

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 $\delta^{13}\text{C}_{\text{DIC}}$. 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 $\delta^{13}\text{C}_{\text{DIC}}$ 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 $\delta^{13}\text{C}_{\text{DIC}}$ 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 $\delta^{13}\text{C}_{\text{DIC}}$ 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 m² 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 $\delta^{13}\text{C}_{\text{DIC}}$)

- 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

Lines 238-245: this is part of an introduction.

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

Lines 247 ff: the groups of rivers with high, intermediate and small discharges may be indicated.

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

Lines 255-260: this part is redundant, shorten.

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

Lines 320-337: introduction, see above. This part and the entire chapter have to be carefully checked. Atmospheric CO₂ has a value of -7 to -8 ‰ but dissolved in water; Line 323: this is not clear: CO₂ has a δ¹³C of -7 to -8 ‰ but when dissolved in water it is around 0 ‰ if the main anion is HCO₃⁻

Apologies for the mistake. Here, we meant to say that DIC originated by dissolution of atmospheric CO₂ (-7 to -8‰) is close to 0‰. It was corrected now. P. 4, L. 92

Chapter 4.4 may be substantially shortened and I would suggest to shift the discussion on the source of DIC to the earlier chapter 4.1.

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.

Lines 440ff is this contradicting the earlier discussion that groundwater is low in the SW and that this substantially is responsible for low concentrations?

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.

Lines 450ff: the whole discussion on lithology could also be better in chapter 4.1. May be a shorter discussion on the reason for the different yields (sediment/rock types and elevation; dams) would be sufficient.

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/iiodc>)

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 rely 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.

Cumbersomeness 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 δ¹³C_{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 $\pm 6 \mu\text{mol l}^{-1}$. 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 $30.86 \pm 1.23 \text{ }^\circ\text{C}$ to $31 \pm 1 \text{ }^\circ\text{C}$ (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 mentioningthe (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 (\pm 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

Lines 185-186: give mean±SD values.

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.

Lines 286-289: Grammatically sentence not correct.

The sentence has been modified. P. 14, L. 320-322.

Lines 304-307: Please comment, if not speculate, on whether these soil characteristics are limited only to surface or extended to the vertical strata as well, which can give an insight into whether the source of low DIC in these surface and ground waters are same or different.

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.

Lines 310-312: Weathering 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)

Lines 316-318: better integrate these with statements made at lines 365-471 and attribute to intense precipitation, presence of less weathering lateritic soils and soil organic carbon.

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

Lines 325-330: both the statements correspond to the weathering but the contribution of $\delta^{13}\text{C}_{\text{DIC}}$ values were differently reported. Pls check.

Though, both the statements correspond to the weathering of silicate and carbonate rocks by carbonic acid, the resulted $\delta^{13}\text{C}$ of DIC is different because it is based on the formation mechanism of carbonic acid. Carbonic acid can be formed by dissolution of soil CO_2 or atmospheric CO_2 , both of which have different $\delta^{13}\text{C}$ values. Weathering of silicate and carbonate rocks by soil CO_2 yield the $\delta^{13}\text{C}_{\text{DIC}}$ values of -17 to 21‰ and -7 to -8‰, respectively. Respective $\delta^{13}\text{C}_{\text{DIC}}$ values would be -7 to -8‰ and -3 to -4‰ if weathering occurs by dissolution of atmospheric CO_2 . Therefore, the $\delta^{13}\text{C}$ of DIC is dependent on the source for formation of carbonic acid, i.e. soil CO_2 or atmospheric CO_2 .

Lines 352-355: Repetition of statements at lines 326-330 but with clarity here. Avoid repetition.

Lines 352-355 have been deleted during the revision

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:

1. Bhavya, P.S., Sanjeev Kumar, Gupta, G.V.M., Sudharma, K.V., Sudheesh, V. (2018). Spatio-temporal variation in $\delta^{13}\text{C}_{\text{DIC}}$ of a tropical eutrophic estuary (Cochin estuary, India) and adjacent Arabian Sea. *Continental Shelf Research*, 153, 75-85, doi: 10.1016/j.csr.2017.12.006.
2. Gupta, G.V.M., Sarma, V.V.S.S., Robin, R.S., Raman, A.V., Jai Kumar, M., Rakesh, M. and Subramanian, B.R (2008). Influence of net ecosystem metabolism in transferring riverine organic carbon to atmospheric CO_2 in a tropical coastal lagoon (Chilka Lake, India). *Biogeochemistry*, 87: 265-285, doi:10.1007/s10533-008-9183-x

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 ~~found~~observed within the Indian estuaries sampled ($3.4 - 44.1 \text{ mg l}^{-1}$) due to ~~significant variability~~variations in the size of rivers, precipitation pattern and lithology in the catchments. Dilution with high precipitation ($2500 \pm 500 \text{ mm}$) and exchange with ground waters of low DIC resulted in very low concentrations of DIC in the estuaries located in the southwest of India ($6.6 \pm 2.1 \text{ mg l}^{-1}$) than the estuaries located in the southeast ($36.3 \pm 6.3 \text{ mg l}^{-1}$), northwest ($30.3 \pm 8.9 \text{ mg l}^{-1}$) and northeast ($19.5 \pm 6.2 \text{ mg l}^{-1}$) ~~regions~~ of India. ~~Though the range of~~ stable ~~carbon isotopes~~isotopic composition of DIC ($\delta^{13}\text{C}_{\text{DIC}}$) 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 ($\delta^{13}\text{C}_{\text{DIC}}$) in the east-flowing rivers.~~ It is estimated that the Indian monsoonal estuaries annually export $\sim 10.4 \text{ Tg}$ (~~$1 \text{ Tg} = 10^{12} \text{ g}$~~) 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~~ This is ~~mainly due to the fact that the Bay of~~

28 ~~Bengal receives higher~~consistent with the freshwater flux which is three times higher in the Bay
29 of Bengal ($\sim 378 \text{ km}^3 \text{ yr}^{-1}$) of freshwater from the catchment area of about 0.96 million km^2 ;
30 ~~whereas~~than the Arabian Sea ~~which receives only~~($\sim 122 \text{ km}^3 \text{ yr}^{-1}$). ~~of freshwater from the~~
31 ~~catchment area of only 0.23 million km^2 . ~~Though~~ Despite the discharge from the Indian
32 monsoonal rivers account for only 1.3% of global freshwater discharge, they disproportionately
33 export 2.5% of the total DIC export by the world major rivers and 9.4% of the Asian rivers to
34 oceans. The yield of DIC (DIC export normalized by the catchment area of the river) was found
35 to be higher in the SW estuaries ($10.8 \pm 6.6 \text{ g m}^{-2} \text{ yr}^{-1}$) than the SE ($5.8 \pm 2.3 \text{ g m}^{-2} \text{ yr}^{-1}$), NE
36 ($8.6 \pm 5.7 \text{ g m}^{-2} \text{ yr}^{-1}$) and NW ($9.5 \pm 4.0 \text{ g m}^{-2} \text{ yr}^{-1}$) other estuaries. Despite the SW estuaries though
37 they export only 0.3 Tg yr^{-1} of DIC, which is more than an order of magnitude lower than that of
38 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.,
39 higher yield of DIC from the SW estuaries ~~It is attributed, due~~ to intense precipitation (~ 3000
40 mm), favorable natural vegetation of tropical moist deciduous and tropical wet evergreen and
41 semi evergreen forests, and tropical wet climate, high soil organic carbon and the dominance of
42 red loamy soils in catchments of the SW rivers. This study, therefore, ~~reveals~~revealed that
43 significant variability ~~in of~~ the ~~lithology and~~hydrological (precipitation), lithological (bed rock
44 and soils) and environmental (vegetation and climate) conditions ~~over in~~ the catchments
45 strongly controls the concentrations and yield of DIC from the Indian monsoonal estuaries.~~

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

48 1. Introduction

49 Dissolved inorganic carbon (DIC) is the major constituent of carbon species and accounts
50 for $\sim 38\%$ of the total fluvial carbon transport to the global oceans (Meybeck, 1993; Cai, 2011;

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51 | Jarvie et al., 2017). World major river systems export annually ~~about~~ 33-400 Tg ($1\text{Tg}=10^{12}\text{g}$) of
52 | DIC to the global oceans (Ludwig et al., 1998; Mackenzie et al., 2004; Lerman et al., 2007).
53 | Chemical weathering of carbonate and silicate rocks and soils ~~and exchange with the ground~~
54 | ~~water~~ in the drainage basin are the major sources of DIC into rivers (Meybeck, 1987; Gaillardet
55 | et al., 1999, Dessert et al., 2001; Viers et al., 2007; Raymond et al., 2008; Tamooh et al., 2013).
56 | ~~However, The DIC concentrations in the estuaries are largely influenced by (i) the hydrological~~
57 | ~~(precipitation and runoff), lithological (type and dominance of rocks and soils) and~~
58 | ~~environmental (temperature, climate and vegetation) conditions, and (ii) anthropogenic activities~~
59 | ~~(deforestation and land use change) in the catchment, and (iii) physical and biological processes~~
60 | ~~such as exchange with ground water (Finlay, 2003; Shin et al., 2011; Maher et al., 2013), and~~
61 | ~~atmosphere, ie CO₂, autotrophic production and heterotrophic decompositionutilization of~~
62 | ~~organic matter (McConnaughey et al., 1994; Abril et al., 2003; Finlay and Kendall, 2007,~~
63 | ~~Hotchkiss et al., 2015; Zou, 2016). besides in-stream processes, such as oxidation of organic~~
64 | ~~carbon by heterotrophic bacteria (Mayogra et al., 2005; Battin et al., 2008; Hotchkiss et al.,~~
65 | ~~2015; Samantha et al. 2015; Zou, 2016) and dissolution of atmospheric carbon dioxide (CO₂).~~
66 | Weathering of carbonate and silicate rocks in the catchment, ~~and~~ uptake of DIC by aquatic plants
67 | ~~and algae during photosynthesis reaction~~ in rivers are the sinks ~~of for the the~~ atmospheric CO₂
68 | (e.g. Berner et al., 1983; Raymond et al., 2008); while the oxidation of organic carbon is the
69 | source of CO₂ to the atmosphere. ~~DIC in rivers and estuaries is therefore strongly linked to the~~
70 | ~~carbon eyele. However, d~~Due to human interferences, DIC fluxes from the world major rivers
71 | ~~have beenare~~ found to increase dramatically in the last century ~~, for example, the Mississippi~~
72 | ~~river (-90%); (Cai, 2003; Raymond and Cole, 2003; Raymond et al., 2008; Ren et al., 2015). It~~
73 | has been noted that substantial alterations in ~~DIC the~~ lateral transport ~~of DIC occurred~~ from land

74 | to sea occurred after the industrialization (Regnier et al., 2013; Bauer et al., 2013). The increase
75 | in riverine DIC flux ~~was has reported to have~~ a significant impact on the chemical composition
76 | (Williamson et al., 1994; Raymond and Cole, 2003; Findlay, 2010; Tank et al., 2010) and carbon
77 | budget in the coastal waters (Cole et al., 2007; Dhillon and Inamdar, 2013). ~~Theus~~, identification
78 | of major sources of DIC in the estuaries and quantification of their export fluxes and its riverine
79 | ~~export flux estimates~~ to the coastal oceans are important ~~for in better~~ understanding the carbon
80 | cycling ~~and its budget on both~~ in the regional ~~and as well~~ global scales (Campeau et al., 2017).

81 | Fluvial carbon fluxes from rivers in the tropical region (30° N to 30°S) ~~is are~~ critical for
82 | global carbon budgets because they contribute significant fraction ~~of to the~~ global DIC (48-64%),
83 | freshwater discharge (66.2%) and suspended sediment load (73.2%) to the world oceans, despite
84 | they occupy only ~43% of the world's land area (Huang et al., 2012). Further~~more~~, humid
85 | tropical climate ~~in over~~ the tropical region supports the export of ~~more~~ fluvial carbon ~~fluxes~~
86 | the continental land masses than the other climates in the world (Meybeck 1993; Ludwig et al.,
87 | 1998). However, the fluvial DIC fluxes from rivers in ~~the is tropical~~ region, except a few large
88 | river systems, were not to the global oceans are unknown included in estimating estimations of
89 | ~~the fluvial carbon fluxes to global oceans~~ due to the paucity of data.

90 | Numerous studies have been documented on DIC export flux from the world major
91 | rivers, for example, the Mississippi (Raymond and Cole, 2003; Raymond et al., 2008; Cai et al.,
92 | 2008), Changjiang and Pearl (Cai et al., 2008), Congo (Wang et al., 2013) and large river
93 | systems in the world (e.g. Gaillardet et al., 1999; Raymond et al., 2013). ~~Although-Though~~ some
94 | measurements were carried out on DIC in the Indian estuaries, for example, Mandovi and Zuari
95 | (Sarma et al., 2001), Godavari ~~estuary~~ (Sarma et al., 2011), Cochin (Gupta et al., -2009; Bhavya
96 | et al., 2016, 2018), Hooghly (Mukhopadhyay et al., 2002; Samanta et al., 2015), Mahanadi

97 (Pattanaik et al., 2017) and [Chila lake, a brackish water estuarine system \(Gupta et al., 2008\)](#)
98 [Indian estuaries \(Sarma et al., 2012\)](#), however, they ~~focus was mainlyed only~~ on [internal cycling](#)
99 [of carbon and exchange at the air-water interface. exchange of CO₂](#). [Carbon export fluxes from](#)
100 [the Chilka lake \(Gupta et al., 2008\) and Cochin estuary \(Gupta et al., 2009\) on east and west](#)
101 [coast of India respectively have been](#) were reported but their sources were not evaluated.
102 ~~Nevertheless, no estimations have been made so far on DIC export fluxes to the north Indian~~
103 ~~Ocean from the Indian subcontinent have been made so far. For the first time, we made an effort~~
104 ~~here to identify the major sources of DIC in the Indian monsoonal estuaries and to estimate their~~
105 ~~export fluxes to the north Indian Ocean.~~

106 ~~The stable isotopic composition of DIC ($\delta^{13}\text{C}_{\text{DIC}}$) is a well established and widely used~~
107 ~~tracer to identify the major sources of DIC in rivers~~ [the aquatic system \(e.g. Singh et al., 2005;](#)
108 [Tamooh et al., 2013; Samanta et al., 2015; Zou, 2016\)](#) ~~because each of the DIC sources have~~
109 ~~due to distinct $\delta^{13}\text{C}_{\text{DIC}}$ ratios~~ [isotopic composition of different sources \(Deines et al., 1974\).](#) ~~The~~
110 ~~isotopic composition of~~ [DIC originated by dissolution of atmospheric CO₂ is about -7 to -8‰](#)
111 [\(Coplen et al., 2002\)](#) whereas it is about -26 to -27‰ if ~~the~~ [DIC is derived from oxidation of](#)
112 [organic matter produced by C₃ plants \(O'Leary, 1988\).](#) ~~The $\delta^{13}\text{C}$ of DIC generated by soil CO₂~~
113 ~~dissolved carbonic acid weathering of silicates is about -17 to -21‰ (Solomon and Cerling,
114 ~~1987) while it is close~~ [in the range of -10 to -9‰ for carbonate rocks because half of the carbon](#)
115 [comes from carbonate rocks \(0‰, Land, 1980\) during weathering. Whereas, the weathering of](#)
116 ~~silicate and carbonate and silicate minerals yield $\delta^{13}\text{C}_{\text{DIC}}$ values in the range of -7 to -8‰ and -3~~
117 ~~to -4‰, respectively, if the carbonic acid formed by the dissolution of atmospheric CO₂.~~
118 ~~Although, Despite distinct isotopic composition of DIC is expected for different sources, DIC~~
119 ~~derived from different sources have distinctly different $\delta^{13}\text{C}_{\text{DIC}}$~~ [the identification of DIC sources](#)~~

120 ~~values, however, the interpretation the $\delta^{13}\text{C}_{\text{DIC}}$ values for identification of its sources is still~~
121 ~~challenging (Amiotte-Suchet et al., 1999; Campeau et al., 2017) due to the isotopic fractionations~~
122 ~~associated with complex mixture of sources and processes such as photosynthesis (O'Leary,~~
123 ~~1988; Finlay, 2004; Parker et al., 2005, 2010), respiration (Finlay, 2003; Waldron et al., 2007),~~
124 ~~DOC photo-oxidation (Opsahl and Zepp, 2001; Vahatalo and Wetzel, 2008), anaerobic~~
125 ~~metabolism (Waldron et al., 1999; Maher et al., 2015) and equilibration with atmospheric CO_2 .~~
126 We made an effort for the first time to identify the major sources of DIC in the Indian monsoonal
127 estuaries and quantify their export fluxes to the north Indian Ocean. The main objectives of this
128 study are to (i) identify the major sources and (ii) examine ~~the~~ potential reasons responsible for
129 DIC variability ~~in concentrations of DIC~~ in the Indian monsoonal estuaries during the discharge
130 (wet) period, and (iii) estimate the DIC export fluxes to the north Indian Ocean by the Indian
131 monsoonal rivers.

132 **2. Study region, sampling and Sampling methods**

133 **2.1 Study Area**

134 The Indian peninsula bifurcate the north Indian Ocean into the Bay of Bengal and the
135 Arabian Sea. Although these two basins occupy ~~ies~~ the same latitudinal belt, their oceanographic
136 processes were reported to be remarkably different ~~and attributed due~~ to higher freshwater flux
137 into the Bay of Bengal ($1.63 \times 10^{12} \text{ m}^3 \text{ yr}^{-1}$) than Arabian Sea ($0.3 \times 10^{12} \text{ m}^3 \text{ yr}^{-1}$; Subramanian,
138 1993; Gauns et al., 2005). ~~significant differences in the freshwater influx and associated~~
139 ~~physical and biological changes (Gauns et al., 2005). This is because the glacial and peninsular~~
140 ~~rivers transport $1.63 \times 10^{12} \text{ m}^3 \text{ yr}^{-1}$ of freshwater to the Bay of Bengal (Subramanian, 1993)~~
141 ~~whereas only $0.3 \times 10^{12} \text{ m}^3 \text{ yr}^{-1}$ to the Arabian Sea.~~ The large freshwater influx leads to the
142 formation of a strong vertical salinity stratification in the Bay of Bengal (Varkey et al., 1996);

143 ~~which results in the suppression of~~that prevents vertical mixing of nutrient rich sub-surface water
144 with ~~that of surface~~ (Prasanna Kumar et al., 2004), ~~makes the Bay of Bengal~~. As a result, the
145 ~~Bay of Bengal is considered to be~~ -relatively less productive - (Prasannakumar et al., 2002) than
146 the adjacent Arabian Sea, which is one of the highly productive zones in the world (Madhupratap
147 et al., 1996; Smith, 2001; Barber et al., 2001) due to injection of nutrients into surface through
148 the seasonal upwelling and convective mixing (Shetye et al., 1994; Madhupratap et al., 1996;
149 Muraleedharan and Prasannakumar, 1996).

150 Discharge from the Indian ~~peninsular-monsoonal~~ rivers is largely fed by the monsoon
151 induced precipitation over the Indian subcontinent, which receives >80% of its annual rainfall
152 during the southwest (SW) monsoon period (June-September) (Soman and Kumar, 1990).
153 Though some amount of rainfall occurs during the NE monsoon (December-March), it ~~will does~~
154 not generate discharge as it will be stored within the ~~dams and~~ reservoirs for domestic, industrial
155 and irrigation purposes. Discharge from the Indian ~~peninsular-monsoonal~~ rivers ~~is therefore~~
156 mainly occurs only during the SW monsoon season (Vijith et al., 2009; Sridevi et al., 2015) ~~and~~
157 hence, ~~termed these rivers are called~~ as monsoonal rivers. Since the major portion of the annual
158 freshwater discharge from ~~the~~ Indian monsoonal rivers ~~is limited to only few months (June—~~
159 ~~October) in a year~~ occurs only during the SW monsoon, ~~unlike the European and American~~
160 ~~rivers,~~ the entire estuary ~~may be~~ is filled with a freshwater ~~without any vertical salinity gradient~~
161 (Vijith et al., 2009; Sridevi et al., 2015) ~~during this period~~. As ~~virtually there is no~~ discharge is
162 small during the ~~dry period~~ rest of the year, the discharge during ~~the~~ SW monsoon (wet period) is
163 ~~considered to be~~ equivalent to the annual discharge ~~from of~~ the monsoonal rivers. Based on ~~the~~
164 rainfall intensity, forest cover, vegetation and soil type in the catchment, estuaries sampled in the
165 present study were categorized into 4 groups, namely the northeast (NE), southeast (SE),

166 southwest (SW) and northwest (NW) estuaries of India (Fig. 1). The SW region of India is
167 characterized by the intense rainfall during ~~the~~-SW monsoon (~3000 mm) following ~~the~~-NE
168 (1000-2500 mm), SE (300-500 mm) and NW (200-500 mm) regions of India (Soman and
169 Kumar, 1990). The SW rivers drain red loamy soils while the NW rivers drain black soils.
170 ~~Except the major rivers Godavari and Krishna, all t~~The rivers reaching ~~the~~-Bay of Bengal (NE
171 and SE estuaries) drain ~~the~~-red loamy and alluvial soils in their upper and lower catchments
172 respectively. ~~except the major rivers. The~~ Godavari and Krishna ~~rivers, which also~~ drain black
173 soils in their upper catchment along with red loamy and alluvial soils in their middle and lower
174 catchments respectively (Geological Survey of India; www. gsi.gov.in). Based on ~~the~~-discharge,
175 ~~the~~ monsoonal estuaries in this study were divided into two types, namely, the major (>150 m³ s⁻¹
176)~~minor and medium~~ (<150 m³ s⁻¹) ~~and major (>150 m³ s⁻¹)~~ estuaries.

177 2.2 Sample collection

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

188 | mid-stream of the estuary using a local mechanized boat to avoid the contamination from river
189 | banks.

190 | ~~In-situ~~In-situ measurements and sample collection ~~was-were done-conducted~~ in the-27
191 | estuaries along the Indian coast (Fig. 1) during the SW monsoon season of the years, 2011 and
192 | 2014. Surface water samples at each location were collected for phytoplankton biomass (Chl-*a*),
193 | DIC and dissolved oxygen (DO). Samples for DIC were collected in air-tight crimp-top glass
194 | bottles and added poison (mercuric chloride) to arrest the biological activity. DO analysis was
195 | carried out at a temporary shore laboratory set up for sample processing after the completion of
196 | sampling on each day. Water samples were filtered through GF/F (nominal pore size of: 0.7µm)
197 | under moderate vacuum and stored in liquid nitrogen for Chl-*a* analysis ~~at the NIO~~.

198 | 2.3. Methods

199 | Temperature and salinity at the sampling locations were measured using a conductivity-
200 | temperature-density (CTD) profiling system (Sea Bird Electronics, SBE 19 plus, United States of
201 | America). Concentrations of DO ~~were-was~~ determined by a Winkler's method (Carritt and
202 | Carpenter, 1966) using an auto titrator (Metrohm, Switzerland) with potentiometric end point
203 | detection. The analytical precision of the method was $\pm 0.07\%$ (RSD). DIC concentrations in
204 | water samples were measured at our Institute laboratory using Coulometer (UIC Inc., USA)
205 | connected to an automatic sub-sampling system. Based on the repeated analysis of samples and
206 | standards, the precision of the method was $\pm 1.8 \mu\text{mol l}^{-1}$. The certified reference materials
207 | (CRM) supplied by Dr. A.G. Dickson, Scripps Institute of Oceanography, USA and internal
208 | standards were used to test the accuracy of our DIC measurements and it was found to be within
209 | ± 0.2 to 0.3% . Chlorophyll-*a* (Chl-*a*) on the filter was extracted into di-methyl formamide
210 | (DMF) and measured the extract fluorometrically using a spectrofluorophotometer (Varian

211 Eclipse, Varian Electronics., UK) following Suzuki and Ishimaru (1990). Annual mean
212 discharge data of the rivers was taken from Meybeck and Ragu (1995, 1996), Central Water
213 Commission, New Delhi (2006, 2012) and Kumar et al. (2005). Catchment area of the rivers
214 was obtained from Water Resources Information System of India (WRIS, [www.india-](http://www.india-wris.nrsc.gov.in)
215 [wris.nrsc.gov.in](http://www.india-wris.nrsc.gov.in)). Soil organic carbon data was taken from Kishwan et al. (2009) and Sreenivas
216 et al. (2016), and the rainfall data was obtained from Soman and Kumar (1990). Dissolved
217 organic carbon (DOC) data for the Indian estuaries was taken from Krishna et al. (2015)-

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

228 **3. Results**

229 3.1. Hydrographic characteristics

230 ~~Prevailing hydrographic conditions in Indian estuaries during the sample collection were given in~~
231 ~~detail elsewhere (Sarma et al., 2012, 2014; Krishna et al., 2015). Briefly, mentioned here for~~
232 ~~ready reference.~~ Surface water temperature was ~~found to be~~ higher in the estuaries located on
233 the east coast (mean $30.869 \pm 1.23^\circ\text{C}$) than the west coast ($27.32 \pm 1.49^\circ\text{C}$) of India. Salinity

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234 varied broadly from near zero (0.106) to 28.78 during the study period. Relatively higher
235 salinities (>20) were recorded by the medium estuaries, which receives relatively lower
236 freshwater discharge from the upstream river, for example, Nagavali (28.78=8), Vaigai (24.63)
237 and Rushikulya (20.70). Mean salinities were lower in the west-flowing NW (0.1±0.02) and SW
238 (2.1±2.8) estuaries than the east-flowing SE (9.5±7.8) and NE (8.5±11) estuaries. Dissolved
239 oxygen saturation varied from as low as 62.63% to as high as 105%, with a mean saturation of
240 89.90±11.4% in the estuaries sampled. The SW estuaries recorded relatively slightly lower DO
241 saturation (82±7%) than the NE (89±15%), NW (93±3%) and SE (96±11%) estuaries.
242 Chlorophyll-*a* (Chl-*a*) concentrations varied broadly from 0.8 to 10.7.5 mg m⁻³, with relatively
243 higher mean concentrations in the SE (4.7±2.5 mg m⁻³) followed by the SW (3.02.8±0.7 mg m⁻³)
244 estuaries (Table 2). ~~However~~ On the other hand, relatively low Chl-*a* was observed in the
245 medium (2.6±1.3 mg m⁻³) than in the major estuaries (3.2±2.1 mg m⁻³).

246 3.1.2 DIC concentrations and $\delta^{13}C$ of DIC ($\delta^{13}C_{DIC}$) in the Indian monsoonal estuaries

247 DIC concentrations in the Indian monsoonal estuaries widely varied from 3.4
248 (Bharathappuzha) to 44.1 mg l⁻¹ (Vellar), with a significant spatial variability (Fig. 2). More than
249 five times higher mean concentrations were observed in the SE (36.3±6.3 mg l⁻¹) and NW
250 estuaries (30.3±8.9 mg l⁻¹) than in the SW ~~estuaries~~ (6.6±2.1 mg l⁻¹); and ~~intermediate~~
251 ~~concentrations were found in the~~ NE estuaries (19.5±6.2 mg l⁻¹). DIC concentrations were found
252 to be similar (~~homoscedastic Student's t-test; p=0.76~~) in the major (22.7±13.6 mg l⁻¹) and
253 medium (21.1±13.2 mg l⁻¹) estuaries (homoscedastic Student's t-test; p=0.76). The $\delta^{13}C_{DIC}$
254 varied from -13.0 to 2.5‰, with a significant spatial variability (Fig. 3) in the estuaries sampled.
255 Relatively depleted $\delta^{13}C_{DIC}$ values were observed in the west flowing estuaries of NW (-

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256 11.1±2.3‰) and SW (-7.4±1.9‰) than the east flowing estuaries of NE (-3.5±2.8‰) and SE (-
257 2.7±5.2‰) regions of India.

258 *3.3. Export fluxes and yield of DIC*

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259 Annual export flux of DIC ~~from the individual estuaries to the~~ coastal ocean ~~from~~
260 ~~individual estuaries~~ varied broadly from 0.01~~09~~ Tg (Chalakkudi) to as high as 2.3~~2~~ Tg (Krishna).
261 Annually, the NE estuaries export higher DIC flux ~~of (4.21 Tg)~~ followed by the SE (3.5~~0~~ Tg) and
262 NW estuaries (2.3~~8-4~~ Tg). ~~Whereas~~In contrast, the SW estuaries recorded the lowest export flux
263 of 0.3~~0~~ Tg which is an order of magnitude lower than that of the export flux by ~~the NE and~~
264 ~~SE~~other estuaries (Fig. 2). The Indian monsoonal estuaries together export about 10.4 Tg yr⁻¹ of
265 DIC to the northern Indian Ocean, of which 7.7 Tg (74.~~2~~%) enters into the Bay of Bengal and the
266 remaining into the Arabian Sea (2.7 Tg). The estuaries, Krishna (2.3~~2~~ Tg), Godavari (1.4~~5~~ Tg)
267 and Haldia (1.4~~6-2~~ Tg) together responsible for the transport of 6465% of total riverine DIC
268 export to the Bay of Bengal ~~by the Indian monsoonal rivers~~. The yield of DIC ranged from 2.7
269 (Bharathappuzha) to 21.6 g m⁻² yr⁻¹ (Mandovi), excluding the exceptionally high yield of 113.4 g
270 m⁻² yr⁻¹ from Haldia estuary. The west flowing rivers to the Arabian Sea are characterized by
271 relatively higher yield of DIC (mean 10.4±5.6 g m⁻² yr⁻¹) than the east flowing rivers to the Bay
272 of Bengal (7.3±4.6 g m⁻² yr⁻¹). Among the estuaries sampled, the SW and SE estuaries recorded
273 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
274 intermediate values noticed in the. ~~The~~ NW (9.5±4.0 g m⁻² yr⁻¹) and NE (8.6±5.7g m⁻² yr⁻¹)
275 estuaries ~~recorded intermediate values~~.

276 4. Discussion

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

289 **4.1 Variability-Distribution and sources of DIC ~~concentrations~~ in the Indian monsoonal** 290 **estuaries**

291 Mean DIC concentration found in this study ($21.9 \pm 13.2 \text{ mg l}^{-1}$; ~~range: 3.4 to 44.1 mg l⁻¹~~)
292 is similar to those observed earlier in the Indian estuaries such as Ganga-Brahmaputra and
293 Hooghly (Singh et al., 2005, Samanta et al., 2015), and in ~~the~~ estuaries elsewhere in the world,
294 for example York, Yangtze, Seri and Xi etc (Raymond and Bauer, 2000, Cai et al., 2008,
295 Ishikawa et al., 2015; Zou, 2016) (Table 1), ~~the Indian estuaries, for example, Ganga-~~
296 Brahmaputra (23 mg l⁻¹; Singh et al., 2005), Hooghly (21.8 mg l⁻¹; Samanta et al., 2015) and
297 Mahanadi (15.0; Pattanaik et al., 2017) and elsewhere in the world, for instance, York river
298 estuary (6-21 mg l⁻¹; Raymond and Bauer, 2000), Yangtze river (28 mg l⁻¹; Cai et al., 2008),
299 British rivers (median 4-43 mg l⁻¹; Jarvie et al., 2017), Seri, central Japan (17.6-21.9 mg l⁻¹;

300 ~~Ishikawa et al., 2015), the Red river, Vietnam (9.1–29.9 mg l⁻¹; Quynh et al., 2016) and Xi river,~~
301 ~~southwest China (18–45.6 mg l⁻¹; Zou, 2016). However, The mean-DIC concentrations in the~~
302 ~~Indian estuaries are higher than those found in some of the the-Asian rivers of tropical region~~
303 ~~(12.7 mg l⁻¹, Huang et al., 2012) and Indian estuaries (21.9±13.2 mg l⁻¹) are higher than the~~
304 ~~global mean of (10.3 mg l⁻¹ (Meybeck and Vorosmarty, 1999) (Table 1), and the Asian rivers~~
305 ~~(12.7 mg l⁻¹) in the tropical region (30°N–30°S; Huang et al., 2012), but lower than those reported~~
306 ~~in the rivers draining into the Gulf of Trieste (N Adriatic;) (37–66 mg l⁻¹; Tamse et al., 2014)~~
307 ~~(Table 1).-~~

308 Among the estuaries sampled along the Indian coast, the SW estuaries are characterized by
309 significantly lower mean concentrations of DIC (6.6±2.1 mg l⁻¹) than the SE (36.3±6.3 mg l⁻¹),
310 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~~
311 ~~considerable spatial variations in the (i) hydrological, lithological and environmental conditions~~
312 ~~in the catchments and (ii) in-stream physical and biogeochemical processes. DIC concentrations~~
313 ~~in estuaries are mainly governed by the hydrological (precipitation and runoff), lithological (type~~
314 ~~and dominance of rocks and soils) and environmental (temperature, climate and vegetation)~~
315 ~~conditions, and anthropogenic activities (deforestation and land use change) in the catchment,~~
316 ~~and in stream physical and biological processes such as exchange with ground water (Finlay,~~
317 ~~2003; Shin et al., 2011; Maher et al., 2013) and atmospheric CO₂, autotrophic production and~~
318 ~~heterotrophic decomposition of organic matter (McConnaughey et al., 1994; Abril et al., 2003;~~
319 ~~Finlay and Kendall, 2007).~~

320 4.1.1. The impact of hydrological conditions

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321 The SW region of India receives the highest amount of rainfall precipitation during the
322 SW monsoon (2500±500 mm) than the SE (400±50 mm), NE (1000±200 mm) and NW
323 (750±250 mm) regions of India (Table 2) (Soman and Kumar, 1990). Though the intense
324 rainfall precipitation in the SW region is expected to cause higher weathering rates and therefore
325 higher DIC (e.g., Gupta et al., 2011), but the observed lower DIC concentrations were found in
326 the SW estuaries. This is attributed could be due to the influence of dilution because the
327 catchment area normalized volume of discharge was found to be higher in the SW estuaries (1.71
328 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
329 three times higher catchment area normalized discharge might have diluted, suggesting that
330 significant dilution of DIC concentrations in the SW estuaries. A strong negative correlation
331 between precipitation in the catchment and DIC concentration in estuaries (r²= 0.89, p<0.001;
332 Fig. 54a) also suggest confirms that DIC concentration in Indian estuaries are rather controlled by
333 the intensity of precipitation over the catchment. Dilution of DIC by heavy precipitation in the
334 SW region can also be seen from relatively depleted δ¹³C_{DIC} values (-7.4±1.9‰) in the SW
335 estuaries because the shorter residence time of soil water depletes the δ¹³C_{DIC} due to preferential
336 dissolution of ¹²CO₂ over ¹³CO₂ (Amiotte-Suchet et al., 1999).

337 - Since many of the hydrological processes are largely dependent on the size of the river
338 and its catchment area, the lower it may govern the DIC concentrations of DIC. The lower
339 concentrations of DIC in the SW estuaries of this study could be may possibly due to the smaller
340 catchment area as size of the rivers. This is because, the SW rivers are small, both in terms of
341 discharge (46 km³ yr⁻¹) and catchment area (total catchment area: 0.02 M km²), than that of SE
342 (102 km³ yr⁻¹ and 0.45 M km², respectively), NE (276 km³ yr⁻¹ and 0.53 M km²) and NW (75 km³
343 yr⁻¹ and 0.21 M km²) rivers (Table 2). However, The DIC concentrations of DIC in the Indian

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344 estuaries showed a significant positive relationship with catchment area ($r^2=0.75$; $p<0.001$; Fig.
345 4a4b) and a negative relationship with volume of discharge ($r^2=0.57$; $p<0.001$; Fig. 4b4c) only in
346 the medium estuaries (discharge: $<150 \text{ m}^3\text{s}^{-1}$), suggesting that an area of catchment and
347 magnitude of discharge controls DIC concentrations largely in the medium estuaries rather than
348 in the major estuaries. It could be due to the influence of in-stream processes as the major rivers
349 are long compared to the medium rivers.

350 Mixing with seawater and exchange of submarine ground water also influences DIC
351 concentrations in the estuaries. Since this study was conducted during the SW monsoon, many
352 of the estuaries are filled with freshwater (salinity >1) due to maximum discharge during this
353 period. On the other hand, higher salinities (>20) were observed in some medium estuaries,
354 namely, Rushikulya, Nagavali and Vaigai recorded higher salinities (>20) due to low flow from
355 upstream river. As a result, the $\delta^{13}\text{C}_{\text{DIC}}$ values were found to be $>0\%$ in these three estuaries
356 (0.1, 0.7 and 2.5‰ respectively). A strong positive correlation was found between $\delta^{13}\text{C}_{\text{DIC}}$ and
357 salinity (Fig. 64d; $r^2=0.71$, $p<0.001$), suggesting that $\delta^{13}\text{C}_{\text{DIC}}$ values in the Indian estuaries
358 are also influenced by the intrusion of marine waters ($\delta^{13}\text{C}_{\text{DIC}}$: -1 to -2%), particularly in
359 medium estuaries. The $\delta^{13}\text{C}_{\text{DIC}}$ values were found to be $>0\%$ in Rushikulya, Nagavali and
360 Vaigai estuaries (0.1, 0.7 and 2.5‰ respectively) suggesting that major contribution of DIC is
361 from intrusion of marine water.

362 As found in many estuaries over the world, submarine ground water exchange submarine
363 groundwater discharge is found to contribute up to 52% of DIC in the Godavari estuary strongly
364 influences DIC concentrations in Indian estuaries, for example, (Rengarajan and Sarma, (2015)
365 due to higher concentrations of DIC by found 3 to 4 times higher DIC concentrations in the in
366 the ground water compared than estuary. the estuarine waters of the Godavari and estimated that

367 ~~submarine ground water discharge contributes up to 52% of DIC concentrations in the Godavari~~
368 ~~estuarine system. –The measured DIC concentrations in ground waters along the entire Indian~~
369 ~~coast (Dr. BSK Kumar, personal communication) showed strong spatial variability with suggest~~
370 ~~relatively lower concentrations in the SW (mean 32±19 mg l⁻¹) than the SE, (106±56), NE~~
371 ~~(92±31) and NW (84±54 mg l⁻¹)-regions of India (Table 2) during discharge period (Dr. BSK~~
372 ~~Kumar, personal communication). Though the DIC concentrations inExchange of SW estuaries~~
373 ~~with ground water with relatively lower DIC ground waters with that of estuarine water were~~
374 ~~higher by about 3 to 5 times than the concentrations found in the Indian estuaries, however,~~
375 ~~exchange of ground water with relatively low DIC concentrations in the SW region could have,~~
376 ~~at least partly, concentrations might have possibly yielded low DIC concentrations. Nevertheless~~
377 ~~it is difficult to ascertain the impact of ground water exchange yielded low DIC in the SW~~
378 ~~estuaries due to lack of submarine ground water discharge rates. can caused the lower DIC~~
379 ~~concentrations in the SW estuarine waters.es. However,~~

380 4.1.2. The impact of in-stream processes

381 ~~Since the~~ Indian monsoonal estuaries ~~were-have been~~ reported ~~as-to be~~ a source of CO₂ to the
382 atmosphere during ~~the~~ discharge period (Sarma et al., 2001, 2011, 2012; Gupta et al., 2008,
383 2009; Bhavya et al., 20162018), ~~due to the microbial decomposition of terrestrial organic matter~~
384 ~~brought by the rivers. This suggests that~~ the DIC input from dissolution of atmospheric CO₂ ~~in~~
385 ~~estuaries~~ can be ruled out. ~~CO₂ release due to, however,~~ heterotrophic decomposition of organic
386 matter adds significant amount of DIC to the Indian estuaries during ~~discharge-this~~ period ~~as~~
387 ~~enhanced bacterial respiration rates were reported in the Indian estuaries (Sarma et al., 2011;~~
388 ~~2012).~~ A fairly good positive correlation between DIC and DOC concentrations ($r^2=0.340$,
389 $p<0.01$; Fig. 4e), except few medium estuaries, suggests that DIC addition through microbial

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390 degradation of ~~particulate~~ organic matter ~~is significant~~ seems to be possible source in the Indian
391 estuaries. ~~Except the NW estuaries, which recorded relatively depleted $\delta^{13}\text{C}$ of DIC ($\delta^{13}\text{C}_{\text{DIC}}$),~~
392 ~~the~~ A positive correlation between $\delta^{13}\text{C}_{\text{DIC}}$ and DOC ~~concentrations~~ was observed, with
393 different slope for NW estuaries ($r^2=0.3543$, $p<0.01$; Fig. 4f), as was observed elsewhere (Xi
394 river, Zou et al., 2016), confirmings that oxidation of organic matter ~~is may be~~ one of the main
395 major DIC sources in the Indian monsoonal estuaries. Similar relationship was also observed in
396 the Xi river (Zou et al., 2016). The range of $\delta^{13}\text{C}_{\text{DIC}}$ (-13.0 to 2.5‰) in the Indian monsoonal
397 estuaries is distinctly enriched than that of the $\delta^{13}\text{C}$ of DIC derived from decomposition of
398 terrestrial C_3 plant derived organic matter (-27 to -26‰, O'Leary, 1988; Fig. 5), suggesting that
399 DIC might have been contributed from decomposition of terrestrial C_4 plants (-17 to -13‰,
400 Krishna et al., 2015) and ~~significant amount of DIC is contributed by $\delta^{13}\text{C}$ enriched sources such~~
401 as weathering of silicate and carbonate rocks. In addition, if weathering occurs due to
402 dissolution of silicate and carbonate rocks due to atmospheric CO_2 , the $\delta^{13}\text{C}_{\text{DIC}}$ yields -8 to -7‰
403 and -4 to -3‰ respectively. On the other hand, the $\delta^{13}\text{C}_{\text{DIC}}$ would be -10 to -9‰ and -21 to -17‰
404 (Solomon and Cerling, 1987) if the dissolution of silicate and carbonate rocks occurs due to soil
405 CO_2 respectively. As discussed above, flux of CO_2 from atmosphere to river cannot be expected
406 due to super-saturation of riverine CO_2 , weathering of silicate and carbonate rocks by dissolution
407 of soil CO_2 may be possible. Though isotopic composition of $\delta^{13}\text{C}_{\text{DIC}}$ derived from
408 decomposition of C_4 plants and weathering due to soil CO_2 are similar and difficult to separate,
409 Sarma et al. (2014) measured isotopic composition of $\delta^{13}\text{C}_{\text{POC}}$ and found that >90% of the POC
410 is contributed by C_3 plants. Hence possible contribution of DIC through decomposition of C_4
411 plants may be negated.

412 ~~by carbonic acid produced by dissolution of atmospheric CO₂ (7 to 8‰ and 3 to 4‰~~
413 ~~respectively) besides the contribution from. On the other hand, autotrophic production removes~~
414 ~~DIC as it converts DIC to organic carbon.~~ Significant negative correlation between chlorophyll-
415 ~~a and DIC concentrations~~ ($r^2=0.47$, $p<0.01$; Fig. 6a), except few SE estuaries where elevated
416 phytoplankton biomass (Chl-a: $>5 \text{ mg m}^{-3}$) was recorded, suggesting that autotrophic removal of
417 ~~DIC is also may be possible sink significant~~ in the Indian monsoonal estuaries during the study
418 period. ~~This process would enrich $\delta^{13}\text{C}_{\text{DIC}}$ due to preferential removal of $^{12}\text{CO}_2$ over $^{13}\text{CO}_2$~~
419 ~~during photosynthesis. A positive relationship was observed between $\delta^{13}\text{C}_{\text{DIC}}$ and Chl-a in the~~
420 ~~Indian estuaries ($r^2=0.50$; $p<0.01$), suggesting that biological removal of DIC enriched $\delta^{13}\text{C}_{\text{DIC}}$.~~
421 ~~In contrast, Significance of DIC addition by~~ heterotrophic decomposition of organic matter
422 ~~(respiration) depletes $\delta^{13}\text{C}_{\text{DIC}}$ due to release of $^{12}\text{CO}_2$ over $^{13}\text{CO}_2$ during this process. Due to lack~~
423 ~~of respiration rates data, we could not able to evaluate its influence. Nevertheless, the dissolved~~
424 ~~oxygen saturation stores the net effect of biological production and heterotrophic respiration,~~
425 ~~and removal by autotrophic production in the Indian estuaries was confirmed. In order to confirm~~
426 ~~the net biological influence on $\delta^{13}\text{C}_{\text{DIC}}$, the same is correlated with DO saturation and found by a~~
427 ~~fairly good significant~~ positive correlation ~~between $\delta^{13}\text{C}_{\text{DIC}}$ and dissolved oxygen saturation~~
428 ($r^2=0.49$, $p<0.01$; Fig 6b), (depleted $\delta^{13}\text{C}_{\text{DIC}}$ values at low % of DO saturation), except NW
429 estuaries, which recorded depleted $\delta^{13}\text{C}_{\text{DIC}}$ ($<-10.0\%$) ~~confirming that biological processes~~
430 ~~enriched $\delta^{13}\text{C}_{\text{DIC}}$ in the Indian monsoonal estuaries.~~ This is because the microbial decomposition
431 ~~of organic matter results in depleted $\delta^{13}\text{C}_{\text{DIC}}$ due to preferential release of ^{12}C over ^{13}C in to DIC~~
432 ~~pool while removal of DIC by autotrophic production enriches the residual DIC due to~~
433 ~~preferential uptake of ^{12}C over ^{13}C during photosynthesis reaction.~~ CO₂ out gassing due to
434 ~~heterotrophic decomposition of organic matter and equilibrium with atmospheric CO₂ results in~~

435 ~~the enrichment of $\delta^{13}\text{C}_{\text{DIC}}$ in reservoirs/dams and stored water bodies (Shin et al., 2001; Brunet et~~
436 ~~al., 2005; Bouvillion et al., 2009; Zeng et al., 2011; Tamooh et al., 2013). As many of the east~~
437 ~~flowing river (e.g. Godavari, Krishna and Cauvery etc) were dammed at many locations for~~
438 ~~domestic, industrial and irrigation purposes., —Hence, relatively enriched $\delta^{13}\text{C}_{\text{DIC}}$ in these east-~~
439 ~~flowing rivers indicate that either DIC is originated from weathering of carbonate rocks by~~
440 ~~carbonic acid produced from dissolution of atmospheric CO_2 and/or storage of water in~~
441 ~~dams/reservoirs as many of them estuaries might have been influenced by storage of water~~
442 ~~besides sources of DIC (e.g. Godavari, Krishna and Cauvery etc) were dammed at many~~
443 ~~locations for domestic, industrial and irrigation purposes. —A significant positive correlation~~
444 ~~between DIC concentrations and $\delta^{13}\text{C}_{\text{DIC}}$ ($r^2=0.77$; $p<0.001$; Fig. 76c), excluding the positive~~
445 ~~$\delta^{13}\text{C}_{\text{DIC}}$ values, indicate that significant contribution of DIC is from oxidation of particulate~~
446 ~~organic carbon in dams/reservoirs or stored water bodies. Therefore, DIC in the Indian estuaries~~
447 ~~are contributed by weathering of silicate and carbonate rocks due to soil CO_2 , biological~~
448 ~~production, organic matter decomposition and exchange of CO_2 to the atmosphere.~~

449 *4.1.3. The impact of catchment lithology*

450 ~~As found in many estuaries over the world, submarine ground water exchange strongly~~
451 ~~influences DIC concentrations in Indian estuaries, for example, Rengarajan and Sarma (2015)~~
452 ~~found 3 to 4 times higher DIC concentrations in the ground water compared the estuarine waters~~
453 ~~of the Godavari and estimated that submarine ground water discharge contributes up to 52% of~~
454 ~~DIC concentrations in the Godavari estuarine system. —The measured DIC concentrations in~~
455 ~~ground waters along the entire Indian coast (Dr. BSK Kumar, personal communication) showed~~
456 ~~strong spatial variability with relatively lower concentrations in the SW (mean $32\pm 19 \text{ mg l}^{-1}$)~~
457 ~~than the SE (106 ± 56), NE (92 ± 31) and NW ($84\pm 54 \text{ mg l}^{-1}$) regions of India during discharge~~

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458 ~~period. Though the DIC concentrations in ground waters were higher by about 3 to 5 times than~~
459 ~~the concentrations found in the Indian estuaries, however, exchange of ground water with~~
460 ~~relatively low DIC concentrations in the SW region could have, at least partly, caused the lower~~
461 ~~DIC concentrations in the SW estuaries.~~

462 Spatial distribution of bedrock and soils over the Indian subcontinent shows that
463 Narmada and Tapti rivers in the NW India and upper reaches of Godavari and Krishna rivers
464 drain over the igneous rocks (Deccan traps) while the other rivers flow ~~through~~ over the
465 metamorphic rocks (Pre-Cambrian), which are the predominant rock type in south India.
466 However, Haldia and lower reaches of the SE rivers drain over the sedimentary rocks
467 (Geological Survey of India, <https://www.gsi.gov.in>). ~~Although, the higher~~ chemical
468 weathering rates were reported ~~to be higher for in the~~ Deccan Trap basalts (Das et al., 2005;
469 Singh et al., 2005), ~~however,~~ higher DIC concentrations were also ~~found~~ observed in estuaries
470 draining over the metamorphic rocks, suggesting that ~~strong influence of other~~ factors may also
471 be governing the concentrations of DIC, other than the bedrocks in the catchment. The broad
472 range of $\delta^{13}\text{C}_{\text{DIC}}$ found in this study (-13.0 to 2.5‰) not only also indicates that DIC contribution
473 is from variable sources such as DIC contribution from weathering of carbonate and silicate
474 rocks by carbonic acid derived from dissolution of soil CO_2 (-10 to -9‰ and -21 to -17‰
475 respectively, Solomon and Cerling, 1987), decomposition of terrestrial organic matter (-267 to -
476 276‰, O'Leary, 1988) and marine water (0 to 2‰,) (Fig. 5), but also suggests processes on
477 $\delta^{13}\text{C}_{\text{DIC}}$ in the Indian monsoonal estuaries.

478 ~~—~~ Spatial distribution of soils shows that lateritic soils, which are poor in lime and silicate,
479 occupied the catchment of the SW rivers. Chemical weathering rates are relatively lower in the
480 lateritic than the non-lateritic soils and the consumption of atmospheric/soil CO_2 through silicate

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481 weathering is lower by ~2 times in the former than the latter (Boeglin and Probst, 1998).

482 ~~Though the~~ upper reaches of the east flowing rivers (NE and SE) drain over the lime-poor red
483 and yellow soils, ~~however, they are dominated by~~ while the lower reaches drain predominantly
484 the lime-rich alluvial soils ~~in their lower reaches~~. Upper reaches of Krishna and Godavari also
485 drain over the lime-rich black soils. The dominance of lateritic soils, which are relatively less
486 susceptible to chemical weathering than the non-lateritic soils, ~~over in~~ the catchments of the SW
487 rivers could ~~have, at least in part, could be possible reason for~~ lowered the DIC concentrations in
488 SW estuaries. The enriched $\delta^{13}\text{C}_{\text{DIC}}$ in the SW estuaries ($-7.4 \pm 1.9\%$) may also be due to less
489 contribution of DIC from lateritic soils as these soils are poor in lime (-10 to -9%) and silicate ($-$
490 21 to -17%) and less susceptible to chemical weathering rates, ~~during the study period.~~
491 Altogether DIC concentrations in the Indian monsoonal estuaries appears to be controlled by the
492 precipitation, area and lithology of the catchment, SGD, and autotrophic production and
493 heterotrophic decomposition of organic matter within the rivers.

494 ~~The SW region of India receives highest amount of rainfall during the SW monsoon~~
495 ~~($2500 \pm 500\text{mm}$) than the SE (400 ± 50), NE (1000 ± 200) and NW ($750 \pm 250\text{mm}$) regions of India~~
496 ~~(Soman and Kumar, 1990). Though the intense rainfall in the SW region is expected to cause~~
497 ~~higher weathering rates and therefore higher DIC (e.g., Gupta et al., 2011), the observed lower~~
498 ~~DIC concentrations in the SW estuaries could be due to the dilution. The catchment area~~
499 ~~normalized volume of discharge was found to be higher in the SW estuaries ($1.71 \text{ m}^3 \cdot \text{m}^{-2}$) than in~~
500 ~~the SE (0.17), NE (0.6) and NW ($0.32 \text{ m}^3 \cdot \text{m}^{-2}$) estuaries, suggesting that significant dilution of~~
501 ~~DIC concentrations in the SW estuaries. A strong negative correlation between precipitation in~~
502 ~~the catchment and DIC concentration in estuaries ($r^2 = 0.89$, $p < 0.001$; Fig. 5) confirms that DIC~~

503 ~~concentration in Indian estuaries are rather controlled by the intensity of precipitation over the~~
504 ~~catchment.~~

505 **4.2 $\delta^{13}\text{C}$ of DIC in the Indian monsoonal estuaries**

506 ~~The stable isotopic composition of DIC ($\delta^{13}\text{C}_{\text{DIC}}$) is a well-established and widely used~~
507 ~~tracer to identify the major sources of DIC in rivers (e.g. Singh et al., 2005; Tamooh et al., 2013;~~
508 ~~Samanta et al., 2015; Zou, 2016) because each of the DIC sources have a distinct $\delta^{13}\text{C}_{\text{DIC}}$ ratios~~
509 ~~(Deines et al., 1974). DIC originated by dissolution of atmospheric CO_2 is about -7 to -8‰~~
510 ~~(Coplen et al., 2002) whereas it is about -26 to -27‰ if DIC is derived from oxidation of organic~~
511 ~~matter produced by C_3 plants (O'Leary, 1988). The $\delta^{13}\text{C}$ of DIC generated by soil CO_2 -dissolved~~
512 ~~carbonic acid weathering of silicates is about -17 to -21‰ (Solomon and Cerling, 1987) while it~~
513 ~~is close to -9‰ for carbonate rocks because half of the carbon comes from carbonate rocks (0‰,~~
514 ~~Land, 1980) during weathering. Whereas, the weathering of carbonate and silicate minerals~~
515 ~~yield $\delta^{13}\text{C}_{\text{DIC}}$ values -7 to -8‰ and -3 to -4‰, respectively, if the carbonic acid formed by the~~
516 ~~dissolution of atmospheric CO_2 . Although, DIC derived from different sources have distinctly~~
517 ~~different $\delta^{13}\text{C}_{\text{DIC}}$ values, however, the interpretation the $\delta^{13}\text{C}_{\text{DIC}}$ values for identification of its~~
518 ~~sources is still challenging (Amiotte-Suchet et al., 1999; Campeau et al., 2017) due to the~~
519 ~~isotopic fractionations associated with complex mixture of sources and processes such as~~
520 ~~photosynthesis (O'Leary, 1988; Finlay, 2004; Parker et al., 2005, 2010), respiration (Finlay,~~
521 ~~2003; Waldron et al., 2007), DOC photo-oxidation (Opsahl and Zepp, 2001; Vahatalo and~~
522 ~~Wetzel, 2008), anaerobic metabolism (Waldron et al., 1999; Maher et al., 2015) and equilibration~~
523 ~~with atmospheric CO_2 .~~

524 ~~The range of $\delta^{13}\text{C}_{\text{DIC}}$ found in this study (-13.0 to 2.5‰) was similar to those reported~~
525 ~~earlier in various rivers, for example, Brahmaputra (Singh et al., 2005), Rhine (Buhl et al., 1991),~~

526 ~~Ottawa (Telmer et al., 1999), St. Lawrence (Yang et al., 1996), Nanpan and Beipan rivers,~~
527 ~~southwest China (Zou, 2016) and Tana river, Kenya (Tamooh et al., 2013). The range of $\delta^{13}\text{C}_{\text{DIC}}$~~
528 ~~in this study indicates a variety of sources, including silicate and carbonate weathering and~~
529 ~~marine waters, contributes DIC to the Indian monsoonal estuaries during the study period.~~
530 ~~Relatively depleted $\delta^{13}\text{C}_{\text{DIC}}$ in the west flowing river estuaries of NW (mean $-11.1 \pm 2.3\%$) and~~
531 ~~SW (mean: $-7.4 \pm 1.9\%$) regions suggest that DIC is contributed from silicate and carbonate~~
532 ~~weathering by the carbonic acid, produced from the dissolution of both soil CO_2 and atmospheric~~
533 ~~CO_2 . Zou (2016) found the $\delta^{13}\text{C}_{\text{DIC}}$ values in the range of -13.9 to -8.1% in the Nanpan and~~
534 ~~Beipan rivers of SW China and were attributed to dominant contribution of DIC from weathering~~
535 ~~of carbonate minerals. Relatively enriched $\delta^{13}\text{C}_{\text{DIC}}$ in the east flowing river estuaries of NE (-6.5~~
536 ~~to -0.7 ; mean: $-3.5 \pm 2.8\%$) and SE (-7.9 to -2.5% ; $-2.7 \pm 5.2\%$) indicates that major contribution of~~
537 ~~DIC is from chemical weathering of carbonate rocks by atmospheric CO_2 dissolved carbonic~~
538 ~~acid or acid from non-carbon sources (Li et al., 2008). Weathering of carbonate minerals by acid~~
539 ~~sources other than carbonic acid causes enrichment compared to weathering by carbonic acid due~~
540 ~~to lack of contribution from $\delta^{13}\text{C}$ depleted carbonic acid of soil CO_2 (-17 to -21%) or~~
541 ~~atmospheric CO_2 (-7 to -8%) origin to the $\delta^{13}\text{C}$ enriched carbonate rocks (0% , Land 1980).~~

542 ~~In addition to the sources, hydrological and biological processes also influence the~~
543 ~~$\delta^{13}\text{C}_{\text{DIC}}$ in streams/rivers. For example, heavy precipitation in the SW region ($2500 \pm 500\text{mm}$)~~
544 ~~than the other regions tends to cause depletion in $\delta^{13}\text{C}_{\text{DIC}}$ values due to shorter residence time of~~
545 ~~soil water (Amiotte Suchet et al., 1999) while CO_2 out gassing causes enrichment due to~~
546 ~~accumulation of ^{13}C during diffusive efflux (Clark and Fritz, 1997) in stored water bodies. Many~~
547 ~~of the east flowing rivers are major and are dammed at many locations (e.g. Godavari, Krishna~~
548 ~~and Cauvery) for domestic, industrial and irrigation purposes. CO_2 out gassing due to~~

549 ~~heterotrophic decomposition of organic matter and autotrophic production significantly alters the~~
550 ~~$\delta^{13}\text{C}_{\text{DIC}}$ signatures in reservoirs (Shin et al., 2001). Further, equilibrium with atmospheric CO_2 in~~
551 ~~the reservoirs due to no/lean flow leads to enrichment in the $\delta^{13}\text{C}_{\text{DIC}}$ values (Brunet et al., 2005;~~
552 ~~Bouvillion et al., 2009; Zeng et al., 2011; Tamooh et al., 2013). Hence, relatively enriched~~
553 ~~$\delta^{13}\text{C}_{\text{DIC}}$ in the NE and SE estuaries could also be due to the storage of water in reservoirs/dams.~~
554 ~~A significant positive correlation between DIC concentrations and $\delta^{13}\text{C}_{\text{DIC}}$ ($r^2=0.77$; $p<0.001$;~~
555 ~~Fig. 7), excluding the positive values, indicate that significant contribution of DIC from~~
556 ~~oxidation of particulate organic carbon in dams/reservoirs or stored water bodies. Shin et al.~~
557 ~~(2011) attributed the stream $\delta^{13}\text{C}_{\text{DIC}}$ values of $-6.9\pm 1.6\%$ and $-7.8\pm 1.5\%$ in silicate and~~
558 ~~carbonate dominated catchments, respectively, in tributaries of the Han River, South Korea to~~
559 ~~CO_2 -out gassing. Positive $\delta^{13}\text{C}_{\text{DIC}}$ values ($>0\%$) were observed only in Rushikulya (0.1%),~~
560 ~~Nagavali (0.7%) and Vaigai (2.5%) in which relatively higher salinities (>20) were found~~
561 ~~during the study period. This is concurrent with earlier observations in the Indian estuaries,~~
562 ~~Hooghly (Samanta et al., 2015) and Cochin (Bhavya et al., 2016) where relatively enriched~~
563 ~~$\delta^{13}\text{C}_{\text{DIC}}$ were found at higher salinities. A strong positive correlation was found between $\delta^{13}\text{C}_{\text{DIC}}$~~
564 ~~and salinity (Fig. 6; $r^2=0.71$, $p<0.001$), suggesting that $\delta^{13}\text{C}_{\text{DIC}}$ values in the Indian estuaries are~~
565 ~~influenced by the intrusion of marine waters ($\delta^{13}\text{C}_{\text{DIC}}=-1$ to 2%).~~

566 **4.3.2 Total DIC export by the Indian monsoonal rivers to the north Indian Ocean**

567 Indian monsoonal rivers annually export ~ 10.4 Tg of DIC to the north Indian Ocean.
568 Nearly three fourth of this amount (7.7 Tg) reaches to the Bay of Bengal while the remaining
569 into the Arabian Sea. ~~It is mainly attributed to the~~ This is consistent with the higher magnitude of
570 annual freshwater discharge ~~because to~~ the Bay of Bengal ~~annually receives~~ (378 km^3) ~~of~~
571 freshwater from the catchment area of about 0.96 M km^{-2} , ~~whereasthan~~ the Arabian Sea

572 ~~receives only~~ (122 km³ ~~of freshwater~~ from the catchment area of ~~only~~ 0.23 M km²). ~~Although~~
573 ~~the increase in volume of discharge dilutes the DIC flux from rivers (Jarvie et al., 1997; Shanley~~
574 ~~et al., 2002), bicarbonate fluxes to the Gulf of Mexico were reported to increase with the volume~~
575 ~~of discharge from the Mississippi river (Raymond and Oh, 2007) due to small dilution factor.~~

576 ~~F~~The total DIC export by the Indian monsoonal estuaries (10.4 Tg yr⁻¹) is only 2.5% of
577 the total DIC export by the world major rivers (400 Tg yr⁻¹), and 9.4% of the export by the Asian
578 rivers (111 Tg yr⁻¹; Huang et al., 2012). The DIC export from the Indian estuaries is far less than
579 the DIC export by the American (61.4 Tg yr⁻¹) and African (17.7 Tg yr⁻¹) rivers and major rivers
580 draining to the tropical Atlantic from South America and Africa (53 Tg yr⁻¹, Araujo et al. 2014).
581 It is mainly due to the fact that ~~the volume of~~freshwater discharge from the Indian monsoonal
582 rivers is very low (~500 km³ ~~yr⁻¹~~) compared to the American (11,799 km³ ~~yr⁻¹~~) and African
583 (3,786 km³ ~~yr⁻¹~~) rivers. However, the Indian monsoonal rivers are exporting ~~DIC~~
584 disproportionately higher DIC to the north Indian Ocean because they account for only 1.3% of
585 the global river discharge but export 2.5% of the global riverine DIC to the oceans.

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

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

593 Krishna et al. (2015) reported that Indian monsoonal estuaries export 2.32 Tg yr⁻¹ of
594 dissolved organic carbon (DOC) to the north Indian Ocean. ~~When combined~~ The total fluvial

595 dissolved carbon flux (DIC+DOC) would be 12.74 Tg yr⁻¹ ~~in which. This indicate that the total~~
596 ~~fluvial dissolved carbon export to the north Indian Ocean by the Indian monsoonal estuaries is~~
597 ~~predominantly contributed by~~ DIC ~~flux contributed up to (~81% and it is) than DOC~~; consistent
598 with earlier reports elsewhere in the world, for example, the British rivers (80%, Jarvie et al.,
599 2017). Since the catchment area of the Indian monsoonal rivers ranged widely from as low as
600 0.001 M km² to as high as 0.313 M km², the export fluxes of DIC were normalized with the
601 catchment area of the river (yield) to obtain DIC yield from each river in order to examine
602 various factors controlling the lateral DIC export to the north Indian Ocean.

603 **4.4.3 Yield of DIC from the Indian monsoonal rivers**

604 The yield ~~of DIC (export normalized by catchment area) of DIC~~ found in this study (mean
605 8.7±5.2 g m⁻² yr⁻¹) is similar those found earlier in the rivers from ~~the~~ tropical region (~~30°N–~~
606 ~~30°S~~) of the Asian continent (~~9.79 g m⁻² yr⁻¹; Huang et al., 2012~~), but significantly higher than
607 ~~those reported from tropical region of~~ the American (~~3.33 g m⁻² yr⁻¹~~) and African (~~0.63 g m⁻² yr⁻¹~~)
608 continents ~~of this tropical region (Table 4)~~ (Huang et al., 2012). ~~The yield of DIC from river~~
609 ~~catchment were reported to be controlled by the hydrological (precipitation, runoff and~~
610 ~~groundwater exchange) and environmental (temperature, type and dominance of soils, soil~~
611 ~~organic carbon, natural vegetation and forest cover) conditions and anthropogenic activities (land~~
612 ~~use change and deforestation) in the catchment (Raymond et al., 2008; Huang et al., 2012).~~
613 ~~Although~~ The SW estuaries annually export relatively ~~less-lower~~ DIC to the north Indian Ocean
614 (0.30 Tg) due to their ~~lower~~ volume of discharge (46 km³ yr⁻¹) ~~from-and~~ relatively smaller
615 catchment area (0.02 M km²) than the SE (~~3.50 Tg, 102 km³ yr⁻¹ and 0.43 M km² respectively~~),
616 NE (~~4.21 Tg, 276 and 0.53~~) and NW (~~2.38 Tg, 75 km³ yr⁻¹ and 0.21 M km²~~) estuaries (Table 2 &
617 3), strikingly in contrast, the higher yield of DIC was found in the former (10.8±6.6 g m⁻² yr⁻¹)

618 than the latter (5.8 ± 2.3 , 8.6 ± 5.7 and 9.5 ± 3.9 $\text{g m}^{-2} \text{yr}^{-1}$, respectively Table 4). This suggests that
619 strong control of catchment and/or in stream processes on yield of DIC from the monsoonal
620 rivers. However, DIC yield showed a significant positive correlation with the volume of
621 discharge ($r^2=0.6667$, $p<0.001$; Fig. 6d) in medium estuaries, and no such relationship was found
622 in the major estuaries. Significant negative relationships were observed between DIC yield and
623 catchment area in the medium ($r^2= -0.5249$, $p<0.001$; Fig. 6e) and major estuaries ($r^2= -0.439$,
624 $p<0.001$; Fig. 6f). This suggests that high precipitation over small catchments increases the DIC
625 yield from the Indian estuaries because the dense precipitation increases the seouring extraction
626 of DIC from soils and rocks in their catchment. Therefore, high precipitation ($2500 \pm 500 \text{mm}$)
627 over the small catchment (0.02 M km^2) could have increased DIC yield from the SW estuaries.
628 A strong linear relationship between the yield of DIC and the intensity of precipitation ($r^2=0.64$,
629 $p<0.001$ Fig. 8a6g) confirms that dense precipitation increases the export yield of DIC from SW
630 estuaries. This could be one the reasons for the observed higher yield of DIC in the SW estuaries
631 which receives high precipitation ($2500 \pm 500 \text{mm}$) over the small catchment area (0.02 M km^2).
632 Ground water exchange do not appears to be controlling DIC yield from the Indian
633 monsoonal estuaries because the groundwater DIC concentrations were lower in the SW (32 ± 19
634 mg l^{-1}) than the other regions SE (106 ± 56), NE (92 ± 31) and NW ($84 \pm 54 \text{mg l}^{-1}$). Existing natural
635 vegetation of tropical moist deciduous and tropical wet evergreen and semi evergreen forests in
636 the SW region also could also have increased DIC yield from the SW estuaries compared to the
637 other estuaries as this vegetation favors the export fluxes of DIC. The drainage basins of the
638 Indian monsoonal estuaries rivers are largely under the tropical dry and wet climate except the
639 SW rivers, Narmada and Tapti. The rivers Narmada and Tapti are under the arid and semiarid

640 climate while the SW rivers are under the tropical wet climate which was also reported to
641 facilitate the riverine export of material from drainage basin to the coastal ocean.

642 ~~Catchments of the SW rivers are largely occupied by the cation deficient lateritic soils~~
643 ~~and therefore precipitation of carbonate minerals in soils is poor. As a result, the soil inorganic~~
644 ~~carbon content in surface (100cm) soils of the catchment of SW rivers was lower than in~~
645 ~~catchments of the other monsoonal rivers studied (Sreenivas et al., 2016). On the other hand, the~~
646 ~~authors (Sreenivas et al., (2016) and Krishwan et al. (2009) found that the soil organic and~~
647 ~~inorganic carbon contents in the surface (100cm) soils of the catchment of SW rivers were~~
648 ~~higher and lower, respectively, in the former than the latter catchments of the SE, SW and NE~~
649 ~~rivers (Table 2). The relationship between soil inorganic and organic carbon is primarily~~
650 ~~dependent on the soil characteristics in the catchment. For example, Guo et al. (2016)~~
651 ~~demonstrated that increase in the soil organic carbon content enhanced the soil inorganic carbon~~
652 ~~in the cropland of upper Yellow river delta, China. A strong positive relationship between soil~~
653 ~~organic and inorganic carbon was also found in the Yanqi river basin, northwest China (Wang et~~
654 ~~al., 2015), and soils in the America (Stevenson et al., 2005) and Canada (Landi et al., 2003).~~
655 ~~This indicates that more dissolution of soil carbonates by acidic conditions formed by release of~~
656 ~~CO₂ through decomposition of soil organic carbon in catchments of the SW rivers. Hence, On~~
657 ~~the other hand, a negative relationship was found between soil organic and inorganic carbon in~~
658 ~~the North China Plain (Huang et al., 2006) and west Loess Plateau (Zeng et al., 2008). The~~
659 ~~negative relationship is mainly due to the higher production of CO₂ by decomposition of soil~~
660 ~~organic carbon and root respiration resulting in the formation of acidic conditions that lead to~~
661 ~~dissolution of soil carbonates. The higher soil organic carbon in the catchment of the SW than in~~
662 ~~catchment of the SE, NE and NW rivers (Kishwan et al., 2009; Sreenivas et al., 2016) could have~~

663 | ~~elevated the yield of DIC from SW estuaries through dissolution of soil carbonates. therefore,~~
664 | ~~produces more CO₂ through microbial decomposition and causes dissolution of soil carbonates~~
665 | ~~leading to the higher yield of DIC from the SW estuaries.~~ A significant linear correlation
666 | between soil organic carbon content and DIC yield in this study ($r^2=0.65$, $p<0.001$; Fig. ~~8b6h~~)
667 | ~~suggests confirms~~ that strong influence of soil organic carbon content in the catchment on DIC
668 | yield from the Indian monsoonal rivers. ~~However, The~~ basin scale studies are required for
669 | comprehensive understanding of the influence of environmental and anthropogenic factors on
670 | DIC export fluxes from the Indian monsoonal rivers.

671 | 5. Summary

672 | In order to examine the spatial variability ~~of in the sources and distribution of~~ dissolved
673 | inorganic carbon (DIC) ~~concentrations and to identify its major sources~~ in the Indian monsoonal
674 | estuaries, and to estimate the riverine export fluxes of DIC to the north Indian Ocean, we
675 | sampled a total of 27 major and medium estuaries along the Indian coast during wet period. An
676 | order of magnitude variability was found in DIC concentrations among the estuaries sampled
677 | (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
678 | SW region of India. It is attributed to significant spatial variability in the size of rivers,
679 | precipitation pattern and lithology in their catchments. Magnitude of discharge, catchment area
680 | and in-stream processes ~~are~~ appears to be ~~important factors for the controlling factors for~~
681 | ~~concentration and yield of DIC in the~~ medium estuaries rather than ~~the~~ major estuaries. ~~in~~
682 | ~~controlling the concentration and yield of DIC, This is~~ probably due to a significant spatial
683 | variability in lithology and hydro-geological and environmental conditions in the catchments.
684 | Indian monsoonal estuaries annually export ~10.4 Tg of DIC to the north Indian Ocean, of which
685 | 7.7 Tg enters in to the Bay of Bengal while the Arabian Sea receives only 2.7 Tg. It is mainly

686 | attributed to the volume of river discharge as the former receives $\sim 378 \text{ km}^3 \text{ yr}^{-1}$ while the latter
687 | receives only $122 \text{ km}^3 \text{ yr}^{-1}$ ~~of freshwater~~ from the Indian monsoonal rivers. The range of $\delta^{13}\text{C}_{\text{DIC}}$
688 | found in this study suggests that major contribution of DIC is ~~largely contributed~~ from
689 | weathering of silicate and carbonate minerals by carbonic acid formed by dissolution of both soil
690 | and atmospheric CO_2 . However, relatively enriched $\delta^{13}\text{C}_{\text{DIC}}$ in the east-flowing river estuaries
691 | indicated the storage of water in dams/reservoirs and intrusion of marine waters. Dense rainfall
692 | ~~(2500±500mm)~~ and higher soil organic carbon content ~~(101.4 g ha⁻¹)~~ in the catchment of SW
693 | rivers than in the catchment of the other rivers resulted in higher yield of DIC from the former
694 | than the latter.

695 **6. Acknowledgements**

696 | We would like to thank the Director, CSIR - National Institute of Oceanography (NIO), Goa, and
697 | the Scientist-In-Charge, NIO-Regional Centre, Visakhapatnam for their kind support and
698 | encouragement. We also acknowledge Dr. M. Dileep Kumar, NIO, Goa for his guidance and
699 | encouragement. The work is part of the Council of Scientific and Industrial Research (CSIR),
700 | funded research project. This publication has NIO contribution number

701 **7. Data Availability**

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

704

705 **References**

706

707 Abril, G., Etcheber, H., Delille, B., Frankignoulle, M. and Borges A. V.: Carbonate dissolution
708 in the turbid and eutrophic Loire estuary, *Mar. Ecol. Progr. Ser.*, 259, 129-138, 2003.
709
710 Amiotte-Suchet, P. et al.: $\delta^{13}\text{C}$ pattern of dissolved inorganic carbon in a small granitic
711 catchment: the Strengbach case study (Vosges mountains, France), *Chem. Geol.*, 159, 129–145,
712 doi:10.1016/s0009-2541(99)00037-6, 1999.
713
714 Araujo, M., Noriega, C., and Lefevre, N.: Nutrients and carbon fluxes in the estuaries of major
715 rivers flowing into the tropical Atlantic, *Front. Mar. Sci.*, 1, 1-16, 2014.
716
717 Barber, R. T., Marra, J., Bidigare, R. C., Codispoti, L. A., Halpern, D., Johnson, Z., Latasa, M.,
718 Goericke, R., and Smith, S. L.: Primary productivity and its regulation in the Arabian Sea during
719 1995, *Deep Sea Res., Part II*, 48, 1127–1172, 2001.
720
721 Battin, T. J. et al.: Biophysical controls on organic carbon fluxes in fluvial networks, *Nature*
722 *Geosci.*, 1, 95–100, doi:10.1038/ngeo101, 2008.
723
724 Bauer, J. E., Cai, W. J., Raymond, P. A., Bianchi, T. S., Hopkinson, C. S., and Regnier, P.A.G.:
725 The changing carbon cycle of the coastal ocean, *Nature*, 504, 61–70, doi:10.1038/Nature12857,
726 2013.
727
728 Berner, R. A., Lasaga, A. C., Garrels, R. M.: The carbonate–silicate geochemical cycle and its
729 effect on atmospheric carbon dioxide over the past 100 million years, *Am. J. Sci.*, 283, 641-683,
730 1983
731
732 Bhavya, P.S., Kumar, S., Gupta, G.V.M., Sudharma, K.V., Sudheesh, V., and Dhanya, K.R.:
733 Carbon isotopic composition of suspended particulate matter and dissolved inorganic carbon in
734 the Cochin estuary during post-monsoon, *Curr.Sci.*, 100, 1539-1543, 2016.
735
736 Boeglin, J. L., and Probst, J. L.: Physical and chemical weathering rates and CO₂ consumption in
737 a tropical lateritic environment: the upper Niger basin, *Chem. Geol.*, 148; 137-156, 1998.

738 Bouillon, S., Abril, G., Borges, A. V., Dehairs, F., Govers, G., Hughes, H. J., Merckx, R.,
739 Meysman, F. J. R., Nyunja, J., Osburn, C., and Middelburg, J. J.: Distribution, origin and cycling
740 of carbon in the Tana River (Kenya): a dry season basin-scale survey from headwaters to the
741 delta, *Biogeosciences*, 6, 2475–2493, doi:10.5194/bg-6-2475-2009, 2009.
742
743 Brunet, F. *et al.* $\delta^{13}\text{C}$ tracing of dissolved inorganic carbon sources in Patagonian rivers
744 (Argentina). *Hydrol. Process.*, 19, 3321–3344, doi:10.1002/hyp.5973, 2005.
745
746 Buhl, D., Neuser, R. D., Richter, D. K., Riedel, D., Roberts, B., Strauss, H. and Veizer, J.: Nature
747 and nurture; Environmental isotope story of the river Rhine. *Naturwissenschaften*, 78, 337–346,
748 1991.
749

750 Cai, W. J., Guo, X. H., Chen, C. T. A., Dai, M. H., Zhang, L. J., Zhai, W. D., Lohrenz, S. E.,
751 Yin, K. D., Harrison, P. J., and Wang, Y. C.: A comparative overview of weathering intensity and
752 HCO₃ flux in the world's major rivers with emphasis on the Changjiang, Huanghe, Zhujiang
753 (Pearl) and Mississippi Rivers, *Cont. Shelf Res.*, 28, 1538-1549, 2008.

754 Cai, W. J.: Riverine inorganic carbon flux and rate of biological uptake in the Mississippi River
755 plume, *Geophys. Res. Lett.*, 30, 1032, 2003.

756
757 Cai, W. -J.: Estuarine and coastal ocean carbon paradox: CO₂ sinks or sites of terrestrial carbon
758 incineration? *Annu Rev Mar Sci*, 3, 123-145, 2011.

759
760 Campeau, A., Wallin, M. B., Giesler, R., Löfgren, S., Mörrh, C. -M., Schiff, S., Venkiteswaran,
761 J. J. and Bishop, K.: Multiple sources and sinks of dissolved inorganic carbon across Swedish
762 streams, refocusing the lens of stable C isotopes, *Nature, Scientific Reports*, 7, 9158,
763 DOI:10.1038/s41598-017-09049-9, 2017.

764
765 Carritt, D. E. and Carpenter, J. H.: Comparison and evaluation of currently employed
766 modifications of the Winkler method for determining dissolved oxygen in seawater: A NASCO
767 report, *J. Mar. Res.*, 24, 286-318, 1966.

768
769 Central Water Commission, *Integrated Hydrological Data Book*, 680 pp., New Delhi, 2012.

770
771 Central Water Commission: *Integrated Hydrological Data Book*, 383 pp., New Delhi, 2006.

772
773 Christopher, P. B., Luettich Jr. R. A., Powers, S. P., Peterson, C. H., and McNinch, J. E.:
774 Estimating the spatial extent of bottom-water hypoxia and habitat degradation in a shallow
775 estuary, *Mar. Ecol. Prog. Ser.*, 230, 103-112, 2002.

776
777 Clark, I. D., and Fritz, P.: *Environmental isotopes in hydrogeology*, CRC Press, 1997.

778
779 Cole, J. J., Prairie, Y. T., Caraco, N. F., McDowell, W. H., Tranvik, L. J., Striegl, R. G., Duarte,
780 C. M., Kortelainen, P., Downing, J. A., Middelburg, J. J., and Melack, J.: Plumbing the global
781 carbon cycle: Integrating inland waters into the terrestrial carbon budget, *Ecosystems*, 10, 171-
782 184, 2007.

783
784 Coplen, T. B. et al.: *Compilation of minimum and maximum isotope ratios of selected elements
785 in naturally occurring terrestrial materials and reagents*, U.S. Department of the Interior and U.S.
786 Geological Survey, 2002.

787
788 Das, A., Krishnaswami, S. and Bhattacharya, S. K.: Carbon isotope ratio of dissolved inorganic
789 carbon (DIC) in rivers draining the Deccan Traps, India: Sources of DIC and their magnitudes.
790 *Earth Planet. Sci. Lett.*, 236, 419-429, doi:10.1016/j.epsl.2005.05.009, 2005.

791
792 Deines, P., Langmuir, D., and Harmon, R. S.: Stable carbon isotope ratios and the existence of a
793 gas phase in the evolution of carbonate ground waters, *Geochim. Cosmochim. Acta*, 38, 1147-
794 1164, doi:10.1016/0016-7037(74)90010-6, 1974.

793 Dessert, C., Dupre, B., Francois, L. M., Schott, J., Gaillardet, J., Chakrapani, G., and Bajpai, S.:
794 Erosion of Deccan Traps determined by river geochemistry: impact on the global climate and the
795 $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of seawater. *Earth and Planet. Sci. Lett.*, 188, 459–474, 2001.
796
797 Dhillon, G. S., and Inamdar, S.: Extreme storms and changes in particulate and dissolved organic
798 carbon in runoff: Entering uncharted waters?, *Geophys. Res. Lett.*, 40, 1322–1327,
799 doi:10.1002/grl.50306., 2013.
800
801 DOE: Hand book of methods for the analysis of the various parameters of the carbon dioxide
802 system in seawater, version 2, edited by Dickson, A.G. and Goyet, C., Rep. ORNL/CDIAC-74,
803 Oak Ridge Natl. Lab., Oak Ridge, Tenn, doi:10.1029/2002GL016312, 1998.
804
805 Findlay, S.: Stream microbial ecology, *J. North Am. Benthol. Soc.*, 29, 170–181,
806 doi:10.1899/09-023.1, 2010.
807
808 Finlay, J. C. and Kendall, C.: Stable isotope tracing of temporal and spatial variability in organic
809 matter sources to freshwater ecosystems, In: *Stable Isotopes in Ecology and Environmental*
810 *Science*, 2nd edn., edited by: Michener, R. H. and Lajtha, K., Blackwell Publishing, Malden,
811 USA, 283–333, 2007.
812
813 Finlay, J. C.: Controls of streamwater dissolved inorganic carbon dynamics in a forested
814 watershed, *Biogeochem.*, 62, 231–252, 2003.
815
816 Finlay, J. C.: Patterns and controls of lotic algal stable carbon isotope ratios, *Limnol. Oceanogr.*,
817 49, 850–861, 2004.
818
819 Gaillardet, J., Dupre, B., Louvat, P., and Allegre, C. J.: Global silicate weathering and CO_2
820 consumption rates deduced from the chemistry of large rivers. *Chem. Geol.* 159, 3–30, 1999.
821
822 Gauns, M., Madhupratap, M., Ramaiah, N., Jyothibabu, R., Fernandes, V., Paul, J. T., and
823 Kumar, S. P.: Comparative accounts of biological productivity characteristics and estimates of
824 carbon fluxes in the Arabian Sea and the Bay of Bengal, *Deep Sea Res., Part II*, 52, 2003–2017,
825 2005.
826
827 Guo, Y., Wang, X., Li, X., Wang, J., Xu, M., and Li, D.: Dynamics of soil organic and inorganic
828 carbon in the cropland of upper Yellow River Delta, China, *Nature Scientific Reports*, 6, 36105,
829 DOI: 10.1038/srep36105, 2016.
830
831 Gupta, G. V. M., Thottathil, S. D., Balachandran, K. K., Madhu, N. V., Madeswaran, P., and Nair,
832 S.: CO_2 supersaturation and net heterotrophy in a tropical estuary (Cochin, India): influence of
833 anthropogenic effect, *Ecosystems*, 12, 1145–1157, doi:10.1007/s10021-009-9280-2, 2009.
834
835 Gupta, H., Chakrapani, G. J., Selvaraj, K., and Kao, S.-J.: The fluvial geochemistry,
836 contributions of silicate, carbonate and saline–alkaline components to chemical weathering flux
837 and controlling parameters: Narmada River (Deccan Traps), India, *Geochim. Cosmochim. Acta*,
838 75, 800–824, 2011.

839 Gurumurthy G. P., Balakrishna K., Riotte J., Braun J.-J., Audry S., Shankar H. N. U. and
840 Manjunatha B. R.: Controls on intense silicate weathering in a tropical river, southwestern India,
841 Chem. Geol., 300–301, 61–69, 2012.
842
843 Hotchkiss, E. R. *et al.* Sources of and processes controlling CO₂ emissions change with the size
844 of streams and rivers, Nature Geoscience 8, doi:10.1038/Ngeo2507, 2015.
845
846 Huang, B., Wang, J. G., Jing, H. Y., and Xu, S. W.: Effects of long- term application fertilizer on
847 carbon storage in calcareous meadow soil, J. Agro-Environ. Sci., 25, 161–164, 2006.
848
849 Huang, T-H., Fu, Y-H., Pan, P-Y., and Arthur, C.T.: Fluvial carbon fluxes in tropical rivers,
850 Current Opinion in Environmental Sustainability, 4, 162–169, 2012.
851
852 Ishikawa, N.F., Tayasu, I., Yamane, M., Yokoyama, Y., Sakai, S., and Ohkouchi, N.: Sources of
853 dissolved inorganic carbon in two small streams with different bedrock geology; Insights from
854 carbon isotopes. Radiocarbon, 57, 439–448, 2015.
855
856 Jarvie, H. P., Neal, C., Leach, D. V., Ryland, G. P., House, W. A., Robson, A. J.: Major ion
857 concentrations and the inorganic carbon chemistry of the Humber rivers, Sci. Total Environ.,
858 194, 285–302, 1997.
859
860 Jarvie, H.P., King, S.M., and Neal, C.: Inorganic carbon dominates total dissolved carbon
861 concentrations and fluxes in British rivers: Application of the THINCARB model –
862 Thermodynamic modeling of inorganic carbon in freshwaters, Sci. Tot. Environ., 575, 496-512,
863 2017.
864
865 Kishwan, J., Pandey, R., and Dhadwal, V. K.: India's forest and tree cover: Contribution as a
866 carbon sink, Tech. Pap. 130, ICFRE BL-23, 2009.
867
868 Krishna, M. S., Prasad, V. R., Sarma, V. V. S. S., Reddy, N. P. C., Hemalatha, K. P. J., and
869 Rao Y. V.: Fluxes of dissolved organic carbon and nitrogen to the northern Indian Ocean from
870 the Indian monsoonal rivers, J. Geophys. Res. Biogeosci., 120, 2067–2080, 2015.
871
872 Kumar, R., Singh, R. D., and Sharma, K. D.: Water resources of India, Curr. Sci., 89, 794–811,
873 2005.
874
875 Land, L. S.: The isotopic and trace element geochemistry of dolomite: the state of the art.
876 Concepts and Models of Dolomitization, 63, 485, doi:10.2110/pec.80.28.0087, 1980.
877
878 Landi, A., Mermut, A. R., and Anderson, D. W.: Origin and rate of pedogenic carbonate
879 accumulation in Saskatchewan soils, Canada. Geoderma 117, 143–156, 2003.
880
881 Lerman, A., Wu, L. L., and Mackenzie, F. T.: CO₂ and H₂SO₄ consumption in weathering and
882 material transport to the ocean, and their role in the global carbon balance, Mar. Chem., 106,
883 326-350, 2007.

884
885 Li, S.-L., Calmels, D., Han, G., Gaillardet, J. and Liu, C.-Q.: Sulfuric acid as an agent of
886 carbonate weathering constrained by $\delta^{13}\text{C}_{\text{DIC}}$: examples from Southwest China, Earth Planet. Sci.
887 Lett., 270, 189–199, 2008.
888
889 Ludwig, W., Amiotte-Suchet, P., Munhoven, G., and Probst, J. L.: Atmospheric CO_2 consumption
890 by continental erosion: present-day controls and implications for the last glacial maximum.
891 Global Planet Change, 17, 107-120, 1998.
892
893 Mackenzie, F. T., Lerman, A., and Andersson, A.J.: Past and present of sediment and carbon
894 biogeochemical cycling models. Biogeosciences 1, 11 -32, 2004.
895
896 Madhupratap, M., Prasanna Kumar, S., Bhattathiri, P. M. A., Kumar, M. D., Raghukumar, S.,
897 Nair, K. K. C., and Ramaiah, N.: Mechanism of the biological response to winter cooling in the
898 northeastern Arabian Sea, Nature, 384, 549–552, 1996.
899
900 Maher, D. T., Cowley, K., Santos, I. R., Macklin, P., and Eyre, B. D.: Methane and carbon
901 dioxide dynamics in a subtropical estuary over a diel cycle: Insights from automated *in situ*
902 radioactive and stable isotope measurements, Mar. Chem., **168**, 69–79,
903 doi:10.1016/j.marchem.2014.10.017, 2015.
904
905 Maher, D. T., Santos, I. R., Golsby-Smith, L., Gleeson, J., and Eyre, B. D.: Groundwater-derived
906 dissolved inorganic and organic carbon exports from a mangrove tidal creek: The missing
907 mangrove carbon sink?, Limnol. Oceanogr., 58, 475-488, 10.4319/lo.2013.58.2.0475, 2013.
908
909 Mayorga, E., Aufdenkampe, A. K., Masiello, C. A., Krusche, A. V., Hedges, J. I., Quay, P. D.,
910 Richey, J. E., and Brown, T. A.: Young organic matter as a source of carbon dioxide out gassing
911 from Amazonian Rivers, Nature, 436, 538–541, 2005.
912
913 McConnaughey, T. A., LaBaugh, J. W., Rosenberry, D. O., Striegl, R. G., Reddy, M. M.,
914 Schuster, P. F., and Carter, V.: Carbon budget for a groundwater-fed lake: calcification supports
915 summer photosynthesis, Limnol. Oceanogr., 39, 1319–1332, 1994.
916
917 Meybeck, M., and Ragu, A.: GEMS/water contribution to the Global Register of River Inputs
918 (GLORI), Provisional Final Rep., 245 pp., UNEP/WHO/UNESCO, Geneva, Switzerland, 1995.
919
920 Meybeck, M., and Ragu, A.: River discharges to the oceans. An assessment of suspended solids,
921 major ions, and nutrients, Environ. Inf. and Assess. Rep., 250, 1996.
922
923 Meybeck, M., and Vorosmarty, C. J.: Global transfer of carbon by rivers, Global Change News
924 Lett, 37, 41974, 1999.
925
926 Meybeck, M.: Global chemical weathering of surficial rocks estimated from river dissolved
927 loads, Am. J. Sci., 287, 401–428, 1987.
928
929

930 Meybeck, M.: Riverine Transport of atmospheric carbon-sources, global typology and budget.
931 Water Air Soil Pollut., 70, 443-463, 1993.
932
933 Mukhopadhyay, S. K., Biswas, H., De, T. K., Sen, S., and Jana, T. K.: Seasonal effects on the
934 air-water carbon dioxide exchange in the Hooghly estuary, NE coast of Bay of Bengal, India. J
935 Environ Monit., 4, 549-552, 2002.
936
937 Muraleedharan, P. M., and Prasanna Kumar, S.: Arabian Sea upwelling—A comparison between
938 coastal and open ocean regions, Curr. Sci., 71, 842–846, 1996.
939
940 O’Leary, M. H.: Carbon Isotopes in Photosynthesis, BioScience, 38, 328–336, doi:
941 10.2307/1310735, 1988.
942
943 Opsahl, S. P. and Zepp, R. G.: Photochemically-induced alteration of stable carbon isotope ratios
944 ($\delta^{13}\text{C}$) in terrigenous dissolved organic carbon, Geophys. Res. Lett., 28, 2417–2420,
945 doi:10.1029/2000gl012686, 2001.
946
947 Parker, S. R., Poulson, S. R., Gammons, C. H., and DeGrandpre, M. D.: Biogeochemical
948 controls on diel cycling of stable isotopes of dissolved O_2 and dissolved inorganic carbon in the
949 Big Hole River, Montana, Environ. Sci. Tech., 39, 7134–7140, doi:10.1021/es0505595, 2005.
950
951 Parker, S. R., Poulson, S. R., Smith, M. G., Weyer, C. L., and Bates, K. M.: Temporal variability
952 in the concentration and stable carbon isotope composition of dissolved inorganic and organic
953 carbon in two Montana, USA Rivers, Aquat Geochem., 16, 61–84, doi:10.1007/s10498-009-
954 9068-1, 2010.
955
956 Pattanaik, J. K., Balakrishnan, S., Bhutani, R. and Singh, P.: Estimation of weathering rates and
957 CO_2 drawdown based on solute load: Significance of granulites and gneisses dominated
958 weathering in the Kaveri River basin, Southern India, Geochim. Cosmochim. Acta, 121, 611-
959 636, 2013.
960
961 Pattanaik, S., Sahoo, R. .K, Satapathy, D. R., Panda, C. R., Choudhury, S. B. et al.: Intra-annual
962 Variability of CO_2 Flux in the Mahanadi Estuary- A Tropical Estuarine System, India. Ann Mar
963 Sci., 1, 005-012, 2017.
964
965 Prasanna Kumar, S., Muraleedharan, P. M., Prasad, T. G., Gauns, M., Ramaiah, N., de Souza, S.
966 N., Sardesai, S., and Madhupratap, M.: Why is the Bay of Bengal less productive during summer
967 monsoon compared to the Arabian Sea?, Geophys. Res. Lett., 29, 2235,
968 doi:10.1029/2002GL016013, 2002.
969
970 Quynh, L.P.T., Binh, P.T.X., Thuy, D.T., Nghia, L.D., and Cuong, H.T.: Relationship of
971 dissolved inorganic carbon (DIC) concentrations with some environmental variables in the Red
972 River water in the period 2008 – 2015, J. Viet. Env., 8, 102-106, 2016.
973
974 Raymond, P. A. et al.: Global carbon dioxide emissions from inland waters, Nature, 503, 355–
975 359, doi:10.1038/nature12760, 2013.

976
977 Raymond, P. A., and Cole, J. J.: Increase in the export of alkalinity from North America's largest
978 river, *Science*, 301, 88–91, doi:10.1126/science.1083788, 2003.
979
980 Raymond, P. A., and Oh, N.-H.: An empirical study of climatic controls on riverine C export
981 from three major U.S. watersheds, *Global Biogeochem. Cycles*, 21, GB2022,
982 doi:10.1029/2006GB002783, 2007.
983
984 Raymond, P. A., Oh, N. H., Turner, R. E., and Broussard, W.: Anthropogenically enhanced
985 fluxes of water and carbon from the Mississippi River, *Nature*, 451, 449–452,
986 doi:10.1038/Nature06505, 2008.
987
988 Raymond, P.A., and Bauer, J.: Atmospheric CO₂ evasion, dissolved inorganic carbon production,
989 and net heterotrophy in the York River estuary, *Limnol.Oceanogr.*, 45, 1707-1717, 2000.
990 Regnier, P., et al.: Anthropogenic perturbation of the carbon fluxes from land to ocean, *Nat.*
991 *Geosci.*, 6, 597–607, doi:10.1038/ngeo1830, 2013.
992
993 Ren, W., Tian, H., Tao, B., Yang, J., Pan, S., Cai, W.-J., Lohrenz, S. E., He, R., and Hopkinson,
994 C. S.: Large increase in dissolved inorganic carbon flux from the Mississippi River to Gulf of
995 Mexico due to climatic and anthropogenic changes over the 21st century, *J. Geophys. Res.*
996 *Biogeosci.*, 120, 724–736, doi:10.1002/2014JG002761, 2015.
997
998 Rengarajan, R., and Sarma, V. V. S. S.: Submarine groundwater discharge and nutrient addition
999 to the coastal zone of the Godavari estuary, *Mar. Chem.*, 172, 57-69, 2015.
1000
1001 Samanta, S., Dalai, T. K., Pattanai K. J. K., Rai, S. K., and Mazumdar, A.: Dissolved inorganic
1002 carbon (DIC) and its $\delta^{13}\text{C}$ in the Ganga (Hooghly) River estuary, India: Evidence of DIC
1003 generation via organic carbon degradation and carbonate dissolution. *Geochim.Cosmochim.Acta*,
1004 165, 226–248, 2015.
1005
1006 Sarma, V. V. S. S., et al.: Emission of carbon dioxide from the Indian monsoonal estuaries,
1007 *Geophys. Res. Lett.*, 39, L03602, doi:10.1029/2011GL050709, 2012.
1008
1009 Sarma, V. V. S. S., et al.: High CO₂ emissions from the tropical Godavari estuary (India)
1010 associated with monsoon river discharges, *Geophys. Res. Lett.*, 38, L08601,
1011 doi:10.1029/2011GL046928, 2011.
1012
1013 Sarma, V. V. S. S., Kumar, M. D., and Manerikar, M.: Emission of carbon dioxide from a
1014 tropical estuarine system, Goa, India, *Geophys. Res. Lett.*, 28, 1239–1242,
1015 doi:10.1029/2000GL006114, 2001.
1016
1017 Sarma, V. V. S. S., Krishna, M. S., Prasad, V. R., Kumar, B. S. K., Naidu, S. A., Rao, G. D.,
1018 Viswanadham, R., Sridevi, T., Kumar, P. P., and Reddy, N. P. C.: Distribution and sources of

1019 particulate organic matter in the Indian monsoonal estuaries during monsoon, *J. Geophys. Res.*
1020 *Biogeosci.*, 119, doi:10.1002/2014JG002721, 2014.
1021
1022 Shanley, J. B., Kendall, C., Smith, T. E., Wolock, D. M., and McDonnell, J. J.: Controls on old
1023 and new water contributions to stream flow at some nested catchments in Vermont, USA,
1024 *Hydrol. Process.*, 16, 589–609, 2002.
1025
1026 Shetye, S. R., Gouveia, A. D., Shenoi, S. S. C.: Circulation and water masses of the Arabian Sea,
1027 *Proc. Indian Acad. Sci. Earth Planet. Sci.*, 103, 107–123, 1994.
1028
1029 Shin, W. J., Chung, G. S., Lee, D., and Lee, K. S.: Dissolved inorganic carbon export from
1030 carbonate and silicate catchments estimated from carbonate chemistry and $\delta^{13}\text{C}_{\text{DIC}}$. *Hydrol. Earth*
1031 *Syst. Sci.*, 15, 2551 – 2560, 2011.
1032
1033 Singh, S.K., Sarin, M.M., and France-Lanord, C.: Chemical erosion in the eastern Himalaya:
1034 Major ion composition of the Brahmaputra and ^{13}C of dissolved inorganic carbon. *Geochim.*
1035 *Cosmochim. Acta*, 69, 3573-3588, 2005.
1036
1037 Smith S. L.: Understanding the Arabian Sea: reflections on the 1994–1996 Arabian Sea
1038 expedition, *Deep Sea Res. Part II* 48, 1385–1402, 2001.
1039
1040 Solomon, D. K., and Cerling, T. E.: The annual carbon dioxide cycle in a montane soil:
1041 observations, modeling and implications for weathering, *Water Resources Res.*, 23, 2257-2265,
1042 1987.
1043
1044 Soman, M. K., and Kumar, K. K.: Some aspects of daily rainfall distribution over India during
1045 the southwest monsoon season, *Int. J. Clim.*, 19, 299–311, 1990.
1046
1047 Sreenivas, K., Dadhwal, V. K., Suresh, K., Sri Harsha, G., Tarik, M., Sujatha, G, Suresh, J. R.,
1048 G., Fyzee, M., and Ravisankar, T.: Digital mapping of soil organic and inorganic carbon status in
1049 Indi, *Geoderm.*, 269. 160-173, 10.1016/j.geoderma.2016.02.002, 2016.
1050
1051 Sridevi, B., Sarma, V.V.S.S., Murty, T.V.R., Sadhuram, Y., Reddy, N.P.C., Vijayakumar, K.,
1052 Raju, N.S.N., Jawahar Kumar, Ch., Raju, Y.S.N., Luis, R., Kumar, M.D., Prasad, K.V.S.R. :
1053 Variability in stratification and flushing times of the Gautami–Godavari estuary, India, *J. Earth.*
1054 *Sys., Sci.*, 124, 993-1003, 2015.
1055
1056 Stevenson, B. A., Kelly, E. F., McDonald, E. V., and Busacca, A. J.: The stable carbon isotope
1057 composition of soil organic carbon and pedogenic carbonates along a bioclimatic gradient in the
1058 Palouse region, Washington State, USA. *Geoderma*, 124, 37–47, 2005.
1059
1060 Subramanian, V.: Sediment load of Indian rivers, *Curr. Sci.*, 64, 928–930, 1993.
1061
1062 Suzuki, R., and Ishimaru, T.: An improved method for the determination of phytoplankton
1063 chlorophyll using N,N-dimethyl formamide, *J. Oceanogr.*, 46, 190–194, 1990.

1064
1065 Tamooch, F., Borges, A. V., Meysman, F. J. R., Meersche, K. V. D., Dehairs, F., Merckx, R. et
1066 al.: Dynamics of dissolved inorganic carbon and aquatic metabolism in the Tana River Basin,
1067 Kenya, *Biogeosciences Discuss.*, 10, 5175–5221, 2013.
1068
1069 Tamse, S., Ogrinc, N., Walter, L.M., Turk, D., and Faganeli, J.: River Sources of Dissolved
1070 Inorganic Carbon in the Gulf of Trieste (N Adriatic): Stable Carbon Isotope Evidence, *Estuaries
1071 and Coasts*, DOI 10.1007/s12237-014-9812-7, 2014.
1072
1073 Tank, J. L., Rosi-Marshall, E. J., Griffiths, N. A., Entekin, S. A., and Stephen, M. L.: A review
1074 of allochthonous organic matter dynamics and metabolism in streams, *J. North Am. Benthol.
1075 Soc.*, 29, 118–146, doi:10.1899/08-170.1, 2010.
1076
1077 Telmer, K., Veizer, J.: Carbon fluxes, pCO₂ and substrate weathering in a large northern river
1078 basin, Canada: carbon isotope perspectives, *Chem. Geol.*, 159, 61–86, 1999.
1079
1080 Vähätalo, A. V., and Wetzel, R. G.: Long-term photochemical and microbial decomposition of
1081 wetland-derived dissolved organic matter with alteration of ¹³C:¹²C mass ratio, *Limnol.
1082 Oceanogr.*, 53, 1387–1392, doi:10.4319/lo.2008.53.4.1387, 2008.
1083
1084 Varkey, M. J., Murty, V. S. N., and Suryanarayana, A.: Physical oceanography of the Bay of
1085 Bengal and Andaman Sea, *Oceanogr. Mar. Biol.*, 34, 1–70, 1996.
1086
1087 Viers, J., Dupre, B., Polve, M., Schott, J., Dandurand, J-L, and Braun, J-J.: Chemical weathering
1088 in the drainage basin of a tropical watershed (Nsimi-Zoetele site, Cameroon): comparison
1089 between organic-poor and organic-rich waters. *Chem. Geol.*, 140, 181-206, 1997.
1090
1091 Viers, J., Oliva, P., Dandurand, J. L., Dupré, B., Gaillardet, J., Heinrich, D. H., and Karl, K. T.:
1092 Chemical weathering rates, CO₂ consumption, and control parameters deduced from the
1093 chemical composition of Rivers, *Treatise on Geochemistry Pergamon, Oxford*, 2007.
1094
1095 Vijith, V., Sundar, D., and Shetye, S. R.: Time-dependence of salinity in monsoonal estuaries,
1096 *Estuar. Coast. Shelf Sci.*, 85, 601–608, doi:10.1016/j.ecss.2009.10.003, 2009.
1097
1098 Waldron, S., Hall, A. J., and Fallick, A. E.: Enigmatic stable isotope dynamics of deep peat
1099 methane, *Glob. Biogeochem. Cy.*, 13, 93–100, doi:10.1029/1998gb900002, 1999.
1100
1101 Waldron, S., Scott, E. M., and Soulsby, C.: Stable isotope analysis reveals lower-order river
1102 dissolved inorganic carbon pools are highly dynamic, *Environ. Sci. Technol.*, 41, 6156–6162,
1103 doi:10.1021/es0706089, 2007.
1104
1105 Wang, J. P., Wang, X. J., Zhang, J., and Zhao, C. Y.: Soil organic and inorganic carbon and
1106 stable carbon isotopes in the Yanqi Basin of northwestern China, *Eur. J. Soil Sci.*, 66, 95–103,
1107 2015.
1108

1109 Wang, Z. A., Biennu, D.J., Mann, P.J., Hoering, K.A. Poulsen, J.R., Spencer, R.G.M., and
1110 Holmes, R.M.: Inorganic carbon speciation and fluxes in the Congo River, *Geophys. Res. Lett.*,
1111 40, 511–516, 2013.

1112
1113 Williamson, C. E., Zagarese, H. E., Schulze, P. C., Hargreaves, B. R., and Seva, J.: The impact
1114 of short-term exposure to UV-B radiation on zooplankton communities in north temperate lakes,
1115 *J. Plankton Res.*, 16, 205–218, doi:10.1093/plankt/16.3.205, 1994.

1116
1117 Yang, C., Telmer, K., and Veizer, J.: Chemical dynamics of the “St. Lawrence” riverine system:
1118 $\delta\text{DH}_2\text{O}$, $\delta^{18}\text{O}_{\text{H}_2\text{O}}$, $\delta^{13}\text{C}_{\text{DIC}}$, $\delta^{34}\text{S}_{\text{sulfate}}$, and dissolved $^{87}\text{Sr}/^{86}\text{Sr}$, *Geochim. Cosmochim. Acta*, 60,
1119 851–866, 1996.

1120
1121 Zeng, F- W., Masiello C. A., and Hockaday, W. C.: Controls on the origin and cycling of
1122 riverine dissolved inorganic carbon in the Brazos River, Texas, *Biogeochemistry*, 104, 275–291,
1123 doi:10.1007/s10533-010-9501-y, 2011.

1124
1125 Zeng, J., Guo, T. W., Bao, G. X., Wang, Z., and Sun, J. H.: Effections of soil organic carbon and
1126 soil inorganic carbon under long-term fertilization, *Soil and Fertilizer Sciences in China* 2, 11–
1127 14, 2008.

1128
1129 Zou, J.: Sources and Dynamics of Inorganic Carbon within the Upper Reaches of the Xi River
1130 Basin, Southwest China, *PLoS One*, 11, e0160964. doi:10.1371/journal.pone.0160964, 2016.

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1135 **Figure 1:** Map showing the study region. Estuaries of the rivers sampled in this study were
1136 indicated by solid black line.

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1138 **Figure 2:** Concentration (mg l^{-1}), export flux (Tg yr^{-1}) and yield ($\text{g m}^{-2} \text{yr}^{-1}$) of dissolved
1139 inorganic carbon (DIC) in the Indian monsoonal estuaries. Estuaries geographically located in
1140 the northeastern (NE), southeastern (SE), southwestern (SW) and northwestern (NW) regions of
1141 India were also shown. Estuaries draining into the Bay of Bengal and the Arabian Sea were also
1142 provided

1143
1144 **Figure 3:** Spatial variability in stable carbon isotopes of dissolved inorganic carbon ($\delta^{13}\text{C}_{\text{DIC}}$, ‰)
1145 in the Indian monsoonal estuaries during discharge period.

1146 **Figure 4:** (a) Positive correlation between dissolved inorganic carbon (DIC) concentration and
1147 catchment area, and (b) negative correlation between DIC concentrations and annual mean
1148 discharge (km^3) of the minor-medium rivers.

1149
1150 **Figure 5:** Inverse correlation between mean dissolved inorganic carbon concentration in
1151 estuaries (DIC , mg l^{-1}) and annual mean rainfall (mm) in catchments of the rivers in the NE, NW,
1152 SE and SW regions of India.

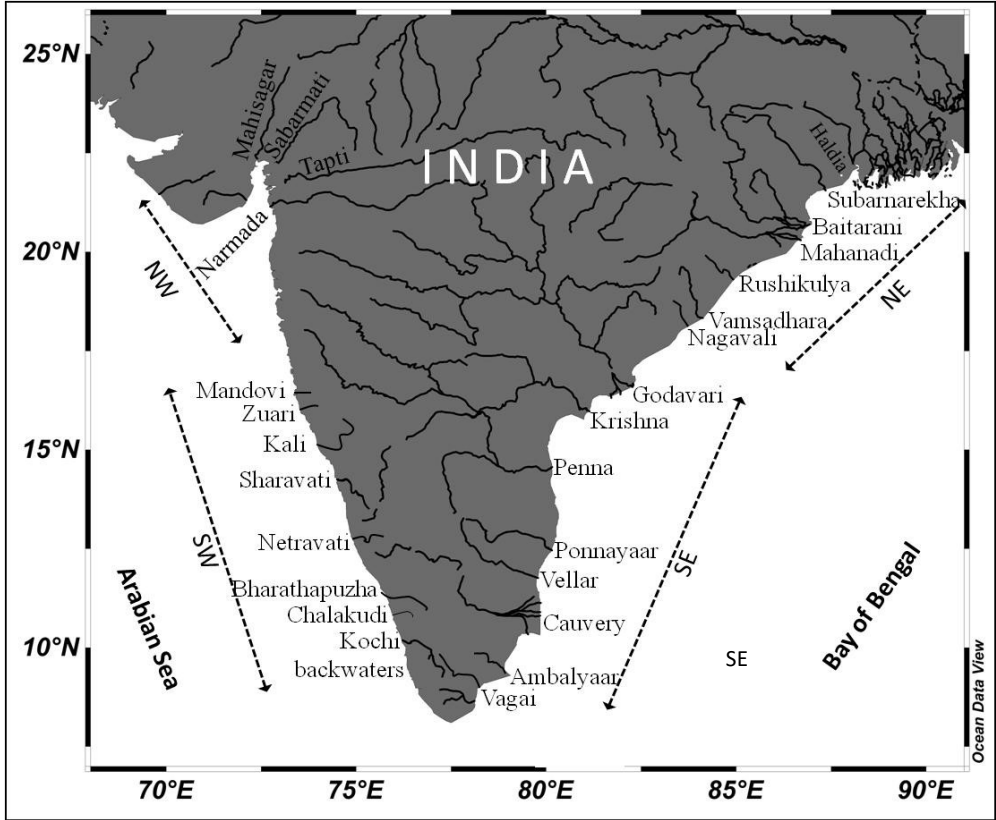
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1154 **Figure 6:** Significant positive correlation between stable carbon isotopes of dissolved inorganic
1155 carbon ($\delta^{13}\text{C}_{\text{DIC}}$, ‰) and salinity in the Indian monsoonal estuaries during the study period.

1156 **Figure 7:** Significant positive correlation between stable carbon isotopes of dissolved inorganic
1157 carbon ($\delta^{13}\text{C}_{\text{DIC}}$, ‰) and concentrations of DIC in the Indian monsoonal estuaries (filled
1158 diamonds), SW estuaries (filled squares) and high saline estuaries (hollow triangles) during the
1159 study period.

1160 **Figure 8:** Relationship of dissolved inorganic carbon (DIC) yield ($\text{g m}^{-2} \text{yr}^{-1}$) with that of (a)
1161 rainfall (mm) and (b) soil organic carbon (kg ha^{-1}) in the catchment area of the NE, NW, SE and
1162 SW rivers
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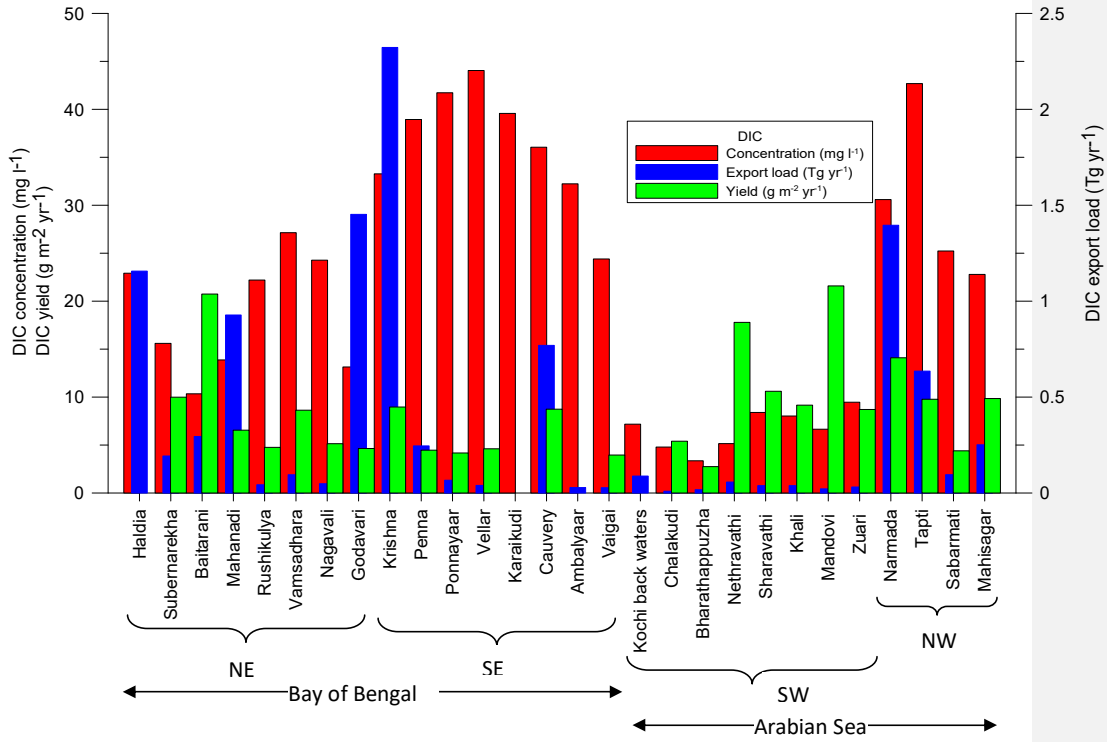
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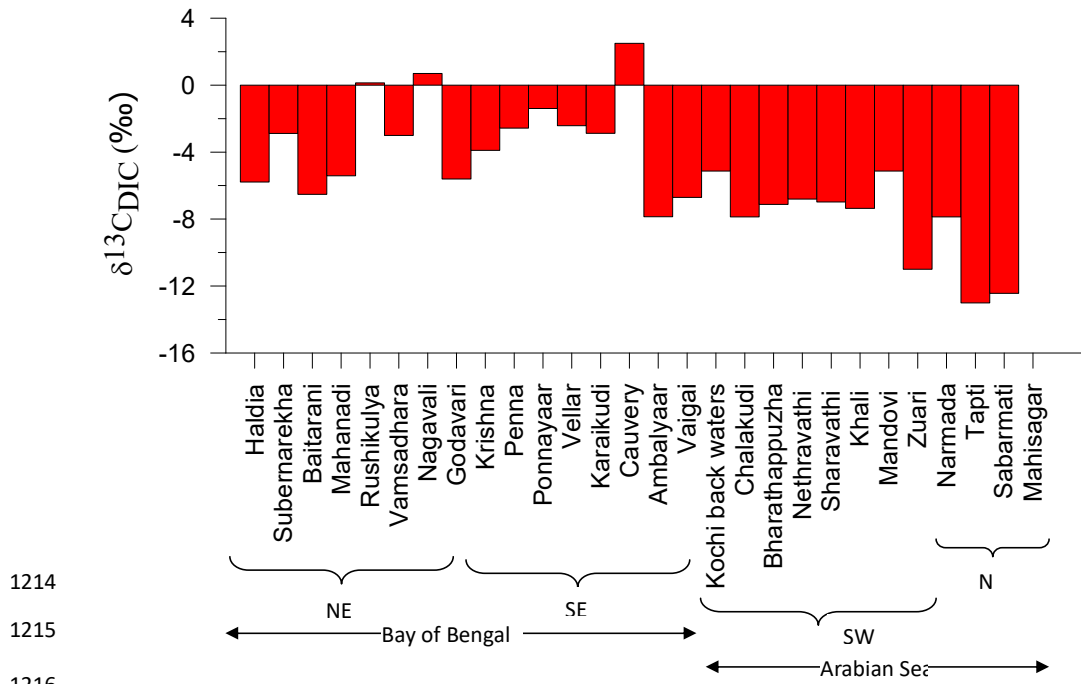
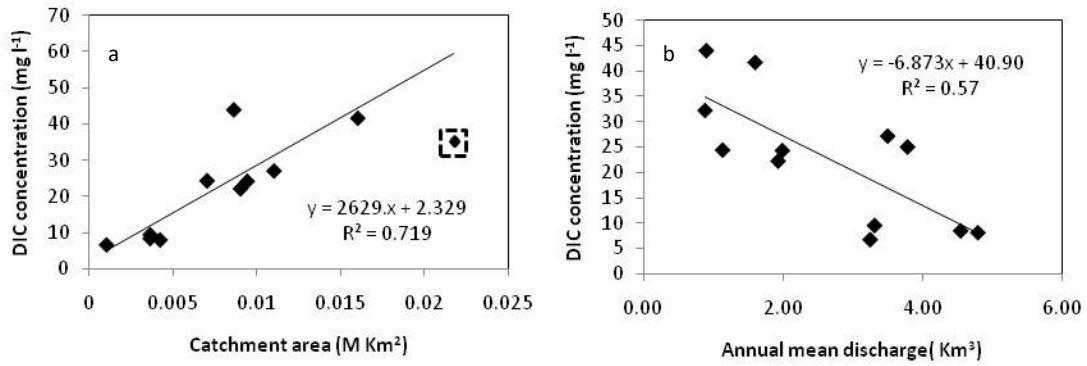


Fig. 3:

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Fig. 4:

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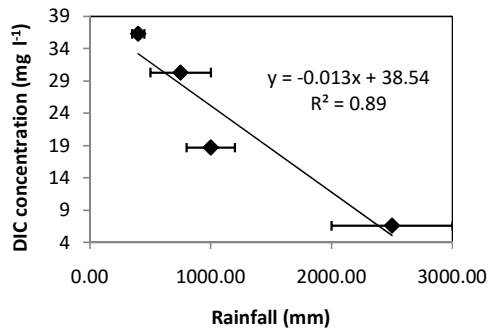
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Fig. 5:

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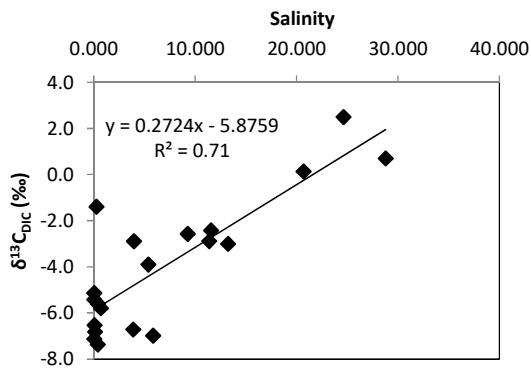
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Fig. 6:

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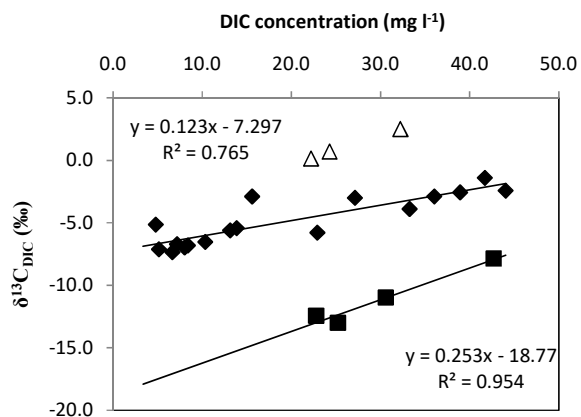


Fig. 7:

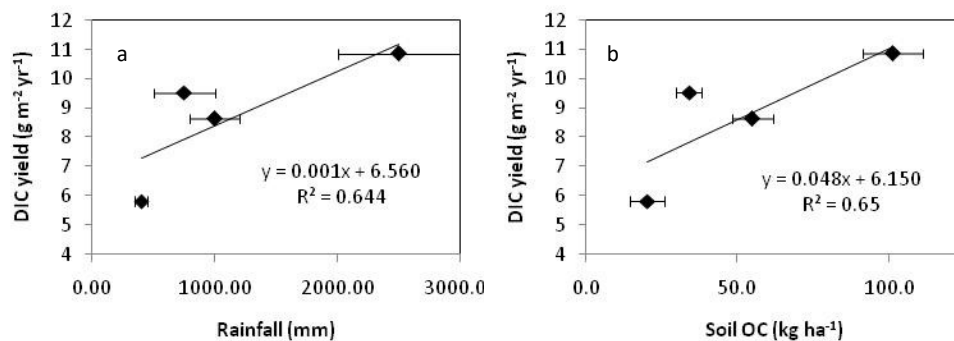


Fig. 8:

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