Editor's comment:
Comment:
Two reviewers and Dr. Akhand raised a number of critical issues on your manuscript. I also agree to their primary concerns; above all the lack of clear study objectives and arguments unsupported by data (primarily those regarding the roles of phytoplankton productivity and OC mineralization in the Hooghly and exogenous C inputs in the Sundarbans). As reviewer 2 mentioned, major differences in measured parameters appear to derive simply from the dominant influence of fresh inputs to the Hooghly estuary. However, many of your arguments have been based on the assumption of anthropogenic impacts on the Hooghly C dynamics, without providing detailed descriptions and data on anthropogenic sources of C.

Response: Based on reviewer's comments, we have reanalysed our data which suggested significant impact of freshwater input on estuarine DIC and POC (line 454-461 and 577-582 of the revised manuscript) but not on DOC and pCO_2 (line 534-540 and 646-652 of the revised manuscript). We have discussed it in the revised manuscript. In the revised manuscript, organic carbon mineralization in the Sundarbans has been established with stronger evidence (ECO_2-AOU relationship) as suggested by the reviewer – 2 (line 619-633 of the revised manuscript). As we don't have any direct data, probability of CO_2 supply through pore-water exchange is supported based on higher low tide CO_2 compared to high tide at Matla estuary (of Sundarbans) as suggested by Akhand et al. (2016) (line 638-641 of the revised manuscript). For primary productivity, earlier works have been used to support our study (line 391-393 and 417-419 of the revised manuscript). Evidence for anthropogenic POC input was found from our study in the Hooghly based on stable isotope values (line 565-567 of the revised manuscript) but for DOC we have to rely only on concentration based interpretation due to lack of isotopic data (line 521-523 of the revised manuscript).

Given the large number of critical issues to address, a thorough revision beyond major revisions would be required to reconsider your manuscript for publication in Biogeosciences. The revised manuscript will be sent to the reviewers to make sure that you have adequately addressed all the raised issues and minor technical corrections. Regarding your assumption on the Hooghly as an anthropogenically modified system, I wondered if your summary of published water quality data (as shown in the supplement) would provide sufficient data supporting your arguments on the dominant role of C sources of anthropogenic origin driving the reported C dynamics in the Hooghly estuary. Please compile (and discuss) more concentration and isotope data on C species of anthropogenic origin, from your own surveys or the literature, to relate them to your interpretations.

Response: Based on reviewers as well as your comment, we have modified the entire manuscript and we think it is much better manuscript now. We have added more information (including quantification of anthropogenic discharge to the estuary on daily basis) in tabular form (Table 1 in the revised manuscript) to establish stronger anthropogenic influence on the Hooghly estuary and the same is proved from our study as well (line 564-567 of the revised manuscript). We hope now our justification and the manuscript will be well accepted to the editor.

I would like to ask you to make all the changes easily identifiable in a marked-up manuscript and a point-by-point reply to the reviewers' and my own comments to facilitate the second round of review. It would be a good idea to indicate line numbers of the revised manuscript when you respond to the reviewer comments.

Response: We are happy to do the needful during the submission procedure.

Response to the reviewer - 1

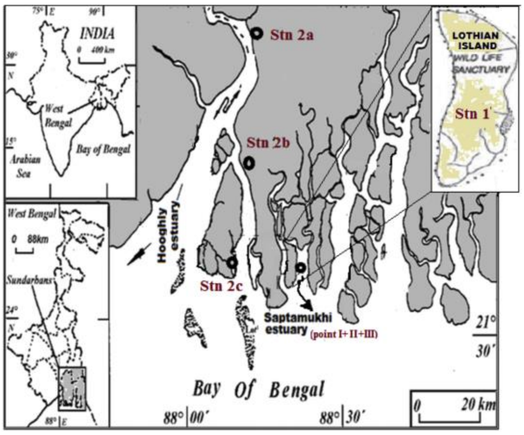
Review of Dutta et al., The authors made measurements of organic and inorganic carbon parameters, along with isotopes and other ancillary measurements in an attempt to determine the sources and distribution of DIC, DOC, and POC in an estuary in the Hooghly-Sundarbans system (shortly written as C biogeochemistry by the authors). Although the ms falls within the scopes of BG and covers a good data range from various sites of the estuarine system but finally it ends up in a disappointment because of poor writing and hesitations of choosing a concrete aim. Unfortunately, the manuscript reads like a data dump, with incomplete descriptions of the methods, presentation of the data, and some speculation about processes but with major processes left out; nothing seems conclusive. The manuscript is still in quite a rough stage, as detailed with a non-exhaustive list of examples below, and does not seem ready for publication.

Response: We are thankful to the reviewer for his constructive criticism and comments. We are now happy to include his suggestions in the revised manuscript at suitable places.

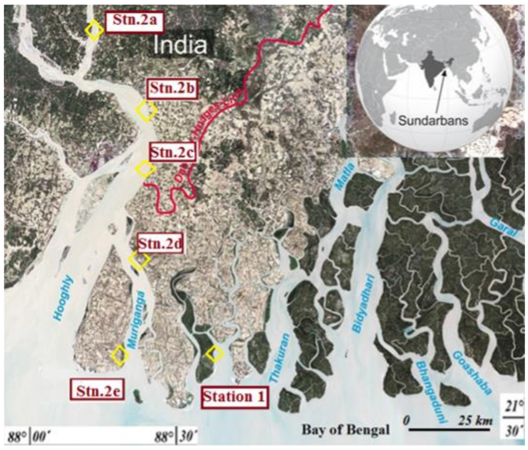
Specific comments:

The problem lies within the title. It seems the authors are in serous dilemma to show the data what actual basis: on C dynamics in polluted vs non-polluted system or only focus on mangroves and compare with sidechain Hooghly in a specific season or discuss on DIC mainly and less focus on DOC and POC or avoid already published articles on the same systems on same parameters on same season! (e.g. Samanta 2015, Ray 2018, 2015) Unfortunately nothing was clear due to poor writing and unclear intention.

***Response*:** The main objective of the present study is to bring out contrast in different components of the carbon cycle of anthropogenically affected Hooghly estuary and mangrove-dominated estuaries of Sundarbans during postmonsoon. We have tried to focus on each component depending on the variabilities and scope of our data. We would respectfully disagree with the reviewer that we have tried to avoid the earlier works by Samanta et al., (2015) and Ray et al. (2015, 2018). We have cited their works and used the findings of these authors in our manuscript to interpret our data. We would also like to submit that whereas Samanta et al. (2015) is a nice study with comprehensive focus on only DIC in the Hooghly estuary; Ray et al. (2015, 2018) covers more number of parameters with limited spatial coverage. In a vast mangrove ecosystem as Sundarbans, Ray et al. (2015, 2018) have covered just one location during both studies. We have tried our best to cover the Hooghly-Sundarbans system on wider scale with multiple parameters to comprehensively study C dynamics in this system. We have made relevant changes in line nos: 127-155 of the revised manuscript with clearly stated intention (line no 142 – 145 of the revised manuscript).



Ray et al. (2015)



Ray et al. (2018)

Figure 1. Spatial coverage in the Hooghly-Sundarbans by Ray et al. (2015, 2018)

Other major comments

I would suggest authors to give details of the sampling stations e.g. how or what type of anthropogenic input is there in the Hooghly? From where it is more coming from (upstream?).

Response: Surface runoff in freshwater region such as waste water discharge from the City of Kolkata (St. H2) and jute industry (located in between many locations of St. H1 to H3) is a major source of anthropogenic inputs to the Hooghly. We have included previously published nutrients concentration as an evidence for higher anthropogenic input in the Hooghly. Relevant modification is done in line no : 120-126 along with addition of a new Table -1 showing contrast between the Hooghly and Sundarbans .

Its better to segment the study sites of Hooghly as upper/mid/lower stretch and Sundarbans as west/central and east. I anticipate the upper and mid stretches are human or industrial impacted compared to lower, so one of ideas in designing the story would be to explain variations of results within Hooghly first between e.g. H1-6 and H6-11 and then compare with S,T,M series. That would read the paper interesting otherwise its just mimicking the findings already shown by Samanta 2015, Ray 2018.

Response: During this postmonsoonal study, based on the present salinity range and gradient, it is difficult to divide the Hooghly estuary into upper/mid/lower stretch like other estuaries with sharp salinity gradient from fresh to marine zone. Although reviewer has suggested human and industrial influence along with lower estuarine region as a basis for such demarcation, we believe that such demarcation would be qualitative as no quantitative information are available to us to support such demarcation. Therefore, we are inclined to divide Hooghly estuary as freshwater zone (H1-H6) and mixing zone (H6 – H11) based on our salinity data. For the Sundarbans, the spatial extent is not wide enough (less than 100 km^2) to divide them into west, central and east zones. If we apply this criterion, we would be left with 3 data points from each region (upper/middle/lower), which is not enough for data analysis and further interpretation to understand the characteristics of individual estuaries (S, T and M). Therefore, in the revised manuscript, we discussed first the freshwater and mixing region in the Hooghly estuary and then compare it with the Sundarbans. We hope that the reviewer will agree to our suggestion.

Authors argued on C- data limitation of previous reports but it is found that Samanata’15 covered even much higher sites from Hooghly than the present report (c.a 35 vs 13 surface water and 8 vs 8 ground water) and Ray '18 was also not far (>10 in S series vs 10 S,T,M). So this argument on data imitation does not hold true!

Response: We agree with the reviewer that DIC is extensively discussed by Samanta et al. (2015) for the Hooghly estuary with much better spatial and seasonal coverage compared to our study. The author also reported $\delta^{13}C_{POC}$ at some locations (n = 26). DIC and pCO_2 for the Hooghly and Matla estuaries have also been reported by Akhand et al. (2016). The first report for the Hooghly-Sundarbans system with different components of C cycle with their isotopic compositions were reported by Ray et al. (2015). However, this study is limited by spatial coverage (3 stations from Hooghly and one from Sundarbans). Unless reviewer is referring to paper other than Ray et al. (2018) published in The Science of Total Environment, his argument about Ray et al. (2018) having large sampling locations (>10 in S series vs 10 S,T,M as pointed out by the reviewer) appears to be not correct. The map of the sampling location of Ray et al. (2018) is shown above. In the light of the above, we would like to argue that the present study has much larger spatial coverage (13 stations from Hooghly and 11 from Indian Sundarbans, line 183-190 of the revised manuscript) with multiple parameters and is better equipped to decipher the differences in C biogeochemistry of the contrasting systems such as Hooghly and the estuaries of Sundarbans.

Result section is only meant for results and it should be avoided to define data set and add citations in Results that fully present in the paper. It is proposed to move those parts of the Result section to discussion (LN 229-234, 248-49, 257-59, 267-71).

Response: Thanks for the suggestion. We followed it throughout the revised manuscript.

This is over-speculative to argue on contributions of pore water on the overlying DIC concentrations based on only one measurement (Tab 3, Lothian PW).

Response: We agree with the reviewer that it is not enough to quantify contribution of pore water on adjoining estuarine water DIC pool based on a single measurement in this large mangrove ecosystem (Sundarbans). We are sure reviewer will appreciate that it is a logistics challenge to perform sampling in the Sundarbans. To perform sampling, permission is needed from the forest department. Also, very few islands are open for scientific investigations and some of them are tiger infested. During the present sampling, we had planned to cover at least all littoral zones of the Lothian Island. However, we were not permitted by the forest security service as conditions were not conducive to carry out investigation at mid and upper littoral zones. Therefore, we had to restrict our measurement in lower littoral zone only. Our advective DIC flux across mangrove sediment-estuary interface can be considered as first-time baseline value. We have indicated the reason for one sampling location in the manuscript as (line no 212-215 of the revised manuscript):

"Pore-water was also collected from lower littoral zone of the Lothian Island (one of the virgin island of the Indian Sundarbans) by digging a hole (~30 cm below the water table). It was not possible to collect pore-water samples from mid and upper littoral zones due to logistic problems."

LN342- 345: This is unclear why ΔDICM2 is shown as micromole instead of permil.

Response: As you can see from the formula, the units of numerator and denominator is (µM x ‰) and ‰, respectively. The ‰' gets cancelled keeping ΔDIC_{M2} unit as 'µM'.

Authors should better calculate the amount of DOC and POC added or subtracted from the system applying conservative mixing (same way they did for DIC) and explain in-depth details of their mixing pattern (same applies to DIC).

Response: Thanks to the reviewer for this suggestion. Using similarly calculated end members values or taken from the same references as DIC, added or removed POC and DOC in the Hooghly were calculated in the revised manuscript. For Sundarbans, mangrove derived POC and DOC addition/removal was calculated using the same expressions as DIC. Additionally, very similar to DIC, a mixing plot between ΔPOC and $\Delta\delta^{13}C_{POC}$ was plotted to explore influencing processes. However, for DOC it was not possible to perform this analysis due to unavailability of $\delta^{13}C_{DOC}$ data during the present study. We have used interrelationships between various parameters to justify removal or addition. We have included the above information in the revised manuscript (line no 543-614 of the revised manuscript).

Based on the above, additions to the POC section is as following: "To decipher processes involved in POC modification, estimated ΔC for POC (ΔPOC) in the Hooghly indicated both net addition (n = 3) and removal (n = 3) of POC in the freshwater region (ΔPOC = – 0.45 to 0.48), whereas removal (n = 6) dominated over addition (n = 1) in the mixing region (ΔPOC = – 0.39 to 0.07). In an estuary, POC may be added through freshwater and surface runoff mediated inputs, phytoplankton productivity, and DOC flocculation. The removal of POC is likely due to settling at subtidal sediment, export to adjacent continental shelf region, modification via conversion to DOC and mineralization in case of oxygenated estuary.

The plot between $\Delta\delta^{13}C$ for POC ($\Delta\delta^{13}C_{POC}$) and ΔPOC (Fig. 5d) indicated different processes to be active in different regions of the Hooghly estuary. Decrease in ΔPOC with increase in $\Delta\delta^{13}C_{POC}$ (RR; n = 4 for mixing region and n = 1 for freshwater region) suggested modification of POC due to aerobic respiration (or mineralization). This process did not appear to significantly impact estuarine CO_2 pool as evident from the POC - pCO_2 relationship (freshwater region: p = 0.29, mixing region: p = 0.50; Fig. 5e). Decrease in both ΔPOC and $\Delta\delta^{13}C_{POC}$ (SD; n = 2 for mixing region and n = 2 for freshwater region) supported settling of POC to sub-tidal sediment. Despite high water residence time (~ 40 days during postmonsoon, Samanta et al., 2015), this process may not be effective in the Hooghly due to unstable estuarine condition (described earlier). Increase in ΔPOC with decrease in $\Delta\delta^{13}C_{POC}$ (SR, FR & PP; n = 2 for freshwater region) indicated increase of POC via surface and freshwater runoff as well as phytoplankton productivity. Increase in both ΔPOC and $\Delta\delta^{13}C_{POC}$ (n = 1 for mixing region and n = 1 for freshwater region) may be linked to DOC to POC conversion by flocculation.

In the Sundarbans, negative and lower ΔPOC_{M2} (–209 to –28μM) compared to ΔPOC_{M1} (–35 to 327μM) suggested DIC like behavior, i.e., simultaneous removal or modification along with addition of mangrove derived POC. No evidence for *in situ* POC-DOC exchange was obvious based on POC-DOC relationship; however, signal for POC mineralization was evident in the Sundarbans from POC - pCO_2 relationship (r^2 = 0.37, p = 0.05, Fig.5f). Similar to the Hooghly, despite high water residence time in mangroves (Alongi et al., 2005, Singh et al., 2016), unstable estuarine condition may not favor efficient settlement of POC at sub-tidal sediment. The export of POC from the Hooghly-Sundarbans system to the northern BOB, without significant *in situ* modification, is also a possibility. This export has been estimated to be ~0.02 - 0.07Tg and ~ 0.58Tg annually for the Hooghly and Sundarbans, respectively (Ray et al. 2018)."

Please look later for explanation related to DOC.

LN349 Are the ground and pore water discharge not being considered as 'biogeochemical' process?

Response*:* We believe it is better to leave ground and pore water discharge from the realm of biogeochemical processes, as no biogeochemical processes are associated with them. It may be described as hydrological processes. We found the "The driving forces of pore-water and groundwater flow in permeable coastal sediments: A review" published by Santos et al. (2012) in the Estuarine Coastal and Shelf Science as a nice review work in this field.

Section 4.3. This part is weakly written and over-speculative without supporting any evidence e.g. the argument of DOC photo-oxidation or conversion of DOC to POC as removal process. While it requires suitable ambient condition for DOC photo-oxidation such as high water residence time, stable environmental condition (not expected in mangroves), the same applies to adsorption/desorption of DOC-POC. Part of that exchange is mediated by charged complexes, repulsion - attraction interactions, and therefore subject to salinity effects. So, when river water rich in DOC first mixes with saline water, at least a portion of DOC is lost from solution (removed) and incorporated into POC (Fe-oxide colloids usually are extracted at the same time). Once the salinity exceeds 2 - 3, however, the effect of salinity on coagulation behaviour is largely complete. Another point is no detailed explanation on distribution pattern with salinity was given, authors should highlight the reasons of the mild upward gradient along Hooghly and steep downward trend along the Sundarban.

Response: We are thankful to the reviewer for insightful comment on the DOC study. We have included these points in the revised manuscript (line no 463-540 of the revised manuscript).

The section on DOC is evolved as following in the revised version: "During the present study, DOC concentrations in the Hooghly estuary were higher compared previously reported by Ray et al. (2018) (226.9 ± 26.2 to 324 ± 27µM), whereas DOC in the Sundarbans were comparable with Ray et al. (2018) (262.5 ± 48.2µM). The marine and freshwater mixing did not appear to exert major control over DOC in the Hooghly-Sundarbans system as evident from lack of significant correlations between DOC and salinity (Hooghly freshwater: r^2 = 0.33, p = 0.23; Hooghly mixing region: r^2 = 0.10, p = 0.50; Sundarbans: r^2 = 0.27, p = 0.10, Fig.4a). Our observations showed similarity with other Indian estuaries (Bouillon et al., 2003) with opposite reports from elsewhere (Raymond and Bauer, 2001a, Abril et al., 2002). This indicates that DOC in this sub-tropical estuarine system is principally controlled by processes other than mixing of two water masses.

Although it is difficult to accurately decipher processes influencing DOC without $\delta^{13}C_{DOC}$ data, some insights may be obtained from estimated ΔC of DOC (ΔDOC). The estimated ΔDOC in the Hooghly indicated both net addition (n = 3) and removal (n = 3) of DOC in the freshwater zone (ΔDOC = – 0.16 to 0.11); whereas, only net addition was evident throughout the mixing zone (ΔDOC = 0.08 to 1.74). In the Sundarbans, except lower Thakuran (St. T3, $\Delta\ DOC_{M1}$ = – 20µM), net addition of mangrove derived DOC was estimated throughout (ΔDOC_{M1} = 2 - 134µM).

In an estuary, DOC can be added through *in situ* production (by benthic and pelagic primary producers), lysis of halophobic freshwater phytoplankton cells and POC dissolution. DOC can be removed through bacterial mineralization, flocculation as POC, and photo-oxidation (Bouillon et al., 2006). At the Hooghly - Sundarbans system, no evidence for freshwater phytoplankton ($\delta^{13}C$: – 33 to – 40‰; Freitas et al., 2001) was found from $\delta^{13}C_{POC}$, ruling out its potential effect on DOC. Although an indirect signal for phytoplankton productivity was observed in the freshwater region from $\delta^{13}C_{DIC}$ and POC relationship (r^2 = 0.68, p = 0.05), further evaluation of its impact on DOC was not possible due to lack of direct primary productivity measurements. Contradictory results exist regarding influence of phytoplankton productivity on DOC. Some studies did not find direct link between DOC and primary productivity (Boto and Wellington, 1988), whereas primary productivity mediated significant DOC formation (~ 8 - 40%) has been reported by others (Dittmar & Lara 2001a, Kristensen & Suraswadi 2002).

The DOC - pCO_2 relationship suggested inefficient bacterial DOC mineralization in the Hooghly (freshwater zone: p = 0.69, mixing zone: p = 0.67, Fig. 4b). However, significant positive relationship between these two in the Sundarbans (r^2 = 0.45, p = 0.02, Fig. 4c) indicated increase in aerobic bacterial activity with increasing DOC. In mangrove ecosystems, leaching of mangrove leaf litter as DOC is fast as ~ 30% of mangrove leaf litter leaching as DOC is reported within initial 9 days of degradation (Camilleri and Ribi, 1986). In the Sundarbans, mangrove litter fall peaks during postmonsoon (Ray et al. 2011) and its subsequent significant leaching as DOC was evident during the present study from comparatively higher DOC compared to POC (DOC:POC = 0.50 - 3.39, mean: 1.79 ± 0.94%). Our interpretation for Sundarbans corroborated with that reported by Ray et al. (2018) for the same system as well as Bouillon et al. (2003) for the Godavari estuary, South India.

Despite high water residence time in the Hooghly (~ 40 days during postmonsoon, Samanta et al., 2015) and in mangrove ecosystem like Sundarbans (Alongi et al., 2005, Singh et al., 2016), DOC photo-oxidation may not be so potent due to unstable estuarine condition in the Hooghly-Sundarbans system (Richardson number < 0.14) with intensive vertical mixing and longitudinal dispersion coefficients of 784 $m^2 s^{-1}$ (Goutam et al., 2015, Sadhuram et al., 2005). The unstable condition may not favor DOC - POC interconversion as well but mediated by charged complexes and repulsion - attraction interactions, the interconversion partly depends upon variation in salinity. More specifically, the interconversion is efficient during initial mixing of fresh (river) and seawater and the coagulation is mostly complete within salinity range 2 - 3. This appeared to be the case in the Hooghly, where DOC and POC was negatively correlated in the freshwater region (r^2 = 0.86, p = 0.007, Fig.4d), which was missing in the mixing region (p = 0.43) and in the Sundarbans (p = 0.84).

Although estimated ΔDOC indicated largely net DOC addition to the Hooghly-Sundarbans system, except leaf litter leaching in the Sundarbans, no significant evidence for other internal sources was found. This suggested potential contribution from external sources that may include industrial effluents and municipal wastewater discharge (i.e., surface runoff) in the freshwater region of the Hooghly (Table 1). However, there is no direct DOC influx data to corroborate the same. Relatively higher DOC compared to POC (DOC/POC > 1) at some locations (H2, H5, H6) may stand as a signal for higher DOC contribution at those locations, but it is not prudent to pinpoint its sources due to lack of isotopic data. Although anthropogenic inputs are mostly confined to freshwater region, relatively higher DOC in the mixing zone of the Hooghly compared to freshwater region suggested DOC input via some additional pathway, possibly groundwater discharge. The contribution of groundwater to the Hooghly estuary within the salinity range observed during the present study has been reported (Samanta et al., 2015). However, there is no report of groundwater mediated DOC influx to the estuary. For mangrove-dominated ecosystems like Sundarbans, a recent study by Maher et al. (2013) estimated ~ 89 - 92% of the total DOC export to be driven by groundwater advection. To understand spatial variability of DOC chemistry in the Hooghly-

Sundarbans system, a thorough investigation related to groundwater and surface runoff mediated DOC flux is warranted.

Overall, on an average ~ 40% higher DOC in the Hooghly compared to the Sundarbans appeared to be due to cumulative effect of freshwater contributions, higher anthropogenic inputs, influence of biogeochemical processes and groundwater contribution. However, DOC inputs via other pathways may be dominant over freshwater mediated input as evident from insignificant DOC - salinity relationship during the present study. To quantitatively understand the relative control of the above-mentioned contributors to the DOC pool in the Hooghly-Sundarbans system, the individual components need to be studied in detail."

Section 4.4 LN410 only freshwater runoff, no surface run off that adds POC too in upstream?

Response: We have included possibility of surface runoff mediated POC addition in the revised manuscript (line no 546-557 of the revised manuscript).

The section is looking like: No significant SPM-salinity or POC-salinity relationship was observed during the present study (Fig. 5a & 5b), except for a moderate negative correlation between POC and salinity ($r^2 = 0.62$, $p = 0.06$) in the freshwater region of the Hooghly. This inverse relationship may be linked to freshwater mediated POC addition. Also, as described earlier, contribution of POC via surface-runoff is also a possibility in this region due to presence of several industries and large urban population (St: H2: Megacity Kolkata) that discharge industrial effluent and municipal wastewater to the estuary on regular basis (Table 1). Primary signal for surface runoff mediated POC addition was evident in the freshwater zone where ~ 61% and ~ 43% higher POC at 'H3' and 'H4' compared to an upstream location (St. H2) was observed. However, based on the present data, it is not possible to decouple freshwater and surface runoff mediated POC input to the Hooghly estuary."

LN440-446 this part is totally redundant as there was not an iota of signal of CH4 from the observed d13 POC (13CH4 is ~ 55-60 permil)

Response: We have removed the section from the revised manuscript.

Does the author have Chl-a or nutrient data (even from literature) to support higher marine input in POC in Sundarban and 13C values of mangrove leaf, and soil from Hooghly to denote higher terrigenous contribution to the POC pool? Authors are suggested to read carefully the works of Samanta'15 and Ray'18 and use their values to support some of the arguments.

Response: We are thankful to the reviewer for this suggestion. The modification is looking as following (line no – 561 – 577 of the revised manuscript):

"In general, wide range for $\delta^{13}C$ (rivers ~ –25 to –28‰; marine plankton ~ –18 to –22‰; C_3 plant ~ –23 to –34‰; C_4 plants ~ – 9 to –17‰) have been reported by different researchers in different ecosystems (Smith and Epstein, 1971, Hedges et al., 1997, Zhang et al., 1997, Dehairs et al., 2000, Bouillon et al., 2002). In the Hooghly, our measured $\delta^{13}C_{POC}$ suggested influx of POC via freshwater runoff as well as terrestrial C_3 plants. Additionally, the estuary was also anthropogenically stressed during postmonsoon with measured $\delta^{13}C_{POC}$ within the range reported for sewage ($\delta^{13}C$ ~ –28 to –14 ‰, Andrews et al., 1998). In the mixing zone of the Hooghly, significantly lower $\delta^{13}C_{POC}$ at 'H11' and 'H12' compared to other sampling locations may be linked to localized ^{13}C depleted organic C influx to the estuary from adjacent mangrove and anthropogenic discharge, respectively. In the estuaries of Sundarbans, isotopic signatures of POC showed similarity with terrestrial C_3 plants. Interestingly, despite being mangrove-dominated estuary (salinity: 12.74 - 16.55) no clear signature of either freshwater or mangrove ($\delta^{13}C$: mangrove leaf ~ –28.4‰, soil ~ –24.3‰, Ray et al., 2015, 2018) borne POC was evident from $\delta^{13}C_{POC}$ values, suggesting towards the possibility of significant POC modification within the system. Modification of POC within the estuaries of Indian sub-continent have been reported earlier (Sarma et al., 2014)."

points of concerns

terminology > I counted 'biogeochemistry' was used over 25 times in the 16 pages ms! too much. Additionally, this is not clear to me what does it actually mean by C biogeochemistry?

Response: We have taken care of it throughout the revised manuscript.

Is it C-components distributions in different phases (solid suspended and dissolved) under varying biogeochemical processes? If so please specify at least once

Response: We have included it in line no 143 of the revised manuscript.

> d13C values are not 'depleted' or 'enriched' (LN256, 428..). When referring to d13C values, they can be described as higher or lower when comparing different samples, or one could describe differences as e.g. a certain C pool is enriched or depleted in 13C versus another C pool or sample.

Response: We have taken care of it throughout the revised manuscript.

> r2 not R2

Response: We have taken care of it throughout the revised manuscript.

Inconsistent use of [POC] in the discussion, if the bracket is used for POC then it should also appear for DIC and DOC

Response: Brackets have been removed for all cases in the revised manuscript.

unit Random use of units: DOC in mg/L, DIC in mM, POC in uM. These should be harmonized. Use DOC in uM for better compare with other studies

Response: To maintain uniformity, all dissolved and particulate C parameters are presented as 'μM' in the revised version.

Sampling Define sampling strategy neatly, Its written postmonsoon was chosen due to high litterfall, but there is no account of litter source identified for DOC or POC or any impact positive or negative on estuarine C biogeochemistry authors assumed. That is to be addressed in the discussion. Mention the H, S, T, M series in the text Mention general tidal nature while sampling (height, HT/LT, depth).

Response: The leaf litter fall is the main source of organic carbon in mangrove sediment, which peaks during postmonsson (Ray et al., 2011). It is expected that high litter fall might influence C components in the Sundarbans (line no – 145 – 147 of the revised manuscript). The signal for influence of litter fall on DOC was evident from the DOC:POC ratio (as leaching) in the Sundarbans (line no – 498 – 502 of the revised manuscript), but no direct signature for mangrove leaf litter on POC was found (modification is also a possibility, see POC section for more details) (line no – 571 – 575 of the revised manuscript). We have included these points in the revised manuscript. Details on 'H, S, T and M' are included in the revised manuscript (line no – 184 – 186 and 190 of the revised manuscript). All samples were collected during the low tide phase as intertidal mangrove sediment - water interaction through groundwater discharge is maximum during low tide phase. Therefore, low tide is ideal sampling time to understand impact of mangroves on adjoining estuarine systems. To assess contrasting features between the Sundarbans and Hooghly, sampling was also conducted during low tide in the Hooghly estuary (line no – 186 – 191 of the revised manuscript).

Methods

Comment – 1: specify> pore size of filters used for DOC, SPM relative uncertainty in POC methods;

Response: Pre-combusted (500^{o}C for 6 hours) Whatman GF/F (pore size: 0.7µm) was used for DOC filtration and SPM collection. Uncertainty for POC was < 10%. Related information are included in line no – 221 – 222 and 243 of the revised manuscript.

Comment – 2: technique of pore water collection; ground water (from tube pump?)

Response: We have included collection techniques for pore-water and groundwater in the revised manuscript on following lines (line no – 212 – 218 of the revised manuscript):

"Pore-water was also collected from lower littoral zone of the Lothian Island (one of the virgin island of the Indian Sundarbans) by digging a hole (~30 cm below the water table). It was not possible to collect pore-water samples from mid and upper littoral zones due to logistic problems. After purging water at least twice in the bore, sample was collected from the bottom of the bore through syringe and transferred to the glass vial (Maher et al., 2013). Twelve groundwater samples were collected from the nearby locations of the Hooghly-Sundarbans system via tube pump."

Figures

Again weak representation: font sizes of x, y axis digits (and titles) to be increased much (too much stress to eyes now!); use box to cover legends, its confusing with data points and legends, remove break in y axis in Fug 3e and 4a), black star coding was used both for sundarban and observed d13DIC and grey round coding was used for Hooghly and observed DIC, these symbols must be changed to give separate identity of them in all figs <overall IMPROVE CLARITY of ALL FIGURES>

Response: We have presented high resolution figures in the revised manuscript to present each region and each ecosystem.

Data use a consistent number of decimals (1) to report d13C data, and Salinity considering the analytical error on the measurements.

Response: Both salinity and δ^{13}C data will be presented up to two decimals in the revised manuscript.

Minor comments

Comment – 1: First sentence of abstract is redundant

Response: We have removed it from the revised manuscript.

Comment – 2: LN65 Use current reference for the riverine export flux (works of Pete Raymond, Huang)

Response: We are thankful to the reviewer for suggestion. We have included Huang et al. (2012) in the revised manuscript (line no – 65-69 of the revised manuscript).

Comment – 3: Many references are out of place e.g. the comparison of present data with Khura (LN 231, 249 Miyajima paper) was unlikely as two environments are totally different even if compared authors should mention conservative data like S in Khura estuary for better comparison.

Response: Salinity of Khura estuary is presented in the revised manuscript (line no – 399-405 of the revised manuscript)

Comment – 4: LN234: Pro-vide values of Samanta et al 2015

Response: We have included postmonsoon DIC (1.70 – 2.25mM) and $\delta^{13}C_{DIC}$ (–11.4 to –4.0‰) values of Hooghly estuary as reported by Samanta et al. (2015) in the revised manuscript (line no – 369 of the revised manuscript).

Finally, I think it is necessary to stand back and consider how to best weave the entire story together in the discussion more efficiently and succinctly

Thanks for the valuable suggestions. We have almost entirely rewritten the manuscript and hope it will be well accepted to the reviewer.

Response to Reviewer 2

Review of Dutta et al "The postmonsoon carbon biogeochemistry of estuaries under different levels of anthropogenic impact". Submitted to Biogeosciences. This study presents data from a single cruise in 2 Indian estuaries to try and decipher differences in carbon cycling between a 2 Indian estuaries with differing levels of anthropogenic influence. After reading and rereading this paper several times, it is unclear what the purpose of this study is. There is no defined hypothesis to be tested, and while the title suggests there will be some kind of comparative analysis to look at anthropogenic impact on carbon cycling (an interesting and important topic), I am left a little underwhelmed with the analysis undertaken. The entire manuscript is based single sampling campaigns, which while not ideal is not the main issue. The main area of concern is the lack of any direction in the paper, and the somewhat descriptive and qualitative nature. I suggest that the authors define their hypothesis more clearly, and use the data to test this hypothesis.

Thanks to reviewer for going through our manuscript and providing valuable suggestions which will help to improve the quality of the revised version. We understand the concern he has raised and we have tried to improve the manuscript accordingly. As we have said in the response to reviewer 1, the main objective of the present study is to bring out contrast in different components of the carbon cycle of anthropogenically affected Hooghly estuary and mangrove-dominated estuaries of the Sundarbans during postmonsoon (line 142-145 of the revised manuscript). As suggested by the reviewer later in comments, we have introduced a table (Table 1) bringing out the differences in basic characteristics of these two systems, which will help the readers to appreciate the differences in anthropogenically affected and mangrove-dominated system. As suggested by the reviewer, in the revised version, given the contrasting nature of the estuaries, we also propose to bring out a central hypothesis. The central hypothesis of this study is: "Considering different nature and quantity of supplied OM within these two contrasting system, we hypothesize C metabolism between these two estuaries to be very different with higher CO_2 exchange flux from anthropogenically influenced estuary compared to mangrove-dominated estuary (line 147-151 of the revised manuscript)." Given the larger spatial coverage of the mangrove-dominated estuary during the present (so far only one estuary in this system has been studied), there is a need for this hypothesis to be tested on wider spatial level.

I have a series of comments below, some minor some major that may help.

Abstract I am not convinced that the data as presented can be used to draw such strong conclusions as to the drivers of carbon dynamics in the studied estuaries. For example, Ln 35-38 The evidence supporting these processes is weak at best – no measurements of production, carbonate dissolution nor pore-water exchange were measured, and the spatial trends in concentrations and isotopes (and relationships between carbon variables and DO etc.) were not strong enough to draw any distinct conclusions on the importance of these mechanisms.

Same goes for lines 45-47.

Response: Based on the specific comments of the reviewers, we have reanalysed the data and reassessed the role of processes he is referring to in sentences mentioned above. In the response to comments below, he will find that we have either discarded the descriptive part or backed the processes active with reanalysis of the data during the present study.

Line 49 – 52. I am unconvinced that the observed trends are shown to be directly linked to anthropogenic influence. Yes, the estuaries appear to differ, but what else might be driving this. For example, looking at salinity and pCO$_2$ in the 2 different estuaries – the highest salinity in the "anthropogenically" impacted estuary is lower than the lowest salinity in the "undisturbed" estuaries. Could the observed differences simply be related to freshwater input? What are the nutrient concentrations in the 2 estuaries? How different are they in hydrodynamics (looks like the geomorphology is distinctly different between the 2 estuary types from Fig 1). These are just a few of the alternative reasons to look at for explaining the differences observed

Response: Based on the comments from both reviewers, we have provided a table in the response below (Table 1 in the revised manuscript), which will help readers to understand the basic differences between the two estuaries. The present study was carried out during postmonsoon season, which brings significant amount of freshwater inputs to the region. Moreover, the Hooghly undergoes sever anthropogenic stress as it passes through industrial areas as well as one of the most densely populated region in India (included in table). We revisited the data in light of the comments from both reviewers and in responses we discuss the changes and processes active in the two estuaries, which led to observed difference.

Introduction

Ln 59 – 60 What is meant by "record biogeochemical and hydrological processes"?

Response: We meant physical/hydrological processes such as mixing between marine and freshwater, tide and wave action, sediment transport etc. and biogeochemical processes such as primary productivity, organic matter decomposition etc. We believe the reviewer was concerned with 'record'. We have modified the sentence in the revised manuscript as follows (line 60-61 of the revised manuscript):

"Situated at the interface of land and sea, estuaries are highly susceptible to anthropogenic inputs and undergo intricate biogeochemical and hydrological processes."

Ln 67 – Richey is not correct ref for this statement (Richey paper is on Amazon)

Response: Thanks to point this out. We have modified the section as follows (line 65-69 of the revised manuscript):

"Tropical rivers, which constitute ~ 66% of global river water discharge, deliver ~ 0.53Pg C to the estuaries annually (Huang et al., 2012). The majority of this exported C is in dissolved form [dissolved inorganic C (DIC): 0.21PgCyr^{-1} and dissolved organic C (DOC): 0.14PgCyr^{-1}] with some contribution as particulate [particulate organic C (POC): 0.13PgCyr^{-1} and particulate inorganic C (PIC): 0.05PgCyr^{-1}] (Huang et al., 2012)."

Ln 68 – 70 – Still large uncertainties on estuarine CO_2 flux – look at error bars on Cai, 2011 estimate

Response: We have pointed out this issue in the revised manuscript (lines 70- 72 of the revised manuscript).

Ln 76 What is meant by "biogeochemical characteristic"?

Response: We meant with regards to cycling of bio-available elements, such as C, N and P. We have changed this sentence to more specific (line 102-105 of the revised manuscript).

Ln 78 – 79 Not always – see Cotovicz Jr, L. C., Knoppers, B. A., Brandini, N., Costa Santos, S. J., & Abril, G. (2015). A strong co₂ sink enhanced by eutrophication in a tropical coastal embayment (guanabara bay, rio de janeiro, brazil). Biogeosciences, 12(20), 6125-6146. doi:10.5194/bg-12-6125-2015

Response: Thanks for this reference. In the revised version, the section may look like (line 105-110 of the revised manuscript):

"In anthropogenically affected estuarine systems, heterotrophy generally dominates over autotrophy (Heip et al., 1995; Gattuso et al., 1998) and a substantial fraction of biologically reactive OM gets mineralized within the system (Servais et al., 1987; Ittekkot, 1988; Hopkinson et al., 1997; Moran et al., 1999). However, this is not always the case as observed in Guanabara Bay, Brazil, which acts as a strong CO_2 sink enhanced by eutrophication (Cotovicz Jr. et al., 2015)."

Among others

Ln 81 – 84 There has been a lot of work on mangrove carbon cycling work done since Dittmar and Larra's work in the early 2000's. Might be worth looking at more recent papers to see how far our understanding has come since then.

Response: We have modified this section in the revised manuscript. Following information may be added (line 76-96 of the revised manuscript).:

"Mangroves covering 137,760 km^2 along tropical and sub-tropical estuaries and coastlines (Giri et al. 2011) are among the most productive natural ecosystems in the world with net primary productivity of 218 ± 72Tg C yr^{-1} (Bouillon et al. 2008). Fine root production coupled with litter fall and wood production are primary sources of mangrove derived C to intertidal sediment (Bouillon et al., 2008). The fate of this mangrove derived C remains poorly understood. Despite taking C burial and CO_2 emission flux across mangrove sediment-atmosphere interface into account, estimates of global mangrove C budget revealed a significant imbalance (~72%) between mangrove net primary productivity and its sinks (Bouillon et al., 2008). Earlier studies reported mangroves to be responsible for ~10% of the global terrestrial derived POC and DOC export to the coastal zones (Jennerjahn and Ittekkot, 2002; Dittmar et al. 2006). However, recent studies proposed DIC exchange as major C export pathway from mangrove forests, which was ~70% of the total mineralized C transport from mangrove forests to coastal waters (Maher et al., 2013; Alongi, 2014; Alongi and Mukhopadhyay, 2014). Another study reported groundwater advection from mangrove to be responsible for 93-99% of total DIC export and 89-92% of total DOC export to the coastal ocean (Maher et al., 2013). Upon extrapolating these C export fluxes to the global mangrove area, it was found that the calculated C exports were similar to the missing mangrove C sink (Sippo et al., 2016). The remaining C that escapes export gets buried in sub-surface sediment layers and participates in anaerobic processes (linked to production of biogenic trace gases like CH_4) or undergoes long-term sequestration (Jennerjhan and Ittekkot 2002; Barnes et al., 2006; Kristensen and Alongi, 2006; Donato et al., 2011; Linto et al., 2014)".

.

Ln 104- 106 Give some quantitative data to support your "anthropogenically influenced" argument. What are nutrient concentrations like? Population density? Land use? Freshwater inflow? Etc etc. A table compiling this data would give the reader an instant understanding of the differences.

Response: Thanks to the reviewer for bringing this point. Reviewer 1 has also asked to include some information in this context from literature. Texts or a table comparing the Hooghly and Sundarbans during postmonsoon based on nutrients concentration, Chla, population density and freshwater inflow will be introduced in the revised manuscript. The information is presented in Table-1 of the revised manuscript as follows:

Parameters	Hooghly	Sundarbans
Nutrients (postmonsoon)	DIN: 14.72 ± 1.77 to 27.20 ± 2.05µM DIP: 1.64 ± 0.23 to 2.11 ± 0.46µM DSi: 77.75 ± 6.57 to 117.38 ± 11.54µM (Mukhopadhyay et al., 2006)	DIN: 11.70 ± 7.65µM DIP: 1.01 ± 0.52µM DSi: 75.9 ± 36.9µM (Biswas et al., 2004)
Chl*a* (postmonsoon)	Chl-a: 2.35 – 2.79 mgm^{-3} (Mukhopadhyay et al., 2006)	Chl*a*: 7.88 ± 1.90 mgm^{-3} (Dutta et al., 2015)
Population density	North 24 Parganas and Hooghly: 2500 km^{-2}, Kolkata: 22000 km^{-2}, Howrah: 3300km^{-2}, South 24 Parganas: 820 km^{-2}	No major Cities and town
Freshwater discharge (postmonsoon)	3070 - 7301 million m^{3} (Rudra et al., 2014)	No information available
Catchment area	6 x 10^{4}km^{2} (Sarkar et al., 2017)	No information available
Industrial and municipal wastewater discharge	1153.8Million L d^{-1} (Ghosh, 1973; Khan, 1995)	No information available
Dissolved metal flux	Increased from 230 – 1770% annually (Samanta and Dalai, 2018)	No information available

Line 117 What is meant by positive and negative feedback here? These terms are not really applicable to biogeochemistry as a whole, but may be related to specific mechanisms/cycles.

Response: In the revised manuscript we have changed this statement as follows (line 145-147 of the revised manuscript):

"The postmonsoon sampling was chosen because of relatively stable estuarine condition for wider spatial coverage and peak mangrove leaf litter fall during this season (Ray et al., 2011), which may have influence on estuarine C dynamics."

Ln 137-140 Clearly there is freshwater input – the salinities are very low. In fact, my thoughts are that these freshwater inputs are a main driver of the observed differences.

Response: The freshwater input in the estuaries of Sundarbans is evident from the salinity values (12.64-16.69) during the study period. However, if you see the salinity values in the Hooghly estuary during the same season (0.04-10.37), the extent of freshwater input in Hooghly is far greater. This difference gets further widened during premonsoon. Because of this reason, we stated 'no perennial source of freshwater and limited anthropogenic input during monsoon". We have changed the sentence as (line 183-186 of the revised manuscript):

"Covering upper, middle, and lower estuarine regions, the present study was carried out during low tide condition in three major estuaries of the Indian Sundarbans [Saptamukhi (S1-S3), Thakuran (T1-T3), and Matla (M1-M3); Fig. 1a] along with its related waterways (S4 & M4)."

Ln 159 Assume the filters were GF/F filters – add these details.

Response: Yes, as reviewer stated it was Whatman GF/F filters. We have included it in the revised manuscript (line 207 of the revised manuscript).

Ln 161 Accuracy of TAlk measurements. Were CRMs measured (hope so!). Also add accuracy/precision etc of all other parameters.

Response: Uncertainties were as follows (line 201-244 of the revised manuscript):

Water temperature: ±0.1^{o}C, Salinity: ±0.1, DO: ±0.1 mgL^{-1}, DIC: <1%, $\delta^{13}C_{DIC}$: < ±0.10‰, DOC: ±52 µgL^{-1}, POC: <10%, $\delta^{13}C_{POC}$: < ±0.10‰, pCO_2: ± 1%. Yes, accuracy of TAlk was tested using Dickson standard (CRM: Bottle – 131) and uncertainty was found to be ±1µmolkg^{-1}.

Ln 196 – 198 What were the input parameters for measuring pCO2? What disassociation constants were used etc?

Response: The pCO_2 was calculated using TAlk, pH, water temperature and salinity and the dissociation constants were calculated following Millero, (2013). We have included it in the revised manuscript (line 247-248 of the revised manuscript).

Ln 205 – 208 Why use L&M relationship? Need some kind of justification here other than saying it is conservative.

Response: Unfortunately, we don't have data on estuarine current velocity which along with wind speed is used for flux calculation as it is believed that turbulence of estuary might have an effect on air-water trace gas flux calculation. Based on only wind velocity, the L&M relationship is one of the most reliable and tested methods for flux calculations, which has been used in previous studies in the region as well (Biswas et al., 2004) We have included it in line 257-258 of the revised manuscript).

Results

Do not compare and contrast your data with previous studies in the results. Just report your data.

Response: We have removed the comparison part from the result to the discussion.

Discussion

Ln 289 – 293 What are the implications for these findings? Need to dig deeper or remove.

Response: Our intension was to present influence of salinity on pH and provide the information at the beginning that this region is a bicarbonate dominated system. We have removed the sentences in the revised version.

Ln 306-311 (and Fig 3b) How was the conservative d13C-DIC mixing line calculated? Looks like you have simply added a linear relationship between the 2 endmembers, the relationship is generally not linear (See Fry, B. (2002). Conservative mixing of stable isotopes across estuarine salinity gradients: A conceptual framework for monitoring watershed influences on downstream fisheries production. Estuaries, 25(2), 264-271. Also as you do not have any mineralogy of carbonates – I would avoid using the term "calcite" precipitation, change to "carbonate" precipitation.

Response: Concentrations and stable isotopic compositions of dissolved or particulate C (presented as C) during conservative mixing (C_{CM} and $\delta^{13}C_{CM}$) were computed as follows (Carpenter et al., 1975, Mook and Tan, 1991):

$$C_{CM} = C_F F_F + C_M F_M$$

$$\delta^{13}C_{CM} = \frac{S_S\,[C_F\,\delta^{13}C_F - C_M\,\delta^{13}C_M] + S_F\,C_M\,\delta^{13}C_M - S_M\,C_F\,\delta^{13}C_F}{S_S\,(C_F - C_M) + S_F\,C_M - S_M\,C_F}$$

Here, 'S' denotes salinity, the suffixes CM, F, M and S denote conservative mixing, freshwater end member, marine end member and sample, respectively. F_F = freshwater fraction = 1 – (S_S / S_M) and F_M = marine water fraction = (1– F_F). This is a commonly used expression for such studies and has been followed by many other workers (Samanta et al. (2015); Bouillon et al. (2003)). The following expressions have been included in line 273-282 of the revised manuscript)

We have changed 'calcite precipitation' as 'carbonate precipitation' in the revised manuscript (line 385-386 and 1063-1064 of the revised manuscript).

Ln 323-325 – What does DO tell you about primary production? Looks like DO is generally under-saturated?

Response: The influence of primary productivity (PP) and/or CO_2 outgassing on DIC at the mixing zone was evident from mixing plot between ΔDIC and $\Delta\delta^{13}C_{DIC}$. We tried to go further and decouple these two processes based on TAlk - DIC relationship. However, as suggested by the reviewer, due to lack of PP measurements and level of DO indicate that it may not be a stretch. We have removed this part from the revised manuscript.

Ln 335-338 Describe all the terms in this equation in the following text

Response: We have described all terms in the revised manuscript (line no 273-306 of the revised manuscript) Additionally, in the revised manuscript, $\delta^{13}C_{Mangrove}$ will be changed as -28.4‰ as reported by Ray et al. (2015) for the Sundarbans system (line no 304 of the revised manuscript).

Ln 359 Where do the TAlk/DIC numbers come from? The stoichiometric relationship should be based on the slope of the line over the whole estuary, rather than individual data points – therefore not sure how you have a range here.

Response: Thanks to the reviewer for this suggestion. Based on his advice, we have made the changes in the revised manuscript (line 425-437 of the revised manuscript):

"High pCO_2 and DIC along with low pH and TAlk/DIC are general characteristics of groundwater, specially within carbonate aquifer region (Cai et al., 2003). Although all the parameters of groundwater inorganic C system (like pH, TAlk and pCO_2) were not measured during the present study, groundwater DIC were ~5.57 and ~3.61 times higher compared to mean surface water DIC in the Sundarbans and Hooghly, respectively. The markedly higher DIC in groundwater as well as similarity in its isotopic composition with estuarine DIC may stand as a signal for influence of groundwater on estuarine DIC, with possibly higher influence at the Sundarbans than Hooghly as evident from the slope of the TAlk - DIC relationships (Hooghly: 0.98, Sundarbans: 0.03). In the Sundarbans, to the best of our knowledge, no report exists regarding groundwater discharge. Contradictory reports exist for the Hooghly, where Samanta et al. (2015) indicated groundwater contribution at low salinity regime (salinity < 10, same as our salinity range) based on 'Ca' measurement, which was not observed based on 'Ra' isotope measurement in an earlier study (Somayajulu et al., 2002)."

Ln 364 – 368 Give details on this calculation. Just using the discharge rate and pore water DIC concentration I get a different value.

Response: Advective DIC flux from intertidal mangrove sediment to estuarine water column (F_{ISW}) was computed using the relation (Reay et al., 1995); $F_{ISW} = \Phi.\nu.C$; where, Φ = porosity of sediment = 0.58 (Dutta et al., 2013), ν = average linear velocity = $d\Phi^{-1}$ (d = specific discharge), C = DIC concentration in intertidal sediment pore water.

So ultimately: F_{ISW} = d.C. During postmonsoon, d = 0.008 cm min^{-1} (Dutta et al., 2015a). Therefore, F_{ISW} = (0.008 cm min^{-1} x 13.43 $mmolL^{-1}$) = 0.107mmol.cm.min^{-1}/1000cm^3= 0.000107 mmol cm^{-2} min^{-1} = 1.07 mmol m^{-2} min^{-1}.

In Sundarbans, tides are semidiurnal in nature, so depending upon changes in hypsometric gradient discharge of pore water will be effective during low period only (i.e. 12 hours). So, F_{ISW} =1.07 $mmolm^{-2}min^{-1}$ = (1.07 x 60 x 12 $mmolm^{-2}d^{-1}$) =770.4 $mmolm^{-2}d^{-1}$. There is a marginal difference in the manuscript, which will be corrected. We have included all details in line 441-449 of the revised manuscript.

Ln 383-390 – Not sure that looking at pCO_2 VS DOC gives any indication as to the importance of pore-water exchange! Could also simply be freshwater input from upstream, surface water runoff, or simply leaching/respiration.

Response: We have suggested to modify the DOC section which does not include the above argument. Please see response to reviewer 1 which deals with DOC (section 4.3, line 463-540 of the revised manuscript).

Ln 412 Give details about the "jute" industry.

Response: This is an industry based on fiber of Corchorus plants, which is used in fabrics for packaging a wide range of agricultural and industrial commodities that require bags, sacks, packs, and wrappings. Locally this is known as *Jute* industry. We included some information on jute industry in line 124-126 of the revised manuscript.

Ln 424-426 The POC isotopes could simply be related to the relative amount of freshwater inputs in each system (this can also be applied to most of the other differences observed)

Response: Related to this following sections have included in the revised manuscript (line 565-575 of the revised manuscript):

"In the Hooghly, our measured $\delta^{13}C_{POC}$ suggested influx of POC via freshwater runoff as well as terrestrial C_3 plants. Additionally, the estuary was also anthropogenically stressed during postmonsoon with measured $\delta^{13}C_{POC}$ within the range reported for sewage ($\delta^{13}C$ ~ –28 to –14 ‰, Andrews et al., 1998). In the mixing zone of the Hooghly, significantly lower $\delta^{13}C_{POC}$ at 'H11' and 'H12' compared to other sampling locations may be linked to localized ^{13}C depleted organic C influx to the estuary from adjacent mangrove and anthropogenic discharge, respectively.
In the estuaries of Sundarbans, isotopic signatures of POC showed similarity with terrestrial C_3 plants. Interestingly, despite being mangrove-dominated estuary (salinity: 12.74 - 16.55) no clear signature of either freshwater or mangrove ($\delta^{13}C$: mangrove leaf ~ –28.4‰, soil ~ –24.3‰, Ray et al., 2015, 2018) borne POC was evident from $\delta^{13}C_{POC}$ values, suggesting towards the possibility of significant POC modification within the system."

Ln 431-446 I am unsure why anaerobic respiration (which is energetically les favourable than aerobic respiration) would be more important in a well oxygenated estuary. The authors should expand this to explain things more clearly or remove.

Response: We have removed anaerobic respiration part from the revised manuscript.

Ln 447-451 What is the importance/implications of this – expand or remove.

Response: The intension was to quantitatively explore dominant OC form (DOC or POC) in total OC pool and dominant dissolved C form (DIC or DOC) in total dissolved C pool in the estuary. We have removed it from the revised manuscript.

Ln 455 – 460 These sections seem to contradict each other. Initially it is stated mangrove inputs are insignificant – then pore-water exchange of mangrove derived CO_2 is highlighted as important?

Response: For the revised manuscript, ECO_2 - AOU relationship (as suggested by the reviewer) was investigated (please see response to a later comment). The significant positive relationship between the two (ECO_2 = 0.154AOU + 1.22, r^2 = 0.76, p = 0.005, n = 8) suggested influence of OM respiration on pCO2 in the Sundarbans. Although, the calculated slope (0.154) was markedly lower compared to the slope for Redfield respiration in HCO_3^- rich environment [ΔCO_2: (-ΔO_2) = 124/138 = 0.90, Zhai et al., 2005] indicating effect of OM mineralization in controlling pCO_2 to be not so potent. Therefore, possibility of pore-water mediated CO_2 influx cannot be totally neglected in mangroves. Although based on present dataset (only low tide phase sampling) it is not possible to justify the argument, a signal for it was also observed from 24 hours pCO_2 observation in the Matla estuary (Sundarbans) by Akhand et al. (2016). We have added it in the revised version line 619-641 of the revised manuscript.

references for this process, might be more appropriate to use some of those here

Response: We have included some other mangrove references in the revised manuscript, such as Call et al. (2015), Bouillon et al. (2007) (line 639-640 of the revised manuscript).

Ln 463 – 466 How about plotting ECO2 vs AOU (in molar units). Look at the slope of the line. This will give a better indication of the importance or aerobic vs anaerobic R.

Response: Regarding this following modification is done in the revised manuscript (line 620-652 of the revised manuscript)

"In the Sundarbans, barring three locations (S3, T3 and M2), a significant negative correlation between pCO_2 and %DO (r^2 = 0.76, p = 0.005; Figure not given) suggested presence of processes, such as OM mineralization, responsible for controlling both CO_2 production and O_2 consumption in the surface estuarine water. Furthermore, significant positive correlation between ECO_2 and AOU (ECO_2 = 0.057AOU + 1.22, r^2 = 0.76, p = 0.005, n = 8; Fig.6a) confirmed the effect of aerobic OM mineralization on CO_2 distribution, particularly in the upper region of the Sundarbans. Our observations were in agreement with a previous study in the Sundarbans (Akhand et al., 2016) as well as another sub-tropical estuary, Pearl River estuary, China (Zhai et al., 2005). However, relatively lower slope for ECO_2 - AOU relationship (0.057) compared to the slope for Redfield respiration in HCO_3^- rich environment [$(CH_2O)_{106}(NH_3)_{16}H_3PO_4 + 138O_2 + 18HCO_3^{2-} \rightarrow 124CO_2 + 140H_2O + 16NO_3^- + HPO_4^{2-}$; ΔCO_2: (-ΔO_2) = 124/138 = 0.90, Zhai et al., 2005] suggested lower production of CO_2 than expected from Redfield respiration. This may be linked to formation of low molecular weight OM instead of the final product (CO_2) during aerobic OM respiration (Zhai et al., 2005). Moreover, pCO_2 - salinity relationship (p = 0.18, Fig.6b) confirmed no significant effect of fresh and marine water contribution on variability of pCO_2 in the Sundarbans. Other potential source of CO_2 to mangrove-dominated Sundarbans could be groundwater (or pore water) exchange across intertidal mangrove sediment-water interface. Although based on our own dataset, it is not possible to confirm the same. However, relatively higher pCO_2 levels during low-tide compared to high-tide at Matla estuary in the Sundarbans (Akhand et al. 2016) as well as in other mangrove systems worldwide (Rosentreter et al., 2018, Call et al., 2015, Bouillon et al., 2007) suggested groundwater (or pore water) exchange to be a potential CO_2 source in such systems.

Unlike Sundarbans, ECO_2 - AOU relationship did not confirm significant impact of OM respiration on CO_2 in either freshwater ($p = 0.50$) or mixing regions ($p = 0.75$) of the Hooghly (Fig. 6c). Overall, pCO_2 in the freshwater region of the Hooghly was significantly higher compared to the mixing zone (Table 3), which may be linked to CO_2 supply in the freshwater region through freshwater or surface runoff from adjoining areas (Table - 1). Inter-estuary comparison of pCO_2 also revealed ~1291 µatm higher pCO_2 in the Hooghly compared to the Sundarbans, which was largely due to significantly higher pCO_2 in freshwater region of the Hooghly (Table 2 & 3). Lack of negative correlation between pCO_2 - salinity in freshwater region (Fig. 6d) of the Hooghly suggested limited contribution of CO_2 due to freshwater inputs. Therefore, CO_2 supply via surface runoff may be primary reason for higher pCO_2 in the Hooghly estuary."

Ln 470 – 473 How was gas exchange and the differences between CO2 and O2 coupled into this calculation? Also how does this value compare to your air-water CO2 fluxes (you will need to normalize your volumetric rates to surface area for comparison)

Response: In both freshwater and mixing zone of the Hooghly estuary, no evidence for significant impact of aerobic OM respiration on pCO_2 was found. Therefore, we have remove this section from the revised version.

Ln 480 – I think your global value for mangrove systems (63 umol/m2/d) should be 63 mmol/m2/d – which is much higher than the fluxes measured in this study.

Response: We are thankful to the reviewer for pointing this out. We have rechecked the value from Call et al. (2015). The actual value (range) is ~ 43-59 mmol C $m^{-2}d^{-1}$. We have corrected it in the revised manuscript (line 659-661 of the revised manuscript).

Conclusions:

Comment: Point 1 – this variability is likely simply linked to the variability in salinity (and therefore freshwater inputs) between the studied estuaries.

Response: Freshwater inputs definitely has a role to play in the variabilities of DIC and POC as observed. However, these variabilities are also linked to *in situ* processes in the estuaries as described in our responses to both the reviewers.

Comment: Point 2 – Unconvinced that primary production has been shown to be the main controlling factor on DIC. Without any measurements of PP or some more thorough analysis of other potential mechanisms, this statement is far too strong

Response: We have changed the conclusion as (line 682-686 of the revised manuscript):

"Coupled with freshwater contribution, inorganic and organic C metabolism appeared to be dominant processes affecting DIC in the Hooghly. However, in the Sundarbans, significant DIC removal over addition was noticed. Influence of groundwater on estuarine DIC biogeochemistry was also observed with relatively higher influence at the Sundarbans."
.

Comment: Point 3 – I see no strong conclusive evidence of either of these points. Again statement is too strong without measurements of DOC flocculation or porewater exchange of DOC

Response: We have changed the conclusion as (line 687-689 of the revised manuscript):

“Higher DOC level in the Hooghly appeared to be regulated by coupled interactions among anthropogenic inputs, biogeochemical processes and groundwater contribution rather than freshwater mediated inputs”

Comment: Point 4 Assume this is based on isotopes? Again this could simply be related to the marked differences in freshwater content within each of the estuaries.

Response: We have changed the conclusion as (line 690-692 of the revised manuscript):

“Signatures of freshwater runoff, terrestrial C_3 plants, and anthropogenic discharge were found in POC of the Hooghly, whereas evidence for only C_3 plants were noticed at the Sundarbans with possible POC modification.”

Short Comment (Akhand)

In the present study, large spatial extent has been covered which includes Hooghly River and other rivers of Indian part of Sundarban. My comments regarding the present study are as follows: 1. From the sampling strategy (line no. 150 to 153), it is apparent that only one-time discrete sampling has been done in all the sites in duplicate, whereas from the third objective of the study it is clear that the authors had the aim to quantify and characterise the air-water CO_2 flux for the post-monsoon season. The authors concluded “During post monsoon, the entire Hooghly-Sundarbans system acted as a source of CO_2 to the regional atmosphere.” How can it be concluded (even quali-tatively) from such discrete data without performing at least one complete diurnal sampling at each site within post-monsoon season, while four months (October, November, December and January) are generally considered as post monsoon season in this region?

Response: As we have stated in response to the reviewer 1, the aim of the present study is to decipher the contrast in different components of C cycle of anthropogenically affected Hooghly estuary and mangrove-dominated Sundarbans (line 142-145 of the revised manuscript). While it would normally be ideal to have both large spatial and temporal coverage including measurements of several parameters along with their isotopic compositions to decipher the same, it is rarely possible due to severe logistics and technical limitations at different levels. We are sure, working in this region, you are aware of that. As we have said in response to reviewer 1, there is only one location in the Sundarbans so far (Ray et al., 2018) from where measurements for all components of C exists. We have strived to make it more representative by larger spatial coverage. We are also aware that four months are generally considered as postmonsoon; however, in light of the limitations mentioned above and advantage of spatial coverage, the conclusions of the present study can be considered as representative of the postmonsoon. Moreover, in the comment below, you are stating that one of the findings of the present study is similar to the one you observed, i.e., both Hooghly and Sundarbans are source of CO_2 to the regional atmosphere. Although your findings on Sundarbans remains limited only to Matla estuary, which can hardly be representative of the vast Sundarbans. Compared to that, the present data set is better placed to represent Sundarbans.

2. The study area and sampling locations are quite similar with the recent work of Akhand et al. (2016). Moreover, the third objective and one of the conclusions of the present study is also very similar to the Akhand et al. (2016). For example, the authors stated, “The entire Hooghly-Sundarbans system acted as source of CO_2 to the regional atmosphere with 17 times higher emission from the Hooghly compared to Sundarbans”, whereas one of the key findings of Akhand et al. (2016) is “River dominated Hugli Estuary emits 14 times more CO_2 than the marine-dominated Matla Estuary”. Surprisingly, despite of such degree of similarity between two studies, there is no comparison of data with Akhand et al. (2016) and not even mentioning of Akhand et al. (2016) in the present work.

Response: We are familiar of Akhand et al. (2016), which deals with CO_2 dynamics in the Hooghly-Sundarbans, especially diurnal observation in Matla estuary. We appreciate your effort in performing 24 hours measurements in this turbulence estuary of the Indian Sundarbans. Akhand et al. (2016) covered four locations in the lower Hooghly estuary and 3 locations from Matla estuary; whereas we covered 13 locations from Hooghly and 11 locations from the Sundarbans including all major estuaries of the Indian Sundarbans (Saptamukhi, Thakuran and Matla) and their related waterways. Given the disparity in sampling designs and locations direct data comparison between these two studies will not be ideal. However, we have included the said study in the introduction section a recent work on Hooghly-Sundarbans system (line 133-134 of the revised manuscript).

3. Reviewer 2 already mentioned that line no. 455 to 460 are self-contradictory. I want to add that I agree with the authors statement that in the estuarine water of Sundarban, an important source of CO_2 is mangrove sediment pore-water exchange during tidal pumping. This fact is also well established from the diurnal dataset of Akhand et al. (2013) and Akhand et al. (2016) in Sundarban. But, it is not clear to me, how this phenomenon can prove the exogenous origin of CO_2?

Response: We have included Akhand et al. (2016) in the revised manuscript to support our statement in the revised manuscript (line 637-641 of the revised manuscript). “Exogenous” means outside the estuary not outside the mangrove ecosystem. We will clarify it in the revised manuscript. For Sundarbans, “Exogenous” CH_4 is already established and for more details please see Dutta et al. (2015) published in Marine Chemistry.

Moreover, except Hooghly and its distributary Muriganga, all other rivers (Saptamukhi, Thakuran, Matla, Gosaba and Bidya) in the Indian part of Sundarban have lost their original connections with the Ganga because of siltation and their estuarine character is now maintained by the monsoonal runoff only (Cole and Vaidyaraman, 1966). So, the central part of Sundarban (which comprises a major part of Indian Sundarban) experiences lack of freshwater (Chakrabarti1998; Mitra et al. 2009). Hence, the source of the exogenous nature of CO_2 input in the Indian part of Sundarban needs more clarifications.

Response: It is obvious that compared to Hooghly, the estuaries of Sundarbans lack freshwater. However, it does not appear to be completely cut off from the source as can be seen from salinity range (salinity: 12.74-16.69) during the study period. However, no correlation between pCO_2 and salinity ruled out significant role of freshwater contribution on CO_2 of the estuary. The sources of CO_2 in the Sundarbans include *in situ* OM respiration along with possibility of supply through pore water exchange during tidal pumping. Following reviewer-2 suggestion, analysis of ECO_2-AOU relationship indicated CO_2 production by OM respiration in the Sundarbans during the study period. Unfortunately, our dataset is not sufficient to prove supply of CO_2 through pore-water exchange. We have used Akhand et al. (2016) in the revised manuscript to support the argument (line 637-641 of the revised manuscript).

4. In line no. 479 to 481 authors stated “FCO_2 measured for the estuaries of Sundarbans was markedly higher than global mean FCO_2 (□63 µmol m^{-2} d^{-1}) observed in mangrove creek and other similar estuaries (Call et al., 2015)”. Reviewer 2 already correctly identified that the value should be 63 m mol $m^{-2}d^{-1}$. It might be a typo by the authors, but it may convey wrong message to the global audience about Sundarban’s mangrove surrounding water. Because, one of the key findings of Akhand et al. (2016) is that the fCO_2 (water) value of the Matla, a mangrove dominated estuary of Sundarban, is at the lower end of the reported data from other mangrove ecosystems of the world. Biswas et al. (2004) also found that the Sundarban’s mangrove dominated water is acting as a sink for atmospheric CO_2 for all the four post monsoon months, while sampling in the three river-mouths. Also see Rosentreter et al. (2018), where they estimated world average flux of □57.5 mmol m^{-2} d^{-1} of CO_2 from the mangrove surrounding water, and also commented that the CO_2 efflux from the estuarine water of

Sundarban is much lower side than the world average even sinks for atmospheric CO_2 in some cases.

Response: We are thankful to the reviewer -2 for pointing this out. After getting his comment we have rechecked the values with Call et al. (2015) and responded so in response to him. We believe that you are stretching it a bit too far for an unintentional typo in a manuscript undergoing peer-review process. As you said “It might be a typo by the authors.”. It was just that. The above value is clarified in line 660 of the revised manuscript.

References: Akhand, A., Chanda, A., Dutta, S., Manna, S., Sanyal, P., Hazra, S., Rao, K.H. and Dadhwal, V.K., 2013. Dual character of Sundarban estuary as a source and sink of CO_2 during summer: an investigation of spatial dynamics. Environmental Monitoring and Assessment, 185(8), pp.6505-6515.

Akhand, A., Chanda, A., Manna, S., Das, S., Hazra, S., Roy, R., Choudhury, S.B., Rao, K.H., Dadhwal, V.K., Chakraborty, K. and Mostofa, K.M.G., 2016. A comparison of CO_2 dynamics and air-water fluxes in a river dominated estuary and a mangrove dominated marine estuary. Geophysical Research Letters, 43(22).

Biswas, H., Mukhopadhyay, S.K., De, T.K., Sen, S. and Jana, T.K., 2004. Biogenic controls on the air-water carbon dioxide exchange in the Sundarban mangrove environment, northeast coast of Bay of Bengal, India. Limnology and Oceanography, 49(1), pp.95-101.

Chakrabarti, P.S., 1998. Changing courses of Ganga, Ganga–Padma river system, West Bengal, India–RS data usage in user orientation, river behavior and control. Journal of River Research Institute, 25, pp.19-40.

Cole, C. V., and P. P. Vaidyaraman. "Salinity distribution and effect of freshwater flows in the Hooghly River." In Proceedings of Tenth Conference on Coastal Engineering, Tokyo, Japan, September, pp. 1312-1434. 1966.

Mitra, A., Banerjee, K., Sengupta, K. and Gangopadhyay, A., 2009. Pulse of Climate Change in Indian Suindarbans: A Myth or Reality?. National Academy Science Letters (India), 32(1), p.19.

Rosentreter, J.A., Maher, D.T., Erler, D.V., Murray, R. and Eyre, B.D., 2018. Seasonal and temporal CO_2 dynamics in three tropical mangrove creeks–A revision of global mangrove CO_2 emissions. Geochimica et Cosmochimica Acta, 222, pp.729-745.

Response: We have included Akhand et al. (2016) and Rosentreter et al. (2018) in the revised manuscript (line 713-716 and 970-973 of the revised manuscript).

The postmonsoon carbon biogeochemistry of estuaries under different levels of anthropogenic impacts

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Abstract

The ~~different aspects of carbon biogeochemistry were studied during the postmonsoon at the Hooghly-Sundarbans estuarine system, a part of the Ganga-Brahmaputra river system located in the northeastern India. The~~present study focused on understanding ~~the~~ differences in postmonsoon carbon (C) biogeochemistry of two adjacent estuaries undergoing different levels of anthropogenic stress by investigating anthropogenically influenced Hooghly estuary and mangrove-dominated estuaries of the Sundarbans~~.~~ in the north-eastern India. The salinity of well oxygenated (%DO: 91 - 104%) estuaries of the Sundarbans varied over a narrow range (12.74 - 16.69) ~~during postmonsoon~~ relative to the Hooghly (0.04 - 10.37). ~~Phytoplankton productivity and~~ Apart from freshwater contribution, mixing model suggested carbonate precipitation and~~/or~~ dissolution ~~were dominant~~ to be major processes controlling DIC ~~dynamics~~ in ~~different parts~~the in the freshwater region of the Hooghly, whereas ~~signal for~~ phytoplankton productivity and CO_2 outgassing dominated mixing zone. The signatures of significant DIC removal over addition through mangrove derived ~~DIC removal~~ organic C mineralization was observed in the Sundarbans. ~~Influence of~~ The DOC in the Hooghly was ~ 40% higher compared to the Sundarbans, which was largely due to cumulative effect of anthropogenic inputs, biogeochemical processes and groundwater ~~on estuarine DIC biogeochemistry was also observed in both the estuaries with relatively higher influence at the Hooghly~~contribution rather than ~~Sundarbans. In both estuarine systems, DOC behaved non-conservatively with ~ 40% higher DOC level~~ freshwater mediated inputs. The measured $\delta^{13}C_{POC}$ in the Hooghly ~~compared to the Sundarbans. No significant~~ suggested organic matter contributions from different sources (freshwater runoff, terrestrial C_3 plants and anthropogenic discharge), whereas evidence ~~of phytoplankton production on DOC level was found in these estuaries, however signal~~for only C_3 plants was noticed at the Sundarbans. The significant departure of ~~DOC input through pore-water exchange at the~~$\delta^{13}C_{POC}$ from typical mangrove $\delta^{13}C$ in the mangrove-dominated Sundarbans ~~was observed. Relatively lower $\delta^{13}C_{POC}$ at the Hooghly~~ suggested significant POC modifications. The average pCO_2 in the Hooghly was ~ 1291 µatm higher compared to the Sundarbans ~~suggest relatively higher terrestrial influence at~~with surface run-off and organic matter respiration as dominant factors controlling pCO_2 in the Hooghly ~~with a possibility of *in situ* biogeochemical modifications of POC at the~~and Sundarbans~~. The freshwater run-off coupled with *in situ* aerobic OC mineralization controlled estuarine pCO_2 level at the Hooghly, whereas the same was principally exogenous for the Sundarbans,~~ respectively. The entire Hooghly-Sundarbans

system acted as source of CO_2 to the regional atmosphere with ~17 times higher emission from the Hooghly compared to Sundarbans. The present studyTaken together, the cycling of C in estuaries with different levels of anthropogenic influences are clearly establishes thedifferent with dominance of anthropogenically influenced estuary over relatively pristine mangrove--dominated one in as CO_2 source to the regional greenhouse gas budget and climate change perspective.atmosphere.

1 Introduction

Estuaries connecting terrestrialSituated at the interface of land and marine ecosystems recordsea, estuaries are highly susceptible to anthropogenic inputs and undergo intricate biogeochemical and hydrological processes operating between these two environments. Estuaries play an important role in modulating global carbon (C) cycle and anthropogenic carbon dioxide (CO_2) budget (Bauer et al., 2013; Regnier et al., 2013; LeQuéré et al., 2016). Atmospheric CO_2 is sequestered into terrestrial systems through photosynthesis and weathering reactions and is transported to the ocean via rivers and estuaries. About $1x10^{15}$ gTropical rivers, which constitute ~ 66% of C is discharged annually from the land global river water discharge, deliver ~ 0.53Pg C to the ocean through rivers and estuaries (Degensannually (Huang et al., 1991). Around 40 %2012). The majority of this exported C is discharged as in dissolved form [dissolved inorganic carbonC (DIC)): 0.21PgCyr^{-1} and the rest as dissolved organic carbonC (DOC) and): 0.14PgCyr^{-1}] with some contribution as particulate [particulate organic carbonC (POC) (Richey): 0.13PgCyr^{-1} and particulate inorganic C (PIC): 0.05PgCyr^{-1}] (Huang et al., 2002).2012). Although estuaries are only ~ 4% of the continental shelf regions, CO_2 emission flux from estuarine surface waters is as high as CO_2 uptake in continental shelf regions of the world, albeit with large uncertainty (Borges et al., 2005; Chen and Borges, 2009; Cai et al., 2006; Cai, 2011) suggesting). This suggests estuaries to be not only active pathway for transport of C (Ittekkot and Laane, 1991) but also a hotspot for biogeochemical modification of labile organic matter (OM) (Frankignoulle et al., 1998).

Mangroves covering 137,760 km^2 along tropical and sub-tropical estuaries and coastlines (Giri et al. 2011) are among the most productive natural ecosystems in the world with net primary productivity of 218 ± 72Tg C yr^{-1} (Bouillon et al. 2008). Fine root production coupled with litter fall and wood production are primary sources of mangrove derived C to intertidal sediment (Bouillon et al., 2008). The fate of this mangrove derived C remains poorly understood. Despite taking C burial and CO_2 emission flux across mangrove sediment-atmosphere interface into account, estimates of global mangrove C budget revealed a significant imbalance (~72%) between mangrove net primary productivity and its sinks (Bouillon et al., 2008). Earlier studies reported mangroves to be responsible for ~10% of the global terrestrial derived POC and DOC export to the coastal zones (Jennerjahn and Ittekkot, 2002; Dittmar et al. 2006). However, recent studies proposed DIC exchange as major C export pathway from mangrove forests, which was ~70% of the total mineralized C transport from mangrove forests to coastal waters (Maher et al., 2013; Alongi, 2014; Alongi and Mukhopadhyay, 2014). Another study reported groundwater advection from mangrove to be responsible for 93-99% of total DIC export and 89-92% of total DOC export to the coastal ocean (Maher et al., 2013). Upon extrapolating these C export fluxes to the global mangrove area, it was found that the calculated C exports were similar to the missing mangrove C sink (Sippo et al., 2016). The remaining C that escapes export gets buried in sub-surface sediment layers and participates in anaerobic processes (linked to production of biogenic trace gases like CH_4) or undergoes long-term sequestration (Jennerjhan and Ittekkot 2002; Barnes et al., 2006; Kristensen and Alongi, 2006; Donato et al., 2011; Linto et al., 2014).

Apart from lateral transport of dissolved and particulate C, biogeochemical processes such as primary production, OM mineralization, $CaCO_3$ precipitation / dissolution and water-atmosphere CO_2 exchange occurring in the estuarine water column also regulate inorganic and organic C biogeochemistry of a mangrove-dominated estuary. These processes largely depend upon pH, nutrient availability, euphotic depth variability as well as planktonic and bacterial biodiversity and community compositions. 72 Tg C yr^{-1} (Bouillon et al. 2008a). The biogeochemical characteristic ofThe biogeochemical cycling of bioavailable elements, such as C and N, in a mangrove-dominated estuary is largely different from anthropogenically polluted estuary, where much of the OM is derived from domestic, agricultural and industrial wastes. In anthropogenically affected estuarine systems, heterotrophy generally dominates over autotrophy (Heip et al., 1995; Gattuso et al., 1998) and a substantial fraction of biologically reactive OM gets mineralized within the system (Servais et al., 1987; Ittekkot, 1988; Hopkinson et al., 1997; Moran et al., 1999). However, this is not always the case as observed in Guanabara Bay, Brazil, which acts as a strong CO_2 sink enhanced by eutrophication (Cotovicz Jr. et al., Our understanding about transformation of mangrove derived C and its subsequent export to the adjacent aquatic system appears to be limited, particularly when mangroves are disappearing at alarming rates worldwide (Dittmar and Lara, 2001a; 2001b). A significant fraction of mangrove sequestered C is supplied to intertidal mangrove sediment via litter fall, which undergoes biogeochemical transformations leading to emission of trace gases, like CO_2 and CH_4, from sediments. The rest is exported to adjacent coastal waters or gets buried in sediment layers as long-term sequestration (Jennerjhan and Ittekkot 2002; Barnes et al., 2015). 2006; Kristensen and Alongi, 2006; Donato et al., 2011; Linto et al., 2014). Apart from lateral transport of dissolved and particulate C, biogeochemical processes, such as primary production, OM mineralization, $CaCO_3$ precipitation / dissolution and water-atmosphere CO_2 exchange, occurring in the estuarine water column also regulates inorganic and organic C biogeochemistry of a mangrove-dominated estuary. These processes largely depend upon pH, nutrient availability, euphotic depth variability as well as planktonic and bacterial biodiversity and community compositions. Lack of ample quantitative estimation of above-mentioned biogeochemical processes in many regions of the world restrains mangrove biogeochemists from an in--depth understanding of these processes, which in different ecological settings. It also leads to uncertainty in estimation of C budget of coastal C biogeochemical budget regions on global scale.

In India, research related to C biogeochemistry of estuarine ecosystems have been in focus since last two decades with emphasis on estuaries located in the southern India (e.g., Bouillon et al., 2003; Sarma et al., 2012; Sarma et al., 2014; Bhavya et al., 2017; Bhavya et al. 2018). During the present study, we focused on C biogeochemical differences of two adjacent estuarine systems, i.e., the estuaries of Sundarbans and Hooghly estuary, which are part of Ganga-Brahmaputra river system located in the northeastern India. Characteristically, these two estuaries are very different from each other with the estuaries of Sundarbans being mangrove-dominated and Hooghly as anthropogenically influenced. Biogeochemical studies in these estuaries are limited to rudimentary measurements with focus (Fig. 1). Characteristically, these two estuaries are very different from each other. The Hooghly estuary experiences significantly higher anthropogenic influence compared to mangrove-dominated Sundarbans as evidenced by high nutrient and freshwater input (Table 1). The anthropogenic influences largely include supply of the industrial effluents and domestic sewage on daily basis from industries and major cities (Kolkata and Howrah) located upstream (Table 1). The industries along the Hooghly is principally *jute* (*Corchorus olitorius*) based industry, which produces fabrics for packaging a wide range of agricultural and industrial commodities. on trace gases (Mukhopadhyay et al., 2002; Biswas et al., 2004, 2007; Dutta et al., 2013, 2015a, 2015b, 2017; Ganguly et al., 2008, 2009), with exception of one comprehensive nutrient budget at the Hooghly estuary (Mukhopadhyay et al., 2006). One of the major drawback of these studies are limited number of sampling locations. Given the vast expanse of these estuaries, extrapolation of data from these studies for the entire ecosystem may lead to overestimation/underestimation. During the present study, we focused on studying different aspects of C biogeochemistry of these two estuarine systems during post-monsoon with relatively better spatial coverage compared to previous studies. The post-monsoon sampling was chosen as it identifies as season for peak mangrove leaf litter fall (Ray et al., 2011) that may have positive or negative feedback on estuarine C biogeochemistry as well as relatively stable estuarine condition for spatial sampling. The prime interest of the present study was to understand differences in factors controlling C cycling of these two biogeochemically dissimilar ecosystems and their relative role in exchange of CO_2 across water-atmosphere interface vis-à-vis regional climate change perspective. Specifically, the objectives were to (i) investigate factors controlling DIC and DOC dynamics in the region, (ii) sources of POM in these two contrasting systems, and (iii) partial pressure of CO_2 (pCO_2) and its exchange across water-atmosphere interface at the Hooghly-Sundarbans during postmonsoon period.

The major focus of biogeochemical studies in the Hooghly and Sundarbans has been on trace gases (Mukhopadhyay et al., 2002; Biswas et al., 2004, 2007; Ganguly et al., 2008, 2009; Dutta et al., 2013, 2015, 2017) with exception of one comprehensive study on nutrient budget at the Hooghly estuary (Mukhopadhyay et al., 2006). Recently, attempts have been made to understand different aspects of C cycling in these two estuaries by different workers (Samanta et al., 2015; Ray et al., 2015, 2018; Akhand et al., 2016). Samanta et al. (2015) have comprehensively studied DIC dynamics in the Hooghly estuary, whereas Akhand et al. (2016) focused on DIC and pCO_2 at the Hooghly-Matla estuary. Different aspects of C cycling in Hooghly-Sundarbans system have been reported by Ray et al. (2015, 2018). Barring Samanta et al. (2015), which has wider spatial and temporal coverage with respect to DIC in the Hooghly, other studies are severely limited in spatial coverage with focus on mid to lower part of the Hooghly estuary and a few locations in the Sundarbans (one location by Ray et al., 2015, 2018; three locations by Akhand et al., 2016). Given the vast expanse of these estuaries, extrapolation of data from these studies for the entire ecosystem may lead to overestimation/underestimation.

During the present study, we focused on understanding differences in varied aspects of C cycle (particulate organic, dissolved inorganic and organic along with gaseous form) of the Hooghly and Sundarbans during postmonsoon with relatively better spatial coverage compared to previous studies. The postmonsoon sampling was chosen because of relatively stable estuarine condition for wider spatial coverage and peak mangrove leaf litter fall during this season (Ray et al., 2011), which may have influence on estuarine C dynamics. Considering different nature and quantity of supplied OM within these two contrasting system, we hypothesize C metabolism between these two estuaries to be very different with higher CO_2 exchange flux from anthropogenically influenced estuary compared to mangrove-dominated estuary. Specifically, the major aims of the present study were to: (a) investigate factors controlling DIC and DOC dynamics in the region, (b) sources of POM in these two contrasting systems, and (c) partial pressure of CO_2 (pCO_2) and its controlling mechanisms along with exchange across water-atmosphere interface at the Hooghly-Sundarbans during postmonsoon period.

2 Materials and methods

2.1 Study area

The present study was carried out in mangrove dominated estuaries of Indian Sundarbans and anthropogenically dominated Hooghly estuary in the northeastern India. Sundarbans (21°32' and 22°40'N: 88°05' and 89°E), inscribed as a UNESCO world heritage site, is the largest mangrove forest in the world situated at the land-ocean boundary of the Ganges - Brahmaputra delta and the Bay of Bengal (BOB). Out of 10,200 km^2 area of Sundarbans, 41% is in India and the rest is in Bangladesh. The Indian part of Sundarbans (or Sundarbans Biosphere Reserve) contains 4200 km^2 of mangrove reserve forest and 1800 km^2 of estuarine waterways along with reclaimed areas. The Sundarbans is crisscrossed by several rivers, such as Muriganga, Saptamukhi, Thakuran, Matla, Bidya, Gosaba and Haribhanga, forming a sprawling archipelago of 102 islands covered with thick mangroves mostly composed of *Avicennia alba*, *Avicennia marina* and *Avicennia officinalis*. ~~The present study was carried out in three major estuaries of Indian Sundarbans (Saptamukhi, Thakuran and Matla; Fig. 1a) with no perennial source of freshwater and limited supply of anthropogenic inputs only during monsoon (Dutta et al., 2015a~~Semidiurnal tide with mean depth ~ 6 m is general characteristic of the estuary (Dutta et al., 2015).

The second study site, the Hooghly estuary (21°31′-23°20′N and 87°45′- 88°45′E~~)~~), is the first deltaic offshoot of the Ganges which ultimately mixes with the northern BOB. Like estuaries of Sundarbans, tides are semidiurnal in nature in the Hooghly as well with variable depth along the channel (~ 21 m at Diamond Harbor (H6) to ~ 8 m at the mouth of the estuary; Fig 1b) (CIFRI, 2012). Before mixing with the BOB, the lower estuarine part of the Hooghly divides into two channels, one being main estuarine stream which directly mixes with the BOB and another smaller channel known as Muriganga (mean depth ~ 6 m; Sadhuram et al., 2005). The width of the river at the mouth of the estuary is ~ 25 km (Mukhopadhyay et al., 2006). . The sampling locations in the Hooghly estuary are shown in Fig.1b. Both estuarine systems experience typical tropical climate having three distinct seasons: premonsoon (February - May), monsoon (June - September) and postmonsoon (October - January) with ~ 80% rainfall during monsoon.

Covering upper, middle, and lower estuarine regions, the present study was carried out during low tide condition in three major estuaries of the Indian Sundarbans [Saptamukhi (S1-S3), Thakuran (T1-T3), and Matla (M1-M3); Fig. 1a] along with its related waterways (S4 & M4). The low-tide postmonsoon sampling was preferred as it was ideal time to evaluate the effect of mangroves on the adjoining estuary due to peak mangrove litter fall (Ray et al., 2011) and groundwater (or pore-water) discharge. To compare and bring out the contrast in different components of the C cycle between mangrove-dominated and anthropogenically influenced estuaries, low-tide sampling was also performed at 13 locations (H1 – H13, Fig. 1b) in the Hooghly estuary (stretch: ~150km).

For the purpose of discussion, henceforth, both the estuarine systems will be discussed as 'Hooghly-Sundarbans system' and the estuaries of Sundarbans will be called 'Sundarbans' unless discussed individually.

2.2 Sampling and experimental techniques

During post-monsoonpostmonsoon (November, 2016), estuarine surface water samples were collected in duplicate at different locations of the Hooghly-Sundarbans system using Niskin bottle (Oceantest equipment; capacity: 5L). A brief description of the on and off field sampling and experimental techniques used during the present study are described below.

2.2.1 Sample collection and on board measurements

Water temperature and pH of the collected samples were measured onboard using thermometer ($\pm 0.1^oC$) and portable pH meter (Orion Star A211) fitted with a Ross type combination electrode calibrated (as described by Frankignoulle and Borges, 2001) on the NBS scale (reproducibility: ±0.005 pH units). Salinity (±0.1) and dissolved oxygen (DO: $\pm 0.1 mgL^{-1}$) concentrations were measured onboard following the Mohr-Knudsen and

Winkler titration methods, respectively (Grasshoff et al., 1983). For total alkalinity (TAlk), 50 ml of filtered (0.7 μm filtersWhatman GF/F filter) estuarine water was titrated onboard in a closed cell using 0.1N HCl following potentiometric titration method (Bouillon et al., 2003). Salinity and dissolved oxygen (DO) concentrations were measured onboard following the Mohr-Knudsen and Winkler titration methods, respectively (Grasshoff et al., 1983).Uncertainty in TAlk measurements was ±1 μmolkg^{-1} as estimated using certified reference material (Dickson standard: CRM-131-0215).

For DIC concentrations ([DIC]) and $\delta^{13}C_{DIC}$ measurements, estuarine surface waters and groundwater samples from nearby regions were collected by gently overfilling glass vials fitted with teflon septa followed by addition of . Pore-water was also collected from lower littoral zone of the Lothian Island (one of the virgin island of the Indian Sundarbans) by digging a hole (~30 cm below the water table). It was not possible to collect pore-water samples from mid and upper littoral zones due to logistic problems. After purging water at least twice in the bore, sample was collected from the bottom of the bore through syringe and transferred to the glass vial (Maher et al., 2013). Twelve groundwater samples were collected from the nearby locations of the Hooghly-Sundarbans system via tube pump. After collection, all samples for DIC and $\delta^{13}C_{DIC}$ were preserved immediately by adding saturated $HgCl_2$ solution to arrest the microbial activity. Pore-water sample from Lothian Island (one of the virgin island of Sundarbans) was also collected for [DIC] and $\delta^{13}C_{DIC}$

For both DOC and SPM (suspended particulate matter) measurements. For DOC concentration ([DOC]) measurement, estuarine, surface water samples were filtered *in situ*on board through pre-weighted and pre-combusted (500°C for 6 hours) Whatman GF/F filters followed by(pore size: 0.7μm). Filtrate was kept for DOC analysis in brown bottles followed by immediate preservation by adding via addition of H_3PO_4 (50μL/15 mL sample) (Bouillon et al., 2003). For suspended), whereas the residue was kept for particulate matter (SPM), water samples were filtered onboard through pre-weighted and pre-combusted Whatman GF/F filtersanalysis. Collected DIC, DOC and SPM samples were properly preserved at 4°C4 °C during transportation to the laboratory. Additionally, micrometeorological parameters associated with water-atmosphere CO_2 exchange flux computation were continuously monitored at 10 m height over the estuary using a portable weather monitor (DAVIS - Vintage Pro2 Plus).

__2.2.2 Laboratory techniquesmeasurements__

The DIC concentrations were measured using Coulometer (Model: UIC. Inc. CM –– 5130; () with analytical uncertainty: of ±0.8%) while%. The $\delta^{13}C_{DIC}$ were analyzedmeasured using Gas Bench attached to a continuous flow mass spectrometer (Thermo Delta V). Values of $\delta^{13}C_{DIC}$ are reported with respect to V-PDB) with reproducibilityprecision better than ± 0.10‰. The DOC concentrations were measured using high-temperature catalytic oxidation analyzer (Shimadzu TOC 5000) and variability in [DOC] for duplicate measurements), which was around ± 52 μgcalibrated using potassium hydrogen phthalate (KHP) solution containing 1, 2, 5, 10, 20 mg L^{-1}.$^{-1}$ of DOC (Ray et al., 2018). The analytical error for DOC measurement was < 2%. For SPM concentrationsmeasurement, filter papers containing SPM were dried in hot air oven at 60°C and final weights were noted. The SPM concentrations were calculated based on difference between final and initial weights of the filter paper and volume of water filtered. For measurement of particulate organic carbon concentrations ([POC]) and its isotopic composition ($\delta^{13}C_{POC}$), a section of, SPM containing filter papers were de-carbonated (by HCl fumes) and analyzed using elementalElemental Analyzer attached to the continuous flow mass spectrometer via conflo. The $\delta^{13}C_{POC}$ values are reported relative to V-PDB with reproducibility better than ± 0.10‰.‰, whereas uncertainty for POC was <10%.

__2.2.3 Computation of %DO, CO_2-system and air - water CO_2 flux calculationand %DO__

The pCO_2 was calculated based on surface water temperature, salinity, TAlk, pH and dissociation constants calculated following Millero (2013). The uncertainty for estimated pCO_2 was ± 1%. The %DO and apparent oxygen utilization (AOU) were calculated as follows: %DO = ($[O_2]_{Measured}$ x 100 / $[O_2]_{Equilibrium}$) and AOU = ($[O_2]_{Measured}$ – $[O_2]_{Equilibrium}$); where, $[O_2]_{Equilibrium}$ is the equilibrium DO concentration calculated at *in-situ* temperature and salinity (Weiss, 1970) and $[O_2]_{Measured}$ is the measured DO concentration of estuarine water. pCO_2 (uncertainty: ± 1%) and other associated parameters of the estuarine CO_2 system were calculated by using equations as given by Millero (2013).

CO_2 exchange fluxes (FCO_2 in μmol m^{-2} hr^{-1}) across water-atmosphere boundary of the estuary were calculated as follows:

$$FCO_2 = k \times K_H^{CO2} \times [pCO_{2\,(water)} - pCO_{2\,(atmosphere)}]; where)]$$

Where, K_H^{CO2} = CO_2 solubility. 'k' representsis gas transfer velocity, which is highly variable and remains a matter of debate (Raymond and Cole, 2001). -The 'k' during the present study was computed as a function of wind velocity (following Liss and Merlivat, – (1986). The computation of 'k' based only on wind velocity are known to vary geographically due to variable impacts of fetch limitation and tidal currents (Borges) parametrization. et al., 2004). For the same wind velocity, the parametrization of Liss and Merlivat, (1986) probably provides least 'k' value over other parametrization (Wanninkhof, 1992; Raymond and Cole, 2001; Borges et al., 2004) and therefore, the FCO_2 presented during this study aremay be considered as the conservative estimates. The wind velocity based 'k' estimation for the Hooghly-Sundarbans system has been applied in earlier studies as well (Mukhopadhyay et al., 2002, Biswas et al., 2004). Mean global atmospheric CO_2 mixing ratio in dry air during 2016 (data source: ftp://aftp.cmdl.noaa.gov/products/trends/co2/co2_annmean_gl.txtftp://aftp.cmdl.noaa.gov/products/trends/co2/co2_annmean_gl.txt) was corrected for water vapor partial pressure to calculate $pCO_{2(atmosphere)}$. The fraction, "K_H^{CO2} x [$pCO_{2\ (water)}$ – $pCO_{2\ (atmosphere)}$]" is the departure of free dissolved CO_2 from atmospheric equilibrium that may be termed as "excess CO_2 (ECO_2)" (Zhai et al., 2005).

%DO and apparent oxygen utilization (AOU) were calculated as follows:

$$\%DO = ([O_2]_{Measured} \times 100 / [O_2]_{Equilibrium})$$

$$AOU = ([O_2]_{Measured} - [O_2]_{Equilibrium})$$

Where, $[O_2]_{Equilibrium}$ is the equilibrium DO concentration calculated at *in-situ* temperature and salinity (Weiss, 1970) and $[O_2]_{Measured}$ is the measured DO concentration of surface water.

2.2.4 Mixing model calculation

Considering salinity as a conservative tracer and an ideal indicator for estuarine mixing mechanism (Fry, 2002), conservative mixing model was applied to the Hooghly estuary to understand addition/removal of dissolved and particulate C by *in situ* biogeochemical processes. Concentrations and stable isotopic compositions of dissolved or particulate C (presented as C) during conservative mixing (C_{CM} and $\delta^{13}C_{CM}$) were computed as follows (Carpenter et al., 1975, Mook and Tan, 1991):

$$C_{CM} = C_F F_F + C_M F_M$$

$$S_S [C_F \delta^{13}C_F - C_M \delta^{13}C_M] + S_F C_M \delta^{13}C_M - S_M C_F \delta^{13}C_F$$

$$\delta^{13}C_{CM} = \frac{S_S (C_F \delta^{13}C_F - C_M \delta^{13}C_M) + S_F C_M \delta^{13}C_M - S_M C_F \delta^{13}C_F}{S_S (C_F - C_M) + S_F C_M - S_M C_F}$$

Here, 'S' denotes salinity, the suffixes CM, F, M and S denote conservative mixing, freshwater end member, marine end member and sample, respectively. F_F = freshwater fraction = $1 - (S_S / S_M)$ and F_M = marine water fraction = $(1 - F_F)$. $C_{Sample} > C_{CM}$ indicates C addition, whereas reverse indicates removal. For model calculation, mean salinity, concentrations of C and $\delta^{13}C$ of samples collected at salinity ≤ 0.3 at the Hooghly estuary were considered as end member values for freshwater, whereas respective values for marine end member were taken from Dutta et al. (2010) and Akhand et al. (2012). Quantitative deviations (ΔC and $\Delta\delta^{13}C$) of measured C concentrations and $\delta^{13}C$ from the respective conservative mixing values were estimated as follows (Alling et al., 2012):

$$\Delta C = (C_{Sample} - C_{CM}) / C_{CM}$$

$$\Delta\delta^{13}C = \delta^{13}C_{Sample} - \delta^{13}C_{CM}$$

Plots between ΔC and $\Delta\delta^{13}C$ for DIC and POC have been used to understand processes influencing DIC and POC in the Hooghly-Sundarbans system. However, the above model could not be applied to DOC due to unavailability of $\delta^{13}C_{DOC}$ during the present study.

Unlike Hooghly, direct application of above-mentioned conservative mixing model was not justified for mangrove-dominated Sundarbans due to narrow salinity gradient (see later). However, assuming that apart from conservative mixing only mangrove derived C ($\Delta C_{Mangrove}$) contributes to estuarine C pool, an approach can be taken to quantify $\Delta C_{Mangrove}$. Two different mass balance equations as used by Miyajima et al. (2009) for estimating $\Delta DIC_{Mangrove}$ was extended to calculate $\Delta C_{Mangrove}$ during the present study:

$$\Delta C_{Mangrove} (\Delta C_{M1}) = C_{Sample} - C_{CM}$$

$$\Delta C_{Mangrove} (\Delta C_{M2}) = \frac{C_{Sample} \times [\delta^{13}C_{CM} - \delta^{13}C_{Sample}]}{\delta^{13}C_{CM} - \delta^{13}C_{Mangrove}}$$

For model calculation, $\delta^{13}C_{Mangrove}$ was taken as – 28.4‰ for Sundarbans (Ray et al., 2015) and end members were taken as same as the Hooghly as estuaries of Sundarbans are offshoot of lower Hooghly estuary.

3 Results

3.1 Environmental parameters

During the present study, water temperature did not show any distinct spatial trend and varied from 28 - ~~29 °C~~29°C and 30.5 - ~~33 °C~~33°C for the Sundarbans (Table ~~1~~2) and Hooghly (Table ~~2~~3), respectively. Salinity of the estuaries of Sundarbans varied over a narrow range (12.74 - 16.69; Table ~~1~~2) with minimum at the upper estuarine location throughout. A relatively sharp salinity gradient was noticed at the Hooghly estuary (0.04 - 10.37; Table ~~2~~3). Surface water DO concentrations were marginally higher in the Sundarbans (6.46 - 7.46 mgL^{-1}) than the Hooghly (5.24-7.40 mgL^{-1}). Both pH and TAlk in the Hooghly estuary (pH: 7.31 to 8.29, TAlk: ~~1.80~~1797 to ~~2.86 meqL~~2862 μeqL^{-1}) showed relatively wider variation compared to the estuaries of Sundarbans (pH: 8.01 to 8.13, TAlk: ~~2.01~~2009 to ~~2.29 meqL~~2289 μeqL^{-1}; Table ~~1 &~~ 2 & 3).

3.2 Variability in DIC, $\delta^{13}C_{DIC}$ and DOC

In the Sundarbans, both ~~[~~DIC~~]~~ and $\delta^{13}C_{DIC}$ varied over a relatively narrow range ~~([~~(DIC~~] = 1.68~~ = 1683 to ~~1.92 mM~~1920 µM, mean: ~~1.76 ± 0.07 mM~~1756 ± 73 µM; $\delta^{13}C_{DIC}$ = – 5.93 to – 4.29‰, mean: – 5.04 ± 0.58‰) compared to the Hooghly estuary ~~([~~(DIC~~] = 1.79~~ = 1678 to ~~2.70 mM~~2700 µM, mean: ~~2.08 ± 0.32 mM~~2083 ± 320 µM; $\delta^{13}C_{DIC}$ = – 8.61 to – 5.57‰, mean: – 6.95 ± 0.90‰; Table ~~1 & 2). The present [DIC] and $\delta^{13}C_{DIC}$ values for the mangrove dominated estuaries of the Indian Sundarbans were in the range of that reported for the mangrove surrounding Khura and Trang rivers ([DIC]: ~ 0.25 – 2.25 mM, $\delta^{13}C_{DIC}$: ~ 0 to – 20‰; Miyajima et al., 2009) in peninsular Thailand. The values for the Hooghly estuary were comparable with previously reported values by Samanta et al. (2015).~~2 & 3). Spatially, in the Hooghly, maximum ~~[~~DIC~~]~~ and $\delta^{13}C_{DIC}$ was noticed at freshwater (H1~~-~~ - H6) and mixing (H7~~-~~ - H13) zones, respectively. Different estuaries of the Sundarbans showed different trends with Saptamukhi and Thakuran showing maximum and minimum ~~[~~DIC~~]~~ at the upper and lower estuarine regions, respectively with reverse trend for $\delta^{13}C_{DIC}$. However, for the Matla, no distinct spatial trend was noticed for both ~~[DIC] and $\delta^{13}C_{DIC}$~~DIC and $\delta^{13}C_{DIC}$. In comparison to the estuarine surface waters, markedly higher DIC and depleted $\delta^{13}C_{DIC}$ were observed for the groundwater (Hooghly: DIC = 5655 to 11756 µM, $\delta^{13}C_{DIC}$ = – 12.66 to – 6.67‰; Sundarbans: DIC = 7524 to 13599 µM, $\delta^{13}C_{DIC}$ = – 10.56 to – 6.69‰; Table 4) and pore-water samples (Sundarbans: DIC = 13425 µM; $\delta^{13}C_{DIC}$ = – 18.05‰; Table 4) collected from the Hooghly-Sundarbans system. The DOC in the Sundarbans varied from 154 to 315 µM (mean: 235 ± 49 µM; Table 2) with no distinct spatial variability. In comparison, ~ 40% higher DOC was noticed in the Hooghly (235 - 662 µM; Table 3) reaching peak in the mixing zone.

~~In comparison to the estuarine surface waters, markedly higher [DIC] and depleted $\delta^{13}C_{DIC}$ were observed for the groundwater (Hooghly: [DIC] = 5.66 to 11.76 mM, $\delta^{13}C_{DIC}$ = –12.66 to –6.67‰; Sundarbans: [DIC] = 7.52 to 13.59 mM, $\delta^{13}C_{DIC}$ = –10.56 to –6.69‰; Table 3) and pore water samples (Sundarbans: [DIC] = 13.43 mM; $\delta^{13}C_{DIC}$ = –18.05‰; Table 3) collected from the Hooghly-Sundarbans system.~~

~~The [DOC] in the Sundarbans varied from 1.85 to 3.78 mgL^{-1} (mean: 2.83 ± 0.59 mgL^{-1}; Table 1) with no distinct spatial variability. In comparison, ~ 40% higher [DOC] was noticed in the Hooghly (2.82 – 7.95 mgL^{-1}; Table 2) reaching peak in the mixing zone. The [DOC] measured in the Hooghly-Sundarbans system were in the range of that reported for the Godavari estuary, South India (~1 – 8.50 mgL^{-1}; Bouillon et al., 2003) but higher than that reported for the Pearl river estuary, China (~ 0.72 – 1.92 mgL^{-1}; Callahan et al., 2004).~~

3.3 Variability in particulate matter and $\delta^{13}C_{POC}$

In the Sundarbans, both SPM and ~~[~~POC~~]~~ varied over a wide range (SPM = 80 to 741 mgL^{-1}, mean: 241 ± 197 mgL^{-1}; ~~[~~POC~~]~~ = 80 to 436 µM, mean: 173 ± 111 µM; Table ~~1~~2) with no distinct spatial variability. Compared to that, SPM and ~~[~~POC~~]~~ in the Hooghly were relatively lower and varied from 38 - 289 mgL^{-1} and 95 - 313 µM (Table ~~2~~3), respectively; reaching maximum at the freshwater zone. The $\delta^{13}C_{POC}$ of the Sundarbans varied from –23.82 to –22.85‰ (mean: –23.36 ± 0.32~~‰); $\delta^{13}C_{POC}$ of~~ ‰), whereas in the Hooghly~~, however, was relatively depleted in ^{13}C (–25.95~~ it varied from –26.28 to –24.~~07‰,~~ 06 (mean: –24.87 ± 0.89‰). ~~The observed $\delta^{13}C_{POC}$ of the Sundarbans were within the range of that reported for mangrove dominated Godavari estuary, South India ($\delta^{13}C_{POC}$: ~ –19 to –29‰, Bouillon et al., 2003) and Khura and Trang rivers, Thailand ($\delta^{13}C_{POC}$ ~ –21 to –33‰; Miyajima et al., 2009). For the Hooghly, the observed $\delta^{13}C_{POC}$ were comparable with that previously reported by Samanta et al. (2015).~~

3.4 Variability in pCO_2 and FCO_2

In the Sundarbans, surface water pCO_2 varied from 376 to ~~561 µatm~~561µatm (mean: 464 ± ~~66 µatm~~66µatm; Table ~~1~~2) with no spatial pattern. Compared to the Sundarbans, ~ 3.8 times higher pCO_2 was estimated in the Hooghly estuary (267 ~~– 4678 µatm; Table 2) reaching its~~

peak in the freshwater region. The estimated pCO_2 for the Hooghly-Sundarbans system were in the range of that previously reported for other tidal estuaries of the Indian subcontinent (Cochin estuary: 150-3800 µatm, Gupta et al., 2009; Mandovi – Zuari estuary: 500-3500 µatm, Sarma et al., 2001) and other tropical countries (Changjiang estuary, China: 200-4600 µatm, Zhai et al., 2007).

- 4678µatm; Table 3) reaching its peak in the freshwater region. Except one location at the Sundarbans (M2: – 42 µM) and two mixing zone locations at the Hooghly (H12: –3.26 µM; H13: –3.43 µM), ECO_2 values were always positive atin the Hooghly-Sundarbans system. The calculated FCO_2 at the Hooghly estuary (–19.38 to 717.5 µmolm^{-2}hr^{-1}; mean: 231 µmolm^{-2}hr^{-1}; Table 23) was ~17 times higher than the mangrove dominated estuaries of the Indian Sundarbans (–2.6 to 30.3 µmolm^{-2}hr^{-1}; Table 12). Spatially, in the Hooghly, higher FCO_2 was noticed at the freshwater region (285.2 to 717.5 µmolm^{-2}hr^{-1}), while no such distinct spatial trend was noticed at the Sundarbans.

4. Discussion

4.1 Environmental parameters of the Hooghly-Sundarbans system

Based on the observed salinity gradient, the Hooghly estuary can be divided into two major salinity regimes: (a) fresh-water zone (H1-H6) and (b) mixing zone (H7 – H13; Fig.1b). Due to narrow salinity range, no such classification was possible for the estuaries of Sundarbans. % DO calculations showed relatively well-oxygenated estuarine environment in the Sundarbans (91 - 104%) compared to the Hooghly (71 - 104%; Fig.2a). Salinity independent variation in pH was noticed for both the estuarine systems (p = 0.14 and 0.07 for the Sundarbans and Hooghly, respectively; Fig.2b). The pH range for this tropical estuarine system clearly indicates the dominance of [HCO_3^{2-}] over [CO_3^{2-}] in both the Hooghly (~ 8.0 - 219.4 times) and Sundarbans (~ 9.7 - 13.6 times). 2). Based on the results obtained during the present study, below we discuss different components of C cycle within Hooghly-Sundarbans system.

4.2 Dissolved inorganic carbon1 Major drivers of DIC dynamics

In the Hooghly, both [DIC] DIC concentrations during the present study were relatively higher compared to that reported by Samanta et al. (2015) for the same season, whereas $\delta^{13}C_{DIC}$ values were within the same range (DIC: 1700 - 2250µM; $\delta^{13}C_{DIC}$: –11.4 to – 4.0‰). Statistically significant correlations between DIC - salinity ($R^2$$r^2$ = 0.43, p = 0.015) and

$\delta^{13}C_{DIC}$ – salinity (R^2 r^2 = 0.58, p = 0.003) in the Hooghly suggested potential influence of marine and freshwater mixing on DIC and $\delta^{13}C_{DIC}$ in the estuary (Fig. 3a & 3b). The above-mentioned significant relationships were statistically significant (Fig. 3a), making it an ideal site for during the present study coupled with earlier $\delta^{18}O$ - salinity (Ghosh et al., 2013) and DIC dynamics (Samanta et al., 2015) studies in the Hooghly rationalize application of two end member mixing model (Ghosh et al., 2013, Samanta et al., 2015). For model calculation, average salinity, [DIC] and $\delta^{13}C_{DIC}$ of samples collected at ≤ 0.3 salinity during the present study were considered as values in this estuary to decipher *in situ* processes influencing DIC chemistry.

Based on the methodology discussed earlier, calculated ΔC for freshwater end member, whereas respectiveDIC (ΔDIC ~ – 0.27 to 0.17) predicted dominance of DIC addition (n = 4) over removal (n = 2) in the freshwater region of the Hooghly, whereas only removal was evident in the mixing zone. In case of $\Delta\delta^{13}C$ for DIC ($\Delta\delta^{13}C_{DIC}$), values for marine end member were taken from Dutta et al. (2010) and Akhand et al. (2012). The [DIC] and $\delta^{13}C_{DIC}$ undermostly positive (n = 9), i.e., measured $\delta^{13}C_{DIC}$ was higher compared to estimated $\delta^{13}C_{DIC}$ due to conservative mixing condition and deviations (ΔDIC and $\Delta\delta^{13}C_{DIC}$) between observed and respective conservative mixing values were computed using Alling et al. (2012) to explore the role of *in situ* biogeochemical processes in modulating estuarine DIC dynamics.

. Deviation plot (ΔDIC vs. $\Delta\delta^{13}C_{DIC}$; Fig.3b3c) for samples of the Hooghly showsshowed following patterns: (a) decrease in ΔDIC with increasing $\Delta\delta^{13}C_{DIC}$ (n = 5) indicating phytoplankton productivity and/or outgassing of CO_2 (PP/CO_2 OG) from water-atmosphere interface, (b) decrease in ΔDIC with decreasing $\Delta\delta^{13}C_{DIC}$ (n = 4) indicating calcitecarbonate precipitation, (CP), and [(c]) increase of ΔDIC with increasing $\Delta\delta^{13}C_{DIC}$ (n = 4) representing calcitecarbonate dissolution (CD) within the system.

Based on these calculations, both organic and inorganic C metabolismsprocesses (productivity, $CaCO_3$carbonate precipitation and dissolution) along with physical processes (CO_2 outgassing across water-atmosphere interface) appearappeared to regulate the DIC chemistry in the Hooghly estuary. Spatially, productivity/PP and CO_2 outgassing appears to be dominant process OG appeared to regulate DIC in the mixing zone as most of the samples ((n = 5 out of 7) from this zone fallof the Hooghly. Earlier studies have advocated high phytoplankton productivity in this quadrant, whereas $CaCO_3$ precipitation and/or dissolution are dominantnon-limiting nutrient condition during postmonsoon in the freshwater zone (Fig.

3b). Further, 'ΔTAlk/ΔDIC' can be used as a proxyHooghly (Mukhopadhyay et al., 2002; Mukhopadhyay et al., 2006). However, based on the present data, particularly due to evaluate relative importancelack of biological productivitydirect PP measurements, it was difficult to spatially decouple PP and CO_2 outgassing in the system. For primary productivity ($106CO_2$ + $122H_2O$ + $16HNO_3$ + H_3PO_4 → $(CH_2O)_{106}(NH_3)_{16}H_3PO_4$ + $138O_2$), theoretical ΔTAlk/ΔDIC is around – 0.16 (ΔTAlk = – 17 and ΔDIC = 106, Cao et al., 2011), whereas the same is 0 for CO_2 outgassing as it affects DIC withoutmixing zone. In contrast to the mixing zone, CP and CD appeared to be dominant processes affecting TAlk (Guo et al., 2008). The ΔTAlk/ΔDIC value for the sampling points located in the productivity/CO_2 outgassing quadrant is – 0.17, close to theoretically calculated value for primary productivity. This suggest that primary productivity is the central process regulatingestuarine DIC chemistry in the mixing zonefreshwater region of the Hooghly estuary.

In the mangrove-dominated estuaries of Sundarbans, [our measured $\delta^{13}C_{DIC}$ values were within the range of that reported by Ray et al. (2018), whereas DIC] concentrations were comparatively lower (DIC: 2130 ± 100 μmolkg^{-1}, $\delta^{13}C_{DIC}$: – 4.7 ± 0.7‰). Our data also showed similarity with Khura and Trang river, two mangrove-dominated rivers of peninsular Thailand flowing towards Andaman sea, although from hydrological prospective these two systems are contrasting in nature [Sundarbans: narrow salinity gradient (12.74 - 16.69) vs. Khura and Trang river: sharp salinity gradient (~ 0 – 35); Miyajima et al., 2009]. Like Hooghly, $\delta^{13}C_{DIC}$ - salinity relationship was notstatistically significant (p = 0.18), whereas $\delta^{13}C_{DIC}$ – salinity was found to be significant ($R^2$$r^2$ = 0.55, p = 0.009; Fig. 3c) as observed at other mangrove dominated systems as well (Miyajima et al., 2009). Unlike Hooghly, the narrow salinity gradient limits the application of two end member mixing model) for the Sundarbans to point out individual influencing biogeochemical factors on DIC. However, but DIC - salinity relationship remained insignificant (p = 0.18) (Fig. 3d & 3e).

Given the dominance of mangrove in the Sundarbans, the role of mangrove derived OC mineralization becomesmay be important in regulating DIC chemistry in ecosystems like the Sundarbans. Two different mass balance equations as proposed by Miyajima et al. (2009) have been adopted to quantify mangrove derived this ecosystem. Theoretically, $\Delta C_{Mangrove}$ for DIC ($\Delta DIC_{Mangrove}$) in the Sundarbans:

$$\Delta DIC_{Mangrove} (\Delta DIC_{M1}) = [DIC] – [DIC_{CM}]$$

$$[DIC] \times [\delta^{13}C_{DIC(CM)} – \delta^{13}C_{DIC}]$$

$$\Delta DIC_{Mangrove} (\Delta DIC_{M2}) = \text{———————————}$$

$\delta^{13}C_{DIC(CM)}$ – $\delta^{13}C_{Mangrove}$ (= –27‰)

Where, CM indicates conservative mixing. Since both Sundarbans and Hooghly estuarine system have same marine end member (BOB) and the Sundarbans are connected to the Hooghly estuary through different branches, similar end member values as Hooghly were used for this calculation as well. Theoretically, $\Delta DIC_{Mangrove}$ estimated based on [DIC] (ΔDIC_{M1}) and $\delta^{13}C_{DIC}$ (ΔDIC_{M2}) should be equal. The negative and unequal values of ΔDIC_{M2} (– 4441 to 6662 µM) and ΔDIC_{M1} (–188186 to 11 µM) indicate large DIC out-flux over influx through mangrove derived OC mineralization in this tropical mangrove system. The removal mechanisms of DIC include CO_2 outgassing across estuarine water-atmosphere boundary (see section 4.5),, phytoplankton uptake and export to adjacent continental shelf region (northern BOB, Ray et al., 2018). The evidence for CO_2 outgassing was found at almost all locations covered during the present study (10 out of 11 locations covered; see section 4.4). Also, a recent study by Ray et al. (2018) estimated DIC export (~ 3.69Tg C yr^{-1}) from the estuaries of Sundarbans as dominant form of C export. Although data for primary productivity is not available for the study period, earlier studies have reported postmonsoon as peak season for phytoplankton productivity (Biswas et al., 2007; Dutta et al., 2018).2015). Given the evidences for presence of DIC removal processes in the Sundarbans, a comprehensive study focused on rate measurements of these processes with higher spatial and temporal coverage is desirable to understand the balance between influx and out-flux of DIC in the Sundarbans.

Other than biogeochemical processes, factors such as groundwater and pore-water exchange to the estuary might also play significant role in estuarine DIC chemistry (Tait et al., 2016). High pCO_2, and DIC andalong with low pH, and TAlk/DIC are general characteristiccharacteristics of groundwater, specially within carbonate aquifer region (Cai et al., 2003). Although all the parameters of ground watergroundwater inorganic C system (like pH, TAlk and pCO_2) were not measured during the present study, groundwater [DIC] were ~5.57 and ~3.61 times higher compared to averagemean surface water [DIC] in the Sundarbans and Hooghly, respectively. The markedly higher [DIC] in groundwater as well as similarity in its isotopic composition with estuarine DIC (Table 3) may stand as a signal for influence of groundwater on estuarine DIC biogeochemistry, with possibly higher influence at the Hooghly rather than Sundarbans than Hooghly as evident from the slope of the TAlk/ - DIC valuerelationships (Hooghly: 0.87-1.1498, Sundarbans: 1.12-1.34; Fig.3d). However, unavailability of any data on 0.03). In the Sundarbans, to the best of our knowledge, no report exists regarding groundwater discharge rate from these systems limits us to quantitatively evaluate groundwater mediated DIC flux to the estuary.. Contradictory reports exist for the Hooghly, where Samanta et al. (2015) indicated groundwater contribution at low salinity regime (salinity < 10, same as our salinity range) based on 'Ca' measurement, which was not observed based on 'Ra' isotope measurement in an earlier study (Somayajulu et al., 2002). Pore-water [DIC] in the Sundarbans was ~7.63 times higher than the estuarine water, indicating possibility of DIC input from the adjoining mangrove system to the estuary through pore-water exchange depending upon changes in hypsometric gradient during tidal fluctuation. Although pore water [DIC] was estimated at only one location, considering postmonsoonA first-time baseline value for advective DIC influx from mangrove sediment to the estuary (F_{DIC}) via pore-water exchange was estimated during the present study using the following expression (Reay et al., 1995):

$$F_{DIC} = \text{Sediment porosity x Mean linear velocity x Mean pore water DIC conc.}$$

$$\text{Mean linear velocity} = \text{Pore water specific discharge / Sediment porosity}$$

Using pore-water specific discharge and porosity as 0.008 cm min^{-1} and 0.58 (Dutta et al., 2013, Dutta et al., 2015a2015), respectively, a first-time baseline during postmonsoon and extrapolating the flux value over daily basis (i.e., for advective DIC influx from mangrove sediment to the estuary can be estimated12 hours as ~ 774tides are semidiurnal in nature), mean F_{DIC} during postmonsoon was calculated as ~ 770.4 mmol m^{-2} d^{-1} using Reay et al. (1995).. However, significant impact of pore-water to estuarine DIC may be limited only in mangrove creek water (samples not collected) as evident from narrow variability of estuarine TAlk and DIC as well as no significant correlation between them (Fig. 3dp = 0.93). A comprehensive investigation on ground and pore waters are needed to thoroughly understand their importance in controlling DIC chemistry of the Hooghly-Sundarbans system.

4.3 Dissolved organic carbon dynamics

In estuarine ecosystems, sources of DOC include terrestrial or lateral inputs, *in situ* production by benthic and pelagic primary producers, bacteria, ciliates, flagellates as well as release from zoo-plankton faeces and dead organisms (Wangersky, 1978). During the present study, no significant correlation was found between [DOC] and salinity (Sundarbans: p = 0.10; Hooghly estuary: p = 0.30; Fig.3e) indicating its non-conservative behavior in the Hooghly-Sundarbans system. Similar non-conservative behavior of DOC has been observed in other estuaries of the Indian Subcontinent (Bouillon et al., 2003) with opposite reports from elsewhere as well (Raymond and Bauer, 2001a, Abril et al., 2002). In the Hooghly-Sundarbans system, [DIC] – [DOC] correlation was not significant (Sundarbans: $p = 0.29$, Hooghly: $p = 0.16$) suggesting limited role of phytoplankton production on the estuarine DOC level. In contrast to the Hooghly ($p = 0.56$), significant positive correlation between pCO_2 and [DOC] was observed in the Sundarbans ($p = 0.02$, $n = 11$) suggesting analogous sources of pCO_2 and DOC within the system, possibly through pore water exchange from adjacent mangroves to the estuary as reported from other mangrove systems worldwide (Cai et al., 1999, Ho et al., 2017).

DOC may be removed from system through mineralization by bacteria, oxidation by UV irradiation (photo-oxidation), conversion to POC by flocculation (Bouillon et al., 2006), or export. Considering equal effect of UV mediated DOC photo-oxidation at both estuarine systems, removal of DOC would be principally regulated by biogeochemical and physical processes. The [DOC]-[POC] correlation was found to be significant in the Hooghly ($p = 0.04$, $n = 12$) but not at the estuaries of Sundarbans, possibly indicating interconversion between POC and DOC (via dissolving and flocculation, respectively) to be a significant player in controlling DOC levels in the Hooghly. No evidence for significant DOC mineralization was found at the Hooghly – Sundarbans system based on [DOC] – [DO] (Sundarbans: $p = 0.85$, Hooghly: $p = 0.40$) as well as pCO_2-[DOC] relationships (described earlier). We do not have data to support export of DOC; however, a recent study quantified an annual export of 0.11-0.34 Tg C and 3.03 Tg C as DOC to the northern BOB from the Hooghly and Sundarbans, respectively (Ray et al., 2018).

4.4 Particulate organic matter in the Hooghly – Sundarbans system

No significant correlation was found between SPM concentrations and salinity for both the estuaries (Sundarbans: $p = 0.69$, Hooghly: $p = 0.40$; Fig. not shown). However, [POC] was negatively correlated with salinity in the Hooghly ($R^2 = 0.38$, $p = 0.026$; Fig.4a) but not at the Sundarbans (Fig. 4b), indicating freshwater run-off mediated addition of POC in the Hooghly estuary. Additionally, compared to other sampling locations relatively higher [POC] at 'H1', 'H3' and 'H4' at the Hooghly indicate contribution from nearby jute industry located on both sides of river bank at these locations. The POC formed relatively larger part of SPM in the Hooghly (0.96 – 4.22%; Fig.4a) compared to the Sundarbans (0.66 – 1.23%) (Fig.4b). The lower contribution of POC to the SPM pool in the mangrove dominated Sundarbans may be due to low primary production owing to high SPM load (Ittekkot and Laane, 1991) as observed at mangrove region of the Godavari estuary as well (Bouillon et al., 2003). Although direct measurement of primary productivity was not carried out during the present study, absence of significant correlation between pCO_2 – %DO indirectly points to that effect (Fig. not shown).

Wide range for $\delta^{13}C_{POC}$ From the above discussion it appears that on an average ~ 327 µM higher DIC in the Hooghly compared to the Sundarbans may be due to cumulative interaction between freshwater content to the individual estuaries as well as degree of biogeochemical and hydrological processes. Relatively higher freshwater contribution in the Hooghly compared to the Sundarbans (as evident from salinity) as well as significant negative relationship between DIC - salinity proved significant impact of freshwater on DIC pool in the Hooghly. However, detailed quantification of other biogeochemical and hydrological processes is needed to decipher dominant processes affecting DIC dynamics in the Hooghly-Sundarbans system.

4.2 DOC in the Hooghly-Sundarbans

During the present study, DOC concentrations in the Hooghly estuary were higher compared previously reported by Ray et al. (2018) (226.9 ± 26.2 to 324 ± 27µM), whereas DOC in the Sundarbans were comparable with Ray et al. (2018) (262.5 ± 48.2µM). The marine and freshwater mixing did not appear to exert major control over DOC in the Hooghly-Sundarbans system as evident from lack of significant correlations between DOC and salinity (Hooghly freshwater: $r^2 = 0.33$, $p = 0.23$; Hooghly mixing region: $r^2 = 0.10$, $p = 0.50$; Sundarbans: $r^2 = 0.27$, $p = 0.10$, Fig.4a). Our observations showed similarity with other Indian estuaries (Bouillon et al., 2003) with opposite reports from elsewhere (Raymond and Bauer, 2001a, Abril et al., 2002). This indicates that DOC in this sub-tropical estuarine system is principally controlled by processes other than mixing of two water masses.

Although it is difficult to accurately decipher processes influencing DOC without $\delta^{13}C_{DOC}$ data, some insights may be obtained from estimated ΔC of DOC (ΔDOC). The estimated ΔDOC in the Hooghly indicated both net addition (n = 3) and removal (n = 3) of DOC in the freshwater zone (ΔDOC = – 0.16 to 0.11); whereas, only net addition was evident throughout the mixing zone (ΔDOC = 0.08 to 1.74). In the Sundarbans, except lower Thakuran (St. T3, $\Delta\ DOC_{M1}$ = – 20µM), net addition of mangrove derived DOC was estimated throughout (ΔDOC_{M1} = 2 - 134µM).

In an estuary, DOC can be added through *in situ* production (by benthic and pelagic primary producers), lysis of halophobic freshwater phytoplankton cells and POC dissolution. DOC can be removed through bacterial mineralization, flocculation as POC, and photo-oxidation (Bouillon et al., 2006). At the Hooghly - Sundarbans system, no evidence for freshwater phytoplankton ($\delta^{13}C$: – 33 to – 40‰; Freitas et al., 2001) was found from $\delta^{13}C_{POC}$, ruling out its potential effect on DOC. Although an indirect signal for phytoplankton productivity was observed in the freshwater region from $\delta^{13}C_{DIC}$ and POC relationship (r^2 = 0.68, p = 0.05), further evaluation of its impact on DOC was not possible due to lack of direct primary productivity measurements. Contradictory results exist regarding influence of phytoplankton productivity on DOC. Some studies did not find direct link between DOC and primary productivity (Boto and Wellington, 1988), whereas primary productivity mediated significant DOC formation (~ 8 - 40%) has been reported by others (Dittmar & Lara 2001a, Kristensen & Suraswadi 2002).

The DOC - pCO_2 relationship suggested inefficient bacterial DOC mineralization in the Hooghly (freshwater zone: p = 0.69, mixing zone: p = 0.67, Fig. 4b). However, significant positive relationship between these two in the Sundarbans (r^2 = 0.45, p = 0.02, Fig. 4c) indicated increase in aerobic bacterial activity with increasing DOC. In mangrove ecosystems, leaching of mangrove leaf litter as DOC is fast as ~ 30% of mangrove leaf litter leaching as DOC is reported within initial 9 days of degradation (Camilleri and Ribi, 1986). In the Sundarbans, mangrove litter fall peaks during postmonsoon (Ray et al. 2011) and its subsequent significant leaching as DOC was evident during the present study from comparatively higher DOC compared to POC (DOC:POC = 0.50 - 3.39, mean: 1.79 ± 0.94%). Our interpretation for Sundarbans corroborated with that reported by Ray et al. (2018) for the same system as well as Bouillon et al. (2003) for the Godavari estuary, South India.

Despite high water residence time in the Hooghly (~ 40 days during postmonsoon, Samanta et al., 2015) and in mangrove ecosystem like Sundarbans (Alongi et al., 2005, Singh et al., 2016), DOC photo-oxidation may not be so potent due to unstable estuarine condition in the Hooghly-Sundarbans system (Richardson number < 0.14) with intensive vertical mixing and longitudinal dispersion coefficients of 784 $m^2 s^{-1}$ (Goutam et al., 2015, Sadhuram et al., 2005). The unstable condition may not favor DOC - POC interconversion as well but mediated by charged complexes and repulsion - attraction interactions, the interconversion partly depends upon variation in salinity. More specifically, the interconversion is efficient during initial mixing of fresh (river) and seawater and the coagulation is mostly complete within salinity range 2 - 3. This appeared to be the case in the Hooghly, where DOC and POC was negatively correlated in the freshwater region ($r^2 = 0.86$, $p = 0.007$, Fig.4d), which was missing in the mixing region ($p = 0.43$) and in the Sundarbans ($p = 0.84$).

Although estimated ΔDOC indicated largely net DOC addition to the Hooghly-Sundarbans system, except leaf litter leaching in the Sundarbans, no significant evidence for other internal sources was found. This suggested potential contribution from external sources that may include industrial effluents and municipal wastewater discharge (i.e., surface runoff) in the freshwater region of the Hooghly (Table 1). However, there is no direct DOC influx data to corroborate the same. Relatively higher DOC compared to POC (DOC/POC > 1) at some locations (H2, H5, H6) may stand as a signal for higher DOC contribution at those locations, but it is not prudent to pinpoint its sources due to lack of isotopic data. Although anthropogenic inputs are mostly confined to freshwater region, relatively higher DOC in the mixing zone of the Hooghly compared to freshwater region suggested DOC input via some additional pathway, possibly groundwater discharge. The contribution of groundwater to the Hooghly estuary within the salinity range observed during the present study has been reported (Samanta et al., 2015). However, there is no report of groundwater mediated DOC influx to the estuary. For mangrove-dominated ecosystems like Sundarbans, a recent study by Maher et al. (2013) estimated ~ 89 - 92% of the total DOC export to be driven by groundwater advection. To understand spatial variability of DOC chemistry in the Hooghly-Sundarbans system, a thorough investigation related to groundwater and surface runoff mediated DOC flux is warranted.

Overall, on an average ~ 40% higher DOC in the Hooghly compared to the Sundarbans appeared to be due to cumulative effect of freshwater contributions, higher anthropogenic inputs, influence of biogeochemical processes and groundwater contribution. However, DOC inputs via other pathways may be dominant over freshwater mediated input as evident from insignificant DOC - salinity relationship during the present study. To quantitatively understand the relative control of the above-mentioned contributors to the DOC pool in the Hooghly-Sundarbans system, the individual components need to be studied in detail.

4.3 Major drivers of particulate organic matter

The average POC during the present study was considerably higher compared to that reported by Ray et al. (2018) for the Hooghly-Sundarbans (Hooghly: 40.3 ± 1.1 to 129.7 ± 6.7μM, Sundarbans: 45.4 ± 7.5µM). However, the present POC values were within the range reported for a large set of Indian estuaries (POC: 51 - 750 µM; Sarma et al., 2014). No significant SPM-salinity or POC-salinity relationship was observed during the present study (Fig. 5a & 5b), except for a moderate negative correlation between POC and salinity (r^2 = 0.62, p = 0.06) in the freshwater region of the Hooghly. This inverse relationship may be linked to freshwater mediated POC addition. Also, as described earlier, contribution of POC via surface-runoff is also a possibility in this region due to presence of several industries and large urban population (St: H2: Megacity Kolkata) that discharge industrial effluent and municipal wastewater to the estuary on regular basis (Table 1). Primary signal for surface runoff mediated POC addition was evident in the freshwater zone where ~ 61% and ~ 43% higher POC at 'H3' and 'H4' compared to an upstream location (St. H2) was observed. However, based on the present data, it is not possible to decouple freshwater and surface runoff mediated POC input to the Hooghly estuary. Relatively lower contribution of POC to the SPM pool of the Sundarbans (0.66 - 1.23%) compared to the Hooghly (0.96 - 4.22%; Fig. 5c) may be due to low primary production owing to high SPM load (Ittekkot and Laane, 1991) as observed in the mangrove-dominated Godavari estuary in the southern India (Bouillon et al., 2003).

In general, wide range for $\delta^{13}C$ (rivers ~ –25 to –28‰; marine plankton ~ –18 to –22‰; C_3 plant ~ –23 to –34‰; C_4 plants ~ –_9 to –17‰) have been reported by ~~several~~different researchers in different ~~environments (Hedges et al., 1997, Bouillon et al., 2002, Zhang et al., 1997,~~ ecosystems (Smith and Epstein, 1971, Hedges et al., 1997, Zhang et al., 1997, Dehairs et al., 2000~~). On an average, $\delta^{13}C_{POC}$ at the Hooghly (–24.87 ± 0.89‰) was relatively lower compared to that of Sundarbans (–23.36 ± 0.32‰) suggesting relatively higher influence of terrestrial inputs in the Hooghly.~~, Bouillon et al., 2002). In the Hooghly, our measured $\delta^{13}C_{POC}$ suggested influx of POC via freshwater runoff as well as terrestrial C_3 plants. Additionally, the estuary was also anthropogenically stressed during postmonsoon with measured $\delta^{13}C_{POC}$ within the range reported for sewage ($\delta^{13}C$ ~ –28 to –14 ‰, Andrews et al., 1998). In the mixing zone of the Hooghly, significantly lower $\delta^{13}C_{POC}$ at 'H11' and 'H12' compared to other sampling locations may be ~~attributed~~linked to localized ^{13}C depleted ~~OC~~organic C influx to the estuary from adjacent ~~mangroves~~mangrove and anthropogenic discharge, respectively~~. No significant correlation between $\delta^{13}C_{POC}$ and salinity (Fig. 4c) was observed during the study period~~.

~~Despite being mangrove dominated region, relatively higher $\delta^{13}C_{POC}$ in the Sundarbans compared to mangroves ($\delta^{13}C$ ~ –27‰; Miyajima et al., 2009) suggest marine~~

influence or biogeochemical modification of POC within the estuarine system. Being well-oxygenated system, *in situ* aerobic biogeochemical transformation of POC is very likely to occur within the estuary; however, evidence for *in situ* aerobic POC mineralization was not obvious from the data as relationship between [POC]-pCO_2 was not significant (Fig. not shown). Similar to open ocean environment, the possibility of OC metabolism within isolated anoxic microhabitats of sinking particulate OM exists in the mangrove dominated estuaries of the Indian Sundarbans (Reeburgh et al., 2007), which may favour production of trace gases, such as CH_4. Although CH_4 super saturation (%CH_4: 2483 ± 50 to 3525 ± 1054) as well as impact of SPM load on CH_4 oxidation have been reported in this oxygenated mangrove environment (Biswas et al., In the estuaries of Sundarbans, isotopic signatures of POC showed similarity with terrestrial C_3 plants. Interestingly, despite being mangrove-dominated estuary (salinity: 12.74 - 16.55) no clear signature of either freshwater or mangrove ($\delta^{13}C$: mangrove leaf ~ –28.4‰, soil ~ –24.3‰, Ray et al., 2015, 2018) borne POC was evident from $\delta^{13}C_{POC}$ values, suggesting towards the possibility of significant POC modification within the system. Modification of POC within the estuaries of Indian sub-continent have been reported earlier (Sarma et al., 2014). Inter-estuary comparison revealed relatively lower average $\delta^{13}C_{POC}$ at the Hooghly (mean $\delta^{13}C_{POC}$: –24.87 ± 0.89‰) compared to the Sundarbans (mean $\delta^{13}C_{POC}$: −23.36 ± 0.32‰), which appeared to be due to differences in degree of freshwater contribution, anthropogenic inputs (high in Hooghly vs. little/no in Sundarbans), nature of terrestrial C_3 plant material (mangrove in the Sundarbans vs. others in Hooghly) as well as responsible processes for POC modification within the system.

To decipher processes involved in POC modification, estimated ΔC for POC (ΔPOC) in the Hooghly indicated both net addition (n = 3) and removal (n = 3) of POC in the freshwater region (ΔPOC = – 0.45 to 0.48), whereas removal (n = 6) dominated over addition (n = 1) in the mixing region (ΔPOC = – 0.39 to 0.07). In an estuary, POC may be added through freshwater and surface runoff mediated inputs, phytoplankton productivity, and DOC flocculation. The removal of POC is likely due to settling at subtidal sediment, export to adjacent continental shelf region, modification via conversion to DOC and mineralization in case of oxygenated estuary.

The plot between $\Delta\delta^{13}C$ for POC ($\Delta\delta^{13}C_{POC}$) and ΔPOC (Fig. 5d) indicated different processes to be active in different regions of the Hooghly estuary. Decrease in ΔPOC with increase in $\Delta\delta^{13}C_{POC}$ (RR; n = 4 for mixing region and n = 1 for freshwater region) suggested modification of POC due to aerobic respiration (or mineralization). This process did not appear to significantly impact estuarine CO_2 pool as evident from the POC - pCO_2

relationship (freshwater region: p = 0.29, mixing region: p = 0.50; Fig. 5e). Decrease in both ΔPOC and $\Delta\delta^{13}C_{POC}$ (SD; n = 2 for mixing region and n = 2 for freshwater region) supported settling of POC to sub-tidal sediment. Despite high water residence time (~ 40 days during postmonsoon, Samanta et al., 2015), this process may not be effective in the Hooghly due to unstable estuarine condition (described earlier). Increase in ΔPOC with decrease in $\Delta\delta^{13}C_{POC}$ (SR, FR & PP; n = 2 for freshwater region) indicated increase of POC via surface and freshwater runoff as well as phytoplankton productivity. Increase in both ΔPOC and $\Delta\delta^{13}C_{POC}$ (n = 1 for mixing region and n = 1 for freshwater region) may be linked to DOC to POC conversion by flocculation.

In the Sundarbans, negative and lower ΔPOC_{M2} (–209 to –28µM) compared to ΔPOC_{M1} (–35 to 327µM) suggested DIC like behavior, i.e., simultaneous removal or modification along with addition of mangrove derived POC. No evidence for *in situ* POC-DOC exchange was obvious based on POC-DOC relationship; however, signal for POC mineralization was evident in the Sundarbans from POC - pCO_2 relationship (r^2 = 0.37, p = 0.05, Fig.5f). Similar to the Hooghly, despite high water residence time in mangroves (Alongi et al., 2005, Singh et al., 2016), unstable estuarine condition may not favor efficient settlement of POC at sub-tidal sediment. The export of POC from the Hooghly-Sundarbans system to the northern BOB, without significant *in situ* modification, is also a possibility. This export has been estimated to be ~0.02 - 0.07Tg and ~ 0.58Tg annually for the Hooghly and Sundarbans, respectively (Ray et al. 2018).

4.4 pCO_2 and FCO_2 in the Hooghly-Sundarbans

The estimated pCO_2 for the Hooghly-Sundarbans system were in the range reported for other tidal estuaries of India (Cochin estuary: 150-3800µatm, Gupta et al., 2009; Mandovi - Zuari estuary: 500-3500µatm, Sarma et al., 2001). In the Sundarbans, barring three locations (S3, T3 and M2), a significant negative correlation between pCO_2 and %DO (r^2 = 0.76, p = 0.005; Figure not given) suggested presence of processes, such as OM mineralization, responsible for controlling both CO_2 production and O_2 consumption in the surface estuarine water. Furthermore, significant positive correlation between ECO_2 and AOU (ECO_2 = 0.057AOU + 1.22, r^2 = 0.76, p = 0.005, n = 8; Fig.6a) confirmed the effect of aerobic OM mineralization on CO_2 distribution, particularly in the upper region of the Sundarbans. Our observations were in agreement with a previous study in the Sundarbans (Akhand et al., 2016) as well as another sub-tropical estuary, Pearl River estuary, China (Zhai et al., 2005). However, relatively lower slope for ECO_2 - AOU relationship (0.057) compared to the slope for Redfield respiration in HCO_3^- rich environment [$(CH_2O)_{106}(NH_3)_{16}H_3PO_4$ + $138O_2$ + $18HCO_3^{2-}$ → $124CO_2$ + $140H_2O$ + $16NO_3^-$ + HPO_4^{2-}; ΔCO_2: ($-\Delta O_2$) = 124/138 = 0.90, Zhai et al., 2005] suggested lower production of CO_2 than expected from Redfield respiration. This may be linked to formation of low molecular weight OM instead of the final product (CO_2) during aerobic OM respiration (Zhai et al., 2005). Moreover, pCO_2 - salinity relationship (p = 0.18, Fig.6b) confirmed no significant effect of fresh and marine water contribution on variability of pCO_2 in the Sundarbans. Other potential source of CO_2 to mangrove-dominated Sundarbans could be groundwater (or pore water) exchange across intertidal mangrove sediment-water interface. Although based on our own dataset, it is not possible to confirm the same. However, relatively higher pCO_2 levels during low-tide compared to high-tide at Matla estuary in the Sundarbans (Akhand et al. 2016) as well as in other mangrove systems worldwide (Rosentreter et al., 2018, Call et al., 2015, Bouillon et al., 2007) suggested groundwater (or pore water) exchange to be a potential CO_2 source in such systems.

Unlike Sundarbans, ECO_2 - AOU relationship did not confirm significant impact of OM respiration on CO_2 in either freshwater (p = 0.50) or mixing regions (p = 0.75) of the Hooghly (Fig. 6c). Overall, pCO_2 in the freshwater region of the Hooghly was significantly higher compared to the mixing zone (Table 3), which may be linked to CO_2 supply in the freshwater region through freshwater or surface runoff from adjoining areas (Table - 1). Inter-estuary comparison of pCO_2 also revealed ~1291 µatm higher pCO_2 in the Hooghly compared to the Sundarbans, which was largely due to significantly higher pCO_2 in freshwater region of the Hooghly (Table 2 & 3). Lack of negative correlation between pCO_2 - salinity in freshwater region (Fig. 6d) of the Hooghly suggested limited contribution of CO_2 due to freshwater inputs. Therefore, CO_2 supply via surface runoff may be primary reason for higher pCO_2 in the Hooghly estuary.

Positive2007; Dutta et al., 2015a; Dutta et al., 2017), but impact of SPM (or POC) to estuarine CH_4 production as an evidence for *in situ* anaerobic POC modification is not known in the Sundarbans. Our data were also not sufficient to establish *in situ* anaerobic POC metabolism in this oxygenated mangrove environment which demands comprehensive investigation on the fate of POC.

In both the estuarine systems, DOC was major constituent of TOC (= DOC + POC) with marginal variability between the Hooghly (43.74-82.05%; mean: 66.17%) and the Sundarbans (33.40-77.26%; mean: 60.06%). Also, dominance of inorganic C was noticed over the organic one throughout the Hooghly-Sundarbans system (TOC/DIC: Sundarbans: 0.16 – 0.38, Hooghly: 0.19 – 0.44).

4.5 Exchange flux of CO_2 in the Hooghly-Sundarbans System

In the Sundarbans, absence of significant correlation between pCO_2 – salinity and pCO_2-AOU (Fig. 5a and 5b) indicates mostly exogenous CO_2 in the estuarine waters. Our interpretation was also supported by insignificant mangrove derived OM respiration as described in the DIC section earlier (section 4.2) as well as positive pCO_2 – [DOC] relationship (section 4.3). The primary source of exogenous CO_2 in the Sundarbans may be CO_2 influx to the estuarine water from mangrove sediment pore-water exchange during tidal pumping. Although this component was not measured during the present study, it has been reported to be a source of CO_2 in a similar estuarine-intertidal marsh complex of five rivers in the southeastern USA (Cai et al., 1999). In the Hooghly, significant negative and positive relationships between pCO_2-salinity (R^2 = 0.58, p = 0.002; Fig 5a) and pCO_2 – AOU (R^2 = 0.37, p = 0.028; Fig 5b) provide evidences for both freshwater run-off and *in situ* aerobic OC mineralization mediated influx of CO_2 in the system. The significant impact of aerobic OC mineralization on estuarine pCO_2 levels have been observed in other tropical estuarine systems (Zhai et al., 2005; Dai et al., 2006). Significant OC mineralization mediated CO_2 addition coupled with its insignificant impact on [DIC] and $\delta^{13}C_{DIC}$ in the Hooghly estuary (see section 4.2) might suggest substantial outgassing or export of CO_2 from the system. Using average AOU values and stoichiometric equation of OC respiration [$(CH_2O)_{106}(NH_3)_{16}H_3PO_4 + 138O_2 \longrightarrow 106CO_2 + 16HNO_3 + H_3PO_4 + 122H_2O$], approximate CO_2 generation through OC mineralization (or respiration) at any instant in the Hooghly estuary was estimated around ~ 21.02 μmolCO_2 L^{-1}.

For both the estuaries, positive mean FCO_2 clearly suggestsuggested the Hooghly-Sundarbans system to be a net source of CO_2 to the regional atmosphere during postmonsoon (Fig.5c6e & 6f). Specifically, from regional climate and environmental change perspective, anthropogenically influenced Hooghly estuary was a relatively greater source of CO_2 to the regional atmosphere compared to the mangrove--dominated Sundarbans as evident from significantly higher CO_2 emission flux from the Hooghly ($[FCO_2]_{Hooghly}$: $[FCO_2]_{Sundarbans}$ = 17). However, despite being a CO_2 source, FCO_2 measured for the estuaries of Sundarbans was markedly higher thanwere considerably lower compared to global mean FCO_2 (~63 μmol$m^{-2}d^{-1}$) observed in reported for mangrove creek and other similar-dominated estuaries ((~ 43-59 mmol C m^{-2} d^{-1}; Call et al., 2015). HoweverSimilarly, FCO_2 measured for the

Hooghly estuary waswere relatively lower compared to some Chinese estuarine systems (Pearl River inner estuary: 46 mmol m^{-2} d^{-1}, Guo et al., 2009; Yangtze River estuary: 41 mmol m^{-2} d^{-1}, Zhai et al., 2007).

The inter-estuary variability ofdifference in FCO_2 between Hooghly and Sundarbans may be due to variability in pCO_2 level as well as micrometeorological and physicochemical parameters controlling gas transfer velocity across water-atmosphere interface. Taken together, it appears that difference in land use and degreeQuantitatively, the difference in ‘k’ values for the Hoogly and Sundarbans were not large ($k_{Sundarbans} - k_{Hooghly}$ ~ 0.031 $cmhr^{-1}$). Therefore, large difference in FCO_2 between these two estuarine systems may be due to difference in pCO_2. Taken together, supporting our hypothesis, it appears that differences in land use and degrees of anthropogenic influence have the potential to alter the C biogeochemistry of aquatic ecosystems with anthropogenically stressed aquatic systems acting as a relatively biggergreater source of CO_2 to the regional atmosphere. than mangrove-dominated ones.

Conclusions

The present study focused on investigating different aspects of C biogeochemistry of the anthropogenically affected Hooghly estuary and mangrove dominated estuaries of the Sundarbans during postmonsoon. Following conclusions were deduced from the study:

- With the exception of SPM, physicochemical parameters of the Hooghly estuary varied over a relatively wider range compared to the Sundarbans.
- Phytoplankton productivity was a majorCoupled with freshwater contribution, inorganic and organic C metabolism appeared to be dominant processes affecting DIC in the Hooghly. However, in the Sundarbans, significant DIC removal over addition was noticed. Influence of groundwater on estuarine DIC biogeochemistry was also observed with relatively higher influence at the Sundarbans.
- Higher DOC level in the Hooghly appeared to be regulated by coupled interactions among anthropogenic inputs, biogeochemical processes and groundwater contribution rather than freshwater mediated inputs.
- Signatures of freshwater runoff, terrestrial C_3 plants, and anthropogenic discharge were found in POC of the Hooghly, whereas evidence for only C_3 plants were noticed at the Sundarbans with possible POC modification.

- Organic matter mineralization and surface run-off from adjoining areas appeared to be dominant controlling factor on DIC in the mixing zone of the Hooghly with carbonate precipitation and dissolution being dominant in the freshwater regime. In the Sundarbans, signal for mangrove derived DIC removal was noticed.
- DOC behaved non-conservatively in the Hooghly-Sundarbans system. Evidence for DOC to POC interconversion was observed in the Hooghly. Analogous sources of *p*CO_2 and DOC in the form of pore-water exchange was found in the Sundarbans.
- In the Sundarbans, contribution of terrestrial organic matter to the POM pool was relatively lower and Hooghly, respectively, with higher average *p*CO_2 in the Hooghly compared to the Hooghly with possibility of *in situ* biogeochemical modifications in the Sundarbans.
- During postmonsoon, theThe entire Hooghly-Sundarbans system acted as a source of CO_2 to the regional atmosphere. In the Hooghly estuary, CO_2 is added through freshwater runoff and OC mineralization, whereas CO_2 in the with ~17 times higher emission from the Hooghly compared to Sundarbans is principally exogenous, suggesting dominance of anthropogenically stressed estuarine system over mangrove-dominated one from regional climate change perspective.

Data availability

Data used in the manuscript is presented in tables (Table 1, Table 2, and Table 3) of the manuscript.

Author contributions

MKD and SK designed the study. MKD with RM and PS collected and analyzed samples. MKD and SK interpreted the data and drafted the manuscript. SKM provided facility to measure basic physicochemical parameters and DOC.

Competing interest

The author declares no conflict of interest.

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References

Abril, G., Nogueira, E., Hetcheber, H., Cabeçadas, G., Lemaire, E., and Brogueira, M.J.: Behaviour of organic carbon in nine contrasting European estuaries, Estuarine Coastal Shelf Sci., 54, 241–262, https://doi.org/10.1006/ecss.2001.0844, 2002.

Akhand, A., Chanda, A., Manna, S., Das, S., Hazra, S., Roy, R., Choudhury, S.B., Rao, K.H., Dadhwal, V.K., Chakraborty, K. and Mostofa, K.M.G.: A comparison of CO_2 dynamics and air-water fluxes in a river dominated estuary and a mangrove dominated marine estuary. Geophys. Res. Lett., 43(22), https://doi.org/10.1002/2016GL070716, 2016.

Akhand, A., Chandra, A., Dutta, S., and Hazra, S.: Air- water carbon dioxide exchange dynamics along the estuarine transition zone of Sunderban, northern Bay of Bengal, India, Indian J. Geo-Marine Sci. 41, 111–116, 2012.

Alongi, D.M., Ramanathan, A.L., Kannan, L., Tirendi, F., Trott, L.A., and Prasad, M.B.K.: Human induced disturbance on benthic microbial metabolism in the Pichavaram mangroves, Vellar Coleroon estuarine complex, India, Mar. Biol. 147, 1033-1044, https://doi.org/10.1007/s00227-005-1634-5, 2005.

Alling, V., Porcelli, D., Morth, C- M., Anderson, L. G., Sanchez- Garcia, L., Gustafsson, O., Andersson, P. S., and Humborg, C.: Degradation of terrestrial organic carbon, primary production and out-gassing of CO_2 in the Laptev and East Siberian Seas as inferred from $\delta^{13}C$ values of DIC, Geochim. Cosmochim. Acta, 95, 143–159, https://doi.org/10.1016/j.gca.2012.07.028, 2012.

Alongi, D.M.: Carbon cycling and storage in mangrove forests, Ann. Rev. Mar. Sci. 6, 195–219, 10.1146/annurev-marine-010213-135020, 2014.

Alongi, D.M., and Mukhopadhyay, S.K.: Contribution of mangroves to coastal carbon cycling in low latitude seas. Agric. For. Meteorol., 213, 266-272, doi:10.1016/j.agrformet.2014.10.005, 2014.

Andrews, J. E., Greenway, A.M., and Dennis, P.F.: Combined carbon isotope and C/N ratios as indicators of source and fate of organic matter in a poorly flushed, tropical estuary. Hunts Bay, Kingston Harbour, Jamaica, Estuar. Coast. Shelf Sci., 46, 743–456, https://doi.org/10.1006/ecss.1997.0305, 1998.

Barnes, J., Ramesh, R., Purvaja, R., Nirmal Rajkumar, A., Senthil Kumar, B., and Krithika, K.: Tidal dynamics and rainfall control N_2O and CH_4 emissions from a pristine mangrove creek, Geophys. Res. Lett. 33, L15405. doi:10.1029/ 2006GL026829, 2006.

Bauer, J. E., Cai, W.J., Raymond, P.A., Bianchi, T.S., Hopkinson, C.S., and Regnier, P.A.G.: The changing carbon cycle of the coastal ocean, Nature, 504 (7478), 61–70, doi: 10.1038/nature12857, 2013.

Bhavya, P.S., Kumar, S., Gupta, G.V.M., and Sudheesh, V.: Carbon uptake rates in the Cochin estuary and adjoining coastal Arabian Sea, Estuaries and Coasts, 40, 447, doi: 10.1007/s12237-016-0147-4, 2017.

Bhavya, P.S., Kumar, S., Gupta, G.V.M., Sudharma, K.V., and Sudheesh, V.: Spatial-temporal variation in $\delta^{13}C_{DIC}$ of a tropical eutrophic estuary (CoachinCochin estuary, India), Cont. **Shelf** Res. 153, 75-85, https://doi.org/10.1016/j.csr.2017.12.006, 2018.

Biswas, H., Mukhopadhyay, S. K., De, T. K., Sen, S., and Jana, T. K.: Biogenic controls on the air-water carbon dioxide exchange in the Sundarban mangrove environment, northeast coast of Bay of Bengal, India, Limnol. Oceanogr. 49, 95-101. doi: 10.4319/lo.2004.49.1.0095, 2004.

Biswas, H., Mukhopadhyay, S.K., Sen, S., and Jana, T.K.: Spatial and temporal patterns of methane dynamics in the tropical mangrove dominated estuary, NE Coast of Bay of Bengal, India. J. Marine Syst. 68, 55-64, https://doi.org/10.1016/j.jmarsys.2006.11.001, 2007.

Borges, A. V., Delille, B., and Frankignoulle, M.: Budgeting sinks and sources of CO_2 in the coastal ocean: Diversity of ecosystems counts, Geophys. Res. Lett., 32, L14601, https://doi.org/10.1029/2005gl023053, 2005.

Borges, A. V., Delille, B., Schiettecatte, L.-S., Gazeau, F., Abril, G., and Frankignoulle, M.: Gas transfer velocities of CO_2 in three European estuaries (Randers Fjord, Scheldt and Thames), Limnol. Oceanogr., 49, 1630–1641, https://doi.org/10.4319/lo.2004.49.5.1630, 2004.

Bouillon, S., Borges, A. V., Castañeda-Moya, E., Diele, K., Dittmar, T., Duke, N. C., Kristensen, E., Lee, S. Y., Marchand, C., Middelburg, J. J., Rivera-Monroy, V. H., Smith, T. J., and Twilley, R. R.: Mangrove production and carbon sinks: A revision of global budget estimates, Global Biogeochem. Cy., 22, GB2013, 10.1029/2007GB003052, 2008a2008.

Bouillon, S., Dehairs, F., Velimirov, B., Abril, G., and Borges, A.V.: Dynamics of organic and inorganic carbon across contiguous mangrove and seagrass systems (Gazi Bay, Kenya), J. Geophys. Res., 112, G02018, doi:10.1029/2006JG000325, 2007.

Bouillon, S., Korntheuer, M., Baeyens, W., and Dehairs, F.: A new automated setup for stable isotope analysis of dissolved organic carbon. Limnol. Oceanogr.; Methods 4, 216. doi: 10.4319/lom.2006.4.216, 2006.

Bouillon. S., Frankignoulle, M., Dehairs, F., Velimirov, B., Eiler, A., Etcheber, H., Abril, G., and Borges, A.V.: Inorganic and organic carbon biogeochemistry in the Gautami Godavari estuary (Andhra Pradesh, India) during pre-monsoon: the local impact of extensive mangrove forests, Global Biogeochem. Cy. 17 (4), 1114, doi:10.1029/ 2002GB00202, 2003.

Bouillon, S., Raman, A.V., Dauby, P., and Dehairs, F.: Carbon and nitrogen stable isotope ratios of subtidal benthic invertebrates in an estuarine mangrove ecosystem (Andhra Pradesh, India), Estuar. Coast. Shelf Sci., 54, 901 - 913. https://doi.org/10.1006/ecss.2001.0864https://doi.org/10.1006/ecss.2001.0864, 2002.

Boto, K. G., and Wellington, J.T.: Seasonal variations in concentrations and fluxes of dissolved organic and inorganic materials in a tropical, tidally dominated waterway, Mar. Ecol. Prog. Ser., 50, 151–160, 1988.

Call, M., Maher, D.T., Santos, I.R., Ruiz-Halpern, S., Mangion, P., and Sanders, et al.: Spatial and temporal variability of carbon dioxide and methane fluxes over semidiurnal and spring–neap–spring timescales in a mangrove creek. Geochim. Cosmochim. Acta, 150, 211–225, https://doi.org/10.1016/j.gca.2014.11.023, 2015.

Cai, W.-J.: Estuarine and coastal ocean carbon paradox: CO_2 sinks or sites of terrestrial carbon incineration?, Annu. Rev. Mar. Sci., 3, 123–145, https://doi.org/10.1146/annurev-marine120709-142723, 2011.

Cai, W.-J., Dai, M., and Wang, Y.: Air-sea exchange of carbon dioxide in ocean margins: A province-based synthesis, Geophys. Res. Lett., 33, 2–5, 2006.

Cai, W.-J., Pomeroy, L.R., Moran, M.A., and Wang, Y.C.: Oxygen and carbon dioxide mass balance for the estuarine intertidal marsh complex of five rivers in the southeastern US, Limnol. Oceanogr., 44 (3), 639–649, https://doi.org/10.4319/lo.1999.44.3.0639, 1999.

Cai, W. J., Wang, Y., Krest, J., and Moore, W.S.: The geochemistry of dissolved inorganic carbon in a surficial groundwater aquifer in North Inlet, South Carolina and the carbon fluxes to the coastal ocean, Geochim. Cosmochim. Acta, 67, 631–637, https://doi.org/10.1016/S0016-7037(02)01167-5, 2003.

Carpenter, I.H., Bradford, W.L., and Grant, V.: Processes affecting the composition of estuarine waters. In: Cronin, L.E. (Ed.), Estuarine Research. 1. Academic, pp. 188–214, 1975.

Camilleri, J. C., and Ribi, G.: Leaching of dissolved organic carbon (DOC) from dead leaves, formation of flakes from DOC, and feeding on flakes by crustaceans in mangroves, Mar. Biol., 91, 337– 344, 1986.

Cai, W. J.: Estuarine and coastal ocean carbon paradox: CO_2 sinks or sites of terrestrial carbon incineration?, Annu. Rev. Mar. Sci., 3, 123–145, https://doi.org/10.1146/annurev-marine120709-142723, 2011.

Callahan, J., Dai, M., Chen, R.F., Li, X.L., Lu, Z.M., and Huang, W.: Dissolved organic matter in the Pearl River Estuary, China, Mar. Chem. 89, 211 – 224, https://doi.org/10.1016/j.marchem.2004.02.013, 2004.

Cao, Z., Dai, M., Zheng, N., Wang, D., Li, Q., Zhai, W., Meng, F., and Gan, J: Dynamics of the carbonate system in a large continental shelf system under the influence of both a river plume and coastal upwelling, J. Geophys. Res., 116, G02010, doi:10.1029/2010JG001596, 2011.

Chen, C.-T. A. and Borges, A. V.: Reconciling opposing views on carbon cycling in the coastal ocean: Continental shelves as sinks and near-shore ecosystems as sources of atmospheric CO2, Deep-Sea. Res. Pt. II., 56, 578–590, 2009.

Dai, M.H., Guo, X.H., Zhai, W.D., Yuan, L.Y., Wang, B.W., Wang, L.F., Cai, P.H., Tang, T.T., and Cai, W. J.: Oxygen depletion in the upper reach of the Pearl River estuary during a winter droughtCIFRI,: Present status of Hilsa in Hooghly – Bhagirathi river, Central Inland Fisheries Research Institute. www.cifri.ernet.in/179.pdf, 2012.

Cotovicz Jr., L. C., Knoppers, B. A., Brandini, N., Costa Santos, S. J., and Abril, G.: A strong CO_2 sink enhanced by eutrophication in a tropical coastal embayment (Guanabara Bay, Rio de Janeiro, Brazil), Biogeosciences, 12, 6125-6146, https://doi.org/10.5194/bg-12-6125-2015, 2015.

. Mar. Chem., 102, 159-169, https://doi.org/10.1016/j.marchem.2005.09.020, 2006.

Dehairs, F., Rao, R.G., Chandra Mohan, P., Raman, A.V., Marguillier, S., and Hellings, L.: Tracing mangrove carbon in suspended matter and aquatic fauna of the Gautami- Godavari Delta, Bay of Bengal (India), Hydrobiologia, 431, 225 - 241. doi: 10.1023/A:1004072310525, 2000.

Degens, E.T., Kempe, S., and Richey, J.E.: Summary: Biogeochemistry of major world rivers. In: Degens ET, Kempe S, and Richey JE (Eds.), Biogeochemistry of Major World Rivers, SCOPE 42. Chichester: John Wiley & Sons. pp 323 347, 1991.

Dittmar, T., Hertkorn, N., Kattner, G., and Lara, R.J.: Do mangroves rather than rivers provide nutrients J.: Mangroves, a major source of dissolved organic carbon to coastal environments south of the Amazon River? Evidence from long-term flux measurements, Mar. oceans, Global Biogeochem. Cycles, 20, doi:10.1029/ 2005gb002570, 2006.Ecol. Prog. Ser., 213, 67 – 77, 2001a.

Dittmar, T., and Lara, R.J.: Driving forces behind nutrient and organic matter dynamics in a mangrove tidal creek in north Brazil, Estuarine Coastal Shelf Sci., 52, 249 – 259, https://doi.org/10.1006/ecss.2000.0743, 2001b2001a.

Donato, D.C., Kauffman, J.B., Kurnianto, S., Stidham, M., and Murdiyarso, D.: Mangroves among the most carbon-rich forests in the tropics. Nat. Geosci., 4, 293-297, doi: 10.1038/NGEO1123, 2011.

Dutta, K., Ravi Prasad, G. V., Ray, D. K., and Raghav, K.: Decadal changes of Radiocarbon in the surface Bay of Bengal: Three decades after GEOSECS and one decade after WOCE, Radiocarbon, 52(2–3), 1191–1196, 2010.

Dutta, M. K., Chowdhury, C., Jana, T. K., and Mukhopadhyay, S. K.: Dynamics and exchange fluxes of methane in the estuarine mangrove environment of Sundarbans, NE coast of India, Atmos. Environ. 77, 631–639, doi: 10.1016/j.atmosenv.2013.05.050, 2013.

Dutta, M. K., Mukherkjee, R., Jana, T. K., and Mukhopadhyay, S. K.: Atmospheric fluxes and photo-oxidation of methane in the mangrove environment of the Sundarbans, NE coast of India; A case study from Lothian Island, Agric. For. Meteorol. 213, 33 41, doi: 10.1016/j.agrformet.2015.06.010, 2015b.

Dutta, M. K., Mukherkjee, R., Jana, T. K., and Mukhopadhyay, S. K.: Biogeochemical dynamics of exogenous methane in an estuary associated to a mangrove biosphere; the Sundarbans, NE coast of India, Mar. Chem. 170, 1–10, doi: 10.1016/j.marchem.2014.12.006, 2015a2015.

Dutta, M.K., Bianchi, T.S., and Mukhopadhyay, S.K.: Mangrove methane biogeochemistry in the Indian Sundarbans: a proposed budget, Frontiers in Marine Science, 4, 187. doi: 10.3389/fmars.2017.00187, 2017.

Frankignoulle, M., Abril, G., Borges, A., Bourge, I., Canon, C., Delille, B., Libert, E., and Théate, J.-M.: Carbon dioxide emission from European estuaries, Science, 282, 434–436, https://doi.org/10.1126/science.282.5388.434, 1998.

Frankignoulle, M., and Borges, A. V.: Direct and indirect pCO_2 measurements in a wide range of pCO_2 and salinity values (the Scheldt estuary), Aquat. Geochem. 7, 267 - 273. doi: 10.1023/A:1015251010481, 2011.

Fry, B.: Conservative mixing of stable isotopes across estuarine salinity gradients: a conceptual framework for monitoring watershed influences on downstream fisheries production, Estuaries 25, 264–271, https://doi.org/10.1007/BF02691313, 2002.

Freitas, H.A., Pessenda, L.C.R., Aravena, R., Gouveia, S.E.M., Ribeiro, A.S., and Boulet, R.: Late quaternary vegetation dynamics in the southern Amazon Basin inferred from carbon isotopes in soil organic matter. Quat. Res. 55, 39–46, https://doi.org/10.1006/qres.2000.2192 2001.

Ganguly, D., Dey, M., Mandal, S.K., De, T.K., and Jana, T.K.: Energy dynamics and its implication to biosphere-atmosphere exchange of CO_2, H_2O and CH_4 in a tropical mangrove forest canopy, Atmos. Environ. 42, 4172 – 4184, 2008.

Ganguly, D., Dey, M., Sen, S., and Jana, T.K.: Biosphere-atmosphere exchange of NOx in the tropical mangrove forest, J. Geophys. Res. 114, G04014. http:// dx.doi.org/10.1029/2008JG000852, 2009.

Gattuso, J.-P., Frankignoulle, M., Bourge, I., Romaine, S., and Buddemeier, R. W.: Effect of calcium carbonate saturation of seawater on coral calcification, Glob. Planet. Change, 18,37-46, https://doi.org/10.1016/S0921-8181(98)00035-6, 1998.

Ghosh, P., Chakrabarti, R., and Bhattacharya, S. K.: Short and long-term temporal variations in salinity and the oxygen, carbon and hydrogen isotopic compositions of the Hooghly Estuary water, India, Chem. Geol. 335, 118–127, https://doi.org/10.1016/j.chemgeo.2012.10.051, 2013.

Ghosh, B, B., Ray, P., and Gopalakrishnan, V.: Survey and characterization of waste water discharged into the Hooghly Estuary, J. Inland Fishery Soc. of India, 4, 2–10, 1973.

Giri, C., Ochieng, E., Tieszen, L., Zhu, Z., Singh, A., Loveland, T., Masek, J., and Duke, N.: Status and distribution of mangrove forests of the world using earth observation satellite data. Global Ecol. Biogeogr. 20(1), 154-159, 2011.

Goutam, K.S., Tanaya, D., Anwesha, S., Sharanya, C., and Meenakshi, C.: Tide and mixing characteristics in Sundarbans Estuarine River system, Hydrol. Current Res. 6 (2), https://doi.org/10.4172/2157-7587.1000204, 2015.

Grasshoff, K., Ehrharft,M., and Kremling, K.: .Methods of Seawater Analysis, 2nd Edn.Weinheim: Verlag Chemie, 1983.

Guo, X., Dai, M., Zhai, W., Cai, W.-J., and Chen, B.: CO_2 flux and seasonal variability in a large subtropical estuarine system, the Pearl River Estuary, China. J. Geophys. Res. 114, G03013. http://dx.doi.org/10.1029/2008JG000905, 2009.

Guo, X., Cai, W.-J., Zhai, W., Dai, M., Wang, Y., and Chen, B.: Seasonal variations in the inorganic carbon system in the Pearl River (Zhujiang) estuary. Continental Shelf Research 28, 1424-1434, https://doi.org/10.1016/j.csr.2007.07.011, 2008.

Gupta, G.V.M., Thottathil, S.D., Balachandran, K.K., Madhu, N.V., Madeswaran, P., and Nair, S.: CO_2 supersaturation and net heterotrophy in a tropical estuary (Cochin, India): influence of anthropogenic effect, Ecosystems, 12 (7), 1145-1157, https://doi.org/10.1007/s10021-009-9280-2, 2009.

Hedges, J. I., Keil, R.G., and Benner, R.: What happens to terrestrial organic matter in the ocean?. Org Geochem., 27, 195 - 212. https://doi.org/10.1016/S0146-6380(97)00066-1https://doi.org/10.1016/S0146-6380(97)00066-1, 1997.

Heip, C. H. R., Goosen, N.K., Herman, P.M.J., Kromkamp, J., Middelburg, J.J., and Soetaert, K.: Production and consumption of biological particles in temperate tidal estuaries, Oceanogr. Mar. Biol. Annu. Rev., 33, 1–149, 1995.

Ho, D.T., Ferron, S., Engel, V.C., Anderson, W.T., Swart, P.K., Price, R.M., and Berbero, L.: Dissolved carbon biogeochemistry and export in mangrove-dominated rivers of the Florida Everglades, Biogeosciences, 14, 2543–2559, doi:10.5194/bg-14-2543-2017, 2017.

Hopkinson, C.S., Fry, B., Nolin, A.: Stoichiometry of dissolved organic matter dynamics on the continental shelf of the Northeastern USA, Cont. Shelf Res. 17, 473–489, doi: 10.1016/S0278-4343(96)00046-5, 1997.

Huang T.-H., Fu Y.-H., Pan P.-Y., Arthur Chen, C.-T.: Fluvial carbon fluxes in tropical rivers, Curr. Opin. Environ. Sustain. 4, 162–169, https://doi.org/10.1016/j.cosust.2012.02.004, 2012.

Ittekkot, V., and Laane, R.W.P.M.: Fate of riverine particulate organic matter. In: Degens, E.T.; Kemp, S.; Richey, J.E., eds. Biogeochemistry of major world rivers. Chichester: Wiley; 233-243, 1991.

Ittekkot, V.: Global trends in the nature of organic matter in river suspensions, Nature 332, 436–438, 1988.

Jennerjahn, T., and Ittekkot, C. V.: Organic matter in sediments in the mangrove areas and adjacent continental margins of Brazil: I. Amino acids and hexosamines, Oceanol. Acta 20, 359–369, 1997.

Koné Y. J.-M., and Borges., A.V.: Dissolved inorganic carbon dynamics in the waters surrounding forested mangroves of the Ca Mau Province (Vietnam), Estuar. Coast. Shelf Sci., 77(3), 409-421, https://doi.org/10.1016/j.ecss.2007.10.001, 2008.

Khan, R. A.: The pollution problem of Hooghly estuarine system; Estuarine Ecosystem Series, Zoological survey of India, part 2, 497–542, 1995.

Kristensen, E., and Alongi, D.M.: Control by fiddler crabs (Ucavocans) and plant roots (Avicennia marina) on carbon, iron, and sulphur biogeochemistry in mangrove sediment, Limnol. Oceanogr. 51, 1557–1571, doi: 10.4319/lo.2006.51.4.1557, 2006.

Kristensen, E., and Suraswadi, P.: Carbon, nitrogen and phosphorus dynamics in creek water of a Southeast Asian mangrove forest, Hydrobiologia, 474, 197–211, 2002.

Le Quéré, C., Andrew, R. M., Canadell, J. G., Sitch, S., Korsbakken, J. I., Peters, G. P., Manning, A. C., Boden, T. A., Tans, P. P., Houghton, R. A., Keeling, R. F., Alin, S., Andrews, O. D., Anthoni, P., Barbero, L., Bopp, L., Chevallier, F., Chini, L. P., Ciais, P., Currie, K., Delire, C., Doney, S. C., Friedlingstein, P., Gkritzalis, T., Harris, I., Hauck, J., Haverd, V., Hoppema, M., Klein Goldewijk, K., Jain, A. K., Kato, E., Körtzinger, A., Landschützer, P., Lefèvre, N., Lenton, A., Lienert, S., Lombardozzi, D., Melton, J. R., Metzl, N., Millero, F., Monteiro, P. M. S., Munro, D. R., Nabel, J. E. M. S., Nakaoka, S.-I., O'Brien, K., Olsen, A., Omar, A. M., Ono, T., Pierrot, D., Poulter, B., Rödenbeck, C., Salisbury, J., Schuster, U., Schwinger, J., Séférian, R., Skjelvan, I., Stocker, B. D., Sutton, A. J., Takahashi, T., Tian, H., Tilbrook, B., van der Laan-Luijkx, I. T., van der Werf, G. R., Viovy, N., Walker, A. P., Wiltshire, A. J., and Zaehle, S.: Global Carbon Budget 2016, Earth Syst. Sci. Data, 8, 605–649, https://doi.org/10.5194/essd-8-605-2016, 2016.

Linto N., Barnes, J., Ramachandran, R., Divia, J., Ramachandran, P., and Upstill-Goddard, R. C.: Carbon dioxide and methane emissions from mangrove-associated waters of the Andaman Islands, Bay of Bengal, Estuaries and Coasts, 37, 381–398, https://doi.org/10.1007/s12237-013-9674-4, 2014.

Liss, P. S., and Merlivat, L.: "Air sea gas exchange rates: introduction and synthesis," in The Role of Air Sea Exchange in Geochemical Cycling, ed P. Buat-Menard (Hingham, MA: D. Reidel) 113–129, 1986.

Maher, D., Santos, I., Golsby-Smith, L., Gleeson, J., and Eyre, B.: Groundwater-derived dissolved inorganic and organic carbon exports from a mangrove tidal creek: The missing mangrove carbon sink?, Limnol. Oceanog., 58, 475–488, doi:10.4319/lo.2013.58.2.0475, 2013.

Millero, F.J.: Chemical Oceanography, Fourth Edition, CRC press, Taylor and Francis Group, 2013.

Miyajima T., Tsuboi Y., Tanaka Y., and Koike, I.: Export of inorganic carbon from two Southeast Asian mangrove forests to adjacent estuaries as estimated by the stable isotope composition of dissolved inorganic carbon, J. Geophys. Res., 114, G01024, doi:10.1029/2008JG000861, 2009.

Moran, M.A., Sheldon Jr., W.M., and Sheldon, J.E.: Biodegradation of riverine dissolved organic carbon in five estuaries of the south United States, Estuaries 22, 55 – 64, 1999.

Mook,W.G., and Tan, T.C.: Stable carbon isotopes in rivers and estuaries. In: Degens, E.T., Kempe, S., Richey, J.E. (Eds.), Biogeochemistry of Major World Rivers. SCOPE, John Wiley and Sons Ltd., pp. 245–264, 1991.

Mukhopadhyay, S.K., Biswas, H., De, T.K., and Jana, T.K.: Fluxes of nutrients from the tropical river Hooghly at the land-ocean boundary of Sundarbans, NE coast of Bay of Bengal, India, J. Marine Syst. 62 (1-2), 9-21, https://doi.org/10.1016/j.jmarsys.2006.03.004, 2006.

Mukhopadhyay, S.K., Biswas, H., De, T.K., Sen, S., Sen, B.K., and Jana, T.K.: Impact of Sundarbans mangrove biosphereSeasonal effects on the air–water carbon dioxide and methane mixing ratio atexchange in the Hooghly estuary, NE coast of Bay of Bengal, India, Atmos. J Environ. Monit. 36 (4), 629-638, https://doi.org/10.1016/S1352-2310(01)00521-0,10.1039/b201614a, 2002.

Ray, R., Baum, A., Rixen, T., Gleixner, G., and Jana, T.K.: Exportation of dissolved (inorganic and organic) and particulate carbon from mangroves and its implication to the carbon budget in the Indian Sundarbans, Sci. Total Environ., 621, 535-547. https://doi.org/10.1016/j.scitotenv.2017.11.225, 2018.

Ray, R., Rixen, T., Baum, A., Malik, A., Gleixner, G., and Jana, T.K.: Distribution, sources and biogeochemistry of organic matter in a mangrove dominated estuarine system (Indian Sundarbans) during the pre-monsoon, Estuar. Coast. Shelf Sci. 167, 404–413, http://dx.doi.org/10.1016/j.ecss.2015.10.017, 2015.

Ray, R., Ganguly, D., Chowdhury, C., Dey, M., Das, S., Dutta, M.K., Mandal, S.K., Majumder, N., De, T.K., Mukhopadhyay, S.K., and Jana, T.K.: Carbon sequestration and annual increase of carbon stock in a mangrove forest, Atmos. Environ. 45, 5016-5024, https://doi.org/10.1016/j.atmosenv.2011.04.074, 2011.

Raymond, P. A. and Cole, J. J.: Gas exchange in rivers and estuaries: Choosing a gas transfer velocity, Estuaries, 24, 312–317, https://doi.org/10.2307/1352954, 2001.

Raymond, P.A., Bauer, J.E.: DOC cycling in a temperate estuary: a mass balance approach using natural ^{14}C and ^{13}C, Limnol. Oceanogr. 46, https://doi.org/10.4319/lo.2001.46.3.0655, 655-667, 2001a.

Reay, W.G., Gallagher, D., and Simmons, G.M.: 1995. Sediment water column nutrient exchanges in Southern Chesapeake Bay near shore environments, Virginia Water Resources Research Centre, Bulletin - 181b, 1995.

Regnier, P., Friedlingstein, P., Ciais, P., Mackenzie, F. T., Gruber, N., Janssens, I. A., Laruelle, G. G., Lauerwald, R., Luyssaert, S., Andersson, A. J., Arndt, S., Arnosti, C., Borges,

A. V., Dale, A. W., Gallego-Sala, A., Godderis, Y., Goossens, N., Hartmann, J., Heinze, C., Ilyina, T., Joos, F., LaRowe, D. E., Leifeld, J., Meysman, F. J. R., Munhoven, G., Raymond, P. A., Spahni, R., Suntharalingam, P., and Thullner, M.: Anthropogenic perturbation of the carbon fluxes from land to ocean, Nat. Geosci., 6, 597–607, doi:10.1038/ngeo1830, 2013.

Richey, J.E., Melack, J.M., Aufdenkampe, A.K., Ballester, V.M., and Hess, L.L.: Outgassing from Amazonian rivers and wetlands as a large tropical source of atmospheric CO_2, Nature, 416, 617-620, doi:10.1038/416617a, 2002.

Rosentreter, J.A., Maher, D.T., Erler, D.V., Murray, R. and Eyre, B.D.: Seasonal and temporal CO_2 dynamics in three tropical mangrove creeks - A revision of global mangrove CO_2 emissions. Geochim. Cosmochim. Acta, 222, 729-745, https://doi.org/10.1016/j.gca.2017.11.026, 2018.

Rudra, K.: Changing river courses in the western part of the ganga-Brahmaputra delta. Geomorphology 227, 87–100, doi: 10.1016/j.geomorph.2014.05.013, 2014.

Sadhuram, Y., Sarma, V.V., Ramana Murthy, T.V. and Prabhakara Rao, B.: Seasonal variability of physicochemical characteristics of the Haldia channel of Hooghly estuary, India. J. Earth Syst. Sci.., 114, 37–49, https://doi.org/10.1007/BF02702007, 2005.

Samanta, S., Dalai, T. K., Pattanaik, J. K., Rai, S. K., and Mazumdar, A.: Dissolved inorganic carbon (DIC) and its $\delta^{13}C$ in the Ganga (Hooghly) River estuary, India: Evidence of DIC generation via organic carbon degradation and carbonate dissolution, Geochim. Cosmochim. Acta, 165, 226 – 248, doi: 10.1016/j.gca.2015.05.040, 2015.

Samanta, S., Dalai, T.K.: Massive production of heavy metals in the Ganga (Hooghly) River Estuary, India: global importance of solute-particle interaction and enhanced metal fluxes to the oceans, Geochim. Cosmochim. Acta, 228, 243–258, https://doi.org/10.1016/j.gca.2018.03.002, 2018.

Sarkar, S.K., Mondal, P., Ok, Y.S., Rinklebe, J.: Trace metal in surface sediments of the Hooghly (Ganges) estuary: distribution and contamination risk assessment, Environ. Geochem. Health 39 (6), 1245–1258, DOI: 10.1007/s10653-017-9952-3, 2017.

Sarma, V.V.S.S., Krishna, M.S., Prasad, V.R., Kumar, B.S.K., Naidu, S.A., Rao, G.D., Viswanadham, R., Sridevi, T., Kumar, P.P., and Reddy, N.P.C.: Sources and transformation of particulate organic matter in the Indian monsoonal estuaries during discharge period., J. Geophys. Res.: Biogeosci..119(11), 2095 – 2111, https://doi.org/10.1029/2011GL050709, 2014.

Sarma, V.V.S.S., Kumar, M.D., and Manerikar, M.: Emission of carbon dioxide from a tropical estuarine system, Goa, India, Geophys. Res. Lettrs., 28, 1239-1242, https://doi.org/10.1029/2000GL006114, 2001.

Sarma, V.V.S.S., Viswanadham, R., Rao, G.D., Prasad, V.R., Kumar, B.S.K., Naidu, S.A., Kumar, N.A., D.B. Rao, Sridevi, T., Krishna, M.S., Reddy, N.P.C., Sadhuram, Y., and Murty, T.V.R.: Carbon dioxide emissions from Indian monsoonal Estuaries. Geophys. Res. Lett. 39, L03602, doi:10.1029/ 2011GL050709, 2012.

Servais, P., Billen, G., and Hascoet, M.C.: Determination of the biodegradable fraction of dissolved organic matter in waters, Water Res. 21,445 – 50, https://doi.org/10.1016/0043-1354(87)90192-8, 1987.

Somayajulu B. L. K., Rengarajan R., and Jani R. A.: Geochemical cycling in the Hooghly estuary, India. Mar. Chem., 79, 171–183. DOI: 10.1016/S0304-4203(02)00062-2, 2002.

Smith, B.N., and Epstein, S.: Two categories of $^{13}C/^{12}C$ ratios for higher plants, Plant Physiology, 47, 380 - 384. https://doi.org/10.1104/pp.47.3.380, 1971.

Sippo, J. Z., Maher, D. T., Tait, D. R., Holloway, C., and Santos, I. R.: Are mangroves drivers or buffers of coastal acidification? Insights from alkalinity and dissolved inorganic carbon export estimates across a latitudinal transect, Global Biogeochem. Cy., 30, 753–766. doi:10.1002/2015GB005324, 2016.

Singh, G., Ramanathan, A.L., Santra, S.C., Rajan, R.K.: Tidal control on the nutrient variability in Sundarban mangrove ecosystem, Journal of Applied Geochemistry, 18(4), 495-503, 2016.

Tait, D. R., Maher, D. T., Macklin, P. A., and Santos, I. R.: Mangrove pore water exchange across a latitudinal gradient, Geophys. Res. Lett. 43, 3334–3341. doi: 10.1002/2016GL068289, 2016.

Wangersky, P. J.: Production of dissolved organic matter. In: Marine ecology. Ed. by O. Kinne. Wiley, Chichester, 4, 115-220, 1978.

Wanninkhof, R.: Relationship between wind speed and gas exchange over the ocean, J. Geophys. Res., 97, 7373–7382, https://doi.org/10.1029/92JC00188, 1992.

Weiss, R.F.: The solubility of nitrogen, oxygen and argon in water and seawater, Deep Sea Research and Oceanographic Abstracts, 17(4), 721-735, https://doi.org/10.1016/0011-7471(70)90037-9, 1970.

Zhai, W., Dai, M., and Guo, X.: Carbonate system and CO_2 degassing fluxes in the inner estuary of Changjiang (Yangtze) River, China, Mar. Chem., 107, 342–356, https://doi.org/10.1016/j.marchem.2007.02.011, 2007.

Zhai, W.D., Dai, M.H., Cai, W.J., Wang, Y.C., and Wang. Z.H.: High partial pressure of CO_2 and its maintaining mechanism in a subtropical estuary: The Pearl River estuary, China. Mar. Chem. 93(1): 21 - 32. https://doi.org/10.1016/j.marchem.2004.07.003, 2005.

Zhang J., Yu, Z.G., Liu, S.M., Xu, H., Wen, Q.B., and Shao B. et al.: Dominance of terrigenous particulate organic carbon in the high-turbidity Shuangtaizihe estuary, Chem. Geol., 138, 211 - 219. ~~https://doi.org/10.1016/S0009-2541(97)00012-0, 1997.~~https://doi.org/10.1016/S0009-2541(97)00012-0, 1997.

Data availability

Data used in the manuscript is presented in tables (Table 2, Table 3, and Table 4) of the manuscript.

Author contributions

MKD and SK designed the study. MKD with RM and PS collected and analyzed samples. MKD and SK interpreted the data and drafted the manuscript. SKM provided facility to measure basic physicochemical parameters and DOC.

Competing interest

The author declares no conflict of interest.

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Figure Captions:

Fig. 1: Sampling locations at the (a) estuaries of Sundarbans, and (b) Hooghly estuary.

Fig. 2: Variability of (a) %DO, and (b) pH with - salinity at the Hooghly-Sundarbans systems. (Green, grey and blue colors indicate freshwater region of the Hooghly, mixing region of the Hooghly and Sundarbans, respectively).

Fig. 3: (a) Observed andDIC - salinity in the Hooghly (solid line indicates estimated concentrations due to conservative mixing values of [DIC] and), (b) $\delta^{13}C_{DIC}$ with- salinity atin the Hooghly estuary, [b] (solid line indicates estimated $\delta^{13}C_{DIC}$ due to conservative mixing), (c) ΔDIC and- Δ $\delta^{13}C_{DIC}$ atin the Hooghly estuary (PP = Phytoplankton productivity, OG = outgassing, CD = carbonate dissolution, CP = carbonate precipitation, ROMOM Res. = Respiration of organic matter), (c) [d) DIC] and $\delta^{13}C_{DIC}$ with - salinity atin the estuaries of Sundarbans, [d] TAlk and [DIC], and (e) [DOC] with$\delta^{13}C_{DIC}$ - salinity in the Hooghly-Sundarbans system.

Fig.4: Variability of (a) [POC](Green, grey and %POC/SPM with salinity in blue color indicates freshwater region of the Hooghly, (b) [POC]mixing region of the Hooghly and %POC/SPM with salinity in the Sundarbans, and (c) $\delta^{13}C_{POC}$ in the Hooghly – Sundarbans system with salinity.respectively).

Fig.5: Variability in (a) pCO_2 with salinity, (b) pCO_2 with AOU, and [c] FCO_2 with salinity in the Hooghly-Sundarbans system.

Table – 1**Fig.4**: (a) DOC - salinity in the Hooghly-Sundarbans, (b) DOC - pCO_2 in the Hooghly, (c) DOC - pCO_2 in the Sundarbans, and (d) DOC - POC in the Hooghly-Sundarbans (Green, grey and blue color indicates freshwater region of the Hooghly, mixing region of the Hooghly and the Sundarbans, respectively).

Fig.5: (a) SPM - salinity, (b) POC - salinity, (c) %POC/SPM – salinity, (d) ΔPOC - Δ $\delta^{13}C_{POC}$ in Hooghly (RR - aerobic respiration, SD - deposition at sub-tidal sediment, SR - surface runoff, FR - freshwater runoff and PP - phytoplankton productivity), (e) POC - pCO_2 in Hooghly and (f) POC - pCO_2 in Sundarbans (Green, grey and blue color indicates

freshwater region of the Hooghly, mixing region of the Hooghly and the Sundarbans, respectively).

Fig. 6: (a) ECO_2 - AOU in the Sundarbans (b) pCO_2 - salinity in the Sundarbans (c) ECO_2 - AOU in the Hooghly (d) pCO_2 - salinity in the Hooghly (e) FCO_2 - salinity in the Hooghly, and (f) FCO_2 - salinity in the Sundarbans (Green, grey and blue color indicate freshwater region of the Hooghly, mixing region of the Hooghly and the Sundarbans, respectively).

Table - 1: General characteristic of the Hooghly estuary and the estuaries of Sundarbans.

Parameters	**Hooghly**	**Sundarbans**
Nutrients (postmonsoon)	DIN: 14.72 ± 1.77 to 27.20 ± 2.05µM DIP: 1.64 ± 0.23 to 2.11 ± 0.46µM DSi: 77.75 ± 6.57 to 117.38 ± 11.54µM (Mukhopadhyay et al., 2006)	DIN: 11.70 ± 7.65µM DIP: 1.01 ± 0.52µM DSi: 75.9 ± 36.9µM (Biswas et al., 2004)
Chl*a* (postmonsoon)	Chl-a: 2.35 – 2.79 mgm^{-3} (Mukhopadhyay et al., 2006)	Chl*a*: 7.88 ± 1.90 mgm^{-3} (Dutta et al., 2015)
Population density	North 24 Parganas and Hooghly: 2500 km^{-2}, Kolkata: 22000 km^{-2}, Howrah: 3300km^{-2}, South 24 Parganas: 820 km^{-2}	No major Cities and town
Freshwater discharge (postmonsoon)	3070 - 7301 million m^3 (Rudra et al., 2014)	No information available
Catchment area	6 x $10^4 km^2$ (Sarkar et al., 2017)	No information available
Industrial and municipal wastewater discharge	1153.8Million L d^{-1} (Ghosh, 1973; Khan, 1995)	No information available
Dissolved metal flux	Increased from 230 – 1770% annually (Samanta and Dalai, 2018)	No information available

Table - 2: Physicochemical parameters, inorganic and organic C related parameters, and CO_2 exchange fluxes across water-atmosphere at the estuaries of Sundarbans. Here, water temperature (W_T), DO, isotopic compositions, DIC, DOC, POC, pCO_2 and FCO_2 are presented in '°C' 'mgL^{-1}', '‰', '~~mM', 'mgL^{-1}~~'µM', 'µM', 'µM', 'µatm' and 'µmol m^{-2} hr^{1}', respectively.

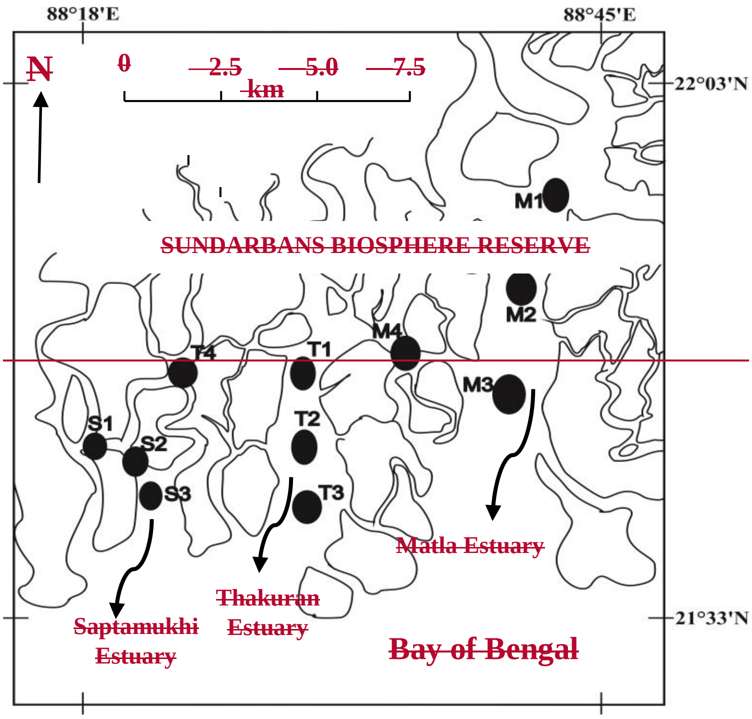
Station	W_T	Salinity	DO	pH	DIC	$\delta^{13}C_{DIC}$	DOC	POC	$\delta^{13}C_{POC}$	pCO_2	FCO_2
S1	28.50	12.74	6.65	8.02	~~1.78~~1780	– 5.59	~~3.34~~ 278	154	– 22.85	536	26.5
S2	28.00	16.02	6.65	8.02	~~1.70~~1703	– 4.33	~~3.20~~ 267	124	– 23.54	561	30.3
S3	28.00	16.69	6.61	8.12	~~1.70~~1700	– 4.29	~~2.36~~ 197	114	– 23.43	395	0.9
S4	29.00	15.25	6.46	8.01	~~1.86~~1861	– 5.27	~~3.78~~ 315	93	– 23.68	543	27.6
T1	29.00	14.30	6.56	8.05	~~1.76~~1757	– 5.57	~~3.11~~ 259	80	– 23.62	490	18.1
T2	29.00	15.51	6.74	8.07	~~1.73~~1727	– 4.79	~~2.19~~ 182	106	– 23.21	456	11.9
T3	28.50	16.55	6.46	8.11	~~1.68~~1683	– 4.39	~~1.85~~ 154	154	– 22.97	403	2.4
M1	28.00	15.14	6.99	8.07	~~1.71~~1711	– 5.93	~~3.38~~ 282	264	– 23.07	443	9.4
M2	28.00	15.14	6.91	8.12	~~1.74~~1735	– 4.63	~~2.62~~ 219	436	– 23.15	376	-2.6
M3	28.00	15.23	7.46	8.13	~~1.74~~1736	– 5.30	~~2.66~~ 222	287	– 23.62	401	1.9
M4	28.50	14.78	6.84	8.04	~~1.92~~1920	– 5.38	~~2.58~~ 215	96	– 23.82	503	20.3

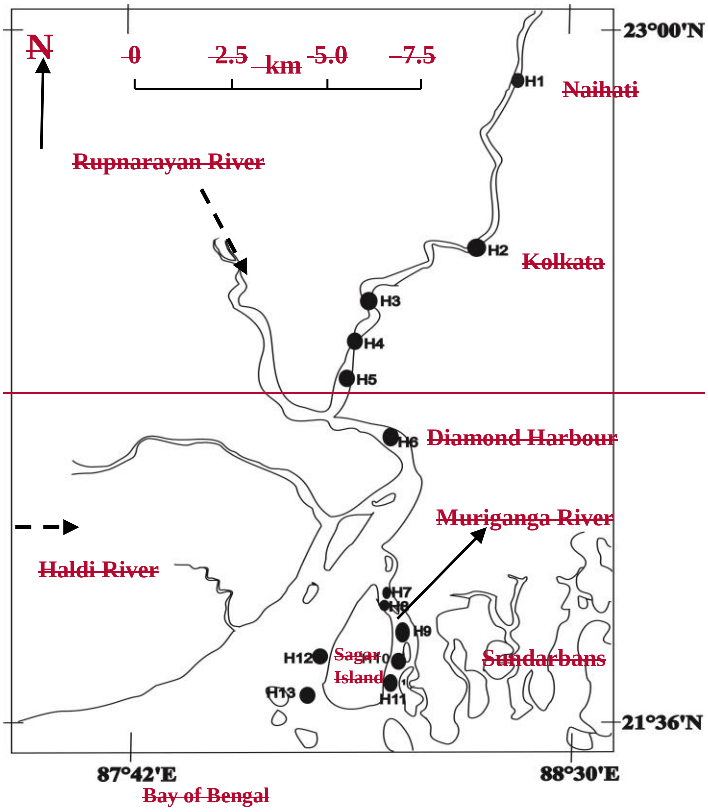
Table - ~~2~~3: Physicochemical parameters, inorganic and organic C related parameters, and CO_2 exchange fluxes across water-atmosphere at the Hooghly estuary. Here, water temperature (W_T), DO, all isotopic compositions, DIC, DOC, POC, pCO_2 and FCO_2 are presented in '°C' 'mgL^{-1}', '‰', '~~mM', 'mgL^{-1}'~~'µM', 'µM', 'µM', 'µatm' and 'µmol m^{-2} hr^{1}', respectively.

Station	W_T	Salinity	DO	pH	DIC	$\delta^{13}C_{DIC}$	DOC	POC	$\delta^{13}C_{POC}$	pCO_2	FCO_2
H1	32.0	0.04	6.29	7.92	~~2.70~~ 2700	– 6.98	~~2.92~~ 244	313	– 25.34	2036	285.2
H2	33.0	0.07	6.11	7.71	~~1.68~~ 1678	– 8.38	~~3.65~~ 304	177	– 25.19	2316	343.8
H3	31.0	0.08	6.45	7.83	~~2.50~~ 2498	– 6.70	~~2.82~~ 235	286	– 25.95	2490	355.4
H4	31.0	0.13	5.24	7.73	~~2.45~~ 2446	– 7.38	~~2.91~~ 243	254	– 25.40	2691	389.2
H5	31.0	0.19	5.38	7.77	~~2.36~~ 2355	– 7.56	~~4.08~~ 340	130	– 25.67	2123	293.1
H6	30.5	0.32	5.66	7.31	~~2.16~~ 2157	– 8.61	~~3.70~~ 308	116	– 24.07	4678	717.5
H7	31.5	5.83	6.71	7.68	~~1.83~~ 1829	– 6.79	~~7.95~~ 662	145	– 24.70	1184	132.0
H8	31.0	5.19	7.14	7.31	~~2.02~~ 2023	– 6.78	~~4.25~~ 354	139	– 23.47	3153	455.8
H9	31.5	9.08	6.62	7.90	~~1.92~~ 1915	– 6.08	~~3.98~~ 332	161	– 23.53	665	44.9
H10	31.5	9.72	6.17	8.08	~~1.79~~ 1787	– 5.78	~~2.99~~ 249	95	– 24.06	452	10.1
H11	31.0	8.43	6.37	8.07	~~1.98~~ 1977	– 7.21	~~4.29~~ 358	95	– 25.94	486	15.6
H12	31.5	5.83	7.40	8.29	~~1.87~~ 1871	– 6.60	~~3.12~~ 260	133	– 26.28	274	-19.3
H13	31.0	10.37	7.00	8.24	~~1.84~~ 1843	– 5.57	~~4.72~~ 394	129	– 24.72	267	-19.8

Table - 34: The [DIC] concentrations and $\delta^{13}C_{DIC}$ of groundwater (GW) and pore-water (PW) samples collected from thearound Hooghly-Sundarbans system.

Ecosystem	**Station**	**[DIC] (mM (µM)**	**$\delta^{13}C_{DIC}$ (‰)**
Hooghly	H3GW	11.7611756	– 12.66
	H4GW	6.236230	– 7.85
	H5GW	6.336327	– 8.96
	H6GW	7.037026	– 11.27
	H7GW	5.665655	– 6.91
	H11GW	9.129115	– 7.67
	H12GW	6.866858	– 7.49
	H13GW	7.267258	– 7.21
	Gangasagar GW	7.257246	– 6.67
Sundarbans	Lothian GW	7.527524	– 6.84
	Lothian PW	13.4313425	– 18.05
	Kalash GW	13.5913599	– 6.69
	Virat Bazar GW	8.308300	– 10.56


88°18'E
88°45'E
22°03'N
21°33'N
N
0
2.5
5.0
7.5
km
SUNDARBANS BIOSPHERE RESERVE
M1
M2
M3
M4
T1
T2
T3
T4
S1
S2
S3
Matla Estuary
Thakuran Estuary
Saptamukhi Estuary
Bay of Bengal


23°00'N
21°36'N
87°42'E
88°30'E
N
0
2.5
5.0
7.5
km
H1
Naihati
Rupnarayan River
H2
Kolkata
H3
H4
H5
H6
Diamond Harbour
Muriganga River
Haldi River
H7
H8
H9
H12
Sagar Island
H11
H13
Sundarbans
Bay of Bengal

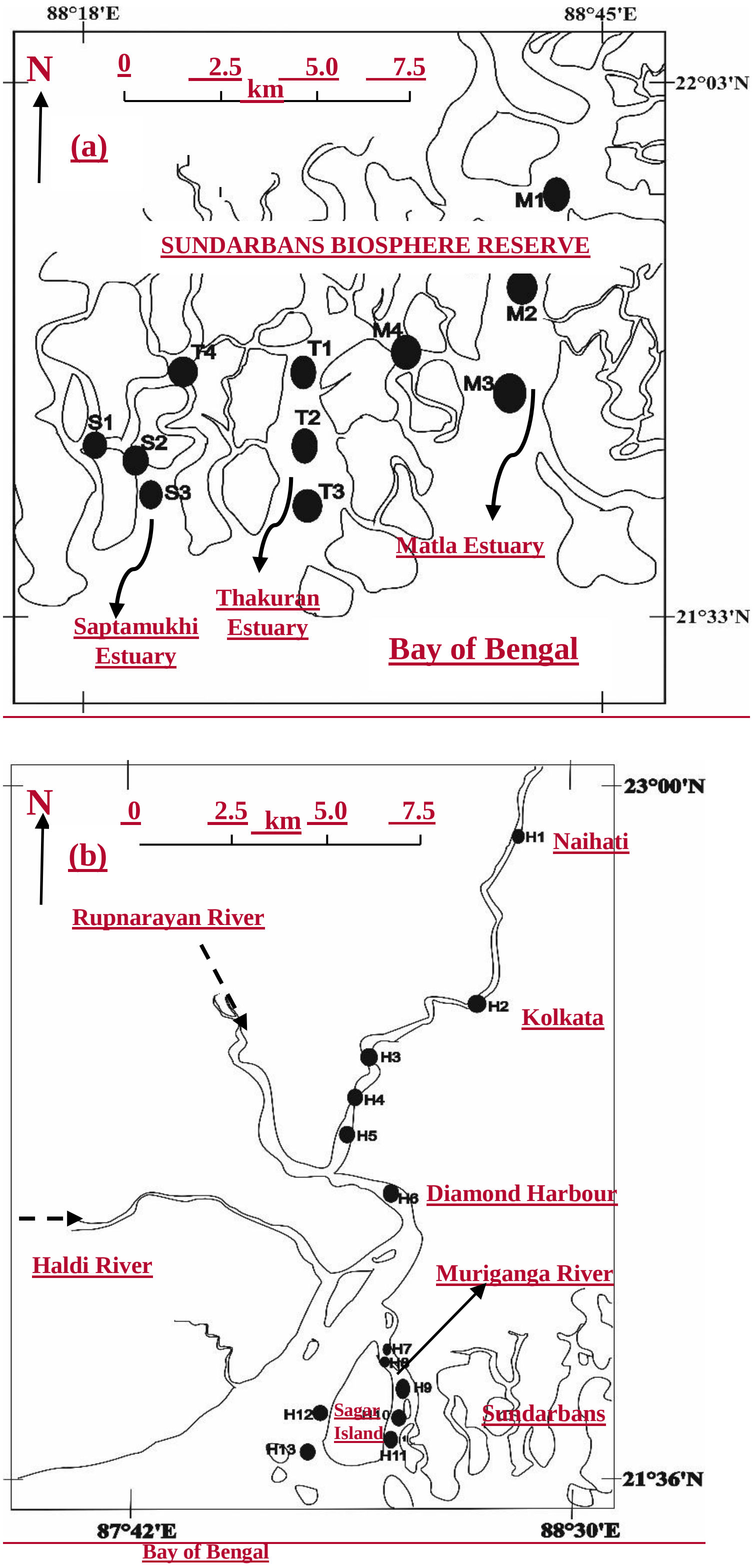
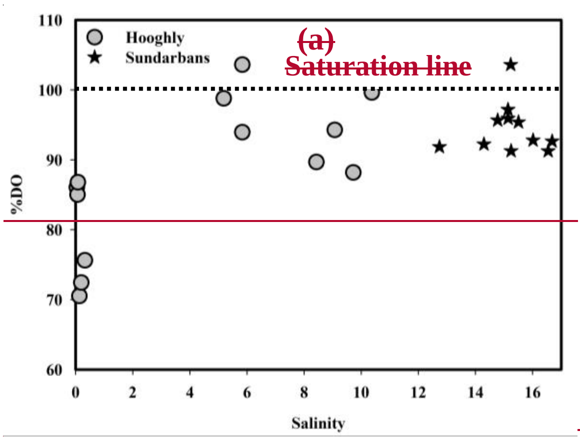
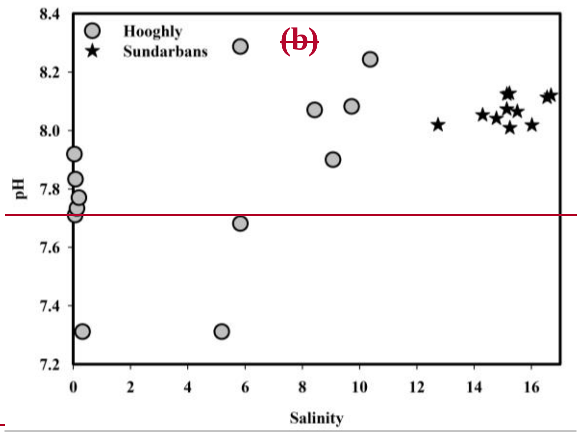

88°18'E
88°45'E
22°03'N
21°33'N
N
0
2.5
5.0
7.5
km
(a)
M1
SUNDARBANS BIOSPHERE RESERVE
M2
M4
T4
T1
M3
S1
T2
S2
S3
T3
Matla Estuary
Thakuran Estuary
Saptamukhi Estuary
Bay of Bengal
23°00'N
N
0
2.5
km
5.0
7.5
(b)
H1
Naihati
Rupnarayan River
H2
Kolkata
H3
H4
H5
H6
Diamond Harbour
Haldi River
Muriganga River
H7
H9
H12
Sagar Island
Sundarbans
H13
H11
21°36'N
87°42'E
88°30'E
Bay of Bengal

Fig.-1


Hooghly
Sundarbans
(a)
Saturation line
%DO
Salinity
110
100
90
80
70
60
0
2
4
6
8
10
12
14
16


Hooghly
Sundarbans
(b)
pH
Salinity
8.4
8.2
8.0
7.8
7.6
7.4
7.2
0
2
4
6
8
10
12
14
16

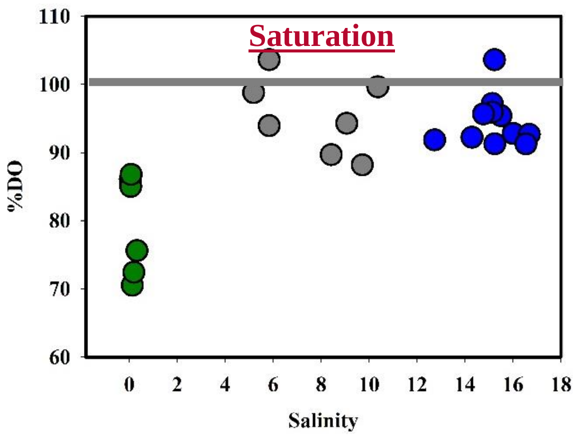
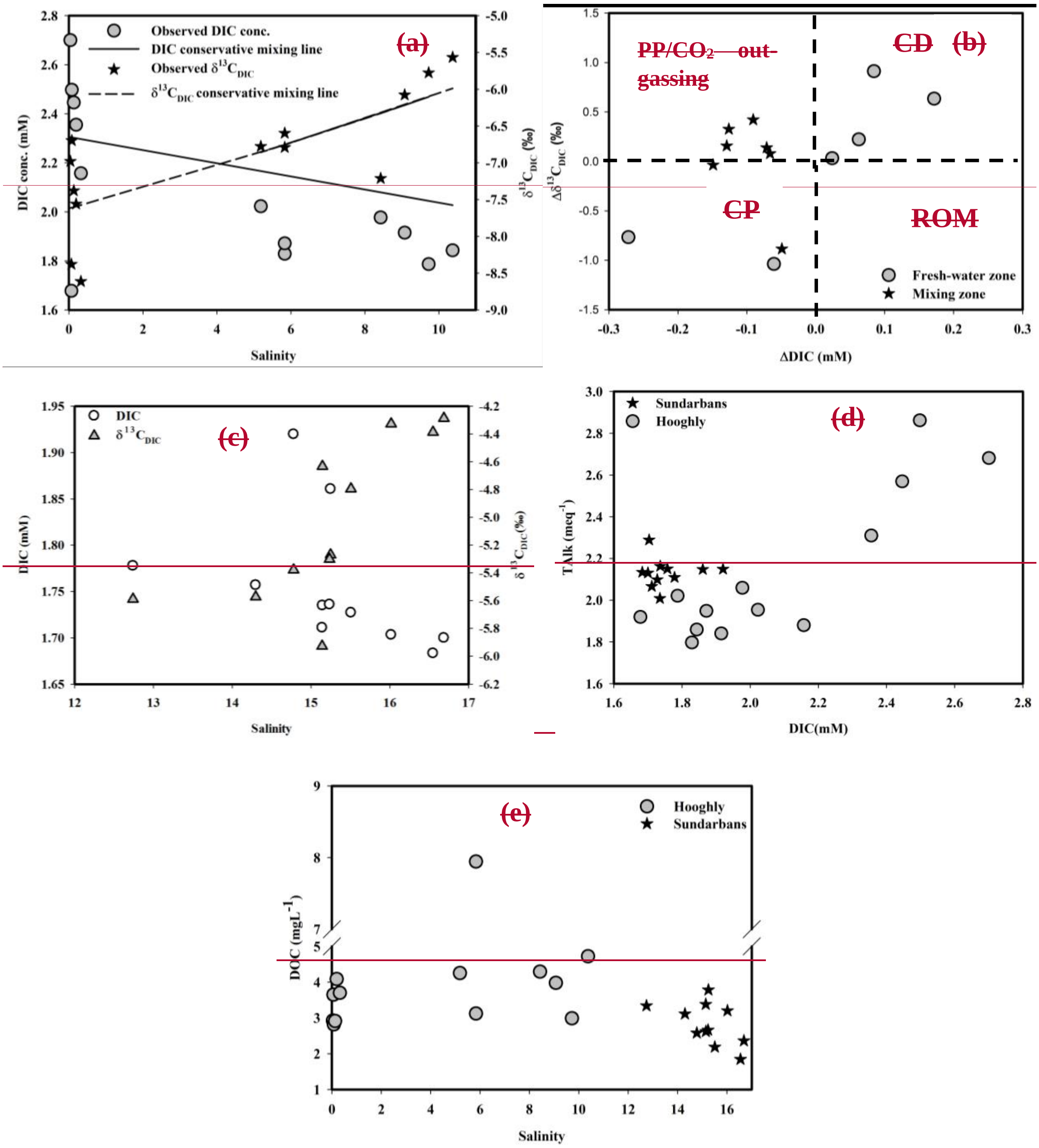

Saturation
%DO
Salinity
110
100
90
80
70
60
0
2
4
6
8
10
12
14
16
18

Fig.-2


(a)
Observed DIC conc.
DIC conservative mixing line
Observed δ13CDIC
δ13CDIC conservative mixing line
DIC conc. (mM)
δ13CDIC (‰)
Salinity
(b)
PP/CO2 out-gassing
CD
CP
ROM
Fresh-water zone
Mixing zone
Δδ13CDIC (‰)
ΔDIC (mM)
(c)
DIC
δ13CDIC
DIC (mM)
δ 13CDIC(‰)
Salinity
(d)
Sundarbans
Hooghly
TAlk (meq-1)
DIC(mM)
(e)
Hooghly
Sundarbans
DOC (mgL-1)
Salinity

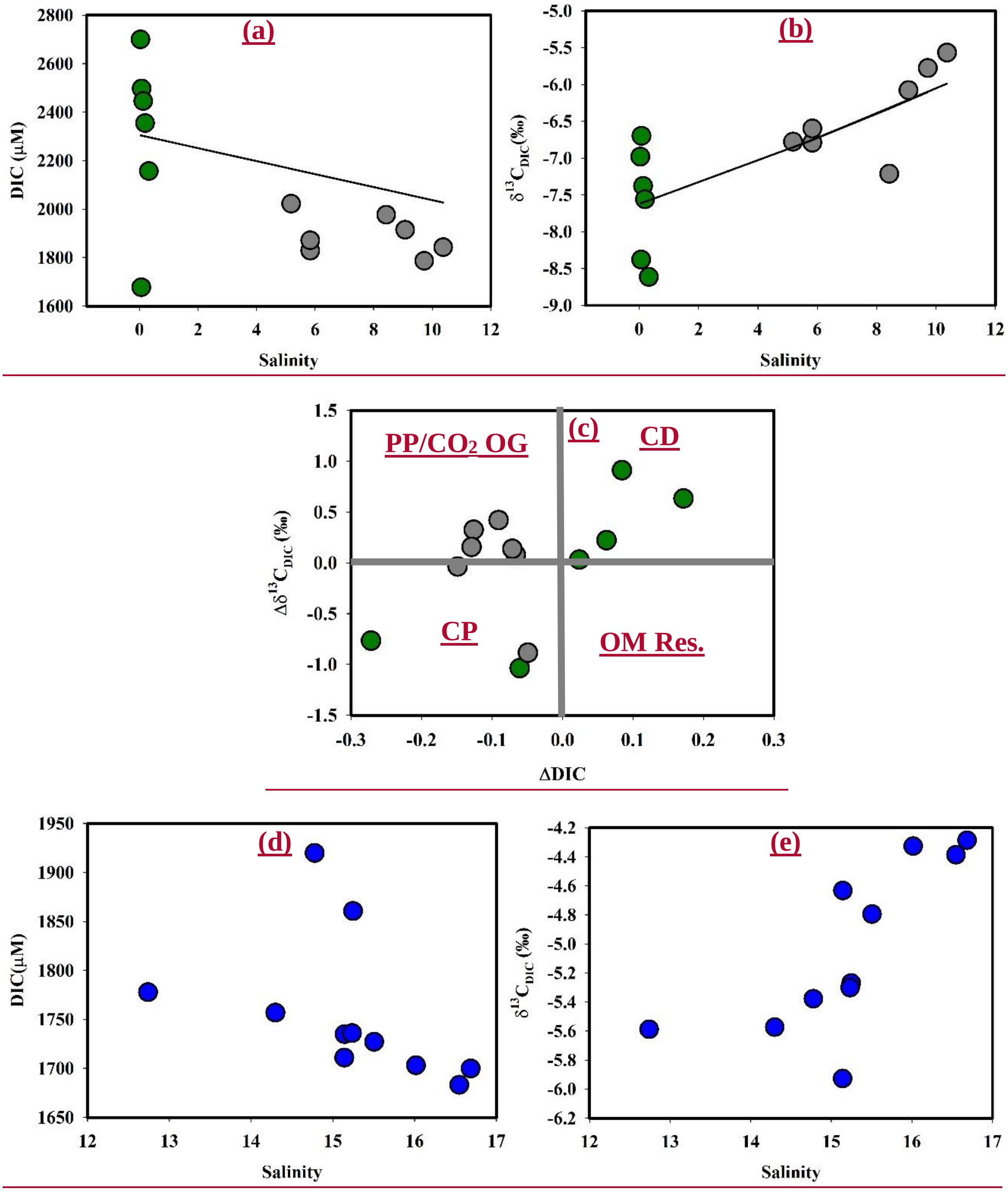
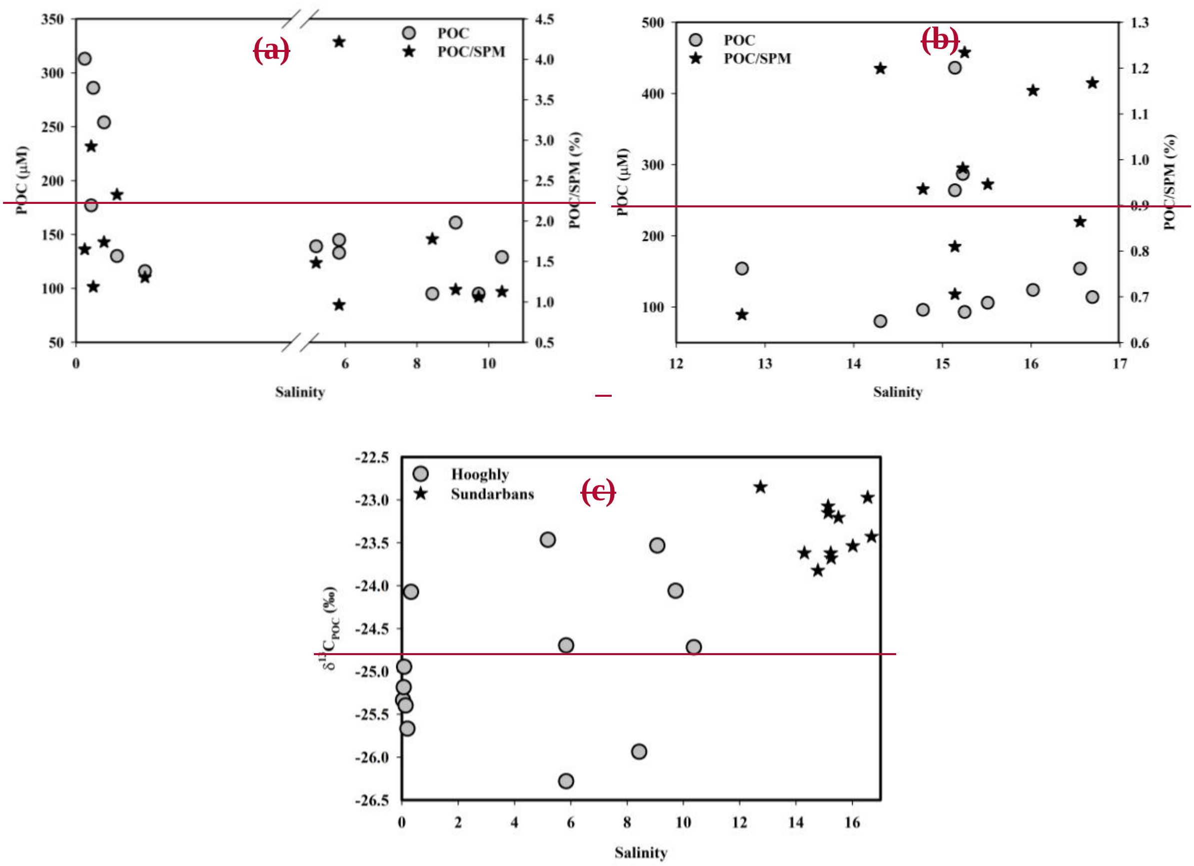

(a)
DIC (μM)
2800
2600
2400
2200
2000
1800
1600
0
2
4
6
8
10
12
Salinity
(b)
$\delta^{13}C_{DIC}$(‰)
-5.0
-5.5
-6.0
-6.5
-7.0
-7.5
-8.0
-8.5
-9.0
0
2
4
6
8
10
12
Salinity
(c)
PP/CO_2 OG
CD
CP
OM Res.
$\Delta\delta^{13}C_{DIC}$ (‰)
1.5
1.0
0.5
0.0
-0.5
-1.0
-1.5
-0.3
-0.2
-0.1
0.0
0.1
0.2
0.3
ΔDIC
(d)
DIC(μM)
1950
1900
1850
1800
1750
1700
1650
12
13
14
15
16
17
Salinity
(e)
$\delta^{13}C_{DIC}$ (‰)
-4.2
-4.4
-4.6
-4.8
-5.0
-5.2
-5.4
-5.6
-5.8
-6.0
-6.2
12
13
14
15
16
17
Salinity

Fig.-3


(a)
POC
POC/SPM
POC (µM)
POC/SPM (%)
Salinity
(b)
POC
POC/SPM
POC (µM)
POC/SPM (%)
Salinity
(c)
Hooghly
Sundarbans
$\delta^{13}C_{POC}$ (‰)
Salinity

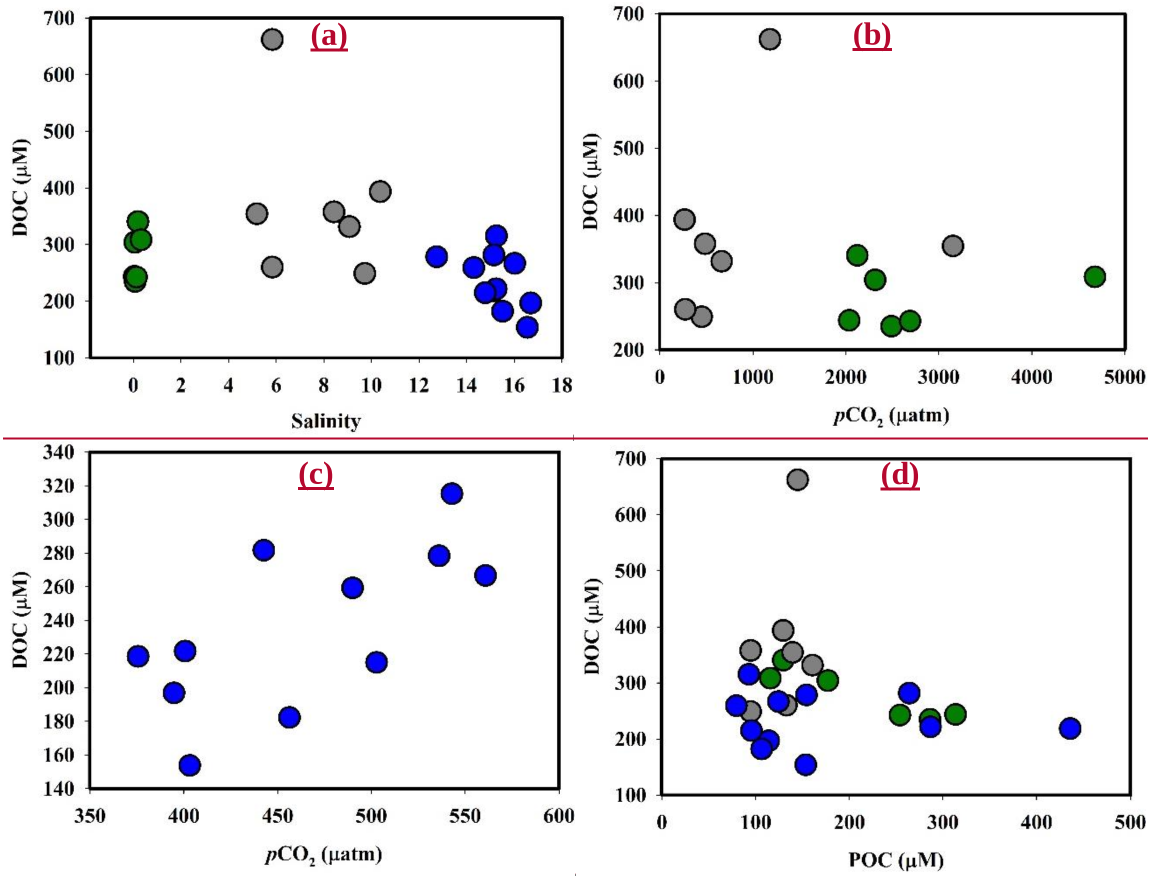
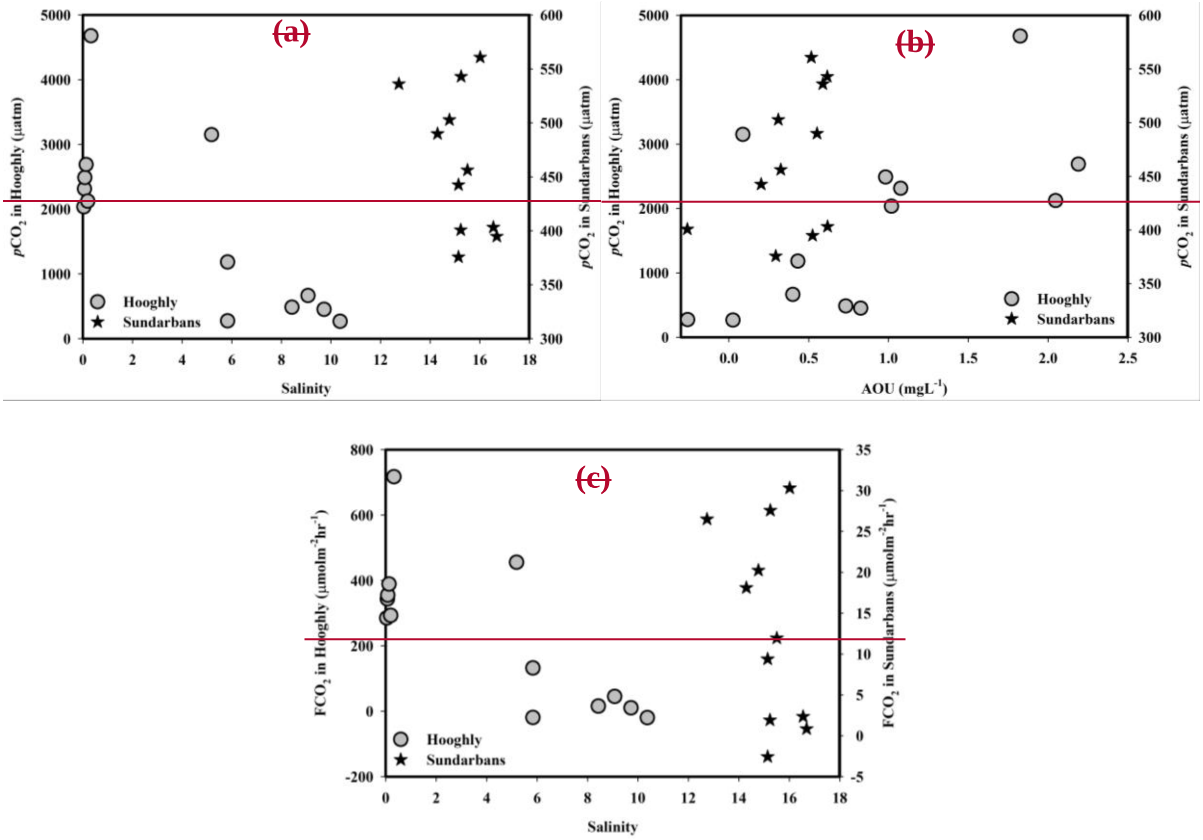
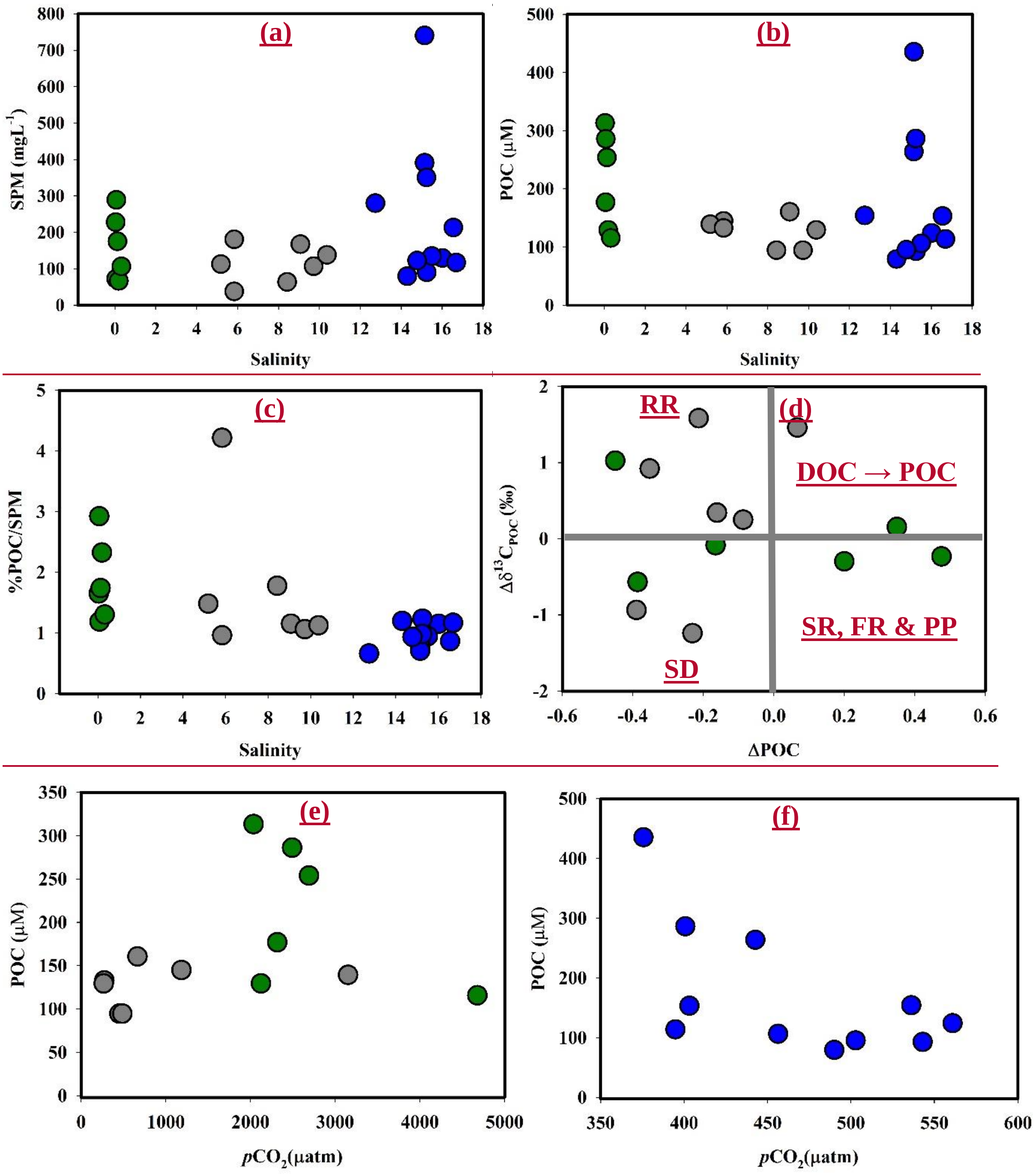

(a)
DOC (µM)
Salinity
(b)
DOC (µM)
pCO$_2$ (µatm)
(c)
DOC (µM)
pCO$_2$ (µatm)
(d)
DOC (µM)
POC (µM)

Fig.-4



(a)
pCO2 in Hooghly (µatm)
pCO2 in Sundarbans (µatm)
Salinity
Hooghly
Sundarbans
0
1000
2000
3000
4000
5000
300
350
400
450
500
550
600
0
2
4
6
8
10
12
14
16
18
(b)
pCO2 in Hooghly (µatm)
pCO2 in Sundarbans (µatm)
AOU (mgL-1)
Hooghly
Sundarbans
0.0
0.5
1.0
1.5
2.0
2.5
(c)
FCO2 in Hooghly (µmolm-2hr-1)
FCO2 in Sundarbans (µmolm-2hr-1)
Salinity
Hooghly
Sundarbans
-200
0
200
400
600
800
-5
0
5
10
15
20
25
30
35



(a)
SPM (mgL⁻¹)
Salinity
(b)
POC (µM)
Salinity
(c)
%POC/SPM
Salinity
(d)
RR
DOC → POC
SR, FR & PP
SD
Δδ13C POC (‰)
ΔPOC
(e)
POC (µM)
pCO2(µatm)
(f)
POC (µM)
pCO2(µatm)

Fig. 5

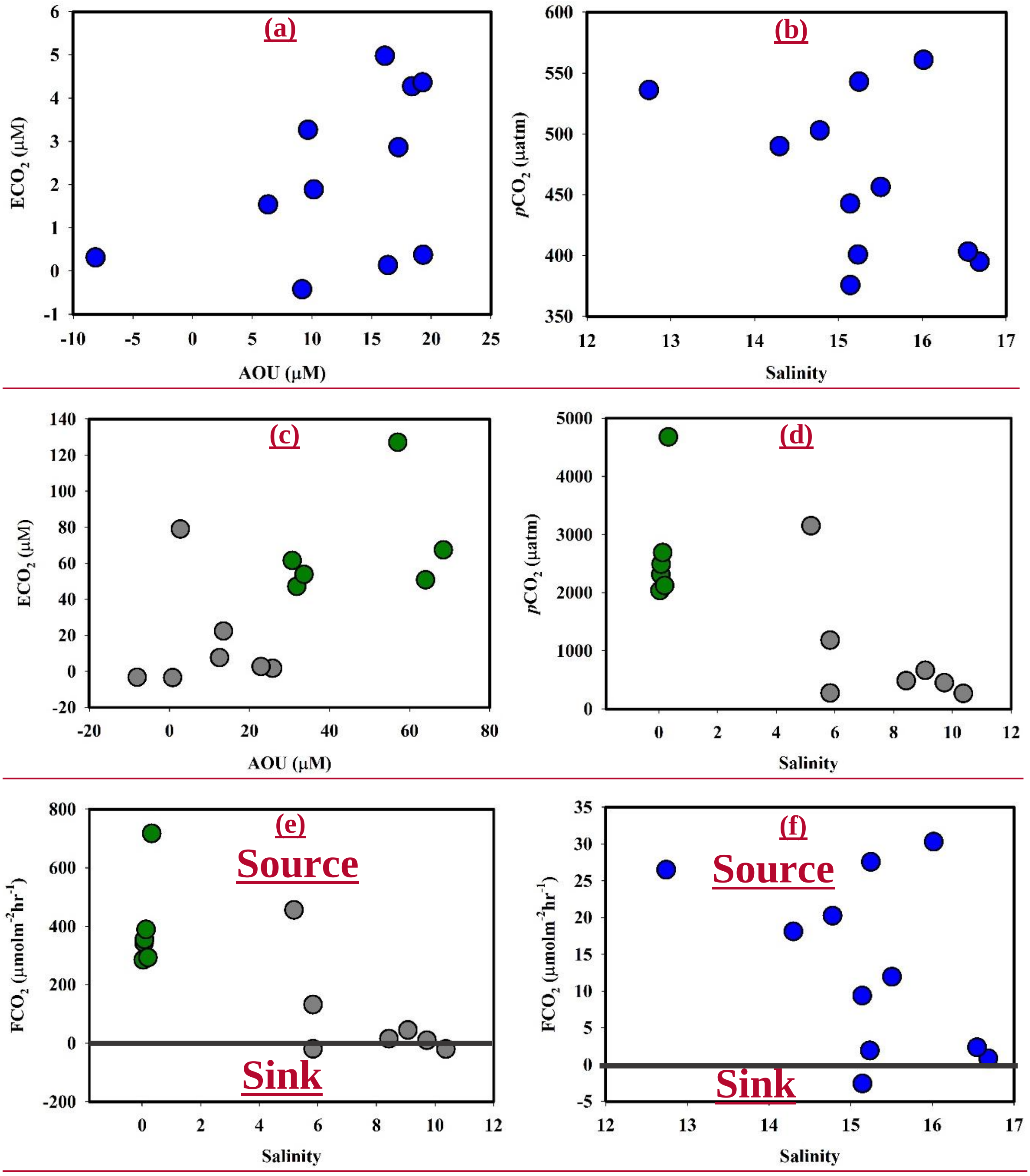

(a)
ECO2 (µM)
AOU (µM)
(b)
pCO2 (µatm)
Salinity
(c)
ECO2 (µM)
AOU (µM)
(d)
pCO2 (µatm)
Salinity
(e)
Source
Sink
FCO2 (µmolm-2hr-1)
Salinity
(f)
Source
Sink
FCO2 (µmolm-2hr-1)
Salinity

Fig.6