

Reply to referee # 2

October 26, 2020

Dear Referee # 2,

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

1 Main concern

The CH₄ model is being promoted as a useful alternative for studying methane, its isotopes, and stratospheric water vapour to the more complete and computationally expensive full chemistry scheme. As a result, I thought that the manuscript could be improved by including some verification of the CH₄ model compared with the (presumably) EMAC simulation from which the sink fields used in the CH₄ set up originated. How do they compare in terms of global mean methane concentration, methane lifetime, methane budget etc..? How does the modelled lifetime compare with other (fullcomplexity) models (e.g., Stevenson et al., <https://acp.copernicus.org/preprints/acp-2019-1219/>) and/or inversion studies? Benchmarking the CH₄ model performance against EMAC and placing its performance in the context of other models/studies would be a valuable addition to the manuscript.

Thank you for this suggestion. In fact the methane (CH₄) mixing ratio of the simplified CH₄ chemistry (CH₄) submodel and the Module Efficiently Calculating the Chemistry of the Atmosphere (MECCA) are by design identical, if the same CH₄ sources are applied and in CH₄ the same educts are prescribed as calculated in MECCA. In that case also the CH₄ lifetime is the same, since it is defined by the sinks. Therefore, from our point of view, a comparison of CH₄ simulated by the CH₄ submodel with that simulated by MECCA is not really meaningful. However, an important factor for the skill of matching the atmospheric CH₄ mixing ratio is the method of how CH₄ emissions are treated. In case of prescribing CH₄ at the lower boundary, the CH₄ mixing ratio in the troposphere represents the chosen condition. In the Earth System Chemistry integrated Modelling (ESCiMo) project (Jöckel et al., 2016) the zonally averaged marine boundary surface data provided by the National Oceanic and Atmospheric Administration/Earth System Research Laboratory (NOAA/ESRL) was used as the lower boundary condition and the simulations consequently reproduced the observations. Jöckel et al. (2016) also show that the CH₄ lifetime in the ECHAM/MESSy Atmospheric Chemistry (EMAC) model is with 8.0 ± 0.6 a rather low, but within the uncertainty range of similar studies. When using emission fluxes as lower boundary condition, reproducing (globally averaged) observations is much more challenging, as current emission inventories are subject to large uncertainties and the exact lifetime of CH₄ is still unknown. For example, we found that inventories derived by inverse modeling are quite dependent on the assumed hydroxyl radical (OH) and hence the CH₄ lifetime (Frank, 2018; Zhao et al., 2020).

The inclusion of optional region and age classes is a valuable addition to the CH₄ model and this information will be useful for estimating emission strengths. The authors cite the example of using a fixed-lag Kalman Filter, which performs an inverse optimization of the emission inventory by comparing simulated and observed mixing ratios of a trace gas. However, although the example provided of the time evolution of a single region class is a nice illustration, it is by no means evidence of the suitability of the CH₄ model as a tool for doing emission inventory optimization. In line with the comment above,

providing a more in-depth assessment of the model performance against observations would greatly strengthen the manuscript and provide evidence of its suitability as a tool for estimating emissions.

Yes, we also think that the estimation of emission strengths is a crucial part of modeling CH₄. The mentioned fixed-lag Kalman Filter and its application in a preproduction has been shown in Frank (2018). In the current publication we present the technical prearrangements, which are part of the CH₄ submodel. As stated before, the performance of simulation results against observations is strongly influenced by the used emission inventory, which is, when targeting emission estimation, not expected to be sufficient a priori. And an in-depth analysis of the application and performance of a full inversion using the concept of the Kalman Filter would be beyond scope of the current manuscript. This will be shown elsewhere in the peer reviewed literature, since work on this is still ongoing. Nevertheless, we include the reference to Frank (2018) in the revised manuscript.

Included in section 3.1:

The third option is implemented for usage by a fixed-lag Kalman filter for inverse optimization. With this option, one age class represents one month and at the end of one month all CH₄ of one age class moves to the next. This option is specifically implemented to be consistent with the Leapfrog time stepping (c.f. option (1)). **A preliminary application of the concept of using the age and emission classes for an inverse optimization using the fixed-lag Kalman Filter has been shown in Frank (2018).**

The authors, in the context of isotopes, also state that the simulation results compare well to observations. Can you include these comparisons with observations, for example?

Yes, we added the comparisons we referred to into the revised supplement.

2 Specific comments

Page 2, line 25 and Page 2, line 33: While methane as a source of stratospheric water vapour (SWV) is unequivocal, it is important to, at least, acknowledge the role of methane as an ozone precursor. From a climate forcing perspective, this indirect forcing is much larger than that from methane-driven changes in SWV but is neglected from the MESSy/CH₄ configuration presented here.

Thank you for this comment. Yes, this is indeed a drawback of the CH₄ submodel and we add a discussion of this into the revised manuscript. Although we must object that the indirect forcing from influencing ozone (O₃) is much larger than that from water vapour (H₂O). From a rapid adjustments perspective the indirect forcing of O₃ and H₂O is of about the same magnitude (Winterstein et al., 2019). Considering slow climate adjustments the effect of H₂O is three times larger (Stecher et al., 2020).

Included paragraph in section 3:

Furthermore, the setup with the CH₄ submodel also lacks any feedback on O₃. In the atmosphere, the O₃ chemistry is influenced by changes in the hydroxyl radical (OH) (reduced by CH₄), H₂O (produced by CH₄) and temperature (influence by radiative forcing of the abundant CH₄). The CH₄ submodel alters H₂O and with that influences the radiation budget and hence the temperature, however, there is no feedback on O₃ when the setup does not include any other chemical mechanism. In a setup where the CH₄ submodel is not used in parallel to MECCA, O₃ climatologies are usually prescribed for the radiation scheme.

Page 8, line 190: Can you be specific about what fraction of the age class is moved to the next class when this option is used?

Thank you for this question, since this seems not clear in the text. The fraction is defined by α . We included this note to the text.

$$M' = \alpha \cdot M, \quad (1)$$

with $\alpha = \frac{\Delta t}{\tilde{T}}$ and \tilde{T} being the user-defined time-span indicating the binning width of the age class. This option carries out a quasi-continuous update of the age classes, as it moves at every time step a fraction (**i.e. defined by α**) of the current age class to the next.

Page 8, line 194: Can you comment on how significant or large is this lack of conservation?

The described procedure is done to avoid the accumulation of small (numerical) errors, which mainly arise from small non-linearities of the large scale advection scheme. The magnitude therefore depends on the applied advection scheme, but is usually of the order of floating point precision. We added this explanation to the text as well.

Included in section 3.1:

In order to reduce numerical errors, the age and emission classes are continuously constrained (i.e., in each model time step) to sum up to the master tracer and are scaled appropriately, if the sum deviates. **The described procedure is done to avoid the accumulation of such numerical errors, which mainly arise from small non-linearities of the large scale advection scheme. The magnitude therefore depends on the applied advection scheme, but is usually of the order of floating point precision.**

Page 14, line 331: Here, you refer to the temperature bias in EMAC leading to a negative bias in water vapour. Is this temperature bias even evident in simulations with specific dynamics or when EMAC is free running?

The negative temperature bias in EMAC is strongest in free running set-ups. It is reduced but is still evident in simulations with specified dynamics as long as the wave-0 (or mean) of the temperature is not included in the nudging procedure, i.e. the temperature bias is not corrected. This is the usually applied procedure for specified dynamics. As soon as the mean temperature is included in the nudging, the bias nearly disappears. For more detailed information on the nudging procedure and the temperature bias, we refer to Jöckel et al. (2016).

Included in section 5.3:

This is associated with a too cold tropopause in EMAC, where a temperature bias of -2 to -6 K is detected in the upper troposphere, **as long as the mean temperature is excluded from the nudging procedure defining the specified dynamics setup** (Jöckel et al., 2016).

A complete listing of the CH₄ chemical mechanism, including isotopes, would make the description more complete rather than only showing the temperature dependent KIEs. This could be added to the Supplementary Material.

As also suggested by the other reviewer we include in the revision the CH₄ sink reactions (R1–R4) in section 1. We also include the corresponding reactions with isotopes deuterium (D) and carbon-13 (¹³C) in the revised supplement.

3 Technical comments

Page 4, line 102: Please change The here presented new submodel for simplified CH₄ chemistry (CH₄) and the auxiliary submodel TRacer SYNChronization (TRSYNC) are implemented based on this framework. to Presented here is a new.... **Agreed.**

Page 7, line 160: Change which can be specified by the user via namelist to which can be specified by the user via a namelist **Agreed.**

Page 7, line 162: Change denotes thereby to thereby denotes **Agreed.**

Page 7, line 167: Change identical to identically **Agreed.**

Figure 2: The onward arrow from tracer e02 a02 should possibly be dotted to be consistent with the one from tracer e01 a02 **Thank you, we changed that for consistency.**

Page 7, line 176: Change fixed-lag to a fixed time lag **Agreed.**

Page 8, line 184: The sentence The implementation of this option is not conform with a Leapfrog time stepping with Asselin-filter and might cause numerical oscillations with negative values Is very awkwardly written please rephrase. **We changed it to: This option is not consistent with a Leapfrog time stepping using an Asselin-filter and might cause numerical oscillations and negative values.**

Page 8, line 193: Again, awkward phrasing with the use of to be conform in the phrase This option is specifically implemented to be conform with the Leapfrog timestepping (c.f. option (1)). Please re-phrase. **We corrected *conform* by *consistent*.**

Page 9, line 220: Replace the here presented CH₄ submodel with the CH₄ submodel presented here **Agreed.**

Page 10, line 232: Replace H₂OISO doubles the hydrological cycle for the water isotopologues with H₂OISO models the hydrological cycle for the water isotopologues or H₂OISO represents the hydrological cycle for the water isotopologues **We changed *doubles* to *duplicates*. We want to point out that the hydrological cycle in H₂OISO is in addition to the cycle in ECHAM.**

Thank you for these suggestions and corrections. We changed the manuscript accordingly.

References

- Frank, F.: Atmospheric methane and its isotopic composition in a changing climate: A modelling study, Ph.D. thesis, Ludwigs Maximillian Universität München, 2018.
- Jöckel, P., Tost, H., Pozzer, A., Kunze, M., Kirner, O., Brenninkmeijer, C. A. M., Brinkop, S., Cai, D. S., Dyroff, C., Eckstein, J., Frank, F., Garny, H., Gottschaldt, K.-D., Graf, P., Grewe, V., Kerkweg, A., Kern, B., Matthes, S., Mertens, M., Meul, S., Neumaier, M., Nützel, M., Oberländer-Hayn, S., Ruhnke, R., Runde, T., Sander, R., Scharffe, D., and Zahn, A.: Earth System Chemistry integrated Modelling (ESCiMo) with the Modular Earth Submodel System (MESSy) version 2.51, *Geosci. Model Dev.*, 9, 1153–1200, doi: 10.5194/gmd-9-1153-2016, URL <http://www.geosci-model-dev.net/9/1153/2016/gmd-9-1153-2016.html>, 2016.
- Stecher, L., Winterstein, F., Dameris, M., Jöckel, P., Ponater, M., and Kunze, M.: Effects of Strongly Enhanced Atmospheric Methane Concentrations in a Fully Coupled Chemistry-Climate Model, *Atmospheric Chemistry and Physics Discussions*, 2020, 1–31, doi: 10.5194/acp-2020-519, URL <https://acp.copernicus.org/preprints/acp-2020-519/>, 2020.

Winterstein, F., Tanalski, F., Jöckel, P., Dameris, M., and Ponater, M.: Implication of strongly increased atmospheric methane concentrations for chemistry–climate connections, *Atmos. Chem. Phys.*, 19, 7151–7163, doi: 10.5194/acp-19-7151-2019, URL <https://www.atmos-chem-phys.net/19/7151/2019/>, 2019.

Zhao, Y., Saunio, M., Bousquet, P., Lin, X., Berchet, A., Hegglin, M. I., Canadell, J. G., Jackson, R. B., Deushi, M., Jöckel, P., Kinnison, D., Kirner, O., Strode, S., Tilmes, S., Dlugokencky, E. J., and Zheng, B.: On the role of trend and variability of hydroxyl radical (OH) in the global methane budget, *Atmospheric Chemistry and Physics Discussions*, 2020, 1–28, doi: 10.5194/acp-2020-308, URL <https://acp.copernicus.org/preprints/acp-2020-308/>, 2020.