Conclusions

The present study summarizes the quasi-instantaneous chemical adjustments of the atmosphere in response to a very strong increase in atmospheric CH_4 . We emphasize that the applied doubling of present day CH_4 surface mixing ratios is not unrealistic as it is even part of the RCP 8.5 scenario. Considering further feedbacks with still uncertain quantitative consequences, it is indeed possible that the presented changes in the atmospheric chemistry will be faced by upcoming generations.

For the range of CH_4 concentrations covered in this study we find that the CH_4 lifetime increases quasi-linearly with enhanced surface mixing ratios. This is ascribed to a strong reduction of OH, which is the main sink of CH_4 in the troposphere. We conclude that the strong reduction of OH will also influence other radiatively active, air quality relevant, and ozone depleting substances in the troposphere. The radical OH is the most important atmospheric detergent and its reduction will enhance the residence time of these substances as well as of CH_4 , and thereby increase the global radiative burden.

Additionally, induced by CH_4 oxidation, SWV will increase substantially [by up to 50 %, when \$CH_4\$ is doubled and more than 250 %, when \$CH_4\$ is increased by a factor of five](#). This leads to [stratospheric-cooling](#) [a stratospheric cooling of several degrees](#), which in turn influences stratospheric chemistry and (to a [less-smaller](#) degree) dynamics. In particular it will lead to an

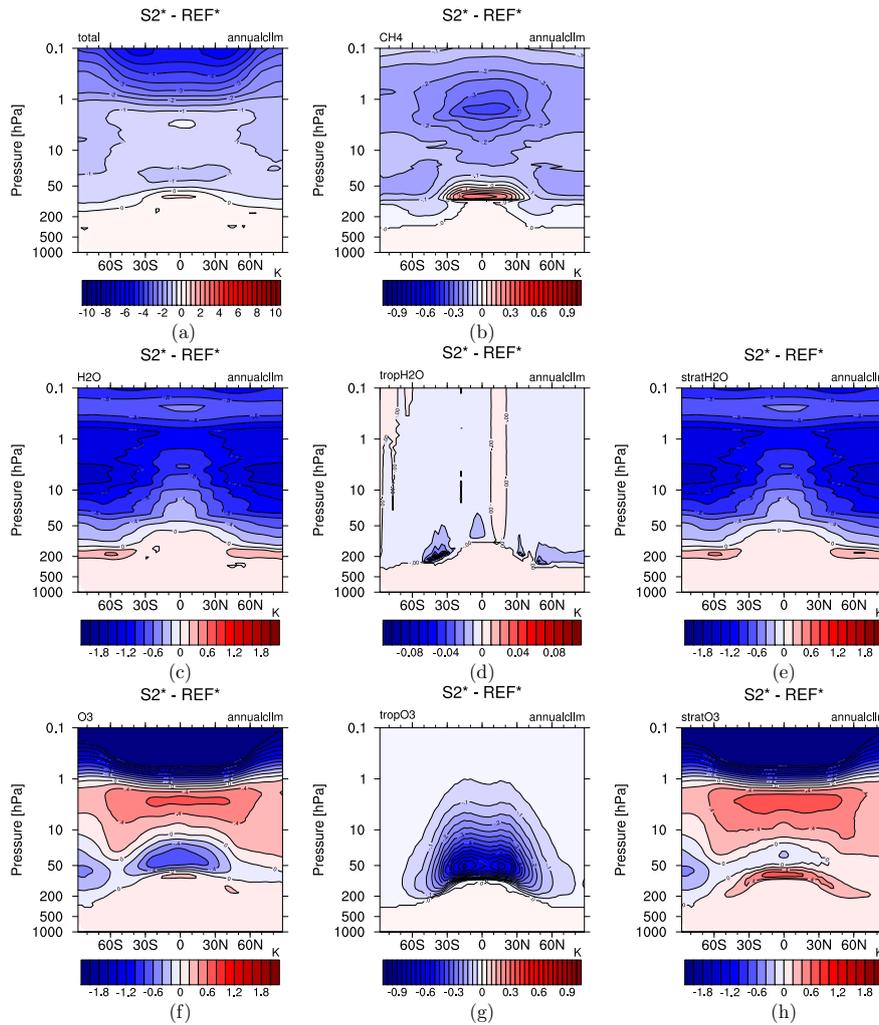


Figure 8. Stratospheric adjusted temperature based on chemical changes in simulation S2* ($2\times\text{CH}_4$) in (a) CH_4 , H_2O and O_3 combined, (b) CH_4 , (c) H_2O , (d) tropospheric H_2O only, (e) stratospheric H_2O only, (f) O_3 , (g) tropospheric O_3 only, (h) stratospheric O_3 only. Note the different color bars in panels (a), (b), (d), and (g).

increase in total O_3 column (not shown) nearly on see Supplement Fig. S5) nearly over the whole globe. Only in the antartic Antarctic spring it causes a strengthening of the ozone depletion. We also detect an O_3 reduction in the lowermost tropical stratosphere, typical for an enhanced tropical up-welling, which indicates small dynamical variations due to the strong increase of CH_4 , although more intense dynamical influences are suppressed by the predefined SST.

- 5 The rapid radiative adjustments of O_3 and SWV are both positive and thus increase the radiative forcing directly induced by CH_4 , consistent with Fig. 8.17 of IPCC (2013). However, the direct CH_4 radiative impact is considerably low biased in the simulations, apparently through a systematic error in the radiation module. This bias remains masked if only the effective

radiative forcing of the twofold and fivefold CH₄ simulations is considered. Individual radiative impact estimates also help interpreting the net stratospheric temperature change in the CH₄ increase simulations. It reveals that the main part of the overall temperature pattern is controlled by cooling from stratospheric H₂O.

5 Since the SSTs are prescribed in the present simulations, tropospheric temperatures and atmospheric dynamics do not represent the situation after adaption of the ocean. This also prohibits the calculation of climate sensitivity parameters. In a future study similar CCM simulations with a mixed layer ocean will be carried out and the contribution of feedbacks associated with SST changes will be investigated accordingly.

Code and data availability. The Modular Earth Submodel System (MESSy) is continuously developed and applied by a consortium of institutions. The usage of MESSy and access to the source code is licensed to all affiliates of institutions, which are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information can be found on the MESSy Consortium Web-site (<http://www.messy-interface.org>).

Author contributions. The simulations were set-up and carried out by P. J. and F. W.. F. T. analyzed the data and compiled the results. M. D. and F. W. structured and composed the manuscript. M. P. and F. W. contrived and carried out the radiative impact calculations. P. J. and M. P. contributed to the text.

15 *Competing interests.* The authors declare that they have no conflict of interest.

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5 MacKenzie, I. A., Nagashima, T., Plummer, D. A., Righi, M., Rumbold, S. T., Stevenson, D. S., Strode, S. A., Sudo, K., Szopa, S., and Zeng, G.: Analysis of present day and future OH and methane lifetime in the ACCMIP simulations, *Atmos. Chem. Phys.*, 13, 2563–2587, <https://doi.org/10.5194/acp-13-2563-2013>, 2013.
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Supplement of: Implication of extreme atmospheric methane concentration for chemistry-climate connections

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2	Difference in the annual cycle of O ₃	5
3	Climatological annual cycle of O ₃ total column change	7
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1 Evaluation of reference with observations

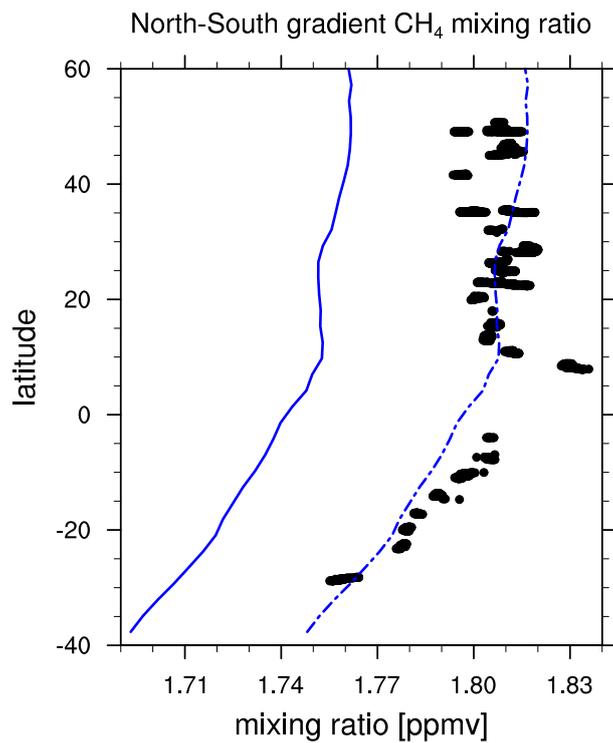


Figure S1. Comparison of observations on the research vessel *Polarstern* Klappenbach et al. (2015) (black) to annual zonal mean methane (CH₄) columns of the reference simulation (blue) in [parts per million volume (ppmv)]. The solid line represents the original column derived from the reference simulations and the dash dotted line are the columns moved by +0.055ppmv (see text for explanation).

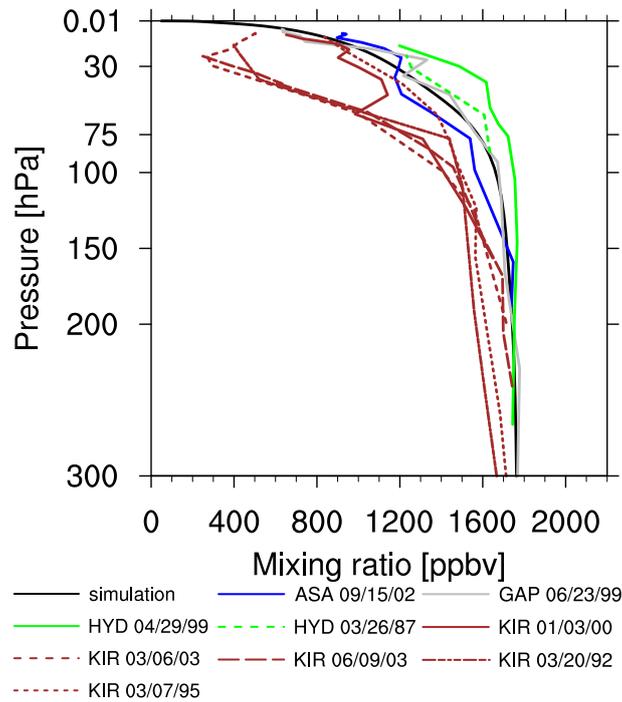


Figure S2. Comparison of vertical global mean CH₄ profile of simulation with balloon borne observations provided by Röckmann et al. (2011). The balloon launch sites are Hyderabad, India (HYD, 17.5° N, 78.60° E), Kiruna, Sweden (KIR, 67.9° N, 21.10° E), Aire sur l'Adour, France (ASA, 43.70° N, -0.30° E) and Gap, France (GAP, 44.44° N, 56.14 E) (see text for explanation).

2 Difference in the annual cycle of O₃

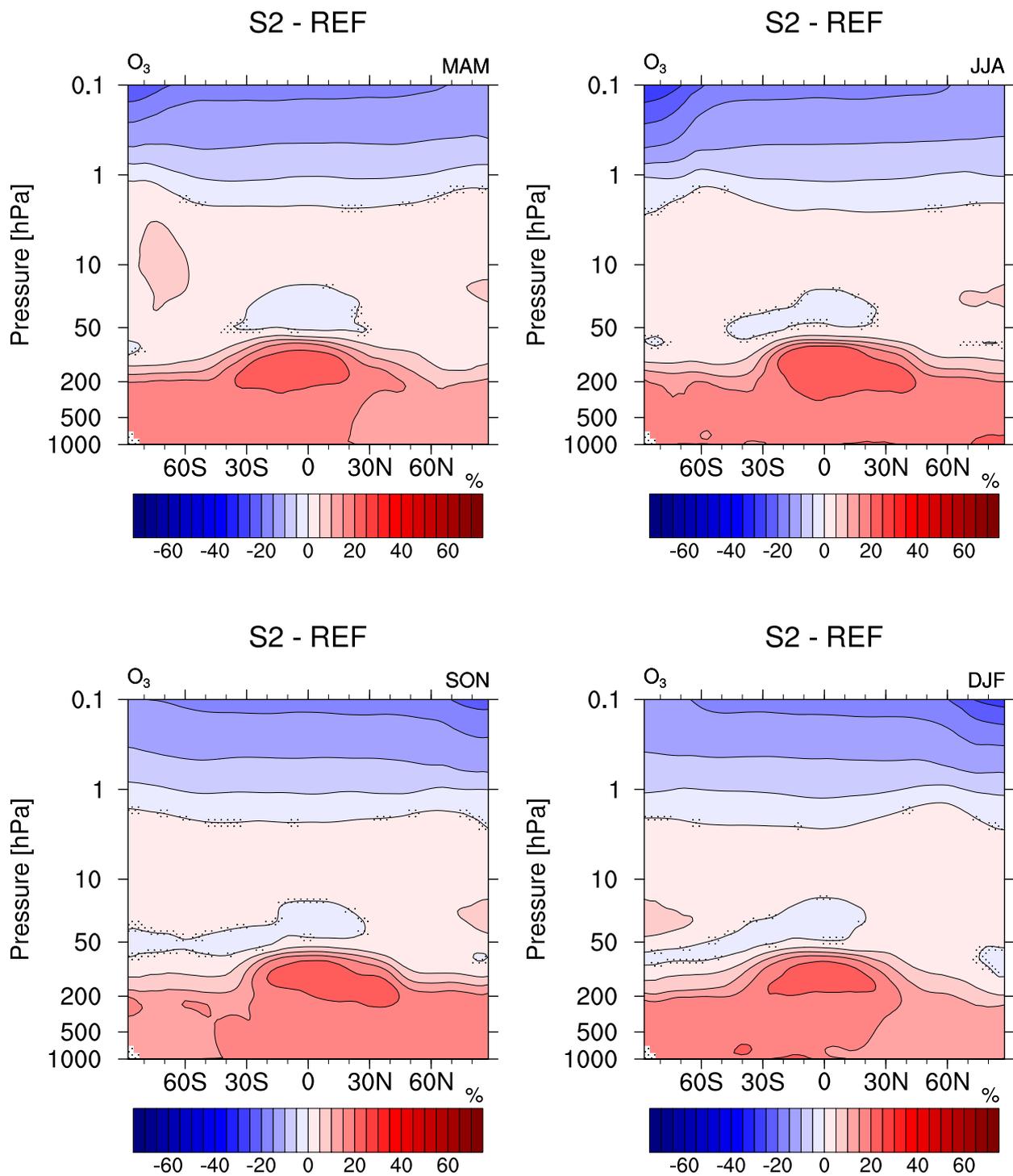


Figure S3. Seasonal differences in ozone (O₃) between ~~S1~~S2 and REF. Non-stippled areas are significant on a 95% confidence level according to a two sided Welch's test.

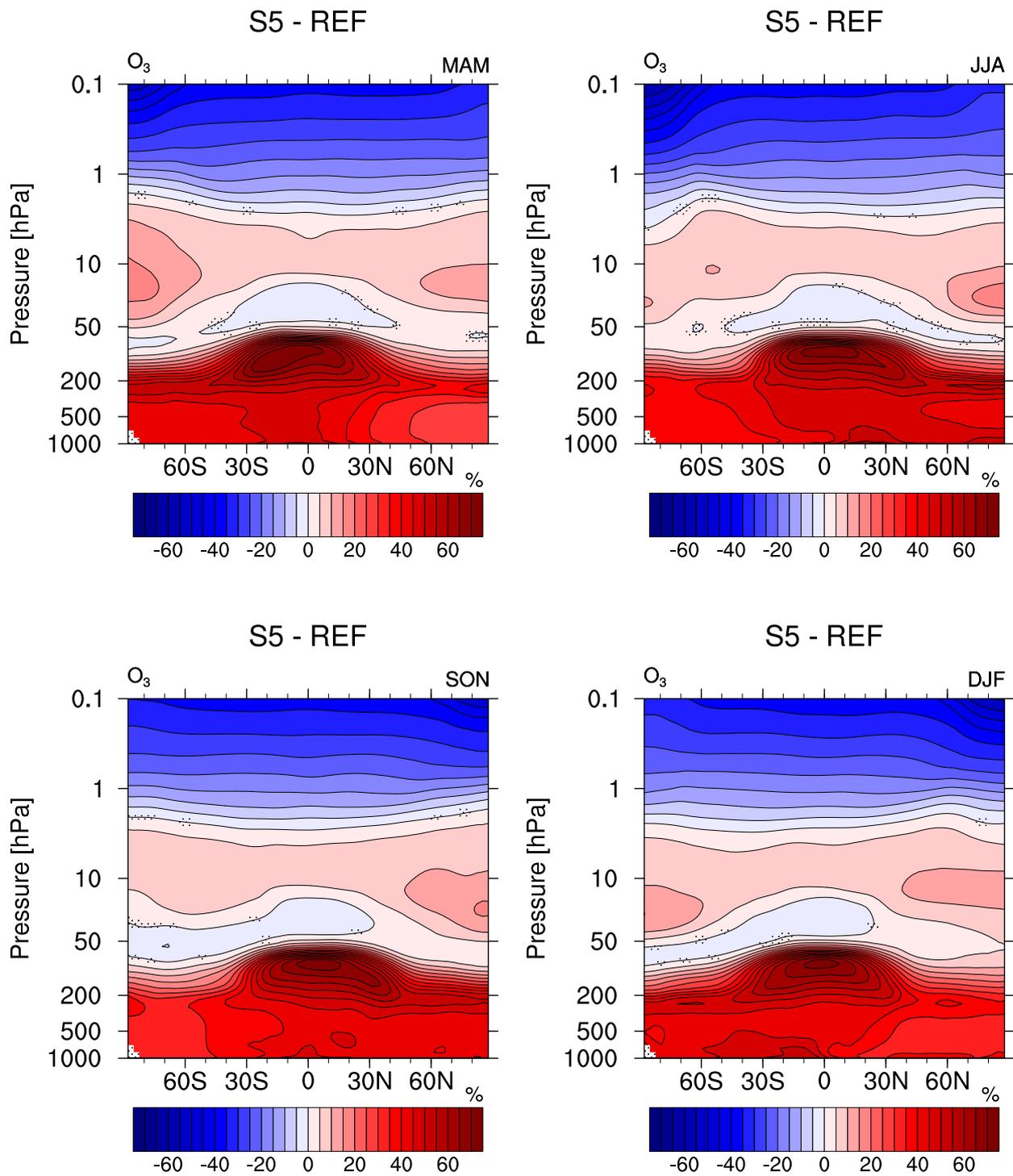


Figure S4. Seasonal differences in O₃ between [S2-S5](#) and REF. Non-stippled areas are significant on a 95% confidence level according to a two sided Welch's test.

3 Climatological annual cycle of O₃ total column change

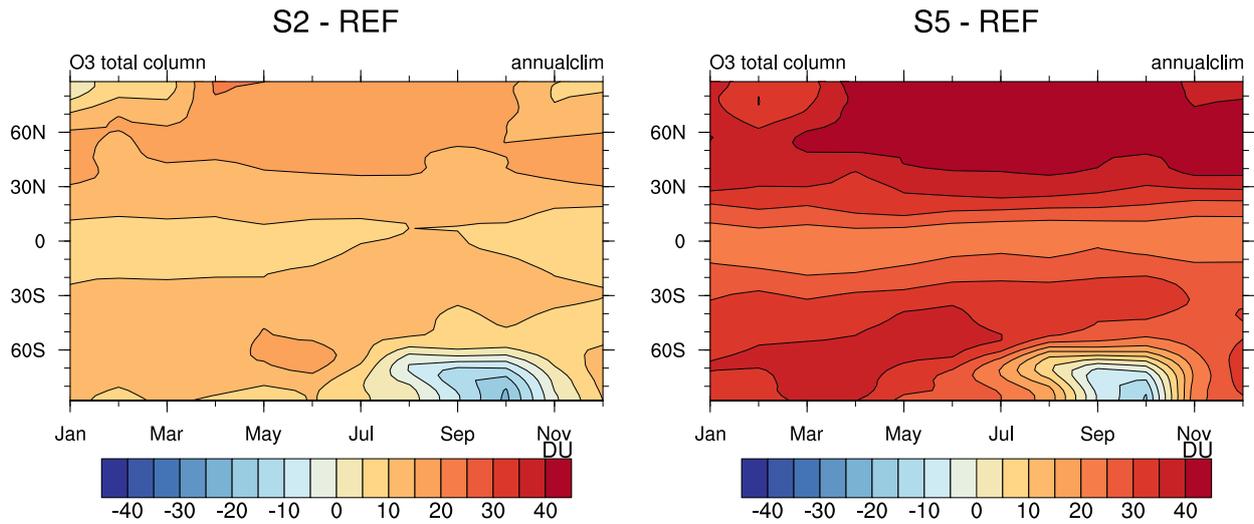


Figure S5. Climatological annual cycle of the total O₃ column change in Dobson Units (DU).

4 Adjusted temperature

— (a) — (b) — (c) — (d) — (e) — (f) — (g) — (h) Stratospheric adjusted temperature based on c

Figure S6. Stratospheric adjusted temperature based on chemical changes in simulation $S2^*-S5^*$ ($5xCH_4$) in (a) CH_4 , H_2O and O_3 together, (b) CH_4 , (c) H_2O , (d) tropospheric H_2O only, (e) stratospheric H_2O only, (f) O_3 , (g) tropospheric O_3 only, (h) stratospheric O_3 only. Note the different color bars in panels (a), (b), (d), and (g).

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Reply to referee # 1

April 10, 2019

Dear Christopher Smith,

thank you very much for such positive comments on our manuscript. We feel particularly honoured for your recommendation to AR6. In the following we reply to your comments point-by-point. The indicated pages of the answers relate to the discussion paper.

1 Specific comments

The authors prefer to use "radiative impact" rather than (effective) radiative forcing, and on page 11 discuss the fact that they are equivalent. Is there a good reason to use RI rather than ERF? Perhaps it is because the reference experiment is present-day rather than pre-industrial.

Indeed we chose to use "radiative impact" over ERF, since ERF is (e.g. in the IPCC) commonly used for the change in radiative forcing from pre-industrial. Since we compare to present-day we decided to use a different notation to avoid confusion.

It would make slightly more sense to me for the perturbation experiments to be called S2 and S5 (for 2x and 5x methane respectively). In this context Figure 3 could be understood without reading the paper.

Thank you, we decided to follow your suggestion, as we agree that this change makes the text more reader-friendly.

page 2, line 32: The way this currently reads implies that in Smith et al. we only used one model. It would be good to highlight that this was a 10-model intercomparison study.

Thank you for this note. It was not our intention to suggest this and are happy to change the text to stress that it is a model intercomparison.

Text added to the manuscript:

Old Smith et al. (2018) investigated the fast radiative feedbacks (adjustments) in a 3×methane (CH₄) simulation without considering the chemical feedback effects.

New Smith et al. (2018) investigated the fast radiative feedbacks (adjustments) in a model-intercomparison using simulations with 3×CH₄ mixing ratio without considering the chemical feedback effects.

page 5, line 10 (near the bottom): "any possible feedback on tropospheric temperature is largely suppressed...": you mean that climate feedbacks (from increasing global mean surface temperatures) are suppressed. There may be some tropospheric temperature change unrelated to surface temperature, due

to changes in tropospheric heating rates in the 2xCH4 and 5xCH4 experiments.

You are right that there are nevertheless some temperature changes. Actually, the feedback on tropospheric temperature is only suppressed concerning the sea surface temperature. There are indeed minor temperature changes over land and in the free atmosphere. In Figure 6, a small but mostly significant tropospheric temperature increase is apparent. However, fixing the SST leads to a stable temperature in the troposphere and the influence on dynamical processes is largely suppressed (e.g. Brewer-Dobson circulation (BDC)).

Text changed in the manuscript:

Old Since SST and SIC are prescribed, any possible feedback on tropospheric temperature is largely suppressed in the present simulations.

New Since SST and SIC are prescribed, the larger part of the feedback on tropospheric temperature is suppressed in the present simulations.

The low direct forcing from methane in ECHAM5, mentioned on page 12: is SW absorption of methane included in ECHAM5? This would account for some of the underestimate if not. See Etminan et al., 2016; <https://agupubs.onlinelibrary.wiley.com/doi/full/10.1002/2016GL071930>. Also, there is a difference between models that include SW absorption of methane in their radiation C2 schemes and those that do not in Smith et al. (see Figure S6 in the supplement to that paper).

We indeed found a shortwave contribution close to zero for the radiative impact directly contributed by CH₄, so this information has been added to the paper together with the reference indicated by the referee. Unfortunately, we could not identify the reason for the longwave part of the underestimation.

Text added to the manuscript:

New Part (but only part) of the underestimation can be attributed to a near zero shortwave absorption contribution that is known from radiation schemes used in other climate models (Etminan et al., 2016; Smith et al., 2018).

The separation of the methane ERF into the "direct" component and the chemistry-driven adjustments (table 1) I like. Stratospheric water vapour and O₃ include stratospheric temperature adjustments, so technically these would include a component of physical adjustment as well as the direct radiative effect of the water vapour and ozone (O₃). The authors are correct that these interactive effects are not present in the participating models in Smith et al.

That is an interesting comment. There are most likely secondary effects due to the changes in stratospheric water vapor (SWV) and O₃ as well as feedbacks on those components. The stratospheric adjusted temperature shown in den supplemental Figures S6 and S7 indicates the expected temperature changes of the chemical species, which are probably the cause of the variations in the meridional transport discussed on page 11. We, for example, also discussed changes in O₃ production and depletion due to stratospheric cooling.

For now, we separated in physical and chemical components based on the intuitive order that "chemical" refers to CH₄, SWV and O₃, including the associated stratospheric temperature adjustment. These are the most important radiatively active species that are directly affected by CH₄ through interactive chemistry, but their radiative impact cannot be quantified reasonably without the temperature adjustment (Hansen et al., 1997; Dessler et al., 2013).

2 Technical corrections

Abstract, line 1 would read better as: Methane (CH₄) is the second most important directly emitted greenhouse gas, the atmospheric concentration of which is influenced by human activities.

Page 3, line 1: "result" ⇒ "resulted"

page 5, line 14: "a large part" ⇒ "some"

Page 11, last line: "(see 2 for a detailed explanation..." not clear what "2" is a reference to here.

Page 14, line 14: "antarctic" ⇒ "Antarctic"

Thank you for these suggestions and corrections. We fully agree and changed the manuscript accordingly. The 2 in the fourth comment refers to the Section 2.

Reply to referee # 2

April 10, 2019

Dear referee,

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

1 Specific comments

I would change the title at least to “Impact of strongly increased methane concentrations for chemistry-climate connections” or even to “Impact of strongly increased methane on atmospheric chemistry and climate”. I don't like the wording extreme because in this case you think its an unrealistic scenario, but at least the 2xmethane (CH₄) scenario is absolutely possible until the end of this century.

Thank you for this comment. You are right that the impression may be wrong at least for the moderate sensitivity simulation S2. We changed the word “extreme” to “strongly increased”.

I don't know why you put the figures S1 and S2 into the supplement. I would recommend to put these both figures directly into the main part of the paper, because you discuss these figures in the main text and even refer in the captions to this text. The same is in my opinion true for Fig. S5 and maybe also Fig. S6, actually. These both figures are in my opinion absolutely not supplemental, but extremely interesting. Maybe you can put S5 directly into the paper, because you discuss the results of the panels of this figure directly in Sect. 3.2, and let S6 in the supplement.

While we are happy, of course, on this appraisal of our figures' relevance, we, however, feel that the length of text and number of figures would get out of balance, if we transferred all the figures to the main paper. All the same, we would like to keep the paper concise and focussed on the main messages. We left S1 and S2 in the supplement, since the evaluation of the reference simulations was a prerequisite for the study but does not give additional scientific information.

Concerning Figure S5 and S6 we agree that these are in fact not supplemental, so we put S5 into the main part of the paper, but kept S6 in the supplement as the patterns of stratospheric temperature adjustment remain largely the same for the two simulations, while the magnitude increases in line with the experimental design. With former Figure S5 now transferred to the main paper (as Figure 8), the text has been somewhat extended to give a full account of this figure's content.

In the last paragraph of Sect. 2 the additional simulations to calculate the single radiation impact of CH₄, ozone (O₃) and stratospheric water vapor (SWV) are explained. Maybe you can add here one or two sentences how the diagnostic for multiple radiation calls in submodel RAD works.

Thank you for this suggestion. We added the text below (and one more reference) to that paragraph.

Text added to the manuscript:

page 4 RAD performs multiple radiation calls with different input within one time step. Only the first call is used for providing the radiative heating feedback to the basemodel, while the other calls produce “perturbed” radiative fluxes and stratospheric temperature changes that are used diagnostically for calculating a stratospheric temperature adjusted RI (Stuber et al., 2001; Dietmüller et al., 2016). In our set-up the first call receives the reference mixing ratios of the chemical species, while the other calls receive climatological means derived from the sensitivity simulations, replacing either all component species combined or each of the three species individually.

You don't explain the higher OH mixing ratios in the lower/middle stratosphere (Fig. 4). I think these aren't a result of the additional water vapour (H_2O), but rather of your mentioned fact that the reaction $\text{CH}_4 + \text{OH}$ is temperature dependent (page 8, lines 10 to 15). In a cooler stratosphere this reaction is slower. Perhaps you can add one or two sentences in Sect. 3.2, but only if you agree.

We think that a full explanation requires additional in-depth analyses, i.e., additional simulations including specific OH budget diagnostics. The reason is that the OH abundance depends on many aspects. Those are, among others:

- The production of OH depends on the the abundance of water vapor and O1D. O1D, in turn, depends on the photolysis rate, which is influenced by the overlying ozone column, the distribution of underlying clouds (reflecting the radiation), and temperature.
- The loss of OH depends on the abundance of reaction partners like CH_4 , CO, and VOCs, and temperature, because the reaction rate coefficients are largely temperature dependent.

Thus, we think a straightforward explanation is currently not possible based on the simulation output we have.

In the conclusions you mention on page 13, line 13 the ozone columns. Please add this figure into the supplement or alternatively don't mention the ozone columns, but instead the stratospheric ozone as shown in Fig. 7.

You are right, that it may confuse the reader, where this result comes from. Since it was not discussed in detail, we add the respective figure on ozone column changes to the supplement and refer to it in the conclusions and in Section 3.2. The new figure is presented at the end of this reply.

Text added to the manuscript:

page 11 The same effect can be observed in the total O_3 column. Overall the total column of O_3 (see Supplement Fig. S5) increases due to the rise of CH_4 in the atmosphere, except in the Southern Hemisphere (SH) polar region, where S2 and S5 show about the same depletion in total O_3 column.

Maybe you can summarize in the conclusions your results of Sect. 3.2 with regard to the middle atmosphere and the relation of CH_4 , H_2O , OH, O_3 and temperature slightly more detailed as on page 13, lines 12 to 14.

We think this is a good idea. According to your suggestion we added some text (see below) in this paragraph.

Text added to the manuscript:

Old: Additionally, induced by CH₄ oxidation, SWV will increase substantially. This leads to stratospheric cooling, which in turn influences stratospheric chemistry and (to a less degree) dynamics. In particular it will lead to an increase in total O₃ column (see Supplement Fig. S5) nearly on the whole globe. Only in the Antarctic spring it causes a strengthening of the ozone depletion.

New: Additionally, induced by CH₄ oxidation, SWV will increase substantially by up to 50 %, when CH₄ is doubled and more than 250 %, when CH₄ is increased by a factor of five. This leads to a stratospheric cooling of several degrees, which in turn influences stratospheric chemistry and (to a smaller degree) dynamics. In particular it will lead to an increase in total O₃ column (see Supplement Fig. S5) nearly over the whole globe. Only in the Antarctic spring it causes a strengthening of the ozone depletion. We also detect an O₃ reduction in the lowermost tropical stratosphere, typical for an enhanced tropical up-welling, which indicates small dynamical variations due to the strong increase of CH₄, although more intense dynamical influences are suppressed by the predefined sea surface temperature (SST).

2 Technical corrections

Page 1, line 2: I would recommend to change a chemistry-climate model (CCM) to the chemistry-climate model EMAC

Page 1, line 6: pollutants ⇒ substances

Page 2, line 11: oceans , ⇒ oceans,

Page 2, line 21: by ⇒ due to

Page 3, line 2: to increase of CH₄ ⇒ caused by the increase of CH₄

Page 3, line 16: which is described in this paper ; will be deleted

Page 4, line 12: being ⇒ of

Page 4, line 13: being ⇒ of about

Page 4, line 20: online simulated emissions ⇒ emissions of other trace gases

Page 4, line 23: employed ⇒ used in submodel RAD

Page 4, line 25: and uses climatological 20 year means of the species of interest, namely CH₄ , O₃ and SWV ⇒ and uses the climatological 20 year means of the species of interest (namely CH₄ , O₃ and SWV) from the corresponding reference or sensitivity simulation (REF, S2 and S5).

Page 4, line 30: The reference simulation REF is set up to represent conditions comparable to the near-present atmospheric conditions in 2010. ⇒ The setup of the reference simulation represents the near-present atmospheric conditions of 2010.

Page 5, line 9: based ⇒ based on

Page 5, lines 9/10: in the simulation data as well ⇒ in all simulations or in our performed simulations

Page 5, line 10: The simulated gradient of the model ⇒ The simulated CH₄ gradient of the REF simulation

Page 5, line 10: during ⇒ from

Page 5, line 19: zonally averaged CH₄ mixing ratio above the tropopause of REF is done ⇒ the zonal mean of CH₄ from REF above the tropopause is done

Page 5, lines 22/23: no line break here

Page 5, line 23: Our simulation ⇒ Our REF simulation or The REF simulation

Page 5, line 25 (or line 4?): turned out to be sufficient for \Rightarrow is suitable for

Page 7, line 3: I think the unit for the reaction coefficient is $\text{cm}^3 \text{s}^{-1}$ instead of s^{-1} ; will be changed

Page 8, line 10: relatively strong relative depletion \Rightarrow relatively stronger depletion

Page 9, line 10: I would insert here a sentence like: The additional H_2O leads to increasing OH in the upper stratosphere and lower mesosphere (Fig. 4.).

Page 10, line 1: -1 - -2 K \Rightarrow -1 to -2 K

Page 10, line 18: Excited \Rightarrow excited

Page 11, line 11: fivefold case (S2). \Rightarrow fivefold case (S5) (see Table 1).

Page 11, line 18: (see 2) \Rightarrow (see Sect. 2)

Page 12, line 12: 1750 (pre-industrial) to 2011 has led \Rightarrow 650 ppbv (pre-industrial) to 1750 ppbv (2011) has led

Page 12, line 17: The stratosphere cools by about -1 - -2 K \Rightarrow The stratosphere additional cools in S1 by about -1 to -2 K

Thank you, we appreciate your awareness for these details and corrected the text accordingly.

Page 4, line 23: in a separate additional simulation \Rightarrow in separate additional simulations

Page 4, line 24: The simulation \Rightarrow Each simulation

Reply: Due to the feature of multiple radiation calls, we were able to perform the additional radiation calculation with only one simulation.

Page 10, line 5: I would insert here a sentence which describes the relation between the O_3 depletion in the upper stratosphere/lower mesosphere (Fig. 7) and the additional cooling.

We believe that this is closely related to your remark concerning temperature dependence of some reaction coefficients, e.g. with respect to OH (see above). We feel that with the available simulation output no more can be done than hinting at the two-way character of the interaction between some chemical species and temperature, and have modified the text as follows:

Text added to the manuscript:

Old: As will be shown below, these temperature changes are mainly induced by the radiative impact of CH_4 in the stratosphere and mesosphere and by the changes in atmospheric chemistry due to enhanced chemically reactive CH_4 .

Furthermore, the extreme ...

New: As will be discussed in detail later in this section, this temperature changes are induced by the radiative cooling from increasing CH_4 and H_2O in the stratosphere and mesosphere, but in particular by the chemically induced O_3 decrease and its associated radiative effect. However, this is clearly a matter of two-way interaction, as the cooling also impacts on chemical reaction rates affecting OH and ozone (see below).

As evident from Fig. 7, the extreme ...

Page 12, line 20: I dont see this pattern: warming in the lower stratosphere, cooling in the middle stratosphere, warming in the upper stratosphere and cooling in the mesosphere, maybe rather: warming in the troposphere, cooling in the lower/middle stratosphere, less cooling in the upper stratosphere and stronger cooling in the mesosphere

Thank you for this comment. The text has been corrected accordingly and also reformulated for a more precise description.

Text added to the manuscript:

Old: This induces a dipole pattern in the total temperature change (warming in the lower stratosphere, cooling in the middle stratosphere, warming in the upper stratosphere and cooling in the mesosphere, see Supplement Fig. S5 and S6).

New: This results in a quadrupole structure of the total temperature change pattern (warming in the troposphere with maximum around the tropopause level, cooling in the lower/middle stratosphere, less cooling in the upper stratosphere and again stronger cooling in the mesosphere, see Fig. 8 and supplement Fig. S6).

Page 13, lines 13/14: Here you discuss a new result: In particular it will lead to an increase in total O_3 column (not shown) nearly on the whole globe. Only in the antarctic spring it causes a strengthening of the ozone depletion. Please integrate this figure either into the paper or at least in the supplement. I also would recommend to discuss this already in Sect. 3.2 after the O_3 paragraph.

Reply: We included the figure in the supplement and added a note in Section 3.2.

3 New figure added to supplement (as Figure S5)

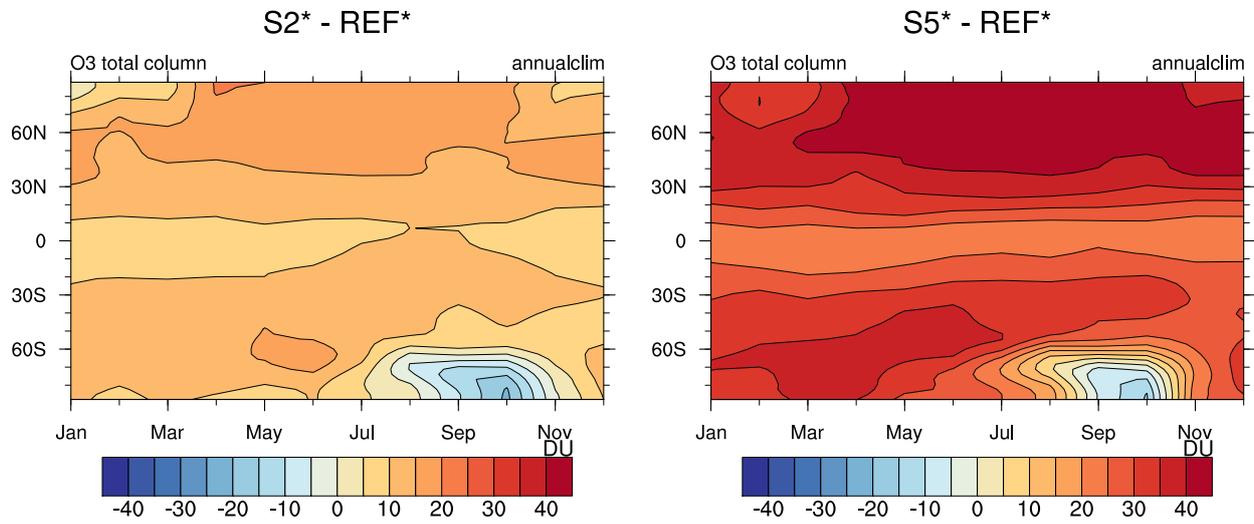


Figure 1: Climatological annual cycle of the total O₃ column change in Dobson Units (DU).