Response to reviewer – 1

Overview

The authors reviewed carbon biogeochemistry in the India Coast, presented its spatiotemporal variability and discussed the potential drivers. As a continuum between land and ocean, estuarine system is important while suffering a great spatiotemporal heterogeneity. Such study could help constrain this variation and better understand its essential role on global carbon budget. However, this manuscript of its current version still saves a large space for improvement, particularly from its data interpretation and content structure perspectives. I am afraid this manuscript will need a thorough revision to fit for the journal.

Response: *Thanks for the encouraging comments about the manuscript and valuable suggestions. We have included your suggestions in the revised manuscripts to improve the quality of the manuscript.*

Major comments

(1) Traceable data is vital for a review article. Unfortunately, there is no clear pathway(s) for data sources in this manuscript. For example, how many sampling stations, how many observations for each estuary, sampling time, etc.? We even do not know if the authors are presenting annual average data or just one-time surveyed data. Why there is no standard deviation for each estuary data in figures? A proper conclusive data table (can be supplementary material) is badly needed to show its rigor and reliability.

Response: *We agree and have added an excel file comprising all data used to prepare the manuscript along with the references as supplementary file. In the file, we have compiled all information related to that data wherever available.*

(2) Following the first major concern, then the data interpretation is problematic. First, the data visualization needs improvement, why only list Sundarbans and Hooghly Estuary sampling stations in Fig.1? Differences on estuaries or dry/wet cycle cannot be well distinguished in both figures and supplementary figures. Second, the way of data processing is also unclear, for example, how do the authors conduct statistical analysis, t-test? two-way ANOVA? any process to meet the assumptions? In supplementary figures, several estuaries are excluded to meet a high p-value relationship seems arbitrary and misleading, same as the threshold 6800 μatm for pCO_2. Is there any reason/accordance to do so?

Response: *The Hooghly and Sundarbans are separately shown in Fig. 1. As you can see three major estuaries (Saptamukhi, Thakuran, and Matla) are included within the Sundarbans. Additionally, the Hooghly and estuaries of Sundarbans are closely associated which are covered thoroughly by Dutta et al., (2019, 2021) from the upstream to downstream and mean data is used in the paper. Considering limited space in the given map and inclusion of the sampling points, the Hooghly-Sundarbans system was plotted as a sub-set in Fig. 1.*

Regarding figures, we have now revised the figures for the revised manuscript. In the original manuscript, we used simple regression analysis to evaluate dependency between C and other parameters. But, yes, as you said each parameter may be related, in the revised manuscript we have included 't' test and PCA analysis wherever applicable as follows:

Table – Results for statistical 't' test analysis between the mean of available data. The analysis is performed at 95% confidence level.

Parameters	Inter-seasonal comparison		Inter-BB & AS estuaries comparison	
	BB estuaries	**AS estuaries**	**Wet Season**	**Dry Season**
Salinity	$p = 0.55$	$p < 0.0001$**	$p = 0.003$**	$p = 0.84$
%DO	$p = 0.60$	$p = 0.31$	$p = 0.07$	$p = 0.001$**
pH	$p = 0.10$	$p = 0.006$**	$p < 0.0001$**	$p = 0.012$**
DIC	$p = 0.59$	NA	$p = 0.008$**	NA
$\delta^{13}C_{DIC}$	$p = 0.69$	NA	$p = 0.000$**	NA
DOC	$p = 0.32$	NA	$p = 0.45$	NA
POC	$p = 0.02$**	$p = 0.02$**	$p = 0.17$	$p = 0.06$
$\delta^{13}C_{POC}$	$p = 0.04$**	NA	$p = 0.21$	NA
pCO_2	$p = 0.07$	NA	$p = 0.10$	NA
FCO_2	$p = 0.41$	NA	$p = 0.29$	NA
CH_4	$p = 0.39$	$p = 0.38$	$p = 0.03$**	$p = 0.11$
FCH_4	$p = 0.05$**	$p = 0.27$	$p = 0.13$	$p = 0.16$

***Statistically significant at $\alpha = 0.05$.*

Principal component analysis (PCA) was performed in order to identify major controlling factors for dissolved and particulate C as well as variability of trace gases (CO_2 and CH_4) in the major estuaries of India. The PCA was performed for 7 parameters (DIC and its isotopic composition, DOC, POC and its isotopic compositions, pCO_2 and CH_4 concentration) based on the availability of other parameters (no of dams, population density, precipitation, estuarine area, discharge, catchment area, salinity, %DO, and pH). Moreover, due to scarcity of data during the dry season the PCA analysis is restricted only during the wet season. The principal component (PC) with eigen values >1 was considered for further analysis and only two factors were identified in this case as given below:

Table: Results for the Principal Component Analysis of estuarine carbon biogeochemistry of India.

	BB estuaries		**AS estuaries**	
Eigen value	2.90	2.07	4.40	1.65
Explained variance (%)	41.48	29.65	62.83	23.58
Cumulative	41.48	71.11	62.63	86.41
Variable	**PC1**	**PC2**	**PC1**	**PC2**
DIC	0.89	0.14	0.92	0.21
$\delta^{13}C_{DIC}$	0.73	0.27	-0.65	0.67
DOC	0.40	0.55	0.58	0.72
POC	-0.76	0.46	0.76	-0.61
$\delta^{13}C_{POC}$	-0.82	-0.35	0.92	-0.21
pCO_2	-0.35	0.80	0.87	0.46

CH_4	*-0.23*	*0.84*	*0.78*	*0.12*

The PC1 accounts for ~41.48% and 62.63% variability for the BB and AS estuaries with strong factor loadings of DIC, $\delta^{13}C_{DIC}$, POC and $\delta^{13}C_{POC}$ for BB estuaries and DIC, $\delta^{13}C_{DIC}$, DOC, POC, $\delta^{13}C_{POC}$, pCO_2 and CH_4 for the AS estuaries. For the BB estuaries, the factor loading might be links with the biological productivity; however, for the AS estuaries the factor loading might be due to POC and DOC decomposition and its associated productions of CO_2 and CH_4. The oxygenated estuary supports aerobic degradation of organic matter producing CO_2 and considered to be restrain anaerobic degradation. However, it should also be noted that CH_4 production in the isolated anoxic microhabitats of sinking particulate organic matter, in well-oxygenated water column, have been observed in the open ocean (see Reeburgh, 2007). The PC2 represents 71.11% and 86.41% of total variance for the BB and AS estuaries with relatively strong factor loading for DOC, pCO_2 and CH_4 for the BB estuaries and $\delta^{13}C_{DIC}$, DOC, and POC for the AS estuaries. The former might be associated with aerobic and anaerobic degradations of DOC, however, the later might be linked with aerobic decomposition of DOC and POC and their impact on variability of $\delta^{13}C_{DIC}$.

Thereafter, Pearson correlation analysis was performed for the factor loadings of the estuarine data with the number of dams, estuarine area, population density, mean annual discharge, catchment area, salinity, pH and %DO. The Pearson correlation matrix for the BB estuaries suggests PC1 is strongly controlled by area of the estuary while PC2 is controlled by the cumulative interactions between salinity, pH as well as catchment area. For the AS estuaries, despite the PC1 is controlled by no of dams, catchment area, salinity and %DO but the PCA analysis failed to highlight any controlling factor for the PC2.

Correlation Matrix (Pearson) for the BB estuaries:

Variables	Dams	Population (/km2)	Area	Discharge	Precipitation	Catchment area (x 103 km2)	Salinity	%DO	pH	PC1	PC2
Dams	**1**	-0.373	**0.805**	**0.934**	-0.052	0.318	-0.368	-0.272	**-0.675**	-0.420	0.528
Population(/km2)	-0.373	**1**	-0.373	-0.362	-0.083	0.155	0.490	0.161	0.293	0.108	-0.209
Area	**0.805**	-0.373	**1**	**0.766**	0.128	0.282	-0.370	-0.439	**-0.675**	**-0.605**	0.596
Discharge	**0.934**	-0.362	**0.766**	**1**	0.169	0.194	-0.496	-0.464	**-0.743**	-0.570	0.541
Precipitation	-0.052	-0.083	0.128	0.169	**1**	-0.402	-0.376	-0.429	-0.135	-0.547	0.047
Catchment area (x 10^3 km2)	0.318	0.155	0.282	0.194	-0.402	**1**	-0.181	0.030	-0.273	0.465	**0.720**
Salinity	-0.368	0.490	-0.370	-0.496	-0.376	-0.181	**1**	**0.680**	**0.748**	0.175	**-0.657**
%DO	-0.272	0.161	-0.439	-0.464	-0.429	0.030	**0.680**	**1**	**0.848**	0.516	-0.539
pH	**-0.675**	0.293	**-0.675**	**-0.743**	-0.135	-0.273	**0.748**	**0.848**	**1**	0.412	**-0.741**
PC1	-0.420	0.108	**-0.605**	-0.570	-0.547	0.465	0.175	0.516	0.412	**1**	0.000
PC2	0.528	-0.209	0.596	0.541	0.047	**0.720**	**-0.657**	-0.539	**-0.741**	0.000	**1**

Values in bolded digits are statistically significant at alpha = 0.05

Correlation Matrix (Pearson) for the AS estuaries:

Variables	Dams	Population (/km2)	Area	Discharge	Catchment area (x 103 km2)	Salinity	%DO	pH	PC1	PC2
Dams	**1**	-0.092	0.518	0.686	-0.713	-0.554	**-0.890**	0.550	**0.821**	0.224
Population (/km2)	-0.092	**1**	0.253	-0.202	-0.550	-0.376	-0.271	0.184	0.457	-0.509
Area	0.518	0.253	**1**	**0.828**	-0.502	-0.491	-0.569	0.560	0.490	-0.552
Discharge	0.686	-0.202	**0.828**	**1**	-0.455	-0.380	-0.645	0.696	0.412	-0.353
Catchment area (x 103 km2)	-0.713	-0.550	-0.502	-0.455	**1**	0.729	**0.938**	-0.699	**-0.940**	0.265
Salinity	-0.554	-0.376	-0.491	-0.380	0.729	**1**	0.687	-0.319	**-0.777**	0.177
%DO	**-0.890**	-0.271	-0.569	-0.645	**0.938**	0.687	**1**	-0.685	**-0.935**	0.100

pH	0.550	0.184	0.560	0.696	-0.699	-0.319	-0.685	**1**	0.501	-0.531
PC1	**0.821**	0.457	0.490	0.412	**-0.940**	**-0.777**	**-0.935**	0.501	**1**	0.000
PC2	0.224	-0.509	-0.552	-0.353	0.265	0.177	0.100	-0.531	0.000	**1**

Values in bolded digits are statistically significant at alpha = 0.05

In the revised manuscript, without eliminating any data we have included the PCA analysis (as mentioned above) to highlight major controlling factors for estuarine C biogeochemistry. We hope it will be well accepted to the reviewers as well as editors.

Also, I have a feeling that the authors messed with riverine and estuarine data. For example, in Line 815 the "~10.30Tg C yr^{-1}" belongs to riverine export fluxes (Krishna et al., 2019) rather than "export fluxes from Indian estuaries", and the following discussion (Lines 819–831) is all about riverine C exports. Accordingly, in Fig. 7 export flux values may put in wrong place. Similarly, I do not think there are so many dams built in coastal estuaries list in Table 1. This is the reason why readers are curious about the data details, if so, I would suggest the authors clarify each estuary area/coordinates and further check about the data.

Response: *Yes, we have crosschecked the export flux data with the Krishna et al. (2019). The data in Line no – 815 of the manuscript belongs to riverine export fluxes to the northern Bay of Bengal. Thanks for pointing it out. We have removed the value in the revised manuscript.*

Yes, in Fig. 7 export fluxes have been placed in the wrong location We have decided to remove the figure from the revised manuscript.

The salinity of the observed estuaries never reached zero (salinity = 0.04 – 23.91). The paper from where most of the data have been picked up is mentioned the data as for "Indian Monsoonal Rivers" but based on the observed salinity we belief the data points are better represented as estuarine data points (better to say freshwater to mixing regime data points) rather than ideal riverine data and that's why we selected the title of the paper as "Carbon biogeochemistry of the Indian estuaries". But for the Hooghly estuary and estuaries of Sundarbans, data for the freshwater to marine regime data were included.

The exact sampling coordinate for all the estuaries are not available from the supportive literature. But based on the availability, we have updated the revised manuscript.

(3) The manuscript structure is organized in a research article format instead of a review. In addition, the separated discussions on DIC, DOC, POC, CO_2, CH_4 read super repetitive and distracting. In fact, carbon biogeochemistry is comprehensive and synthesized, drivers (e.g. hydrologic, biochemical, etc.) on any single carbon species would further impact on other carbon interactivities and then the entire carbon budget. Re-organization of manuscript structure to look at the drivers more synthetically is highly recommended.

Response: *Yes, we have noted the points and have improved the revised manuscript accordingly. All C parameters are interlinked; keeping that in mind we have included PCA in the revised manuscript (as described above).*

(4) Many important information is missing, such as temperature gradient, wind speeds, net ecosystem productions, submarine groundwater discharge rates, two end-members values, etc., these are decisive to estuarine carbon biogeochemistry. Also, I am curious about the anthropogenic impact on estuarine carbon biogeochemistry. It seems the anthropogenic

discharges in this study are mostly referred as sewage discharges to upper rivers, then how to identify the anthropogenic carbon in lower estuarine area proportionally?

Response: *Agreed. All the information is inherently linked with estuarine carbon biogeochemistry. Unfortunately, all the above-mentioned chapters/ parameters have not been thoroughly examined in the estuarine carbon biogeochemistry research in India. Nevertheless, we have tried to better incorporate the above-mentioned factors as much as possible in the revised manuscript.*

Regarding two-end members mixing model analysis, despite Bouillon et al. (2003) and thereafter Samanta et al. (2015) and Dutta et al. (2019, 2021) identified some major governing factors for estuarine carbon biogeochemistry, however, the two end members seasonal values for all the Indian estuaries are not available which precluded us form applying these more broadly across all the data.

Regarding the degree of anthropogenic impact, we simply do not have enough information to comprehensively discuss its impact. This is why we used population density as a proxy for anthropogenic inputs in the paper. For this reason, we have de-emphasized the discussion of anthropogenic inputs by changing this section into a paragraph combined with a new "natural and anthropogenic sources" section.

Yes, the reported $\delta^{13}C_{POC}$ values provided a signal for sewage inputs in the Indian estuaries and the sewage is mainly discharged in the freshwater region of the estuary (like the Hooghly). However, it may be quite difficult to identify it in the lower estuarine area as the possibility exists for its biogeochemical modification within the upper to the lower estuarine stretch. Due to that modification the anthropogenic signal might be masked in the lower estuary and the same has been reported in the anthropogenically stressed Hooghly estuary (Dutta et al. 2019, 2021).

Line comments

Line 210: more details on "statistical analysis".

Response: *As advised 't' test and PCA test are included in the revised manuscript. It is briefly discussed in the methods section of the revised manuscript.*

Line 347: references.

Response: *Added references.*

Line 348: "DIC addition/removal" details.

Response: *Depending upon the physicochemical and micromaterial condition of the estuary, the DIC addition includes organic matter respiration and carbonate dissolution whereas DIC removal includes CO_2 outgassing, phytoplankton productivity and carbonate precipitation. We have included it in the revised manuscript.*

Line 399: should be "riverine DIC" instead of "estuarine DIC".

Response: *Yes, "riverine DIC" will be a much better term to use here. We have replaced it in the revised manuscripts.*

Line 483: the difference between "Terrestrial DOC" and "Riverine DOC"?

Response: *In aqueous systems, DOC can be added both internally as well as externally. The DOC originating from within the river is known as autochthonous DOC and typically comes from aquatic plants or algae. However, DOC originating outside the river is known as allochthonous DOC which typically comes from soils or terrestrial plants and ultimately discharges to the river.*

Line 492: where is "Fig. 12A"?

Response: *Sorry, it is s typographical error. We have removed it from the revised manuscript. Thanks for pointing out the mistake.*

Line 540-543: the purpose for comparing regional DOC/DON to POC/PON? or DOC fraction in global coastal ocean?

Response: *It was included just to compare fractions of C and N in the dissolved and particulate forms. However, we have removed it from the revised manuscript.*

Line 552-553: you cannot say this unless the data about POC/DOC from two end-members.

Response: *Yes, agree. In the revised manuscript "line no 548-558 of the pre-revised manuscript" is modified as "The DOC was dominant over POC throughout all Indian estuaries (DOC/POC >1). Additionally, a recent study on the eastern Indian estuaries showed DOC and POC inter-conversion in the anthropogenically stressed Hooghly estuary and DOC influx via mangrove leaf litter leaching in the mangrove dominated estuaries of Indian Sundarbans estuaries (Dutta et al., 2021, Ray et al., 2018)."*

Line 570: why 6800 µatm threshold?

Response: *The 6800µatm threshold was chosen simply based on the nature of the plotted data, which showed a clear breaking point. The plot showed completely different regimes between <6800 µatm and >6800 µatm conditions. While it is possible to perform a change point analysis to statistically determine thresholds with higher precision, the outcome is likely to be very similar. We are not suggesting that 6800 µatm is a quantitatively significant value to consider with respect to diverging estuarine biogeochemical behaviours, but rather, make the case that by splitting the dataset with this arbitrary threshold we can observe different trends. We have softened our description of this relationship to sound less definitive and more exploratory. Additionally, in the revised manuscript we have used PCA to find out the major controlling factors for carbon dynamics of the Indian estuaries.*

Line 579: further explain "a decrease of aerobic bacterial activity with increasing DOC"

Response: *We mean that the increasing DOC load may inhibit bacterial respiration and decreases the* CO_2 *production rate. We have added this clarification to the manuscript.*

Line 616: further explain "freshwater mixing is not the major driver of POC", as it shows lower salinity with higher POC and 13C values.

Response: *Yes, true. Despite low salinity being integrated with higher POC and $\delta^{13}C_{POC}$ values, the relationship between salinity POC and $\delta^{13}C_{POC}$ are not significant (Fig. S9). This indicates mixing between marine and freshwater is not controlled primarily by the estuarine POC dynamics. We have clarified the statement in the revised manuscript.*

Line 668-671: more direct evidence is needed to evaluate anthropogenic impact on pCO_2 rather than population density. For example, anthropogenic pCO_2 is 100 µatm out of total pCO_2 400 µatm in Estuary A, whereas anthropogenic pCO_2 is 200 µatm out of total pCO_2 1000 µatm in Estuary B.

Response: *Yes, we agree that more direct evidence is needed to evaluate the anthropogenic impact on pCO_2 rather than population density. But no comprehensive information was available on the degree of anthropogenic discharges in the Indian major estuaries and that forced us to use population density as a proxy for anthropogenic inputs. Although % contribution of anthropogenic pCO_2 inputs in total pCO_2 (as you suggested) is the best quantitative way to evaluate the importance of anthropogenic inputs, indeed we don't have enough information to estimate the same. This may be considered as a future research scope. For this reason, we have de-emphasized the discussion of anthropogenic inputs by changing this section into a paragraph combined with a new "natural and anthropogenic sources" section. By not having a stand-alone section about anthropogenic inputs we hope that this limitation is alleviated.*

Line 686: where is "Fig. 21"?

Response: *The "Fig. 21" will be corrected as "Fig. S15" (please see supplementary file) in the revised manuscript.*

Line 695-696: wrong statement "nitrification plays crucial role in increasing pH"

Response: *Yes, it was a typographical error. In the revised manuscript the statement is modified as "nitrification plays crucial role in decreasing pH".*

Line 699: "unlikely"

Response: *Sorry, it will be "likely". We have modified it in the revised manuscript.*

Line 712-713: details for FCO2

Response: *We have elaborated on it in the revised manuscript.*

Link 862: where is "Table 6"

Response: *Thanks for pointing out the mistake. The "Table 6" was missed while uploading the tables during the submission process which is attached below. We will take care of it while uploading the revised version of the paper.*

Table – 6: Contribution of Indian estuaries in global estuarine CO_2 and CH_4 fluxes to the regional atmosphere. Surface area and flux are given in 'km^2' and '$Gg\ yr^{-1}$' unit.

Parameters	***Surface area***	***Total outgassing flux***	***References***
Global estuarine CO_2 flux	*1.40×10^6*	*2.20×10^6*	*Abril and Borges (2004)*
	0.94×10^6	*1.58×10^6*	*Borges (2005)*
	0.94×10^6	*1.17×10^6*	*Borges et al. (2005)*
	0.94×10^6	*1.32×10^6*	*Chen and Borges (2009)*
	1.10×10^6	*0.99×10^6*	*Borges and Abril (2012)*
Mean CO_2 flux	*1.06×10^6*	*1.45×10^6*	*Present study*
Indian estuarine CO_2 flux	***2.70×10^4***	***9.72×10^3***	*Present study*
Contribution by Indian estuaries	***2.54%***	***0.67%***	*Present study*
Global estuarine CH_4 flux	*1.40×10^6*	*1.05×10^3*	*Bange et al. (2004)*
	1.40×10^6	*1.30×10^3*	*Upstill-Goddard et al. (2000)*
	1.40×10^6	*2.40×10^3*	*Middelburg et al. (2002)*
	1.10×10^6	*6.60×10^3*	*Borges and Abril (2012)*
Mean CH_4 flux	*1.33×10^6*	*2.84×10^3*	*Present study*
Indian estuarine CH_4 flux	***2.70×10^4***	***3.27***	*Present study*
Contribution by Indian estuaries	***2.54%***	***0.12%***	*Present study*

Tables and Figures

Table 1: add coordinates, references

Response: *Exact sampling coordinates for all the estuaries are not mentioned in the paper wherefrom the data is derived. Based on the availability we have updated it in the revised manuscript. But references have been added in the revised manuscript.*

Table 2: are they annual averaged numbers? Standard deviation?

Response: *In table – 1, the degree of precipitation and annual discharge are presented as annual average but standard deviations are not available from where the data was picked up. Regarding tidal amplitude, it is not clear in the relevant paper (Sarma et al., 2012) whether it is annual average or not. So, we are unable to clarify it. The other parameters like number of dams, catchment area and population density are not linked with the annual average.*

Table 5: confusing table, please improve

Response: *Sorry, we have removed table – 5 from the revised manuscript as the DIC and DOC export flux data which are the baseline data for the table are for the riverine export flux not for the estuarine export flux.*

Fig. 1: why only zoom in two estuaries? Instead display C3 and C4 plants area, population density is more important to be visualized.

Response: *The reason for zooming into the Hooghly-Sundarbans estuaries in Fig. 1 is mentioned earlier. We agree population density is a part of C biogeochemistry, but the distribution of C3 and C4 is one of the major sources of terrestrial C that ultimately discharges in the river, eventually to the estuary and continental shelf. Moreover, some of the estuaries (like the estuaries of Sundarbans) have very limited anthropogenic influence. Based on that,* *we belief it is better to present distributions of C3 and C4 plants rather than population density. I hope the justification will be acceptable to the reviewer.*

Fig. 2 - Fig. 6: cannot distinguish that data between dry and wet, standard deviation needed.

Response: *We have revised the figure in the revised manuscript but presentation of standard deviation is not possible in all the cases due to the scarcity of replicate datasets. We have added standard deviation where possible in the revised manuscript.*

Fig. 6: estuarine export fluxes values should be river-borne C, the figure is unnecessary if only two components are evaluated.

Response: *Yes, we have removed the figure from the revised manuscript.*

For all supplementary figures: there is no spatial information, reason why exclude several estuarine data, the number of observations are too small, standard deviations? Data interpretation seems unconvincing due to potential data manipulation.
Citation: https://doi.org/10.5194/bg-2022-200-RC1

Response: *Yes, we agree with the reviewer. For complete understanding, spatial coverage is very important in the case of estuary as elemental biogeochemistry changes along estuarine salinity gradient. In this regard, although some of the Indian estuaries are spatially covered, only their mean value is presented in the paper during data presentation. Some of the estuaries like the Hooghly-Sundarbans system are spatially well discussed. We have added some information on it in the revised manuscripts.*

We have included a PCA in the revised manuscript now without eliminating a single data to find out major driving forces.

Yes, Indian estuaries are not been very thoroughly studied to date. So, until now there is a scarcity of published data, which makes it difficult to present standard deviations in all cases. But in the revised manuscript we have added standard deviation wherever possible.

We have tried to improve data interpretation in the revised manuscript wherever possible.