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

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Abstract. Rivers are an important 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 rivers and to quantify their export flux to the north Indian Ocean, 27 major and medium rivers were sampled during the discharge period. Significant spatial variability in concentrations of DIC (3.4 – 73.6 mg l^{-1}) was observed and it is attributed to spatial variations in the precipitation pattern, size of rivers, pollution, and lithology of the catchments. The stable isotopic composition of DIC ($\delta^{13}C_{DIC}$) also showed strong spatial variability (-13.0 to -1.4‰) in the Indian monsoonal rivers with relatively depleted $\delta^{13}C_{DIC}$ values in rivers of the northwestern of India (-11.1±2.3‰) and enriched values in the southeastern India (-3.5±2.3‰). Results of the least square linear regression models of Keeling and Miller-Tans plots indicated that chemical weathering of carbonate and silicate minerals by soil CO_2 is the major source of DIC in the Indian monsoonal rivers. Spatial variability in the deviation of $\delta^{13}C_{DIC}$ from the approximated $\delta^{13}C$ of source may probably due to dominant autotrophic production in rivers of the southeast region whereas, heterotrophic decomposition of organic matter largely influences in the other Indian monsoonal rivers. It is estimated that the Indian monsoonal rivers annually export ~10.3 Tg of DIC to the northern Indian Ocean, of which the major fraction (75%) 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 to the Bay of Bengal (~378 km^3 yr^{-1}) than to the Arabian Sea (122 km^3 yr^{-1}). Despite discharge from the 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. Despite rivers from the SW region of India export an order of magnitude lower DIC (0.3 Tg yr^{-1}) than the rivers from other regions of India, the highest yield of DIC was found in the former and it 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 rivers from SW region.

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 (weathering of carbonate and silicate rocks) and biogenic (decomposition of organic matter in soils) sources (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 in 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. 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 are strongly coupled to atmospheric CO_2 because they act as either sink or source of atmospheric CO_2 (e.g. Berner et al., 1983; Mook and Tan, 1991; Gaillardet et al., 1999; Richey et al., 2002). The DIC in

rivers and its export to the coastal oceans is thus intimately linked to the global carbon cycle (Campeau et al., 2017)

Riverine export fluxes of DIC to coastal regions of the world oceans have been estimated on the global (Gaillardet et al., 1999) 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 world major river systems to the gloabal ocean has been estimated as ~327 - 385 Tg ($1Tg=10^{12}g$) (Ludwig et al., 1998; Meybeck and Vorosmarty, 1999). However, many of the regional studies on DIC export fluxes were limited only to the major river systems (e.g. Gaillardet et al., 1999; Joesoef et al., 2017), 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 the regional scales. However, DIC measurements are still lacking in several medium rivers from different regions of the world in general and Asia in particular.

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 the 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.

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). 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 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 $\delta^{13}C$of DIC derived from different sources is well separable (Deines et al., 1974), the isotopic fractionation by in-stream physical and biological processes alters the $\delta^{13}C$ of DIC source (Fig. 2). For example, photosynthesis and degassing enriches (O'Leary, 1988; Finlay, 2004; Parker et al., 2005, 2010; Polsenaere and Abril, 2012; Venkiteswaran et al., 2014) 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). Though, 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), the isotopic equilibration between the DIC and CO_2 in the atmosphere significantly influences the $\delta^{13}C_{DIC}$ in rivers (Abongwa and Atekwana, 2014; Deirmendjian and Abril, 2018) due to

104 selective fluxes of $^{12}CO_2$ and $^{13}CO_2$ at the water-air interface. Hence, the influence of
105 biogeochemical processes within the rivers must be considered while interpreting the $\delta^{13}C_{DIC}$
106 results for identification of DIC sources. The main objectives of this study are to (i) identify
107 the major sources of DIC in the Indian monsoonal rivers, (ii) estimate the export flux and
108 yield of DIC to the north Indian Ocean and (iii) examine the major processes in the
109 catchments and within the rivers controlling DIC in the Indian monsoonal rivers.

110 **2. Study region, sampling and methods**

111 **2.1 Study Area**

112 The Indian peninsula bifurcates the north Indian Ocean into the Bay of Bengal and the
113 Arabian Sea. Although these two basins occupy the same latitudinal belt, their
114 oceanographic processes were reported to be remarkably different due to higher freshwater
115 flux into the Bay of Bengal (1.63×10^{12} m^3 yr^{-1}) than to the Arabian Sea (0.3×10^{12} m^3 yr^{-1};
116 Subramanian, 1993; Gauns et al., 2005). The large freshwater influx leads to the formation
117 of a strong vertical salinity stratification in the Bay of Bengal (Varkey et al., 1996) that
118 prevents vertical mixing of nutrient rich sub-surface water with the surface (Prasanna Kumar
119 et al., 2004). As a result, the Bay of Bengal is considered to be relatively less productive
120 (Prasannakumar et al., 2002) than the adjacent Arabian Sea, which is one of the highly
121 productive zones in the world (Madhupratap et al., 1996; Smith, 2001; Barber et al., 2001)
122 due to injection of nutrients into the surface through the seasonal upwelling and convective
123 mixing (Shetye et al., 1994; Madhupratap et al., 1996; Muraleedharan and Prasannakumar,
124 1996).

125 Discharge from the Indian monsoonal rivers is largely fed by the monsoon induced
126 precipitation over the Indian subcontinent, which receives >80% of its annual rainfall during
127 the southwest (SW) monsoon period (June-September) (Soman and Kumar, 1990). Though
128 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 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, rivers sampled in the present study were categorized into 4 groups, namely the northwest (NW), southwest (SW), southeast (SE) and northeast (NE) 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 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 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 rivers in this study were divided into two types, namely, the major (>150 $m^3 s^{-1}$) and medium (<150 $m^3 s^{-1}$) rivers.

2.2 Sample collection

Water samples were collected from the freshwater regions of the estuaries 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

154 different years and the mean was used for export flux estimations. Further, samples were
155 collected in mid-stream of the river using a local mechanized boat to avoid the contamination
156 from river banks.

157 *In-situ* measurements and sample collection were conducted in 27 rivers of the Indian
158 subcontinent (Fig. 1) during the SW monsoon season of the years, 2011 and 2014. Surface
159 water samples at each location were collected for phytoplankton biomass (Chl-*a*), DIC and
160 dissolved oxygen (DO). Samples for DIC were collected in air-tight crimp-top glass bottles
161 and added poison (mercuric chloride) to arrest the biological activity. DO analysis was
162 carried out at a temporary shore laboratory set up for sample processing after the completion
163 of sampling on each day. Water samples were filtered through GF/F (nominal pore size of
164 0.7μm) under moderate vacuum and stored in liquid nitrogen for Chl-*a* analysis.

165 **2.3. Methods**

166 Temperature and salinity at the sampling locations were measured using a
167 conductivity-temperature-density (CTD) profiling system (Sea Bird Electronics, SBE 19 plus,
168 United States of America). Concentration of DO was determined by a Winkler's method
169 (Carritt and Carpenter, 1966) using an auto titrator (Metrohm, Switzerland) with
170 potentiometric end point detection. The analytical precision of the method was ±0.07%
171 (RSD). Dissolved oxygen saturation is computed following formulations given by Garcia and
172 Gordon (1992). DIC concentrations in water samples were measured at our Institute
173 laboratory using Coulometer (UIC Inc., USA) connected to an automatic sub-sampling
174 system. Based on the repeated analysis of samples and standards, the precision of the method
175 was ±0.02 mg l^{-1}. The certified reference materials (CRM) supplied by Dr. A.G. Dickson,
176 Scripps Institute of Oceanography, USA and internal standards were used to test the accuracy
177 of our DIC measurements and it was found to be within ± 0.2 to 0.3%.. Potetiometric Gran
178 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 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 at near zero salinity (river end member) with the annual discharge.

205 Spatial variability of DIC concentrations within the river was minimized to a large extent by
206 collecting samples from 2 to 3 locations in each river while the inter-annual variability by
207 collecting samples during discharge periods of two years. However, variability in DIC
208 concentrations within the discharge period results in some uncertainties in our estimations of
209 DIC export fluxes. Time series measurements in the Godavari estuary (our unpublished
210 results) revealed that the variability in DIC concentrations within the discharge period is up to
211 10%. Therefore, the error associated with our DIC flux estimates may be about 10%. DIC
212 flux normalized by catchment area (yield) was calculated by dividing the total DIC export
213 flux of the river by its catchment area.

214 **3. Results**

215 **3.1. Hydrographic characteristics**

216 Surface water temperatures were higher in rivers from the NE and SE regions (mean
217 30.9±1.2^{o}C) than the rivers from SW and NW regions (27.3±1.5^{o}C) of India Dissolved
218 oxygen saturation varied from as low as 63% to as high as 105%, with a mean saturation of
219 90±11%. The rivers from SW region of India recorded more under-saturation of DO (82±7%)
220 than the rivers located in the NE (89±15%), NW (93±3%) and SE (96±11%) regions of India.
221 Chlorophyll-*a* (Chl-*a*) concentrations varied broadly from 0.8 to 7.5 mg m^{-3}, with relatively
222 higher mean concentrations in rivers of the SE region (4.7±2.5 mg m^{-3}) followed by the SW
223 (2.8±0.7 mg m^{-3}) regions of India. On the other hand, relatively low Chl-*a* was observed in
224 the medium (2.6±1.3 mg m^{-3}) than the major estuaries (3.2±2.1 mg m^{-3}).

225 **3.2 DIC concentrations and $\delta^{13}C_{DIC}$**

226 DIC concentrations in the Indian monsoonal rivers widely varied from 3.4
227 (Bharathapuzha) to 73.6 mg l^{-1} (Vellar), with a significant spatial variability (Fig. 3a; Table
228 1). Highest mean DIC concentration was observed in rivers of the SE region (37.4±6.3 mg l^{-1})
229) 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 NW (28.4±8.9 mg l^{-1}) and NE (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}) 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 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 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 -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 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 rivers varied broadly from 0.01 Tg (Chalakudi) to as high as 2.33 Tg (Krishna) (Fig. 3b; Table 1). Among the rivers sampled, rivers of the NE region of India export higher DIC (6.52 Tg yr^{-1}) while the lowest was found from rivers of the SW region (0.24 Tg yr^{-1}) (Table 1). The Indian monsoonal rivers together export about 10.32 Tg yr^{-1} of DIC to the northern Indian Ocean, of which 7.81 Tg (75%) enters into the Bay of Bengal and the remaining into the Arabian Sea (2.51 Tg). The yield of DIC ranged from 2.8 (Bharathapuzha) to 20.7 g m^{-2} yr^{-1} (Baitarani) (3c; Table 1), excluding the exceptionally high yield of 119 g m^{-2} yr^{-1} from Haldia river. The

255 mean yield was found to be more or less similar in rivers from all the four regions of India,
256 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 NE (7.7 g m^{-2} yr^{-1})
257 regions. Despite the export flux of DIC is lowest from rivers of the SW region (0.24 Tg yr^{-1}),
258 interestingly, the yield from rivers of this region is on par with (even slightly higher than) the
259 other Indian monsoonal rivers (Table 1; Fig. 3b&c). Yields of DIC found in this study are
260 similar to those found earlier in rivers elsewhere in the world (Huang et al., 2012).

261 **4. Discussion**

262 **4.1 Distribution of DIC in the Indian monsoonal rivers**

263 Distribution of DIC in the Indian monsoonal rivers showed large spatial variability,
264 with the lowest values in rivers from the SW region of India (Fig. 3a). DIC concentrations in
265 rivers are known to be influenced by the intensity of precipitation over the catchment, basin
266 lithology (Giesler et al., 2013; Lofgren et al., 2014), length of the fluvial network (Hotchkiss
267 et al., 2015) and in-stream physical and biological processes (Mook and Tan, 1991; Raymond
268 et al., 2008). The spatial distribution of rainfall over the Indian subcontinent
269 (www.imd.gov.in) shows that the SW region receives the highest annual rainfall (~3000 mm)
270 than the rest of India (Soman and Kumar, 1990).

271 The intense precipitation over the SW region is expected to cause higher weathering
272 rates and thus higher DIC in rivers (e.g., Gupta et al., 2011), but lower DIC concentrations
273 were found in rivers of this region. It could be due to the influence of dilution because the
274 dense precipitation over the small catchment area (Table 1) might have diluted DIC
275 concentrations in rivers of this region. In order to understand the influence of the density of
276 rainfall on DIC in rivers, we normalized the volume of discharge from the river with its
277 catchment area. The catchment area normalized volume of discharge was found to be much
278 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
279 (0.6 m^3 m^{-2}) and NW (0.32 m^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. 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.

Rivers of the SW region are relatively small in size, both in terms of catchment area (total catchment area: $20x10^3$ km^2) and the length of the river (mean length: 126 km), than the rivers from other regions (SE, NE and NW) of India (Table 1). 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. 4b) also support this argument.

The major physical and biological processes controlling DIC concentrations in rivers are the exchange of CO_2 with the atmosphere, 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 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). 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; Bouillon 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.

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). 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. A significant positive correlation was found between total

alkalinity (TA) and $\delta^{13}C_{DIC}$ (r^2=0.52; p<0.01; Fig. 4h), 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.

4.2 Major sources of DIC in the Indian monsoonal rivers

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 as 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), 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 the measured $\delta^{13}C_{DIC,}$ but correlation coefficient (r^2) is only 0.51 (Fig. 5c), suggesting that significant spatial variability in the influence of in-stream processes on the $\delta^{13}C_{DIC.}$ The Miller-Tans plot of CO_2 ($\delta^{13}C$-CO_2 x CO_2 as a function of CO_2) showed highly significant linear regression model with a slope of -10.7‰ (r^2=0.97; p<0.001; Fig. 5d). These results indicated that chemical weathering of carbonate and silicate minerals by soil CO_2 (-10 to -9‰) is the major source of DIC in the Indian rivers. contributed by. 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 under-saturation

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.

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

Indian monsoonal rivers annually export ~10.3 Tg of DIC to the north Indian Ocean. Nearly three fourth of this amount (7.8 Tg) reaches to the Bay of Bengal while the Arabian Sea receives only one fourth (2.5 Tg). 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 970x10^3 km^2 than the Arabian Sea (122 km^3 yr^{-1} from the catchment area of 244x10^3 km^2). The total DIC export by the Indian monsoonal rivers (10.3 Tg yr^{-1}) is lower 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 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 (400 Tg yr^{-1}). 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) of this region for higher dissolution.

Krishna et al. (2015) reported that Indian monsoonal 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 12.6 Tg yr^{-1} in which DIC flux contributed up to ~81%. The predominance of DIC has also been found in rivers elsewhere in the world, for example, the British rivers (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 $1x10^3$ km^2 to as high as $313x10^3$ 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 DIC export to the north Indian Ocean.

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 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). The yield was highest (8.8±5.6 g m^{-2} yr^{-1}) in rivers from the SW region of India, despite they export relatively lower DIC (0.3 Tg yr^{-1}) due to their low volume of discharge (46 km^3 yr^{-1}) and relatively smaller catchment ($20x10^3$ km^2) than the rivers from SE, NE and NW regions of India (Table 1. DIC yield showed a significant positive correlation with the volume of discharge (r^2=0.67, $p<0.001$; Fig. 6a) in medium rivers and no such relationship was found in the major rivers. Significant negative relationship was observed between DIC yield and catchment area of river (r^2 = -0.49, $p<0.001$; Fig. 6b and r^2 = -0.43, $p<0.001$; Fig. 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 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 (~3000 mm) over the small catchment ($20x10^3$ km^2) could have increased DIC yield from the rivers of SW region of India.

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 rivers in the SW region were higher and lower, respectively, than the catchments of the rivers from SE, SW and NE region of India. Decomposition of soil organic matter releases excess CO_2 and, the increase in soil CO_2 leads to the formation of acidic conditions in soils. This would increase the DIC yield by more dissolution of soil carbonates and chemical weathering of carbonate and silicate rocks (Zou et al., 2016). A significant linear correlation was found between soil organic carbon content and DIC yield in this study (r^2=0.65, p<0.001; Fig. 6e), suggesting that higher soil organic carbon in the catchment of the rivers from SW region could have elevated the yield of DIC from rivers of this region. The basin scale studies are, however, required for comprehensive understanding of the influence of environmental and anthropogenic factors on export fluxes and yield of DIC from the Indian monsoonal rivers.

5. Summary

In order to examine the spatial variability in the sources and distribution of dissolved inorganic carbon (DIC) in the Indian monsoonal rivers, and to estimate their export fluxes of DIC to the north Indian Ocean, we sampled a total of 27 major and medium rivers during wet period. An order of magnitude variability was found in DIC concentrations among the rivers sampled (3.4 - 73.6 mg l^{-1}), with a lower mean concentration of 6.6±2.1 mg l^{-1} in rivers

453 located in the SW region of India. It is attributed to significant spatial variability in the size
454 of rivers, precipitation pattern, pollution and lithology in their catchments. The approximated
455 $\delta^{13}C$ of DIC source from the Keeling and Miller-Tans plots (-2.0 and -3.0‰ respectively)
456 and, the calculated $\delta^{13}C$ of CO_2 suggested that DIC in the Indian rivers is mainly originated
457 from chemical weathering of carbonate minerals, but largely affected by autotrophic
458 production in rivers from the southeast region of India and heterotrophic decomposition of
459 organic matter in rivers from other regions of India. Indian monsoonal rivers together export
460 ~10.3 Tg yr^{-1} of DIC to the north Indian Ocean, of which 7.8 Tg yr^{-1} enters in to the Bay of
461 Bengal while the Arabian Sea receives only 2.5 Tg yr^{-1}. It is mainly attributed to the volume
462 of river discharge as the former receives ~378 km^3 yr^{-1} while the latter receives only 122 km^3
463 yr^{-1} from the Indian monsoonal rivers. Dense rainfall and higher soil organic carbon content
464 in the catchment of rivers from the SW region than in the catchment of the other Indian rivers
465 resulted in highest yield of DIC from the former than the latter.

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473 **7. Data Availability**

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

476 **References**

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Figure captions

Figure 1: Map showing the study region. Rivers sampled in this study were indicated by solid black line. Distribution of soils in catchments of the Indian monsoonal rivers sampled

was also shown. Rivers draining the four regions, i.e., northwest (NW), southwest (SW), southeast (SE) and northeast (NE) were shown by solid black arrows.

Figure 2: Schematic diagram showing the typical range of $\delta^{13}C$ of different sources of dissolved inorganic carbon (DIC) in rivers. Various major processes influencing the $\delta^{13}C$ of DIC ($\delta^{13}C_{DIC}$) within the rivers were also shown. Hollow 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 3: Spatial variability in the concentration (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 rivers studied. Rivers geographically located in the northwest (NW), southwest (SW), southeast (SE) and northeast (NE) regions of India were also shown. Rivers draining into the Bay of Bengal (east-flowing rivers) were shown by gray shade while rivers draining into the Arabian Sea (west-flowing) were shown by no shade.

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 (DO) saturation and (f) dissolved organic carbon (DOC) concentration. Linear relationships observed between (g) DIC concentrations and population density in the catchment and (h) total alkalinity and $\delta^{13}C$ of DIC in the Indian monsoonal rivers during the study period. Ovals with dashed line indicate the outliers which were not included in the regression equations. Rivers of the northwest region of India showed linear relationships as shown by the other Indian rivers but with a different slope (Fig. f-h)

Figure 5: 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 (Miller-Tans plot) in the Indian monsoonal rivers. (c) Linear relationship between calculated $\delta^{13}C$ of CO_2 and the measured $\delta^{13}C_{DIC}$ and (d) Miller-Tans linear regression model of $\delta^{13}C\text{-}CO_2$ x CO_2 as a function of CO_2 concentration in the Indian monsoonal rivers.

Figure 6: Significant relationships of dissolved inorganic carbon (DIC) yield with (a) river discharge in medium estuaries, (b) catchment areas of the medium rivers, (c) catchment areas of the major rivers, (d) rainfall over the catchment of all the rivers sampled and (e) soil organic carbon (OC) content in catchments of the Indian monsoonal rivers studied. Since the data on soil OC is not available for each watershed (e) was plotted using the available soil OC data on regional scale (NW, SW, SE and NE regions of India), Hence, it contains only four points.

Table captions

Table 1: Catchment area, discharge, length 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. Rivers located in the northern region (north of 16^{o}N) of India were shown by the shaded (grey) area. Of these rivers, Mahisagar, Sabarmati, Tapti and Narmada are located in the northwestern (NW) whereas, rivers from Godavari to Hyadri are located in the northeastern (NE) region of India.

River name	Catchment area ($x10^3$ km^2)	Annual Discharge (km^3)	Length (km)	Popula-tion (No. km^{-2})	No. of major Dams	Eleva-tion (m)	Rain-fall (mm)	pH	DIC Conc. (mg l^{-1})	DIC export flux (Tg yr^{-1})	DIC yield (g m^{-2} yr^{-1})	$\delta^{13}C_{DIC}$ (‰)	Cal. $\delta^{13}C\text{-}CO_2$ (‰)
West flowing rivers (Arabian Sea)													
MAHISAGAR	34.8	11.0	580	507	138	500	785	7.3	22.8	0.3	7.2	-12.4	-21.5
SABARMATI	21.7	3.8	371	1702	62	1173	787	7.2	24.6	0.1	4.3	-13.0	-
TAPTI	65	14.9	724	208	375	752	888	7.1	35.6	0.5	8.2	-7.9	-16.4
NARMADA	99	45.6	1312	184	281	1317	1120	7.5	30.6	1.4	14.1	-11.0	-
ZUARI	1	3.2	34	92	3	-	3500	7.0	4.8	0.0	4.4	-5.1	-13.2
MANDOVI	3.6	3.3	81	62	2	600	3500	6.5	4.5	0.0	14.5	-7.4	-13.6
KHALI	4.2	4.8	184	111	6	600	3200	7.2	5.7	0.0	6.5	-7.0	-15.5
SHARAVATHI	3.6	4.5	128	109	3	700	4000	6.5	8.4	0.0	10.6	-6.8	-13.1
NETRAVATHI	3.2	11.1	103	103	-	1000	3923	6.3	5.1	0.1	17.8	-7.1	-12.4
BHARATHAPUZHA	6.2	5.1	209	-	13	1964	2500	6.0	3.4	0.0	2.8	-7.9	-11.6
CHALAKUDY	1.7	1.9	144	-	6	1250	3600	6.3	4.8	0.0	5.4	-5.1	-10.3
East flowing rivers (Bay of Bengal)													
VAIGAI	7	1.1	258	499	2	1200	850	-	26.0	0.0	4.2	-7.9	-
AMBALAYAAR	-	0.9	-	-	-	-	-	8.6	32.2	0.0	-	-	-
CAUVERY	88	21.3	800	393	122	1341	1075	7.5	40.5	0.9	9.8	-2.9	-11.8
VELLAR	8.6	0.9	210	457	3	900	980	7.4	73.6	0.1	7.7	-2.4	-11.1
PONNAIYAR	16	1.6	396	291	4	900	969	7.4	43.4	0.1	4.3	-1.4	-10.3
PENNA	55	6.3	597	186	61	1439	510	8.3	37.1	0.2	4.3	-2.6	-11.9
KRISHNA	259	69.8	1300	260	736	1903	784	8	33.5	2.3	9.0	-3.9	-
GODAVARI	313	110.5	1465	193	978	1067	1300	-	13.1	1.5	4.6	-5.6	-9.6
NAGAVALI	9.4	2.0	256	150	4	1300	1000	-	15.5	0.0	3.3	-	-13.1
VAMSADHARA	11.0	3.5	254	130	3	370	1400	8.2	27.2	0.1	8.6	-3.0	-12.4
RUSIKULYA	9.0	1.9	175	360	13	1000	1000	-	19.8	0.0	4.2	-	-
MAHANADI	141.6	66.9	858	282	280	890	1406	6.9	13.3	0.9	6.3	-5.4	-13.2
BAITARANI	14.2	28.5	414	324	8	900	1450	6.0	10.3	0.3	20.7	-6.5	-10.2
SUBARNALEKHA	29.2	12.4	395	338	12	600	1800	8.0	13.6	0.2	5.8	-2.9	-12.0
HYADRI	10.2	50.5		191				7.1	24.1	1.2	-	-5.8	-14.0

Table 1

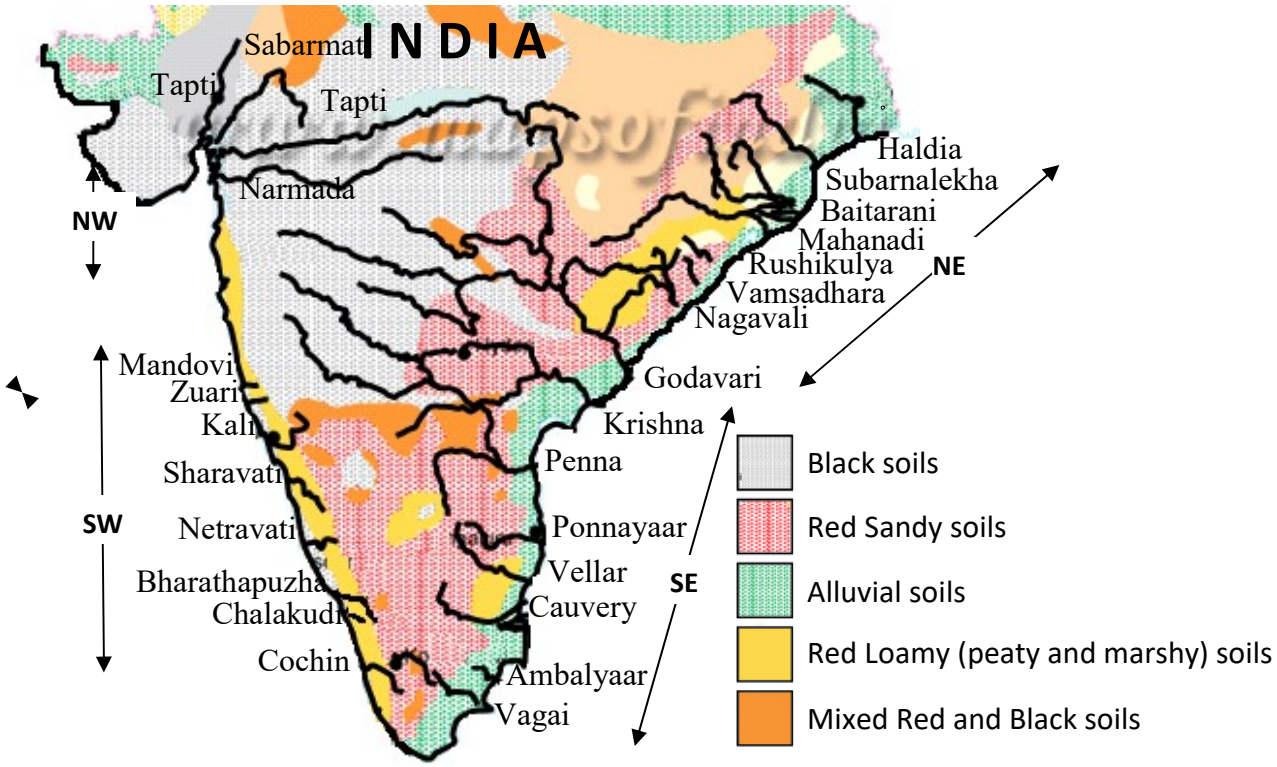

INDIA
Sabarmati
Tapti
Tapti
Narmada
NW
Haldia
Subarnalekha
Baitarani
Mahanadi
Rushikulya
Vamsadhara
Nagavali
NE
Mandovi
Zuari
Kali
Sharavati
Netravati
Bharathapuzha
Chalakudi
Cochin
SW
Godavari
Krishna
Penna
Ponnayaar
Vellar
Cauvery
Ambalyaar
Vagai
SE
Black soils
Red Sandy soils
Alluvial soils
Red Loamy (peaty and marshy) soils
Mixed Red and Black soils

Fig. 1

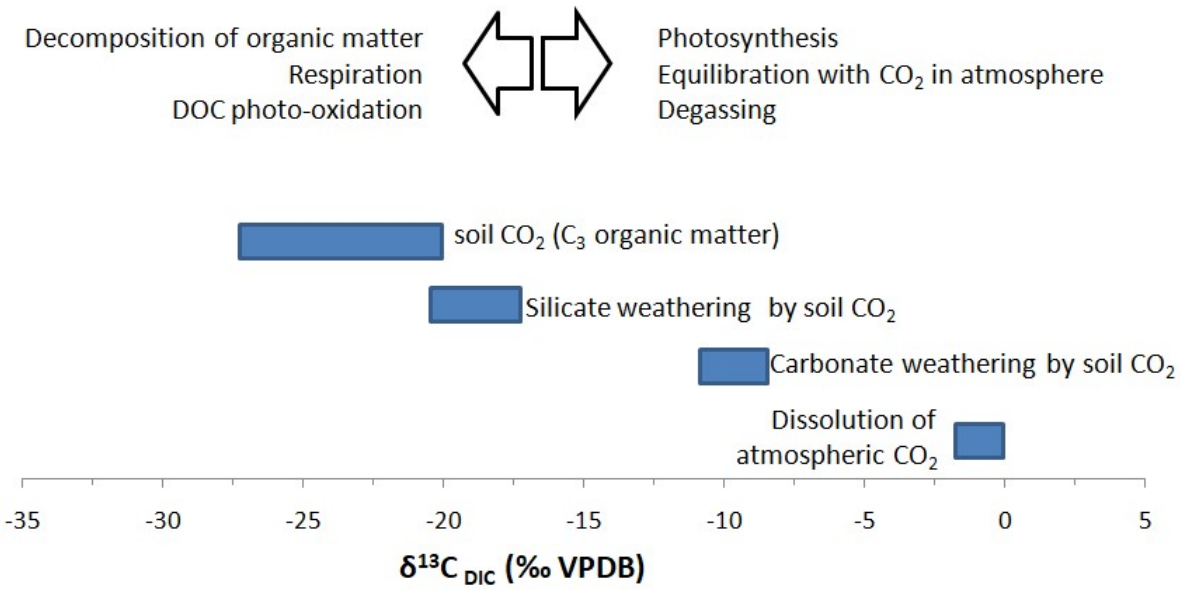
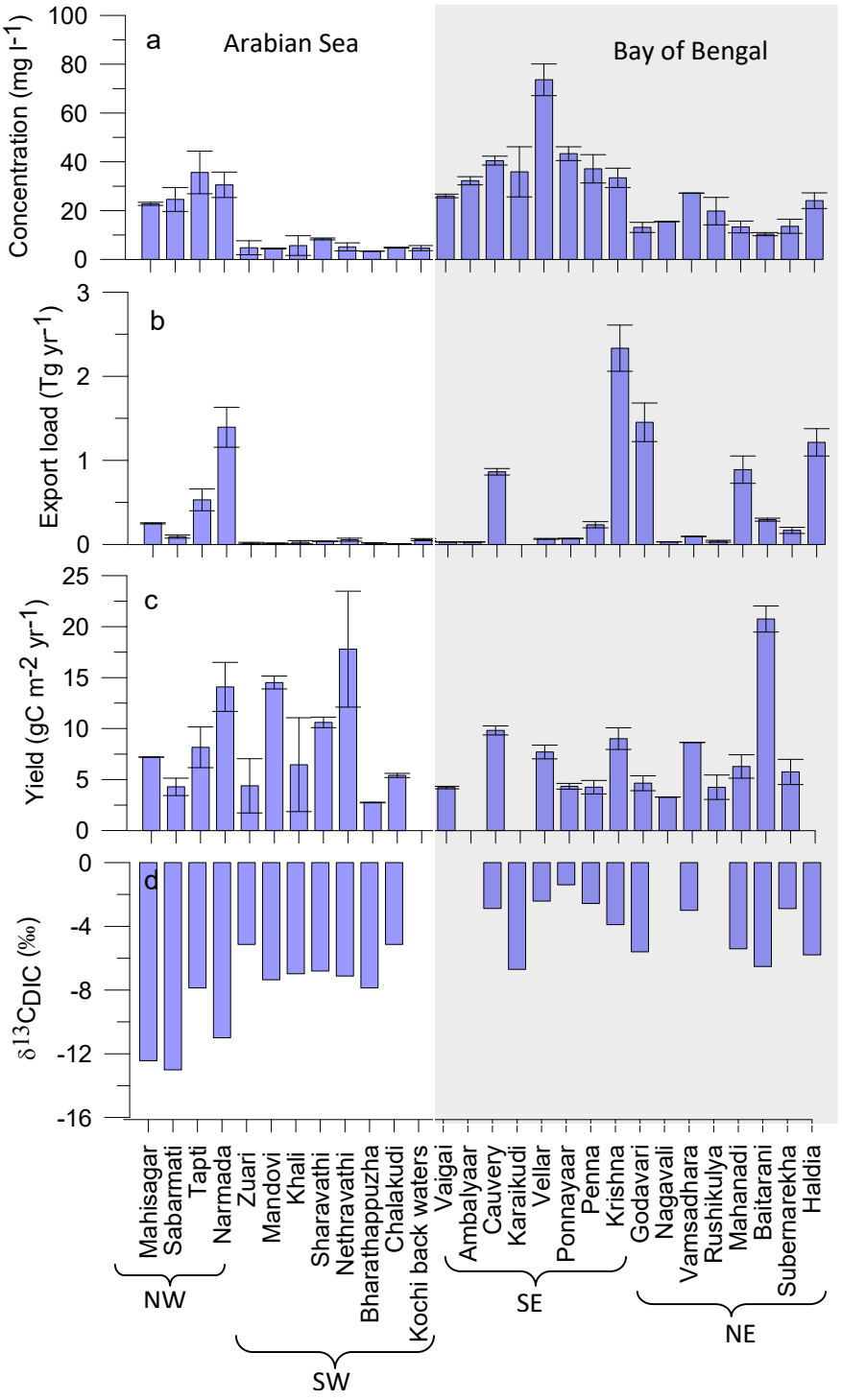

Decomposition of organic matter
Respiration
DOC photo-oxidation
Photosynthesis
Equilibration with CO_2 in atmosphere
Degassing
soil CO_2 (C_3 organic matter)
Silicate weathering by soil CO_2
Carbonate weathering by soil CO_2
Dissolution of atmospheric CO_2
-35
-30
-25
-20
-15
-10
-5
0
5
$\delta^{13}C_{DIC}$ (‰ VPDB)

Fig. 2


a
Arabian Sea
Bay of Bengal
Concentration (mg l-1)
100
80
60
40
20
0
b
Export load (Tg yr-1)
3
2
1
0
c
Yield (gC m-2 yr-1)
25
20
15
10
5
0
d
δ13CDIC (‰)
0
-4
-8
-12
-16
Mahisagar
Sabarmati
Tapti
Narmada
Zuari
Mandovi
Khali
Sharavathi
Nethravathi
Bharathappuzha
Chalakudi
Kochi back waters
Vaigai
Ambalyaar
Cauvery
Karaikudi
Vellar
Ponnayaar
Penna
Krishna
Godavari
Nagavali
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Rushikulya
Mahanadi
Baitarani
Subernarekha
Haldia
NW
SW
SE
NE

Fig. 3

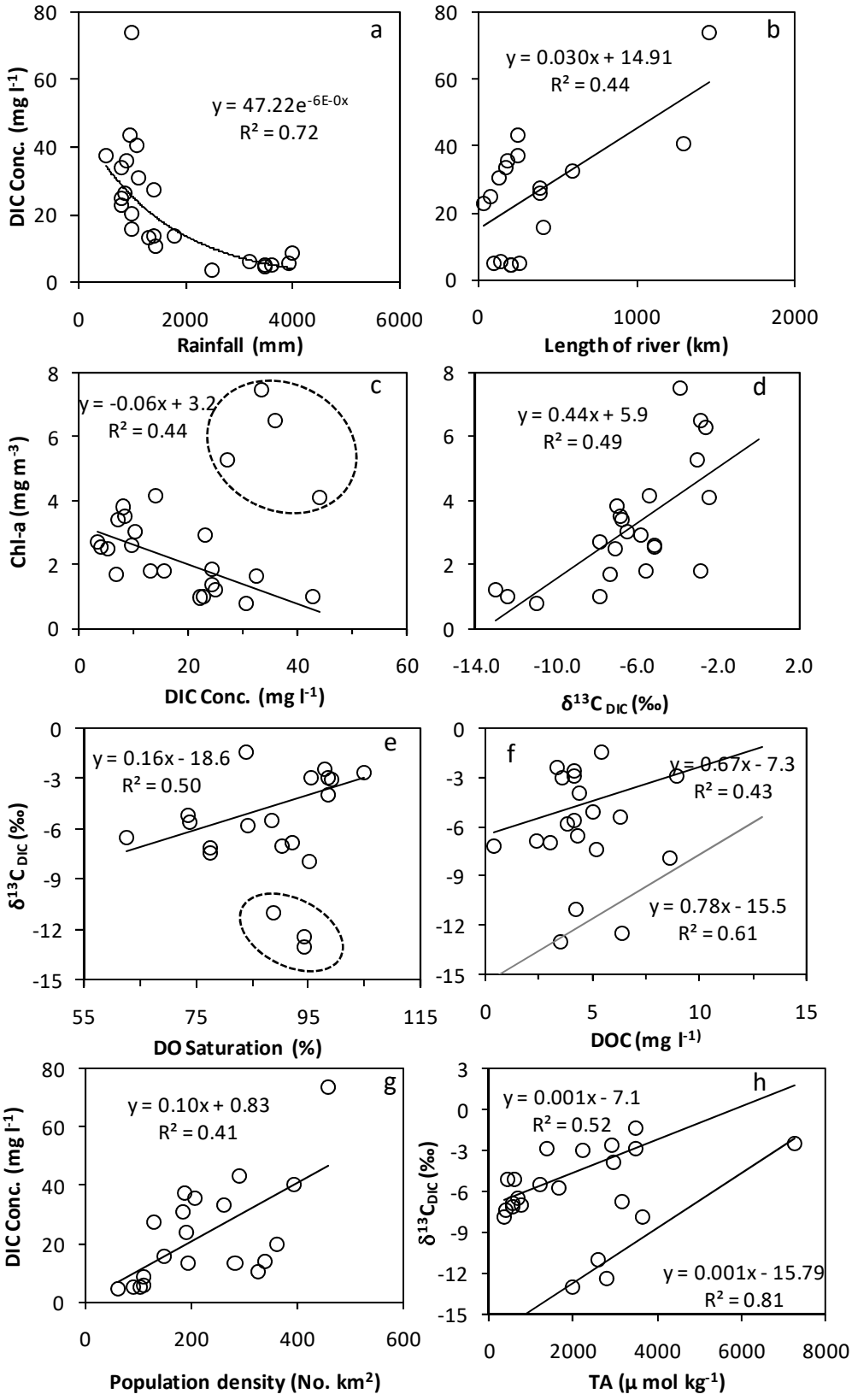
a
y = 47.22e-6E-0x
R² = 0.72
DIC Conc. (mg l-1)
Rainfall (mm)
b
y = 0.030x + 14.91
R² = 0.44
Length of river (km)
c
y = -0.06x + 3.2
R² = 0.44
Chl-a (mg m-3)
DIC Conc. (mg l-1)
d
y = 0.44x + 5.9
R² = 0.49
δ13C DIC (‰)
e
y = 0.16x - 18.6
R² = 0.50
δ13C DIC (‰)
DO Saturation (%)
f
y = 0.67x - 7.3
R² = 0.43
y = 0.78x - 15.5
R² = 0.61
DOC (mg l-1)
g
y = 0.10x + 0.83
R² = 0.41
DIC Conc. (mg l-1)
Population density (No. km²)
h
y = 0.001x - 7.1
R² = 0.52
y = 0.001x - 15.79
R² = 0.81
δ13C DIC (‰)
TA (µ mol kg-1)

Fig. 4

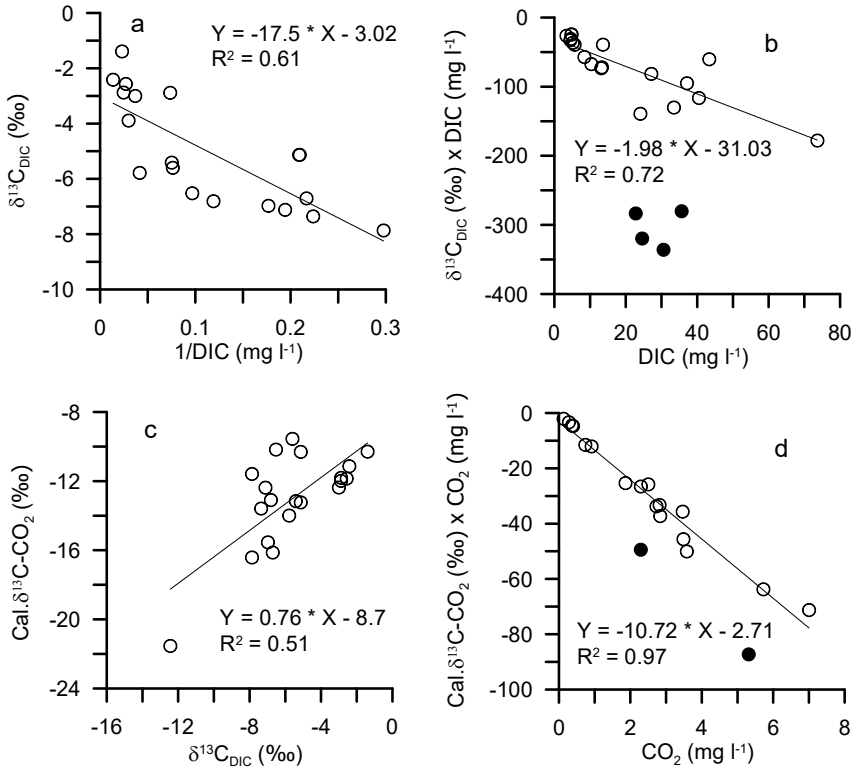
a
Y = -17.5 * X - 3.02
R² = 0.61
δ13CDIC (‰)
1/DIC (mg l-1)
b
Y = -1.98 * X - 31.03
R² = 0.72
δ13CDIC (‰) x DIC (mg l-1)
DIC (mg l-1)
c
Y = 0.76 * X - 8.7
R² = 0.51
Cal.δ13C-CO2 (‰)
δ13CDIC (‰)
d
Y = -10.72 * X - 2.71
R² = 0.97
Cal.δ13C-CO2 (‰) x CO2 (mg l-1)
CO2 (mg l-1)

Fig. 5

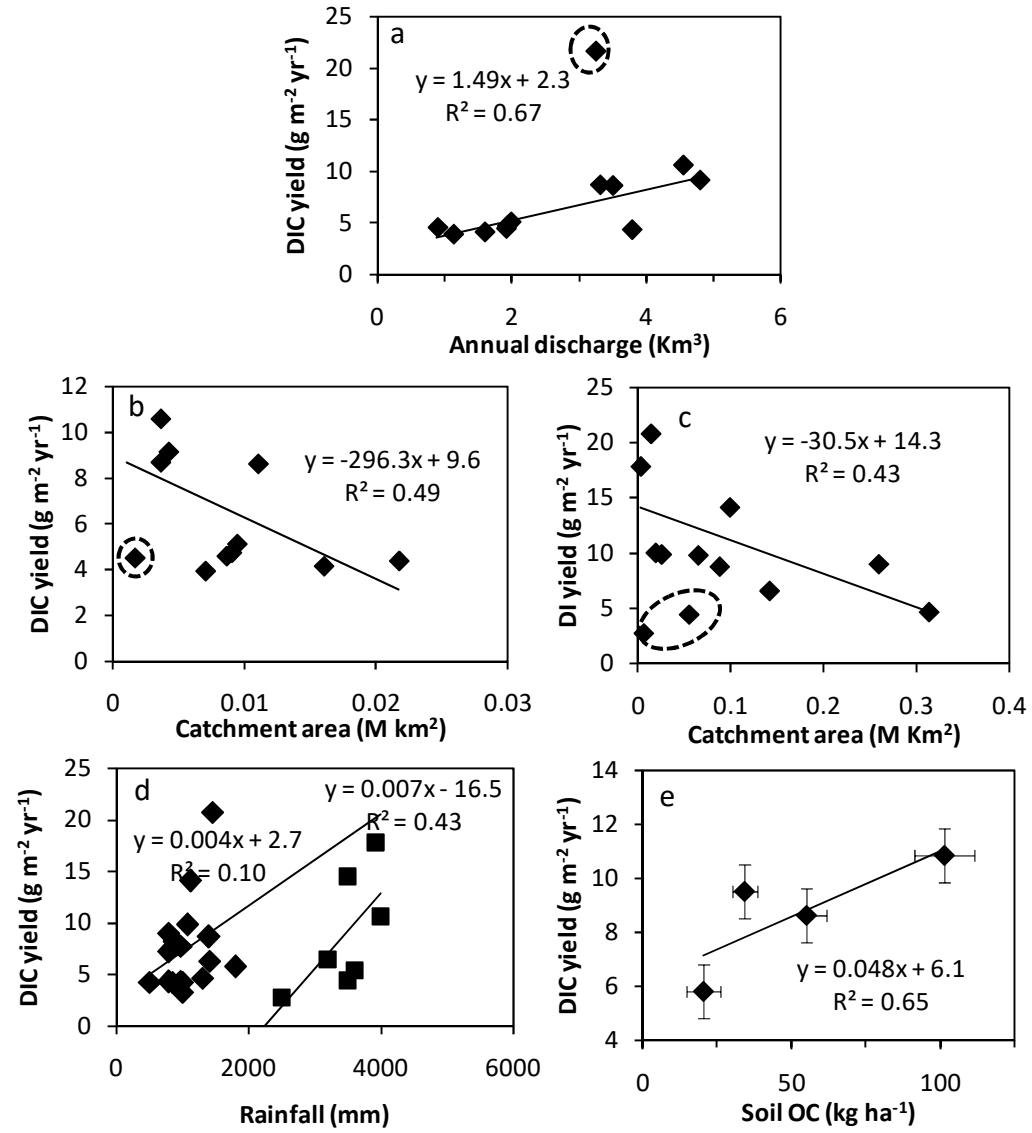
a
y = 1.49x + 2.3
R² = 0.67
DIC yield (g m-2 yr-1)
Annual discharge (Km3)
b
y = -296.3x + 9.6
R² = 0.49
DIC yield (g m-2 yr-1)
Catchment area (M km2)
c
y = -30.5x + 14.3
R² = 0.43
DI yield (g m-2 yr-1)
Catchment area (M Km2)
d
y = 0.007x - 16.5
R² = 0.43
y = 0.004x + 2.7
R² = 0.10
DIC yield (g m-2 yr-1)
Rainfall (mm)
e
y = 0.048x + 6.1
R² = 0.65
DIC yield (g m-2 yr-1)
Soil OC (kg ha-1)

Fig. 6: