The paper by Krishna et al. provides a very large and almost complete data set on DIC concentrations in the estuaries of Indian rivers and the $\delta^{13}$CDIC. The authors use the data to calculate DIC discharge to the Arabian Sea and Bay of Bengal and DIC yields from the catchments. It is a very important and valuable data set collected in 2011 and 2014 and should be published and made available also in the light of ongoing global change affecting the amount and sources of riverine DIC discharged to the ocean basins.

Thank you very much.

In the methods section the authors state that they have done multiple sampling in the estuaries. The standard deviation should be given in Figure 2.

Standard deviation has been given in Figure 2 as you suggested

In the methods the authors indicate that they have DO and chl-a data which they present only in a summarized form in Figures of the paper. These also need to be made available in an attachment.

This data will be made available

The interpretation of the data is rather convoluted and therefore difficult to follow. It needs restructuring and would benefit from one or more tables.

The discussion part has been restructured following your suggestions given below

Furthermore, an introduction to the use of $\delta^{13}$C$_{DIC}$ as an indicator of DIC source is missing. Such information is given in lines 320-337. This could fit into the introduction.

Description on the use of $\delta^{13}$C$_{DIC}$ as an indicator of DIC source has been shifted into the introduction section. P. 4-5, L. 89-105.

Generally chapters 4.1 and 4.2 may be merged if the discussion is organized differently, may be as suggested below. Now some aspects of the $\delta^{13}$C$_{DIC}$ results are mentioned in 4.1 and 4.2 repeats some of the earlier arguments. In order to better organize the discussion a Table would help showing the average rainfall in the four regions, the volume of discharge per m-2 and soil OC (lines 308 ff). To my mind the last paragraph of chapter 4.1. may rather be the starting point of the discussion.

The structure of the discussion has been modified by considering your suggestions. In order to avoid repetitions, the chapters 4.1 and 4.2 are now merged and it was started with the last paragraph of the chapter 4.1, as you suggested. Average rainfall, volume of discharge and soil OC data of the four regions have been provided in a separate Table (Table 2)

In general I would suggest to follow a clear structure in the discussion (Chapter 4.1.), discussing consecutively (for example). (Most of these points are already mentioned in lines 237-245):

- dilution effects and mixing effects with sea water (this point may well be discussed in the beginning to exclude certain samples from detailed source discussions using $\delta^{13}$CDIC)
- The impact of rock weathering: carbonate vs. silicates, which rock types dominate the catchment?
- The impact of soil organic matter

Structure of the discussion chapter has been revised following your suggestions. It has been re-organized in three sub-sections (4.1.1 to 4.1.3) under the section 4.1. The first sub section 4.1.1 has been dedicated to discuss the impact of hydrological conditions (including mixing and dilution) while the influence of in-stream process were discussed in the second sub-section 4.1.2. Subsections 4.1.3 dealt with the impact of lithology and soils in the catchment. By re-organizing in this way, many repetitions have been deleted.

In Schulte et al. I found a very good sketch of the contrasting impacts of these two mechanisms on $\delta^{13}\text{C}_{\text{DIC}}$ which may provide a helpful concept
- Primary production and respiration in the river/catchment
- possible anthropogenic impacts.
- More points..?

A sketch showing the $\delta^{13}\text{C}_{\text{DIC}}$ range of DIC derived from different sources to rivers has been provided now (Figure 5). The influence of physical and biogeochemical processes such as photosynthesis and decomposition of organic matter on $\delta^{13}\text{C}_{\text{DIC}}$ has also been shown in this figure.

The following collections of data could be given as Tables:
lines 225-233: Table of DIC concentrations selected rivers;
lines 390-396: Table of DIC discharge and
lines 415-418: Table of DIC yields of various regions/rivers.

Data in lines 225-233 was provided in Table 1
Data in lines 390-396 was provided in Table 3
Data in lines 415-418 was provided in Table 3

Detailed comments:

Line 109ff: the studied rivers are perennial so that there is most probably some discharge during the non-monsoonal months and some of the river catchments may even receive winter rains so that these sentences have to be formulated a bit differently. The term “monsoonal rivers” is OK but the discharge during the other seasons may be stated as “small”.

This paragraph has been modified and the discharge during the dry period was stated as small.  P. 6, L. 134.
There is a small amount of discharge during the winter monsoon but will be stores in dams/reservoirs.  P. 6, L.127-130.

Lines 177/178: delete sentence

The sentence was deleted
This part has been shifted to an introduction chapter. P. 2-3, L. 50-57.

Discharge from all four groups of rivers was provided in Table 2.

This section (L. 255-260) has been modified. P. 15, L. 326-328.

Apologies for the mistake. Here, we meant to say that DIC originated by dissolution of atmospheric CO$_2$ (-7 to -8‰) is close to 0‰. It was corrected now. P. 4, L. 92

Chapter 4.4 has been revised and shortened considerably. Many of the redundancies were deleted, and the discussion on source of DIC has been deleted as it has already been discussed in the chapter 4.1.

This was deleted during revision because it is a repetition. The discussion on the impact of submarine ground water discharge (SGD) on DIC has already been made in the chapter 4.1.

The impact of lithology on DIC has been discussed in the sub-section 4.1.3 of the chapter 4.1 as you suggested. Here, we mention only the role of soil organic carbon content in increasing DIC yield from the SW rivers in a short paragraph. P. 20-21, L. 458-467.

The authors promise to send the data on request by E Mail. However, they should be made available in a data bank or as an attachment to the paper.

Data is accessible through the website of our Institute’s data centre (http://www.nio.org/iodc)
The abstract is quite good and may be retained even after changing the discussion. Likewise, changes of the summary would be also rather small after a revision.

Thank you. Summary will be refined.

REVIEWER 2

The manuscript by Moturi et al. presents an extensive database on the DIC fluxes to the Indian Ocean from the monsoonal rivers in the Indian subcontinent. It is clearly the outcome of very hard work, which resulted in this important dataset. As such, this work is valuable, and I can imagine that if published, this dataset would be used by modelers and other researchers.

Thank you very much.

However, at its current form, the manuscript suffers from two essential drawbacks, which in my opinion, should be corrected before the manuscript can be published or even properly reviewed.

My first and main concern is with the quality of the presentation, namely the writing. The manuscript is heavily burdened by numerous grammatical mistakes, redundancies, and unintelligible sentences. Since English is not my native language, I am well aware of how hard it is to write in a foreign language, and therefore, I strongly urge the authors to have their manuscript edited by a native English speaker and/or by a professional editor.

Since English is not our native language, we have a limited control on the English language and therefore there are mistakes in grammar, phrasing, syntax and connecting words in the manuscript. Most of such mistakes were corrected during the revision.

My second concern is with the somewhat superficial interpretation of the data. The authors relay heavily on correlations to investigate the relations between different characteristics of the rivers, but correlations do not necessarily imply cause/effect relations or, as the author argue in the discussion section. Therefore, the conclusions the authors draw are rather general, and do not go much beyond the data itself. Consequently, the manuscript has more resemblance to a report, and may be more appropriate to publication as such. I would recommend the authors to consider more carefully how this dataset can advance what we have already learned from previous works. At its present form, it is very hard to evaluate the scientific contribution of this work, and therefore, I recommend that this discussion paper be withdrawn, and perhaps submitted ab-initio after it has been thoroughly edited and revised.

We have used correlations between various parameters to explain the impact and its statistical significance of different processes within the catchment and rivers on the export and yield of DIC from the Indian monsoonal rivers to the northern Indian Ocean. Though the correlations do not necessarily imply cause/effect relations, as you said, we performed these correlations only to understand the major processes controlling the DIC export fluxes from Indian monsoonal rivers. We have not based only on correlations alone but we described different sources and processes responsible for distribution and export fluxes of DIC from the India monsoonal rivers.
General comments:
Grammar: The text is laden with grammatical errors. The first sentence in the abstract, for example, is flawed. So are lines 19-22, 65-68, 177-178 and many more.

The first sentence in the abstract has been corrected. P. 1, L. 10-11
Lines 19-22 have been revised. P. 1, L. 19-22
Lines 65-68 were corrected. P. 3, L. 70-73.
Lines 177-178 were deleted as the Reviewer 1 suggested deleting these lines.

The usage of connectors (“However”, “Though”, “Despite” etc.) is wrong throughout the text.

Mistakes associated with English language such as grammar, phrasing, syntax and connecting words were rectified.

Writing: Many sentences in the text are excessively long and incomprehensible (e.g. lines 30-35, lines 225-232). Reading sentences that contain more than 80 words, 11 values and more than 10 references (lines 225-232) is extremely demanding, and prevents the reader from understanding the messages that the authors try to convey.

Long and incomprehensible sentences have been modified to obtain the clarity in conveying the message to the readers.
For example, the sentence in lines 30-35 has been split into two sentences. P. 2, L. 29-37.
Lines 225-232 have been modified and restricted to 64 words (reduced from 117 words) by providing DIC concentrations of different rivers in a table (Table 1).
P. 12, L. 265-269.

Cumbersoness and redundancies: Far too many results are incorporated in the text, instead of being presented as figures (e.g. lines 225-232, 247-250). This makes the manuscript cumbersome and turns the reading into a very demanding task.

Many of the results mentioned in the text have been given in Tables. For example, DIC concentrations mentioned in lines 225-232 are now given in a table (Table 1).
Results mentioned in line 248-249, 309 and 425-428 were given in Table 2.
Results mentioned in lines 390-396 and 415-418 were provided in Table 3

Some statements repeat themselves along the text (e.g. lines 176-177 and lines 216-217), making the text needlessly long.

Lines 176-177 were deleted as the same was mentioned in the discussion.
P. 11, L. 254-255.

In some parts of the manuscript, there are no references to existing figures. Instead, the authors re-cite the values (see previous comment), whereas in others, the authors refer to relationships which should have been presented as figures (e.g. lines 263-264. See also detailed comment #22).
All figures have been cited in text. Relationships mentioned in the text have been presented as figures. Figure 4 (4a to 4f) and figure 6 (6a to 6h).

Units: The authors report most of their DIC data in mg l-1. This unit is somewhat archaic, and unclear. To what does the “mg” refer? Bicarbonate? Carbon? The more explicit concentration units of mol l-1 or mol kg-1 are much more common in the current literature. The authors themselves use mol kg-1 in the methods section. In the same section, they use percentage to describe the accuracy. This usage of multiple units for the same parameter is needlessly confusing. I recommend reporting all the results in mol l-1 or mol kg-1.

DIC concentrations throughout the manuscript were expressed in mg l^{-1}. Though it is old, we used mg l^{-1} because we expressed DIC export in Tg yr^{-1} and DIC yield in g m^{-2} yr^{-1}. Further, same y-axis has been used for both DIC concentration and yield in figure 2. In the unit mg l^{-1}, ‘mg’ refers to the total dissolved inorganic carbon but not only bicarbonate. In the methods section also, units for precision have been changed mg l^{-1} in order to maintain the consistency throughout the manuscript. Accuracy of the method is generally expressed as percentage of error, the units (%) used for accuracy of our method for determination of DIC remain unchanged.

Error propagation and significant figures: In the methods sections, the authors report the analytical errors associated with their concentrations measurements. However, they do not propagate these errors to the DIC fluxes. In addition, the authors report too many significant figure compared to the error they report.

The errors associated with flux and yield estimations have been provided in figure 2 in the form of error bars. Significant figure was followed.

Figures and missing figures: Figures 1 and 2 are clear and informative. The rest of the figures are correlations, and could be presented in one or two panels. For some reason the authors did not include figures for some of the correlations they describe in the text. I cannot understand why.

All the correlations mentioned in the text have been presented as figures in two panels (figures 4 and 6).

Specific comments: (please note that I did not include all the grammatical errors in the text).

Line 10: change to “rivers are an/a important/significance source of: : :”

The sentence has been modified as ‘Rivers an important source of ...’ P. 1, L. 10

Line 19-22: revise this sentence. The usage of connectors is grammatically wrong. Use “enriched” instead of “caused the enrichment”. Also, the “stable isotopic composition” cannot be “enriched”. Use either “enriched in 13C” or “increase _13CDIC values”

The sentence has been revised. It has been split into two sentences. ‘caused the enrichment’ has been changed to ‘enriched δ^{13}C_{DIC}’ P. 1, L. 19-22.

Line 25: The sentence is grammatically wrong
The sentence has been modified. P. 1, L. 25-26.

Line 30: define “yield of DIC”

‘yield of DIC’ has been defined. P. 2, L. 29.

Lines 30-35: This sentence is too long and unintelligible

This sentence has been split into two sentences. P. 2, L. 29-37.

Line 56: “The Mississippi river”

It was deleted during revision.

Line 65: How do the fresh water discharge, and suspended sediment load relate to the fluvial carbon fluxes?

Freshwater discharge significantly influences the fluvial carbon fluxes to estuaries and coastal region as it scours terrestrial carbon from rocks and soils. However, suspended load was deleted to convey the message more clearly. P. 3, L. 70-73.

Line 67: The sentence is grammatically wrong

It was corrected. P. 3, L. 70-73

Line 71: change “estimating” to “estimations”

The sentence was modified. P. 4, L. 76-77.

Lines 73-76: Most of the rivers mentioned in this paragraph are located between 30°S - 30°N. So why do the authors claim for “: : :paucity of data” (line 72) for this region?

Here, we meant to say that many of the medium rivers from this region were not included in the global DIC estimations due to the paucity of data. The rivers mentioned in lines 73-79 (Mississippi, Congo, Changjiang and Pearl) are only a few of the large rivers in the world. However, to obtain the clarity, the sentence has been modified. P. 4, L. 76-77.

Lines 76-81: The sentence is grammatically wrong

The sentence has been modified. P. 4, L. 81-86.

Line 82: The phrasing of this sentence is awkward, consider revising

The sentence has been modified. P. 5, L. 106-107.

Line 154: The units here are different from the units used in the text. Please be consistent. It is advised to use mol kg-1 throughout the text
Unit ‘mg l$^{-1}$’ has been used to express DIC concentrations throughout the text as explained earlier. It has been changed here also. P. 8, L. 174

Line 155: change “Scripts” to “Scripps”

Sorry for the mistake. It has been corrected to ‘Scripps’ P. 8, L. 175

Line 157: If the CRM from Andrew Dickson lab is used, 0.3 % equals approximately ±6 µmol l$^{-1}$. This is considerably larger than the precision the authors report in line 154. This error should be propagated along with other sources of error, to calculate the error on the flux estimations

0.2 to 0.3% is the error associated with the accuracy of DIC determination while the value given in line 154 is the precision of the method. However, the precision has also been changed to mg l$^{-1}$ to maintain the consistency. As mentioned earlier, errors associated with determination of DIC concentrations and standard deviations of the mean values were propagated to DIC export flux and yield calculations. These errors have been shown in figure 2 in the form of error bars.

Line 177-178: This sentence’s phrasing is awkward, consider rephrasing

This sentence has been deleted, as suggested by the Reviewer 1

Line 179: If the error is in the second significant figure, it makes no sense to report 4 significant figure. Change 30.86±1.23 °C to 31±1 °C (and throughout the rest of the manuscript)

Results have been presented up to the significant figure for all the parameters.

Line 205: remove the comma after “The estuaries”

‘Comma’ has been removed. P. 11, L. 243

Lines 216-217: This was already stated in lines 176-177.

The sentence in lines 176-177 has been deleted to avoid the repetition because it was mentioned in the discussion. P. 11, L. 254-255.

Line 232: These values were already mentioned in line 224.

The value has been deleted. P. 12, L. 269

Line 236: I suggest that the mean values be added to figure 1 or to figure 2

Mean DIC concentration in the each region was provided in figure 2 as you suggested

Lines 247-250: There are way too many values in this sentence.

These values were deleted from the text and were provided in a table (Table 2)
Lines 255-277: The authors describe 4 correlations here. None of them is shown in a figure, whereas other correlations are. Why did the authors chose not to show there correlations in figures? Since the readers cannot see the fit the authors used, there is no point in mentioning the (very poor) $R^2$ values.

**Figures were provided for all the correlations mentioned in the text. They have been presented in two panels (Figure 4 and 6).**

Line 328: The sentence is grammatically wrong

**The sentence has been corrected. P. 5, L. 97-99.**

Line 501: Add the NIO number or remove this sentence

**Contribution number will be added only after the manuscript has been accepted for publication (during galley proof correction)**

**REVIEWER 3**

This paper presents a hard work from the extensive field coverage of 27 Indian monsoonal estuaries twice during the discharge period of two different years. In the growing concern of climate change when many of the biophysical and biogeochemical models are suffering from the lack of data sets from the tropical rivers, I am sure this paper once published will significantly fill that gap and heavily used by many researchers. However, the manuscript requires to provide clarity and corrections on certain issues before it is published.

**Thank you very much**

The DIC concentrations and fluxes are influenced by the rainfall variability among the four regions, the discussion will be benefited if it starts with this information.

**Yes. Concentration and fluxes of riverine DIC are strongly influenced by the variability in rainfall over the catchment of the river (region). We have started the discussion with the rainfall variability over the four regions (NE, SE, SW and NW) of India and its impact on distribution of DIC concentrations in Indian estuaries. P. 12-13, L. 278-293**

From Figure 1, it is apparent that many of the east flowing rivers, especially in the central and southern regions, are sourced from the western catchments but none in the vice versa direction. This is important and highlighted because high rainfall SW regions have less discharge and DIC fluxes but much of this rainfall might be sourcing the less rain fed SE rivers and contribute to high DIC fluxes. I strongly suggest the authors to include a Table of all rivers sampled (grouped into four regions) with details of their size-class (large and medium), catchment size, length of the river, soil organic carbon, discharge rate, mean DIC concentration, export flux, yield, etc. for better utilizing the hard work of this study by scientific community.

**A table (Table 2) has been provided in which the rivers (grouped into four regions) were given along with their characteristics such as the catchment area, annual mean**
discharge, soil OC and precipitation. Mean (±SD) values of concentrations, export flux and yield of DIC from each group was also provided.

Many of the statements are repeated throughout the manuscript which makes it length, for example, parts of section 4.2 and 4.4 carry some common information. Restructuring of discussion by appropriately merging relevant subsections will improve the focus and clarity.

The discussion chapter has been completely re-organized by merging the sections 4.1 and 4.2 (as suggested by the Reviewer 1) to avoid repetitions. Under this section, the impacts of hydrological conditions, in-stream processes, catchment lithology and soil organic carbon on DIC concentrations has been discussed, and this information has not been repeated. The repeated information in the section 4.4 was deleted, and this section has been considerably shortened.

Number of figures can also be minimized, for example, merge figs.4 & 5 and 6 &7.

As suggested by the Reviewer 2, figures for all the correlations mentioned in the text were given now. However, to minimize the number of figures, as you suggested, all the correlation figures were merged and given in two panels, i.e., figures 4 (4a-4f) and 6 (6a-6h).

The manuscript requires thorough editing for English grammar for better reading.

Many of the mistakes in English language have been corrected and the quality of English language has been improved.

Specific Comments:

Line 43: delete ‘about’.

‘about’ has been deleted. P. 2, L. 46.

Line 54: it is an obvious statement, delete.

The sentence has been deleted

Lines 55-57: how much increase? Specify ‘Mississippi river’.

As per Ren et al., (2015) the total increase in DIC export throughout the 21st century from the Mississippi River to Gulf of Mexico would be over 90% due to the combined effect of climate-related changes along with rising atmospheric CO₂. However, this sentence was modified during revision. P. 3, L. 60-62.

Lines 76-81: include carbon studies from Gupta et al. (2008) in the Chilka lake, a brackish water estuarine system. Also, include Bhavya et al. (2018) for Cochin estuary.
Studies on Chilka lake by Gupta et al. (2008) and Cochin estuary by Bhavya et al. (2018) have been included. P. 4, L. 81-88.

Lines 81-82: Carbon export fluxes from the Chilka lake (Gupta et al., 2008) and Cochin estuary (Gupta et al., 2009) on east and west coast of India respectively were earlier reported.

This sentence was added. P. 4, L. 86-88.

Lines 95-102 & 120-124: Too big sentences.

Big sentences in lines 95-102 and 120-124 have been modified. They have been split into two sentences to obtain the clarity. P. 5-6, L. 117-124 and P. 6-7 L. 142-146, respectively.

Lines 132-134: Year 2011 was a normal monsoon year but 2014 was an El-Nino year.

The mean values of normal monsoon and weak monsoon (El Nino) provides better mean concentrations rather than the mean of two normal monsoon years (expected to be higher side than long term mean) or two weak monsoon years (expected to be lower than long term mean). Therefore, field sampling in this study was conducted one during the normal monsoon year and the other during the weak monsoon year.

Please comment or speculate the variability in light of having used discharge data of earlier years from the published literature. Authors may refer to Indian Annual Rainfall Statistics reports available online at www.imd.gov.in.

Since the aim of our study is to estimate the export fluxes of DIC, the long term mean values of discharge provide relatively better estimates than the discharge values in a year or two due to strong inter-annual variability in discharge (as it depends on the strength of the monsoon). IMD provides data on annual rainfall statistics over the Indian subcontinent but not the volume of discharge from each river which is crucial for flux quantification. However, we have used the IMD rainfall statistics over the Indian subcontinent to discuss the spatial variability in DIC concentrations in the Indian estuaries.

Lines 134-137: These are contradicting the statements made at lines 130-131.

Lines 130-131 mean to say that it is from starting point (origin) to ending point (estuary) of the river, i.e. entire length of the river; whereas Line 134-137 means that it is the length of the estuary (upper and lower estuaries) but not the entire length of the river. However, these sentences have been modified to obtain the clarity. P. 7, L. 149-154.

Line 139: replace was with ‘were’.

Sorry for the mistake. ‘was’ was replaced with ‘were’ P. 7, L. 157

Line 174: specify the source of catchment area.

Source of the catchment area of rivers has been given. P. 9, L. 195-196
Mean±SD values for chlorophyll-a have been provided. P. 10, L. 222-223

Lines 207-208: delete ‘by the Indian monsoonal rivers’.
‘by the Indian monsoonal rivers’ was deleted. P. 11, L. 245

Lines 216-17: repeated statement

The sentence in lines 216-217 is a repeated one with the lines 176-177. Since the sentence in lines 176-177 was deleted, the sentence in lines 216-217 retained.

Lines 250-254: Provide full details in a Table for better usage of this work by many researchers.

These details have been provided in Table 2.

Line 256: Include Gupta et al., 2008 for Chilka lake. Bhavya et al. 2016 covers only dry season (postmonsoon), replace it with Bhavya et al. 2018 for all seasons.

Gupta et al. (2008) has been included and Bhavya et al. (2016) has been replaced with Bhavya et al. (2018). P. 15, L. 326-328

Lines 260-262: Rather relationship with TOC (DOC+POC) is better.

A positive relationship between DOC and DIC indicates that addition of DIC by heterotrophic decomposition of POC which gives both DOC and DIC during heterotrophic transformation. Because of this reason we have taken only DOC instead of TOC

Lines 282-286: It seems this ground water regional variation is following the variability of DIC in the regional estuaries. Does this mean the cause factors for DIC variation are also applicable for its variation in the ground water? Please make a statement on this.

Since ground waters are one of the important sources of DIC in estuaries, it is possible that ground water DIC concentrations will have significant impact on DIC concentrations in estuaries. However, due to the influence of other factors such as hydrology, lithology and environmental characteristics of the catchment on DIC concentrations in estuaries, it is very difficult to make a statement that ground water is the only cause factor for variability of DIC concentrations in estuaries. However, we discussed the influence of ground water (SGD) on DIC concentrations. P. 14, L. 315-324.

Line 285: provide units for all the values.

These values have been deleted from the text and provided in a table (Table 2) as suggested by the other Reviewer.
The sentence has been modified. P. 14, L. 320-322.

We considered the characteristics (lithology) of only surface rocks/soils in the catchment area of the river/region from available soil maps of India. We have not discussed the vertical strata of the rocks/soils. It is possible that vertical strata of the rocks could have influenced the low DIC concentrations in ground waters of the SW region. However, we have not focussed on the reasons for spatial variability in ground water DIC concentration as it is not the scope of this study.

Weighing rates may be high due to highest precipitation but DIC flux from the weathering of lateritic soils to the SW estuaries (refer lines 304-307) could have been far lower than other regions.

DIC concentrations and export flux are far lower in the SW region than the other regions and it could be due to dilution, SGD with low DIC concentrations and the dominance of lateritic soils in the catchment of SW rivers. However, the dense rainfall over the SW region increases the scouring of DIC from soils and therefore causes elevated yield of DIC (DIC export normalized by the river catchment area) from SW rivers. It has been discussed in the section 4.1 (4.1.1 to 4.1.3)

A statement has been given on the sources/controlling processes of DIC in the Indian estuaries. P. 17, L. 375-377

Though, both the statements correspond to the weathering but the contribution of d13CDIC values were differently reported. Pls check.

Though, both the statements correspond to the weathering of silicate and carbonate rocks by carboxylic acid, the resulted δ13C of DIC is different because it is based on the formation mechanism of carbonic acid. Carbonic acid can be formed by dissolution of soil CO₂ or atmospheric CO₂, both of which have different δ13C values. Weathering of silicate and carbonate rocks by soil CO₂ yield the δ13CDIC values of -17 to 21‰ and -7 to -8‰, respectively. Respective δ13CDIC values would be -7 to -8‰ and -3 to -4‰ if weathering occurs by dissolution of atmospheric CO₂. Therefore, the δ13C of DIC is dependent on the source for formation of carboxylic acid, i.e. soil CO₂ or atmospheric CO₂.
Lines 395-396: Are these discharge per day or year?

These discharges are per year. It has been mentioned in the text. P. 18, L. 415-416

Line 401: Relatively higher export fluxes…….. compared to what?

The sentence has been modified. P. 19, L. 420-424.

Line 405: When combined…..with DIC export flux?

The total fluvial dissolved carbon flux is the sum of DIC and DOC. It has been clearly mentioned in the text. P. 19, L. 426-429

Lines 424-425: SW region is having highest rainfall but lowest discharge rate from smallest catchment area. If so, large amount of rainfall might be happening over the non-catchment area. What would be the fate of this? Please discuss on the possibility of its seeping into the ground water and its contribution of DIC flux to the SW coast of India, its relativity with respect to surface flux?

The density of rainfall is high in the SW than the other regions of India. However, lower river discharge from the SW rivers is mainly due to the small catchment area of the SW rivers than the other peninsular rivers. We have discussed the influence of ground water on the concentrations and export flux of DIC from the Indian monsoonal rivers. However, we could not quantify the ground water DIC flux to the coastal waters (SW and SE coast of India) and their contribution to surface DIC export flux because we have not determined the ground water exchange rates, which is not within the scope of the present study.

Lines 432-433: include -ve sign for the r^2 values for having the negative relationships.

Negative sign '-' has been given for the r^2 value, if the relationship is inverse.

Lines 441-442: Reference to the comment for lines 424-425. Does low DIC concentration in the ground water of SW region is also due to high dilution rate and possible lateritic soil strata? Please comment on what would be the ground water discharge rate and its associated DIC export flux to the SW coastal AS compared to the other regions.

This comment is similar to the one mentioned above. Low DIC concentration in the ground waters of the SW region could also be due to high dilution and different soil characteristics (lithology). Though we have discussed possible influence of ground water exchange on the concentrations and export flux of DIC from the Indian monsoonal rivers, we could not quantify the ground water DIC flux to the coastal waters (Arabian Sea and Bay of Bengal) and their contribution to surface DIC export flux because we have not determined the ground water exchange rates, which is not within the scope of the present study.

Line 469: soil organic carbon content….what is the source for this data?
Soil organic carbon data has taken from Kishwan et al., 2009 and Sreenivas et al., 2016. This has been mentioned in the text also. P. 9, L. 196-197

Suggested Literature:


These two references have been cited for Cochin estuary and Chilka lake respectively.
Export fluxes of dissolved inorganic carbon to the Northern Indian Ocean from the Indian monsoonal rivers

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Abstract. Rivers are an important strong source of dissolved inorganic carbon (DIC) to the adjacent coastal waters. In order to identify the examine the spatial variability in the distribution and major sources of DIC in the Indian monsoonal estuaries and to quantify their export flux to the north Indian Ocean, 27 major and medium estuaries along the Indian coast were sampled during the discharge period. An order of magnitude significant variability in DIC concentrations of DIC was observed within the Indian estuaries sampled (3.4 - 44.1 mg l\(^{-1}\)) due to significant variability variations in the size of rivers, precipitation pattern and lithology in the catchments. Dilution with high precipitation (2500±500 mm) and exchange with ground waters of low DIC resulted in very low concentrations of DIC in the estuaries located in the southwest of India (6.6±2.1 mg l\(^{-1}\)) than the estuaries located in the southeast (36.3±6.3 mg l\(^{-1}\)), northwest (30.3±8.9 mg l\(^{-1}\)) and northeast (19.5±6.2 mg l\(^{-1}\)) regions of India. Though the range of stable carbon isotopes isotopic composition of DIC (\(\delta^{13}C_{\text{DIC}}\)) indicates that DIC is largely contributed by weathering of silicate and carbonate minerals however, the storage of water in dams/reservoirs and intrusion of marine waters appears to be responsible for caused the enriched ment in stable carbon isotopic composition of DIC (\(\delta^{13}C_{\text{DIC}}\)) in the east-flowing rivers. It is estimated that the Indian monsoonal estuaries annually export ~10.4 Tg (1Tg=10\(^{12}\) g) of DIC to the northern Indian Ocean, of which the major fraction (74.2%) enters into the Bay of Bengal and the remaining reaches to the Arabian Sea. This is mainly due to the fact that the Bay of

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Bengal receives higher consistent with the freshwater flux which is three times higher in the Bay of Bengal (~378 km$^3$ yr$^{-1}$) of freshwater from the catchment area of about 0.96 million km$^2$, whereas than the Arabian Sea which receives only (~122 km$^3$ yr$^{-1}$) of freshwater from the catchment area of only 0.23 million km$^2$. Though Despite the discharge from the Indian monsoonal rivers account for only 1.3% of global freshwater discharge, they disproportionately export 2.5% of the total DIC export by the world major rivers and 9.4% of the Asian rivers to oceans. The yield of DIC (DIC export normalized by the catchment area of the river) was found to be higher in the SW estuaries (10.8±6.6 g m$^{-2}$ yr$^{-1}$) than the SE (5.8±2.3 g m$^{-2}$ yr$^{-1}$), NE (8.6±5.7 g m$^{-2}$ yr$^{-1}$) and NW (9.5±4.0 g m$^{-2}$ yr$^{-1}$) other estuaries. Despite the SW estuaries, they export only 0.3 Tg yr$^{-1}$ of DIC, which is more than an order of magnitude lower than that of the export by the NE (4.2 Tg yr$^{-1}$), and SE estuaries (3.5 Tg yr$^{-1}$) and NW (2.4 Tg yr$^{-1}$) estuaries.

Higher yield of DIC from the SW estuaries is attributed due to intense precipitation (~3000 mm), favorable natural vegetation of tropical moist deciduous and tropical wet evergreen and semi evergreen forests, and tropical wet climate, high soil organic carbon and the dominance of red loamy soils in catchments of the SW rivers. This study, therefore, revealed that significant variability in the lithology and hydrological (precipitation), lithological (bed rock and soils) and environmental (vegetation and climate) conditions over in the catchments strongly controls the concentrations and yield of DIC from the Indian monsoonal estuaries.

**Keywords:** dissolved inorganic carbon, export flux, Indian rivers, Bay of Bengal, Arabian Sea, North Indian Ocean

1. **Introduction**

Dissolved inorganic carbon (DIC) is the major constituent of carbon species and accounts for ~38% of the total fluvial carbon transport to the global oceans (Meybeck, 1993; Cai, 2011;...
Jarvie et al., 2017). World major river systems export annually about 33-400 Tg (1Tg=10^{12} g) of DIC to the global oceans (Ludwig et al., 1998; Mackenzie et al., 2004; Lerman et al., 2007).

Chemical weathering of carbonate and silicate rocks and soils, and exchange with the ground water in the drainage basin are the major sources of DIC into rivers (Meybeck, 1987; Gaillardet et al., 1999, Dessert et al., 2001; Viers et al., 2007; Raymond et al., 2008; Tamooh et al., 2013).

However, the DIC concentrations in the estuaries are largely influenced by (i) the hydrological (precipitation and runoff), lithological (type and dominance of rocks and soils) and environmental (temperature, climate and vegetation) conditions, and (ii) anthropogenic activities such as exchange with ground water (Finlay, 2003; Shin et al., 2011; Maher et al., 2013), and atmosphere, ie CO_{2}, autotrophic production and heterotrophic decomposition/ utilization of organic matter (McConnaughey et al., 1994; Abril et al., 2003; Finlay and Kendall, 2007, Hotchkiss et al., 2015; Zou, 2016). Besides in-stream processes, such as oxidation of organic carbon by heterotrophic bacteria (Mayogra et al., 2005; Battin et al., 2008; Hotchkiss et al., 2015; Samantha et al., 2015; Zou, 2016) and dissolution of atmospheric carbon dioxide (CO_{2}). Weathering of carbonate and silicate rocks in the catchment, and uptake of DIC by aquatic plants and algae during photosynthesis reaction in rivers are the sinks of atmospheric CO_{2} (e.g. Berner et al., 1983; Raymond et al., 2008); while the oxidation of organic carbon is the source of CO_{2} to the atmosphere. DIC in rivers and estuaries is therefore strongly linked to the carbon cycle. However, due to human interferences, DIC fluxes from the world major rivers have been found to increase dramatically in the last century, for example, the Mississippi river (~90%; Cai, 2003; Raymond and Cole, 2003; Raymond et al., 2008; Ren et al., 2015). It has been noted that substantial alterations in DIC the lateral transport of DIC occurred from land
to sea occurred after the industrialization (Regnier et al., 2013; Bauer et al., 2013). The increase in riverine DIC flux was reported to have a significant impact on the chemical composition (Williamson et al., 1994; Raymond and Cole, 2003; Findlay, 2010; Tank et al., 2010) and carbon budget in the coastal waters (Cole et al., 2007; Dhillon and Inamdar, 2013). The identification of major sources of DIC in the estuaries and quantification of their export fluxes and its riverine export flux estimates to the coastal oceans are important for better understanding the carbon cycling and its budget on both regional and as well global scales (Campeau et al., 2017).

Fluvial carbon fluxes from rivers in the tropical region (30°N to 30°S) are critical for global carbon budgets because they contribute significant fraction of the global DIC (48-64%), freshwater discharge (66.2%) and suspended sediment load (73.2%) to the world oceans, despite they occupy only ~43% of the world’s land area (Huang et al., 2012). Furthermore, humid tropical climate in over the tropical region supports the export of more fluvial carbon fluxes from the continental land masses than the other climates in the world (Meybeck 1993; Ludwig et al., 1998). However, the fluvial DIC fluxes from rivers in the tropical region, except a few large river systems, were not to the global oceans are unknown included in estimating estimations of the fluvial carbon fluxes to global oceans due to the paucity of data.

Numerous studies have been documented on DIC export flux from the world major rivers, for example, the Mississippi (Raymond and Cole, 2003; Raymond et al., 2008; Cai et al., 2008), Changjiang and Pearl (Cai et al., 2008), Congo (Wang et al., 2013) and large river systems in the world (e.g. Gaillardet et al., 1999; Raymond et al., 2013). Although some measurements were carried out on DIC in the Indian estuaries, for example, Mandovi and Zuari (Sarma et al., 2001), Godavari estuary (Sarma et al., 2011), Cochin (Gupta et al., 2009; Bhavya et al., 2016–2018), Hooghly (Mukhopadhyay et al., 2002; Samanta et al., 2015), Mahanadi
(Pattanaik et al., 2017) and Chila lake, a brackish water estuarine system (Gupta et al., 2008) Indian estuaries (Sarma et al., 2012), however, their focus was mainly on internal cycling of carbon and exchange at the air-water interface, exchange of CO$_2$. Carbon export fluxes from the Chilka lake (Gupta et al., 2008) and Cochin estuary (Gupta et al., 2009) on east and west coast of India respectively have been reported but their sources were not evaluated. Nevertheless, no estimations have been made so far on DIC export fluxes to the north Indian Ocean from the Indian subcontinent have been made so far. For the first time, we made an effort here to identify the major sources of DIC in the Indian monsoonal estuaries and to estimate their export fluxes to the north Indian Ocean.

The stable isotopic composition of DIC ($\delta^{13}$C$_{DIC}$) is a well-established and widely used tracer to identify the major sources of DIC in rivers, the aquatic system (e.g. Singh et al., 2005; Tamooh et al., 2013; Samanta et al., 2015; Zou, 2016) because each of the DIC sources has a due to distinct $\delta^{13}$C$_{DIC}$ ratios. isotopic composition of different sources (Deines et al., 1974). The isotopic composition of DIC originated by dissolution of atmospheric CO$_2$ is about -7 to -8‰ (Coplen et al., 2002) whereas it is about -26 to -27‰ if the DIC is derived from oxidation of organic matter produced by C$_3$ plants (O’Leary, 1988). The $\delta^{13}$C of DIC generated by soil CO$_2$ dissolved carbonic acid weathering of silicates is about -17 to -21‰ (Solomon and Cerling, 1987) while it is close in the range of -10 to -9‰ for carbonate rocks because half of the carbon comes from carbonate rocks (0‰, Land, 1980) during weathering. Whereas, the weathering of silicate and carbonate and silicate minerals yield $\delta^{13}$C$_{DIC}$ values in the range of -7 to -8‰ and -3 to -4‰, respectively, if the carbonic acid formed by the dissolution of atmospheric CO$_2$.

Although, despite distinct isotopic composition of DIC is expected for different sources, DIC derived from different sources have distinctly different $\delta^{13}$C$_{DIC}$ the identification of DIC sources...
values, however, the interpretation the δ$^{13}$C$_{DIC}$ values for identification of its sources is still challenging (Amiotte-Suchet et al., 1999; Campeau et al., 2017) due to the isotopic fractionations associated with complex mixture of sources and processes such as photosynthesis (O’Leary, 1988; Finlay, 2004; Parker et al., 2005, 2010), respiration (Finlay, 2003; Waldron et al., 2007), DOC photo-oxidation (Opsahl and Zepp, 2001; Vahatalo and Wetzel, 2008), anaerobic metabolism (Waldron et al., 1999; Maher et al., 2015) and equilibration with atmospheric CO$_2$.

We made an effort for the first time to identify the major sources of DIC in the Indian monsoonal estuaries and quantify their export fluxes to the north Indian Ocean. The main objectives of this study are to (i) identify the major sources and (ii) examine the potential reasons responsible for DIC variability in concentrations of DIC in the Indian monsoonal estuaries during the discharge (wet) period, and (iii) estimate the DIC export fluxes to the north Indian Ocean by the Indian monsoonal rivers.

2. Study region, sampling and Sampling methods

2.1 Study Area

The Indian peninsula bifurcate the north Indian Ocean into the Bay of Bengal and the Arabian Sea. Although these two basins occupy the same latitudinal belt, their oceanographic processes were reported to be remarkably different and attributed due to higher freshwater flux into the Bay of Bengal (1.63 x 10$^{12}$ m$^3$ yr$^{-1}$) than Arabian Sea (0.3 x 10$^{12}$ m$^3$ yr$^{-1}$; Subramanian, 1993; Gauns et al., 2005). Significant differences in the freshwater influx and associated physical and biological changes (Gauns et al., 2005). This is because the glacial and peninsular rivers transport 1.63 x 10$^{12}$ m$^3$ yr$^{-1}$ of freshwater to the Bay of Bengal (Subramanian, 1993) whereas only 0.3x10$^{12}$ m$^3$.yr$^{-1}$ to the Arabian Sea. The large freshwater influx leads to the formation of a strong vertical salinity stratification in the Bay of Bengal (Varkey et al., 1996).
which results in the suppression of that prevents vertical mixing of nutrient rich sub-surface water with that of surface (Prasanna Kumar et al., 2004), makes the Bay of Bengal. As a result, the Bay of Bengal is considered to be relatively less productive (Prasannakumar et al., 2002) than the adjacent Arabian Sea, which is one of the highly productive zones in the world (Madhupratap et al., 1996; Smith, 2001; Barber et al., 2001) due to injection of nutrients into surface through the seasonal upwelling and convective mixing (Shetye et al., 1994; Madhupratap et al., 1996; Muraleedharan and Prasannakumar, 1996).

Discharge from the Indian peninsular monsoonal rivers is largely fed by the monsoon induced precipitation over the Indian subcontinent, which receives >80% of its annual rainfall during the southwest (SW) monsoon period (June-September) (Soman and Kumar, 1990). Though some amount of rainfall occurs during the NE monsoon (December-March), it will not generate discharge as it will be stored within the dams and reservoirs for domestic, industrial and irrigation purposes. Discharge from the Indian peninsular monsoonal rivers is therefore mainly occurs only during the SW monsoon season (Vijith et al., 2009; Sridevi et al., 2015) and hence, termed these rivers are called as monsoonal rivers. Since the major portion of the annual freshwater discharge from the Indian monsoonal rivers is limited to only few months (June-October) in a year, occurs only during the SW monsoon, unlike the European and American rivers, the entire estuary may be filled with freshwater without any vertical salinity gradient (Vijith et al., 2009; Sridevi et al., 2015) during this period. As virtually there is no discharge is small during the dry period rest of the year, the discharge during the SW monsoon (wet period) is considered to be equivalent to the annual discharge from of the monsoonal rivers. Based on the rainfall intensity, forest cover, vegetation and soil type in the catchment, estuaries sampled in the present study were categorized into 4 groups, namely the northeast (NE), southeast (SE),
southwest (SW) and northwest (NW) estuaries of India (Fig. 1). The SW region of India is characterized by the intense rainfall during the SW monsoon (~3000 mm) following the NE (1000-2500 mm), SE (300-500 mm) and NW (200-500 mm) regions of India (Soman and Kumar, 1990). The SW rivers drain red loamy soils while the NW rivers drain black soils. Except the major rivers Godavari and Krishna, all the rivers reaching the Bay of Bengal (NE and SE estuaries) drain the red loamy and alluvial soils in their upper and lower catchments respectively. The Godavari and Krishna rivers, which also drain black soils in their upper catchment along with red loamy and alluvial soils in their middle and lower catchments respectively (Geological Survey of India; www.gsi.gov.in). Based on discharge, the monsoonal estuaries in this study were divided into two types, namely, the major (>150 m$^3$s$^{-1}$) and minor (<150 m$^3$s$^{-1}$) estuaries.

2.2 Sample collection

Estuaries are known to be biologically active spots in the aquatic ecosystem and therefore significant modification of DIC (through autotrophic primary production or heterotrophic respiration) is possible. Hence, water samples were collected from mouth of the estuaries rather than from mid or upstream rivers to obtain reliable export fluxes of DIC to the coastal ocean. Further, to minimize the inter-annual variability in DIC concentrations, sampling was conducted in discharge period of two different years, i.e., 2011 and 2014, and the mean DIC concentration in each estuary was used for export flux estimations. Each estuary was sampled at 3 to 5 locations between the upstream river upper (near zero salinity head) and mouth of the estuary lower (mouth) estuaries in order to minimize the spatial variability in DIC concentrations, and the mean concentrations were used for flux estimates. Further, samples were collected in
mid-stream of the estuary using a local mechanized boat to avoid the contamination from river banks.

**In-situ** In-situ measurements and sample collection was done in the estuaries along the Indian coast (Fig. 1) during the SW monsoon season of the years, 2011 and 2014. Surface water samples at each location were collected for phytoplankton biomass (Chl-a), DIC and dissolved oxygen (DO). Samples for DIC were collected in air-tight crimp-top glass bottles and added poison (mercuric chloride) to arrest the biological activity. DO analysis was carried out at a temporary shore laboratory set up for sample processing after the completion of sampling on each day. Water samples were filtered through GF/F (nominal pore size of 0.7µm) under moderate vacuum and stored in liquid nitrogen for Chl-a analysis at the NIO.

2.3. Methods

Temperature and salinity at the sampling locations were measured using a conductivity-temperature-density (CTD) profiling system (Sea Bird Electronics, SBE 19 plus, United States of America). Concentrations of DO was determined by Winkler’s method (Carritt and Carpenter, 1966) using an auto titrator (Metrohm, Switzerland) with potentiometric end point detection. The analytical precision of the method was ±0.07% (RSD). DIC concentrations in water samples were measured at our Institute laboratory using Coulometer (UIC Inc., USA) connected to an automatic sub-sampling system. Based on the repeated analysis of samples and standards, the precision of the method was ±1.8 µmol l$^{-1}$. The certified reference materials (CRM) supplied by Dr. A.G. Dickson, Scripps Institute of Oceanography, USA and internal standards were used to test the accuracy of our DIC measurements and it was found to be within ± 0.2 to 0.3%. Chlorophyll-a (Chl-a) on the filter was extracted into di-methyl formamide (DMF) and measured the extract fluorometrically using a spectrofluorophotometer (Varian...

Total export flux of DIC from each river was estimated by multiplying the mean concentrations of DIC in an estuary with the mean annual discharge. Spatial variability in DIC concentrations in estuaries was minimized to a large extent by collecting samples from head to mouth of the estuary while the inter-annual variability by collecting samples during discharge periods of two years. However, variability in DIC concentrations within the discharge period results in some uncertainties in our estimations of DIC export fluxes. Time series measurements in the Godavari estuary (our unpublished results) revealed that the variability in DIC concentrations within the discharge period is up to 10%. Therefore, the error associated with our DIC flux estimates can may be about ±10%. DIC fluxes normalized by catchment area (yield) were calculated by dividing the total DIC export flux of the river by its catchment area.

3. Results

3.1. Hydrographic characteristics

 Prevailing hydrographic conditions in Indian estuaries during the sample collection were given in detail elsewhere (Sarma et al., 2012, 2014; Krishna et al., 2015). Briefly, mentioned here for ready reference. Surface water temperature was found to be higher in the estuaries located on the east coast (mean 30.86±1.23°C) than the west coast (27.32±1.40°C) of India. Salinity
varied broadly from near zero (0.106) to 28.28 during the study period. Relatively higher salinities (>20) were recorded by the medium estuaries, which receive relatively lower freshwater discharge from the upstream river, for example, Nagavali (28.78), Vaigai (24.64) and Rushikulya (20.70). Mean salinities were lower in the west-flowing NW (0.1±0.02) and SW (2.1±2.8) estuaries than the east-flowing SE (9.5±7.8) and NE (8.5±11) estuaries. Dissolved oxygen saturation varied from as low as 62.6% to as high as 105%, with a mean saturation of 89.9±11.4% in the estuaries sampled. The SW estuaries recorded relatively slightly lower DO saturation (82±7%) than the NE (89±15%), NW (93±3%) and SE (96±11%) estuaries.

Chlorophyll-a (Chl-a) concentrations varied broadly from 0.8 to 10.75 mg m$^{-3}$, with relatively higher mean concentrations in the SE (4.7±2.5 mg m$^{-3}$) followed by the SW (3.0±2.8±0.7 mg m$^{-3}$) estuaries (Table 2). However, on the other hand, relatively low Chl-a was observed in the medium (2.6±1.3 mg m$^{-3}$) than in the major estuaries (3.2±2.1 mg m$^{-3}$).

DIC concentrations in the Indian monsoonal estuaries widely varied from 3.4 (Bharathappuzha) to 44.1 mg l$^{-1}$ (Vellar), with a significant spatial variability (Fig. 2). More than five times higher mean concentrations were observed in the SE (36.3±6.3 mg l$^{-1}$) and NW estuaries (30.3±8.9 mg l$^{-1}$) than in the SW estuaries (6.6±2.1 mg l$^{-1}$) and intermediate concentrations were found in the NE estuaries (19.5±6.2 mg l$^{-1}$). DIC concentrations were found to be similar (homoscedastic Student’s t-test; p=0.76) in the major (22.7±13.6 mg l$^{-1}$) and medium (21.1±13.2 mg l$^{-1}$) estuaries (homoscedastic Student’s t-test; p=0.76). The $\delta^{13}$C$_{DIC}$ varied from -13.0 to 2.5‰, with a significant spatial variability (Fig. 3) in the estuaries sampled. Relatively depleted $\delta^{13}$C$_{DIC}$ values were observed in the west flowing estuaries of NW (-...
11.1±2.3‰) and SW (-7.4±1.9‰) than the east flowing estuaries of NE (-3.5±2.8‰) and SE (-
2.7±5.2‰) regions of India.

3.3. Export fluxes and yield of DIC

Annual export flux of DIC from the individual estuaries to the coastal ocean from
different estuaries varied broadly from 0.01 Tg (Chalakudi) to as high as 2.32 Tg (Krishna).

Annually, the NE estuaries export higher DIC flux of 4.2±1 Tg followed by the SE (3.59 Tg) and
NW estuaries (2.38±1 Tg). In contrast, the SW estuaries recorded the lowest export flux
of 0.3 Tg which is an order of magnitude lower than that of the export flux by the NE and
SE estuaries (Fig. 2). The Indian monsoonal estuaries together export about 10.4 Tg yr⁻¹ of
DIC to the northern Indian Ocean, of which 7.7 Tg (74.2%) enters into the Bay of Bengal and the
remaining into the Arabian Sea (2.7 Tg). The estuaries, Krishna (2.32 Tg), Godavari (1.45 Tg)
and Haldia (1.46±2 Tg) together responsible for the transport of 64.5% of total riverine DIC
export to the Bay of Bengal by the Indian monsoonal rivers. The yield of DIC ranged from 2.7
(Bharathappuzha) to 21.6 g m⁻² yr⁻¹ (Mandovi), excluding the exceptionally high yield of 113.4 g
m⁻² yr⁻¹ from Haldia estuary. The west flowing rivers to the Arabian Sea are characterized by
relatively higher yield of DIC (mean 10.4±5.6 g m⁻² yr⁻¹) than the east flowing rivers to the Bay
of Bengal (7.3±4.6 g m⁻² yr⁻¹). Among the estuaries sampled, the SW and SE estuaries recorded
higher (10.8±6.6 g m⁻² yr⁻¹) and lower (5.8±2.3 g m⁻² yr⁻¹) yields of DIC respectively whereas
intermediate values noticed in the NW (9.5±4.0 g m⁻² yr⁻¹) and NE (8.6±5.7g m⁻² yr⁻¹)
estuaries recorded intermediate values.

4. Discussion
Hydrographic characteristics of the Indian monsoonal estuaries during the study period were described elsewhere (Sarma et al., 2012, 2014; Krishna et al., 2015). Strong flow from the upstream rivers due to the SW monsoon-induced heavy precipitation over the catchment makes most of the estuaries less saline (near zero) during the study period, except the minor-medium estuaries, Nagavali, Vaigai and Rushikulya, during the study period. No vertical salinity stratification was observed in estuaries in all estuaries sampled during the study period; and it is consistent with earlier observations in the Indian estuaries during discharge period Godavari and Mandovi estuaries (Vijith et al., 2009; Sridevi et al., 2015). This is the unique feature of the Indian estuaries as strong stratification occurs in unlike the European and American estuaries following discharge (Christopher et al., 2002). This difference is mainly caused by high discharge in shorter period in the Indian than other estuaries in the world (Vijith et al., 2009).

4.1 Variability—Distribution and sources of DIC concentrations in the Indian monsoonal estuaries

Mean DIC concentration found in this study (21.9±13.2 mg l\(^{-1}\); range: 3.4 to 44.1 mg l\(^{-1}\)) is similar to those observed earlier in the Indian estuaries such as Ganga-Brahmaputra and Hooghly (Singh et al., 2005, Samanta et al., 2015), and in the estuaries elsewhere in the world, for example York, Yangtze, Seri and Xi etc (Raymond and Bauer, 2000, Cai et al., 2008, Ishikawa et al., 2015; Zou, 2016) (Table 1). The Indian estuaries, for example, Ganga-Brahmaputra (23 mg l\(^{-1}\); Singh et al., 2005), Hooghly (21.8 mg l\(^{-1}\); Samanta et al., 2015) and Mahanadi (15.0; Pattanaik et al., 2017) and elsewhere in the world, for instance, York river estuary (6.21 mg l\(^{-1}\); Raymond and Bauer, 2000), Yangtze river (28 mg l\(^{-1}\); Cai et al., 2008), British rivers (median 4.43 mg l\(^{-1}\); Jarvie et al., 2017), Seri, central Japan (17.6-21.9 mg l\(^{-1}\);
Ishikawa et al., 2015), the Red river, Vietnam (9.1 - 29.9 mg l\(^{-1}\); Quynh et al., 2016) and Xi river, southwest China (18.45 - 64.9 mg l\(^{-1}\); Zou, 2016). However, the mean DIC concentrations in the Indian estuaries are higher than those found in some of the Asian rivers of tropical region (12.7 mg l\(^{-1}\), Huang et al., 2012) and Indian estuaries (21.9 ± 13.2 mg l\(^{-1}\)) are higher than the global mean of 10.3 mg l\(^{-1}\) (Meybeck and Vorosmarty, 1999) (Table 1), and the Asian rivers (12.7 mg l\(^{-1}\)) in the tropical region (30°N - 30°S; Huang et al., 2012), but lower than those reported in the rivers draining into the Gulf of Trieste (N Adriatic; 37-66 mg l\(^{-1}\); Tamse et al., 2014) (Table 1).

Among the estuaries sampled along the Indian coast, the SW estuaries are characterized by significantly lower mean concentrations of DIC (6.6 ± 2.1 mg l\(^{-1}\)) than the SE (36.3 ± 6.3 mg l\(^{-1}\)), NE (19.5 ± 6.2 mg l\(^{-1}\)) and NW (30.3 ± 8.9 mg l\(^{-1}\)) estuaries (Table 2). This could be due to considerable spatial variations in the (i) hydrological, lithological and environmental conditions in the catchments and (ii) in-stream physical and biogeochemical processes. DIC concentrations in estuaries are mainly governed by the hydrological (precipitation and runoff), lithological (type and dominance of rocks and soils) and environmental (temperature, climate and vegetation) conditions, and anthropogenic activities (deforestation and land use change) in the catchment, and in-stream physical and biological processes such as exchange with ground water (Finlay, 2003; Shin et al., 2011; Maher et al., 2013) and atmospheric CO\(_2\)-autotrophic production and heterotrophic decomposition of organic matter (McConnaughey et al., 1994; Abril et al., 2003; Finlay and Kendall, 2007).

4.1.1. The impact of hydrological conditions
The SW region of India receives the highest amount of rainfall during the SW monsoon (2500±500 mm) than the SE (400±50 mm), NE (1000±200 mm), and NW (750±250 mm) regions of India (Table 2) (Soman and Kumar, 1990). Though the intense rainfall in the SW region is expected to cause higher weathering rates and therefore higher DIC (e.g., Gupta et al., 2011), the observed lower DIC concentrations were found in the SW estuaries. This is attributed to the influence of dilution because the catchment area normalized volume of discharge was found to be higher in the SW estuaries (1.71 m³ m⁻²) than in the SE (0.17 m³ m⁻²), NE (0.6 m³ m⁻²) and NW (0.32 m³ m⁻²) estuaries. About three times higher catchment area normalized discharge might have diluted the significant dilution of DIC concentrations in the SW estuaries. A strong negative correlation between precipitation in the catchment and DIC concentration in estuaries (r²= 0.89, p<0.001; Fig. 5a) also suggests that DIC concentration in Indian estuaries are rather controlled by the intensity of precipitation over the catchment. Dilution of DIC by heavy precipitation in the SW region can also be seen from relatively depleted δ¹³C_DIC values (-7.4±1.9‰) in the SW estuaries because the shorter residence time of soil water depletes the δ¹³C_DIC due to preferential dissolution of $^{12}CO_2$ over $^{13}CO_2$ (Amiotte-Suchet et al., 1999).

Since many of the hydrological processes are largely dependent on the size of the river and its catchment area, the lower it may govern the DIC concentrations of DIC. The lower concentrations of DIC in the SW estuaries of this study could be possibly due to the smaller catchment area as size of the rivers. This is because, the SW rivers are small, both in terms of discharge (46 km³ yr⁻¹) and catchment area (total catchment area: 0.02 M km²), than that of SE (102 km³ yr⁻¹ and 0.45 M km², respectively), NE (276 km³ yr⁻¹ and 0.52 M km²) and NW (75 km³ yr⁻¹ and 0.21 M km²)-rivers (Table 2). However, the DIC concentrations of DIC in the Indian
Estuaries showed a significant positive relationship with catchment area ($r^2=0.75$; $p<0.001$; Fig. 4a4b) and a negative relationship with volume of discharge ($r^2=0.57$; $p<0.001$; Fig. 4b4c) only in the medium estuaries (discharge: $<150$ m$^3$s$^{-1}$), suggesting that an area of catchment and magnitude of discharge controls DIC concentrations largely in the medium estuaries rather than in the major estuaries. It could be due to the influence of in-stream processes as the major rivers are long compared to the medium rivers.

Mixing with seawater and exchange of submarine groundwater also influenced DIC concentrations in the estuaries. Since this study was conducted during the SW monsoon, many of the estuaries are filled with freshwater (salinity $>1$) due to maximum discharge during this period. On the other hand, higher salinities ($>20$) were observed in some medium estuaries, namely, Rushikulya, Nagavali and Vaigai recorded higher salinities ($>20$) due to low flow from upstream river. As a result, the $\delta^{13}$C$_{DIC}$ values were found to be $>0\%$ in these three estuaries (0.1, 0.7 and 2.5$\%$ respectively). A strong positive correlation was found between $\delta^{13}$C$_{DIC}$ and salinity (Fig. 6d; $r^2=0.71$, $p<0.001$), suggesting that $\delta^{13}$C$_{DIC}$ values in the Indian estuaries are also influenced by the intrusion of marine waters ($\delta^{13}$C$_{DIC}$: -1 to 2$\%$), particularly in medium estuaries. The $\delta^{13}$C$_{DIC}$ values were found to be $>0\%$ in Rushikulya, Nagavali and Vaigai estuaries (0.1, 0.7 and 2.5$\%$ respectively) suggesting that major contribution of DIC is from intrusion of marine water.

As found in many estuaries over the world, submarine groundwater exchange influences DIC concentrations in Indian estuaries, for example, (Rengarajan and Sarma, 2015) due to higher concentrations of DIC by 3 to 4 times higher DIC concentrations in the groundwater compared than estuary. The estuarine waters of the Godavari and estimated that
submarine ground water discharge contributes up to 52% of DIC concentrations in the Godavari estuarine system. The measured DIC concentrations in ground waters along the entire Indian coast (Dr. BSK Kumar, personal communication) showed strong spatial variability with suggest relatively lower concentrations in the SW (mean 32±19 mg l⁻¹) than the SE (106±56), NE (92±31) and NW (84±54 mg l⁻¹)-regions of India (Table 2) during discharge period (Dr. BSK Kumar, personal communication). Though the DIC concentrations in Exchange of SW estuaries with ground water with relatively lower DIC ground waters with that of estuarine water were higher by about 3 to 5 times than the concentrations found in the Indian estuaries, however, exchange of ground water with relatively low DIC-concentrations in the SW region could have, at least partly, concentrations might have possibly yielded low DIC concentrations. Nevertheless it is difficult to ascertain the impact of ground water exchange yielded low DIC in the SW estuaries due to lack of submarine ground water discharge rates. can caused the lower DIC concentrations in the SW estuarine waters. However, 4.1.2. The impact of in-stream processes

Since the Indian monsoonal estuaries were have been reported as-to be a source of CO₂ to the atmosphere during the discharge period (Sarma et al., 2001, 2011, 2012; Gupta et al., 2008, 2009; Bhavya et al., 2016,2018), due to the microbial decomposition of terrestrial organic matter brought by the rivers. This suggests that, the DIC input from dissolution of atmospheric CO₂ in estuaries can be ruled out. CO₂ release due to, however, heterotrophic decomposition of organic matter adds significant amount of DIC to the Indian estuaries during discharge this period as enhanced bacterial respiration rates were reported in the Indian estuaries (Sarma et al., 2011; 2012). A fairly good positive correlation between DIC and DOC concentrations (r²=0.340, p<0.01; Fig. 4e), except few medium estuaries, suggests that DIC addition through microbial
degradation of particulate organic matter is significant, seems to be possible source in the Indian estuaries. Except the NW estuaries, which recorded relatively depleted $\delta^{13}$C of DIC ($\delta^{13}$C$_{\text{DIC}}$), the $\Delta$ positive correlation between $\delta^{13}$C$_{\text{DIC}}$ and DOC _concentrations_ was observed, with a different slope for NW estuaries ($r^2=0.3543$, $p<0.01$; Fig. 4f), as was observed elsewhere (Xi river, Zou et al., 2016), confirming that oxidation of organic matter is _may be_ one of the main major DIC sources in the Indian monsoonal estuaries. Similar relationship was also observed in the Xi river (Zou et al., 2016). The range of $\delta^{13}$C$_{\text{DIC}}$ (-13.0 to 2.5‰) in the Indian monsoonal estuaries is distinctly enriched than that of the $\delta^{13}$C of DIC derived from decomposition of terrestrial C$_3$ plant derived organic matter (-27 to -26‰, O'Leary, 1988; Fig. 5), suggesting that DIC might have been contributed from decomposition of terrestrial C$_4$ plants (-17 to -13‰, Krishna et al., 2015) and significant amount of DIC is contributed by $\delta^{13}$C-enriched sources such as weathering of silicate and carbonate rocks. In addition, if weathering occurs due to dissolution of silicate and carbonate rocks due to atmospheric CO$_2$, the $\delta^{13}$C$_{\text{DIC}}$ yields -8 to -7‰ and -4 to -3‰ respectively. On the other hand, the $\delta^{13}$C$_{\text{DIC}}$ would be -10 to -9‰ and -21 to -17‰ (Solomon and Cerling, 1987) if the dissolution of silicate and carbonate rocks occurs due to soil CO$_2$ respectively. As discussed above, flux of CO$_2$ from atmosphere to river cannot be expected due to super-saturation of riverine CO$_2$, weathering of silicate and carbonate rocks by dissolution of soil CO$_2$ may be possible. Though isotopic composition of $\delta^{13}$C$_{\text{DIC}}$ derived from decomposition of C$_4$ plants and weathering due to soil CO$_2$ are similar and difficult to separate, Sarma et al. (2014) measured isotopic composition of $\delta^{13}$C$_{\text{POC}}$ and found that $>90\%$ of the POC is contributed by C$_3$ plants. Hence possible contribution of DIC through decomposition of C$_4$ plants may be negated.
by carbonic acid produced by dissolution of atmospheric CO$_2$ (7 to 8‰ and 3 to 4‰ respectively) besides the contribution from. On the other hand, autotrophic production removes DIC as it converts DIC to organic carbon. Significant negative correlation between chlorophyll-
$\text{a}$ and DIC concentrations ($r^2 = -0.47$, $p<0.01$; Fig. 6a), except few SE estuaries where elevated phytoplankton biomass (Chl-a: >5 mg m$^{-3}$) was recorded, suggesting that autotrophic removal of DIC is also a possible sink significant in the Indian monsoonal estuaries during the study period. This process would enrich $\delta^{13}$C$_{\text{DIC}}$ due to preferential removal of $^{12}$CO$_2$ over $^{13}$CO$_2$ during photosynthesis. A positive relationship was observed between $\delta^{13}$C$_{\text{DIC}}$ and Chl-a in the Indian estuaries ($r^2 = 0.50$, $p<0.01$), suggesting that biological removal of DIC enriched $\delta^{13}$C$_{\text{DIC}}$.

In contrast, significance of DIC addition by heterotrophic decomposition of organic matter (respiration) depletes $\delta^{13}$C$_{\text{DIC}}$ due to release of $^{12}$CO$_2$ over $^{13}$CO$_2$ during this process. Due to lack of respiration rates data, we could not able to evaluate its influence. Nevertheless, the dissolved oxygen saturation stores the net effect of biological production and heterotrophic respiration. The net biological influence on $\delta^{13}$C$_{\text{DIC}}$, the same is correlated with DO saturation and found by a fairly good significant positive correlation between $\delta^{14}$C$_{\text{DIC}}$ and dissolved oxygen saturation ($r^2 = 0.49$, $p<0.01$; Fig 6b), (depleted $\delta^{13}$C$_{\text{DIC}}$ values at low % of DO saturation), except NW estuaries, which recorded depleted $\delta^{13}$C$_{\text{DIC}}$ (<-10.0‰) confirming that biological processes enriched $\delta^{13}$C$_{\text{DIC}}$ in the Indian monsoonal estuaries. This is because the microbial decomposition of organic matter results in depleted $\delta^{13}$C$_{\text{DIC}}$ due to preferential release of $^{12}$C over $^{13}$C in to DIC pool while removal of DIC by autotrophic production enriches the residual DIC due to preferential uptake of $^{12}$C over $^{14}$C during photosynthesis reaction. CO$_2$ out gassing due to heterotrophic decomposition of organic matter and equilibrium with atmospheric CO$_2$ results in
the enrichment of $\delta^{13}$C$_{\text{DIC}}$ in reservoirs/dams and stored water bodies (Shin et al., 2001; Brunet et al., 2005; Bouvillion et al., 2009; Zeng et al., 2011; Tamooh et al., 2013). As many of the east flowing river (e.g. Godavari, Krishna and Cauvery etc) were dammed at many locations for domestic, industrial and irrigation purposes. Hence, relatively enriched $\delta^{13}$C$_{\text{DIC}}$ in these east flowing rivers indicate that either DIC is originated from weathering of carbonate rocks by carbonic acid produced from dissolution of atmospheric CO$_2$ and/or storage of water in dams/reservoirs as many of them estuaries might have been influenced by storage of water besides sources of DIC (e.g. Godavari, Krishna and Cauvery etc) were dammed at many locations for domestic, industrial and irrigation purposes. A significant positive correlation between DIC concentrations and $\delta^{13}$C$_{\text{DIC}}$ ($r^2=0.77, p<0.001$; Fig. 7c), excluding the positive $\delta^{13}$C$_{\text{DIC}}$ values, indicate that significant contribution of DIC is from oxidation of particulate organic carbon in dams/reservoirs or stored water bodies. Therefore, DIC in the Indian estuaries are contributed by weathering of silicate and carbonate rocks due to soil CO$_2$, biological production, organic matter decomposition and exchange of CO$_2$ to the atmosphere.

4.1.3. The impact of catchment lithology

As found in many estuaries over the world, submarine ground water exchange strongly influences DIC concentrations in Indian estuaries, for example, Rengarajan and Sarma (2015) found 3 to 4 times higher DIC concentrations in the ground water compared the estuarine waters of the Godavari and estimated that submarine ground water discharge contributes up to 52% of DIC concentrations in the Godavari estuarine system. The measured DIC concentrations in ground waters along the entire Indian coast (Dr. BSK Kumar, personal communication) showed strong spatial variability with relatively lower concentrations in the SW (mean 32±19 mg l$^{-1}$) than the SE (106±56), NE (92±31) and NW (84±54 mg l$^{-1}$) regions of India during discharge.
period. Though the DIC concentrations in ground waters were higher by about 3 to 5 times than the concentrations found in the Indian estuaries, however, exchange of ground water with relatively low DIC concentrations in the SW region could have, at least partly, caused the lower DIC concentrations in the SW estuaries.

Spatial distribution of bedrock and soils over the Indian subcontinent shows that Narmada and Tapti rivers in the NW India and upper reaches of Godavari and Krishna rivers drain over the igneous rocks (Deccan traps) while the other rivers flow through over the metamorphic rocks (Pre-Cambrian), which are the predominant rock type in south India. However, Haldia and lower reaches of the SE rivers drain over the sedimentary rocks (Geological Survey of India, https://www.gsi.gov.in). Although the higher chemical weathering rates were reported to be higher for in the Deccan Trap basalts (Das et al., 2005; Singh et al., 2005), however, higher DIC concentrations were also found observed in estuaries draining over the metamorphic rocks, suggesting that strong influence of other factors may also be governing the concentrations of DIC, other than the bedrocks in the catchment. The broad range of $\delta^{13}C_{\text{DIC}}$ found in this study (-13.0 to 2.5‰) not only also indicates that DIC contribution is from variable sources such as DIC contribution from weathering of carbonate and silicate rocks by carbonic acid derived from dissolution of soil CO$_2$ (-10 to -9‰ and -21 to -17‰ respectively, Solomon and Cerling, 1987), decomposition of terrestrial organic matter (-267 to -236‰, O’Leary, 1988) and marine water (0 to 2‰, ) (Fig. 5)—but also suggests processes on $\delta^{14}C_{\text{DIC}}$ in the Indian monsoonal estuaries—.

Spatial distribution of soils shows that lateritic soils, which are poor in lime and silicate, occupied the catchment of the SW rivers. Chemical weathering rates are relatively lower in the lateritic than the non-lateritic soils and the consumption of atmospheric/soil CO$_2$ through silicate
weathering is lower by ~2 times in the former than the latter (Boeglin and Probst, 1998).

Though the upper reaches of the east flowing rivers (NE and SE) drain over the lime-poor red and yellow soils, however, they are dominated by while the lower reaches drain predominantly the lime-rich alluvial soils in their lower reaches. Upper reaches of Krishna and Godavari also drain over the lime-rich black soils. The dominance of lateritic soils, which are relatively less susceptible to chemical weathering than the non-lateritic soils, over-in the catchments of the SW rivers could have, at least in part, could be possible reason for lowered the DIC concentrations in SW estuaries. The enriched $\delta^{13}$C$_{\text{DIC}}$ in the SW estuaries (-7.4±1.9‰) may also be due to less contribution of DIC from lateritic soils as these soils are poor in lime (-10 to -9‰) and silicate (-21 to -17‰) and less susceptible to chemical weathering rates during the study period. Altogether DIC concentrations in the Indian monsoonal estuaries appears to be controlled by the precipitation, area, and lithology of the catchment, SGD, and autotrophic production and heterotrophic decomposition of organic matter within the rivers.

The SW region of India receives the highest amount of rainfall during the SW monsoon (2500±500mm) than the SE (400±50), NE (1000±200) and NW (750±250mm) regions of India (Soman and Kumar, 1990). Though the intense rainfall in the SW region is expected to cause higher weathering rates and therefore higher DIC (e.g., Gupta et al., 2011), the observed lower DIC concentrations in the SW estuaries could be due to the dilution. The catchment area normalized volume of discharge was found to be higher in the SW estuaries (1.71 m$^3$ m$^{-2}$) than in the SE (0.17), NE (0.6) and NW (0.32 m$^3$ m$^{-2}$) estuaries, suggesting that significant dilution of DIC concentrations in the SW estuaries. A strong negative correlation between precipitation in the catchment and DIC concentration in estuaries ($r^2 = 0.89$, p<0.001, Fig. 5) confirms that DIC
concentration in Indian estuaries are rather controlled by the intensity of precipitation over the catchment.

4.2 δ¹³C of DIC in the Indian monsoonal estuaries

The stable isotopic composition of DIC (δ¹³CDIC) is a well-established and widely used tracer to identify the major sources of DIC in rivers (e.g. Singh et al., 2005; Tamooh et al., 2013; Samanta et al., 2015; Zou, 2016) because each of the DIC sources have a distinct δ¹³C DIC ratios (Deines et al., 1974). DIC originated by dissolution of atmospheric CO₂ is about −7 to −8‰ (Coplen et al., 2002) whereas it is about −26 to −27‰ if DIC is derived from oxidation of organic matter produced by C₃ plants (O’Leary, 1988). The δ¹³C of DIC generated by soil CO₂-dissolved carbonic acid weathering of silicates is about −17 to −21‰ (Solomon and Cerling, 1987) while it is close to −9‰ for carbonate rocks because half of the carbon comes from carbonate rocks (0‰, Land, 1980) during weathering. Whereas, the weathering of carbonate and silicate minerals yield δ¹³C DIC values −7 to −8‰, and −3 to −4‰, respectively, if the carbonic acid formed by the dissolution of atmospheric CO₂. Although, DIC derived from different sources have distinctly different δ¹³C DIC values, however, the interpretation the δ¹³C DIC-values for identification of its sources is still challenging (Amiotte-Suchet et al., 1999; Campeau et al., 2017) due to the isotopic fractionations associated with complex mixture of sources and processes such as photosynthesis (O’Leary, 1988; Finlay, 2004; Parker et al., 2005, 2010), respiration (Finlay, 2003; Waldron et al., 2007), DOC photo-oxidation (Opsahl and Zepp, 2001; Vahatalo and Wetzel, 2008), anaerobic metabolism (Waldron et al., 1999; Maher et al., 2015) and equilibration with atmospheric CO₂.

The range of δ¹³C DIC found in this study (−13.0 to 2.5‰) was similar to those reported earlier in various rivers, for example, Brahmaputra (Singh et al., 2005), Rhine (Buhl et al., 1991),
Ottawa (Telmer et al., 1999), St. Lawrence (Yang et al., 1996), Nanpan and Beipan rivers, southwest China (Zou, 2016) and Tana river, Kenya (Tamooh et al., 2013). The range of δ\(^{13}\)C\(_{\text{DIC}}\) in this study indicates a variety of sources, including silicate and carbonate weathering and marine waters, contribute DIC to the Indian monsoonal estuaries during the study period. Relatively depleted δ\(^{13}\)C\(_{\text{DIC}}\) in the west flowing river estuaries of NW (mean -11.1±2.3‰) and SW (mean: -7.4±1.9‰) regions suggest that DIC is contributed from silicate and carbonate weathering by the carbonic acid, produced from the dissolution of both soil CO\(_2\) and atmospheric CO\(_2\). Zou (2016) found the δ\(^{13}\)C\(_{\text{DIC}}\) values in the range of -13.9 to -8.1‰ in the Nanpan and Beipan rivers of SW China and were attributed to dominant contribution of DIC from weathering of carbonate minerals. Relatively enriched δ\(^{13}\)C\(_{\text{DIC}}\) in the east flowing river estuaries of NE (-6.5 to 0.7; mean: -3.5±2.8‰) and SE (-7.9 to 2.5‰; -2.7±5.2‰) indicates that major contribution of DIC is from chemical weathering of carbonate rocks by atmospheric CO\(_2\)-dissolved carbonic acid or acid from non-carbon sources (Li et al., 2008). Weathering of carbonate minerals by acid sources other than carbonate acid causes enrichment compared to weathering by carbonate acid due to lack of contribution from δ\(^{14}\)C-depleted carbonate acid of soil CO\(_2\) (-17 to -21‰) or atmospheric CO\(_2\) (-7 to -8‰) origin to the δ\(^{13}\)C-enriched carbonate rocks (0‰, Land 1980).

In addition to the sources, hydrological and biological processes also influence the δ\(^{13}\)C\(_{\text{DIC}}\) in streams/rivers. For example, heavy precipitation in the SW region (2500±500mm) than the other regions tends to cause depletion in δ\(^{13}\)C\(_{\text{DIC}}\) values due to shorter residence time of soil water (Amiotte Suchet et al., 1999) while CO\(_2\) out gassing causes enrichment due to accumulation of \(^{13}\)C during diffusive efflux (Clark and Fritz, 1997) in stored water bodies. Many of the east flowing rivers are major and are dammed at many locations (e.g. Godavari, Krishna and Cauvery) for domestic, industrial and irrigation purposes. CO\(_2\)-out gassing due to
heterotrophic decomposition of organic matter and autotrophic production significantly alters the δ\(^{13}\)C\(_{\text{DIC}}\) signatures in reservoirs (Shin et al., 2001). Further, equilibrium with atmospheric CO\(_2\) in the reservoirs due to no/lean flow leads to enrichment in the δ\(^{13}\)C\(_{\text{DIC}}\) values (Brunet et al., 2005; Bouvillion et al., 2009; Zeng et al., 2011; Tamooh et al., 2013). Hence, relatively enriched δ\(^{13}\)C\(_{\text{DIC}}\) in the NE and SE estuaries could also be due to the storage of water in reservoirs/dams. A significant positive correlation between DIC concentrations and δ\(^{13}\)C\(_{\text{DIC}}\) (\(r^2=0.77; p<0.001\); Fig. 7), excluding the positive values, indicate that significant contribution of DIC from oxidation of particulate organic carbon in dams/reservoirs or stored water bodies. Shin et al. (2011) attributed the stream δ\(^{13}\)C\(_{\text{DIC}}\) values of -6.9±1.6‰ and -7.8±1.5‰ in silicate and carbonate dominated catchments, respectively, in tributaries of the Han River, South Korea to CO\(_2\) out gassing. Positive δ\(^{13}\)C\(_{\text{DIC}}\) values (>0‰) were observed only in Rushikulya (0.1‰), Nagavali (0.7‰) and Vaigai (2.5‰) in which relatively higher salinities (>20) were found during the study period. This is concurrent with earlier observations in the Indian estuaries, Hooghly (Samanta et al., 2015) and Cochin (Bhavya et al., 2016) where relatively enriched δ\(^{13}\)C\(_{\text{DIC}}\) were found at higher salinities. A strong positive correlation was found between δ\(^{13}\)C\(_{\text{DIC}}\) and salinity (\(r^2=0.71; p<0.001\)), suggesting that δ\(^{13}\)C\(_{\text{DIC}}\) values in the Indian estuaries are influenced by the intrusion of marine waters (δ\(^{13}\)C\(_{\text{DIC}}\): -1 to 2‰).

4.3.2 Total DIC export by the Indian monsoonal rivers to the north Indian Ocean

Indian monsoonal rivers annually export ~10.4 Tg of DIC to the north Indian Ocean. Nearly three fourth of this amount (7.7 Tg) reaches to the Bay of Bengal while the remaining into the Arabian Sea. It is mainly attributed to the annual freshwater discharge because to the Bay of Bengal annually receives (378 km\(^2\) of freshwater from the catchment area of about 0.96 M km\(^2\), whereas the Arabian Sea

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receives only \( (122 \text{ km}^3) \) of freshwater from the catchment area of only \( 0.23 \text{ M km}^2 \). Although the increase in volume of discharge dilutes the DIC flux from rivers (Jarvie et al., 1997; Shanley et al., 2002), bicarbonate fluxes to the Gulf of Mexico were reported to increase with the volume of discharge from the Mississippi river (Raymond and Oh, 2007) due to small dilution factor.

The total DIC export by the Indian monsoonal estuaries (10.4 Tg yr\(^{-1}\)) is only 2.5% of the total DIC export by the world major rivers (400 Tg yr\(^{-1}\)), and 9.4% of the export by the Asian rivers (111 Tg yr\(^{-1}\); Huang et al., 2012). The DIC export from the Indian estuaries is far less than the DIC export by the American (61.4 Tg yr\(^{-1}\)) and African (17.7 Tg yr\(^{-1}\)) rivers and major rivers draining to the tropical Atlantic from South America and Africa (53 Tg yr\(^{-1}\), Araujo et al. 2014).

It is mainly due to the fact that the volume of freshwater discharge from the Indian monsoonal rivers is very low (~500 km\(^3\) yr\(^{-1}\)) compared to the American (11,799 km\(^3\) yr\(^{-1}\)) and African (3,786 km\(^3\) yr\(^{-1}\)) rivers. However, the Indian monsoonal rivers are exporting DIC disproportionately higher DIC to the north Indian Ocean because they account for only 1.3% of the global river discharge but export 2.5% of the global riverine DIC to the oceans.

Disproportionate

Though American and African rivers account for 30% and 10% of the global river discharge, they export only 15% and 4.4% of global riverine DIC to oceans, respectively.

Higher DIC fluxes from the tropical regions are mainly attributed to the favourable climatic conditions, lithology and land use cover (Huang et al., 2012) in this region for higher dissolution as. Relatively higher export fluxes from the Indian rivers could be due to higher weathering rates of silicate and carbonate minerals were reported in the drainage basins of the Indian rivers (Das et al., 2005; Gurumurty et al., 2012; Pattanaik et al., 2013).

Krishna et al. (2015) reported that Indian monsoonal estuaries export 2.32 Tg yr\(^{-1}\) of dissolved organic carbon (DOC) to the north Indian Ocean. When combined...
dissolved carbon flux \((\text{DIC}+\text{DOC})\) would be 12.7 Tg yr\(^{-1}\) in which—This indicate that the total fluvial dissolved carbon export to the north Indian Ocean by the Indian monsoonal estuaries is predominantly contributed by DIC flux contributed up to \((-81\%\text{ and it is})\) than DOC, consistent with earlier reports elsewhere in the world, for example, the British rivers (80%, Jarvie et al., 2017). Since the catchment area of the Indian monsoonal rivers ranged widely from as low as 0.001 M km\(^2\) to as high as 0.313 M km\(^2\), the export fluxes of DIC were normalized with the catchment area of the river (yield) to obtain DIC yield from each river in order to examine various factors controlling the lateral DIC export to the north Indian Ocean.

4.4.3 Yield of DIC from the Indian monsoonal rivers

The yield of DIC (export normalized by catchment area) of DIC found in this study (mean 8.7±5.2 g m\(^{-2}\) yr\(^{-1}\)) is similar those found earlier in the rivers from the tropical region \((30^\circ\text{N}–30^\circ\text{S})\) of the Asian continent \((9.79\text{ g m}^{-2}\text{ yr}^{-1};\text{ Huang et al., 2012})\), but significantly higher than those reported from tropical region of the American \((3.33\text{ g m}^{-2}\text{ yr}^{-1})\) and African \((0.63\text{ g m}^{-2}\text{ yr}^{-1})\) continents of this tropical region (Table 4) (Huang et al., 2012). The yield of DIC from river catchment were reported to be controlled by the hydrological (precipitation, runoff and groundwater exchange) and environmental (temperature, type and dominance of soils, soil organic carbon, natural vegetation and forest cover) conditions and anthropogenic activities (land use change and deforestation) in the catchment (Raymond et al., 2008; Huang et al., 2012). Although the SW estuaries annually export relatively less DIC to the north Indian Ocean \((0.30\text{ Tg})\) due to their lower volume of discharge \((46\text{ km}^3\text{ yr}^{-1})\) from and relatively smaller catchment area \((0.02\text{ M km}^2)\) than the SE \((3.50\text{ Tg},102\text{ km}^3\text{ yr}^{-1}\text{ and }0.43\text{ M km}^2\text{ respectively}),\) NE \((4.21\text{ Tg},276\text{ and }0.53)\) and NW \((2.38\text{ Tg},75\text{ km}^3\text{ yr}^{-1}\text{ and }0.21\text{ M km}^2\text{ respectively})\) estuaries (Table 2 & 3), strikingly in contrast, the higher yield of DIC was found in the former \((10.8±6.6\text{ g m}^{-2}\text{ yr}^{-1})\)
than the latter (5.8±2.3, 8.6±5.7 and 9.5±3.9 g m⁻² yr⁻¹, respectively; Table 4). This suggests that strong control of catchment and/or in-stream processes on yield of DIC from the monsoonal rivers. However, DIC yield showed a significant positive correlation with the volume of discharge (r²=0.6667, p<0.001; Fig. 6d) in medium estuaries and no such relationship was found in the major estuaries. Significant negative relationships were observed between DIC yield and catchment area in the medium (r²=−0.5249, p<0.001; Fig. 6e) and major estuaries (r²=−0.439, p<0.001; Fig. 6f). This suggests that high precipitation over small catchments increases the DIC yield from the Indian estuaries because the dense precipitation increases the scouring-extraction of DIC from soils and rocks in their catchment. Therefore, high precipitation (2500±500mm) over the small catchment (0.02 M km⁻²) could have increased DIC yield from the SW estuaries. A strong linear relationship between the yield of DIC and the intensity of precipitation (r²=0.64, p<0.001 Fig. 8a-g) confirms that dense precipitation increases the export yield of DIC from SW estuaries. This could be one the reasons for the observed higher yield of DIC in the SW estuaries which receives high precipitation (2500±500mm) over the small catchment area (0.02 M km⁻²).

Ground water exchange do not appears to be controlling DIC yield from the Indian monsoonal estuaries because the groundwater DIC concentrations were lower in the SW (32±19 mg l⁻¹) than the other regions SE (106±56), NE (92±31) and NW (84±54 mg l⁻¹). Existing natural vegetation of tropical moist deciduous and tropical wet evergreen and semi evergreen forests in the SW region also could have increased DIC yield from the SW estuaries compared to the other estuaries as this vegetation favors the export fluxes of DIC. The drainage basins of the Indian monsoonal estuaries-rivers are largely under the tropical dry and wet climate except the SW rivers, Narmada and Tapti. The rivers Narmada and Tapti are under the arid and semiarid
climate while the SW rivers are under the tropical wet climate which was also reported to facilitate the riverine export of material from drainage basin to the coastal ocean.

Catchments of the SW rivers are largely occupied by the cation deficient lateritic soils and therefore precipitation of carbonate minerals in soils is poor. As a result, the soil inorganic carbon content in surface (100cm) soils of the catchment of SW rivers was lower than in catchments of the other monsoonal rivers studied (Sreenivas et al., 2016). On the other hand, the authors (Sreenivas et al., 2016) and Krishwan et al. (2009) found that the soil organic and inorganic carbon contents in the surface (100cm) soils of the catchment of SW rivers were higher and lower, respectively, in the former than the latter catchments of the SE, SW and NE rivers (Table 2). The relationship between soil inorganic and organic carbon is primarily dependent on the soil characteristics in the catchment. For example, Guo et al. (2016) demonstrated that increase in the soil organic carbon content enhanced the soil inorganic carbon in the cropland of upper Yellow river delta, China. A strong positive relationship between soil organic and inorganic carbon was also found in the Yanqi river basin, northwest China (Wang et al., 2015), and soils in the America (Stevenson et al., 2005) and Canada (Landi et al., 2003). This indicates that more dissolution of soil carbonates by acidic conditions formed by release of CO₂ through decomposition of soil organic carbon in catchments of the SW rivers. Hence, On the other hand, a negative relationship was found between soil organic and inorganic carbon in the North China Plain (Huang et al., 2006) and west Loess Plateau (Zeng et al., 2008). The negative relationship is mainly due to the higher production of CO₂ by decomposition of soil organic carbon and root respiration resulting in the formation of acidic conditions that lead to dissolution of soil carbonates. If the higher soil organic carbon in the catchment of the SW than in catchment of the SE, NE and NW rivers (Kishwan et al., 2009; Sreenivas et al., 2016) could have
elevated the yield of DIC from SW estuaries through dissolution of soil carbonates. Therefore, reduces more CO$_2$ through microbial decomposition and causes dissolution of soil carbonates leading to the higher yield of DIC from the SW estuaries. A significant linear correlation between soil organic carbon content and DIC yield in this study ($r^2=0.65$, p<0.001; Fig. S6h) suggests that strong influence of soil organic carbon content in the catchment on DIC yield from the Indian monsoonal rivers. However, the basin scale studies are required for comprehensive understanding of the influence of environmental and anthropogenic factors on DIC export fluxes from the Indian monsoonal rivers.

5. Summary

In order to examine the spatial variability of sources and distribution of dissolved inorganic carbon (DIC) concentrations and to identify its major sources in the Indian monsoonal estuaries, and to estimate the riverine export fluxes of DIC to the north Indian Ocean, we sampled a total of 27 major and medium estuaries along the Indian coast during wet period. An order of magnitude variability was found in DIC concentrations among the estuaries sampled (3.4 - 44.1 mg l$^{-1}$), with a lower mean concentrations of 6.6±2.1 mg l$^{-1}$ in estuaries located in the SW region of India. It is attributed to significant spatial variability in the size of rivers, precipitation pattern and lithology in their catchments. Magnitude of discharge, catchment area and in-stream processes are important factors for controlling the concentration and yield of DIC in the medium estuaries rather than the major estuaries. This is probably due to a significant spatial variability in lithology and hydro-geological and environmental conditions in the catchments. Indian monsoonal estuaries annually export ~10.4 Tg of DIC to the north Indian Ocean, of which 7.7 Tg enters in to the Bay of Bengal while the Arabian Sea receives only 2.7 Tg. It is mainly
attributed to the volume of river discharge as the former receives ~378 km$^3$ yr$^{-1}$ while the latter receives only 122 km$^3$ yr$^{-1}$ of freshwater from the Indian monsoonal rivers. The range of $\delta^{13}$C$_{\text{DIC}}$ found in this study suggests that major contribution of DIC is largely contributed from weathering of silicate and carbonate minerals by carbonic acid formed by dissolution of both soil and atmospheric CO$_2$. However, relatively enriched $\delta^{13}$C$_{\text{DIC}}$ in the east-flowing river estuaries indicated the storage of water in dams/reservoirs and intrusion of marine waters. Dense rainfall (2500±500mm) and higher soil organic carbon content (101.4 g ha$^{-1}$) in the catchment of SW rivers than in the catchment of the other rivers resulted in higher yield of DIC from the former than the latter.

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7. Data Availability

The data set used in the current study can be obtained from the corresponding author by an e-mail request.

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Figure 1: Map showing the study region. Estuaries of the rivers sampled in this study were indicated by solid black line.

Figure 2: Concentration (mg l⁻¹), export flux (Tg yr⁻¹) and yield (g m⁻² yr⁻¹) of dissolved inorganic carbon (DIC) in the Indian monsoonal estuaries. Estuaries geographically located in the northeastern (NE), southeastern (SE), southwestern (SW) and northwestern (NW) regions of India were also shown. Estuaries draining into the Bay of Bengal and the Arabian Sea were also provided.

Figure 3: Spatial variability in stable carbon isotopes of dissolved inorganic carbon (δ¹³CDIC, ‰) in the Indian monsoonal estuaries during discharge period.

Figure 4: (a) Positive correlation between dissolved inorganic carbon (DIC) concentration and catchment area, and (b) negative correlation between DIC concentrations and annual mean discharge (km³) of the minor-medium rivers.

Figure 5: Inverse correlation between mean dissolved inorganic carbon concentration in estuaries (DIC, mg l⁻¹) and annual mean rainfall (mm) in catchments of the rivers in the NE, NW, SE and SW regions of India.
**Figure 6:** Significant positive correlation between stable carbon isotopes of dissolved inorganic carbon ($\delta^{13}$C$_{DIC}$, ‰) and salinity in the Indian monsoonal estuaries during the study period.

*Figure 7:* Significant positive correlation between stable carbon isotopes of dissolved inorganic carbon ($\delta^{13}$C$_{DIC}$, ‰) and concentrations of DIC in the Indian monsoonal estuaries (filled diamonds), SW estuaries (filled squares) and high saline estuaries (hollow triangles) during the study period.

**Figure 8:** Relationship of dissolved inorganic carbon (DIC) yield (g m$^{-2}$ yr$^{-1}$) with that of (a) rainfall (mm) and (b) soil organic carbon (kg ha$^{-1}$) in the catchment area of the NE, NW, SE and SW rivers.
Fig. 1:
Fig. 2

DIC concentration (mg l⁻¹)
DIC yield (g m⁻² yr⁻¹)

DIC export load (Tg yr⁻¹)
Yield (g m⁻² yr⁻¹)

Arabian Sea
Bay of Bengal
NE
SE
NW
SW

Haldia
Subernarekha
Baitarani
Mahanadi
Rushikulya
Vamsadhara
Nagavali
Godavari
Krishna
Penna
Ponnayaar
Vaigai
Ambalyaar
Karaikudi
Cauvery
Chalakudi
Bharathappuzha
Nethravathi
Sharavathi
Khali
Mandovi
Zuari
Namadgi
Tapi
Sabarmati
Mahi

DIC
Fig. 3:
Fig. 4:

\[ y = 2629x - 2.329 \]
\[ R^2 = 0.719 \]

Fig. 5:

\[ y = -0.013x + 38.54 \]
\[ R^2 = 0.89 \]

Fig. 6:

\[ y = 0.2724x - 5.8759 \]
\[ R^2 = 0.71 \]
Fig. 7:

\[ y = 0.123x - 7.297 \quad R^2 = 0.765 \]

\[ y = 0.253x - 18.77 \quad R^2 = 0.954 \]

Fig. 8:

\[
\begin{align*}
\text{Dic yield (g m}^{-2}\text{yr}^{-1}) & \quad \text{Rainfall (mm)} \\
\text{Dic yield (g m}^{-2}\text{yr}^{-1}) & \quad \text{Soil OC (kg ha}^{-2}\text{)}
\end{align*}
\]

\[
\begin{align*}
\bar{y} = 0.001x + 6.560 \\
R^2 = 0.644
\end{align*}
\]

\[
\begin{align*}
\bar{y} = 0.048x + 6.150 \\
R^2 = 0.65
\end{align*}
\]