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Key Points:
- Hypolimnetic methane storage was positively related to the mean summer rate of gross primary productivity across five lakes and 24 lake-years
- Yet within each lake, there was no relationship between hypolimnetic methane storage and mean summer gross primary productivity
- Predictions of lake methane responses to shifting trophic status should incorporate varying lake responses and the temporal scale of change

Supporting Information:
- Supporting Information S1

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Methane and Primary Productivity in Lakes: Divergence of Temporal and Spatial Relationships

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Abstract In lakes, the production and emission of methane (CH_4) have been linked to lake trophic status. However, few studies have quantified the temporal response of lake CH_4 dynamics to primary productivity at the ecosystem scale or considered how the response may vary across lakes. Here, we investigate relationships between lake CH_4 dynamics and ecosystem primary productivity across both space and time using data from five lakes in northern Wisconsin, USA. From 2014 to 2019, we estimated hypolimnetic CH_4 storage rates for each lake using timeseries of hypolimnetic CH_4 concentration through the summer season. Across all lakes and years, hypolimnetic CH_4 storage ranged from <0.001 to 7.6 mmol CH_4 m^{-2} d^{-1} and was positively related to the mean summer rate of gross primary productivity (GPP). However, within-lake temporal responses to GPP diverged from the spatial relationship, and GPP was not a significant predictor of interannual variability in CH_4 storage at the lake scale. Using these data, we consider how and why temporal responses may differ from spatial patterns and demonstrate how extrapolating cross-lake relationships for prediction at the lake scale may substantially overestimate the rate of change of CH_4 dynamics in response to lake primary productivity. We conclude that future predictions of lake-mediated climate feedbacks in response to a shifting distribution of trophic status should incorporate both varying lake responses and the temporal scale of change.

Plain Language Summary Many lakes produce substantial amounts of methane, a potent greenhouse gas. Previous research has found that more methane is produced from lakes with high algal biomass. However, little is known about how methane dynamics from a single lake respond to annual changes in algal biomass. By examining lake methane dynamics and metrics of algal biomass from five lakes across 5 years, we find that within-lake responses of methane dynamics do not align with the across-lake patterns we see when comparing different lakes. Within-lake methane responses were different between lakes and were undetectable in some lakes. Understanding the temporal scale of how lakes respond to changes in algal biomas is important for predicting the role of lakes for producing methane under future environmental change scenarios.

1. Introduction

Lakes are important, but often overlooked, contributors to atmospheric greenhouse gas concentrations, in part due to the emission of methane (CH_4) gas (Bastviken et al., 2011; DelSontro et al., 2018; Wik et al., 2016). CH_4 is a potent greenhouse gas with 34 times the global warming potential of carbon dioxide (CO_2) and, in lakes, is primarily produced as a product of anaerobic decomposition in sediments (Bastviken et al., 2004). Previous research has made important advances in estimating lake contributions to atmospheric CH_4 concentration (Anthony et al., 2010; Bastviken et al., 2011; Beaulieu et al., 2019) and has highlighted a positive relationship between lake trophic status and increases in CH_4 production and emissions (Deemer et al., 2016; DelSontro et al., 2018; Huttunen et al., 2003; West et al., 2016).

The relationship between lake CH_4 dynamics and trophic status is often attributed to a combination of both extreme anoxic conditions common in productive systems and an increase in autochthonous organic matter settling to sediments fueling anaerobic decomposition. Field surveys consistently report positive relationships between proxies for lake trophy, such as chlorophyll *a* concentration and CH_4 production and emissions across the landscape (Deemer et al., 2016; DelSontro et al., 2018; West et al., 2016). Additionally, experiments confirm that CH_4 production from lake sediments increases with algal biomass input (Sepulveda-Jauregui et al., 2018; West et al., 2012, 2015), indicating a clear role of organic matter supply for fueling methanogenesis. However, cross-system comparative studies and laboratory experiments may not fully represent how a single lake responds to temporally varying primary productivity at the ecosystem scale.

Both comparative studies and laboratory experiments often consider large differences in trophic status, which is helpful for understanding patterns across the landscape or potential mechanisms driving methanogenesis, but is not necessarily representative of the shifts in lake primary productivity over time. Given inter-annual variation in temperature, precipitation, and other factors that influence lake primary productivity, changes in trophic status are often assessed at the decadal time scale (Leech et al., 2018; Schindler et al., 2012). Additionally, studies which provide a snapshot of lake trophic status and CH_4 dynamics do not consider time lags that are likely present in organic matter sedimentation and biogeochemistry (Bain et al., 2012; Winfrey & Winfrey & Zeikus, 1978). For example, sediment mixing processes, such as sediment focusing and lateral transfer by wind or wave action, causes modern-day production of organic matter to mix with legacy autochthonous and allochthonous organic matter in sediments (Larson & MacDonald, 1993). This integrative nature of sedimentation has the potential to create complex relationships or obscure relationships between modern-day primary productivity and CH_4 dynamics. Thus, further research is needed to quantify the temporal change in lake CH_4 dynamics and determine whether and how it responds to shifts in primary productivity at the ecosystem scale.

Previous studies considering lake CH_4 responses to primary productivity have often focused on either lake CH_4 production or emission (Deemer et al., 2016; DelSontro et al., 2018; West et al., 2012, 2016). In lakes, CH_4 is produced primarily in pelagic anoxic sediments and is then emitted from the lake ecosystem through ebullition and diffusive emissions (Bastviken et al., 2008). These are useful components to consider as they represent the beginning (production) and end (emissions) of the CH_4 cycle in lakes. However, because production and emission are both spatially and temporally heterogeneous within a lake (Loken et al., 2019), quantifying temporal change is very labor intensive and rates of CH_4 production and emission are often only measured in a fraction of the ecosystem (West et al., 2016).

CH_4 stored in the hypolimnion over the course of a stratified season (hereafter referred to as CH_4 storage) is a useful metric for studying the relationship between CH_4 dynamics and lake productivity, because it captures processes at the ecosystem scale and it often covaries with other aspects of the CH_4 cycle (Bastviken et al., 2004). In dimictic lakes common to temperate and boreal regions, CH_4 is stored in the hypolimnion layer during the stratified season, when the temperature and density of lake water prevents mixing between the top and bottom layers (Bastviken et al., 2004). During autumn mixing, this previously stored CH_4 is often emitted and can be a significant fraction of the total annual CH_4 emissions of a lake (Encinas Fernández et al., 2014; Kankaala et al., 2007; Riera et al., 1999). However, CH_4 oxidation, ebullition, and diffusion all cause loss from CH_4 storage prior to autumn mixing, and the magnitude of loss is likely related to the physical and chemical variables that influence water column stability (Bastviken et al., 2004). Despite this, CH_4 storage is integrated over the length of the stratified season and all hypolimnetic sediments, and thus less subject to spatial and temporal heterogeneous than measures of diffusive emissions or ebullition.

In the present study, we measured rates of CH_4 storage, lake primary productivity, and other lake chemical and physical characteristics from five freshwater lakes across 5 years. First, we compare estimates of hypolimnetic CH_4 storage to other previously described CH_4 processes, including production and emission from these lakes. Second, we test the relationship between lake CH_4 storage and primary productivity both across lakes and within lakes. We hypothesized that there would be significant inter-annual variation in CH_4 storage and that it would be correlated with yearly differences in lake primary productivity both across and within lakes. Finally, we consider how temporal changes in CH_4 dynamics may differ from spatial patterns and evaluate the consequences of substituting space for time in predictive models of lake CH_4 dynamics under changing lake primary productivity.

2. Materials and Methods

2.1. Study Site

We sampled five north-temperate, freshwater lakes at the University of Notre Dame Environmental Research Center (UNDERC) near Land O'Lakes, WI, including Crampton (CR), East Long (EL), Hummingbird (HB), Morris (MO), and West Long (WL). Lakes are all within an area of 25 km^2 and were sampled from 2014 to

2019, excluding 2017 due to equipment failure. EL was also not sampled during 2019 due to a separate whole-lake manipulation. These lakes span a gradient of hydrology, size, and productivity (supporting information Table S1) and are situated in watersheds of primarily wetland and forest ecosystems. This region has an average summer (1 May to 15 September) air temperature of 17.1°C and average total summer precipitation of 509.8 mm, estimated from a meteorological station in WL from 2014 to 2019 (Solomon et al., 2018). Additionally, lakes are comparable to the many small, slightly acidic lakes that dominate the north temperate region (Hanson et al., 2007).

2.2. Hypolimnetic CH_4 Storage Rates

In each lake, we measured CH_4 concentration from one point in the middle of the hypolimnion at the deepest point in the lake from 1 May to 15 September every year. Because of a concentration gradient in which CH_4 often increases from the bottom of the thermocline to the sediments (Bastviken et al., 2008), we sampled the middle depth of the hypolimnion as a conservative estimate of hypolimnetic CH_4 concentration. This single point measurement does not account for horizontal variability in the hypolimnion, which may lead to error in our estimates of CH_4 storage. However, previous research has considered sampling at the deepest point in the lake to be a useful measurement for comparing across lakes and years (Bastviken et al., 2008; Encinas Fernández et al., 2014).

During each sampling event, we identified the hypolimnion depth using water temperature profiles, in which changes in water temperature of more than 1°C identified the depth of thermal stratification. We collected replicate ($n = 2$) water samples from one depth in the middle of the hypolimnion using a Van Dorn water sampler with an airtight valve. In most lakes and years, the middle of the hypolimnion was constant (Table S1). In CR, the hypolimnion sample depth was more variable, but was only correlated with time in 1 year. Immediately following sampling, we performed headspace equilibrations of water samples and determined CH_4 concentrations on an Agilent 6890 Gas Chromatograph as described in West et al. (2016).

To estimate annual summer rates of lake CH_4 storage for each lake, we regressed hypolimnion CH_4 concentration with day of year and included lake and year as fixed effects: CH_4 concentration~day * lakeID * year. We inferred CH_4 storage rates from the slope of the relationship, accounting for lake and year effects, and converted volumetric rates to areal rates (mmol CH_4 m^{-2} day^{-1}) to account for differences in hypolimnion volume.

Finally, to compare estimates of CH_4 storage to other previously described rates of CH_4 production and emissions from these lakes, we utilized estimates measured during a single year of our study (West et al., 2016).

2.3. Gross Primary Productivity

We estimated gross primary productivity (GPP) daily in each lake using the open water diel dissolved oxygen method as described in Solomon et al. (2013). High-frequency dissolved oxygen (DO) sensors (miniDOT Logger, PME Incorporated) and temperature sensors (Onset HOBO Pendants, Onset Computer Corporation) were deployed at the deepest point in each lake and logged on 10-min intervals. DO sensors were deployed just below the water surface at 0.5 m. Temperature sensors were deployed throughout the epilimnion and into the hypolimnion, starting at 0.1 m and increasing in depth by 0.25 m increments. Meteorological data, including wind speed, wind direction, air temperature, PAR, relative humidity, and barometric pressure, were collected from a single floating platform sensor station (Onset HOBO Met Station, Onset Computer Corporation) deployed at the deepest point in West Long (WL) lake. Rates of GPP (mmol C m^{-2} day^{-1}) were converted from units of oxygen to units of carbon by assuming a respiratory quotient of 1 and reported in areal rates based on epilimnion layer depth to account for differences in epilimnion volume across lakes.

2.4. Statistical Analyses

To determine whether rates of CH_4 storage were correlated with lake primary productivity across all years and lakes, we first fit a linear regression between the summer rates of CH_4 storage and the mean summer GPP rates. To determine lake-specific responses to GPP, we fit a second model using an analysis of covariance (ANCOVA) design: CH_4 storage~GPP * lakeID. A significant GPP effect would indicate an overall relationship between CH_4 storage and GPP across all lake and years, while a significant lake ID effect would indicate that lake characteristics alter the long-term mean rate of CH_4 storage for each lake. Additionally,

Table 1
Coefficient of Determination (R^2), p Value, and Akaike Information Criterion (AIC) Score of Candidate Models for Predicting Lake Hypolimnetic Methane Storage as Functions of Mean Annual Gross Primary Productivity (GPP) and Lake ID

Predictors	R^2	p value	AIC	Ecological interpretation
Lake ID	0.54	0.003	16.95	CH_4 storage is not related to mean annual GPP. Lake characteristics alter long-term mean CH_4 storage.
GPP + Lake ID	0.54	0.009	18.93	CH_4 storage is related to mean annual GPP similarly across all lakes, but lake characteristics alter the long-term mean rate of CH_4 storage.
GPP	0.28	0.008	22.02	CH_4 storage is related to mean annual GPP similarly across all lakes. Lake characteristics do not alter long-term mean CH_4 storage.
GPP * Lake ID	0.61	0.06	23.19	CH_4 storage is related to mean annual GPP, but lake characteristics alter both the long-term mean rate of CH_4 storage and the response of CH_4 storage to GPP.

Note. Models are ordered by increasing AIC score.

a significant interaction between GPP and lake ID would indicate that lake characteristics also alter the relationship between CH_4 storage and GPP for each lake. We thus inferred lake-specific responses to GPP by summing the overall effect of GPP and the lake-specific interaction effects from the ANCOVA model.

Finally, to identify the best predictive model for rates of CH_4 storage, we conducted a stepwise regression using the *stepAIC* function from the *MASS* package (Venables & Ripley, 2002), which evaluates model fit based on Akaike information criterion (AIC) scores. Candidate models were all subsets of the ANCOVA model and are outlined in Table 1 with the ecological interpretation described. Use of AIC scores also allowed us to compare candidate models that were not subsets of each other.

All statistical analyses were performed in R (R Core Team, 2018). Data and code are available at the Zenodo repository (https://doi.org/10.5281/zenodo.3946997) and in Solomon et al. (2018).

3. Results

3.1. Hypolimnetic Methane Storage Rates

Hypolimnetic CH_4 concentration increased linearly across the summer season in all lakes, and rates of CH_4 storage were highly variable both across lakes and across years (Figure 1). CH_4 storage rates ranged from <0.001 mmol CH_4 m^{-2} d^{-1} to 7.6 mmol CH_4 m^{-2} d^{-1} and were highest on average in MO and lowest in CR (Table S1). In CR lake, CH_4 storage rates were largely driven by late season accumulation in 2014 and 2018, evident by an increase in CH_4 concentration (Figure 1). Technical replicates support these late-season observations in CR, and analyses with or without these data did not change the overall results of the study, as variation in CH_4 storage rates in CR lake was much lower than any other lake (Table S1).

In all lakes, CH_4 storage represented an important component of the overall CH_4 budget. Compared to emissions estimated during a single year of our study (West et al., 2016), CH_4 storage was higher than emission estimates in all lakes (Figure 2). However, the average proportion of CH_4 production that could be accounted for by CH_4 storage was quite variable between lakes, ranging from 1.0% to 43.8%. Rates of CH_4 oxidation were not measured and may at least partially account for the difference between production and other CH_4 fates (storage and emissions).

3.2. Gross Primary Productivity

Across all lakes and years, mean summer GPP also varied substantially, ranging from 14.9 to 58.9 mmol C m^{-2} d^{-1} (Figure S1; Table S1). At the lake scale, interannual variation in mean summer GPP was greater in some lakes than others. In EL, HB, and WL, the range of GPP was approximately 10 mmol C m^{-2} day^{-1}, while in MO it was 31.5 mmol C m^{-2} day^{-1}, and in CR 16.7 mmol C m^{-2} day^{-1}. Additionally, mean summer GPP was significantly different across lakes ($F_{20,4} = 7.71$, $p < 0.05$); however, the ranges did overlap between lakes. Finally, mean summer GPP was not significantly different across years ($F_{20,4} = 0.11$, $p = 0.97$), indicating changes in GPP were not synchronous across lakes and presumably not driven by interannual variation in temperature and precipitation.

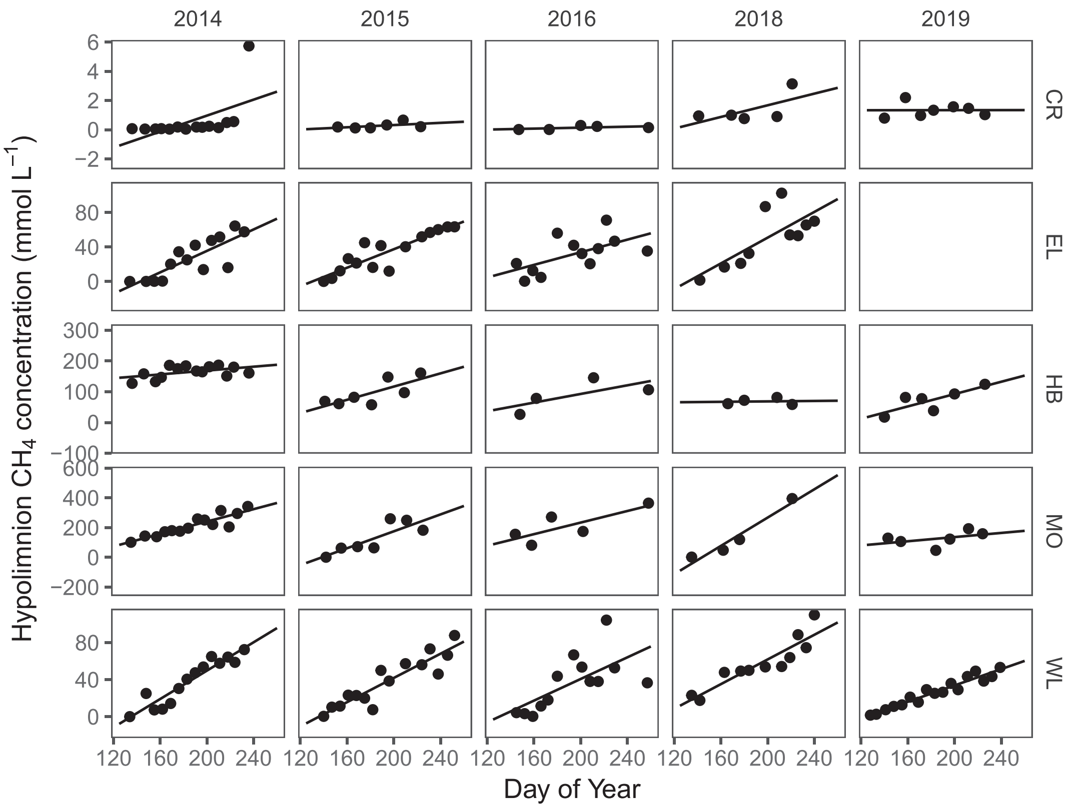

Figure 1. Hypolimnion methane (CH_4) concentration (mmol L^{-1}) from May–September in five lakes across 5 years. Data were collected from the middle of the hypolimnion in each lake, approximately weekly or biweekly through the length of the summer season. Black lines represent the lake-year specific relationships between hypolimnetic CH_4 concentration and day of year that were estimated jointly from a multiple regression accounting for lake and year effects.

3.3. Influence of Gross Primary Productivity on CH_4 Storage

As hypothesized, we found that rates of CH_4 storage were significantly positively related to mean annual GPP across all lakes and years ($R^2 = 0.28$, $p < 0.05$; Figure 3a). However, differences in CH_4 storage across years within a lake were not related to GPP; neither the main GPP effect ($F_{14,1} = 0.01$, $p = 0.89$) nor the interaction between GPP and lake ID ($F_{14,4} = 0.58$, $p = 0.67$) was significant, indicating that within-lake temporal differences in CH_4 storage were not related to GPP. Instead, lake ID was the only significant predictor of CH_4 storage ($F_{14,4} = 4.88$, $p < 0.05$). In addition, the stepwise regression analysis revealed that the lake-specific intercept model best predicted CH_4 storage based on AIC scores (Table 1; Figure 3b).

4. Discussion

CH_4 emissions from freshwater lakes are an important contributor to atmospheric greenhouse gas concentrations, especially in the context of increasing eutrophication of inland waters. Here, we reported new estimates for rates of CH_4 storage in five lakes across 5 years and showed that storage is a substantial fraction of the lake CH_4 budget (Figure 2). Additionally, across all lake-years in our dataset, we saw a positive relationship between lake primary productivity and CH_4 storage (Figure 3a), supporting previously described spatial patterns. However, our findings also suggest that primary productivity is not a reliable predictor of temporal CH_4 dynamics within a given lake (Table 1). Thus, we conclude that temporal responses of lake CH_4 storage to changes in primary productivity diverge from the spatial pattern and, at time scales of 5 year or less, are dampened in comparison.

CH_4 storage was a meaningful component of the overall CH_4 budget in each lake and should be considered when determining the influence of primary productivity on lake CH_4 dynamics. CH_4 storage rates varied significantly across lakes and across years and were within the range of previously reported rates for similar

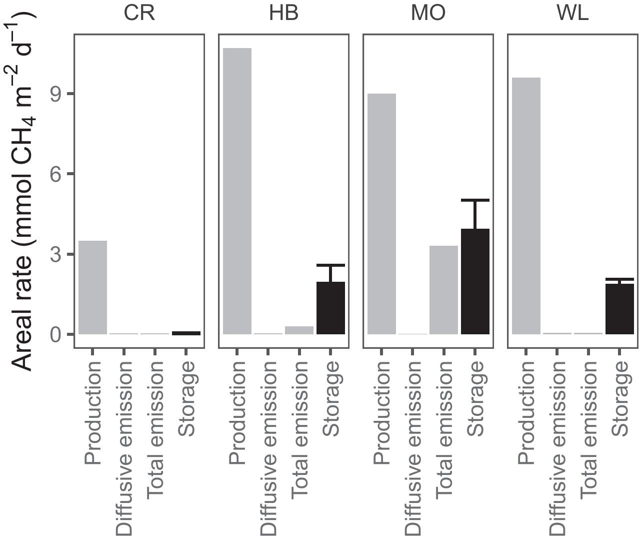

Figure 2. Estimated average rates of methane (CH_4) processes for four freshwater lakes: Crampton (CR), Hummingbird (HB), Morris (MO), and West Long (WL). CH_4 storage is a substantial fraction of the overall CH_4 budget CH_4 compared to estimates of CH_4 diffusive or total emissions in all four lakes. Gray bars denote processes previously estimated in West et al. (2016). Black bars denote long-term mean CH_4 storage estimated in the present study with error bars representing one standard error. CH_4 oxidation rates were not measured and may account for the difference between CH_4 production and fates. East Long (EL) is excluded as it was not included in the 2016 study.

lakes (Bastviken et al., 2008; Encinas Fernández et al., 2014; Michmerhuizen et al., 1996). Additionally, even after considering that up to 54–88% may be oxidized during autumn turnover mixing (Encinas Fernández et al., 2014; Kankaala et al., 2006, 2007; Utsumi et al., 1998), rates of CH_4 storage were comparable to other pathways of emission (Figure 2).

Previous studies have identified a relationship between lake primary productivity and CH_4 dynamics (Deemer et al., 2016; DelSontro et al., 2018; West et al., 2016) and have suggested that ongoing eutrophication of inland waters will substantially increase lake CH_4 emissions (Beaulieu et al., 2019; West et al., 2015). However, previous work used proxies for lake trophic status, and few studies have quantified these relationships with direct measures of lake primary productivity. In line with these observations, we found that CH_4 storage rates were positively correlated with mean summer GPP across all lakes and years. The slope of this relationship can be interpreted as a conversion efficiency of GPP-C to CH_4-C of 0.085 (Figure 3a). However, we encourage caution in interpreting and extrapolating these estimates, as mean summer GPP was not a reliable predictor of within-lake variation in CH_4 storage (Table 1; Figure 3b). At the lake scale, the response of CH_4 storage to changes in GPP varied. We note that the range of GPP in our study lakes is smaller than that of other studies (DelSontro et al., 2018), which may result in the dampened interactions we see here. However, lakes did span a meaningful range of GPP (doubling in most lakes over the course of the 5 years), and so other mechanisms should also be considered for understanding the disconnect between spatial patterns and temporal responses at this scale.

Within-lake temporal responses to GPP may be related to other lake physiochemical variables that influence CH_4 dynamics, including sediment temperature (DelSontro et al., 2018; Duc et al., 2010; Zeikus & Winfrey, 1976), extent of anoxia (Encinas Fernández et al., 2014; Liu et al., 1996), and lake bathymetry (West et al., 2016). While sediment temperature is roughly consistent across our stratified study lakes, both the extent of anoxia and lake bathymetry varied between lakes and have the potential to influence rates of CH_4 storage and its response to primary productivity. The extent of anoxia strongly influences both CH_4

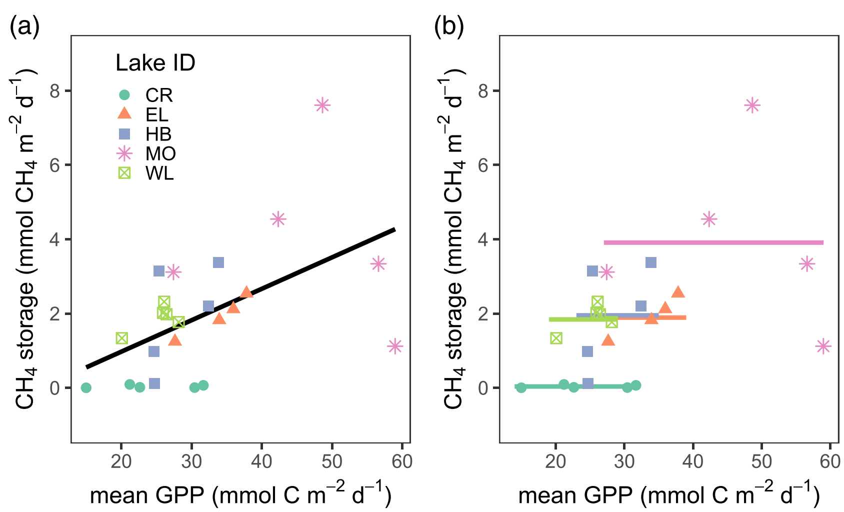

Figure 3. (a) Rates of hypolimnetic methane (CH_4) storage are significantly positively related to the rates of mean summer gross primary productivity (GPP) across all lakes and years ($R^2 = 0.28$, $p < 0.05$). (b) However, a lake-specific intercept-only model best predicts CH_4 storage based on coefficient of determination (R^2) and Akaike information criterion (AIC) score ($R^2 = 0.54$, $p < 0.05$; Table 1).

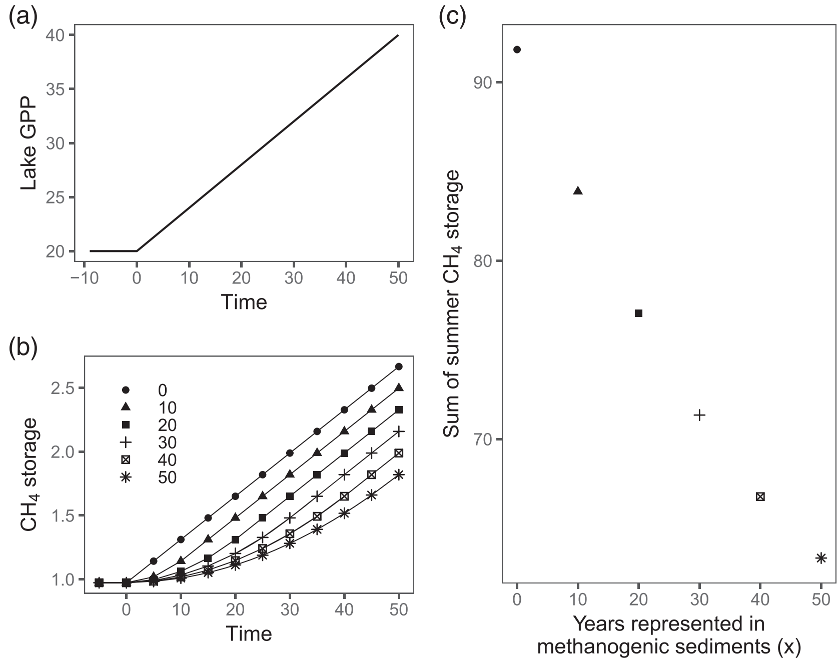

Figure 4. (a) Simulation experiment to evaluate the consequence of mean summer gross primary productivity (GPP) doubling from a historical state over 50 years. (b) Rate of CH_4 storage predicted using simulated GPP and Equation 1, where shape indicates different values for the number of years represented in the active methanogenic zone of sediments (x). (c) The sum of summer CH_4 storage rates over 50 years accounting for different extents of active methanogenesis in the sediment.

production (Sobek et al., 2009) and oxidation (Encinas Fernández et al., 2014)—when oxygen is depleted, production increases, and oxidation decreases—while lake depth is negatively related to the probability of ebullition and loss from hypolimnion storage (West et al., 2016). Thus, it could be hypothesized that deep, strongly stratified lakes would have the highest rates of CH_4 storage and the strongest response to primary productivity, while shallow, less stratified lakes would have lower rates of CH_4 storage and be less prone to changes in primary productivity. Interestingly, in our study, EL lake was the only lake in which CH_4 storage was positively related to interannual variability in GPP (Figure 3; blue points) and is both deep and strongly stratified compared to the other lakes (Table S1). However, further research is needed to confirm the extent to which anoxia and bathymetry interact with lake primary productivity across different lakes and to quantify the influence of these factors on lake temporal responses.

Lake-specific temporal responses in CH_4 storage may also diverge from spatial patterns due to the temporal scale at which sediments change. Mixing of surface sediments by gravity, waves, and currents causes both vertical and horizontal mixing, often down to 1 cm (Larson & MacDonald, 1993; Robbins, 1982). In many small, temperate lakes, 1 cm of surface sediment can account for the last 2–5 years of sediment deposition, dependent on lake sedimentation rates and these physical mixing dynamics (Leavitt & Carpenter, 1989). Additionally, in anoxic pelagic sediments, methanogenic activity has been reported down to >10 cm (West et al., 2012; Zepp Falz et al., 1999), which translates to deposition from the last 20–50 years. Thus, yearly changes in primary productivity are likely integrated with past primary productivity to dictate organic matter availability in methanogenic sediments.

To visualize the potential influence of sediment mixing on CH_4 storage responses to GPP in a single lake, we can describe this mathematically with Equation 1:

$$CH_4\ \text{storage}_t = \beta_0 + \beta_1 * \frac{\sum_{t-x}^{t} \text{GPP}}{x}, \quad (1)$$

where CH_4 storage at a given time (t) is linearly predicted by the average GPP over some interval of time (x). Here, the extent of sediment mixing in a given lake is represented by x, the interval of time over which GPP is averaged. In other words, x denotes the number of years that a given year's GPP contributes to implicit sediment organic matter pool in the active methanogenic zone. Using regression coefficients estimated from the spatial relationship between CH_4 storage and GPP from our empirical results (Figure 3a), we can assign $\beta_0 = -0.72$ and $\beta_1 = 0.085$ to the above equation. We note that this representation of sediment mixing dynamics assumes instantaneous mixing and the subsequent equalizing of sediment carbon quality across years of deposition. While previous research concludes that carbon age does influence quantity and quality (Gälman et al., 2008), this simple model simulation allows us to explicitly consider how different extents of sediment mixing (x) may change our predictions for how CH_4 dynamics respond to changes in GPP.

In this thought experiment, the mean summer GPP of one lake has doubled linearly from a historical state over 50 years (Figure 4a). We simulated CH_4 storage rates based on Equation 1 and with x values ranging from 0 to 50 years. This parameterization represents a gradient of scenarios from no sediment mixing ($x = 0$) to high extent of sediment mixing ($x = 50$) that are plausible from previous empirical studies (Leavitt & Carpenter, 1989; West et al., 2012; Zepp Falz et al., 1999). From these simulations, we see that, at all non-zero values of x, CH_4 storage has a dampened response to primary productivity compared to expectations where sediment mixing extent is not represented ($x = 0$) and where prediction of CH_4 storage is based on purely spatial patterns (solid circle points, Figure 4b). Additionally, over the 50 years, the difference in the sum of summer CH_4 storage between a scenario with no sediment mixing and one with 50 years of sediment mixing is nearly 30 mmol CH_4 m^{-2} d^{-1}, representing a 31% reduction (Figure 4c). Finally, we see that within the first 5 years of changes in GPP, similar to the time scale of our empirical results, there is little to no response of CH_4 storage to GPP (Figure 4b).

By integrating the sedimentation of organic material over time and considering different extents of sediment mixing, our expectation for how CH_4 dynamics respond to increases in primary productivity changes dramatically. Thus, it could be hypothesized that lakes with different extents of sediment mixing may have very different responses to modern-day changes in primary productivity. Lakes with long-term systematic increases in GPP would be most suited to test this hypothesis, as non-directional interannual variation in GPP would also have a dampening effect on this relationship and may obscure the effect of sediment mixing. Further, given the depth of active methanogenic sediments in many stratified lakes, systems with decadal-long records of carbon and CH_4 budgets that include carbon sedimentation and burial rates would be needed to test this hypothesis and constrain the influence of sediment mixing. We suggest that CH_4 storage is a useful metric for long-term monitoring given the single point estimates used here and may be a clear step forward toward quantifying the long-term temporal link between lake primary productivity and CH_4 dynamics across different lakes.

The use of cross-system comparative studies for investigating lake CH_4 dynamics has provided major advances in our understanding of what regulates lake CH_4 production and emission. However, we show that the next step forward is to investigate processes that may alter the response of lake CH_4 processes to modern-day primary productivity, as this relationship was not consistent at the lake scale. We hypothesize a few mechanisms for why lake-specific responses may diverge from the spatial pattern, but further research is needed to directly test these hypotheses and quantify the influence of anoxia, lake bathymetry, and sediment mixing on lake CH_4 responses. Further, we suggest caution when making predictions for future scenarios of environmental change, as assuming temporal responses will mirror spatial patterns—and not accounting for differences in lake characteristics—may lead to substantial misestimation of the rate of change. As lakes undergo changes in trophic status, lake CH_4 responses are likely to change, and we conclude that incorporating the temporal scale of change is key to accurately representing lake CH_4 dynamics with environmental change.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Data supporting the analyses and conclusions are available at the Zenodo repository (https://doi.org/10.5281/zenodo.3946997) and at the cited references.

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