Reply to Anonymous Referee #1

by Thomas Kleinen

We very much thank the reviewer for taking the time to review our manuscript. We especially appreciate the very valuable suggestions for improving the manuscript which appear to come from a perspective that is quite distinct from our modelling focus. I have included the reviewer's comments in blue colour, while our reply is in black.

Summary

Kleinen et al. modelled the transient evolution of atmospheric methane mole fraction during the last deglaciation with a fully coupled Earth System model (ESM). As mentioned in the Introduction section of this paper, so far studies of glacial-interglacial methane cycles have been limited to simple box model exercises or ESM runs on steady state time slices. This manuscript provides a valiant first attempt to bridge the knowledge gap and provide valuable insights into the transient dynamics of Earth's methane system. Some of the highlight findings from this manuscript include

- Modeled prediction and improved mechanistic understanding about which part of the Earth's wetland region is most responsible for CH4 emissions associated with changes in AMOC during D-O#1 (OD-Bolling transition) and Younger Dryas-Preboreal transition. This provides a testable hypothesis for future CH4 interpolar gradient measurements from ice cores once the issue with in situ production in dusty Greenland ice (Lee et al., 2020) is dealt with. Furthermore, this in itself is also a good benchmark on how good this model is in predicting future CH4 emission (Kleinen et al., 2021).
- Constraints on how the CH4 lifetime (and oxidative capacity of the atmosphere) responded to CH4 emissions and how it can feedback back into the atmospheric CH4 burden during periods of abrupt CH4 rises
- Further emphasis on the importance of tropical wetlands for the global CH4 cycle, in agreement with top-down results from ice cores (e.g., Rhodes et al., 2015; Bock et al., 2017) and modern/recent top-down results (Lunt et al., 2019; Shaw et al., 2022).

I find the manuscript to be very well-written and enjoyable to read. The model input and results are discussed in sufficient details. I would highly recommend this manuscript for publication after some minor revisions. Kleinen et al. is sitting on a trove of important first results, and I think some of the additional data they already have from this experiment (such as interpolar gradient, simulated CH4 mole fraction in the tropics, latitude binned CH4 sink(s?), further details below) can be presented in a way that is more accessible and useful for future ice core/paleo CH4 studies. Furthermore, the discussion section of this manuscript is a bit short, and I think after conducting these experiments, Kleinen et al. is in a unique position to provide us with further insights about the glacial-interglacial methane dynamics and the role of some of the smaller CH4 sources (either quantitatively or qualitatively, will be elaborated further below).

Thank you very much. We appreciate the fact that the reviewer seems to regard our results highly and will be happy to take his or her suggestions to improve our manuscript.

General comments

One of the peculiar things about Kleinen et al. simulations is the relatively low fire emissions (Figure 3c, lower than 10 Tg CH4/yr during the Holocene), which I think disagree with most paleodata we have. From CH4 stable isotopes Bock et al. (2017) calculated certain acceptable solutions for total geological + fire CH4 emissions during the Holocene (Fig. 2 of their paper). If geological emissions is small (as constrained by the 14CH4 data and assumed in this study), then fire emissions has to be fairly large (on the order of 22-55 Tg CH4/yr) (Dyonisius et al., 2020) to balance and produce such a heavy d13C-CH4 and dD-CH4 signature recorded in ice core.

Measurements of other trace gases in ice cores that are co-emitted by fires (mainly CO, ethane and acetylene) (Wang et al., 2010; Nicewonger et al., 2020) also predict Holocene fire emissions (say around ~1000 CE) that is higher/comparable to modern day fire emissions (that is anthropogenic + wildfire total emissions corresponding to ~40Tg CH_4 per year). On the other hand, the global charcoal index (which is a bit more qualitative than trace gases in ice core) predict Holocene fire emissions that is a bit lower than modern (e.g., Marlon et al., 2008). So I think it is fair to say that the paleofire proxies are a bit all over the place, as they don't even agree with one another. However, even the charcoal record does not predict late Holocene fire emissions so low that it is less than ~1/3 rd of total anthropogenic + wildfire emissions today.
Other than being a sizeable portion of the natural CH_4 budget, fire emissions are obviously important because NO_x, aerosol, CO, and NHMC (non methane hydrocarbon) emissions that affect the oxidative capacity of the atmosphere and CH_4 lifetime. I understand that the low fire emissions used in Kleinen et al. simulations are simply the result from the well-cited SPITFIRE model (Lasslop et al., 2014) they used and there is nothing wrong with that. It might be prohibitedly expensive to rerun the transient experiment or conduct sensitivity analysis with larger fire emissions, I'm not sure. If a simple sensitivity analysis is not possible, I think at least Kleinen et al. should address this disagreement and maybe qualitatively discuss how their results would've changed if Holocene fire emissions as predicted by paleofire proxies mentioned above (and by extension maybe also LGM?) were a bit higher. On a similar vein, the glacial-interglacial variability in CH_4 uptake by soil seems a bit low (only +- couple of Tg CH_4/yr over the whole deglaciation). Recent findings (for example Oh et al., 2020) suggest a much more dynamic soil uptake (at least in the high latitude) that can respond on decadal timescale to offset high arctic CH_4 emissions associated with modern warming. Again, I do not expect Kleinen et al. to rerun the transient experiment with more sensitive/variable soil uptake parameter, but it would be nice if this is maybe qualitatively addressed in the discussion section.

The reviewer touches upon two important points we apparently did not address sufficiently in our manuscript, the emissions from wildfires and the soil uptake.
The relatively low emissions from wildfires are, as the reviewer assumes, the result of applying the SPITFIRE model. The SPITFIRE model is – by ESM standards – a relatively sophisticated and well-published fire model. It may, however, not be ideal for this particular application. In the development of the model, the main focus lay on the description of fires in the modern period, which are mainly ignited by humans. The description of wildfires with ignition by natural factors, mainly lightning, is somewhat less sophisticated, however, and this may well be the reason for the relatively low fire emissions in glacial climate. In particular, the model assumes that lightning does not change in glacial climate – an assumption that is likely untrue. However we do not have a better description of lightning strike available, and the lightning model we use for the estimation of lightning NO_x does not show more lightning strikes in glacial than in preindustrial climate.
The reviewer is quite right, unfortunately it is prohibitively expensive to re-run the model experiment (it takes 3-4 months and several 10k€). Thus we will have to discuss this issue more carefully than we have done in the reviewed manuscript, which we will do in the revised version.

With regard to the uptake of methane by soils, we are aware of the Oh et al. results. In our model, however, the soil uptake of methane is not limited by the rate at which microbes are able to process the methane, but it is rather limited by the rate of diffusion of methane from the atmosphere into the surface soil. Thus we are not able to model this directly, but we will certainly discuss this in the revised version of our manuscript.

In Section 2.3 where Kleinen et al. discuss atmospheric methane sink, it is also not immediately clear whether they explicitly include CH_4 sink from reaction with chlorine (Allan et al., 2007). I presume that the chlorine sink is somewhere in there, considering ECHAM/MESSy model used in this study have been previously used to argue that the CH_4 sink from tropospheric Cl reaction at the present is low (Gromov et al., 2018). It would be nice if this is explicitly clarified in the manuscript. Furthermore, although the Cl sink might be low, it has important effect on CH4 stable isotopes. If Kleinen et al. have a proper quantitative attribution to the temporal evolution of each individual CH4 sink (e.g., relative contributions from tropospheric Cl sink vs. reaction with OH, and other CH4 sinks such as stratosphere destruction, O(1)D)) during the deglaciation, an additional figure showing these parameters and short discussion would greatly benefit future studies of CH4 mole fraction and isotopes in ice core. It would also be highly beneficial to see a similar figure to figure 11a (CH4 flux by latitude band) but for CH4 sink/lifetime if such parameter exists and saved in the model runs. Finally, it is also okay if it these sink attributions are not explicitly available, but that should also be mentioned/discussed if Kleinen et al. think the relative importance of one vs. other can potentially change during the deglaciation.

The methane sink formulation we are using is a highly parameterised simplification derived from the ECHAM/Messy model. As a result, we can only determine the combined effect of all the different sink terms, but not the individual contributions. We will extend the discussion of the methane sink to the different terms, though. With regard to the Figure of methane sink by latitude, we will try to include such a Figure in the revised manuscript. I believe all the necessary output should be available.

Interpretations of CH4 studies from ice cores are often limited to 2 or 3 box models due to the practical limitation that we only have measurements from Greenland and Antarctic ice cores. A peculiar feature in some time slice paleo CH4 reconstruction from models (e.g., Murray et al., 2014) is that the CH4 mole fraction in the tropics is higher than in CH4 mole fraction in both poles during the LGM. Unfortunately, we cannot reliably measure and reconstruct tropical CH4 mole fraction from tropical/low-latitude alpine ice cores due to in situ production from organics in alpine ice cores. If CH4 mole fraction in the tropics (say in 30S to 30N lat bin) is indeed higher than the northern hemisphere, then obviously the 2, 3 box model inversions commonly used in ice core studies (e.g., Chappellaz et al., 1997; Baumgartner et al., 2012) are inaccurate. The CH4 mole fraction in the tropics is a balance between CH4 emissions (which is highest in the tropics) and removal by OH (which is also highest in the tropics) – both of which can only be addressed with fully coupled CTM-ESM like the one used by Kleinen et al. It would greatly benefit the paleo-CH4 community, both experimentalists and modelers if Kleinen et al. can add to their figure 2 their reconstructed CH4 concentration over the tropics (despite the lack of data constraints) and provide some additional discussion about how reasonable they think their LGM simulation is (with focus on whether CH4 in the tropics is higher/lower than CH4 in Greenland during the LGM).

Thank you very much for this excellent suggestion – we didn’t think of it as it is never discussed in the literature, but it’s easily feasible and may be quite valuable.

I’m also interested in the fact that in the transient simulations, the first abrupt CH4 spike seen by Kleinen et al. in both base and MWM scenario coincidentally occurred at 16 ka, concurrent with Heinrich stadial 1 (HS1) event. It might not be immediately obvious at first, but there is also a small and abrupt CH4 spike at 16 ka associated with HS1 (Rhodes et al., 2015). It has been argued that this small HS1 CH4 increase is due to southward movement in ITCZ activating/intensifying emissions from southern hemisphere wetlands (Seltzer et al., 2017; Rhodes et al., 2015).
In page 10 line 223, Kleinen et al. mentioned that they unfortunately do not have this equivalent HS1 event in their simulation, at least in term of AMOC signature. They argued that the CH4 rise they see at 16ka is actually D-O#1/OD-BO happening too early in the model. But I think the 16 ka coincidence warrants further investigation and discussion. I’m especially interested if Kleinen et al. think that there are any “Heinrich-like” events recorded somewhere in either the base or MWP simulations – for example, maybe the weakened AMOC state at ~15.5-14ka in the MWM simulation (figure 1)?. There are other indicators for Heinrich stadial on top of AMOC strength

(like for example sea ice extent, Antarctic temperature, etc.) to check.
Heinrich events are particularly important in term of NH ice sheet evolution during the deglaciation. It would be very interesting to see additional discussion in this manuscript (doesn't have to be very long) on whether there is a Heinrich-like event in these simulations. If there is any, how this Heinrich like events affect the spatial distribution of CH4 emissions and if there is not, how the lack of 'Heinrich-like' event in the simulations affect the robustness of the interpretation (in term of say, sensitivity of CH4 emissions to AMOC changes driven by melting NH ice sheet).

Strictly speaking our current experimental setup is not capable of producing Heinrich events, as a Heinrich event in our current understanding is a dynamical interplay of ocean and ice-sheet dynamics. With prescribed ice sheets, it is not possible to obtain this, a dynamically coupled ice sheet model is required instead (see Ziemen et al. (2019, Clim. Past., https://doi.org/10.5194/cp-15-153-2019) for an example). We are currently working on this, but we are not there yet.
Having said that, we do obtain events resembling Heinrich events in our current experiment, the Younger Dryas transition in our MWM experiment being an example of one, and there may be other events showing some of the characteristics of H events in our deglaciation experiment – we will look at more diagnostics before submitting the revised manuscript, and will certainly extend the discussion of H events for the revision.

Finally, if possible, I would highly recommend the authors to add the relatively simple time series data, especially the ones produced by their simulations (time series data to plot figure 3a, 3b, 3c, 4c, 7a,7b, 11a,11b) in the supplementary section of this paper, or somewhere online and easily accessible.

Thank you. Yes, it is indeed planned to make the full output of the model available on the Earth System Grid. Processing of the output is currently ongoing but rather time-consuming as it involves very large amounts of data, so publication of the data set may happen at a slightly later date than publication of the paper. We had not yet considered also publishing the aggregated time-series output, but we will certainly rethink that as it makes the output accessible to a wider audience.

Line comments
Page 1 line 13: "four points in time". Not sure where the 4th abrupt CH4 transition is. I can only see 3 abrupt transitions during Termination 1, OD-BO CH4 rise, Allerod-YD CH4 drop, and YD-PB CH4 rise.
Oooops, thanks for pointing that out. Will correct.

Page 2 line 41: "We investigated methane emissions [...]5000 years apart from the LGM"; "apart" is not clear, I would change it to "before the LGM"
Thanks. We will correct that.

Page 3 line 69-70 "Methane emissions from wildfires ..." word by word repeated in page 4 line 100. Alter one of either sentence by a little bit.
Thanks. We will correct that.

Page 5 line 140: I might have missed it, but I think "PFT" is not defined anywhere in this paper
That is entirely possible. We will check and correct.

Page 8 figure 1: The purpose of this figure is to provide broad overview of the model parameters and metric in term of T1 deglaciation. In my opinion, plotting some actual data on top would greatly help readers evaluate these model metrics. For example, for global mean temperature (fig. 1a) I think it would be nice to see Shakun et al. (2012) temperature reconstruction on top. It would be nice if atmospheric CO2 is plotted on the second y-axis of fig. 1c. Finally, another important

metric that should be easily trackable in transient climate model simulation is mean ocean temperature (as integrator of various other metrics such as ice volume, sea level, AMOC strength etc). Mean ocean temperature can be plotted next to noble gas based mean ocean temperature reconstruction from ice cores (e.g., Baggenstos et al., 2019).
Thanks for the suggestion. We will incorporate this.

Page 9 fig2: As I mentioned above, CH_4 concentration over the tropics would be very beneficial to plot here despite the lack of data constraints. Furthermore, the CH_4 interpolar gradient (see Eq. 1 in Brook et al., 2000) is a commonly calculated analytical metric in ice core (Baumgartner et al., 2012; Sowers, 2010; Brook et al., 2000) and it would be great if it the CH_4 interpolar gradient from the simulation runs can be calculated and presented in this figure. Finally, the missing Greenland ice core data at ~16 – 14ka is fair; but between 10-2ka, since it is the Holocene (which is not as dusty as the LGM), Greenland mole fraction from ice core is only minimally affected by in situ production (Lee et al., 2020). As such, I would highly recommend the authors to plot composite Greenland CH_4 mole fraction by Beck et al. (2018).
Thanks for the suggestion. We will extend the Figure.

Page 22 line 389: I would disagree with the assumption that all oceanic CH_4 emission is geologic. There is a small amount of CH_4 emissions from the open ocean due to decomposition/cycling of organic matter (Weber et al., 2019). This would likely have small impact on the overall result of the paper, but needs to be acknowledged.
Thanks for that correction, our formulation was a little hasty. We will correct this.