Anonymous review # 1 comments and reply.

Reviewer # 1,

Thank you for your time and contribution in reviewing this manuscript. All general and main text comments have been updated in the manuscript text as recommended.

Review of 2022-549: “Characterization of atmospheric methane release in the outer Mackenzie River Delta from biogenic and thermogenic sources.” by Wesley et al.

Wesley et al. have studied hotspot of methane from the Mackenzie Delta, measuring stable isotopes from atmospheric methane above know aquatic and terrestrial hotspot of methane. Their δ^{13}C-CH4 signatures indicate that both biogenic and thermogenic sources are found in the delta.

I find that study very interesting as methane hotspots are rarely characterized, especially I appreciate the effort of verifying data obtained by airborne eddy covariance. Although the airborne study has been realized in 2013 and the discrete sampling in 2019. In this study they characterized very few hotspots (8) and it would be worthwhile to continue this study and add more data to be able to derive a regional pattern. I recommend for publication with very minor correction.

General comments:

Lake 1 is also known under the name “Swiss Cheese lake”, it would be useful for the reader not familiar with this area to mention that in the supplement (for example in table S1).

Main text:

L168-170: The sentence starting with “Sampling transect locations...”is repeated L231: There is a “-“ missing before “53”

Reviewer Nicolas R. Hasson comments and reply:

Dr. Hasson,

Thank you for your time and thorough review of this manuscript. The authors agree that the incorporation of recent (2021- present) literature will strengthen the support for our findings and have decided to incorporate the majority of your suggestions.

Review:

"Characterization of atmospheric methane release in the outer Mackenzie River Delta from biogenic and thermogenic sources" (egosphere-2022-549) Submitted on 26 Jun 2022.

Reviewer:

Nicholas R. Hasson (NRH), University of Alaska Fairbanks (no-competing conflicts)

Assessment:

The study provides significant insight into the findings of Kohnert et al. (2017), which have spurred a multitude of subsequent airborne and ground measurements across the Arctic, a practice that continues to this day. The results suggest a variety of methane production sources in the MRD, with source isotope signatures ranging from -42 ‰ (biogenic) to -88 ‰ (thermogenic), range values indicates that methane in the MRD is likely being produced by both biogenic and thermogenic sources and suggest some mixing, strongly linked to seasonality. This is perfectly in line with what we know about methane production and the interpretation, which seems scientifically valid, does add important reference results to the limited data from airborne/ground coupled surveys on methane hotspot investigations.

I would like the authors to consider recent work (post-Kohnert, 2017) on characterizing hotspots in MRD has provided significant results (e.g. Elder et al., 2020, 2021; Baskaren et al. 2022). Please consider these more-recent updated references on magnitude and occurrence of methane hotspots in MRD, and how this enhances, supports, or disagrees with your findings. Additionally, consider thermogenic and biogenic isotopic signatures from recent similar work (Kleber et al., 2023; Sullivan et al., 2021; Elder et al., 2018), which may support or not the interpretation concluded here. It may be important to briefly mention how so.

However, these results significantly add to import investigation on the discussion on source attribution (e.g. biogenic vs. thermogenic) sources of methane hotspots, particularly important for airborne validation and coupling of ground truth observation, yet data has remained limited. Therefore, this warrant publications with minor technical adjustments, such as the inclusion of additional supporting reference material that supports or challenges these results and/or provides valuable recent observations (post-Kohnert, 2017) that alludes to the behavior of hotspots in MRD. The comments should be viewed as recommended suggestions for these reference materials, and the authors have the discretion to merely incorporate the reference material without my proposed text modifications. However, I have provided some examples of how the text could be altered to either support or challenge interpretations, which may further enrich these crucial ground truths for ongoing campaigns carrying out similar observations.

In summary, these findings are important and significant results for future endeavors focusing on airborne/ground verification of methane hotspots in the Arctic.

Review Summary:

The authors present novel data and likely is the first to measure stable carbon isotope signatures of atmospheric methane at hotspots in the Mackenzie River Delta (MRD). The results suggest a variety of methane production sources in the MRD, with source isotope signatures ranging from - 42 ‰ (biogenic) to -88 ‰ (thermogenic). Of the eight sites investigated, two had a thermogenic origin, four were biogenic, and two were possibly a result of oxidation of mixed biogenic/thermogenic sources. This is supported by other recent studies.

These results also suggest methane migration from below the thin permafrost at most sites, including from the Taglu gas field, over an area of approximately 20 km north to south, which suggest complex permafrost distribution (e.g. due to the hydrology of river taliks, pingo- systems, coastal settings, and together a mix of likely through-taliks and permafrost degradation). The study was able to validate airborne eddy covariance hotspot locations using walking transects to measure atmospheric methane variation. However, authors point out these methods only provide a snapshot of methane sources during site visits, and a comprehensive understanding of annual methane production is yet to be established.

Authors suggest that future research should include year-round flux measurements and stable carbon isotope measurements to fully quantify the annual methane emission from both biogenic and thermogenic sources. Additionally, combining portable methane analyzers with flux chamber and isotopic measurements could help to better identify and quantify sources, particularly at sites where both biogenic and thermogenic sources are likely. Geophysical mapping atop these transects would provide a useful coupling to permafrost distribution and potential source attribution between biogenic and thermogenic sources.

I highly recommend adding some of the added reference support material **(#1-13)** and considering comments #1-23. Particularly important, **reference material post-Kohnert et al. results from MRD, e.g., Elder et al. (2020, 2021) and Baskaren et al. (2022).** I would like the **authors to consider (Kleber et al., 2023; Sullivan et al., 2021; Elder et al., 2018)**, which may serve to challenge or support these results, and it may be important briefly mentioning how so.

I hope these added suggestions and reference materials can serve to enhance your otherwise significant results. Nice work.

Review by section:

1 Introduction - Please consider comments/suggestions **#1-11**

#1NRH: please consider additional reference supports (A,B), which updates the magnitudes of methane hotspots (including from MRD) and the spatial/temporal distribution, which have been found to follow a power law series as a function of distance to stand water; arctic hotspot methane law (e.g. <40 m from wetland boundary).

#2NRH: Furthermore, extreme hotspots have been shown to range from **48 mg to 1008 mg CH_4 m^{-2} hr^{-1}**, including from MRD region. However, I mentioned from a recent meta analysis on terrestrial sources of methane (e.g. >60 N)(Kuhn et al, 2021) shows the roughly average yearly CH_4 emission from terrestrial permafrost areas to be 2.22 mg m^{-2} hr^{-1}. Therefore, your reported {4-5 mg m^{-2} h^{-1} values} **are considered high or roughly double the average hourly mean.**

#3NRH**: I would label extreme hotspots,** including from MRD region, to include the sources found more recently (post-Kohnert study) by Elder et al. (2020,2021) and Baskaren et al. (2022); the work here highlights the importance of your work and significant of the Kohnert et al. follow up (e.g. isotopic fingerprinting associated with hotspots), but currently, lacks these updates references. I've organized potential text below:

A. Additional references for spatial and topographic methane hotspots in MRD

#4NRH: I highly recommend adding recent reference support regarding methane hotspot detection in Mackenzie Delta Region (MRD). I've added a potential way to include citation in the text (line 91-95)

Suggested text inclusion
"In the MRD, arctic CH4 hotspots have been found to exhibit a power law relationship with the distance to the nearest standing water (Elder et al., 2020). The geomorphic factors controlling CH4 hotspots in the MRD reveal a spatial decay in the correlation between distance to water and land cover or vegetation type (Baskaran et al., 2022).

Suggested supporting reference #1:
Elder, C. D., Thompson, D. R., Thorpe, A. K., Hanke, P., Walter Anthony, K. M., & Miller, C. E. (2020). Airborne mapping reveals emergent power law of Arctic methane emissions. Geophysical Research Letters, 47, e2019GL085707. https://doi.org/10.1029/2019GL085707

Suggested supporting reference #2:
Baskaran, Latha & Elder, Clayton & Bloom, A. & Ma, Shuang & Thompson, David & Miller, Charles. (2022). Geomorphological patterns of remotely sensed methane hot spots in the Mackenzie Delta, Canada. Environmental Research Letters. 17. https://dx.doi.org/10.1088/1748-9326/ac41fb

Response to Comment 1- 4

The following text has been added to line 45:

"In the MRD, Arctic CH_4 the frequency of CH_4 hotspots decreases exponentially as distance to standing water increases (Elder et al., 2020; Baskaran et al., 2022)."

The following text has been added at line 78:

"Elder et al. (2021) observed diffusive flux averaging 48.75 mg CH_4 m^{-2} hr^{-1} and peaking at 1,008 mg CH_4 m^{-2} hr^{-1} directly over thawed permafrost on the edge of a thermokarst lake in interior Alaska."

The following Text has been added to the paragraph from lines 94:

"According to a recent meta-analysis, the cut off value of 5 mg CH4 m^{-2} hr^{-1} used by (Kohnert et al., 2017) is approximately double the mean flux rate for Arctic and boreal regions (Kuhn et al., 2021)".

B. Additional references for methane hotspot magnitudes

#5NRH**:**: Please add updated reference to the context of “extreme methane hotspots” equating to {4-5 mg m^{-2} h^{-1}}. For example, Elder et al. (2021) reported “Ground-based chamber measurements confirmed average daily CH4 fluxes of 1,170 mg m^{-2} d^{-1}, with extreme daily maxima up to 24,200 mg CH4 m^{-2} d^{-1}”. Converting to hourly, this equates to **48 mg to 1008 mg m^{-2} hr^{-1}**. **Which are considered extreme.**

Suggested supporting reference #3:
Elder, C. D., Thompson, D. R., Thorpe, A. K., Chandanpurkar, H. A., Hanke, P. J., Hasson, N., et al.(2021). Characterizing methane emission hotspots from thawing permafrost. Global Biogeochemical Cycles, 35, e2020GB006922. https://doi.org/10.1029/2020GB006922

#6NRH**:** line 94-96 from your text, I recommend adding more recent hotspot values, e.g.,...

... “hotspots identified {were high, when considering the} maximum published value of around 4 mg CH4 m^{-2} hr^{-1} detected for biogenic fluxes north of 61°N (Friborg et al., 2000; Sturtevant et al., 2012; Sachs et al., 2008).

#7NRH**:** <additionally> ...{However, recent hotspot observations from >60°N show more extreme hotspots ranging from **48 mg to 1008 mg CH4 m^{-2} hr^{-1}** , including from the MRD (Elder et al., 2020; 2021; Baskaran et al. 2022)}...<additionally>...{and recent meta analysis shows the **yearly daily average of terrestrial hotspots to range 5.04 to 29.3 mg CH4 m^{-2} h^{-1}**, when considering other emission inventories >4 mg **CH4 m^{-2} h^{-1}**. }. Futhermore, from a more holistic reference gathering, I made a rough calculation of the terrestrial database of methane emissions (Kuhn et al., 2021), including 105 previous methane flux investigations, I calculated the average CH4 emission to be 53.3 mg m^{-2} d^{-1} (range: 705 to -9.8; mg m^{-2} d^{-1} ;N=545). When converting to hourly, the average yearly CH4 emission from terrestrial permafrost areas showed 2.22 mg m^{-2} hr^{-1}.

#8NRH**:** Therefore, your reported {4-5 mg m^{-2} h^{-1} values} **are considered high or roughly double the average hourly mean.** However, under the context of eddy covariance open-air mixing values, which are very high in this context (e.g. versus chamber-derived emissions).

#9NRH**:** I would mentioned this. >Values can be higher, with recently reported the yearly daily average of 705 to 121 mg m^{-2} d^{-1} or ranging 5.04 to 29.3 mg m^{-2} h^{-1}. Together with the >60°N extreme hotspots ranging from **48 mg to 1008 mg CH4 m^{-2} hr^{-1}** , including from the MRD (Elder et al., 2020; 2021; Baskaran et al. 2022)}

Suggested supporting reference #4:

Elder, C. D., Thompson, D. R., Thorpe, A. K., Chandanpurkar, H. A., Hanke, P. J., Hasson, N., et al. (2021). Characterizing methane emission hotspots from thawing permafrost. *Global Biogeochemical Cycles*, 35, e2020GB006922. https://doi.org/10.1029/2020GB006922

Suggested supporting reference #5:

McKenzie Kuhn, Ruth Varner, David Bastviken, Patrick Crill, Sally MacIntyre, et al. 2021. BAWLD-CH4: Methane Fluxes from Boreal and Arctic Ecosystems. Arctic Data Center doi:10.18739/A2DN3ZX1R.

#10NRH**:** Note, the isotopic composition of CH4 hotspots characterized in Elder et al. (2021), was later investigated and showed biogenic origin: "**CH4 hotspot revealed a ^{14}C age of 35,360 YBP (δ^{13}C -73.8 ‰)**(Hasson et al. 2022)." This supports your biogenic source attributions of hotspots detected by airborne observations, although from Alaska.

Response to Comment 5 -10

These are very interesting results by Hasson et al (2022) and Elder et al (2021) that do indeed support the results in this study, they should be mentioned here and further included in the discussion.

The following has been added to line 95:

"More recent work has shown that exceptionally high flux rates averaging 48.75 mg CH_4 m^{-2} hr^{-1} can be attributed to biogenic production, with a stable carbon isotope signature of -73.8 ‰ (Elder et al., 2021; Hasson, 2022).

Suggested supporting reference#6

Hasson, N., et al. (2022). Methane emissions show exponential inverse relationship with electrical resistivity from discontinuous permafrost wetlands in Alaska. AGU Fall Meeting Abstracts. AAGUFM.B15E..06H https://ui.adsabs.harvard.edu/abs/2022AGUFM.B15E..06H

#11NRH**:** Note, it may also be useful to discuss the Elder et al. (2021) results, which goes on to upscale these observations from arctic and pan-arctic hotspot detection, including MRD:

e.g., "Emissions from the hotspot accounted for ~40% of total diffusive CH4 emissions from the entire study area. Combining these results with hotspot statistics from our 70,000 km^{-2} airborne survey across Alaska and northwestern Canada (e.g. MRD), we estimate that terrestrial thermokarst hotspots currently emit 1.1 (0.1 – 5.2) Tg CH4 yr-1, or roughly 4% of the annual pan- Arctic wetland budget from just 0.01% of the northern permafrost land area." This support MRD hotspot signficance.

Response to comment #11

The following has been added to line 80:

"Terrestrial thermokarst hotspots are estimated to account for roughly 4% of the pan-Arctic CH_4 budget but make up only 0.01% of the northern permafrost land area (Elder et al., 2021)"

No further comments.

Section 2: Setting - Please consider adding reference material for context/support **#12-13**

#12NRH: I recommend the additional references to support your setting, e.g., at line #116-118:

< “Permafrost is generally seen as being continuous under land areas in MRD, but it is mostly missing under lakes that don't freeze all the way to the bottom during winter (Nguyen et al., 2009).” <adding further support context> “Additionally, it has been demonstrated in permafrost regions >60 N, **river taliks extend beyond the river plane by connecting through-taliks with nearby wetlands** (Minsley et al., 2012; see Figs 4,5), showing how “discontinuous” permafrost conditions could be present adjacent to MRD tributaries”>

#13NRH: **why not include** something like <"The airborne geophysical data detailed in Minsley et al. (2012), illustrates the complexity of river through-taliks and hydrology (e.g. Yukon river) which may mimic areas like the Mackenzie River Delta (MDR). This complexity leads to the presence of discontinuous permafrost near rivers, indicating that the MDR might be fragmented near river tributaries, allowing gas-conduits to form along hydrological through-taliks. These intricate river taliks have the potential to connect with lakes, as observed in the Yukon and Noatak rivers in Alaska, and have been identified in close proximity to some of the largest geological seep sources (Sullivan, 2021). Recent studies have further uncovered that methane-rich groundwater springs are transporting deep thermogenic and biogenic methane gas, such as in Svalbard, north of 79°N latitude. This gas reaches the surface and is found to be supersaturated with methane at levels up to 600,000 times greater than what would be expected for equilibrium with the atmosphere (Klebar et al., 2023).">...

- For example, river talik networks can fragment “continuous permafrost” into “discontinuous permafrost” by through-taliks near large watersheds (e.g. MDR), which may help support why hotspots of deeper thermogenic emissions would occur here in “continuous zone permafrost”.

- For example, Sullivan et al. shows the river talik is at work here. and is along the Noatak river, suggesting “microbially produced fossil CH4 is being vented though a narrow thaw conduit below Esieh Lake through pockmarks on the lake bottom. This is one of the highest flux geologic CH4 seep fields known in the terrestrial environment and potentially the highest flux single methane seep”,

- For example: recent work (2023) highlights the context of your investigation on thermogenic sources in settings section, e.g., : “

Response to comments 12 and 13

The following has been added to line 114:

“This landscape is a prime location for the formation of Arctic river taliks (Ensom et al., 2012) which can be sources of high-rate geologic CH_4 seeps (Sullivan et al., 2021). These river taliks can also form connecting through taliks with nearby wetlands which can create a network of discontinuous permafrost (Minsley et al., 2012).”

Suggested supporting reference #7-9

Minsley, B. J., et al. (2012), Airborne electromagnetic imaging of discontinuous permafrost, Geophys. Res. Lett., 39, L02503, doi:10.1029/2011GL050079.

Sullivan, TD, Parsekian, AD, Sharp, J, et al. Influence of permafrost thaw on an extreme geologic methane seep. Permafrost and Periglac Process. 2021; 32: 484–502. https://doi.org/10.1002/ppp.2114

Kleber, G.E., Hodson, A.J., Magerl, L. et al. Groundwater springs formed during glacial retreat are a large source of methane in the high Arctic. Nat. Geosci. 16, 597–604 (2023). https://doi.org/10.1038/s41561-023-01210-6

No further comments.

Section 2: Study Location - Please consider reference support **#14**

#14NRH :Line 175 reference suggestion (context)...e.g., **context for the geophysical extent of through-taliks near sites (1) channel seep, Pingo 2, Pingo 1, and Site 9...**

>"However, nearby reports along the arctic coastal shelf have indicated the lack of ice-bonded permafrost beneath the coastline, implying the existence of extensive through-taliks that intrude sea-to-land. These through-taliks are potentially connecting subpermafrost aquifers on land, alluding to the poorly understood and highlight complex dynamics of coastal subsurface hydrology."> impacting potentially (1) channel seep, Pingo 2, Pingo 1, and Site 9...

<u>Suggested supporting reference #10</u>
Micaela N. Pedrazas et al., Absence of ice-bonded permafrost beneath an Arctic lagoon revealed by electrical geophysics.Sci. Adv.6,eabb5083(2020).DOI:10.1126/sciadv.abb5083

Response to comment 13:

The authors do not believe that this reference is good support for through taliks at the near shore sites.

No further comments.

Section 3: Methods, Sample collection and analysis - comments/suggestions **#14-15**

Please consider added reference to warrant justification of sampling protocols on hotspots, which support authors approach, which does support initial hypothesis (e.g. distance from standing water=larger source attribution target).

NRH: The authors used similar method is based on protocols by Andersen et al., 2018. Authors point out that this method are only able to recover a small fraction of the hotspot (e.g. 5-10% by area). Note, as previous aforementioned studies allude to (e.g. Elder et al., 2021, Hasson et al., 2022), even though hotspots can dominate the local diffusive CH4 budget (e.g. 90% total), these arise from only a tiny fraction of the area (e.g. 15% of observations), suggesting that although Kohnert et al. observations show large coverage (e.g. 1 km2), these hotspots may come from discrete areas (e.g. 150-200 m transects), suggesting this study may have captured these disproportionally large sources from a disproportionally small area, similar to Elder et al. 2021

#14NRH: The importance of these sampling protocols rest on the hypothesis that the sample locations chosen (e.g. near wetlands and pingos, etc.) are the source of these hotspots. **This is not necessarily unwarranted bias,** since prior work has shown that hotspots follow a power law series from distance to standing water (e.g. 0-100 m)(e.g. Elder et al. 2020, 2021; Baskaran et al. (2022)). Its interesting that the hotspots from pingos are near the lowlands (e.g. supporting Baskaran et al., analysis of topographical controls). Given the considerable expensive of field work by helicopter and challenging environment, which limits sampling observational time, future work may also consider transects <100 m from wetlands or areas with complex through-taliks and complex permafrost hydrology (e.g. Pingos, tributaries, coastline).

The authors may wish to highlight this fact for MRD hotspots to minimize effects of bias or rational for the interpretation of data.

Therefore, if sampling protocols reference the power law findings by Elder et al., 2020,Baskaran et al. 2022, the bias near wetlands has a justified reason. Furthermore, discrete changes from upland to lowlands (escarpment bluffs, river terraces, lake terraces, etc.) have been shown to be statistically related to high occurrence of MRD hotspots (Baskaran et al. (2022)). **The authors sampling protocol near distance to standing water or topographic changes near targets is a good approach.**

Response to Comment 14:

The authors agree that this is not an unwarranted bias but would like to be clear to the reader that they had to make a choice when sampling that could have biased the results to sources from wetlands and pingos.

#15NRH: Suggestion, it might be advantages to show the sampling transects as a function of distance to standing water (e.g. Channel seep was ~50 m from standing water, etc.). Does this spatial relationship tell us anything more about the data (e.g. isotopic signature, concentrations?). Perhaps the authors can mention any relationship that is found, e.g. similar to the pingo lowland having higher concentration than pingo upland, so future studies can plan sampling around these known spatial relationships between geomorphology and water table position.

Response to Comment 15

There was a relationship as the 2 definitive thermogenic sites are both aquatic seeps, increasing the possibility of through taliks. The authors believe this is adequately addressed in the discussion.

No additional comments on methods here.

Section 3: Methods, Determination of CH4 source stable isotope value **- #16-17**

NRH: Authors justify mass balance approach, considering limitations and assumptions. In this context, sampling minimized bias by sampling upwind and downwind of the estimated locations, along 600-800 m transects, which provided a large spatial and temporal range estimate of source attributions. Wind regime was observed using handled anemometer with accuracy of +/- 0.1 m/s. From the paragraph (L239-256), the authors acknowledge the limitations of the Keeling plot analysis under certain field conditions. They accept that the method's assumption of only two components being measured (the source released at the surface/atmosphere interface and the background regional atmospheric signature) can be challenging in broad areas or windy conditions that may cause mixing.

Given these constraints, its acceptable to assume these limitations when applying the Keeling plot analysis, which depend on the specific context of the hypothesize: the authors clarify earlier that hotspots may form from discrete areas near wetlands and pingos. It seems than the significance of the data is determined by the wind direction from source target (wind direction from or away source target). The authors have acknowledged the limitations and adapted their methodology, accordingly, attempting to collect samples as close to the known point source of emissions as possible when observable ebullition was present.

Their acceptance of the limitation when appraising large hotspots, based on their assumption that the atmospheric point samples taken within these hotspot source regions could represent a mixed δ13C-CH4 signature, seems a reasonable approach given the constraints of field conditions and the necessity to identify broad trends.

#16NRH: I recommend complementary rose wind plots that show the wind direction versus source target over time, and how that may contribute to the interpretations of results. Showing wind rose plots (over time, e.g. 30 minutes) shows how the upwind effects total wind mixing effects on dilute

concentrations or may allude to the source (e.g. increased as wind shifted NE, versus SW). Although, currently, the authors do show in Figure 3 the wind direction (red arrow vector), **but perhaps a wind rose representing the time domain of sampling could be shown.** Although, not necessary, if this “red arrow” on wind represents a mean over the duration of time sampled or constant direction.

#17NRH: **Does the red vector arrow show the mean or constant direction of wind over the sampling period?** If so, mentioned this in Figure 3 caption (e.g. wind direction represents average during sampling time).

Response to Comment 16 and 17

The following has been added to Figure 3 Caption: “The red vector arrow indicates the constant wind direction during the sampling period.”

No further comments.

Section 4: Results

NRH: In summary, the results show trends of elevated methane and carbon dioxide concentrations in specific sites and a variability in stable carbon isotope signatures, indicating possible differences in the sources of these greenhouse gasses at each site. The results highlight the observation of elevated atmospheric CH4 concentrations at four of the five (Pingo 1, Pingo 2, Wetland 2, Wetland 3) where walking transects were performed.

The results (as expected, from hypothesis) show that proximity to source of seep, e.g., ebullition (closest to standing water) which had substantially elevation methane concentrations. From this, I am curious of the relationship between distance to standing water and/or topographic upland versus lowland relationship to both concetrations and isotopic signatures elsewhere. Also noteworthy, Keeling plot values show -88.4‰ for wetlands in the fall, during maximum thaw, or when thermal lag from summer months penetrates deeper in talik sources, given the latency heat of water. Whereas Keeling plot values show -56.7‰ during summer months, when the cold season thermal lag can be substantial in the subsurface.

#18NRH: Perhaps this suggest age dependency on talik versus time of thermal lag in subsurface and seasonal thermal lags associated with age. Alternatively, hydrology or seasonality of groundwater surge (higher in summer, lower in august) may result in more mixing effects. It might be useful to discuss this later.

Line 273-274: > Keeling plot values were -88.4‰ for Wetland 1 when sampled in the fall, but - 56.7 ‰ when sampled in the summer.>

#19NRH: Please add month (x), e.g., fall (X), summer (X) in Line 273-274. No further comments.

Response to Comment 18 and 19

“October” and “July” have been added to line 247

Section 5.1: Results - Consider comments on reference material #20-21 - Outer delta pingos

NRH: The study discovered high methane concentrations in airborne hotspots in the northwestern outer MRD. The carbon isotope signatures from these sites suggest a likely mixture of thermogenic and biogenic methane sources. Despite the proximity of these high readings to pingo features, notably, **the highest values were detected in the surrounding low-lying shrub tundra, not the features themselves.**

#20NRH: This suggest topographical relationships to hotspots or water table position by Elder et al., Baskaren et al., etc.

NRH (Line 315): indeed, I agree, this is exciting result and assumptions about the permafrost through-taliks, pingo hydrology, and complex permafrost and absence of permafrost does warrant geophysical transects. It seems understanding the source attribution is a function of the assumptions about permafrost or absence of permafrost, which can be demarcated by low- frequency EM geophysical transects.

Response to Comment 20:

The following text has been added to line 282:

“This interpretation would be consistent with recent findings in the region that water table depth, hydrology and topography are critical factors driving emissions at hotspots in the MRD (Elder et al., 2020; Baskaran et al., 2022; Hodson et al., 2020a, b).”

- Wetland sites

<sampling at Wetland 1 revealed a seasonal variability in the carbon isotope signature, with a value of -88.3‰ in October (suggesting a biogenic source) and -53.4‰ during the summer (indicating a mixed source). This could be due to methane oxidation or a blend of biogenic and thermogenic sources.> >The lack of observed methane ebullition during summer suggests a potential seasonal variation in methane flux in these wetland settings.>Therefore, contributions to the atmospheric methane at Wetland 1 during summer could be from both biogenic and thermogenic sources, with oxidation and varying production pathways also being plausible.>

>Similar observations for seasonal variability in terrestrial sources are not well documented in the literature, although transport of CH4 from anaerobic soils with sedge vegetation has been observed to bypass the aerobic zone, limiting oxidation during the growing season (Olefeldt et al., 2013; King et al., 1998). Therefore, it is possible that there were contributions to the atmosphere from biogenic and thermogenic sources at Wetland 1, but oxidation and varying production pathways cannot be ruled out as the reason for the signature derived during the summer sampling>

#21NRH:Interesting result. This may also allude to thermal lag times moving through different depths seasonally or perhaps more important here, the mixing blend of biogenic and thermogenic sources are enhanced by summer peak water levels, which can transport various methane sources significantly in the watershed. **Groundwater discharge as a driver of methane emissions and mixing from Arctic lakes and transporting methane from upland active layer thaw to lowland area has been shown to be significant,** supporting oxidation and blending of biogenic and thermogenic source.

... **Ground water discharge is known to follow as seasonal trend, which perhaps alludes to the shifts from summer to fall data, with greater mixing of biogenic and thermogenic sources in summer, and more stable (deeper) biogenic sources emanating directly from the talik, without ground water mixing.** For example, Please see Olid et al(2022) and Paytan et al. (2015).

<u>Suggested supporting reference #11</u>

Olid, C., Rodellas, V., Rocher-Ros, G. et al. Groundwater discharge as a driver of methane emissions from Arctic lakes. Nat Commun 13, 3667 (2022). https://doi.org/10.1038/s41467-022-31219-1

<u>Suggested supporting reference #12</u>
Lecher, Alanna & Dimova, Natasha & Sparrow, Katy & Garcia-Tigreros, Fenix & Murray, Joseph & Tulaczyk, Slawek & Kessler, John. (2015). Methane transport from the active layer to lakes in the Arctic using Toolik Lake, Alaska, as a case study. Proceedings of the National Academy of Sciences of the United States of America. 112. 10.1073/pnas.1417392112.

Response to Comment 21:

The following has been added to line 315:

"Groundwater inputs to lakes in permafrost areas are higher during the summer months (Olid et al., 2022), which would increase the possibility of thermogenic inputs during the summer."

No further comments.

Section 5.2: Results - Consider more recent reference support #21 and counter evidence #22,23 (if applicable), and further recent support for thermogenic versus biogenic signatures in arctic from #24

Line 368: >Some of the largest occurrences of atmospheric release of CH4 in Arctic environments have been reported in association with large gas seeps of thermogenic CH4, causing high-rate ebullition (Walter Anthony et al., 2012)>

#21NRH: consider also adding the more recent literature for support for both biogenic sourced geological gas and thermogenic >

Kleber, G.E., Hodson, A.J., Magerl, L. et al. Groundwater springs formed during glacial retreat are a large source of methane in the high Arctic. Nat. Geosci. 16, 597–604 (2023). https://doi.org/10.1038/s41561-023-01210-6

Sullivan, TD, Parsekian, AD, Sharp, J, et al. Influence of permafrost thaw on an extreme geologic methane seep. Permafrost and Periglac Process. 2021; 32: 484–502. https://doi.org/10.1002/ppp.2114

#22NRH: I am curious how these results of δ13C-CH4 source signatures (e.g. -42.0 to 44.7 ‰) challange large gas seeps of thermogenic CH4, when Elder et al. (2018) showed these signatures associated with alaskan lakes dominated by young carbon, which may challenge this interpretation.

For example, you may wish to add this edits:

<Some of the largest occurrences of atmospheric release of CH4 in Arctic environments have been reported in association with large gas seeps of thermogenic CH4, causing high-rate ebullition (Walter Anthony et al., 2012). **However, arctic coastal plain CH4 emissions have been associated with younger carbon sources (Elder et al., 2018)**”>

Response to Comment 21 and 22:

The following has been added at line 327:

“but biogenic contributions from microbially produced fossil CH_4 (Sullivan et al., 2021) and young carbon can also occur at seeps with high flux rates (Elder et al., 2018)”

#23NRH: you may want to download Elder et al. supp section and take a look at those corresponding δ13C-CH4 signatures and similarities or dissimilarities to your data.

Suggested supporting reference #13

Elder, C.D., Xu, X., Walker, J. et al. Greenhouse gas emissions from diverse Arctic Alaskan lakes are dominated by young carbon. Nature Clim Change 8, 166–171 (2018). https://doi.org/10.1038/s41558-017-0066-9

#24NRH (Line 389-400): Nice summary paragraph. Kelebar et al. (2023) supports similar interpretations and may be used as additional reference.

Kleber, G.E., Hodson, A.J., Magerl, L. et al. Groundwater springs formed during glacial retreat are a large source of methane in the high Arctic. Nat. Geosci. 16, 597–604 (2023). https://doi.org/10.1038/s41561-023-01210-6

Response to Comment 23 and 24:

The following has been added at line 353:

“Stable carbon isotope values even higher (more enriched in ^{13}C) than those reported in this study (-38.8 ‰) were observed below lake ice on the north slope of Alaska (Elder et al., 2018). These higher values were attributed to oxidation of biogenic CH_4 but, were measured in an environment where high rates of oxidation are likely.”

No further comments.

6 Conclusion

NRH: The authors present novel data and likely is the first to measure stable carbon isotope signatures of atmospheric methane at hotspots in the Mackenzie River Delta (MRD). The results suggest a variety of methane production sources in the MRD, with source isotope signatures ranging from -42 ‰ (biogenic) to -88 ‰ (thermogenic). Of the eight sites investigated, two had a thermogenic origin, four were biogenic, and two were possibly a result of oxidation of mixed biogenic/thermogenic sources. This is supported by other recent studies.

These results also suggest methane migration from below the thin permafrost at most sites, including from the Taglu gas field, over an area of approximately 20 km north to south, which suggest complex permafrost distribution (e.g. due to the hydrology of river taliks, pingo- systems, coastal settings, and together a mix of likely through-taliks and permafrost degradation).

The study was able to validate airborne eddy covariance hotspot locations using walking transects to measure atmospheric methane variation. However, authors point out these methods only provide a snapshot of methane sources during site visits, and a comprehensive understanding of annual methane production is yet to be established.

Authors suggest that future research should include year-round flux measurements and stable carbon isotope measurements to fully quantify the annual methane emission from both biogenic and thermogenic sources. Additionally, combining portable methane analysers with flux chamber and isotopic measurements could help to better identify and quantify sources, particularly at sites where both biogenic and thermogenic sources are likely. Geophysical mapping atop these transects would provide a useful coupling to permafrost distribution and potential source attribution between biogenic and thermogenic sources.

The study offers valuable insights into the Kohnert et al. (2017) results, which inspired many further airborne and ground measurements across the arctic (e.g. ABoVE domain). Therefore, these results should be published as presented, with additional comments as recommended, and may further enhance these valuable ground truths for current campaigns conducting similar observations.

I highly recommend adding some of the added reference support material (#1-13) and considering comments #1-23. Particularly important, reference material post-Kohnert et al. results from MRD, e.g., Elder et al. (2020, 2021) and Baskaren et al. (2022). I hope these added suggestions and reference support materials can serve to enhance your significant results.

Nice work. NRH 8-5-2023 End.

Assignment: Accepted subject to minor technical corrections or additional reference material, as suggested.

References:

Baskaran, L., Elder, C., Bloom, A. A., Ma, S., Thompson, D., & Miller, C. E. (2022). Geomorphological patterns of remotely sensed methane hot spots in the Mackenzie Delta, Canada. *Environmental Research Letters*, *17*(1), 015009. https://doi.org/10.1088/1748-9326/ac41fb

Elder, C. D., Thompson, D. R., Thorpe, A. K., Chandanpurkar, H. A., Hanke, P. J., Hasson, N., James, S. R., Minsley, B. J., Pastick, N. J., Olefeldt, D., Walter Anthony, K. M., & Miller, C. E. (2021). Characterizing Methane Emission Hotspots From Thawing Permafrost. *Global Biogeochemical Cycles*, *35*(12). https://doi.org/10.1029/2020GB006922

Elder, C. D., Thompson, D. R., Thorpe, A. K., Hanke, P., Anthony, K. M. W., & Miller, C. E. (2020). Airborne Mapping Reveals Emergent Power Law of Arctic Methane Emissions. *Geophysical Research Letters*, 10.

Elder, C. D., Xu, X., Walker, J., Schnell, J. L., Hinkel, K. M., Townsend-Small, A., Arp, C. D., Pohlman, J. W., Gaglioti, B. V., & Czimczik, C. I. (2018). Greenhouse gas emissions from diverse Arctic Alaskan lakes are dominated by young carbon. *Nature Climate Change*, *8*(2), 166–171. https://doi.org/10.1038/s41558-017-0066-9

Hasson, N. (2022). *Methane emissions show exponential inverse relationship with electrical resistivity from discontinuous permafrost wetlands in Alaska*. AGU Fall Meeting. https://ui.adsabs.harvard.edu/abs/2022AGUFM.B15E..06H

Kohnert, K., Serafimovich, A., Metzger, S., Hartmann, J., & Sachs, T. (2017). Strong geologic methane emissions from discontinuous terrestrial permafrost in the Mackenzie Delta, Canada. *Scientific Reports*, *7*(1), 5828. https://doi.org/10.1038/s41598-017-05783-2

Kuhn, M. A., Varner, R. K., Bastviken, D., Crill, P., MacIntyre, S., Turetsky, M., Walter Anthony, K., McGuire, A. D., & Olefeldt, D. (2021). BAWLD-CH_4: A comprehensive dataset of methane fluxes from boreal and arctic ecosystems. *Earth System Science Data*, *13*(11), 5151–5189. https://doi.org/10.5194/essd-13-5151-2021

Minsley, B. J., Abraham, J. D., Smith, B. D., Cannia, J. C., Voss, C. I., Jorgenson, M. T., Walvoord, M. A., Wylie, B. K., Anderson, L., Ball, L. B., Deszcz-Pan, M., Wellman, T. P., & Ager, T. A. (2012). Airborne electromagnetic imaging of discontinuous permafrost: AEM IMAGING OF PERMAFROST. *Geophysical Research Letters*, *39*(2), n/a-n/a. https://doi.org/10.1029/2011GL050079

Sullivan, T. D., Parsekian, A. D., Sharp, J., Hanke, P. J., Thalasso, F., Shapley, M., Engram, M., & Walter Anthony, K. (2021). Influence of permafrost thaw on an extreme geologic methane seep. *Permafrost and Periglacial Processes*, *32*(3), 484–502. https://doi.org/10.1002/ppp.2114