Biogeosciences Discuss., https://doi.org/10.5194/bg-2018-4
Manuscript under review for journal Biogeosciences
Discussion started: 9 January 2018

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

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Abstract. Rivers are strong source of dissolved inorganic carbon (DIC) to the adjacent coastal waters. In order to identify the major sources of DIC in the Indian monsoonal estuaries and their export flux to the north Indian Ocean, 27 major and medium estuaries along the Indian coast were sampled during discharge period. An order of magnitude variability in DIC concentrations was found within the Indian estuaries sampled (3.4 - 44.1mg l^{-1}) due to significant variability in the size of rivers, precipitation pattern and lithology in the catchments. Dilution with high precipitation (2500±500 mm) and exchange with ground waters of low DIC resulted in very low concentrations of DIC in estuaries located in the southwest of India (6.6±2.1 mg l^{-1}) than the estuaries located in the southeast (36.3±6.3 mg l^{-1}), northwest (30.3±8.9 mg l^{-1}) and northeast (19.5±6.2 mg l^{-1}) regions of India. Though the range of stable carbon isotopes of DIC ($\delta^{13}C_{DIC}$) indicates that DIC is largely contributed by weathering of silicate and carbonate minerals, however, the storage of water in dams/reservoirs and intrusion of marine waters caused the enrichment in stable carbon isotopic composition of DIC ($\delta^{13}C_{DIC}$). It is estimated that the Indian monsoonal estuaries annually export ~10.4 Tg (1Tg=10^{12} g) of DIC to the northern Indian Ocean, of which the major fraction (74.2%) enters into the Bay of Bengal and the remaining reaches to the Arabian Sea. It is mainly due to the fact that the Bay of Bengal receives ~378 km^3 yr^{-1} of freshwater from the catchment area of about 0.96 million km^{-2}, whereas the Arabian Sea receives only 122 km^3 yr^{-1} of freshwater from the catchment area of only 0.23 million km^2.

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Though the discharge from the Indian monsoonal rivers account for only 1.3% of global freshwater discharge, they disproportionately export 2.5% of the total DIC export by the world major rivers and 9.4% of the Asian rivers to oceans. The yield of DIC was found to be higher in the SW estuaries (10.8±6.6 g m^{-2} yr^{-1}) than the other estuaries though they export only 0.3 Tg yr^{-1} of DIC, which is more than an order of magnitude lower than the export by the NE (4.2 Tg yr^{-1}) and SE estuaries (3.5 Tg yr^{-1}), due to intense precipitation, favorable natural vegetation and tropical wet climate, high soil organic carbon and dominance of red loamy soils in catchments of the SW rivers. This study, therefore, reveals that significant variability in the lithology and hydrological and environmental conditions over the catchments strongly controls the concentrations and yield of DIC from the Indian monsoonal estuaries.

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

1. Introduction

Dissolved inorganic carbon (DIC) is the major constituent of carbon species and accounts for ~38% of the total fluvial carbon transport to the global oceans (Meybeck, 1993; Cai, 2011; Jarvie et al., 2017). World major river systems export annually about 33-400 Tg (1Tg=10^{12}g) of DIC to the global oceans (Ludwig et al., 1998; Mackenzie et al., 2004; Lerman et al., 2007). Chemical weathering of carbonate and silicate rocks and soils, and exchange with the ground water in the 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), besides in-stream processes, such as oxidation of organic carbon by heterotrophic bacteria (Mayogra et al., 2005; Battin et al., 2008; Hotchkiss et al., 2015; Samantha et al. 2015; Zou, 2016) and dissolution of atmospheric carbon dioxide (CO_2). Weathering of carbonate and

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silicate rocks in the catchment, and uptake of DIC by aquatic plants and algae during photosynthesis reaction in rivers are the sinks of 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. DIC in rivers and estuaries is therefore strongly linked to the carbon cycle. However, due to human interferences, DIC fluxes from the world major rivers have been found to increase dramatically in the last century, for example, Mississippi (Cai, 2003; Raymond and Cole, 2003; Raymond et al., 2008; Ren et al., 2015). It has been noted that substantial alterations in DIC lateral transport occurred from land to sea after the industrialization (Regnier et al., 2013; Bauer et al., 2013). The increase in riverine DIC flux was reported to have a significant impact on the chemical composition (Williamson et al., 1994; Raymond and Cole, 2003; Findlay, 2010; Tank et al., 2010) and carbon budget in the coastal waters (Cole et al., 2007; Dhillon and Inamdar, 2013). Thus, identification of major sources of DIC and its riverine export flux estimates to the coastal oceans are important for better understanding the carbon cycling and its budget on both regional and global scales (Campeau et al., 2017).

Fluvial carbon fluxes from rivers in the tropical region (30^{o} N to 30^{o}S) is critical for global carbon budgets because they contribute significant fraction of global DIC (48-64%), freshwater discharge (66.2%) and suspended sediment load (73.2%) to the world oceans, despite they occupy only ~43% of world's land area (Huang et al., 2012). Further, humid tropical climate in the region supports the export of more fluvial carbon fluxes from the continental land masses than the other climates in the world (Meybeck 1993; Ludwig et al., 1998). However, DIC fluxes from rivers in this region were not included in estimating the fluvial carbon fluxes to global oceans due to the paucity of data.

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Numerous studies have been documented on DIC export flux from the world major rivers, for example, Mississippi (Raymond and Cole, 2003; Raymond et al., 2008; Cai et al., 2008), Changjiang and Pearl (Cai et al., 2008), Congo (Wang et al., 2013) and large river systems in the world (e.g. Gaillardet et al., 1999; Raymond et al., 2013). Although some measurements were carried out on DIC in the Indian estuaries, for example, Mandovi and Zuari (Sarma et al., 2001), Godavari estuary (Sarma et al., 2011), Cochin (Gupta et al., 2009; Bhavya et al., 2016), Hooghly (Mukhopadhyay et al., 2002; Samanta et al., 2015), Mahanadi (Pattanaik et al., 2017) and Indian estuaries (Sarma et al., 2012), however, they focused only on air-water exchange of CO_2. Nevertheless, no estimations on DIC export fluxes to the north Indian Ocean from the Indian subcontinent have been made so far. For the first time, we made an effort here to identify the major sources of DIC in the Indian monsoonal estuaries and to estimate their export fluxes to the north Indian Ocean. The main objectives of this study are to (i) identify the major sources and (ii) examine the potential reasons responsible for variability in concentrations of DIC in the Indian monsoonal estuaries during the discharge (wet) period, and (iii) estimate the DIC export fluxes to the north Indian Ocean by the Indian monsoonal rivers.

2. Study region and Sampling

2.1 Study Area

The Indian peninsula bifurcate the north Indian Ocean into the Bay of Bengal and the Arabian Sea. Although these two basins occupies the same latitudinal belt, their oceanographic processes were reported to be remarkably different and attributed to significant differences in the freshwater influx and associated physical and biological changes (Gauns et al., 2005). This is because the glacial and peninsular rivers transport 1.63 x 10^{12} m^3 yr^{-1} of freshwater to the Bay of Bengal (Subramanian, 1993) whereas only 0.3x10^{12} m^3 yr^{-1} to the Arabian Sea. The large

96 freshwater influx leads to formation of a strong vertical salinity stratification (Varkey et al.,
97 1996), which results in the suppression of vertical mixing of nutrient rich sub-surface water with
98 that of surface, makes the Bay of Bengal relatively less productive (Prasannakumar et al., 2002)
99 than the Arabian Sea, which is one of the highly productive zones in the world (Madhupratap et
100 al., 1996; Smith, 2001; Barber et al., 2001) due to injection of nutrients into surface through the
101 seasonal upwelling and convective mixing (Shetye et al., 1994; Madhupratap et al., 1996;
102 Muraleedharan and Prasannakumar, 1996).

103 Discharge from the Indian peninsular rivers is fed by the monsoon induced precipitation
104 over the Indian subcontinent, which receives >80% of its annual rainfall during the southwest
105 (SW) monsoon period (June-September) (Soman and Kumar, 1990). Though some amount of
106 rainfall occurs during the NE monsoon (December-March), it will not generate discharge as it
107 will be stored in dams and reservoirs for domestic, industrial and irrigation purposes. Discharge
108 from the Indian peninsular rivers is therefore occurs only during the SW monsoon season (Vijith
109 et al., 2009; Sridevi et al., 2015) and hence, termed as monsoonal rivers. Since the freshwater
110 discharge from the Indian monsoonal rivers is limited to only few months (June – October) in a
111 year, unlike the European and American rivers, the entire estuary may be filled with freshwater
112 without any vertical salinity gradient (Vijith et al., 2009; Sridevi et al., 2015) during this period.
113 As virtually there is no discharge during the dry period, the discharge during SW monsoon (wet
114 period) is equivalent to the annual discharge from the monsoonal rivers. Based on the rainfall
115 intensity, forest cover, vegetation and soil type in the catchment, estuaries sampled in the present
116 study were categorized into 4 groups, namely the northeast (NE), southeast (SE), southwest
117 (SW) and northwest (NW) estuaries of India (Fig. 1). The SW region of India is characterized
118 by the intense rainfall during the SW monsoon (~3000 mm) following the NE (1000-2500 mm),

SE (300-500 mm) and NW (200-500 mm) regions of India (Soman and Kumar, 1990). The SW rivers drain red loamy soils while the NW rivers drain black soils. The rivers reaching the Bay of Bengal (NE and SE estuaries) drain the red loamy and alluvial soils in their upper and lower catchments respectively, except the major rivers Godavari and Krishna, which also drain black soils in their upper catchment along with red loamy and alluvial soils in their middle and lower catchments (Geological Survey of India; www. gsi.gov.in). Based on the discharge, monsoonal estuaries in this study were divided into two types, namely, the minor (<150 m^3s^{-1}) and major (>150 $m^3 s^{-1}$) estuaries.

2.2 Sample collection

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

In-situ measurements and sample collection was done in the 27 estuaries (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: 0.7µm) under moderate vacuum and stored in liquid nitrogen for Chl-*a* analysis at the NIO.

3. Methods

Temperature and salinity at the sampling locations were measured using a CTD system (Sea Bird Electronics, SBE 19 plus, United States of America). Concentrations of DO were determined by Winkler's method (Carritt and Carpenter, 1966) using an auto titrator (Metrohm, Switzerland) with potentiometric end point detection. The analytical precision of the method was ±0.07% (RSD). DIC concentrations in water samples were measured at our Institute laboratory using Coulometer (UIC Inc., USA) connected to an automatic sub-sampling system. Based on the repeated analysis of samples and standards, the precision of the method was ±1.8 µmol l^{-1}. The certified reference materials (CRM) supplied by Dr. A.G. Dickson, Scripts Institute of Oceanography, USA and internal standards were used to test the accuracy of our DIC measurements and it was found to be within ± 0.2 to 0.3%. Chlorophyll-*a* (Chl-*a*) on the filter was extracted into di-methyl formamide (DMF) and measured the extract fluorometrically using a spectrofluorophotometer (Varian Eclipse, Varian Electronics., UK) following Suzuki and Ishimaru (1990). Annual mean discharge data of rivers was taken from Meybeck and Ragu (1995, 1996), Central Water Commission, New Delhi (2006, 2012) and Kumar et al. (2005). Soil organic carbon data was taken from Kishwan et al. (2009) and rainfall data was obtained from Soman and Kumar (1990). Dissolved organic carbon (DOC) data for the Indian estuaries 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 with the mean annual discharge. Spatial variability in DIC concentrations in estuaries was minimized to a large extent by collecting samples from head to mouth of the estuary while the inter-annual variability by collecting samples during discharge periods of two years. However, variability in DIC concentrations within the discharge period results in some uncertainties in our estimations of DIC export fluxes. Time series measurements in the Godavari estuary (our unpublished results) revealed that the variability in DIC concentrations within the discharge period is up to 10%. Therefore, the error associated with our DIC flux estimates can be about ±10%. DIC fluxes normalized by catchment area (yield) were calculated by dividing the total DIC export flux of the river by its catchment area.

3. Results

Prevailing hydrographic conditions in Indian estuaries during the sample collection were given in detail elsewhere (Sarma et al., 2012, 2014; Krishna et al., 2015). Briefly, mentioned here for ready reference. Surface water temperature was found to be higher in estuaries located on the east coast (mean 30.86±1.23^{o}C) than the west coast (27.32±1.49^{o}C) of India. Salinity varied broadly from near zero (0.06) to 28.78 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 (28.78), Vaigai (24.63) and Rushikulya (20.70). Dissolved oxygen saturation varied from as low as 62.6% to as high as 105%, with a mean saturation of 89.9±11.4% in the estuaries sampled. Chlorophyll-*a* (Chl-*a*) concentrations varied broadly from 0.8 to 10.7 mg m^{-3}, with relatively higher mean concentrations in the SE (4.7 mg m^{-3}) followed by the SW (3.0 mg m^{-3}) estuaries. However, 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}).

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Manuscript under review for journal Biogeosciences
Discussion started: 9 January 2018

188 **3.1 Concentrations and $\delta^{13}C$ of DIC ($\delta^{13}C_{DIC}$) in the Indian monsoonal estuaries**

189 DIC concentrations in Indian estuaries widely varied from 3.4 (Bharathappuzha) to
190 44.1mg l^{-1} (Vellar), with a significant spatial variability (Fig. 2). More than five times higher
191 mean concentrations were observed in the SE (36.3±6.3 mg l^{-1}) and NW estuaries (30.3±8.9 mg
192 l^{-1}) than in the SW estuaries (6.6±2.1 mg l^{-1}), and intermediate concentrations were found in the
193 NE estuaries (19.5±6.2 mg l^{-1}). DIC concentrations were found to be similar (homoscedastic
194 Student's t-test; p=0.76) in the major (22.7±13.6 mg l^{-1}) and medium (21.1±13.2 mg l^{-1})
195 estuaries. The $\delta^{13}C_{DIC}$ varied from -13.0 to 2.5‰, with a significant spatial variability (Fig. 3) in
196 the estuaries sampled. Relatively depleted values were observed in the west flowing estuaries of
197 NW (-11.1±2.3‰) and SW (-7.4±1.9‰) than the east flowing estuaries of NE (-3.5±2.8‰) and
198 SE (-2.7±5.2‰) regions of India.

199 Annual export flux of DIC from the individual estuaries to coastal ocean varied broadly
200 from 0.009 Tg (Chalakudi) to as high as 2.32 Tg (Krishna). Annually, the NE estuaries export
201 higher DIC flux of 4.21 Tg followed by the SE (3.50 Tg) and NW estuaries (2.38 Tg). Whereas,
202 the SW estuaries recorded the lowest export flux of 0.30 Tg which is an order of magnitude
203 lower than the export flux by the NE and SE estuaries (Fig. 2). The Indian monsoonal estuaries
204 together export about 10.4 Tg yr^{-1} of DIC to the northern Indian Ocean, of which 7.7 Tg (74.2%)
205 enters into the Bay of Bengal and the remaining into the Arabian Sea (2.7 Tg). The estuaries,
206 Krishna (2.32 Tg), Godavari (1.45 Tg) and Haldia (1.16 Tg) together responsible for the
207 transport of 64% of total riverine DIC export to the Bay of Bengal by the Indian monsoonal
208 rivers. The yield of DIC ranged from 2.7 (Bharathappuzha) to 21.6 g m^{-2} yr^{-1} (Mandovi),
209 excluding the exceptionally high yield of 113.4 g m^{-2} yr^{-1} from Haldia estuary. The west flowing
210 rivers to the Arabian Sea are characterized by relatively higher yield of DIC (mean 10.4±5.6 g m^{-}

^{2} yr^{-1}) than the east flowing rivers to the Bay of Bengal (7.3±4.6 g m^{-2} yr^{-1}). Among the estuaries sampled, the SW and SE estuaries recorded higher (10.8±6.6g m^{-2} yr^{-1}) and lower (5.8±2.3g m^{-2} yr^{-1}) yields of DIC respectively. The NW (9.5±4.0 g m^{-2} yr^{-1}) and NE (8.6±5.7g m^{-2} yr^{-1}) estuaries recorded intermediate values.

4. Discussion

Hydrographic characteristics of the Indian monsoonal estuaries during the study (discharge) period were described elsewhere (Sarma et al., 2012, 2014; Krishna et al., 2015). Strong flow from the upstream rivers due to the SW monsoon-induced precipitation over the catchment makes most of the estuaries less saline (near zero), except the minor estuaries, Nagavali, Vaigai and Rushikulya, during the study period. No vertical salinity stratification was observed in estuaries during the study period, consistent with earlier observations in the Indian estuaries during discharge period (Vijit et al., 2009; Sridevi et al., 2015), unlike the European and American estuaries (Christopher et al., 2002).

4.1 Variability of DIC concentrations in the Indian monsoonal estuaries

Mean DIC concentration found in this study (21.9±13.2 mg l^{-1}; range: 3.4 to 44.1 mg l^{-1}) is similar to those observed earlier in the Indian estuaries, for example, Ganga-Brahmaputra (23 mg l^{-1}; Singh et al., 2005), Hooghly (21.8 mg l^{-1}; Samanta et al., 2015) and Mahanadi (15.0; Pattanaik et al., 2017) and elsewhere in the world, for instance, York river estuary (6-21 mg l^{-1}; Raymond and Bauer, 2000), Yangtze river (28 mg l^{-1}; Cai et al., 2008), British rivers (median 4-43 mg l^{-1}; Jarvie et al., 2017), Seri, central Japan (17.6-21.9 mg l^{-1}; Ishikawa et al., 2015), the Red river, Vietnam (9.1-29.9 mg l^{-1}; Quynh et al., 2016) and Xi river, southwest China (18-45.6mg l^{-1}, Zou, 2016). However, mean DIC concentrations in the Indian estuaries (21.9±13.2

mg l^{-1}) are higher than the global mean of 10.3 mg l^{-1} (Meybeck and Vorosmarty, 1999) and the Asian rivers (12.7 mg l^{-1}) in the tropical region (30°N-30°S; Huang et al., 2012), but lower than the rivers draining into the Gulf of Trieste (N Adriatic) (37-66 mg l^{-1}; Tamse et al., 2014).

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. DIC concentrations in estuaries are mainly governed by the hydrological (precipitation and runoff), lithological (type and dominance of rocks and soils) and environmental (temperature, climate and vegetation) conditions, and anthropogenic activities (deforestation and land use change) in the catchment, and in-stream physical and biological processes such as exchange with ground water (Finlay, 2003; Shin et al., 2011; Maher et al., 2013) and atmospheric CO_2, autotrophic production and heterotrophic decomposition of organic matter (McConnaughey et al., 1994; Abril et al., 2003; Finlay and Kendall, 2007). Since many of these processes are largely dependent on the size of the river and its catchment, the lower DIC concentrations in the SW estuaries of this study could be due to the size of the rivers. This is because, the SW rivers are small both in terms of discharge (46 km^3 yr^{-1}) and catchment area (total catchment area: 0.02 M km^2) than the SE (102 km^3 yr^{-1} and 0.45 M km^2, respectively), NE (276 km^3 $yr^{-1}$and 0.53 M km^2) and NW (75 km^3 yr^{-1} and 0.21 M km^2) rivers. However, DIC concentrations showed significant positive relationship with catchment area (r^2=0.75; $p<0.001$; Fig. 4a) and negative relationship with volume of discharge (r^2=0.57; $p<0.001$; Fig. 4b) 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 the major estuaries.

Biogeosciences Discuss., https://doi.org/10.5194/bg-2018-4
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Indian monsoonal estuaries were reported as a source of CO_2 to the atmosphere during discharge period (Sarma et al., 2001, 2011, 2012; Gupta et al., 2009; Bhavya et al., 2016) due to the microbial decomposition of terrestrial organic matter brought by the rivers. This suggests that the DIC input from dissolution of atmospheric CO_2 in estuaries can be ruled out, however, heterotrophic decomposition of organic matter adds significant amount of DIC to the Indian estuaries during discharge period. A fairly good positive correlation between DIC and DOC concentrations (r^2=0.34, $p<0.01$), except few medium estuaries, suggests that DIC addition through microbial degradation of particulate organic matter is significant in the Indian estuaries. Except the NW estuaries, which recorded relatively depleted $\delta^{13}C$ of DIC ($\delta^{13}C_{DIC}$), the positive correlation between $\delta^{13}C_{DIC}$ and DOC concentrations (r^2=0.35, $p<0.01$), as was observed elsewhere (Xi river, Zou et al., 2016), confirms that oxidation of organic matter is one of the main DIC sources in the Indian estuaries. On the other hand, autotrophic production removes DIC as it converts DIC to organic carbon. Significant negative correlation between chlorophyll-*a* and DIC concentrations (r^2=0.47, $p<0.01$), except few SE estuaries 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 estuaries during the study period. Significance of DIC addition by heterotrophic decomposition and removal by autotrophic production in the Indian estuaries was confirmed by a fairly good positive correlation between $\delta^{13}C_{DIC}$ and dissolved oxygen saturation (r^2=0.49, $p<0.01$), (depleted $\delta^{13}C_{DIC}$ values at low % of DO saturation), except NW estuaries, which recorded depleted $\delta^{13}C_{DIC}$ (<-10.0‰). This is because the microbial decomposition of organic matter results in depleted $\delta^{13}C_{DIC}$ due to preferential release of ^{12}C over ^{13}C in to DIC pool while removal of DIC by autotrophic production enriches the residual DIC due to preferential uptake of ^{12}C over ^{13}C during photosynthesis reaction.

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

290 Spatial distribution of bedrock and soils over the Indian subcontinent shows that
291 Narmada and Tapti rivers and upper reaches of Godavari and Krishna rivers drain the igneous
292 rocks (Deccan traps) while the other rivers flow through the metamorphic rocks (Pre-Cambrian),
293 the predominant rock type in south India. However, Haldia and lower reaches of the SE rivers
294 drain the sedimentary rocks (Geological Survey of India, https://www.gsi.gov.in). Although, the
295 chemical weathering rates were reported to be higher for Deccan Trap basalts (Das et al., 2005;
296 Singh et al., 2005), however, higher DIC concentrations were also found in estuaries draining the
297 metamorphic rocks, suggesting that strong influence of factors other than the bedrocks in the
298 catchment. Spatial distribution of soils shows that lateritic soils, which are poor in lime and
299 silicate, occupied the catchment of the SW rivers. Chemical weathering rates are relatively lower
300 in the lateritic than the non-lateritic soils and the consumption of atmospheric/soil CO_2 through

silicate weathering is lower by ~2 times in the former than the latter (Boeglin and Probst, 1998). Though the upper reaches of the east flowing rivers (NE and SE) drain the lime-poor red and yellow soils, however, they are dominated by the lime-rich alluvial soils in their lower reaches. Upper reaches of Krishna and Godavari also drain the lime-rich black soils. The dominance of lateritic soils, which are relatively less susceptible to chemical weathering than the non-lateritic soils, over the catchments of the SW rivers could have, at least in part, lowered the DIC concentrations in SW estuaries during the study period.

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

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

The stable isotopic composition of DIC ($\delta^{13}C_{DIC}$) is a well-established and widely used tracer to identify the major sources of DIC in rivers (e.g. Singh et al., 2005; Tamooh et al., 2013; Samanta et al., 2015; Zou, 2016) because each of the DIC sources have a distinct $\delta^{13}C_{DIC}$ ratios (Deines et al., 1974). DIC originated by dissolution of atmospheric CO_2 is about -7 to -8‰

(Coplen et al., 2002) whereas it is about -26 to -27‰ if DIC is derived from oxidation of organic matter produced by C_3 plants (O'Leary, 1988). The $\delta^{13}C$ of DIC generated by soil CO_2 dissolved carbonic acid weathering of silicates is about -17 to -21‰ (Solomon and Cerling, 1987) while it is close to -9‰ for carbonate rocks because half of the carbon comes from carbonate rocks (0‰, Land, 1980) during weathering. Whereas, the weathering of carbonate and silicate minerals yield $\delta^{13}C_{DIC}$ values -7 to -8‰ and -3 to -4‰, respectively, if the carbonic acid formed by the dissolution of atmospheric CO_2. Although, DIC derived from different sources have distinctly different $\delta^{13}C_{DIC}$ values, however, the interpretation the $\delta^{13}C_{DIC}$ values for identification of its sources is still challenging (Amiotte-Suchet et al., 1999; Campeau et al., 2017) due to the isotopic fractionations associated with complex mixture of sources and processes such as photosynthesis (O'Leary, 1988; Finlay, 2004; Parker et al., 2005, 2010), respiration (Finlay, 2003; Waldron et al., 2007), DOC photo-oxidation (Opsahl and Zepp, 2001; Vahatalo and Wetzel, 2008), anaerobic metabolism (Waldron et al., 1999; Maher et al., 2015) and equilibration with atmospheric CO_2.

The range of $\delta^{13}C_{DIC}$ found in this study (-13.0 to 2.5‰) was similar to those reported earlier in various rivers, for example, Brahmaputra (Singh et al., 2005), Rhine (Buhl et al., 1991), Ottawa (Telmer et al., 1999), St. Lawrence (Yang et al., 1996), Nanpan and Beipan rivers, southwest China (Zou, 2016) and Tana river, Kenya (Tamooh et al., 2013). The range of $\delta^{13}C_{DIC}$ in this study indicates a variety of sources, including silicate and carbonate weathering and marine waters, contributes DIC to the Indian monsoonal estuaries during the study period. Relatively depleted $\delta^{13}C_{DIC}$ in the west flowing river estuaries of NW (mean -11.1±2.3‰) and SW (mean: -7.4±1.9‰) regions suggest that DIC is contributed from silicate and carbonate weathering by the carbonic acid, produced from the dissolution of both soil CO_2 and atmospheric

347 CO_2. Zou (2016) found the $\delta^{13}C_{DIC}$ values in the range of -13.9 to -8.1 ‰ in the Nanpan and
348 Beipan rivers of SW China and were attributed to dominant contribution of DIC from weathering
349 of carbonate minerals. Relatively enriched $\delta^{13}C_{DIC}$ in the east flowing river estuaries of NE (-6.5
350 to 0.7; mean: -3.5±2.8‰) and SE (-7.9 to 2.5‰; -2.7±5.2‰) indicates that major contribution of
351 DIC is from chemical weathering of carbonate rocks by atmospheric CO_2 dissolved carbonic
352 acid or acid from non-carbon sources (Li et al., 2008). Weathering of carbonate minerals by acid
353 sources other than carbonic acid causes enrichment compared to weathering by carbonic acid due
354 to lack of contribution from δ^{13}C-depleted carbonic acid of soil CO_2 (-17 to -21‰) or
355 atmospheric CO_2 (-7 to -8‰) origin to the δ^{13}C-enriched carbonate rocks (0‰, Land 1980).

356 In addition to the sources, hydrological and biological processes also influence the
357 $\delta^{13}C_{DIC}$ in streams/rivers. For example, heavy precipitation in the SW region (2500±500mm)
358 than the other regions tends to cause depletion in $\delta^{13}C_{DIC}$ values due to shorter residence time of
359 soil water (Amiotte-Suchet et al., 1999) while CO_2 out gassing causes enrichment due to
360 accumulation of ^{13}C during diffusive efflux (Clark and Fritz, 1997) in stored water bodies. Many
361 of the east flowing rivers are major and are dammed at many locations (e.g. Godavari, Krishna
362 and Cauvery) for domestic, industrial and irrigation purposes. CO_2 out gassing due to
363 heterotrophic decomposition of organic matter and autotrophic production significantly alters the
364 $\delta^{13}C_{DIC}$ signatures in reservoirs (Shin et al., 2001). Further, equilibrium with atmospheric CO_2 in
365 the reservoirs due to no/lean flow leads to enrichment in the $\delta^{13}C_{DIC}$ values (Brunet et al., 2005;
366 Bouvillion et al., 2009; Zeng et al., 2011; Tamooh et al., 2013). Hence, relatively enriched
367 $\delta^{13}C_{DIC}$ in the NE and SE estuaries could also be due to the storage of water in reservoirs/dams.
368 A significant positive correlation between DIC concentrations and $\delta^{13}C_{DIC}$ (r^2=0.77; $p<0.001$;
369 Fig. 7), excluding the positive values, indicate that significant contribution of DIC from

oxidation of particulate organic carbon in dams/reservoirs or stored water bodies. Shin et al. (2011) attributed the stream $\delta^{13}C_{DIC}$ values of -6.9±1.6‰ and -7.8±1.5‰ in silicate and carbonate dominated catchments, respectively, in tributaries of the Han River, South Korea to CO_2 out gassing. Positive $\delta^{13}C_{DIC}$ values (>0‰) were observed only in Rushikulya (0.1‰), Nagavali (0.7‰) and Vaigai (2.5‰) in which relatively higher salinities (>20) were found during the study period. This is concurrent with earlier observations in the Indian estuaries, Hooghly (Samanta et al., 2015) and Cochin (Bhavya et al., 2016) where relatively enriched $\delta^{13}C_{DIC}$ were found at higher salinities. A strong positive correlation was found between $\delta^{13}C_{DIC}$ and salinity (Fig. 6; r^2=0.71, $p<0.001$), suggesting that $\delta^{13}C_{DIC}$ values in the Indian estuaries are influenced by the intrusion of marine waters ($\delta^{13}C_{DIC}$: -1 to 2‰).

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

Indian monsoonal rivers annually export ~10.4 Tg of DIC to the north Indian Ocean. Nearly three fourth of this amount (7.7 Tg) reaches to the Bay of Bengal while the remaining into the Arabian Sea. It is mainly attributed to the magnitude of discharge because the Bay of Bengal annually receives 378 km^3 of freshwater from the catchment area of about 0.96 M km^{-2}, whereas the Arabian Sea receives only 122 km^3 of freshwater from the catchment area of only 0.23 M km^2. Although the increase in volume of discharge dilutes the DIC flux from rivers (Jarvie et al., 1997; Shanley et al., 2002), bicarbonate fluxes to the Gulf of Mexico were reported to increase with the volume of discharge from the Mississippi river (Raymond and Oh, 2007) due to small dilution factor.

The total DIC export by the Indian monsoonal estuaries (10.4 Tg yr^{-1}) is only 2.5% of the total DIC export by the world major rivers (400 Tg yr^{-1}), and 9.4% of the export by the Asian

392 rivers (111Tg yr^{-1}; Huang et al., 2012). The DIC export from the Indian estuaries is far less than
393 the DIC export by the American (61.4 Tg yr^{-1}) and African (17.7 Tg yr^{-1}) rivers and major rivers
394 draining to the tropical Atlantic from South America and Africa (53Tg yr^{-1}, Araujo et al. 2014).
395 It is mainly due to the fact that the volume of discharge from the Indian monsoonal rivers is very
396 low (~500 km^3) compared to the American (11,799 km^3) and African (3,786 km^3) rivers.
397 However, the Indian monsoonal rivers are exporting DIC disproportionately to the north Indian
398 Ocean because they account for only 1.3% of the global river discharge but export 2.5% of the
399 global riverine DIC to the oceans. Disproportionate DIC fluxes from the tropical regions are
400 mainly attributed to the favourable climatic conditions, lithology and land use cover (Huang et
401 al., 2012) in this region. Relatively higher export fluxes from the Indian rivers could be due to
402 higher weathering rates of silicate and carbonate minerals in the drainage basins of the Indian
403 rivers (Das et al., 2005; Gurumurty et al., 2012; Pattanaik et al., 2013)

404 Krishna et al. (2015) reported that Indian monsoonal estuaries export 2.32 Tg yr^{-1} of
405 dissolved organic carbon (DOC) to the north Indian Ocean. When combined the total fluvial
406 dissolved carbon flux would be 12.71 Tg yr^{-1}. This indicate that the total fluvial dissolved
407 carbon export to the north Indian Ocean by the Indian monsoonal estuaries is predominantly
408 contributed by DIC (~81%) than DOC, consistent with earlier reports elsewhere in the world, for
409 example, the British rivers (80%, Jarvie et al., 2017). Since the catchment area of the Indian
410 monsoonal rivers ranged widely from as low as 0.001 M km^2 to as high as 0.313 M km^2, the
411 export fluxes of DIC were normalized with the catchment area of river to obtain DIC yield from
412 each river in order to examine various factors controlling the lateral DIC export to the north
413 Indian Ocean.

414 **4.4 Yield of DIC from the Indian monsoonal rivers**

The yield of DIC found in this study (mean 8.7±5.2 g m^{-2} yr^{-1}) is similar those found earlier in rivers from the tropical region (30°N – 30°S) of the Asian continent (9.79 g m^{-2} yr^{-1}; Huang et al., 2012), but significantly higher than the American (3.33 g m^{-2} yr^{-1}) and African (0.63 g m^{-2} yr^{-1}) continents of this tropical region (Huang et al., 2012). The yield of DIC from river catchment were reported to be controlled by the hydrological (precipitation, runoff and groundwater exchange) and environmental (temperature, type and dominance of soils, soil organic carbon, natural vegetation and forest cover) conditions and anthropogenic activities (land use change and deforestation) in the catchment (Raymond et al., 2008; Huang et al., 2012). Although the SW estuaries annually export relatively less DIC to the north Indian Ocean (0.30 Tg) due to their lower volume of discharge (46 km^3 yr^{-1}) from relatively smaller catchment area (0.02 M km^2) than the SE (3.50 Tg, 102 km^3 yr^{-1} and 0.43 M km^2 respectively), NE (4.21 Tg, 276 and 0.53) and NW (2.38 Tg, 75 km^3 yr^{-1} and 0.21 M km^2) estuaries, strikingly, the higher yield of DIC was found in the former (10.8±6.6 g m^{-2} yr^{-1}) than the latter (5.8±2.3, 8.6±5.7 and 9.5±3.9 g m^{-2} yr^{-1}, respectively). This suggests that strong control of catchment and/or in-stream processes on yield of DIC from the monsoonal rivers. However, DIC yield showed significant positive correlation with the volume of discharge (r^2=0.66, $p<0.001$) in medium estuaries and no such relationship was found in the major estuaries. Significant negative relationships were observed between DIC yield and catchment area in the medium (r^2=0.52, $p<0.001$) and major estuaries (r^2=0.49, $p<0.001$). This suggests that high precipitation over small catchments increases the DIC yield from the Indian estuaries because the dense precipitation increases the scouring of DIC from soils and rocks in their catchment. A strong linear relationship between the yield of DIC and the intensity of precipitation (r^2=0.64, $p<0.001$ Fig. 8a) confirms that dense precipitation increases the export yield of DIC. This could be one the reasons for the observed

438 higher yield of DIC in the SW estuaries which receives high precipitation (2500±500mm) over
439 the small catchment area (0.02 M km^2).

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

450 Catchments of the SW rivers are largely occupied by the cation deficient lateritic soils
451 and therefore precipitation of carbonate minerals in soils is poor. As a result, the soil inorganic
452 carbon content in surface (100cm) soils of the catchment of SW rivers was lower than in
453 catchments of the other monsoonal rivers studied (Sreenivas et al., 2016). On the other hand, the
454 authors (Sreenivas et al., 2016) and Krishwan et al. (2009) found that the soil organic carbon was
455 higher in the former than the latter. The relationship between soil inorganic and organic carbon is
456 primarily dependent on the soil characteristics in the catchment. For example, Guo et al. (2016)
457 demonstrated that increase in the soil organic carbon content enhanced the soil inorganic carbon
458 in the cropland of upper Yellow river delta, China. A strong positive relationship between soil
459 organic and inorganic carbon was also found in the Yanqi river basin, northwest China (Wang et
460 al., 2015), and soils in the America (Stevenson et al., 2005) and Canada (Landi et al., 2003). On

the other hand, a negative relationship was found between soil organic and inorganic carbon in the North China Plain (Huang et al., 2006) and west Loess Plateau (Zeng et al., 2008). The negative relationship is mainly due to the higher production of CO_2 by decomposition of soil organic carbon and root respiration resulting in the formation of acidic conditions that lead to dissolution of soil carbonates. The higher soil organic carbon in the catchment of the SW than in catchment of the SE, NE and NW rivers (Kishwan et al., 2009; Sreenivas et al., 2016) therefore, produces more CO_2 through microbial decomposition and causes dissolution of soil carbonates leading to the higher yield of DIC from the SW estuaries. A significant linear correlation between soil organic carbon content and DIC yield in this study (r^2=0.65, $p<0.001$; Fig. 8b) suggests that strong influence of soil organic carbon content in the catchment on DIC yield from the Indian monsoonal rivers. However, basin scale studies are required for comprehensive understanding of the influence of environmental and anthropogenic factors on DIC export fluxes from the Indian monsoonal rivers.

5. Summary

In order to examine the variability of dissolved inorganic carbon (DIC) concentrations and to identify its major sources in the Indian monsoonal estuaries, and to estimate the riverine export fluxes of DIC to the north Indian Ocean, we sampled a total of 27 major and medium estuaries along the Indian coast during wet period. An order of magnitude variability was found in DIC concentrations among the estuaries sampled (3.4 - 44.1mg l^{-1}), with a lower mean concentrations of 6.6±2.1 mg l^{-1} in estuaries located in the SW region of India. It is attributed to significant variability in the size of rivers, precipitation pattern and lithology in their catchments. Magnitude of discharge, catchment area and in-stream processes are appears to be important factors for medium estuaries rather than major estuaries in controlling the concentration and

yield of DIC, probably due to a significant variability in lithology and hydro-geological and environmental conditions in the catchments. Indian monsoonal estuaries annually export ~10.4 Tg of DIC to the north Indian Ocean, of which 7.7 Tg enters in to the Bay of Bengal while the Arabian Sea receives only 2.7 Tg. It is mainly attributed to the volume of river discharge as former receives ~378 km^3 yr^{-1} while the latter receives only 122 km^3 $yr^{-1}$of freshwater from the Indian monsoonal rivers. The range of $\delta^{13}C_{DIC}$ found in this study suggests that DIC is largely contributed from weathering of silicate and carbonate minerals by carbonic acid formed by dissolution of both soil and atmospheric CO_2. However, relatively enriched $\delta^{13}C_{DIC}$ in the east-flowing river estuaries indicated the storage of water in dams/reservoirs and intrusion of marine waters. Dense rainfall (2500±500mm) and higher soil organic carbon content (101.4 g ha^{-1}) in the catchment of SW rivers than in the catchment of the other rivers resulted in higher yield of DIC from the former than the latter.

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

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

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

Biogeosciences Discuss., https://doi.org/10.5194/bg-2018-4
Manuscript under review for journal Biogeosciences
Discussion started: 9 January 2018

947 **Figure 4:** (a) Positive correlation between dissolved inorganic carbon (DIC) concentration and
948 catchment area, and (b) negative correlation between DIC concentrations and annual mean
949 discharge (km^3) of the minor rivers.
950
951 **Figure 5:** Inverse correlation between mean dissolved inorganic carbon concentration in
952 estuaries (DIC, mg l^{-1}) and annual mean rainfall (mm) in catchments of the rivers in the NE, NW,
953 SE and SW regions of India.
954
955 **Figure 6:** Significant positive correlation between stable carbon isotopes of dissolved inorganic
956 carbon ($\delta^{13}C_{DIC}$, ‰) and salinity in the Indian monsoonal estuaries during the study period.

957 **Figure 7:** Significant positive correlation between stable carbon isotopes of dissolved inorganic
958 carbon ($\delta^{13}C_{DIC}$, ‰) and concentrations of DIC in the Indian monsoonal estuaries (filled
959 diamonds), SW estuaries (filled squares) and high saline estuaries (hollow triangles) during the
960 study period.
961
962 **Figure 8:** Relationship of dissolved inorganic carbon (DIC) yield (g m^{-2} yr^{-1}) with that of (a)
963 rainfall (mm) and (b) soil organic carbon (kg ha^{-1}) in the catchment area of the NE, NW, SE and
964 SW rivers
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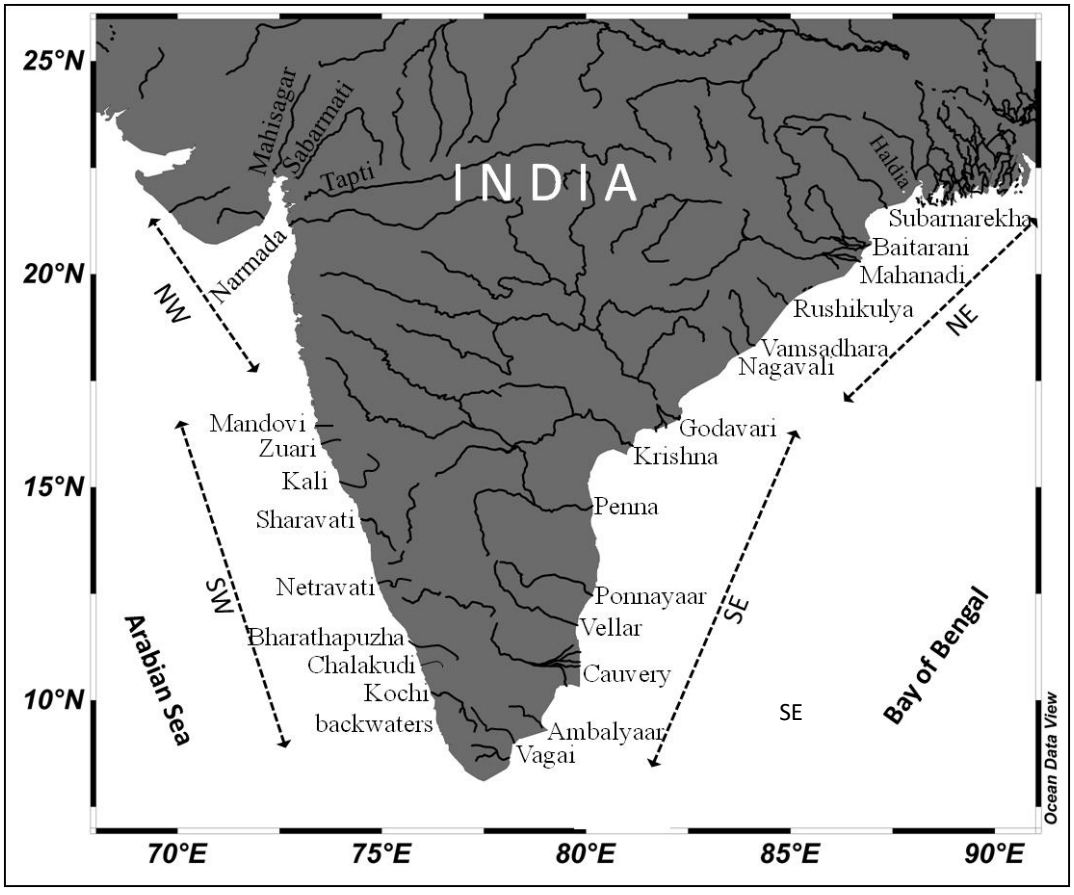

Fig.1:

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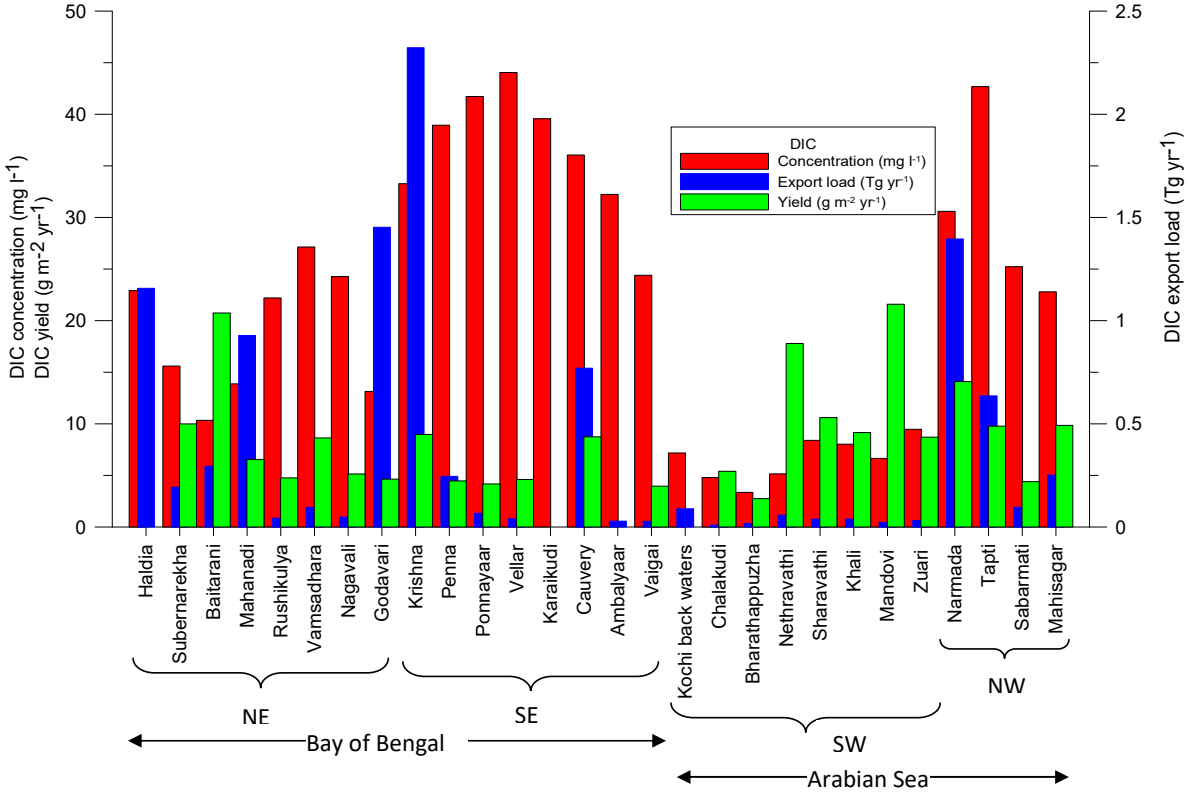

Fig. 2

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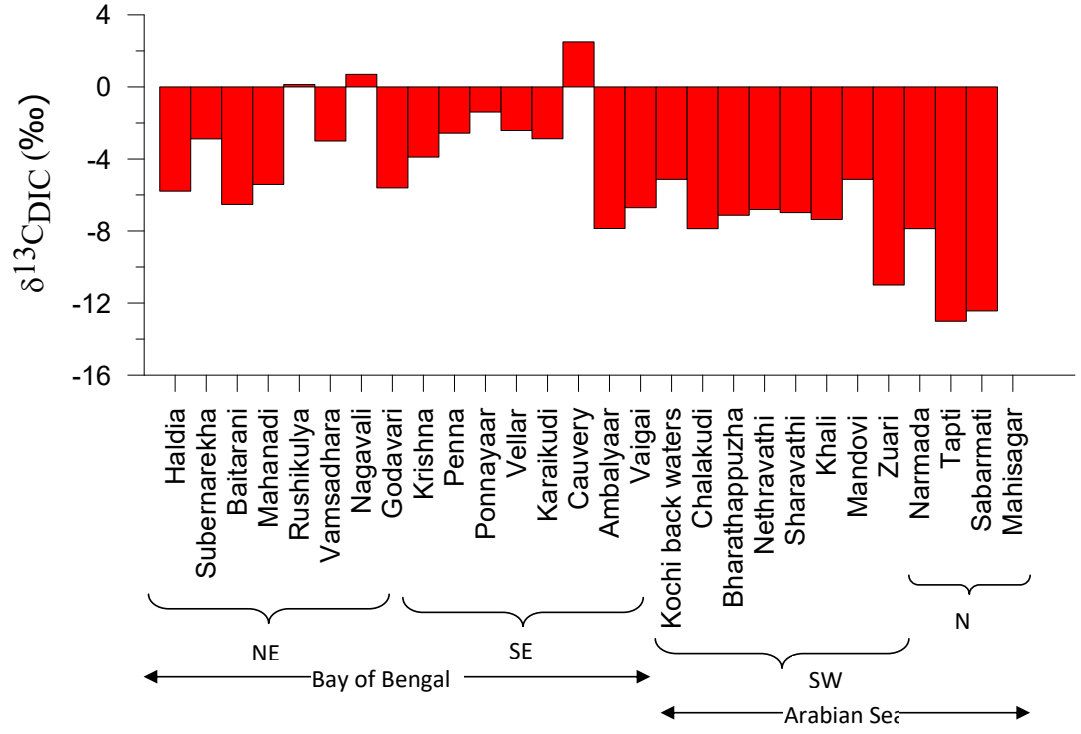

Fig. 3:

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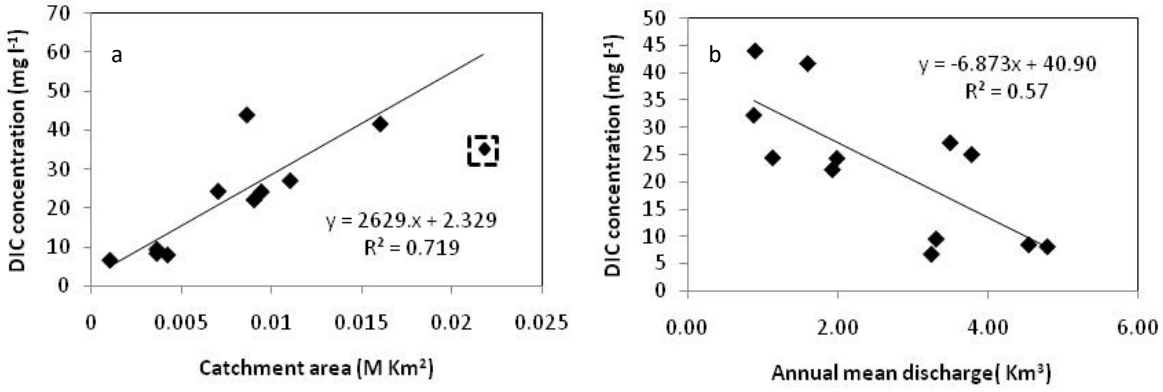

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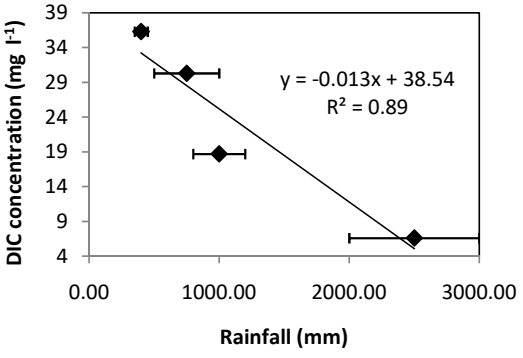

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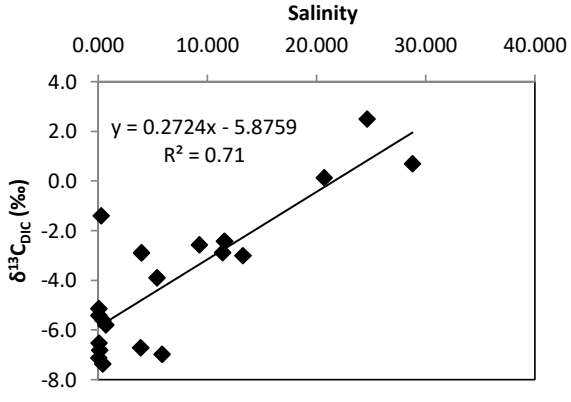

Fig. 6:

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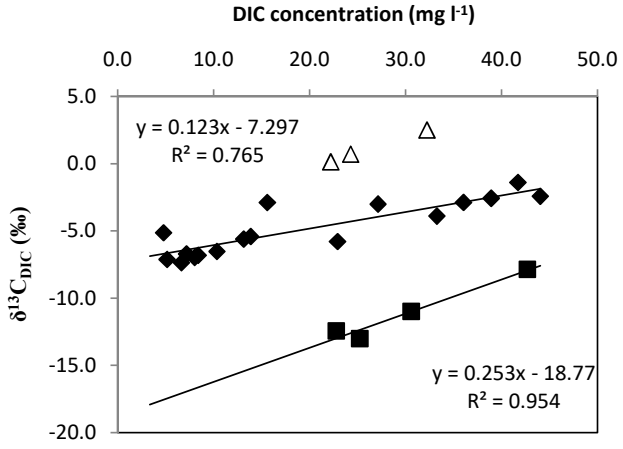

Fig. 7:

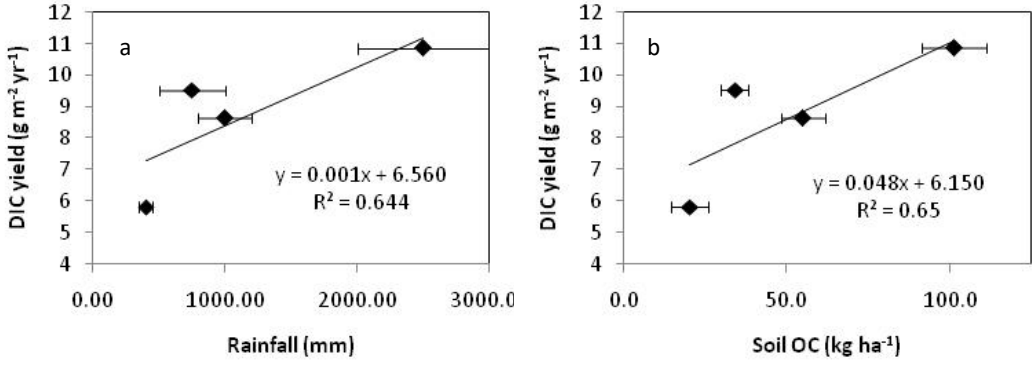

Fig. 8: