Low CO₂ evasion rate from the mangrove-surrounding waters of the Sundarbans

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Abstract Globally, water bodies adjacent to mangroves are considered significant sources of atmospheric CO₂. We directly measured the partial pressure of CO₂ in water \([pCO_2(\text{water})]\) and related biogeochemical parameters with high temporal resolution, covering both diel and tidal cycles, in the mangrove-surrounding waters around the northern Bay of Bengal during the post-monsoon season. Mean \(pCO_2(\text{water})\) was marginally oversaturated in two creeks \((470 \pm 162 \, \mu\text{atm}, \text{mean} \pm \text{SD})\) and undersaturated in the adjoining estuarine stations \((387 \pm 58 \, \mu\text{atm})\) compared to atmospheric \(pCO_2\), and was considerably lower than the global average. We further estimated the \(pCO_2(\text{water})\) and buffering capacity of all possible sources of the mangrove-surrounding waters and concluded that their character as a CO₂ sink or weak source is due to the predominance of marine water from the Bay of Bengal with low \(pCO_2\) and high buffering capacity. Marine water with high buffering capacity suppresses the effect of \(pCO_2\) increase within the mangrove system and lowers the CO₂ evasion even in creek stations. The \(\delta^{13}C\) of dissolved inorganic carbon (DIC) in the mangrove-surrounding waters indicated that the DIC sources were a mixture of mangrove plants, pore-water, and groundwater, in addition to marine water. Finally, we showed that the CO₂ evasion rate from the estuaries of the Sundarbans is much lower than the recently estimated world average. Our results demonstrate that mangrove areas having such low emissions should be considered when up-scaling the global mangrove carbon budget from regional observations.

Keywords Air–water CO₂ flux · Mangrove-surrounding waters · Estuary · \(pCO_2(\text{water})\) · Low CO₂ emission

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Introduction

The carbon stocks within several coastal ecosystems (e.g. mangroves, tidal marshes and seagrass beds), collectively referred to as “blue carbon”, have drawn attention in the context of global climate change (Donato et al. 2011; McLeod et al. 2011; Pendleton et al. 2012), and initiatives to characterize the functioning and long-term future of these blue-carbon ecosystems have also begun (Macreadie et al. 2019). These ecosystems are known to be carbon sinks; however, mangroves deserve special mention owing to their large soil organic carbon pool and for being a center for active deposition of both autochthonous and allochthonous organic matter (Breithaupt et al. 2012; Sanders et al. 2016a, 2016b).

Mangroves, despite covering only 0.1% of the Earth’s total land area (Jennerjahn and Ittekkot 2002), are one of the most productive ecosystems in the world, with high carbon-sequestration potential. The carbon (C) stock in mangroves per unit area (956 Mg-C ha⁻¹) is much higher than that in other carbon-rich ecosystems such as salt marshes (593 Mg-C ha⁻¹), seagrass beds (142 Mg-C ha⁻¹), peatland (408 Mg-C ha⁻¹), or even rain forests (241 Mg-C ha⁻¹) (Alongi 2014; Donato et al. 2011; Twilley et al. 1992).

Although mangrove ecosystems as a whole are net sinks for atmospheric CO₂, the waters adjacent to mangroves (as well as the associated sediments) emit substantial amounts of CO₂ because they have high organic carbon loading, which is mainly attributed to mangrove biomass, terrestrial detritus, the microphytobenthos, and phytoplankton (Borges et al. 2003; Kristensen and Alongi 2006; Krumins et al. 2013). The diagenetic processes change the balance of exported DIC and total alkalinity (TAlk) from mangroves, which promotes carbonate buffering in the water bodies adjacent to mangroves (Sippo et al. 2016; Joesoef et al. 2017; Maher et al. 2018).

The Sundarbans is the world’s largest mangrove forest and has a wide variety of mangrove flora and associated fauna. The Sundarbans mangroves encompass a complex network of channels, creeks and large estuaries. In the last decade, the air–water CO₂ flux has been well studied in the Indian section of the Sundarbans (hereafter, the Indian Sundarbans) in terms of spatial variability (Akhand et al. 2013b, 2016; Biswas et al. 2004). In this regard, Reiman and Xu (2019) reported significant underestimation of pCO₂(water) and the air–water CO₂ flux due to the use of a single daily pCO₂(water) value, compared to the diurnal dataset. Rosentreter et al. (2018) emphasized the importance of the temporal resolution of sampling, which can lead to considerable uncertainty. They argued that data acquisition at hourly or greater intervals often misses the tidal maxima and minima, and deducing the mean CO₂ flux from such data might lead to under- or overestimation of fluxes.

The mineralization of dissolved and particulate organic carbon (DOC and POC, respectively) leads to additional DIC in water bodies adjacent to mangroves (Gattuso et al. 1998; Maher et al. 2013, 2015). The mineralization of organic carbon in mangrove sediments is facilitated through several pathways such as sulfate reduction, iron reduction, and aerobic respiration (Borges et al. 2003; Kristensen and Alongi 2006; Krumins et al. 2013). The diagenetic processes change the balance of exported DIC and total alkalinity (TAlk) from mangroves, which promotes carbonate buffering in the water bodies adjacent to mangroves (Sippo et al. 2016; Joesoef et al. 2017; Maher et al. 2018).

Although the mangrove-surrounding waters (or “mangrove water” or other similar terms) usually act as a source of CO₂, there are still uncertainties with respect to the global budget of these emissions. Recent measurements in mangrove-surrounding waters showed negative fluxes in some regions and during certain times of the year (Akhand et al. 2013b, 2016; Biswas et al. 2004). In this regard, Reiman and Xu (2019) reported significant underestimation of pCO₂(water) and the air–water CO₂ flux due to the use of a single daily pCO₂(water) value, compared to the diurnal dataset. Rosentreter et al. (2018) emphasized the importance of the temporal resolution of sampling, which can lead to considerable uncertainty. They argued that data acquisition at hourly or greater intervals often misses the tidal maxima and minima, and deducing the mean CO₂ flux from such data might lead to under- or overestimation of fluxes.

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The hydrology of the Indian Sundarbans is characterized mainly by marine water from the Bay of Bengal (BoB) and to some extent by monsoonal runoff (Cole and Vaidyaraman 1966; Mitra et al. 2009; Sarkar et al. 2004). At present, all rivers in the Indian Sundarbans, except for the Hooghly and its distributary the Muriganga, have lost their direct connections with the main flow of the River Ganga because of siltation in their upper reaches. However, these disconnected rivers receive reduced amounts of riverine freshwater from the Hooghly River through a large number of waterways such as the Hatania-Doania Canal (Ray et al. 2018a).

A unique quality of the BoB is that, despite being part of the open ocean, it has low salinity (Rao and Sivakumar 2003; Prasad 2004; Pant et al. 2015). The main reason for this low salinity is that several large perennial rivers (such as the Ganges, Brahmaputra, Meghna, Mahanadi, and others) discharge into the BoB (Sandeep et al. 2018). The BoB has low $p$CO$_2$ and is considered to be a sink for CO$_2$ (Akhand et al. 2013a; Goyet et al. 1999; Kumar et al. 1996). Hence, the tidally driven estuaries of the Indian Sundarbans are affected more by the low-$p$CO$_2$ water (nearly equal to or below atmospheric equilibrium) of the BoB than by high-$p$CO$_2$ riverine freshwater.

We hypothesized that despite being mangrove-surrounding waters, the CO$_2$ sink or weak source character of the Indian Sundarbans is mainly caused by the predominance of water with low-$p$CO$_2$ and high-buffering-capacity from the BoB, with higher TAlk than DIC (Goyet et al. 1999; Sabine et al. 2002). To examine this hypothesis, we obtained high-temporal-resolution data for $p$CO$_2$(water) and other related biogeochemical parameters. In addition, we measured TAlk and DIC along with stable isotopic signatures of DIC to quantify and identify their sources in the surface water. Finally, we compared the estimated fluxes, considering the annual evasion rate over the entire estuary, with global average data. We used the Matla Estuary for comparison, as this estuary flows through the central part of the Indian Sundarbans as well as for a long distance covering almost the entire north–south extent. Moreover, the estuaries of the Sundarbans have been exhaustively studied in the recent past from the perspective of air–water CO$_2$ flux (Biswas et al. 2004; Akhand et al. 2013b, 2016; Dutta et al. 2019) as discussed previously. Our objectives were to reduce the uncertainty of flux estimation by direct and continuous measurement of $p$CO$_2$(water) in order to confirm the previously observed low $p$CO$_2$(water) of earlier studies and identify plausible reasons behind such low $p$CO$_2$(water).

Materials and methods

Study area

The Sundarbans is situated in the lower stretch of the Ganges–Brahmaputra-Meghna (GBM) Delta, extending into both India (40%) and Bangladesh (60%) and facing the BoB to the south. The present study was carried out in the Indian Sundarbans, which comprises an area of 10,200 km$^2$ out of which 4200 km$^2$ is demarcated as reserve forest (Ray et al. 2015). Sampling was conducted at six stations in a small north–south section of the estuary approximately 9 km long (width varying between 0.70 and 0.85 km) to the west of Dhanchi Island, and in two tributary creeks (≈ 20 m wide) on the adjoining island (Fig. 1). For details about the study area see Supplementary Material S1. Dhanchi Island covers an area of about 33 km$^2$, and its southern tip ends at the BoB. To the east of the island flows the Thankuran Estuary (7 km wide).

Sample collection

We collected samples at a single station in each of two creeks (hereafter referred to as stations C1 and C2) on the western side of Dhanchi Island, and at six subtidal locations in the adjoining estuary (hereafter referred to as E1, E2, E3, E4, E5 and E6) (Fig. 1). The estuarine stations covered the north-to-south stretch of Dhanchi Island. The abbreviations “C” and “E” stand for “creek” and “estuary”, respectively. Creek stations are more influenced by dense mangrove vegetation than estuarine stations.

Surface $p$CO$_2$(water), salinity, water temperature, dissolved oxygen, and chlorophyll-$a$ were monitored with sensors (for details see section Analytical Protocol). Data for a 24-h diurnal cycle for all of these parameters were acquired at 1-min intervals at each of the eight stations between 27 January and 6 February 2018. Surface water samples were collected at two peak low and high tides; i.e. four times from each station during a complete tidal and diurnal cycle for
the analyses of the parameters TAlk, DIC, nutrients (PO₄³⁻ and SiO₃²⁻), and δ¹³CDIC. In addition, surface water samples were collected from a station named Diamond Harbor (salinity 0.39, measured with the same salinity sensor used for diurnal sampling described later in section Analytical Protocol), which served as the riverine freshwater end-member. The samples were preserved as described in the next paragraph and sent to the laboratory for analysis.

Samples for TAlk and DIC were collected into 250-mL Duran bottles (SCHOTT AG, Mainz, Germany), filtered through glass-fiber filters (GF/F; Whatman, Maidstone, Kent, UK) and poisoned with mercuric chloride (200 μL saturated aqueous solution per bottle) to prevent changes in TAlk and DIC due to biological activity. Samples for nutrients analyses were collected for input parameters of CO₂SYS software (Lewis and Wallace 1998). The samples were filtered through 0.2-μm polytetrafluoroethylene filters (DISMIC–25HP; Advantec, Durham, NC, USA) into acid-washed 50-mL polyethylene bottles and frozen at −20 °C until analysis.

Analytical protocol

TAlk and DIC concentrations were determined using a batch-sample analyzer (ATT-05; Kimoto Electric Co., Ltd., Osaka, Japan) implementing the Gran Plot method (Dickson et al. 2007). The accuracy of TAlk and DIC was estimated to be ±1 μmol kg⁻¹ water and ±2 μmol kg⁻¹ water, respectively by triplicate measurements of the certified reference material (CRM) for TAlk and DIC (Kanso Company Ltd., Osaka, Japan).

The pCO₂(water) was measured with a CO₂ analyzer (non-dispersive infrared sensor) through an equilibrator system (CO₂-09, Kimoto Electric Co., Ltd.) (Kayanne et al. 1995; Tokoro et al. 2014) using a gas-permeable membrane (Saito et al. 1995). The instrument was calibrated every day at the beginning.
of the measurements using certified reference gases [pure N₂ gas (0 ppm) and span gas (600 ppm CO₂ gas with a N₂ base; Chemtron Science Laboratories, Mumbai, India)]. The estimated accuracy and precision of the pCO₂ analyzer were ± 5 μatm and ± 2 μatm, respectively, which was in parity with the previous works (Saito et al. 1995, 1999; Kayanne et al. 2005) where the same pCO₂ measurement system was used.

Stable isotope signatures (δ¹³C(DIC)) were measured with an isotope-ratio mass spectrometer (Delta Plus Advantage; Thermo Electron, Bremen, Germany) coupled with an elemental analyzer (Flash EA 1112; Thermo Electron) following the method of Miyajima et al. (1995). Stable isotope ratios are expressed in δ notation as the deviation from standards in parts per thousand (%) according to the following equation:

\[
δ^{13}C_{DIC} = \left[ \frac{R_{sample}}{R_{standard}} - 1 \right] \times 10^3
\]  

where \( R \) is ¹³C/¹²C. Vienna Pee Dee Belemnite (VPDB) was used as the isotope standard for carbon. The analytical precision of the Delta Plus Advantage mass-spectrometer system, based on the standard deviation of the internal reference replicates, was < 0.2‰ for δ¹³C(DIC).

Salinity and temperature were measured with a CT-sensor (INFINITY-CT; JFE Advantech CO., Ltd, Nishinomiya, Japan). Dissolved oxygen (DO) concentration was measured at hourly intervals with a portable DO meter (FiveGo Series; Mettler Toledo, Giessen, Germany). Chlorophyll-a concentration (Chl-a) and turbidity were measured in situ with a fluorometer (COMPACT-CLW, JFE Advantech CO., Ltd). The Chl-a fluorescence sensor was adjusted in a uranine solution by the manufacturer to yield a constant calibration factor. We have corrected the Chl-a data using the regression line between sensor data and 12 spectrophotometrically measured Chl-a data collected during sampling. Chl-a was measured following standard spectrophotometric procedures using Shimadzu UV–Visible 1600 double-beam spectrophotometer (Parsons et al. 1992). Nutrients concentrations (PO₄³⁻ and SiO₃²⁻) in sample filtrates were measured using an Auto Analyzer (QuAAtro; BL TEC K. K., Osaka, Japan; for nutrient data, see Table S1, Supplementary Material).

Estimation of air–water CO₂ flux

The air–water CO₂ flux (\( F_{CO_2} \), μmol CO₂ m⁻² h⁻¹) was determined by the following equation:

\[
F_{CO_2} = k \times K_0 \times \Delta pCO_2
\]

where \( k \) is the gas transfer velocity (cm h⁻¹), \( K_0 \) denotes the solubility coefficient of CO₂ (mol m⁻³ atm⁻¹), and \( \Delta pCO_2 \) denotes the difference in fugacity (≈ partial pressure) of CO₂ between water and air \([pCO_2(\text{water}) - pCO_2(\text{air})]\). \( K_0 \) is computed based on the equation given by Weiss (1974). The \( pCO_2(\text{air}) \) was considered to be 408 μatm, i.e. the global mean \( pCO_2(\text{air}) \) observed during the study period (https://www.co2.earth/historical-co2-datasets). The mole fraction of CO₂ was converted to partial pressure of CO₂ in air \([pCO_2(\text{air})]\) by using the virial equation of state (Weiss 1974). A positive \( F_{CO_2} \) value indicates CO₂ efflux from the water to the atmosphere and vice versa. The parameter \( k \) was calculated by using three different empirical gas transfer velocity models based on wind speed: Liss and Merlivat (1986) (LM86), Raymond et al. (2000) (R00) and Ho et al. (2011) (H11). These formulae were selected for the present study because the estuarine channels in the Indian Sundarbans are very wide and there is no hindrance to the free-flowing wind, which makes the wind velocity the most important component regulating the CO₂ fluxes. The equations for the gas transfer velocity calculations are given below:

\[
k = 0.17 \times U_{10}^{0.5} \quad \text{for } U_{10} \leq 3.6 \text{ m } s^{-1} \quad \text{(LM86)}
\]

\[
k = (2.85 \times U_{10} - 9.65) \times \left( \frac{660}{S_c} \right)^{0.5} \quad \text{for } 3.6 < U_{10} < 13 \text{ m } s^{-1} \quad \text{(LM86)}
\]

\[
k = 2.78 \times U_{10}^{0.46} \times \left( \frac{660}{S_c} \right)^{0.5} \quad \text{(R00)}
\]

\[
k = (a + 0.266 \times U_{10}^{2}) \times \left( \frac{660}{S_c} \right)^{0.5} \quad \text{(H11)}
\]
where $U_{10}$ is the wind speed at 10-m height and $a$ is a constant accounting for gas transfer from bottom-shear-driven turbulence (assumed to be zero in the present study area, in accordance with Ho et al. [2011] and their study in the wide Hudson Bay, USA). $Sc$ is the Schmidt number of CO$_2$ as given by Jähne et al. (1987). Only Eq. (3a) was used to compute the gas transfer velocity according to $LM86$, because the mean wind speed was always $< 3.6$ m s$^{-1}$ during the present study. Wind speed data were acquired by using a handheld anemometer (AM 4201; Lutron Inc., Singapore) and corrected for the 10-m measurement height (Kondo 2000).

Data analysis

The Hooghly River is the main “artery” (Ray et al. 2018a) and only possible source of riverine freshwater to the Indian Sundarbans. For this reason, the near-zero salinity region of the Hooghly River has been widely used in previous works (Dutta et al. 2019; Ray and Shahraki 2016; Ray et al. 2015, 2018a) as the freshwater end-member for the Indian Sundarbans. In accordance with these previous studies, we defined the observed salinity at the Diamond Harbor station (salinity approx. 0) as a proxy for the riverine freshwater end-member. Figure 1 includes a schematic showing the pathway of riverine freshwater input to the study area through the Hatania-Doania Canal, which connects the Indian Sundarbans with the Hooghly River (Ray et al. 2018a).

To characterize the pathway of mineralization of organic matter in this study, we normalized both TAlk and DIC with respect to salinity. We analyzed the stoichiometric relationship (the slope) between salinity-normalized TAlk ($nTAlk$) and salinity-normalized DIC ($nDIC$). DIC was normalized according to the following equation (Friis et al. 2003):

$$nDIC = \left[ \frac{DIC_{\text{meas}} - DIC_{S=0}}{S_{\text{meas}}} \right] \times S_{\text{mean}}$$

(6)

where $DIC_{\text{meas}}$ is the measured DIC, $DIC_{S=0}$ is the DIC of the riverine freshwater end-member (i.e. where salinity = 0), $S_{\text{meas}}$ is the measured salinity, and $S_{\text{mean}}$ is the mean salinity, which is used for normalization (25.0 for this study). TAlk was also normalized using the same equation, replacing $DIC_{\text{meas}}$ and $DIC_{S=0}$ with $TAlk_{\text{meas}}$ and $TAlk_{S=0}$, respectively.

Excess DIC for the creek and estuarine stations was calculated from the difference between the “in situ DIC” and the expected DIC in water when $pCO_2$ (water) is equal to $pCO_2$ (air) (Abril et al. 2000). The latter was determined by using CO$_2$SYS software with the in situ salinity, temperature, measured alkalinity and an atmospheric $pCO_2$ of 408 µatm as input parameters.

$pH$ was estimated from the measured TAlk and DIC values ($n = 31$) using CO$_2$SYS software (version 2.5) (Lewis and Wallace 1998). The uncertainty in $pH$ estimation was 0.012 (estimated from the accuracy of the input parameters, TAlk and DIC, stated in section Analytical Protocol, by using the error computation tool of the CO$_2$SYS software).

We estimated the $pCO_2$ (water) and Revelle factor of the sampled waters and all possible sources of the estuarine water—namely pore-water, groundwater, the riverine freshwater end-member, and offshore water of the northern BoB—by using CO$_2$SYS software (Lewis and Wallace 1998). For the riverine freshwater end-member, we used the measured TAlk and DIC of the present study and data from previous studies (Akhand et al. 2016; Dutta et al. 2019) to calculate the Revelle factor. For groundwater, the $pCO_2$ (water) and Revelle factor were calculated by using measured TAlk and DIC (Akhand et al., unpublished data; see details of the sample collection in Table S2, Supplementary Material). We used the pore-water pH data of Mandal et al. (2009) and pore-water DIC data of Dutta et al. (2019) (from samples collected from various sites in the Indian Sundarbans, depth reported as 30 cm below the water table) as input parameters in CO$_2$SYS to compute the $pCO_2$ (water) and Revelle factor of pore-water. For the northern BoB, we used the measured pH and TAlk data of Akhand et al. (2012) and Sarma et al. (2012) [we used only the data from the station nearest to the Indian Sundarbans from Sarma et al. (2012)]. For waters farther offshore in the BoB (almost to latitude 10°N) we used the TAlk and DIC data of Goyet et al. (1999) to compute the Revelle factor. Unlike for the other possible sources, we could estimate only a single value of the Revelle factor and the $pCO_2$ (water) for pore-water and water from offshore in the BoB. We believe that as the data for pore-water is close to that of ground water and data from the far offshore BoB is close to the data from the northern BoB, these representative single data will not hamper any interpretation of the present study.
Oxygen saturation was calculated according to the solubility equation given by Benson and Krause (1984). Apparent oxygen utilization (AOU) was calculated according to the formula, \( \text{AOU} = C_{\text{obs}} - C_{\text{sat}} \), where \( C_{\text{obs}} \) is the observed DO concentration and \( C_{\text{sat}} \) is the oxygen concentration at saturation.

Statistical analyses were done using SPSS software version 16.0 (SPSS Inc., Chicago, USA). We checked for normality of the biogeochemical parameters—namely salinity, water temperature, DO, \( p\text{CO}_2\) (water), pH, TAlk, DIC, TAlk/DIC, \( \delta^{13}\text{C}_{\text{DIC}} \) and Chl-\( a \)—by applying the Shapiro–Wilk test. All of these parameters exhibited non-normal distributions. In order to test the significance of differences in these parameters between the creek and estuarine stations, we applied the non-parametric Mann–Whitney U test (also known as the Wilcoxon rank sum test).

**Results**

**Physicochemical setting**

Salinity and water temperature were significantly higher in the creeks than in the estuary (salinity, \( p < 0.001 \); water temperature, \( p < 0.001 \); Mann–Whitney U test) but varied within very narrow ranges (Table 1). DO concentrations were significantly higher in the estuary than in the creeks (\( p < 0.001 \), Table 1). Chl-\( a \) concentrations were significantly higher in the creeks than in the estuary (\( p < 0.001 \), Table 1).

**Carbonate-chemistry parameters**

There were no significant differences in mean TAlk and DIC between the creeks and estuary, but some creek samples had high values during ebb tide (TAlk, 2732 \( \mu \text{mol kg}^{-1} \); DIC, 2683 \( \mu \text{mol kg}^{-1} \)) (Fig. 2a, b). No significant difference was found in TAlk/DIC (\( p > 0.05 \)) (Fig. 2c). \( p\text{CO}_2\) (water) and \( \delta^{13}\text{C}_{\text{DIC}} \) were significantly different between creeks and estuary (\( p < 0.001 \); the mean \( p\text{CO}_2\) (water) was oversaturated in the creeks (470 \( \pm 162 \) \( \mu \text{atm} \), mean \( \pm \) SD), and undersaturated in the estuary (387 \( \pm 58 \) \( \mu \text{atm} \)) with respect to \( p\text{CO}_2\) (air) (408 \( \mu \text{atm} \)) (Fig. 3 and Table 1), whereas, the mean pH was slightly lower in the creeks (7.91 \( \pm 0.16 \)) than in the estuary (7.96 \( \pm 0.06 \)); however, the difference was not significant (\( p > 0.05 \), Table 1). \( \delta^{13}\text{C}_{\text{DIC}} \) varied over a wide range, from \( -1.5\% \) to \(-7.6\% \) with a mean of \(-3.4\% \pm 1.9\% \) and \(-1.9\% \pm 0.2\% \) in the creeks and estuary, respectively (Fig. 2d). Both Chl-\( a \) (Fig. 4a, b) and turbidity (Fig. 4c, d) showed significant positive correlations with \( p\text{CO}_2\) (water) (\( p < 0.001 \)).

The Revelle factor varied over a wide range among the possible sources of the mangrove-surrounding waters in the estuary (Table 2). The mean Revelle factors at the creek and estuarine stations were 12.8 \( \pm 2.1 \) (range, 11.2–17.9) and 12.4 \( \pm 1.4 \) (11.5–16.3), respectively. The Revelle factor of the riverine freshwater end-member was 26.7. The estimated Revelle factors of the northern BoB and the waters farther offshore in the BoB (almost to latitude 10\(^\circ\)N) were 7.5–8.1 and 8.9, respectively, whereas those of pore-water and groundwater were 17.6 and 14.9, respectively.

\( \text{DIC} \) showed significant negative correlations with \( \delta^{13}\text{C}_{\text{DIC}} \) both in the creeks (\( r = -0.990, \ p < 0.001 \)) and the estuary (\( r = -0.980, \ p < 0.001 \)) (Fig. 5a). There were significant negative correlations between TAlk/DIC and excess DIC both in the creeks (\( r = -0.995; \ p < 0.001 \)) and the estuary (\( r = -0.980; \ p < 0.001 \)) (Fig. 5b). nDIC and nTAlk were significantly correlated in the creeks (\( r = 0.995; \ p < 0.001 \)) and in the estuary (\( r = 0.992; \ p < 0.001 \)), with slopes of 0.84 and 0.81, respectively (Fig. 5c). There was no significant correlation between AOU and excess DIC (\( p < 0.001 \), Fig. 5d).

Air–water \( \text{CO}_2 \) flux

Both the creek and estuarine stations exhibited diel and tidal variation in air–water \( \text{CO}_2 \) fluxes, and acted as both sinks and sources of atmospheric \( \text{CO}_2 \) at different times over the diel cycle (Fig. 5). The creeks acted as net sources of \( \text{CO}_2 \) with a mean flux of 13 \( \pm 34 \) \( \mu \text{mol m}^{-2} \text{ h}^{-1} \), 107 \( \pm 284 \) \( \mu \text{mol m}^{-2} \text{ h}^{-1} \) and 69 \( \pm 180 \) \( \mu \text{mol m}^{-2} \text{ h}^{-1} \) according to Eqs. (3a), (4), and (5), respectively. In contrast, the estuary acted as a net sink for \( \text{CO}_2 \) with a mean flux of \(-4 \pm 12 \) \( \mu \text{mol m}^{-2} \text{ h}^{-1} \), \(-37 \pm 95 \) \( \mu \text{mol m}^{-2} \text{ h}^{-1} \) and \(-23 \pm 64 \) \( \mu \text{mol m}^{-2} \text{ h}^{-1} \) according to Eqs. (3a), (4), and (5), respectively.
Table 1 Mean ± standard deviation and range (in parentheses) of all high-temporal-resolution physicochemical and carbonate-chemistry parameters measured with sensors at creek (C) and estuarine (E) stations, and the estimated pH

| Parameter                  | Station | Tidal phase | C1 (Average of C1 and C2) | C2 | C3 | C4 | C5 | C6 | E1 (Average of E1 to E6) |
|----------------------------|---------|-------------|--------------------------|----|----|----|----|----|-------------------------|
| Water temperature (°C)     | 4th Day after NT | 20.94 ± 0.46 (20.27–22.52) | 21.41 ± 0.75 (19.44–22.52) | 21.18 ± 0.66 (19.44–22.52) | 21.13 ± 0.41 (20.41–22.35) | 21.32 ± 0.19 (21.09–22.01) | 21.96 ± 0.19 (21.50–22.44) | 21.62 ± 0.14 (21.37–21.90) | 22.29 ± 0.23 (21.99–22.83) |
| Salinity                  | 4th Day after NT | 24.83 ± 0.16 (24.22–25.12) | 25.92 ± 0.46 (25.01–27.62) | 25.37 ± 0.65 (24.22–27.62) | 25.01 ± 0.07 (24.89–25.21) | 25.24 ± 0.08 (25.07–25.59) | 25.83 ± 0.09 (25.60–26.04) | 25.57 ± 0.12 (25.27–25.79) | 26.05 ± 0.15 (25.80–26.34) |
| Dissolved oxygen (mg L⁻³) | 4th Day after NT | 5.1 ± 0.2 (4.8–5.4) | 5.0 ± 0.2 (4.7–5.4) | 5.4 ± 0.2 (5.1–5.7) | 5.9 ± 0.2 (5.5–6.2) | 5.4 ± 0.2 (5.0–5.6) | 6.0 ± 0.2 (5.6–6.6) | 5.6 ± 0.2 (5.3–6.0) | 5.9 ± 0.1 (5.7–6.3) |
| Chlorophyll-a (mg m⁻³)    | 4th Day after NT | 1.83 ± 0.48 (0.33–6.46) | 1.79 ± 0.61 (0.84–5.00) | 1.71 ± 0.90 (0.33–6.46) | 1.15 ± 0.31 (0.52–6.41) | 1.14 ± 0.23 (0.33–3.10) | NA | 1.40 ± 0.23 (0.79–3.98) | 1.23 ± 0.38 (0.63–3.23) |
| pCO₂(water) (µatm)        | 4th Day after NT | 417 ± 92 (315–709) | 523 ± 197 (369–1204) | 470 ± 162 (315–1204) | 356 ± 53 (311–610) | 344 ± 13 (320–390) | 425 ± 39 (368–576) | 355 ± 14 (331–410) | 397 ± 14 (380–450) |
| pH                        | 4th Day after NT | 7.98 ± 0.05 (7.93–8.03) | 7.85 ± 0.21 (7.54–7.97) | 7.91 ± 0.16 (7.54–8.03) | 7.89 ± 0.13 (7.70–8.00) | 7.99 ± 0.02 (7.96–8.00) | 7.96 ± 0.01 (7.95–7.98) | 7.99 ± 0.01 (7.99–8.00) | 7.97 ± 0.01 (7.96–7.98) |

For tidal and diurnal variability of these variables see Fig. 3 and Supplementary Figure S1

ST Spring Tide, NT Neap Tide, NA data not available
Discussion

Sinks and low effluxes of CO₂ in comparison with the global average

Our results show that the estuary clearly acted as a net CO₂ sink and the mangrove creeks acted as a weak source of CO₂. The uncertainty of the estimated \( pCO_2 \) (water) and air–water CO₂ flux was lower compared to previous studies in the Sundarbans because of the higher temporal resolution (cf. Rosentreter et al. 2018) and direct estimation of \( pCO_2 \) (water) (cf. David et al. 2018). We compared the \( pCO_2 \) (water) and air–water CO₂ flux with values from other studies around the world (Table 3), focusing on the recent studies that used high-temporal-resolution direct measurements of \( pCO_2 \) (water) or air–water CO₂ flux. It is evident that no previous study of mangrove-surrounding waters reported such a mean low \( pCO_2 \) (water) as that in the present study, although several previous studies reported values in the lower range of \( pCO_2 \) (water) from the mangrove creeks that were equal to or lower than the atmospheric CO₂ concentration (Call et al. 2015; Sippo et al. 2016; Rosentreter et al. 2018). The low \( pCO_2 \) (water) observed in the present study indicates that sometimes the mangrove-surrounding waters act as a sink or weak source of atmospheric CO₂. Nevertheless, our high-temporal-resolution and direct \( pCO_2 \) measurements can reduce the uncertainty in the data for the air–water CO₂ flux of mangrove-surrounding waters showing high spatio-temporal variability.

The air–water CO₂ flux data for the mangrove-surrounding waters obtained by the present study and previous studies in the Sundarbans are markedly lower than the recently reported global average. Even the annual mean CO₂ flux for the entire Matla Estuary (approximately 6.3 ± 0.9 mmol m⁻² d⁻¹; Akhand et al.
Fig. 3 Time-series plots of diel variation of salinity, $p$CO$_2$(water) (µatm), and CO$_2$ flux [µmol m$^{-2}$ h$^{-1}$; according to the wind parameterization of Ho et al. (2011)] at the two creek stations (C1 and C2) and the six estuarine stations (E1, E2, E3, E4, E5 and E6). FT, flood tide; ET, ebb tide. The shaded regions show the night-time portion of the diel cycle. (Color figure online)
Fig. 4 Relationship between chlorophyll-α concentration and $pCO_2$ (water) in a the creek stations and b the estuarine stations. Relationship between turbidity (FTU, Formazin Turbidity Unit) and $pCO_2$ (water) in c the creek stations and d the estuarine stations. Red straight lines indicate regression lines. (Color figure online)

Table 2 Revelle factors and $pCO_2$ (water) values at the study site and in potential sources of mangrove-surrounding waters in the Indian Sundarbans

| Water types                          | Revelle factor | $pCO_2$ (water) (μatm) | Source of estimate                  |
|--------------------------------------|----------------|------------------------|-------------------------------------|
| Potential Sources                    |                |                        |                                     |
| Riverine freshwater end-member       | 26.7–33.6      | 1421–4678              | Present study                       |
|                                      |                |                        | Akhand et al. (2016)                |
|                                      |                |                        | Dutta et al. (2019)                 |
| Pore-water of Sundarbans             | 17.6           | ≈5423                  | Mandal et al. (2009)                |
|                                      |                |                        | Dutta et al. (2019)                 |
| Groundwater of Sundarbans            | 14.9–18.4      | 506–1219               | Akhand et al. (unpublished data)    |
| Northern Bay of Bengal                | 7.5–8.1        | 149–299                | Sarma et al. (2012)                 |
|                                      |                |                        | Akhand et al. (2012)                |
| Bay of Bengal (≈10°N latitude)       | 8.9            | ≈340                   | Goyet et al. (1999)                 |
| Study sites                          |                |                        |                                     |
| Creek stations                       | 11.0–14.1      | 315–1204               | Present study                       |
| Estuarine stations                   | 10.9–12.5      | 311–636                | Present study                       |
2016) is much less than the world average for emissions from mangrove-associated water (56.8 ± 8.9 mmol m⁻² d⁻¹; Rosentreter et al. 2018) (Fig. 6).

Predominance of marine water and low pCO₂

The waters around mangroves usually exhibit significant diel and tidal variability in terms of both pCO₂(water) and air–water CO₂ flux, with higher pCO₂(water) values during low tides and vice versa (Zablocki et al. 2011). However, the continuous high-temporal-resolution measurements in this study showed that these changes in pCO₂(water) with the tide occurred only at the creek stations (Fig. 3) and not at the estuarine stations, except for station E1. The high pCO₂(water) during low tide is generally attributed to pCO₂-rich pore-water as well as groundwater, and the low pCO₂(water) during flood tide results from the dilution of mangrove-derived water (Akhand et al. 2016; Call et al. 2015; Maher et al. 2013). The absence of pCO₂(water) maxima during low tide at our estuarine stations suggests that the pore-water, which seems to have a prominent effect in the creeks, did not play a significant role in regulating the diel variation of pCO₂(water) at these estuarine stations. The most plausible reason behind this observation might be that the higher marine water volume in the estuarine stations and higher water residence time in the creek stations exceed the effect of pore-water in the estuarine stations even during the low tide period.

The Revelle factor is the ratio of the relative change of DIC in marine water; thus, it reflects the carbonate

![Fig. 5](image-url)
buffering capacity of the water mass and is implicitly related to the TAlk/DIC ratio (Egleston et al. 2010). A low Revelle factor can indicate high carbonate buffering capacity and the potential for CO$_2$ uptake (Bates et al. 2012; Sabine et al. 2004). Among the possible sources of water in the mangrove-dominated estuary in this study, the riverine freshwater end-member, groundwater, and pore-water had both...
$p\text{CO}_2$(water) values and Revelle factors higher than the observed values in the creeks and estuary (Table 2). In contrast, the waters from the northern BoB and farther offshore had $p\text{CO}_2$(water) and an estimated Revelle factor lower than the waters at the study site (Table 2). These results indicate that the low-$p\text{CO}_2$ waters of the BoB have higher buffering capacity than all of the other possible sources of water in this estuary. Thus, we infer that the low $p\text{CO}_2$(water) of the Sundarbans may be due mainly to the predominance of low-$p\text{CO}_2$ (Akhand et al. 2012, 2013a; Goyet et al. 1999; Sarma et al. 2012) and high buffering capacity.

Another reason behind the low $p\text{CO}_2$(water) of this study site, in addition to the predominance of low-$p\text{CO}_2$ water from the BoB, might be that the Indian Sundarbans receives comparatively less riverine freshwater input than other river-dominated estuaries (Chakrabarti 1998; Mitra et al. 2009). River-dominated estuaries having large riverine freshwater inputs have shown higher $p\text{CO}_2$(water) than marine-dominated estuaries having less riverine freshwater inputs (Akhand et al. 2016; Jiang et al. 2008; Maher and Eyre 2012). The rapid export of material because of estuarine geometry and the meso- to macro-tidal nature of this estuary further enhance the low $p\text{CO}_2$(water) character. Specifically, Ray et al. (2018a) and references therein have shown that the rapid transport of material from Sundarbans mangroves to the BoB is due to (i) the shorter water residence time, (ii) large tidal amplitudes, and (iii) the funnel-shaped geometry of the estuary, which tends to amplify the tide, in turn facilitating faster material transport.

Effects of mangroves on air–water CO$_2$ flux

The measured TA$_{\text{alk}}$ and DIC in the present study are in agreement with the reported ranges for TA$_{\text{alk}}$ and DIC for other mangrove-associated waters that act as net sources of CO$_2$ (David et al. 2018; Linto et al. 2014; Ray et al. 2018b; Zablocki et al. 2011) despite the sink and weak source behaviors of the Indian Sundarbans. Some high TA$_{\text{alk}}$ and DIC values at creek stations during ebb tide suggest that the TA$_{\text{alk}}$ and DIC were added from the mangrove ecosystem. The significant negative correlation between TA$_{\text{alk}}$/DIC and excess DIC in both creeks and estuary suggests that higher TA$_{\text{alk}}$/DIC suppressed excess DIC and contributed to the CO$_2$ sink or weak source character (Fig. 5b). Exports of higher TA$_{\text{alk}}$-to-DIC ratios from mangroves to coastal waters, with a TA$_{\text{alk}}$/DIC ratio of 1.2, led to an overall increase in pH and thus had a buffering effect (Sippo et al. 2016). The TA$_{\text{alk}}$/DIC at our sites was 1.08 $\pm$ 0.06 (mol mol$^{-1}$), indicating a relatively high buffering capacity in the mangrove-surrounding waters of the Sundarbans, but it was not markedly higher than that observed in other parts of the world (1.00–1.10; Sippo et al. 2016). Increased pH and under-saturation of $p\text{CO}_2$ in mangrove-surrounding waters can also occur as a result of the remnant TA$_{\text{alk}}$ after CO$_2$ outgassing and outwelling of mangrove TA$_{\text{alk}}$, as discussed by Sippo et al. (2016).

Our results showed that diagenetic processes in this mangrove system reduced TA$_{\text{alk}}$/DIC ratios, which increased $p\text{CO}_2$ in the mangrove-surrounding waters (Fig. 6). The diagenesis of organic carbon in mangroves sediments takes place through several anaerobic pathways that supply TA$_{\text{alk}}$ and DIC and change the pore-water TA$_{\text{alk}}$/DIC (Borges et al. 2003; Bouillon et al. 2007b; Koné and Borges 2008). The relationships between nDIC and nTA$_{\text{alk}}$ in this study, with slopes of 0.84 (creeks) and 0.81 (estuary) (Fig. 5c), suggest denitrification, as observed in the waters around mangroves in the Mekong Delta, Vietnam (Alongi et al. 2000) and Gaji Bay, Kenya (Bouillon et al. 2007a). Denitrification has previously been reported in the mangrove sediment of the Indian Sundarbans and Bangladesh (Neogi et al. 2016) Sundarbans. Das et al. (2020) reported a mean denitrification rate of 14.72 nmol N$_2$O-N h$^{-1}$ (g dry wt$^{-1}$) in the sediments of Lothian Island (Indian Sundarbans). The absence of a significant correlation between AOU and excess DIC (Fig. 5d) suggests that aerobic respiration was not the major diagenetic pathway for organic matter (OM) degradation in the present study. A primary assumption of the method of delineating the diagenetic pathway of OM mineralization using the nDIC:nTA$_{\text{alk}}$ ratio is that the C, N, and P of the OM follows the Redfield ratio (Krumins et al. 2013). However, the organic matter in the mangrove-surrounding waters might not always match the Redfield ratio, which implies an uncertainty in this stoichiometric analysis. Still, our principal purpose was to understand the nDIC:nTA$_{\text{alk}}$ ratio from the perspective of the carbonate buffering capacity, rather than determining the exact organic matter mineralization pathway.
nDIC:nTAlk ratios indicate that the diagenetic pathways at the study site provide a lower buffering capacity than the marine water, and increased excess DIC and pCO$_2$ (Fig. 5b). Marine water with high buffering capacity suppresses the effect of pCO$_2$ increase in the mangrove system and the lowers CO$_2$ evasion even in the creek stations (Fig. 5b).

The effect of mangroves on carbonate chemistry in coastal waters is variable depending on the biogeochemical processes in the mangrove systems. We found that the mean pH value at the more marine-influenced estuarine stations was higher than that at the more mangrove-influenced creek stations (Table 1); however, the difference of pH was not found to be significant between creek and estuarine stations ($p > 0.05$). pH values reported in the offshore waters of the BoB (8.13–8.46) were much higher (by 0.1–0.4 units; Akhand et al. 2012, 2013a; Sarma et al. 2012) than the pH values observed in both the creek and the estuarine stations (Table 1). These results also support our suggestion that the low pCO$_2$(water) of the mangrove-surrounding waters can be attributed to the effect of BoB water with low pCO$_2$ (Akhand et al. 2012, 2013a; Goyet et al. 1999; Sarma et al. 2012;) and higher buffering capacity rather than to the effect of mangroves.

Biological uptake of DIC is also an important factor in explaining low pCO$_2$(water) as well as the air–water CO$_2$ flux. Both buffering capacity and Revelle factor can be affected by biological factor(s) (Hauck and Volker, 2015; Hauck et al. 2015). In the present study, higher mean Chl-$_a$ concentrations (Table 1 and Supplementary Fig. S1) were associated with pCO$_2$(water) above saturation at the creek stations and vice versa at the estuarine stations (Table 2). However, there was no significant negative relationship between Chl-$_a$ concentration and pCO$_2$(water) at any of the stations, rather a significant positive correlation was found (Fig. 4a and b). These results indicate that biological control of pCO$_2$(water) was less important than physical mixing in this study. However, the negative excess DIC at several creek stations (Fig. 5b) might be related to higher phytoplankton production, which would contribute to reducing the CO$_2$ effluxes. The significant positive correlation between Chl-$_a$ and pCO$_2$(water), along with significant positive correlation between turbidity and pCO$_2$(water) (Fig. 4c and d), indicates that high Chl-$_a$ with turbidity led to a high organic matter degradation rate and high DIC input as observed in some previous studies (Abril et al. 2000; Fay and McKinley 2017; Tishchenko et al. 2018).

We found a significant negative correlation between DIC and δ$^{13}$C$_{DIC}$ (Fig. 5a), with higher DIC values and lower δ$^{13}$C$_{DIC}$ values in the creeks than in the estuary (Fig. 2b and d). This suggests that the sources of DIC have lighter δ$^{13}$C values and were supplied from the mangroves. The most plausible explanation for the DIC sources is therefore a combination of mineralization of mangrove tissue (δ$^{13}$C$_{TOC}$ = –28.08% to –26.31%; Ray et al. 2015), POC in the water around mangroves (δ$^{13}$C$_{POC}$ = –23.3% to –22.3%; Dutta et al. 2019; Ray et al. 2015), and marine phytoplankton (–22.0% to –20.0%; Rosentreter et al. 2018). Groundwater DIC and pore-water DIC (–18.0%, pore-water DIC in Sundarbans, Dutta et al. 2019; –14.5% to –10.0 %, ground water/pore-water DIC, Maher et al. 2013) are also possible DIC sources. We assumed that air–water CO$_2$ fluxes had a negligible effect on isotopic fractionation, as the fluxes were at near-equilibrium. This type of mixed source for DIC has been reported in other mangrove environments (Maher et al. 2017; Rosentreter et al. 2018; Sea et al. 2018). Under such a scenario, the carbon dynamics in the mangrove-surrounding waters cannot be explained solely by mangrove-derived DIC loading, but must also be regulated by other allochthonous sources (Rosentreter et al. 2018).

Conclusion

The results of this study suggest that mangrove-surrounding waters can act as a sink or a weak source for atmospheric CO$_2$, contrary to most previous studies. There have been previous studies of CO$_2$ dynamics in the Sundarbans, but the precision and temporal resolution of the data were too coarse to determine source/sink characteristics. The present study successfully overcame these problems and reduced the uncertainties by analyzing the diel variability of pCO$_2$(water) at eight sites, covering tidal maxima and minima. Our findings further show that the CO$_2$ sink or weak source character of the Sundarbans’ mangrove-surrounding waters was caused by predominance of the low-pCO$_2$ and high-
buffering-capacity waters of the BoB. The TA\textit{k} export from the mangroves also buffers the increase in $p$CO$_2$ owing to the DIC addition. The marine water with high TA\textit{k}/DIC ratio buffers the effect of $p$CO$_2$ increase in the mangrove systems, lowering CO$_2$ evasion in the study site. Finally, we argue that areas with such low emissions should be included when the global mangrove carbon budget is estimated by scaling-up regional observations.

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**Authors’ contributions** AA, AC, KW, SH, and TK designed the study, AA, AC, and SD did the field work and sample collection. AA, AC, KW, SD, TT, and KC did the chemical and data analyses. AA, AC, KW, SH, and TK wrote the manuscript. All authors read and approved the manuscript.

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**Data availability** Dataset is archived in the Zenodo database ([https://doi.org/10.5281/zenodo.4572499](https://doi.org/10.5281/zenodo.4572499); Akhand et al. 2021_Biogeochemistry).

**Code availability** Not applicable.

**Declarations**

**Conflict of interest** The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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