



Seasonal Variation of Methane Fluxes in a Mangrove Ecosystem in South India: An Eddy Covariance-Based Approach

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Abstract

The atmospheric methane (CH_4) concentration has increased in recent years due to natural and anthropogenic causes. Hence, it is essential to quantify the potential sources of CH_4 to understand the factors responsible for its fluxes on a local to regional scale through in situ observations. Coastal wetlands, particularly the mangrove ecosystems in the tropical and subtropical coasts, are significant sources of CH_4 . In this study, we used an eddy covariance-based technique to measure the CH_4 fluxes in a mangrove ecosystem in Pichavaram, South India. The daily mean CH_4 flux ranged from 12 to 26 $\text{nmol m}^{-2} \text{s}^{-1}$ during the wet season and from 6 to 20 $\text{nmol m}^{-2} \text{s}^{-1}$ during the dry season. The monthly mean flux during the wet period was between 0.8 and 1.8 $\text{g CH}_4 \text{m}^{-2} \text{month}^{-1}$, and in the dry season, it was between 0.4 and 0.6 $\text{g CH}_4 \text{m}^{-2} \text{month}^{-1}$. The visual correlogram and structural equation modelling technique revealed that air temperature, creek water dissolved oxygen, soil organic carbon, and redox potential are important factors that control the CH_4 fluxes. The results suggest that the Pichavaram mangrove wetland acts as a source for CH_4 . Our results also indicate that tidal inundation and seasonal variations in atmospheric temperature and water salinity are key factors affecting the CH_4 flux in the Pichavaram mangrove ecosystem.

Keywords Tropical mangroves · Eddy covariance · Methane efflux · Air temperature · Salinity · South India

Introduction

Methane (CH_4) is the second most abundant anthropogenic greenhouse gas (GHG) in the atmosphere (Myhre et al. 2013; Etminan et al. 2016; Allen et al. 2018), contributing to indirect

warming with a radiative forcing of 0.48 Wm^{-2} . In contrast, carbon dioxide (CO_2) contributes about 1.66 Wm^{-2} (IPCC 2013; Myhre et al. 2013). Recent estimates have revealed that the CH_4 level in the atmosphere has increased 2.5 times from 720 ppb during the pre-industrial period (Etheridge et al. 1998). Methane is emitted from various sources, including wetlands, and during anthropogenic activities such as agriculture, power production, and urban waste disposal (Saunois et al. 2017; Turner et al. 2019; Rosentreter et al. 2021). The increase in atmospheric temperature (T_{Air}) due to global warming leads to a rise in CH_4 emissions from these sources (Gedney et al. 2004; Ma et al. 2017; Zhang et al. 2017). Moreover, CH_4 concentrations have increased rapidly from 1775 to 1857 ppb between 2007 and 2018 (Nisbet et al. 2016, 2019; Saunois et al. 2020), and this trend is found to be strong in tropical and subtropical regions (Nisbet et al. 2016). In India, CH_4 observations and flux measurements are sparse; only a handful of such observations are available from a few urban and semi-urban areas (Ganesan et al. 2017; Guha et al. 2018; Tiwari et al. 2020). The available records show that the values are relatively high and display large seasonality, if compared with the observations in Mauna Loa in Hawaii and Seychelles in the Indian Ocean (Sreenivas

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et al. 2016; Metya et al. 2021a, b). Due to limited observational CH_4 flux data and the limited spatial scale of chamber-based measurements (Saunois et al. 2020), it is difficult to estimate a regional-scale CH_4 budget for India. Therefore, there is an urgent need to increase ecosystem-level data on CH_4 fluxes, adopting methods that integrate small-scale variability as the eddy covariance (EC) technique and focusing on ecosystems that store large amounts of organic carbon as wetlands.

Mangrove forests are important ecosystems in tropical and subtropical coasts and store large amounts of carbon in their sediments (Atwood et al. 2017). They are being used to mitigate the effects of climate change in many areas (Siikamäki et al. 2012; Wylie et al. 2016) as the rates of primary productivity are high in these forests ($218 \pm 72 \text{ TgC year}^{-1}$) (Bouillon et al. 2008) and they store vast amounts of carbon in their above- and below-ground biomass (Donato et al. 2011). However, the mangrove sediments produce GHGs, which are released to the atmosphere (Chen et al. 2014).

The anoxic conditions of sediments, their organic matter content (SOM), redox potential (Eh), and the hydrogeophysical environment are key potential drivers of CH_4 emission from mangrove forests. The anoxic conditions that prevail during the wet season cause more CH_4 production through methanogenesis than during the dry season (Jacotot et al. 2018). The decomposition of SOM depends on tidal inundation and rainfall that influence the Eh of the sediment, which in turn alters the rate of GHG emission (Kristensen et al. 2008a). Although mangrove forests act as a sink for CO_2 (Gnanamoorthy et al. 2020; Liu et al. 2020), the sediment and water in the creeks generally act as a source for CO_2 and CH_4 , which are released to the atmosphere (Borges et al. 2003; Jacotot et al. 2018). Methane emission from mangrove soil is highly variable but generally higher than that from creek water. Moreover, enhanced emission has been observed in the locations where roots and pneumatophores are present (Sotomayor et al. 1994; Purvaja et al. 2004; Kristensen et al. 2008b).

A majority of the studies on mangrove CH_4 budget across the globe have focused only on sediment and aquatic fluxes (McNicol et al. 2017). In this regard, the soil-to-atmosphere CH_4 flux estimation showed broad variability, from 6 to $828 \text{ mmol m}^{-2} \text{ day}^{-1}$, as observed by various investigators in India, Brazil, Puerto Rico, China, Taiwan, Tanzania, Thailand, and Australia using the static chamber technique (Lyimo et al. 2002; Chang and Yang 2003; Purvaja et al. 2004; Lekphet et al. 2005; Allen et al. 2007; Wang et al. 2016; Nobrega et al. 2016; He et al. 2019). The recent global estimates of CH_4 emission from mangrove sediments and from creek water are $0.315 \pm 0.123 \text{ Tg CH}_4 \text{ year}^{-1}$ and $0.232 \pm 0.059 \text{ Tg CH}_4 \text{ year}^{-1}$, respectively (Rosentreter et al. 2018). All these studies only showed soil–water–atmosphere CH_4 fluxes attained from the static chamber, measured in

spatially limited areas. Chamber-based methods, if not properly replicated, cannot provide precise estimates of CH_4 budgets due to large temporal and spatial variability of CH_4 fluxes (Wille et al. 2008; Jha et al. 2014). There is also a significant data gap in direct quantification of net CH_4 flux from an ecosystem, and the seasonal-scale variability is not well documented. Therefore, long-term, continuous, direct monitoring of ecosystem-scale CH_4 fluxes can improve our understanding and estimation of regional CH_4 budgets (Liu et al. 2020).

An EC flux tower provides an instantaneous trace gas exchange between the land surface and the atmosphere. This system is widely used in various ecosystems due to the advantage of direct and near continuous flux monitoring under natural, unmanipulated conditions (Baldocchi 2003; Aubinet et al. 2012). However, the CH_4 flux observation is still considered to be limited in comparison to CO_2 flux estimation (Knox et al. 2019). Recently, Liu et al. (2020) studied the CH_4 emission rate in the mangroves of the subtropical region of Hong Kong, China, using the EC technique. Such studies are rare in the Indian context, and to our knowledge, EC-based CH_4 flux estimation has been reported only from the mangrove ecosystem of the Sundarban delta using 2 months of EC data (Jha et al. 2014). To fill this gap, an EC flux tower was set up in the south Indian mangrove wetland of Pichavaram under the MetFlux India Project (Deb Burman et al. 2020; Chakraborty et al. 2020). The main objective of this study is to examine the diurnal and seasonal (dry and wet periods) variations in CH_4 fluxes using the EC method and their relationship with various environmental and physico-chemical parameters that control CH_4 emission in a mangrove environment.

Materials and Methods

Research Site

The research work was conducted in the mangrove forest located in the Cauvery Vellar-Coleroon estuarine (wetland) region of Pichavaram (Vellar and Coleroon) (lat. $11^\circ 25' 36'' \text{ N}$; long. $79^\circ 47' 38'' \text{ E}$) (Fig. 1). The mangrove ecosystem cover pattern shows that 813 ha are covered by the naturally dense mangrove forest, 68 ha by sparse mangrove, 664 ha by marshy vegetation, and 340 ha by mudflats. The mangrove wetland is comprised of 51 small islands colonised by mangrove plants (Selvam et al. 2002). Nearly 12 true mangroves species exist in the study area. There are two distinct distribution zones of mangrove species: the *Rhizophora* spp. zone and the *Avicennia* spp. zone. The *Rhizophora* spp. zone is found in the fringes of the creeks and is narrow in width (6–18 m). The *Avicennia* spp. zone makes up the interior of the forests and ranges in width from 60 to 80 m. The height

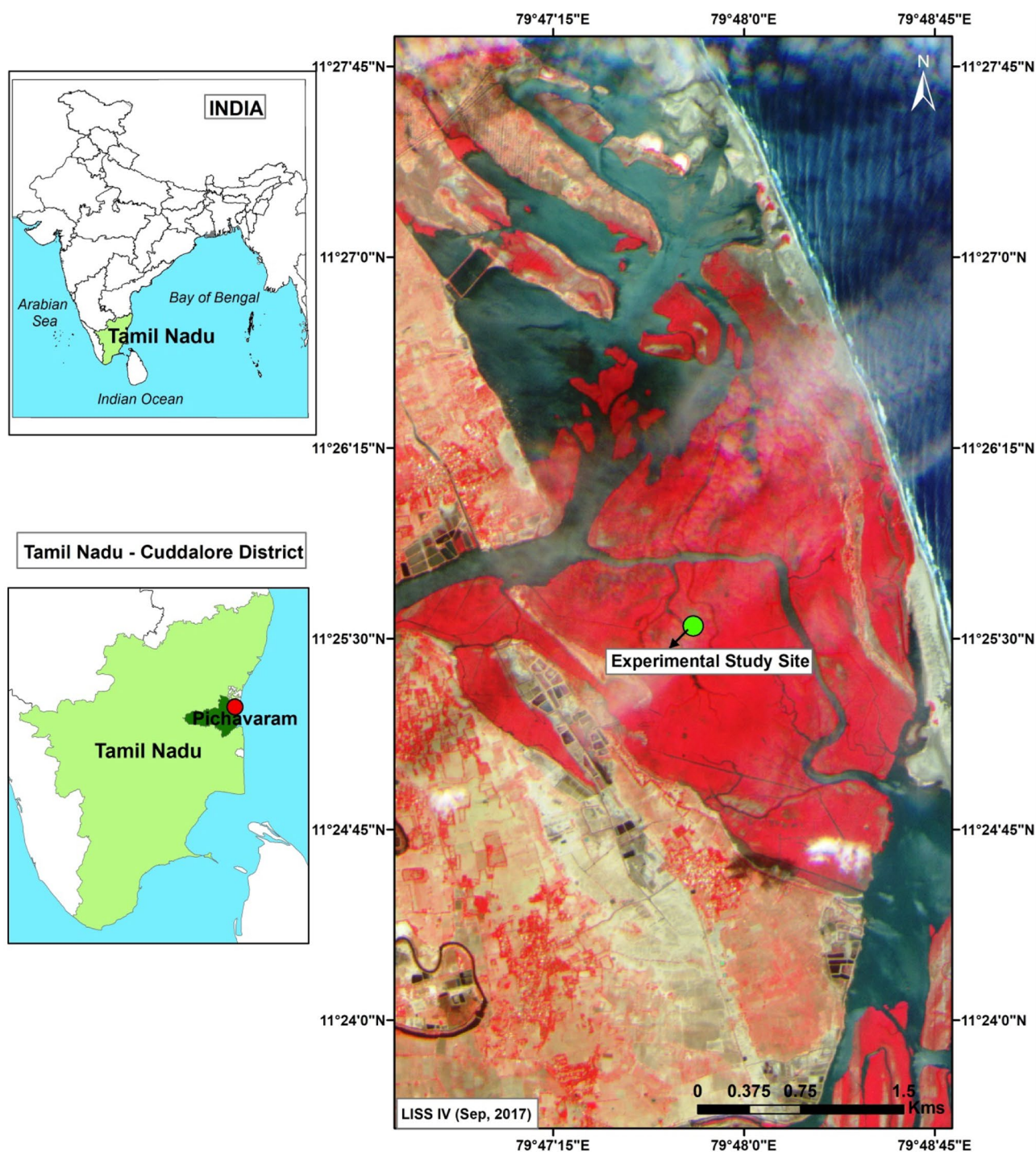


Fig. 1 The study area map indicates the EC flux tower location using a light green circle on the right-side panel. The upper-left portion shows the map of India, while the green-shaded area in lower left

denotes the state of Tamil Nadu. A zoomed-in version of the research site is shown on the right side, where the red colour indicates the Pichavaram mangroves

of the mangrove trees varies between 3 and 7.5 m. The mean leaf area index (LAI) in *Rhizophora* spp. is $4 \text{ m}^2 \text{ m}^{-2}$, and for the *Avicennia* zone, it is $2 \text{ m}^2 \text{ m}^{-2}$. The water column in the creeks is generally shallow, ranging between 0.3 and 3.0 m (Selvam 2003). The Pichavaram mangrove wetland

is hot in the summer months and relatively cooler during the post-monsoon period (Kathiresan 2000). It experiences dry conditions during April–August and wet conditions during September–November (the monsoon season). The peak rainfall occurs during the northeast monsoon season

(October–December) in the southeastern part of peninsular India (IMD 1973; Rajeevan et al. 2012), which is characteristically different from other parts of India. The tidal table predicted for the nearest location of the EC tower is the Cuddalore Port, India. The period of inundation for Pichavaram has been derived using the tides4fishing software package.

Eddy Covariance Tower and Instrumentation

The iron-based, triangle-shaped (10 m height) flux tower was erected in the observation site to monitor the CH₄, CO₂, and H₂O and energy fluxes in 2016. The EC flux tower is encircled by abundant natural mangrove vegetation with prevailing species such as *Rhizophora mucronata*, *R. apiculata*, and *Avicennia marina*. The canopy height ranges from 3 to 4 m. Three solar-power systems ensure uninterrupted power supply to the sensors (one 24 V and two 12 V power output) using batteries (500 AH). The CH₄ flux measurements were carried out at the 10-m level. A three-axis ultrasonic anemometer (Wind Master Pro, Gill Instruments Limited, Hampshire, UK) measured the high-frequency wind velocity and sonic temperature (T_s), and the open path methane analyser (model: LI-7700, LI-COR Biosciences, NE, USA) measured atmospheric CH₄ concentration. The raw data from the anemometer and open path methane analyser were logged by an analyser interface unit (model: LI-7550, LI-COR Biosciences, NE, USA) at the rate of 10 Hz. The supplementary environmental parameters were also monitored above the canopy at 10-s interval, averaged over 30 min, and logged on a CR3000 data logger (Campbell Scientific Inc., Logan, UT, USA). The meteorological parameters – net radiation (using a net radiometer; model: NR01, Hukseflux, Netherlands) and photosynthetic active radiation (PAR) (model: SQ-100 and 300 series, Apogee instruments Inc., UT, USA) – were measured at the 6-m level. Further meteorological parameters, such as air temperature, relative humidity, wind direction, wind speed, and rainfall, were measured at various levels, namely, 2 m, 6 m, and 10 m (using a Vaisala weather transmitter; model: WXT520, Helsinki, Finland). The soil heat flux was monitored at two depths (2.5 cm and 5 cm) (model: HFP01SC-20, Hukseflux,

Netherlands). The LAI was recorded using a portable LAI-2200 plant canopy analyser in the four different directions of each location (LI-COR Biosciences, NE, USA). An enclosed path infrared gas analyser (IRGA) sensor (model: Li-7200, LI-COR Biosciences, NE, USA) was also used to measure the CO₂ concentration and its fluxes. The results of CO₂ fluxes are presented in Gnanamoorthy et al. (2020).

CH₄ Flux Data Analysis

The EC technique is considered to be a reliable method of estimating CH₄ fluxes in the mangrove (Liu et al. 2020) and other ecosystems (Knox et al. 2019).

The following successive equation is used to compute CH₄ fluxes (Baldocchi 2003):

$$fCH_4 = \overline{\rho_a} \cdot \overline{\omega' s'} \quad (1)$$

where fCH_4 is the CH₄ flux (nmol m⁻² s⁻¹); ρ_a is the air density; ω' and s' are vertical wind speed and the CH₄ concentration variations, respectively; and the over bar in the equation denotes the averaging time of 30 min.

The collected raw EC data (from April 2018 to November 2018) were processed using the EddyPro software (6.1.0, LI-COR Biosciences, NE, USA); this gave the values for concentration and fluxes of CH₄ averaged on a 30-min time frame. The raw flux data usually contain different kinds of noises, and hence, they are subjected to a variety of correction schemes to obtain reliable data. The FLUXNET community has developed protocols that employ various correction schemes (Baldocchi et al. 2001; Reichstein et al. 2005), which we have followed here and are described in Table 1.

Further processing is done to eliminate sensor errors and unusual atmospheric situations, such as heavy rain events (Liu et al. 2020). The night-time fluxes are underestimated due to low turbulence (Baldocchi 2003). To address this issue, the friction velocity (u^*) threshold is fixed at 0.13 ms⁻¹, and data beyond this limit are excluded using an average value test technique (Zhu et al. 2006). The received signal strength indicator (RSSI) is a measure of the cleanliness of the mirrors of the CH₄ sensor. Data having RSSI

Table 1 Methods used to calculate the 30-min flux measurements in the EddyPro software

Flux data correction scheme	References
Triple-coordinate rotation to eliminate errors due to sensor tilt and non-homogenous terrain at the site	Baldocchi et al. (2000); Wilczak et al. (2001)
WPL calibration to compensate air-density variations	Webb et al. (1980)
Removal of large spikes due to instrument error and unrealistic values	Sabbatini et al. (2018)
Quality control tests for the flux values	Foken et al. (2004); Göckede et al. (2008)
Flux footprint estimation	Kljun et al. (2004); Kormann and Meixner (2001)

lower than 20% are considered imprecise and hence rejected (Fortuniak et al. 2017).

Gap Filling of CH₄ Fluxes

Estimation of continuous time series of the flux data using EC is difficult due to several reasons, such as low turbulence, extreme rain events, and technical snags, for example, power and sensor malfunction. No standard universal method is currently available to fill the gap for the CH₄ fluxes (Nemitz et al. 2018). To generate continuous time series on the daily and monthly scale of CH₄ fluxes, we used the marginal distribution sampling (MDS) method for gap filling algorithm (Reichstein et al. 2005; Papale et al. 2006), which is considered a reliable method by the FLUXNET community and is incorporated in the REddyProc package (Wutzler et al. 2018).

As this method is used widely for gap filling of CO₂ and H₂O fluxes (Deb Burman et al. 2021), the MDS algorithm can be used for CH₄ fluxes as well (Rinne et al. 2007; Mejjide et al. 2011; Hatala et al. 2012a, b; Alberto et al. 2014; Ge et al. 2018; Tang et al. 2018).

Estuarine Soil and Water Parameters

In addition to the EC data, soil samples were collected bimonthly from three locations in and around the flux tower footprint area, encompassing the natural mangrove stands. Mangrove soil was collected at the following depths using a stainless steel corer: 0–15 cm, 15–30 cm, 30–60 cm, and 60–90 cm. The sampled soil was stored in plastic bags and then transported to the laboratory for further analysis. Soil parameters such as temperature (T_{Soil}), pH, Eh, and salinity (soil sal) were measured in the field using digital pH, Eh, and electrical conductivity metres (Hanna instruments). The soil organic carbon (SOC) was estimated using the Walkley–Black (WB) method (Walkley and Black 1934).

Surface water samples from the creek canal at four different locations around the flux tower were collected once a week using a Niskin water sampler and transferred in clean borosilicate bottles. We used a water quality monitoring system (Hydrolab Quanta Multi-Probe Meter, TX, USA) to measure in situ water temperature (WT), dissolved oxygen (DO), and salinity (W_{sal}). The pH was measured at three-digit precision using a pH metre (EUTECH waterproof, cyber scan pH-610-pH/mV/temperature metre-ECPH-WP61042K) calibrated on the NBS scale. The biological oxygen demand (BOD) was measured using Winkler's method. Chlorophyll “a” (Chl) was analysed using the method outlined by Strickland and Parsons (1972). The total alkalinity (Talk) was determined by the standard method of Gran titration (Gran 1952). We filtered the water sample through a cellulose acetate filter paper (0.45 µm) to measure

the alkalinity. The filtered water was stored in 250-mL borosilicate bottles, and 100 µL of saturated mercuric chloride solution was added to arrest microbial growth (Cai and Wang 1998; Jiang et al. 2008). While collecting the creek water, all sampling bottles were allowed to overflow for at least twice their volume to reduce contact with the air. Afterwards, the sampled bottles were stored at 4 °C for further analysis. For total dissolved inorganic carbon (DIC) and dissolved organic carbon (DOC) measurements, water samples were collected using separate clean high-density (HD) polythene bottles and stored in an icebox. Total organic carbon (TOC) and DOC were measured using an automated TOC analyser (Innoax Lab 0463). Particulate organic carbon (POC) was obtained using GF/F84 filters (0.45 µm), dried at 65 °C, and analysed on elemental analyser (Perkin Elmer 2400).

Statistical Analysis

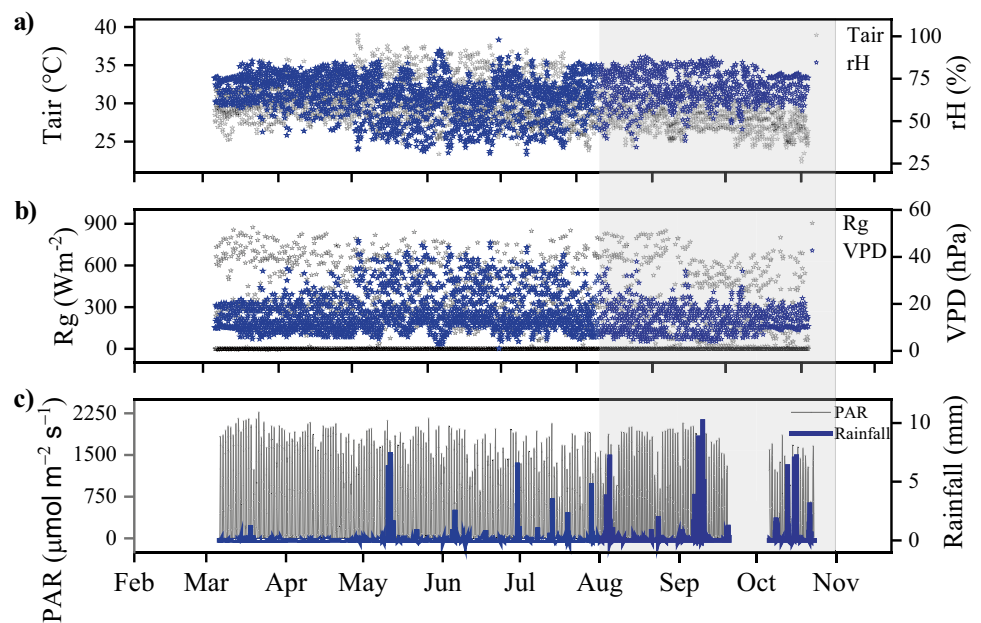
The statistical analyses (linear regression) were carried out using the Origin software (version 2018, Origin Lab Co., Northampton, MA, USA). Visualisation of a correlation matrix graph was generated using a package “Performance Analytics” of the R Team Cooperation (Peterson et al. 2018). Finally, the fitting of the structural equation model (SEM) was accomplished using the Amos 21.0 software (IBM, Armonk, NY, USA). The SEM is a multivariate statistical analysis technique which, in this case, describes how the CH₄ fluxes were influenced by the environmental parameters in the mangrove ecosystem. The goodness of model fit (Hou et al. 2004) was determined to test the robustness of the result. The index includes CMIN/DF (minimum discrepancy divided by degrees of freedom), root mean square error of approximation (RMSEA), Bentler–Bonett normed fit index (NFI), comparative fit index (CFI), goodness-of-fit index (GFI), incremental fit index (IFI), Akaike information criterion (AIC), and expected cross-validation index (ECVI).

Results

Atmospheric and Estuarine Parameters

Meteorological parameters varied monthly over the study period (Fig. 2a–c). The mean monthly T_{Air} during the study period varied from 27.3 (November 2018) to 31 °C (June 2018), while the monthly T_{Soil} ranged from 26.8 (November) to 30.5 °C (June) (Table S1). The relative humidity varied from 30 to 99%. The daily maximum net radiation on the measurement days varied between 412.4 and 906 Wm⁻². The total precipitation was 351 mm, with the highest rainfall (113 mm) observed in November (Fig. 2c). The noon time mean PAR was highest in the hot season (April–July; range: 1482–1911 µmol m⁻² s⁻¹) and lower in the wet season

Fig. 2 Half-hour time series of (a) atmospheric temperature (TAir) and relative humidity (rH); (b) net radiation (Rg) and vapour pressure deficit (VPD); (c) photosynthetic active radiation (PAR) and averaged daily rainfall data from April 2018 to November 2018. The grey shade indicates the wet season and the white area of each panel indicates the dry season



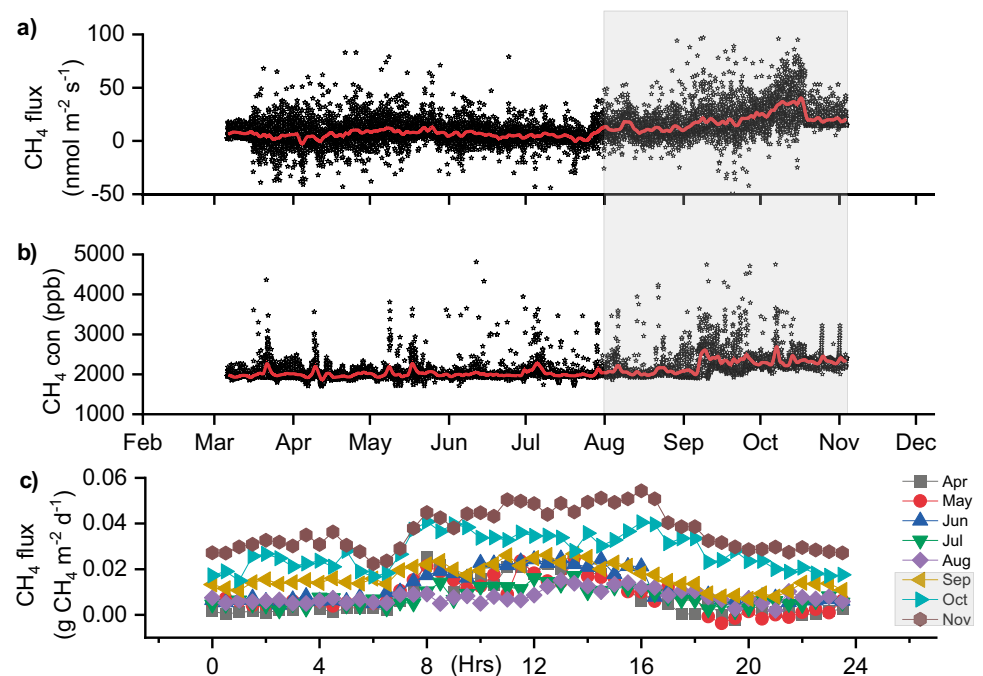
(October–November; range: 1397–1603 $\mu\text{mol m}^{-2} \text{s}^{-1}$). Other measured environmental parameters were temperature, pH, salinity, Talk, DO, BOD, Chl, TOC, DIC, DOC, and POC for water and temperature, pH, salinity, Eh, and SOC for soil (Table S1).

Diurnal and Seasonal Variability of the CH_4 Fluxes

Figure 3a shows the seasonal variation of CH_4 fluxes. Higher values were observed in the wet season. The mean

daily fluxes, regardless of the months and time of day, are mostly positive, indicating the ecosystem is a net CH_4 source (Fig. 3a, c). A typical diurnal pattern of CH_4 flux was observed: maximum post-noon (13:00–16:00 h) and minimum in the early morning (1:00–6:00 h) and evening (19:00–21:00 h) (Fig. 3c). During the dry season (April–August) and wet season (September–November), the daily maximum CH_4 flux was observed in the post-noon. The average daily variability of CH_4 flux was lower in the dry season ($6.4 \pm 4.1 \text{ nmol m}^{-2} \text{s}^{-1}$) and higher during the

Fig. 3 Half-hour continuous (a) methane (CH_4) flux and (b) CH_4 concentration ($\text{CH}_4 \text{ con}$) are plotted on a monthly scale and (c) mean monthly diurnal (24 h) patterns of CH_4 flux. Mean diurnal flux was calculated based on the sum of the average hourly CH_4 flux multiplied by the total days in the month (c). Red-colour streaks in panels (a)–(b) indicate daily means. The grey shade indicates the wet season, and the white area of each panel indicates the dry season (a–b)



wet season ($19.63 \pm 5.3 \text{ nmol m}^{-2} \text{ s}^{-1}$) (Fig. 3c). The highest values recorded in October and November were 1.33 and $1.77 \text{ g CH}_4 \text{ m}^{-2} \text{ month}^{-1}$, respectively. The values observed during April–August were low, between 0.36 and $0.59 \text{ g CH}_4 \text{ m}^{-2} \text{ month}^{-1}$.

The mean daily CH_4 concentration in the study area was 2098 ± 275 ppb (Fig. 3b). The maximum mean daily value of 2346 ppb was recorded in November and the minimum of 1980 ppb during May. The diurnal patterns of CH_4 concentration reached a maximum value in the morning hours (04:00–07:30 h) and minimum post-noon (13:00–16:00 h), and it was opposite to CH_4 flux values. The averaged CH_4 concentration during the dry and wet periods was 2009 ± 183 ppb and 2248 ± 336 ppb, respectively.

Footprint Analysis

We calculated the footprint of the CH_4 fluxes to characterise the flux source area in the mangrove forest, as suggested in Baldocchi (1997). The footprint calculation was demonstrated using a parameterisation scheme outlined by Kljun et al. (2004). The wind rose diagrams (Fig. 4a–c) illustrate the predominant wind direction and footprint, and the CH_4 concentration is calculated to get the location of the EC flux measurements. The estimated footprint or fetch (90%) varied from 118 to 448 m, with a mean distance of 204 m from the EC tower (Fig. 4a). The dominant wind direction at the research site was southwest-west-northwest (Fig. 4b), and the wind speed was between 1.05 and 7.1 m s^{-1} with a mean value of 2.5 m s^{-1} during the entire study period (Fig. 4c).

The CH_4 flux was recorded predominantly from the west, and especially from the southwest, with values of $11\text{--}30 \text{ nmol m}^{-2} \text{ s}^{-1}$ (Fig. 4c). Significantly higher values of CH_4 fluxes ($40 \text{ nmol m}^{-2} \text{ s}^{-1}$) have been observed with westerly/southwesterly airflow due to the presence of dense mangrove vegetation (Fig. 4c). The distribution of mean CH_4 concentration depends on the wind direction, which has been calculated for the whole study period (Fig. 4b). Higher CH_4 concentration from the natural mangrove forest was observed when the wind speed was low ($< 2 \text{ m s}^{-1}$) (Fig. 5). The relationship between CH_4 concentration and wind direction was the same as CH_4 flux values throughout the study period, while the monthly averaged CH_4 concentration values were between 1980 and 2346 ppb (Fig. 4c).

Estuarine Factors Controlling the CH_4 Fluxes

Figure 5a–b show the CH_4 fluxes with respect to tidal inundation as it is one of the most important factors controlling the fluxes (Purvaja and Ramesh 2001). Methane flux varies highly depending on tidal inundation patterns. The study site receives two high tides and low tides every day

(semi-diurnal tides). The CH_4 flux was relatively higher during the low tide than during the high tide condition. During low tide, CH_4 emissions reached a maximum at the beginning of the rising tide and continued until the end of the tide (Fig. 5). The results suggest that high CH_4 flux was observed after high tide patterns during both dry and wet seasons. Figure 6a shows a reasonably good negative correlation between CH_4 flux and water salinity ($R^2 = 0.27$, $p < 0.05$). This tendency was only partly sustained when the data were grouped by salinity class. The averaged CH_4 emission was significantly lower for euryhaline conditions (> 40) than for mesohaline conditions (5–18). Oligohaline conditions (0.5–5) showed considerably higher efflux than both euryhaline and mesohaline conditions. The peak variability overlapped with the highest emission in the salinities between 3 and 5. The relationship between CH_4 flux and air temperature showed increasing CH_4 fluxes with increasing temperature ($R^2 = 0.60$, $p < 0.05$; Fig. 6b).

Both CH_4 flux and CH_4 concentration showed negative correlations with air temperature, soil temperature, WT, wind direction, soil pH, and POC as illustrated in the correlogram (Fig. 7). While CH_4 flux was positively correlated with Eh and chlorophyll, CH_4 concentration showed a positive correlation with CH_4 flux and chlorophyll content in the water (Fig. 7).

Structural Equation Modelling

Based on the features of CH_4 flux from the mangrove forest, a conceptual model of the major factors affecting CH_4 fluxes and CH_4 concentration has been established (Fig. 8). The SEM contains two latent variables (CH_4 flux and CH_4 concentration) and five quantifiable variables (DO, Eh, TAIR, Wsal, and SOC). Typical model fitting index analysis was predicted using conceptual models and assumptions; the primary model was tailored using Amos 21.0 (IBM SPSS). By successive frequent fitting, evaluation, and alteration of the model, the concluding normalised coefficient correction model with better fitting indices was attained (Fig. 8; Table 2). The SEM, absolute fitting index, relative fitting index, and reduced index were used in this study on the basis of earlier endorsements (Fang et al. 2019; Hou et al. 2004). The output of the fitting index investigation for the model is given in Table 2. The fitting index of the model was usually acceptable, meeting all fitting index necessities. The relationship model of CH_4 efflux in mangrove forests attained using the statistical technique was reasonable. Methane flux was significantly affected by Eh and CH_4 concentration ($p < 0.05$) but not by SOC and TAIR ($p > 0.05$), and the respective path coefficients were 1.92 and 0.96 indicating the significant impact of mangrove forests on flux

Fig. 4 (a) Wind rose and fetch of the eddy covariance (EC) system. The wind rose figure displays the footprint (fetch) and direction at the research site. Specifically, the stripes reveal the direction, while the colours indicate the maximum footprint value of 90%. (b) Methane concentration (CH_4 con) and direction calculated for the research site. The stripes display the direction, while the colours show the CH_4 concentration variations. (c) CH_4 flux in relation to wind speed and wind direction during the research period. Green and blue colours indicate the wind speed and direction, respectively

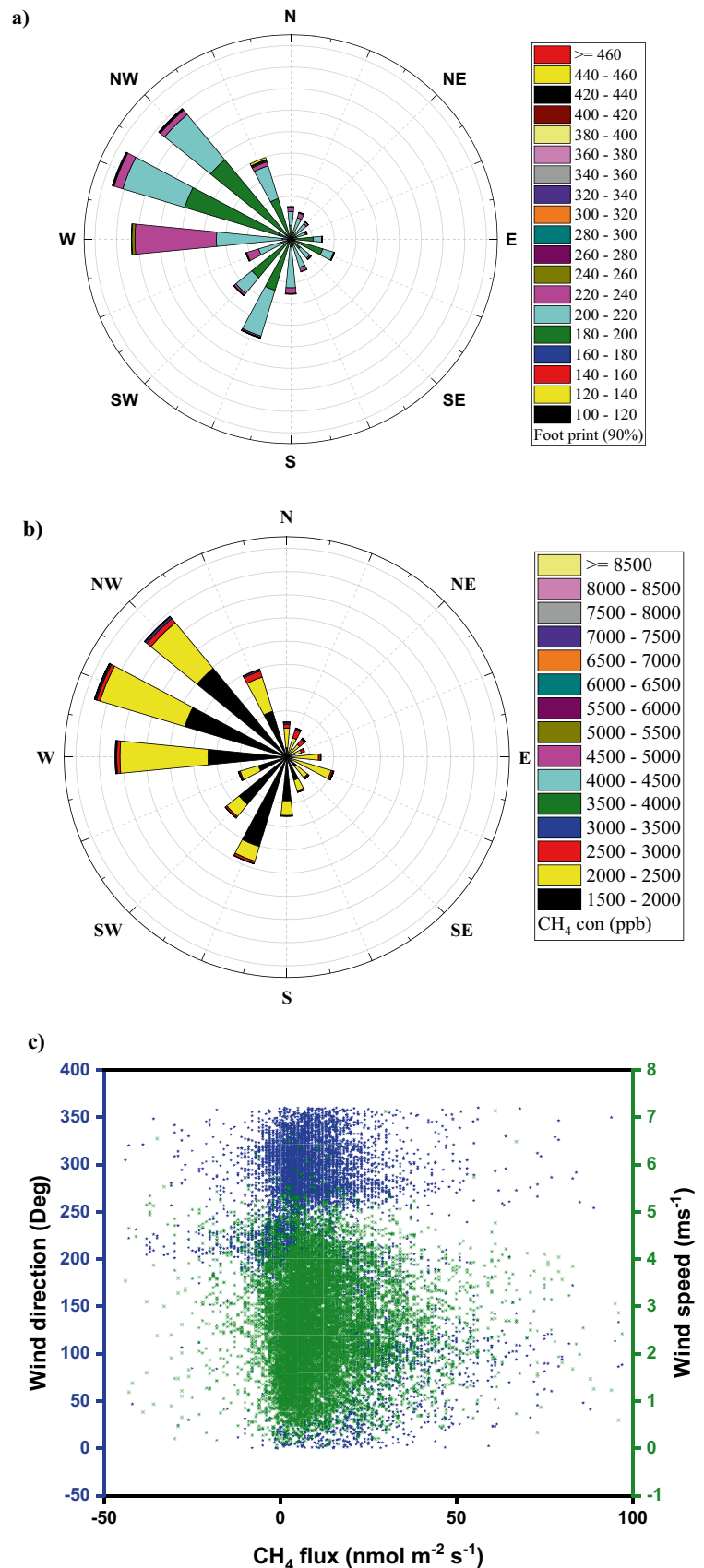
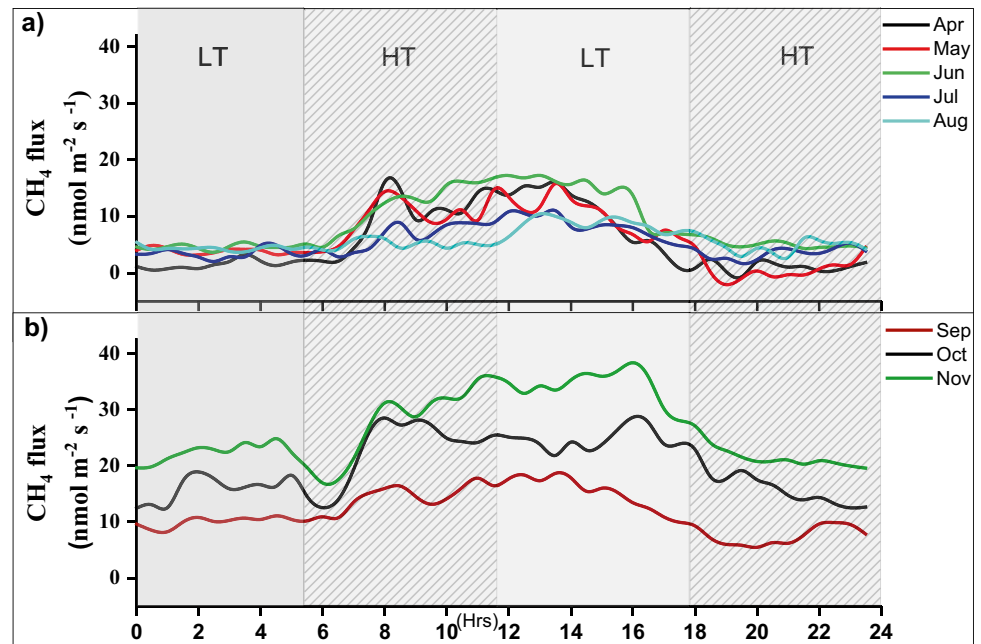


Fig. 5 The effects of high tide (HT) and low tide (LT) on diurnal methane flux during the dry season (a) and the wet season (b) in the Pichavaram study site. The x-axis represents the daily hours (h)

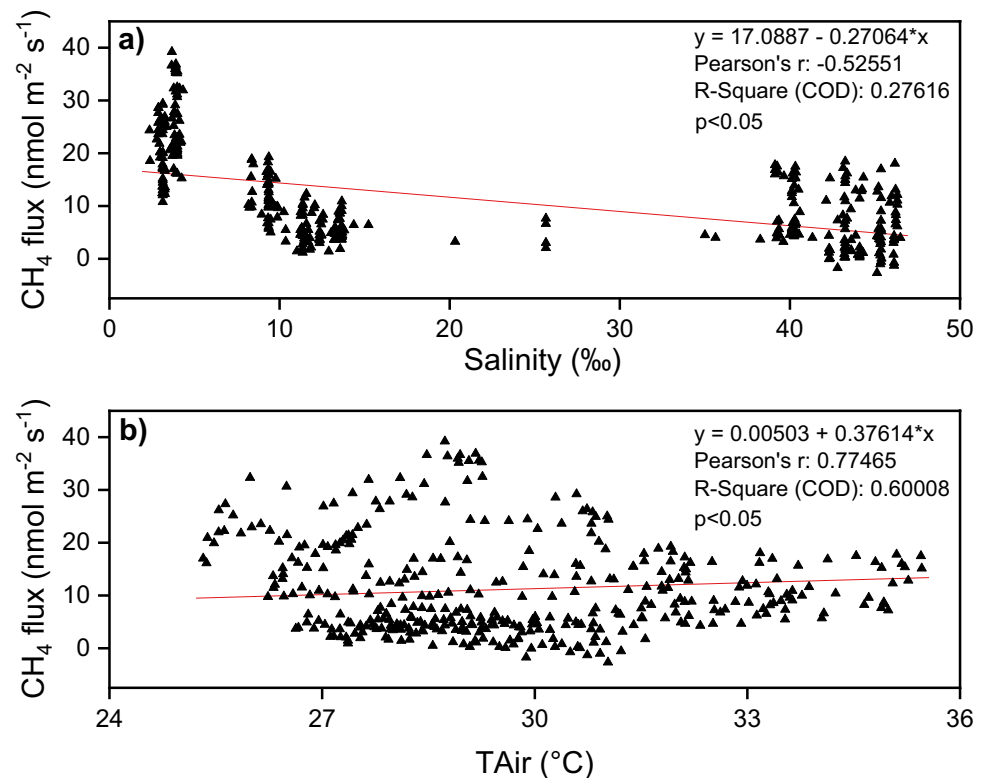


exchange rate (Fig. 8). Furthermore, water salinity was the most significant factor controlling CH_4 concentration (path coefficient -0.68 ; $p < 0.01$), whereas SOC and TAir were non-significant ($p > 0.05$). More significant relationships were found between the parameters for Eh and DO and for water salinity and DO (path coefficients 0.82 and -0.65 , respectively; $p < 0.001$).

Discussion

Biogeochemical processes in mangrove ecosystem are complex and small-scale spatial and temporal variability, which makes the estimation of ecosystem-level CH_4 fluxes complicate (Barr et al. 2010; Jha et al. 2014; Liu et al. 2020). In the past, the chamber-based method was

Fig. 6 Correlation between the salinity (a) and air temperature (b) with the methane flux observed in the study site



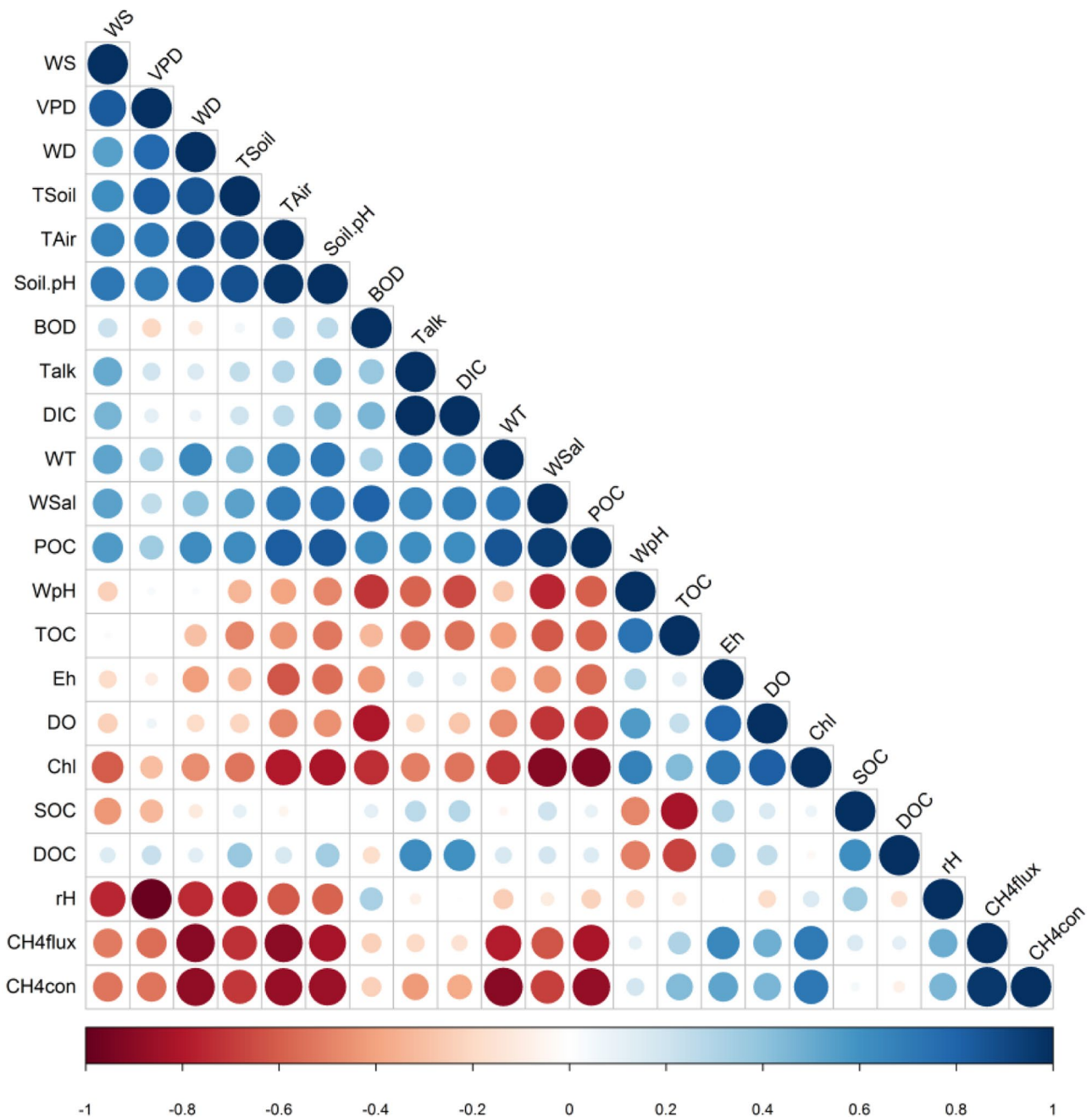


Fig. 7 Correlation matrix of the methane (CH_4) emissions against atmospheric/soil/water index. Positive correlations are displayed in blue and negative correlations in red. The colour intensity and the size of the circle are proportional to the correlation coefficients

widely used to estimate the CH_4 fluxes between soil and atmosphere to measure the fluxes on a plot-base (field) scale or field survey in several ecosystems (Livingston and Hutchinson 1995; Purvaja and Ramesh 2001; Acosta et al. 2019). The EC method is an advanced technique that provides a more precise estimation of fluxes from a large area, especially from natural forest ecosystems and agricultural wetlands (Baldocchi 2014; Petrescu et al. 2015; Morin 2018; Knox et al. 2019).

The published CH_4 emissions, using EC and the closed static chamber technique, from subtropical and tropical mangroves are highly variable (Table 3). The highest daily CH_4 flux measured via EC was observed during the summer months in the Sundarban mangroves in east India (Jha et al. 2014). Methane fluxes from the Sundarbans are likely higher than those measured in the Pichavaram mangroves because seasonal rainfall pattern, tidal inundation range, and temperature in the latter are different from those in the Sundarbans.

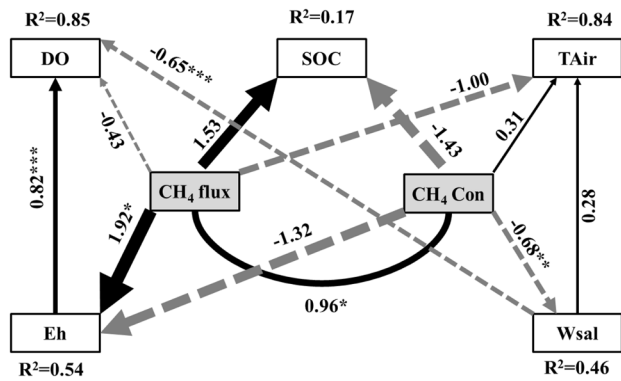


Fig. 8 Standardised coefficients correction model for key driving features of methane (CH_4) efflux from mangrove forests: the structural equation model (SEM) considered plausible pathways through which dissolved oxygen (DO), soil redox potential (Eh), air temperature (TAir), water salinity (Wsal), and soil organic carbon (SOC) influence CH_4 flux and concentration. The arrow width indicates the strength of standardised path coefficients. The solid black line arrows and the grey dashed lines represent positive path coefficients and negative path coefficients, respectively. Numbers on the arrow indicate significant standardised path coefficients (* $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$) proportional to the arrow width. R^2 indicates the variance of endogenous variable explained by the model. $\chi^2 = 4.908$, $df = 9$, probability level = 0.842, RMSEA = 0.000

Moreover, in the same study site, the closed static chamber studies also reported a higher rate of CH_4 emission than that reported by the present study (Purvaja and Ramesh 2001; Purvaja et al. 2004). Most of the chamber-based data showed higher rates of CH_4 emission in the mangrove wetland due to the measurement techniques which allows for a buildup of CH_4 inside the chamber volume, when compared with emissions measured by the EC method (Allen et al. 2007; Konnerup et al. 2014; He et al. 2019). In contrast, lower CH_4 flux ranges were also observed by micrometeorological and chamber methods in the Indian Sundarban mangroves (Mukhopadhyay et al. 2002; Biswas et al. 2007). Most of the previous estimations of CH_4 emission rates were done using closed static chamber techniques, which are relatively less precise due to smaller area and discontinuous estimations in terms of temporal and spatial variability (Pavelka et al. 2007; Jha et al. 2014). Reported CH_4 effluxes in a subtropical mangrove forest in Hong Kong, China, were higher in

the dry as compared to the wet season, which contrasts our findings, but the mean CH_4 flux rate was comparable with our results (Liu et al. 2020). All these findings show that mangrove forests are not a strong source of CH_4 due to the inhibiting influences of sulphate and salinity (Purvaja and Ramesh 2001; Livesley and Andrusiak 2012).

CH_4 emission rates are high, particularly during the wet season (Purvaja et al. 2004; Zheng et al. 2018). Correspondingly, the overall average monthly CH_4 fluxes, presented as part of the present study, generally increased from August onwards, and the maximum change was observed in November, coinciding with the northeast monsoon (Fig. 3a, c). The studies conducted on these factors suggest that the CH_4 fluxes depend on seasons and abiotic factors such as soil type, soil temperature, water salinity, soil pH, Eh, tidal patterns, and soil moisture. Similarly, biotic factors such as vegetation, microbial diversity, and micro- and macro-algal mats on the sediment also control the CH_4 fluxes (Biswas et al. 2007; Allen et al. 2007; Purvaja et al. 2004; Krithika et al. 2008; Dutta et al. 2013; Bai et al. 2014; Jha et al. 2014; Chauhan et al. 2015; Wang et al. 2016). Methanogenesis is associated with anoxic environments which are most likely to occur during the wet season (Großkopf et al. 1998; Schwarz et al. 2007). During the monsoon season, anthropogenic organic matter from agriculture runoff and aquaculture farm organic waste discharging into the mangroves are additional sources of substrate for the methanogenic bacteria, which are liable for CH_4 emissions (Krupadam et al. 2007). Hence, to reduce CH_4 emissions, the anthropogenic input, particularly aquaculture waste discharged into the mangrove wetland from adjoining shrimp farms, should be diminished (Purvaja and Ramesh 2001). In the present study, CH_4 emissions were at their peak during the wet season and then dropped during the dry season. The months of August and September, representing a transition period from the dry to the wet season, showed a sharp increase in soil respiration, resulting in increased CH_4 fluxes (Gnanamoorthy et al. 2019). Intense emissions occurred in the wet season resulting in a higher variability due to the inundation of mangrove soil during the monsoon and spring tides. The influence of tides and freshwater significantly contributes to the CH_4 emission in the mangroves, but limited studies are available in this regard (Barnes et al. 2006; Purvaja et al. 2004; Kristensen et al. 2008b; Chauhan

Table 2 Structural equation model coefficients fitting indices

Model fitting index		Evaluation standard	Output results
Absolute fitting indices	CMIN/DF	< 3	0.545
Relative fit indices	NFI	> 0.9	0.920
	CFI	> 0.9	1.000
Compact indices	IFI	> 0.9	1.079
	AIC	The smaller, the better	56.908
	ECVI	The smaller, the better	8.130

Table 3 Reported methane fluxes in the mangrove wetlands of the tropical and subtropical regions (daily and hourly fluxes are presented for static chamber and eddy covariance measurements, respectively)

Measurement techniques	Mangrove sites	Range/mean values (g CH ₄ m ⁻² day ⁻¹)	Reference
Static chamber	Pichavaram, India	47.23–324.48	Purvaja and Ramesh (2001)
Micrometeorological	Sundarban, India	–16–32	Mukhopadhyay et al. (2002)
Static chamber	Pichavaram, India	7.4–63.7	Purvaja et al. (2004)
Static chamber	Sundarban, India	0.032–2.15	Biswas et al. (2007)
Static chamber	Pichavaram, India	15.04–23.83	Senthilkumar (2008)
Eddy covariance	Sundarban, India	150.22	Jha et al. (2014)
Eddy covariance	Mai Po Nature Reserve mangrove, Hong Kong SAR, China	40	Liu et al. (2020)
Eddy covariance	Pichavaram, India	8–37	Present study
	Mangrove sites	Short-term values (mg CH ₄ m ⁻² h ⁻¹)	Reference
Static chamber	Chelmer, Southeast Queensland, Australia	0.003–17.37	Allen et al. (2007)
Static chamber	Ciénaga Grande de Santa Marta, Colombia	0–31.57	Konnerup et al. (2014)
Static chamber	Dongzhaigang National Nature Reserve, China	0.84–5.49	He et al. (2019)
Eddy covariance	Pichavaram, India	0.326–1.538	Present study

et al. 2015). Our study reported 6–14% higher in concentration of CH₄ emission than the values observed by Metya et al. (2021a) from Sinthagad a hilly area in western India. Further, it varies with a study from Sundarban mangroves wherein emission was lower by 25% (Jha et al. 2014).

In mangrove wetlands, abiotic and biotic factors are recognised as controlling aspects of CH₄ emissions (Jha et al. 2014; Liu et al. 2020). Though both abiotic and biotic factors influence CH₄ emissions, seasonal factors are the main influencers of CH₄ efflux. If CH₄ production is constant with the season, studies have to find a way to predict for different pathways such as ebullition from emission from leaves, sediments, and import and export by the tidal water. During the study period, between July and August, the mangrove area was swamped with freshwater from the Coleroon River up to a depth of 90–120 cm. This suggests that when pneumatophores and soil are inundated, the gas transfer between the aerenchyma and the atmosphere is impeded and vice versa. The CH₄ efflux values ranged from 0.36 to 0.38 g CH₄ m⁻² month⁻¹, which is lower than the June and September efflux estimates. Recently, a multiyear study of EC-based ecosystem CH₄ fluxes was examined for temporal variations and control of the biophysical drivers in estuarine mangroves of Hong Kong, China. The study found that the daily mangrove CH₄ flux peaked during the summer with the combination of higher temperature and lower salinity, whereas it was minor in the winter months (Liu et al. 2020).

Furthermore, the water level plays a significant role in determining soil temperature (Jha et al. 2014; Wang et al. 2016) and water salinity, and it was well correlated with the SEM (Fig. 8). Creek water parameters, particularly the increase in TOC concentration and sudden drop in water

salinity from 40.5 in June to 11.5 in July, were also visualised and correlated in the present study (Figs. 7–8). Several studies revealed that the CH₄ fluxes were influenced by salinity (Purvaja and Ramesh 2001; Krithika et al. 2008; Dutta et al. 2013; Chauhan et al. 2015).

Salinity is one of the unique factors regulating the CH₄ flux in the mangroves wetlands; moreover, under hypersaline conditions, leaves have an adaptation strategy to reduce the transpiration process (Wu et al. 2012). Therefore, the mangrove plants can reduce transpiration, which significantly inhibits the CH₄ fluxes in the mangrove ecosystem. A good relationship was observed between creek water salinity and CH₄ flux in the present study. The hypersalinity reduces the CH₄ flux during the dry period. However, physiological controls and transport mechanisms are quite unclear for CH₄ fluxes in mangrove trees (Purvaja et al. 2004; He et al. 2019). The impact of tidal inundation on CH₄ efflux has a strong seasonal variation that can largely be attributed to the dry and wet seasons. The results of Jha et al. (2014) clarified that the emitted CH₄ flux during high tides was about 80% more than that during low tides. In contrast, the present study showed diurnal variability (Fig. 5).

Conclusions

This study reports CH₄ fluxes estimated using an EC technique in a South Indian tropical mangrove ecosystem. We have characterised the diurnal and seasonal patterns. Ecosystem-scale observations of the Pichavaram mangroves showed them to be a minor source for CH₄ during the wet season when compared with the dry season. The Pichavaram

mangroves are also characterised by a lower emission rate than that seen in the tropical mangroves of Sundarbans, India, as well as in a subtropical forest near Hong Kong, China. Furthermore, statistical results indicate that air temperature, tidal inundation patterns, and water salinity were important for describing the variability of CH_4 flux in the site. Overall, the results suggest that the Pichavaram mangrove wetland acts as a minor source for CH_4 . However, it may become a larger source as anthropogenic inputs of organic matter are increased and sea levels rise in the future. The study also has significance in quantifying the carbon sequestration rate of the wetland by eliminating the carbon lost through CH_4 emission. Hence, it is very important to have continuous, long-term data to understand the variations in CH_4 emission mechanisms under different environmental conditions.

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Declarations

Conflict of Interest The authors declare no competing interests.

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