Dear Referee # 2,

Thank you for your time and contribution in reviewing this manuscript. Responses to general and specific comments are below.

Review of Characterization of atmospheric methane release in the outer Mackenzie River Delta from biogenic and thermogenic sources
Synopsis:

The authors present results of ground-based measurements of atmospheric methane concentrations and ^{13}C isotope signatures made in the Mackenzie River Delta, NWT, CN in 2019 and 2021 near locations which had anomalously high fluxes during airborne eddy covariance surveys in 2012 and 2013. They interpret their findings to mean that these previously determined "hotspot" areas were generated either by thermogenic CH_4 sources, biogenic CH_4 sources, or mixtures of both sources. They conclude that their study only provided a snapshot and that more robust methods are needed to more confidently confirm the origins of the CH_4 hotspots identified by the airborne surveys. The authors' dataset is indeed rare, is collected in a climate-sensitive environment, and could be valuable to the scientific community. However, their incomplete methods and lack of description regarding some of the assumptions they make about their sampling approach greatly limit the interpretation of their results. I believe this study will make an important contribution, but not before the authors fully articulate some critical shortcomings about their sampling and whether or not it can actually be used to interpret previously-discovered methane emission hotspots in the Mackenzie River Delta.

General Comments:
This study highlights the challenges of relating remotely sensed patterns to those which can be observed from the ground. A proper ground-truthing of remotely sensed signals requires careful consideration of the limitations of both approaches. Aside from a comment that it is impractical to sample the airborne-derived hotspots from the ground, there was little discussion regarding how representative their ground sampling was and whether it could be used to interpret the airborne-derived hotspots at all. The significant spatial disparity between the previous airborne survey and the present ground survey, the minimum spatial resolution at which the airborne survey can be confidently interpreted, and the limited description/presentation of the ground-based survey's methods all raise considerable uncertainty in the study's findings. At a minimum, this study should more explicitly (and quantitatively) describe their findings in space relative to the remotely- observed hotspots. For example, can the walking transect data be represented in figure 1?
Were samples collected up-wind or down-wind of the hotspots? ...etc.
How representative are their discrete samples of the airborne-observed hotspots to which they attempt to attribute their findings? From Figure 1, one can see that the airborne hotspots are several kilometers across. Are the authors attempting to use singular samples from discrete locations to describe CH_4 sources from kilometers-wide "hotspots?" The ground-based sampling methods should further discuss the assumptions, strategy, and potential biases involved in their plan. For example, was sampling meant to describe individual features (e.g.

pingos) on the ground or the large-area hotspots more generally? How do the authors reconcile these scale differences? If they intended to apply isotopic signatures and flux rates to individual features on the ground, why not use chambers or other methods to explicitly isolate the air from those sources? If they intended on sampling the air to infer something about the surrounding landscape, why not conduct a more formal wind analysis or "footprint" determination and then describe the features within the footprint? It seems that the approaches fall somewhere in between these two objectives, but as a result, fail to robustly describe the environment in which they were made.
The authors have not clearly articulated their assumptions regarding separating background air from the air that contains mixtures of CH_4 originating from their hotspots. Can their methods sufficiently distinguish between the two? How do they know that the background air wasn't already a mixture of other local sources? How do variable wind conditions (velocity **and direction**) affect their sampling approach? There is little description regarding the direction of the wind and how this could affect their goal to interpret the hotspots.
No reference is made to Figure S1 or Figure S2 in the manuscript text. This was surprising, especially since Figure S2 contains the walking transect data.

There are several biases outlined here which are not effectively addressed in the manuscript. Formal wind analysis and flux chamber measurements would improve the quality of this data but we were unable to collect these measurements. We attempted to select methods which would help to characterize these large hotspots with limited space, weight and time on the ground due to the remoteness of these sites. We believe the following changes, along with the changes made to specific comments will highlight and address these biases.

Samples were collected upwind and downwind of any features so that we would be certain to pick up background and peak concentrations if the feature were a source. Wind speed and direction is shown of Figure S2, which can be moved to the main manuscript. We have refrained from trying to make correlations with wind speed because we do not have high-rate wind measurements. The following could be added to the end of the first paragraph in the sample collection section of the methods (3.2, line 165):

"Walking transect locations were selected by completing one transect up wind and one down wind of the estimated location of the highest flux concentration, in order to obtain background concentrations at each site. Discrete point samples were taken parallel to each walking transect, 3-5 m further away from the center of the hotspot. This sampling method was designed to cover the most area, with the fewest samples, in the shortest amount of time possible. By using this method we were still only able to cover a small fraction of the hotspot (5-10% by area), so samples were collected around potential sources of CH_4, such as pingos or wetlands. This increased the likelihood of pinpointing the source."

The description of the walking transects can be rewritten as follows:

"Walking transects were carried out by filling a 30 m coil of 4 mm inside diameter aluminium Synflex tubing while walking at a steady pace across the ground. A constant flow rate of 20 standard cubic centimetres per minute (CCM) was maintained by attaching a small pump and a flow controller to the coil of tubing. Samples were analyzed using a Picarro G2210i analyser immediately on return from the field site. Five walking transects using Synflex tubing took approximately 20 minutes each to fill and covered a distance between 600-800 m. Methane and CO_2 concentrations were measured every 1-2 seconds on the air samples collected in aluminium tubing. This allowed for consideration of spatial variability in methane concentrations at each site. Mixing of the air sample inside the tube between collection and analysis is limited due to the small diameter of the tubing. A similar method was used during drone-based CH_4 measurements (Andersen et al., 2018)."

It would be quite difficult to add more information to Figure 1 and still effectively convey it to the reader. Figure S2 can be moved to the main manuscript and the raw data from the walking transects can be added to the supplement.

Specific Comments:

Line 39: A more recent/updated reference is available for the global CH_4 budget. Saunois, M., Stavert, A.R., Poulter, B., Bousquet, P., Canadell, J.G., Jackson, R.B., Raymond, P.A., Dlugokencky, E.J., Houweling, S., Patra, P.K. and Ciais, P., 2020. The global methane budget 2000–2017. Earth system science data, 12(3), pp.1561-1623.

Reply: "Current global estimates of wetland CH_4 flux to the atmosphere range between 153-227 Tg CH_4/yr (Saunois et al., 2016). Non-wetland, freshwater systems are also significant contributors of CH_4 to the atmosphere on a global scale (Kirschke et al., 2013) which is estimated between 60-180 Tg CH_4/yr (Saunois et al., 2016)."

Can be changed to:

"Current global estimates of wetland CH_4 flux to the atmosphere range between 101-179 Tg CH_4/yr (Saunois et al., 2020). Non-wetland, freshwater systems are also significant contributors of CH_4 to the atmosphere on a global scale (Kirschke et al., 2013) which is estimated between 117-212 Tg CH_4/yr (Saunois et al., 2020)."

Line 76: Without context, a "hotspot" can have an ambiguous definition. Since 5 mg CH_4 $m^{-2}d^{-1}$ is not a particularly high flux in the context of typical wetland/lake emissions, I think that the authors need to emphasize on what spatial scale this would be considered a high flux or a hotspot. This would also help set the stage for placing the new observations in the proper spatial context.

Reply: This sentence can be rewritten to improve context. Please note that the flux rate threshold used to define geologic emission in Kohnert et al (2017) and for inclusion into this study was 5 mg CH_4 $m^{-2}h^{-1}$, not 5 mg CH_4 $m^{-2}d^{-1}$.

"These authors assumed that these hotspots were primarily of geologic origin (CH_4 produced beneath the permafrost, including thermogenic CH_4 from natural gas reserves that has the potential to migrate through discontinuities in the permafrost) since the inferred flux rates of the hotspots identified were significantly greater than the maximum values of around 4 mg CH_4 $m^{-2}h^{-1}$ detected for biogenic fluxes north of 61°N (Friborg et al., 2000; Sachs et al., 2008; Sturtevant et al., 2012). These fluxes also occurred in the summer period when most lakes were fully oxygenated, reducing biogenic emissions."

Lines 81 - 86: Check verb tenses in this paragraph. It currently reads like this work has not happened yet- as one would write in a proposal.
Reply: The paragraph will be changed to past tense.

Line 102: Bowen et al. (2008) is plural, so probably change "has" to "have" Lines 119 - 121: Results should be moved to the results section.
Reply: "Has" can be changed to "have" and lines 119-121 can be moved to the results section.

Line 125: What does "focused" mean here? It's also misspelled. This sentence could use a rewrite to improve clarity.
Reply: This sentence can be rewritten as follows:

"Four additional sites (Pingo 3, Wetland 1, Lake 1, Channel Seep) were at locations where point source, aquatic seeps where concentrated ebullitions of CH_4 flux were seen in open water in the summer, or in holes in newly formed ice in the fall. Channel Seep and Lake 1 were previously known to researchers in the field party, while Pingo 3 and Wetland 1 sites were identified by holes in the ice which were observed from the helicopter while passing overhead during the fall. Discrete point samples were taken for stable carbon isotope ratio ($\delta^{13}C$-CH_4) analysis."

Line 155: Could sampling only in wetlands bias the observations towards one emission pathway (or source) over another? Do thermogenic emissions only occur in wetlands and/or lakes? Or can they occur in dry areas as well? Some discussion and communication of assumptions around this sampling strategy are warranted (see general comment).
Reply: While this was the best method to cover very large sites that were accessible only by helicopter. We agree that this sampling method could bias the results toward the obvious features which we sampled. The following can be added to clarify:

"Sampling transect locations were selected within the general hotspot area where the airborne eddy covariance fluxes were highest. Samples were concentrated around features such as wetland areas or pingos that were considered possible sources of CH_4. Pingos and wetlands can be sources of both biogenic and thermogenic CH_4 but in dry areas biogenic emissions are far less likely than thermogenic emissions. Unfortunately, this has the potential to bias sampling to areas where biogenic emissions are more likely."

Lines 159 – 164: Is this an “air core?” or something else? This description is somewhat vague. It raises several questions. What is the objective of the “walking transect?” How was the tubing filled? What are “analytical determinations?” Mixing of what?
Reply: Yes, these are essentially air cores, these samples were intended to give a continuous transect of CH_4 and CO_2 concentrations. The description of the methods can be rewritten as follows in order to clarify:

“Walking transects were carried out by filling a 30 m coil of 4 mm inside diameter aluminium Synflex tubing while walking at a steady pace across the ground. A constant flow rate of 20 standard cubic centimetres per minute (CCM) was maintained by attaching a small pump and a flow controller to the coil of tubing. Samples were analyzed using a Picarro G2210i analyser immediately on return from the field site. Five walking transects using Synflex tubing took approximately 20 minutes each to fill and covered a distance between 600-800 m. Concentrations of CH_4 and CO_2 were made every 1-2 seconds on the air samples collected in aluminium tubing which allowed for consideration of spatial variability in methane concentrations at each site. Mixing of the air sample inside the tube between collection and analysis is limited due to the small diameter of the tubing. A similar method was used during drone-based CH_4 measurements (Andersen et al., 2018).”

Line 168: The aerial EC hotspot sites? Or on walking transects, or both? Be more specific about the spatial representativeness of the observations.
Reply: The following could be added to the end of the first paragraph in the sample collection section of the methods (3.2, line 165): “Walking transect location was selected by completing one transect up wind and one down wind of the estimated location of the highest flux concentration, in order to obtain background concentrations at each site. Discrete point samples were taken parallel to each walking transect, 3-5 m further away from the center of the hotspot. This sampling method was designed to cover the most area, with the fewest samples, in the shortest amount of time possible. By using this method we were still only able to cover a small fraction of the hotspot (5-10% by area), so samples were collected around potential sources of CH_4, such as pingos or wetlands. This increased the likelihood of pinpointing the source.”

Line 175: How were fluxes derived from chamber data? How many vials were analyzed per flux observation? For how long? Linear extrapolation? Was non-linearity observed? If so, how was this handled in data QA/QC? These parameters are typically reported alongside chamber flux data.
Reply: We had originally included the flux chamber measurements because they have considerable value. There is a lack of published chamber measurements in the Mackenzie River Delta as well as the Canadian Arctic. The chamber flux data will be removed from the manuscript since it does not add significantly and distracts from the main narrative presented in this manuscript.

Line 221: What is “reasonable confidence?” Can you be more quantitative? Correlation statistic? Otherwise, this is not very informative.
Reply: The sentence on line 221 can be changed to the following:

“Estimates of source stable carbon isotope signatures ($\delta^{13}C\text{-}CH_4$) derived from Keeling plots were -53.0 (+/- 1.01) ‰ for Pingo 1 and -63.6 (+/- 1.87) ‰ for Pingo 2.”

Line 222: Phrases like “substantially enriched” and “relatively close” are not very informative and weaken the impact of the claims being made. Please quantify where possible.
Reply: The sentence on line 222 can be changed to the following:

“These signatures are enriched in ^{13}C compared to typical biogenic sources with $\delta^{13}C\text{-}CH_4 < -70$ ‰ and within 14 ‰ of our assumed threshold of -50 ‰ based on thermogenic gas found in the Taglu hydrocarbon reservoir.”

Line 266: How was “background” determined?
Reply: Site 9 data can be added to Table 1. Background was determined to be 1.995 ppm CH_4, which was the average value recorded at the ECCC weather station at Inuvik during the time of the fieldwork.

“No elevated atmospheric methane values were detected during walking transects at Site 9” can be changed to:

“Atmospheric CH_4 values during walking transects at Site 9 peaked at 2.044 ppm,”

Cited Literature:

Andersen, T., Scheeren, B., Peters, W., & Chen, H. (2018). A UAV-based active AirCore system for measurements of greenhouse gases. *Atmospheric Measurement Techniques*, *11*(5), 2683–2699. https://doi.org/10.5194/amt-11-2683-2018

Friborg, T., Christensen, T. R., Hansen, B. U., Nordstroem, C., & Soegaard, H. (2000). Trace gas exchange in a high-Arctic valley: 2. Landscape CH_4 fluxes measured and modeled using eddy correlation data. *Global Biogeochemical Cycles*, *14*(3), 715–723. https://doi.org/10.1029/1999GB001136

Kirschke, S., Bousquet, P., Ciais, P., Saunois, M., Canadell, J. G., Dlugokencky, E. J., Bergamaschi, P., Bergmann, D., Blake, D. R., Bruhwiler, L., Cameron-Smith, P., Castaldi, S., Chevallier, F., Feng, L., Fraser, A., Heimann, M., Hodson, E. L., Houweling, S., Josse, B., … Zeng, G. (2013). Three decades of global methane sources and sinks. *Nature Geoscience*, *6*(10), Article 10. https://doi.org/10.1038/ngeo1955

Sachs, T., Wille, C., Boike, J., & Kutzbach, L. (2008). Environmental controls on ecosystem-scale CH_4 emission from polygonal tundra in the Lena River Delta, Siberia. *Journal of Geophysical Research*, *113*, G00A03. https://doi.org/10.1029/2007JG000505

Saunois, M., Bousquet, P., Poulter, B., Peregon, A., Ciais, P., Canadell, J. G., Dlugokencky, E. J., Etiope, G., Bastviken, D., Houweling, S., Janssens-Maenhout, G., Tubiello, F. N., Castaldi, S., Jackson, R. B., Alexe, M., Arora, V. K., Beerling, D. J., Bergamaschi, P., Blake, D. R., … Zhu, Q. (2016). The global methane budget 2000–2012. *Earth System Science Data*, *8*(2), Article 2. https://doi.org/10.5194/essd-8-697-2016

Saunois, M., Stavert, A. R., Poulter, B., Bousquet, P., Canadell, J. G., Jackson, R. B., Raymond, P. A., Dlugokencky, E. J., Houweling, S., Patra, P. K., Ciais, P., Arora, V. K., Bastviken, D., Bergamaschi, P., Blake, D. R., Brailsford, G., Bruhwiler, L., Carlson, K. M., Carrol, M., … Zhuang, Q. (2020). The Global Methane Budget 2000–2017. *Earth System Science Data*, *12*(3), 1561–1623. https://doi.org/10.5194/essd-12-1561-2020

Sturtevant, C. S., Oechel, W. C., Zona, D., Kim, Y., & Emerson, C. E. (2012). Soil moisture control over autumn season methane flux, Arctic Coastal Plain of Alaska. *Biogeosciences*, *9*(4), 1423–1440. https://doi.org/10.5194/bg-9-1423-2012