

Spatial variations in dissolved inorganic nutrients in the groundwaters along the Indian coast and their export to adjacent coastal waters

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Abstract

Submarine Groundwater Discharge (SGD) is one of the main external nutrient sources to the coastal waters. The concentration of nutrients in groundwaters are a few folds higher than that of adjacent coastal waters, therefore SGD enhances nutrients levels in the coastal waters and influences coastal biota. In order to examine the spatial and seasonal variability in nutrient concentrations and exchange to the coastal waters, groundwater samples were collected at ~90 locations along the Indian coast during the wet and dry seasons. This study revealed that dissolved inorganic nitrogen (DIN) and dissolved inorganic phosphates (DIP) and urea were found to be high during the dry than wet period. Higher concentrations of DIN and DIP were observed during both wet and dry periods in the groundwater along the east than the west coast of India. The State-wise mean amount of fertilizer used during Kharif (wet) and Rabi (dry) period in each Indian State showed significant correlation with mean concentrations of DIN and urea. The observed linear relationship of DIN with bacterial respiration and inverse relationship with DO saturation and ammonium in groundwater suggested that decomposition of organic matter and nitrification contributed to the DIN pool in the groundwater. The mean rate of SGD fluxes varied between $1.6 \times 10^4 \text{ m}^3/\text{d}$ and $1.75 \times 10^{11} \text{ m}^3/\text{d}$ in the Indian coastal region. The annual mean SGD flux of DIN and DIP was estimated to be 0.103 ± 0.02 and $0.021 \pm 0.01 \text{ Tg}$ ($1 \text{ Tg} = 10^{12} \text{ g}$) to the western coastal Bay of Bengal (east coast of India) and 0.06 ± 0.03 and $0.015 \pm 0.01 \text{ Tg/y}$ to the eastern coastal Arabian Sea (west coast of India) respectively. The estimated SGD flux of DIN and DIP to the Indian coastal waters amounted to 0.163 ± 0.04 and $0.036 \pm 0.02 \text{ Tg/y}$ respectively and it is almost close to that of nutrients discharged by rivers (0.22 ± 0.05 and $0.11 \pm 0.03 \text{ Tg/y}$ respectively). Among the external sources of nitrogen and phosphorus, such as river discharge, atmospheric deposition, the contribution by SGD is highly significant in the Bay of Bengal (30 and 17% respectively) than in case of Arabian Sea (24 and 25% respectively).

Key words: Nutrients, Fertilizers, nitrification, groundwater, Indian coast

Introduction

Submarine groundwater discharge (SGD) is the flow of water to the coastal ocean from the seabed either driven by hydraulic forcing from inland aquifers or by ocean forces, such as waves, tides, and buoyancy, and plays a significant role on water and chemical budgets of the ocean (Taniguchi et al., 2002; Moore, 2010). The SGD to the coastal ocean is critical as it contains high concentrations of solutes and nutrients. It has been estimated that SGD contributes to ~10% of the freshwater input by rivers to the world's ocean but similar amounts of solutes such as carbon, nutrients, trace elements by rivers (Taniguchi et al., 2002; Slomp and Van Cappellen, 2004; Liu et al., 2014; Windom et al., 2006; Kim et al., 2005). Recently Luijendijk et al. (2020) estimated that SGD accounts 0.6% of the total freshwater input and ~2% of the solute input to the ocean and further suggested that the SGD has significant impact on the coastal ecosystem but not of the offshore region. The flow of excess nutrients into coastal waters through SGD is a major concern (Vitousek et al., 1997; Castro et al., 2003) and can lead to a range of impacts on coastal ecosystem such as eutrophication (Knee and Paytan, 2011; Rabalais et al., 2009), deoxygenation (eg: Breitberg et al., 2018) and global nutrient budget (Galloway et al., 2008; Slomp and Cappellen, 2004). The discharge of groundwater nutrients further change the composition of the phytoplankton by modifying the nitrogen to phosphorus (N/P) ratio. For instance, algae in the coastal waters are characterized by an N/P ratio of 16 ('Redfield ratio') for their growth (Redfield et al., 1963). Many coastal waters are characterized by an N/P ratio that is below Redfield and are thought to be N-limited (Howarth and Marino, 2006). As P is mostly retained in soils, whereas N is typically not (Buss et al., 2004; Lorah et al., 2009), therefore, SGD is usually characterized by N/P ratios of >16, especially in agricultural areas. Therefore, the regions influenced by SGD can change the limiting nutrient from N to P.

Coastal waters play a significant role in the global carbon and nitrogen cycle (Herbert 1999; Bauer et al., 2013). The main external sources of N and P to the coastal ecosystems are atmospheric deposition, river discharge, SGD and nitrogen fixation (Howarth et al., 2002). Since nutrient concentrations in coastal groundwater are several orders of magnitude greater than surface waters, SGD could be a significant factor in the eutrophication of coastal waters (Valiela et al., 1990, 1992; Luijendijk et al., 2020). Input of high N and P through SGD may be linked to formation of harmful algal blooms and eutrophication in the coastal waters (LaRoche et al., 1997; Anderson et al., 2002; Lecher et al., 2015). Kwon et al. (2014), based on ²²⁸Ra isotopic studies, found that global SGD is 3 to 4 times higher than that of freshwater discharge from rivers.

The seasonal eutrophication and suboxia/hypoxia occur along the Indian coast and it is mainly attributed to coastal upwelling, anthropogenic nutrients inputs by rivers and salinity stratification

(Naqvi et al., 2000; Sarma et al., 2013). The role of SGD on coastal eutrophication along the Indian coast is unclear. The regions of SGD along the Indian coast are identified using different techniques and tracers (Krishna et al., 2016; Suresh Babu et al., 2009; Moore, 1997; Jacob et al., 2009; Rahaman and Singh, 2012). Moore (1997) made a pioneering work on the SGD water flux and estimated that groundwater flux from the Ganga-Brahmaputra region to the Bay of Bengal accounts to ~30% of the annual river discharge from the Ganga-Brahmaputra river system. Basu et al. (2001) reported $2 \times 10^{11} \text{ m}^3/\text{y}$ of groundwater flows directly into the Bay of Bengal from the Ganga-Brahmaputra river system. Rengaragan and Sarma (2015) estimated that ~30% of the coastal nutrients (N,P and Si) are contributed by SGD. Recently Luijendijk et al. (2020) found that Indian coast is a high-risk zone for eutrophication due to discharge of high magnitude of nutrients (N and P) from groundwater to the coastal waters. Recently, Kaliraj et al. (2019) identified that the seawater intrusion is found to be greater in the eastern coastal regions up to the extent of 2-3 km. The groundwater level along the east coast, at most of the locations, is between 0 and 10 m mean sea level (msl) whereas the groundwater level is between 20 and 40 m msl along the west coast due to the topographic elevation (Manivannan and Elango, 2019). It is found that the groundwater levels are below msl (0 to -2 m msl) in about 1900 km^2 of the Indian coast and it was increased by about 400 km^2 during the past decade (Manivannan and Elango, 2019). Freshwater discharge from groundwater to the coastal regions is reported in some States, such as Andhra Pradesh, Karnataka, Maharashtra, Gujarat, West Bengal, Kerala, and Tamil Nadu (Suresh Babu et al., 2009; Rahaman and Singh, 2012; Antony and Ramanujam, 2014; Rengarajan and Sarma, 2015; Debnath and Mukherjee, 2016; Chidambaram et al., 2017; George et al., 2018; Prakash et al., 2018; Srinivasamoorthy et al., 2018, Yadav et al., 2019). These studies suggested that a significant amount of groundwater discharges along the Indian coast and it may input significant concentration of nutrients, minerals, metals and organic components into the coastal waters. Since India has 7500 km long coast line, the nutrients flux through SGD may be significant and its influence on coastal ecosystem needs evaluation. Therefore, the main aims of this study are to 1) examine the variability in nutrient concentrations in the groundwaters close to the Indian coast during the wet and dry periods, 2) evaluate the possible sources of nutrients to the groundwater along the Indian coast and 3) estimate the fluxes of nutrients using available SGD rates in the literature to the adjacent coastal waters (Bay of Bengal and Arabian Sea) to account its contribution to the external sources of N and P to the coastal regions.

2. Materials and Methods

2.1. Study area

Indian subcontinent has diverse terrain – varied from the Himalayan peaks to the Indian Ocean coastline. India has a coastline of 7,517 kms, which covers 9 major states - West Bengal, Odisha, Andhra Pradesh, Tamil Nadu, Kerala, Karnataka, Goa, Maharashtra, and Gujarat. Groundwater samples were collected from 90 wells along the East and West coast of India (Fig.1) during wet/monsoon (August 2015) and dry/non monsoon (April 2017) periods.

2.2. Hydroclimatology

Indian subcontinent receives rainfall during summer (June to September) and winter (November to February) (Soman and Kumar, 1990) with the highest precipitation received in the western India, mainly in the southwest region, during former and southeastern region during latter period. The annual mean rainfall is ~1500 mm with 70% of it occurs during summer. The highest rainfall occurs in the southwest and northeastern India followed by northwest and southeast region during summer (Soman and Kumar, 1990).

The characteristics of soil are important on elemental composition of groundwaters. In India, most common soil types are Alluvial, Black, Red, Yellow, Laterite, and Arid soils (Fig. 1; Bhattacharyya and Kumar, 2015). Red and Yellow soils are the most abundant soil types in India, and these soils can be found along the coastal states, such as Goa, Kerala, Karnataka, Tamil Nadu, Andhra Pradesh, and Odisha (Fig. 1). Red and Yellow soils contain 0.08% of nitrogen and 0.09% of phosphorus (Shahid and Naseem 2017). Alluvial soil constitutes the largest and most important soil group of India and mostly along the coastal region. These soils are deficit in nitrogen, phosphorus and organic matter. The black soils are very dark and have a very high clay content. Black soil occurs in the coastal states of Maharashtra, Gujarat, Andhra Pradesh and some parts of Tamil Nadu. This soil type mainly covers the central, western and southern states of India. Black soil type has low nitrogen and phosphorus content (Shahid and Naseem 2017).

2.3. Agricultural practices and fertilizer utilization in India

Indian agricultural practice is mainly based on the nature of soil and water availability. India has two main cropping seasons, namely Kharif (April to September) and Rabi (October to March). The major Kharif crops, include rice, sorghum, millet, maize, cotton, sugar cane, soybean and groundnut, whereas wheat, barley, gram, linseed, rapeseed and mustard are grown during Rabi season. The fertilizer demand (mainly Urea and NPK-Nitrogen, phosphorus and potash) for crops grown during Kharif period is double to that of Rabi season (www.fert.nic.in). The seasonal mean consumption

rates were 5 to 20% higher during the wet (Kharif) than dry (Rabi) season (www.fert.nic.in). The mainly used fertilizers are urea (46% nitrogen), diammonium phosphate (DAP; 18% nitrogen and 46% phosphate) and NPK (nitrogen, phosphorus and potash in 16% each) in India. Therefore, the fertilizer consumption varies with season which may have impact on the seasonal variability in nutrient concentrations in the groundwater. The annual mean fertilizer consumption in the Indian subcontinent is 165 kg/hectare, and it is ~10% of fertilizers used in the Asia. The fertilizer usage per hectare of agricultural land in India is roughly equal to that of North America (168 kg/hectare), South America (197 kg/hectare), but it is half of that of Australia (303 kg/hectare) (<https://www.indexmundi.com/facts/indicators/AG.CON.FERT.ZS/rankings>; <https://ourworldindata.org/fertilizer>).

2.4. *Sampling and analytical methods*

Groundwater samples were collected from the open wells, which are <300 m away from the coast, along both east and west coast of India. The details of position, well depth, rainfall and soil characteristics of the each well sampled are given in Table 1. Temperature was measured with a mercury thermometer and conductivity was measured with a Conductivity meter (HI2003) with accuracy $\pm 1\%$. Water samples were collected using 5 L Niskin bottle and subsampled first for dissolved oxygen (DO) followed by nutrients. DO samples were collected in 125 ml glass stoppered bottles. Care was taken to avoid trapping of gas bubbles while subsampling for DO. The DO in the sample was fixed with winker reagents. After precipitate is settled, 1 ml of concentrated sulfuric acid was added and liberated iodine was titrated with hypochlorite and the end point was identified using potentiometer following Carritt and Carpenter (1966). Water samples for nutrients were collected in 100 ml plastic bottles and frozen until analysis in the laboratory. Nutrients were measured using spectrophotometric method following Grashoff et al. (1992) using Skalar autoanalyzer. The concentration of dissolved inorganic nitrogen (DIN) is computed as sum of nitrate, nitrite, and ammonium. Dissolved Urea was analyzed using spectrophotometer method following Mulvenna and Savidge (1992). The measurement precision of nitrate + nitrite, phosphate, ammonium and urea were ± 0.02 , 0.01, 0.02 and 0.04 $\mu\text{mol l}^{-1}$ respectively. Bacterial respiration (BR) was measured following DO incubation method (Kamler, 1969 and Gyllenberg, 1973;1981). The water samples were collected in six 125 ml glass bottles for DO measurements. The DO in the bottle was fixed in three bottles using winker reagent and other three bottles were incubated in dark for 24 hours and DO was fixed after incubation period. The loss of DO in the incubated bottle from that of initial was taken as respiration rate. The DO saturation was estimated following Garcia and Gordon (1992). The percentage of DO saturation was computed as

$$\text{DO saturation (\%)} = (\text{DO}_{\text{measured}}/\text{DO}_{\text{saturation}}) \times 100$$

Soil samples were collected at selected locations given in Table 1. The upper few centimeters of the soil were removed using the plastic spoon and the soil sample was collected in the plastic ziplock bags and frozen the samples at 3°C until analysis. The samples were dried at 60° C and about 0.5 mg of the dried sample was dissolved in 50 ml of nutrients-free deionized water and sonicated for 1 hour. The extracted water was filtered through GF/F filter and nutrients extracted into the water was analysed following same procedure described above.

The data of the amounts of fertilizers used in the Indian States are obtained from the Ministry of Agriculture website (www.fert.nic.in).

2.5. Graphical representation and statistical analyses of data

The software Arc GIS was used for graphical representation of 3 dimensional plots of our data and Statistica software is used for statistical analysis of rank correlation matrix (Spearman's r) and student t-test.

3. Results

3.1. Hydrographic condition in Indian groundwaters

The Indian coastal groundwater temperature during study period varied between 26.0 and 34.0°C being warmer during the wet (30.2±1 °C) than dry period (26.4±4 °C) (Table 2). The water conductivity was higher during the dry (6.04±8 mS/cm) than wet period (1.76±3 mS/cm; $t=4.26$, $p<0.0001$, $n=86$) and higher conductivity was observed in the groundwater along the east than the west coast of India (Table 2). DO saturation levels ranged between 15 and 111% in the Indian coastal groundwater with higher saturation during the wet (61±27%) than dry period (55±24%). In contrast, higher bacterial respiration rates were found during the dry (3.5±3 mg/l/d) than wet period (0.9±0.6 mg/l/d; Table 2).

3.2. Spatial and temporal distribution of dissolved inorganic nutrients in the groundwater and soil along the Indian coast

The concentration of ammonium in the groundwater varied from 0.01 to 0.29 mg/l with insignificant difference in mean concentrations between wet (0.06±0.06 mg/l) and dry periods (0.05±0.06mg/l) (Table 2) along both east and west coast of India. The State-level mean concentration of ammonium was higher in the States located along the east coast (0.06-0.12 mg/l) than west coast (0.02-0.06 mg/l) during wet period whereas it was 0.08 to 0.11 and 0.01 to 0.04 mg/l respectively during dry period (Table 3). Like groundwater, ammonium concentrations in the soil was higher in the east

(0.41 ± 0.35 mg/g) and west (0.15 ± 0.31 mg/g) coasts of India during wet period (Table 4). The concentrations of nitrate in the groundwater varied from 0.09 to 150 mg/l being higher during the dry (27.2 ± 32 mg/l) than wet period (10.8 ± 8 mg/l) (Table 2). During both periods, the concentration of nitrate displayed insignificant differences between east and west coast of India (Fig 2; Table 2). During wet period, the concentration of nitrate decreased from north to south (Odisha; 16.6 ± 7 mg/l followed by Andhra Pradesh (12.06 ± 8 mg/l), Tamil Nadu (11.0 ± 7 mg/l) with the exception of West Bengal (6.59 ± 4 mg/l) along the east and west coast of India (Gujarat; 15.57 ± 7 mg/l followed by Maharashtra (13.6 ± 9 mg/l), Goa (6.8 ± 10 mg/l), Karnataka (5.52 ± 4 mg/l) and Kerala (5.8 ± 9 mg/l) (Table 3). In the case of dry period, State-mean concentration of nitrate increased from north to south (2.63 to 63.39 mg/l) along the east coast but significant pattern was not observed along the west coast of India (Table 3). The concentration of nitrate in soil during wet period varied from 0.01 to 0.85 mg/g and higher concentrations were found in the east (0.22 ± 0.20 mg/g) than west coast of India (0.32 ± 0.24 mg/g) (Table 4).

The concentration of dissolved inorganic phosphate (DIP) in the groundwater varied from 0.07 to 2.17 mg/l (Fig. 3) with higher concentrations during the dry (0.76 ± 0.5 mg/l) than wet period (0.54 ± 0.3 mg/l) (Table 2). During both the periods, the concentration of DIP in the east coast was higher (0.65 ± 0.3 mg/l; 0.94 ± 0.5 mg/l respectively during wet and dry periods) than west coast of India (0.43 ± 0.2 mg/l; 0.60 ± 0.3 mg/l respectively) (Table 2). The State-mean concentration of DIP decreased from north to south along both east and west coast of India during dry period but it was increased along east coast and decreased along west coast during wet period (Table 3). DIP in the soil varied from 0.01 to 0.28 mg/g during the wet period with higher along the east coast (0.13 ± 0.08 mg/g) than in the west coast (0.07 ± 0.07 mg/g) (Table 4).

The concentration of urea varied from 0.01 to 27.9 mg/l, with higher concentrations during the dry (7.21 ± 5 and 6.17 ± 4 mg/l on the east and west coasts respectively) than wet period (1.07 ± 1 and 0.71 ± 0.6 mg/l respectively) (Table 2; Fig 4). The State-mean concentration of urea decreased from north to south along east coast whereas it decreased along west coast only during dry period. In case of wet period, State-mean concentration decreased from north to south along west coast but no significant trend is noticed along east coast (Table 3). Urea in soil varied from 0.01 to 0.81 mg/g during the wet period and the highest concentrations were found in the east coast (0.37 ± 0.25 mg/g) than in the west coast of India (0.19 ± 0.2 mg/g) (Table 4).

During wet period, nitrate in the groundwater showed a significant linear positive correlation with nitrate in the soil ($r^2=0.60$, $p<0.01$; Fig. 5a). Nitrate in groundwater linearly correlated with bacterial respiration ($r^2=0.59$, $p<0.001$; Fig. 5b) and inversely correlated with DO saturation ($r^2=0.49$,

$p < 0.001$; Fig. 5c) and ammonium ($r^2 = 0.50$, $p < 0.001$; Figure is not shown). During dry period, nitrate in water showed a significant linear positive relation with bacterial respiration ($r^2 = 0.62$, $p < 0.001$; Fig. 6a) and inverse relation with ammonium ($r^2 = 0.56$, $p < 0.001$; Fig. 6b), urea ($r^2 = 0.65$, $p < 0.001$; Fig. 6c), and DO saturation ($r^2 = 0.63$, $p < 0.001$; Fig. 6d). DIP in groundwater displayed a significant positive correlation with DIP in the soil ($r^2 = 0.42$, $p < 0.05$, $n = 45$) (Fig. 5d). Urea in the groundwater showed a significant correlation with urea in soil ($r^2 = 0.41$, $p < 0.05$; Fig. 5e). Urea in the groundwater showed significant inverse relation with ammonium in the groundwater ($r^2 = 0.58$, $p < 0.01$; Fig. 5f). Ammonium in the groundwater showed a linear positive correlation with ammonium in soils ($r^2 = 0.61$, $p < 0.01$) during wet period (Figure is not shown). The State mean concentration of DIN and DIP in the groundwater showed relationship with state-wise fertilizer consumption in India during wet (kharif) and dry (Rabi) period (Fig 7) and it is significant for DIN but not for DIP.

4. Discussion

4.1. Variability in concentrations and sources of nutrients in the groundwater along the Indian coast

The concentrations of nutrients (ammonium, nitrate, phosphate and urea) in the groundwater along the Indian coast varied significantly with space and time (Fig. 2-4). The concentration of ammonium (0.01 and 0.29 mg/l) in the Indian coastal groundwater is consistent with the same observed in the groundwater elsewhere in the world (0.01-0.19 mg/l; Table 5). Ammonium concentrations in the groundwater displayed linear relationship with ammonium in soil indicating that soil may be a significant source of ammonium in the groundwater. Ammonium in the groundwater displayed inverse relation with urea (Fig. 5f) indicating that decomposition of urea may be contributed to ammonium in the groundwater suggesting that agricultural fertilizers may be possible source of ammonium to the ground water. The mean concentration of nitrate in the groundwater in India (19.3 mg/l) is higher than that observed in United Kingdom, United States of America, Australia, New Zealand, most parts of Europe, and southeast Asia, but lower than that observed in Germany (25.1 mg/l) and Africa (52.8 mg/l) (Table 5). The concentrations of nitrate displayed significant positive relationship with soil nitrate (Fig. 5a), bacterial respiration (Fig. 5b; and 6a) and inverse relation with dissolved oxygen saturation (Fig. 5c and 6d) and ammonium (Fig. 6b) during both wet and dry periods suggesting that organic matter decomposition, nitrification, and soil nitrogen may be major sources of nitrate to the groundwater.

The mean concentrations of DIP (0.65 ± 0.4 mg/l) in the groundwater along the Indian coast is consistent with that reported elsewhere in the world (0.76 ± 0.5 mg/l), but significantly lower than that found in Africa (3.35 mg/l) and Canada (11.6 mg/l) and French Polynesia (2.5 mg/l) (Table 5). Both

DIP and urea in the groundwater displayed linear relation with phosphate (Fig. 5d) and urea in the soil (Fig. 5e) suggesting that soil may be a significant source to the groundwater DIP and Urea. Microbial decomposition of soil organic matter may be a significant source of DIP to the ground water; however, the relationship of DIP in the groundwater with bacterial respiration and DO saturation is insignificant indicating adsorption of DIP in groundwater on to sediment particles (Spiteri et al., 2008; Buss et al., 2004; Lorah et al., 2009). A wide range of variability in nutrient concentrations in the groundwater along the Indian coast could be due to variations in soil characteristics, amount of fertilizer used, precipitation and seasonality in the groundwater levels.

4.1.1. Influence of soil characteristics

In India, most common soil types are Alluvial, Black, Red, and Yellow along the Indian coast (Fig. 1; Table 1; Bhattacharyya and Kumar, 2015). Alluvial soil is the most abundant soil type along the east coast of India (Fig.1) whereas Red and Yellow soils are abundant along the west coast of India, except in Gujarat where alluvial soil is dominant. All these soils contain very low content of nitrogen, phosphorus and organic matter (Shahid and Naseem 2017).

Despite Alluvial soil is dominant along the east coast of India, the State-wise mean concentrations of nitrate were high in the groundwater collected from the Odisha, Andhra Pradesh, and Tamil Nadu States, but low in the West Bengal during the wet period (Table. 3). Along the west coast, Red and Yellow soils are dominant, but high concentration of nitrate were observed in the ground waters from Maharastra but low in Karnataka, Goa and Kerala (Table 3) during wet period. In case of dry period, high nitrate concentration was observed in Andhra Pradesh, Tamil Nadu, Goa, and Gujarat (28.8 to 63.39 mg/l) but lower concentration are observed in Odisha, and West Bengal and all these regions were dominated by Alluvial soil. Similar behavior was observed in case of DIP distribution. Interestingly both DIN and DIP were high in Odisha, Andhra Pradesh, Tamil Nadu, Maharastra and Gujarat during wet period whereas the states with high concentration of DIP were not same during the dry period. The inconsistency between the nature of soil and concentration of nitrogen and phosphorus content in the soil indicating that it may not be the sole reason for spatial variations in DIN and DIP concentrations in the coastal groundwater.

4.1.2. Influence of agricultural fertilizer use

Coastal States are known to produce more rice followed by sugar cane throughout the year in India due to abundant supply of water and 40-80% of the agricultural land is cultivated using groundwater (Table 6). Since Indian coastal regions are enriched with alluvial, Red and Yellow soils, which is poor in phosphate and nitrogen (Shahid and Naseem 2017), and farmers use excessive fertilizers for

better yield.. The State-wise mean amount of fertilizer used during Kharif (wet) and Rabi (dry) period in each state showed significant correlation with mean concentrations of DIN ($r^2=0.45$ and $r^2=0.35$; $p<0.01$ during the wet and dry periods respectively). Adsorption of phosphate on to the sediment particles, which removes groundwater phosphate (Buss et al., 2004; Lorah et al., 2009; Spiteri et al., 2008) is held responsible for not getting a similar relationship of high significance between DIP in the groundwater and the fertilizer used. However with the soil nutrients, both nitrate, and phosphate of the groundwater showed significant linear correlations with nitrate ($r^2=0.60$, $p<0.01$; Fig. 5a), phosphate ($r^2=0.42$, $p<0.05$; Fig. 5d) and urea ($r^2=0.41$, $p<0.05$; Fig. 5e) during the wet period . About 6 to 7 times decrease in urea concentrations were observed from dry (6.6 ± 3 mg/l) to wet period (0.9 ± 0.7 mg/l) suggesting that fertilizer (urea) input was significantly higher during former than the latter period except in Odisha, Karnataka and Maharashtra States. The strong correlation between fertilizer utilization in the State and mean concentrations in the groundwater suggests fertilizers are one of the major sources of nutrients in the coastal groundwater in India.

4.1.3. Role of internal processes on nutrients in the ground water

Several processes in the ground water, such as bacterial respiration, nitrification and denitrification may contribute in accumulation of nutrients in groundwater. A positive relation was observed between nitrate with bacterial respiration ($r^2=0.64$, $p<0.001$ and 0.62 , $p<0.001$ respectively during the wet and dry periods; Fig. 5b, and 6a) and inverse relation with DO saturation ($r^2=0.55$, $p<0.001$ and 0.63 , $p<0.001$; Fig. 5c and 6d) and ammonium in the groundwater ($r^2=0.62$, $p<0.001$ and 0.69 , $p<0.001$; Fig. 6b), which suggests that decomposition of organic matter and nitrification also contributed to the nitrate concentrations in the groundwater during the wet and dry periods. The concentration of nitrate was more than twice during dry period (27.2 ± 32 mg/l) than wet period (10.8 ± 8 mg/l) in the east and west coast of India and it is consistent with higher bacterial respiration rates during the former period (3.5 ± 2 mg/l/h) than the latter (0.85 ± 0.5 mg/l/h) (Table 2). Therefore, higher concentrations of nutrients during dry period may be due to higher decomposition of soil organic matter resulting in release of more nitrate into the groundwaters. Though bacterial respiration of organic matter releases both nitrogen and phosphorus, the removal of phosphate in association with particles prevented a significant relation to be observed between bacterial respiration and phosphate using either period. This resulted in weak concentration gradient of phosphate between dry (0.76 ± 0.5 mg/l) and wet periods (0.54 ± 0.3 mg/l) compared to nitrate (27.23 ± 32 and 10.86 ± 8 mg/l during dry and wet periods respectively) (Table 2). The mean N:P ratios during dry period (52 ± 73) was much above the Redfield value but was much lower during wet period (25 ± 25). The difference in N:P ratios during the wet and dry period was mainly driven by concentration of nitrate

as variability of phosphate is not significantly different (Table 2). The strong correlation between bacterial respiration, ammonium, and DO saturation with mean concentrations in the groundwater nitrate suggests that in situ soil organic carbon decomposition, and nitrification seems to be contributing to the accumulation of nutrients in the groundwater.

4.2. Nutrient export from groundwater to the adjacent coastal regions

The concentration of nitrate in the coastal water varied between 0.007 and 0.03 mg/l along the Indian coast (De Sousa et al., 1996; Gauns et al., 2005; Sarma et al., 2013) and the concentration of nutrient in the groundwater is an order of magnitude higher than coastal waters (Table 2 and 3). Therefore, the exchange of groundwater with adjacent coastal water may have significant impact on coastal ecosystem. Manivannan and Elango, (2019) found that the groundwater levels are below msl (0 to -2 m msl) in about 1900 km² of the Indian coast and it was increased by about 400 km² during the past decade. They further noticed that about 7% of the total coastal area of India is readily affected by seawater intrusion. On the other hand, ~57% of groundwater levels along the Indian coast line are above the msl (0-10m; Manivannan and Elango, 2019) where significant amount of groundwater may be exchanged with coastal waters due to hydrological gradient.

The rate of SGD flux is estimated using different tracers, mainly Ra²²⁸, Rn²²² and Sr⁸⁷, at several locations along the Indian coast (Table 7). The mean SGD fluxes varied between 1.6 x 10⁴ m³/d in Kerala coast and 1.75 x 10¹¹ m³/d in the Karnataka coast (Table 7). The SGD fluxes estimated along the Indian coast are consistent with the rates found in different regions in the south-east Asia (Table 8). In order to quantify the amount of nutrients discharged to the Indian coastal waters, nutrients flux was computed based on the rate of SGD given in Table 7 and mean concentration of DIN and DIP in the coastal groundwater. The annual mean flux of DIN and DIP is observed to be 0.103±0.05 and 0.021±0.01 Tg (1 Tg=10¹² g) to the western coastal Bay of Bengal (east coast of India) and 0.06±0.05 and 0.015±0.05 Tg/y to the eastern coastal Arabian Sea (West coast of India) respectively. The rate of nutrient flux through SGD is higher along the eastern than western Indian coastal water that may be due to high permeability of the sediments in the former region as mostly rocky environment exists in the latter coast. However, the strong salinity stratification due to freshwater discharge by rivers in the along the east prevent vertical mixing of nutrients supply to the upper ocean along the east coast of Indian (Sarma et al., 2012; 2013). In contrast, weak stratification due to less river discharge aids vertical mixing and nutrients supply to the surface in the western Indian coastal region (De Sousa et al., 1996; Gauns et al., 2005). Although nutrients supply to surface layers through vertical mixing is low in the coastal BoB, the supply of nutrients through SGD being large (almost twice than that of in the Arabian Sea) can compensate nutrient deficiency to a large extent.

Nevertheless, the flux of DIN through SGD (0.163 ± 0.07 Tg/y) is close to that of riverine fluxes to the Indian coast (0.22 ± 0.05 Tg/y) whereas DIP flux through SGD (0.036 ± 0.02 Tg/y) lower than that of riverine flux (0.11 ± 0.03 Tg/y; Krishna et al., 2016).

Apart from nutrients through SGD, both Bay of Bengal and Arabian Sea receives external nutrient inputs through atmospheric deposition, river discharge, nitrogen fixation. The source of N and P through atmospheric deposition is estimated by Srinivas and Sarin (2013) as 0.09 and 0.014 Tg/y to the Arabian Sea and 0.12 and 0.021 Tg/y to the Bay of Bengal respectively. The discharge of N and P to the Arabian Sea and Bay of Bengal were estimated to be 0.1 and 0.03 Tg/y respectively to the Arabian Sea whereas 0.12 and 0.08 Tg/y to the Bay of Bengal respectively (Krishna et al., 2016) (Table 9). Based on these estimates, the external nitrogen supply to the Arabian Sea is dominated by riverine (40%) source followed by atmospheric (36%) and SGD (24%) (Table 9). In case of Bay of Bengal, all these sources almost contributed equally (30-35%). The contribution of P by river (51%) is more significant and both atmospheric and SGD sources (24-25%) are equivalent in the Arabian Sea. Similar P by river is higher (61%) in the Bay of Bengal followed by equal contribution by atmospheric and SGD inputs (17%) (Table 9).

Gauns et al. (2005) reported equal primary production along the east and west coast of India during non-monsoon period and this could not be explained through physical processes as strong stratification occurs in the former than latter period. This paradox may be now explained through nutrients supply by SGD. On the other hand, occurrence of phytoplankton blooms, including harmful algal blooms (HABs), were reported associating with seasonal suboxia along the west coast of India during summer monsoon (Naqvi et al., 1998) and central west coast of India during winter (Gomes et al., 2014). Seasonal occurrence of oxygen minimum zone is reported in the Bay of Bengal (Sarma et al., 2013). The potential reasons for occurrence of such blooms and oxygen minimum zone were attributed to coastal upwelling, riverine nutrient flux and untreated sewage (Naqvi et al., 2000; Gomes et al., 2014; Gupta et al., 2016; Sarma et al., 2013). The nutrients brought by SGD may also be included as a potential cause. However, the mechanism of formation of algal blooms and its link to SGD needs further evaluation. A caution is warranted on the estimate of SGD along the Indian coast due to several assumptions made such as excluding diurnal variability in SGD (Kobayashi et al., 2017; Taniguchi et al., 2019), monthly variability in variability in nutrients concentrations in the groundwater and spatial variations in the SGD rates. In addition to this, the behavior of N species is very complicated and it is governed on the redox conditions and remineralization of organic matter in the groundwater and coastal waters. For instance, ammonium is often found to be removed by nitrification in oxic conditions (Spiteri et al., 2008; Anwar et al., 2014; Anschutz et al., 2016).

Dissolved N can be removed under the anoxic conditions as molecular nitrogen. This would modify N fluxes to the coastal regions. Similarly, phosphate adsorbs on the particles and removes to the sediment. Therefore, our estimation may be relatively on the higher side due to neglecting the above said processes in the estimations due to lack of data. This study suggests that impact of SGD on coastal ecosystem and primary production may be significant and needs an in-depth studies in the future.

Summary

Submarine Groundwater Discharge (SGD) is a major pathway of transport of dissolved inorganic nutrients (DIN, DIP) to coastal waters, where the scarcity of DIN limits the primary production. The concentrations of DIN and DIP in the groundwater ranged from 0.09 to 240 mg/l and 0.18 to 3.08 mg/l, with higher during the dry (27.3 ± 32 mg/l and 0.8 ± 0.5 mg/l) compared to the wet period (11.0 ± 8 mg/l and 0.5 ± 0.3 mg/l) respectively. The mean concentration of DIN in the groundwater in India (17.5 mg/l) is somewhat higher than that observed in several developed nations. The concentrations of nitrate in groundwater displayed significant positive relationship with soil nitrate, bacterial respiration and inverse relation with dissolved oxygen saturation and ammonium suggesting that organic matter decomposition, nitrification, and soil nitrogen are major sources of nitrate in the groundwater. Furthermore, the amount of fertilizer used in each coastal State of India showed strong correlation with mean concentration of nitrate suggesting that fertilizers are one of the major sources of nutrients in the coastal groundwater in India. The flux of DIN and DIP through SGD (0.163 ± 0.07 and 0.036 ± 0.02 Tg/y respectively) is almost close to that of riverine fluxes to the Indian coast (0.22 ± 0.05 and 0.11 ± 0.03 Tg/y respectively) suggesting that significant amount of nutrients is brought to the coastal waters through SGD and it can impact coastal primary production and ecosystem dynamics, which needs evaluation.

Recently Chen et al. (2019) noticed that increasing leaf area of vegetation from 2000 to 2017 in India and China due to dominant driver of CO₂ fertilization. China alone accounts for 25% of the global net increase in leaf area with only 6.6% of global vegetated area. The greening in China is from forests (42%) and croplands (32%), but in India is mostly from croplands (82%) with minor contribution from forest (4.4%). The food production in India has increased by over 35% since 2000 mostly owing to an increase in harvested area through multiple cropping facilitated by fertilizer use and surface and or groundwater irrigation (Chen et al., 2019). In such case the accumulation of excess fertilizers in the groundwater is expected and would increase its discharge to the coastal waters. Therefore, long-term observations needed to be set up to continuously monitoring groundwater nutrient levels and its impact on the coastal ecosystem.

Ethics approval and consent to participate

Not applicable.

Consent of publication

Not applicable

Availability of data and material

The datasets used in this study are available from the corresponding author on request.

Competing interests

The authors declare that we have no competing interests

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Authors' contributions:

BSKK, RV, DBR, MHKP – collection of samples, analysis of samples

BSKK – preparation of initial draft of the manuscript and computations.

VRK, NS – Data processing, drawing figures, interpretation of data

VVSSS – Concept of the work, execution of the sampling and analysis plan, interpretation of data and finalization of manuscript.

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Table 1: The location, well depth, mean rainfall and soil type in the region of ground water samples are given. The stations with asterisk (*) denotes where soil samples are collected.

S.no	Latitude	Longitude	Station Name	well depth (m)	Above mean sea level (m)	Rain fall ^s (mm)	Soil type
1	22.1889	88.7132	WB1	80	6	136	Alluvial
2	22.2271	88.4461	WB2	78	7	101	Alluvial
3	22.2181	88.0949	WB3	75	7	97.6	Alluvial
4	22.4632	87.9733	WB4	70	7	144	Alluvial
5	22.0428	88.0657	WB5	70	32	86	Alluvial
6	22.0129	87.8173	WB6	65	6	113	Alluvial
7	21.8712	87.7571	WB7 [#]	70	9	125	Alluvial
8	21.7852	87.7441	WB8	75	7	158	Alluvial
9	21.7205	87.6637	WB9	76	6	140	Alluvial
10	21.6234	87.5206	WB10	80	8	120	Alluvial
11	21.6701	87.2859	OD11	35	11	116.5	Laterite
12	21.3413	86.7631	OD12 [#]	30	14	113	Laterite
13	20.8119	86.7609	OD13 [#]	40	19	97	Alluvial
14	20.5014	86.3277	OD14 [#]	40	6	80	Alluvial
15	20.0989	86.1886	OD15	35	8	76	Alluvial
16	19.8422	85.9032	OD16	35	20	73	Alluvial
17	19.9016	85.3656	OD17	30	7	65	Alluvial
18	19.5972	85.1211	OD18	35	10	97	Alluvial
19	19.3503	84.9882	OD19	35	8	125	Alluvial
20	19.1611	84.7112	OD20	38	5	52	Alluvial
21	18.9408	84.5648	AP21	200	5	132	Alluvial
22	17.7275	83.3302	AP22	100	8	167	Alluvial
23	17.3167	82.5695	AP23	30	6	170	Alluvial
24	16.9569	82.2552	AP24 [#]	25	16	174	Alluvial
25	16.3966	81.8095	AP25 [#]	20	6	134	Alluvial
26	16.1777	81.1385	AP26	15	20	94	Alluvial
27	15.7809	80.3801	AP27	20	22	77	Alluvial
28	15.5046	80.0365	AP28 [#]	16	18	60	Alluvial
29	14.2971	80.0904	AP29	13=	19	91	Alluvial
30	13.5888	80.0343	AP30 [#]	50	20	123	Alluvial
31	12.9399	80.2401	TN31	30	18	79	Alluvial
32	12.3821	80.0894	TN32 [#]	16	19	35	Alluvial
33	11.7479	79.7687	TN33	23	22	45	Alluvial
34	11.0311	79.8503	TN34 [#]	20	20	20	Alluvial
35	10.3765	79.8497	TN35	20	15	16	Alluvial
36	10.0388	79.2333	TN36 [#]	20	14	31	Alluvial
37	9.3316	78.9741	TN37	10	17	45	Alluvial
38	9.2353	78.7875	TN38 [#]	18	20	90	Alluvial
39	8.7571	78.1566	TN39	15	22	153	Alluvial
40	8.0885	77.5493	TN40 [#]	18	25	178	Alluvial
41	8.3945	77.0885	KL41	200	13	124	Red and Yellow
42	8.8634	76.6935	KL42 [#]	120	12	71	Red and Yellow
43	9.3183	76.4217	KL43	30	8	116	Red and Yellow
44	9.9994	76.2929	KL44	25	5	146	Red and Yellow
45	10.4192	76.1052	KL45 [#]	20	5	126	Red and Yellow
46	10.9248	75.9139	KL46	15	5	146	Red and Yellow
47	11.4696	75.6548	KL47 [#]	15	6	165	Red and Yellow
48	11.8847	75.3725	KL48	15	10	225	Laterite
49	12.2428	75.1449	KL49 [#]	20	13	195	Laterite
50	12.6821	74.9032	KL50	20	10	205	Laterite
51	12.7884	74.8593	KA51 [#]	80	13	119	Laterite

52	12.8748	74.8292	KA52	60	9	172	Laterite
53	13.2228	74.7845	KA53 [#]	50	13	182	Laterite
54	13.3889	74.7431	KA54	50	10	187	Laterite
55	13.6189	74.6926	KA55 [#]	50	14	193	Red and Yellow
56	13.8183	74.6343	KA56	40	14	126	Red and Yellow
57	14.0271	74.5291	KA57 [#]	25	25	120	Red and Yellow
58	14.2417	74.4468	KA58	15	5	101	Red and Yellow
59	14.4308	74.4231	KA59 [#]	30	7	112	Red and Yellow
60	14.6572	74.3121	KA60	30	5	120	Red and Yellow
61	14.9111	74.0816	GA61 [#]	15	11	135	Laterite
62	14.9592	74.0551	GA62	15	8	150	Laterite
63	15.0021	74.0786	GA63 [#]	15	12	146	Laterite
64	15.0421	73.9991	GA64	15	15	135	Laterite
65	15.1261	73.9495	GA65 [#]	10	16	128	Laterite
66	15.2068	73.9504	GA66	20	11	132	Laterite
67	15.7103	73.7257	GA67 [#]	20	16	145	Laterite
68	15.7243	73.6882	MH68 [#]	10	12	150	Red and Yellow
69	16.1077	73.4725	MH69	20	15	175	Red and Yellow
70	16.4731	73.3943	MH70 [#]	45	35	190	Red and Yellow
71	16.9938	73.317	MH71	25	25	205	Red and Yellow
72	17.5015	73.1855	MH72	35	44	145	Red and Yellow
73	17.9758	73.0918	MH73 [#]	15	58	86.5	Laterite
74	18.6471	72.8783	MH74	25	10	95	Laterite
75	19.3753	72.8275	MH75	25	6	103	Laterite
76	19.9562	72.8231	MH76	20	20	124	Laterite
77	20.1356	72.8065	MH77 [#]	50	12	94	Laterite
78	20.5981	72.9424	GJ78 [#]	30	6	111	Alluvial
79	21.1242	72.7861	GJ79	30	20	125	Alluvial
80	22.2507	73.1852	GJ80 [#]	35	22	106	Alluvial
81	21.6102	72.2693	GJ81	30	30	113	Alluvial
82	21.0591	71.7591	GJ82 [#]	30	17	95	Alluvial
83	20.7613	71.0551	GJ83	35	30	73	Alluvial
84	20.8979	70.3932	GJ84	40	25	82	Alluvial
85	21.6439	69.6032	GJ85 [#]	30	14	67	Alluvial
86	22.2399	68.9323	GJ86	25	12	98	Alluvial
87	22.4717	70.0786	GJ87 [#]	25	16	78	Alluvial

[#]indicates that where the soil samples are collected during wet period.

^{\$}http://imd pune.gov.in/Clim_RCC_LRF/Annual_Climate_Summary/annual_summary_2015.pdf

Table 2: Mean (\pm SD) of groundwater properties along Indian coast during wet and dry periods
 ***, **, * represents $p < 0.001$, 0.05, 0.1 respectively.

Parameter	Wet Period	Coast	Wet period	Dry Period	Coast	Dry period	t-value	df																																																																																																																																										
Temperature ($^{\circ}$ C)	30.21 \pm 1.4	East	30.92 \pm 1.3	26.39 \pm 4.4	East	27.61 \pm 4.6	7.65***	87																																																																																																																																										
		West	29.61 \pm 1.2		West	25.35 \pm 4.1			Conductivity (mS/cm)	1.76 \pm 3.2	East	2.60 \pm 4	6.04 \pm 8.7	East	8.84 \pm 9	4.26***	86	West	1.06 \pm 1	West	3.67 \pm 7	pH	6.53 \pm 0.7	East	6.95 \pm 0.3	6.67 \pm 0.6	East	7.06 \pm 0.3	1.29*	87	West	6.17 \pm 0.8	West	6.33 \pm 0.7	DO Saturation (%)	61.16 \pm 27	East	64.56 \pm 28	54.69 \pm 24	East	44.27 \pm 22	1.62**	84	West	58.13 \pm 26	West	63.54 \pm 23	BR (mg/l/d)	0.85 \pm 0.5	East	0.76 \pm 0.5	3.54 \pm 2	East	4.33 \pm 3.1	7.86***	87	West	0.93 \pm 0.5	West	2.87 \pm 2.9	Ammonium (mg/l)	0.06 \pm 0.06	East	0.09 \pm 0.07	0.05 \pm 0.06	East	0.09 \pm 0.07	1.32*	85	West	0.04 \pm 0.04	West	0.02 \pm 0.02	Nitrite(mg/l)	0.12 \pm 0.2	East	0.14 \pm 0.1	0.05 \pm 0.09	East	0.07 \pm 0.13	3.5***	86	West	0.10 \pm 0.2	West	0.03 \pm 0.04	Nitrate(mg/l)	10.86 \pm 8	East	11.47 \pm 7	27.23 \pm 32	East	29.53 \pm 38	3.4***	86	West	10.33 \pm 9	West	25.33 \pm 25	DIN(mg/l)	11.04 \pm 8	East	11.69 \pm 7	27.33 \pm 32	East	29.76 \pm 25	3.1***	86	West	10.46 \pm 9	West	25.45 \pm 25	DIP(mg/l)	0.54 \pm 0.3	East	0.65 \pm 0.3	0.76 \pm 0.5	East	0.94 \pm 0.5	3.6***	86	West	0.43 \pm 0.2	West	0.60 \pm 0.3	DIN:DIP	25 \pm 25	East	20.62 \pm 15	52 \pm 72	East	48.46 \pm 69	4.9***	86	West	28.69 \pm 31	West	55.67 \pm 75	Urea(mg/l)	0.88 \pm 0.9	East	1.07 \pm 1	6.64 \pm 5	East	7.21 \pm 5	9.6***
Conductivity (mS/cm)	1.76 \pm 3.2	East	2.60 \pm 4	6.04 \pm 8.7	East	8.84 \pm 9	4.26***	86																																																																																																																																										
		West	1.06 \pm 1		West	3.67 \pm 7			pH	6.53 \pm 0.7	East	6.95 \pm 0.3	6.67 \pm 0.6	East	7.06 \pm 0.3	1.29*	87	West	6.17 \pm 0.8	West	6.33 \pm 0.7	DO Saturation (%)	61.16 \pm 27	East	64.56 \pm 28	54.69 \pm 24	East	44.27 \pm 22	1.62**	84	West	58.13 \pm 26	West	63.54 \pm 23	BR (mg/l/d)	0.85 \pm 0.5	East	0.76 \pm 0.5	3.54 \pm 2	East	4.33 \pm 3.1	7.86***	87	West	0.93 \pm 0.5	West	2.87 \pm 2.9	Ammonium (mg/l)	0.06 \pm 0.06	East	0.09 \pm 0.07	0.05 \pm 0.06	East	0.09 \pm 0.07	1.32*	85	West	0.04 \pm 0.04	West	0.02 \pm 0.02	Nitrite(mg/l)	0.12 \pm 0.2	East	0.14 \pm 0.1	0.05 \pm 0.09	East	0.07 \pm 0.13	3.5***	86	West	0.10 \pm 0.2	West	0.03 \pm 0.04	Nitrate(mg/l)	10.86 \pm 8	East	11.47 \pm 7	27.23 \pm 32	East	29.53 \pm 38	3.4***	86	West	10.33 \pm 9	West	25.33 \pm 25	DIN(mg/l)	11.04 \pm 8	East	11.69 \pm 7	27.33 \pm 32	East	29.76 \pm 25	3.1***	86	West	10.46 \pm 9	West	25.45 \pm 25	DIP(mg/l)	0.54 \pm 0.3	East	0.65 \pm 0.3	0.76 \pm 0.5	East	0.94 \pm 0.5	3.6***	86	West	0.43 \pm 0.2	West	0.60 \pm 0.3	DIN:DIP	25 \pm 25	East	20.62 \pm 15	52 \pm 72	East	48.46 \pm 69	4.9***	86	West	28.69 \pm 31	West	55.67 \pm 75	Urea(mg/l)	0.88 \pm 0.9	East	1.07 \pm 1	6.64 \pm 5	East	7.21 \pm 5	9.6***	86	West	0.71 \pm 0.6	West	6.17 \pm 4								
pH	6.53 \pm 0.7	East	6.95 \pm 0.3	6.67 \pm 0.6	East	7.06 \pm 0.3	1.29*	87																																																																																																																																										
		West	6.17 \pm 0.8		West	6.33 \pm 0.7			DO Saturation (%)	61.16 \pm 27	East	64.56 \pm 28	54.69 \pm 24	East	44.27 \pm 22	1.62**	84	West	58.13 \pm 26	West	63.54 \pm 23	BR (mg/l/d)	0.85 \pm 0.5	East	0.76 \pm 0.5	3.54 \pm 2	East	4.33 \pm 3.1	7.86***	87	West	0.93 \pm 0.5	West	2.87 \pm 2.9	Ammonium (mg/l)	0.06 \pm 0.06	East	0.09 \pm 0.07	0.05 \pm 0.06	East	0.09 \pm 0.07	1.32*	85	West	0.04 \pm 0.04	West	0.02 \pm 0.02	Nitrite(mg/l)	0.12 \pm 0.2	East	0.14 \pm 0.1	0.05 \pm 0.09	East	0.07 \pm 0.13	3.5***	86	West	0.10 \pm 0.2	West	0.03 \pm 0.04	Nitrate(mg/l)	10.86 \pm 8	East	11.47 \pm 7	27.23 \pm 32	East	29.53 \pm 38	3.4***	86	West	10.33 \pm 9	West	25.33 \pm 25	DIN(mg/l)	11.04 \pm 8	East	11.69 \pm 7	27.33 \pm 32	East	29.76 \pm 25	3.1***	86	West	10.46 \pm 9	West	25.45 \pm 25	DIP(mg/l)	0.54 \pm 0.3	East	0.65 \pm 0.3	0.76 \pm 0.5	East	0.94 \pm 0.5	3.6***	86	West	0.43 \pm 0.2	West	0.60 \pm 0.3	DIN:DIP	25 \pm 25	East	20.62 \pm 15	52 \pm 72	East	48.46 \pm 69	4.9***	86	West	28.69 \pm 31	West	55.67 \pm 75	Urea(mg/l)	0.88 \pm 0.9	East	1.07 \pm 1	6.64 \pm 5	East	7.21 \pm 5	9.6***	86	West	0.71 \pm 0.6	West	6.17 \pm 4																					
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		West	58.13 \pm 26		West	63.54 \pm 23			BR (mg/l/d)	0.85 \pm 0.5	East	0.76 \pm 0.5	3.54 \pm 2	East	4.33 \pm 3.1	7.86***	87	West	0.93 \pm 0.5	West	2.87 \pm 2.9	Ammonium (mg/l)	0.06 \pm 0.06	East	0.09 \pm 0.07	0.05 \pm 0.06	East	0.09 \pm 0.07	1.32*	85	West	0.04 \pm 0.04	West	0.02 \pm 0.02	Nitrite(mg/l)	0.12 \pm 0.2	East	0.14 \pm 0.1	0.05 \pm 0.09	East	0.07 \pm 0.13	3.5***	86	West	0.10 \pm 0.2	West	0.03 \pm 0.04	Nitrate(mg/l)	10.86 \pm 8	East	11.47 \pm 7	27.23 \pm 32	East	29.53 \pm 38	3.4***	86	West	10.33 \pm 9	West	25.33 \pm 25	DIN(mg/l)	11.04 \pm 8	East	11.69 \pm 7	27.33 \pm 32	East	29.76 \pm 25	3.1***	86	West	10.46 \pm 9	West	25.45 \pm 25	DIP(mg/l)	0.54 \pm 0.3	East	0.65 \pm 0.3	0.76 \pm 0.5	East	0.94 \pm 0.5	3.6***	86	West	0.43 \pm 0.2	West	0.60 \pm 0.3	DIN:DIP	25 \pm 25	East	20.62 \pm 15	52 \pm 72	East	48.46 \pm 69	4.9***	86	West	28.69 \pm 31	West	55.67 \pm 75	Urea(mg/l)	0.88 \pm 0.9	East	1.07 \pm 1	6.64 \pm 5	East	7.21 \pm 5	9.6***	86	West	0.71 \pm 0.6	West	6.17 \pm 4																																		
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Table 3: State-wise mean (standard deviation) concentration of nutrients in the Indian coast during dry (wet) period

State	Nitrate	NH4	Urea	DIP
West Bengal	2.63±2 (6.59±4)	0.11±0.09 (0.12±0.08)	4.46±2.3 (1.22±1.1)	1.34±0.6 (0.46±0.1)
Odisha	20.70±11 (16.16±7)	0.08±0.08 (0.08±0.09)	4.65±1.4 (0.36±0.36)	1.05±0.5 (0.66±0.3)
Andhra Pradesh	28.80±37 (12.06±8)	0.08±0.05 (0.08±0.07)	9.11±7.4 (1.87±1.7)	0.85±0.5 (0.77±0.3)
Tamil Nadu	63.39±53 (11.09±7)	0.09±0.08 (0.06±0.05)	11.6±6.2 (0.91±0.06)	0.54±0.2 (0.78±0.3)
Kerala	12.18±11 (5.80±9)	0.04±0.03 (0.05±0.04)	11.7±7.0 (0.66±0.5)	0.35±0.1 (0.22±0.2)
Karnataka	21.98±19 (5.52±4)	0.01±0.01 (0.05±0.06)	5.83±2.3 (0.92±1)	0.47±0.2 (0.29±0.1)
Goa	60.31±33 (6.8±10)	0.01±0.01 (0.02±0.01)	4.70±1.8 (0.16±0.3)	0.54±0.2 (0.28±0.1)
Maharashtra	12.24±8 (13.6±9)	0.01±0.01 (0.06±0.03)	3.81±1.4 (0.89±0.4)	0.68±0.3 (0.49±0.3)
Gujarat	30.63±24 (15.57±7)	0.01±0.02 (0.04±0.02)	2.20±0.8 (0.84±0.6)	1.00±0.4 (0.69±0.2)

Table 4 : Mean and (\pm SD) of soil properties along east and west coast of India during wet period.

Parameter	Coast-wise	Wet Period	Wet Period
Ammonium (mg/g)	East	0.41 \pm 0.35	0.26 \pm 0.35
	West	0.15 \pm 0.31	
Nitrite (mg/g)	East	0.03 \pm 0.05	0.04 \pm 0.06
	West	0.03 \pm 0.03	
Nitrate (mg/g)	East	0.22 \pm 0.20	0.28 \pm 0.23
	West	0.32 \pm 0.24	
DIN (mg/g)	East	0.66 \pm 0.60	0.58 \pm 0.64
	West	0.50 \pm 0.58	
DIP (mg/g)	East	0.13 \pm 0.08	0.09 \pm 0.08
	West	0.07 \pm 0.07	
Urea (mg/g)	East	0.37 \pm 0.25	0.27 \pm 0.24
	West	0.19 \pm 0.20	

Table 5: Comparison of nitrate, nitrite, ammonium, DIP, DIN ocean in groundwater in and around the study site and across world.

Country	Area	NO3 (mg/l)	NO2 (mg/l)	NH4 (mg/l)	PO4 (mg/l)	DIN (mg/l)	N/P	References
UK	Hampshire	6.58	0.0017	0.01	0.04	6.6	164.8	Stuart and Smedley (2009)
UK	Oxford and Wilts	4.47	0.04	0.19	1.78	4.7	2.6	Cobbing et al., (2004)
UK	Otter	9.55	0.004	0.11	0.07	9.7	138.1	Bearcock and Smedley (2012)
USA	Kirckwood-Cohansey	3.6	0.03		0.01	3.6	363.0	Wieben, Baker and Nicholson (2013)
USA	South west of Jerome	7.5				7.5		Baldwinetal (2000)
Africa	North east Algeria	52.8	3.5		3.35	56.3	16.8	Halimi et al., (2016)
Australia	over all	11.5			0.082	11.5	140.2	Sundharam and Jane (2009)
New Zealand	over all	1.3			0.025	1.3	52.0	Sundharam and Jane (2009)
Australia	Great barrier Reef	11.3		0.01	0.06	11.3	188.5	Bhaskaran et al., (2002), Lenahan (2012), Thorburn et al., (2003)
Canada	Nanfei River basin				11.6			Qian et al., (2011)
China	Continuum	14.1	0.03	0.15		14.3		Yang et al., (2012)
Europe	Over all	10.5		0.1	0.08	10.6	132.5	Nixon et al., (2003)
Europe	Germany	25.1				25.1		Thomus and Dietrich (2006)
Japan	Arike Bay	2	0.002	0.06	0.035	2.1	58.9	Yasumoto et al., (2007)
Malaysia	Over all	5.5			0.06	5.5	91.7	Khan et al., (2007)
Moorea	French Polynesia	2.5			2.14	2.5	1.2	Knee et al., (2016)
Sri Lanka	Girivawa/Talawa	1.5			0.45	1.5	3.3	Young et al., (2010)
Pakistan	Over all	10.2				10.2		Tahir and Rasheed (2008)
India	Gujarat	0.23			0.08	0.2	2.9	Rupal et al., (2012)
India	Tamil Nadu	31	0.2	3.6	0.9	34.8	38.7	Vetrimuguran et al., (2013)
India	Wet period	10.8	0.12	0.06	0.5	11.2	25	Present Study
India	Dry period	27.2	0.05	0.05	0.7	27.5	52	Present Study

Table 6: The soil type, major crops and land use in different states in India

State	Total area (km ²)	Agriculture land (km ²)	% agricultural land	% irrigated land	% land using ground-water	Major crops	Types of soils
WB	88,752	603,45	68	62	38	Rice, Jute, Tea, Tobacco	Alluvial
OD	155,707	87460	56	26	74	Rice, Sugar cane, Jute, Oil Seeds	Red and Yellow, Laterite, Alluvial
AP	162,970	97782	60	38	62	Rice, Sugar cane, Cotton and Ground nuts	Alluvial, Red, Yellow and Black
TN	130058	50000	38	50	50	Rice, Millets, Cotton, Sorghum	Red and Yellow, Alluvial, Laterite
KL	38863	28000	72	22	78	Rice, Coffee, Rubber	Red and Yellow, laterite
KA	191,791	123,100	64	27	73	Rice, Cotton, Tobacco, Sugar cane, Coffee	
GA	3702	580	16	40	60	Rice, Vegetable, Fruits	Laterite
MH	307713	49234	16	16	84	Rice, Cotton, Sunflower	Black, Red and Yellow, Laterite
GJ	196,024	126,600	65	41	59	Rice, Wheat, Jowar, Cotton	Alluvial, Black

Table 7: SGD fluxes to Indian Ocean at different coastal regions along the India.

Region	Seepage rate (m³/d)	Method	Reference
West Bengal, India	5.4 x 10 ⁸	Sr ⁸⁷	Chakrabarthy et al., 2018
Odisha, India	3.1 x 10 ⁸	Ra ²²⁸	Rengarajan and Sarma 2015
Andhra Pradesh, India	3.1 x 10 ⁸	Ra ²²⁸	Rengarajan and Sarma 2015
Tamil Nadu, India	9.8 x 10 ⁸	Ra ²²⁸	Rengarajan et al., 2002; Barath Kumar et al., 2018; Jha et al., 2011)
Kerala, India	1.6 x 10 ⁴	Rn ²²²	Jacob et a., 2009
Karnataka, India	1.7 x 10 ¹¹	Rn ²²²	Sartandel et al., 2014; Kavitha, Chandrashekara and Paramesh 2017
Goa, India	4.7 x 10 ⁸	Sr ⁸⁷	Rehaman and Singh 2012
Maharashtra, India	4.7 x 10 ⁸	Sr ⁸⁷	Rehaman and Singh 2012
Gujarat, India	4.7 x 10 ⁹	Ra ²²⁸	Sartandel et al., 2014; Suman, Bhat and Krishnaswamy 1969

Table 8: Comparison of SGD fluxes to ocean in different coastal hydrological system in and around the study site and across world

Region	Method	SGD rate (m ³ /d)	Reference
Gautami Godavari Estuary	²²⁸ Ra	1.55-7.44 x 10 ⁶	Rengarajan and Sarma 2015
Kakinada Bay	²²⁸ Ra	314.7 x 10 ⁶	Rengarajan and Sarma 2015
Ganga-Brahmaputra Estuary	²²⁶ Ra	6.3-63.0	Moore 1997
Thiruvananthapuram	²²² Rn	1.6 x 10 ⁴	Jacob et a., 2009
Narmada estuary	⁸⁷ Sr	100 - 5500	Rahaman and Singh 2012
Western Bengal Basin, India	Water Balance study	5.9 x 10 ⁷	Mukherjee et al., 2007(a,b)
West Bengal coast, India	⁸⁷ Sr	54 x 10 ⁸	Chakrabarthy et al., 2018
Odisha coast, India	²²⁸ Ra	314.7 x 10 ⁶	Rengarajan and Sarma 2015
Andhra Pradesh coat, India	²²⁸ Ra	314.7 x 10 ⁶	Rengarajan and Sarma 2015
Tamil Nadu coast, India	²²⁸ Ra	9.8 x 10 ⁸	Rengarajan et al., 2002; Barathkumar et al., 2016; Hemalatha, Rajaram and Puranik 2011)
Kerala coast, India	²²² Rn	1.6 x 10 ⁴	Jacob et a., 1993
Karnataka coat, India	²²² Rn	1.75 x 10 ¹¹	Sartandet et al., 2014; Kavitha, Chandrashekara and Paramesh 2017)
Goa coat, India	⁸⁷ Sr	4.75 x 10 ⁸	Rahaman and Singh 2012
Maharashtra coast, India	⁸⁷ Sr	4.75 x 10 ⁸	Rahaman and Singh 2012
Gujarat coast, India	²²⁸ Ra	4.72 x 10 ⁹	Sartandel et al., 2014; Suman et al., 1969)
Bengal Basin (India-Bangladesh)	³ He/ ³ H	1.5 x 10 ⁷	Dowling et al., 2003
South Chain Sea Shelf	²²⁸ Ra	2.2 x 10 ⁸	Liu et al., 2012
Eastern Hainan Island-Barmen Bay	²²⁶ Ra	2.9 x 10 ⁵	Su et al., 2011
Yellow Sea	²²⁶ Ra	6.3 x 10 ⁸	Kim et al.2003
Manila Bay (Philippines)	⁸⁷ Sr	2.36 x 10 ⁶	Taniguchi et al., 2008
Global SGD flux	²²⁸ Ra	1 x 10 ¹¹ 12 X10 ¹³	Burnett et al., 2003; Kwon et al., 2014

Table 9: External N and P sources from various reservoirs to the northern Indian Ocean and their contribution to the total external sources (given in bracket as percentage).

Basin	Nutrient	River (Tg/y)	Atmosphere (Tg/y)	SGD (Tg/y)
Arabian Sea	Nitrogen	0.10 (40%)	0.09 (36%)	0.06 (24%)
	Phosphorus	0.03 (51%)	0.014 (24%)	0.015 (25%)
Bay of Bengal	Nitrogen	0.12 (35%)	0.12 (35%)	0.103 (30%)
	Phosphorus	0.08 (66%)	0.021 (17%)	0.021 (17%)

Figure 1: Map showing groundwater sampled in this study. Black stars indicate that, the locations where SGD flux rate taken from the literature and red circles indicates the locations where SGD flux calculated based on the available data.

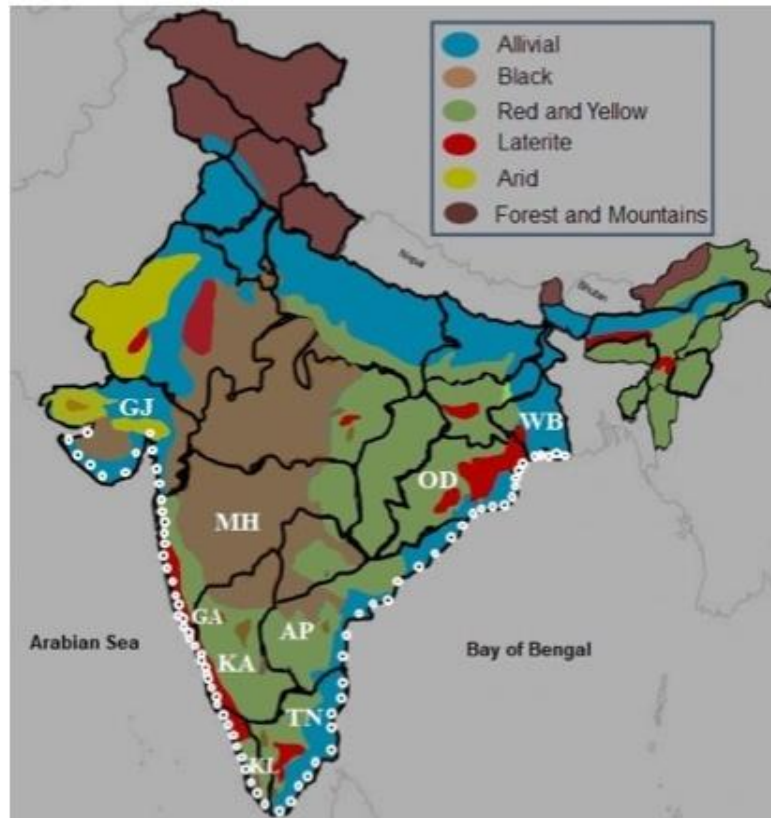


Figure 2 : Variability of DIN in groundwaters along the east and west coast of India during a) wet and b) dry periods.

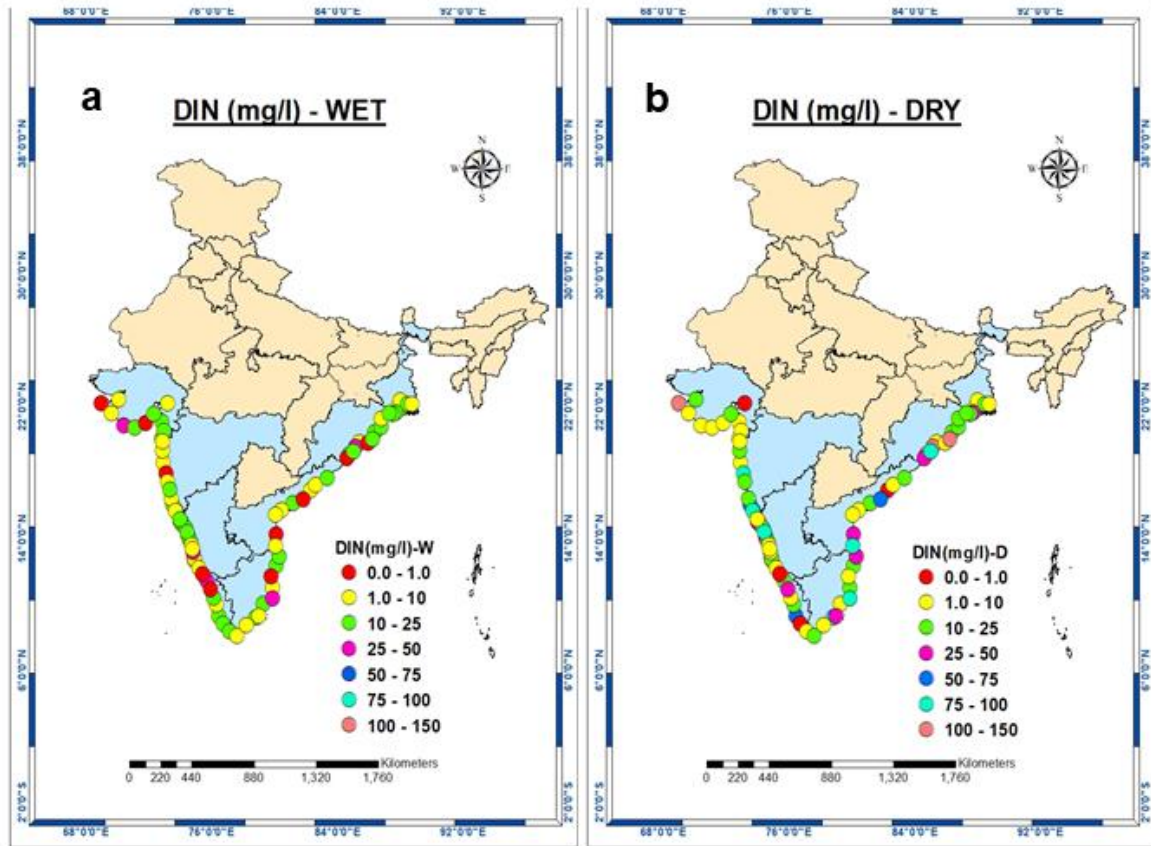


Figure 3: Variability of DIP in ground waters along the east and west coast of India during a) wet and b) dry periods.

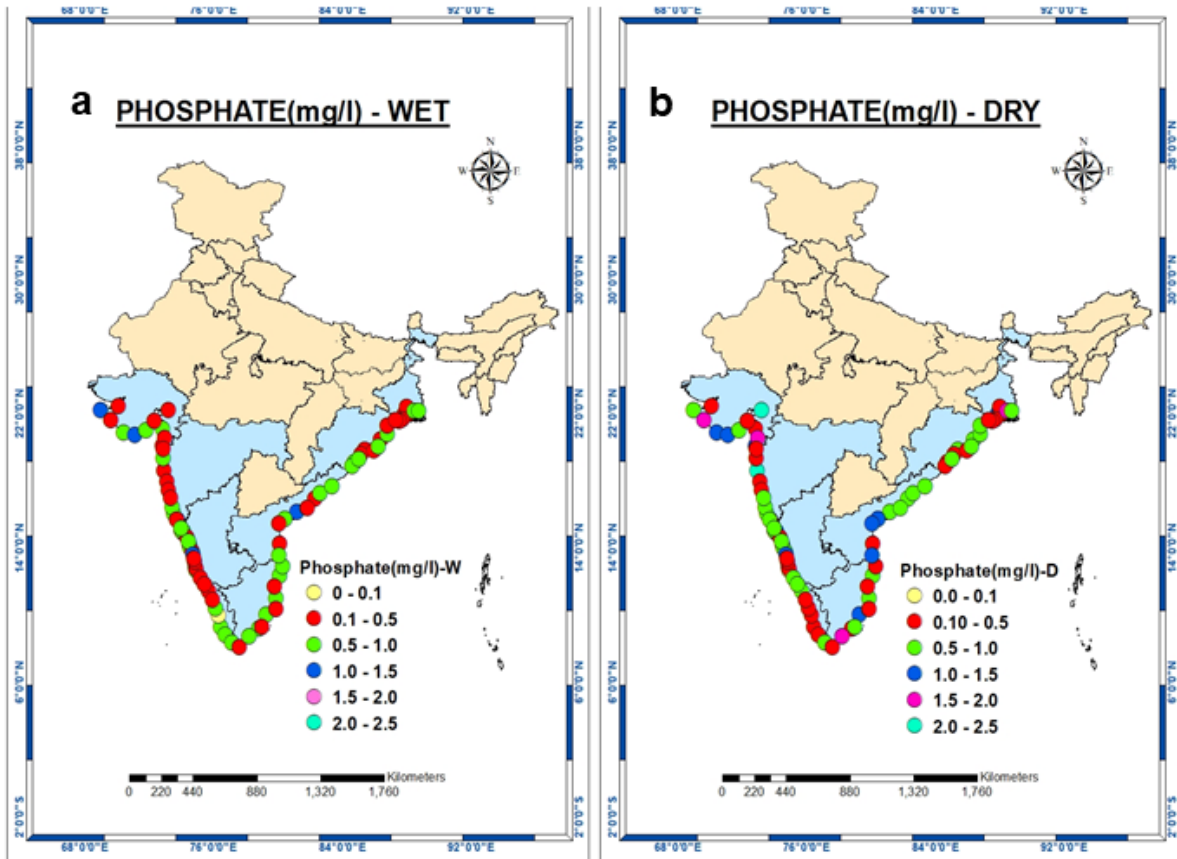


Figure 4 : Variability of urea in groundwaters along the east and west coast of India during a) wet and b) dry periods.

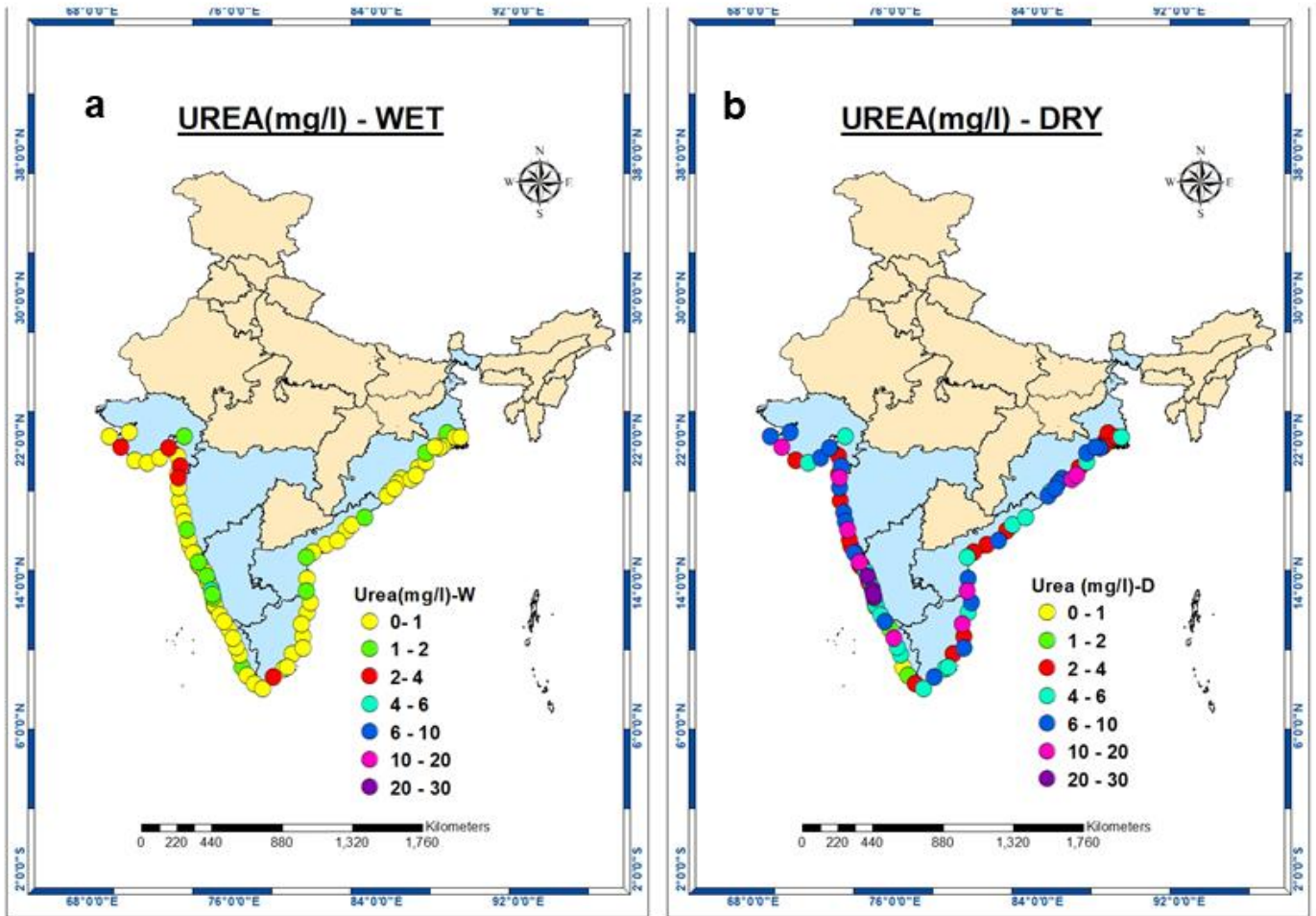


Figure 5: Relationship between nutrients in the ground water with soil and bacterial respiration in the ground water during wet period

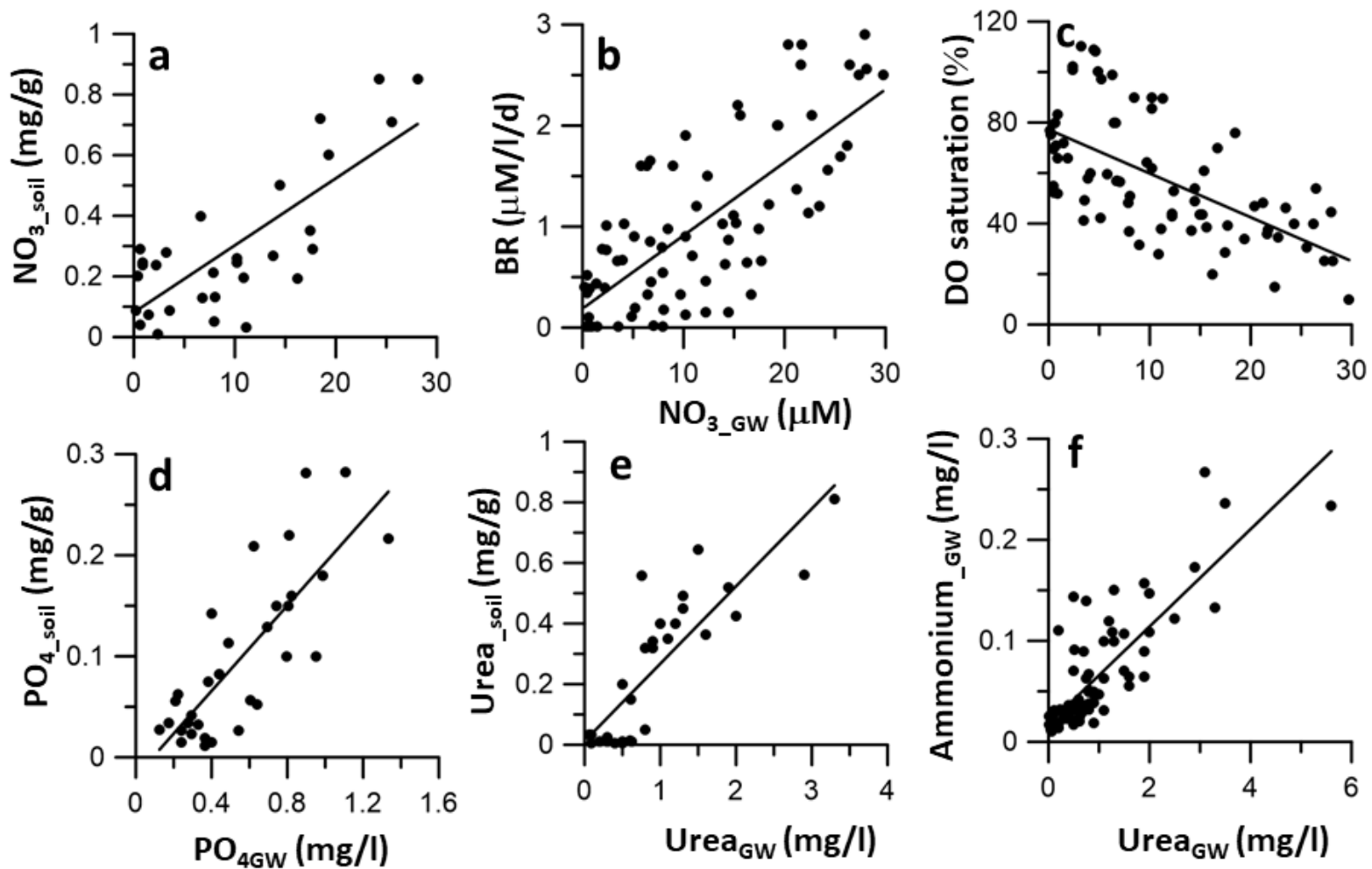


Figure 6: Relationship of nitrate in the ground water with bacterial respiration, ammonium, urea and DO saturation in the ground water during dry period.

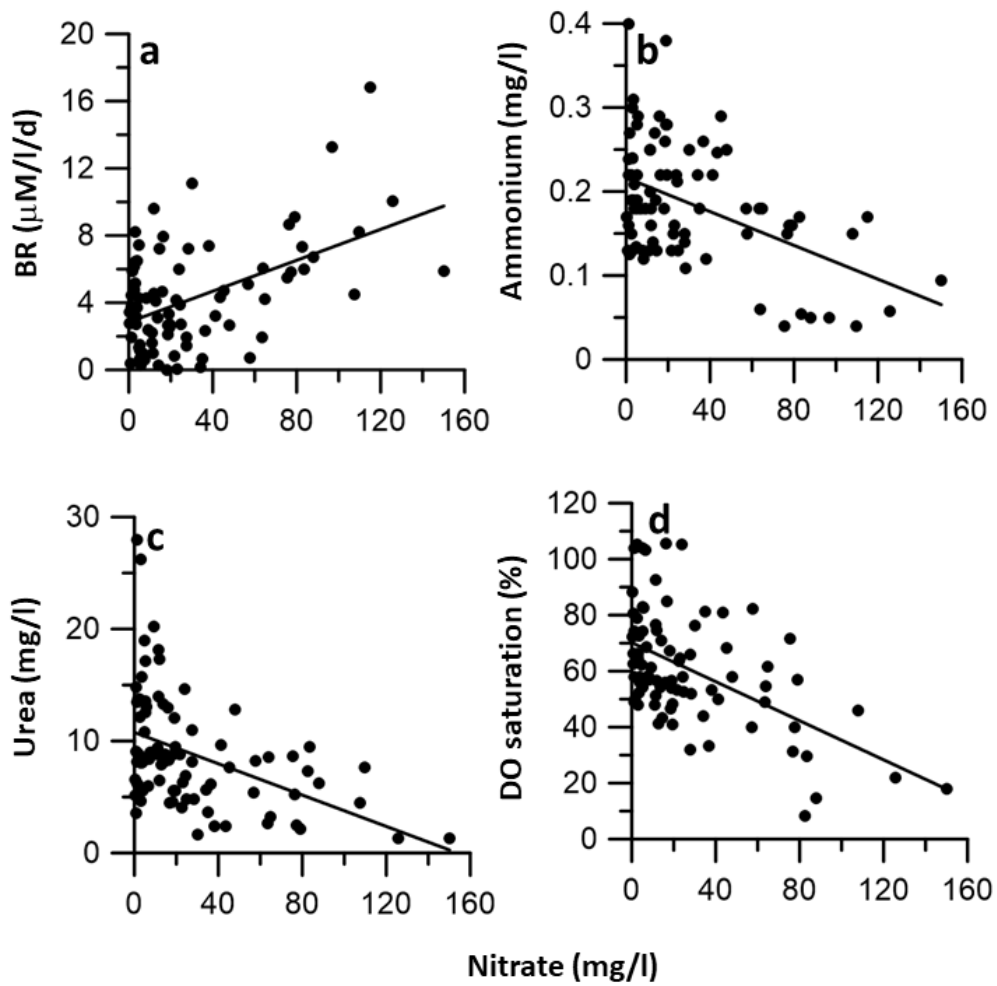


Figure 7: Relationship between State wise fertilizer consumption in Indian coastal states, and mean concentrations DIN in each state during a) Kharif (wet) and b) Rabi (dry) seasons.

