

Replies to Referee #2 (bold italics).

General

The manuscript 'Using d13C-CH4 and dD-CH4 to constrain Arctic methane emissions' by Warwick and co-workers presents a study of atmospheric CH4 and its isotopes (d13C and dD) in the Arctic. Model simulations of all three components together with observations are used in a qualitative way to draw conclusions on the main arctic emission sources and their seasonal behavior. The analysis applies state-of-the-art modeling techniques and the methods and results are generally presented with a clear language and structure. The work clearly adds an important piece of information entangling the contributions of different methane sources and will probably help to further improve process models that are of paramount importance to understand future climate-emission feedbacks in the Arctic. I only have a few minor comments that can be addressed in the revised text but will, most likely, not require any major changes in the analysis.

We thank the referee for their very helpful comments and suggestions which have helped improve the manuscript.

Major comments

1. One possibly more important remark has to do with the chosen wetland emissions in the presented simulations. Why was the rather old Feng et al. dataset chosen as a reference? As can be seen in Figure 1 the LPJ-Bern emissions already follow the suggested delay in summer-time emissions. Related to this: What is the influence of the spatial distribution of the chosen wetland emissions. Could the suggested shift in emissions also stem from an erroneous distribution in space rather than in time? How different is the spatial distribution of LPJ-Bern as compared to Feng et al?

We followed the TRANSCOM model comparisons (Patra et al., 2011, Saito et al. 2013) in using the Fung et al. (1991) wetland dataset. Although there are now quite a few published wetland methane emission datasets available (see Fig. 1), uncertainties are large and it is not clear which dataset may be the most accurate or best performing.

We performed a simple analysis to investigate whether an erroneous emission distribution, rather than emission seasonality, could influence the modelled seasonality of methane mixing ratios etc. at the chosen measurement sites. Our model includes 4 wetland methane tagged tracers: north European, north American, north Asian and tropical. We tried varying the relative quantities of northern emissions (e.g. decreasing north American and/or north European while increasing Asian/Siberian emissions) and varying the relative quantities of northern vs. tropical emissions. However, the model results were very similar to either the INC_WET or NO_WET scenarios and we were unable to capture observed seasonalities in mixing ratios and/or isotopic ratios (see also our reply to referee #1's comment no. 3).

It is possible that inaccuracies in the emission distributions within our tagged regions could also impact modelled mixing ratio seasonalities. For example, if the model had a greater proportion of emissions >50°N located at very high latitudes, total emissions >50°N during May could be reduced (see new Fig. 2b). However, this would also have the unwanted impact of reducing emissions during October (Fig. 2b) and would alter the modelled latitudinal gradient (which is currently well captured, Fig 4). Therefore we believe it would be very difficult to correct the modelled atmospheric seasonal cycles by altering only wetland emission distributions and not seasonalities.

There are differences between the spatial distribution of emissions in the LPJ-Bern model and Fung et al., (1991), however the main emission hotspots are in broadly the same locations (West Siberia, Northern Europe, Hudson Bay lowlands). The differences in spatial distribution do not appear to be the cause of the different summed 50-90°N seasonalities between the 2 datasets, as the delayed seasonal cycle in the LPJ_Bern dataset relative to the Fung dataset is a consistent feature across all high latitude northern locations. In the Fung dataset, summed zonal mean September emissions are lower than corresponding emissions in the peak emission months of June, July and August, across all latitudes >50N (see Fig. 2). However, In the LPJ-Bern dataset, zonally summed June emissions are lower than corresponding emissions in the peak emission months of July, August and September, across all latitudes >50°N (not shown). We have added this information to the manuscript at the end of Section 6.2.2 (P10, I21-29).

Minor comments

2. P1, I18/19: Clarify if by inventories you are referring to purely anthropogenic emissions here.

This is now clarified in the text to read ‘anthropogenic or wetland emission inventories’. Whether anthropogenic or wetland emissions are implicated depends upon the seasonality and isotopic fractionation of the ESAS source.

3. P1, I26: What is the status of the Nisbet et al. publication? If not yet published another reference is needed here.

The Nisbet paper has now been published in GBC. The manuscript and reference list has been updated.

4. P2, I33: ‘In this study’: Does this still refer to Berchet et al. or the current study?

This refers to Berchet study. The text has been changed to clarify this.

5. P3, I6/7: The given reference is rather old. Please give some newer references and a total amount of emissions here. See for example Kirschke et al. 2013 for some numbers and additional references.

A newer reference (EDGAR v4.2) has been added.

6. P3, I8: The additional stratospheric sink by Cl and O1D should be mentioned here as well, although it probably adds little to the seasonality.

The stratospheric Cl and O1D sink is now listed here.

7. P3, I22-24: You should mention the work by Rigby et al. here as well, who already ran a CH₄ isotope model (both d¹³C and dD) to evaluate the benefits of atmospheric isotopic observations: Rigby et al. 2012, JGR, VOL. 117, D12312, doi:10.1029/2011JD017384.

This study has been mentioned elsewhere in the manuscript, however we agree it would be appropriate to mention it here again and have added another reference to the Rigby study at this point.

8. P3, I30f: In figures 5, 8 and 10 more than these 4 sites are used for comparison. Please mention which other sites are used in the figures.

Further information regarding the additional measurement locations in figs 5, 8, and 10 has now been added to Section 2. Their locations are now shown in Figure 3.

9. P4, I16-18: For which period are these values given?

The period 2005 to 2009. This information has been added to the text.

10. P5, I2: Which functional relationship was actually used to calculate the OH reaction rate coefficient? Reference or equation.

Relevant references for the OH, Cl and O1D reaction rate coefficients have now been added to the manuscript in Section 4.

11. P5, I9f: To put the importance of these other reactions into perspective, could you give an average lifetime of CH₄ wrt to stratospheric and MBL reactions as well?

Lifetimes for the MBL and stratospheric reactions have now been included in Section 4.

12. P5, Table S2: Table S2 should be integrated in the main paper, since it is an essential information for the study. However, in the table it should be clarified which source is taken from which reference and how seasonality is considered.

Table S2 has been moved into the main paper (Table 1), and each source referenced.

13. P5, I24f: Which influence may the spin-up still have on the results. The spin-up basically leads to an initial state in which sources and sinks are in equilibrium. Is this adequate for the study period or could it be important to start from a background that is not in perfect equilibrium (due to steadily increasing emissions in reality).

In this study we have spun up the model using anthropogenic emission data from 2005, and compared to atmospheric observations from 2005 to 2009. During the period 2000-2007, methane mixing ratios remained approximately constant in the atmosphere (excluding seasonal variations) and the global growth rate was close to zero suggesting the methane budget was approximately in equilibrium.

Prior to the year 2000, and post 2007, atmospheric methane levels increased, indicating a disequilibrium in the methane budget. This disequilibrium is not represented in our scenarios. Due to the long lifetime of methane, it is possible that changes in emissions pre-2000 could influence atmospheric mixing ratios post-2005, however this would be more likely to impact inter-annual methane trends than the seasonal variations considered in this paper.

Due to uncertainties in interannual methane emission trends, that the period considered in the paper occurs towards the end of an apparent period of equilibrium in the methane budget, and that we are considering seasonal variations rather than year to year trends, we believe that a spin up using yearly constant methane emissions is justified in this case.

14. P6, I22: 'with the seasonal cycle'. Does this refer to the observed seasonal cycle?

Text has been changed to read 'observed seasonal cycle'.

15. P9, I2f: Fig. 1 should be mentioned here again, which shows the emission profiles of Fung and Fung delayed.

A reference to figure 1 has now been included.

16. P9, I6: 'forward': To me this is confusing. I would call it shifting the seasonal cycle backward.

We wrote 'one month forward' as April emissions have been moved to May, May to June etc.. In order to be less confusing we have removed the phrase 'forward in the year' and instead said 'delayed by one month'.

17. p10, l1-3: Berchet et al. 2016 clearly showed a strong seasonal cycle for emissions from ESAS, with a summertime peak in the order of what was suggested by Shakhova et al. 2012 for the whole year (see Fig. 5 in Berchet et al. 2016), i.e. 10-15 Tg/yr. In contrast, Berchet et al. suggest close to zero emissions in winter. How could this seasonality, that was not considered in the current model analysis, change the drawn conclusions concerning the impossibility to accommodate the ESAS flux as suggested by Shakhova et al.?

We believe this point is already partially covered by summary points (a) and (b) in Section 6.3. They outline that, to accommodate a large ESAS source, our model requires a reduction in either high latitude wetland emissions or high latitude anthropogenic emissions, depending on whether ESAS emissions are considered to be seasonal or aseasonal, and the value chosen for their $\delta^{13}\text{C}$ isotopic composition.

A strong summertime peak for the ESAS emissions would resemble the seasonality for high latitude northern wetlands, which are also predicted to peak in the summer. Therefore, including such a seasonal cycle for ESAS emissions would make it harder to distinguish between ESAS and high latitude wetland emissions in our model simulations, particularly if ESAS were assigned a very negative $\delta^{13}\text{C}$ isotopic signature ($\sim -70\text{‰}$), similar to high latitude wetlands.

We have added further information to the manuscript regarding the possible impacts of a seasonal ESAS source in Section 6.3 (P12, l7-9 and P12, l30 – P13).

18. Section 6.2: Please comment on the good agreement of the delayed temporal development used in your sensitivity run and that simulated by LPJ-Bern (as seen in Fig. 1). Does their model version already include possible CH₄ emissions after freezing of the top soil?

The WETCHIMP-WSL model intercomparison (Bohn et al. 2015) compared emissions from all the WETCHIMP models in the West Siberian region. The late peak in LPJ-Bern emissions was also identified in this study, and was found to be predominantly due to a late peak in wet mineral soil emission intensity, despite a very late peak in CH₄-producing area. We have added this information to the discussion in Section 6.2.2 (final paragraph).

19. P11, l21-23: This thought should be given some more discussion. Would it still be possible to accommodate a 50 % reduction in high-latitude anthropogenic emissions (as compared with the BASE run) within the range given by previous studies (e.g. Kirschke et al.).

The -50% value given was an error. Anthropogenic emissions >50N total 36 Tg/yr in our scenario. If hydrate emissions are increased by 12 Tg/yr (from 5 to 17 Tg/yr), a 12Tg/yr reduction in anthropogenic emissions would be equate to a 33% reduction. This has been corrected in the text.

A reduction of this magnitude would remain within the range of top-down and bottom studies studies presented in Kirschke et al. review paper, although be very close to the lower estimates given for the agri-waste and fossil sources. (Biomass burning only represents a small proportion of emissions at these latitudes.) This information has been added to the manuscript in Section 6.3 (P13,114-19).

20. Figure 2: Should be part of the supplement. It is not essential to the study, but 'only' demonstrates that the applied model seems to perform reasonably well in terms of OH degradation.

Figure 2 has been moved to supplementary information.

21. Figures 6 and 9: Why is there no observed dD for Ny-Alesund? If it was not observed there, it should be mentioned somewhere in the text.

dD was observed at Ny-Alesund. However after quality control, there was not sufficient data to be able to plot a seasonal cycle. This has now been explained in the caption for Fig. 6.

22. Adding a table summarizing all model runs (BASE and sensitivities) and their specific settings would be nice. All the sensitivities should be given an abbreviation/name (not done in all cases) in this table and in the text so that it is easier to quickly identify what the specifics of a certain run

A Table summarising all model runs has now been added to the manuscript (Table 2).

References

Patra, P. K., Houweling, S., Krol, M., Bousquet, P., Belikov, D., Bergmann, D., Bian, H., Cameron-Smith, P., Chipperfield, M. P., Corbin, K., Fortems-Cheiney, A., Fraser, A., Gloor, E., Hess, P., Ito, A., Kawa, S. R., Law, R. M., Loh, Z., Maksyutov, S., Meng, L., Palmer, P. I., Prinn, R. G., Rigby, M., Saito, R., and Wilson, C.: TransCom model simulations of CH₄ and related species: linking transport, surface flux and chemical loss with CH₄ variability in the troposphere and lower stratosphere, Atmos. Chem. Phys., 11, 12813-12837, doi:10.5194/acp-11-12813-2011, 2011.

Saito, R., et al., TransCom model simulations of methane: Comparison of vertical profiles with aircraft measurements, J. Geophys. Res., 118, 3891-3904, doi:10.1002/jgrd.50380, 2013.