



Riverine and submarine groundwater nutrients fuel high primary production in a tropical bay



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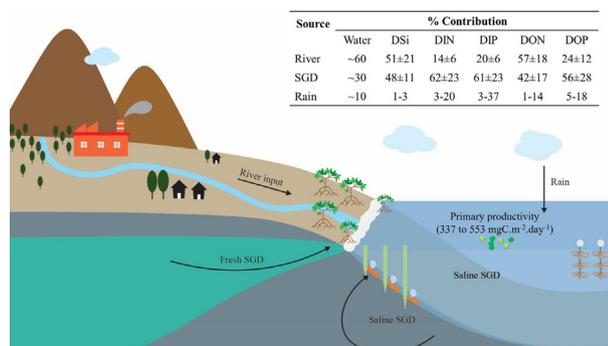
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HIGHLIGHTS

- Tapi River is the main source of nitrate, nitrite and dissolved organic nitrogen to Bandon Bay.
- Ammonia and dissolved phosphorus are mainly originated via submarine groundwater seepage.
- Dissolved organic nutrients are the potential key sources supporting high primary production in Bandon Bay.

GRAPHICAL ABSTRACT



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ABSTRACT

River discharge has long been recognized as a major source of nutrients supporting high primary production (PP) in Bandon Bay, while submarine groundwater discharge (SGD) and atmospheric deposition have largely been overlooked. In this study, we evaluated contributions of nutrients via river, SGD and atmospheric deposition, and their roles on PP in the bay. Contribution of nutrients from the three sources during different time of the year was estimated. Nutrients supply from Tapi-Phumduang River accounted for two-fold the amount from SGD while very little supply was from atmospheric deposition. Significant seasonal difference in silicate and dissolved inorganic nitrogen were observed in river water. Dissolved phosphorus in river water was mainly (80 % to 90 %) of DOP in both seasons. For the bay water, DIP in wet season was two-fold higher than in dry season while dissolved organic phosphorus (DOP) was only one half of those measured in dry season. In SGD, dissolved nitrogen was mostly inorganic (with 99 % as NH_4^+), while dissolved phosphorus was predominantly (DOP). In general, Tapi River is the most important source of nitrogen (NO_3^- , NO_2^- , and DON), contributing >70 % of all considered sources, especially in wet season, while SGD is a major source for DSi, NH_4^+ and phosphorus, contributing 50 % to 90 % of all considered sources. To this end, Tapi River and SGD deliver a large quantity of nutrients and support high PP in the bay (337 to 553 $\text{mg-C m}^{-2} \text{ day}^{-1}$).

1. Introduction

Nutrient has been one of the key indicators for evaluating the water quality and health status of aquatic environment system (EPA, 2019), and it can directly control the level of marine primary productivity

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(Yoshikawa et al., 2017). Global socio-economic development has dramatically increased nutrient loads to rivers (Caraco and Cole, 2001; Seitzinger et al., 2005) thereby elevating the fluxes of nutrients to coastal areas. Meanwhile in some areas groundwater can be an important source of nutrients and other dissolved constituents to the coastal ocean (Burnett et al., 2007; Santos et al., 2021). Increased in the input of nutrients and changes in nutrient ratios owing to anthropogenic activities typically result in eutrophication, which can impact on the primary production and aquatic food webs, causes severe hypoxic events in coastal environments (Diaz and Rosenberg, 2008; Liu et al., 2009), and leads to other socio-economic problems such as threats to fisheries and tourism (Bricker et al., 2008).

As a semi-enclosed bay in the southern part of the Gulf of Thailand (GOT), Bandon Bay is one of the largest cultivation areas of oysters (*Crassostrea lugubris*, *Saccostrea commercialis* and *Crassostrea belcheri*), blood cockle (*Anadara granosa*), green mussel (*Perna viridis*) and white shrimp (*Litopenaeus vannamei*). The area produces >40 % of the total mollusk production in Thailand and has been designated as shellfish production area for export after 1986 (Yakupitiyage and Kaewnern, 2008). However, rapid population growth and economic development in the last few decades have led to deforestation of the mangroves along with massive development of mariculture in the areas surrounding the bay. These result in poor water quality and increased sedimentation (Tookwinas and Youngvanisset, 1998). Moreover, land-based activities surrounding Bandon Bay such as wastewater discharge from domestic, industrial, agricultural and aquacultural activities might further worsen the health of the ecosystem therein (Jarernpornnipat et al., 2003). Climate change may also impact Bandon Bay by making eutrophication and oxygen depletion more severe and frequent. All of these factors can directly affect mollusk culture and their production (Jarernpornnipat et al., 2003).

This study presents the biogeochemistry of Tapi-Phumduang River system and Bandon Bay in wet and dry seasons during 2019–2020. Nutrients in river/estuary, bay, and rain waters and submarine groundwater samples were collected and analyzed. The fluxes of river, submarine groundwater discharge (SGD), and rain and the associated nutrient inputs into Bandon Bay were reported and compared. In addition, its role as a pathway for the cycling of nutrients on the primary production was analyzed and discussed. The results of this study have led to a better understanding of the nutrient sources and dynamics in the region and their relationship with the sustainability of the ecosystems in the coastal areas of Bandon Bay and the adjacent coastal waters.

2. Materials and methods

2.1. Study area

The study area was located in Tapi-Phumduang River basin (Fig. 1), between latitude 7°58.2' to 9°31.0' N and longitude 97°28.4' to 99°46.0' E, covering the total area of 13,454 km² (HII, 2012). Bandon Bay is a shallow bay connected to the GOT and receiving freshwater discharges from Tapi River, the longest river in southern Thailand with the length of 232 km. The river composes of two main rivers, Tapi and Phumduang, joining to become Tapi-Phumduang River at ~30 km upstream of the river mouth before emptying into Bandon Bay (Yakupitiyage and Kaewnern, 2008). Tapi River (catchment area: ~7329 km²) is originated from Khao Luang Mountain range in Nakhon Si Thammarat Province and flow northward, while Phumduang River (catchment area: ~6125 km²) originates from Phuket Mountain range to the west of Surat Thani Province. Phumduang River flow is regulated by Rajjaprappa Dam located at the headwater (Wattayakorn et al., 2001). Downstream of the confluence, Tapi-Phumduang River passes through Surat Thani municipality, where most of the province's population reside. This is where most anthropogenic input enters the river before draining to the bay. No wastewater treatment plant exists in Tapi-Phumduang River basin. Apart from the main river, there are other 18 fringing creeks that supply freshwater to Bandon Bay.

Average tidal range at Tapi River mouth is 1.0 m, with the amplitudes of 0.70 m during neap tides to 1.90 m during spring tides. The

Tapi-Phumduang River Basin is essentially a tropical rain forest climate influenced by monsoon winds, which high temperature and high rainfall. Average temperature was 26.9 °C with monthly temperature ranging from 20.9 °C to 35.2 °C. During December through May (dry season), the catchment area receives little or no rainfall. Meanwhile, the annual rainfall in this area is 1520 mm with a monthly average of 1.8 mm in dry season (February) and 136.4 mm in wet season (HII, 2012).

Bandon Bay is located between latitude 9°10' to 9°40' N and longitude 99°20' to 99°60' E, covering 1070 km², with approx. 80 km of shoreline and only about 20 km² (20 km) of mangrove swamps remains in Bandon Bay (Jarernpornnipat et al., 2003). The average depth of the Bandon Bay was 2.9 m, varying from <1 m to 5 m near the bay mouth (Jarernpornnipat et al., 2003) and 7–8 m at the river channel (Chinfak et al., 2021). The area approximately 480 km², so-called inner bay, is exploited for aquaculture which mainly includes oysters, blood cockles and green mussels (Jarernpornnipat et al., 2003). In the normal situation, the bay receives most of the surface freshwater runoff from Tapi River, with 600 and 150 m³/s discharges in wet and dry seasons, respectively (Wattayakorn et al., 2001; Sunthawanic et al., 2020). The river system receives effluents from intensive agriculture (palms, rubber, and seasonal plantation), fishery industries, large urbanization, as well as intensive inland aquaculture (*Litopenaeus vannamei* and *Oreochromis niloticus*). Nutrient loading from the river and creeks results in high primary production which subsequently supports an extensive production of shellfish therein, including those of oysters, cockles, green mussels, short-necked clam, mud crab and white shrimp (Jarernpornnipat et al., 2003). Additionally, sea-based sources such as tourism (sea-farm homestay) and fishery activities may contribute and alter water quality of the bay.

2.2. Sample collection

Sample collection was performed in July–August 2019 (wet season) and March–April 2020 (dry season). River water, seawater, groundwater, and rainwater were collected for chemical analyses from 29 stations (Table 1 and Fig. 1).

Water samples were collected using 2.5 L water sampler at 0.5 m below surface. The anchor station was set up at the middle of Tapi river mouth (D1), and at a water house in the bay (D2) surrounded by cockle farms. Diurnal sampling was carried out at 3 h-interval over a complete tidal cycle (25 h). Rainwater samples were collected at station D2, using pre-cleaned 1 L glass beaker in August 2019 and March 2020 and to represent wet and dry seasons, respectively. The method of collection was slightly modified from Gioda et al. (2008). The rain collectors were placed on the rooftop of a stilt house in the bay approximately 5 m above sea surface to avoid splash or throughfall contamination from sea spray. Samples were collected by discarding the first 5 min of rain and collecting the rainwater until the end of the event. Two heavy rain events were collected in each season because we could obtain enough water sample for analysis during those heavy rain events. The water samples from shallow wells (W1–W4) were collected using a 2.5 L water sampler to collect water at 0.5 m below the water surface. Water samples from submarine groundwater discharge (SGD1–SGD3) were collected in mangrove swamps using piezometers (Fig. 2). Meanwhile, ambient seawater samples were collected during both sampling periods. Groundwater seepage fluxes were measured by seepage meters (Fig. 2).

The piezometer was a 5-cm diameter tube made of Plexiglas. Of its 2.5-m length, the last 20 cm of one end was perforated with 1.6-mm diameter holes around the cylinder and was capped with conical shape polyethylene (PE) for easy insertion into the sediment. During deployment, the piezometer was inserted 1 m deep into the sediment. At the start, we siphoned out any water in piezometer well and allowed new groundwater to refill for at least an hour. Seepage water samples were later collected using a 50 ml-PE syringe.

The seepage meter, made according to Lee (1977), was a rigid plexiglas cylinder measuring 40 cm in diameter and height with one open end. This open end of the cylinder was inserted into the bottom sediment with the

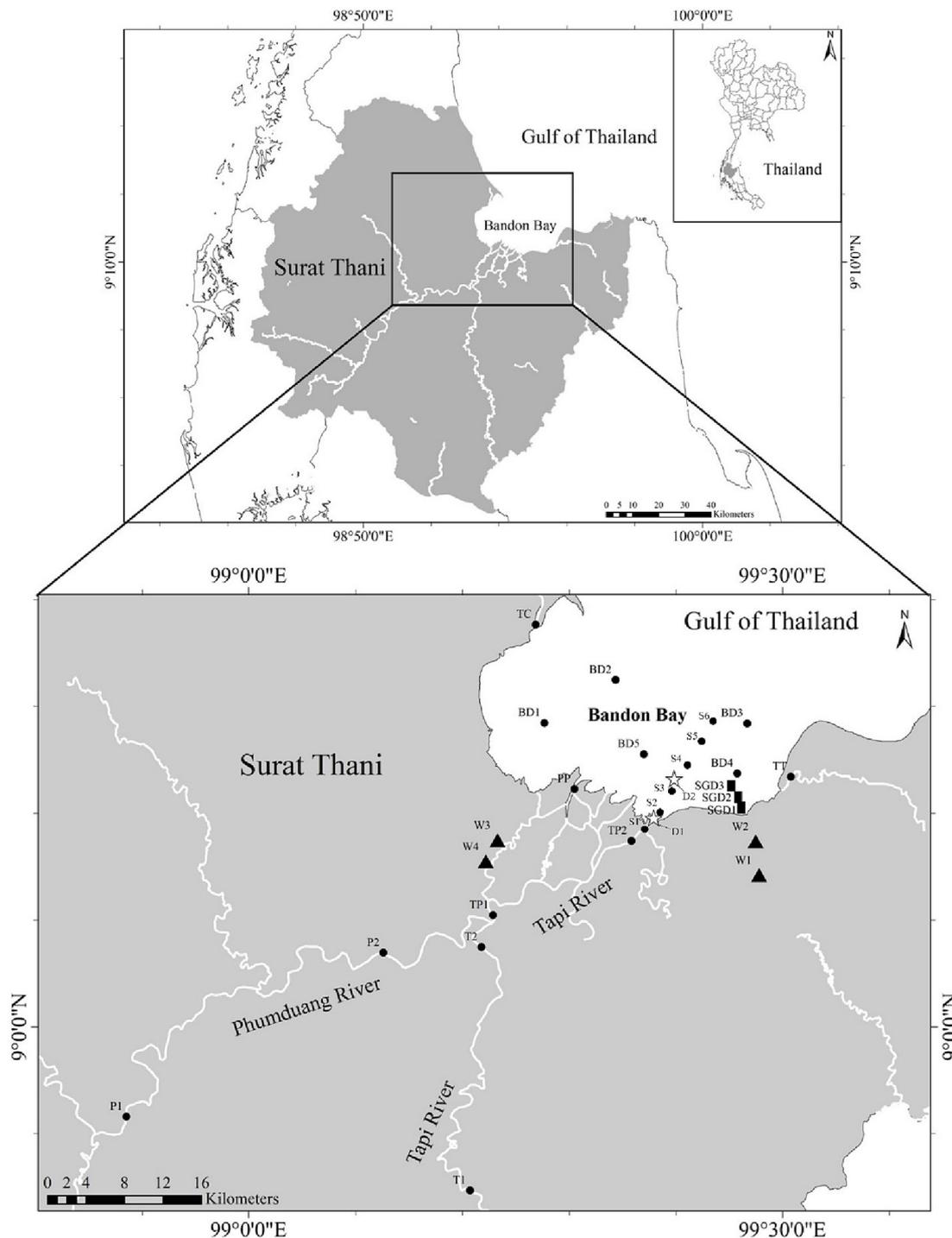


Fig. 1. Station locations in Tapi-Phumduang River system and Bandon Bay: water and sediment sampling stations (black circles), shallow well sampling station (black triangles), seepage meter stations (black rectangles) and two anchor stations for diurnal study (open stars).

head space of approximately 1 cm. Seepage groundwater was collected in a 2-L PE plastic bag which was connected to a small port at the top of the meter to collect diffused seepage water. The bag was pre-cleaned with 1 L of seawater to prevent possible short-term artifacts relating to the expansion of the collector bags (Shaw and Prepas, 1989). A series of three seepage meters were installed on the mud flat during low tide. The three seepage meters were placed perpendicular to the coast. The nearest to shore was 100 m from the shoreline, and other two were placed at the 50 m-interval from the first one. After installation, meters were pumped to dry and allowed the water to refill covering one tidal cycle (24 h). The seepage flux (in $m^3 m^{-2} d^{-1}$ or $m d^{-1}$) was calculated from the volume and

collection time in a specific surface area. Meanwhile, ambient seawater samples were collected during both sampling periods.

All water samples for nutrient analysis were collected in 1-L acid-cleaned PE bottles. The samples were filtered at low pressure through a cellulose acetate filter (0.45 μm pore size) directly into 50 ml pre-acid cleaned HDPE Nalgene bottles. The filtrates were preserved by adding one drop of saturated $HgCl_2$. All preserved samples were stored in a dark and cool environment during transportation to the laboratory (Liu et al., 2011).

Chlorophyll a (Chl-a) were determined in all water samples except groundwater and rainwater. The water was filtered through Whatman GF/F filters (0.7 μm , 47 mm diameter) with a known volume of water.

Table 1

Description of sampling sites and sample collection methods. All site numbers are similar to those in Fig. 1.

Station	No.	Sample collection methods
River (6 stations: $n = 6$)		
Phumduang River @ upstream	P1	-Using a 2.5 L water sampler to collect surface water (0.5 m below surface) during low tide.
Phumduang River @ downstream	P2	
Tapi River @ upstream	T1	
Tapi River @ river	T2	
Tapi-Phumduang River @ river	TP1	
Tapi-Phumduang River @ downstream	TP2	
Mangrove creek (3 station: $n = 3$)		
Tha Chana creek @ upstream	TC	
Phunphin creek @ upstream	PP	
Tha Thong creek	TT	
Salinity gradient (6 station: $n = 6$)		
Tapi-Phumduang river @ downstream	S1	
Tapi-Phumduang river @ river mouth	S2	
Tapi-Phumduang river @ estuary	S3	
Bandon Bay @ inner bay	S4	
Bandon Bay @ inner bay	S5	
Bandon Bay @ outer bay	S6	
Bay (5 station: $n = 5$)		
Bandon Bay @ inner bay	BD1	
Bandon Bay @ outer bay	BD2	
Bandon Bay @ outer bay	BD3	
Bandon Bay @ inner bay	BD4	
Bandon Bay @ inner bay	BD5	
Time series (2 stations: $n = 19$)		
Tapi-Phumduang River @ river mouth ($n = 10$)	D1	-Using a 2.5 L water sampler to collect surface water at 3 h-interval over a complete tidal cycle (25 h)
Bandon Bay @ inner bay ($n = 9$)	D2	-Using the dark-light bottles to determine the primary productivity during neap tide.
Rainwater (share station with bay time series: $n = 2$)		
Bandon Bay @ inner bay	D2	-Using pre-cleaned 1 L glass beaker to collect rainwater during heavy rain
Shallow well (4 stations: $n = 4$)		
Shallow well @ Kanchanadit	W1	-Using a 2.5 L water sampler to collect surface well water after siphoned for 30 min.
Shallow well @ Kanchanadit	W2	
Shallow well @ Phunphin	W3	
Shallow well @ Phunphin	W4	
Seepage water (3 stations: $n = 6$)		
Seepage @ Kanchanadit ($n = 2$)	SGD1	-Using a 50 ml-PE syringe to collect seepage water form piezometer after siphoned for at least an hour.
Seepage @ Kanchanadit ($n = 2$)	SGD2	-Using a seepage meter attached with a 2-L PE plastic bag to collect seepage water for the seepage flux calculation.
Seepage @ Kanchanadit ($n = 2$)	SGD3	

The filters were preserved by adding a few drops of saturated $MgCO_3$, kept frozen, and transported to the laboratory for immediate analysis.

2.3. Physicochemical characteristics

Physicochemical characteristics including temperature, salinity, dissolved oxygen (DO), pH and turbidity were measured in situ prior to

sampling using an In-Situ® Aqua TROLL 600 Multiparameter probe. Water depth during sampling was measured by Hondex® portable depth sounder.

2.4. Determination of primary productivity

Primary productivity (PP) incubation experiments were set up at the diurnal station in the bay (D2) in both wet and dry seasons. Rates of oxygen consumption and production were determined using dark-light bottles method. Dissolved oxygen (DO) concentration in each bottle was measured by Winkler method (Strickland and Parsons, 1972). Water at 0.5-m depth was collected and carefully filled into three pre-cleaned BOD bottles. DO concentration in the first bottle was determined immediately and served as an initial condition. The second bottle (dark bottle) was wrapped with aluminum foil to prevent light penetration and hence served as a control to measure respiration. The third bottle (light bottle) allowed water to be exposed to light. The dark and light bottles were installed back to 0.5-m depth and kept incubated at natural light conditions for 4 h from 10 am to 2 pm. Net productivity, DO produced in the light bottles, is the difference between the amount of DO produce through photosynthesis and that consumed through aerobic respiration. The amount of DO in the dark bottle was DO left after aerobic respiration. The total amount of DO production is called gross productivity. Three replicates were made in each season.

2.5. Chemical analyses

Dissolved inorganic nutrients including dissolved inorganic nitrogen (DIN) [nitrite (NO_2^-), nitrate (NO_3^-), ammonium (NH_4^+)], dissolved inorganic phosphorus (DIP) [phosphate (PO_4^{3-})], and dissolved silica or silicate (dissolved silica or DSI) were determined based on colorimetric method following standard methods described in Strickland and Parsons (1972) and Grasshoff et al. (1999). Total dissolved phosphorus (TDP) and total dissolved nitrogen (TDN) was analyzed based on colorimetric method after hot persulfate oxidation (Strickland and Parsons, 1972). Dissolved organic nitrogen (DON) was calculated by subtracting DIN from TDN, as well as dissolved organic phosphorus (DOP). Urea as part of DON was determined by the colorimetric method described in Grasshoff et al. (1999).

Analysis of Chl-a, a main algal pigment, was analyzed using a fluorometric method after extraction in 90 % acetone. The extracted solutions were excited by the blue light wavelengths, with resulting fluorescence in red which was detected by a photomultiplier. Correction of the significant fluorescence by phaeopigments was performed by acidifying the sample to convert all Chl-a to phaeopigments. By applying a measured conversion for the relative strength of chlorophyll and phaeopigment fluorescence, the two values can be used to calculate Chl-a concentrations (Knap et al., 1994).

2.6. Statistical analyses

The Kolmogorov-Smirnov and Shapiro-Wilk methods were used to ensure the normal distribution of data. Then these data were used to compare between sources (e.g., river, bay, rain, well, seepage waters) by using one-way ANOVA with post-hoc multiple comparisons (Tukey's HSD test). For seasonal comparison, t -test dependent ($n < 30$) was used to compare data between two seasons. Pearson's two-tailed correlation analysis was used to test significant correlation among variables. The correlation was considered statistically significant at 95 % ($p < 0.05$) and 99 % ($p < 0.01$). All statistical analyses were performed by IBM SPSS® statistical software 26.0. In addition, ArcGIS® 10.3.1 was used to create the values distribution maps.

3. Results

3.1. Physicochemical conditions

As the water at the time of sampling was shallow (2 to 7 m in the river and < 1 to 4 m in the bay), a very small difference in physicochemical conditions (temperature, salinity, and pH) between surface and bottom waters

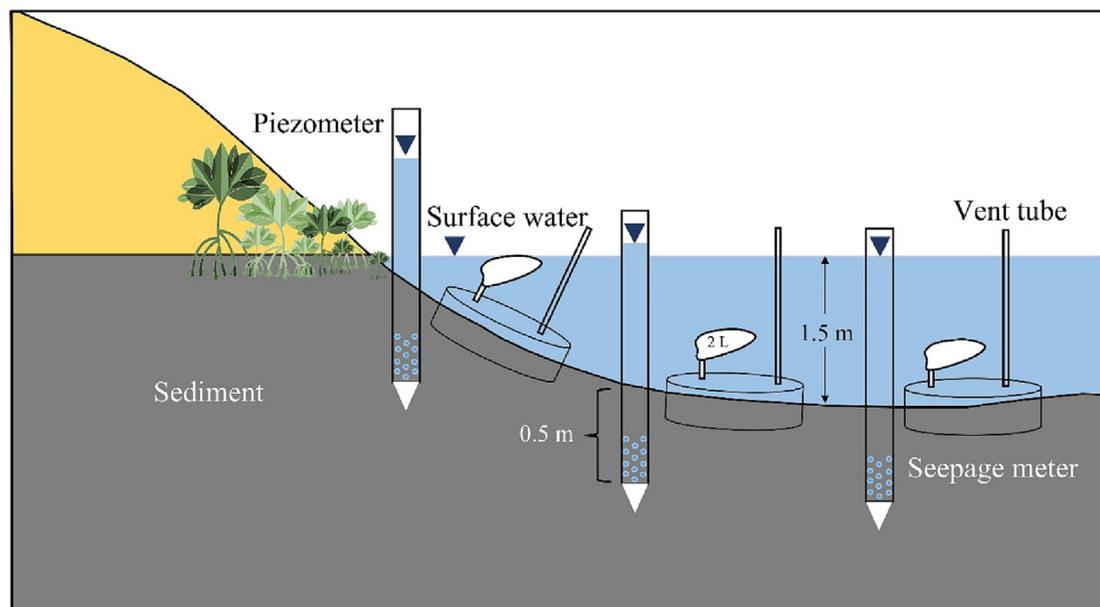


Fig. 2. Concept diagram of piezometers and seepage meters in mud flats adjacent to mangrove swamps.

of the river was observed. Hence, we used surface water at 0.5 m to represent the whole for the water column in this study. Physicochemical conditions in the river including mangrove creek, bay, shallow well water, submarine groundwater, and rain waters were summarized in Table 2.

On the scale of the entire river system and bay, the average surface water temperatures did not show any significant difference between wet and dry seasons. Salinity generally ranged from 0 in the upstream to 4 at the river mouths with the exception of Thachana mangrove creek (approx. 26 in both seasons). Low salinity in the bay stations reflected high influence of freshwater from the rivers. Salinity of the outer bay water (BD2 and BD3) ranged from 26 to 33, while in the inner bay (BD1, BD4 and BD5) was 18 to 20. Slightly higher DO was found in the bay (5 to 7 mg L^{-1}) than those in the rivers (4 to 7 mg L^{-1}). In general, pH in the rivers was generally lower and more variable than those in the bay (Table 2) except for station T1, which is located at the upstream reach of Tapi River (8.60 in wet season and 8.35 in dry season). At station T1, we also found the highest concentration of Chl-*a*. Average pH of rainwaters was 6.65 and 5.93 in wet and dry seasons, respectively. Turbidity was observed to be highest at the Tapi river mouth (TP2). Temperature in shallow well water was slightly lower than those in river water and seawater, and there was no significant difference between seasons. Salinity of SGD collecting in the bay was generally higher than those measuring in the shallow well, indicating the recirculation of seawater. The pH values in the shallow well waters (5.34 to 8.01) were lower than those in the SGD waters (7.35 to 7.76). DO concentrations in shallow wells were low and did not differ between seasons (Table 2).

3.2. Spatial and temporal variation of nutrients

Dissolved nutrients both inorganic and organic forms measured in the river, bay, shallow well, SGD water samples are reported in Table 2. The spatial distribution of nutrients in rivers and Bandon Bay were illustrated in Fig. 3.

3.2.1. Nutrient concentration in Tapi-Phumduang River

Nutrient concentration along the main river varied greatly in both space and time. DSI in the main river ranged from 61 to 213 μM . It was found that NO_3^- ($16.9 \pm 7.39 \mu\text{M}$) occupied 71 to 96 % of the DIN in wet season, while NH_4^+ ($7.13 \pm 2.63 \mu\text{M}$) was a dominant nitrogen species in dry season accounting 36 to 75 % of DIN. In general, NO_2^- , as an intermediated species, was low in the river (mostly $<1.0 \mu\text{M}$).

In wet season, DIN represented approx. 40 % of TDN, while only 14 % in dry season, indicating an enrichment of DON in the river during dry season, especially in the upstream sections. DON in stations T1 and T2 in Tapi river were 300 and 157 μM (accounted approx. 95 % of TDN), and in P1 and P2 were 70.5 and 91.9 μM (accounted approx. 85 % of TDN), respectively. The concentration of DIP was lower than 1 μM in both seasons. Average DOP concentrations, accounting for 80 to 90 % of TDP, during wet and dry seasons were 2.49 ± 1.16 and $3.02 \pm 0.22 \mu\text{M}$, respectively. The nutrient fluxes from rivers into the estuary system (Table 3) were estimated from the nutrient concentrations multiplied by the average freshwater discharges of the Tapi River (600 and $155 \text{ m}^3/\text{s}$ for wet and dry seasons, respectively).

The concentration of urea, one form of DON, in the main river, was $0.83 \pm 0.27 \mu\text{M}$ (2 % of DON) and $4.04 \pm 1.62 \mu\text{M}$ (8 % of DON), in wet and dry season, respectively. Higher concentration of urea was observed in all upstream stations in both seasons, especially in Tapi River (T1 and T2).

3.2.2. Nutrient concentration in mangrove creek

All available nutrients (DSi , NO_2^- , NO_3^- and NH_4^+) in the wet season were higher than in dry season. Average DSI was approx. 100 μM in the wet season and $\sim 40 \mu\text{M}$ in the dry season. NO_2^- in all stations was $<1 \mu\text{M}$, while NO_3^- and NH_4^+ varied from 0.24 to 10.7 μM and 1.53 to 4.33, respectively. In most stations, PO_4 was $<1 \mu\text{M}$ except at the Thathong channel in wet season (2.39 μM), which is higher than river and bay waters. DON accounted for 70 % to 90 % of TDN and DOP accounted for 50 % to 90 % of TDP. Urea, which contributed approx. 2 % to 6 % of DON, was generally lower in wet season (0.57 to 1.16 μM) compared to dry season (2.06 to 3.32 μM).

3.2.3. Nutrient concentration in Bandon Bay

In general, nutrients in bay water were lower than those examined in the rivers. Unlike the rivers, only DSI and NO_3^- showed significant difference between seasons ($P < 0.01$). DSI and NO_3^- concentrations in the wet season were four times higher than in the dry season. No significant difference of NH_4 concentration between wet and dry seasons (0.73 to 2.60 μM). NO_2^- concentration in all stations was $<0.5 \mu\text{M}$, indicating active N-conversion in bay water. Similar to river water, dissolved N species in the bay water was predominated by DON, which represented 80 % to 90 % of TDN in wet season and 70 % to 80 % in dry season. The concentration of DIP and DOP was low in both seasons with the average of <1 to

Table 2
Nutrient concentrations in Tapi-Phumduang River system, mangrove creeks, Bandon Bay, shallow wells, submarine groundwater, and rain waters during wet (July 2019) and dry (March 2020) seasons.

	Temp (°C)	Salinity	pH	DO (mg/l)	Turbidity (NTU)	DSi (μM)	NO ₂ ⁻ (μM)	NO ₃ ⁻ (μM)	NH ₄ ⁺ (μM)	DIN (μM)	Urea (μM)	DON (μM)	DIP (μM)	DOP (μM)	Chl a (μg/L)
Wet season															
Tapi-Phumduang River system (n = 6)															
Min-Max	29.4–32.2	0.02–0.42	7.44–8.60	4.08–6.93	9.68–144	161–213	0.13–3.90	6.89–27.3	0.19–5.72	7.21–33.0	0.51–1.27	20.3–50.3	0.12–0.85	0.68–3.76	1.24–4.63
Average ± SD	31.0 ± 0.91	0.10 ± 0.14	7.84 ± 0.40	5.70 ± 0.85	46.16 ± 45.8	180 ± 19.6	1.38 ± 1.32	16.9 ± 7.39	1.51 ± 1.96	19.8 ± 9.30	0.83 ± 0.27	28.8 ± 10.1	0.37 ± 0.26	2.49 ± 1.16	2.55 ± 1.09
Mangrove creeks (n = 3)															
Min-Max	29.8–32.6	3.1–26.4	7.1–7.9	4.8–5.1	17.4–79.5	35.2–160	0.32–0.79	0.24–10.7	1.94–4.33	2.90–13.5	0.57–1.16	32.4–63.3	0.30–2.39	0.70–3.15	0.78–1.58
Average ± SD	31.0 ± 1.2	11.8 ± 10.4	7.50 ± 0.30	4.90 ± 0.10	38.4 ± 29.1	98.4 ± 51.1	0.40 ± 0.09	4.44 ± 4.51	2.87 ± 1.05	9.93 ± 4.97	0.89 ± 0.24	43.0 ± 14.4	1.14 ± 0.90	2.28 ± 1.12	1.24 ± 0.34
Bandon Bay (n = 5)															
Min-Max	28.3–29.6	20.4–33.1	8.03–8.27	5.16–6.58	8.14–68.0	4.80–22.0	0.13–0.33	0.13–0.97	0.73–2.60	1.84–3.11	0.16–0.44	12.1–18.2	0.46–0.91	0.50–1.26	1.78–5.25
Average ± SD	28.8 ± 0.48	26.8 ± 4.65	8.16 ± 0.08	5.86 ± 0.53	23.5 ± 22.9	12.84 ± 5.98	0.21 ± 0.08	0.50 ± 0.34	1.66 ± 0.73	2.37 ± 0.57	0.27 ± 0.27	15.1 ± 2.43	0.64 ± 0.17	0.96 ± 0.28	3.58 ± 1.38
Shallow wells (n = 4)															
Min-Max	28.6–30.8	0.01–0.30	5.34–8.01	1.01–5.75	9.26–29.6	246–497	0.15–1.05	6.83–47.2	0.29–1.86	7.27–50.1	–	42.9–73.6	0.24–2.96	2.87–14.9	–
Average ± SD	28.6 ± 0.09	0.13 ± 0.11	6.85 ± 0.98	2.96 ± 1.79	16.8 ± 7.87	365 ± 90.0	0.56 ± 0.37	25.6 ± 17.2	0.98 ± 0.65	27.2 ± 17.8	–	59.4 ± 13.0	1.77 ± 0.99	7.82 ± 5.12	–
SGD (n = 3)															
Min-Max	–	17–20	7.35–7.76	–	–	215–317	0.09–0.15	0.13–0.31	131–145	131–145	–	23.1–32.0	4.52–5.97	21.7–24.8	–
Average ± SD	–	18.3 ± 1.09	7.56 ± 0.31	–	–	255 ± 44.7	0.11 ± 0.03	0.20 ± 0.08	139 ± 5.79	139 ± 5.82	–	28.7 ± 5.40	5.30 ± 1.52	23.1 ± 7.91	–
Rain (n = 2)															
Min-Max	–	–	6.20–6.70	–	–	2.16–2.24	0.07–0.11	0.59–0.95	5.11–7.08	5.78–8.15	–	1.55–2.17	0.44–0.49	0.83–0.97	–
Dry season															
Tapi-Phumduang River system (n = 9)															
Min-Max	29.6–30.8	0.04–3.24	7.23–8.35	4.55–6.83	11.5–204	61.4–85.0	0.34–0.69	1.91–8.04	1.97–10.8	5.53–18.9	2.22–6.47	29.4–300	0.27–0.53	2.72–3.36	1.46–7.64
Average ± SD	30.2 ± 0.46	0.62 ± 1.17	7.65 ± 0.37	5.82 ± 0.80	58.1 ± 66.9	71.1 ± 8.71	0.49 ± 0.13	4.56 ± 2.34	7.13 ± 2.63	12.2 ± 4.34	4.04 ± 1.62	115 ± 92.5	0.44 ± 0.09	3.02 ± 0.22	3.61 ± 2.26
Mangrove creeks (n = 3)															
Min-Max	29.2–30.2	0.16–26.3	7.72–7.84	5.52–6.99	14.9–23.2	22.2–66.2	0.27–0.46	1.91–2.47	1.53–3.69	3.70–6.63	2.06–3.32	37.4–110	0.36–0.71	3.04–3.68	1.26–2.58
Average ± SD	29.6 ± 0.42	10.3 ± 11.43	7.77 ± 0.05	6.32 ± 0.61	18.2 ± 3.60	43.1 ± 18.0	0.40 ± 0.09	2.11 ± 0.25	2.45 ± 0.91	4.96 ± 1.23	2.87 ± 0.58	62.7 ± 33.5	0.53 ± 0.14	3.29 ± 0.28	1.72 ± 0.61
Bandon Bay (n = 5)															
Min-Max	29.7–30.4	18.4–33.3	7.79–7.90	5.82–6.72	6.47–46.6	25.4–60.0	0.08–0.31	1.86–2.27	1.43–2.17	3.41–4.74	0.23–0.72	11.6–19.7	0.22–0.53	1.01–1.62	1.16–3.85
Average ± SD	30.0 ± 0.24	27.9 ± 5.75	7.85 ± 0.04	6.27 ± 0.29	28.1 ± 15.7	45.9 ± 12.6	0.22 ± 0.08	2.13 ± 0.14	1.76 ± 0.31	4.11 ± 0.47	0.42 ± 0.20	16.2 ± 2.74	0.37 ± 0.12	1.22 ± 0.22	2.44 ± 1.14
Shallow wells (n = 4)															
Min-Max	27.8–29.0	0.10–1.25	5.78–6.79	1.72–3.85	21.70–50.0	73.4–118	0.84–4.24	2.86–14.4	0.14–1.10	7.54–17.0	–	40.2–69.1	1.02–9.56	2.52–5.01	–
Average ± SD	28.5 ± 0.50	0.45 ± 0.47	6.32 ± 0.45	2.71 ± 0.76	34.7 ± 10.0	97.1 ± 18.0	1.95 ± 1.53	6.59 ± 4.77	0.38 ± 0.42	8.91 ± 5.09	–	46.7 ± 13.8	3.56 ± 3.51	5.43 ± 3.70	–
SGD (n = 3)															
Min-Max	–	28.0–29.0	7.47–7.58	–	–	103–155	0.11–0.23	0.28–0.33	63.6–68.0	64.2–68.5	–	49.6–56.3	2.13–3.29	2.27–2.73	–
Average ± SD	–	28.7 ± 0.43	7.54 ± 0.09	–	–	133 ± 45.2	0.18 ± 0.05	0.31 ± 0.43	66.0 ± 1.81	66.5 ± 1.77	–	52.6 ± 2.78	2.77 ± 0.48	3.53 ± 1.47	–
Rain (n = 2)															
Min-Max	–	–	5.7–5.9	–	–	3.36–4.40	0.07–0.11	1.26–1.52	14.2–15.0	15.8–16.3	–	3.16–4.61	0.53–0.58	1.26–1.55	–

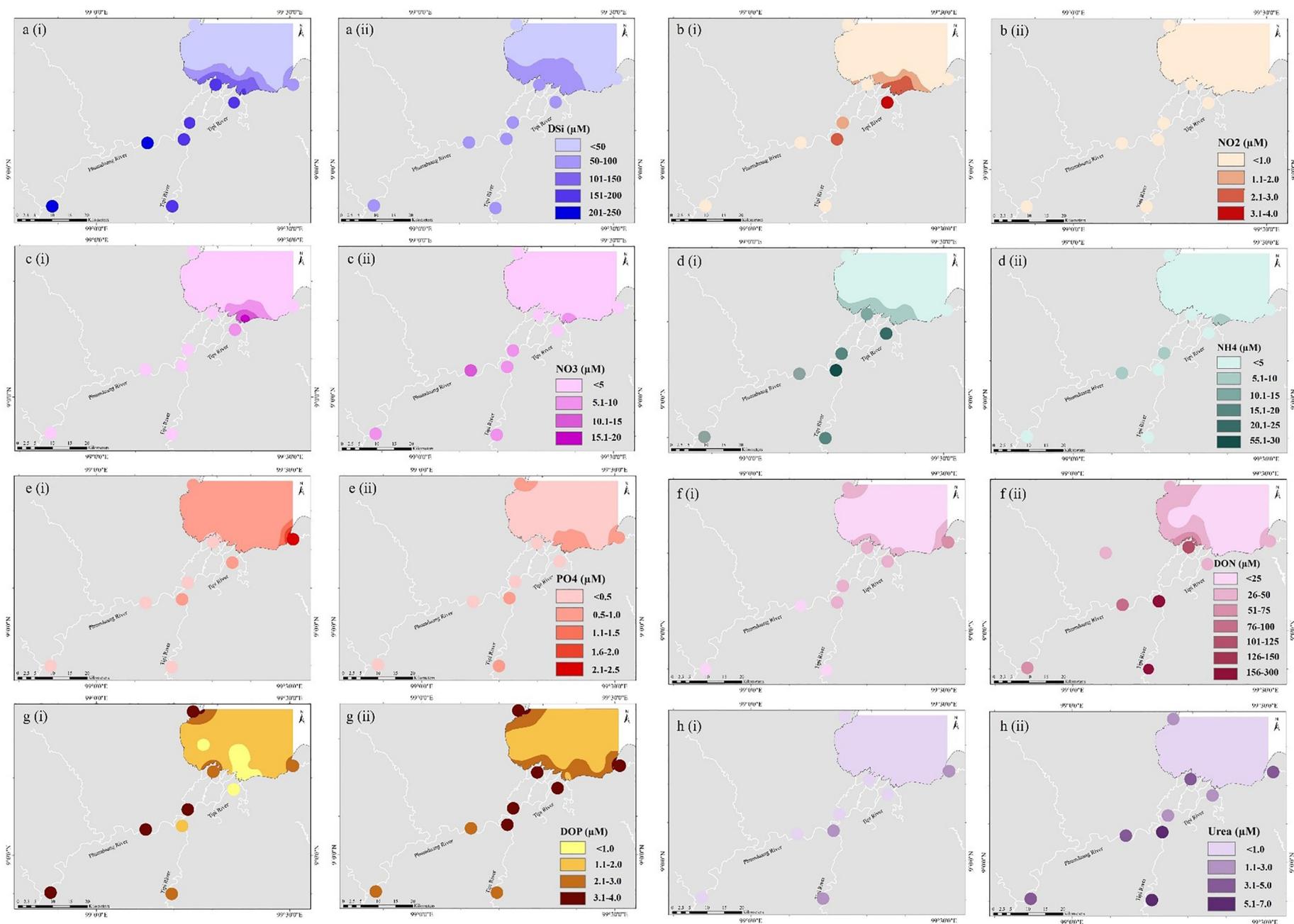


Fig. 3. Distribution of nutrients in the study areas: (a) DSI, (b) NO_2^- , (c) NO_3^- ; (d) NH_4^+ , (e) PO_4^{3-} or DIP, (f) DON, (g) DOP, and (h) urea during (i) wet season in 2019 and (ii) dry season in 2020.

Table 3
Nutrient fluxes via Tapi-Phumduang River, rainwater, and submarine groundwater into Bandon Bay.

	River conc. (μM)	River flux (moles/day)	Rain conc. (μM)	Rain flux (moles/day)	Seepage conc. (μM)	Seepage flux (moles/day)	% Contribution		
							River	Rain	SGD
Wet season									
DSi	91.3 \pm 40.7	7.89 $\times 10^4$	2.20 \pm 0.04	4.80 $\times 10^3$	255 \pm 44.7	5.51 $\times 10^4$	57	3	40
NO ₂ ⁻	0.95 \pm 0.27	8.24 $\times 10^2$	0.09 \pm 0.02	2.02 $\times 10^2$	0.11 \pm 0.02	2.42 $\times 10^1$	78	19	2
NO ₃ ⁻	6.78 \pm 3.47	5.86 $\times 10^3$	0.77 \pm 0.18	1.68 $\times 10^3$	0.20 \pm 0.08	4.27 $\times 10^1$	76	22	2
NH ₄ ⁺	5.65 \pm 2.72	4.88 $\times 10^3$	6.10 \pm 0.99	1.33 $\times 10^4$	138 \pm 5.77	2.99 $\times 10^4$	10	28	62
DIN	13.4 \pm 6.19	1.66 $\times 10^4$	6.96 \pm 1.18	1.52 $\times 10^4$	139 \pm 5.80	4.88 $\times 10^4$	15	20	65
DON	19.6 \pm 3.93	1.69 $\times 10^4$	1.86 \pm 0.31	4.06 $\times 10^3$	28.7 \pm 3.96	3.97 $\times 10^3$	70	5	25
DIP	0.72 \pm 0.18	6.23 $\times 10^2$	0.47 \pm 0.02	1.02 $\times 10^3$	5.30 \pm 0.60	1.15 $\times 10^3$	22	37	41
DOP	3.65 \pm 1.25	3.15 $\times 10^3$	0.90 \pm 0.07	1.97 $\times 10^3$	23.1 \pm 1.28	5.00 $\times 10^3$	31	18	49
Dry season									
DSi	85.9 \pm 18.6	1.92 $\times 10^4$	3.88 \pm 0.52	1.12 $\times 10^3$	133 \pm 21.6	2.43 $\times 10^4$	44	1	55
NO ₂ ⁻	0.46 \pm 0.16	1.03 $\times 10^2$	0.09 \pm 0.02	2.59 $\times 10^0$	0.18 \pm 0.05	3.21 $\times 10^1$	76	1	23
NO ₃ ⁻	5.41 \pm 2.27	1.21 $\times 10^3$	1.39 \pm 0.13	4.00 $\times 10^1$	0.31 \pm 0.02	5.75 $\times 10^1$	93	3	4
NH ₄ ⁺	2.05 \pm 0.87	4.58 $\times 10^2$	14.6 \pm 0.37	4.20 $\times 10^2$	66.0 \pm 1.81	1.21 $\times 10^4$	4	3	93
DIN	7.92 \pm 2.84	1.77 $\times 10^3$	16.1 \pm 0.26	4.63 $\times 10^2$	66.5 \pm 1.77	1.22 $\times 10^4$	13	3	84
DON	30.0 \pm 5.61	6.68 $\times 10^3$	3.88 \pm 0.72	1.12 $\times 10^2$	52.6 \pm 2.79	9.62 $\times 10^3$	41	1	59
DIP	0.44 \pm 5.60	1.01 $\times 10^2$	0.56 \pm 0.03	1.60 $\times 10^1$	2.77 \pm 0.48	5.07 $\times 10^2$	16	3	81
DOP	0.55 \pm 0.20	1.24 $\times 10^2$	1.41 \pm 0.15	4.05 $\times 10^1$	3.53 \pm 1.47	6.46 $\times 10^2$	15	5	80

$\sim 1 \mu\text{M}$ (Fig. 3 and Table 2). In the wet season, DIP was two-fold higher than in the dry season, while DOP was only one half of those presented in the dry season. DOP was predominant species of phosphorus in the dry season, while in the wet season DIP and DOP were presented in approximately equal amounts.

Urea concentration in bay water was significantly lower ($<1 \mu\text{M}$) than in the river waters ($p < 0.05$). Average urea concentration was 0.42 ± 0.09 and $0.27 \pm 0.04 \mu\text{M}$ in wet and dry seasons, respectively. In bay water, urea accounted for only 2 % of DON, which is lower than those observed in river water (Fig. 3).

3.2.4. Nutrient concentration in shallow wells

Average DSi, NO₃⁻, and DIN were significantly higher in the wet season than in the dry season. Concentrations of NO₃⁻ in the wet season accounted for 87 % to 97 % of DIN, while only 35 % to 87 % of DIN in the dry season. Average NO₂⁻ and NH₄⁺ concentrations were generally $<1 \mu\text{M}$, which varied between 0.15 to 0.84 μM and 0.14 to 1.86 μM in wet and dry seasons, respectively. DIP concentration varied from as low as 0.24 μM in the wet season to as high as 9.56 μM in the dry season. The higher DIP was observed at W2, which is located at the small urban center during wet and dry seasons. DOP during the wet season ranged from 2.87 to 14.9 μM , accounting for 50 % to 92 % of TDP and was higher than in the dry season. Similar to phosphorus, TDN was predominated by organic form in both seasons. DON ranged from 40.2 to 73.6 μM , which contributed 46 % to 91 % of TDN in wet season and 82 to 91 % of TDN in dry seasons.

3.2.5. Nutrient concentration in submarine groundwater discharge

In general, SDG contained DSi, DIP, and NH₄⁺ higher than other water bodies (river, rain, and bay). DSi in SDG ranged from 100 μM in dry season to 317 μM in wet season. The concentrations of NH₄⁺ ranged from 131 to 145 μM in the wet season and 63.6 to 68.0 μM in the dry season. Most dissolved nitrogen in SDG was predominated in dissolved inorganic forms, which accounted for 83 % of TDN in wet season and for 56 % in dry season, while NH₄⁺ accounted for 99 % of DIN. Average of DIP was higher in wet season than in dry season, with the range from 2.13 μM in dry season to 5.97 μM in wet season. In contrast to nitrogen, DOP was a predominant form in TDP. DOP accounted for 81 % of TDP in wet season and dropped to 53 % in dry season. The estimated fluxes of nutrients to Bandon Bay via SDG were calculated from nutrient concentration multiplied by seepage rate. Specific seepage rates ($16.5 \pm 1.20 \text{ m}^3 \text{ m}^{-1} \text{ d}^{-1}$) were integrated over distance (50–100 m) offshore to provide an estimate of the total seepage per unit width of shoreline per day ($0.28 \pm 0.02 \text{ m}^2 \text{ d}^{-1}$). Assuming muddy along the mangrove swamp shoreline is the source of groundwater (Kristensen and Suraswadi, 2002; Wang et al., 2021; Santos et al., 2021), the

uncertainty of nutrient fluxes from SDG can be calculated following Burnett et al. (2007) by multiply the specific seepage rate with an estimated shore-line length of mangrove swamp in Bandon Bay (20 km). The results have shown in Table 3.

3.2.6. Nutrient concentration in rainwater

Nutrient concentrations in rainwater varied depending on rain events and seasons. Nutrients in rainwater collected at the first rain event in dry season were higher than those of wet season. In general, rainwater contained lower DSi, and NO₂⁻ than other water resources, with an average of 3 μM and 0.1 μM for DSi and NO₂⁻, respectively. Level of DIP in rainwater was similar to river and bay waters, but much lower than in submarine groundwater. Notably, NH₄⁺ in rainwater, accounted for 89 % of DIN, was found higher than in river and bay waters, and being a predominant species among TDN. DON presented only 20 % of TDN, while DOP was about 70 % of TDP, with the concentration comparable to bay water in both seasons. To our knowledge, no published data on dry and wet nutrient depositions in Bandon Bay is available. The uncertainty of nutrient fluxes from rainwater to the bay was determined from the average nutrient concentration and rainfall rate following Xing et al. (2017). Daily fluxes of nutrients from rainwater in wet and dry seasons are reported in Table 3.

3.2.7. Nutrient distribution during estuarine mixing

Concentration of nutrients at the river mouth varied according to the mixing of two-end members. Highest concentration of most nutrients (DSi, NO₂⁻, NO₃⁻, and DIP) was observed in the river end and decreased with an increase of salinity, implying riverine input. While DOP has shown an increasing trend with salinity in wet season, and slightly decreasing with salinity was observed in dry season.

Strong conservative mixing within the estuarine was revealed for DSi, NO₂⁻, NO₃⁻, NH₄⁺, and DIP (except in wet season by negative correlation with salinity ($R^2 > 0.70$), especially in wet season. Most nutrients showed negative relation with salinity, except DON and DOP showed no clear relation with salinity. This indicates that DON and DOP were regenerated from organic matter degradation or be supplied from groundwater. At the mixing zone (salinity 5–15), NH₄⁺ and DIP showed additional behavior in wet season. While the correlation with DOP, and DON was no significantly. Variation of each nutrient concentration as function of salinity has shown in Fig. 4.

3.2.8. Nutrients in tidal cycles

The investigation was carried out at the river mouth (D1) and center of the bay (D2) stations during spring tide in which water level variability was highest (Fig. 5). At the river mouth, surface salinity ranging from 3.44 to

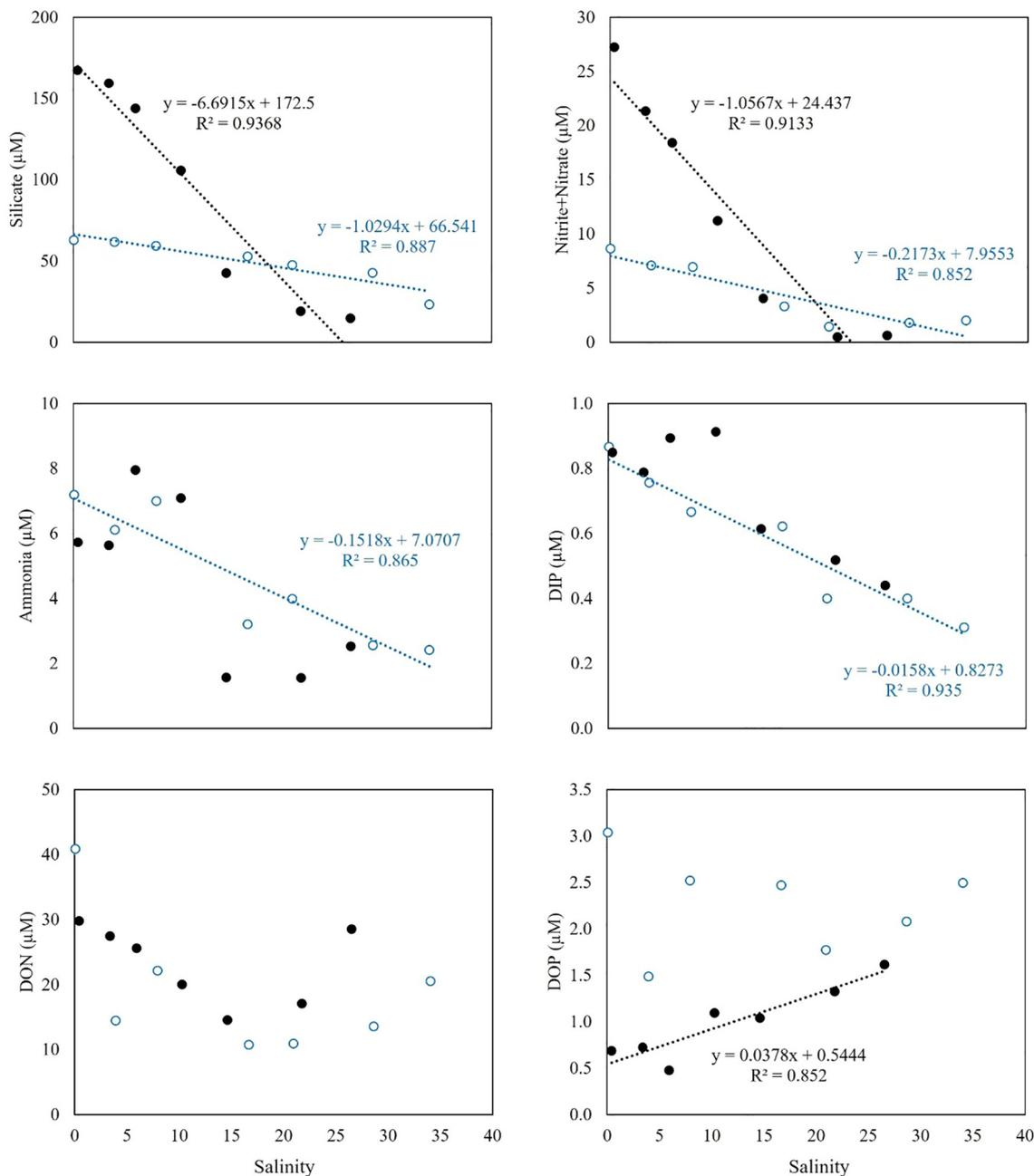


Fig. 4. Variation of nutrient concentrations along the salinity gradient during wet season in 2019 (black circle) and dry season in 2020 (open circle) along with the linear regression lines (when $R^2 > 0.8$).

18.54 in wet season and from 3.72 to 15.14 in dry season were observed. Salinity at the bay station ranged from 13.91 to 27.84 and 20.19 to 32.82 in wet season and dry season, respectively. Water pH ranged from 7.00 to 7.6 at the river mouth station and remained ~8.0 at the bay station. Dissolved oxygen concentration at both stations ranged from 6 to 9 mg/l with the saturation of 70 % to 116 % (average 92 %).

In general, all nutrients showed an increase in concentration during low tide (Fig. 5), reflecting rivers as an important source of nutrients. At the river mouth station, DON and DOP showed weak relations with the tidal cycle. At bay station, the dilution with seawater during high tide did a few changes of NH_4^+ , DIP, DON, and DOP concentrations, especially in dry season, indicating seawater exchange with the GOT and bay activities such as oyster and blue mussel cultures and SGD may be the other sources of these nutrients into the bay. While DIN, NO_3^- , and NO_2^- were higher

during low tide, indicating the river is the primary sources of these nutrient species input to the bay. DSi concentrations at river mouth and center bay stations were high fluctuations, especially during low tide.

3.3. Chlorophyll-a (Chl-a) and primary productivity (PP)

Spatial distributions of Chl-a were examined in Tapi-Phumduang River system and Bandon Bay during wet and dry seasons (Fig. 6). In the rivers and channels, Chl-a concentration in dry season is generally higher than in wet season. The concentrations of Chl-a varied from $1.24 \mu\text{g L}^{-1}$ in wet season to $7.64 \mu\text{g L}^{-1}$ in dry season. The highest concentration of Chl-a was found at the upstream stations of Tapi River (T1 and T2), according to phytoplankton bloom events during the sampling period. The Chl-a concentration in mangrove channels was lower than in the main river,

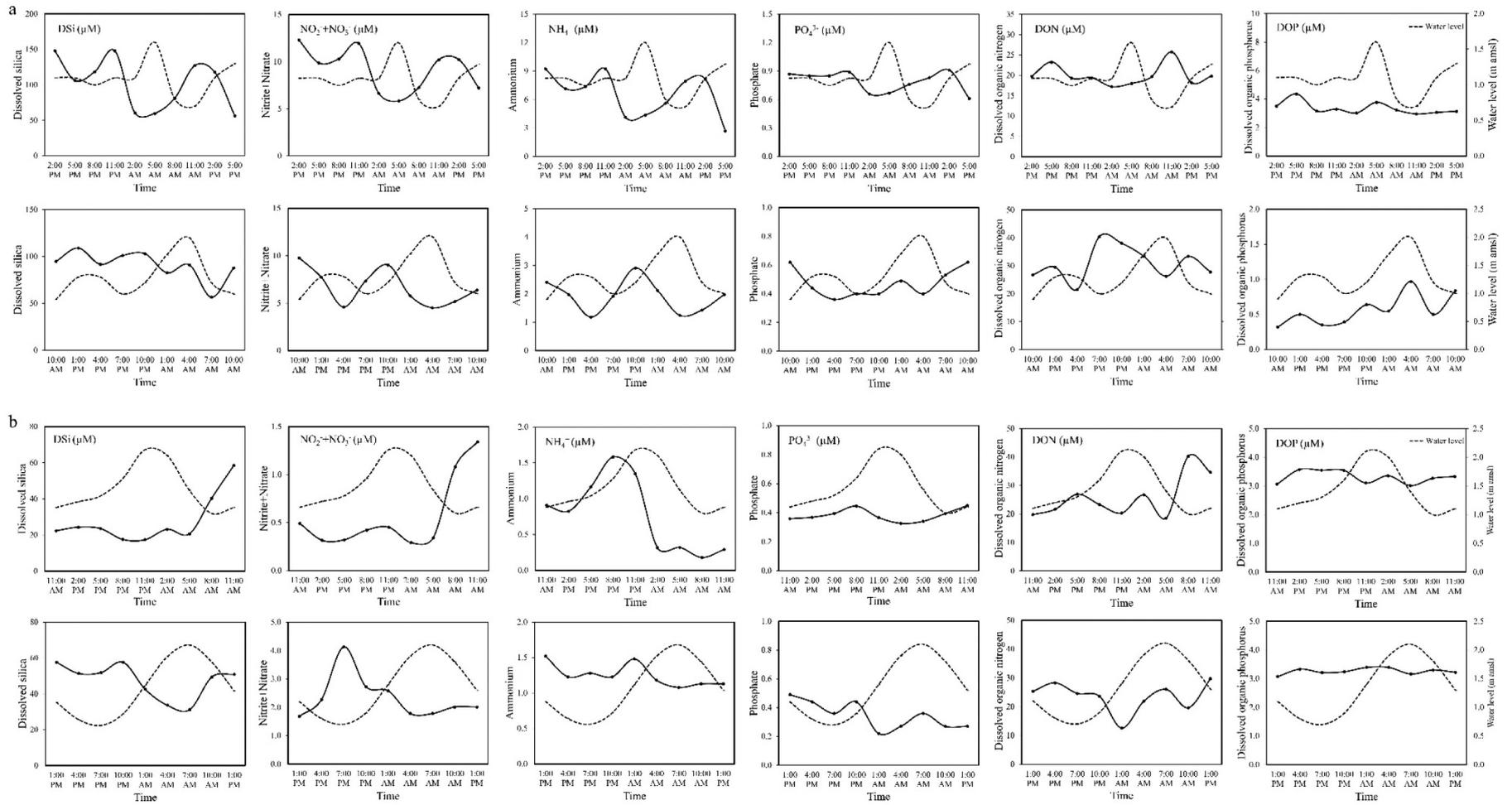


Fig. 5. a. Nutrient dynamics over one tidal cycle at Station D1 (Tapi river mouth) during wet season in 2019 (top row) and dry season in 2020 (bottom row). Dash lines indicate water level (in meter above MSL) and solid lines indicate nutrient concentrations.

b. Nutrient dynamics over one tidal cycle at Station D2 (Bandon Bay) during wet season in 2019 (top row) and dry season in 2020 (bottom row). Dash lines indicate water level (in meter above MSL) and solid lines indicate nutrient concentrations.

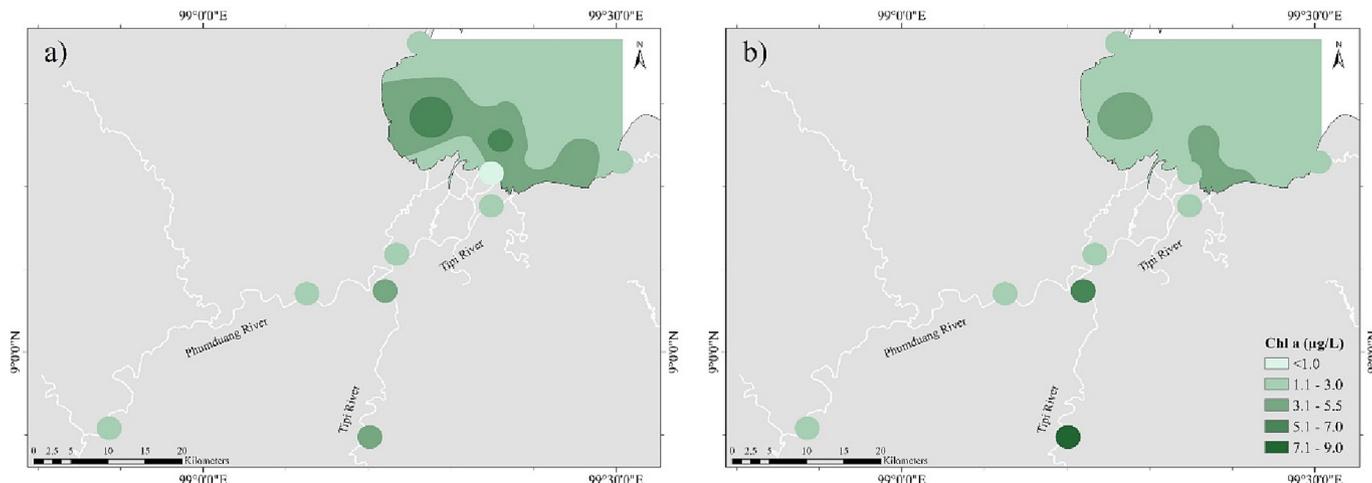


Fig. 6. Distribution of chlorophyll-a in Tapi-Phumduang River system and Bandon Bay during (a) wet season in 2019 and (b) dry season in 2020.

ranging from 0.78 to 2.58 $\mu\text{g L}^{-1}$ with no significantly found during wet and dry season. The Chl-a showed a strong positive relationship with DON ($p < 0.05$, $r^2 = 0.77$) in the river system and the channels (Table 4). Pearson's correlation confirmed the relationship between Chl-a and DON ($p < 0.01$).

In the bay, fluctuation of Chl-a concentration was observed. Slightly higher Chl-a concentration was observed in wet season (1.78 to $5.25 \mu\text{g L}^{-1}$) than in dry season (1.16 to $3.85 \mu\text{g L}^{-1}$). Chl-a concentration in the inner bay were significantly higher than in the outer bay ($p < 0.05$). The Chl-a showed a strong positive relationship DON ($r^2 = 0.68$) and DOP ($r^2 = 0.75$) (Table 4). Pearson's correlation confirmed the relationship between Chl-a and organic nutrient ($p < 0.05$) (Fig. 7).

In Bandon Bay, average concentrations of NH_4^+ and NO_3^- in seawater were of the same magnitude. Assuming both NH_4^+ and NO_3^- may be N sources for photosynthesis reaction in the bay. The photosynthetic quotient (PQ) using in this study is assumed from the average of 1.45 and 1.1 which equal to 1.275. The DO values obtained were converted to PP, (in mg-C L^{-1})

using Eq. (1) as described in Cullen (2001) and consequently, multiply by water depth (1 m) and photosynthesis time (assuming 12 h).

$$\text{PP} (\text{mgC L}^{-1}) = \text{DO} (\text{mg L}^{-1}) \times 0.375/\text{PQ} \tag{1}$$

As a result, PP values at the surface bay water were ranged 452 to $553 \text{ mg-C m}^{-2} \text{ day}^{-1}$ (494 ± 32.1) and 338 to $472 \text{ mg-C m}^{-2} \text{ day}^{-1}$ (404 ± 39.5) in wet and dry seasons, respectively. The PP in wet season was slightly higher than dry season.

4. Discussion

4.1. Nutrient transport in the river system

Nutrient concentrations in the Tapi-Phumduang River basin show a wide range of variation (Table 2) likely caused by seasonal fluctuation in natural and anthropogenic activities. Tapi-Phumduang River was deemed

Table 4
Correlation analysis of chlorophyll-a and physiochemical parameters in surface water of Tapi-Phumduang River system and Bandon Bay.

	Temp	Salinity	DO	pH	Turbidity	DSi	DIN	DIP	DON	DOP	Urea	Chl a
Tapi-Phumduang River												
Temp	1											
Salinity	-0.18	1										
DO	-0.03	-0.25	1									
pH	0.56*	-0.15	0.61**	1								
Turbidity	0.10	-0.12	-0.08	0.02	1							
DSi	0.33	-0.46	-0.05	0.08	-0.02	1						
DIN	0.42	-0.46	-0.41	0.12	0.25	0.50*	1					
DIP	0.48*	0.21	-0.35	0.10	0.29	-0.23	0.07	1				
DON	0.04	-0.20	0.27	0.23	-0.23	-0.28	-0.12	0.04	1			
DOP	-0.48*	0.11	0.50*	-0.06	-0.35	-0.23	-0.69**	-0.67**	0.01	1		
Urea	-0.22	-0.21	0.32	0.07	-0.18	-0.54*	-0.25	-0.08	0.86**	0.24	1	
Chl a	0.29	-0.30	0.37	0.45	-0.23	0.04	0.07	-0.13	0.77**	0.05	0.66**	1
Bandon Bay												
Temp	1											
Salinity	-0.28	1										
DO	0.77**	-0.70*	1									
pH	-0.66*	-0.11	-0.28	1								
Turbidity	-0.25	0.13	-0.42	-0.40	1							
DSi	0.71*	-0.05	0.52	-0.84**	0.02	1						
DIN	0.58	0.12	0.18	-0.93**	0.46	0.74*	1					
DIP	-0.48	-0.19	-0.31	0.51	0.19	-0.59	-0.48	1				
DON	0.32	-0.19	0.45	-0.19	-0.49	0.47	-0.06	-0.02	1			
DOP	0.18	0.54	-0.27	-0.32	0.02	0.15	0.34	-0.71*	-0.35	1		
Urea	0.47	-0.58	0.63	-0.50	0.19	0.70*	0.52	-0.13	0.20	-0.32	1	
Chl a	-0.34	-0.42	-0.01	0.18	0.53	-0.25	-0.11	-0.21	0.68*	0.75**	0.41	1

* Significant correlation at the 0.05 level (2-tailed).

** Significant correlation at the 0.01 level (2-tailed).

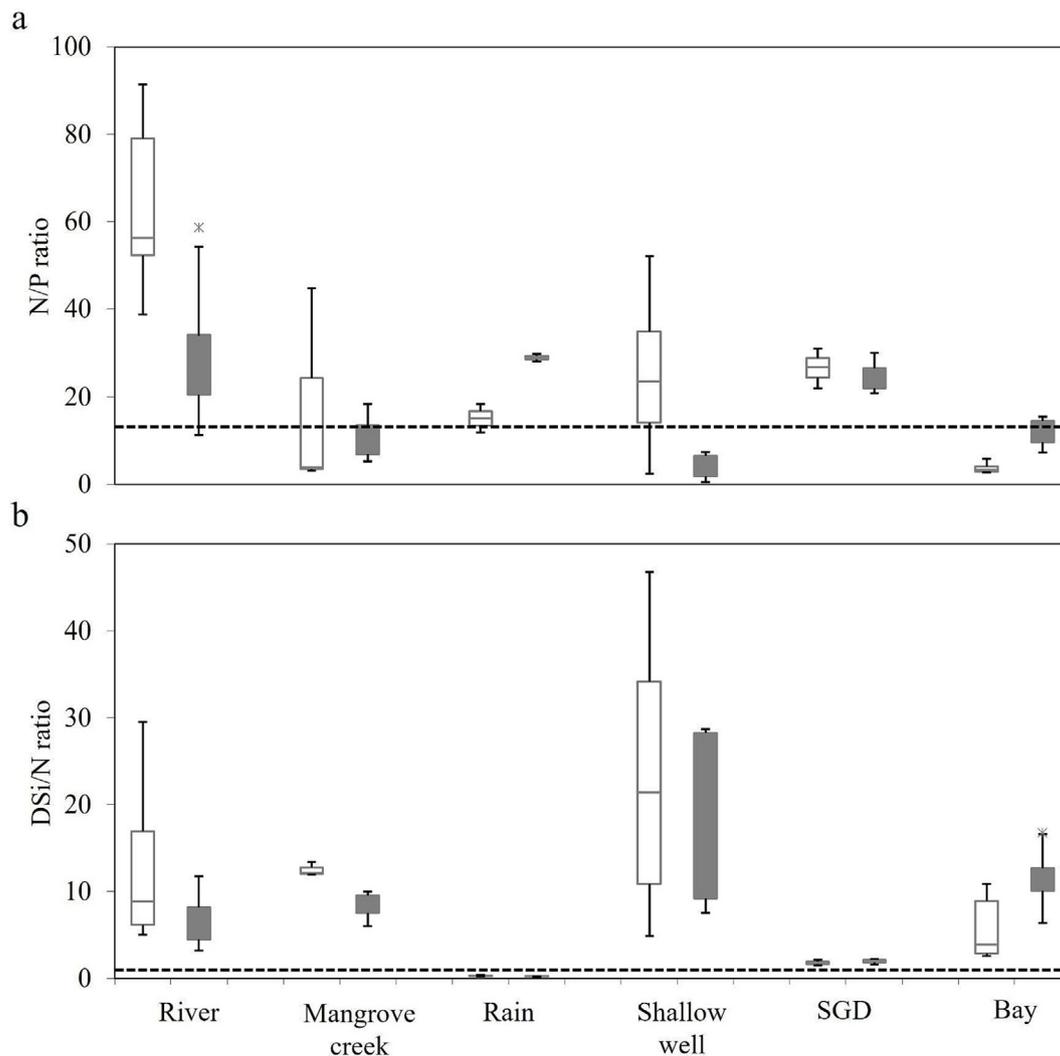


Fig. 7. Ratios of (a) DIN:DIP and (b) DSi:DIN in rivers, mangrove creeks, rain, shallow well SGD and bay in wet season 2019 (white bars) and dry season 2020 (grey bars). Dashed line for N/P ratio (top) is 16 (Redfield ratio) and dashed line for DSi/N ratio (bottom) is 1.

as a less polluted river according to Thailand water quality classification and the classification by DMCR (2020). Higher concentrations of DON at the upstream river (300 μM , $\sim 77\%$ of TDN) are similar to the previous study (316 μM) by Dupra et al. (2000), and portion of DON to TDN was comparable to the global rivers ($>70\%$) by Seitzinger et al. (2005), and mountainous river systems (70%–88%) of tropical Western Peninsular India (Pradhan et al., 2015a, 2015b).

Approximately 30% and 40% of total land use areas of Surat Thani Province covered by dense forest and agriculture area, respectively (Bunruamkaew and Murayama, 2012). Caraco and Cole (1999) and Caraco and Cole (2001) reported that mostly DON exported from watersheds around the world is from natural sources. Numerous studies conducted in forested ecosystems have shown that DON losses can be substantial (Campbell et al., 2000; Perakis and Hedin, 2002). $>70\%$ of the TDN in streams and rivers of temperate South American forest regions consisted of DON (Perakis and Hedin, 2002), and largely similar DON (75% to 83%) were also found in stream waters of the forests and undergone reforestation in northern Thailand (Miller et al., 2005). In addition, DON accounts for 40% to 90% of the TDN in rivers of the SE USA, while DON comprised only 2% of the TDN in the running river water of a forested area in NE USA (Alberts and Takács, 1999).

Concurrent increase in DON concentration with increased population density might be caused by the anthropogenic nitrogen addition to the watersheds through the fertilizer applications or through wastewater disposal,

which may be exported in organic form (Kroeger et al., 2006). Wiegner et al. (2006) reported DON made up a substantial fraction of the TDN pool where the watershed land use was dominated by agriculture: Pocumoke River (83%) and Choptank River (94%), USA. Roughly 78% of DON can be extracted from soil (Miller et al., 2005), and 58% of DON may originate from fertilizers loaded to the watershed (Caraco and Cole, 1999) due to excessive nitrogen fertilizer applied to crop soils that is often not used by the plants and is carried in the runoff polluting groundwater, rivers, and finally into the coastal areas (Tirado et al., 2008). The impact of DON losses from agricultural fields on water quality has already been shown for the Chesapeake Bay area where the large concentrations of DON were related to the surrounding area of agricultural land (Jordan and Weller, 1997).

According to Czerwionka (2016), DON originating from municipal waste could contribute to as high as 73% of the TDN pool and this municipal wastewater DON is made up of primarily aliphatic compounds, which are more easily utilizable for microorganisms compared to their aromatic counterpart from forest soil. To this end, we postulate that most DON observed in this study may derive from dense forest areas upstream. However, an inverse relation between DON concentration and the concentration of DIN, which may indicate that DON can be turned to DIN by microbial activities (Czerwionka, 2016). In addition, DON can be produced by the autolysis of settled phytoplankton cells or the hydrolysis of other highly-bioavailable fraction of particulate organic nitrogen (Hargreaves, 1995).

Moreover, the active or passive release from submerged macrophytes and benthic algae can be another significant input of DON to shallow freshwaters and wetlands (Jansson, 1979).

Urea is one of the most important components of DON pool in freshwater and marine environments (Siuda and Kiersztyn, 2015), yet it constitutes only a small percentage of DON pool in majority of aquatic environments (Berman and Bronk, 2003). Less urea concentration was generally observed in mountainous river systems such as the tropical Western Peninsular India (1 and 3 μM) compared to anthropogenic sources (fertilizer and domestic wastes), which regenerated of urea from the breakdown of DON in the freshwater region (Pradhan et al., 2015a, 2015b).

Urea is an important nitrogen source for aquatic micro-organisms released to freshwater from both natural and anthropogenic sources, such as fertilizers, herbicides, pesticides, and excretion of mammals and other animals (Fisher et al., 2016). Low contributions of urea to DON (2 % and 8 %) in Tapi River, indicated urea was likely produced from the breakdown of DON in the natural rather than anthropogenic source; this is similar to what was reported in the Neuse River Estuary, USA (Twomey et al., 2005). Selemeni et al. (2017) reported a slightly higher percentage of urea to DON (8 to 16 %) measured in areas of low use of fertilizers in Pangani River Basin's ecosystem, Tanzania. While higher urea to DON was observed in intensive agricultural and urban development of Qu'Appelle lakes (up to 50 % of DON) in the central North America (Bogard et al., 2012). Low content of urea in Tapi River system was either due to low use of urea as fertilizers or urea decomposition. Bogard et al. (2012) reported 50–99 % of the total dissolved N pool in predominated forest basin was non-urea DON.

DIN loading to streams is directly related to the extent of agriculture in the catchment (Heggie and Savage, 2009). High concentrations of NO_3^- , making up a large fraction of DIN, in rivers can be primarily attributed to anthropogenic nutrient sources (Li et al., 2016; Yuan et al., 2012), particularly to runoff of fertilizers not utilized by target plants (Bu et al., 2011; Falco et al., 2010). More than 50 % of the applied N may have been transported to the river because of the low fertilizer uptake efficiency in tropical regions (Pradhan et al., 2015a, 2015b). In Thailand, water pollution with high NO_3^- derived from fertilizer runoff is more widespread with increasing population since the use of chemical fertilizers in Thailand started in 1961. The exponential increase in fertilizer use was reported in the 1970s. In fact, the fertilizer uses greatly increased >100 times from 18 thousand tons in 1961 to 2 million tons in 2004. In spite of this massive increase in chemical fertilizer use, the yield of rice and maize increased barely 1-time, indicating a tremendous loss of fertilizers into the environment due to their imbalance use and poor management (Tirado et al., 2008).

Meanwhile, the high NH_4^+ export to waterways is often related to human population density and anthropogenic activities in the watershed including sewage from industrial emission or leakage of manure and fertilizers from agricultural activities (Caraco and Cole, 1999; Du et al., 2017). Discharge of high levels of NH_4^+ from agricultural lands to rivers either via sewer systems or soil can cause severe pollution problems (Eryuruk et al., 2018). In addition, wastewaters from poultry and piggery (especially from farms with no water treatment systems) contain high concentrations of organic substances and NH_4^+ can become one of the major pollution sources and cause severe pollution problems (Kizito et al., 2015; Peng et al., 2020).

The high concentration of DIP, up to 2.39 μM , in mangrove creeks this study (Fig. 3e) is likely from intensive aquaculture pond and agriculture in the nearby areas. However, the DIP concentrations measured in our systems were lower to those from Nanliu (3.7 μM) and Lianzhou Rivers (5.5 μM) in China, where intensive shrimp ponds directly drained into the waterways (Kaiser et al., 2013). Pulatsu et al. (2004) suggested that intensive aquaculture system with supplementary feeds, fertilizers and metabolic wastes are the main source of inorganic phosphorus and particulate loads to surrounding environments. In addition, higher DIP can be accumulated in sediment aquaculture pond and made more available in the water column (Boyd, 2002). Excreta from fish and shrimp farming was also

expected to produce wastes characterized by large proportion of DIP into the water column (La Rosa et al., 2002; Mateka, 2015). Moreover, most DIP could be attributed to the application of phosphorus-containing fertilizers such as triple superphosphate. Kaiser et al. (2013) reported high DIP concentration is consistent with high fertilizer application to agriculture. In Thailand, there is an over-application of phosphorus fertilizer and low utilization efficiency of phosphorus by plants (>45 %) results in significant loss of applied phosphorus to aquatic systems (Tirado et al., 2008). Consequently, phosphorus loss from agriculture soil is now the most important sources of phosphorus to aquatic systems, and is the biggest contributor to aquatic ecosystem eutrophication (Chen et al., 2008).

Generally, river represents the major pathways of DSI from the natural sources in the surrounding area to the ocean (Gago et al., 2005). In Tapi River, DSI conservative decrease along the river suggests point source. Tapi River water has originated from the Nakhon Sri Thammarat and Phuket mountains, where mostly composed with quartzite, argillite, and siliceous slate (Brown et al., 1951). Weathering process is the mainly delivered DSI to the world oceans by rivers, which accounts for about 66 % of the world DSI input (Tréguer and De La Rocha, 2013). This process is constrained by the interaction of rock types, plate tectonic activities and climate (Liu et al., 2011).

Rain events can result in nutrient inputs derived from land to the river. In Surat Thani Province, approximately 70 % of annual precipitation occurs during wet season (June to January), and the average monthly rainfall is 139 mm. Liu et al., 2011 suggested that river discharges can be enhanced by rainfall and weathering rates are affected by precipitation and temperature, which can lead to higher nutrient concentrations during the wet season. Not surprised that dissolved inorganic nutrients in Tapi River are higher in wet season than in dry season (Fig. 3), suggesting that rainfall might be an important factor affecting nutrient supply to Bandon Bay.

4.2. Bay nutrient dynamics

Mixing along the salinity gradient shows that DSI, $\text{NO}_2^- + \text{NO}_3^-$, and DON are conservative while other dissolved nutrients (NH_4^+ , DIP, DOP) and some DON behave non-conservatively additional input/output processes within the bay along with the exchange with the GOT (Fig. 4).

Intensive shrimp farming pond was observed along the coastline of Bandon Bay (Jarernpornnipat et al., 2003). Pond aquaculture had been shown to be a major source of nutrient to estuarine systems (Wolanski et al., 2000; Herbeck et al., 2011). However, influence of shrimp pond effluents is probably low in Bandon Bay due to low exchange of shrimp pond water and low nutrients concentrations at the floodgate of shrimp pond (PP station). Yakupitiyage and Kaewner (2008) reported low nitrogen and phosphorus concentrations (10–25 %) exported from shrimp culture to Bandon Bay. Our data suggest that shrimp farming pond does not represent a major source of NH_4^+ , DIP, DOP, and DON.

Coastal around Bandon Bay consists of extensive mariculture includes the cultivation of blood cockles, oysters, and green mussels (Ratchatapattanukul et al., 2017). Li et al. (2016) reported that bivalve aquaculture and their activities such as shellfish harvesting were the major source of DIN and DIP with contributing for 64 % and 81 % of total influx to the bay, while bivalves also in turn become another source of nutrients through excretion in organic form (Magni and Montani, 2005). Large amount of organic matter (feces or pseudo-feces) accumulated on the seabed below farm in the intensive shellfish aquaculture (Mirto et al., 2000; Grant et al. (2012), and most feces or pseudo-feces are returned into water columns herein during harvesting (Li et al., 2016). In addition, excretion by bivalves can be a strong source of NH_4^+ and organic nutrients in coastal system (Tang et al., 2005). Dame et al. (1991) reported ammonia excretion by dense bivalve population was controlled influence on nitrogen concentrations in some coastal regions including the intensive mussel culture area in eastern shore of Nova Scotia (Strain, 2002). Hence, DON and DOP leaching from feces or pseudo-feces and NH_4^+ excretion form bivalve cultivation might be an important source of DON and DOP, and NH_4^+ in Bandon Bay.

A few changes of NH_4^+ even during tidal cycle (Fig. 5b) indicate that NH_4^+ is not only river and aquaculture activities but has another strong source within the bay. Maximum concentration in the inner bay, despite strong dilution of river discharge (Fig. 3d(i)), suggest a NH_4^+ in this area supported by groundwater seepage. Santos et al. (2021) and Alongi (2020) reported NH_4^+ was the commonly predominated measured in groundwater seepage, large NH_4^+ was the results of the POM and OM conversions to NH_4^+ by microbial process through the aerobic and anaerobic mineralization processes, while the dissimilatory nitrate reduction to ammonia process was also can directly converted nitrate back to ammonia (Decleure et al., 2015). It is not surprising that NH_4^+ dominates nitrogen fraction (99 % of DIN) in SGD observed in this study. Wang et al. (2021) suggested that muddy sediment served as a strong source of NH_4^+ to the estuarine waters in China. Similarly, Kristensen and Suraswadi (2002) posited that groundwater seepage was the main sources of dominated NH_4^+ in water of Bangrong mangrove forest in Thailand, and further suggested that SGD was the main sources of dissolved nutrients into the water column. To this end, SGD may be the potential sources of nutrients in Bandon Bay.

In addition, DIP was also higher and relatively constant in saline groundwater with salinity >10 to 30, while the relationship between concentration of DIP and salinity was unclear due to groundwater was affected by various factors such as groundwater contamination with DIP, effects of DIP adsorption-desorption equilibria at the ambient salinity, DIP remineralization by decomposing organic matter in coastal aquifers (Cho et al., 2018). In general, higher DOP was also observed in saline groundwater in the tidal beach, similarly the previous observed in the upper GOT by Burnett et al. (2007), revealing a significant input of DOP into receiving coastal water.

4.3. Water and nutrient contribution estimates into Bandon Bay

While we recognize that these flux calculations have large uncertainties and thus serve as rough estimates due to limited number of samples with only one discharge were used to estimate nutrient fluxes. However, the results are still compelling in terms of illustrating the significance of river, rain, and seepage groundwater inputs and their contribution (Table 3). To compare the different sources of nutrients to the bay, we quantified total daily inputs and their contribution from the river, wet atmospheric deposition, and SGD (Table 3). Other potential sources cannot be directly quantified by the methods used in this study. Mangrove potentially influence nutrient dynamics in estuaries (Kristensen and Suraswadi, 2002), but the small areas of mangrove forest remain in Bandon Bay due to excessive cutting of the mangrove forest along with uncontrolled massive development of mariculture (Jarernpornpipat et al., 2003). Kaiser et al. (2015) have shown that the mangrove nutrient uptake and release are unlikely to have a significant effect on estuarine water composition in the Nanlui Estuary due to the small area of mangrove forest remaining in this area. Currently, the input of domestic wastewater is probably less importance as most of the river catchment is dominated dense forest and agriculture with small urban and industrial areas. Shrimp pond effluent has been shown to be an important source of nutrient input to the coastal bay (Herbeck et al., 2011; Trott and Alongi, 2000). Nakorn et al. (2017) reported effluent from large scale shrimp pond in Bandon Bay were treated and re-used.

Tapi River is most important of the quantified nutrient sources to Bandon Bay. It supplies 76 % in wet season to 93 % in dry season of the quantified NO_3^- , while strong support of DON (>70 %) during wet season and no significantly differences during wet and dry season (76 % to 78 %) of NO_2^- inputs. Furthermore, it contributes half of the DSi (45 % to 50 %) input during wet and dry seasons. Consequently, the river remains the major sources of these nutrients into Bandon Bay. Nutrient fluxes from our study and those from other major rivers and seepage groundwater were shown in Table 5. The daily river export of inorganic (DSi, DIN, DIP) and organic nutrients (DON and DOP) were significantly lower than larger rivers that contributed large amount water to the upper GOT ($p < 0.01$) such as Chao Phraya River (Burnett et al., 2007) and all inorganic nutrients

calculated were also significantly lower than other larger Chinese river ($p < 0.01$) such as Danao River (Wang et al., 2017) and Daya River (Gao et al., 2018; Wang et al., 2018). While DIN and DIP exports were comparable to the smaller rivers in USA (Null et al., 2012; Dulai et al., 2016) and South Korea (Hwang et al., 2016).

This study estimated that average SGD flow ranged from 15 % to 45 % of the total discharge into Bandon Bay during dry and wet seasons. It has been reported that total discharge of SGD flowing into an estuary in Florida, USA ranged from 9 % to 80 % (Peterson et al., 2010). Makings et al. (2014) estimated that SGD represented 50 % of the total discharge flowing of the Caboolture River estuary in Australia. Moreover, our estimated the percentage of SGD discharge into Bandon Bay was significantly higher than Delaware River discharged into Delaware Bay (6 to 9 %) in New Jersey, USA (Schwartz, 2003). In this study, SGD showed to be the major sources of NH_4^+ , DIN, DIP, DOP and DSi into Bandon Bay in both seasons. It supplies 62 % to 93 % of the quantified NH_4^+ and 65 % to 84 % of DIN, approximately 41 % to 81 % of DIP, 49 % to 80 % of DOP, and contributes half of DSi input (45 % to 55 %) during study period. The large percentage of nitrogen and phosphorus contribution via SGD in this study was relatively higher than that reported in the upper GOT (70 % and 33 % of the total input), where strong influenced by large four main rivers with high nutrient loading from industrial and domestic sources (Burnett et al., 2007). However, the amount of DSi, DIN and DIP contributed by SGD were significantly lower than those found in the upper GOT conducted by Burnett et al. (2007), and China bays such as Daya Bay (Gao et al., 2018; Wang et al., 2018), and Maowei Bay (Chen et al., 2018), and China lagoons such as Hainan Island (Wang and Jinzhou, 2016). While these nutrients were comparable to the Geoje Bay, Korea (Hwang et al., 2016) and were also significantly higher than those observed in Kaneohe Bay, USA (Dulai et al., 2016). Groundwater usually has higher nutrient concentration than the receiving seawater and some of these concentrations also greater than those in river or streams (Rocha et al., 2022). Moore (1999) suggested the concentration of nitrogen, phosphorus, and silicate in SGD often higher than those in river water when compensating for the lower flux of groundwater relative to surface water, because groundwater nutrient concentration can be high compared to other potential sources, even a relatively small volumetric flux could provide significant nutrient subsidies (Slomp and Van Cappellen, 2004; Boehm et al., 2006; Wu et al., 2013).

Atmospheric deposition is an importance of nitrogen and phosphorus (Volk et al., 2012; Zheng et al., 2019). Rainwater is an important source of nutrients to the Bandon Bay, and the percentage of all nutrients contributed via rain were approximately 10 % of the total input, which was lower than the Middle Coastal Bays (14 %) in Maryland (Wells et al., 2002) and the Lianzhou Bay (25 %) in China (Kaiser et al., 2013), and these values must be regarded as an underestimate of atmospheric input. Volk et al. (2012) suggest that the wet deposition accounts for <50 % of the total atmospheric deposition, these values must be regarded as an underestimate of atmospheric input. In addition, concentration of nutrient in wet deposition was highly variable from one event to the next depending on frequency and amount of precipitation (Jung et al., 2011).

4.4. Organic nutrients supporting the primary production (PP) in Bandon Bay

DON and DOP entering coastal system through physical transport and coastal processes are characterized by significant terrestrial DON and DOP inputs. Phytoplankton and bacteria utilization of DON as alternative source of nitrogen has long been recognized (Admiraal et al., 1987; Bronk et al., 2007). Both of which are well known to use DON as nitrogen source (Veuger et al., 2004; Bronk et al., 2007). Since phytoplankton biomass is higher in the coastal regions, higher production of DON is expected (Sarma et al., 2013). Liu et al. (2011) found higher DON concentrations (14–26 μM) in the Hainan Island of China during highest discharge period. However, the concentration of DON in the coastal region and its possible advection to the open sea region depends on the quality of DON and biological activity (Sarma et al., 2019). Another source of terrestrial DON and DOP is SGD. Groundwater is generally transporting large amounts of

Table 5

Comparison between nutrient fluxes from our study and those from other major rivers and submarine groundwater.

Study site	Method	Nutrient fluxes ($\times 10^4$ mol/day)								Reference
		DSi	NO ₂ ⁻	NO ₃ ⁻	NH ₄ ⁺	DIN	DON	DIP	DOP	
River										
Chao Phraya River (wet), Thailand		42.6				225	101	18.9	5.57	Burnett et al. (2007)
Chao Phraya River (dry), Thailand		25.6				48.9	31.7	3.29	0.37	
San Francisco, USA						0.45		0.30		Null et al. (2012)
Kaneohe Watershed, USA		3.64–20.8				0.08–0.19		0.01–0.03		Dulai et al., 2016
Geoje River		3.91				1.61		0.01	05	Hwang et al. (2016)
Daya River						10.8		1.67		Gao et al. (2018)
DanAo River		10.8		3.34				3.65		Wang et al. (2017)
Daya River		11.8		3.84				3.65		Wang et al. (2018)
Tapi River (wet), Thailand		7.89	0.08	0.59	0.49	1.66	1.69	0.06	0.32	This study
Tapi River (dry), Thailand		1.92	0.01	0.12	0.05	0.18	0.67	0.01	0.01	This study
SGD										
Upper gulf of Thailand (wet)	meter*	64.5				48.3	38.4	10.4	1.69	Burnett et al. (2007)
Upper gulf of Thailand (dry)	meter*	11.3				11.9	10.7	2.33	0.49	
Kaneohe Bay, USA	Rn	1.62–5.72				0.05–0.13		0.001–0.02		Dulai et al., 2016
Hainan Island (LY lagoon), China	Ra	2510				5.87		1.60		Wang and Jinzhou (2016)
Hainan Island (XH lagoon), China	Ra	2560				157		8.5		
Geoje Bay, Korea	Rn	23.8				8.18		0.12		Hwang et al. (2016)
Tropical Bay, China	Rn	940				450		5.3		Chen et al. (2018)
Daya Bay						195–206		5.72–6.04		Gao et al. (2018)
Daya Bay, China	meter*	35.4–113		10.5–19.9				0.40–1.22		Wang et al. (2018)
Bandon Bay (wet), Thailand	meter*	5.51	0.02	0.04	3.0	4.88	0.4	0.12	0.50	This study
Bandon Bay (dry), Thailand	meter*	2.43	0.03	0.06	1.21	1.22	0.4	0.05	0.06	This study

Note: *seepage meter.

nitrogen and phosphorus into the coastal water due to the natural processes and anthropogenic inputs include weathering and leaching of soils, fertilizers, and seepage of wastes. Wattayakorn et al. (2004) found that approximately 40 % of the coastal organic nutrients were supported via SGD.

The PP in surface water ranged from 435 to 555 mg-C m⁻² day⁻¹ in the study region despite of low DIN concentrations (<5 μM). In fact, PP in this study is within in the average ranges of the previous reported study using a ¹³C-based method (435 to 555 mg-C m⁻² day⁻¹) by Yoshikawa et al. (2017) and are considered moderate compared to the various reported in the estuaries worldwide 52 to 1499 mg-C m⁻² day⁻¹ (Boynton et al., 1982). The Chl-a in displayed significant correlation with DON ($r^2 = 0.77$; $p < 0.01$) in the river. On the other hand, DOP and DON displayed significant correlations ($r^2 = 0.75$; $p < 0.01$ and $r^2 = 0.68$) with Chl -a in the bay, where DIP (<1 μM) and DIN (<5 μM) were low (Figs. 3e(i) and 3e(ii)). Based on the ratio the change in concentrations of nutrients in the Western Atlantic (N: P: Si = 16: 1: 15) (Redfield et al., 1963), the primary limiting nutrient for PP was likely to be nitrogen during the study period (4: 1: 6 in the wet season and 12: 1: 11 in the dry season). Burnett et al. (2007) reported the nitrogen-limited conditions were generally observed in the GOT.

The significant relation of the Chl-a with DOP and DON suggest that DON and DOP are transported into the bay through transfer by tidal exchange or generated within the bay. This pathway is more clearly revealed in this study with higher concentrations of DON and DOP during low tide (Fig. 5b) and these concentrations continued increasing with increasing salinity (Fig. 4), indicate possible source from both river waters, submarine groundwater, and exchange with the open sea. In contrast, low DIN and DIP concentrations that were observed in the bay region even though enriched DIN and DIP supported via SGD suggest the occurrence of low DIN and DIP concentrations in the bay regions, where intrusion of seawater from the GOT during high tide. De Galan et al. (2004) reported that DON contributes up to 50 % of TDN in the coastal zone, while it is up to 90 % in the open ocean region (Mahaffey et al., 2004; Torres-Valdés et al., 2009). Therefore, sources from river water or SGD or GOT seawater intrusion are possible. This pathway is more clearly revealed in this study with less change in concentrations of DON and DOP throughout mixing with the GOT seawater during high tide. This study suggests that organic nutrients both from river water and SGD are strong significantly supporting PP in Bandon Bay.

In addition to this, DON is produced in situ through biological processes in the aquatic systems.

Pujo-Pay et al. (1997) reported that DON can be provided by phytoplankton during its metabolism, cell death and lysis. Collos et al. (1992) reported that DON releases from phytoplankton (diatom) and its consequent uptake by dividing cells during the dark incubation, which could provide a competitive advantage over co-occurring primary producers that would not be able to take up these compounds.

5. Conclusion

To the best of our knowledge, this work represents the first comprehensive biogeochemical observations on nutrient dynamics in Bandon Bay and their input fluxes via various sources (river runoff, SGD, and rainwater) into the bay. Our study confirms the importance of river runoff, SGD, and rainwater for the nutrient delivery to coastal water, thereby supporting the PP in Bandon Bay. Complex biochemical systems and seasonal fluctuation causes nutrient concentrations in Tapi-Phumduang River system, SGD, rainwater, and bay water showed a large range of variation. However, all nutrient concentrations in these areas are generally at the levels of average global conditions and do not exceed Thai water quality standard. River runoff and SGD liberate new nutrients into the Bandon Bay and considered as the two main sources of nutrients discharged into Bandon Bay. Urbanization with intensive agriculture and little industrial activities are the main sources of nutrient pollution from river system, which may cause negative impacts on quantity/quality of coastal water and productivity therein. Intensive shrimp pond surrounding the bay may not be strong direct nutrient sources, but shrimp pond expansion has direct negative consequences on the density and distribution of mangrove areas. This can hamper the mangrove's ecosystem services and reduce their function such as filters for anthropogenic nutrient fluxes through nutrient uptake. Mariculture and their harvest activity are also other sources of new nutrient via the nutrient resuspension during the harvesting process and animal excretion. In addition, heavy coastal weathering, and erosion during strong monsoon season and macro tidal may influence of the nutrient concentration and distribution in Bandon Bay. Nutrient sources in Bandon Bay were primarily from riverine input and SGD, followed by rainwater. The nutrient exported to the bay can be influenced by several meteorological factors including

rainfall and the timing of monsoon. High nutrient load into the coastal environment can result in nutrient imbalance and effect phytoplankton production and composition, and eventually cause water pollution. Although there are many sources of water and nutrients released into Bandon Bay, nutrient levels in the bay were not significant differences between seasons. This can be attributed to the fact that nutrients released from various sources are quickly removed during estuarine mixing and taken up by phytoplankton. High PP in Bandon Bay plays an important role in keeping nutrients at low levels and maintaining relatively good water quality.

CRedit authorship contribution statement

N.C., J.Z., and P.S. conceived the original idea. N.C., J.Z., P.S., W.Y., C.C., and J.D. designed the study. N.C., J.Z., P.S., W.Y., and C.C. carried out the field sampling. N.C. conducted the lab analyses with the help from J.Z., P.S., and S.J. for the analytical techniques. N.C., J.Z., P.S., W.Y., C.C., and J.D. conducted the data analyses and prepared the visualization and data presentation. The original draft was prepared by N.C. in consultation with J.Z., P.S., W.Y., and C.C. and all authors discussed the results and contributed to the final manuscript.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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