Authors' response to anonymous referee #2

In the following, *referee's comments are in italic*, authors' responses in normal font.

We thank the anonymous referee #2 for providing the valuable feedback. We apologize that the current form of the paper did not meet expectations.

Please note that some of the Figure and Table numbers have changed due to modification.

Article:

Table 1 *new

Table 1 -> Table 2

Table 2 -> Table 3

Table 4 *new

Figure 5 *removed

Supplementary:

Table 3 * removed

Table 3 * new

Table 4 *new

Figure 4-> Figure 9

Figure 4 *new

Figure 5 *new

Figure 6 *new

Figure 7 *new

Figure 8 *new

Figure 5 -> Figure 10

Figure 6 *removed

Figure 7 *removed

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 uncertainites? 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.

We apologize for the excessive amount of unnecessary information in the article. We have now reduced the number of details and rewritten some part of the article to better meet your expectations. The major uncertainties are not the sources itself but the magnitude of each source. Analysing $\delta^{13}C$ seasonal cycles of various sources, e.g., anthropogenic (enriched with $\delta^{13}C$) vs biogenic (depleted in ^{13}C) can tell us whether some of the components are underestimated or overestimated by inventories. We have now formulated the goals of the study and the motivations of used methods better in the Introduction section to clarify, what new this study brings. Please see Lines 71-89. Please find more information what is new in this study from the beginning of Reply to anonymous referee #1. We also apologize for the typos/grammar errors, and now we have had the text checked by native English speakers.

L3: change to 'These include emissions' etc.

This has been corrected.

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.

We apologize for the unclear expression that was misleading. We have now rephrased the sentence: Due to surface fluxes, the anti-correlations between CH_4 and $\delta^{13}C$ are not perfect and experience large variation (p=-0.35 to -0.91) in north of 30° S.

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?

We apologize for the unclear expression for this. We have now rephrased the text as follows: Anthropogenic CH_4 emission seasonal cycles also have uncertainties. Although some countries report emission magnitudes to e.g., UNFCCC, often only annual values are reported, and emissions from e.g., rice paddies may not properly consider e.g., temperature dependencies and soil properties (Yan et al., 2009).

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

We thank you for the feedback. We have now added this: The estimated magnitude of soil sink also varies; from bottom-up estimates is 11–49 Tg CH_4 yr^{-1} and from top-down estimates 27–45 Tg CH_4 yr^{-1} −1 (Saunois et al., 2020, and references there in).

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

This has been corrected and the new sentence is: Sinks enrich the atmosphere in the $^{13}CH_4$, due to their kinetic isotopic effect (KIE), and they have a strong seasonality, mainly due to the seasonality of OH radical in troposphere, and Cl and O(1D) in stratosphere.

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.

We apologize for the unfortunate mistake of references here. We have now corrected it as: The kinetic isotopic effects (KIE) $k(^{12}CH_4)/k(^{13}CH_4)$= 1.004 and 1.013 are used for $^{13}CH_4$ Oh and O(1D), respectively (Saueressig et al., 2001), and 1.066 (Crowley et al., 1999) is used for Cl.

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.

Thank you for your feedback. We have now rephrased the sentence and added information about the magnitude of the soil sink used in this study: Yearly totals are varying 32.7–33.8 Tg CH_4 yr^{-1}.

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?

We acknowledge the stratosphere-troposphere exchange. However, this study focuses on the modelled values in the troposphere and therefore we don't address the topic of stratosphere much. However, the STE effect can be significant, but would need another study. Further discussion can be found from lines 521-525: Other than the tropospheric sinks, the stratosphere-troposphere exchange also affects to some extent (Wang et al., 2002), and therefore the stratospheric sinks of Cl and O(1D) could contribute to the tropospheric seasonality. In the stratosphere, the effect of emissions is negligible, and the seasonality is largely driven by the atmospheric sinks. This was true in our simulations as well. The chemical sinks strongly enrich the $\delta^{13}C$ in the stratosphere. Therefore, the stratospheric air that returns to the troposphere can affect the tropospheric seasonality of $\Delta\delta^{13}C$ in mid and high latitudes.

L154: 'EFWW' should be 'EFMM'

This has been corrected.

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?

We apologize for the unclear expressions and have now clarified this in the article: We first examined the filled values (grids with no initial value assigned) by applying the values from Monteil et al. (2011) and Thompson et al. (2018) (Table 2).

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.

We apologize for the misallocation of Table 2 (now Table 3).

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.

We apologize that this section did not meet expectations and we have now rewritten the section.

L321: "The effect of the soil sink is small at these latitudes, so probably the seasonal cycle of d13C 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?

We have rewritten this section. The phase ellipse at the SH high latitudes do not follow the theoretical KIE line, and that can be due to the effect of other atmospheric sinks, i.e., stratospheric Cl and O(1D), and horizontal long-range and vertical transport. However, we should not have discarded the effect of soil sink completely, and the sentence/section is rewritten as follows (please see lines 323-333): At latitudes south of 30° S, the SIM_E5 phase ellipses' eccentricities are very high, with strong anti-correlation compared to those in the NH (Fig. 3). This indicates that ΔCH_4 and $\Delta\ \delta^{13}C$ are close to the perfect inverse phases, i.e., the seasonal cycles are330preliminary driven by the atmospheric sinks and little affected by the seasonal cycle of emissions. However, the phase ellipse from SIM_NS does not exactly follow the KIE line, indicating the effect of sinks other than OH, i.e., stratospheric Cl, O(1D) and soil sinks, and horizontal long-range and vertical transport.

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'.

You are correct and this has now been corrected. We apologize for the mistake in the description.

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…".

This sentence has been rephrased.

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?

We apologize for the unclear text. It is clear, that there is no seasonality in the emissions and sinks. We have now rephrased this sentence: Such a case would be when the CH_4 fluxes have no seasonal cycle and only the atmospheric sinks derive the seasonality of the mixing ratios. In that case, the ΔCH_4 maximum (minimum) occurs simultaneously as $\Delta\ \delta^{13}C$ minimum (maximum), and we expect d_{min} = 0 and d_{max} = 366/2 = 183.

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.

We apologize that this section did not meet expectations and we have now rewritten the section.

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.

It is possible that the transport patterns also influence the seasonality at Alert or other locations at high northern latitudes. However, we would need to do new research to investigate this, and this study is not focused on this topic and therefore unable to provide answers to this question. We shortly address stratosphere (lines 521-525 and 551-552).

Lines 521-525: Other than the tropospheric sinks, the stratosphere-troposphere exchange also affects to some extent (Wang et al., 2002), and therefore the stratospheric sinks of Cl and O(1D) could contribute to the tropospheric seasonality. In the stratosphere, the effect of emissions is negligible, and the seasonality is largely driven by the atmospheric sinks. This was true in our simulations as well. The chemical sinks strongly enrich the δ^{13}C in the stratosphere. Therefore, the stratospheric air that returns to the troposphere can affect the tropospheric seasonality of Dd3 C in mid and high latitudes.

Lines 551-552: The focus of the study was the troposphere. Nevertheless, the tropospheric cycles are affected by the stratosphere-troposphere exchange, and this calls for further studies.

However, there are also other reasons such as the used wetland isotopic signature, see discussion lines 397-403: Although we have used a recently published spatial distribution of source signatures where available, there are still large uncertainties in the modelled δ^{13}C values due to, e.g., the vegetation types, especially for the tropical wetlands (Ganesan et al., 2018; Fisher et al., 2011). We found that δ^{13}C values in high northern latitude sites, e.g., Alert, were overestimated by the model. Isotope signatures in autumn in northern wetlands can be as low as -79.4 ‰ (Hornibrook, 2009), and using more negative isotopic signatures could lead to a better match with the observations. Wetland CH_4 emissions and their seasonality are large globally (Supplementary Fig. S1 and Table 1) and therefore any error in the source signature is expected to have a large impact on the seasonality of δ^{13}C.

L476: typo – 'Delta13'

This has been corrected

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.

We apologize for the format of the discussion, and we have now edited this section to meet expectations.

Line 557: reference missing?

This has been corrected.

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.

To address this argument and topic further, we would need to make new runs to investigate this topic. This study cannot provide an answer. It is true that the selection of KIE value has an impact on the results, but we didn't test different KIE values for reaction with OH. We used the value of KIE = 1.004 based on Saueressig (2001) as recommended in Burkholder et al. 2019. Please see lines 605-615 for changes.

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?

Thank you for your feedback. We have now added the simulation SIM_NS to the plot to provide more information.

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

This has been corrected