



Phosphate removal through adsorption on suspended matter in the Bay of Bengal: Possible implications to primary production

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MS received 24 July 2020; revised 14 October 2020; accepted 14 October 2020

Low primary production in the Bay of Bengal (BoB) could not be explained as this region receives nutrients from atmospheric deposition, rivers, eddies, and mixing due to depression/cyclones. In addition to nutrients, BoB also receives significant amount of suspended particulate matter (SPM) from the major rivers and their concentrations are in an order of magnitude higher than elsewhere in the open ocean region. Here we show that the removal of phosphorus (P) through adsorption on SPM may be a potential mechanism to decrease in primary production in the northern BoB. Significant fraction of P removal (5–50% of total dissolved inorganic phosphate) in association with SPM was observed in the BoB. The magnitude of removal of P through SPM is linearly related with dissolved inorganic phosphate (DIP), particulate organic carbon (POC) in the water column suggesting that P is removed in association with organic matter. The fraction of P removed from that of DIP showed inverse relation with salinity, and linear relation with SPM concentration suggesting that SPM brought by river discharge removed P from the water column. The P removed by SPM in the mixed layer showed inverse relation with mixed layer integrated primary production in the open sea region but its impact is negligible in the coastal waters. The laboratory experiment by measuring primary production in the presence of different quantities of SPM concentrations confirmed decrease in primary production due to removal of phosphate in the BoB.

Keywords. Phosphate; adsorption; suspended matter; river discharge; Bay of Bengal.

1. Introduction

The Bay of Bengal (BoB) receives significant amount of fresh water from major rivers, such as Ganges, Brahmaputra, Irrawaddy–Salween river systems. The annual supply of riverine nutrients to the BoB amounts to 133×10^9 mol (Sarin *et al.* 1989; Krishna *et al.* 2016). In addition, nutrients brought by cyclonic eddies (CE) (Prasanna Kumar

et al. 2004; Sarma *et al.* 2016, 2019; Sherin *et al.* 2018) and atmospheric dust deposition (Srinivas *et al.* 2011) are reported to support primary production in the BoB. Despite nutrients supply, low primary production in the BoB is attributed to light limitation due to cloud cover (Gomes *et al.* 2000; Madhupratap *et al.* 2003; Prasanna Kumar *et al.* 2010), and weak nutrients supply due to strong stratification caused by river discharge

(Prasanna Kumar *et al.* 2002; Gauns *et al.* 2005; Ramaiah *et al.* 2010). BoB receives ~ 1350 million tons of SPM from the major rivers (Milliman and Meade 1983; Subramanian 1993; Anon 1993) resulting in high turbidity in the northern BoB (Gomes *et al.* 2000; Madhupratap *et al.* 2003). Dissolved inorganic phosphorus is an important nutrient for all living organisms and plays an essential role in regulating the primary production in the marine environments (Froelich 1988). Phosphate removal through adsorption on sediment particles was reported in the riverine and estuarine environments (e.g., Froelich 1988; Borggaard *et al.* 2004; Li *et al.* 2013) and its role has been neglected in the open sea regions due to low SPM load. In this paper, we report that removal of phosphate, in association with SPM, limits primary production due to high SPM load in the BoB. Major objectives of this study are: (a) to quantify the amount of P removal from water column in association with SPM and (b) to evaluate its influence on primary production in the BoB.

2. Material and methods

2.1 Sampling

Two cruises were undertaken in the western and central BoB between 10° and 20°N during 23 March to 6 April, 2018 and 11–29 June, 2019

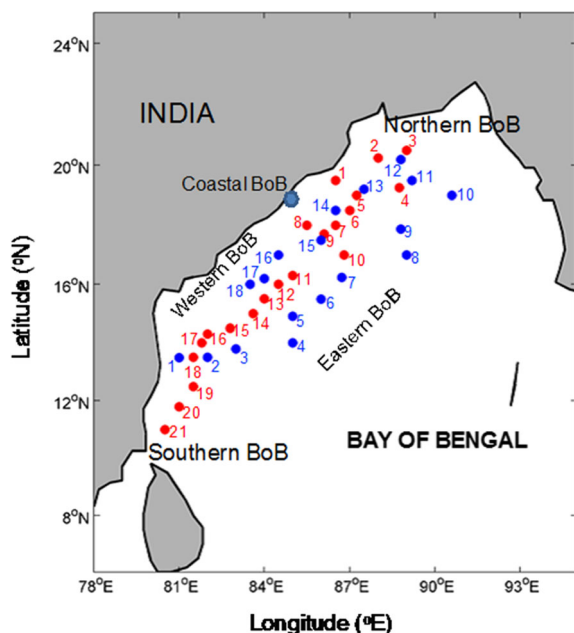


Figure 1. Map showing the station locations in the coastal BoB and offshore regions during May (red) and June (blue). Station numbers are given and regions are identified.

onboard *ORV Sindhu Sankalp* (cruise No. SSK118) and *ORV Sagar Nidhi* (Cruise No. SN142), respectively (figure 1). The monthly time series sampling was conducted in the coastal BoB at fixed location (figure 1) and samples were collected in the upper 30 m of water column, where the depth of water column was 40 m. Water column temperature and salinity (SBE 911 plus, USA), were measured using sensors attached to Rosette (SBE 32, USA). The detection limits for temperature, salinity, and pressure are 0.001°C , 0.001, and 0.1 m, respectively. The sampling was carried out in the upper 200 m of water column at standard depths using 10 L Niskin bottles attached to the CTD Rosette, but here we report only data collected in the upper 25 m where the impact of phosphate removal is significant.

2.2 Analytical techniques

2.2.1 Nutrients, SPM and POC

Water samples from the Niskin bottles were subsampled for nutrients in 100 ml plastic bottles and were filtered through $0.2\ \mu\text{m}$ polycarbonate filter (Millipore) and concentration of nutrients were measured onboard following standard spectrophotometric procedures using an autoanalyzer (Ecolab, USA) (Grasshoff *et al.* 1983). The detection limits for nitrate, nitrite, ammonium (dissolved inorganic nitrogen; DIN) and phosphate (dissolved inorganic phosphate DIP) were 0.02, 0.01, 0.02 and $0.01\ \mu\text{M}$, respectively. Suspended particulate matter (SPM) was determined by dry weight of the material retained on a $0.22\ \mu\text{m}$ polycarbonate filter (Millipore) after filtering ca. 3–5 l of water under a moderate vacuum. About 5 l of sample were filtered through pre-combusted (at 450°C for 6 hrs) GF/F filters and dried at 50°C for 12 hr. These filters were exposed to the concentrated hydrochloric acid for 12 hrs to remove inorganic carbon on the filter. The content of carbon in particulate organic matter (POM) retained on the filter was determined using Elemental analyser (Vario pyrocube; M/s. Elementar Analysensysteme, GmbH, Germany).

2.2.2 Phosphate adsorbed on SPM

The SPM collected on the $0.22\ \mu\text{m}$ polycarbonate filters were cut into four quarters and each quarter is placed in the 10 ml of aged surface seawater

bottles collected in the BoB. The concentration of phosphate (P) in the aged surface seawater is under detection limits ($0.008 \mu\text{M}$). The aged seawater with filter was placed in a sonicator for 30 min and centrifuged for 10 min at 2000 rpm. The concentrations of P in the extracted water, after filtration was passed through $0.7 \mu\text{m}$ grass fibre (GF/F) filter, was measured colorimetrically. The contribution of adsorbed phosphate to the DIP is computed as $(\text{adsorbed P/DIP}) * 100$. In order to examine the impact of sonication on organic matter, the filter stored bottle was shaken for 6 hrs and centrifuged through $0.7 \mu\text{m}$ GF/F and measured colorimetrically. The P released through sonication and shaken methods did not yield significant difference.

2.2.3 Collection and analysis of atmospheric aerosols

Atmospheric aerosols (referred as total suspended particulates (TSP)) were collected for ~ 24 hr on pre-combusted quartz fibre filters (395.5 cm^2 , PALLFLEX) using a High Volume Sampler (Envirotech 430) at a flow rate of $1.2 \text{ m}^3 \text{ min}^{-1}$. An aliquot of the filter containing aerosols was transferred to a container containing 50 mL of Milli-Q water and sonicated for 30 min using an ultrasonicator to extract nutrients. Nutrients in extract were measured using Ion Chromatography (850 Metrohm Professional, Switzerland) and details of the methodology are given in Yadav *et al.* (2016).

2.2.4 P removal experiment

About 100 mg of sediment, from Godavari upper river, middle estuary, lower estuary and coastal BoB, was added to 100 ml of $2 \mu\text{M}$ potassium dihydrogen phosphate solution, prepared in the nutrients free aged seawater, and placed on the shaking water bath for 2 hrs at room temperature ($\sim 26^\circ\text{C}$). The concentration of P in the water before and after addition of sediment was measured. The data are reported as percentage of concentration of phosphate after addition of sediment divided by concentration of phosphate before addition.

2.2.5 Impact of P removal on primary production experiment

In order to examine the impact of P removal on primary production, surface waters from the

coastal BoB was collected in 1 l transparent Nalgene bottles and 100, 200, and 500 mg of the dry sediment from the same location were added and kept on shaking water bath at room temperature for 2 hrs. The water was allowed to settle for 12 hrs and supernatant water was transferred to another Nalgene bottle using 10 mm Teflon tube and 1 ml of $0.2 \text{ mM } ^{13}\text{C}$ -enriched sodium bicarbonate is added and incubated from dawn to dusk for 12 hrs under sunlight along with control sample, in which no sediment is added. After incubation, samples are filtered through pre-combusted GF/F filters and dried at 60°C for overnight. These filters were exposed to the concentrated hydrochloric acid for 12 hrs to remove inorganic carbon on the filter. The content and atomic ratios of carbon on the material retained on the filter were measured using Elemental analyser connected to Isotope Ratio Mass Spectrometer (IRMS; M/s. Elementar Analysensysteme, GmbH, Germany).

2.2.6 Computation of N^*

N^* (proxy for nitrogen fixation) was computed following Gruber and Sarmiento (1997) using the following equation:

$$N^* = (N - 16\text{DIP} + 2.90) \times 0.87,$$

where N and DIP are dissolved inorganic nitrogen and dissolved total inorganic phosphate (in μM), respectively.

3. Results

3.1 Monthly variability in hydrographic properties in the coastal BoB

The monthly variability in hydrographic characteristics of the coastal BoB has been described in detail in Sarma *et al.* (2018), we describe here in brief. Cooler temperature and lower saline waters were observed during winter and contrasting conditions are noticed during summer (figure 2a). Coastal upwelling occurs during March to August due to favourable winds. Significantly lower salinity is noticed during September to December in the upper 30 m of water column (figure 2b). The higher SPM load is noticed between January and July with maximum during April and May associated with high salinity and cooler waters (figure 2c). The concentration of DIN was higher during March to June and the least concentrations

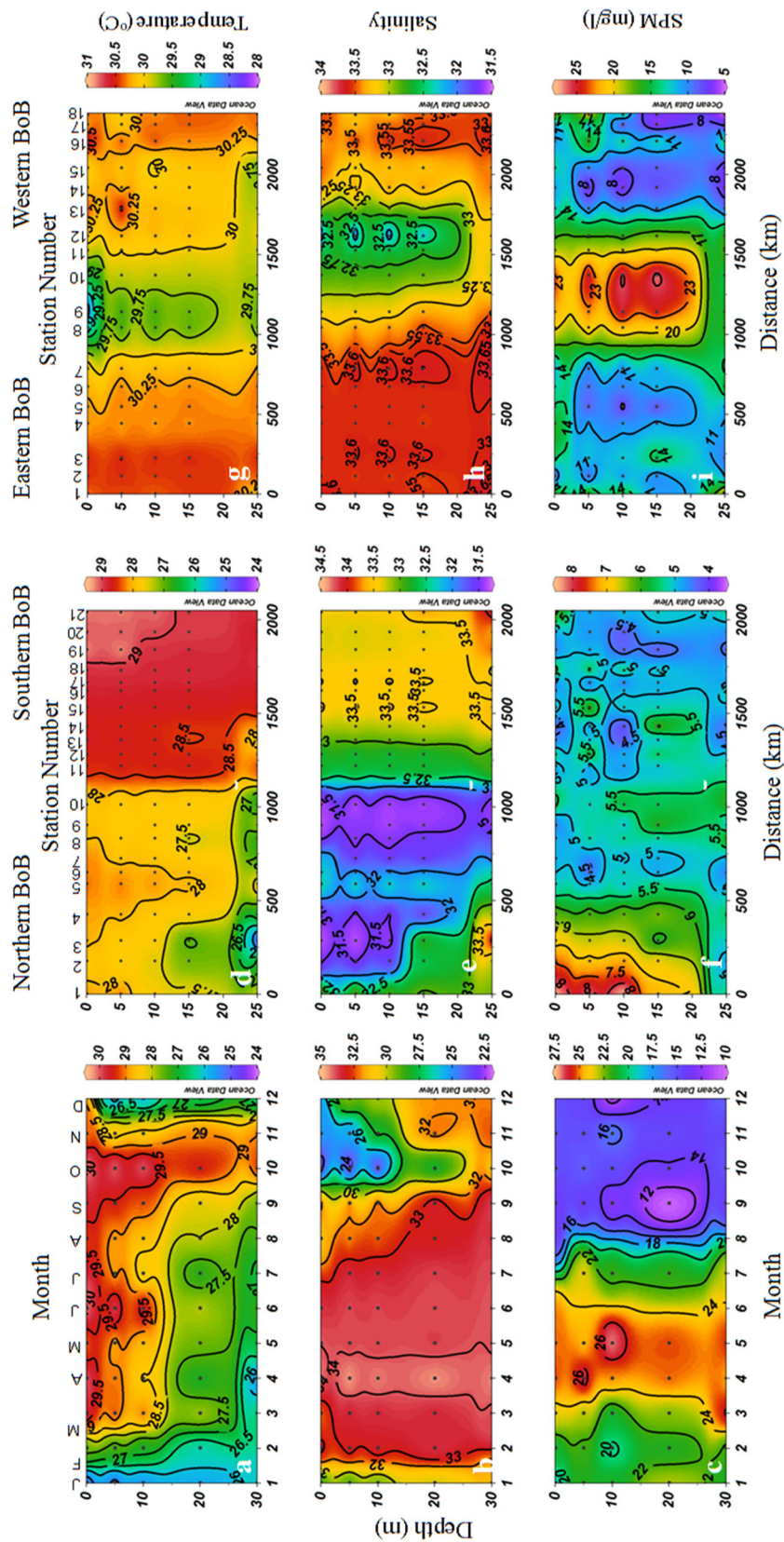


Figure 2. Monthly variability in (a) temperature (°C), (b) salinity, and (c) suspended particulate matter (SPM; mg l⁻¹) in the coastal BoB and spatial variations in (d) temperature (°C), (e) salinity and (f) SPM (mg l⁻¹) during March and (g) temperature (°C), (h) salinity and (i) SPM (mg l⁻¹) during June in the offshore BoB. The station number and regions are also given. The x-axis represents months for coastal region and distance (km) from station 1 in the offshore regions.

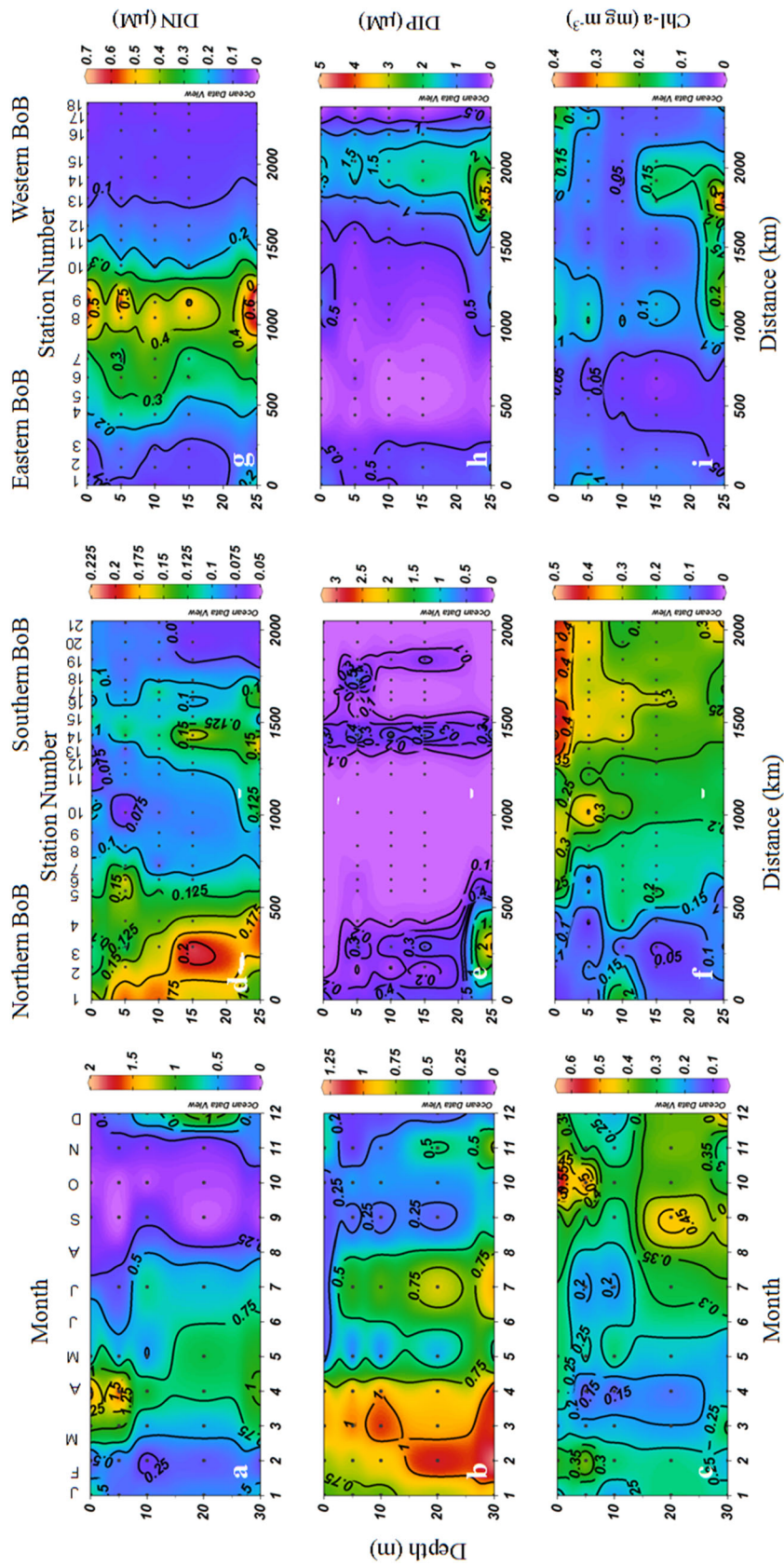


Figure 3. Monthly variability in (a) DIN (μM), (b) DIP (μM), (c) Chl-*a* (mg m^{-3}) in the coastal BoB and (d) DIN (μM), (e) DIP (μM) and (f) Chl-*a* (mg m^{-3}) during March and (g) DIN (μM), (h) DIP (μM) and (i) Chl-*a* (mg m^{-3}) during June in the offshore BoB.

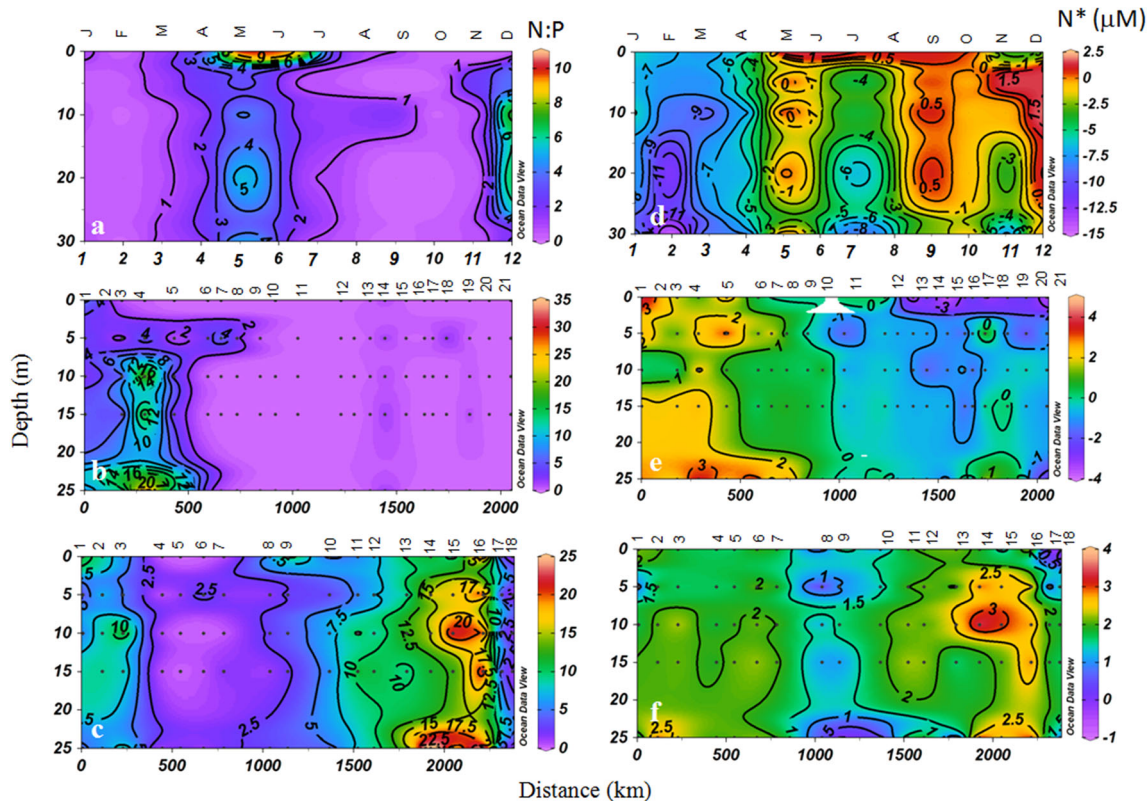


Figure 4. Monthly variability in (a) N:P ratio and (b) N^* values in the coastal BoB and spatial variability in (c) N:P ratio and (d) N^* values (μM) during March and (e) N:P ratios and (f) N^* values (μM) during June in the offshore BoB.

are noticed between September and November (figure 3a). Similarly, the concentrations of DIP were also high between January and August and low concentrations were found between September and December (figure 3b). In contrast, lower concentrations of Chl-*a* are observed during January to August and increase in Chl-*a* is noticed during September to December (figure 3c). The N:P ratios are mostly less than Redfield ratio and relatively higher ratios (1–10) were found during March to August and lower ratios were found during other periods (figure 4a). The higher N^* values are found during September to December and it is consistent with low phosphate concentration (figure 4b).

3.2 Spatial variability in hydrographic properties in offshore BoB

The hydrographic characteristics of the water column during March–April 2018 is described elsewhere (Sarma *et al.* 2019). Relatively cooler waters were observed in the north and warmed towards south during March (figure 2d). In case of June, relatively cooler waters were observed in the north and western Bay compared to eastern Bay during June (figure 2g). The low saline water was

occupied in the northern Bay compared to the south during March (figure 2e) and both salinity and temperature was increased gradually towards south. Similarly, low saline water observed in the northern Bay during June 2019 was associated with cooler water and it was decreased towards south (figure 2h). The eastern Bay was found to contain relatively high saline water than western Bay (figure 2h). The higher SPM concentration was noticed in the northern Bay during the month of March and June associated with low saline water compared to south (figure 2f, i). The concentration of SPM was relatively higher during June 2019 in the northern Bay than during the month of March. The mean concentrations of dissolved inorganic nitrogen (DIN) and DIP in the upper 30 m were low in the northern (0.3 ± 0.1 and 0.08 ± 0.02 μM , respectively) than southern BoB (0.07 ± 0.02 and 0.38 ± 0.1 μM , respectively) during March 2018 (figure 3d, e), whereas higher concentration of DIN was observed in the western Bay than eastern Bay associating with lower salinity during the month of June 2019 (figure 3g, h). The concentration of DIP was significantly lower during the month of June than March with relatively higher concentrations in the western Bay (0.1 ± 0.05 μM) than eastern

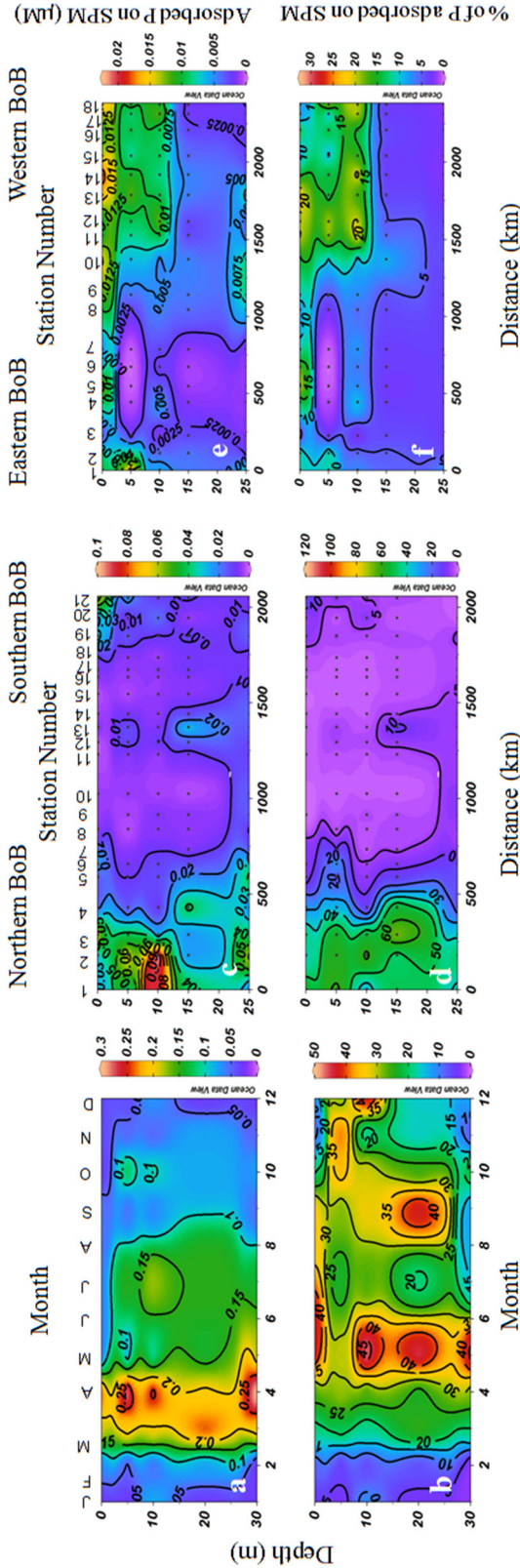


Figure 5. Monthly variability in (a) P adsorbed on SPM (μM) and (b) percentage of contribution of P adsorbed on SPM to DIP in the coastal BoB. Spatial variations in (c) P adsorbed on SPM (μM) and (d) percentage of contribution of P adsorbed on SPM to DIP during March and (e) P adsorbed on SPM (μM) and (f) percentage of contribution of P adsorbed on SPM to DIP during June in the offshore BoB.

Bay ($0.03 \pm 0.01 \mu\text{M}$). Relatively higher Chl-*a* was observed in the northern ($0.16 \pm 0.1 \text{ mg m}^{-3}$) than southern Bay ($0.08 \pm 0.05 \text{ mg m}^{-3}$) during March, whereas the highest Chl-*a* was observed in the northern Bay ($0.45 \pm 0.1 \text{ mg m}^{-3}$) associating with low salinity. The higher Chl-*a* was observed in the eastern BoB associating with high SPM than western Bay. The N/P ratio ranged between 0.8 and 32 in the study region and above the Redfield value of 16 was observed in the upper 30 m of water column in the northern BoB than south during March (<5) (figure 4b). The higher N:P ratio (>15) was observed in the western (18 ± 2) than the eastern Bay (7 ± 3) and low salinity region in the north during the month of June. The N^* values ranged between -4 and $3.2 \mu\text{M}$ in the study region and positive values ($>0 \mu\text{M}$) were observed in the northern than southern BoB. The positive N^* values in the northern BoB were associated with high N:P ratios during March. The N^* values ranged between -1 and $3.8 \mu\text{M}$ with higher values in the western Bay associated with higher N:P ratios during the month of June (figure 4d, f).

3.3 Spatial and seasonal variability in particle associated phosphate in the BoB

The P adsorbed on SPM ranged between 0.02 and $0.38 \mu\text{M}$ in the coastal region with higher concentrations during March to August than September to December in the coastal BoB (figure 5a). The P adsorbed on SPM in offshore region varied between 0.001 and $0.12 \mu\text{M}$ and $0.001\text{--}0.02 \mu\text{M}$ during March and June, respectively (figure 5b, c). The removal of P per gram of SPM is higher in the coastal BoB ($1.5\text{--}24 \mu\text{mol/g}$) with maximum during March and April, followed by March ($0.02\text{--}11 \mu\text{mol/g}$) and June ($0.02\text{--}2.6 \mu\text{mol/g}$) in the offshore region. The contribution of P adsorbed on SPM to the DIP amounted to 5–48% in the coastal BoB (figure 5d) with higher levels during March–June (figure 5e, f). The contribution of removal was higher (10–62%) in the northern BoB during March and 10–25% in the western BoB during June. The concentration of DIP showed significant linear relationship with amounts of P adsorbed on SPM in the BoB with variable slopes in the coastal ($y = 0.2$), March (0.2 and 0.05 in the southern and northern BoB) and June (0.03) (figure 6a) in the offshore region. The concentration of POC showed linear relation with concentration of P adsorbed on

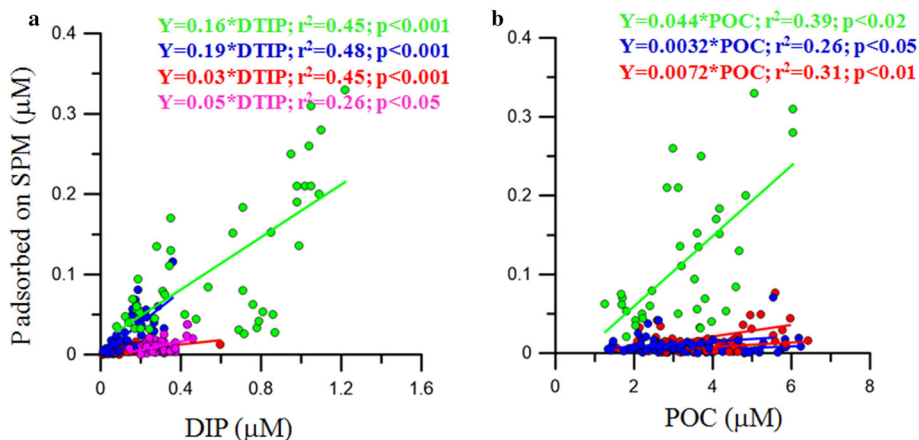


Figure 6. Relationship P adsorbed on SPM (μM) with (a) DIP (μM) and (b) POC (μM) in the coastal BoB (green), during March (red) and June (blue)

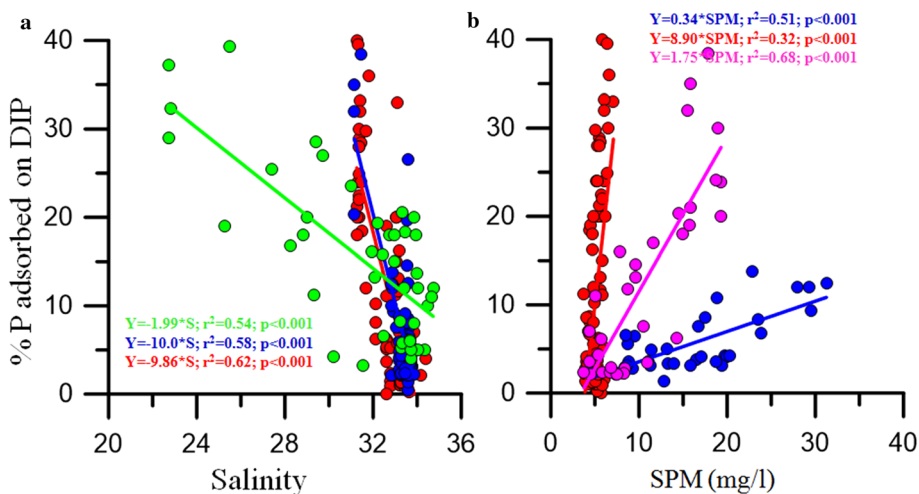


Figure 7. Relationship of percentage of contribution of P adsorbed on SPM (μM) to DIP (μM) with (a) salinity and (b) SPM (mg l^{-1}) in the coastal BoB (green), June (red), northern BoB (red) and southern BoB (purple) during March.

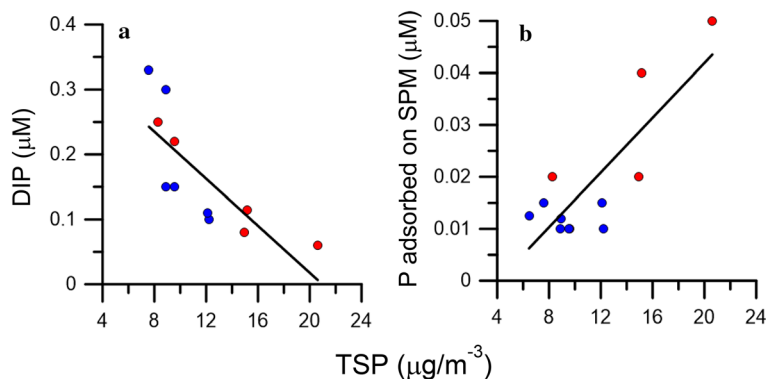


Figure 8. Relationship of TSP ($\mu\text{g m}^{-3}$) with (a) DIP (μM) and (b) P adsorbed on SPM (μM) in the offshore regions during March (red) and June (blue).

SPM in the coastal region (0.033) with lower slopes during June (0.002) than March (0.015) (figure 6b). The salinity in the upper 30 m showed significant inverse relation with contribution of P

adsorbed on SPM to DIP with variable slopes in the coastal (-2.01) than offshore regions (-9.85) (figure 7a). The concentration of SPM showed significant linear relation with contribution of P

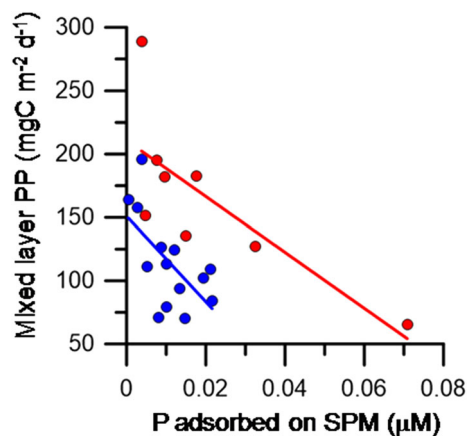


Figure 9. Relationship between P adsorbed on SPM (μM) with mixed layer integrated primary production ($\text{mgC m}^{-3} \text{d}^{-1}$) during March (red) and June (blue).

adsorbed on SPM with higher slope during March (8.9) than June (0.34 and 1.75 in eastern and western BoB), whereas no significant relation was found in the coastal region (figure 7b).

3.4 Atmospheric total suspended particulate matter

The total suspended particulate matter (TSP) in the atmosphere varied from 8.5 to $20.6 \mu\text{g m}^{-3}$ in the study region with relatively higher in the north ($17.2 \pm 2 \mu\text{g m}^{-3}$) than in the south ($8.8 \pm 1 \mu\text{g m}^{-3}$) during March and higher in the west ($18.2 \pm 2 \mu\text{g m}^{-3}$) than eastern Bay ($14.8 \pm 3 \mu\text{g m}^{-3}$) during June (figure not shown). The concentration of inorganic nitrogen in the TSP ranged from 0.08 to $0.28 \mu\text{gN m}^{-3}$ in the study region with lower concentrations during June than that in March. The depositional flux of inorganic nitrogen, computed following Yadav *et al.* (2016), varied from 4.9 to $17.2 \mu\text{gN m}^{-2} \text{d}^{-1}$ and with higher during March than June. The TSP showed inverse relation with DIP in the mixed layer ($r^2 = 0.85$; $p < 0.001$) and linear relation was noticed with adsorbed P on SPM ($r^2 = 0.62$; $p < 0.05$) (figure 8).

3.5 Primary production

3.5.1 Observations

The primary production integrated to the mixed layer varied between 65 and $289 \text{mgC m}^{-2} \text{d}^{-1}$ during March, whereas it ranged from 70 to $196 \text{mgC m}^{-2} \text{d}^{-1}$ during June in the BoB.

Relatively lower primary production was observed in the northern and eastern BoB associated with high SPM and low DIP concentrations. The mixed layer primary production displayed inverse relationship with mixed layer mean P adsorbed on the SPM ($r^2 = 0.42$; $p < 0.05$ and $r^2 = 0.59$; $p < 0.001$, respectively, during March and June) (figure 9).

3.5.2 Experiments

The primary production in the control samples, which contains 20mg/l of SPM, was $68 \text{mgC m}^{-3} \text{d}^{-1}$ and it decreased by 27% ($49.5 \text{mgC m}^{-3} \text{d}^{-1}$) in the sample containing 100mg/l of SPM and further decreased by 53 and 102% by addition of 200 and 500mg/l of SPM. The amount of SPM added to the bottle showed inverse relation with the primary production ($r^2 = 0.89$; $p < 0.001$).

4. Discussion

4.1 Possible mechanisms responsible for anomalous N/P ratios in the BoB

The anomalous N/P ratio was noticed in the northern and western BoB associated with low saline waters and high SPM load. Similarly high N:P ratios were associated with high salinity, cooler water and high SPM load in the coastal BoB. The high SPM in the coastal waters was associated with coastal upwelling during March to June (Sarma *et al.* 2013). Though N:P ratios are not above the Redfield values due to low concentrations of DIN, but they are significantly higher than offshore regions (Sherin *et al.* 2018). Such high N:P can be explained by several processes such as (1) atmospheric nitrogen fixation, (2) deposition of atmospheric nitrogen (both these sources enhance N:P ratio due to excess nitrogen input from atmosphere), and (3) removal of P from the water column in association with mineral particles (this would enhance N:P ratio due to removal of P without modifying N). The occurrence of nitrogen fixation is reported in the oligotrophic regions associated with low DIN concentration (e.g., Fong *et al.* 2008; Löscher *et al.* 2016) that enhances N/P ratio (Bethoux and Copin-Montegant 1986). In order to examine the possible nitrogen fixation in the BoB, the N^* was computed following Gruber and Sarmiento (1997). The concentration of N^* was positive (ranged between 0 and $4 \mu\text{M}$) in the northern BoB during March and western Bay during June and

September to December in the coastal BoB. The positive N^* was associated with low P and high N/P ratios suggesting that higher N^* values might have occurred due to anomalously high N/P ratios (figure 4). Loscher *et al.* (2019) reported no occurrence of nitrogen fixation in the BoB and attributed to possible limitation of trace metals other than iron during winter when phosphate concentrations were below detection limits. Recently, Sarma *et al.* (2020a) noticed weak nitrogen fixation in the Bay of Bengal and their spatial variability is controlled by availability of phosphate. They further noticed that addition of phosphate enhanced nitrogen fixation by 2–7 times. Nevertheless, low phosphate concentrations found in the study region (figure 3) suggests weak nitrogen fixation may be possible and it may not significantly modify N/P ratios. An alternative source of nitrogen would be deposition of atmospheric nitrogen contributing to high N/P ratio (Herut and Krom 1996; Kouvarakis *et al.* 2001). The BoB receives high amount of atmospheric dust through Indo-Gangetic Plain (IGP) resulting in deposition of high concentrations of DIN in the northern BoB compared to the adjacent basin Arabian Sea (Kumar *et al.* 2008; Srinivas *et al.* 2011). The Aerosol Optical Depth (AOD) is the highest over the northern BoB followed by South China Sea than elsewhere in the world and the rate of increase in AOD is the highest over the BoB than elsewhere in the world (Zhang and Reid 2010). Srinivas *et al.* (2011) found that atmospheric deposition of nitrogen supports 13% of the primary production in the BoB, whereas Yadav *et al.* (2016) reported 3–33% of primary production in the coastal BoB. The concentrations of inorganic nitrogen in the atmospheric dust varied between 0.08 and 0.28 $\mu\text{g m}^{-3}$ in the study region and the depositional flux was ranged from 4.8 to 17.1 $\mu\text{g m}^{-2} \text{d}^{-1}$ with insignificant variation between northern ($9.8 \pm 5 \mu\text{gN m}^{-2} \text{d}^{-1}$) and southern BoB ($9.2 \pm 2 \mu\text{gN m}^{-2} \text{d}^{-1}$). The concentration of P in the atmospheric dust was below detection limits during the study periods. Srinivas *et al.* (2011) observed higher inorganic nitrogen ($1.8 \mu\text{g m}^{-3}$) than organic nitrogen ($0.08 \mu\text{g m}^{-3}$) in the atmospheric dust over BoB during March–April in the BoB. Since concentration of dissolved inorganic nitrogen is low in the atmospheric dust and it is unlikely to bring significant modification in the N/P ratios in the BoB.

On the other hand, the isotopic composition of particulate nitrogen gets depleted due to either nitrogen fixation or atmospheric nitrogen deposition (Montoya *et al.* 2002; Stueken *et al.* 2015). The

concentration of particulate nitrogen was higher in the northern BoB with relatively enriched isotopic value of nitrogen in the BoB ($5.8 \pm 0.2\%$; Sarma *et al.* 2019), which is close to deep water value (5.5 to 6.0‰; Sigman *et al.* 1997; Bristow *et al.* 2017; Sherin *et al.* 2018) suggesting that neither nitrogen fixation nor atmospheric nitrogen deposition contribute to anomalous N/P ratios in the study region.

The third possible mechanism could be removal of P from seawater through adsorption onto the mineral particles of riverine origin. The riverine mineral particles are rich in iron hydroxides and clay minerals, which have a high affinity for P in seawater (e.g., Bhattacharya 1970; Sarin *et al.* 1989; Jha *et al.* 1993). It has been found that sedimentation of atmospheric dust may also remove P through adsorption from the water column (Feely *et al.* 1990; Wheat *et al.* 1996; Bjerrum and Canfield 2002). In contrast, several investigators (e.g., Pan *et al.* 2002; Ridame *et al.* 2003; Krom *et al.* 2004) reported that atmospheric dust or riverine mineral particle may act as a source of P to the seawater. In order to examine the role of particle (atmospheric or riverine origin) on adsorption/desorption of P from water is examined.

4.2 Possible loss of P through mineral adsorption

The adsorption of P and its removal to sediment in the freshwater, estuarine and coastal regions have been reported earlier (e.g., House *et al.* 1998; Pant and Reddy 2001; Al-Enezi *et al.* 2016). P can rapidly react with particle surfaces in natural systems (Froelich 1988; Borggaard *et al.* 2004) and sorption and desorption reactions between water and sediment particles regulates P concentrations in the rivers, estuaries and coastal waters (Wauchope and McDowell 1984; Richardson 1985; Price *et al.* 2010). The adsorption and desorption of P on the particle is dependent on the concentration of iron oxides, clay content, organic matter and pH (Spiteri *et al.* 2006; Lee and Kim 2015; Zhang *et al.* 2016). In addition to this, the ratio between the concentration of P in water and that required to attain zero equilibrium with respect to SPM, where both adsorption and desorption is equal (Pan *et al.* 2002), would decide the direction of the sorption process.

Several investigators found that Saharan dust is a significant source of P to the Mediterranean Sea

as the concentration of P on Saharan dust is quite high (112 $\mu\text{mol P/g}$) compared to that in the surface seawater (Pan *et al.* 2002; Ridame *et al.* 2003). In the present case, no measurable concentration of P was observed in the atmospheric dust particles over the BoB, whereas Srinivas and Sarin (2013) reported $\sim 30 \pm 19 \text{ nmol P/m}^3$ over the BoB. Since the concentration of P in the surface waters varied from 0.1 to 0.5 and 0.2 to 1.2 μM , respectively, in the offshore and coastal regions and absent (less than detection limits) in the atmospheric dust particles, indicates possible adsorption of P on to the particles can be considered as the main mechanism for P removal. This is further confirmed by positive relation between TSP and P adsorbed on to the particles.

River-borne SPM may also influence P in water through adsorption–desorption equilibria. Pan *et al.* (2002) reported that whether river-borne SPM acts as a source or sink for P is dependent on the ratio concentration of P between water and P required for zero equilibrium. The BoB received 3.5–8.2 and 7.5–23.5 mg l^{-1} of SPM during March and June in the offshore region, respectively, whereas 11.2–26.5 mg/l in the coastal BoB. The concentration of SPM in the BoB is significantly higher in the Indian Ocean due to discharge of significant amount of suspended load from the Ganges and Brahmaputra rivers (Milliman and Meade 1983; Anon 1993; Subramanian 1993). The magnitude of adsorption on the SPM seems to amount to 0.01–0.12 μM during March, 0.01–0.02 μM during June in the offshore region and 0.02–0.38 μM in the coastal BoB (Supplementary table S1; figure 5a, c, e). Relatively high PO_4 was adsorbed on the SPM in the BoB in association with high concentrations of SPM (figures 2 and 5) in the coastal and northern BoB than south. The magnitude of adsorbed P on SPM accounted for 5–60% of DIP in the northern BoB during March and up to 20% during June in the offshore and varied between 5 and 45% in the coastal BoB. Despite the higher concentration of SPM during June in comparison to March in the coastal BoB than in offshore region, the removal of P per gram of SPM is lower during June (0.02–2.6 $\mu\text{mol/g}$) and March (0.02–11 $\mu\text{mol/g}$) in the offshore region than coastal BoB (1.5–24 $\mu\text{mol/g}$) suggesting that the composition of SPM, mainly clay content, concentration of iron oxides and pH of waters are important than concentration of SPM alone in the removal of P from the water column (e.g., Bhattacharya 1970; Sarin *et al.* 1989; Jha

et al. 1993). pH of the water plays a vital role in enhancing Fe^{2+} oxidation and removal of dissolved P (Davison and Seed 1983; Spiteri *et al.* 2006). The annual mean pH in the coastal waters was low (8.07 ± 0.1) followed by March (8.10 ± 0.04) and June 8.16 ± 0.05 (Sarma *et al.* 2018, 2019; Our unpublished data during June). Hernandez and Burnham (1974) found that P desorption from soil increase with increase in pH. Therefore, lower P per gram of SPM during June in the offshore region may also have been caused by desorption of P at higher pH, in addition to clay content and ion concentrations. Nevertheless, as adsorption seems to be a significant process in time and space in the Bay, the removal of phosphate from the water column seems to be a source of anomalous N/P ratios in the BoB (figure 5).

The adsorption and desorption of P on particle is dependent on the ratio between aqueous P concentration and zero equilibrium P concentration (Pan *et al.* 2002) and the latter is found to vary between 0.02 and 0.5 μM in various environments (e.g., Slomp *et al.* 1998; Zhang *et al.* 2016). Our laboratory experiment conducted using coastal sediment to find the zero equilibrium P concentration revealed that to be $0.06 \pm 0.03 \mu\text{M}$ (Supplementary figure S1). Since the sediment composition in the BoB, is expected to be variable in space and time, the zero equilibrium P concentrations will also be variable. Nevertheless, the concentration of P in the upper 25 m of water column is mostly above the experimental zero equilibrium concentration suggesting that P adsorption on SPM is possible.

The concentration of P adsorbed on the SPM showed linear relation with concentration of DIP in the water column (figure 6a) and also concentration of POC suggesting that availability of more phosphate and organic matter in the water column facilitated removal of P from the water column (figure 6b). However, the slope of the relationship between DIP and P adsorbed displayed significant spatial variability with higher slope in the coastal region (0.16) and northern BoB (Stns. 1–10; slope = 0.19) than south during March (stns. 11–21; slope = 0.05) followed by June (0.03) in the offshore region (figure 6a). This suggests that more P is removed in the coastal and northern BoB, where relatively low pH (8.07–8.11) and high SPM occurred than in June (8.16). In case of relationship between DIP and POC, higher slope is found in the coastal region (0.044) than June (0.007) and March (0.0032) in the offshore regions and this difference

may lie in quality of organic matter as more humic substances contribution is expected in the coastal than offshore region. Zhou *et al.* (2015) found higher removal of phosphate associated with higher concentrations of humic substances in laboratory experiments. The fraction of P removed from DIP in the water column displayed inverse relationship with salinity (figure 7a) with significantly different slopes in the coastal (1.99 ; $r^2 = 0.54$; $p < 0.001$) than offshore (9.86 – 10 ; $p < 0.001$) indicating that SPM brought by the river discharge removed P from the water column and the variable slopes indicate that rate of removal is different between the coastal and offshore regions and is caused by quality of organic matter and pH. The relationship between P removed and concentration of SPM in the water column also displayed variable slopes (northern Bay (0.34), southern Bay (1.75) during March and 8.90 during June; figure 7b) in the offshore region but insignificant relation is found in the coastal region (figure not shown) indicating that the rate of removal is variable in space and time. In addition to this, the concentration of TSP in air showed inverse relation with DIP ($r^2 = 0.85$; $p < 0.001$; figure 8a) but a weaker one with adsorbed P ($r^2 = 0.62$; $p < 0.05$; figure 8b) suggesting the possible removal by TSP as well. The variability in the concentrations of P removal to TSP relationship may also have been caused by pH of the water as it was higher during June than in March. Though we have no direct evidence for P removal by TSP, the lack of P on TSP suggests possible removal from the water column.

Increased P adsorption on soil with increased soil organic carbon was reported and suggested that organic carbon in the soil controls bioavailability of P (Yang *et al.* 2019; and references therein). Our experiment to study P removal from seawater by sediments from different salinity domains of Godavari estuary suggested that the coastal sediments remove lower P (20%) than riverine sediments (64%) (Supplementary figure S2) as the concentration of organic matter is higher in river sediment (7.8 mg/g) than in the coastal sediment (3.1 mg/g). An increase in pH from river (6.95) to coastal region (8.11) leads to desorption of P from the sediment particles (Davison and Seed 1983; Spiteri *et al.* 2006). Though some desorption of P is expected in the region, our study suggests that both deposition of TSP and SPM flux from rivers seem to make adsorption as the dominant process of P removal, the extent of which is governed by salinity, pH and organic matter in the water. The

removal of P from the water column may significantly influence primary production in the BoB.

4.3 Influence of P removal on primary productivity

The mixed layer integrated primary production displayed significant inverse relationship with the concentration of phosphate removed by the SPM indicating that phosphate seems to be limiting primary production to some extent in the offshore BoB (figure 9). Such relationship was absent in the coastal water (figure is not shown) as P was removed significantly in the coastal waters; however, enough concentration of P still available in the water column to support primary production. The slopes of the relationship between P removed on SPM and mixed layer primary production are significantly different due to various factors such as light, magnitude of SPM in the water and other physical conditions on primary production during different periods. Light is reported to control primary production in the BoB during summer monsoon (June–September) due to heavy cloud cover over the BoB (Gomes *et al.* 2000; Madhupratap *et al.* 2003) and contrasting conditions occurs during spring (March–May) in the BoB. In order to further confirm the influence of phosphate removal on primary production, the laboratory experiments were conducted. In this experiment, surface water was incubated to measure the primary production and 100 mg of SPM is added to one bottle and without addition to the other (control). It was noticed that ~ 0.25 μM of phosphate is removed from the water in the SPM added bottle due to adsorption. This led to decrease in surface primary production by ~ 8 $\text{mgC m}^{-3} \text{d}^{-1}$ in the SPM added bottle than control (Supplementary figure S3) suggesting that removal of P in association with SPM decreased primary production. Nevertheless, this study suggested that river-borne mineral particles and atmospheric dust remove P from the water column through adsorption leading to anomalously high N/P ratios resulting in decrease in primary production in the BoB. However, this impact on primary production may be negligible in the coastal waters, where high concentration of phosphate is available, but it is highly significant in the open sea regions, especially in the northern BoB, where low concentrations of phosphate and high concentration of SPM is recorded.

5. Summary and conclusion

The BoB is known to be one of the low productive regions in the world and it is attributed to salinity stratification, weak nutrient inputs, and light limitation. The study carried out in the western and coastal BoB suggested that anomalously high N/P ratios are observed associating with high SPM and atmospheric dust inputs. Though N^* was positive in the northern and western BoB, higher isotopic composition of nitrogen in the particulate organic matter suggests nitrogen fixation may be unlikely. The atmospheric deposition of nitrogen is also low and it may be unlikely to contribute to anomalously high N/P ratio. The P adsorbed on the SPM ranged between 5 and 60% of DIP in the BoB indicating that significant fraction of P may be removed from the water column. The amount of P removed through adsorption on SPM displayed strong relation with SPM and TSP indicating that river-borne and atmosphere-borne particles significantly remove P from the water column. The amount of P removed by SPM also showed significant relation with particulate organic carbon suggesting that it is removed in association with organic matter resulting in high N/P ratios. The mixed layer integrated primary production display significant inverse relationship with amount of P removed on SPM suggesting that significant decrease in primary production in the offshore region of BoB due to adsorption P on SPM. The laboratory experiments using coastal sediments suggested that zero equilibrium concentration of P is much lower than the upper ocean P concentration indicating possible adsorption of P on SPM. Since no detectable P is found on atmospheric dust particles deposited over the BoB, it is possible that these particle might have removed P through adsorption on their surface. Therefore, the low primary production in the northern BoB may potentially be due to the unavailability of phosphate than hitherto hypothesized as light limitation and weak supply of nutrients.

In addition, P data collected from BoB since past 5 decades are represented as bioavailable dissolved inorganic phosphate (BDIP) as samples were never recommended to filter before nutrient analysis (IOC 1994). In the P analysis, acid is used (mixed reagent), hence the P adsorbed on the particles is released and thus, it represents total dissolved inorganic phosphate (TDIP) and therefore BDIP is overestimated as particle adsorbed P may not be bioavailable. Based on such data, several

investigators interpreted that light is the controlling factor for phytoplankton biomass in the BoB (e.g., Prasanna Kumar *et al.* 2010). Recently Sarma *et al.* (2020b) found that light does not inhibit primary production in the BoB. In addition to this, occurrence of *Trichodesmium* spp. was reported during March–April period in the southern BoB (Hedge *et al.* 2008); however, recent study suggests that nitrogen fixation does not occur in the BoB during winter (Loscher *et al.* 2019) and weak fixation during summer (Sarma *et al.* 2020a) in the BoB. Sarma *et al.* (2020a) further noticed that addition of dissolved inorganic phosphate enhanced nitrogen fixation by 2–7 times and attributed to severe phosphate limitation in the BoB. The present study suggests that P removal through SPM may be major controlling factor for inhibiting nitrogen fixation in the BoB and decreased primary production, especially in the northern BoB, where impact of river discharge is more. The estimated P removal was 5–60% of dissolved inorganic phosphate in the BoB during a weak discharge period. It is likely to increase during summer and post-monsoon periods, when river discharge and SPM increase by an order of magnitude.

It was hypothesized that rivers may carry excess fertilizers to the BoB, as countries surrounded by BoB consumes significant amount of fertilizer, leading to coastal eutrophication, change in biodiversity beyond national jurisdiction (www.un.org) and formation of dead zones (Bristow *et al.* 2017). The highest aerosol optical depth was observed over the BoB and dry deposition of nutrients from atmospheric dust is proposed to enhance primary production (Patra *et al.* 2007; Srinivas *et al.* 2011; Yadav *et al.* 2016). Both river and atmospheric dust bring nutrients along with mineral particles and the present study suggests that significant fraction of P is removed from the water column in association with mineral particles resulting in limitation of primary production than hitherto hypothesized as eutrophication. The atmospheric dust is increasing in the northern BoB at the rate never observed elsewhere in the world due to rapid industrialization in the south and south-eastern countries (Zhang and Reid 2010). This may lead to increase in particle concentrations in the upper ocean and removal of P from the water column and severe limitation of P to the primary production in future. Modelling efforts coupling with atmospheric processes are rather scarce and such efforts are required for the BoB to examine the potential changes in foodweb dynamics in future.

Acknowledgements

We would like to thank Director, CSIR-NIO for his support and allotment of ship time onboard *Sindhu Sankalp* (SSK 118) to conduct these experiments. We would like to thank Director, INCOIS for granting the project under O-MASCOT on 'Influence of eddies on oxygen minimum zone and carbon sequestration in the Bay of Bengal'. We would like to thank Ministry of Earth Science for allotting ship time onboard *ORV Sagar Nidhi* (SN142). We would like to thank Ship Cell and ship's staff for their support during cruises. We would like to thank the two anonymous reviewers for their suggestions to improve the presentation of the manuscript. This has NIO contribution number 6608.

Author statement

Mr D N Rao made measurements of PO₄ extraction from the suspended matter during March 2018 and analysed the nutrients concentration. He also analysed coastal nutrient samples for the entire year. Mr V R D Ghosh analysed nutrients data during June 2019 and conducted experiment on phosphate adsorption on sediments from different locations, Mr P Sam has analysed PO₄ extraction from suspended matter during June 2019 and conducted primary production experiment with and without suspended matter, Ms K Yadav collected atmospheric dust samples in the coastal and offshore region and analysed the elemental composition, and Dr V V S S Sarma conceptualized the idea, receiving funding, preparation of first draft of the manuscript. All authors contributed in finalizing the manuscript.

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Corresponding editor: JOYDIP MUKHOPADHYAY