Atmospheric Chemistry and Physics Discussions

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Comment on acp-2021-622

Eric Saboya (Referee)

Referee comment on "Using carbon-14 and carbon-13 measurements for source attribution of atmospheric methane in the Athabasca oil sands region" by Regina Gonzalez Moguel et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-622-RC1, 2021

This research article demonstrates how combined measurements of carbon-13 and carbon-14 in atmospheric methane (CH_4) can be used to constrain the relative contributions of CH_4 emissions from sources in the Athabasca oil sands region in Canada. Isotopic measurements of carbon-13 and carbon-14 were made over a short field campaign and were used to infer the dominant CH_4 sources through Keeling plot analyses and back-trajectories of air-masses arriving at the measurement site.

The research article addresses an important and interesting question about how useful combined measurements of carbon-13 and carbon-14 are for attributing CH_4 sources. There have been rapid advancements in the instrumentation used for making precise atmospheric measurements of methane isotopes, particularly of carbon-14 in CH_4, which are becoming less labour intensive and easier to deploy (e.g. Zazzeri et al., 2021). In principle, the method presented is appropriate for addressing the aims of the research article. There are some specific points that I think should be addressed or clarified within the research article to help communicate your findings to the reader.

The HYSPLIT back-trajectories used were generated for a height of 50 metres above ground level (line 151). Back-trajectories at this height may not correspond to the back-trajectories at a height of 3 m, where measurements were made. The meteorology could be different between these heights and so will the back-trajectories. As the results rely heavily on the back-trajectories to evaluate the source origin, back-trajectories should be generated at a height that is representative of the measurements and the results re-evaluated.

It would be useful to have an indication of where the wetlands are in relation to the measurement site, as well as any landfills, and Fort McMurray (line 204), which is mentioned but not included on Figure 1.

It is unclear whether your results are consistent with the emissions inventories for the mining and tailings pond emissions. An explicit comment about this in your conclusion would be useful.

Emphasise the value of the carbon-14 measurements in your conclusions. Why is it important to use carbon-14 over, say, δD measurements with carbon-13?

The span of the sampling campaign is inconsistent in the manuscript. Line 96: "13-23 August" referring to the full campaign; Line 224 "20-24 August" referring to a subsample of measurements (end date different to line 96); Data presented in Table 1 and Figure 3 cover 16-23 August. You should check the length of the sampling campaign that covers the measurements presented is consistent throughout the paper to avoid confusion.

The calibration problem with the G2201-i isotopic analyser (line 278) should be moved from your results to the methods section. If the data is not presented nor reliable it should not be mentioned in the research article.

Could you please clarify what you mean by 'simple linear regression' (line 157) Is this, for example, an ordinary least square or orthogonal distance regression? It would also be useful to know whether the measurement errors (shown in Figure 3) are included when calculating the linear regression parameters and if these uncertainties are included in the uncertainties of the y-intercept and gradient?

On line 237 you state there is "one datapoint with a higher $\delta^{13}C$ than the background air". What is defined as the background air? Does this refer to clean-air measurements at another location?

It is stated differences between your study and Baray et al. (2018) is "mainly due to the uncertainty in the isotopic signatures of the CH_4 sources" (line 265). Are there any other main sources of uncertainty in your approach and the Baray et al. (2018) approach? Maybe consider the fetch of your measurements made at 3 metres? Is the 8-day sampling period representative of emissions in the AOSR?

On line 279, in this instance p-values do not describe whether a relationship is linear. Please remove.

Figure 3. Please include the appropriate numerical symbols for the y-intercept on panels A, B, and C. Please explain the meaning of the y-intercept on panel C in the caption and text. Figure 3C is also not mentioned in the text, you should refer to it somewhere.

The labelling of Table 1 and Table 2 in the manuscript do not correspond to how these tables are presented. Please check the labelling.

Table 2. Please include uncertainties for the concentration values. Please clarify if sample 11 was in fact made at 3:55 am (if so I admire your commitment) and that the $\Delta^{14}C$ value in sample 8 is that low.

I find it hard to interpret the spatial scale of Figure 2B. Adding a spatial scale bar or latitude-longitude values to the gridlines will make this clearer.

On line 155, I would alter "simple linear mixing" to "simple mixing", linear doesn't make sense to me in this context.

On line 23 (and throughout) what do mean by "10 ≤ 1 ‰"? This doesn't make sense mathematically.

On line 47, as per ACP guidelines latin phrases should not be in italics.

On line 82, Miller et al. (2020) is missing from your list of references.

Units are presented using inconsistent notation. Please stick to index notation (as per ACP guidelines), leaving a space between the value and the unit (or symbol) and between different units e.g. 5 kg m^{-2} s^{-1} not 5kg$m^{-2}s^{-1}$

On line 109, What is th^{-1}? Please clarify.

On line 217, you are missing the numerical symbol for value.

This research article was, on the whole, well-written and clear to follow. As mentioned, the method is appropriate, and the analysis is also well-thought out.