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 dont like the wording extreme because in this case you think its an unrealistic scenario, but at least the 2xmethane (CH_4) 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 CH_4 , ozone (O_3) 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 CH_4 +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 explanantion 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 CH_4 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 O_3 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 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 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 O_3 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 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 predefinded 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 CH_4 ⇒ caused by the increase of CH_4

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 CH_4 , O_3 and SWV ⇒ and uses the climatological 20 year means of the species of interest (namely CH_4 , O_3 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 CH_4 gradient of the REF simulation

Page 5, line 10: during ⇒ from

Page 5, line 19: zonally averaged CH_4 mixing ratio above the tropopause of REF is done ⇒ the zonal mean of CH_4 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 ⇒ is suitable for

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

Page 8, line 10: relatively strong relative depletion ⇒ 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 ⇒ -1 to -2 K

Page 10, line 18: Excited ⇒ excited

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

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

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

Page 12, line 17: The stratosphere cools by about -1 - -2 K ⇒ 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 ⇒in separate additional simulations

Page 4, line 24: The simulation ⇒ 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)

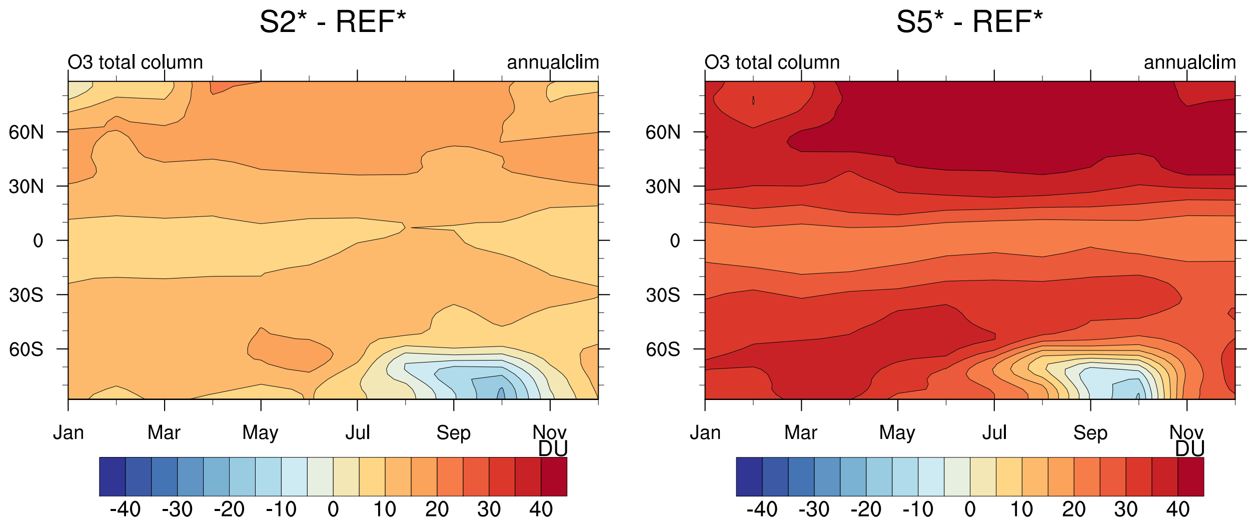

Figure 1: Climatological annual cycle of the total O_3 column change in Dobson Units (DU).