Major comments

Method for DIC fluxes calculation

L200 “Total export flux of DIC… multiplying the mean concentration of DIC in an estuary with the annual discharge. Spatial variability of DIC in estuaries was minimized to a large extend by collecting samples from head to mouth of the estuary” Estuarine saline samples contain a fraction of seawater; their DIC is thus a mixture of riverine DIC and marine DIC, with proportions depending to the value of salinity. The concentrations of DIC in these estuarine samples are affected by dilution with seawater and cannot be multiplied by the freshwater discharge to derive a flux to the ocean. According to all textbooks on estuarine hydrology and chemistry, the DIC concentration at the zero salinity end-member must be used. Concentration versus salinity plots can be used to obtain an “apparent zero end-member” (AZE). The method used here is underestimating the real DIC flux when freshwater DIC cc is higher than seawater DIC cc (most cases in these indian rivers), and, inversely, overestimating the flux when river DIC < ocean DIC.

The samples collected at stations of only near zero salinities (riverine end members) were used in this study and saline samples (salinity >2) were excluded. In a very few medium estuaries where samples were not collected at near zero salinities, the riverine end member was calculated from the DIC versus salinity plot, as you suggested. As we have considered only riverine end members for this study, the text was modified accordingly and confined only to rivers. The export fluxes of DIC are estimated by multiplying DIC concentrations at riverine end member with the discharge of the river.

The use of the term “estuary” all through the MS in often inappropriate. At many places in the MS, in the abstract, introduction and discussion, you mix the two notions of river and estuary. You use the word “estuary” when it should be “river”. See below

Since we have considered only the riverine end member samples for this study, we have used only ‘river’ throughout the manuscript, i.e., in the abstract, introduction and discussion.

Interpretation of 13C DIC

In general, the discussion of the 13C DIC signal is confusing and too superficial. It must be strengthen by more quantitative analysis that allows to evaluate the respective contribution of processes under some specific conditions. You mention weathering of silicate/carbonate and “storage” in dams as major processes. However the relationship between DIC cc and 13C and lithology is only verbal. You must relate DIC concentrations and 13C-DIC signature with lithology and the proportion of carbonate versus silicate rocks on the respective watershed.

The discussion on the 13C DIC was changed completely and re-written. It was given in a separate section 4.2. Since the 13C DIC measured in the riverine end member samples is a mixture of different carbonate species (HCO3, CO3 and CO2) which are largely

dependent on pH and temperature. Further, in-stream physical and biological processes alter the 13C of source. In order to separate the influence of different in-stream processes on 13C of DIC source, we tried to approximate the 13C of DIC source using two different graphical model techniques, Keeling plot and Miller-Tans plot and interpreted the outcome of the models for DIC sources. Deviations of the measured 13C DIC from the approximated 13C of DIC source were attributed to the influence of in-stream processes. Further, to filter the influence of pH and DIC speciation on bulk13C DIC, we approximated the 13C of CO2 using a set of isotopic fractionation enrichment factors across the DIC speciation. With this approach, we found that 13C DIC in most (~75%) of the Indian rivers is influenced by heterotrophic decomposition of organic matter, whereas in the rivers from southeast region of India, it is controlled by autotrophic production. Overall, we tried to explain the 13C DIC in a more quantitative manner.

Include lithology in the map in figure 1 and provide in a table the proportion of carbonate and silicate rocks in each watershed. The table should also include classical information for each watershed (discharge, drainage, presence of dams, eventually population density). All information necessary for the quantitative information of the data should be included.

Lithology was included in the map (Fig. 1), as you suggested. A table (Table 1) with all the necessary information such as catchment area, discharge, length of the river, number of dams on the river and population density in catchment area of the river was provided. However, we could not get the information on proportions of silicate and carbonate rocks in each watershed, and thus not provided in the table.

Sections 4.1.2 and 4.1.3 on the origin of DIC is extremely speculative and sometimes repetitive. We must know what are major processes and minor processes. Pollution by megacities is ignored although it is probably a major driving force on DIC in these rivers.

As mentioned above, the discussion on 13C DIC was changed completely and it was re-written to present the results in a more quantitative way. The major processes influencing the 13C DIC were identified using graphical model approach (Keeling and Miller-Tans plots). To filter the influence of pH and DIC speciation on the bulk 13C DIC, we have approximated the 13C of CO2. Influence of pollution on DIC was also included now as it is also important to be considered.

Detailed comments

Abstract
L14: spatial and/or temporal variability?

It is spatial variability. It was mentioned the text.

L17: “exchange of groundwater with low DIC” this is speculation because groundwater are supposed to have high DIC concentrations

Yes. DIC concentrations are higher in ground waters than the DIC in rivers/estuaries. Spatial variability of ground water DIC showed the lowest mean values in the SW region than the other regions of India. We mean to say here that exchange of ground waters with relatively lower DIC in this region (but not lower than the estuarine DIC) than the other regions. However, the sentence was modified during revision.

L21: “intrusion of marine waters”: intrusion of seawater occurs in all estuaries (not only east-flowing estuaries), and are more or less important depending of sampling strategy and salinity of the samples. Comparison between systems can be done only on the basis of the zero salinity end-member.

DIC concentrations at stations of only near zero salinities in each river (riverine end member) were considered for this study. Since the saline samples were not included in this study, the sentence was modified accordingly.

L27-28: “9.4% of the Asian rivers” unintelligible sentence

Since the Indian rivers are part of the Asian rivers, this part of the sentence was deleted.

Intro
L44 “DIC is the major constituent of carbon species” where, please rephrase

It was rephrased as “Dissolved inorganic carbon (DIC) is one of the major constituent of carbon species in rivers’.

L46 “33-400 Tg of DIC” do you mean 330-400 Tg? Please proofread you MS to remove such typos before re-submitting

Sorry for the mistake, it was 330 only but not 33. However, it was given more precisely in the revised text.

L50: should be “rivers” instead of “estuary” here

Since only the riverine end members were used for this study, ‘river’ was used throughout the manuscript.

L51-55: rephrase and specify what process is more important quantitatively

During re-structuring the manuscript, the major processes controlling DIC concentrations in the Indian monsoonal rivers were discussed in the 'discussion' chapter. Hence, it was deleted from here.

L62-65: this is too general, how does human alterations of riverine DIC relate with findings of your study?

These two sentences were deleted as our findings are not related with human alterations. However, the influence of population density on DIC concentrations in the Indian monsoonal rivers was discussed in the section 4.1 and shown in Fig. 4g

L70-88: this is too general, and not focussed in relation with the present study. Please rewrite

Entire 'Introduction' chapter was re-written. During this process, this section (L.70-88) was deleted.

L88 "their sources" of DIC?

Yes. It is 'their sources of DIC'. However, this sentence was deleted from the text as it is related to the estuarine export of DIC (including saline samples) to the coastal ocean.

L90 aquatic systems

Corrected to 'aquatic systems'

L91-93 "due to distinct isotopic composition of different sources" not only as 13C-DIC is also affected by fractionation by internal processes such as aquatic primary production and gas exchange. The figure 5 should be used here (as new figure 2) to support this statement

The section was modified as ".Though the δ^{13}Cof DIC derived from different sources is well separable (Deines et al., 1974), the isotopic fractionation by in-stream physical and biological processes alters the δ^{13}C of DIC source (Fig. 2). For example, photosynthesis and equilibration with atmospheric CO_2 enriches (O'Leary, 1988; Finlay, 2004; Parker et al., 2005, 2010) while the heterotrophic decomposition of organic matter and photo-oxidation of dissolved organic carbon depletes the δ^{13}C of DIC (Opsahl and Zepp, 2001; Finlay, 2003; Waldron et al., 2007; Vahatalo and Wetzel, 2008) (Fig. 2)".

The figure 5 was given as figure 2 to explain various in-stream processes influencing the 13C of DIC.

L100: "despite distinct isotopic composition of DIC is expected for different sources, the identification of DIC sources is still challenging due to isotopic fractionation associated with complex mixture of sources and processes such as photosynthesis" awkward sentence. Mixture does not fractionate. All through the MS you must prioritize what is more important among the different processes that impact 13C-DIC

The sentence was shifted to section 4.2 (sources of DIC). It was modified as "Though, the $\delta^{13}C_{DIC}$ is a promising tool to decipher the sources of DIC, its interpretation for

source material identification in rivers is still challenging because multiple physical and biological processes within the rivers significantly alter the $\delta^{13}C$ of DIC source. The influence of major in-stream processes on the $\delta^{13}C_{DIC}$ must be separated before interpreting the results for major sources of DIC, failing which leads to erroneous conclusions. In order to identify and separate DIC sources, we used here two different graphical mixing model techniques, Keeling plot (Keeling, 1958; Pataki et al., 2003) and Miller-Tans plots (Miller and Tans, 2003). These models approximate the hypothetical $\delta^{13}C$ of source material as an intercept (in Keeling plot) and slope (in Miller-Tans plot) of the least square linear regression equations (Pataki et al., 2003; Campeau et al., 2017). The deviations from the approximated $\delta^{13}C$ of source can be interpreted to the influence of in-stream processes".

Study area
We need a table with all necessary information about each watershed

A table (Table 1) was provided with all necessary information of each watershed.

L121_124 Not sure information on upwelling in the Arabian sea is necessary here

Since the rivers draining into the Arabian Sea were also covered in this study, the information on the Arabian was also provided along with the Bay of Bengal.

L138 rivers instead of "estuaries"?

It was changed to 'rivers' as you suggested

L147 when talking about discharge, better refer to "rivers" rather than "estuaries"

The term 'rivers' was used throughout the manuscript
Sample collection
We need to know about salinity of your samples. Comparing data with high salinity from an estuary with data with low salinity in another estuary is meaningless. Salinity must appear somewhere in the MS, and what should be compared is DIC and 13C-DIC at the zero salinity end-member.

Salinity samples were not included in this study. Hence, discussion on DIC and 13C-DIC was confined to freshwater regions.
L200: use the AZE DIC concentration to calculate fluxes

Mean of the DIC concentrations measured at stations of near zero salinity in each river was used for DIC export flux estimations. In some cases, where measurements at near zero salinity were not conducted, apparent zero end member was estimated from the relationship of DIC with salinity, as you suggested.

L214: here you mix different information (river composition and dilution with seawater) and the fact that salinity is high in an estuary is due to your sampling strategy. All interpretation

should refer to river end-member, and should not include different proportion of dilution with seawater

DIC concentrations at near zero salinity were used for this study. No saline samples were included in this study and thus no contribution of DIC from seawater. Hence, comparison, discussion and interpretations made in this study were confined only to the river end member.

Section 3.3: re-calculate DIC fluxes using the zero salinity end member

DIC fluxes were re-calculated using the measured DIC concentrations at near zero salinity (river end member). For few medium rivers, where samples were not collected at near zero salinity stations, the riverine end member was obtained from the DIC versus salinity plot, as you suggested.

L258 "no vertical salinity stratification was observed in all estuaries sampled… Mandovi estuary" awkward sentence

The sentence was deleted during revision as the saline samples were not included in this study. Hence, stratification in estuaries is irrelevant here.

L273 "among the estuaries sampled along the indian coast, the SW estuaries are characterized by lower mean concentrations of DIC than the SE, NE…" awkward sentence, please rephrase

The sentence was modified during the revision. '**Distribution of DIC in the Indian monsoonal rivers showed large spatial variability, with the lowest values in rivers from the SW region of India (Fig. 3a)'.**

L279 "the SW region of India receive the highest amount of precipitation during the SW moosoon than the SE, NE and NW regions of India" awkward sentence, please rephrase

The sentence was corrected and rephrased as 'The spatial distribution of rainfall over the Indian subcontinent (www.imd.gov.in) shows that the SW region receives the highest annual rainfall (~3000 mm) than the rest of India (Soman and Kumar, 1990). '

L286 "About three times higher catchment area normalized discharge might have diluted DIC concentration in the SW estuaries" not clear, please rewrite

It was modified to obtain clarity. 'In order to understand the influence of the density of rainfall on DIC in rivers, we normalized the volume of discharge from the river with its catchment area. . The catchment area normalized volume of discharge was found to be much higher in rivers from the SW region (1.71 m^3 m^{-2}) than the rivers from SE (0.17 m^3 m^{-2}), NE (0.6 m^3 m^{-2}) and NW (0.32m^3 m^{-2}) regions of India. About three times higher catchment area normalized discharge might have diluted DIC concentrations in the rivers of the former region.

L289 rivers instead of “estuaries”

It was changed to rivers. The sentence was re-written as ‘A strong exponential decrease in DIC concentrations with increasing rainfall over the catchment (r^2= 0.72, p<0.001; Fig. 4a) also suggests that DIC concentration in the Indian rivers are strongly influenced by density of precipitation over the catchment.’

L290-293 not sure dilution will alter the 13C-DIC. You mention “residence time of soil water” but this is not clear: what process will deplete 13C DIC ? Be more precise when referring to Amiotte Suchet 1999

The sentence was deleted during the revision. Discussion on 13C –DIC was provided in a separate section 4.2 (sources of DIC) to maintain the focus of the study.

L296: you mention here “lower catchment area” but the info on catchment areas is missing in the MS

Catchment area of each river was provided in the Table 1

L300-304: rewrite and provide more quantitative evidence for the occurrence of in stream processes. Here the interpretation of the data is only verbal. What instream process are you referring to?

It was re-written and some part of the paragraph was deleted. It was re-written as ‘Since the contribution of DIC from in-stream processes, such as decomposition of organic matter, has been demonstrated to increase along the course of fluvial network (Hotchkiss et al., 2015), possibly due to increase in the residence time of water (Catalan et al., 2016), the lowest DIC concentrations found in rivers from the SW region may also, at least partly, be due to their small size. Fairly good positive correlation between DIC concentrations and length of the rivers (r^2=0.38, p<0.01) also support this argument.’

L309-311. Comparing low salinity 13C signature in a given estuary with high salinity 13C-DIC signature in another estuary makes no sense. The fact that 13C-DIC correlates with salinity in a given estuary is a truism. These types of statements must be avoided in all the MS

The section on ‘mixing with seawater’ was deleted as the saline samples were not included in the present study.

L320 “exchange of SW estuaries with…” unintelligible sentence: how can you exchange en estuary? All the literature converges to the conclusion that groundwater have high DIC concentrations, so your statement here is pure speculation

The sentence was deleted as the estuarine (saline) samples were not included in this study

L328 “DIC input from the dissolution of atmospheric CO2 can be ruled out”. Isotopic equilibration of DIC with the atmosphere occurs even if waters are oversaturated with respect to the atmosphere.

Isotopic exchange is true, but here, we meant to say that the addition of DIC to rivers through the dissolution of atmospheric CO_2 is more unlikely. However, the sentence was modified to avoid confusion. It was modified as ‘Since the Indian monsoonal estuaries have been reported to be a source of CO_2 to the atmosphere during the discharge period due to heterotrophic decomposition of organic matter (Sarma et al., 2001, 2011, 2012; Gupta et al., 2008, 2009; Bhavya et al., 2018), the DIC input from the dissolution of atmospheric CO_2 may be unlikely. On the other hand, organic matter decomposition is expected to add significant amount of DIC as enhanced bacterial respiration rates were reported during this period (Sarma et al., 2011; 2012’.

L331-336 this is speculation : the fact that 13C DIC correlates positively with DOC does not necessarily mean that oxidation of organic matter predominates. High 13C DIC can be due to fractionation by phytoplankton and high DOC to the exudation/hydrolysis of phytoplanktonic cells. The correlation can also be indirect, High DOC and high 13C DIC coming from the nature of soils and rocks (or pollution) on the watersheds, or can also be due to the dilution of seawater in the samples. The highest value of DOC in fig 4e at about 13 mg L-1 may also come from contamination by sewage. See also below my comments on figure 4.

Yes. Uptake of DIC by phytoplankton enriches 13C DIC and exudation of phytoplankton cells increases DOC concentration, leading to a positive relationship between the two. Yet, it is also due to carried forward signatures from the catchment as you suggested. Hence the section was re-written as follows:

On the other hand, organic matter decomposition is expected to add significant amount of DIC as enhanced bacterial respiration rates were reported during this period (Sarma et al., 2011; 2012). In contrast, significant negative correlation between chlorophyll-*a* and DIC (r^2=-0.44, $p<0.01$; Fig. 4c), except few SE rivers where elevated phytoplankton biomass (Chl-a: >5 mg m^{-3}) was recorded, suggesting that autotrophic removal of DIC is also significant in the Indian monsoonal rivers during the study period. A significant positive relationship was observed between the $\delta^{13}C_{DIC}$ and Chl-a (r^2=0.49; $p<0.01$; Fig. 4d), supporting this argument because preferential uptake of ^{12}C than ^{13}C during photosynthesis leaves the residual DIC enriched in ^{13}C. On the other hand, $\delta^{13}C_{DIC}$ showed significant positive correlation with DO saturation (r^2=0.50, $p<0.01$; Fig. 4e) (depleted $\delta^{13}C_{DIC}$ values at more under saturation of DO) and DOC concentrations (r^2=0.43, $p<0.01$; Fig. 4f) as was observed in the Xi river (Zou et al., 2016). Altogether, enriched $\delta^{13}C_{DIC}$ are associated with higher DOC, less under saturation of DO and higher phytoplankton biomass (Chl-a) while the depleted $\delta^{13}C_{DIC}$ are associated more under saturation of DO and less DOC. This suggests that both autotrophic removal and heterotrophic addition control DIC in the Indian rivers during the discharge period, with a considerable spatial variability. However, influence of these processes on DIC concentrations is difficult to separate with this bulk $\delta^{13}C_{DIC}$ data set, as the $\delta^{13}C_{DIC}$ in rivers is also influenced by pollution, catchment lithology and outgassing of CO_2 (Shin et

al., 2011; Brunet et al., 2005; Bouvillion et al., 2009; Zeng et al., 2011; Tamooh et al., 2013). Excluding Sabarmati and Mahisagar rivers, DIC concentrations showed fairly good linear relationship with population density over the catchment of the river (r^2=0.41, p<0.01; Fig. 4g), suggesting that considerable influence of pollution from the mega cities and industries on DIC in the Indian rivers.

L338 "is distinctly enriched than that…" unintelligible language

The sentence was deleted during revision

L339-342: the contribution of C4 plant material is speculative. Many other processes may enrich DIC in 13C such as carbonate weathering (which also increases alkalinity), gas exchange and mixing with seawater DIC.

This section was modified as 'These results indicated that DIC in the Indian rivers is largely contributed by chemical weathering of carbonate and silicate minerals by soil CO_2 (-10 to -9‰). Deviations of the measured $\delta^{13}C_{DIC}$ (-13.0 to -1.4‰) from that of the approximated $\delta^{13}C$ of DIC source (-3.0 to -2.0‰) and $\delta^{13}C$ of CO_2 (-10.7‰) could be due to the influence of in-stream process. In more than 75% of the Indian rivers sampld, the deviation from the $\delta^{13}C$ of DIC source is towards negative side (depletion) ($\delta^{13}C_{DIC}$ < -3.0‰), suggesting that heterotrophic decomposition of organic matter is the dominant process controlling DIC in these rivers. While, no (or very little) deviation was observed only in rivers from the SE region of India (mean $\delta^{13}C_{DIC}$: -3.1‰) could be due to the competition between autotrophy, degassing and heterotrophy as these processes influences the $\delta^{13}C_{DIC}$ in opposite directions (Fig. 2); the former two processes causes enrichment while the latter depletes $\delta^{13}C_{DIC}$.. Relatively higher phytoplankton biomass (mean Chl-a: 4.6 mg m^{-3}) and less unsaturation of DO (98.7%) was observed in these rivers compared to the mean of the rest of the Indian rivers (2.4 mg m^{-3} and 87.5% respectively), suggesting that autotrophy is one of the dominant processes controlling DIC in rivers from the SE region of India. Total number of dams on the rivers from this (SE) region (mean 155, Table 1) is not significantly higher from that of the mean of total number of dams on the Indian rivers sampled (mean 135) , suggesting that degassing due to storage of water may not be the dominant process responsible for enrichment in $\delta^{13}C_{DIC}$ of these rivers".

We need some quantitative analysis here: what are the contributions of silicate and carbonate rocks on the respective watersheds, and does 13C DIC correlate with alkalinity? High alkalinity is an indication of carbonate weathering

Yes. 13C DIC is correlated with alkalinity and the same was given in figures (Fig. 4h). Since the discussion on 13C-DIC was changed completely, this paragraph was modified during the revision.

L326-377. All the section 4.1.2 is extremely speculative, discussion on processes are only verbal and not quantitative. All the possible processes are mentioned but without giving more emphasis on one process compared to another. Entire section must be rethought and re-written.

It was completely re-written as mentioned in above responses to your comments.

L360 "we could not able to evaluate" please proofread your MS before submitting!

It was deleted during the revision

L362 "the same is correlated with DO saturation and found significant…" revise language "confirming that biological processes enriched 13C DIC in the Indian estuaries. CO2 outgassing due to heterotrophic decomposition…. In reservoirs/dams…" This is extremely confusing and speculative. In fact you mention all possible processes but you don't know from your analysis which one is most important. In addition "biological activity" is too general and what most probably makes the 13C become enriched is primary production (autotrophy) and not heterotrophy.
Pollution by megacities is ignored although it is probably a major driving force on DIC in these rivers.

The sentence was deleted, and the entire section was re-written. Influence of pollution on DIC in the Indian rivers was also discussed. During re-structuring of the manuscript, DIC concentrations and 13C-DIC were separated in to two sections, i.e., 4.1 and 4.2, to maintain the focus of our study.

L388 your are repeating the same statements as before

Repeated statement was deleted

L395 "…consumption of atmospheric/soil CO2 through silicate weathering is lower by 2 times…" is this a rule that applies everywhere? Rephrase

The sentence was modified to obtain clarity. Lateritic soils, which are poor in lime and silicate, occupied the catchment of the rivers in the SW region of India. Relatively lower chemical weathering rates of the lateritic than the non-lateritic soils could be one of the reasons for the observed lower DIC concentration the rivers from SW region of India.

End of section 4.1.3 is too long and repetitive

The discussion of isotopes (end of the section 4.1.3) was deleted as it is a repetition. In order to avoid the repetitions on $\delta^{13}C$ of DIC, the entire discussion on this topic has been given in a separate section, 4.2.

L427 80% of DIC and 20% of DOC (what about POC?) is different from other rivers in the world and not "consistent with earlier reports elsewhere"

Here, we meat to say the fluvial dissolved (not particulate) carbon fluxes. However, the sentence was modified to avoid confusion. Contributions of DIC and DOC to dissolved carbon fluxes vary from region to region. For example, DIC dominates, as was observed in this study, in the British rivers and high altitude Swedish rivers while DOC

dominates in the low land Swedish rivers. It is not uniform over world.

L435 “similar those found…” revise language

The sentence was revised. The yield of DIC found in this study (mean 8.7±5.2 g m^{-2} yr^{-1}) is close to those found in rivers from the tropical region of Asia, but significantly higher than those reported from tropical region of the American and African continents (Huang et al., 2012).

L451: you repeating the same statements

As the statement was mentioned previously, it was deleted here to avoid repetition.

L465: “significant correlation between soil organic carbon and DIC yield … confirms that strong influence of soil and DIC yield…” a correlation does not necessarily prove a process, correlation can be indirect. However, this is probably one interesting finding that deserves more discussion: soil OC oxidation will release DIC in the form of excess CO2, but it wil also enhance weathering and the export of DIC in the form of alkalinity. Deserve more discussion

Yes. Correlation does not necessarily prove a process, but we used correlations to understand the major processes controlling DIC in the Indian rivers. Soil OC was found to be higher in the SW region compared to the SE, NE and NW regions of India. It might have increased the yield of DIC from rivers of the former region as decomposition of soil OC releases CO2 leading to formation of acidic conditions which subsequently increases the dissolution of carbonate minerals and chemical weathering of carbonate and silicate rocks.

L489 (and abstract) “storage of water in dams” you must consider the probable occurrence of phytoplanktonic production that would decrease DIC and increase 13C DIC.

Phytoplankton production in rivers was found to be the dominant processes controlling the 13C DIC in rivers from the southeast region of India. Whereas, heterotrophic decomposition was found to be dominant in the other Indian rivers studied. However, the average number of dams on the rivers from SE region is close to the average number of dams on the Indian rivers, suggesting that influence of water storage in dams on 13C DIC in these rivers is minimal. It could be due to competition between different processes that enrich (autotrophic production and degassing) and depletes (heterotrophic decomposition of organic matter) the 13C DIC in dams.

Owing to the population density in India, DIC is probably largely affected by the release of sewage waters

Yes. DIC concentrations in Indian rivers are affected by the pollution, Population density in the catchment area of the rivers showed linear relationship with DIC concentrations in river as shown in Fig. 4g.

Fig1: please show lithology and calculate the respective proportion of carbonate and silicate in each watershed

Distribution of soils was shown on the map in Fig. 1. However, we could not get the information on respective proportions of carbonate and silicate in each watershed.

Fig2 and 3 can be combined. Please place the west on the left and the east on the right as in figure 1. Reading would be improve by showing DIC yields on a seprated panel

Fig.2 and 3 were combined and given in one panel as Fig. 3. Concentrations, export flux and yield of DIC were shown separately (Fig. 3a,b,c respectively) for better reading. West-flowing rivers were placed on the left and east-flowing rivers were shown on the right, as you suggested.

Figs 4 and 6 are very confusing, number of data points are different from one panel to another, avoid mixing different information in such figure. Prepare one figure with a limited number of variable on the X axis. It is not clear why fig 4a, 6g and 6 h have only 4 data points: insert these ones in a different figure.

Some of the figures were removed and some were modified. All the figures were given in one panel (Fig. 4 a-g) to obtain clarity.

What do ovals represent is not mentioned in the captions

Ovals represent the outliers and were not considered in the regression equations.

In fig 4 you show 13C DIC versus salinity, and 13C dic versus DOC. What about DOC and DIC versus salinity? Salinity plots should appear in a separate figure in order to calculate zero salinity end-members

Since DIC concentrations at stations of only near zero salinities were considered for this study, figures with salinity were deleted.

Fig5 could appear before when mentioning the processes controlling 13C DIC in rivers in the introduction

Figure 5 was given as figure 2 while explaining the in-stream processes influencing 13C DIC in rivers

Table 1 is useless; what is the meaning of comparing DIC concentrations in world rivers with Indian estuaries that include saline samples?

This table was deleted

Table 2 should show the information for individual rivers, not only regions. Quantitative

information on the lithology in the watershed should be included and used for quantitative analysis of the data

A table (Table 1) was provided with all necessary data of each river including the catchment area, discharge, length of the river, number of dams along the course of the river and, population density and annual mean precipitation over the catchment.

Export fluxes of dissolved inorganic carbon to the Northern Indian Ocean from the Indian monsoonal rivers

Moturi S. Krishna[1], Rongali Viswanadham[1], Mamidala H. K. Prasad[1], Vuravakonda R. Kumari[1], Vedula V. S. S. Sarma[1]

[1]CSIR-National Institute of Oceanography, Regional Centre, Visakhapatnam, 530017, India

Correspondence to: M. S. Krishna (moturi@nio.org)

Abstract. Rivers are an important source of dissolved inorganic carbon (DIC) to the adjacent coastal waters. In order to examine the spatial variability in the distribution and major sources of DIC in the Indian monsoonal ~~estuaries~~ rivers and to quantify their export flux to the north Indian Ocean, 27 major and medium ~~estuaries~~ rivers ~~along the Indian coast~~ were sampled during the discharge period. Significant spatial variability ~~in~~ of DIC concentrations (3.4 – 73.6 mg l^{-1}) ~~of DIC~~ was observed within the Indian ~~estuaries~~ rivers sampled. It is attributed to ~~(3.4 - 44.1mg l^{-1})~~ ~~due to~~spatial variations in the size of rivers, precipitation pattern, pollution, and lithology ~~in~~ of the catchments. ~~Dilution with high precipitation (2500±500 mm) and exchange with ground waters of low DIC resulted in very low concentrations of DIC in the estuaries located in the southwest of India (6.6±2.1 mg l^{-1}) than the estuaries located in the southeast (36.3±6.3 mg l^{-1}), northwest (30.3±8.9 mg l^{-1}) and northeast (19.5±6.2 mg l^{-1}) of India.~~ The stable isotopic composition of bulk DIC ($\delta^{13}C_{DIC}$) indicates that predominant contribution of DIC is ~~largely contributed by~~ from chemical weathering of ~~silicate and~~ carbonate and silicate minerals by soil CO_2. As the in-stream processes significantly alter the $\delta^{13}C_{DIC}$ in rivers, two different graphical mixing model techniques, Keeling plot and Miller-Tans plot, were used to approximate the $\delta^{13}C$ of DIC source. Least square linear regression models of both Keeling and Miller-Tans plots approximated similar $\delta^{13}C$ of DIC source (-2.0 and -3.0‰ respectively). Further, the $\delta^{13}C$ of CO_2 was approximated in order to filter the influence of pH and DIC speciation on the measured

bulk $\delta^{13}C_{DIC}$. Our results indicated that DIC in the Indian rivers is contributed by chemical weathering of carbonate minerals but largely influenced by autotrophic production in rivers from the southeast region and heterotrophic decomposition of organic matter in the other Indian rivers sampled. ~~The storage of water in dams/reservoirs and intrusion of marine waters appears to be responsible for the enriched $\delta^{13}C_{DIC}$ in the east flowing rivers.~~ It is estimated that the Indian monsoonal ~~estuaries~~ rivers annually export ~10.43 Tg of DIC to the northern Indian Ocean, of which the major fraction (74%) enters into the Bay of Bengal and the remaining reaches to the Arabian Sea. This is consistent with the freshwater flux which is three times higher in the Bay of Bengal (~378 km^3 yr^{-1}) than the Arabian Sea (122 km^3 yr^{-1}). Despite the discharge from Indian monsoonal rivers account for only 1.3% of the global freshwater discharge, they disproportionately export 2.5% of the total DIC export by the world major rivers. ~~and 9.4% of the Asian rivers to oceans. The yield of DIC (DIC export normalized by the catchment area of the river) was found to be higher in the SW estuaries (10.8±6.6 g m^{-2} yr^{-1}) than the SE (5.8±2.3g m^{-2} yr^{-1}), NE (8.6±5.7g m^{-2} yr^{-1}) and NW (9.5±4.0 g m^{-2} yr^{-1}) estuaries.~~ Despite ~~the~~ rivers from the SW region of India ~~estuaries~~ export an order of magnitude lower DIC (~~only~~ 0.3 Tg yr^{-1}) than the rivers from other regions of India, ~~of DIC, which is more than an order of magnitude lower than that of the export by the NE (4.2 Tg yr^{-1}), SE (3.5 Tg yr^{-1}) and NW (2.4 Tg yr^{-1}) estuaries~~., highest~~r~~ yield of DIC was found in the former and it is ~~from the SW estuaries is~~ attributed to intense precipitation (~3000 mm), favorable natural vegetation of tropical moist deciduous and tropical wet evergreen and semi evergreen forests, tropical wet climate, high soil organic carbon and the dominance of red loamy soils in catchments of the ~~SW~~ rivers from SW region. ~~This study, therefore~~Our study ,demonstrates ~~revealed~~ that significant spatial variability of the hydrological ~~(precipitation)~~, lithological ~~(bed rock and soils)~~ and environmental ~~(vegetation and~~

~~climate)~~ conditions in the catchments and in-stream processes (autotrophic production and heterotrophic decomposition of organic matter) strongly controls the ~~concentrations and yield of~~ DIC in~~from~~ the Indian monsoonal ~~estuaries~~rivers.

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

1. Introduction

Dissolved inorganic carbon (DIC) is one of the major constituent of carbon species in rivers. DIC in rivers mainly originates from the geogenic (chemical weathering of carbonate and silicate rocks) and biogenic (decomposition of organic matter in soils) sources as well (Meybeck, 1987; Mook and Tan, 1991; Gaillardet et al., 1999, Dessert et al., 2001; Viers et al., 2007; Raymond et al., 2008; Tamooh et al., 2013). The former consumes atmospheric carbon dioxide (CO_2) while the latter releases CO_2 fixed by the terrestrial plants. In addition to these major sources from the catchment, DIC is also contributed by various physical and biological processes within the rivers. For instance, heterotrophic decomposition of organic matter, photo-oxidation of dissolved organic carbon (DOC), autotrophic respiration and dissolution of atmospheric CO_2 contribute DIC to rivers. ~~However, DIC contribution from dissolution of atmospheric CO_2 in river is reportedly negligible ().~~ On the other hand, autotrophic production by aquatic plants (photosynthesis) and evasion of CO_2 to atmosphere with draw DIC from rivers. All these processes in the catchments and within the rivers as well are strongly coupled to atmospheric CO_2 as they act as either sink ~~(chemical weathering, photosynthesis and dissolution of CO_2)~~ or source ~~(decomposition of organic matter and autotrophic respiration)~~ of atmospheric CO_2 (e.g. Berner et al., 1983; Mook and Tan, 1991; Gaillardet et al., 1999; Raymond et al., 2008). ~~issolution of atmospheric CO_2also contributes to DIC in rivers~~ The DIC in rivers and its~~riverine~~

export ~~of~~ to the coastal oceans is thus intimately linked to the global carbon cycle (Campeau et al., 2017) ~~the hydrosphere. More than one third (and accounts for ~ 38%) of the total fluvial carbon transport to the global oceans (Meybeck, 1993; Cai, 2011; Jarvie et al., 2017) is in the form of DIC.~~

Export fluxes of DIC from rivers to ~~the~~ coastal regions of the world oceans have been estimated on the global (Gaillardet et al., 1999; Raymond et al., 2013) and regional scales (Richey et al., 2002; Wallin et al., 2013; Crawford et al., 2014; Campeau et al., 2014; Kokic et al., 2015) to understand the component of DIC in the global carbon budget. Annual export flux of DIC from the w~~W~~orld major river systems to the gloabal ocean ~~export annually~~has been estimated as ~ ~~330~~327 - ~~400~~ 385 Tg (1Tg=10^{12}g) ~~of DIC to the global oceans~~ (Ludwig et al., 1998; ~~Mackenzie et al., 2004; Lerman et al., 2007~~Meybeck and Vorosmarty, 1999). However, many of the regional studies on DIC export fluxes were limited to the major river systems only (e.g. Gaillardet et al., 1999; Raymond et al., 2013), for example, the Mississippi (Raymond and Cole, 2003; Raymond et al., 2008; Cai et al., 2008), Changjiang and Pearl (Cai et al., 2008) and Congo (Wang et al., 2013) rivers etc. Regional studies on the riverine export fluxes of DIC are very important for the global carbon cycle and budget as the export fluxes are largely dependent on the hydrological, lithological and environmental conditions, which are highly variable on regional scales. However, DIC measurements are still lacking in several medium rivers from different regions of the world in general and ~~the~~ Asia~~n rivers~~ in particular. ~~Chemical weathering of carbonate and silicate rocks and soils in the drainage basin are the major sources of DIC into rivers (Meybeck, 1987; Gaillardet et al., 1999, Dessert et al., 2001; Viers et al., 2007; Raymond et al., 2008; Tamooh et al., 2013). The DIC concentrations in rivers the estuaries are largely influenced by (i) the hydrological (precipitation and runoff), lithological (type and dominance of~~

~~rocks and soils) and environmental (temperature, climate and vegetation) conditions, (ii) anthropogenic activities (deforestation and land use change) in the catchment and (iii) physical and biological processes such as exchange with ground water (Finlay, 2003; Shin et al., 2011; Maher et al., 2013), atmosphere, autotrophic production and heterotrophic utilization of organic matter (McConnaughey et al., 1994; Abril et al., 2003; Finlay and Kendall, 2007, Hotchkiss et al., 2015; Zou, 2016) in rivers and estuaries. Weathering of carbonate and silicate rocks in the catchment, uptake of DIC by aquatic plants in rivers are the sinks for the atmospheric CO_2 (e.g. Berner et al., 1983; Raymond et al., 2008) while the oxidation of organic carbon is the source of CO_2 to the atmosphere. Due to human interferences, DIC fluxes from the world major rivers are found to increase dramatically in the last century (Cai, 2003; Raymond and Cole, 2003; Raymond et al., 2008; Ren et al., 2015). It has been noted that substantial alterations in the lateral transport of DIC from land to sea occurred after the industrialization (Regnier et al., 2013; Bauer et al., 2013). The increase in riverine DIC flux has a significant impact on the chemical composition (Williamson et al., 1994; Raymond and Cole, 2003; Findlay, 2010; Tank et al., 2010) and carbon budget in the coastal waters (Cole et al., 2007; Dhillon and Inamdar, 2013). The identification of major sources of DIC in the estuaries and quantification of their export fluxes to the coastal oceans are important in understanding the carbon cycling both in the regional as well global scales (Campeau et al., 2017).~~

Studies on the sources and export fluxes of DIC from the Indian rivers are very limited. Though DIC measurements were conducted in some Indian estuaries, for example, Mandovi and Zuari (Sarma et al., 2001), Godavari (Sarma et al., 2011), Cochin (Gupta et al., 2009; Bhavya et al., 2018), Hooghly (Mukhopadhyay et al., 2002; Samanta et al., 2015), Mahanadi (Pattanaik et al., 2017) and Chilka (Gupta et al., 2008; Muduli et al., 2013), they were confined only to the

internal cycling of DIC and exchange of CO_2 at the air-water interface, but not focused on sources and export fluxes of DIC. The major sources of DIC in the Indian rivers remain unclear, except only a couple of rivers, Krishna (Das et al., 2005; Laskar et al., 2014) and Ganges (Samanta et al., 2015), . Further, the quantity of annual DIC export by the Indian rivers to the coastal regions is unknown. Here, we made an attempt to understand the major sources of DIC in the Indian monsoonal rivers (Fig. 1) using $\delta^{13}C_{DIC}$ as a potential tracer, and to estimate the riverine export flux of DIC to the north Indian Ocean from the Indian subcontinent.

~~Fluvial carbon fluxes from rivers in the tropical region (30°N to 30°S) are critical for global carbon budgets because they contribute significant fraction to the global riverine DIC (48-64%) and freshwater discharge (66.2%) to the world oceans despite they occupy only ~43% of the world's land area (Huang et al., 2012). Furthermore, humid tropical climate over the tropical region supports the export of fluvial carbon from the continental land masses than the other climates in the world (Meybeck 1993; Ludwig et al., 1998). However, the fluvial DIC fluxes from rivers in the tropical region, except a few large river systems, to the global ocean are unknown due to the paucity of data.~~

~~Numerous studies have been documented on DIC export flux from the world major rivers, for example, the Mississippi (Raymond and Cole, 2003; Raymond et al., 2008; Cai et al., 2008), Changjiang and Pearl (Cai et al., 2008), Congo (Wang et al., 2013) and large river systems in the world (e.g. Gaillardet et al., 1999; Raymond et al., 2013). Though some measurements were carried out on DIC in the Indian estuaries, for example, Mandovi and Zuari (Sarma et al., 2001), Godavari (Sarma et al., 2011), Cochin (Gupta et al.,2009; Bhavya et al., 2018), Hooghly (Mukhopadhyay et al., 2002; Samanta et al., 2015), Mahanadi (Pattanaik et al., 2017) and Chila lake, a brackish water estuarine system (Gupta et al., 2008), the focus was mainly on internal~~

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~~cycling of carbon and exchange at the air-water interface. 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 reported but their sources were not evaluated.~~

The stable isotopic composition of DIC ($\delta^{13}C_{DIC}$) is widely used to identify the major sources of DIC in the aquatic systems (e.g. Singh et al., 2005; Tamooh et al., 2013; Samanta et al., 2015; Zou, 2016). ~~due to distinct isotopic composition of different sources (Deines et al., 1974).~~ The isotopic composition of DIC originated by dissolution of atmospheric CO_2 is about 0‰ (Coplen et al., 2002) whereas it is about -27 to -26‰ if the DIC is derived from oxidation of organic matter produced by C_3 plants (O'Leary, 1988). The $\delta^{13}C$ of DIC generated by ~~soil CO_2 dissolved~~ carbonic acid (formed by soil CO_2 dissolution) weathering of silicates is about -21 to -17‰ (Solomon and Cerling, 1987) while it is in the range of -10 to -9‰ for carbonate rocks because half of the carbon comes from carbonate rocks (0‰, Land, 1980) during weathering. The weathering of silicate and carbonate minerals yield $\delta^{13}C_{DIC}$ in the range of -8 to -7‰ and -4 to -3‰, respectively, if the carbonic acid formed by the dissolution of atmospheric CO_2. Though the isotopic composition of DIC derived from different sources is ~~distinctly different~~is well separable (Deines et al., 1974), the isotopic fractionation by ~~internal~~in-stream physical and biological processes alters the $\delta^{13}C_{DIC}$ of source material (Fig. 2). For example, photosynthesis and equilibration with atmospheric CO_2 enriches (O'Leary, 1988; Finlay, 2004; Parker et al., 2005, 2010) while the heterotrophic decomposition of organic matter and photo-oxidation of dissolved organic carbon depletes the $\delta^{13}C$ of DIC (Opsahl and Zepp, 2001; Finlay, 2003; Waldron et al., 2007; Vahatalo and Wetzel, 2008) (Fig. 2). Of these processes, equilibrium with atmospheric CO_2 is of less significance because rivers are generally in disequilibrium with atmospheric CO_2 (Raymond et al., 2013) and emit CO_2 to atmosphere due to oversaturation

(Oquist et al., 2009; Campeau et al., 2017). Nevertheless, the influence of internal processes within the rivers must be considered ~~in the~~while interpreting~~ation~~ the ~~of~~ $\delta^{13}C_{DIC}$ results for identification of DIC sources.

~~Despite distinct isotopic composition of DIC is expected for different sources, the identification of DIC sources is still challenging (Amiotte Suchet et al., 1999; Campeau et al., 2017) due to isotopic fractionations associated with complex mixture of sources and processes such as photosynthesis (O'Leary, 1988; Finlay, 2004; Parker et al., 2005, 2010), respiration (Finlay, 2003; Waldron et al., 2007), DOC photo oxidation (Opsahl and Zepp, 2001; Vahatalo and Wetzel, 2008), anaerobic metabolism (Waldron et al., 1999; Maher et al., 2015) and equilibration with atmospheric CO_2.~~

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

2. Study region, sampling and methods

2.1 Study Area

The Indian peninsula bifurcates the north Indian Ocean into the Bay of Bengal and the Arabian Sea. Although these two basins occupy the same latitudinal belt, their oceanographic

processes were reported to be remarkably different due to higher freshwater flux into the Bay of Bengal (1.63 x 10^{12} m^3 yr^{-1}) than to the Arabian Sea (0.3 x 10^{12} m^3 yr^{-1}; Subramanian, 1993; Gauns et al., 2005). The large freshwater influx leads to the formation of a strong vertical salinity stratification in the Bay of Bengal (Varkey et al., 1996) that prevents vertical mixing of nutrient rich sub-surface water with the surface (Prasanna Kumar et al., 2004). As a result, the Bay of Bengal is considered to be relatively less productive (Prasannakumar et al., 2002) than the adjacent Arabian Sea, which is one of the highly productive zones in the world (Madhupratap et al., 1996; Smith, 2001; Barber et al., 2001) due to injection of nutrients into the surface through the seasonal upwelling and convective mixing (Shetye et al., 1994; Madhupratap et al., 1996; Muraleedharan and Prasannakumar, 1996).

Discharge from the Indian monsoonal rivers is largely fed by the monsoon induced precipitation over the Indian subcontinent, which receives >80% of its annual rainfall during the southwest (SW) monsoon period (June-September) (Soman and Kumar, 1990). Though some amount of rainfall occurs during the northeast (NE) monsoon (December-March), it does not generate discharge as it will be stored within the dam reservoirs for domestic, industrial and irrigation purposes. Discharge from the Indian monsoonal rivers mainly occurs during the SW monsoon season (Vijith et al., 2009; Sridevi et al., 2015) hence, these rivers are called as monsoonal rivers. Since the major portion of the annual freshwater discharge ~~from Indian monsoonal rivers~~ occurs only during the SW monsoon, the entire estuary is filled with freshwater (Vijith et al., 2009; Sridevi et al., 2015) during this period. As discharge is small during the rest of the year, the discharge during the SW monsoon (wet period) is considered to be equivalent to the annual discharge of the monsoonal rivers. Based on rainfall intensity, forest cover, vegetation and soil type in the catchment, estuaries sampled in the present study were categorized into 4

groups, namely the ~~northeast~~ northwest (~~NE~~NW), ~~southeast~~ southwest (~~SE~~SW), ~~southwest~~ southeast (~~SW~~SE) and ~~northwest~~ northeast (~~NW~~NE) ~~estuaries~~ rivers of India (Fig. 1). The SW region of India is characterized by the intense rainfall during SW monsoon (~3000 mm) following the NE (1000-2500 mm), SE (300-500 mm) and NW (200-500 mm) regions of India (Soman and Kumar, 1990). The SW rivers drain red loamy soils while the NW rivers drain black soils. Except the major rivers Godavari and Krishna, all the rivers reaching Bay of Bengal (NE and SE ~~estuaries~~rivers) drain red loamy and alluvial soils in their upper and lower catchments respectively. The Godavari and Krishna rivers drain black soils in their upper catchment ~~along with~~whereas red loamy and alluvial soils in their middle and lower catchments respectively (Geological Survey of India; www.gsi.gov.in). Based on discharge, the monsoonal ~~estuaries~~ rivers in this study were divided into two types, namely, the major (>150 $m^3 s^{-1}$) and medium (<150 $m^3 s^{-1}$) ~~estuaries~~rivers.

2.2 Sample collection

Water samples were collected ~~within the~~ from the freshwater regions of the estuaries rather than from ~~mid or upstream rivers~~ head waters to obtain reliable export fluxes of DIC to the coastal ocean. Samples were collected at 2 to 3 locations to minimize the spatial variability within the freshwater zone of the estuary. Further, to minimize the inter-annual variability in DIC concentrations, sampling was conducted in two different years and the mean ~~is~~ was used for export flux estimations. ~~Each estuary was sampled at 3 to 5 locations between the upper (head) and lower (mouth) estuaries in order to minimize the spatial variability in DIC concentrations, and the mean concentrations are used for flux estimates.~~ Further, samples were collected in mid-stream of the ~~estuary~~ river using a local mechanized boat to avoid the contamination from river banks.

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

2.3. Methods

Temperature and salinity at the sampling locations were measured using a conductivity-temperature-density (CTD) profiling system (Sea Bird Electronics, SBE 19 plus, United States of America). Concentration of DO was determined by a Winkler's method (Carritt and Carpenter, 1966) using an auto titrator (Metrohm, Switzerland) with potentiometric end point detection. The analytical precision of the method was ±0.07% (RSD). Dissolved oxygen saturation is computed following formulations given by Garcia and Gordon (1992). DIC concentrations in water samples were measured at our Institute laboratory using Coulometer (UIC Inc., USA) connected to an automatic sub-sampling system. Based on the repeated analysis of samples and standards, the precision of the method was ±0.02 mg l^{-1}. The certified reference materials (CRM) supplied by Dr. A.G. Dickson, Scripps Institute of Oceanography, USA and internal standards were used to test the accuracy of our DIC measurements and it was found to be within ± 0.2 to 0.3%.. Potetiometric Gran titration method (Metrohm, Switzerland) was used for determination of pH and total alkalinity and followed the standard operating procedures given by Department of Energy (DOE) (1998).

The stable carbon isotopic composition of DIC in the water was measured on Gas Bench coupled with isotope ratio mass spectrometer (EA-IRMS-Delta V, Finnigan,Germany). 50 ml air-tight bottles with rubble septa were filled with 0.5 ml of high purity ortho-phosphoric acid and purged with high purity helium. About 1 ml of water sample is injected to the bottle and incubated at constant temperature of 50°C for 12 hours. The CO_2 extracted into the head space is injected to the IRMS through gas bench. The results are expressed relative to conventional standards, that is, pee dee belemnite (PDB) limestone for carbon (Coplen, 1996) as δ values, defined as:

$$\delta R = [(Xsample – Xstandard)/Xstandard) \times 10^3] \text{ ‰}$$

where R refers to ^{13}C and X stands for $^{13}C/^{12}C$. The high-purity tank of CO_2 was used as working standard for carbon. These gases were calibrated with IAEA standards. Standard deviation on 20 aliquots of the same sample was lower than 0.05‰ for $\delta^{13}C$. Chlorophyll-*a* (Chl-*a*) on the filter was extracted into di-methyl formamide (DMF) and measured the extract fluorometrically using a spectrofluorophotometer (Varian Eclipse, Varian Electronics., UK) following Suzuki and Ishimaru (1990). Annual mean discharge data of the rivers was taken from Meybeck and Ragu (1995, 1996), Central Water Commission, New Delhi (2006, 2012) and Kumar et al. (2005). Catchment area of the rivers was obtained from Water Resources Information System of India (WRIS, www.india-wris.nrsc.gov.in). Soil organic carbon data was taken from Kishwan et al. (2009) and Sreenivas et al. (2016), and the rainfall data was obtained from Soman and Kumar (1990). Dissolved organic carbon (DOC) data for the Indian ~~estuaries~~ rivers was taken from Krishna et al. (2015)

Total export flux of DIC from each river was estimated by multiplying the mean concentrations of DIC ~~in an estuary~~at near zero salinity (river end member) with the annual

discharge. Spatial variability of DIC concentrations ~~in estuaries~~within the river was minimized to a large extent by collecting samples from ~~head to mouth of the estuary~~2 to 3 locations in each river while the inter-annual variability by collecting samples during discharge periods of two years. However, variability in DIC concentrations within the discharge period results in some uncertainties in our estimations of DIC export fluxes. Time series measurements in the Godavari estuary (our unpublished results) revealed that the variability in DIC concentrations within the discharge period is up to 10%. Therefore, the error associated with our DIC flux estimates may be about 10%. DIC flux~~es~~ normalized by catchment area (yield) ~~were~~ was calculated by dividing the total DIC export flux of the river by its catchment area.

3. Results

3.1. Hydrographic characteristics

Surface water temperature~~s~~ ~~was~~ were higher in rivers from the NE and SE regions (mean 30.9±1.2°C) than in rivers from the SW and NW regions (27.3±1.5°C) of India~~estuaries located on the east coast (mean 30.9±1.2°C) than the west coast (27.3±1.5°C) of India. Salinity varied broadly from near zero (0.1) to 28.8 during the study period. Relatively higher salinities (>20) were recorded by the medium estuaries, which receives relatively lower freshwater discharge from the upstream river, for example, Nagavali (28.8), Vaigai (24.6) and Rushikulya (20.7). Mean salinities were lower in the west-flowing NW (0.1±0.02) and SW (2.1±2.8) estuaries than the east-flowing SE (9.5±7.8) and NE (8.5±11) estuaries.~~ Dissolved oxygen saturation varied from as low as 63% to as high as 105%, with a mean saturation of 90±11% in the ~~estuaries~~ rivers sampled. The rivers from the SW region of India ~~estuaries recorded slightly lower DO~~more unsaturation of DO (82±7%) than the rivers located in the NE (89±15%), NW (93±3%) and SE (96±11%) ~~estuaries~~regions of India. Chlorophyll-*a* (Chl-*a*) concentrations varied broadly from

0.8 to 7.5 mg m^{-3}, with relatively higher mean concentrations in the rivers of ~~-~~the SE region (4.7±2.5 mg m^{-3}) followed by the SW (2.8±0.7 mg m^{-3}) ~~estuaries~~regions of India. On the other hand, relatively low Chl-*a* was observed in the medium (2.6±1.3 mg m^{-3}) than in the major estuaries (3.2±2.1 mg m^{-3}).

3.2 DIC concentrations and $\delta^{13}C_{DIC}$

DIC concentrations in the Indian monsoonal ~~estuaries~~ rivers widely varied from 3.4 (Bharatha~~p~~puzha) to ~~44.1~~ 73.6 mg l^{-1} (Vellar), with a significant spatial variability (Fig. ~~2~~3a; Table 1). ~~More than five times~~ ~~h~~Highest~~r~~ mean DIC concentration~~s~~ ~~were~~ was observed in rivers of the SE region (~~36.3~~37.4±6.3 mg l^{-1}) while the lowest was found in the SW region (5.2±2.1 mg l^{-1}) of India. Intermediate values were found in rivers of the~~and~~ NW ~~estuaries~~ (~~30.3~~28.4±8.9 mg l^{-1}) ~~than in the SW (6.6±2.1 mg l^{-1})~~ and NE ~~estuaries~~ (~~19.5~~17.1±6.2 mg l^{-1}) regions of India. DIC concentrations were found to be similar in the major (22.7±13.6 mg l^{-1}) and medium (21.1±13.2 mg l^{-1}) ~~estuaries~~ rivers (homoscedastic Student's t-test; p=0.76). Mean DIC concentration found in this study (21.4±16.3 mg l^{-1}) is similar to those observed earlier in the major river systems ~~in~~of India (Brahmaputra; Singh et al., 2005) and elsewhere in the world, for example, British rivers (Jarvie et al., 2017) and Swedish rivers (Campeau et al., 2017) . However, DIC concentrations in the present study are higher than ~~those found in some of the Asian rivers of tropical region (12.7 mg l^{-1}; Huang et al., 2012) and~~ the global mean DIC (10.3 mg l^{-1}, Meybeck and Vorosmarty, 1999) (Table 1), but lower than those reported in the rivers draining into the Gulf of Trieste (N Adriatic; 37-66 mg l^{-1}, Tamse et al., 2014). ~~The $\delta^{13}C_{DIC}$ varied from -13.0 to 2.5~~ 1.4‰, ~~with a significant spatial variability (Fig. 3d) in the estuaries~~ rivers ~~sampled. Relatively depleted $\delta^{13}C_{DIC}$ values were observed in the west flowing estuaries~~rivers of the NW region ~~(-11.1±2.3‰) while enriched $\delta^{13}C_{DIC}$ and was found in rivers of~~ SW ~~(-7.4±1.9‰) than the east flowing estuaries of~~

~~NE (-3.5±2.8‰) and~~the SE ~~region (-2.76±5.2‰) regions of India. The $\delta^{13}C_{DIC}$ values found in this study are well within the range of values reported earlier in rivers of India (Das et al., 2005) and elsewhere in the world, for example, Swedish streams (-27.6 to -0.6‰; Campeau et al., 2017).~~

The $\delta^{13}C_{DIC}$ varied from -13.0 to -1.4‰, with a significant spatial variability (Fig. 3d; Table 1) in the rivers sampled. Relatively depleted $\delta^{13}C_{DIC}$ values were observed in the rivers of the NW region (-11.1±2.3‰) while enriched $\delta^{13}C_{DIC}$ was found in rivers of the SE region (-3.5±2.3‰) of India (Fig. 3d). The $\delta^{13}C_{DIC}$ values found in this study are well within the range of values reported earlier in rivers of India (Das et al., 2005) and elsewhere in the world, for example, Swedish streams (-27.6 to -0.6‰; Campeau et al., 2017) and rivers from Italy and Slovenia (-12.8 to -7.7‰, Tamse et al., 2014).

3.3. Export fluxes and yield of DIC

Annual export flux of DIC to the coastal ocean from the individual ~~estuaries~~ rivers varied broadly from 0.01 Tg (Chalakudi) to as high as 2.33 Tg (Krishna) (Fig. 3b; Table 1). Among the rivers sampled, ~~The~~ rivers of the NE region of India ~~estuaries~~ export higher DIC ~~flux~~ (~~4.2~~6.52 Tg yr^{-1}) while the lowest was found from rivers of the SW region of India (0.24 Tg yr^{-1}) (Table 1). ~~followed by the SE (3.5 Tg yr^{-1}) and NW estuaries (2.4 Tg yr^{-1}). In contrast, the SW estuaries recorded the lowest export flux of 0.3 Tg yr^{-1} which is an order of magnitude lower than that of the export flux by other estuaries (Fig. 2).~~ The Indian monsoonal ~~estuaries~~ rivers together export about 10.~~4~~32 Tg yr^{-1} of DIC to the northern Indian Ocean, of which 7.~~7~~81 Tg (~~74~~75%) enters into the Bay of Bengal and the remaining into the Arabian Sea (2.~~7~~51 Tg). ~~The estuaries~~

~~Krishna (2.3 Tg yr^{-1}), Godavari (1.5 Tg yr^{-1}) and Haldia (1.2 Tg yr^{-1}) together responsible for the transport of 65% of total riverine DIC export to the Bay of Bengal.~~ The yield of DIC ranged from 2.~~7~~ 8 (Bharatha~~p~~puzha) to ~~21.6~~20.7 g m^{-2} yr^{-1} (~~Mandovi~~Baitarani) ~~(Fig.~~ 3c; Table 1), excluding the exceptionally high yield of ~~113~~119~~.4~~ g m^{-2} yr^{-1} from Haldia ~~estuary~~river. The mean yield was found to be more or less similar in rivers from all the four regions of India, i.e, NW (8.4 g m^{-2} yr^{-1}), SW (8.8 g m^{-2} yr^{-1}), SE (6.6 g m^{-2} yr^{-1}) and SE (7.7 g m^{-2} yr^{-1}) regions. ~~The west flowing rivers to the Arabian Sea are characterized by relatively higher yield of DIC (mean 10.4±5.6 g m^{-2} yr^{-1}) than the east flowing rivers to the Bay of Bengal (7.3±4.6 g m^{-2} yr^{-1}).~~ Despite the export flux of DIC is lowest from rivers of the SW region (0.24 Tg yr^{-1}), interestingly, the yield from rivers of this region is on par (even slightly higher) with the other Indian monsoonal rivers (Table 1; Fig. 3b&c). The yields of DIC found in this study are similar to those observed in rivers elsewhere in the world (Huang et al., 2012). ~~Among the estuaries sampled, the SW and SE estuaries recorded higher (10.8±6.6 g m^{-2} yr^{-1}) and lower (5.8±2.3 g m^{-2} yr^{-1}) yields of DIC respectively whereas intermediate values were noticed in the NW (9.5±4.0 g m^{-2} yr^{-1}) and NE (8.6±5.7g m^{-2} yr^{-1}) estuaries.~~

~~*3.4 Stable C isotopes of DIC ($\delta^{13}C_{DIC}$)*~~

~~The $\delta^{13}C_{DIC}$ varied from -13.0 to -1.4‰, with a significant spatial variability (Fig. 3d; Table 1) in the rivers sampled. Relatively depleted $\delta^{13}C_{DIC}$ values were observed in the rivers of the NW region (-11.1±2.3‰) while enriched $\delta^{13}C_{DIC}$ was found in rivers of the SE region (-2.6±5.2‰) of India (Fig. 3d). The $\delta^{13}C_{DIC}$ values found in this study are well within the range of values reported earlier in rivers of India (Das et al., 2005) and elsewhere in the world, for example, Swedish streams (-27.6 to -0.6‰; Campeau et al., 2017) and rivers from Italy and Slovenia (-12.8 to -7.7‰, Tamse et al., 2014).~~

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4. Discussion

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

4.1 Distribution ~~and sources~~ of DIC in the Indian monsoonal ~~estuaries~~rivers

~~Mean DIC concentration found in this study (21.9±13.2 mg l^{-1}) is similar to those observed earlier in the Indian estuaries such as Ganga Brahmaputra and Hooghly (Singh et al., 2005, Samanta et al., 2015), and in estuaries elsewhere in the world, for example York, Yangtze, Seri and Xi etc (Raymond and Bauer, 2000, Cai et al., 2008, Ishikawa et al., 2015; Zou, 2016) (Table 1). The DIC concentrations in the Indian estuaries are higher than those found in some of the Asian rivers of tropical region (12.7 mg l^{-1}, Huang et al., 2012) and the global mean (10.3 mg l^{-1}, Meybeck and Vorosmarty, 1999) (Table 1), but lower than those reported in the rivers draining into the Gulf of Trieste (N Adriatic; 37-66 mg l^{-1}, Tamse et al., 2014) (Table 1).~~ Distribution of DIC ~~concentrations~~ in the Indian monsoonal rivers showed large spatial

variability, with the lowest values in rivers from the SW region of India (Fig. 3a). DIC concentrations in rivers are known to be influenced by the intensity of precipitation over the catchment, basin lithology, length of the fluvial network (Hotchkiss et al., 2015) and in-stream physical and biological processes (Mook and Tan, 1991; Raymond et al., 2008). The spatial distribution of rainfall over the Indian subcontinent (www.imd.gov.in) shows that the SW region receives the highest annual rainfall (~~2500±500~~~3000 mm) than the rest of the India (Soman and Kumar, 1990). ~~Among the estuaries sampled along the Indian coast, the SW estuaries are characterized by significantly lower mean concentrations of DIC (6.6±2.1 mg l^{-1}) than the SE (36.3±6.3 mg l^{-1}), NE (19.5±6.2 mg l^{-1}) and NW (30.3±8.9 mg l^{-1}) estuaries (Table 2). This could be due to considerable spatial variations in the (i) hydrological, lithological and environmental conditions in the catchments and (ii) in-stream physical and biogeochemical processes.~~

~~*4.1.1. The impact of hydrological conditions*~~

~~The SW region of India receives the highest amount of precipitation during the SW monsoon (2500±500 mm) than the SE (400±50 mm), NE (1000±200 mm) and NW (750±250 mm) regions of India (Table 2) (Soman and Kumar, 1990).~~ The intense precipitation ~~in~~ over the SW region is expected to cause higher weathering rates and ~~therefore~~ thus higher DIC in rivers (e.g., Gupta et al., 2011), but lower DIC concentrations were found in rivers ~~from~~of this (~~the~~ SW) region.~~ estuaries~~. ~~This is attributed~~It could be due to the influence of dilution because the dense precipitation over the small catchment area of the rivers from this region (Table 1) might have diluted DIC concentrations in rivers of this region. In order to understand the influence of the density of rainfall on DIC in rivers, we normalized the volume of discharge from the river

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with its catchment area. The catchment area ~~-~~normalized volume of discharge was found to be much higher in ~~the SW estuaries~~ rivers from the SW region ($1.71\ m^3\ m^{-2}$) than ~~in~~ the rivers from~~the~~ SE ($0.17\ m^3\ m^{-2}$), NE ($0.6\ m^3\ m^{-2}$) and NW ($0.32\ m^3\ m^{-2}$) ~~estuaries~~regions of India. About three times higher catchment area normalized discharge might have diluted DIC concentrations in the ~~SW estuaries~~rivers of the former region. A strong ~~negative correlation~~exponential decrease in DIC concentrations with increasing ~~the~~ rainfall over the catchment ~~between precipitation in the catchment and DIC concentration in estuaries~~ (r^2= ~~-~~ 0.~~89~~72, p<0.001; Fig. 4a) also suggests that DIC concentration in Indian ~~estuaries~~ rivers are ~~rather controlled~~strongly influenced by the ~~intensity~~ density of precipitation over the catchment. ~~Dilution of DIC by heavy precipitation in the SW region can also be seen from relatively depleted $\delta^{13}C_{DIC}$ values (-7.4±1.9‰) in the SW estuaries because the shorter residence time of soil water depletes the $\delta^{13}C_{DIC}$ due to preferential dissolution of $^{12}CO_2$ over $^{13}CO_2$ (Amiotte-Suchet et al., 1999).~~

~~Since many of the hydrological processes are largely dependent on the size of the river and its catchment area, these two factors may govern the concentrations of DIC in estuaries. The lower concentrations of DIC in the SW estuaries may possibly due to smaller catchment area as~~~~The~~ ~~r~~Rivers ~~from~~ of the SW ~~rivers~~ region are relatively small in size, both in terms of ~~discharge (46 $km^3\ yr^{-1}$) and~~ catchment area (total catchment area: ~~0.02~~20x10^3 ~~M~~ km^2) and the length of the river (mean length: 126 km), than the rivers from other regions (~~that of~~ SE, NE and NW) of India (Table 1) ~~rivers (Table 2)~~. Since the contribution of DIC from in-stream processes, such as decomposition of organic matter, has been demonstrated to increase along the course of the fluvial network (Hotchkiss et al., 2015), possibly due to increase in the residence time of water (Catalan et al., 2016), the lowest DIC concentrations found in rivers from the SW region

may also, at least partly, be due to their small size. Fairly good positive correlation between DIC concentrations and length of the rivers (r^2=0.38, p<0.01; Fig. 4~~e~~b) also support this argument. ~~The concentrations of DIC in the Indian estuaries showed a significant positive relationship with catchment area (r^2=0.76; p<0.001; Fig. 4b) and a negative relationship with volume of discharge (r^2= 0.57; p<0.001; Fig. 4c) only in the medium estuaries (discharge: <150 m^3s^{-1}), suggesting that an area of catchment and magnitude of discharge controls DIC concentrations largely in the medium estuaries rather than in the major estuaries. It could be due to the influence of in-stream processes as the major rivers are long compared to the medium rivers.~~

~~Mixing with seawater and exchange of submarine ground water also influence DIC concentrations in the estuaries. Since this study was conducted during the SW monsoon, many of the estuaries are filled with freshwater (salinity >1) due to maximum discharge during this period. On the other hand, higher salinities (>20) were observed in some medium estuaries, namely, Rushikulya, Nagavali and Vaigai recorded higher salinities (>20) due to low flow from upstream river. A strong positive correlation was found between $\delta^{13}C_{DIC}$ and salinity (Fig. 4d; r^2=0.71, p<0.001), suggesting that DIC in the Indian estuaries is also influenced by the intrusion of marine waters particularly in medium estuaries. The $\delta^{13}C_{DIC}$ values were found to be >0‰ in Rushikulya, Nagavali and Vaigai estuaries (0.1, 0.7 and 2.5‰ respectively) suggesting that major contribution of DIC in these estuaries is from intrusion of marine water.~~

~~As found in many estuaries over the world, submarine groundwater discharge is found to contribute up to 52% of DIC in the Godavari estuary (Rengarajan and Sarma, 2015) due to higher concentrations of DIC by 3 to 4 times in in the ground water than estuary. The measured DIC concentrations in ground waters along the entire Indian coast suggest relatively lower concentrations in the SW (mean 32±19 mg l^{-1}) than the SE, NE and NW regions of India~~

~~(Table 2) during discharge period (Dr. BSK Kumar, personal communication). Exchange of SW estuaries with ground water with relatively lower DIC concentrations might have possibly yielded low DIC concentrations. Nevertheless it is difficult to ascertain the impact of ground water exchange yielded low DIC in the SW estuaries due to lack of submarine ground water discharge rates.~~

~~*4.1.2. The impact of in stream processes*~~

The major physical and biological processes ~~within the rivers that~~ controlling the DIC concentrations in rivers are the exchange with atmospheric CO_2, autotrophic removal and heterotrophic addition of DIC. Since the Indian monsoonal estuaries have been reported to be a source of CO_2 to the atmosphere during the discharge period due to heterotrophic decomposition of organic matter (Sarma et al., 2001, 2011, 2012; Gupta et al., 2008, 2009; Bhavya et al., 2018), the DIC input from the dissolution of atmospheric CO_2 ~~can be ruled out~~is ~~thus~~may be unlikely. On the other hand, ~~CO_2 release due to heterotrophic decomposition of~~ organic matter decomposition is expected to add~~s~~ significant amount of DIC ~~to the Indian estuaries during this period~~ as enhanced bacterial respiration rates were reported ~~in the Indian estuaries~~ during the study period ~~(~~Sarma et al., 2011; 2012). In contrast, significant negative correlation between chlorophyll-*a* and DIC (r^2=-0.44, $p<0.01$; Fig. 4c), except few SE ~~estuaries~~rivers where elevated phytoplankton biomass (Chl-a: >5 mg m^{-3}) was recorded, suggesting that autotrophic removal of DIC is also significant in the Indian monsoonal rivers during the study period. A significant positive relationship was observed between the $\delta^{13}C_{DIC}$ and Chl-a (r^2=0.~~50~~49; $p<0.01$; Fig. 4d), supporting this argument because preferential uptake of ^{12}C than ^{13}C during photosynthesis leaves the residual DIC enriched in ^{13}C. On the other hand, $\delta^{13}C_{DIC}$ showed significant positive correlation with DO saturation (r^2=0.50, $p<0.01$; Fig. 4e)~~,~~ (depleted $\delta^{13}C_{DIC}$ values at more under

saturation of DO) and DOC concentrations (r^2=0.43, p<0.01; Fig. 4f) as was observed in the Xi river (Zou et al., 2016). Altogether, enriched $\delta^{13}C_{DIC}$ are associated with higher DOC, less under saturation of DO and higher phytoplankton biomass (Chl-a) while the depleted $\delta^{13}C_{DIC}$ are associated more under saturation of DO and less DOC. This suggests that both autotrophic removal and heterotrophic addition control DIC in the Indian rivers during the discharge period, with a considerable ~~regional~~spatial variability. However, influence of these processes on DIC concentrations is difficult to separate with this bulk $\delta^{13}C_{DIC}$ data set, as the $\delta^{13}C_{DIC}$ in rivers is also influenced by pollution, catchment lithology and outgassing of CO_2 (Shin et al., 2011; Brunet et al., 2005; Bouvillion et al., 2009; Zeng et al., 2011; Tamooh et al., 2013). Excluding Sabarmati and Mahisagar rivers, DIC concentrations showed fairly good linear relationship with population density over the catchment of the river, suggesting that considerable influence of pollution from the mega cities and industries on DIC in the Indian rivers. ~~A fairly good positive correlation between DIC and DOC concentrations (r^2=0.34, p<0.01; Fig. 4e), except few medium estuaries, suggests that DIC addition through microbial degradation of organic matter seems to be possible source in the Indian estuaries. A positive correlation between $\delta^{13}C_{DIC}$ and DOC was observed, with different slope for NW estuaries (r^2=0.43, p<0.01; Fig. 4f), confirming that oxidation of organic matter may be one of the major DIC sources in the Indian monsoonal estuaries. Similar relationship was also observed in the Xi river (Zou et al., 2016). The range of $\delta^{13}C_{DIC}$ (-13.0 to 2.5‰) in the Indian monsoonal estuaries is distinctly enriched than that of the $\delta^{13}C$ of DIC derived from decomposition of terrestrial C_3 plant derived organic matter (-27 to -26‰, O'Leary, 1988; Fig. 5), suggesting that DIC might have been contributed from decomposition of terrestrial C_4 plants (-17 to -13‰, Krishna et al., 2015) and weathering of silicate and carbonate rocks. In addition, if weathering occurs due to dissolution of silicate and~~

carbonate rocks due to atmospheric CO_2, the $\delta^{13}C_{DIC}$ yields -8 to -7‰ and -4 to -3‰ respectively. On the other hand, the $\delta^{13}C_{DIC}$ would be -10 to -9‰ and -21 to -17‰ (Solomon and Cerling, 1987) if the dissolution of silicate and carbonate rocks occurs due to soil CO_2 respectively. As discussed above, flux of CO_2 from atmosphere to river cannot be expected due to super saturation of riverine CO_2, weathering of silicate and carbonate rocks by dissolution of soil CO_2 may be possible. Though isotopic composition of $\delta^{13}C_{DIC}$ derived from decomposition of C_4 plants and weathering due to soil CO_2 are similar and difficult to separate, Sarma et al. (2014) measured isotopic composition of $\delta^{13}C_{POC}$ and found that >90% of the POC is contributed by C_3 plants. Hence possible contribution of DIC through decomposition of C_4 plants may be negated.

Significant negative correlation between chlorophyll *a* and DIC (r^2= 0.44, $p<0.01$; Fig. 6a), except few SE estuaries where elevated phytoplankton biomass (Chl a: >5 mg m^{-3}) was recorded, suggesting that autotrophic removal of DIC may be possible sink in the Indian monsoonal estuaries during the study period. This process would enrich $\delta^{13}C_{DIC}$ of residual DIC due to preferential removal of $^{12}CO_2$ over $^{13}CO_2$ during photosynthesis. A positive relationship was observed between $\delta^{13}C_{DIC}$ and Chl a in the Indian estuaries (r^2=0.50; $p<0.01$), suggesting that biological removal of DIC enriched $\delta^{13}C_{DIC}$. In contrast, heterotrophic decomposition of organic matter (respiration) depletes $\delta^{13}C_{DIC}$ due to release of $^{12}CO_2$ over $^{13}CO_2$ during this process. Due to lack of respiration rates data, we could not able to evaluate its influence. Nevertheless, the dissolved oxygen saturation stores the net effect of biological production and heterotrophic respiration. In order to confirm the net biological influence on $\delta^{13}C_{DIC}$, the same is correlated with DO saturation and found significant positive correlation (r^2=0.50, $p<0.01$; Fig. 6b), (depleted $\delta^{13}C_{DIC}$ values at low DO saturation), except NW estuaries, which recorded

~~depleted $\delta^{13}C_{DIC}$ (<-10.0‰) confirming that biological processes enriched $\delta^{13}C_{DIC}$ in the Indian monsoonal estuaries. CO_2 out gassing due to heterotrophic decomposition of organic matter and equilibrium with atmospheric CO_2 results in the enrichment of $\delta^{13}C_{DIC}$ in reservoirs/dams and stored water bodies (Shin et al., 2011; Brunet et al., 2005; Bouvillion et al., 2009; Zeng et al., 2011; Tamooh et al., 2013). As many of the east flowing river (e.g. Godavari, Krishna and Cauvery etc) were dammed at many locations for domestic, industrial and irrigation purposes., relatively enriched $\delta^{13}C_{DIC}$ in these estuaries might have been influenced by storage of water besides sources of DIC. A significant positive correlation between DIC concentrations and $\delta^{13}C_{DIC}$ (r^2=0.76; p<0.001; Fig. 6c), excluding the positive $\delta^{13}C_{DIC}$ values, indicate that significant contribution of DIC is from oxidation of organic carbon in dams/reservoirs or stored water bodies. Therefore, DIC in the Indian estuaries are contributed by weathering of silicate and carbonate rocks due to soil CO_2, biological production, organic matter decomposition and exchange of CO_2 to the atmosphere.~~

~~*4.1.3. The impact of catchment lithology*~~

~~Spatial distribution of bedrocks and soils over the Indian subcontinent shows that Narmada and Tapti rivers located in the NW region of India and upper reaches of Godavari and Krishna rivers drain over the igneous rocks (Deccan traps) while the other rivers flow over the metamorphic rocks (Pre Cambrian), which are the predominant rock type in south India. However, Haldia and lower reaches of the SE rivers drain over the sedimentary rocks (Geological Survey of India, https://www.gsi.gov.in). Though higher chemical weathering rates were reported in the Deccan Trap basalts (Das et al., 2005; Singh et al., 2005), higher DIC concentrations were also observed in estuaries draining over the metamorphic rocks, suggesting that other factors may also be governing the concentrations of DIC, than the bedrocks in the~~

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~~catchment. The broad range of $\delta^{13}C_{DIC}$ found in this study (-13.0 to 2.5‰) also indicates that DIC contribution is from variable sources such as weathering of carbonate and silicate rocks by carbonic acid derived from dissolution of soil CO_2 (-10 to -9‰ and -21 to -17‰ respectively, Solomon and Cerling, 1987), decomposition of organic matter and marine water (0 to 2‰) (Fig. 5).~~

Spatial distribution of soils shows that rivers of the NW region of India and upper reaches of Krishna and Godavari rivers drain the lime-rich black soils (Fig. 1) while rivers from the SW region drain red loamy soils. Whereas, the east-flowing rivers drain the lime-poor red sandy soils in the upper but lime-rich alluvial soils in the lower reaches (Fig.1). L~~l~~ateritic soils, which are poor in lime and silicate, occupied the catchment of the rivers in the SW region of India. ~~rivers.~~ Relatively lower c~~C~~hemical weathering rates ~~are relatively lower in~~of the lateritic than the non-lateritic soils ~~and the consumption of atmospheric/soil CO_2 through silicate weathering is lower by ~2 times in the former than the latter (Boeglin and Probst, 1998)~~could be one of the reasons for the observed lower DIC concentration the rivers from SW region of India. A significant positive correlation was found between total alkalinity (TA) and $\delta^{13}C_{DIC}$ (r^2=0.52; $p<0.01$; Fig. 4~~gh~~), suggesting that significant contribution of DIC is from weathering of carbonate minerals in the catchment. Though the higher chemical weathering rates were reported for the Deccan Trap basalts (Das et al., 2005; Singh et al., 2005) which occupied the catchments of rivers of the NW region of India and upper reaches of Godavari and Krishna, higher DIC concentrations were also observed in rivers draining over the metamorphic rocks. This suggests that the influences of factors other than bedrock are also significant on the concentrations of DIC in the Indian rivers. ~~The upper reaches of the east flowing rivers (NE and SE) drain over the lime-poor red and yellow soils, while lower reaches drain predominantly the lime rich alluvial soils. Upper reaches~~

~~of Krishna and Godavari also drain over the lime-rich black soils. The dominance of lateritic soils, which are relatively less susceptible to chemical weathering than the non lateritic soils in the catchments of the SW rivers could be possible reason for lower DIC concentrations in SW estuaries. The enriched $\delta^{13}C_{DIC}$ in the SW estuaries (-7.4±1.9‰) may also be due to less contribution of DIC from lateritic soils as these soils are poor in lime (~10 to ~9‰) and silicate (~21 to ~17‰) and less susceptible to chemical weathering rates.~~

4.2 Major sources of DIC in the Indian ~~M~~monsoonal rivers

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~~The range of $\delta^{13}C_{DIC}$ (-13.0 to -1.4‰) in the Indian monsoonal rivers is distinctly different from that of the $\delta^{13}C$ of DIC derived from dissolution of soil CO_2 produced by the decomposition of terrestrial C_3 plant organic matter (~ 22‰, O'Leary, 1988; Campeau et al., 2017; Fig. 5), suggesting that contribution of DIC from dissolution of soil CO_2 is of less significance. However, the observed range of $\delta^{13}C_{DIC}$ is close to the range of $\delta^{13}C_{DIC}$ derived from weathering of silicate and carbonate rocks by atmospheric CO_2 (~8 to ~7‰ and ~4 to ~3‰ respectively) and soil CO_2 (~10 to ~9‰ and ~21 to ~17‰, repectively) (Solomon and Cerling, 1987), suggesting the major fraction of DIC is of geogenic origin in general and weathering of carbonate mineral in particular.~~

Though, the $\delta^{13}C_{DIC}$ is a promising tool to decipher the sources of DIC, its interpretation for source material identification in rivers is still challenging due to the influence of multiple physical and biological processes within the rivers, which significantly alter the $\delta^{13}C_{DIC}$ of source. The influence of major in-stream processes on the $\delta^{13}C_{DIC}$ must be separated before interpreting the $\delta^{13}C_{DIC}$ results for interpretation of major sources of DIC, failing which leads to erroneous conclusions. In order to identify and separate DIC sources, we used here two

~~independent~~different graphical mixing model techniques, Keeling plot (Keeling, 1958; Pataki et al., 2003) and Miller-Tans plots (Miller and Tans, 2003). These models approximate the hypothetical $\delta^{13}C$ of source material as an intercept (in Keeling plot) and slope (in Miller-Tans plot) of the least square linear regression equations (Pataki et al., 2003; Campeau et al., 2017). The deviations from the approximated $\delta^{13}C$ of source ~~material~~ can be interpreted to the influence of in-stream processes. Further, we approximated the $\delta^{13}C$ of CO_2 using a set of enrichment factors of isotopic fractionation across the carbonate species (Zhang et al., 1995) in order to filter the impact of DIC speciation and pH on the bulk $\delta^{13}C_{DIC}$ values. This approach has already been used by Quay et al. (1992), Mayorga et al. (2005) and recently by Campeau et al. (2017).

Significant negative relationships were observed in both Keeling plot ($\delta^{13}C_{DIC}$ as a function of 1/DIC; Fig. 5a) and Miller-Tans plot ($\delta^{13}C_{DIC}$ x DIC as a function of DIC; Fig. 5b) (r^2=0.61, p<0.01 and r^2=0.72, p<0.01 respectively) of DIC in the Indian rivers, except the rivers draining the Deccan Trap basalts. Both graphical mixing models, Keeling and Muller-Tans plots, approximated the similar $\delta^{13}C$ of source material (-3.0‰ and -2.0‰ respectively; Fig. 5a&b~~-d~~), suggesting that weathering of carbonate minerals is the predominant source of DIC in the Indian monsoonal rivers rather than biogenic soil CO_2. Calculated $\delta^{13}C$ of CO_2 ranged from -21.5 to 9.6‰ in the Indian rivers with a mean value of -13.0±2.7‰. Calculated $\delta^{13}C$ of CO_2 is linearly correlated with measured bulk $\delta^{13}C_{DIC,}$ but correlation coefficient (r^2) is only 0.51 (Fig. 5c), suggesting that significant spatial variability in the influence of in-stream processes on $\delta^{13}C_{DIC.}$ The Miller-Tans plot of CO_2 ($\delta^{13}C$-CO_2 x CO_2 as a function of CO_2) showed highly significant ~~(r^2=0.97; p<0.001)~~ linear regression model with a slope of -10.7‰ ~~(~~(r^2=0.97; p<0.001; Fig. 5~~c~~d). These results indicated that DIC in the Indian rivers is largely contributed by chemical weathering of carbonate and silicate minerals by soil CO_2 (-10 to -9‰). D~~. However, d~~eviations

of the ~~bulk~~measured $\delta^{13}C_{DIC}$ ~~observed in the rivers~~ (-13.0 to -1.4‰) from that of the approximated $\delta^{13}C$ of DIC source (-3.0 to -2.0‰) and $\delta^{13}C$ of CO_2 (-10.7‰) could be due to the influence of in-stream process. In more than 70% of the Indian rivers sampld, ~~Since~~ the deviation from the $\delta^{13}C$ of DIC source is towards negative side (depletion) ~~in many rivers~~ ($\delta^{13}C_{DIC}$ < -3.0‰), suggesting that h~~, H~~eterotrophic decomposition of organic matter is the dominant process that controls DIC in these rivers~~ in 70% of the Indian rivers sampled because $\delta^{13}C_{DIC}$ in these rivers is depleted than the approximated $\delta^{13}C$ of DIC source~~. While, no (or very little) deviation was observed only in~~ rivers from the SE region of India (mean $\delta^{13}C_{DIC}$: -3.1‰) could be due to the competition between autotrophy, degassing and heterotrophy as these ~~two~~ processes influences the $\delta^{13}C_{DIC}$ in opposite directions (Fig. 2); the former two processes causes enrichment while the latter depletes $\delta^{13}C_{DIC}$. ~~It is consistent with~~Relatively higher phytoplankton biomass (mean Chl-a: 4.6 mg m^{-3}) and ~~near~~less unsaturation of DO (98.7%) was observed in these rivers compared to the mean of the rest of the Indian rivers ~~from other region of India~~ (2.4 mg m^{-3} and 87.5% respectively), suggesting that autotrophy is one of the dominant processes controlling the DIC in rivers from the SE region of India. Total number of dams on the rivers from this (SE) region (mean 155, Table 1) is not significantly higher from that of the mean of total number of dams on the Indian rivers sampled (mean 135) , suggesting that degassing due to storage of water may not be the dominant process responsible for enrichment in $\delta^{13}C_{DIC}$ of these rivers.

4.~~2~~ 3 Total DIC export by the Indian monsoonal rivers to the north Indian Ocean

Indian monsoonal rivers annually export ~10.~~4~~ 3 Tg of DIC to the north Indian Ocean. Nearly three fourth of this amount (7.~~7~~ 8 Tg) reaches to the Bay of Bengal while the remaining

into the Arabian Sea. This is consistent with the higher magnitude of freshwater discharge to the Bay of Bengal (378 km^3 yr^{-1}) ~~-~~from the catchment area of about ~~0.96~~970x10^3 ~~M~~ km^2 than the Arabian Sea (122 km^3 yr^{-1} from the catchment area of ~~0.23~~244x10^3 ~~M~~ km^2). The total DIC export by the Indian monsoonal ~~estuaries~~ rivers (10.~~3~~4 Tg yr^{-1}) is only 2.5% of the total DIC export by the world major rivers (400 Tg yr^{-1}). ~~, and 9.4% of the export by the Asian rivers (111 Tg yr^{-1}; Huang et al., 2012).~~ The DIC export from the Indian ~~estuaries~~ monsoonal rivers is far less than the DIC export by the American (61.4 Tg yr^{-1}) and African (17.7 Tg yr^{-1}) rivers and major rivers draining to the tropical Atlantic from South America and Africa (53 Tg yr^{-1}, Araujo et al. 2014). It is mainly due to the fact that freshwater discharge from the Indian monsoonal rivers is very low (~500 km^3 yr^{-1}) compared to the American (11,799 km^3 yr^{-1}) and African (3,786 km^3 yr^{-1}) rivers. However, the Indian monsoonal rivers are exporting disproportionately higher DIC ~~to the north Indian Ocean~~ because they account for only 1.3% of the global river discharge but export 2.5% of the global riverine DIC export to the oceans. Though American and African rivers account for 30% and 10% of the global river discharge, they export only 15% and 4.4% of global riverine DIC to oceans, respectively. Disproportinately higher DIC flux from the Indian rivers could be due to relativley higher weathering rates of silicate and carbonate minerals in their drainage basins (Das et al., 2005; Gurumurty et al., 2012; Pattanaik et al., 2013). Higher DIC fluxes from the tropical regions are mainly attributed to the favourable climatic conditions, lithology and land use cover (Huang et al., 2012) ~~in~~ of this region for higher dissolution. ~~Relativley as higher weathering rates of silicate and carbonate minerals were reported in the drainage basins of the Indian rivers (Das et al., 2005; Gurumurty et al., 2012; Pattanaik et al., 2013)~~

Krishna et al. (2015) reported that Indian monsoonal ~~estuaries~~ rivers export 2.32 Tg yr^{-1} of dissolved organic carbon (DOC) to the north Indian Ocean. The total fluvial dissolved carbon flux (DIC+DOC) would be 12.7 Tg yr^{-1} in which DIC flux contributed up to ~81%. The predominance of DIC has also been found in rivers ~~and it is consistent with earlier reports~~ elsewhere in the world, for example, the British rivers (~~80%,~~ Jarvie et al., 2017) and high altitude Swedish rivers (Campeau et al., 2017). Since the catchment area of the Indian monsoonal rivers ranged widely from as low as ~~0.00~~1x10^3 ~~M~~ km^2 to as high as ~~0.313~~313x10^3 ~~M~~ km^2, the export fluxes of DIC were normalized with the catchment area of the river (yield) in order to examine various factors controlling the ~~lateral~~ DIC export to the north Indian Ocean.

4.3 Yield of DIC from the Indian monsoonal rivers

The yield ~~(export flux normalized by catchment area)~~ of DIC found in this study (mean 8.7±5.2 g m^{-2} yr^{-1}) is ~~similar~~close to those found ~~earlier~~ in ~~the~~ rivers from the tropical region of ~~the~~ Asia~~n continent~~, but significantly higher than those reported from tropical region of the American and African continents ~~(Table 3)~~ (Huang et al., 2012). The yield was highest (8.8±5.6 g m^{-2} yr^{-1}) in rivers from the SW ~~estuaries~~ region of India, ~~annually~~despite they export relatively lower DIC ~~to the north Indian Ocean~~ (0.3 Tg yr^{-1}) due to their low volume of discharge (46 km^3 yr^{-1}) and relatively smaller catchment area (~~0.02~~20x10^3 ~~M~~ km^2) than the rivers from SE, NE and NW ~~estuaries~~ regions of India (Table 1 ~~2 & 3), in contrast, the higher yield of DIC was found higher in the former (10.8±6.6 g m^{-2} yr^{-1}) than the latter (Table 3)~~. DIC yield showed a significant positive correlation with the volume of discharge (r^2=0.67, p<0.001; Fig. ~~6d~~6a) in medium ~~estuaries~~ rivers and no such relationship was found in the major ~~estuaries~~rivers. Significant negative relationship~~s were~~ was observed between DIC yield and catchment area of river (~~in the medium (~~r^2 = -0.49, p<0.001; Fig. ~~6e~~6b~~)~~ and ~~major estuaries (~~r^2 = -0.43, p<0.001;

Fig. ~~6f~~6c for medium and major rivers respectively), suggesting the smaller rivers export more DIC per unit area of catchment compared to the major river systems, and thus inclusion of DIC data from medium rivers in the world significantly alters the global estimations of DIC. ~~This suggests that high precipitation over small catchments increases DIC yield because the dense precipitation increases the extraction of DIC from soils and rocks in their catchment.~~ A fairly good linear relationship between the yield of DIC and the intensity of precipitation (r^2=0.43, $p<0.01$ Fig. 6d) was observed only in the rivers which receives >2000mm of annual mean precipitation. Higher precipitation over the catchment increases the yield of DIC because the dense precipitation enhances the extraction of DIC from soils and rocks in their catchment Therefore, high precipitation (~~2500±500~~~3000 mm) over the small catchment (~~0.02~~20x10^3 ~~M~~ km^2) could have increased DIC yield from the rivers of SW ~~estuaries~~region of India. ~~A strong linear relationship between the yield of DIC and the intensity of precipitation (r^2=0.64, $p<0.001$ Fig. 6g~~6d) ~~confirms that dense precipitation increases the export yield of DIC from~~ rivers of the ~~SW~~ region. ~~estuaries~~.

~~Existing natural vegetation of tropical moist deciduous and tropical wet evergreen and semi evergreen forests in the SW region~~ of India ~~could also have increased DIC yield from~~ rivers ~~of this region the SW estuaries as this vegetation favors the export fluxes of DIC. The drainage basins of the Indian monsoonal rivers are largely under the tropical dry and wet climate except the SW rivers, Narmada and Tapti. The rivers Narmada and Tapti are under the arid and semiarid climate while the SW rivers are under the tropical wet climate which was also reported to facilitate the riverine export of material from drainage basin to the coastal ocean.~~

Sreenivas et al. (2016) and Krishwan et al. (2009) found that the soil organic and inorganic carbon contents in the surface (100cm) soils in the catchment of ~~SW~~ rivers in the SW

714 region were higher and lower, respectively, than the catchments of the rivers from SE, SW and
715 NE ~~rivers~~ region of India ~~(Table 2)~~. Decomposition of soil organic matter releases excess CO_2
716 and the increase in ~~d~~ soil CO_2 leads to the formation of acidic conditions in soils. This would
717 increase the DIC yield by ~~This indicates that~~ more dissolution of soil carbonates ~~by acidic~~
718 ~~conditions formed by release of CO_2 through decomposition of soil organic carbon in catchments~~
719 ~~of the SW rivers~~and chemical weathering of carbonate and silicate rocks ~~by the acidic~~
720 ~~conditions~~. A significant linear correlation was found between soil organic carbon content and
721 DIC yield in this study (r^2=0.65, p<0.001; Fig. 6e), suggesting that ~~Hence, the~~ higher soil
722 organic carbon in the catchment of the rivers from SW region ~~than the SE, NE and NW rivers~~
723 ~~(Kishwan et al., 2009; Sreenivas et al., 2016)~~ could have elevated the yield of DIC from rivers of
724 this region. ~~SW estuaries through dissolution of soil carbonates. A significant linear correlation~~
725 ~~between soil organic carbon content and DIC yield in this study (r^2=0.65, p<0.001; Fig. 6h)~~
726 ~~confirms that strong influence of soil organic carbon content in the catchment on DIC yield from~~
727 ~~the Indian monsoonal rivers.~~ The basin scale studies are, however, required for comprehensive
728 understanding of the influence of environmental and anthropogenic factors on ~~DIC~~ export fluxes
729 and yield of DIC from the Indian monsoonal rivers.

730 **5. Summary**

731 In order to examine the spatial variability in the sources and distribution of dissolved
732 inorganic carbon (DIC) in the Indian monsoonal ~~estuaries~~rivers, and to estimate their ~~riverine~~
733 export fluxes of DIC to the north Indian Ocean, we sampled a total of 27 major and medium
734 ~~estuaries~~ rivers ~~along the Indian coast~~ during wet period. An order of magnitude variability was
735 found in DIC concentrations among the ~~estuaries~~ rivers sampled (3.4 - ~~44.1~~73.6 mg l^{-1}), with a
736 lower mean concentration of 6.6±2.1 mg l^{-1} in ~~estuaries~~ rivers located in the SW region of India.

It is attributed to significant spatial variability in the size of rivers, precipitation pattern, pollution and lithology in their catchments. The approximated $\delta^{13}C$ of DIC source from the Keeling and Miller-Tans plots (-2.0 and -3.0‰ respectively) and, the calculated $\delta^{13}C$ of CO_2 which filters the influence of pH and DIC speciation, suggested that DIC in the Indian rivers is mainly originated from chemical weathering of carbonate minerals but largely affected by autotrophic production in rivers from the southeast region of India and heterotrophic decomposition of organic matter in the other Indian rivers sampled. ~~Magnitude of discharge, catchment area and in-stream processes appears to be the controlling factors for concentration and yield of DIC in the medium estuaries rather than the major estuaries. This is probably due to a significant spatial variability in lithology and hydro-geological and environmental conditions in the catchments.~~ Indian monsoonal ~~estuaries~~ rivers together ~~annually~~ export ~10.~~4~~ 3 Tg yr^{-1} of DIC to the north Indian Ocean, of which 7.~~7~~ 8 Tg yr^{-1} enters in to the Bay of Bengal while the Arabian Sea receives only 2.5~~7~~ Tg yr^{-1}. It is mainly attributed to the volume of river discharge as the former receives ~378 km^3 yr^{-1} while the latter receives only 122 km^3 yr^{-1} from the Indian monsoonal rivers. ~~The range of $\delta^{13}C_{DIC}$ found in this study suggests that major contribution of DIC is from weathering of silicate and carbonate minerals by carbonic acid formed by dissolution of soil CO_2. However, relatively enriched $\delta^{13}C_{DIC}$ in the east flowing river estuaries indicated the storage of water in dams/reservoirs and intrusion of marine waters.~~ Dense rainfall ~~-~~and higher soil organic carbon content in the catchment of rivers from the SW ~~rivers~~ region than in the catchment of the other Indian rivers resulted in higher yield of DIC from the former than the latter.

6. Acknowledgements

We would like to thank the Director, CSIR - National Institute of Oceanography (NIO), Goa, and the Scientist-In-Charge, NIO-Regional Centre, Visakhapatnam for their kind support

and encouragement. We also acknowledge Dr. M. Dileep Kumar, NIO, Goa for his guidance and encouragement. The work is part of the Council of Scientific and Industrial Research (CSIR), funded research project. This publication has NIO contribution number .....

7. Data Availability

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

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Figure captions

Figure 1: Map showing the study region. ~~Estuaries of the r~~Rivers sampled in this study were indicated by solid black line. Distribution of soils in catchments of the Indian monsoonal rivers sampled were shown. The rivers draining the four regions northwest (NW), southwest (SW), southeast (SE) and northeast (NE) were shown by solid black arrows.

Figure 2: Schematic diagram showing the $\delta^{13}C$ of different end members of dissolved inorganic carbon (DIC) sources. Various major processes influencing the $\delta^{13}C$ of DIC ($\delta^{13}C_{DIC}$) within the rivers were also shown. Black arrows (⇦ and ⇨) indicates the direction of change in $\delta^{13}C_{DIC}$ due to the influences different in-stream process mentioned against arrows.

Figure ~~2~~3: Spatial variability in c~~C~~oncentration (mg l^{-1}; 3a), export flux (Tg yr^{-1}; 3b) and yield (g m^{-2} yr^{-1}; 3c) of dissolved inorganic carbon (DIC) and its stable isotopes ($\delta^{13}C_{DIC}$, 3d) in the Indian monsoonal ~~estuaries~~rivers studied. ~~Estuaries~~ Rivers geographically located in the northwest~~eastern~~ (~~NE~~NW), southwest (SW), southeast~~ern~~ (SE) and~~,~~ ~~southwestern (SW) and~~ northeast~~western~~ (~~NW~~NE) regions of India were also shown. Rivers draining into the Bay of Bengal (east-flowing rivers) were shown by gray shade ~~Estuaries draining into the Bay of Bengal and the Arabian Sea were also provided~~while rivers draining into the Areabian Sea (west-flowing) were shown by no shade.

~~**Figure 3:** Spatial variability in stable carbon isotopes of dissolved inorganic carbon ($\delta^{13}C_{DIC}$, ‰) in the Indian monsoonal estuaries during the study period.~~

Figure 4: (a) Exponential decrease and (b) linear increase of dissolved inorganic carbon (DIC) concentrations with increasing the rainfall over the catchment and length of the river respectively. (c) Inverse and (d) linear relationships of chlorophyll-a (Chl-a) with concentrations and $\delta^{13}C$ of DIC respectively. Significant linear relationships of $\delta^{13}C$ of DIc with (e) dissolved oxygen saturation, (f) dissolved organic carbon (DOC) concentration, (g) total alkalinity and (h) DIC in the Indian monsoonal rivers during the study period. Ovals with dashed line indicate that outliers which were not included in the regression equations. Rivers of the northwest region of India showed linear relationships as shown by other Indian rivers but with a different slope (Fig. f-h) ~~Correlation between mean DIC concentration and precipitation in the four regions (NE, SE, SW and NW) of India. Relationship of DIC concentrations with (b) catchment area and (c) discharge volume of rivers in the medium estuaries. (d) Correlation between $\delta^{13}C_{DIC}$ and salinity in the in the Indian monsoonal estuaries during the study period.~~

Figure 5: ~~A schematic showing the range of $\delta^{13}C$ values of dissolved inorganic carbon (DIC) derived from various sources. Different physical and biogeochemical processes influencing the $\delta^{13}C$ of DIC were also shown. The forward arrow (⇨) indicates enrichment while the reverse arrow (⇦) indicates depletion in the $\delta^{13}C$ of DIC~~Least square linear regression models of (a) $\delta^{13}C_{DIC}$ as a function of 1/DIC (Keeling plot) and (b) $\delta^{13}C_{DIC}$xDIC as a function of DIC

concentrations (Millte-Tans plot) in the Indian monsoonal rivers. Linear relationship between calculated $\delta^{13}C$ of CO_2 and the measured bulk $\delta^{13}C_{DIC}$ (c) and Miller-Tans linear regression model of $\delta^{13}C$- CO_2 x CO_2 as a function of CO_2 concentration (d) in the Indian rivers sampled in this study.

Figure 6: Significant relationships dissolved inorganic carbon (DIC) yield with (~~between (~~a) river discharge in medium estuaries, (b) catchment areas of the medium rivers, (c) cathment areas of the major rivers, (d) rainfall over the catchment of all rivers sampled and (e) soil organic carbon (OC) content in catchments of the Indian monsoonal rivers studied. Since the data on soil OC data is not available for each watershed (e) was ploted using the available soil OC data on regional scale (NW, SW, SE and NE regions of India), Hence, it contains only four points. ~~chlorophyll-a and concentration of dissolved inorganic carbon (DIC), (b) $\delta^{13}C_{DIC}$ and dissolved oxygen (DO) saturation and (c) $\delta^{13}C_{DIC}$ and concentrations of DIC in the Indian monsoonal estuaries during the study period. The relationships of DIC yield from medium estuaries with (d) volume of discharge and (e) catchment area of the rivers. (f) Correlation between the yield of DIC and catchment area of the rivers in the major estuaries. Relationships of DIC yield from the Indian monsoonal estuaries with (g) precipitation and (h) soil organic carbon in the four regions (NE, SE, SW and NW) of India.~~

Table captions

Table 1: Catchment area, discharge, lengh and elevation of each river and, annual mean rainfall, number of dams and population density in each watershed of the Indian monsoonal rivers sampled. Concentrations, export fluxes and yields of dissolved inorganic carbon (DIC) and its stable isotopes ($\delta^{13}C_{DIC}$) of Indian rivers were given. Measured pH and calculated $\delta^{13}C$ of CO_2 from isotopic fractionation factors across DIC speciation were also provided. ~~Mean or range of concentrations of dissolved inorganic carbon (DIC, mg l^{-1}) in the Indian monsoonal estuaries and elsewhere in the world.~~

~~**Table 2:** Total catchment area (million square kilometre), annual discharge volume (km^3) and export flux (Tg yr^{-1}) of the estuaries located in the NE, SE, SW and NW regions of India. Mean (±SD) values of concentration, $\delta^{13}C$ and yield of DIC in estuaries of these four regions were given. Annual mean rainfall (mm) and soil organic carbon content (ton ha^{-1}) in surface soils of these regions were also shown. Mean (±SD) concentrations of DIC in the ground waters of these four regions were also provided.~~

~~**Table 3:** Export flux (Tg yr^{-1}) and yield (gC m^{-2} yr^{-1}) of dissolved inorganic carbon (DIC) from the Indian monsoonal rivers and from the rivers elsewhere in the world.~~

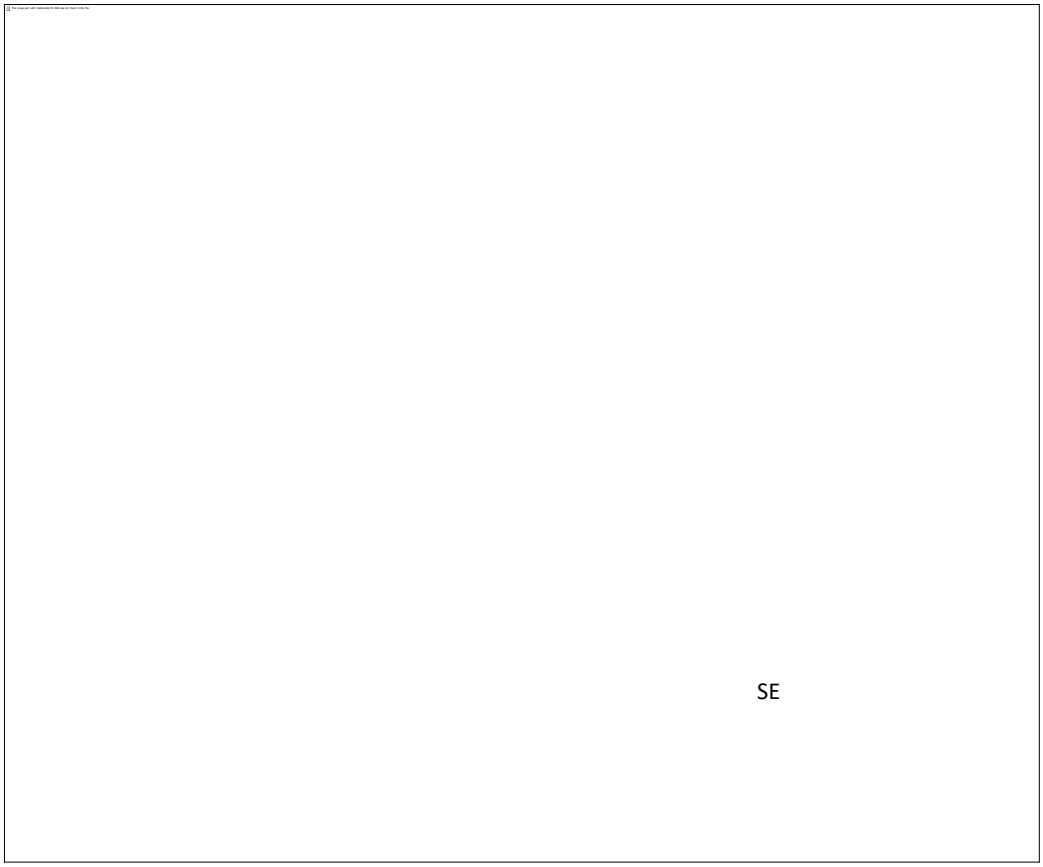

Fig.1:

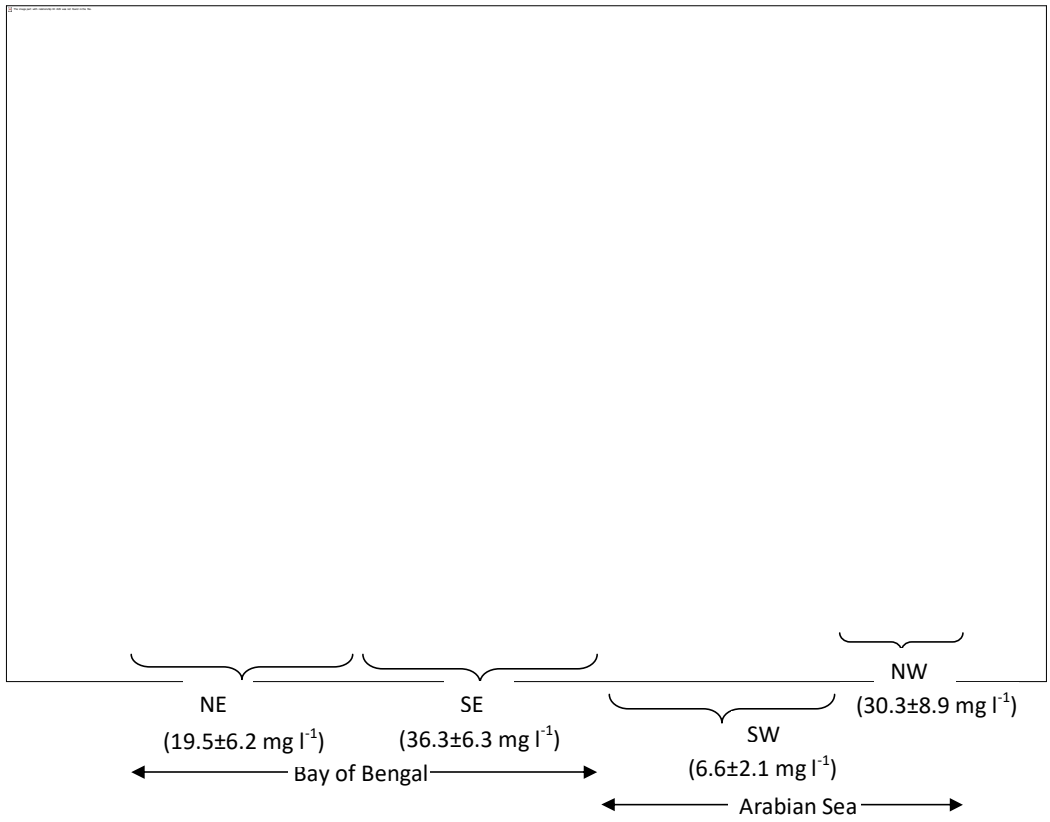
NE
(19.5±6.2 mg l-1)
SE
(36.3±6.3 mg l-1)
SW
(6.6±2.1 mg l-1)
NW
(30.3±8.9 mg l-1)
Bay of Bengal
Arabian Sea

Fig. 2

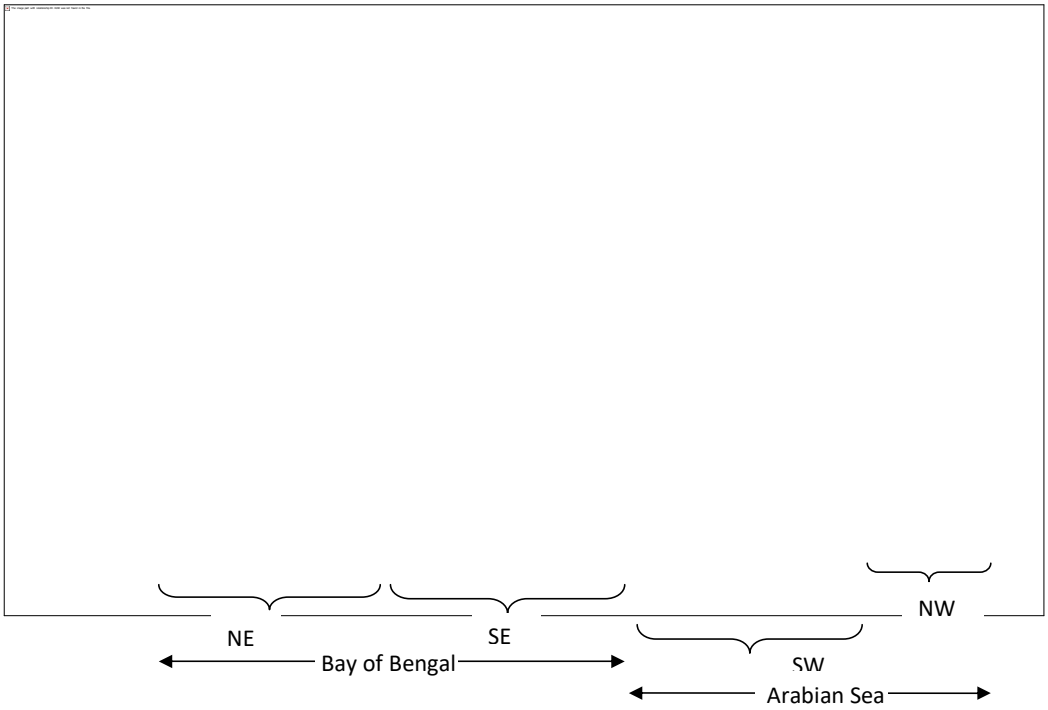

Fig. 3:

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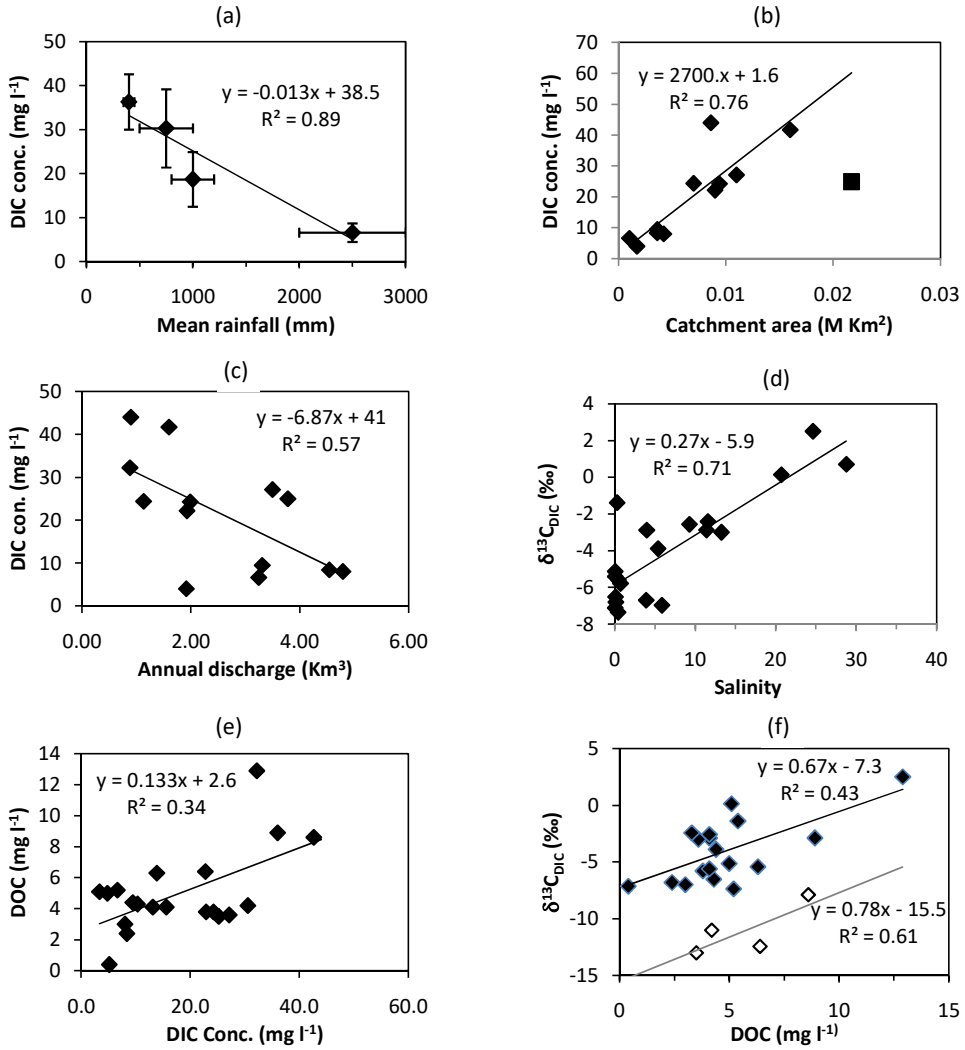

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Fig. 4

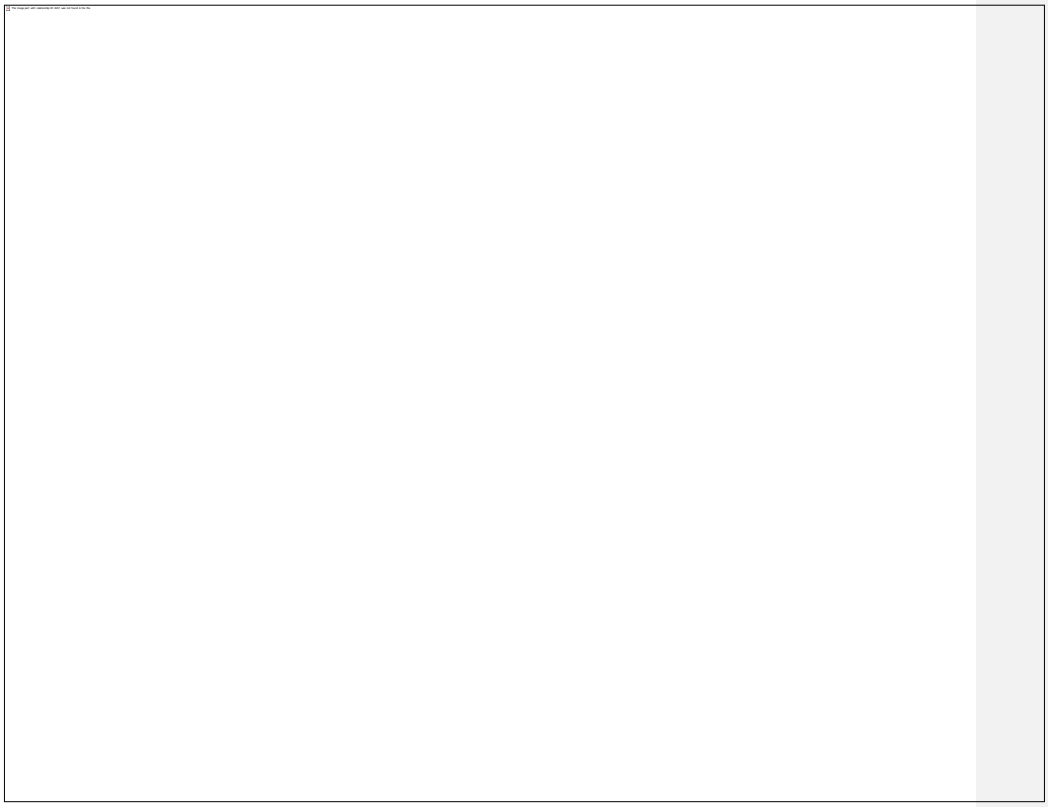

Fig. 5

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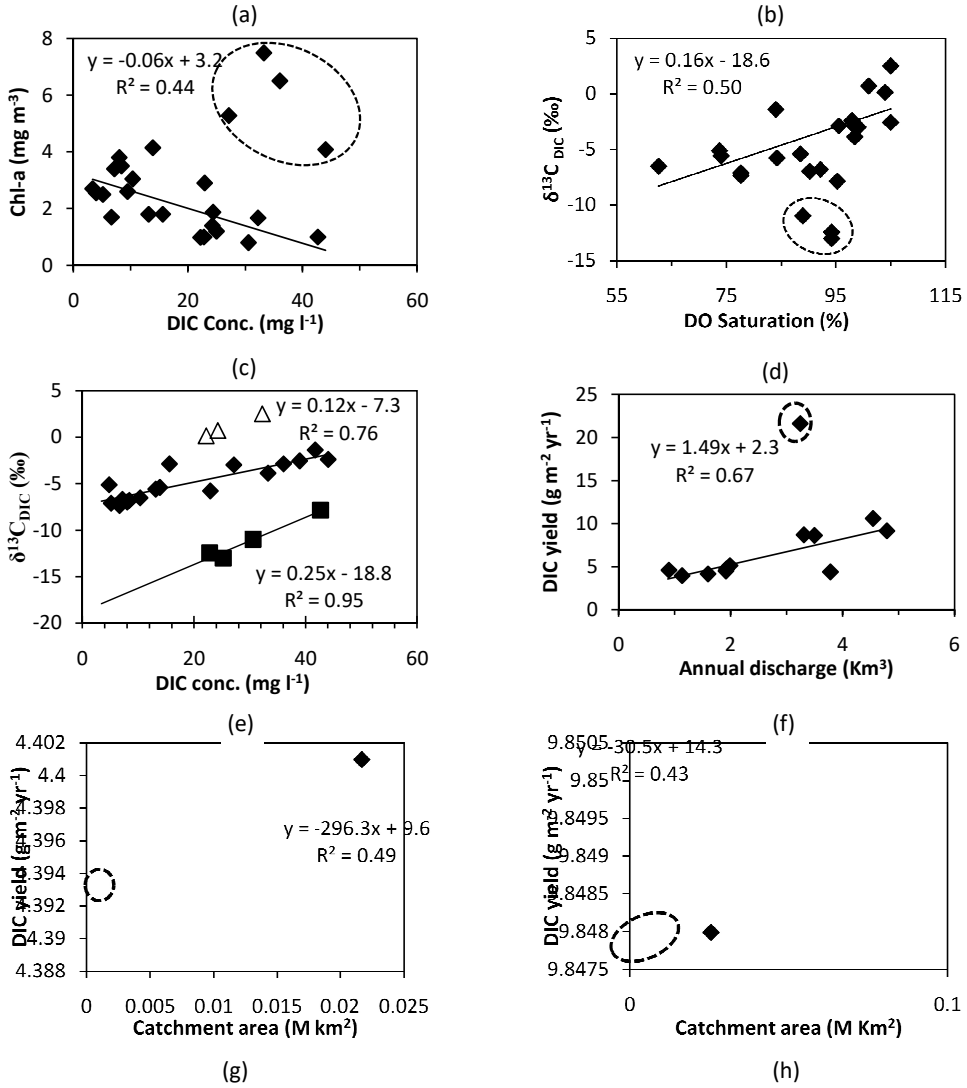

(g) (h)

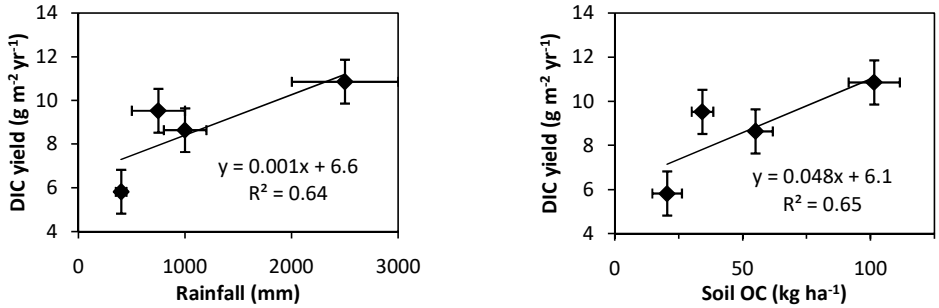

Fig. 6:

S. No.	Mean DIC conc. (mg l^{-1})	River	Reference
1	23	Ganga-Brahmaputra	Singh et al., 2005
2	22	Hooghly	Samanta et al., 2015
3	15	Mahanadi	Pattanaik et al., 2017
4	6-21	York river estuary	Raymond and Bauer, 2000
5	28	Yangtze river	Cai et al., 2008
6	4 - 43	British rivers	Jarvie et al., 2017
7	18 - 22	Seri, central Japan	Ishikawa et al., 2015
8	9 - 30	Red river, Vietnam	Quynh et al., 2016
9	18 - 46	Xi river, southwest China	Zou, 2016
10	37 - 66	rivers draining into the Gulf of Trieste	Tamse et al., 2014
11	10.3	Global mean	Meybeck and Vorosmarty, 1999
12	12.7	Asian rivers in tropical region	Huang et al., 2012
13	3.4 to 44	Indian estuaries	Present study

Table 1

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S. No.	Region of India	Total catchment area (M km^2)	Annual Discharge (km^3)	Mean±SD of DIC conc. (mg l^{-1})	Mean±SD DIC yield (g $m^{-2}yr^{-1}$)	Annual DIC export flux (Tg)	Mean±SD $\delta^{13}C_{DIC}$ (‰)	Mean (±SD) annual rainfall (mm)	Mean±SD GW DIC (mg l^{-1})*	Soil OC (ton ha^{-1})
1	NE	0.53	276	19.5±6.2	8.6±5.7	4.2	-3.5±2.8	1000±200	92±31	55
2	SE	0.45	102	36.3±6.3	5.8±2.3	3.5	-2.7±5.2	400±50	106±56	20
3	SW	0.02	46	6.6±2.1	10.8±6.6	0.3	-7.4±1.9	2500±500	32±19	101
4	NW	0.21	75	30.3±8.9	9.5±4.0	2.4	-11.1±2.3	750±250	84±54	34

*data has been taken from Dr. BSK Kumar, personnel communication.

Table 2

S. No.	Rivers	DIC export flux (Tg yr^{-1})	DIC yield (gC m^{-2} yr^{-1})	Reference
1	World major rivers	385	2.58	Meybeck and Vorosmarty, 1999
2	Asian rivers	111	9.79	Huang et al., 2012
3	American rivers	61.4	3.3	Huang et al., 2012
4	African rivers	17.7	0.63	Huang et al., 2012
5	Rivers draining to the tropical Atlantic from South America and Africa	53	-	Araujo et al. 2014
6	Tropical rivers	210	3.3	Huang et al., 2012
7	Indian monsoonal rivers*	10.4	8.7	Present study

Table 3