High spatiotemporal variability of methane concentrations challenges estimates of emissions across vegetated coastal ecosystems

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Abstract
Coastal methane (CH4) emissions dominate the global ocean CH4 budget and can offset the “blue carbon” storage capacity of vegetated coastal ecosystems. However, current estimates lack systematic, high-resolution, and long-term data from these intrinsically heterogeneous environments, making coastal budgets sensitive to statistical assumptions and uncertainties. Using continuous CH4 concentrations, δ13C-CH4 values, and CH4 sea–air fluxes across four seasons in three globally pervasive coastal habitats, we show that the CH4 distribution is spatially patchy over meter-scales and highly variable in time. Areas with mixed vegetation, macroalgae, and their surrounding sediments exhibited a spatiotemporal variability of surface water CH4 concentrations ranging two orders of magnitude (i.e., 6–460 nM CH4) with habitat-specific seasonal and diurnal patterns. We observed (1) δ13C-CH4 signatures that revealed habitat-specific CH4 production and consumption pathways, (2) daily peak concentration events that could change >100% within hours across all habitats, and (3) a high thermal sensitivity of the CH4 distribution signified by apparent activation energies of ~1 eV that drove seasonal changes. Bootstrapping simulations show that scaling the CH4 distribution from few samples involves large errors, and that ~50 concentration samples per day are needed to resolve the scale and drivers of the natural variability and improve the certainty of flux calculations by up to 70%. Finally, we identify northern temperate coastal habitats with mixed vegetation and macroalgae as understudied but seasonally relevant atmospheric CH4 sources (i.e., releasing ≥ 100 μmol CH4 m−2 day−1 in summer). Due to the large spatial and temporal heterogeneity of coastal environments, high-resolution measurements will improve the reliability of CH4 estimates and confine the habitat-specific contribution to regional and global CH4 budgets.

KEYWORDS
blue carbon, carbon cycle, climate change, coastal greenhouse gas emissions, methane fluxes
1 | INTRODUCTION

Methane (CH$_4$) is the second most important greenhouse gas (GHG) driving global climate change (Shindell et al., 2009). Past research has shown that coastal marine environments dominate the global ocean CH$_4$ budget and contribute 5–28 Tg CH$_4$ yr$^{-1}$ to total global CH$_4$ emissions (Rosentreter et al., 2021b; Weber et al., 2019). However, a scarcity of systematic, high-resolution, and long-term measurements has hampered our ability to confine CH$_4$ emissions from a wide range of heterogeneous and dynamic coastal environments impeding efforts to evaluate the potential of coastal ecosystems to mitigate climate change by storing carbon (Rosentreter et al., 2021a).

Particularly in coastal sediments, CH$_4$ can be produced in large amounts due to the organic carbon surplus of primary production from submerged (e.g., seagrass and macroalgae) and partially emerged (e.g., mangroves and salt marshes) vegetation (Duarte et al., 2005; Ortega et al., 2019; Snelgrove et al., 2018) and the accumulation of allochthonous particulate organic matter (Barnes & Goldberg, 2020). CH$_4$ emissions from mangroves that only occur around the tropics (70% when excluding mangroves that only occur around the tropics) have been the focal point for global coastal ecosystems, seagrass meadows, salt marshes, and mangrove forests (Al-Haj & Fulweiler, 2020). However, stretching ecosystems as “blue carbon” storage reservoirs by counteracting carbon emissions from terrestrial ecosystems in Al-Haj and Fulweiler (2020) employed flux chamber measurements or discrete sampling. Chamber measurements produce time-averaged flux estimates (often for a period between 24 and 48 h). In contrast, discrete samples have no time-weighted average, but due to logistical reasons, are usually taken at frequencies of no more than one to five samples per day and location (Banerjee et al., 2018; Dutta et al., 2015; Nirmal Rajkumar et al., 2008). These studies resulted in significant advances in our understanding of CH$_4$ emission from coastal systems. Yet, the strong influence of physical forcing (e.g., wind, waves, currents, tides) on the main CH$_4$ emission pathways (diffusion and ebullition) over short timescales (minutes to hours) can lead to a high CH$_4$ concentration and flux variability within one diel cycle, as has been shown in lake environments (Siezko et al., 2020) and tidal influenced estuarine systems (Rosentreter et al., 2018). In the past decade, methods have been developed to improve the spatial and temporal resolutions of CH$_4$ concentration and flux measurements in aquatic systems. For example, using real-time in situ measurements based on a gas equilibrator coupled to cavity ring-down spectroscopy (CRDS), Call et al. (2015) and (2019) showed variability across days to weeks and Rosentreter et al. (2018) documented seasonal CH$_4$ variability in mangrove creeks. These high-resolution efforts have facilitated an improved understanding of different pathways, sources, and sinks in mangrove forests, yet the amplitude and underlying mechanisms of this variability in other coastal marine ecosystems are largely unknown. In addition, seasonal sampling becomes especially important for annual estimates from underrepresented northern temperate and high-latitude regions, but time-series measurements are often discontinued in winter due to harsh weather conditions.

Although high-resolution measurements are critical for reliably capturing the magnitude of the coastal CH$_4$ variability, these sampling campaigns can be time-consuming and expensive. As such, it is desirable to determine the sampling effort required to obtain a high-accuracy, representative mean dissolved CH$_4$ concentration for various coastal environments.

We explored the spatial and temporal variability of CH$_4$ across various heterogeneous coastal environments by systematically measuring CH$_4$ concentrations in three widely distributed yet understudied northern temperate coastal habitats (Figure 1a). The
CH₄ distribution in shallow (<4 m water depth) mixed-vegetated, algae-dominated, and adjacent bare sediment habitats was assessed during five sampling campaigns spanning an entire year (Figure 1b), including an ice-covered period in late winter/early spring. We performed in situ real-time monitoring of CH₄ concentrations using CRDS to account for the temporal variability by diel cycles and peak events (Call et al., 2015; Maher et al., 2013; Rosentreter et al., 2018). This state-of-the-art technique also permits high temporal resolution measurements of stable carbon isotope ratios of CH₄ (δ¹³C·CH₄) that help elucidate the controls and formation and removal pathways of the coastal carbon cycle (Maher et al., 2015). All measurements were complemented with benthic vegetation and physicochemical data to (a) provide spatially and temporally resolved CH₄ distribution and emission data from major northern temperate nearshore benthic environments; (b) identify potential biotic and abiotic drivers in shaping the temporal variability of CH₄; and (c) test whether current methods are sufficient in resolving the CH₄ distribution within highly heterogeneous and dynamic coastal settings both spatially and temporally.

2 | MATERIALS AND METHODS

2.1 | Study area

This study compares three distinct nearshore shallow (<4 m water depth) coastal habitats located on the island of Askö in the Baltic Sea (58°49'15.4"N 17°38'08.8"E). The habitats are representative for globally pervasive coastal ecosystems and were identified according to their dominant type of substrate and vegetation: (1) mixed-vegetated communities of vascular plants and algae on sediments (hereafter "mixed-vegetated" habitat); (2) mixed turf- and macroalgae on rocks with pockets of sediments (hereafter "algae-dominated" habitat), and (3) surrounding soft sediments without major macrovegetation cover (hereafter "bare sediments"). Each habitat was assessed visually, and the percent cover of the underlying substrate and macrovegetation was recorded within a 5-m radius. Taxa that could not be identified underwater were sampled and confirmed in the laboratory. Benthic surveys were repeated in April and September 2020. Overall, the mixed-vegetated habitat was characterized by coarse sediments with 60–90% total vegetation cover. The vegetation was dominated in equal parts by vascular plants (e.g., Phragmites australis, Stuckenia pectinata, and Ruppia spiralis) and benthic algae (e.g., Chara aspera and heterogenous assemblages of filamentous algae). The "algae-dominated" habitat was situated on rocks and boulders with pockets of permeable sediments with 80–95% total vegetation cover comprised of the macroalgae Fucus vesiculosus, and Ulva spp., the encrusting Hildenbrandia rubra, and various filamentous algae. No vascular plants were identified in this habitat. The surrounding bare sediment habitat with fine soft sediments had 7–10% total vegetation, of which were mainly dislodged F. vesiculosus and filamentous algae. The study was conducted at the SW facing side of the island, which is dominated by rocky cliffs and shallow embayments and is relatively open to the Baltic Sea. The habitats were fully submerged at all times due to the absence of tides in this region of the Baltic Sea (Medvedev et al., 2016). The average of measured salinities (i.e., per sampling period and habitat) in the studied area ranged from 6.2 to 7.0 over the course of the year, and, thus, reflected brackish water conditions typical for the central Baltic Sea.
While the Baltic Sea receives freshwater inflows from land and has limited saltwater inflows from the Danish straits, locally at the study site on the island in the outer Stockholm archipelago, there were no major freshwater inputs from rivers or streams, which is reflected by relatively constant salinity throughout the measurement period.

### 2.2 Experimental design

We quantified the partial pressures of surface water and atmospheric CH$_4$ and CO$_2$ along with the related C-isotopes (i.e., $\delta^{13}$C-CH$_4$ and $\delta^{13}$C-CO$_2$) in the three habitats during five measurement periods in 2020 and 2021 (i.e., May 18–29; July 6–17; August 17–29; November 30 to December 8, 2020; March 1–6, 2021). For the measurements, we used an adapted version of the Water Equilibration Gas Analyzer System (WEGAS) (details in Humborg et al., 2019) coupled to a CRDS. The system consists of four major components: (i) a submersible seawater intake pump at around 0.3 m water depth mounted to a movable raft that can be deployed noninvassively over the various habitats; (ii) a water handling system comprised of a showerhead equilibrator (1 L headspace volume) and a thermosalinograph (Seabird TSG 45) fed via a hose by the seawater prised of a showerhead equilibrator (1 L headspace volume) and a vasively over the various habitats; (iii) a gas handling system with circulation pumps for the showerhead and ambient air; and (iv) the CRDS gas analyzer for CH$_4$ and CO$_2$ concentration and related C-isotope measurements (model G2201-i, Picarro Inc.). The use of a large seawater intake pump results in the combined measurement of CH$_4$ from ebullition (bubbles) and the dissolved form in the water. The individual contribution of the two forms can, however, not be resolved using the current system. For CH$_4$ and CO$_2$ analyses, gas in the showerhead of the equilibrator was measured for 35 min, followed by gas measurements of ambient air for 10 min (i.e., one complete cycle was 45 min). These measurement cycles (i.e., 35 min, water; and 10 min, air measurements) ran continuously during the five measurement periods mentioned above. The raft with the water intake pump was moved between the defined habitats every 24 h from the shore with ropes. Measurements in March were distinct from the other sampling periods due to the ice cover that had been present for 4–6 weeks prior to the time of sampling. Here, holes were drilled into the ice and the pump lowered to measure “under-ice” concentrations. We validated the CRDS analyzer’s performance according to the manufacturer’s instructions with “ALPHAGAZ™ Stable Isotope Ratio Gases” for Picarro instruments. Specifically, before each deployment period, we injected three standards with varying CO$_2$ and CH$_4$ bulk concentrations, and varying $\delta^{13}$C-CO$_2$ and $\delta^{13}$C-CH$_4$ isotope values (i.e., low = 1.00 ppm CH$_4$, -24.20‰ $\delta^{13}$C-CH$_4$, 250.00 ppm CO$_2$, -5.00‰ $\delta^{13}$C-CO$_2$; natural = 1.77 ppm CH$_4$, -48.30‰ $\delta^{13}$C-CH$_4$, 399.00 ppm CO$_2$, -8.50‰ $\delta^{13}$C-CO$_2$; and high = 10.00 ppm CH$_4$, -68.60‰ $\delta^{13}$C-CH$_4$, 1000.00 ppm CO$_2$, -20.10‰ $\delta^{13}$C-CO$_2$). Measurements with each standard ran for 10 min, and three-point calibration lines were constructed whose regression coefficients were used to scale the unknown sample data if needed.

Concentration and isotope measurement at 1 Hz frequency were averaged and logged every 10 s. The recorded data were filtered by removing data from the transition period between ambient air and water measurements due to the response time of CRDS to sharp changes in concentrations of CH$_4$ and CO$_2$. Data were also removed during improper functioning (e.g., low water flow). For this study, we used 210,059 (averaged from 2,100,590 measurements at 1 Hz) data points each for CH$_4$, CO$_2$, $\delta^{13}$C-CH$_4$, and $\delta^{13}$C-CO$_2$ for statistical purposes. CH$_4$ concentrations in water (in ppm obtained by the CRDS) were converted to molar concentrations (i.e., CH$_4$ in nM) and CO$_2$ was converted to pressure units (i.e., pCO$_2$ in $\mu$atm) (Humborg et al., 2019). Alongside CRDS measurements, several other environmental and meteorological variables were recorded. Surface water temperature, pH, and dissolved oxygen concentrations at the point of water intake were logged every 15 min using a multiparameter sonde (model EXO2, YSI) that was calibrated prior to each deployment. Water temperature and salinity were also recorded with every CRDS measurement with a thermosalinograph (Seabird TSG 45) that was positioned before the showerhead equilibrator. Wind data observations (wind speed and direction) and air temperature were obtained from a Metek uSonic-3 heated 3D sonic anemometer, and a Vaisala HMP155 shielded temperature probe mounted on a 1.5-m high meteorological mast. The mast was located at the waterline in a coastal bay, approximately 400 m to the northwest of the sampled habitats. Mean winds were adjusted to a 10-m reference height assuming a logarithmic profile with neutral stability (Haugen, 1973):

$$U_{10} = U + \left( \frac{U^*}{\kappa} \right) \times \log \left( \frac{10}{z_o} \right)$$

where $U$ is the measured wind speed at height $z_o$, $U^*$ is the measured friction velocity by the 3D sonic anemometer, and $\kappa$ is the von Karman constant (0.4).

### 2.3 Exploration of the CH$_4$ distribution variability

We used a generalized linear model (GLM) to examine differences across habitats within each month. Due to positive-skewed data and overdispersion, a quasi-Poisson model was constructed using the glm() function in $\alpha$ (R Core Team, 2021) with “Month” (i.e., March, May, July, August, December) and “Habitat” (i.e., Mixed-vegetated, Algae-dominated, and Bare sediments) as factors. We used the R package “emmeans” (Lenth et al., 2019) for pairwise post hoc multiple comparisons with Bonferroni-adjusted p-values. Results and model details are presented in Table S1. The relationships among CRDS and environmental data were initially assessed using principal component analysis (PCA) using the R packages “FACTOmR” (Husson et al., 2016) and “FACTOextra” (Kassambara & Mundt, 2017). PCA is a multivariate technique used to emphasize variation and to visualize patterns in a dataset, particularly when there are many variables. Upon the visual inspection of the PCA, we calculated Spearman coefficients for correlations between
The saturation of CH$_4$ drivers (i.e., water temperature, salinity, dissolved oxygen and CO$_2$ concentrations, and pH).

The thermal sensitivity of the CH$_4$ distribution was further explored by applying principles of the metabolic theory of ecology (MTE) (Sibly et al., 2012), calculating the activation energy (Ea) based on Arrhenius equations in the seasonal thermal regime. The activation energies (Ea in eV) were estimated by fitting a linear regression equation between the natural logarithm of CH$_4$ concentrations and the reciprocal of temperature (1/kT), where k is the Boltzmann’s constant (8.62 × 10$^{-5}$ eV K$^{-1}$) and T is the measured water temperature in Kelvin. EAs allow for a comparison of temperature dependencies across systems and metabolic processes (Sibly et al., 2012).

We applied the Rayleigh model to estimate the fraction of CH$_4$ that was oxidized in surface water in each habitat and sampling month, as:

$$\delta^{13}C_{CH4}(CRDS) = \delta^{13}C_{CH4}(S) + \epsilon x (1 - f)$$

where $\delta^{13}C_{CH4}(CRDS)$ is the isotopic composition of surface water CH$_4$ measured with the CRDS system, $\delta^{13}C_{CH4}(S)$ is the isotopic value of the CH$_4$ source in sediments, $-67\%$ that has been measured in local sediments, $\epsilon$ is the isotope fractionation factor for CH$_4$ oxidation of $-20\%$ (Bastviken et al., 2002), and $f$ represents the fraction of remaining CH$_4$ in surface water, that is, $1 - f$ is the fraction of oxidized CH$_4$. The Rayleigh model assumes a closed system when CH$_4$ oxidation occurs, which means CH$_4$ oxidation is the only sink of CH$_4$ in water column and is faster than the refreshment of CH$_4$ supplied to the surface water. This is an oversimplification given the high variability of coastal systems. The true fraction of CH$_4$ oxidized in surface water could, thus, be underestimated due to the contribution of $^{13}C$-depleted CH$_4$ source mixing with surface water CH$_4$ with higher $\delta^{13}C_{CH4}$ values in a partially open system.

### 2.4 Sampling effort evaluation of dissolved CH$_4$ concentrations

We used a bootstrapping exercise to determine the minimum number of concentration measurements per day required to obtain a high-accuracy, representative daily mean dissolved CH$_4$ concentration across the various coastal habitats and sampling months. The assumption of these simulations is that our high-resolution sampling effort (i.e., one sample per second) can sufficiently capture the temporal variations of surface water CH$_4$ concentrations for each habitat. We randomly sampled the population of measured CH$_4$ concentrations assuming a variable sample size, from 1 to 600 samples a day (with sample replacement). This sampling was repeated 200 times for each sample size, and for each simulation, we calculated the resulting mean CH$_4$ concentration.

### Table 1: Methane (CH$_4$) concentrations and saturations in the three studied nearshore coastal habitats

| Month       | Habitat                  | CH$_4$ (nM) | CH$_4$ saturation (%) |
|-------------|--------------------------|-------------|-----------------------|
|             | Mean (±SD)               | CV (%)      | Median (IQR)          | Range                  | Median (IQR) | N  |
| March*      | Mixed-vegetated          | 90 (±17)    | 19                     | 84 (78–96)             | 68–152       | 1659 (1530–1892) | 6904 |
|             | Algae-dominated          | 68 (±4)     | 6                      | 67 (66–70)             | 57–82        | 1320 (1289–1371) | 4495 |
|             | Bare sediments           | 74 (±5)     | 7                      | 74 (71–76)             | 60–102       | 1438 (1389–1493) | 6083 |
| May         | Mixed-vegetated          | 56 (±17)    | 30                     | 56 (42–69)             | 17–103       | 1369 (1034–1672) | 19,573 |
|             | Algae-dominated          | 41 (±15)    | 37                     | 40 (27–49)             | 17–101       | 980 (731–1159)  | 17,894 |
|             | Bare sediments           | 40 (±8)     | 20                     | 41 (35–45)             | 20–75        | 980 (908–1071)  | 18,056 |
| July        | Mixed-vegetated          | 119 (±33)   | 28                     | 112 (99–144)           | 58–204       | 3056 (2720–4059) | 7182 |
|             | Algae-dominated          | 80 (±24)    | 30                     | 71 (66–85)             | 45–169       | 1949 (1800–2335) | 10,885 |
|             | Bare sediments           | 69 (±17)    | 25                     | 70 (54–82)             | 34–115       | 1977 (1517–2249) | 11,961 |
| August      | Mixed-vegetated          | 190 (±74)   | 39                     | 174 (150–211)          | 53–460       | 5275 (4624–6563) | 21,801 |
|             | Algae-dominated          | 144 (±54)   | 38                     | 153 (97–189)           | 41–274       | 4755 (2847–5850) | 23,597 |
|             | Bare sediments           | 161 (±53)   | 33                     | 151 (133–185)          | 41–324       | 4570 (3991–5835) | 19,210 |
| December    | Mixed-vegetated          | 18 (±7)     | 39                     | 19 (12–24)             | 6–37         | 426 (258–526)  | 17,253 |
|             | Algae-dominated          | 13 (±3)     | 23                     | 12 (11–15)             | 9–23         | 252 (230–332)  | 11,878 |
|             | Bare sediments           | 9 (±2)      | 22                     | 9 (8–10)               | 6–17         | 191 (163–214)  | 10,587 |
| Annual      | Mixed-vegetated          | 97 (±79)    | 81                     | 77 (34–143)            | 6–460        | 1707 (849–4287) | 75,413 |
|             | Algae-dominated          | 79 (±61)    | 77                     | 65 (32–116)            | 9–274        | 1508 (803–3250) | 68,749 |
|             | Bare sediments           | 79 (±64)    | 81                     | 57 (38–122)            | 6–324        | 1424 (937–3690) | 65,897 |

The saturation of CH$_4$ is relative to the dissolved equilibrium with ambient air. Abbreviations: CV, coefficient of variation; IQR, interquartile range; N, number of individual observations (10 s average of 1 Hz measurements); SD, standard deviation. The asterisk denotes under-ice sampling in March.
2.5 | Sea–air flux computation

The sea–air flux (F) of CH₄ was calculated as:

$$F = k \times K_0 \times (pCH₄_{sea} - pCH₄_{air})$$

where $k$ (m s⁻¹) is the gas transfer velocity, $K_0$ (mol m⁻³ atm⁻¹) is the aqueous-phase solubility of CH₄, and $pCH₄_{sea}$ and $pCH₄_{air}$ are the measured partial pressures (atm) of CH₄ in the near-surface water and in the air, respectively. We compared our site-specific atmospheric CH₄ concentration measurements to data of the closest ICOS atmospheric monitoring station (i.e., Utö–Baltic Sea station; station ID: UTO). Locally measured $pCH₄$ ranged from 1.884 to 2.124 (mean ± SE = 1.969 ± 0.063) over the study period, which compares to 1.921–2.112 (mean ± SE = 1.978 ± 0.001) from the ICOS Utö–Baltic Sea station (Laurila, 2021). Despite similar mean values over the study period, locally measured $pCH₄$ reflects better the site-specific variability and is, thus, more suitable for the computation of air–sea fluxes, especially if concentration gradients (i.e., water to atmosphere) are small. The solubility is determined from Wiesenburg and Guinasso (1979) as:

$$Sc = \beta + A_1 + A_2(100/T) + A_3 ln(T/100) + B_1 + B_2(T/100) + B_3(T/100)^2$$

where $\beta$ is the dimensionless (mL of gas dissolved per mL of H₂O) Bunsen solubility coefficient, $A_1$, $A_2$, $A_3$, and $B_1$, $B_2$, $B_3$ are constants from Table 1 in Wiesenburg and Guinasso (1979), $T$ is the measured water temperature (K), and $S$ the measured salinity (%). Assuming CH₄ behaves as an ideal gas, $K_0$ is related to $\beta$ by $K_0 = \beta (R \times T_{STD})^{-1}$, where $R$ (m³ atm⁻¹ mol⁻¹ K⁻¹) is the ideal gas constant and $T_{STD}$ (K) is the standard temperature in Kelvin.

The gas transfer velocity ($k$) used is that determined by Wanninkhof (2014) as:

$$k = 0.251 \times U^2 \times \left( \frac{Sc_{balticsea}}{660} \right)^{-0.5}$$

where $U$ is the wind speed (m s⁻¹) at 10 m height and $Sc_{balticsea}$ is the Schmidt number at the measurement site, which is dependent on temperature, salinity, and gas molecule. Sc was corrected for the corresponding temperature that was measured simultaneously with partial pressures of CH₄ ($pCH₄$) according to coefficients taken from Table 1 in Wanninkhof (2014). Furthermore, the Schmidt number for Baltic Sea brackish water (i.e., $Sc_{balticsea}$) with measured salinity ($S_{balticsea}$) was calculated by interpolation of Sc for fresh water (salinity 0‰) and seawater (salinity 35‰) following (Gü lözw et al. 2013) and (Jähne et al. 1987):

$$Sc_{balticsea} = \frac{(Sc_{seawater} - Sc_{freshwater}) \times Sc_{balticsea} + Sc_{freshwater}}{35}$$

All fluxes are expressed in μmol CH₄ m⁻² day⁻¹. Other variables (e.g., currents, waves, water depth) can also be used to predict $k$ in coastal environments, but the studied location does not have any significant permanent or tidal currents, and estuarine models may not provide better results for our setting. Furthermore, Lundevall-Zara et al. (2021) tested other wind-based $k$ models in similar habitats of the same location and concluded that calculated average $k$-values from different models were close to those of the Wanninkhof (2014) relationship for the range of wind velocities encountered on the island of Askö. Thus, for a better comparability across studies, we have therefore decided to use this relationship.

2.6 | Estimating annual sea–air fluxes of CH₄

We estimated sea–air fluxes of CH₄ across all habitats over the entire annual cycle. Based on the strong temperature dependencies of CH₄ concentrations, we calculated CH₄ concentrations outside of the measurement periods using the Arrhenius equations from Figure 3b and the 15-min interval surface water temperature measurements from March 3, 2020 to March 3, 2021 (Figure 1b), as:

$$\ln(\text{CH}_4(nM)) = a + b \times \left( \frac{1}{T} \right)$$

with $\ln(\text{CH}_4)$ as the natural logarithm of the CH₄ concentration in nM, $a$ and $b$ as intercept and slope, respectively, and the reciprocal of temperature ($1/K$), where $k$ is the Boltzmann’s constant ($8.62 \times 10^{-5} \text{ eV K}^{-1}$) and $T$ is the measured water temperature in Kelvin. For the mixed-vegetated, algae-dominated, and the bare sediment habitat, the intercepts were 47.14, 47.36, and 51.66, respectively, and the slopes were −1.02, −1.03, and −1.13, respectively.

We determined the difference between the measured and the estimated CH₄ concentrations per habitat and month (where measured data were available) as the percentage of the calculated value (i.e., the percent error). Overall, there was a good agreement of the temperature-based calculated CH₄ concentrations with the actual measured concentrations across all habitats in May, July, and August (i.e.; mostly <10% deviation of the means; Table S2). Calculated CH₄ concentrations in March and December tended to be underestimated by 20–50% relative to the measured concentrations. The data show that temperature can be a good proxy to estimated CH₄ concentrations if enough in situ data are available. However, it also becomes apparent that, when absolute concentrations are low, disparities of few nanomole in the CH₄ concentration likely contributed to large differences (Table S2).

Sea–air fluxes of CH₄ were then calculated based on equations provided above, assuming an average salinity of 6.6 (i.e., the average of measured salinities ranging from 6.2 to 7.0 over the course of the year). Wind speed data from the study location matching the CH₄ concentrations (measured and calculated) was available for 21,445 out of 34,932 data points (61%). The remaining wind speed data were estimated using a Monte-Carlo simulation on the distribution (mean ± SD, 2.25 ± 2.01 m/s) of available wind speed data from that year.
3 | RESULTS

3.1 | CH₄ concentrations and δ¹³C-CH₄ values across coastal habitats

We report a high spatial and temporal variability of surface water CH₄ concentrations across the mixed-vegetated, algae-dominated, and bare sediment habitats that span two orders of magnitude, ranging from 6 to 460 nM (Figure 1c; Table 1). During all sampling periods, the highest concentrations were always observed in the mixed-vegetated habitat, followed by algae-dominated, and surrounding bare sediment habitats (Table 1). A generalized linear model (GLM) with pairwise post hoc multiple comparisons confirmed that CH₄ concentrations differed significantly across habitats within each sampling month (Table S1), with an exception of the algae-dominated and bare sediment habitats in May. In addition, differences between the algae-dominated and bare sediment habitats were minor (expressed by odds ratios close to 1 as effect size statistics) in May, July, and August (Table S1). There were strong seasonal variations of CH₄ concentrations with similar patterns across all habitat types. In general, the highest CH₄ concentrations were observed in August, followed by July, March, May, and December (Figure 1c; Table 1). The δ¹³C-CH₄ values of surface water varied by >7‰ over the sampling months in all habitat types. Across all habitats, CH₄ was most enriched in ¹³C in December (average of −55‰) and became most depleted in March, approaching −63‰ (Figure 1d). Differences in δ¹³C-CH₄ values across habitats in the same month occurred only in some cases and were smaller than the annual temporal variation (Table S3).

CH₄ concentrations also varied greatly during each sampling month and in each habitat type (Figure 1c). Most variability within months was ascribed to CH₄ concentration changes independent of the time of the day (Figure 2a-c). "Peak events" with >100% change of the CH₄ concentrations occurred within hours and were observed in all habitats and during all sampling campaigns (Figure 2d-f). We used the coefficient of variation (CV) as a standardized measure that describes the dispersion of the CH₄ distribution around the mean to quantify and compare the within-month variability. Overall, the CVs ranged from 5% to 39%, with the lowest variability of CH₄ concentrations in March when the surface water was covered with ice and the highest variability generally occurring in July and August (Table 1). An exception to the seemingly random CH₄ variability within one diel cycle was the mixed-vegetated habitat in August, when CH₄ consistently peaked during midday (mean ± SD, 333 ± 93 nM at 13:00 h local time), and was lowest at night (141 ± 24 nM at 02:00 h local time; Figure 2a).

3.2 | Correlation of surface water CH₄ with environmental variables

Principal component analysis (PCA) revealed distinct separation of the CH₄ and environmental data across months and to a lesser extent across habitats (Figure 3a). The first two principal components explained 43.2% of the variation in the data. Separation was greatest along principal component axis 1 (PC1 = 24.6%) that split the groups into five distinct clusters representing the sampling months March, May, July, August, and December. Variations in CH₄ concentrations (32.1%), temperature (23.4%), salinity (14.8%), and oxygen (11.0%) contributed most to the separation of the data along principal component axis 1. Data points within each month spread predominantly along principal component axis 2 (PC2 = 18.4%), and their variation was driven by the time of the day, CO₂ concentrations, and pH.

Upon the visual inspection of the PCA (Figure 3a), we computed correlation matrices based on Spearman's rank correlation coefficient of CH₄ concentrations in each habitat with temperature, salinity, CO₂, O₂, and pH (Figure S1). Temperature showed the...
The strongest positive association with CH₄ concentrations in the algae-dominated habitat ($r^2(68,740) = .82$, $p < .0001$), followed by bare sediments ($r^2(65,812) = .71$, $p < .0001$), and the mixed-vegetated habitat ($r^2(75,410) = .70$, $p < .0001$). Weaker and negative associations were also apparent for O₂, CO₂, and salinity in all habitat types (Figure S1). pH was negatively associated with CH₄ concentrations in the mixed-vegetated and algae-dominated habitat but positively in the surrounding bare sediments. Given the strong association with temperature, we further explored the thermal sensitivity of the CH₄ distribution by calculating the apparent activation energy ($E_a$ in eV) based on Arrhenius equations in each habitat within the seasonal thermal regime (Figure 1b). Estimated $E_a$s were similar across all habitats with positive (i.e., higher CH₄ concentrations at higher temperature) values of 1.13 eV ($F(1,59,812) = 277,552.7$, $p < .0001$, $r^2 = .82$) in bare sediments, 1.03 eV ($F(1,64,252) = 256,516.8$, $p < .0001$, $r^2 = .80$) in algae-dominated, and 1.02 eV ($F(1,65807) = 204,754.7$, $p < .0001$, $r^2 = .80$) in the mixed-vegetated habitat, respectively (Figure 3b).

The $δ^{13}$C-CH₄ signatures provided an additional dimension to reveal the main processes controlling CH₄ variability given the isotope fractionation associated with CH₄ production and consumption (i.e., oxidation) (Barker & Fritz, 1981). $δ^{13}$C-CH₄ values as a function of CH₄ concentrations reflected temporal variations across seasons (Figure 4). In all habitats, the lowest CH₄ concentrations with the highest $δ^{13}$C-CH₄ values were observed in December, while the highest CH₄ concentrations and the lowest $δ^{13}$C-CH₄ values were found in August and March. The Rayleigh model, assuming that the supply of CH₄ is much slower than oxidation in water column, was applied to estimate the fraction of CH₄ that was oxidized in surface water, suggesting 20% of CH₄ loss through oxidation in August and March, and up to 50% in December in all habitats (Table S3).
3.3 | Sea-air fluxes of CH₄

Surface waters were supersaturated throughout all measurement periods and habitats relative to CH₄ in ambient air. The median (IQR) CH₄ saturation in the mixed-vegetated habitat was 1706 (848-4286)%, 1508 (802-3250)% in algae-dominated, and 1423 (937-3689)% in adjacent bare sediment habitats. The resulting sea-air flux rates of CH₄ were highly variable and ranged from 0.1 to 3852 μmol CH₄ m⁻² day⁻¹ during ice-free periods and primarily reflected differences in water column CH₄ concentrations between habitats and months (Table S4). The median (IQR) CH₄ flux rates were highest during measurement periods in July, with 138 (81–245), 98 (26–172), and 77 (36–119) μmol CH₄ m⁻² day⁻¹ in the mixed-vegetated, algae-dominated, and bare sediment habitat, respectively. The high CH₄ flux rates in July coincided with high wind speeds during this month (Table S4). CH₄ flux rates were lowest in all habitats in December, with median values of 0.7–3.5 μmol CH₄ m⁻² day⁻¹. No fluxes were computed for the ice-covered period. The estimated annual median (IQR) sea-air fluxes were 12 (3–43), 10 (2–32), and 7 (2–29) μmol CH₄ m⁻² day⁻¹ in the mixed-vegetated, algae-dominated, and bare sediment habitat, respectively.

4 | DISCUSSION

Our high-resolution measurements revealed differences in the distribution of surface water CH₄ concentrations across neighboring coastal habitats over short spatial (within meters) scales and exceptionally high temporal variability that could only be detected with continuous measurement techniques during several seasons. Acknowledging this high spatiotemporal variability is critical to confine CH₄ emissions from coastal environments and the variability associated with their habitat heterogeneity.

4.1 | Temperature sensitivity of coastal CH₄ distribution

Median CH₄ concentrations measured across the here-studied habitats were 4–13 times higher than those observed in deeper waters of the open Baltic Sea (Schmale et al., 2010; Wilson et al., 2018), up to three times higher than previously published data for coastal Baltic areas (Humborg et al., 2019; Ma et al., 2020), and substantially higher than globally compiled nearshore CH₄ concentrations (Weber et al., 2019) (Table S5). The magnitude highlights that vegetated coastal ecosystem are characterized by excessive organic matter loads from primary production, trapping and accumulation of allochthonous organic matter, and sedimentary conditions that can favor CH₄ production (Dale et al., 2019; Wallenius et al., 2021). However, we also report an exceptionally high spatiotemporal variability of surface water CH₄ concentrations.

A first major source of this variability was attributed to seasonal differences in CH₄ concentrations. The significant correlation between CH₄ concentrations and temperature over the sampling months suggests that temperature mainly regulates seasonal variations. Like most other forms of metabolism, methanogenesis is temperature-dependent, and the calculated apparent activation energies (EA = −1 eV, across all habitats) were in line with previous global estimates of ecosystem-scale CH₄ fluxes with an EA of 0.96 eV (Yvon-Durocher et al., 2014). Thus, the higher CH₄ concentrations in late summer are likely due to increased production under warmer water temperatures. Historical data from the nearby oceanographic observation station “2507 Landsort Norra” between 2010 and 2020 confirmed that the annual surface water temperature curve from our study area is representative of previous years (Sveriges meteorologiska och hydrologiska institut, 2022). We infer that the observed temperature sensitivity is primarily driven by natural temperature variations rather than a warming effect. Both aerobic CH₄ oxidation together with anaerobic CH₄ oxidation in sediments may also increase in summer due to temperature controlling their rates (Treude et al., 2005; Zehnder & Brock, 1980) and the increased supply of CH₄ supply by molecular diffusion. However, in summer, the overall production of sedimentary CH₄ likely outweighed the relative contribution CH₄ oxidation pathways. In support of this, parallel measured δ¹³C-CH₄ values combined with the Rayleigh model revealed that the relative contribution of CH₄ production versus oxidation shifted across seasons. CH₄ oxidizing bacteria favor isotopically lighter CH₄, leaving the residual CH₄ with heavier isotopes. Low CH₄ concentrations accompanied by high δ¹³C-CH₄ values suggest that up to 50% of CH₄ was oxidized in winter, indicating an important role of CH₄ oxidation in removing CH₄ relative to CH₄ production. This microbial oxidation efficiency decreased to 20–30% in summer due to a boosted supply of CH₄ to the water column relative to its oxidation. High oxygen concentrations mediated by the photosynthetic activity of algae and plant communities during the day and increased light exposure in summer may have contributed to inhibiting CH₄ oxidation (Murase & Sugimoto, 2005; Rudd et al., 1976).

4.2 | Ice-cover effects on CH₄ dynamics

An exception to the overall seasonal trend was observed in March (i.e., late winter/early spring). Measurements during this month were marked by ice cover that, to this point, had been present for 4–6 weeks. Analogous to many northern lakes (Denfeld et al., 2018), we observed an accumulation of CH₄ under the ice, with mean concentrations six times higher than in December (last month without ice cover). More negative δ¹³C-CH₄ values in March (~62 to ~64‰) suggest CH₄ supply with overall low oxidation. This observation corroborates studies showing suppressed methanotrophic activity at very cold temperatures (e.g., Phelps et al., 1998). Calculations of the Rayleigh model confirmed that <20% of the surface water CH₄ was oxidized during this period. However, the CH₄ depleted in ¹³C could also be a result of varying fractionation during methanogenesis at lower temperatures or mixed CH₄ formation pathways. The CH₄ accumulation under ice will likely result in enhanced outgassing events.
following ice break (Ducharme-Riel et al., 2015; Karlsson et al., 2013). Whereas under-ice CH$_4$ accumulation is a well-studied feature of northern lakes, these dynamics have not been described for northern temperate coastal regions with regular sea ice every year. Our data suggest the necessity to include the ice-covered period and CH$_4$ outgassing during ice breakup in future coastal CH$_4$ sampling strategies and the annual CH$_4$ budget of northern temperate and high-latitude regions (Omstedt et al., 2004).

### 4.3 Physical forcing may drive short-term CH$_4$ variability

A second major source of variability in the CH$_4$ concentrations was short-term variations that occurred within hours (Figure 2d–f). Most of this variability was independent of the time of the day and without an apparent and reoccurring diel pattern. However, fluctuations of the CH$_4$ concentrations were so strong that the minimum and maximum values within one habitat and sampling campaign (time window max. 12 days) could differ by up to one order of magnitude (Table 1). The dispersion of the CH$_4$ probability distribution around the mean concentration was on average 30% during the ice-free months and, thus, much higher than the reported global open ocean CH$_4$ variability with CVs ranging between 2% and 11% (Wilson et al., 2018). While we could not find any direct correlation to the available environmental data, one possible explanation for the high variability could be the physical influence of the open coastal setting through wind and/or wave action. A wave-induced pumping effect on the pore water pressure can transport solutes from deeper to surface layers (Precht & Huettel, 2004; Yang et al., 2019); Thus, varying CH$_4$ release rates from permeable coastal sediments in very shallow waters may cause variable near-surface CH$_4$ concentrations, as has been shown relevant even for lake systems (Hofmann et al., 2010). In support of this, the CVs of the CH$_4$ distribution were much lower across all habitats in March (mean CV = 10%), when, due to ice cover, the influence of waves and winds on the water column and sediments was likely minor and no CH$_4$ escaped to the atmosphere.

### 4.4 Reoccurring diel CH$_4$ patterns in summer

A reoccurring diel pattern in CH$_4$ concentration changes was only observed in the mixed-vegetated habitat in August, with the highest concentrations consistently toward midday and lowest at night. This marked diel variation may be attributed to plant-mediated transport of CH$_4$ by convective throughflow from rooted submerged plants, which were only present in the mixed-vegetated habitat. The convective transport through pressure gradients can account for up to 60% of the total CH$_4$ transport from sediments during daylight hours and high photosynthetic activity (Kim et al., 1998; van den Berg et al., 2020). In the early stages of plant growth, molecular diffusion through dead/live plants into the standing water column can be the primary transport mechanism (Kim et al., 2001). Most plants at the mixed-vegetated site were fully submerged; thus, a sediment–plant–water flux is likely. However, Phragmites stems (comprising ~10% of the total vegetation in the mixed-vegetated site) possibly facilitated a sediment–plant–air flux of CH$_4$ (van den Berg et al., 2020), which will have remained undetected with our approach. Abiotic CH$_4$ photoproduction from organic matter degradation may also play a role in shaping site-specific CH$_4$ dynamics in oxygenated surface waters (Li et al., 2020; Zhang & Xie, 2015). However, given that reoccurring and pronounced diel cycles were only visible in one of the three neighboring habitats, benthic/plant-mediated pathways seem more likely to have caused the patterns observed. Overall, the contribution of plant-mediated fluxes and the relation to seasonal succession patterns of submerged vegetation, along with the contribution of CH$_4$ photoproduction in shallow coastal waters with high incident irradiance remain uncertain and need further investigation.

### 4.5 Spatial distribution of CH$_4$ reflects coastal ecosystem mosaic

Shallow coastal habitats are heterogeneous, and the variation in spatial structure and temporal change of benthic communities defines the expression of ecosystem functions in form and magnitude (Snelgrove et al., 2014). Reflecting the coastal ecosystem mosaic (Sheaves, 2009), some of the measurements across the studied neighboring habitats were not further than 30–50 m apart. Yet, despite their proximity, we observed significant differences in the distribution of CH$_4$ in the water column and the magnitude of the resulting sea–air fluxes. Surface water CH$_4$ concentrations are likely related to variable CH$_4$ production and oxidation rates, as indicated by varying δ$^{13}$C-CH$_4$ values across sites during some months (Figure S3). These differences may be ascribed to different quantities and qualities of organic matter deposited within local sediments and differences of sediment properties (e.g., porosity) (reviewed in Rosentreter et al., 2021a). The presence of rooted vegetation may also play a role in the small-scale variability, as roots provide substrate via root litter and exudates and transport oxygen into the sediments. In addition, while the employed system measures CH$_4$ in the dissolved form and from ebullition (bubbles), the individual contribution of the two phases cannot be resolved but may contribute to differences between the habitat types. It becomes apparent that more research is required to determine the spatial scale of this variability and to understand better the controls on substrate availability for methanogenesis. In particular, links between biodiversity metrics (i.e., abundance and biomass) of primary and secondary producers and CH$_4$ production and consumption pathways need to be better constrained as has been shown relevant for seafloor metabolism (i.e., gross primary production and community respiration) in shallow waters (Rodil et al., 2021). Likewise, integrating knowledge on the structure...
of sediment microbial communities associated with the different habitats is imperative to improve the prediction of CH$_4$ production and oxidation pathways from different coastal habitats (Wallenius et al., 2021).

4.6 | High sampling intensity is required to capture coastal CH$_4$ variability

Particularly the high temporal variability on timescales from hours to days complicates our ability to generalize the distribution of CH$_4$ in nearshore coastal environments and obstructs efforts to confine diffusive flux calculations that are based on concentration measurements. Therefore, we conducted a bootstrapping analysis on our continuous data to determine the minimum number of individual concentration samples per day required to obtain a high accuracy, representative mean dissolved CH$_4$ concentrations. The data exploration shows that collecting one discrete water sample a day, a typical approach used to describe CH$_4$ concentration differences across geolocations (Banerjee et al., 2018; Dutta et al., 2015; Nirmal Rajkumar et al., 2008), results in a large uncertainty, with a potential to over- or underestimate the mean CH$_4$ concentration by almost 70%. Specifically, taking only one sample per day from the mixed-vegetated habitat in August would result in a mean CH$_4$ concentration with a 5th–95th percentile of 90–320 nM based on 200 simulations. Increasing the sampling intensity to five samples per day reduces the uncertainty to 30%. In comparison, 50 samples per day instead would narrow this uncertainty to a 5th–95th percentile of 171–209 nM (10% uncertainty), closer to the observed true mean CH$_4$ concentration of 191 nM during this period (Figure 5a). A similar pattern was apparent in all other habitats and sampling periods (Table S6). Consequently, the data collection and sampling strategy are detrimental to accurately capturing the temporal variability and assure justified mean CH$_4$ concentrations that are the basis for flux computations. Thus, near-continuous measurements using CRDS (Hartmann et al., 2018; Humborg et al., 2019; Maher et al., 2013) or similar systems to determine in situ CH$_4$ concentrations in surface waters are desirable when addressing complex pathways and transformations of CH$_4$ in coastal ecosystems. For annual estimates, seasonal measurements that reflect local climatological patterns will be required.

4.7 | Northern temperate coastal habitats are seasonal CH$_4$ emission hotspots

The high-resolution CH$_4$ concentration measurements allowed us to establish annual CH$_4$ emission estimates across all habitat types. The diffusive CH$_4$ fluxes suggest that northern temperate coastal habitats with mixed vegetation, algal dominance, and their adjacent bare sediment areas are net sources of atmospheric CH$_4$ throughout the year. As a result of extended periods of low temperature and temporal ice cover, the median annual fluxes were at the lower end compared to coastal wetland and tidal flat CH$_4$ emissions globally (Rosentreter et al., 2021b). However, in summer, CH$_4$ emissions of ≥100 μmol CH$_4$ m$^{-2}$ day$^{-1}$ across all habitats were comparable to, or even higher than, those reported from similar (Lundevall-Zara et al., 2021) or other vegetated coastal ecosystems (Al-Haj & Fulweiler, 2020; Rosentreter et al., 2021b). During these periods, large amounts of carbon are turned over in the habitats studied here (Attard et al., 2019a, 2019b), and macrophyte tissues become a direct component of local sediment organic matter pools (Marcelina et al., 2018) that favor local CH$_4$ production (Dale et al., 2019; Wallenius et al., 2021). Despite these seasonally relevant CH$_4$ emissions, there is still a paucity of data from northern temperate coastal habitats in general, and they are exceedingly underrepresented in current global CH$_4$ budgets. Yet, just in the Baltic Sea, the potential distribution

![Figure 5](image_url)
area in waters of <5 m depth is almost 30,000 km² (HELCOM, 2013; Jakobsson et al., 2019), and, thus, equals 22% of the global areal extent of mangroves (Bunting et al., 2018) or 19% that of seagrass meadows (McKenzie et al., 2020). Thus, we: (a) postulate that nearshore habitats in northern temperate regions are understudied but seasonally relevant emitters of CH₄ to the atmosphere; (b) encourage including these habitats in future coastal CH₄ emission estimates, while also recognizing their pronounced seasonality; and (c) hypothesize that including these habitats amplifies the global ocean CH₄ budget significantly, especially when considering that macroalgae habitats alone contribute most (>50%) to the total global extent of coastal vegetation (Duarte et al., 2013).

4.8 Uncertainties in coastal CH₄ distribution and future research directions

Variations of surface water CH₄ concentrations and resulting sea–air fluxes reflecting the heterogeneous nature of coastal environments currently complicate generalizing regional patterns and upscaling attempts globally. Given the CH₄ distribution patterns identified in this study, we encourage several aspects to be considered to refine large-scale coastal CH₄ emission budgets.

First, studies currently used for global coastal CH₄ budgets have a site-selective bias due to their particular relevance in providing a service (e.g., they are interesting from a blue carbon perspective) and for other practical reasons like the accessibility of the study area. Here, we provided evidence that northern temperate coastal habitats, which are presently understudied for their contribution to CH₄ fluxes (e.g., algal communities on rocky shores), can be seasonally relevant sources of atmospheric CH₄. Similar measurements should be extended to additional coastal environments and geolocations to confirm the global relevance of their CH₄ emissions. The spatial heterogeneity of coastal habitats provides an opportunity for measurements along environmental gradients, with great potential to increase inference across scales (Snelgrove et al., 2014).

Second, new technical approaches have to be embraced to better understand the high temporal variability of the CH₄ distribution and the underlying processes in coastal environments. The use of continuous rather than time-averaged measurements helps to account for short-term temporal variations by diel cycles or peak events (Call et al., 2015; Maher et al., 2013; Rosentreter et al., 2018), and reduces uncertainties when establishing diel budgets. The high-resolution measurements across multiple seasons and the identification of dependencies on environmental variables have also bearings for predicting future CH₄ emissions under various changing environmental conditions.

Third, net annual CH₄ fluxes are influenced by temporal variations throughout the year. Thus, to increase confidence when compiling data for global coastal CH₄ budgets, better seasonal coverage of coastal CH₄ needs to be combined with the recognition that reported mean values (both CH₄ concentrations and emissions) might be biased toward sampling in a particular period only. As the seasonal behavior of CH₄ is highly site-specific, the variations need to be considered for each habitat type and geolocation.

Lastly, measurements of CH₄ emission from northern temperate and high-latitude coastal habitats should be acknowledged in future emission budgets. Climate change occurs particularly fast in northern hemisphere mid-latitude (Cohen et al., 2014) and high-latitude (Screen & Simmonds, 2010; Serreze et al., 2009) regions. Specifically, as Earth approaches an average warming of 2°C, some northern hemisphere high-latitude regions are expected to reach 4°C annual warming, outpacing the global average (Overland et al., 2014; Post et al., 2019). Although we could show a nonlinear behavior of CH₄ emissions with temperature, future studies aiming at resolving questions associated with climate change need to consider interannual rather than seasonal variations in CH₄ emissions and the balance of all important carbon pathways that influence CH₄ production pathways (Yvon-Durocher et al., 2014).

5 Conclusion

We conducted seasonal sampling campaigns of dissolved CH₄ concentrations and δ¹³C-CH₄ values using a fast-response automated gas equilibrator and CRDS system across three globally pervasive vegetated and nonvegetated coastal habitats. As the first study to compare high-resolution measurements across neighboring habitats, we highlight unprecedented spatiotemporal variability of the CH₄ distribution driven by habitat-specific CH₄ production and consumption pathways, seasonal temperature dependencies, and short-term fluctuations. A bootstrapping analysis on the continuous data revealed that scaling the CH₄ distribution from few samples involves large errors, and at least ~50 samples per day are needed to achieve accurate emission estimates. Failing to include such high-resolution measurements in future global CH₄ assessments may result in a continued systematic bias of regional and global estimates due to the lack of measurements representative for the coastal ecosystem mosaic—a highly heterogeneous environment in space and time. Ultimately, a better understanding of the habitat-specific contribution to the global CH₄ emission budget would improve efforts to address climate change, such as by revealing the net potential of coastal blue carbon habitats to sequester carbon.

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Conflict of Interest

The authors declare that they have no competing interests.
AUTHOR CONTRIBUTIONS
F.R., C.H., and A.N. involved in conceptualization; F.R., X.S., M.C.G., and C.H. involved in methodology; C.H., A.N., J.P., and V.B. involved in material and logistics; F.R., X.S., J.P., S.B., E.B., F.N., and C.H. investigated the study; F.R., X.S., J.P., C.H., and A.N. contributed in data interpretation and data analysis; F.R., X.S., C.H., and A.N. wrote the original draft; all authors reviewed and edited the manuscript.

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REFERENCES
Al-Haj, A. N., & Fulweiler, R. W. (2020). A synthesis of methane emissions from shallow vegetated coastal ecosystems. Global Change Biology, 26, 2988–3005. https://doi.org/10.1111/gcb.15046
Attard, K. M., Rodil, I. F., Berg, P., Norkko, J., Norkko, A., & Glud, R. N. (2019a). Seasonal metabolism and carbon export potential of a key coastal habitat: The perennial canopy-forming macroalga Fucus vesiculosus. Limnology and Oceanography, 64, 149–164. https://doi.org/10.1002/no.11026
Attard, K. M., Rodil, I. F., Glud, R. N., Berg, P., Norkko, J., & Norkko, A. (2019b). Seasonal ecosystem metabolism across shallow benthic habitats measured by aquatic eddy covariance. Limnology and Oceanography Letters, 4, 79–86. https://doi.org/10.1002/loi2.10107
Banerjee, K., Paneerselvam, A., Ramachandran, P., Ganguly, D., Singh, G., & Ramesh, R. (2018). Seagrass and macrophyte mediated CO2 consumption in anoxic marine sediments. Biogeosciences, 15, 79–86. https://doi.org/10.5194/bg-15-79-2018
Barker, J. F., & Fritz, P. (1981). Carbon isotope fractionation during microbial methane oxidation. Nature, 289, 289–291. https://doi.org/10.1038/292289a0
Barnes, R. O., & Goldberg, E. D. (1976). Methane production and consumption in anoxic marine sediments. Geology, 4, 297. https://doi.org/10.1130/0091-7613(1976)4<297:MPACIA>2.0.CO;2
Bastviken, D., Ejeertsson, J., & Tranvik, L. (2002). Measurement of methane oxidation in lakes: A comparison of methods. Environmental Science and Technology, 36, 3354–3361. https://doi.org/10.1021/es010311p
Bunting, P., Rosenqvist, A., Lucas, R., Rebelo, L.-M., Hilarides, L., Thomas, N., Hardy, A., Itoh, T., Shimada, M., & Finlayson, C. (2018). The global Mangrove watch—A new 2010 global baseline of Mangrove extent. Remote Sensing, 10, 1669. https://doi.org/10.3390/rs10011669
Call, M., Maher, D. T., Santos, I. R., Ruiz-Halpern, S., Mangion, P., Sanders, C. J., Erlér, D. V., Oakes, J. M., Rosentreter, J., Murray, R., & Eyre, B. D. (2015). Spatial and temporal variability of carbon dioxide and methane fluxes over semi-diurnal and spring-neap–spring tidescales in a mangrove creek. Geochimica Et Cosmochimica Acta, 150, 211–225. https://doi.org/10.1016/j.gca.2014.11.023
Call, M., Santos, I. R., Dittrar, T., de Rezende, C. E., Asp, N. E., & Maher, D. T. (2019). High pore-water derived CO2 and CH4 emissions from a macro-tidal mangrove creek in the Amazon region. Geochimica et Cosmochimica Acta, 247, 106–120. https://doi.org/10.1016/j.gca.2018.12.029
Cardinale, B. J., Srivastava, D. S., Emmett Duffy, J., Wright, J. P., Downing, A. L., Sankaran, M., & Jouseau, C. (2006). Effects of biodiversity on the functioning of trophic groups and ecosystems. Nature, 443, 989–992. https://doi.org/10.1038/nature05202
Cohen, J., Screen, J. A., Furtado, J. C., Barlow, M., Whittledon, D., Coumou, D., Francis, J., Dethloff, K., Entekhabi, D., Overland, J., & Jones, J. (2014). Recent arctic amplification and extreme mid-latitude weather. Nature Geoscience, 7, 627–637. https://doi.org/10.1038/ngeo2234
Dale, A. W., Flury, S., Fossing, H., Regnier, P., Ray, H., Scholze, C., & Jørgensen, B. B. (2019). Kinetics of organic carbon mineralization and methane formation in marine sediments (Aarhus Bay, Denmark). Geochimica Et Cosmochimica Acta, 252, 159–178. https://doi.org/10.1016/j.gca.2019.02.033
Denfeld, B. A., Baulch, H. M., del Giorgio, P. A., Hampton, S. E., & Karlsson, J. (2018). A synthesis of carbon dioxide and methane dynamics during the ice-covered period of northern lakes. Limnology and Oceanography Letters T. 3, 117–131. https://doi.org/10.1002/lol2.10079
Dias, G. M., Christofoletti, R. A., Kitazawa, K., & Jenkins, S. R. (2018). Environmental heterogeneity at small spatial scales affects population and community dynamics on intertidal rocky shores of a threatened bay system. Ocean and Coastal Management, 164, 52–59. https://doi.org/10.1016/j.ocecoaman.2017.12.001
Duarte, C. M., Losada, I. J., Hendriks, I. E., Mazzarrasa, I., & Marbà, N. (2013). The role of coastal plant communities for climate change mitigation and adaptation. Nature Climate Change, 3, 961–968. https://doi.org/10.1038/nclimate1970
Duarte, C. M., Middelburg, J. J., & Caraco, N. (2005). Major role of marine vegetation on the oceanic carbon cycle. Biogeoosciences, 2, 1–8. https://doi.org/10.5194/bg-2-1-2005
Ducharme-Riel, V., Vachon, D., del Giorgio, P. A., & Prairie, Y. T. (2015). The relative contribution of winter under-ice and summer hypolimnetic CO2 accumulation to the annual CO2 emissions from Northern Lakes. Ecosystems, 18, 547–559. https://doi.org/10.1007/s10021-015-9846-0
Dutta, M. K., Mukherjee, R., Jana, T. K., & Mukhopadhyay, S. K. (2015). Biogeochemical dynamics of exogenous methane in an estuary associated to a mangrove biosphere. Marine Chemistry, 170, 1–10. https://doi.org/10.1016/j.marchem.2014.12.006
Gülzow, W., Rehder, G., Schneider v. Deimling, J., Seifert, T., & Tóth, Z. (2013). One year of continuous measurements constraining methane emissions from the Baltic Sea to the atmosphere using a ship of opportunity. Biogeoosciences, 10, 81–99. https://doi.org/10.5194/bg-10-81-2013
Hartmann, J. F., Gentz, T., Schiller, A., Greule, M., Grossart, H. P., Ionescu, D., Keppler, F., Martínez-Cruz, K., Sepulveda-Jauregui, A., & Isenbeck-Schröter, M. (2018). A fast and sensitive method for the continuous in situ determination of dissolved methane and its 13C-isotope ratio in surface waters. Limnology and Oceanography: Methods, 16, 273–285. https://doi.org/10.1002/lom3.10244
Haugen, D. A. (1973). Workshop on micrometeorology. American Meteorological Society.
HELCOM. (2013). Red List of Baltic Sea underwater biotopes, habitats, and biotope complexes.
Hewitt, J. E., Thrush, S. F., & Dayton, P. D. (2008). Habitat variation, species diversity and ecological functioning in a marine system. Journal of Experimental Marine Biology and Ecology, 366, 116–122. https://doi.org/10.1016/j.jembe.2008.07.016
Hofmann, H., Federwisch, L., & Peeters, F. (2010). Wave-induced release of methane: Littoral zones as source of methane in lakes. Limnology and Oceanography, 55, 1990–2000. https://doi.org/10.4319/lo.2010.55.5.1990
Holland, K. T., & Elmore, P. A. (2008). A review of heterogeneous sediments in coastal environments. *Earth-Science Rev*, 89, 116–134. https://doi.org/10.1016/j.earscirev.2008.03.003

Humborg, C., Geibert, M. C., Sun, X., McCrackin, M., Mörrth, C.-M., Stranne, C., Jakobsson, M., Gustafsson, B., Sokolov, A., Norkko, A., & Norkko, J. (2019). High emissions of carbon dioxide and methane from the coastal Baltic sea at the end of a summer heat wave. *Frontiers in Marine Science*, 6, 1–14. https://doi.org/10.3389/fmars.2019.00493

Husson, F., Josse, J., Le, S., Mazet, J., & Husson, M. F. (2016). Package ‘FactoMineR’. An R Package, 96, 698.

Jähne, B., Münnich, K. O., Bösinger, R., Dutzi, A., Huber, W., & Libner, P. (1987). On the parameters influencing air-water gas exchange. *Journal of Geophysical Research*, 92, 1937. https://doi.org/10.1029/JC092iC02p01937

Jakobsson, M., Stranne, C., O'Regan, M., Greenwood, S. L., Gustafsson, B., Humborg, C., & Weidner, E. (2019). Bathymetric properties of the Baltic Sea. *Ocean Science*, 15, 905–924. https://doi.org/10.5194/os-15-905-2019

Karlsson, J., Giesler, R., Persson, J., & Lundin, E. (2013). High emission of carbon dioxide and methane during ice thaw in high latitude lakes. *Geophysical Research Letters*, 40, 1123–1127. https://doi.org/10.1002/grl.50152

Kassambara, A., & Mundt, F. (2017). Package ‘factoextra’. Extr. Vis. results Multivar. data Anal. 76.

Kim, J., Verma, S. B., & Billesbach, D. P. (2001). Seasonal variation in methane emission from a temperate Phragmites-dominated marsh: Effect of growth stage and plant-mediated transport. *Global Change Biology*, 5, 433–440. https://doi.org/10.1046/j.1365-2486.1999.00237.x

Kim, J., Verma, S. B., Billesbach, D. P., & Clement, R. J. (1998). Diel variation in methane emission from a midlatitude prairie wetland: Significance of convective throughflow in Phragmites australis. *Journal of Geophysical Research Atmospheres*, 103, 28029–28039. https://doi.org/10.1029/98JD02441

Koch, E. W. (2001). Beyond light: Physical, geological, and geochemical parameters as possible submersed aquatic vegetation habitat requirements. *Estuaries*, 24, 1. https://doi.org/10.2307/1352808

Laurila, T. (2021). ICOS ATC CH4 Release, Utö - Baltic sea (57.0 m), 2018-03-09–2021-01-31. https://hdl.handle.net/11676/m67T4

Lundevall-Zara, M., Lundevall-Zara, E., & Brüchert, V. (2021). Sea-air exchange of methane in shallow inshore areas of the Baltic Sea. *Frontiers in Marine Science*, 8, 1–20. https://doi.org/10.3389/fmars.2021.657459

Ma, X., Sun, M., Lennartz, S. T., & Bange, H. W. (2020). A decade of methane measurements at the Boknis Eck time series station in Eckernförde Bay (southwestern Baltic Sea). *Biogeosciences*, 17, 3427–3438. https://doi.org/10.5194/bg-17-3427-2020

Maher, D. T., Cowley, K., Santos, I. R., Macklin, P., & Eyre, B. D. (2015). Methane and carbon dioxide dynamics in a subtropical estuary over a diel cycle: Insights from automated in situ radioactive and stable isotope measurements. *Marine Chemistry*, 168, 69–79. https://doi.org/10.1016/j.marchem.2014.10.017

Marxer, M. T., Santos, I. R., Leuven, J. R. F. W., Oakes, J. M., Erler, D. V., Carvalho, M. C., & Eyré, B. D. (2013). Novel use of cavity ring-down spectroscopy to investigate aquatic carbon cycling from microbial to ecosystem scales. *Environmental Science and Technology*, 47, 12938–12945. https://doi.org/10.1021/es402777s

Marcelina, Z., Adam, S., & Pierre, R. (2018). Spatial and temporal variability of organic matter sources and food web structure across benthic habitats in a low diversity system (southern Baltic Sea). *Journal of Sea Research*, 141, 47–60. https://doi.org/10.1016/j.seares.2018.05.007

McKenzie, L. J., Nordlund, L. M., Jones, B. L., Cullen-Unsworth, L. C., Roelfsema, C., & Unsworth, R. K. F. (2020). The global distribution of seagrass meadows. *Environmental Seagrass Ecosystems: An International Journal*, 15, 074041. https://doi.org/10.1088/1748-9326/ab70d6

McLeod, E., Chmura, G. L., Bouillon, S., Salm, R., Björk, M., Duarte, C. M., Lovelock, C. E., Schlesinger, W. H., & Silliman, B. R. (2011). A blueprint for blue carbon: Toward an improved understanding of the role of vegetated coastal habitats in sequestering CO₂. *Frontiers in Ecology and the Environment*, 9, 552–560. https://doi.org/10.1890/110004

Medvedev, I. P., Rabinovich, A. B., & Kulikov, E. A. (2016). Tides in three encased basins: The Baltic, Black, and Caspian Seas. *Frontiers in Marine Science*, 3, 46. https://doi.org/10.3389/fmars.2016.00046

Murase, J., & Sugimoto, A. (2005). Inhibitory effect of light on methane oxidation in the pelagic water column of a mesotrophic lake (Lake Biwa, Japan). *Limnology and Oceanography*, 50, 1339–1343. https://doi.org/10.4319/lo.2005.50.4.1339

Nirmal Rajkumar, A., Barnes, J., Ramesh, R., Purvaja, R., & Upstill-Griffiths, W. O. (2005). Inhibitory effect of light on methane photoproduction to the oceanic methane paradox. *Nature Geoscience*, 12, 748–754. https://doi.org/10.1038/s41561-019-0421-8

Overland, J. E., Wang, M., Walsh, J. E., & Stoewe, J. C. (2014). Future Arctic climate changes: Adaptation and mitigation time scales. *Earth's Future*, 2, 68–74. https://doi.org/10.1002/2013EF000162

Phelps, A. R., Peterson, K. M., & Jeffries, M. O. (1998). Methane ef- flux from high-latitude lakes during spring ice melt. *Journal of Geophysical Research Atmospheres*, 103, 29029–29036. https://doi.org/10.1029/98JD00044

Post, E., Alley, R. B., Christensen, T. R., Macias-Fauria, M., Forbes, B. C., Gooseff, M. N., Iler, A., Kerby, J. T., Laidre, K. L., Mann, M. E., Olafsson, J., Stoewe, J. C., Ulmer, F., Virginia, R. A., & Wang, M. (2019). The polar regions in a 2°C warmer world. *Nature Geoscience*, 8, 479–486. https://doi.org/10.1038/s41561-019-0421-Z

Roelfsema, C., & Unsworth, R. K. F. (2020). The global distribution of macroalgae to oceanic carbon sequestration. *Frontiers of Marine Science*, 7, 1–10. https://doi.org/10.3389/fmars.2020.00046

R Core Team. (2021). R: A language and environment for statistical computing. R Foundation for Statistical Computing.

Reeburg, W. S. (1983). Rates of biogeochemical processes in anoxic sediments. *Annual Review of Earth and Planetary Sciences*, 11, 269–298. https://doi.org/10.1146/annurev.ea.11.050183.001413

Rodil, I. F., Attard, K. M., Gustafsson, C., & Norkko, A. (2021). Variable contributions of seafloor communities to ecosystem metabolism across a gradient of habitat-forming species. *Marine Environment Research*, 167, 105321. https://doi.org/10.1016/j.marenvres.2021.105321

Rosentreter, J. A., Al-Haj, A. N., Fulweiler, R. W., & Williamson, P. (2021a). Methane and nitrous oxide emissions complicate coastal blue carbon assessments. *Global Biogeochemical Cycles*, 35, 044034. https://doi.org/10.1029/2020GB006858

Rosentreter, J. A., Borges, A. V., Deemer, B. R., Holgerson, M. A., Liu, S., Song, C., Melack, J., Raymond, P. A., Duarte, C. M., Allen, G. H., Olefeldt, D., Poulter, B., Battin, T. I., & Eyré, B. D. (2021b). Half of global methane emissions come from highly variable aquatic ecosystem sources. *Nature Geoscience*, 14(4), 225–230. https://doi.org/10.1038/s41561-021-00715-2
WALLENIUS, A. J., DALCIN MARTINS, P., SLOMP, C. P., & JETTEN, M. S. M. (2021). Anthropogenic and environmental constraints on the microbial methane cycle in coastal sediments. *Frontiers in Microbiology*, 12, 631621. https://doi.org/10.3389/fmicb.2021.631621

WANNINKHOF, R. (2014). Relationship between wind speed and gas exchange over the ocean revisited. *Limnology and Oceanography: Methods*, 12, 351–362. https://doi.org/10.4319/lom.2014.12.351

WEBER, T., WISEMAN, N. A., & KOCK, A. (2019). Global ocean methane emissions dominated by shallow coastal waters. *Nature Communications*, 10, 1–10. https://doi.org/10.1038/s41467-019-12541-7

WIEBUSCH, D. A., & GUINASSO, N. L. (1979). Equilibrium solubilities of methane, carbon monoxide, and hydrogen in water and sea water. *Journal of Chemical and Engineering Data*, 24, 356–360. https://doi.org/10.1021/jc60083a006

WILSON, S. T., BANGE, H. W., ARÉVALO-MARTÍNEZ, D. L., BARNES, J., BORGES, A. V., BROWN, I., BULLISTER, J. L., BURGOS, M., CAPPÉLLE, D. W., CASSO, M., DE LA PAZ, M., FARIAS, L., FENWICK, L., FERRÓN, S., GARCÍA, G., GLOCKZIN, M., KARL, D. M., KOCK, A., LAPERRIERE, S., ... REHDER, G. (2018). An intercomparison of oceanic methane and nitrous oxide measurements. *Biogeosciences*, 15, 5891–5907. https://doi.org/10.5194/bg-15-5891-2018

YANG, Z., ZHU, Y., LIU, T., SUN, Z., LING, X., & YANG, J. (2019). Pumping effect of wave-induced pore pressure on the development of fluid mud layer. *Ocean Engineering*, 189, 106391. https://doi.org/10.1016/j.oceaneng.2019.106391

YVON-DUCHER, G., ALLEN, A. P., BASTVIKEN, D., CONRAD, R., GUDASZ, C., ST-PIERRE, A., THANH-DUC, N., & DEL GIORGIO, P. A. (2014). Methane fluxes show consistent temperature dependence across microbial to ecosystem scales. *Nature*, 507, 488–491. https://doi.org/10.1038/nature13164

ZEHNDER, A. J. B., & BROCK, T. D. (1980). Anaerobic methane oxidation: Occurrence and ecology. *Applied and Environment Microbiology*, 39, 194–204. https://doi.org/10.1128/AEM.39.1.194-204.1980

ZHANG, Y., & XIE, H. (2015). Photomineralization and photomethanification of dissolved organic matter in Saguenay River surface water. *Biogeosciences*, 12, 6823–6836. https://doi.org/10.5194/bg-12-6823-2015

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