Reply to Referee #1 (bold italics).

Overview:

The manuscript "Using δ1 C-CH4 and δD-CH4 to constrain Arctic methane emissions" by Warwick et al. describes the results of a modelling study of Arctic wetland and hydrate emissions, in which the simulated concentrations of CH4, along with the associated δ13C-CH4 and δD-CH4 ratios, are compared to observations made at a number of high-latitude Northern Hemisphere measurement sites. The latitudinal gradient of these isotopologues is also assessed in comparison to observations. Finally, in an attempt to improve our current understanding of methane emissions from the Arctic, the effect of changes made to the wetland and hydrates emission inventories in the region is investigated.

Overall the manuscript is very well written, with few technical corrections necessary. The figures are generally quite clear and well chosen, although some small alterations are necessary for a couple of them. The methods used in this manuscript provide a neat way of assessing the accuracy of some of the current methane inventories used in atmospheric models, and the improvement in the comparisons with observations after the seasonal cycle of the wetland emissions is altered is striking. Using the three isotope ratios of methane as a 'triple check' on the seasonal cycle of the emissions works well and provides extra clues as to the timing and magnitude of emissions in the region. Finally the examination of the magnitude of hydrate emissions in the Arctic, whilst brief, does indicate that some recent estimates of emissions from this source may be too large.

My main reservation is that the conclusions drawn are dependent on a single (fairly old) wetland inventory, and there is no discussion on the impact that this fact might have on results. Is the relative geographical distribution of wetland emissions important for your conclusions to be substantiated? See general comments for more details.

I recommend this manuscript for publication after these revisions have been carried out.

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

Our response to the comment on our use of the Fung et al., 1991 emission inventory is included in the replies to the general comments below.

Comments:

1. Page 1, lines 20-29: These paragraphs could use some extra references. You describe the recent changes in the methane growth rate without referring to any sources for this information ('2007... rapid methane increase','growth was strongest in the tropics', etc.), and there is also no reference for the assertion

that fossil fuel changes could play a role in the global growth rate or that Arctic emissions are poorly quantified.

Several references have now been added to this paragraph (see P1, l20-28), including those for the role of fossil fuel changes. The poor quantification of Arctic emissions is discussed (and referenced) by source type in the following paragraphs of the introduction.

2. Page 5, lines 5-9: In this work, you have used observations averaged over 2005-2009 and model meteorology for 2009 only, but in order to show that the OH fields used in the study are to some extent accurate, you show comparisons with MCF concentrations at one site for the year 2011. To be consistent with the meteorology used for the later figures, can you show 2009 concentrations here? MCF measurements should also be available at Alert, Canada. Does the model also capture the seasonal cycle that far north?

   The figure has been changed to include observed MCF mixing ratios for 2009. As for the 2011 data, there is good agreement between modelled and observed seasonal cycles. As requested by Referee #2, we have now moved this figure into the supplementary information (Fig. S1).

   MCF measurements are also available at Alert, however this data contains significantly more noise and lacks a smooth seasonal cycle. The model predicts a smooth MCF seasonal cycle at Alert, similar to that modelled at Barrow, but with a slightly smaller amplitude. The modelled MCF seasonal cycle at Alert compares well to the observed Alert seasonal cycle, minus the noise (i.e. capturing the approx. timing of peak/minimum mixing ratios). However, due to the additional noise in the Alert observations we have chosen to present only the Barrow data.

3. Page 5, line 16: My main reservation with this study is related to the emissions inventories used. The Fung wetland inventory is now 25 years old, and whilst it generally does a good job, I think it is worth at least discussing the idea that the distribution of emissions in this inventory may not be correct. Since all of your observation sites are located in the US and Europe, are the observed seasonal cycles sensitive to the significant emissions from Siberia, or is the cycle only of the local emissions important?

   In our model simulations, high latitude northern wetland emissions from Asia, Europe and America were coloured or 'tagged' separately. Our results show us that modelled seasonal cycles at presented measurement sites are predominantly influenced by high northern latitude wetland emissions from America and/or Europe, with little sensitivity to Siberian emissions. We found that altering the Fung

emission distribution in a simple way via varying the relative emission strengths associated with these regional tracers offered no improvement with the comparison to observations. Increasing the European and/or American contributions while reducing the Asian (Siberian) contribution gave a result similar to INC_WET, and vice versa to NO_WET. This information has been added to relevant sections of the manuscript (5.2.1, 6.2.1 and 7).

In the introduction we discuss the large uncertainties associated with high northern latitude wetland emissions. Given the large variability in the spatial distribution and global magnitude of emissions in both process models and inversion studies (e.g. see Table 3, Melton et al., 2013), it is hard to determine which inventory may contain the most accurate spatial distribution of emissions. Although the Fung emissions are now 25 years old, and have been proceeded by newer wetland emission estimates, it is not clear that newer estimates are necessarily better (or worse). Note that the Fung wetland emissions were used in a variety of models in the TRANSCOM studies in 2011 and 2013 (Patra et al, 2011; Saito et al., 2013).

4. Ideally, you'd carry out a supplementary model-run in which an alternative wetland scenario is used. The Bousquet (2011) inversion inventory, for example, assimilated observations of CH4 made throughout the Arctic, and would likely, therefore, be able to capture the seasonal cycle of Arctic CH4 well. However, according to Figure 1, it does not show the same delayed seasonal cycle and large magnitude of autumnal emissions required in your FUNG_DEL cycle in order to capture the seasonal cycle of CH4 . Also, as far I can tell, it has not been compared to observations of methane isotopologues before, and doing so may back up your conclusions that significant emissions deeper into the autumn are necessary.

   Unfortunately it is not possible to perform any further comparable simulations due to changes in computer platform.

   The Bousquet inversion inventory provides estimates of wetland emissions from 1993 to 2009, and it is the average of the years 1993 to 2004 that was shown in Figure 1 (these years were chosen originally to aid comparison to the WETCHIMP data). However, when considering the year by year data, there is a large inter-annual variation in total 50-90°N wetland spring-time emissions in the Bousquet dataset, with negative or very low total wetland emissions from latitudes >50°N occurring during May in many recent years. Years in which total May emissions >50°N are either negative or very low (similar to winter values) in the Bousquet dataset are: 2002, 2005, 2006, 2007, 2008 and 2009. Therefore emission

data derived from the Bousquet atmospheric methane inversions supports our result for very low high latitude wetland emissions in May (and thus a later spring/summer kick-off in wetland emissions) for the 2005-2009 period. We have now changed Figure 1 to show Bousquet average emissions from the years 2005-2009 (as these are the years that we later use for observational data).

As well as updating Figure 1, we have added a comment about the varying seasonality of the Bousquet dataset to Section 7.

5. Related to this, I note that you used the GFEDv2 biomass burning inventory. Version 4 of this inventory is now available, and any changes to the impact that the heavier δ13C-CH4 has at these locations might affect your conclusions. However, I accept that the relative contribution of biomass burning emissions compared to wetland emissions at these latitudes is probably very small and therefore unlikely to have an effect unless emissions are local to the measurements.

   We agree that ideally these simulations could be updated to use version 4 for biomass burning emissions. However, as outlined above, no further comparable model simulations are now possible. An analysis of our tagged tracers demonstrates that biomass burning emissions have a negligible impact on seasonal cycles and the latitudinal gradient at these latitudes. Therefore we do not believe updating the biomass burning emission inventory would alter our conclusions.

6. If further simulations are not possible, I think a discussion of the effect of your choices on your results should be included in the results section.

   We assume this comment is aimed principally at our choice of wetland emission dataset (an issue also brought up by Reviewer 2). We have replied to these comments above and added further information to the manuscript about spatial/temporal emission distributions in both the Bousquet dataset (as requested by Reviewer 1) and the LPJ-Bern dataset (as requested by reviewer 2), see Section 6.2.2, Section 7.

   In addition, in Figure 2, we have added a new panel providing more information on the latitudinal variation in emission seasonal cycles in the Fung et al. dataset. We found that varying the spatial distribution of high latitude northern wetland emissions in the Fung dataset in a simple way did not improve the comparison with atmospheric observations (please see also our reply to referee #1’s comment no. 3 and referee #2’s comment no. 1).

7. Page 5, line 16: This is the first mention of the BASE scenario. You should make explicit that here, 'BASE' refers to the control experiment that uses the emissions described in the previous section, rather than some model set-up.Page 6, line 6-10: This paragraph needs a little more detail. You have not previously described the locations of those measurements made further south than Cold Bay (perhaps they could also be included in Figure 4?). You say that the gradient in δD-CH 4 is captured, and also that it is underestimated in the NH mid latitudes. Can you explain more clearly? It looks to me that perhaps the δD-CH 4 is mostly captured quite well as far to 50S, but that using the South Pole value as a baseline is shifting the model away from the observations. Perhaps it's the SH gradient that isn't captured, rather than the NH gradient?

   The text has been changed to read 'BASE control scenario' to make this clearer.

   We agree with the referee's comments regarding the δD-CH 4 latitudinal gradient. The paragraph discussing this at the end of Section 5.1 has been expanded.

   A description of the other measurement sites used in this study has been added to Section 2 and their locations plotted on the previous Figure 4 (now Figure 3).

8. Page 8, line 12: It's a shame that there are no δD-CH 4 ratios included here for completeness, but since the changes to the wetland emissions in this section of the study don't improve simulated CH 4 or δC 13 -CH 4 concentrations, I understand the reluctance to carry out the runs.

   As previously mentioned, unfortunately no further comparable runs are possible at this point.

9. Page 8, line 18: The name "WETLD_X2" is a little misleading, as emissions have been increased only by 50%. Can you change this name?

   We have changed the name of this scenario to INC_WETLD.

10. Page 11, line 15: Are the model lines here full zonal means across all longitudes? If so, is there any impact on the comparisons at the sites in the Arctic if you compare only at the measurement locations? I think the plot would be too busy if you included these comparisons within it, but you could mention it in the text if there is any effect.

Yes, these are full zonal means. We did initially include the measurement location data, but as suggested by the reviewer, the plot became too busy and so it was removed before submission. As requested, we have now added some text to Section 6.3 describing the impact of using model data from measurement station locations rather than zonal means (and to make it clear zonal means are plotted).

11. Figure 3: I think that this plot could be a little clearer. Can you include the locations of the measurement sites here (or in Figure 4)? Is this an annual mean or is it the peak summertime emissions? Can you differentiate between regions where wetland emissions are zero and where they're just smaller than the lowest value in your colourbar?

    We have added the location of the measurement sites to the previous Figure 4 (now Figure 3) as it enables all sites to be included. The Figure caption has been updated to clarify annual mean emissions are shown. The colourbar has been changed to differentiate regions where wetland emissions are zero.

12. Perhaps you could include a similar second panel showing the standard deviation of the emissions, or the month during which emissions peak (or at least mention it in the text)? i.e. do emissions peak in July everywhere in the Artic, or does it vary by region?

    We have now included a second panel in Figure 3 that shows zonally summed emissions for each of the summer emission months (seasonality does not vary greatly with longitude). For latitudes < ~70°N, emissions peak in July. For latitudes > ~70°N, emissions are fairly constant for the June-August period, and decline slightly for September.

13. Figure 10: Can you differentiate the lines more clearly in this plot? The difference between the dash, dot, dot-dash and dot-dot-dash lines is not obvious enough in a plot of this size (especially as they only deviate in a small subsection of latitudes).

    We have now tried using various different line types and colours for this figure. In our opinion the best improvement was obtained by swapping some of the line types in the legend and increasing the length of the y-axis.

Technical corrections:

Page 1, line 11 and throughout: I find the use of the term 'coloured' throughout the manuscript to describe the different tracers a bit odd, although I accept that it can be a difficult idea to describe well. I'd suggest changing to the term 'tagged' or similar for clarity.

Text changed to read 'tagged'.

Page 2, line 3: "to-date" -> "to date" (no hyphen)

Changed.

Page 12, line 29: "May-time emissions" -> "May emissions"/"emissions than predicted in May"

Changed.

Page 13, line 13: "currently lacking" -> "currently-lacking"

Changed.

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.