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Reply on RC2

Haiyue Tan et al.

Author comment on "An integrated analysis of contemporary methane emissions and concentration trends over China using in situ and satellite observations and model simulations" by Haiyue Tan et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-464-AC3, 2021

Response to Reviewer #3

Comment [3-1]: This work attempts to attribute the sources contributing to the atmospheric CH_4 mixing ratio and their trends in China using the GEOS-Chem model simulations driven by two commonly used global anthropogenic emission inventories. It uses in-situ and satellite observations of CH_4 mixing ratios to explain the model results. Study also performs sensitivity test with OH to reproduce observed CH_4 mixing ratios and trends over China.

The discussion on the differences between the model results and observations is not sufficient. Authors can address some missing pieces of information or address some limitations in the results shown. I recommend the manuscript send back for the major revisions with following major/minor comments.

Response [3-1]: We sincerely thank the reviewer for the constructive comments, and time spent reviewing the manuscript. Each comment has been implemented in the revised manuscript.

The main comments raised by the reviewer concern about whether our model evaluations support the use of CEDS v2017-05-18 emissions rather than EDGAR v4.3.2 for simulating the CH_4 trends over China in the time period of 2007-2018. This study does not attempt to provide a conclusive judgement that the CH_4 emission estimates in CEDS are more accurate than those in EDGAR. Instead based on analyses of a series of global model simulations accounting for the interannually varying OH and considering their latest available years (2012 for EDGAR and 2014 for CEDS), we find that the rising CH_4 levels over China over 2007-2018 can be better captured in the model simulation with the CEDS inventory. This provides an important constraint on the trends of CH_4 emissions over China. The comments from the reviewer are valuable and help us better interpret the model simulations. Please see our point-to-point responses below.

Comment [3-2]: Major comments: The study claims "model simulation using the CEDS inventory and interannually varying OH levels can best reproduce observed CH_4 mixing ratios and trends over China". I don't agree to a certain extent.

First of all, EDGAR v4.3.2 provides global emission estimates, at source-sector level, for the historic period from 1970 until 2012. How did the author estimate the EDGAR emissions beyond 2012? It appears the emissions are extrapolated (?) till 2018 for this study. Similarly, in case of CEDS inventory the emission estimates are during 1970-2014. How does the emissions are calculated beyond 2014 in this case too?

Response [3-2]: Thank you for pointing it out. For both EDGAR and CEDS, we did not extrapolate them, and instead as we now state in Section 2.3: "For all the datasets of emissions (using EDGAR or CEDS) and sinks as described above, the closest available year will be used for simulation years beyond their available time ranges." This is mainly because of the large uncertainties in global CH_4 emission estimates (Saunois et al., 2020) as well as the slow trends of emissions in China after 2010 compared to 2000s (Sheng et al., 2021; Liu et al., 2021). As methane has a lifetime of about 9 years, the changes of CH_4 mixing ratios after 2012 are strongly affected by its emissions before, which drives the model differences with EDGAR vs. with CEDS.

We now state here in Section 2.3: "the closest available year will be used for simulation years beyond their available time ranges as recent studies suggested weak trends in Chinese CH_4 emissions after 2010 (Sheng et al., 2021; Liu et al., 2021). Since CH_4 has a long lifetime of about 9 years, model results in the later years (e.g., after 2012 for EDGAR and after 2014 for CEDS) are strongly affected by the emissions in earlier years".

Comment [3-3]: In Figure 6, over 'DSI' and 'LLN', simulations from both inventories are comparable at least till the year 2016 (it appears that, trends are affected by later years simulations for GCE). Over 'SDZ', EDGAR performs better than CEDS, however, over 'WLD', CEDS is better than EDGAR. Overall, these results are not very conclusive to say CEDS is better.

Response [3-3]: As we discussed above, simulated CH_4 levels at these Chinese sites after the year 2016 would still be strongly affected by emissions in earlier years. The high bias at SDZ in the CEDS model simulation likely suggest that regional CH_4 emissions around this site (i.e., North China) are too high in CEDS.

We now state in the Section 3.2, "These results can be explained by the higher CH_4 emission estimates and increases in CEDS than EDGAR since 2007, and may also reflect the regional CH_4 emissions around SDZ (i.e., North China) are too high in CEDS. Further evaluations of the two model simulations with CH_4 column mixing ratio measurements (since 2011) at six TCCON sites in Asia (Wunch et al., 2011) show similar results, with small biases of 0.2%–1.0% in CH_4 mixing ratios for GCC and negative biases of 2.6%–3.7% for GCE (Fig. S3). This again reflects the higher Chinese CH_4 emission estimates in years around 2012 in CEDS than EDGAR, which then affect the model simulations afterwards by using their emissions of the latest available years."

Comment [3-4]: In Figure 7-8, the trend correlations for model simulations using EDGAR and CEDS with GOSAT are not significantly different.

Response [3-4]: Both correlation coefficients (r) and mean biases over China between model simulations and GOSAT are presented in Figs. 7 and 8. The correlation coefficients show the similarity of spatial distributions between observed CH_4 mixing ratio (or trend) and model results, and are not significantly different for the two model simulations, however, the mean biases indicate that model results with EDGAR underestimate the GOSAT observed trends in CH_4 mixing ratios over China.

We now state in the text: "As for the CH_4 trends during 2010–2017 over China, both GCC and GCE show similar spatial patterns as those observed by GOSAT with moderate correlations of 0.2–0.5, while GCC model results have smaller biases of -1.7–0.4 ppbv a^{-1}, compared to GCE results that underestimate the trends by 2.6–4.7 ppbv a^{-1}."

Comment [3-5]: In Figure 9, it appears for HIPPO observations, both simulations (performed using EDGAR and CEDS emission inventory) are within observed standard deviation. But for ATOM observations, CEDS inventory performs better than EDGAR. One reason to me is EDGAR extrapolated (?) emissions are used for the model simulations comparison with ATOM observations, whereas, in case of HIPPO observations actual EDGAR emission estimates are used. So, this Figure is also not very conclusive to say CEDS inventory is better, moreover, almost all the observations from HIPPO and ATOMS are over Pacific and American continent.

Response [3-5]: Thanks for the comment. As responded above, both EDGAR and CEDS emissions were not extrapolated, and we do not think this would affect our analyses much due to the long lifetime of CH_4. We suggest as stated in the text that "changes in the model bias for the comparisons with HIPPO and ATom measurements reflect their simulated trends in the CH_4 mixing ratios". HIPPO and ATom are two aircraft campaigns well designed for measuring global-scale atmospheric composition. The measurements over Pacific account for influences of Asian outflows, and thus can be applied for evaluating model CH_4 simulations with information on Asian CH_4 sources. Please also see our response to the next comment.

Comment [3-6]: Figure 10, mixes both aircraft observations, which is not correct in my opinion. This figure is confusing.

Response [3-6]: Thanks for raising the concern, yet we think this comparison is valid for long lived tracers such as CH_4. Both HIPPO and ATom campaigns provide measurements over the Pacific region in all four seasons and with extensive vertical profiling, supporting the analyses shown in Figure 10.

We now state in Section 2.1, "Both campaigns provide global-scale measurements of atmospheric composition in all seasons, and conduct continuous profiling between ~0.15 km and 8.5 km altitude with many profiles extending to nearly 14 km". We state in Section 3.2, "Since both HIPPO (2009–2011) and ATom (2016–2018) provide measurements over the Pacific (black box in Fig. 1), we calculate the differences between HIPPO and ATom measurements as the observed CH_4 concentration trends over this region, and these trends also largely reflect the influences from upwind Asian CH_4 sources and levels". In addition, we have revised the legends of Figure 10 for clarification following one minor comment below.

Comment [3-7]: Another issue is the source attribution of CH_4. The attribution of CH_4 sources with tagged tracer needs more evidences. The source contribution should be provided along with confidence interval. Is there any relevant study to support this analysis for CH_4?

Response [3-7]: Thanks for the comment. The tagged tracer approach has been applied in a number of studies. We now stated in Section 2.3, "The tagged CH_4 tracer approach has been recently applied in GEOS-Chem to quantify source contributions in U.S. Midwest

(Yu et al., 2021) and the GFDL-AM4.1 model with focus on the global CH_4 budget (He et al., 2020)".

We have now added error-bars on the left panels of Figure 11 to present the standard deviations of contributions from different regions as a metric of confidence level. The values are up to 11% for the contributions to CH_4 mixing ratios and up to 0.4 ppbv a^{-1} to the trends over China.

We also acknowledge that the source attribution results can heavily rely on the regional and sectorial estimates of underlying CH_4 emissions. We have stated in the last section that "It shall be noted that our source attribution results can be biased by the use of CEDS and the uncertainty in the interannual variations of OH levels", and discussed the uncertainties in this paragraph.

Comment [3-8]: Minor comments: Some places in the manuscript authors use 'Fig.' and somewhere 'Figure'. Please use uniform convention.

Response [3-8]: Thank you for pointing it out. The uses of 'Fig.' and 'Figure' have followed the request of ACP format, i.e., the abbreviation "Fig." should be used when it appears in running text and should be followed by a number unless it comes at the beginning of a sentence.

Comment [3-9]: Line 55; please add a reference after "a lifetime of 9.14 (±10%) years"

Response [3-9]: We now add the reference in the text: "Over 90% of atmospheric CH_4 is lost via oxidation by OH in the troposphere, leading to a lifetime of 9.14 (±10%) years against this sink (IPCC, 2013)."

Comment [3-10]: Fig2: How do you define the regions for tagged CH_4 tracer simulations?

Response [3-10]: The nine regions (China, India, Europe, South America, North America, Africa, Oceania, Rest Asia and the rest world) are defined mainly following Bey et al. (2001) with some modifications. We now state in the text: "The regions used for the tagged simulation are shown in Fig. 2, mainly based on Bey et al. (2001) with additional tagged regions for China and India in Asia. "

Comment [3-11]: In Figure 6-10, please mention the model configure and OH field configuration used for simulations in the caption for better clarity.

Response [3-11]: We now clarify in the caption of Figs. 6-10: "GCE (with EDGAR anthropogenic emissions and interannually varying OH; red lines) and GCC (with CEDS and interannually varying OH; blue lines)"

Comment [3-12]: Fig10: Legends needs to be adjusted properly.

Response [3-12]: Thanks for pointing it out. The legends in Fig. 10 have been adjusted from “Trend = …”, “bias (GCE) = …”, “bias (GCC) = …” to “Obs. …”, “GCE …”, “GCC …” to better present the mean values.

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