Export of dissolved inorganic nutrients to the northern Indian Ocean from the Indian monsoonal rivers during discharge period

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Abstract
Coastal regions are highly productive due to the nutrients largely supplied by rivers. To examine the contribution of dissolved inorganic nutrients (DIN) by Indian rivers to coastal waters, data were collected near the freshwater heads of 27 monsoonal rivers of peninsular India during three weeks in late July to mid-August, the middle of the principal runoff period of the southwest monsoon of 2011. Twelve researchers in four groups, equipped with car and portable laboratory equipment, sampled mid-stream of each estuary using mechanized boat, and filtered and partly analyzed the water in the evening. The estimated exports were 0.22±0.05, 0.11±0.03, and 1.03±0.26 Tg yr⁻¹ for dissolved inorganic nitrogen, phosphorus and silicate, respectively. Higher amounts of DIN reach the Bay of Bengal than the Arabian Sea due to the higher volume (~76%) of discharge to the former. In contrast, the export of dissolved inorganic nitrogen is almost same to the Bay of Bengal (0.12±0.03 Tg yr⁻¹) and Arabian Sea (0.10±0.02 Tg yr⁻¹) principally due to the polluted Narmada and Tapti rivers in the northwest. Including input from the glacial rivers, Ganges, Brahmaputra and Indus, it is estimated that the northern Indian Ocean receives ~1.84±0.46, 0.28±0.07 and 3.58±0.89 Tg yr⁻¹ of nitrate, phosphate and silicate, respectively, which are significantly lower than the earlier estimates of DIN export from the Indian rivers based on DIN measured in the mid or upstream rivers. Such low fluxes in this study were attributed to efficient retention/elimination of DIN (~91%) before reaching the coastal ocean. Hence, this study suggests that the importance of sampling locations for estimating nutrient fluxes to the coastal ocean. Riverine DIN export of 1.84±0.46 Tg yr⁻¹ would support 12.2±3.1 Tg C yr⁻¹ of new production in coastal waters of the northern Indian Ocean that results in a removal of 12.2±3.1 Tg atmospheric CO₂ yr⁻¹.

Keywords: export flux, nutrients, rivers, northern Indian Ocean, Bay of Bengal, Arabian Sea
1. INTRODUCTION

River discharge is one of the major sources of dissolved inorganic nutrients (DIN), such as nitrite, nitrate, ammonium, phosphate and silicate to the coastal environment (Dyer, 1973; McClanahan, 1988; Seitzinger et al., 2006; Singh and Ramesh, 2011). The coastal regions also receive DIN by the aerosol deposition (Krishnamurti et al., 1998; Duce et al., 2008; Srinivas et al., 2011; Singh et al., 2012), coastal upwelling (Rixen et al., 2000), eddies (Prasanna Kumar et al., 2004) and ground water exchange (Moore, 1996). Surplus amounts of nutrient supply by the above said processes make the coastal oceans highly productive. However, anthropogenic activities, such as extensive use of fertilizers in agricultural fields, plantation of leguminous plants, deposition of oxidized form of nitrogen (N), and domestic and industrial sewage significantly influences the riverine export of N and phosphorus (P) to rivers, and subsequently to the coastal systems (Lima et al., 2010). For instance, Green et al. (2004) reported that >6-folds increase in the riverine export of dissolved inorganic N to the global ocean from pre-industrial (2.1 Tg yr\(^{-1}\); 1Tg = 10\(^{12}\)g) to the contemporary period (14.5 Tg yr\(^{-1}\)). Further, significant variations in the precipitation pattern (IPCC, 2001) and warming of the earth surface in the next century (Houghton et al., 1996) due to human interference is likely to cause dramatic changes in the riverine DIN loading because of changes in hydraulic residence time and basin temperature (Green et al., 2004) of the rivers. The rapidly increasing N load to estuaries and coastal regions causes several severe environmental problems, including eutrophication (Nixon, 1995; Justic et al., 1995a,b; Cloern, 2001; Rabalais et al., 2002, Lima et al., 2010) and changes in the ecosystem function and community structure (Tilman, 1984; Aber et al., 1993; Bowman et al., 1995).

Studies on concentrations and fluxes of DIN from major rivers to establish a land-sea connection with reference to global biogeochemistry (Giraud et al., 2008) are myriad. In view of the importance and growing demand for the basic data on river discharge, several programmes, such as the Global Water Quality Monitoring Programme of UNEP/WHO/UNESCO/WMO (GEMS/WATER; Fraser et al., 1995), World Register of River Inputs (WORRI), Global River Index (GLORI; Milliman et al., 1995) and GEMS/WATER Global Register of River Inputs (GEMS-GLORI; Meybeck and Ragu, 1996) registered the characteristics of drainage basins and major water quality attributes of discharge from the world major rivers, respectively. Several studies have reported DIN fluxes from major rivers to the global ocean. For example, 30 major rivers in the world transport 0.81 and 6.72 Tg yr\(^{-1}\) of dissolved inorganic P and N, respectively (Meybeck, 1982). Delivery of nutrients to the coastal ocean was also extensively studied in ~200 coastal systems.
around the world during an International programme on Land-Ocean Interactions in the Coastal Zone (LOICZ), a core project of International Geosphere-Biosphere Programme (IGBP). Smith et al. (2003) estimated export loads of DIN from about 165 rivers in the world and Alvarez-Cobelas (2008) reported nutrient fluxes from about 645 rivers worldwide.

On the regional scale, DIN fluxes were reported from several major rivers in the world, for instance, Amazon (Richey and Victoria, 1993), Mississippi (Goolsby et al., 1999; Howarth et al., 1996), Changjiang (Liu and Shen, 2001), European rivers (Billen et al., 2011), Chinese and Korean rivers (Liu et al., 2009) and rivers draining into the eastern Atlantic coast (Canton et al., 2012) and the north Atlantic Ocean (Howarth et al., 1996). However, studies on DIN fluxes to the northern Indian Ocean from the Indian rivers are very limited. Dileep Kumar et al. (1992) reported that the major rivers transport 0.05 and 0.06 Tg yr\(^{-1}\) of inorganic phosphate and nitrate, respectively, to the northern Indian Ocean. Singh and Ramesh (2011) estimated dissolved inorganic N fluxes to the northern Indian Ocean from few major rivers using the data collected by Subramanian (2008) and Lambs et al. (2005). Though some studies reported DIN concentrations in the Indian estuaries, they were confined to either a single or a group of few rivers (Zingde, 1999; 2005; Lambs et al., 2005; Mukhopadhyay et al., 2006; Subramanian, 2008; Sarma et al., 2010; Pednekar et al., 2011; Parab et al., 2013; Dixit et al., 2013; Anand et al., 2014). No efforts have been made so far to examine the spatial variations in nutrients concentrations in the Indian estuaries and their flux to the coastal zones of the Bay of Bengal and Arabian Sea. We made here, the first ever effort to collect DIN data from 27 estuaries in the Indian subcontinent to estimate export fluxes of DIN to the northern Indian Ocean. The objectives of this study are (i) to examine the spatial variability of DIN concentrations in the Indian monsoonal estuaries and (ii) to estimate export fluxes of DIN to the northern Indian Ocean during the southwest monsoon (SWM). Since, we confined to inorganic components only, the dissolved organic nitrogen was not included in the present estimations of dissolved N fluxes to the northern Indian Ocean. India has the second-highest consumption of fertilizers in the world and the excess is expected to end up in the river and then to the coastal oceans (Sarma et al., 2014). About 78% of the fertilizer application occurs in the catchment regions of the Bay of Bengal. Thus, it can be hypothesized that the Bay of Bengal may receive significantly higher input compared to Arabian Sea.

2. STUDY AREA

Unlike Pacific and Atlantic Oceans, the northern Indian Ocean is surrounded by the Asian land masses on the three sides and opened to the south Indian Ocean in the south. The Indian land mass
separates the northern Indian Ocean into the Bay of Bengal and the Arabian Sea which show remarkably different oceanographic features. The Arabian Sea receives relatively less precipitation and total freshwater influx from rivers (0.3x10^{12} \text{ m}^3 \text{ yr}^{-1}), but is known to be one of the highly productive regions in the world (Madhupratap et al., 1996; Smith, 2001; Kumar et al., 2010) due to supply of nutrients through seasonal upwelling and convective mixing. In contrast, the Bay of Bengal receives enormous amount of freshwater (1.63 x 10^{12} \text{ m}^3 \text{ yr}^{-1}; Subramanian, 1993) from the glacial and monsoonal rivers of India, Bangladesh and Myanmar resulting in strong vertical salinity stratification (Varkey et al., 1996) that suppresses vertical mixing resulting in low biological production (Prasannakumar et al., 2002).

Major and medium-sized monsoonal rivers draining into the Bay of Bengal and the Arabian Sea were shown in Figure 1 (Table 1). Discharge from the Indian monsoonal rivers is limited to few months only because these rivers are fed by the SWM induced precipitation over the catchment, unlike the glacial rivers such as Ganges, Brahmaputra, Irrawaddy, Salween and Indus. The glacial rivers are largely fed by melting of glaciers and hence, discharges continuously. However, glacial rivers also show remarkably high discharge (50% of the annual discharge) during SWM (June – September) due to the influence of monsoonal precipitation over the catchments (e.g. Ittekkot et al., 1991). Therefore, the variations in discharge from all these rivers are rather governed by spatial variations in the precipitation pattern over the catchment area (Vijith et al., 2009).

The Indian subcontinent receives more than 80% of its annual rainfall during SWM (CPCB, 1995) period (June-September) and the entire estuary may be filled with freshwater without vertical salinity gradient (Vijith et al., 2009; Sridevi, 2013). Though the northeast monsoon (NEM) (December - March) yields some rainfall, discharges do not reach the coastal ocean as many of the rivers were dammed to meet domestic, industrial and irrigation needs. Therefore, virtually there is no discharge from the monsoonal rivers during dry period (January-May). As a result, the discharge during SWM (wet period) is almost equivalent to the annual discharge and so, that value for the wet period is used herein.

The estuaries studied here were categorized into 4 groups based on their geographic location on the subcontinent and intensity of rainfall; northeast (NE), southeast (SE), southwest (SW) and northwest (NW) estuaries. The SW region receives maximum rainfall during SWM (~3000 mm) followed by NE (1000-2500 mm), SE (300-500 mm) and NW (200-500 mm) regions of India (Soman and Kumar, 1990).


2.1 Sample collection

In order to obtain reliable export fluxes of DIN to the coastal ocean, in this study, samples were collected from estuaries rather than from mid or upstream rivers. A total of 12 researchers in four groups, equipped with car, CTD, Niskin sampler, filtration units, vacuum pumps, auto titrator for DO analysis etc. have participated in the field work for in-situ measurements and sample collection in the 27 Indian monsoonal estuaries during SWM. Estuaries located in the NE, SE, SW and NW regions of the Indian subcontinent were sampled by the groups 1, 2, 3 and 4, respectively, during 28 July – 18 August, 2011. Each estuary was sampled at 3 to 5 locations between upstream river (near zero salinity) and mouth of the estuary in order to minimize the spatial variation in concentrations. Samples were collected in mid stream of the estuary using a local mechanized boat to avoid contamination from the bank. Surface water samples from each station were collected for DIN, dissolved oxygen (DO), chlorophyll-a (Chl-a) and total suspended matter (TSM).

At the end of the sampling on each day, we set up a temporary shore laboratory for filtration of samples for DIN, TSM and Chl-a, and DO analysis by auto titrator (Metrohm, Switzerland) using a potentiometric end point detection. Filtered water samples for nutrients were preserved with poison (Mercuric Chloride) for analysis at our Institute laboratory by auto analyzer. The SWM commences generally in the last week of May in the southwest coast of India and progresses towards the northeast by the end of June. Concentrations of DIN were found to be higher in the initial discharge that normally occurs in the month of June and decline with time (Sarma et al., 2010; Parab et al., 2013). Earlier time series observations on DIN in the Godavari (Sarma et al., 2010), Mandovi (Pednekar et al., 2011) and Zuari (Anand et al., 2014) estuaries suggested that the variability in DIN concentrations during discharge period, except the higher concentrations during initial discharge, was within ±20%. In the present study, the samples were collected during July - August to avoid the possibility of overestimation of DIN fluxes by using higher concentrations in the initial discharge period. DIN concentrations measured in estuaries of the glacial rivers of India, Bangladesh, Myanmar and Pakistan, such as the Ganges, Brahmaputra, Irrawaddy, Salween and Indus were taken from published reports (Karim and Veizer, 2000; Lambs et al., 2005; Chapman et al., 2015) in order to estimate the total DIN flux reaching the northern Indian Ocean. Mean annual discharge data was taken from Meybeck and Ragu (1995, 1996), Central Water Commission, New Delhi (2006, 2012) and Kumar et al. (2005).
3. MATERIALS AND METHODS

Temperature, salinity and depth were measured using a CTD system (Sea Bird Electronics, SBE 19 plus, United States of America). DO concentrations were determined following Winkler’s method (Carritt and Carpenter, 1966) using auto titrator (Metrohm, Switzerland) with potentiometric end point detection at the temporary shore laboratory. The analytical precision of the method, expressed as standard deviation, was ±0.07%. DIN concentrations in the filtered water samples (0.22 µm polycarbonate filters, Millipore) were measured at our Institute laboratory by colorimetric method following Grashoff et al. (1992) using auto analyzer. The precision were ±0.02, ±0.02, ±0.01 and ±0.02 µmol L⁻¹ for ammonium, nitrate+nitrite, phosphate and silicate, respectively. About 500 ml of sample was filtered through glass fiber filter (GF/F; nominal pore size: 0.7µm) at a gentle vacuum and Chl-a on the filter was extracted into N,N-dimethyl formamide (DMF) at 4°C in the dark for 12 h. Fluorescence intensity of the extract was measured using spectrofluorophotometer (Carry Eclipse, Varian Electronics, UK) following Suzuki and Ishimaru (1990). Based on repeated analysis of samples, the analytical precision of the method was estimated to be ±4%. TSM was determined as weight difference of material retained on pre-weighted polycarbonate filters (0.22 µm; Millipore) after passing 500-1000 ml of water sample.

Total export of DIN from each river was estimated by multiplying the mean concentrations of DIN in an estuary with the mean annual discharge. The uncertainty in these estimations is mainly due to variability of DIN concentrations within the discharge period of sample collection and inter-annual variability in their concentrations. As examples, daily measurements of DIN concentrations in a major monsoonal estuary, the Godavari, for three years (2007-2009) were utilized. The intra-annual (Sarma et al., 2010) and inter-annual (Acharyya et al., 2012) variability in DIN concentrations during discharge (SWM) period ranged between 10 – 25%. Therefore, the error in our DIN flux estimations due to onetime sampling can be up to ±25%, which is close to the error associated with the estimations of DIN fluxes from few Indian rivers (±21%) by Singh and Ramesh (2011). DIN fluxes normalized by catchment area were calculated by dividing the total export of DIN with that of catchment area of the river.

4. RESULTS

The hydrography of the monsoonal estuaries during the study period was reported elsewhere (Sarma et al., 2014). Salinity in many of these estuaries was close to zero, except few estuaries, we measured <13 (an average of our stations). Minor estuaries, Nagavali, Vaigai and Rushikulya recorded higher
salinities of 28.8±5.6, 27.9±4.3 and 20.7±5.8 respectively. Chl-a varied from 0.8 to 10.7 mg m⁻³, with a higher values in the SW estuaries (>5 mg m⁻³), except Ponnayaar and Vellar. The monsoonal estuaries were under saturated with reference to DO (mean 87%), except few minor estuaries, Rushikulya, Nagavali, Penna, Ambalyaar and Vaigai (104%) during the study period. TSM ranged from 6.5 to 905 mg L⁻¹, with a higher suspended matter in the NW estuaries (mean 458 mg L⁻¹).

4.1 Concentrations of DIN in the Indian monsoonal estuaries during SWM

Monsoonal estuaries from India displayed a wide range of concentrations (Fig. 2a) from 0.01-0.23, 0-0.16, 0.04-0.98, 0.02-0.42 and 0.40-7.56 mg L⁻¹ for ammonium, nitrite, nitrate, phosphate, and silicate, respectively, during study period. Relatively higher concentrations of nitrate were found in the NW estuaries (mean 0.84 mg L⁻¹) followed by SW (0.28 mg L⁻¹), NE (0.17 mg L⁻¹) and SE (0.05 mg L⁻¹). The range of DIN concentrations observed in this study are within the range of those reported earlier in the Indian estuaries, Kochi (Martin et al., 2008), Godavari (Sarma et al., 2010), Mandovi (Pednekar et al., 2011), Mahanadi (Dixit et al., 2013) and Zuari (Anand et al., 2014), during discharge period. The range of nitrate concentrations in this study (0.04 - 1.0 mg L⁻¹), except polluted Narmada and Tapti, during discharge period is close to the range of concentrations reported by Lambs et al. (2005) in the major estuaries from India, and also in estuaries elsewhere in the world (Ohowa et al., 1997; Burns et al., 2008; Liu et al., 2009; Lima et al., 2010). However, the mean nitrate concentration (0.27 mg L⁻¹) is remarkably lower than the mean nitrate concentration (10.1 mg L⁻¹) in the rivers draining into the northern Indian Ocean (Subramanian, 2008). This is mainly due to the location of sampling, because Subramanian (2008) collected samples along the course of rivers whereas, we worked the samples from estuaries.

The mean concentrations of phosphate and silicate observed (0.02-0.42 and 0.40-7.56 mg L⁻¹, respectively) are close to those reported earlier for the Indian rivers (Ramesh et al., 2015) and elsewhere in the world (Xincheng and Haunting, 2001; Lima et al., 2010; Lebo and Sharp, 1993), but higher than those reported for Chinese and Korean estuaries (Liu et al., 2009) and the Delaware estuary (Sharp, 1994). Concentrations of phosphate were tended to be higher in the SW estuaries (mean 0.38 mg L⁻¹) than the other estuaries (mean 0.18 mg L⁻¹). Both natural processes such as weathering of P, and anthropogenic processes, such as deforestation, sewage and mining of P and its usage as detergents, fertilizer in agricultural and aquaculture practices, influence the surface transport of P to estuaries and coastal ocean (Mackenzie et al., 1998; Bennett et al., 2001; Fixen and West, 2002). Relatively lower consumption of P-containing synthetic fertilizers such as di-ammonium phosphate (DAP) in the SW region (mean 17.1 kg hectare⁻¹) than the other regions of the Indian
subcontinent (22.1 kg hectare\(^{-1}\)) suggest a strong influence of P leaching from soils in addition to the contribution from fertilizers. The likely reason could be the higher elevation of catchments of the SW rivers which originates from the Western Ghats. When compared to forest catchments, high-elevation catchments are more sensitive to environmental changes (Baron, 1992). The weathering of rocks results in release of soluble alkali phosphates and colloidal calcium phosphate to rivers and subsequently to estuaries (Ramesh et al., 2015).

Concentrations of silicate were tended to be higher by about four times in the SE estuaries (mean 4.24 mg L\(^{-1}\)) than in the other monsoonal estuaries (mean 0.95 mg L\(^{-1}\)). Major source of riverine export of silica to the coastal ocean is a chemical weathering of soils and rocks in the catchment area (e.g. Wollast and Mackenzie, 1983; Gaillardet et al., 1999; Durr et al., 2011). Soils in catchment areas of the SE rivers are composed mainly of red sandy soils and alluvial soils both of which are rich in silicate. Alluvial soils are soft so that the rivers transport more silt particles, which contain Si-rich quartz and feldspar, to the estuaries and coastal regions during river discharge. Moreover, relatively less forest cover in the SE region than the SW and NE regions may enhances the erosion and weathering of soils and rocks in catchment area of the SE rivers.

4.2 Total export and fluxes of DIN to the northern Indian Ocean during SWM

Total export of DIN to the northern Indian Ocean by the individual monsoonal rivers was in the range of 0.013-4.8, 0.004-1.34, 0.033-78.2, 0.066-21.2 and 0.786-362.9 (x10\(^9\)) g yr\(^{-1}\) for ammonium, nitrite, nitrate, phosphate and silicate, respectively (Fig. 2b). The respective total export to the northern Indian Ocean was estimated at 0.021, 0.009, 0.190, 0.11 and 1.026 Tg yr\(^{-1}\) from the Indian rivers. The Godavari transport highest nitrate (0.08 Tg yr\(^{-1}\)), while the Krishna transport highest amounts of phosphate (0.02 Tg yr\(^{-1}\)) and silicate (0.36 Tg yr\(^{-1}\)) among the monsoonal rivers. These rivers export similar amount of dissolved inorganic N (ammonium+nitrite+nitrate) to the Bay of Bengal (0.12±0.03 Tg yr\(^{-1}\)) and the Arabian Sea (0.10±0.02 Tg yr\(^{-1}\)), but they export extremely higher amounts of phosphate and silicate to the former (0.08±0.02 and 0.91±0.23 Tg yr\(^{-1}\), respectively) than the latter (0.03±0.007 and 0.12±0.03 Tg yr\(^{-1}\)). Including the glacial rivers of India, Bangladesh, Myanmar and Pakistan, it is estimated that the Bay of Bengal and Arabian Sea receives ~1.74 and 0.10 Tg yr\(^{-1}\) of nitrate, respectively. Excluding Irrawaddy and Salween, the rivers draining the Indian subcontinent export 1.62±0.40, 0.27±0.06 and 3.58±0.89 Tg yr\(^{-1}\) of nitrate, phosphate and silicate, respectively to the northern Indian Ocean. These amounts are significantly lower than the
earlier reports on export of nitrate (9.3 Tg yr\(^{-1}\); Singh and Ramesh, 2011) and silicate (14.0 Tg yr\(^{-1}\); Dileep Kumar et al., 1992) by the Indian rivers to the northern Indian Ocean.

Export fluxes of DIN normalized by catchment area from Indian monsoonal rivers were ranged from 1.5-757, 0.4-97.7, 3.8-1919 kg N km\(^{-2}\) yr\(^{-1}\), 7.3-2055 kg P km\(^{-2}\) yr\(^{-1}\) and 84-3643 kg Si km\(^{-2}\) yr\(^{-1}\) for ammonium, nitrite, nitrate, phosphate and silicate, respectively (Fig. 2c; Table 1). Relatively higher export fluxes of dissolved inorganic N (mean 755 kg N km\(^{-2}\) yr\(^{-1}\)), phosphate (612 kg P km\(^{-2}\) yr\(^{-1}\)) and silicate (1388 kg Si km\(^{-2}\) yr\(^{-1}\)) were observed in the SW estuaries than the other estuaries (203 kg N km\(^{-2}\) yr\(^{-1}\), 163 kg P km\(^{-2}\) yr\(^{-1}\) and 760 kg Si km\(^{-2}\) yr\(^{-1}\), respectively). However, DIN fluxes from the Indian estuaries are far less than the global fluxes of dissolved silicate (1600-6100 kg Si km\(^{-2}\) yr\(^{-1}\)) to the coastal zones in the world (Durr et al., 2011). These fluxes are similar to those reported elsewhere in the world for nitrate (Caraco and Cole, 1999), phosphate (Howarth et al., 1996) and silicate (Durr et al., 2011).

5. DISCUSSION

A wide range of concentrations, total export and fluxes of dissolved inorganic N, phosphate and silicate suggests a large spatial variability among the Indian monsoonal rivers during SWM. Such variability is mainly driven by significant variability in discharge (range: 0.9 - 111 km\(^{3}\) yr\(^{-1}\)), catchment area (0.001 – 0.313 x10\(^{6}\) km\(^{2}\)), and density of human population (111-711 persons km\(^{-2}\)), soil type and climatic condition in watersheds of the monsoonal rivers studied. Moreover, catchment areas of these rivers are experiencing variable climatic (temperature) and hydrological (precipitation and runoff) conditions, and anthropogenic activities (land use change and consumption of fertilizers) (Soman and Kumar, 1990; Jaga and Patel, 2012). For instance, mean annual temperatures are relatively higher in the SE (>27.5°C) than the SW (<22.5°C), NE and NW (22.5-27.5°C) regions of India (http://www.mapsofindia.com/maps/india/annualtemperature.htm). Basin characteristics (Jordan et al., 1997; Haggard et al., 2003; Buck et al., 2004), catchment size (Brainwood et al., 2004; Buck et al., 2004) and runoff (Schubel and Pritchard, 1986) were reported to control the variability of DIN concentrations in rivers elsewhere. A large spatial variability in concentrations and export of dissolved inorganic N, phosphate and silicate was also reported from major rivers in the world and varied dramatically from one region to the other due to changes in catchment type, run off, environmental conditions and anthropogenic activities in the catchment (Meybeck, 1982; Alvarez-Cobelas et al., 2009; Durr et al., 2011).
5.1 Total export of dissolved inorganic N to the northern Indian Ocean

The monsoonal estuaries from India export ~0.22±0.05 Tg yr⁻¹ of dissolved inorganic N to the northern Indian Ocean. Most of this amount (0.19±0.05 Tg yr⁻¹) is exported by the estuaries located in northern India (0.11±0.03 and 0.08±0.02 Tg yr⁻¹ by the NE and NW estuaries, respectively) than the southern Indian estuaries which export only 0.03 Tg yr⁻¹ (0.01 and 0.02 Tg yr⁻¹ by the SE and SW estuaries, respectively). It is attributed to use of N-containing fertilizers in agricultural fields of the Indian subcontinent as the fertilizer consumption is ~5 times higher in northern India (18 Tg yr⁻¹) than southern India (3.5 Tg yr⁻¹) (Ministry of Agriculture, Government of India, [http://eands.dacnet.nic.in/latest_2006.htm](http://eands.dacnet.nic.in/latest_2006.htm)). It is also consistent with the nitrogen isotopic composition of particulate nitrogen in the Indian estuaries during discharge period (Sarma et al., 2014). Large amount of rainfall (>80% of the annual rainfall) over the Indian subcontinent during SWM washes out the excess fertilizers applied to the agricultural fields and DIN from soils in the watershed to rivers and subsequently to estuaries (Acharryya et al., 2012; Sarma et al., 2014). Linear correlation between TSM and nitrate concentrations (r²=0.75; p<0.001; Fig. 3) confirms terrestrial origin of nitrate to the Indian estuaries during study period. Subramanian (2008) attributed the higher nitrate input from the glacial river Ganges (0.9 Tg yr⁻¹) to the Bay of Bengal to use of fertilizers in the fields of its basin. Shuiwang et al. (2000) also observed the strong influence of application of chemical fertilizers in catchment of the major Chinese rivers, Huanghe, Zhujiang and Changjiang on DIN fluxes from these rivers.

Although the Bay of Bengal receives ~76% (378 km³) of freshwater discharge from ~80% (0.98x10⁶ km³) of the catchment area compared to the Arabian Sea which receives only 24% (121.6 km³) of discharge from ~20% (0.24x10⁶ km³) of catchment area of the monsoonal rivers during SWM, the total export of dissolved inorganic N by the monsoonal rivers from India was found to be similar to the Arabian Sea (0.10±0.02 Tg yr⁻¹) and the Bay of Bengal (0.12±0.03 Tg yr⁻¹). Despite the west-flowing rivers are smaller with reference to both catchment area (0.001-0.10 x10⁶ km²) and volume of discharge (1.92-45.6 km³) than the east-flowing rivers (0.007-0.313 x10⁶ km² and 0.88-110.5 km³, respectively), the similar export of dissolved inorganic N to the Arabian Sea is due to major contribution (~0.08±0.02 Tg yr⁻¹) from the NW rivers (Narmada through Mahisagar). It is mainly due to the contribution from the heavily polluted Narmada and Tapti estuaries by industrial and port activities, and consumption of more fertilizers in the northern India as discussed above. It could also be, at least in part, due to the more intense erosion of soils in catchment areas of the NW rivers than the other regions. Remarkably higher TSM in the NW (458 mg L⁻¹) than the SW (41), SE (35) and
NE (77 mg L\(^{-1}\); except Godavari) estuaries confirms that occurrence of rigorous soil erosion in catchment areas of the NW rivers. It is mainly due to less precipitation (200-500mm) and lack of dense forest cover in the NW region (Soman and Kumar, 1990). Further, catchment areas of the NW rivers is dominated by the black soils which were reported to have relatively higher rate of soil erosion (20 ton hectare\(^{-1}\) yr\(^{-1}\)) than the red (10-15 ton hectare\(^{-1}\) yr\(^{-1}\)) and alluvial (5-10 ton hectare\(^{-1}\) yr\(^{-1}\)) soils (Singh et al., 1992). Relatively higher population density in watershed of the west-flowing rivers (about 1410 persons km\(^{-2}\)) than the east flowing rivers (about 608 persons km\(^{-2}\)) (Central Water Commission, 2012) suggests that the higher anthropogenic interference in the watershed of the former than latter. Based on model results from the large rivers in the world, Caraco and Cole (1999) demonstrated that anthropogenic loading is dominant nitrate export to the coastal ocean. More than 50% of the 1000-fold variation found in the nitrate export by the world rivers was attributed to human interferences alone (Cole et al., 1993). Similarly, 100-fold variation in the continental United States (Jordan and Weller, 1996) and ~20-fold variation in the sub-regions adjoining the North Atlantic (Howarth et al., 1996) were also attributed to anthropogenic N loading in watershed of the rivers. Hence, both natural processes and anthropogenic activities in the watershed of the west-flowing rivers control the export of dissolved inorganic N to the Arabian Sea.

Including the large input (1.63 Tg yr\(^{-1}\)) from the glacial rivers, Ganges, Brahmaputra, Irrawaddy and Salween, the Bay of Bengal receives ~1.74±0.43 Tg yr\(^{-1}\) of nitrate, while the Arabian Sea receives only ~0.10±0.02 Tg yr\(^{-1}\) including input from the glacial river Indus (0.2 Tg yr\(^{-1}\)). Together, glacial and monsoonal rivers export ~1.84±0.46 Tg yr\(^{-1}\) of nitrate to the northern Indian Ocean. Based on DIN measured along the course (mid or upstream) of the rivers (Subramanian, 2008), Singh and Ramesh (2011) reported four times higher nitrate export (9.3 Tg N yr\(^{-1}\)) to the northern Indian Ocean. However, the export is estimated at only 2.1 Tg N yr\(^{-1}\) if the data of Lambs et al. (2005), in which DIN were measured in estuaries, are used (Singh and Ramesh, 2011). Our estimate of 1.84±0.46 Tg N yr\(^{-1}\) is close to the latter one (2.1 Tg N yr\(^{-1}\)), since the DIN concentrations in estuaries rather along the course or mid-river were used for flux estimates in both studies. These results suggest strongly that the location of sampling for DIN concentrations is important for the estimation of DIN export to the coastal ocean. Clearly, the retention/elimination of DIN by biogeochemical processes, including utilization by phytoplankton, along the course of the rivers before reaching estuaries is critical. Therefore, this study suggests that the likelihood of over estimation in export of DIN to the coastal ocean if concentrations in the course of the rivers are measured.
5.2 Total export of phosphate and silicate to the northern Indian Ocean

The total export of both phosphate and silicate from the monsoonal rivers from India was higher to the Bay of Bengal (0.08±0.02 and 0.91±0.23 Tg yr\(^{-1}\), respectively) than to the Arabian Sea (0.03±0.007 and 0.12±0.03 Tg yr\(^{-1}\)) due to higher discharge to the former (t-test; p=0.14). For instance, the major rivers such as Godavari, Krishna, Mahanadi, Haldia and Cauvery draining into the Bay of Bengal export 0.07±0.02 Tg yr\(^{-1}\) of phosphate and 0.81±0.20 Tg yr\(^{-1}\) of silicate to the Bay of Bengal. The present estimate of phosphate export from the monsoonal rivers from India (0.08±0.02 Tg yr\(^{-1}\)) is nearly double the phosphate export from the monsoonal rivers (0.04 Tg yr\(^{-1}\)) reported by Ramesh et al. (2015). This is mainly due to the fact that they did not include the many of the medium-sized east- and west-flowing Indian monsoonal rivers, which carry considerable amount of phosphate, and only a few major rivers (Mahanadi, Godavari, Krishna, Cauvery, Periyar, Narmada and Tapti) were considered for their study. Including the input from the glacial rivers Ganges, Brahmaputra and Indus, the Indian rivers export 0.28±0.07 Tg yr\(^{-1}\) of phosphate to the northern Indian Ocean. This is significantly higher than the phosphate export to the northern Indian Ocean of 0.05 Tg yr\(^{-1}\) reported by Dileep Kumar et al. (1992). These authors estimated phosphate export from only few rivers (Ganges, Mahanadi, Narmada, Vellar, Mandovi and Zuari). Also, the glacial rivers (Brahmaputra and Indus), major (Godavari and Krishna) and medium monsoonal rivers (Haldia, Baitarani, Penna, Netravari etc.) were not included in their study. In contrast, we estimated the phosphate export from the glacial rivers, Ganges, Brahmaputra and Indus, and 27 monsoonal rivers (major and medium) from India to the northern Indian Ocean. The Bay of Bengal receives ~75% of the total input of phosphate (~0.21±0.05 Tg yr\(^{-1}\)), whereas the Arabian Sea receives only 25% (~0.07±0.02 Tg yr\(^{-1}\)) from the Indian estuaries due to higher runoff to the former (Subramanian, 1993; Gauns et al., 2005).

We estimate that the monsoonal rivers export ~1.03±0.25 Tg yr\(^{-1}\) of silicate to the northern Indian Ocean of which 0.91±0.23 Tg yr\(^{-1}\) enter the Bay of Bengal and 0.12±0.03 Tg yr\(^{-1}\) the Arabian Sea. Including input from the glacial rivers, the total export of silicate to the northern Indian Ocean is estimated to be 3.58±0.89 Tg yr\(^{-1}\). This amount is only one fourth of the silicate export (14.0 Tg yr\(^{-1}\)) from the glacial and monsoonal rivers to the northern Indian Ocean reported by Dileep Kumar et al. (1992). Again, it is due to the differences in sampling locations for measurement of concentrations as discussed earlier. Of the total riverine export of 3.58±0.89 Tg yr\(^{-1}\), about 93% (3.35±0.88 Tg yr\(^{-1}\)) reaches the Bay of Bengal and the remaining 7% enter the Arabian Sea. The difference is principally due to major contribution (2.45±0.61 Tg yr\(^{-1}\)) from the Himalayan rivers dominated by silicate-
weathering, Ganges and Brahmaputra (e.g. Krishnaswami and Singh, 2005), and from the east-flowing monsoonal rivers (0.72±0.18 Tg yr⁻¹) that drain alluvial soils in the SE region of the subcontinent.

5.3 Export fluxes of DIN normalized by catchment area from the Indian monsoonal estuaries

Since the variability in catchment area (0.001 – 0.313 x10⁶ km²) and discharge volume (0.9 - 111 km³) of the monsoonal estuaries is large, export fluxes of DIN were normalized by the respective catchment areas in order to understand the factors controlling their fluxes to the coastal ocean. Though human interferences and fertilizer consumption in the watershed should strongly control the total export of dissolved inorganic N to northern Indian Ocean, their impact on export fluxes (per unit area basis) seem to be quite low. Though India has the second-highest consumption of fertilizers (26.5 Tg yr⁻¹) in the world, with a share of 15.3% of world's N consumption (Jaga and Patel, 2012), export fluxes of dissolved inorganic N from the Indian estuaries (357 kg N km⁻² yr⁻¹) are lower than the export fluxes from many rivers in the world (Howarth et al., 1996; House et al., 1997; Garnier et al., 2002; Green et al., 2004; Billen et al., 2005; El Boukhary, 2005; Durr et al., 2011; Carey and Fulweiler, 2011, 2013). It is attributed to the elimination and/or retention of N in soils of the watershed (Dillon et al., 1990; Howarth et al., 1996, 2006; Arheimer, 1998; Lepisto et al., 2001; Boyer et al., 2002; Billen et al., 2011) and differences in sampling locations for DIN measurements.

The annual application of ~17.66 Tg of synthetic N-fertilizers (Jaga and Patel, 2012) in the agricultural fields in India versus the transport of 1.62±0.32 Tg yr⁻¹ to the northern Indian Ocean result in a retention and/or elimination of ~91% of anthropogenic N rather than transporting to the coastal ocean. It is consistent with the conclusion of Bouwman et al. (2005) that only 4-5% of the total fertilizer input to the agricultural system in the world is transported to coastal oceans. It is due to elimination of N in different stages such as crop removal, denitrification in the soils and ground water systems and utilization by algae on the course of the rivers. More efficient (~91%) retention and/or elimination of anthropogenic N in watersheds of the Indian rivers than by the North American and Western European watersheds (74%; Howarth et al., 1996, 2006; Boyer et al., 2002; Alexander et al., 2002), European watersheds (range: 50-90%, mean: 82%; Billen et al., 2011) and mean retention in global watershed (~80%; Caraco and Cole, 1999) appears to lower the export fluxes of dissolved inorganic N from the Indian rivers.
5.4 Importance of sampling sites in rivers for estimation of DIN export fluxes

Large amounts of the riverine DIN are likely to be consumed within the course of the rivers enroute to coastal ocean (Billen et al., 1991; Singh and Ramesh, 2011). Due to efficient retention and/or elimination of N in rivers before reaching estuaries, it is possible that DIN fluxes are over estimates when the concentrations of DIN in rivers were used. For instance, DIN measured in rivers (Dileep kumar et al., 1992; Subramanian, 2008) results in three times higher export fluxes of inorganic N (mean 3290 kg N km$^{-2}$ yr$^{-1}$) and silicate (mean 4969 kg Si km$^{-2}$ yr$^{-1}$) to the northern Indian Ocean by the Indian rivers than the fluxes (981 kg N km$^{-2}$ yr$^{-1}$ and 936 kg Si km$^{-2}$ yr$^{-1}$) yielded from DIN measured in estuaries (Lambs et al., 2005). The concentrations of DIN in estuaries/near river mouth provide better estimates of riverine DIN fluxes to coastal ocean since the concentrations in estuaries are a net result of both the processes that add DIN to- and remove DIN from the river system (Darracq et al., 2008; Yan et al., 2010; Gao et al., 2012). Therefore, collection of samples for DIN measurement in the estuaries rather than mid or upstream rivers in the present study could also be the potential reason for the lower DIN fluxes from the Indian rivers than many rivers in the world. Selection of sampling location for DIN measurement is, thus, critical to obtain reliable estimates of export fluxes of DIN to the coastal oceans, and many of the earlier estimates from global rivers based on the DIN in rivers may become substantially lower if DIN are measured in estuaries.

5.5 Impact of hydrological conditions on concentration, total export and fluxes of DIN to the northern Indian Ocean

Concentrations of ammonium, nitrite, nitrate, phosphate and silicate did not show relationship with neither catchment area nor freshwater discharge indicating that size of the river is not controlling the concentrations of DIN in these estuaries. In contrast, a significant linear relationships of total export of dissolved inorganic N, phosphate and silicate with catchment area ($r^2=0.52$, $p<0.001$, Fig.4a; $r^2=0.80$, $p<0.001$, Fig.4b; $r^2=0.85$, $p<0.001$, Fig.4c, respectively) and the length of the river ($r^2=0.75$, $p<0.001$, Fig.5a; $r^2=0.61$, $p<0.001$, Fig. 5b and $r^2=0.66$, $p<0.001$, Fig. 5c, respectively) were observed, indicating that export of DIN to the northern Indian Ocean is strongly controlled by the length and catchment area of river, despite concentration varies spatially. However, DIN fluxes normalized by catchment area showed no correlation with discharge ($r^2=0.00$, $p=0.89$; $r^2=0.001$, $p=0.89$ and $r^2=0.05$, $p=0.27$, respectively), indicating that export fluxes of DIN (per unit area basis) are independent of volume of freshwater discharge. Despite low discharge (46.28 km$^3$) occurs from a small catchment area (0.024 x10$^6$ km$^2$) of the SW rivers compared to the other monsoonal rivers (453 km$^3$ and 1.17x10$^6$ km$^2$, respectively), the higher export fluxes of dissolved inorganic N (755 kg
N km\(^{-2}\) yr\(^{-1}\)), phosphate (612 kg P km\(^{-2}\) yr\(^{-1}\)) and silicate (1388 kg Si km\(^{-2}\) yr\(^{-1}\)) from the SW estuaries than from the other monsoonal estuaries (203 kg N km\(^{-2}\) yr\(^{-1}\), 163 kg P km\(^{-2}\) yr\(^{-1}\) and 760 kg Si km\(^{-2}\) yr\(^{-1}\), respectively) (t-test; p=0.03, p=0.04 and p=0.003, respectively) is attributed to strong influence of hydrological conditions (precipitation and runoff) and anthropogenic activities in the watershed. Relatively higher rainfall (~3000 mm) in small catchments of SW rivers (0.024x10\(^6\) km\(^2\)) during SWM results in a very high annual discharge from unit area of catchment (1.71 m\(^3\) m\(^{-2}\) yr\(^{-1}\)) than the NE (0.6), SE (0.17) and NW (0.32 m\(^3\) m\(^{-2}\) yr\(^{-1}\)) rivers. This is because the latter regions receive relatively less rainfall over large catchment areas (Soman and Kumar, 1990). High density rainfall and elevated catchments of the SW rivers causes erosion of more DIN from soils, resulting in higher export fluxes of DIN from the SW estuaries than from SE, NE and NW estuaries during SWM.

### 5.6 Impact of riverine DIN export on the ecosystem of Indian coastal waters

It is well known that primary production in the sunlit upper ocean is strongly limited by the availability of DIN in most of the oceans including the northern Indian Ocean (Broecker, 1974; Falkowski, 1997; Prasanna Kumar et al., 2002; Madhupratap et al., 2003). As the supply of dissolved inorganic N through atmospheric deposition is relatively minor in the northern Indian Ocean (Duce et al., 2008; Srinivas et al., 2011; Singh et al., 2012), the riverine loading to this region is, therefore, important for primary production. Based on Redfield ratio of C:N:P (106:16:1 by atoms), export of 1.84±0.46 Tg yr\(^{-1}\) of dissolved inorganic N from the rivers of India, Bangladesh, Myanmar and Pakistan would support ~12.2±3.1 Tg yr\(^{-1}\) of new carbon production (11.5±2.9 and 0.66±0.17 Tg C yr\(^{-1}\) in the Bay of Bengal and the Arabian Sea, respectively). The river-borne nutrients (1.74±0.43 Tg yr\(^{-1}\)) support ~17% of total primary production (66 Tg yr\(^{-1}\); Fernandes et al., 2008) in the Bay of Bengal (upper 10m), whereas it is <1% (104 Tg yr\(^{-1}\); Bhattathiri et al., 1996) in the Arabian Sea. These results suggest that riverine export of DIN results in removal of significant fraction of atmospheric CO\(_2\) through the biological pump in the Bay of Bengal but not in the Arabian Sea. However, further studies are required to estimate the impact of riverine DIN fluxes on primary production in the Indian coastal waters.

### 6. SUMMARY

River discharge is the major pathway of transport of dissolved inorganic nutrients (DIN) to coastal waters of the northern Indian Ocean, where the scarcity of DIN limits the primary production. In this study, it is estimated that the monsoonal and glacial rivers export 1.84±0.46, 0.28±0.07 and 3.58±0.89 Tg yr\(^{-1}\) of nitrate, phosphate and silicate, respectively, to the northern Indian Ocean. Of
this export, the contribution from monsoonal rivers is $0.22\pm0.05$, $0.11\pm0.03$ and $1.03\pm0.26$ Tg yr$^{-1}$, respectively. Our estimations of DIN export by the Indian rivers to the northern Indian Ocean are lower than the earlier reports on DIN export from these rivers. The differences are attributed to the choice of sampling location, as the earlier studies collected DIN along the course of rivers. Significant removal of DIN on the course of the rivers before reaching estuaries leads to possible over estimation of DIN export to the coastal ocean. Hence, in this study, relatively lower fluxes of DIN to the northern Indian Ocean from the Indian rivers reflect efficient retention/elimination of DIN (~91%) in the watershed and on the course of the river before reaching to the coastal ocean. The monsoonal rivers export larger amounts of phosphate (73%; $0.08\pm0.02$ Tg yr$^{-1}$) and silicate (88%; $0.91\pm0.23$ Tg yr$^{-1}$) to the Bay of Bengal than to the Arabian Sea ($0.03\pm0.007$ and $0.12\pm0.03$ Tg yr$^{-1}$ respectively) due to intense discharge to the former during SWM. However, the similar export of dissolved inorganic N to the Bay of Bengal ($0.12\pm0.03$ Tg yr$^{-1}$) and the Arabian Sea ($0.10\pm0.02$ Tg yr$^{-1}$) is attributed to large contribution from the polluted NW rivers Narmada and Tapti.

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REFERENCES


Fig. 1: Map showing monsoonal estuaries sampled in this study. Major and medium-sized rivers were indicated by the thick and thin black lines, respectively. Monsoonal and glacial rivers were indicated by black and white letters, respectively.
Fig. 2: Spatial distribution of (a) concentrations (mg L\(^{-1}\)), (b) total export (tons yr\(^{-1}\)) and (c) export fluxes (kg km\(^{-2}\) yr\(^{-1}\)) of ammonium, nitrite+nitrate, phosphate and silicate in the monsoonal estuaries from India during southwest monsoon (SWM). The NW, SW, SE and NE represent estuaries located in the northwestern, southwestern, southeastern and northeastern parts of India, respectively. Estuaries draining into the Arabian Sea and the Bay of Bengal were also shown.
Fig. 3: Linear correlation between nitrate and TSM concentrations (mg L$^{-1}$) in the monsoonal estuaries from India during southwest monsoon. Mahisagar estuary shown by rectangle was not included in the linear fit.
Fig. 4: Correlation between catchment area of rivers and total export of dissolved (a) inorganic N, (b) phosphate and (c) silicate in the Indian monsoonal estuaries during southwest monsoon (SWM).

Fig. 5: Correlation between the length of rivers and total export of dissolved (a) inorganic N, (b) phosphate and (c) silicate in the Indian monsoonal estuaries during southwest monsoon (SWM).
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</tbody>
</table>
Table 1: Catchment area (x10^6 km^2), annual mean discharge (km^3) and length (km) of the Indian monsoonal rivers were given. Measured concentrations (mg L^{-1}) in estuaries during southwest monsoon (SWM), and estimated total export (tons year^{-1}) of dissolved inorganic N (nitrite+nitrate+ammonium), phosphate and silicate to the coastal ocean from the monsoonal rivers were also provided. Export fluxes normalized by catchment area of dissolved inorganic N (kg N km^{-2}yr^{-1}), phosphate (kg P km^{-2}yr^{-1}) and silicate (kg Si km^{-2}yr^{-1}) from the Indian monsoonal estuaries during SWM were also given. Estuaries draining into the Bay of Bengal and the Arabian Sea were shown separately.