Review of Kangasho et al.

The study of Kangasaho et al. investigates the influence of emission sources and atmospheric sinks of CH4 on the seasonal cycle of CH4 and d13C based on simulations with the atmospheric transport model TM5.

Similar studies, which laid the foundations to our understanding of the seasonal cycles of CH4 and d13C and their phase relations, have already been published in the early 2000s. Bergamaschi et al. (2000), for example, interpreted measurements at Izaña using global simulations with the TM2 model. Allan et al. (2001) also performed TM2 simulations and focused on the interpretation of the seasonal cycles in the extratropical southern hemisphere. Together they established a theoretical understanding of the phase relations between the seasonal cycles of CH4 and d13C and of phase ellipses also used here. In those studies, CH4 and d13C were simulated separately for different emission sources and then added up to the totals, which enabled a thorough analysis and understanding of the results. In the study of Bergamaschi et al. (2000), this information was further used in a simple inversion framework to adjust the emission sources to better match the CH4 and d13C observations at Izaña.

In the study of Kangasaho et al., instead, only two separate tracers were simulated (total CH4 and total d13C), while the influence of different emission sources or different seasonal emission cycles was analyzed through a set of sensitivity runs (see Table 2). This setup seems much less flexible and has clear limitations with respect to the interpretability of the results.

It is difficult to see what we can learn from this study that we didn't know already before. The main goal seems to be to reassess the seasonal cycles given our present best knowledge of sources and sinks and their isotopic signatures and to evaluate whether the seasonal emission cycles proposed in the EDGAR emission inventories are consistent with observed seasonal cycles in CH4 and d13C (and their phase relations).

However, the goals of the paper are not clearly stated and the chosen approaches are rather poorly motivated. The paper provides mainly technical descriptions of the model setup and simulations but fails to motivate the design choices. Another major weakness is a lengthy and unfocused presentation and discussion of the results with long descriptions of what is seen in figures but failing to extract the essential information in a concise manner.

Thus, I cannot recommend publication of the manuscript in its present form. It either needs

- a much clearer explanation of the goals of the study
- a much clearer motivation of the simulation setup and explanation of the methods used to analyze the results
- a much shorter and more concise presentation and discussion of the results. The present results section is unnecessarily lengthy and unfocused (see also points below)

Another, probably better option, would be to redesign the simulation experiment, notably by not only simulating two separate tracers but simulating CH4 and d13C from the different sources individually as in those earlier studies. Such an experiment could also include tracers with seasonally varying and seasonally constant emissions.

Further major points:

Section 3.1.1 is very hard to follow and in fact painful to read. The text basically describes what is seen in the figure but adds a lot of numbers that often seem quite irrelevant and certainly do not help telling a concise story. If needed, such numbers could be summarized in a table (latitude band, amplitude of seasonal cycle, month of maximum, month of minimum, etc.), but actually I don't see the point in presenting all these details at all. This section could easily be cut to one third by only focusing on the main points and leaving out all the fine details. There is also a lot of speculation in this section ("we suspect that", "this indicates that", "this could explain" etc.) which is related to the weaknesses of the simulation design mentioned earlier.

The main take home messages from Figure 2 could simply be: (i) the seasonal cycles of CH4 and d13C are to first order in antiphase, i.e. maxima in CH4 correspond to minima in d13C and vice versa (consistent with what we know from previous studies). Minima (maxima) in d13C, however, tend to be xx months earlier than the maxima (minima) in CH4. (ii) at high latitudes in the southern hemisphere, where emissions are small and the seasonal cycle is primarily determined by sinks, differences between the different sensitivity simulations are very small. (iii) At northern mid- to high latitudes, differences in the seasonal timing of emissions has a pronounced influence on the seasonal cycle of CH4 and especially of d13C. In the simulation with constant emissions (E5_WETNS), for example, wetland emissions are higher in winter to spring and lower in summer to autumn compared to the reference simulation (E5). This leads to higher CH4 north of 30°N in spring and lower in autumn and, because of the strongly depleted isotopic signature, to a minimum in d13C in early spring (not seen in E5) and a maximum in autumn which is delayed compared to E5 by xx months. (iv) At low southern latitudes (0-30°S), there is a double peak structure in CH4 due to xyz and, in case of constant emissions, also in d13C. When seasonal variations in emissions is accounted for (ref. simulation E5), however, the double-peak in d13C disappears because d13C is pulled down by strong wetland emissions in the southern hemisphere summer and autumn and pushed up by strong biomass burning emissions in the southern hemisphere spring. A few further details may be added, but further discussions could be presented in the context of the comparison with the observations in Section 3.2.

Similarly, the presentation of the results in terms of phase ellipsoids needs to be cut strongly and could be presented along the same line of arguments as above.

One of the conclusions drawn in the study is that the seasonal cycle of wetland emissions is the dominant driver of the seasonal cycle of d13C in the NH and tropics (see corresponding sentence in the abstract). However, this seems to be primarily based on the comparison between simulations E5_NS and E5_WETNS. Comparing simulations E432 and E432_EFMMNS would tell a different story, namely that a seasonal cycle in agricultural CH4 emissions could also perturb the d13C seasonal cycle. Which effect is closer to reality should be decided based on the comparison against observations and not just on the comparison between individual sensitivity runs. Note that in all simulations using EDGAR 5 (E5, E5_WETNS, E5_NS) the agriculture sector has no seasonal variability, too, and therefore it is clear that this sector cannot contribute to differences between these runs. The conclusion drawn on page 12, line

280 (and again in the abstract and conclusions section), is therefore not valid. The main difference between simulations E5_WETNS and E5_NS (at least in the southern hemisphere) is that in E5_WETNS biomass burning emissions have a seasonal cycle but those of E5_NS have not. Comparing these two simulations tells a lot about the influence of biomass burning emissions (see e.g. panel for latitude band 0-30°S in Fig. 2), but this is hardly addressed in the paper.

Section 3.3 describes seasonal cycles in the stratosphere. This section has several weaknesses. First, it is poorly connected to the rest of the paper focusing on the troposphere. How stratosphere-troposphere-exchange affects the seasonal cycles of CH4 and d13C would actually be an interesting topic, but the simulation setup does not allow addressing this question. Second, averaging the seasonal cycles over a large vertical range from 88 hPa to the top of the atmosphere does not seem a good idea, as there are probably significant differences in seasonality across this altitude range. The authors should rather focus on a single level (e.g. 80 hPa) or present vertical cross-sections/vertical profiles of the evolution in the stratosphere. Third, the discussion of the results is not convincing. For example, the authors argue that the strong amplitude of the seasonal cycles at the South Pole compared to the North Pole is due to the Brewer-Dobson circulation being stronger in the Southern Hemisphere, but actually the Brewer-Dobson circulation is stronger in the Northern Hemisphere, see e.g. Rosenlof (1995; https://agupubs.onlinelibrary.wiley.com/doi/10.1029/94JD03122).

Section 4.1 needs to be rewritten and shortened as well. Rather than discussing the results in the context of other studies, it basically presents a review of existing literature. This is not what I expect from a discussion section. Section 4.2 is slightly better but could also be shortened.

Minor points:

The concept of phase ellipsoids should be better explained in the introduction section or somewhere else in the paper. Without this it is difficult to follow the discussion in section 3.1.2. Why do sinks produce a line but emissions an ellipsoid? Actually, to my knowledge only seasonally changing emissions produce an ellipsoid.

Sections 2.2.1 and 2.2.2 presenting the anthropogenic and natural emissions are quite long. A lot of the information could be put in a table (category, data source, total emissions per category, amplitude of seasonal cycle, etc.) so that the text could be shortened by referring to the numbers in the table rather than listing all these numbers in the text.

What is the basis for the selection of the three observation sites ALT, NWR and SPO? Data availability? Distance from sources? Would there be other stations that could have been looked at? Why were they not considered?

Figure 2: It would be good to highlight the line corresponding to SIM_E5 as a thick black line as a reference. The discussion in Section 3.1.1 should mainly refer to this line and discuss other simulations with respect to their deviations from this reference.

Small points, typos and grammar:

The article needs substantial editing for grammar:

For example, wrong prepositions are used at many places, e.g. "in 60-90°N" instead of "AT 60-90°N" or "the effect in the d13C cycle" instead of "the effect ON the d13C cycle". The suggested corrections to the abstract below give an indication of the magnitude of this problem. I do not provide any corrections related to grammar for the rest of the manuscript.

Affiliations: Shouldn't it be INSTAAR rather than NSTAAR?

Abstract:

Page 1, Line 3: What do you mean by "examined"? This is a very vague term.

P1, L3: "Those includes" -> "Those include"

P1, L5: "in addition to other sources" -> in addition to other natural sources"

P1, L6: "global uniform value" -> "globally uniform value"

P1, L9: "in north of" -> "north of"

P1, L11: "timing of d13C seasonal minimum" -> "timing of the d13C seasonal minimum"

P1, L11, "days in 60°" -> "days at 60°"

P1, L14-15: "In light of this research, comparison to the observation" -> "These comparisons"

P1, L19: In what sense should the "proportion of biogenic to fossil based emissions" be revised? Please be more specific.

P2, L32: "much studied" -> "studied intensively"

P2, L36: Why should these emission cycles depend on "political decisions"?

P2, L52: "also warned that" -> "also cautioned that"

P3, L58: Sinks do not have a strong seasonality due to the KIE but simply due to the seasonality of OH (or Cl) radicals.

P3, L63: The d13C maximum was two months EARLIER not later than the CH4 minimum in those studies.

P3, L79: "that are a coarsening from" -> "corresponding to a subset of"

P3, L79: ", and vertical mixing was calculated" -> ". Convective vertical mixing was calculated"

P4, L86: reactions of CH4 with OH, Cl and O1D are chemical but not photochemical reactions.

P4, L90: It is not quite clear from the description whether the levels of Cl and O1D are prescribed in the stratosphere or whether the reaction rates are prescribed (which is not exactly the same). Please clarify.

P4, L117: I wouldn't say that the values in the stratosphere are "not important", but they are certainly "less important". Troposphere and stratosphere are linked through stratosphere-troposphere-exchange.

P5, L132: "enriched 13CH4" -> "enriched in 13CH4". The seasonal cycle "is" dominated, not "are" dominated.

P5, L135: What do you mean by "keeping the same annual totals"? The same as in 2010?

P5, L143: "due to the differences" -> "due to the following differences"

P5, L143-146: These sentences are a bit confusing and could probably be formulated more elegantly.

P5, L148: Delete "compared to v5.0".

P5, L150: "increasing trend" -> "increasing emissions"

P7, L183: For some sources, spatially varying isotopic signatures were used following Feinberg et al. (2018). It would be good to know in more detail how Feinberg et al. collected this information. Certainly, it was not generated "based on global chemistry-climate simulations with SOCOL".

Table 2: Replace "vary globally" by "varying globally" in the table caption.

P8, L184: Geological and wetland emissions should not be described together in a single sentence, because these are completely independent sources.

P8, L199: What are the values proposed by Monteil and how do they differ from those of Thompson? Why do you not list the values of Monteil also in Table 1?

P9,L202: It should probably be "In contrast" rather than "In addition".

P9, L203: I don't understand the statement "decimal values instead of rounded integers" at all (I can only guess what this means).

P9, L212: Why "In contrast"? NWR is also far away from sources and measures background air like SPO. I don't see the contrast.

P9, L213: "Rockies" -> "Rocky Mountains"

P10, L227: "we examined the seasonal cycle" -> "we examined the impact of the seasonal cycle"

P10, L252: "in 90°S – 60°S approximately" -> "at 90°S-60°S is approximately"

P12, L269: "in spring" -> "from spring"

P12, L274: "two maxima peaks" -> "two maxima"

P12, L275: "In south of" -> "South of"

P12, L277: "closer follows" -> "more closely follows"

P12, L282: shouldn't it be "increased" rather than "decreased"?

P12, L296: "only one month" -> "only during one month"

P13, L305: "the strong effect" -> "the strong contribution"

P13, L308: "and magnitude of autumn" -> "and the magnitude of the autumn"

P13, L318: "against that of" -> "against those of"

P15, L342: On page 4 the KIE of the reaction with OH had a value of 1.004. Here it has a value of -3.98 per mil. How do these numbers relate?

P17, L408: I can't see in Figure 4 that the modelled amplitude is larger "mainly due to a deeper minimum". The maximum seems to be equally overestimated.

P18, L429: "not only the cause" -> "not the only cause"

P18, L437: "depletion better than in ALT" -> "depletion better"

P18, L445: "reach at maximum" -> "reach a maximum"

P18, L446: "After DOY=75 that" -> "After DOY=75"

P18, L449: "reach its maximum" -> "reach their maximum"

P19, L466-467: I don't understand why the first sentence leads to the conclusion presented in the second sentence. It should rather be the other way round: "Because" chemistry time scale is shorter than transport d13C stabilized more quickly at mid- to high latitudes and not "therefore".

P19, L476: Remove the "Delta" and add a "C" after d13.

P20, L491: This sentence needs clarification. A factor 1.5 to 7 change in the amplitude of the seasonal cycle of d13C due to a simple change of the isotopic signature (of what??) by 1 per mil sounds like a huge effect, which could make any model simulations useless.

P21, L504: "within in a country" -> "within a country"

P22, L567: Here a "(reference)" is missing.

P24: L614-615: The last two sentences in this section contradict the previous sentence concluding that the chlorine sink is not important.

P24, L624: "are reverse of deltaCH4 cycles" -> "are opposite to those of deltaCH4"

P25, L633: "moel" -> "model"

P25, L633: "extend" -> "extent"