

Interactive comment on “A very limited role of tropospheric chlorine as a sink of the greenhouse gas methane” by Sergey Gromov et al.

Anonymous Referee #2

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The paper presents an interesting new angle on evaluating previous indirect claims of a globally significant role of Cl for the removal of CH₄, by analyzing the isotopic composition of the reaction product CO. The results indicate that the previous estimates are strongly overestimated.

The paper is well focused on bringing over a clear message and I appreciate that the authors try to keep it short. However, in particular in section 2 and Fig 2, where the new evidence is presented, the information is very dense, and the authors should help the reader by adding more explanation and making more explicit statements.

The derivation of equations 2 and 3 should be shown explicitly, either in the main paper or in an appendix, and more information / explanation should be added. A parameter λ_a is introduced, but what is it? An additional parameter μ is introduced, is

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this necessary? It replaces the parameter ΔS that is mentioned in the sentence above Eq. 2 to express the sensitivity, but it does not appear in the equation. This should be motivated better.

Fig 2 shows results obtained with Eq. 2 and 3, but the authors should help the reader by describing this complex figure step by step, linking it to the equations and the data. For example, why is Fig 2 shown as function of α , what does this signify, and how does ΔS enter this figure? What are the units of the numbers in Fig 2a (permil). I cannot fully follow the argumentation of paragraph 25, but it appears important for the paper. The error ranges given in lines 213 ff do not correspond to the error ranges indicated in Fig. 2.

Minor points:

Line 100: It would be useful to spell out precisely what the issue is. It is mentioned indirectly in the following lines (the CH₄ derived fraction would be too dominant, line 102), but please provide the line of argumentation explicitly: 1) the bottom-up budget of CO isotopic composition is too negative in ¹³C compared to observations, 2) the most negative course is CH₄, 3) to close the isotope budget required lowering the yield in previous studies, and 4) making CH₄-derived CO even more depleted in ¹³C would aggravate the problem.

Line 165: Can you comment on the difference in Cl levels compared to Hossaini et al. (2016)?

Line 171: On which basis do you “expect” a factor 1/5 lower variation of the CH₄-derived CO in the ETSH (I assume compared to the SH)?

Line 176: Provide some more details on η_C . This is a complicated quantity, and relevant here, so some background should be provided in the paper itself rather than referring to Gromov (2013). Specify next sentence: Is it the difference between the atmospheric isotopic composition and a global averaged source mix or the sources at

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this point and space in time?

Line 192: It is not clear what you want to indicate here: “In a statistical sense...” Do you refer to the differences derived for different Δ_m values, or the difference between the stations. And further, what does this mean?

Fig 2, and caption. What is the unit of the numbers shown as labels in Fig 2a? Is it useful to show a yield from a personal communication (M. Krol, correct spelling) in the figure without relating it to a reference?

Line 205 ff: Explain better the meaning of the sentence “Importantly, ...”. It is not immediately clear that you are less sensitive when you add a sink than when you replace a sink.

Either reword or remove lines 227 – 229. This is a confusing statement.

Line 230 – 232: Motivate where the number of “at least one-third” comes from.

Line 233: Motivate the value of 2 per mill.

Line 234 ff: Phrasing the quantification in terms of λ values is confusing. We know very well that the yield of CO is higher than 0.12. Is it not more instructive to compare the model results with the experimental data in Fig 2? I.e. discuss the “vertical” offset, which would simply imply less change in the CI sink, rather than the “horizontal” offset, which projects the real cause of the discrepancy to an unrealistic change in λ ?

Line 235 to 240: Move this paragraph to the description of Figure 2a. Now it is used only in the discussion of the unrealistically low λ value, but it is helpful for Fig 2a in general.

Line 251/52: Given the timescales for equilibration of the mole fraction and the isotope reservoirs (Tans, P. P.: A note on isotopic ratios and the global atmospheric methane budget, *Gl. Biogeochem. Cycles*, 11, 77-81, 1997), it seems highly unlikely to me

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that there could be a “stabilization” signal in the isotopic composition before the signal occurs in CH₄, at least when this is interpreted as a manifestation of steady state between sources and sinks. I suggest replacing “hiatus” by the explicit statement of intermittent stop in the annual growth.

Line 259: ... is useful ... For what? And on what basis do you make such a statement? The rest of the paragraph is quite vague, I wonder why you did not investigate the seasonal signal if you suggest that it should be a sensitive indicator for CI.

Line 277: As mentioned above, I suggest changing the line of argumentation away from the totally unrealistic values for λ . You can make the point stronger by staying with the possible λ range.

Line 288: This range of values in the realistic range for λ should be presented and discussed in the main text, see comments above. I consider this a (the?) main result, which is not well motivated and presented. Also, you should link it to the maximum possible CI difference between the two periods in the following paragraph, where the message is less clear because the parameter λ_a is involved again.

Line 289/90: This may be overly optimistic. What about source variations of CO (e.g. bb)? If it is mentioned in the conclusions it should follow from a more detailed discussion in the previous section, but it has not been mentioned before at all.

Line 294: What is λ_a (see comment above) and why does it come back here in the conclusions, whereas most of the discussion was about λ ?

Line 297: This last sentence is not really about your results, and already known, so although the statement is strong, it does not summarize your analysis. Also, one could argue that this implies that there is still at least one big error in the present understanding of the global CO isotope budget parameter, correction of which could offset the budget in a way that there may be room for CI again.

Technical points:

C4

Make “incomplete” comparisons complete. As it is now, one has to go back to the previous sentence to exactly locate the reference for the comparison.

Examples:

line 26: It is easier . . . compared to what? (direct measurements)

line 83: . . . are less complicated . . . compared to what? (the NH)

line 171: . . .much lower. . . compared to what?

Line 152: Rephrase sentence “significant or not. . .”. If a signal is not significant, don’t use it to support a scientific argument. Also, in this sentence you write about effects in both directions, in the next sentence you relate this to CO that would work in a “similar” direction (but here it is only one of them). This is confusing, please clarify.

Caption Fig 1 mentions pluses but they are not visible (probably the small dashes in the boxes where the vertical part of the cross coincides with the vertical line).

Line 172: Reword sentence: “The average fraction of the latter . . .” You write that two values are proportional, but one has a fixed value, so the other one as well.

Line 181: Reword “. . .can be approximated as due to. . .”

Line 211 and line 226: replace cf. *ibid.* by Fig 2.

Line 224: remove “happens to be”, this implicates that this is by chance.

Line 249 and 250: add “period” after HC and LC

Line 291: Why “Nevertheless”? The sentence does not seem to require this logical connection.

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