Spectral evidence for substrate availability rather than environmental control of methane emissions from a coastal forested wetland

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\textbf{ABSTRACT}

Knowledge of the dynamics of methane (CH\textsubscript{4}) fluxes across coastal freshwater forested wetlands, such as those found in the southeastern US remains limited. In the current study, we look at the spectral properties of ecosystem net CH\textsubscript{4} exchange (NEE\textsubscript{CH4}) time series, and its cospectral behavior with key environmental conditions (temperature (T\textsubscript{s}), water table (WTD) and atmospheric pressure (P\textsubscript{a})), and physiological fluxes (photosynthesis (GPP), transpiration (LE), sap flux (J\textsubscript{s})) using data from a natural bottomland hardwood swamp in eastern North Carolina. NEE\textsubscript{CH4} fluxes were measured over five years (2012 - 2016) that included both wet and dry years. During the growing season, strong cospectral peaks at diurnal scale were detected between CH\textsubscript{4} efflux and GPP, LE and J\textsubscript{s}. This suggests that the well understood diurnal cycles in the latter processes may affect CH\textsubscript{4} production through substrate availability (GPP) and transport (sap flow and LE). The causality between different time series was established by the magnitude and consistency of phase shifts. The causal effect of T\textsubscript{s} and P\textsubscript{a} were ruled out because despite cospectral peaks with CH\textsubscript{4}, their phase relationships were inconsistent. The effect of fluctuations in WTD on CH\textsubscript{4} efflux at synoptic scale lacked clear indications of causality, possibly due to time lags and hysteresis. The stronger cospectral peak with ecosystem scale LE rather than J\textsubscript{s} suggested that the evaporative component of LE contributed equally with plant transpiration. Hence, we conclude that while the emission of dissolved gases through plants likely takes place, it may not contribute to higher CH\textsubscript{4} emissions as has been proposed by aerenchymatous gas transport in sedge wetlands. These findings can inform future model development by (i) highlighting the coupling between vegetation processes and CH\textsubscript{4} emissions, and (ii) identifying specific and non-overlapping timescales for different driving factors.

1. Introduction

Wetlands sequester nearly 30% of the global soil carbon despite limited geographical range (~ 8%) (Song et al., 2015). Wetlands also contribute significantly to global methane (CH\textsubscript{4}) emissions, a powerful greenhouse gas with a global warming potential 28 times greater than that of carbon dioxide (CO\textsubscript{2}) (IPCC, 2013). A 2.5-fold increase in CH\textsubscript{4} emissions since the preindustrial times has generated increased scrutiny of this process (Walter et al., 2001; Reeburgh, 2003; Denman et al., 2007; Meng et al., 2012 and Melton et al., 2016) and remains a big uncertainty from the perspective of global CH\textsubscript{4} estimates (Saunois et al., 2016; Melton et al., 2016).

\textit{Abbreviations: CO\textsubscript{2}, Carbon dioxide; WT, Continuous wavelet transformation; COI, Cone of Influence; CWT, Cross-wavelet transformation; DWT, Discrete wavelet transformation; ET, Evapotranspiration; GPP (umol m\textsuperscript{-2} s\textsuperscript{-1}), Gross primary productivity; PAR (umol m\textsuperscript{-2} s\textsuperscript{-1}), Photosynthetically active radiation

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empirical temperature response functions for CH4 production while dynamics of ecosystem CH4 exchange. Improved understanding of the underlying processes and the interaction of different drivers are needed to better capture the magnitude and dynamics of ecosystem CH4 exchange. CH4 exchange in wetlands is a multidimensional process. CH4 production occurs primarily in the anaerobic regions of the soil (Megenial et al., 2004), although production inside plants has also been hypothesized (Covey and Megenial, 2019). The emission of dissolved CH4 in soil water to the atmosphere can occur through diffusion or ebullition (Jeffrey et al., 2019), and may be modified by specialized plant anatomical structure, vascular architecture and rooting volume (Bhullar et al., 2013; Bhullar et al., 2014). The outgassing of CH4-saturated water may also occur in plant vasculature, along its transport pathway to leaves (Nesbit et al., 2009). While the transport of dissolved gases in xylem sap has been established (Teskey et al., 2008; Aubrey and Teskey 2009), its precise quantification is difficult. The diffusion of CH4 through the root and stem spongy tissues (aerenchyma) of some especially wetland-adapted species (e.g. sedges) has been hypothesized to result in elevated emissions compared to non-vegetated surfaces, as the off-gassing of CH4 through the plant allows it to bypass the microbial re-oxygenation in the anaerobic-aerobic interface in the soil (Bubier and Moore, 1994; Joabsson et al., 1999). Although some tree species also have aerenchymatous tissues (e.g. baldcypress, water tupelo and Atlantic white cedar), its role in ecosystem CH4 emissions is unclear. Finally, vegetation structure and activity may also affect CH4 production, as plant-derived carbohydrates may serve as substrates for archaeal methanogens in the soil (Whiting and Chanton, 1993; Christensen et al., 2003; Long et al., 2010). As plant carbohydrate status can affect soil CO2 production (Mitra et al., 2019), presumably through an effect on microbial substrate availability, it could also be an important regulator of archaeal methanogen activity. Other well-documented physical drivers of CH4 production include water table depth (WTD), atmospheric pressure, and temperature. Water table position operates as an ‘on/off’ switch for CH4 production. Water table drawdown aerates deeper soil horizons, altering the balance between methanogenic archaea and methanotrophic bacteria, reducing net CH4 efflux. Fluctuations in atmospheric pressure have been shown to facilitate CH4 release from wetlands, as the dissolution of gases in water is in part controlled by pressure (Tokida et al., 2007). Finally, temperature is often seen as the primary driver of biogeochemical fluxes as it controls the available energy (Arrhenius, 1889; Nahlik and Mitsch, 2011; Sachs et al., 2010), and many models include empirical temperature response functions for CH4 production while water level acts as a simple on/off switch (Olefeldt et al., 2013; Yvon-Durocher et al., 2014; Turetsky, 2014a,b; Vargas et al., 2010; Treat et al., 2018). Despite the universality of the temperature and water table response of CH4 efflux, such simple models oversimplify the system and may limit our ability to interpret observed CH4 flux dynamics. This is due to the fact that instantaneous combinations of water table and soil temperature overlooks well-known phenomena like time-lags and hystereses (Hatala et al., 2012; Moore and Dalva, 1993; Sachs et al., 2008; Meijide et al. 2011; Wille et al. 2008). An alternative to this rudimentary approach is a scale-dependent analysis of NEECH4 (Sturtevant et al., 2016). Given the number of steps to net CH4 emission (described above), decomposing the CH4 signals into low and high-frequency bands may shed light onto CH4 dynamics and its control mechanisms. This methodology has been successfully applied to identify the multi-scale drivers of CO2 flux across different biomes (Katul et al., 2001; Stoy et al., 2005; Vargas et al., 2010; Mitra et al. 2019).

In the current study, we will analyze the spectral properties of ecosystem net CH4 exchange time series, and its co-spectral behavior with key environmental conditions (temperature and water table) and physiological processes (photosynthesis, respiration and transpiration) using a 5-year record from a natural bottomland hardwood swamp in eastern North Carolina. We hypothesize that the functional dependence of CH4 exchange on any of these factors would manifest in co-spectral signatures with them. At the very least, spectral analysis will allow partitioning the sources of variability between fast-changing (radiation, vapor pressure deficit, stomatal conductance, photosynthesis), intermediate or synoptic-scale (atmospheric pressure, soil moisture availability, water table depth), and seasonal processes (seasonal changes in radiation, temperature, phenology). The current study focuses on the variation in CH4 and its drivers at the first two of the three timescales.

2. Methods

2.1. Study site

The study site (35°47′16.32″N; 75°54′13.74″W) is located in Alligator River National Wildlife Refuge on the Albemarle-Pamlico peninsula in Dare County of eastern North Carolina, USA. The site is registered as US-NC4 in the Ameriflux database. Established in 1984, Alligator River National Wildlife Refuge is characterized by a heterogeneous conglomeration of pocosin wetland types (Allen et al., 2011). Overstory vegetation across this freshwater forested wetland consist primarily of water tupelo (Nyssa aquatica), red maple (Acer rubrum), swamp tupelo (Nyssa biflora), along with bald cypress (Taxodium distichum), sweetgum (Liquidambar styraciflua), white cedar (Chamaecyparis thyoides) and loblolly pine (Pinus taeda). The understory vegetation consists of fetterbush (Lyonia lucida), bitter gallberry (Ilex alba), red bay (Persea borbonia), and sweet bay (Magnolia virginiana). The 30-year (1981-2010) mean temperature and precipitation were 16.9°C and 1270 mm yr−1 (measured at Manteo airport, NC, about 32 km from the study site). Canopy height across the site ranged from 15 to 20 m, with leaf area index peak at 4.0 ± 0.3 in early July from a minimum of 1.3 ± 0.3 during the non-growing season (Domec et al., 2015). The pH of soil at the surface varied between 4.2 and 4.8 (Minick et al., 2019b). Bulk density of dry soil was 0.08 ± 0.02 g cm−3 (mean ± SD). The primary soil types are poorly drained Pungo and Belhaven mucks with their corresponding organic carbon content varying between 40 and 100% and 20 and 100%, respectively (Web Soil Survey, Miao et al., 2017). Precipitation is the primary source of

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| List of symbols | 
|------------------|------------------|
| $p(x,y)$ | Joint probabilities |
| $CH_4$ (umol m$^{-2}$ s$^{-1}$) | Methane |
| $NEE_{CH4}$ (umol m$^{-2}$ s$^{-1}$) | Net ecosystem CH4 exchange |
| $NEE_{CO2}$ (umol m$^{-2}$ s$^{-1}$) | Net ecosystem CO2 exchange |
| $P_a$ (kPa) | Atmospheric pressure |
| $T_a$ (°C) | Air temperature |
| $P_s$ (kPa) | Atmospheric pressure |
| $LE$ (W m$^{-2}$) | Ecosystem-scale latent energy |
| $H(x)$ | Entropy of variable $x$ |
| $H(y)$ | Entropy of variable $y$ |
| $J(x,y)$ | Joint entropy of $x$ and $y$ |

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freshwater for this refuge, and astronomical tides are absent due to the unique combination of geomorphic features and lagoon environments (Moorhead and Brinson, 1995). Other features of this site include the presence of micro-topography, the absence of runoff, and low-intensity drainage.

2.2. Gas scale measurements

Ecosystem-scale turbulent fluxes of latent energy (LE), CO₂ and CH₄ exchange were measured with eddy covariance method (Baldocchi et al., 1988). The instrumentation consisted of 3-D sonic anemometer thermometer (Windmaster, Gill Instruments Limited, Hampshire, UK), an open path CH₄ analyzer (LI-7700, LI-COR, Lincoln, NE, USA), and an enclosed-path CO₂ and H₂O analyzer (LI-7200, LI-COR). In addition to turbulent fluxes, we also measured CO₂ concentration (LI-820 infrared gas analyzer and a multi-port system) along the height of the canopy to estimate canopy storage of CO₂, which was added to the turbulent fluxes of CO₂ to quantify net ecosystem exchange of CO₂ (NEE). Detailed information on calculation of CO₂ concentration across our site has already been well documented (Miao et al., 2017).

Net ecosystem exchange of CH₄ (NEE) quantified the net CH₄ sequestered or lost by the ecosystem. The sampling frequency of the raw data was 10 Hz and was recorded using a LI-7550 auxiliary interface unit (LI-COR, Nebraska, United States). The average canopy height at the site was 20 m, and the height of the turbulent flux measurements was 28.2 m in 2012, and 33 m since 2013.

The turbulent fluxes were calculated and corrections applied with Eddypro software (v 6.1.0, LI-COR). The different corrections can be briefly summarized as follows: screening for spikes (Vickers and Mahrt, 1997), rotation of sonic anemometer wind vectors (Wilczak et al., 2001), detrending the raw time series (block averaging), correction of the time lags between scalar concentration and rotated vertical wind speed, and corrections for variations in air density (Webb et al., 1980).

NEE fluxes measured by the LI-7700 open path analyzer were corrected for spectroscopic effects along with the density corrections (Burba et al., 2019). The spectroscopic corrections are required for narrow band laser analyzers to account for the impact of temperature and pressure on the shape of the absorption feature. High (Ibrom et al., 2007) and low pass (Moncrieff et al., 2004) filtering corrections were also applied, and low-quality flux outputs were discarded (Mauder and Foken, 2006). Post-processing of the 30-minute fluxes include filtering the data corresponding to low signal strength (<10%) and integral turbulence characteristics, removal of outliers (Papale et al., 2006) and friction velocity thresholding (Goulden et al., 1996). As the tower is surrounded by homogeneous vegetation, there was no directional variability in NEE. Final data coverage of NEE and NEECO₂ were 27-47% and 60-75% respectively, depending on a year. NEECO₂ (Supp. Figure S1) was partitioned to gross primary productivity (GPP) and ecosystem respiration by modeling nighttime NEECO₂ as a function of air temperature using a Q₁₀ model. This particular function was used to predict day and night ecosystem respiration and GPP was quantified as the sum of NEECO₂ and modeled ecosystem respiration.

2.3. Meteorological measurements

Micrometeorological variables that were measured above canopy included air temperature (Tₐ) and relative humidity (RH; HMP45, Vaisala, Finland), photosynthetically active radiation (PAR, PARLITE, Kipp & Zonen, Delft, Netherlands (KZ)), precipitation (TE525, Texas Electronics) and atmospheric pressure (Pₐ) was measured using a pressure sensor (CS 105, Campbell Scientific, Logan, UT, USA). Belowground measurements include soil temperature at 5 cm (Tₜ₅) and 20 cm depth (Tₜ₂₀; CS107, Campbell Scientific). Water table depth (WTD) was measured with a pressure water level data logger (Infinities, Port Orange, FL, USA). All the meteorological variables were measured at 10 sec interval and averaged every 30 minutes.

2.4. Sap flow measurements

Granier-type sap flux sensors were used to monitor transpiration at the individual tree scale (Granier 1985, 1987). Five trees per each predominant species (pond pine, water tupelo, red maple and bald cypress, accounting for more than 75% of the total tree basal area) were instrumented near the instrument tower. The 20 mm sensors were installed on the north side of each tree (at diameter at breast height ~ 1.3 m) to reduce the effect of radiation on sap flow estimates. The sensors were covered with aluminum foil to protect the sap flux sensors from external perturbations, including precipitation, as well as minimize temperature fluctuations that would arise from factors other than the sap flow.

The temperature difference data from each sap flux sensor was converted to sap velocity (Jₜ, g m⁻² s⁻¹) (Ward et al., 2017). Final Jₜ was the basal-area-weighted sum of Jₜ of the three main canopy species listed above, with the underlying assumption that fluctuations in Jₜ is proportional to transpiration dynamics (Small and McConnell, 2008). Temperature difference data from each sensor were recorded as half-hourly averages using a CR1000 data logger (Campbell Scientific). Further information on sapflow-derived tree transpiration at this site can be found in Domec et al. (2015).

2.5. Spectral analysis

To evaluate the contribution of different environmental and physiological factors with different time constants to the temporal dynamics of NEE, we analyzed the wavelet spectra of NEE and co-spectra of NEE with presumed independent scalar (GPP, LE, Jₜ, Tₛ, WTD, Pₐ) time series in the frequency domain. LE and Jₜ were used as the proxies of CH₄ transport in tree sap and GPP represented the impact of carbon assimilation on NEE. Wavelet transformation generated average wavelet spectrum, defined as the average variance (or energy) associated with specific frequencies for the whole data period. The dominant scale was identified by the maximum of wavelet variance (Kumar and Foufoula-Georgiou, 1997).

Wavelet analysis is advantageous on account of localization in both temporal and scale domain, making it robust to deal with non-stationarities in data. Continuous wavelet transformation with the Morlet (Grinsted et al., 2004) function was used to investigate the oscillation of NEE in space and time. The mathematical theory behind this time series strategy has been extensively documented (Torrance and Compo, 1998; Grinsted et al., 2004; Stoy et al., 2013) and applied in our previous work on soil respiration (Mitra et al., 2019).

Apart from average wavelet spectrum, co-spectral or Cross-wavelet transformation (CWT) analysis also generated heatmap that provided a qualitative assessment if one time series was related to another via phase diagram. This was accomplished by analyzing regions of high correlation (red colored areas in heat maps) and at specific frequency (sub-daily, daily, synoptic) in the heat map. In the heat maps, the vertical axis indicated the different frequencies while the horizontal axis referred to the time of year. The arrows in the heat map only represent a lag at one certain time and frequency. Direction of arrows in the heat maps provide an understanding of the nature of coupling between NEE and causal scalar (GPP, LE, Jₜ, Tₛ, WTD, or Pₐ) time series. Arrows in the heat map pointing at right and left would indicate positive and negative correlation between NEE and drivers, with no lags. NEE lagged the driver when arrows pointed at up-right (positive correlation) and down-left (negative correlation) direction. Arrows pointing at up-left and down-right direction indicate the driver lagging NEE.

Causality was inferred only when NEE (i.e. effect) was either in sync or lagged behind the drivers (i.e. the cause) (Banfi and...
Ferrini, 2012). Another critical aspect identified in the heat map is the cone of influence. Cone of influence is defined as the boundary within which wavelet transformation is not affected by edge effects. Transformations beyond the cone of influence are ignored. Edge effect arises from the lack of sufficient low frequency data at the beginning and end of the data series.

Given the time constant of processes of interest in this study, we constrained the sampling rate for cospectra between 12 hours and 30 days. Data were normalized to have zero mean and unit variance and gaps in the data were padded with zeros. Padding with a constant does not affect co-spectral power (Mitra et al., 2019) unless the gaps are systematically distributed (which was not in our data). We confirmed the absence of spurious spectral peaks by using a null time series of red noise with the same gap structure as the observations, and evaluated the WT and CWT spectra and cospectra, similar to Mitra et al. (2019).

2.6. Lag quantification

Instead of the heat maps, we adopted the non-decimated discrete wavelet transformation (DWT) with Haar basis function (Mahrt, 1991; Katul et al., 2001) to quantify lags between \( \text{NEE}_{\text{CH4}} \) and GPP, LE and \( J_s \) at different scales (Whitcher et al., 2000). For this, the time series of \( \text{NEE}_{\text{CH4}} \), GPP, LE and \( J_s \) was first decomposed into multiple scales (d1 – d11). The different scales (d1-d11) can broadly be classified into diurnal (1 hour – 1.33 days/ d1-d6), synoptic (2.67 - 21.33 days/ d7-d10) and phenological (42.67 days/d11) frequencies. \( \text{NEE}_{\text{CH4}} \) and GPP, LE and \( J_s \) time series at each scale were lagged by ±12 hours. Typically, \( J_s \) lags LE due to water storage within trees (Maltese et al., 2018). This lag time between LE and \( J_s \) was determined using the above-mentioned approach and adjusted prior to the calculation of their individual lags against \( \text{NEE}_{\text{CH4}} \). The highest statistically significant (\( p < 0.05 \)) cross-correlation wavelet coefficient along with the corresponding lag hours have been reported. If more than one independent scalars preceded \( \text{NEE}_{\text{CH4}} \), the variable with the shortest lag time was identified as the controlling factor, following Koebisch et al. (2015).

2.7. Information theory analysis

Wavelet multiresolution analysis was combined with information theory metrics (Shannon, 1948) to have a better understanding of the mutual dependency between \( \text{NEE}_{\text{CH4}} \) and its drivers across different scales. Multiresolution analysis consisted of wavelet transformation of an original time series to compute low and high pass filters (d1 – d11). The Haar basis function (Mahrt, 1991) was used for wavelet transformation.

The information theory metrics of interest was mutual information content. Mutual information content can be defined as the amount of information that can be gained about variable \( x \) when information about another variable \( y \) is provided. Mutual information content is greater than zero, with higher values indicative of a larger reduction in uncertainty. Mathematically, mutual information content (\( MI \)) can be expressed as follows:

\[
MI(x, y) = H(x) + H(y) - J(x, y) = \sum_{i} \sum_{j} p(x_i, y_j) \log \frac{p(x_i | y_j)}{p(x_i)}
\]

Where \( H(x) \) and \( H(y) \) refers to the entropy of each variable and \( J(x, y) \) is the joint entropy, and \( p(x_i, y_j) \) and \( p(x_i | y_j) \) are the joint and conditional probabilities respectively. In this analysis, \( x \) was the \( \text{NEE}_{\text{CH4}} \) time series, and \( y \) was each level of high and low band pass filtered versions of \( \text{NEE}_{\text{CH4}} \), GPP, LE, \( J_s \), \( T_{sd} \), \( P_a \), and WTD. This allowed us to quantify the contribution of each time scale to the predictive uncertainty of \( \text{NEE}_{\text{CH4}} \) surface exchange.

As the different measurements had different magnitude and units, all-time series were normalized to have zero mean and unit variance. This allowed all datasets to be treated equally, before wavelet and cross

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Fig. 1. Seasonal variation of 30-minute air temperature \( (T_a) \) (Fig. 1A – E), soil temperature at 5 cm depth \( (T_{so}) \) (Fig. 1F – J), atmospheric pressure \( (P_a) \) (Fig. 1K-O), water table depth \( \text{WTD} \) (Fig. 1P-T), gross primary productivity \( \text{GPP} \) (Fig. 1U-Y), latent energy \( \text{LE} \) (Fig. 1Z-A4), sap flux density \( (J_s) \) (Fig. A5 – A9) and net ecosystem exchange of methane \( \text{NEE}_{\text{CH4}} \) (Fig. A10-A14) for US-NC4 across different years (2012, 2013, 2014, 2015, 2016).
3. Results

Daily average air temperature ($T_a$) ranged from 5°C in January to 28°C in July (Fig. 1A-E). $T_a$ generally remained above 25°C from the middle of June until the end of July. Soil temperature at 5 cm depth ($T_s$) displayed a similar pattern with a lower amplitude (Fig. 1F-J). The most pronounced changes in WTD were in response to the annual fall storms (Fig. 1P-T). The 5-year study period had normal to above average precipitation compared to the 30-year normal (Supp. Figure S2).

NEECH4 (Fig. 1A10-A14) had a strong seasonal pattern, increasing from the beginning of May to July, and declining rapidly from September to October, where it stayed near annual minimum until April. Although the seasonality patterns of GPP (Fig. 1U-Y), LE (Fig. 1Z-A4), and $J_s$ (Fig. 1A5-A9) were very similar, their cospectra with NEECH4 exhibited distinct differences, including distinct differences in the cospectral peak height (Fig. 2).

Oscillation of NEECH4 was statistically significant at the diurnal scale, with the presence of weak but significant variance also at synoptic scales (Fig. 2A-E). Mutual information between CH4 and its different band-filtered versions decreased with increase in dyadic time scales (Supp. Fig. S3 A-E). Cospectral analysis between NEECH4 and GPP found significant variance at the diurnal scale, and weak but significant interactions at the synoptic scale, as well (Fig. 2F-J). The multiscale analysis found a relative reduction in uncertainty in the estimation of NEECH4 by GPP at diurnal time scales ($\sim$3-6d) (Supp. Fig. S3 F-J). The only exception was in 2013 (Fig. S3G) when the lowest reduction in uncertainty was at the phenological time scale (d9-d11).

Heat maps revealed strong correlation between GPP and NEECH4 during the growing season (May – September) at the diurnal scale (Period $\sim$1) (Fig. 3A, Supp. Figures S4). The two signals demonstrated a strong covariance with no lags (arrow at right) or NEECH4 slightly lagged GPP (arrow at up-right) in 2012 (Fig. 3A), 2014 (Supp. Fig. S4B) and 2016 (Supp. Figure S4D). The relationship between the two time series was inconsistent in 2013 (Supp. Figure S4A), which we attribute to greater data gaps in this year. Based on direction of arrow in the heat map (down-left or left), there were also brief periods when GPP and NEECH4 were negatively correlated (Supp. Fig. S4A & C).

Cospectral analysis between NEECH4 and LE (and $J_s$) yielded similar insight with strong variance at daily scale and weak but significant trends at the monthly time step (Fig. 2K-T). Mutual information analysis found the high-frequency band-filtered versions of LE (scales d4-d6) contributing to a greater reduction of uncertainty of NEECH4 (Supp. Fig. S3 K-O). Mutual information analysis also found a reduction in the uncertainty for NEECH4 by LE at the phenological time scale (d11) (Supp. Figures S3 L-O). Information theory analysis between NEECH4 and $J_s$ also yielded similar outcomes (results not shown). The magnitudes of the diurnal cospectral peaks with NEECH4 were similar, but slightly declining in the order of $GPP > LE > J_s$ (Fig. 2).

Heat maps highlighted strong correlation between NEECH4 and LE at the diurnal scale (Period $\sim$1) (Fig. 3B). Both the fluxes oscillated positively with no lags (arrows pointed at right) in 2012, 2015 and 2016 or NEECH4 lagged (arrows pointed at up-right) LE in 2012 (Fig. 3B, Supp. Fig. S5 & D). Data gaps were too large in 2013 (Supp. Fig. S5A) to establish a consistent relationship. In 2014 and 2015, there were instances when LE fluxes lagged NEECH4 (arrows at down-right, Supp. Fig. S5B & C). LE and NEECH4 were positively coupled at the synoptic scale for brief periods during the growing season in 2012, 2013, and 2014 (Fig. 3B, Supp. Fig. S5A & B). The heat maps between NEECH4 and $J_s$ showed similar peaks, but since the peak magnitude was slightly lower (5-35%) than for LE, the figures are not shown.

$NEECH4$, $T_{s5}$ cospectra were characterized by significant interactions at the synoptic scale (Fig. 2U-Y), and with small but statistically significant cycles also detected at the daily time step in 2013, 2014 & 2015 (Fig. 2V-X). MI analysis found the NEECH4-$T_{s5}$ interaction to be also strong at the same time scales in all years (Supp. Fig. S3 P-T). The diurnal timestep of $T_{s5}$ was also found to significantly reduce uncertainty in NEECH4 in 2014 and 2015 (Supp. Fig. S3 R & S). Based on direction of arrows in the heat maps, the nature of coupling between $T_{s5}$ and NEECH4 was inconsistent at the synoptic scale for all years (Fig. 3C, Supp. Figure S6).

$P_a$ covaried with NEECH4 at synoptic and monthly frequencies
This was supported by the multiscale analysis, which found $MI$ to increase exponentially, highlighting the importance of low-frequency components of $P_a$ driving the dynamics of $NEE_{CH4}$ (Supp. Fig. S3 U-Y). In the heat maps, areas of high common power between $P_a$ and $NEE_{CH4}$ during the early part of the growing season yielded no consistent relationship from periods 4-16 days across all years (Fig. 3D, Supp. Fig. S7). Cospectral analysis between $NEE_{CH4}$ and $WTD$ was characterized by significant interactions at the synoptic and monthly scale (Fig. 2A5-A9). $MI$ analysis also found the longest time scales of water table dynamics (d12, Supp. Fig. S3 Z-A4) driving the variation in $NEE_{CH4}$ exchange. In the heat maps, strong correlation between $WTD$ and $NEE_{CH4}$ corresponding to periods greater than four days were primarily concentrated during the late fall season (Supp. Fig. S7C & D). Arrows in the heat maps could not definitively establish causality as $NEE_{CH4}$ sometimes lagged (arrows pointed at up-right (positive correlation) and down-left (negative correlation) direction. Arrows pointing at up-left and down-right direction indicate the driver lagging $NEE_{CH4}$. Red colored areas in heat maps surrounded by white lines indicate areas with 5% significance level.

The degree of association for $NEE_{CH4}$ with $GPP$, $LE$ and $J_s$ was quantitatively analyzed for dyadic scale 6 (~1.3 days) during the growing season as the strongest variance between the time series were concentrated during that particular phase (Fig. 3A&B, Supp. Fig. S4 &S5). $GPP$ and $NEE_{CH4}$ were positively correlated with the latter (effect) lagging the former (cause) by 0-4.5 hours at scale 6 (Fig. 4A). $J_s$ lagged $LE$ by 0.5 to 1.5 hours (2012 – 2016) at dyadic scale 6. After adjusting for this lag time, the lag analysis for $NEE_{CH4}$ with $LE$ ($& J_s$) found the latter leading the former only in 2012 and 2013, with shorter lag for $LE$ in 2013 (Fig. 4B & C). The lags were shorter in both years for $LE$ than for $GPP$.

4. Discussion

Scalar analysis of CH$_4$ variability has both theoretical and practical

![Fig. 3. Heat maps highlighting for 2012, the cross-wavelet transformation (CWT) between net ecosystem exchange of methane ($NEE_{CH4}$) and different drivers including A) gross primary productivity ($GPP$); B) latent energy ($LE$); C) soil temperature at 5 cm depth ($T_s$); D) atmospheric pressure ($P_a$) and E) water table depth ($WTD$). Arrows in the heat map pointing at right and left would indicate positive and negative correlation between $NEE_{CH4}$ and drivers, with no lags. $NEE_{CH4}$ lagged the driver when arrows pointed at up-right (positive correlation) and down-left (negative correlation) direction. Arrows pointing at up-left and down-right direction indicate the driver lagging $NEE_{CH4}$. Red colored areas in heat maps surrounded by white lines indicate areas with 5% significance level.](image1)

![Fig. 4. Degree of association (lag/lead) of $NEE_{CH4}$ with $GPP$ (A), $LE$ (B), and $J_s$ (C) for dyadic scale 6 (~1.3 days).](image2)
implications. By identifying the requisite scale and time, the predictive relationship between CH₄ and biotic/abiotic drivers can be improved with potential to reduce uncertainty in CH₄ budget (Yates et al., 2007). It can also help to develop a comprehensive theoretical framework with regard to the drivers of NEECH₄. While the full development of these capabilities is beyond the scope of this study, our results offer strong support to the view that the short-term variability in methane is driven by substrate availability from photosynthesis, while correlations with environmental conditions appear merely coincidental, resulting from the same radiation signal that drives GPP.

NEECH₄ exhibited significant variance at sub-diaily and diurnal scale with weak variance at synoptic, and phenological timescales (Fig. 2A-E, Supp. Fig. S3A-E). Although the finding of systemic diurnal structure in NEECH₄ is not universal, it is consistent with the reports of Dacey and Klug (1979), Rambaud et al. (1977), Joabsson et al. (1999), Hatala et al. (2012), Godwin et al. (2013) and Matthes et al. (2014), and follows the spectral properties reported for NEE (Kutul et al., 2001; Braswell et al., 2005; Stoy et al., 2009; Mitra et al., 2019).

Assuming that the power spectrum of response variables (NEECH₄ in this case) is influenced by the power spectra of environmental and biological forcing, and that a cause precedes effect, we will attribute the control of NEECH₄ to different environmental factors and biological processes based on the power cospectra of these different driving processes with NEECH₄ and the phase lag between the two time series. For example, the strong cospectral peak between NEECH₄ and GPP at the daily frequency (Fig. 2F-J, Supp. Fig. S3 F-J) is consistent with earlier observations across a wide range of wetlands, from subarctic peatlands to subtropical marshes (Whiting and Chanton, 1993; Matthes et al., 2014). The consistency in the phase direction (right at both right or up-right) between NEECH₄ and GPP (Fig. 3A, Supp. Fig. S4) suggest a possible causal connection between the two time series. This hypothesis of NEECH₄ regulation by carbohydrate substrate supply from GPP is supported by observations that new photosynthates are the preferred carbon substrate for methanogens (Chanton and Whiting, 1996; Minoda and Kimura, 1994; Hatala et al. 2012; Dorodnikov et al., 2011; Ström et al., 2012).

The length of the time lag between GPP and NEECH₄ (a few hours: Fig. 4A) is consistent with the rate of spread of carbohydrate pressure-concentration waves throughout the plant (Thompson and Holbrook, 2003), and similar to time lags being observed between GPP and soil CO₂ efflux in many earlier reports (Mitra et al., 2019; Liu et al., 2006). It is important to note that studies using tracer analysis invariably observe longer time periods for actual mass flow of carbohydrates (Kuzyakov and Gavrichkova, 2010; Mencuccini and Hölttä, 2010; Wingate et al., 2010). The mass flow of carbohydrates has a finite and rather uniform speed, usually around 20-30 cm hr⁻¹, and thus the delay of assimilation and label detection in target tissues is proportional to the length of the transport pathway, whereas pressure-concentration waves alter carbohydrate availability throughout the plant at a shorter timescale, and the effect is independent of distances involved (Kuzyakov and Gavrichkova, 2010; Thompson and Holbrook, 2003).

There were also periods when GPP and the NEECH₄ time series were either decoupled (Supp. Fig. S4B) or negatively correlated (Supp. Fig. S4A & C). This could be indicative of suppressed CH₄ production and emission (for example, possibly due to increased CH₄ oxidation; Strom et al., 2005; Bouchard et al., 2007), or the dominance of an alternative control mechanism over substrate limitation. One such alternative mechanism could be the transport of dissolved CH₄ with xylem sap, that is then released with transpiration, or that could also diffuse out through the stem along the way (Barba et al., 2019). This would explain the strong covariances of CH₄ at diurnal scale with both LE and Jₜ (Fig. 2K-T, Supp. Fig. S3 K-O). Like GPP, LE and Jₜ are tightly regulated by stomatal conductance, and partitioning their effect on NEECH₄ with spectral analysis tools is challenging. The cospectral peak between LE and GPP (Supp. Figure S9, only shown for 2012) is very similar in shape to the cospectrum of NEECH₄ and GPP, except about 4-fold greater in amplitude. Nevertheless, the lower power of the NEECH₄ cospectrum with Jₜ than with either GPP or LE (Fig. 2) speaks against the dominance of emissions by transport of dissolved CH₄ in the sap. Indeed, the phase angles of the NEECH₄ and LE also exhibited decoupling at the same time as those of NEECH₄ and GPP cospectra (Supp Figs S4A, S4C, S5A, S5C).

Finally, the evidence for GPP-mediated control of NEECH₄ is bolstered by the lag time analysis. It has been proposed that the variable with the shortest lag interval would be the primary driver (Koebsch et al., 2015). There is more and stronger evidence for the substrate availability-based limitation on NEECH₄ than for stomatically controlled release of CH₄ with the transpiration stream (although the latter is not insignificant). The fact that in some years NEECH₄ peaks preceded Jₜ (and LE) suggests that their relationship was correlative, or confounded by other processes (Supp Figures S5B & S5C). Furthermore, the shorter lag time for NEECH₄ vs LE relationship compared to NEECH₄-vs-Jₜ in 2013 (Fig. 4 B & C) suggest that the evaporative component of ET was equally important to the transport pathway with transpiration. It appears that wherever the evaporation of water occurs, CH₄ dissolved in the water gets released, and the trees do not appear to represent a preferred low-resistance pathway for CH₄ emission (Barba et al., 2019).

The relationship of NEECH₄ with environmental drivers Tₛ, Pₛ and WTD did not lend strong support to widely accepted view of biophysical regulation of CH₄ production in wetlands. While these factors covaried with NEECH₄ at diurnal to synoptic time scales, similar to earlier reports (e.g. Tageson et al., 2012; Hanis et al., 2013; Song et al., 2015; Klapstein et al., 2014; Tokida et al., 2007), the phase direction indicating temporal offsets were either inconsistent (Pₛ, Tₛ, WTD) (Supp. Figures S6-S8) or negative at diurnal scale (Tₛ) (Supp. Figure S6C), indicating correlative rather than causal relationship with NEECH₄.

Large rain events, especially during the hurricane season in the fall, were usually associated with significant changes in NEECH₄, and the cospectra of NEECH₄ and WTD showed strong peaks in 2013, 2015 and 2016 (Fig. 2 A5 – A9), but the phase angles (Supp. Fig. S8) failed to indicate consistent causality in this pattern. We therefore conclude that the effect of water table must have been mediated by some other process, possibly plant physiological status. Alternatively, the cospectra may have been obscured by time lags and hysteretic responses. For example, the substrate pools and activation times of different microbial populations may cause delayed responses on account of short-lived peaks in water table (e.g. Kettunen et al., 1999; Blodau and Moore, 2003; Knorr et al., 2008). It is also possible that there is a threshold response of NEECH₄ to fluctuations in the water table, for example, as shown for CO₂ by Miao et al. (2013). Similarly, Brown et al. (2014) showed that maximum methanogen activity occurred at a particular critical water table depth at which the redox potential and substrate availability were at the optimum.

While the evidence for GPP-mediated control of NEECH₄ appears strong and holds up to different angles of scrutiny, it could be asked if omitting CH₄ storage in the canopy profile could have affected the findings. As CH₄ profile was not measured at the study site, the only option for estimating canopy storage was via the single-point rate of change approach (Hollinger et al., 1994), which in our experience is very unreliable for estimating CO₂ storage in tall canopies, including at the current study site. Nevertheless, the inclusion of this storage estimate did not significantly alter the spectral signature of NEECH₄, as well as the cospectra with GPP (data not shown). In addition, the magnitude of the daily cospectral peaks and the lag times remained the similar to those without storage.

In summary, these findings call for re-evaluation of the common correlative patterns as the current way to model CH₄ production in wetlands (e.g. Bridgham et al., 2013; Chu et al., 2014; Turetsky et al., 2014). The spectral analysis reported here has identified certain time (growing season) and frequency (diurnal) patterns that dominate the multidimensional interaction between NEECH₄ and its biotic and abiotic drivers. We posit that the insights of the significance of GPP-mediated
substrate availability for soil microbial activity (Mitra et al., 2019) have the potential to stimulate emergent ideas for model development. One such approach could be time-varying parameters with specific spectral constraints. Second, the role of dissolved \( \text{CH}_4 \) transport in the sap may benefit from the detailed representation of plant hydraulic properties, and stem water storage that are being introduced to ecosystem models like CLM and Noah-MP (Li & Matheny, personal communication).

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Supplementary materials


References


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