The evolution of methane production rates from young to mature thermokarst lakes

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ABSTRACT

Thermokarst lakes, formed by permafrost thaw in the Arctic, are hotspots for methane (CH_4) and carbon dioxide (CO_2) emissions, and are expected to double permafrost carbon emissions by the end of the century. While the implications of ongoing permafrost thaw on CH_4 dynamics within these lakes have been modeled, here we provide empirical data on CH_4 production dynamics as lakes evolve from young recently formed lakes to older lakes that have been present for hundreds of years. Sediment cores (up to 4 m long) were collected from the centers and thermokarst margins of a new thermokarst lake [Big Trail Lake (BTL), <70 years] and from an older thermokarst lake [Goldstream Lake (GSL), ~900 years] from the same interior Alaskan watershed. The highest CH_4 production rates were observed in the uppermost sediments near the sediment-water interface at the thermokarst margins of both lakes, with a steep decrease with sediment depth into the talik. BTL exhibited elevated CH_4 production rates, correlated with higher carbon lability for thermal induced reactions measured by Rock Eval analyses, suggesting its potential use as a proxy for organic carbon breakdown by methanogenesis. In contrast, GSL displayed lower CH_4 production rates, likely due to a longer period of organic carbon degradation and reduced carbon lability. The integrated sediment-column CH_4 production rates were similar (around 7 to 10 mol m^{-2} $year^{-1}$), primarily due to the thinner talik at BTL. Our data support the predictions that formation and expansion of thermokarst lakes over the next centuries will increase CH_4 production in newly thawed Yedoma permafrost sediments, while CH_4 production will decrease as taliks mature and labile organic carbon is used up.

1. Introduction

Permafrost covers one quarter of the northern hemisphere (Obu, 2021; Zhang *et al.*, 2008). Significant warming of the Arctic and subarctic regions, with temperature increases of 2 to 5°C relative to pre-industrial levels (Post *et al.*, 2019), exacerbates the thawing of this permafrost. In turn, Arctic soils, which are currently a store of carbon, become a source of carbon to the atmosphere by emitting greenhouse gases (Schuur *et al.*, 2015). It is estimated that until the end of this century thaw, rapidly thawed permafrost will become an important source of greenhouse gasses (Turetsky *et al.*, 2020). Among these gasses, CH_4 is expected to be a dominant driver of the circumpolar permafrost-carbon radiative, responsible for up to 70% of this effect (Walter Anthony *et al.*, 2018).

Methane release is expected to increase in rapidly thawing permafrost beneath thermokarst lakes, which are CH_4 "hotspots" on the landscape, and expected to double permafrost carbon emissions and increase associated radiative forcing effects by 130% by the end of the century (Elder *et al.*, 2021; Hugelius *et al.*, 2014; Olefeldt *et al.*, 2016; Walter Anthony *et al.*, 2018). Thermokarst lakes are formed when ground ice melts and water accumulates in subsidence areas (Hopkins, 1949). As thaw continues beneath the lake, previously frozen organic carbon within the thawed permafrost soils called taberites (Farquharson *et al.*, 2016; Strauss *et al.*, 2013), becomes available for microbial degradation, which produces CO_2 and CH_4 (Freitas *et al.*, 2025; Heslop *et al.*, 2015; Walter Anthony *et al.*, 2018). In particular, organic-rich permafrost Yedoma soils of Alaska and Siberia are noteworthy reservoirs of old, ^{14}C-depleted, labile soil carbon that is quickly degraded into greenhouse gases upon thaw (Dutta *et al.*, 2006; Estop-Aragonés *et al.*, 2020; Knoblauch *et al.*, 2018; Zimov *et al.*, 1997).

Models have simulated the changes in methane production as thermokarst lakes evolve (Kessler *et al.*, 2012). These models show that while older lakes may have lower surface production rates, their taliks can still contribute significantly to total methane flux. Over centuries, the talik deepens through the Yedoma and into the bedrock, while intra-talik organic carbon in the *in situ* thawed Yedoma sediments (taberites) gradually becomes refractory. At later stages, thawed permafrost organic carbon is no longer available for CH_4 production and the lake is no longer a significant source of permafrost-derived CH_4 to the atmosphere, unless permafrost soil carbon is re-transported from lake margins or the watershed to surface lake sediments (Walter Anthony *et al.*, 2014).

However, while the implications of ongoing permafrost thaw on CH_4 dynamics within these lakes have been modeled, few empirical data exist about changes in CH_4 production in relation to stages of thermokarst-lake evolution. The limited studies on CH_4 production rates in the talik of thermokarst lakes in the Arctic exhibit substantial variability, with fluctuations of 3 to 4 orders of magnitude observed across different environments and studies. For example, the top sediment layer of a non-Yedoma lake on the North Slope of Alaska shows CH_4 production levels of 1000 nmol cm^{-3} d^{-1} (de Jong *et al.*, 2018). CH_4 production rates range from 2 to 35 nmol cm^{-3} d^{-1} in Doughnut and Vault Lakes in discontinuous Yedoma's permafrost in Alaska (Martinez-Cruz *et al.*, 2018), while values as high as 350 nmol cm^{-3} d^{-1} have been reported in Vault Lake, and lowest in Goldstream Lake (Sepulveda-Jauregui *et al.*, 2015). Our recent study in this region on short sediment cores (up to one meter depth) constrained CH_4 production rates in the upper sediments similar to (Martinez-Cruz *et al.*, 2018), based on radiocarbon and CH_4 accumulation incubations (Pellerin *et al.*, 2022). Freitas et al. (2025) showed, by using radiocarbon dating, sediment incubations, and sediment facies classifications, that CH_4 production can also occur deep (~20 m) beneath Yedoma thermokarst lakes in sand and gravel layers. All together, the data raised several questions regarding the evolution of CH_4 production rates throughout the talik, the role of CH_4 oxidation and the lability of organic carbon in thermokarst lake systems, warranting further investigation.

In this study, Yedoma thermokarst lake sediment cores were retrieved up to four meters in depth to measure, calculate and compare CH_4 production rates along the talik between two distinct lake systems. Then, the CH_4 production rates, indicating the susceptibility of the organic carbon to microbial degradation, were compared to the total organic carbon (TOC) and its lability based on thermally induced breakdown. The thermal lability was deduced from Rock-Eval analysis, which involves gradual heating under pyrolysis conditions, followed by combustion of the residual sample (Behar *et al.*, 2001). With the gradual heating, the generation of hydrocarbon, CO and CO_2 are monitored. Following pyrolysis, the residual organic matter is lean in hydrogen and structural changes making it more aromatic and refractory. The long cores enabled a direct comparison of CH_4 production rates, porewater geochemistry and organic carbon lability along geographically proximate yet geomorphologically distinct lakes – a young lake (<70 years) and an old lake (~900 years) (Fig. 1). They also improved our

understanding of how CH_4 production rates and carbon lability are expected to change over time, providing the first empirical data for comparison to model predictions.

2. Methods

Region of study

The Goldstream Valley watershed is located about 10 km northeast of Fairbanks, Alaska (Fig. 1). This area has a subarctic, continental climate with an average annual temperature of -3.3°C and annual precipitation of 280 mm (Douglas *et al.*, 2020). The vegetation is primarily composed of boreal lowland species. The emergent lake and nearby thermokarst environments have been described by Elder *et al.* (2021). Recent studies (Hasson *et al.*, 2022) show that the mean annual ground temperature of permafrost is -0.26°C at a depth of 7.2 m. The Goldstream valley features discontinuous, ice-rich Yedoma type permafrost, originating from late-Pleistocene loess deposits that were remobilized during the Holocene. These deposits form thick layers over relic Goldstream formations, altering the soil chemistry and ice volume distribution (Péwé, 1975). Between 1949 and 2009 the number of thermokarst lakes in the valley has doubled and their total area increased by about 40% (Walter Anthony *et al.*, 2020). These lakes are interconnected by a shifting watershed that feeds into the Tanana River, which is part of the Yukon River Basin.

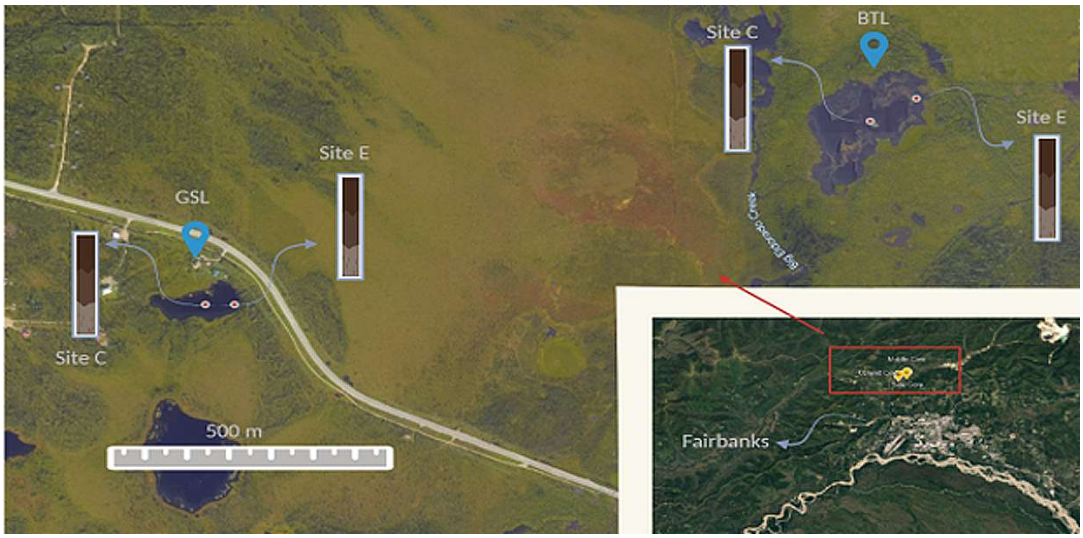

Figure 1: Study sites: The young (~70 years old) Big Trail Lake (BTL) and the old (850-900 years old in its center) Goldstream lake (GSL) in the Goldstream Valley (© Google Earth), including the locations of the collected cores in both lakes (Edge (E), Center (C)).

In this study, two markedly different thermokarst lakes were studied. Big Trail Lake (BTL) (64.9189N, 147.8212W, 609 m^2 in 2009) is an actively expanding lake formed from a wetland and possibly a migrating fluvial channel sometime between 1949 and 1967 (Walter Anthony *et al.*, 2020). Extensive geophysical surveys at BTL showed massive ice (*e.g.* foliated ice wedges) starting roughly at 10-15 m below the irregular talik shape (Walter Anthony *et al.*, 2020). Permafrost and valley hydrology were investigated on the valley scale (Emond *et al.*, 2018). BTL is surrounded by valley-bottom creeks and streams, some supply water to the lake by draining surrounding upland fens and the historical channel; these tributaries sometimes run dry and isolate the lake hydrologically at the surface. The outlet of the lake feeds into Eldorado Creek.

Goldstream lake (GSL) (64.9156N, 147.8495E, 1278m^2), located about 1.5 km from BTL, is actively eroding into relic Yedoma permafrost with likely much less reworked Yedoma, due to its location on the base toe slope of Goldstream valley and slightly elevated above the Goldstream creek watershed at 196 m. Geophysical surveys estimated talik thickness range of 30-40 m (Emond *et al.*, 2018; Péwé, 1975). A previous study cored 20-m of Goldstream Lake sediments and found ice-free sand and gravels beneath 16-m of thawed silt (Walter Anthony *et al.*, 2020). Radiocarbon dates suggest the oldest part of the basin is around 850 to 900 years; however rapid expansion of the eastern margin into Yedoma permafrost has occurred since 1949.

Sampling and profiles

We collected sediment cores using a vibro-corer deployed directly from the lake ice in March 2022. In each lake one core was collected from the center of the lake and from the edge within the water saturated zone with an aluminum liner. It was winched out of the sediment, tilted on its side and cut into ~1 m sections (marked in Table S1), which were quickly capped and transported by a snow machine and vehicle to the University of Alaska Fairbanks for further analysis. In the lab, each section was cut lengthwise and one half was sampled horizontally at intervals of 15 to 25 cm. First, a sub sample of sediment was taken immediately after slicing the core liner for CH_4 concentrations and the stable carbon isotope composition ($\delta^{13}C_{CH4}$ and $\delta^{13}C_{CO2}$); about 3 mL of sediment was taken in a cut-off 3 mL syringe and inserted directly into a 20 mL fully saturated with an anoxic 5 M sodium chloride solution. Another cut-out 3 mL syringe was inserted as well into the sediment at each depth to extract sediment for incubations. For

density measurements, samples were taken into 10 mL vials, weighed, and then re-weighed after drying an aliquot of sediment at 60°C for 4 days. For total organic carbon (TOC) concentration, its carbon stable isotope composition ($\delta^{13}C_{TOC}$) and Rock-Eval analyses a sediment subsample was freeze-dried. Porewater was extracted by Rhizons (Dickens *et al.*, 2007). The Rhizons were inserted into the sediment and vacuum was created with a 10 mL syringe which accumulated the extracted porewater after filtration through 0.22 µm. The filtrated porewater was then stored at 4°C in 2 mL amber glass vials without headspace for measurement of dissolved inorganic carbon (DIC) concentrations and its stable carbon isotope composition ($\delta^{13}C_{DIC}$) within a week from sampling.

Production rate experiments

A 3 mL cut-out syringe was inserted into the sediment in each core at 20 to 30 cm intervals to retrieve about 2 mL of sediment. The sediment was added to 20 mL serum bottle, which was sealed with butyl rubber stopper and crimped with aluminum cap. The bottles were vigorously shaken immediately and purged with 99.999% N_2 gas for 15 minutes to remove oxygen and other gasses. Three serum bottles were taken at each depth. After weighing, the samples were stored in the dark at 4°C. The increase in CH_4 concentrations in the headspace was recorded after 80, 130 and 160 days by GC-FID (see below), allowing the back-calculation of CH_4 production rates.

Methane production rates were measured at each time point (80, 130, 160 days) using a small (100 µL) aliquot of gas from the headspace of the serum bottles. Headspace CH_4 concentration was converted to total CH_4 in the bottle based on the concentration, the volume of sediment and the volume of headspace in each serum bottle. CH_4 production rate was then taken as the increase in CH_4 concentration over time. Since each depth had three serum bottles, the average CH_4 production rate was reported for each depth and the uncertainty on CH_4 production rate was reported as the standard deviation of the mean. After 160 days, $\delta^{13}C_{CH4}$ was also measured. Since any CH_4 dissolved in the porewater at the time of sampling had been removed during the N_2 purge, the $\delta^{13}C_{CH4}$ values represented the newly accumulated CH_4 during the incubation.

Total profile CH_4 production rates, reported in mol m^{-2} $year^{-1}$ were calculated using the CH_4 production rates obtained from the incubation experiments and depth integration

throughout the thawed talik. The inferred talik thickness was based on geophysical surveys and measurements of the taberite depth (Freitas *et al*., 2025; Walter Anthony *et al.*, 2020).

Analytical methods

The headspace of the serum bottles of the CH_4 rate incubations was measured for CH_4 concentrations at each time point using the procedure described by Pellerin et al. (2022). In general, 250 µL aliquot of gas from the headspace and inserted into a Gas Chromatograph (GC, Thermo) equipped with Flame Ionization Detector (FID) and a Packed ShinCarbon ST column (Restek) at 120°C. The GC-FID was calibrated with a standard curve prepared with CH_4 concentration of 1%. CH_4 concentrations in the profiles were measured by injecting 3 mL of 99.999% N_2, while simultaneously removing 3 mL of the saturated 5 M sodium chloride solution. After equilibrating for two weeks upside down, bottles were vigorously shaken and analyzed for CH_4 as descried above. This method had a precision of ±2 µM. The $\delta^{13}C_{CH4}$ and $\delta^{13}C_{CO2}$ values were measured together by PreCon and Gas Bench II interfaced with Delta V Gas Source Isotope Ratio Mass spectrometer (GS-IRMS, Thermo). Internal standards of Tiso-2 (Isometric Instruments, 2.5% CH_4) $\delta^{13}C$ -38.3 ‰, and inhouse (99.8 % CH_4) $\delta^{13}C$ -48.6 ‰ were used, with analytical error of 0.2 ‰. For $\delta^{13}C_{CO2}$ determination, 99.8 % CO_2 lecture bottle (Scott mini mix) with $\delta^{13}C$ of -25.5 ‰ was used and results were reported with analytical error of 0.3 ‰. All $\delta^{13}C$ results were reported compared to Vienna Pee Dee Belemnite (VPDB).

The TOC in the freeze-dried sediment was measured after the removal of carbonates by the addition of 1% H_3PO_4 and drying at 40°C in silver cups until the sample stopped reacting with the acid. Triplicate samples and internal standards were then packed in tin cups and measured on an elemental analyzer and HS2022 IRMS (Sercon). TOC concentrations were measured using concentration calibration and peak heights, and $\delta^{13}C_{TOC}$ was reported vs. VPDB with the reference materials IAEA-600 ($\delta^{13}C$ -27.7 ‰ VPDB), USGS62 ($\delta^{13}C$ -14.8 ‰ VPDB) and USGS63 ($\delta^{13}C$ -1.2 ‰ VPDB). The precision was ±0.3‰.

Organic carbon lability for thermally induced chemical reactions was characterized by Rock-Eval analysis (Rock-Eval 6 Vinci Technologies). The technique determines the proportion of pyrolysable C (PC) and residual carbon (RC). PC is composed of the sum

of three pyrolysates: S1, composed mostly of small volatile molecules, S2, larger hydrocarbon molecules thermally cracking like algal cell walls and S3, derived from oxygen-containing molecules. The residual carbon (RC) is released from the sample during the combustion cycle (Carrie *et al.*, 2012, Sanei *et al.*, 2005). The indices used as proxies to the organic lability are: 1) hydrogen index, which is calculated as HI = S2/TOC x 100 (Behar et al, 2001). Higher HI values indicate a greater hydrogen-rich organic compounds, implying lability. 2) The ratio between PC and RC. Higher ratio indicates that the OM is richer in hydrogen and is more aliphatic; hence, it might be more available to microbial respiration. About 20 mg of the prepared samples were placed in the RE6, which was then ramped at a predetermined rate (25 °C/min) from 200°C to 650°C in the pyrolysis oven. The oven was cooled down and the sample was transferred to the combustion oven, where it was ramped from 200°C up to 850°C at a rate of 25 °C/min.

The DIC and $\delta^{13}C_{DIC}$ were analyzed after acidification of the porewater sample to convert DIC to CO_2. The $\delta^{13}C_{DIC}$ was measured with GS-IRMS (Thermo) interfaced to Gas Bench II. Values were reported relative to VPDB with precision of ±0.1‰. DIC concentrations were measured by integrating the signal of the sample on the IRMS. The signal was calibrated using peak heights $NaHCO_3$ solutions prepared at concentrations of 3 to 10 mM. The precision was ±0.2 mM.

The comparison of $\delta^{13}C_{CH4}$ between the CH_4 in the profiles and CH_4 produced in the rate incubations was used to quantify the contributions of CH_4 in the shallow sediments relative to the transport of CH_4 migrating or diffusing from deeper in the talik. This is assuming that the CH_4 measured in the profiles represents CH_4 that is produced *in situ* plus CH_4 that is produced in greater depths and migrates upward, while that in the incubations (after purging) represents only the *in situ* CH_4 production. The bubbles composition was assumed to be the most negative $\delta^{13}C_{CH4}$ value measured in the profiles because it migrates from deeper in the sediment, as was shown previously (Pellerin *et al.*, 2022).

Equation 1:

$$new\ CH4\ production\ fraction = \frac{\delta^{13}CH_4(PW) - \delta^{13}CH_4(bubble)}{\delta^{13}CH_4(incubation) - \delta^{13}CH_4(bubble)}$$

3. Results

Methane production characteristics from porewater profiles

Microbial respiration was intensive in general, as indicated by the high DIC concentrations in all sites. In the center of BTL, DIC concentrations increase linearly from the surface towards 150 cm depth with a peak of 37 mM at 200 cm, followed by a subsequent decrease back to 25 mM (Fig. 2A). On the other hand, at the edge of BTL, DIC concentrations ranged from 10 to 15 mM (Fig. 2B). Along the talik in the center of the lake the $\delta^{13}C_{DIC}$ values increased from 3 ‰ to 16 ‰ (Fig. 2A). On the edge of the lake the $\delta^{13}C_{DIC}$ values increased in the upper 50 cm from 5 to 17 ‰ and then remained similar (Fig. 2B).

BTL center exhibited CH_4 concentration peaks at 70 and 300 cm (about 1 mM), while CH_4 concentrations in the remaining core sections ranged from 0.3 to 0.5 mM. The $\delta^{13}C_{CH4}$ values ranged from -55.6 to -69 ‰ in the deeper section (Fig. 2A). CH_4 concentrations at BTL edge (Fig. 2B) exhibited values of 0.2 to 0.5 mM at depths of 0 to 150 and 200 to 300 cm, with peak values of up to 1.4 mM observed at depths of 150 and 350 cm. The shallow part exhibited $\delta^{13}C_{CH4}$ values of -55 ‰, which became more negative, with -71.5 ‰, in the deeper section.

At GSL center, DIC concentrations remained around 20 mM, with a maximum of 32 mM observed at depth of 100 to 150 cm (Fig. 2C). The $\delta^{13}C_{DIC}$ values decreased in the upper 100 cm, from -2 to -15 ‰, followed by a slight increase to -12 ‰ (Fig. 2C). At the edge of GSL, DIC concentrations exhibited a linear increase with depth, from 20 to 46 mM (Fig. 2D). The $\delta^{13}C_{DIC}$ values (Fig. 2D) exhibited different trends than BTL, with negative values, decreased from approximately -2 to -6 ‰ within the first 100 cm. Below this depth, there was a slight increase in $\delta^{13}C_{DIC}$ values, to -3 ‰.

Methane concentrations at GSL remained relatively steady at the center (1 mM), with a slight decrease at 150 cm to 0.7 mM (Fig. 2C). CH_4 concentrations edge exhibited varied range of 0.5 to 1.5 mM (Fig. 2D). The highest CH_4 concentration was observed in the deepest and shallowest parts of the core (close to 1.5 mM). The lowest concentration was found at a depth of 150 cm. The $\delta^{13}C_{CH4}$ values at the center of GSL ranged from -68.4 ‰ in the shallow part to -78 ‰ in the deeper part (Fig. 2C). At the edge they ranged from -65 ‰ in the shallow part to -73 ‰ in the deepest section (Fig. 2D).

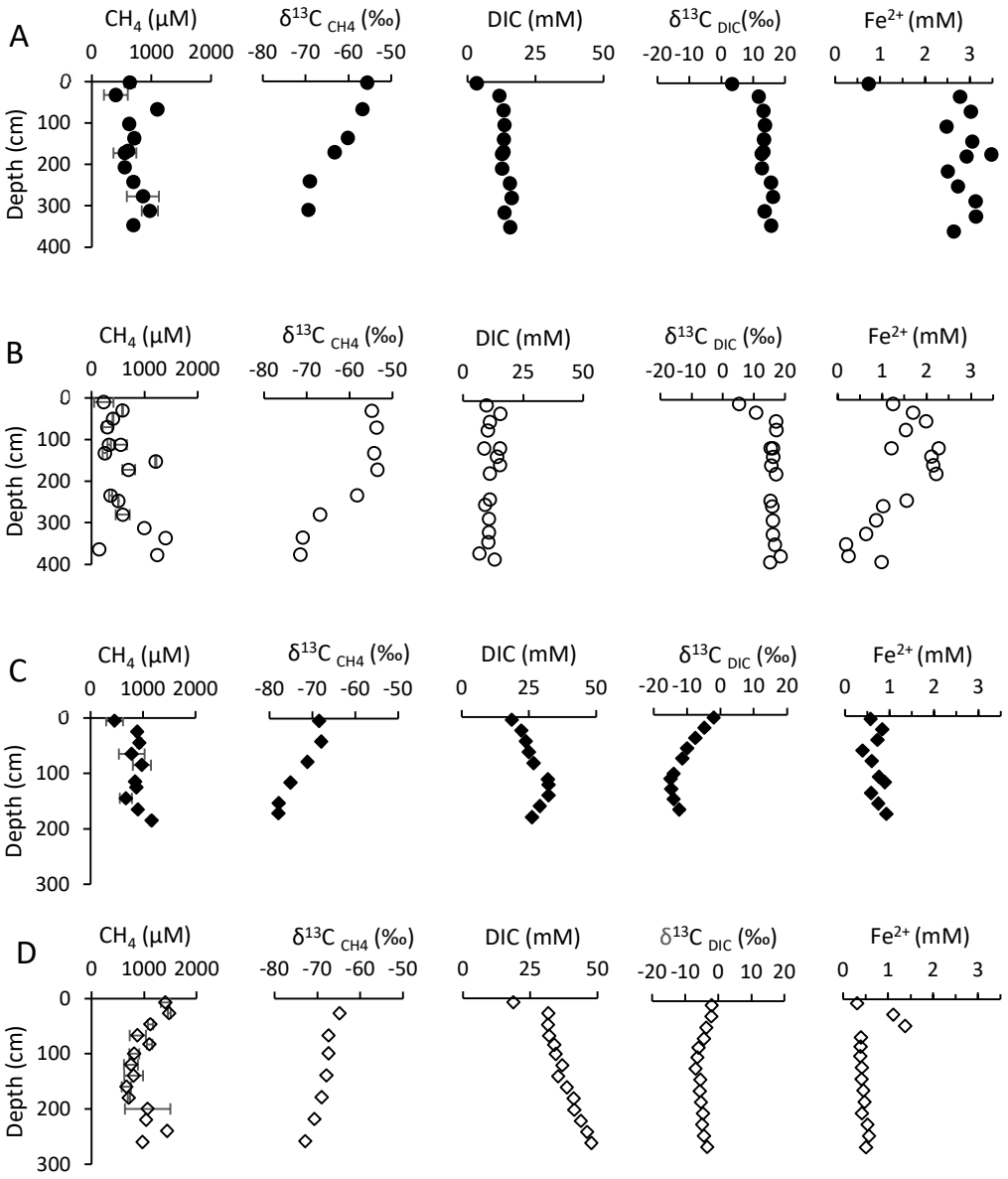

Figure 2: Pore water profiles of GSL (circles) and BTL (diamonds) cores. Panels A and B correspond to BTL Center (full) and Edge (empty) respectively, while panels C and D correspond to GSL Center and Edge respectively. Error bars within markers limit unless depicted otherwise.

Surficial sediments of lakes edges were organic rich (TOC~10 %) with relatively low $\delta^{13}C_{TOC}$ (table S1) and low CH_4 and DIC concentrations. The TOC content decreased significantly with depth on the edge, while the center of the lakes had low TOC content. In BTL in both edge and center, the $\delta^{13}C_{DIC}$ increased significantly with depth, with values typical of methanogenesis. In GSL, on the other hand, there was a significant

decrease in $\delta^{13}C_{DIC}$ with depth but relatively constant DIC and CH_4 concentrations and isotopes in the rate incubations, which suggest small role of CH_4 related processes with some signature of methane oxidation in the upper sediments.

Methane production rates in sediment incubations

Methane production rates were measured by sediment incubation batch experiments. The sediment from BTL and GSL had the highest CH_4 production rates near the sediment-water interface. In BTL the rates within the upper meter ranged from 2 to 20 nmol cm^{-3} day^{-1} in the core taken from the center of the lake (Fig. 3A) and 7 to 35 nmol cm^{-3} day^{-1} in the core taken from the edge of the lake (Fig. 3B). In the deeper sediments, the rates decreased to about 1 nmol cm^{-3} day^{-1} in both sites.

At the center of GSL (Fig. 3C), CH_4 production rates were of 4 to 6 nmol cm^{-3} day^{-1} in the upper one meter of sediment. Deeper, the rates decreased, ranging from 2 to 4 nmol cm^{-3} day^{-1}. At the edge higher rates were measured in the upper 50 cm (yet lower than in BTL) following by a sharp decrease below 50 cm depth (Fig. 3D).

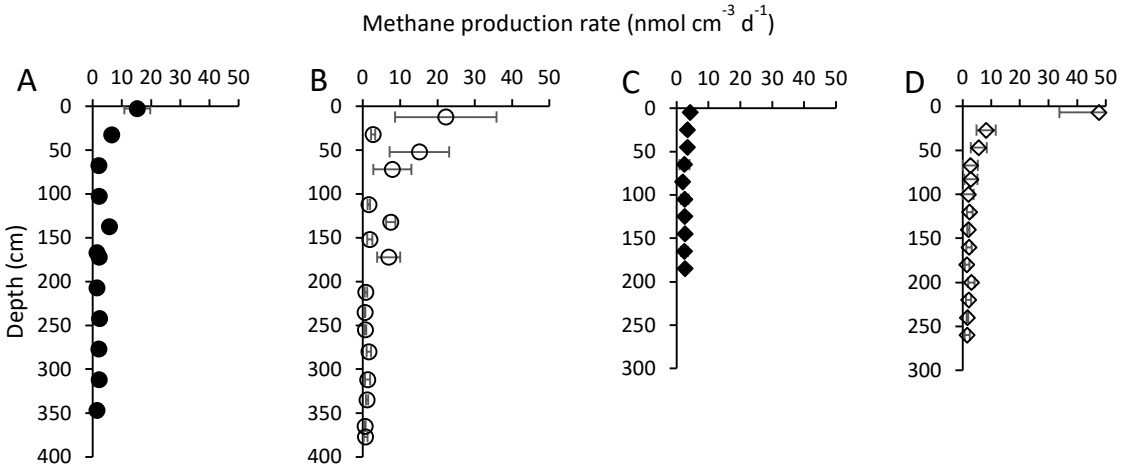

Figure 3: CH_4 production rates (nmol CH_4 cm^{-3} day^{-1}) from incubation experiments of BTL sediments: Center (A) and Edge (B) cores. A steep decrease in rates with depth is observed in both the edge and center sites. Depth to permafrost beneath Big Trail Lake is thought to be 10 to 15 meters (Walter Anthony et al., 2020). CH_4 production rates (nmol CH_4 cm^{-3} day^{-1}) in GSL from incubation experiments of Center (C) and Edge (D) cores. A noticeable decrease in rates with depth is observed at both center and edge of the lake**.** The talik depth in GSL is estimated in the center to be between 15 to 40 m (see below).

Total thawed talik CH_4 production

The total profile-integrated CH_4 production rate throughout the thawed talik indicates the potential flux of CH_4 out of the sediment into the lake water column. This accumulated rate depends on CH_4 production rates at individual depths, facies thicknesses (surface sediments, taberites, *etc*.) and talik thickness. Since we did not have samples below 4 m depth, we extrapolated our data to the depth range of the known taberite thickness which was 10 to 15 m at BTL and ~16 at GSL (Walter Anthony *et al.*, 2020). Different fits were evaluated for calculating the total CH_4 production (Table S3-S5), and a power law regression was chosen as the best fit for extrapolating the total CH_4 production rate (Table S4, Fig. 4).

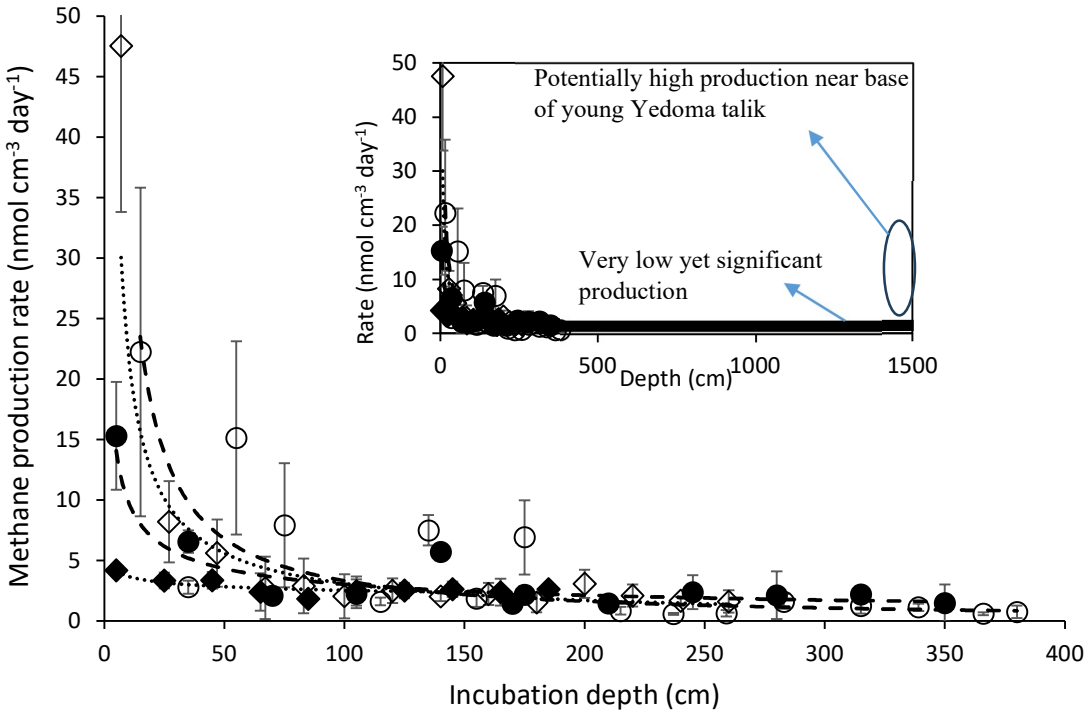

Figure 4: CH_4 production rates as measured in the different cores (symbols) and power law extrapolation (curves, equations in Table S5) down to 4 meters and 15 meters (upper right side). A constant low rates of less than 1 nmol cm^{-3} day^{-1} below 3 meters down to 15 meters is also marked in the upper right side, as well as the potential role of high production of CH_4 near of the base of the young talik.

The total 12 m column CH_4 production calculated for the BTL edge and center cores were about 8.5 (±12%) and 7.4 (±37%) mol m^{-2} $year^{-1}$, respectively. In GSL, talik thickness is greater (<40 m); however, the volumes of thawed silt are potentially

comparable at both lakes, and the main difference is in time since thaw. Because BTL is a younger lake, we assume the talik sediments have thawed within the last 70 years; whereas talik sediments beneath the center of GSL are thought to have been thawed for eight to nine centuries (Walter Anthony *et al.*, 2020). At GSL lake our 15 m profile-integrated CH_4 production rates from the center and the edge were 7 to 11 mol m^{-2} $year^{-1}$ (Table S4). Large uncertainties stem from the extrapolation (Table S5), the exceedingly long tail followed by the real measurements (Fig. 4), potential variability in the composition and thickness of the surface organic-rich sediments and the actual talik depth across both lakes. We also assume that the rates are extremely low down to the base of the talik, whereas there is a potential for high production rates near the base of the young taliks. These high rates together with the high surface rates in the edges of the lakes and the center of BTL can explain the ebullition there and the lack of ebullition at the center of GSL (Walter Anthony *et al.*, 2020).

The source of CH_4

The source of CH_4 in the sediment was estimated by comparing the $\delta^{13}C_{CH4}$ values in the profiles to those measured in the rate incubations (after purging and waiting several months for *in situ* production) (Fig. S1). The $\delta^{13}C_{CH4}$ values in the profiles from BTL were about -55 to -75 ‰ (Edge) and -60 to -70 ‰ (Center), whereas the values from the incubations of sediment from BTL were about 10 to 20 ‰ all along the profiles. The same observation is made in GSL, where $\delta^{13}C_{CH4}$ values of the dissolved CH_4 in the profiles were about -60 to -70 ‰ (Edge) and -70 to -80 ‰ (Center), with more positive values for incubations. The significant difference between the incubations and *in situ* profiles points towards an additional deep source of ^{13}C depleted methane in the natural environment, consistent with talik-sourced ebullition observations (Walter Anthony *et al.*, 2020).

Organic carbon characteristics in the sediments

The TOC profiles in the center of BTL sediments exhibited a gradual decline from 2% to less than 1% and in GSL from approximately 1% (Fig. S2). At the edge of the lakes higher TOC was observed near the top of the cores, up to 13% (BTL) and 9% (GSL) gradually decreasing with depth. The $\delta^{13}C_{TOC}$ in the center was constant near -27 ‰ in the upper 150 cm (Table S1). An increase to -25‰ was observed around 200 and 350

cm. At the edge, $\delta^{13}C_{TOC}$ values were around -28 ‰ at the upper part with an increase observed below 200 cm to -25 ‰.

Rock-Eval data obtained at both center and edge sites of both lakes exhibits a correlation between organic index values and sediment depth. The HI of the BTL was mostly much higher from that of the GSL and is indicative for a mix of Type I-III kerogen for the young lake (BTL), and a Type III kerogen for the mature lake (GSL). The OI on the other hand was extremely high for both lakes, exceeding 150 $mgCO_2\ g^{-1}TOC$. In both the edge and center cores of the BTL there was an inverse dependency between the HI and OI, suggesting that with the lose of H the OM became more oxidized. This relation is missing from the mature lake, in which the OM lost most of its H, presumably due to microbial degradation (Fig. S3). PC/RC ratio in both sites of BTL decreased with depth and stabilized at 250 cm, with the center core generally exhibiting lower ratios compared to the edge. The PC/RC ratio in the edge of GSL slightly decreased with depth. When comparing the two cores the lower values were observed at GSL center core, meaning it is more refractory than the edge (Fig. 5).

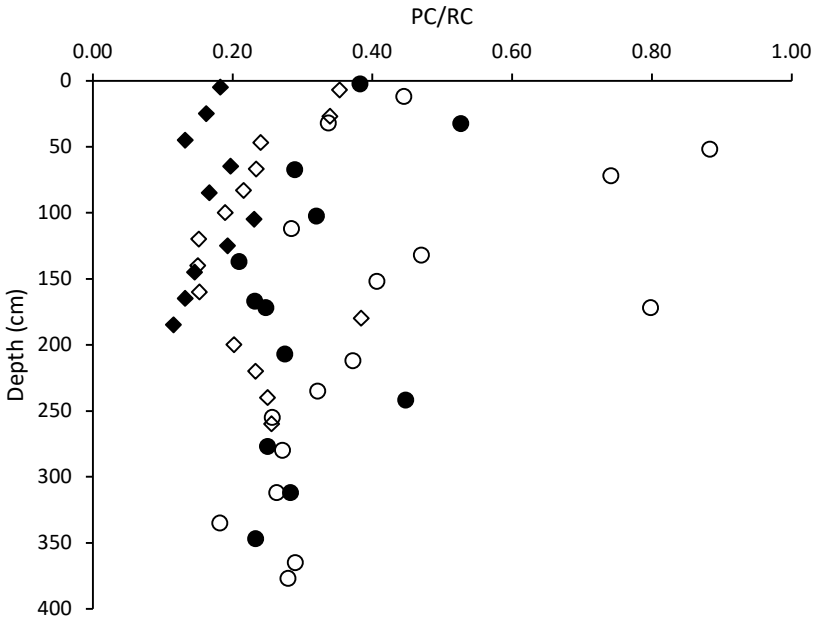

Figure 5: Sediment profiles of the ratio of pyrolysable carbon to refractory carbon (PC/RC). The ratio decreases not only with depth but also with the evolution of the lakes, as organic carbon becomes more refractory. The uncertainty of the data is smaller than symbols sizes (circles – BTL, diamonds – GSL, full symbols – Center, Empty symbols – Edge of lakes).

4. Discussion

Total CH_4 production in the talik

The CH_4 production rates observed in the upper sediments of this study are similar in magnitude to the ones observed in our previous studies (Lotem *et al.*, 2023; Pellerin *et al.*, 2022). Previous studies in which cores depth was limited to about one-meter depth were unable to provide a full understanding of the characteristics of CH_4 dynamics within thermokarst lake taliks. In Vault Lake, another thermokarst lake in central Alaska, a decrease in CH_4 production was observed with depth down to 6 m but CH_4 production rates were significantly higher than most reports (Heslop *et al.*, 2015). Freitas *et al.* (2025) also showed low, but significant cumulative anaerobic respiration throughout the taberal sediments down to the gravel zone at around 16 m in GSL, with scattered values below.

Since we were able to sample up to 4 m in the talik of both BTL and GSL and both in the center and the edge (near the lakeshore) locations, our rates measurements provide important confirmation of conceptual and numerical model predictions. They show high CH_4 production rates in the surface with steep decrease with depth in the talik as the taberite organic carbon becomes more refractory over time. Our findings are also consistent with the findings from 8 m permafrost sediment core from the Lena Delta where highest CH_4 production rates were observed in the first 125 cm (Wagner *et al.*, 2007).

The study also serves as the first empirical test of models predicting changes in CH_4 production rates with the evolution of lakes. It shows that CH_4 production rates are highest in the top sediments and are low, but still significant in the deep talik. We did not measure significant differences between CH_4 production rates at depths deeper than 100 cm at BTL and GSL (Fig. 4). However, other lines of evidence such as the different isotopic enrichments of the DIC pools strongly suggest that there should be differences in CH_4 production rates between BTL and GSL. It may be that the low rates and higher uncertainty on the measurements in the deep talik made it impossible to differentiate between the rates in the two lakes.

The long cores and the steep CH_4 production decrease with depth enable estimating the total production rates with higher certainty than the accumulated rates calculated in the 1 m cores and estimated constant rates along the talik (Pellerin *et al.*, 2022). It should

be noted that despite reaching down to 4 m in the talik, we still needed to extrapolate the rates to the entire thickness of thawed taberal sediments which is 10 to 16 m depths (Walter Anthony *et al.*, 2020). It has been suggested previously that CH_4 production rates may increase with depth of the talik because more recently thawed permafrost might release more labile organic carbon available for degradation (Walter Anthony et al., 2014) due to a rapid turnover time upon thaw (Schadel *et al.*, 2014; Shaver *et al.*, 2006). However, previous studies have not found a significant rise in CH_4 production rates with depth, except near the thaw front at the base of the young talik (Heslop *et al.*, 2015). This potential rise in young talik is marked in figure 4 and may explain the lowest accumulated production rate in the center of GSL. The recent study of (Freitas *et al.*, 2025) showed similar low values in the taberites overlying the sand/gravel zone in GSL at about 16 m and then scattered high values (in part because they were normalized to the total organics that was very low). To extrapolate our rates deeper into the talik, we experimented with several approaches such as using power law decay or constant rate that reflects the lower rates obtained in the incubations. Using the integrated rates with a power law equation, we calculated total talik CH_4 production (Table S4). We decided to present a power law fit since it indicates a declining rate in the deeper talik and reflects the overall trend observed in our incubations (Table S5). However, it is important to highlight that all models that we experimented with yield similar conclusions as to the total talik CH_4 production and the model used to extrapolate CH_4 production rates deep in the talik does not affect our conclusions.

An important finding is that facies thickness and talik depth play a significant role in determining total talik CH_4 production, and not just lake age and location within a lake. This is because despite measurably higher CH_4 production rates in the upper meter of sediment cores, as well as near the edges of the lakes, the low but relatively constant CH_4 production rates observed at depths in all the sites (5 to 10%), impact the integrated talik CH_4 production rates (Fig. 4).

The new total thawed talik CH_4 production rates are of the same magnitude as CH_4 emissions measured by previous studies in BTL (Elder *et al.*, 2021) and other discontinuous permafrost thermokarst lakes (Fig. 6). This similarity supports only a minor role for CH_4 oxidation (aerobically and anaerobically) in the lakes. It also fits the finding in the upper one meter of several thermokarst lakes, which shows that anaerobic

oxidation of CH_4 rates, as deduced from batch experiments, are two orders of magnitude lower than CH_4 production and not a significant sink of CH_4 (Lotem *et al.*, 2023).

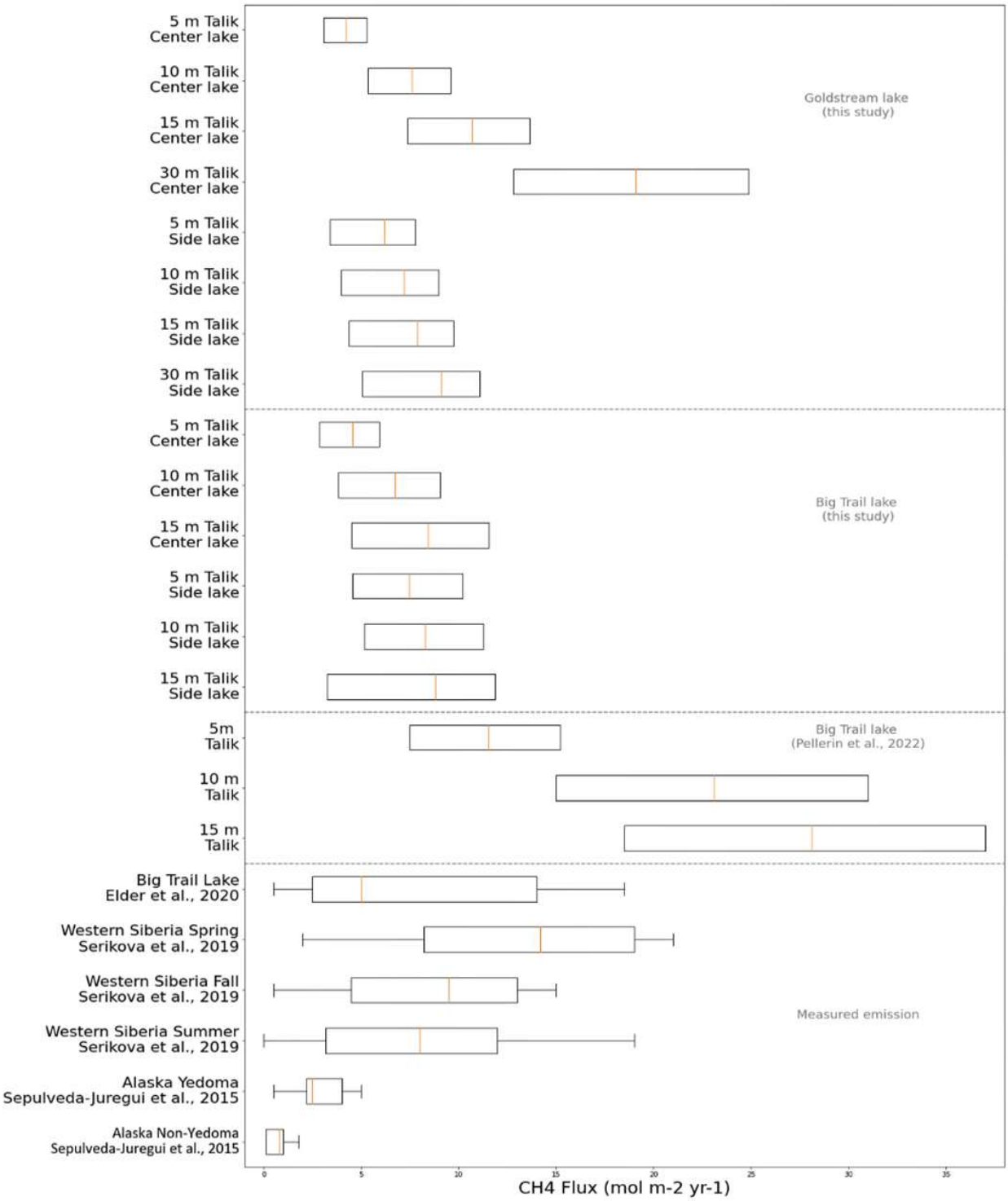

Figure 6: Total talik CH_4 production in BTL and GSL (mol m^{-2} $year^{-1}$), obtained by integrating the CH_4 production rates over the potential range of thawed talik depths in the sediments (from surface to the marked depth) using a power law extrapolation. The middle of the box corresponds to the mean estimate, and the uncertainty is the length of the boxes.

The implication of this observation is that as lakes mature total thawed talik CH_4 production rates can remain similar or even increase, although the carbon becomes less available for microbial degradation. This is when the talik deepens fast enough and offsets the overall drop in carbon lability. This is the case observed for both BTL and the eastern thermokarst margin of GSL. The question is whether there is control and a link between the organic carbon, its nature and CH_4 production rates in BTL and GSL, as discussed below.

Factors influencing CH_4 production rates and potential net emissions

We investigated further the controls of CH_4 production in BTL and GSL taliks. Those lakes are in the same valley system, it seems likely that they have the same sources, and that CH_4 production will be influenced from *in situ* difference. As the isotopic composition is much more sensitive for quantifying sedimentary processes rather than the concentration changes (*e.g.* Sivan *et al.*, 2011), $\delta^{13}C_{DIC}$ and $\delta^{13}C_{CH4}$ can be used to determine the controls on CH_4 production in the lakes. The first clear observation is that methanogenesis is strongly imprinted in the $\delta^{13}C_{DIC}$ of the young, recently thawed talik of BTL, and not in older, GSL talik, which has been thawed longer (Fig. 2). The $\delta^{13}C_{DIC}$ values of BTL are positive (around 10 to 20‰) as methanogenesis produces ^{13}C depleted CH_4 and, by mass balance, ^{13}C enriched DIC. The $\delta^{13}C_{DIC}$ values in the sediments of GSL are about -5 to -10‰ at depth (Fig. 2), as they are probably affected only by slight anaerobic oxidation of CH_4 and other organics with mechanism discussed by Lotem *et al.* (2023).

The $\delta^{13}C_{CH4}$ values of both lakes of -78 to -53‰ (Fig. 2) support this picture. They are typical to freshwater sediments, which are usually dominated by microbial acetoclastic methanogenesis (*e.g.* Whiticar, 1986). Methanogenesis pathways and their importance in this system were quantified by radiocarbon probing (Pellerin *et al.*, 2022) and by the $\delta^{13}C_{CH4}$ vs δD_{CH4} (Liu *et al.*, 2025). The relatively heavy $\delta^{13}C_{CH4}$ values suggest low CH_4 oxidation (*e.g.* Whiticar 1999; Sivan *et al.*, 2011), which results in ^{13}C enrichment in CH_4. The control of methanogenesis on BTL profiles can be evident from looking at the $\delta^{13}C_{CO2}$ values and plotting them against $\delta^{13}C_{CH4,}$ similarly to Krause and Treude (2021) (Fig. S7). The data support acetoclastic methanogenesis and CH_4 oxidation in all sites.

The main difference between the lakes is likely related to the fundamental difference in age of the two taliks, which influence the lability of organic carbon and the resulting biogeochemical cycles. The lability difference of the organics was further investigated by assessing the properties of the organic carbon in the talik of BTL and GSL.

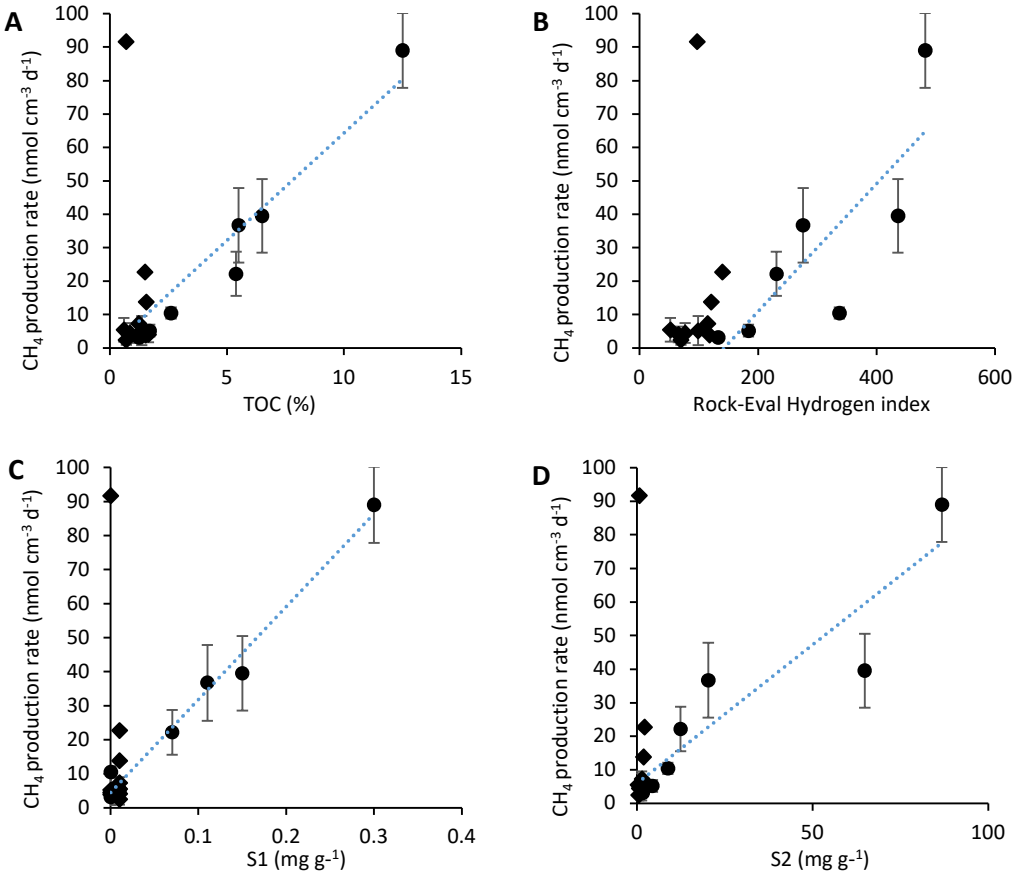

Figure7: Methane production rates (including its uncertainty) as measured in the top upper 100 cm from both center and edge cores compared with TOC content (A), hydrogen index (B, R2=0.67) and the most labile organic carbon compounds: S1, S2 (C, D), as measured on Rock-Eval. A positive relationship (R^2 0.67 to 0.98) is evident in the upper 100 cm for BTL (circles) but not for GSL (diamonds).

Methane production rates are highly correlated to TOC and indices of thermal induced lability in the upper sediments (Fig. 7A) but not correlated with them in the deep talik of BTL (Fig. S4-S5). The correlation is between two independent methods that estimate lability of organic carbon to microbial degradation - the lability based on thermal degradation and the CH_4 production rate. The most labile fraction of pyrolyzed carbon (S1, S2) are correlated with CH_4 production rate in both center and edge sites (Fig. 7B-D). The results demonstrate that the highest CH_4 production rates observed in our study are likely due to both the lability and quantity of organic matter.

The pyrolyzed carbon to residual carbon ratio (PC/RC) is a simple measure of how labile the organic carbon is. As the PC/RC ratio higher, the sample has more hydrogen, and has more pyrolysable fraction (Carrie *et al.*, 2012). Throughout the four cores taken for this study, the RC/PC ratio was strongly dependent on depth in the talik (Fig. 5), which is consistent with older, less labile organic matter deeper in the sediment because of depositional history. However, the most interesting observation was the decreasing trend of lability to thermal induced reactions between sites. BTL edge had the highest PC/RC values which correspond to organic carbon with the highest lability, followed by BTL center, GSL edge, with the lowest value observed at the GSL center (Fig. 5). The lake edges exhibit highest PC/RC ratios compared to lakes centers, suggesting that the centers of the lakes contain more refractory organic carbon, which may be a result of the edges of the lakes being "younger" (the age since thaw). Alternatively, it could be that the edges have additional input of organic carbon from vegetation or runoff from land, as supported by the labile fractions found in the edges (Fig. 7C). Both edge and center of GSL, on average, had lower PC/RC ratios than both the center and edge of BTL. This is interpreted as GSL containing a greater fraction of less labile organic carbon, consistent with the longer time since the permafrost thawed and formed the talik and lake at this site. Carbon thus becomes less labile for microbial degradation as the lake evolves. In permafrost environments, organic carbon lability is not always correlated directly with what is termed the "age" of the organic carbon such as in most marine sediments where lability, depth and age are often directly correlated. In the talik of thermokarst lakes, time since thaw seems to have some control on the lability of the organic carbon and exerts control on CH_4 production rate. All together, our study emphasizes the potential use of the lability of thermal induced reactions as a proxy for organics lability for methanogenesis.

It seems thus that at the onset of thawing, during the formation of a young thermokarst lake a high amount of highly labile organic carbon accumulates and begins to degrade. As thawing progresses, the lakes margins expand and deepen and the refractory organics remain, leading to lower CH_4 production rates. A proposed conceptual mechanism is presented in figure 8. This mechanism can be tested in future studies, which include expanding the study to additional lakes, incorporating seasonal sampling, and integrating microbial community analyses to better understand CH_4 cycling processes.

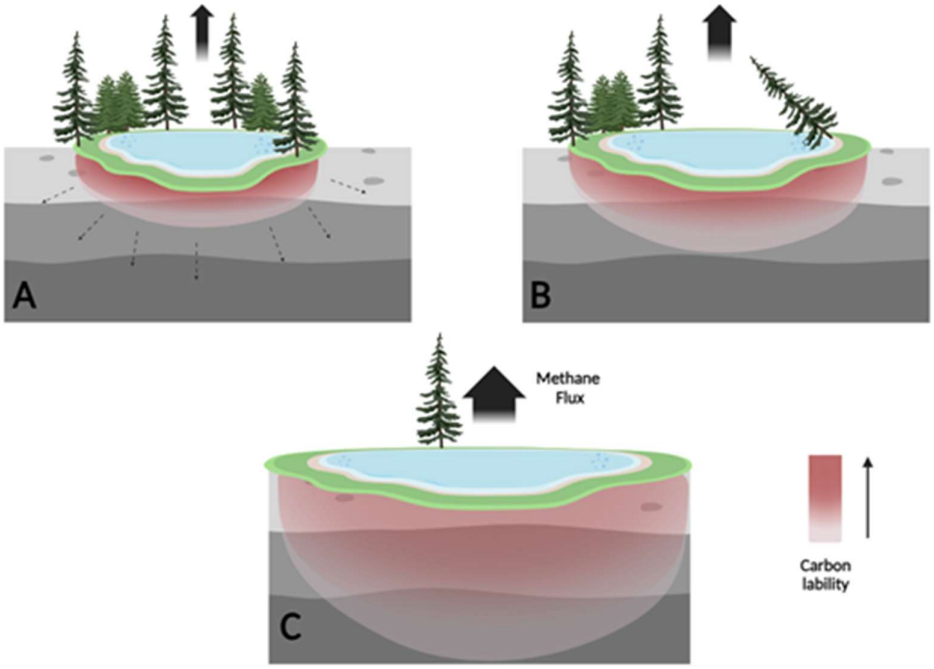

Figure 8: Proposed conceptual mechanism of OM lability and CH_4 emission as thermokarst lakes mature. Laboratory incubations support the conceptual and numerical models of decreasing carbon lability leading to lower CH_4 production as Yedoma thermokarst lakes mature. The gray sediment denotes Yedoma permafrost soil. Red scale color denotes organic carbon lability with talik expansion into in-situ thawed Yedoma (taberites). **(A)** Young active lake: Labile carbon results in high CH_4 production rates with CH_4 production in taberites proportional to the talik thickness. **(B)** Expanding lake: Over decades new organic carbon from surrounding soils and plants is added to surface lake sediments and to freshly thawed taberites at the base of the talik, while the availability of labile organic carbon attenuates in formerly thawed taberites. **(C)** Mature lake: Over centuries the talik deepens (into bedrock) beyond the depth of taberites, while taberite organic carbon becomes refractory. CH_4 emissions may still be significant, depending on exogenous (non-taberite) inputs of organic carbon. At later stages (as suggested by Walter Anthony *et al.*, 2014), the organic carbon is no longer available for methanogenesis and the lake thus is no longer a significant source for permafrost-derived CH_4 to the atmosphere.

Methane origin in the talik

The comparison of $\delta^{13}C_{CH4}$ values between the profile measurements and those obtained from the incubation experiment shows less enriched $\delta^{13}C_{CH4}$ values in the sediment cores across all samples (Fig. S6). These values in the sediment profiles show that the pore water CH_4 is not entirely produced *in situ*, suggesting that the new CH_4 produced in the lake sediments contains a more isotopically enriched $\delta^{13}C_{CH4}$ values compared to the CH_4 diffused from deeper layers, which could be produced by a

different CH_4 production pathway (Pellerin *et al.*, 2022; Liu *et al.*, 2025). CH_4 generated at greater depths within the talik, which subsequently ascends through diffusion or bubble transport, exhibits a more negative $\delta^{13}C_{CH4}$ probably due to different conditions from those prevailing in the top meter of the talik, possibly under lower metabolic rates and using different metabolic pathways (Maltby *et al.*, 2016; Berberish *et al.*, 2020; Pellerin *et al.,* 2022).

By considering the depleted $\delta^{13}C_{CH4}$ values in the sediment cores and the corresponding incubation values at each depth, we can quantitatively assess the fraction of modern CH_4 present in the sediment profile. A comprehensive analysis of the young production fraction in all four cores shows that as depth increases, the contribution of newly produced CH_4 decreases and tends to approach zero across all cores (Fig. S6). Additionally, the new CH_4 fraction observed in Goldstream Lake (both edge and center) is smaller compared to that of BTL. This finding suggests a greater influence of newly produced CH_4 in younger lakes compared to those that have formed and thawed longer. A shift in methanogenesis pathway in the sediments vs the incubations can be an alternative explanation, however less likely, given the observations here and in the previous studies (Pellerin *et al.*, 2022).

5. Conclusions

This study presents the first empirical data quantifying CH_4 production and organic carbon degradation of thermokarst lakes from young to mature. This was achieved by quantifying the evolution of organic carbon degradation and CH_4 production rates throughout the evolution of lakes from a young dynamic lake to a mature one. The findings underscore the vertical variations in CH_4 production rates, the influence of permafrost thawing on microbial activity, and the divergent patterns observed among lakes of different development stages.

Our high-resolution profiles and long-term incubations show highest CH_4 production rates on the edges of the young lake BTL, then the center of BTL, then the edge of mature GSL and the lowest at the center of GSL. The higher rates coincided with higher TOC levels, more labile carbon content for thermal induced chemical reactions, simpler carbon compounds, and a young CH_4 source. These factors provided probably favorable conditions for microbial populations to decompose carbon, resulting in elevated CH_4 production. The high correlation between the two different methods to estimate organic

carbon lability for microbial degradation CH_4 production rates and the lability to thermal induced chemical reactions, emphasizes the potential use of the Rock-eval analyses to estimate the susceptibility of organic carbon for microbial degradation.

While the higher rates in the upper part of the young lake, the increase in talik depth also play a significant role in determining the total CH_4 production rate. Our proposed conceptual mechanism, as depicted in figure 8, considers these two parameters (lake age and thawed talik thickness) when discussing accumulated CH_4 production rates. It can be expected thus that the expansion of thermokarst lakes in the Arctic will continue to influence CH_4 production as the younger lakes expand into deeper permafrost layers.

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Authors contribution

YG, AP, EER and OS participated in sampling campaigns led by KWA and NH. YG conducted incubation experiments. EER and YG measured geochemical samples and processed the data. NH conducted geophysical measurements. OS led manuscript writing with AP. YOR was responsible for Rock Eval6 method development and with AP and OS interpreted the data.

Conflict of interest

The authors declare that they have no conflict of interest.

SUPPORTING INFORMATION

Additional supporting information may be found in the online version of the article at the publisher's website.

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