RESEARCH ARTICLE

International Journal of Civil, Environmental and Agricultural Engineering

DOI: 10.34256/ijceae2022

Estimating groundwater inputs from Sankarabarani River Basin, South India to the Bay of Bengal evaluated by Radium (^{226}Ra) and nutrient fluxes

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Received : 24th May 2020, Accepted : 02nd September 2020

Abstract: Sankarabarani river basin gains significance due to presence of major industrial, agricultural, urban development and tourist related activities has influenced the water quality in the estuarine environment. Investigations about river water quality has been attempted but not more studies focus about the evaluation of groundwater discharge a significant process that connects groundwater and the coastal seawater have been attempted. For the present study, radium (^{226}Ra) a naturally occurring isotope was measured at three locations and used as effective tracers for estimating the groundwater discharge along with nutrient inputs to the Bay. Groundwater samples representing north east monsoon (December, 2017) has been collected during tidal variation in three locations (Location A- away from the coast towards inland, Location B-intermediate between Location A and the coast and Location C-at the estuary). ^{226}Ra mass balance calculated groundwater fluxes irrespective of tidal variations were 2.27×10^{8} m^3/d, 2.19×10^{8} m^3/d and $5.22\times10^{7} m^3/d$ for A, B and C locations respectively. The nutrients like Dissolved inorganic nitrogen (DIN), Dissolved inorganic Phosphate (DIP) and Dissolved Silica (DSi) were found to be influencing the coastal groundwater by contributing fluxes to the sea of about 679.33 T mol/day. The study suggests increasing radium and nutrient fluxes to the Bay altering the coastal ecosystems would result in surplus algal blooms creating hypoxia.

Keywords: Groundwater discharge, radium isotope, nutrients, algal blooms, hypoxia

1. Introduction

Groundwater discharge is an omnipresent coastal process that incorporates flow of fresh groundwater from coastal aquifers prejudiced by inland hydraulic head to the sea, mixing between the fresh and seawater and infiltration of sea water through the sediment pores near the shoreline predisposed by waves, tidal fluctuations, storms and sea bed variations [1-5]. The total flux of groundwater discharge to the bay is found to be influenced by components as discussed. The estimated total groundwater flow to the oceans are non-superior than 6% of the total world surface runoff, but the nutrients, metals, carbon and radium fluxes happening via groundwater is greater than 50% in comparison with river contributed discharge [6-11]. Higher inputs of metals and nutrients by groundwater discharge greatly influences the coastal marine ecosystems like eutrophication, algal blooms, elevated bacteria and gulf hypoxia [5, 12-16].

Methodologies available for measuring groundwater fluxes and associated nutrient exchanges to the bay like application of Darcy's law [17-20] seepage meters, thermal infrared imagery, flow and density dependent modeling and application of geochemical tracers [21-30].

Application of chemical and radionuclide tracers has been used as effective geochemical tracers to quantify groundwater discharge [31]. Radium isotopes are used effectively to quantify groundwater fluxes along with source appropriation due to the fact that radium gets attached to the sediment particles when in connection with fresh water and gets desorbed when comes in contact with saline water [1,21,31]. The four naturally occurring radioactive isotopes, ^{226}Ra ($t_{1/2}$=1602 y), ^{228}Ra ($t_{1/2}$=5.75 y), ^{223}Ra ($t_{1/2}$=11.4 d) and ^{224}Ra ($t_{1/2}$=3.66 d) through U-Th disintegration suggest radium isotopes significant in measuring the fresh and saline water mixing [31]. The rapid existed isotopes (^{223}Ra and ^{224}Ra) due to their decay from thorium gets regenerated and gets attached to the sediment particles, whereas the long-lived isotopes (^{226}Ra and ^{228}Ra) requires additional time to regenerate. This isotopic renaissance makes Ra isotopes to get adsorbed onto freshwater and gets more soluble in saline water environments f ion exchange resulting with cations in sea water [19, 32-33]. Hence, during sea water intrusion, the sediment gets enriched with rapid living Ra isotopes and depleted in extensive survived radium isotopes due to their variations in their ingrowth, cationic solubility and displacement, increasing mineral charge, presence and stability of organic and inorganic complexes [34, 35]. The presence or absence of radium in water environment is controlled by pH of source water, temperature, redox potential, sediment grain size and porosity and the fractionisation of exchangeable radium [36, 37]. Of all the isotopes ^{226}Rn have been shown to be powerful tool to quantify groundwater discharge fluxes due to its long half-life and increasing predisposition during sea water intrusion and adsorption to particles in low saline water [31]. The residence time of water in a bay is considered for calculating the fluxes of excess ^{226}Ra supported groundwater discharge by dividing the radium flux with the estimated 226Ra activity in groundwater [31]

The river Sankarabarani a fan medium river basin drains Bay of Bengal along the southern side of Pondicherry, India. Due to increase in population, anthropogenic related activities (major industries like paper, alcoholic beverages, chemical, pharmaceutical, agricultural and tourist activities) has altered the material instabilities in the river and adjoining regions due to direct discharge of nutrient, metal and toxic elemental fluxes resulting in ecological imbalance [37]. Previous studies [17, 38] has attempted for groundwater fluxes using radon mass balance approach and radon progeny calculations. But attempts relating radium and nutrient fluxes are gloomy. Hence attempt has been made in the present study to determine the magnitude of groundwater discharge in the estuarine system of sankarabarani river using Ra isotopes and nutrients aiming to calculate the relation between groundwater discharge and nutrient inputs.

2. Methods

2.1. Study area

Sankarabarani river initiates at Gingee Hills, Villupuram district of Tamilnadu, India with tributaries annamangalam, nariyar odai, thondiar, pambaiyar and guduviyar joining the main river course and configures Bay of Bengal on the east coast of India at paradise beach a main tourist destination in Pondicherry after traversing a total of 78.6 Km (Figure 1). The climatic condition is humid to tropical with maximum temperature of about 37°C observed during May to June and minimum (27°C) detected between December to January. The study area experiences higher evaporation rates, with little or no river input during summer with higher salinities. The maximum wind velocity observed is about 19 km hr-^{1}. Average annual precipitation averages 1383 mm per year (CGWB 2007) of which 62% is received during northeast monsoon between October to December. The unconsolidated Quaternary alluvial deposits of sands, clays, silts, gravels and kankar represents the shallow potential aquifer with an average thickness of about 34 m thick with hydraulic conductivity of about 5.0 to 10.0 m/day.

The river forms estuary at the configuring point with a mean depth of 5.0 m with semidiurnal tides ranging in heights from 0.32 to 1.31 m generating substantial inflow of saline water into the river [17]. The sediment transport due to the impact of wave, tide, river or wind energy is estimated as 237,000 m^3 yr^{-1} towards north [39]. The annual average fresh water flow is about 0.02 m^3 s^{-1} mainly renowned during rainy seasons. The water column in the river basin is slightly stratified during monsoon due to inflow of terrestrial water flow and during summer the water is vertically mixed due to mixing of saline water. The land use practiced around the study area is generally agriculture, urban and industrial activities. The discharge of agricultural, industrial and septic sewages has contaminated the shallow quaternary aquifers resulting in groundwater inputs with excess ions, heavy metals and nutrients that may influence the estuaries leading to coastal eutrophication and gulf hypoxia. Thus, measuring groundwater inputs using tracers are of primal significance.

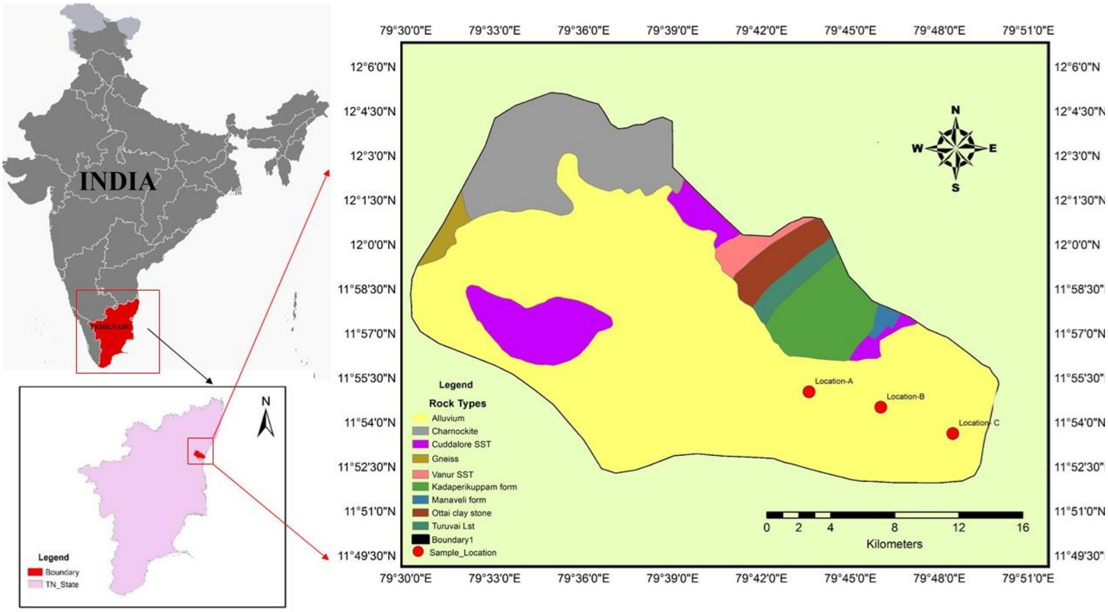

Figure 1. Location of the study area with geology and sampling points

2.2. Water sampling and analysis

Groundwater samples were collected at three locations (A, B, and C) along the shore perpendicular transects representing the fresh, intertidal and saline portions of the aquifers that extends up to 10 Km onshore with reference to tidal variations i.e. during both high and low tides. Water samples were collected from remote bore wells (<30 m depth) tapping alluvial aquifers and purged three well volumes before collection and filtered through 0.45 µm filter and taken in acid rinsed polyethylene containers and immediately shifted to hydrogeological laboratory, Department of Earth Sciences, Pondicherry University and stored at -4°C for nutrient analysis. Physical parameters like total dissolved solids, conductivity, Eh, salinity and DO were measured in the field by using calibrated handheld multiprobe HI9828 (HANNA). Samples for radium analysis were stored in 20 L containers.

The radium 226 in water sample were detected using pre-concentration by co-precipitation technique [40]. Groundwater samples collected in 20 litres pre-acid cleaned containers were transferred to container with 5.0g of analytical grade MnO_2 powder, stirred at a constant rate and then allowed to settle. From the mixture about 5.0 litres of sample were collected and heated in fume wood until clear solution and then treated with 50.0 ml concentrated HCl. To make it exchange to nitric acid medium the solution was evaporated and then treated with 30.0 ml of Conc.HNO_3 and once again heated so as to remove any organic presence in water samples and then treated with 4N HNO_3 to make it up to 70.0 ml. The pre concentrated 70.0 ml sample were then transferred to radon bubbler by vacuum transfer technique and stored for a period of 21 days for suitable in growth time and or equilibrium between ^{226}Ra and ^{222}Rn is established. Water samples with dissolved radon has been transferred to pre-evacuated and background counted ZnS (Ag) scintillation counter and stowed for 180 minutes for attaining equilibrium and fixed to photomultiplier and programmable alpha counting system and counted with emanometry method [41]. The Ra estimation errors were within ±10%. The 226 Ra activity in the samples were calculated using equation recommended by [42-43] as below:

$$226\mathrm{Ra}\ (\mathrm{mBqL}-1) = \frac{6.97 \times 10^{-2} \times (S_{c+}SD)}{V_W X E_c X[1-(\exp - \lambda_c t)] \times [\exp - \lambda_c T] \times [1-(\exp - \lambda_c \theta)}$$

Where, S_c infers sample count – background count, SD represents standard deviation, V_w is the volume of water in L, E_c represents the scintillation cell efficiency, λ being the decay constant for radon as $2.098\times10^{-6}\,s^{-1}$, T represents the counting delay subsequent to sampling, t signifies the counting duration(s) and θ being the time required for radium build-up in the bubbler(s).

3. Result and discussions

3.1. Physicochemical parameters

The physicochemical parameters salinity, pH, DO, and Eh are represented in the Table 1. Influence of sea water is well felt in samples collected in location B and C. Dissolved oxygen is found to be decreasing with decreasing distance towards the coast suggesting lower DO in saline water and consumption in location B indicating varying intensity of geochemical reactions. Salinity is also found to be increasing with distance towards the coast suggesting the influence of sea water infiltration in location C and intermediate salinity noted in location B suggests the intermixing of fresh groundwater with saline water demarcated as the subterranean estuary. Decreasing redox condition noted in location B and C suggests the anoxic water condition and higher redox noted in groundwater samples collected in location A signifies the oxic water condition. Both EC and TDS were found to be increasing towards the coast suggesting the influence of saline water and fresh water mixing in location B and C.

3.2. Radium (^{226}Ra) estimation

The ^{226}Ra activity in the study area samples has been summarised in Table 2 and also represented pictorially in figure 2. The Radium activity of the groundwater from location-A is 0.09 ± 0.01Bq/l during high tide (HT) and 0.63 ± 0.04 Bq/l during low tide (LT). In Location-B Ra during HT and LT were 0.75 ± 0.09Bq/l and 0.77 ± 0.03Bq/l respectively. In location C, the radium activity during HT ranges 1.32 ± 0.16Bq/l and 1.082 ± 0.16Bq/l during LT.

The increasing order of Ra in water samples follow the order Location- C > Location- B > Location-A irrespective of tidal variations. Radium decreased with distance offshore indicating Ra enrichment at the shorelines.

In location-A radium activity is low during high tide due to tidal influence and high during low tide period due to fresh groundwater discharge. During high tides Ra gets absorbs to sediments/aquifer materials and during sea water pumping during low tide it gets dissolved significantly due to variation in the adsorption coefficient of radium between low and high ionic strength water. In location-B radium activity is almost constant in both tides due to fresh groundwater mixing with seawater but found to be higher than location-A suggesting regenerated from aquifer sediments to the overlying water column along with influence of recirculated sea water. In location-C radium activity is very high due to saline influence by tide and also due to discharge influenced by recirculated seawater.

Table 1. Physicochemical parameters for the groundwater samples in three diverse locations and for two tidal events from the study area

Location	Tide	Eh (mV)	TDS (ppm)	EC (µs/cm)	DO (ppm)	Salinity (ppm)	DIN (µmol/l)	DIP (µmol/l)	DSI (µmol/l)
A	High	-45	720	1260	5.5	710	217.405	24.11	476.53
	Low	-39	680	1200	4.2	680	201.43	23,69	476.53
B	High	-72	1450	2580	4.8	1460	182.73	27.37	445.21
	Low	-64	1430	2540	4.5	1450	166.11	23.37	474.45
C	High	-97	5850	10200	2.8	5850	235.63	25.27	444.80
	Low	-89	5780	10100	1.8	5760	126.28	27.37	440.22

Table 2. 226Ra in locations-A, B and C and its variations with tides (values in Bq L-1)

Locations	Sample Type	High Tide	Low Tide
		^{226}Ra (Bq/l)	^{226}Ra (Bq/l)
A	Groundwater	0.0900	0.6297
B	Groundwater	0.7530	0.7763
C	Groundwater	1.3194	1.0828

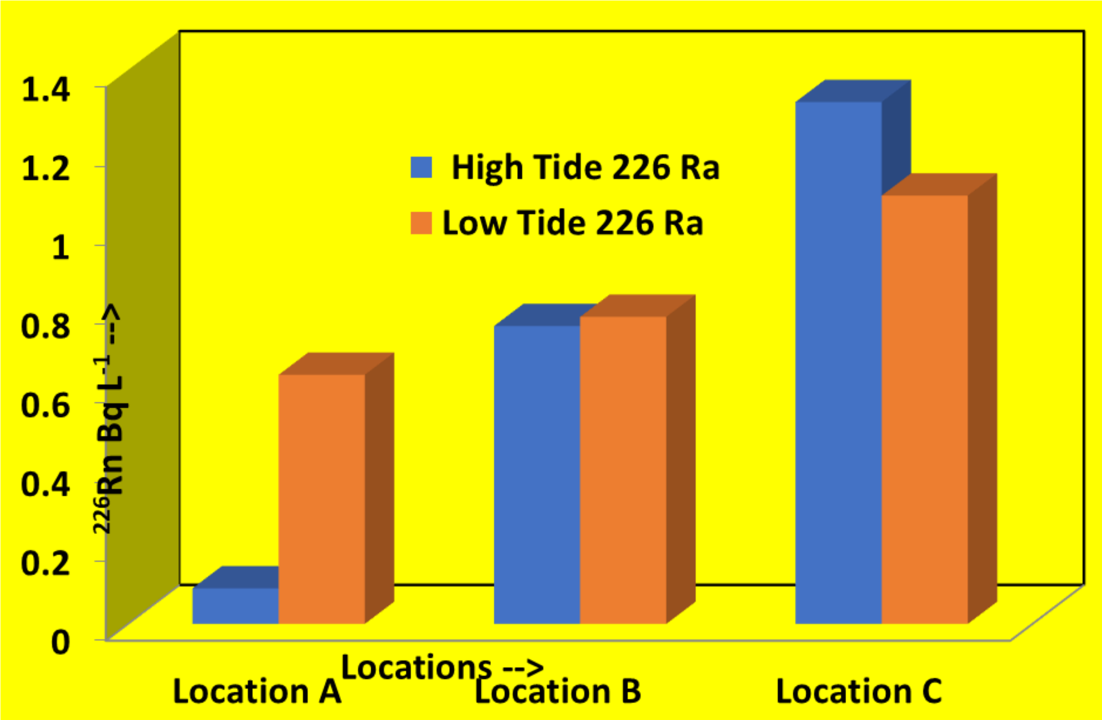

Figure 2. ^{226}Ra variations in locations-A, B and C and its variations with tides (values in Bq L^{-1})

3.3. ^{226}Ra mass balance model

The ^{226}Ra mass-balance model has been attempted to estimate the groundwater discharge related fluxes [31,44]. For the present study area, the likely sources for radium is chiefly due to groundwater discharge and hence the activities of ^{226}Ra above the baseline supported by Bay of Bengal should represent the contribution from groundwater discharge.

$$GD\ (m3\ days-1)\ = \frac{Excess^{226}\ Ra\ (dpm\ m^{-3}).vol\ (m^3)}{\tau(days).gw^{226}\ Ra\ (dpm\ m^{-3})}$$

Here, the excess ^{226}Ra is contributed by seawater, the vol represents the study area volume, τ being the water residence time and gw^{226}Ra is the radium activity in groundwater end member. By considering the conservative behaviour, the calculation of excess ^{226}Ra is as follows:

$$Excess\ 226Ra\ =\ 226_{Ra}Raobs\ x\ \frac{S_{obs}}{S_{sos}}$$

Here, $^{226}Ra_{obs}$ and S_{obs} represents the measured ^{226}Ra activity and groundwater salinity and $^{226}Ra_{sos}$ and S_{sos} signifies the known ^{226}Ra activity and salinity in sea water which is considered for the present study as $^{226}Ra_{SOS}$ = 20 dpm 100 L^{-1} and S_{SOS} = 35.0. The groundwater fluxes calculated by using radium mass balance equation mentioned above shows decreasing trend from the inland area to estuary (Table 3).

In location-A and B, groundwater fluxes noted were 2.27 x $10^8 m^3$/d and 2.19 x $10^8 m^3$/d respectively with minor variations with reference to tides. In location-C, groundwater discharge flux shows a decreasing trend with a flux of 5.1x$10^7 m^3$/d during high tide and 5.2x$10^7 m^3$/d during low tide. Higher groundwater discharge noted in location-A away from the coast might be due to the greater fresh groundwater discharge. Decreasing groundwater fluxes observed in location-B in comparison with location-A signifies the influence of fresh groundwater mixing with seawater intruded into the sediments demarcated as subterranean estuary zone. In location-C still lower groundwater fluxes noted in comparison with location-B and A might be due to the coastal proximity and influence of tides. Higher groundwater fluxes noted at the proximity of the coast signifies the groundwater fluxes influenced by recirculated sea water.

Table 3. Calculated Groundwater fluxes attempted using ^{226}Ra mass balance

Locations	High tide (m^3/d)	Low Tide (m^3/d)
Location-A	2.27 x 10^8	2.27 x 10^8
Location-B	2.19 x 10^8	2.19 x 10^8
Location-C	5.1x10^7	5.2x10^7

3.4. Nutrient flux and budgeting

The nutrients disposed to the sea from the coastal aquifers vary with reference to chemical constituents and quantum of nutrients. It is already known fact about the role of nutrients in altering the biogeochemistry of coastal environment along with primary production. Nutrients to the groundwater environment are mainly from waste disposal, fertilizer application along with deposition from atmosphere [45]. Attempt has been made to quantify the groundwater derived nutrient fluxes. Due to increasing human density fertilizers like urea, sulphate, nitrate and chlorides of ammonium, di-ammonium phosphate (DAP) and N: P: K are applied to the crops for better harvesting [46]. About 7.0 kg/m^3 of fertilizers are used for cultivation purposes in the study area resulting in dissolution and leaching to the aquifers. In view of groundwater flow from aquifers to the coast results in continuous discharge of nutrients to the coast resulting in algal blooms [25,47] resulting in biogeochemical alterations along the coastal regions.

The nutrients DIN (sum of nitrate, nitrite and ammonia), DIP (Dissolved inorganic phosphate) and Dsi (Dissolved Silica) concentrations in water samples are represented in Figure 3. Nutrients in location-A, during high tide follow the order, DSi (476.5µmol/l) > DIN (217.4 µmol/l) > DIP (24.1µmol/l) and during low tide follow the order, DSi (476.5µmol/l) > DIN

(201.4 µmol/l) > DIP (23.6µmol/l). The source of nutrients for location-A is mainly due the fertilizers like (N: P: K and di-ammonium phosphate (DAP) applied to the crops and weathering of silicate minerals at the catchments. In location-B, the dominance of nutrient irrespective of tides follow the order, DSi (475.2 & 474.5µmol/l)> DIN (182.7 & 166.1 µmol/l)> DIP (27.3 & 23.16µmol/l) respectively. Lower nutrients were observed in comparison with location-A might be due to the mixing between fresh groundwater and sea water resulting in reduced aquifer conditions [16]. In location-C, the nutrients follow the order, DSi (444.8 & 440.2µmol/l)> DIN (235.6 & 126.2 µmol/l)> DIP (25.2 & 27.3µmol/l). Silica in groundwater is the most abundant nutrient in the coastal aquifers and found to be influenced by natural processes like rock weathering along with biogenic silica dissolution from sediments [45]. Higher DIN observed in location-A signifies sources from intensive agricultural practices and application of fertilizers [16] and lowering of DIN in location-B and C might be due to suboxic aquifer condition and or nitrate removal through denitrification process. Higher Phosphate observed in location-A might be due greater population density and shorter residence time due to greater hydraulic gradient and its removal in location-B and C might be due to remineralisation and adsorption to the iron oxides with increasing salinisation [48]. In general, the mixing of fresh groundwater with saline water resulted in increasing salinity and decrease of nutrient concentration with distance towards the coast

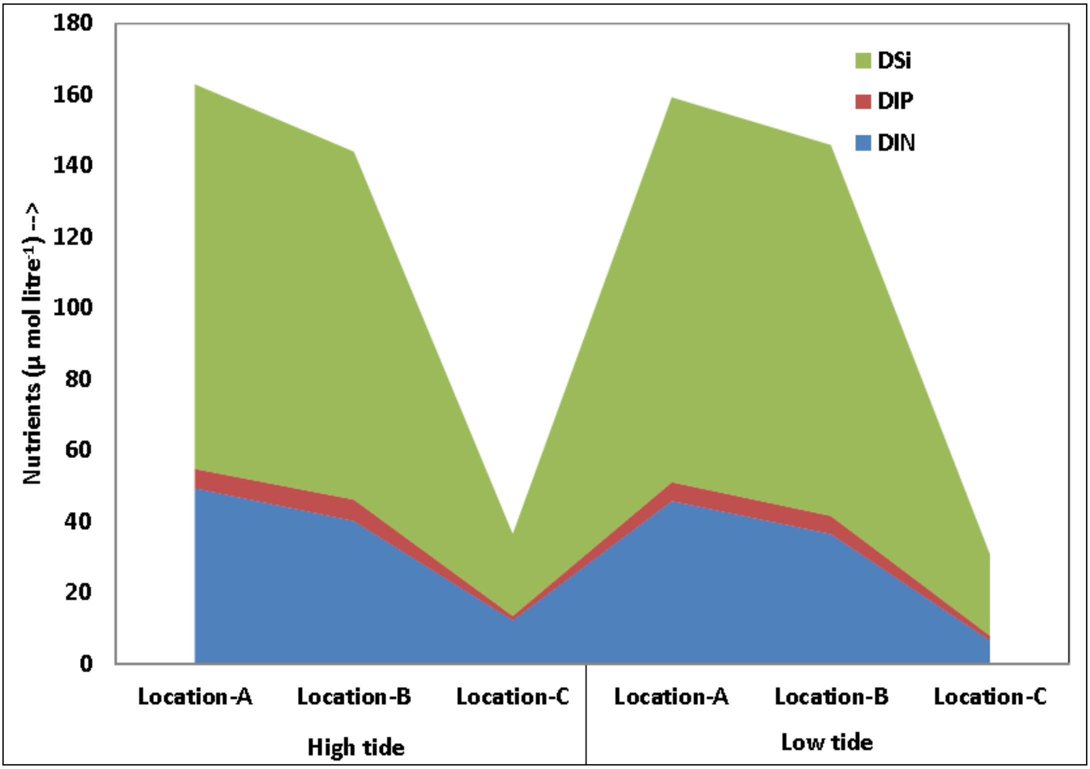

Figure 3. Nutrient variations influenced by tides and locations

3.5. Nutrient fluxes

The nutrient fluxes from groundwater to the Bay of Bengal has been attempted by multiplying the nutrient concentration of each locations with the groundwater flux from the study area as suggested by Daniel Montiel et al, (2018) [49].

$N_{Nutrient\ flux} = F_{GD}$ x Nutrient concentration

Where, N nutrient is the groundwater attributed nutrient flux, FGD is the fresh groundwater discharge and Nutrient concentration is the concentration of the nutrient by each location.

Table 4. Groundwater supported nutrient fluxes with reference to tidal variations and locations.

Groundwater supported Nutrient flux (Tmol per day)				
Tide	Locations	DIN	DIP	Dsi
High tide	Location-A	49.35	5.47	108.17
	Location-B	40.14	6.01	97.81
	Location-C	12.15	1.30	22.95
Low tide	Location-A	45.72	5.37	108.17
	Location-B	36.49	5.13	104.23
	Location-C	6.56	1.42	22.89

The nutrient flux in the location-A (Table 4) is found to be higher due to fresh groundwater discharge and agricultural activities in the study area. Decreasing nutrient fluxes towards coast is mainly due to process of mixing of saline water with groundwater resulting in conversion of oxidative environment to reducing environment that might have acted as sink of nutrients. The other sources might also be due the recirculated saline water observed in location-B and C along with which the nutrients might also be recirculated due the influence of food webs in sedimentation and coastal marine ecosystems [16,45]. The nutrient fluxes through groundwater discharge is found to be contributing of about 77%, 4% and 32% for DSi, DIP and DIN respectively. The total nutrient fluxes from the study area to the bay is calculated to be about 679.33 Tmol/day. The nutrient inputs due to atmospheric deposition is not considered for the present study due to its negligible quantity in comparison with other sources in the coastal regions. Thus, DIN and DSi are noted to contribute for groundwater discharge in the bay in comparison with DIP. Hence, it is concluded that terrestrial sources along with mixing process are contributing nutrients to the bay which will have a significant effect on the coastal eco system in the study area.

4. Summary and conclusion

1. The present study gathers results generated from physiochemical features, groundwater discharge flux and nutrient fluxes from the Sankarabarani River basin, India.
2. The radium isotopes (^{226}Ra) were used to quantify the groundwater flux and nutrients flux were quantified using DIN, DIP and DSi obtained from the samples.
3. According to the physiochemical parameters, the location-A comes under the freshwater zone, location-B demarcated as the subterranean estuary and location-C comes under the influence of saline water, might be due to over drafting, seawater intrusion and impact due to tidal pumping.
4. Radium concentration is increasing towards the coast due to fresh groundwater discharge and mixing of saline water through aquifer.
5. From the radium mass balance, groundwater flux is found to be decreasing towards coast, suggesting the mixing between fresh groundwater with sea water and that gets discharged as recirculated ground or seawater.
6. Nutrients are higher in location-A might be due to agricultural activities and fresh groundwater discharge and found to be decreasing in location-B and C might be due to the seawater mixing influenced by tidal variations. Oxic and anoxic groundwater seems to influence the presence of nutrients in the groundwater environment.
7. The total nutrient fluxes from the land to the sea via groundwater discharge is found to be about 679.33 Tmol/day.
8. It is suggested for long term monitoring to evaluate the groundwater radium and nutrient fluxes and related ecological and environmental consequences.

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Acknowledgements: We sincerely thank Department of Science and Technology (DST), Science and Engineering Research board, New Delhi, India for financial support to carry the present work through Extra Mural Research (Grant No. EMR/2015/001101). The author (PG) also acknowledges DST for granting Junior Research Fellow (JRF) position. The authors thank students and colleagues for their help during the field expeditions.

Conflict of interest: NIL