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Comment on acp-2021-843

Anonymous Referee #2

Referee comment on "Role of emission sources and atmospheric sink on the seasonal cycle of CH₄ and d13-CH₄: analysis based on the atmospheric chemistry transport model TM5" by Vilma Kangasaho et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-843-RC2>, 2021

Review for 'Role of emission sources and atmospheric sink on the seasonal cycle of CH₄ and d13C-CH₄: analysis based on the atmospheric chemistry transport model TM5' by Kangasaho et al.

General Comments:

This study investigates the influence of emission sources on CH₄ and d13C-CH₄ seasonal cycles between 2000 and 2012, comparing results from two different EDGAR emission inventories. A number of model simulations using the TM5 model have been performed, analysed independently and then the results compared to a small subset of atmospheric observations available. The main conclusions outlined in the abstract are that the seasonal cycle of emissions from enteric fermentation and manure management are better represented in EDGAR v5.0 (where they have no seasonal cycle) than in v4.3.2, that the modelled d13C-CH₄ seasonal cycle at Alert was about half that indicated by the observations, and that results from Niwot Ridge indicate that the proportion of biogenic to fossil emissions should be revised.

Whilst I believe this study does contain new useful information concerning the methane budget and our understanding of its seasonal cycles, I cannot recommend it for publishing in its current form. The manuscript as it stands contains a lot of detail in the text which does not add to scientific understanding and is hard to follow in places, which could be made more concise. In addition, the main aims of the simulations and conclusions could be better defined. For example, what are the major uncertainties in the methane budget and how/where can examining its seasonal cycles help reduce these uncertainties? What is new here compared to previous studies analysing the seasonal cycles of CH₄ and d13C-CH₄? There are also a variety of typos/grammar which require correcting.

Specific Comments:

L3: change to 'These include emissions' etc.

L8: The text says that the phase ellipses do not form straight lines due to emissions. However, later in the text one of the conclusions is that even with constant emissions, the phase ellipses are still not straight lines due to the influence of sinks other than OH.

L36: Do the authors mean 'Reported inventories for anthropogenic based thermogenic and biogenic CH₄ emission seasonal cycles mainly depend on political decisions' rather than the emissions themselves?

L55: A literature range for the magnitude of the soil sink would be useful here.

L58: Rephrase the way it is written, it sounds like the seasonality of the sinks is due to the KIE which is incorrect.

L94: Is Crowley et al. 1999 the correct reference here (the only mention of a KIE for CH₄+OH I could see in this paper puts it at ~1.005, based on Cantrell et al. 1990)? Apologies if I've missed something.

L100-105: Is the magnitude of the soil sink used in TM5 known (Tg/yr)? There is quite a large range in the literature, and the magnitude assumed would be relevant for the CH₄ seasonal cycles based on text later in the manuscript.

L117: The d¹³C-CH₄ values in the stratosphere could be relevant for your analysis if seasonal variations in stratosphere-troposphere transport influenced d¹³C-CH₄ seasonal cycles in the troposphere. Is this something that has been considered or might be significant at higher latitudes?

L154: 'EFWW' should be 'EFMM'

L188: I'm not sure I understand exactly what the missing data in grid cells is here. Do the authors mean, for example, there are grid boxes where you have wetland emissions from LPX-Bern, but no isotopic data is available for that grid box from Ganesan et al.? Could this be clarified?

P9: Consider combining section 2.3 and 2.5 (or move location of Table 2)? Table 2 appears in Section 2.3, but is not mentioned in the text until Section 2.5.

L245: Section 3.1.1: There is a lot of detail here that I'm not sure contributes to the science understanding gained from this paper, and most of which can be deduced from looking at Figure 2. I think it would be easier to follow if the amount of detail regarding e.g. exact lag periods, percentage change in seasonal cycle magnitudes between certain simulations, could be reduced, so that the text highlights the most important differences between the simulations that can be seen in Figure 2, and how this adds to our understanding.

L321: "The effect of the soil sink is small at these latitudes, so probably the seasonal cycle of $\delta^{13}C$ is preliminarily driven by the atmospheric sinks at these latitudes". I think a bit more information would be useful here. The ellipse produced for SIM_NS differs in gradient at all latitudes from the theoretical KIE line when only OH is considered. Why do the authors think this is if not the soil sink?

Figure 3 caption: The solid black line is described as 'the KIE line of SIM_NS, and considering only the OH sink', this is confusing. Later in the text (L340), I think is the correct description for the solid black line: 'the theoretical KIE line when only the OH sink is considered'.

L325: "When there are seasonal cycles only in the atmospheric sinks...", I think should be "When seasonal cycles solely arise from seasonal variations in the atmospheric sinks...".

L326: If there are no emissions and no sinks affecting the seasonal cycle, then there would be no seasonal cycle? Could the authors clarify this?

Section 3.1.2: Again there is a lot of detail here, but it's not clear what the take away message is to the reader. As for Section 3.1.1, this section should be made more concise.

L429-432: Could seasonal changes in transport patterns (horizontal or vertical from the stratosphere) also influence the seasonal cycle at Alert? Given the high latitude of Alert, the CH₄ sink there is likely to be relative small all year compared to the tropics.

L476: typo - 'Delta13'

L487: The Discussion section does not discuss the results of the paper, but is instead a mostly a summary of literature available regarding seasonal variations in emissions.

Line 557: reference missing?

L611-613: I disagree that results from this study support the conclusions of Gromov et al. (2018) who concluded a negligible tropospheric Cl sink for CH₄, as the simulations are able to capture much of the seasonal variation in CH₄ and d13C-CH₄ at the South Pole. I think a further simulation including a tropospheric Cl sink and an assessment of its impact on the seasonal cycle at SPO would be required to back this statement up. As the authors point out, the highest Cl concentrations are anticipated to be in the tropics, and may not have a strong influence at SPO. Also, if the d13C-CH₄ seasonal cycle at SPO is mainly controlled by the atmospheric sinks, could the choice of KIE for 13CH₄+OH used in TM5 influence results here? As the authors point out, there are 2 differing values in the literature.

L631-632: To conclude that the comparison between observed and modelled d13C-CH₄ at SPO suggests the emission seasonality in the model is at the right level, I think that Figure 4 needs to show an influence of emission seasonality on the d13C-CH₄ seasonal cycle, which is not clear from the current plot. Perhaps if SIM_NS could also be plotted in Figure 4, this would show more of an impact?

L633: typo, 'moel' should be 'model'