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Sources and export of nutrients associated with integrated multi-trophic aquaculture in Sanggou Bay, China

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ABSTRACT: Field observations were made from 2012 to 2014 at an integrated multi-trophic aquaculture (IMTA) site in Sanggou Bay (SGB), China, to characterize the nutrients associated with aquaculture activities, and to assess the effects of aquaculture on nutrient cycles in the bay. Dissolved inorganic and organic nutrient levels were measured in rivers, groundwater, and SGB. Seasonal variations in nutrient concentrations were detected in the rivers, particularly enrichment of dissolved inorganic nitrogen (DIN) and silicate (DSi). Nutrient concentrations showed considerable seasonal variation, with higher and significantly different concentrations occurring in autumn than in the other seasons. The composition and distribution of nutrients were also affected by the species being cultured. Dissolved organic nitrogen and phosphorus (DON and DOP) accounted for 27 to 87% of total dissolved nitrogen and 34 to 81% of total dissolved phosphorus, respectively. Phosphorus may be a potentially limiting nutrient for phytoplankton growth in summer. Nutrient budgets were developed based on a simple steady-state box model. These showed that bivalve aquaculture was the major source of PO_4^{3-} (contributing 64% of total influx) and led to increased riverine fluxes of PO_4^{3-}. The results indicated that substantial quantities of nitrogen and DSi accumulated in sediments or were transformed into other forms (e.g. phytoplankton cell composition or particles). Large quantities of DIN and PO_4^{3-} were removed from the bay through harvesting of seaweeds and bivalves, which represented up to 64 and 81% of total outflux, respectively. The results show that aquaculture activities play the most important role in nutrient cycling in SGB.

KEY WORDS: Nutrients · IMTA · Budgets · Aquaculture activities · Sanggou Bay

INTRODUCTION

With an annual average increase of 8.7% over the past 40 yr, aquaculture is the fastest-growing food production sector in the world, and is overtaking capture fisheries as a source of food fish (Herbeck et al. 2013). The rapid growth of aquaculture has given rise to a wide variety of environmental problems, including ecosystem degradation and water pollution (Neori et al. 2004). One of the largest of impacts of aquaculture effluents to local ecosystems is imbalance created in nutrient dynamics and eutrophic

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conditions (Marinho-Soriano et al. 2009, Bouwman et al. 2011). In addition, excess nutrients cause stress in the cultivated organisms, with deleterious effects including smaller size, reduced production, and mass mortality (Newell 2004, Mao et al. 2006). Due to increasing concerns about the environmental impacts of aquaculture, a new method of aquaculture with a smaller ecological footprint has been developed. Integrated multi-trophic aquaculture (IMTA) has the potential to mitigate the environmental impacts of aquaculture (Buschmann et al. 2008).

IMTA is described as the cultivation of aquatic species from different trophic levels within a shared water system (Bostock et al. 2010). Such systems significantly increase the sustainability of aquaculture and recycle waste nutrients from high trophic-level species into production of lower trophic-level crops of commercial value (Troell et al. 2009). Seaweeds are used in IMTA systems for their nutrient-absorbing and sequestering properties. Nutrients excreted and egested by bivalves can be absorbed by macroalgae and recycled into valuable biomass (Newell 2004, Buschmann et al. 2008), and this amount of nutrient waste can be effectively removed from the ecosystem. In addition, a number of studies have confirmed that suspension-feeding bivalves can exert top-down control on phytoplankton (Newell & Koch 2004, Wall et al. 2008); larger nanoplankton will be removed in comparison with smaller (<3 µm diameter) picoplankton species, thereby reducing turbidity (Newell 2004). The resulting increased light penetration can potentially enhance the production of benthic plants (Newell & Koch 2004). If high levels of dissolved inorganic nitrogen (DIN) regenerated by bivalves are sufficient to allow the relatively slow-growing nanoplankton to grow fast enough to overcome grazer control, primary production can be stimulated through recycling of nitrogen (Smaal et al. 2001). Some marine IMTA systems have been commercially successful at industrial scales, especially in Asia (China) (Troell et al. 2009).

China is the largest aquaculture producer in the world, with a total production of 34.1 million tons, which accounts for 62% of total global production and 51% of the global value (Yang et al. 2005, FAO 2010, Yuan et al. 2010, Yu et al. 2012). The area devoted to aquaculture increased from 11.2×10^4 ha in 1977 to 218×10^4 ha in 2012 (The People's Republic of China Ministry of Agriculture Fisheries Bureau 2013). The rapid growth of aquaculture has led to eutrophication of coastal waters (Wu et al. 2014), and to the occurrence of aquatic diseases that have resulted in major economic losses (Fei 2004); for example, in 1998, more than 10 billion Chinese Yuan (approximately US$ 1.5 billion) were lost because of mariculture disease (Fei 2004). To improve the environmental sustainability of aquaculture and benefit the local economy, IMTA was developed in China. Sea-ranching and suspended aquaculture are the 2 main forms of IMTA in China, and the latter is used in Sanggou Bay.

Sanggou Bay (SGB) is located in northern China and has been used for aquaculture for over 30 yr (Zhang et al. 2009). It has been estimated that more than 300 t of inorganic nitrogen have been excreted into the bay by cultivated and fouling animals (Troell et al. 2009). Studies of core sediments also indicated that the total nitrogen (TN) content has increased in recent decades as a consequence of aquaculture activities (Song et al. 2012). Bivalves clear seston particles >3 µm in diameter from natural water and are not supplied with additional feed in the bay. The absolute and relative abundances of dinoflagellate cells in the bay are lower inside the scallop culture area than outside (Zhang et al. 2005), and the phytoplankton community has changed as a result; meanwhile, the reduction in phytoplankton biomass has a negative impact on bivalve growth (Duarte et al. 2003, Shi et al. 2011a). In addition, kelp can compete with phytoplankton for nutrients, and 80 000 t of dried kelp can be produced annually through uptake of inorganic nitrogen from the bay (Zhang et al. 2009). In pursuing high levels of productivity, SGB has been subject to a rapid growth in aquaculture, with long-line culture of kelp having expanded to areas more than 8 km away from the coast, where the water depth is between 20 and 30 m (Troell et al. 2009, Fu et al. 2013).

Much attention has been focused on the carrying capacity of shellfish and kelp mariculture (Bacher et al. 2003, Nunes et al. 2003, Shi et al. 2011a), ecology (Song et al. 2007, Hao et al. 2012), nutrient levels (Wang 2012, Zhang et al. 2012), and nutrient fluxes at the sediment–water interface (Jiang et al. 2007, Sun et al. 2010) in SGB, but the effects of aquaculture activities on nutrient cycling have not been well studied in the bay. The objective of this study was to determine the amounts and composition of dissolved nutrients in the bay and associated rivers and groundwater, to assess the sources and transportation of nutrients, to evaluate the impact of aquaculture activities on nutrient cycling, and to discriminate the importance of internal nutrient inputs vs. physical transport, based on the land–ocean interactions in the coastal zone (LOICZ) nutrient model (Gordon et al. 1996).

MATERIALS AND METHODS

Study area

SGB (Fig. 1) is a semi-enclosed water body of approximately 144 km^2 at the eastern end of Shandong Peninsula, and has an average depth of 7.5 m (Zhang et al. 2009). The bay is characterized by semi-diurnal tides having an average tidal range of 2 m, and is connected to the Yellow Sea through an 11.5 km wide channel (Mao et al. 2006, Jiang et al. 2007). It is dominated by land–ocean climate, with water temperatures ranging from 2 to 26°C (Kuang et al. 1996). Approximately 73.3% of annual precipitation in the area (819.6 mm) occurs during the wet season, from June to September. The average river discharge into the bay is 1.7–2.3 × 10^8 m^3 yr^{-1}, and this carries an annual sediment load of 17.1 × 10^4 t.

More than 70% of the area of SGB is currently used for aquaculture (Zhang et al. 2009, 2010, Fu et al. 2013). It is one of the largest aquaculture production sites in China, and is extensively used for the culture of scallops (*Chlamys farreri*), Pacific oyster *Crassostrea gigas*, and seaweeds (*Saccharina japonica* and *Gracilaria lemaneiformis*) (Zhang et al. 2009). These species are grown in both monoculture and polyculture, from suspended longlines (Fang et al. 1996a) (Fig. 1). *S. japonica* monoculture occurs mainly near the mouth of the bay, bivalves are mainly cultured in the western part of the bay, and kelp and bivalve polyculture occurs in the middle part of the bay (Fig. 1). The co-cultivation of abalone *Haliotis discus hannai* with kelp (*S. japonica*) has also been developed, with the abalones held in lantern nets hanging vertically from the longlines. In 2012, production included approximately 84 500 t dry weight of *S. japonica*, 25 410 t wet weight of *G. lemaneiformis*, and approximately 15 000 and 60 000 t wet weight of *C. farreri* and *C. gigas*, respectively (data from Rongcheng Fishery Technology Extension Station). The main cultured species has shifted from scallop to oyster since 1996 because of reduced scallop production as a consequence of disease (Zhang et al. 2009).

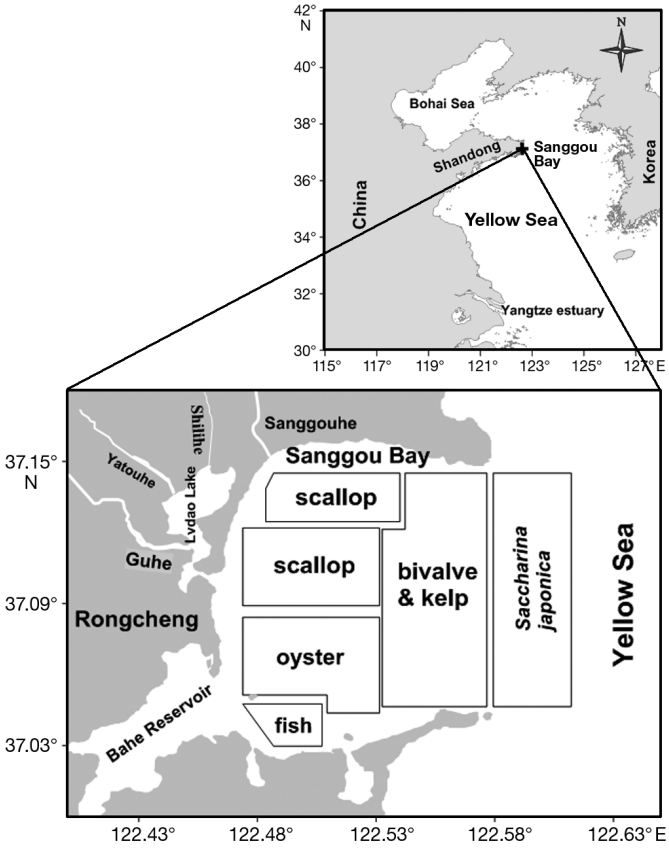


Fig. 1. Location of Sanggou Bay, China, and aquaculture activities, showing the regions of kelp (*Saccharina japonica*) monoculture; scallop, oyster, and fish monoculture; and multi-species aquaculture

To increase production, aquaculture has expanded from the bay to the open sea since the 1990s (Fang et al. 1996a). However, the total aquaculture production of kelp has not increased (Shi et al. 2011a). This may be related to a reduced supply of nutrients resulting from a decrease in the water exchange rate, which has been a consequence of reduced circulation because of the increase in aquaculture activities (Fang et al. 1996b). The hydrodynamic conditions have changed significantly because of the presence of suspended aquaculture (Shi et al. 2011a). Current speeds can be reduced by aquaculture facilities including rafts, and ropes impose drag (Grant & Bacher 2001, Duarte et al. 2003). The renewal of suspended particles for bivalve culture and nutrient regeneration for kelp have also been reduced (Grant & Bacher 2001, Duarte et al. 2003). Compared with the period of farming activities up to 1983, tidal currents had decreased by 50% by 1994 because of large-scale cultivation (Zhao et al. 1996). Based on a 2-dimensional model, Grant & Bacher (2001) estimated a reduction of 41% in the water exchange rate in SGB because of increased bottom friction with expansion of intensive suspended aquaculture. The vertical current has also changed because of suspended aquaculture (Fan & Wei 2010).

Sample collection

Sampling took place during 31 May to 4 June 2012 (early summer), 20 September to 2 October

2012 (early autumn), 22 to 25 April 2013 (spring), 21 to 25 July 2013 (summer), 16 to 17 October 2013 (autumn), and 15 to 17 January 2014 (winter) (Fig. 2). Two anchor stations for monitoring over complete tidal cycles of 25 h were established, one in April 2013 in the northern mouth of the bay (D1), and the other in October 2013 in the southern mouth (D2) (Fig. 2), respectively. At each station, surface water samples were collected by submersing a 1 l acid-cleaned polyethylene bottle from a boat, and bottom water samples were collected using a 5 l polymethyl methacrylate water sampler. River water samples were collected from the river edge in 0.5 l acid-cleaned polyethylene bottles, and groundwater was collected from wells around the bay (Fig. 2).

Water temperature and salinity were measured *in situ* using a WTW MultiLine F/Set3 multi-parameter probe. Each water sample was immediately filtered through a 0.45 µm pore size cellulose acetate filters (pre-cleaned with hydrochloric acid, pH = 2) into a polyethylene bottle that had previously been rinsed 3 times with some of the filtered water sample. The filtrates were fixed by the addition of saturated $HgCl_2$ solution (Liu et al. 2005), and the filters were dried at 45°C and weighed to determine the mass of suspended particulate matter (SPM).

Chemical analysis

Dissolved nutrient concentrations were measured in the laboratory using an Auto Analyzer 3 (Seal Analytical). Total dissolved nitrogen (TDN) and total dissolved phosphorus (TDP) were measured according to the methods of Grasshoff et al. (1999). The DIN

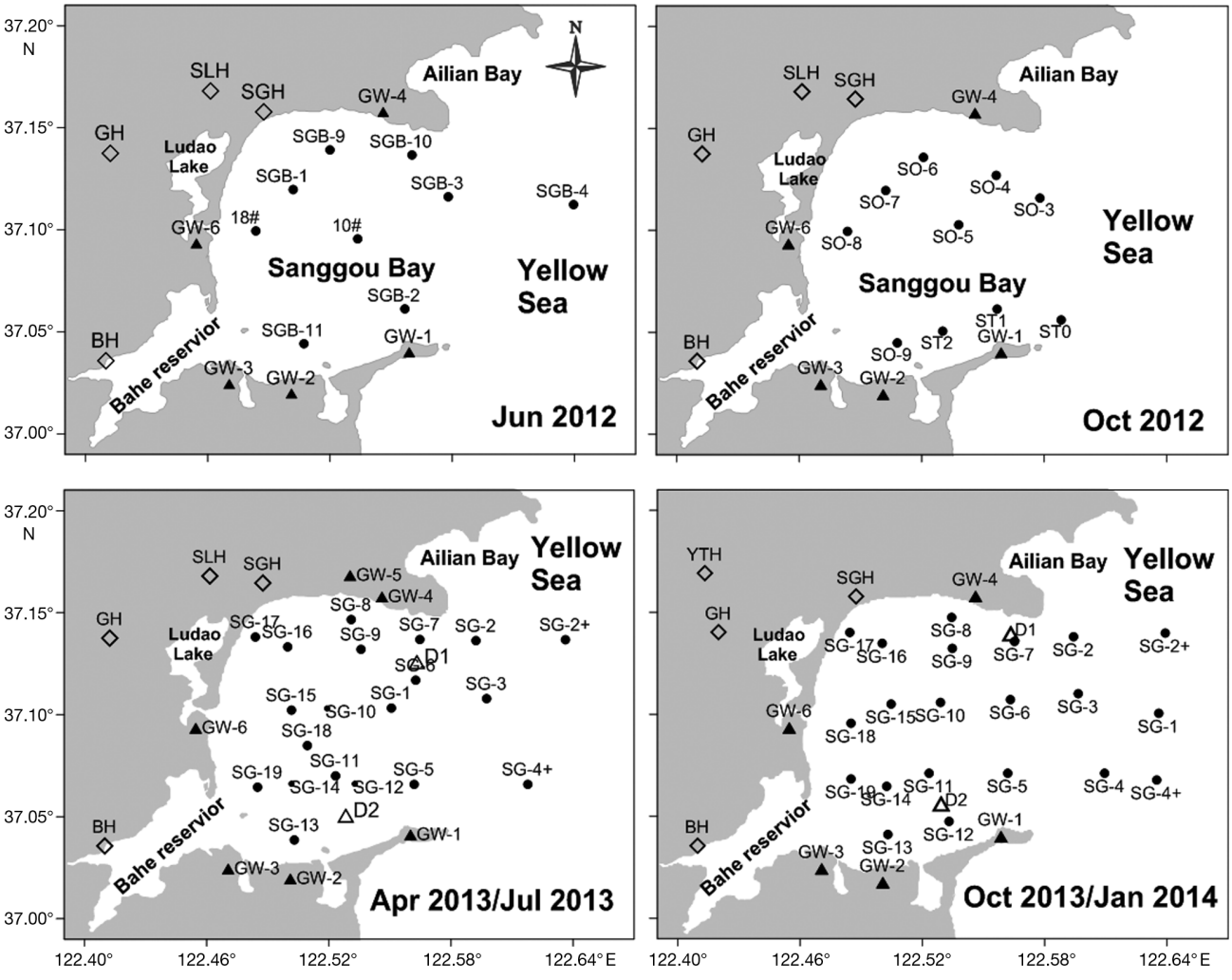


Fig. 2. Sampling stations in Sanggou Bay for the cruises during 2012 to 2014. (◇) River stations; (▲) groundwater stations (BH: Bahe; GH: Guhe; SLH: Shilihe; SGH: Sanggouhe; YTH: Yatouhe); (●) bay stations; (△) anchor stations

concentration was determined as the sum of the NO_3^-, NO_2^-, and NH_4^+ concentrations. The concentrations of dissolved organic nitrogen (DON) and dissolved organic phosphorus (DOP) were estimated by subtracting DIN from TDN and PO_4^{3-} from TDP, respectively. The analytical precision of NO_3^-, NO_2^-, NH_4^+, PO_4^{3-}, dissolved silicate (DSi), TDN, and TDP was <5%.

Statistical analysis

Statistical analyses were performed using the software SPSS 20.0 by IBM. One-way ANOVAs were used to analyze the individual effects of seasons and particular cultivation area on variations in SPM, and 2-way ANOVAs were used to analyze the combined effects of seasons and cultivation area on variations in SPM. Two-way ANOVAs were also used to analyze the effects of surface/bottom and seasons on variations in nutrient concentrations. Based on *a posteriori* homogeneity tests, Tukey's HSD or Tamhane's T2 comparisons were applied to assess the statistical significance of differences ($p < 0.05$) following ANOVA.

Nutrient budgets

Dissolved nutrient budgets for the study system were constructed based on the LOICZ box model (Gordon et al. 1996). This model has been widely used to construct nutrient budgets defining the internal biogeochemical processes and external nutrient inputs of estuarine and coastal ecosystems (Savchuk 2005, Liu et al. 2009). For our model, we assumed that the study system was in a steady state, and the bay was treated as a single well-mixed box. The water mass balance, salinity balance, and the nonconservative fluxes of nutrient elements based on nutrient concentrations and water budgets were estimated according to Eqs. (1) to (3), respectively:

$$V_R = V_{in} - V_{out} = -V_Q - V_P - V_G - V_W + V_E \quad (1)$$

$$V_X(S_1 - S_2) = S_R V_R \quad (2)$$

$$\Delta Y = \text{outflux} - \text{influx} = V_R C_R + V_X C_X - V_Q C_Q - V_P C_P - V_G C_G - V_W C_W \quad (3)$$

where V_R is the residual flow, and V_Q, V_P, V_G, V_W, V_E, V_{in}, V_{out}, V_X, and ΔY are the river discharge, precipitation, groundwater, wastewater, evaporation, inflow of water to the system of interest, outflow of water from the system of interest, the mixing flow between the 2 systems and nonconservative flux of nutrients, respectively. The volume of aquaculture effluent discharged directly into the system of interest was not considered, as the data were limited. We assumed that the salinity of fresh water (V_Q, V_P, and V_E) was 0. In Eq. (2), $S_R = (S_1+S_2)/2$, where S_1 and S_2 are the average salinity of the system of interest and the adjacent system, respectively. The total water exchange time (τ) of the system of interest was estimated from the ratio of V_S to ($V_R + V_X$), where V_S is the volume of the system. In Eq. (3), C_Q, C_P, C_G, C_W, C_R, and C_X are the average concentrations of nutrients in the river discharge, the precipitation, groundwater, wastewater, the residual flow, and the mixing flow, respectively. C_R and C_X equate to $(C_1 + C_2)/2$ and $(C_1 - C_2)$, respectively. C_1 and C_2 are the average concentrations of nutrients in the system of interest and the adjacent system, respectively. Outflux and influx are the total nutrient flux out of and into the system of interest, respectively. A negative or positive sign for ΔY indicates that the system of interest was a sink or a source, respectively.

RESULTS

Hydrographical chacteristics

The surface water temperature (Table 1) reflected the seasonality of this temperate system. The surface water temperature decreased from the mouth to the

Table 1. Seasonal variations in temperature, salinity, and suspended particulate matter (SPM) in Sanggou Bay, China, during the study. Mean values are given in parentheses

Season	Temperature (°C)		Salinity		SPM (mg l^{-1})	
	Surface	Bottom	Surface	Bottom	Surface	Bottom
Spring	6.00–9.60 (7.60)	6.10–9.90 (7.80)	30.2–31.3 (30.8)	30.1–31.4 (30.7)	3.91–31.9 (13.6)	3.59–40.5 (14.9)
Summer	13.3–25.9 (20.0)	13.5–20.6 (17.0)	28.2–30.8 (30.0)	30.2–30.7 (30.4)	3.78–26.4 (13.9)	5.61–92.0 (37.6)
Autumn	17.7–25.0 (20.1)	16.6–23.3 (19.3)	29.1–30.0 (29.6)	29.3–29.9 (29.5)	5.75–29.3 (15.5)	11.9–67.8 (27.4)
Winter	1.80–5.70 (3.50)	0.90–5.30 (3.15)	29.2–30.6 (30.0)	29.2–30.4 (29.9)	2.27–54.0 (15.8)	3.04–54.7 (13.5)

west of the bay in spring and summer, but increased in this direction in autumn and winter. The horizontal distribution of temperature in the near-bottom layer was similar to that in surface water, but the temperatures were generally lower. The salinity of both surface and bottom water gradually increased from the west of the bay to mouth, except in winter. The salinity was lowest in autumn (Table 1).

The SPM concentrations varied considerably among seasons and cultivation areas, as evidenced by the large ranges shown in Table 1 and Fig. 3. The average concentration of SPM showed minor differences between surface and bottom waters in spring and winter, but was significantly less in surface water than in the bottom layer in both summer and autumn between different cultivation areas, especially those involving oyster and scallop monoculture (Fig. 3). A 1-way ANOVA indicated very significant differences in SPM concentration in bottom water of the bay in different seasons ($p < 0.05$). The subsequent post hoc Tamhane's T2 test showed that the concentrations of SPM in bottom water in summer and autumn differed significantly from those in spring and winter. In addition, a 1-way ANOVA indicated highly significant differences between different cultivation areas ($p < 0.05$). The subsequent post hoc Tamhane's T2 test showed that the values of SPM in both bottom and surface waters in the fish, oyster, and scallop cultivation areas differed significantly from those in the kelp, offshore, and bivalve and kelp areas.

Nutrients in rivers

Nutrient concentrations in rivers adjacent to SGB varied greatly during the study period (Table 2). The rivers were generally enriched with DIN relative to PO_4^{3-} (Table 2). The DIN was dominated by NO_3^-, which accounted for 73 to 98% of DIN among all seasons. The NO_2^- concentrations in rivers were generally >2 µM except Bahe river (0.14–1.13 µM; Table 2). The PO_4^{3-} concentration ranged from 0.08 to 6.02 µM in the rivers, with an annual average of 1.45 µM. Seasonal variation of PO_4^{3-} in the Bahe river was similar to that in the Guhe river, and the PO_4^{3-} concentrations in the Bahe and Guhe rivers were lower than in the Shilihe and Sanggouhe rivers (Table 2). The DSi concentrations were high in our study rivers (average 182 µM; Table 2), indicating a high weathering rate associated with rivers adjacent

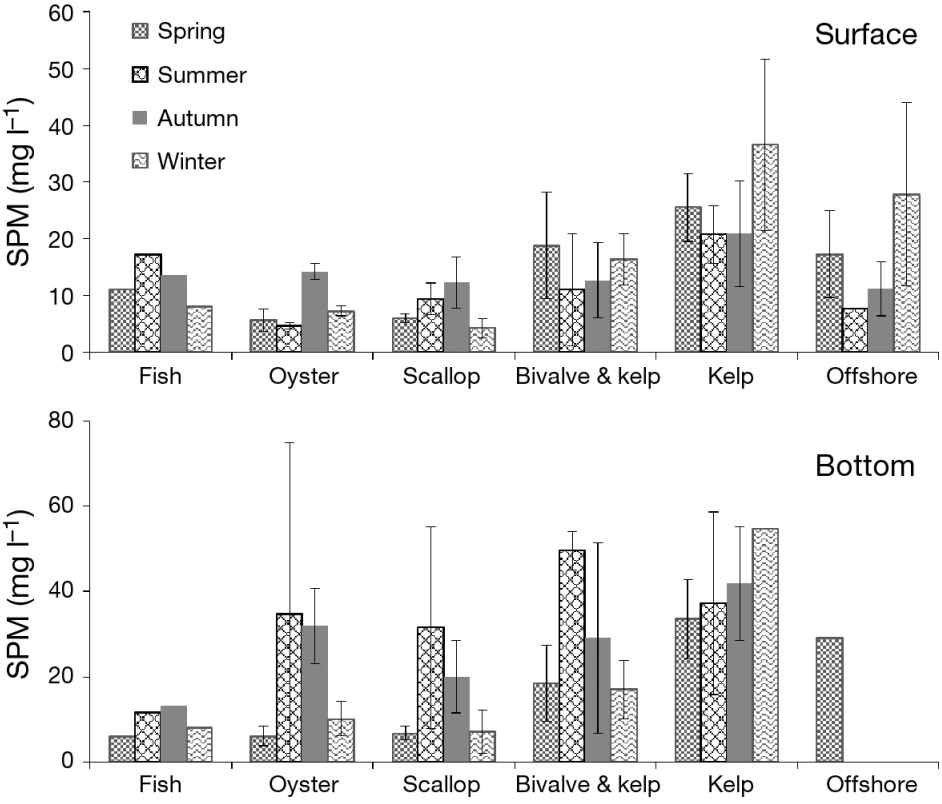


Fig. 3. Suspended particulate matter (SPM) concentrations (mg l^{-1}; mean ± SD) in various cultivation areas in different seasons during the study periods

Table 2. Nutrient concentrations (µM) and molar ratios in surface water in rivers adjacent to Sanggou Bay, China, in different seasons during the study. DSi: dissolved silicate. Dates are given as year-month

River	NH_4^+	NO_2^-	NO_3^-	PO_4^{3-}	DSi	N:P	Si:N
Bahe							
2012-06	2.53	0.15	0.48	0.17	29.0	18	9.0
2012-09	0.65	0.04	0.25	0.17	164	5.5	176
2013-04	2.91	0.32	28.9	2.23	47.9	14	0.4
2013-07	0.65	1.13	88.6	0.32	90.4	21	1.0
2013-10	1.41	0.15	10.5	0.56	246	21	20
2014-01	1.68	0.12	18.5	1.78	142	11	7.0
Guhe							
2012-06	36.3	17.5	309	0.19	72.3	1905	0.2
2012-09	9.86	10.4	530	0.08	102	7157	0.2
2013-04	22.5	5.18	288	1.80	55.6	175	0.2
2013-07	2.56	7.60	590	0.16	208	122	0.3
2013-10	12.6	6.40	240	–	143	–	0.6
2014-01	5.71	3.05	455	0.50	130	927	0.3
Shilihe							
2012-06	103	4.38	283	3.23	364	121	0.9
2012-09	17.8	6.70	600	0.22	282	2821	0.5
2013-04	93.0	17.0	503	2.89	243	212	0.4
2013-07	30.3	10.2	199	3.60	199	63	0.3
Sanggouhe							
2012-06	15.4	2.26	169	0.12	172	1505	0.9
2012-09	4.00	17.5	508	2.60	182	204	0.3
2013-04	8.23	10.2	351	1.44	166	256	0.5
2013-07	25.0	16.6	382	6.02	382	106	0.4
2013-10	2.73	4.40	362	–	318	–	0.9
2014-01	7.25	5.65	569	2.00	236	291	0.4
Yatouhe							
2013-10	4.68	3.20	420	–	189	–	–
2014-01	6.50	4.36	687	0.36	211	–	–

to the SGB. Except for Bahe river, the DIN:PO_4^{3-} molar ratios in the rivers were significantly higher than the Redfield ratio (Table 2), indicating that phytoplankton might be limited by phosphorus despite high NO_3^- values, especially in summer in the Bahe and Guhe rivers. The high concentrations of DIN led to DSi:DIN ratios that were less than or approached a value of 1.

Spatial and temporal variations of nutrients in SGB

The concentrations of dissolved inorganic nutrients decreased gradually from offshore to the inner part of SGB in spring (April 2013; Fig. 4a), while the DON and DOP concentrations showed the opposite horizontal distribution (Fig. 4a). The concentrations of NO_3^- accounted for 53–92% and 56–89% of the DIN in surface and near-bottom layers, respectively. DON contributed 27–46% of TDN in surface water outside the bay, where kelp monoculture occurs, and accounted for 46–87% of TDN inside of the bay. DON represented 40–84% of TDN in the near-bottom layer. For phosphorus compounds, PO_4^{3-} and DOP accounted for approximately 66 and 34% of TDP in the bay, respectively. The molar ratios of DIN:PO_4^{3-} ranged from 7.8 to 31 (average 19 ± 7.9 SD) in surface water, and from 9.4 to 69 in the near-bottom layer, respectively. The average DSi:DIN ratio was higher than the Redfield ratio in both surface (1.3 ± 0.8) and bottom (1.2 ± 0.6) waters. Studies of nutrient uptake kinetics have shown that the threshold values for phytoplankton growth are 1.0 µM DIN and 0.1 µM PO_4^{3-} (Justi et al. 1995). In the western part of the bay, DIP concentrations were lower than the threshold values for phytoplankton growth (Fig. 4a). This suggests that phosphorus may be the most limiting element for phytoplankton growth in the following season.

During June 2012 (Fig. 4b), the levels of dissolved inorganic nutrients were lower than those in spring (Fig. 4a). The NO_3^-, NO_2^-, and NH_4^+ concentrations decreased gradually from offshore to the inner part of the bay, while PO_4^{3-} and DSi concentrations showed the opposite horizontal distribution. With respect to nitrogen compounds, NO_3^- comprised 24–78% of DIN in surface water and 34–72% in bottom water. Surface water was depleted in PO_4^{3-} (0.03–0.17 µM), which led to the DIN:PO_4^{3-} ratios being significantly higher than the Redfield ratio. The DIN:DSi molar ratios ranged from 0.4 to 3.2 (average 1.6 ± 0.7). In July 2013, nutrient concentrations increased significantly from the mouth of the bay to the inner part (Fig. 4c), and were higher in the near-bottom layer than in surface water. The DIN was dominated by NH_4^+, which contributed 32–89% (mean 62%) and 32–69% (mean 52%) to DIN in surface water and the near-bottom layer, respectively. DON comprised 57–88% of the TDN in the entire bay, and DOP accounted for 34–75% and 46–81% of the TDP in surface water and the near-bottom layer, respectively. The molar ratios of DIN:PO_4^{3-} were higher than the Redfield ratio in surface water, and the DSi:DIN ratios were higher than or comparable to the Redfield ratio. The PO_4^{3-} concentrations in surface water at 70% of the stations in June 2012 (Fig. 4b), and in the southeastern part of the bay in July 2013 (Fig. 4c), were lower than the threshold values. This suggested that phytoplankton growth might be limited by P in summer. In the western part of the bay (the main area for bivalve culture) the DIN concentrations were lower than or comparable to the threshold values, suggesting that N might be potentially limiting for phytoplankton growth in this part of the bay.

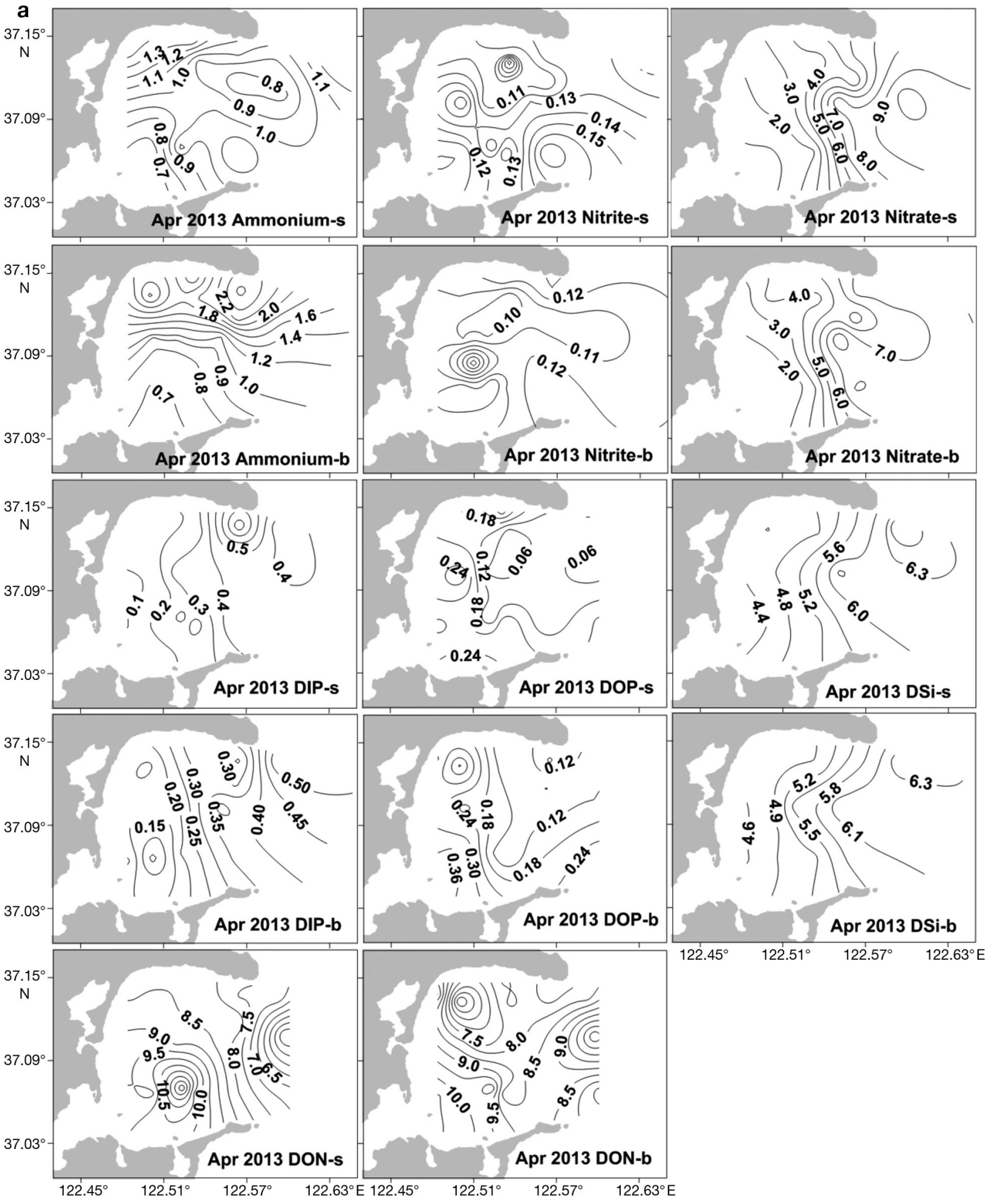


Fig. 4. Horizontal distributions of nutrients (µM) in Sanggou Bay: (a) April 2013; (b) June 2012; (c) July 2013; (d) October 2012; (e) October 2013; (f) January 2014. DIP (DOP): dissolved inorganic (organic) phosphorus, DSi: dissolved silicate, DON: dissolved organic nitrogen. s: surface; b: bottom

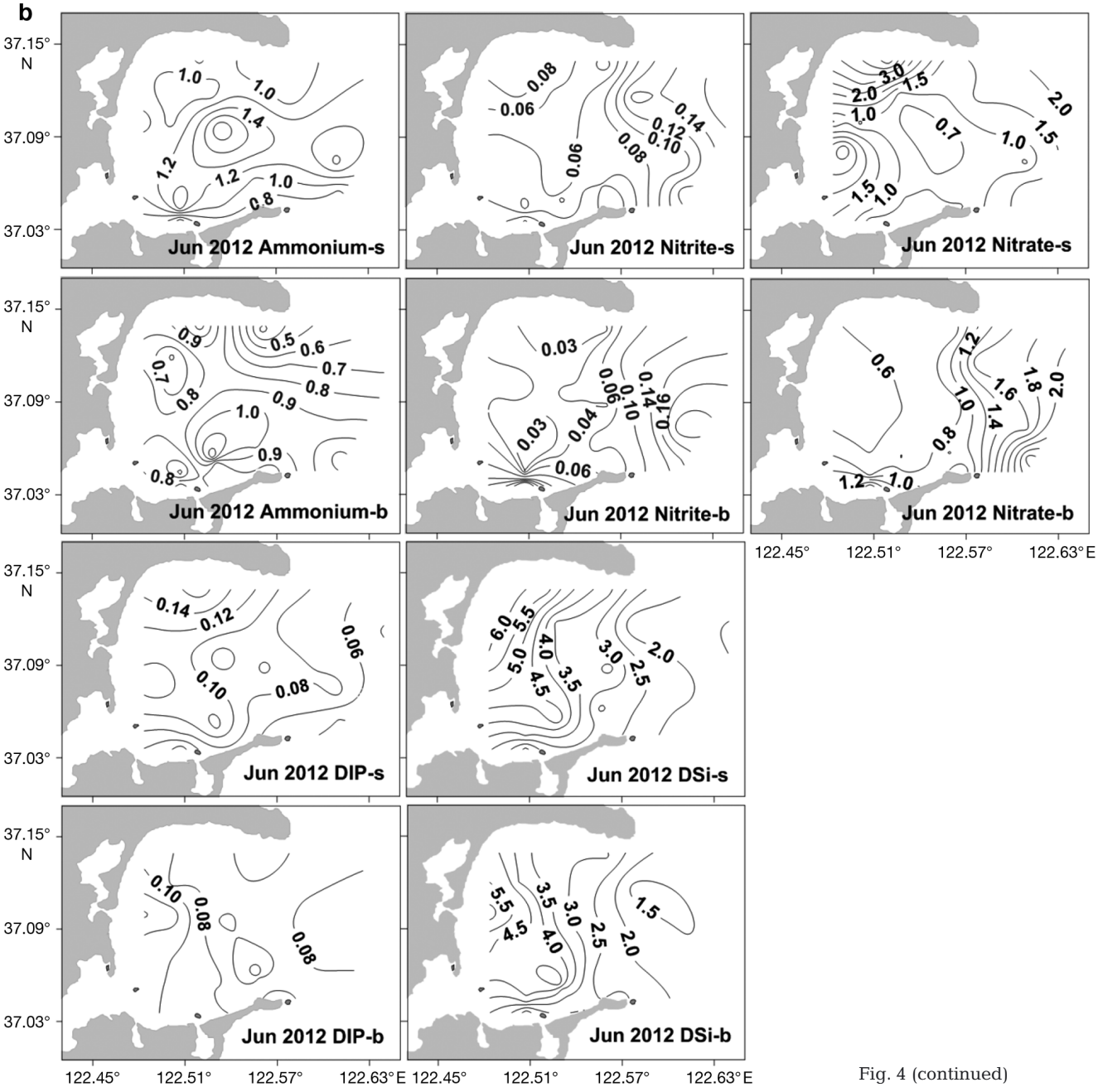
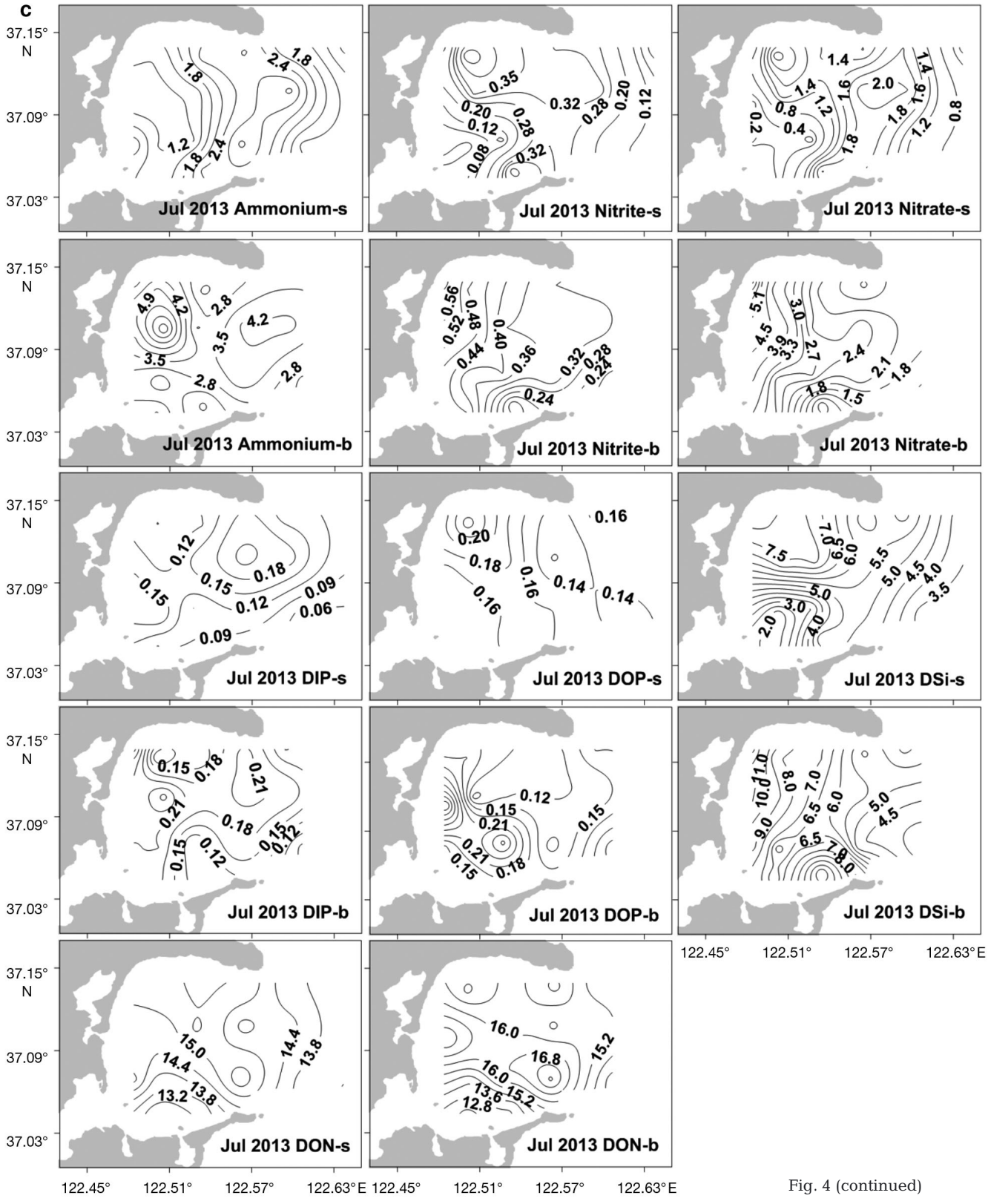


Fig. 4 (continued)

During the September–October 2012 study period, NO_3^- and NH_4^+ concentrations decreased from south to north in the bay; NO_2^-, DSi, and DOP increased gradually from west to east, and the PO_4^{3-} concentration increased from northeast to southwest (Fig. 4d). Throughout the entire bay, NO_3^- comprised 52–86% of DIN, and NH_4^+ comprised 6–38%. In October 2013, the NO_3^-, NO_2^-, DON, DIP, and DSi concentrations decreased from the mouth to the southwestern part of the bay (Fig. 4e). Throughout the entire bay, NO_3^- accounted for 55–84% of DIN. DON comprised 27–48% of TDN inside the bay, and 51–61% in the kelp monoculture area. DOP contributed to 12–36% and 16–50% of TDP in surface water and the bottom layer, respectively. In autumn in both 2012 and 2013, the average DIN:PO_4^{3-} ratios were higher than the Redfield ratio, while the DSi:DIN ratios in the water column were comparable to the Redfield ratio.

In winter, the horizontal distribution of nutrients was similar to that in spring (except for the NO_2^- and NH_4^+ concentrations), with higher concentrations in the near-bottom layer than in surface water (Fig. 4f).



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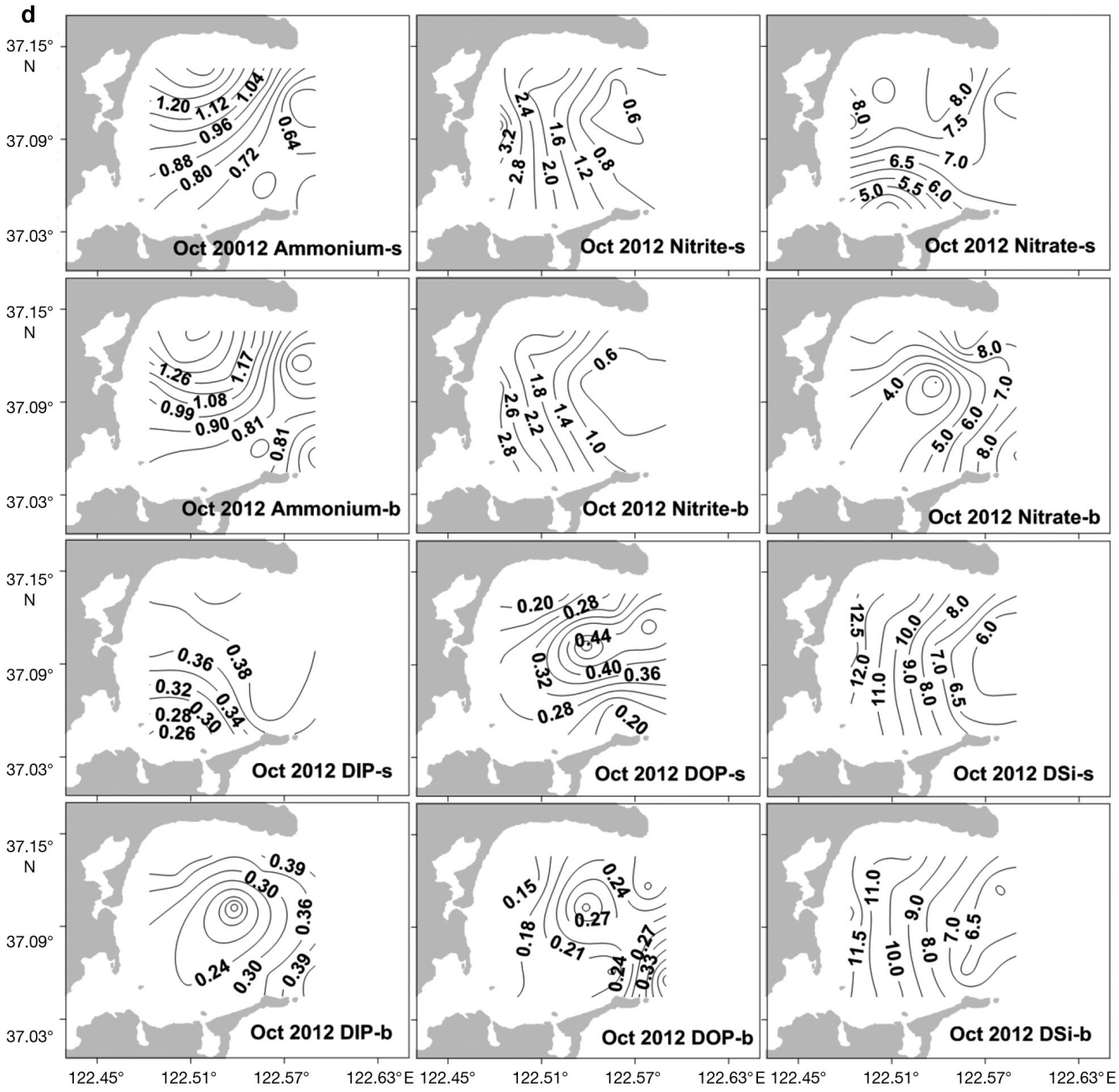

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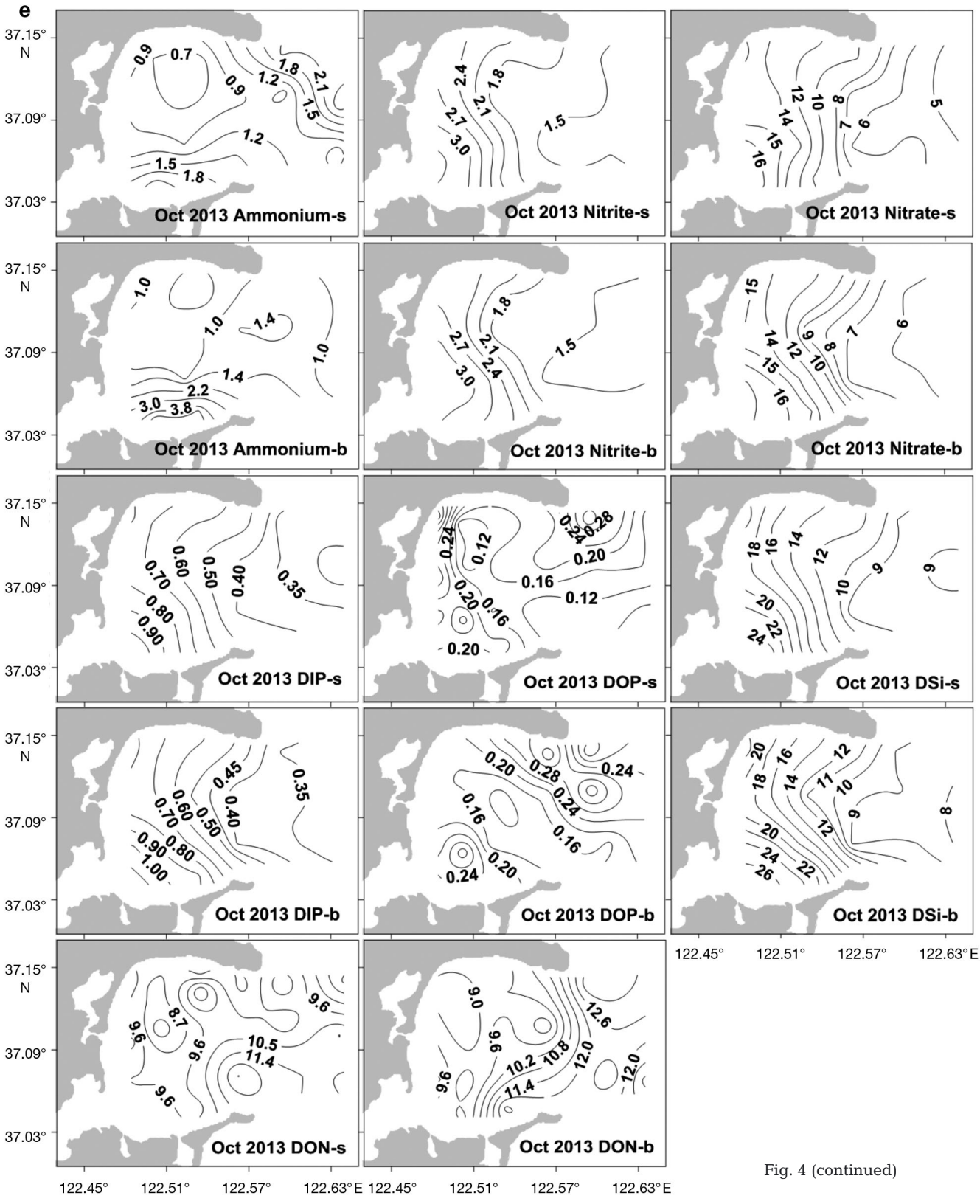
In the entire bay, NO_3^- accounted for 66–92% of DIN. DON was the dominant species of TDN, which represented 53–81% of TDN in the water column, and DOP represented 35–67% of TDP. The molar ratios of DIN:PO_4^{3-} ranged from 20 to 62 and 17 to 46 in surface and bottom waters, respectively. The average DSi:DIN ratio in surface and bottom waters was comparable and significantly lower than the Redfield ratio. The results suggest that phosphorus may be a limiting element for phytoplankton growth in winter.

Seasonality in nutrient concentrations was evident in SGB (Figs. 4 & 5). At all sites, the NO_3^-, PO_4^{3-}, and DSi concentrations were significantly higher in autumn than in the other seasons. The average NO_3^- concentrations in surface (9.44 ± 4.00 µM) and bottom (9.72 ± 4.48 µM) waters in autumn exceeded those in summer by factors of 7.4 and 5.3, respectively. DIN was dominated by NO_3^-, except in summer. The DON concentrations in winter (16.0 ± 1.67 µM) were comparable to those in summer, and were signifi-



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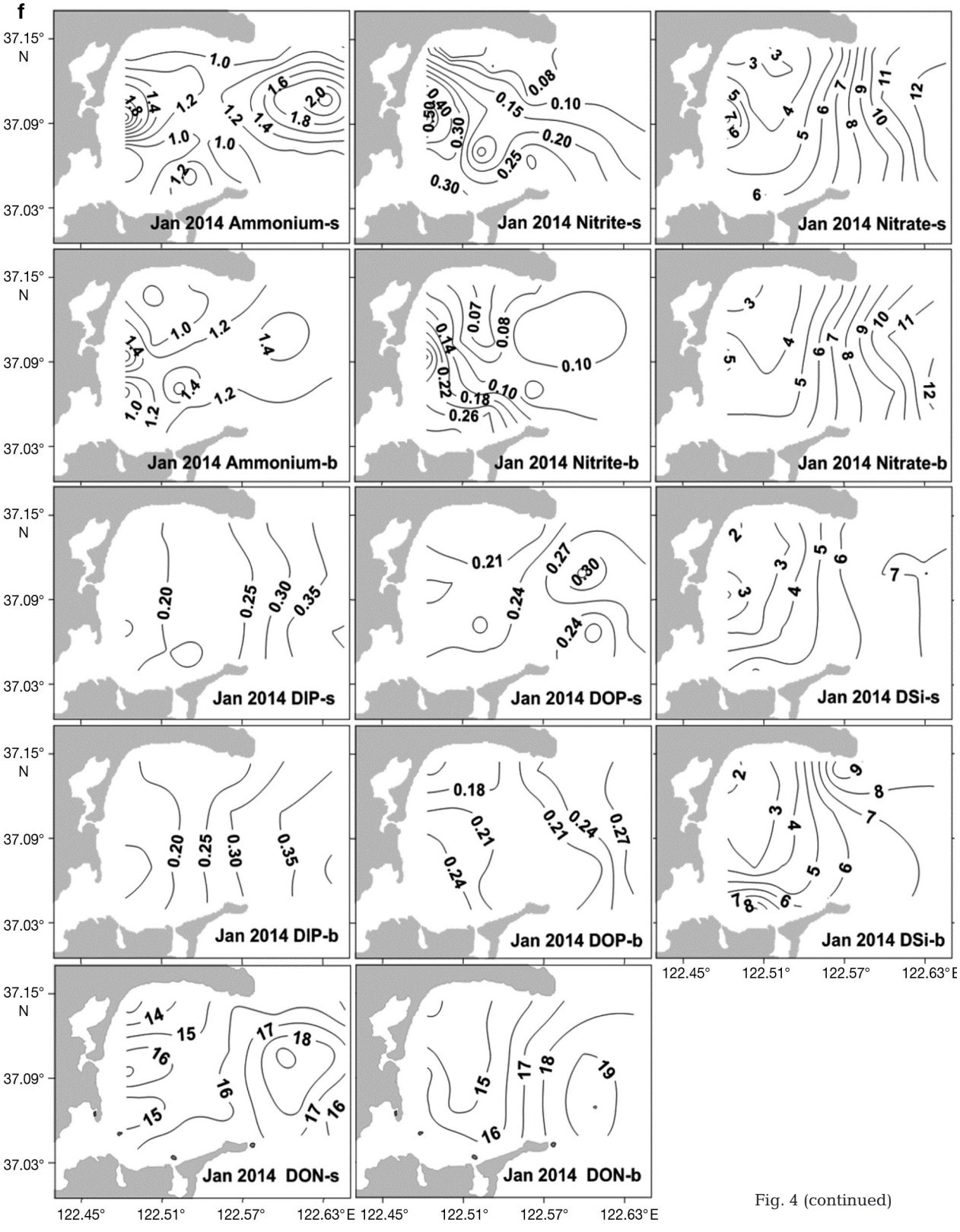

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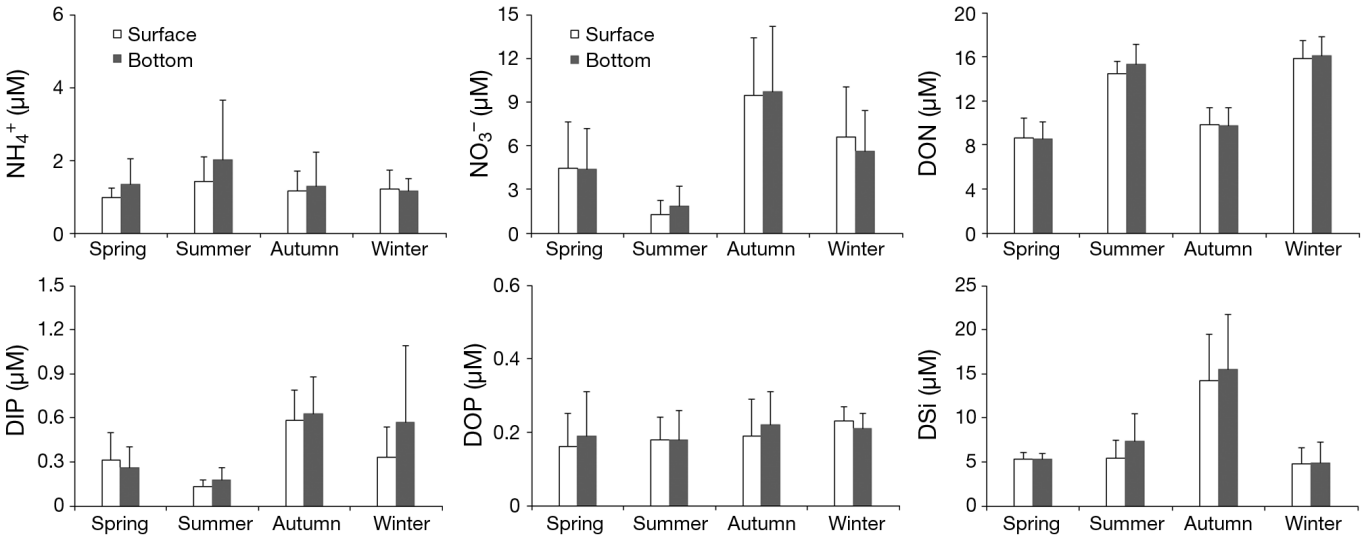


Fig. 5. Seasonal variations in nutrient concentrations (µM) in Sanggou Bay during the study period. DON: dissolved organic nitrogen, DIP (DOP): dissolved inorganic (organic) phosphorus, DSi: dissolved silicate

cantly higher than the concentrations in spring and autumn (Fig. 4). TDN was dominated by DON (59–82%), except in autumn (approximately 40%). Two-way ANOVA indicated highly significant differences in nutrient concentrations among seasons and layers ($p < 0.01$). The subsequent post hoc Tukey's HSD test showed that the nutrient concentrations in autumn differed significantly from those in other seasons ($p < 0.01$). Two-way ANOVA also indicated highly significant differences in nutrient concentrations among seasons and cultivation areas (Fig. 6; $p < 0.01$), suggesting that aquaculture activities significantly affect the nutrient composition in SGB.

Nutrients at the anchor stations

In April 2013, all nutrients changed during the tidal cycle at Stn D1 (Fig. 7a). The maximum concentrations usually occurred during high tide, indicating the outer bay as a nutrient source. The vertical profiles for concentrations of all dissolved inorganic nutrients at Stn D1 showed that the water column was well mixed (Fig. 7a). High concentrations of DON (9.01–13.8 µM) were found throughout the water column, and comprised up to 50% of TDN. The DIN:PO_4^{3-} ratio ranged from 23 to 74 in surface water and from 30 to 132 in near-bottom water, and the DSi:DIN ratio ranged from 0.5 to 0.8 in surface water and from 0.4 to 0.9 in near-bottom water. At Stn D2, the nutrient concentrations were higher in near-bottom waters than in surface water, the exception being NH_4^+ and DOP (Fig. 7a). The DON (8.26–10.5 µM) comprised 66–87% of TDN. The concentrations of DOP (0.08–0.35 µM) represented 25–73% of TDP, and indicated a well-mixed profile. The DIN:PO_4^{3-} ratio increased from 8.0–20 in surface water to 11–37 in near-bottom water, while the DSi:DIN ratio decreased from 1.6–3.2 in surface water to 1.0–1.5 in near-bottom water. The nutrient concentrations at Stn D1 were higher than at D2.

Analysis of the concentrations of all nutrients during 18–19 October 2013 showed that the water column at Stn D1 was well mixed (Fig. 7b). No parameter showed significant differences between day and night, indicating that tidal mixing was the main factor affecting concentration changes. The concentrations of DON were 5.38–10.5 µM, which comprised 26–83% of TDN. The DOP concentrations were 0.05–0.34 µM, which represented 8–39% of TDP. The DIN:PO_4^{3-} ratio was 23–36 (average 27) in surface water, and 22–51 (average 28) in bottom water. The DSi:DIN ratio was 0.7–1.0 (average 0.9) in surface water and 0.5–1.0 (average 0.8) in bottom water. At Stn D2, the concentrations of all nutrients in surface water showed a general decrease with increasing tide height. The DIN:PO_4^{3-} and DSi:DIN ratios in surface water ranged from 22 to 32 and 0.8 to 1.0, respectively. The nutrient concentrations at Stn D1 were lower than at D2.

Water and nutrient budgets in SGB

Domestic wastewater is discharged directly into rivers adjacent to SGB, and so in developing a water

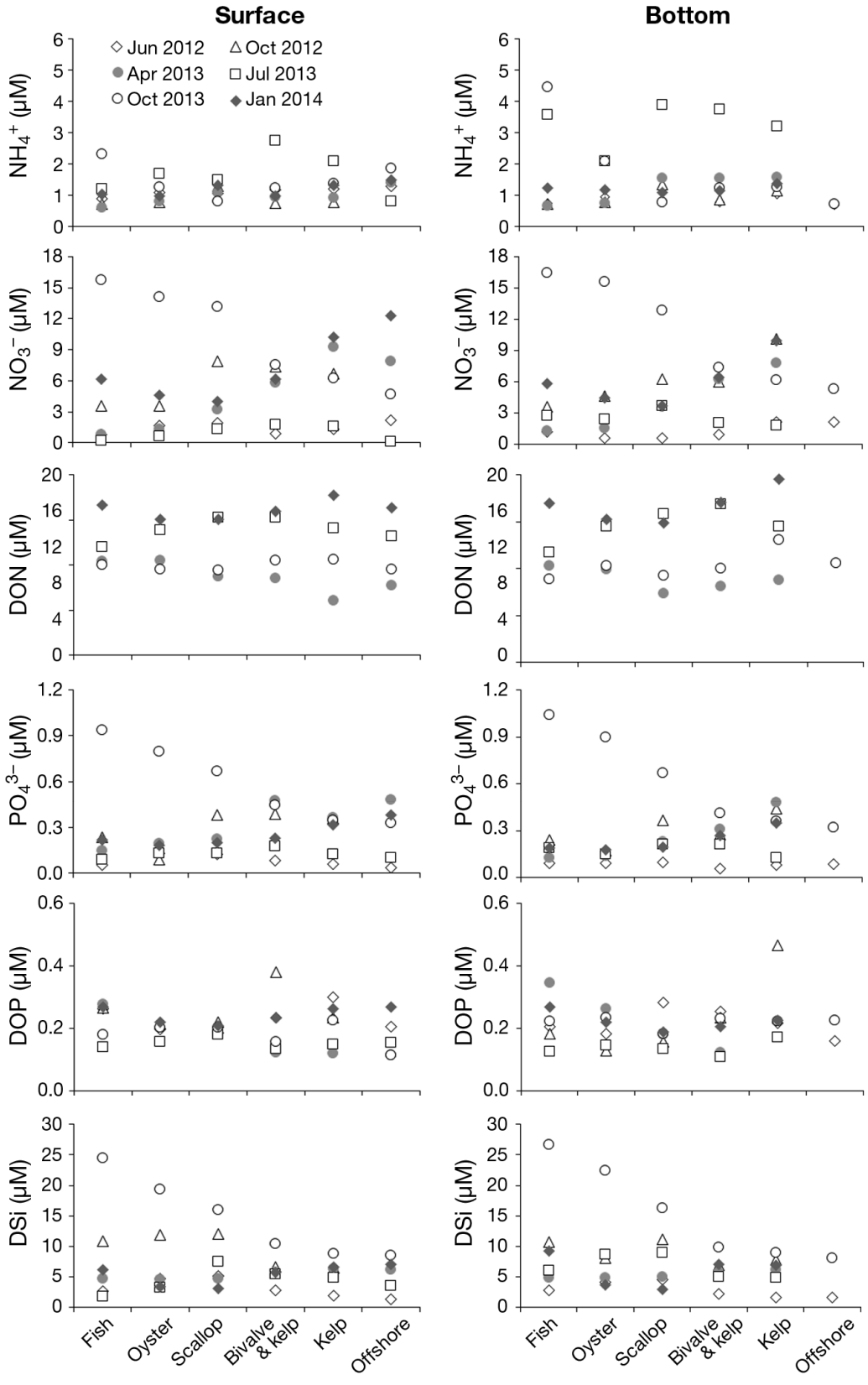


Fig. 6. Nutrient cycles, averaged for various aquaculture regions in Sanggou Bay Left: nutrients in surface water, right: nutrients in the near-bottom layer. DON: dissolved organic nitrogen, DOP: dissolved organic phosphorus, DSi: dissolved silicate

budget for the bay, sewage discharge was included in river discharges. The Guhe is the largest major river that directly empties into SGB. In developing the water budget (Fig. 8), we used the average discharge (V_Q) of the Guhe during 2011. The submarine groundwater discharge (SGD) was estimated based on submarine groundwater measurements made in June 2012. The groundwater discharge into SGB was calculated to be (2.59–3.07) × 10^7 m^3 d^{-1}, based on the naturally occurring ^{228}Ra isotope (Wang et al. 2014). Generally, recirculated seawater accounts for 75 to 90% of total SGD (Moore 1996). Based on Ra isotopes, Beck et al. (2008) reported that recirculated seawater could account for approximately 90% of total SGD, and could increase as a consequence of precipitation (Guo et al. 2008). In our study, groundwater samples were collected during a summer in which substantial rainfall occurred. Based on the assumption that recirculated seawater could account for 90% of total SGD in SGB, the SGD was estimated to be (2.59–3.07) × 10^6 m^3 d^{-1}. As the volume (V_S) of SGB is 10.8 × 10^8 m^3, the total water exchange time (τ) for SGB, estimated from the ratio $V_S/(V_R + V_X)$, was 22.4 d.

Scallop (*Chlamys farreri*) and oyster (*Crassostrea gigas*) are the main shellfish cultured in SGB. Aquaculture wastewater effluents are discharged directly into the bay. The minimum individual wet weight of oysters and scallops at harvest are 40 and 23 g (Nunes et al. 2003), respectively, and 60 000 t of oyster (wet weight) and 15 000 t of scallop are harvested annually from the bay (data from Rongcheng Fishery Technology Extension Station). Based on these data, we estimated that bivalve cultivation involved approximately 2.15 × 10^9 individuals during 2012. Based on excretion rates determined for bivalves and oysters in Sishili Bay (China) (Zhou et al. 2002a), the quantities of DIN and phosphate excreted by scallops were 3.84 and 0.21 µmol h^{-1} $ind.^{-1}$, respectively, and by oysters were 3.57 and 0.25 µmol h^{-1} $ind.^{-1}$, respectively. The bivalve growth

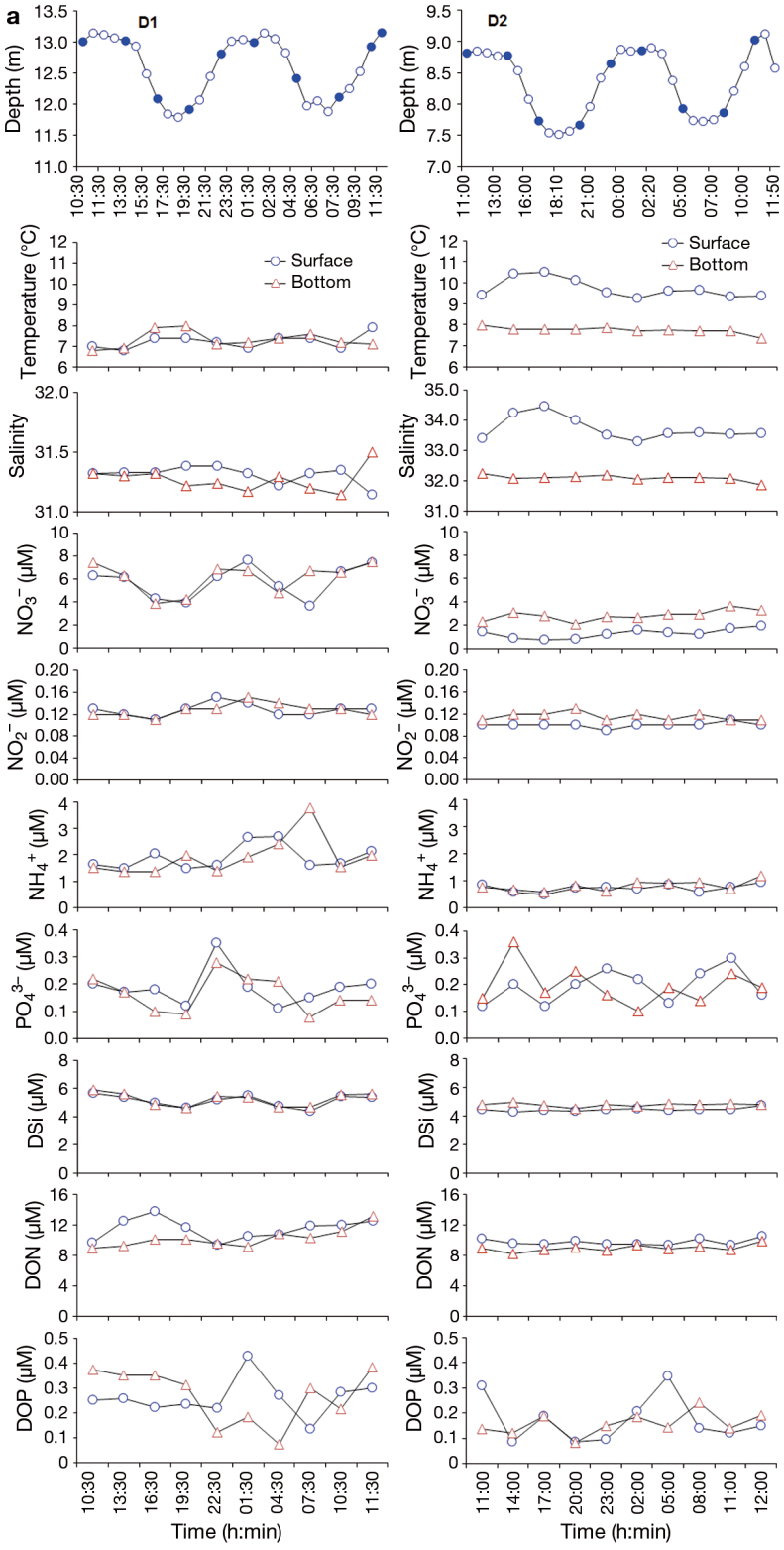
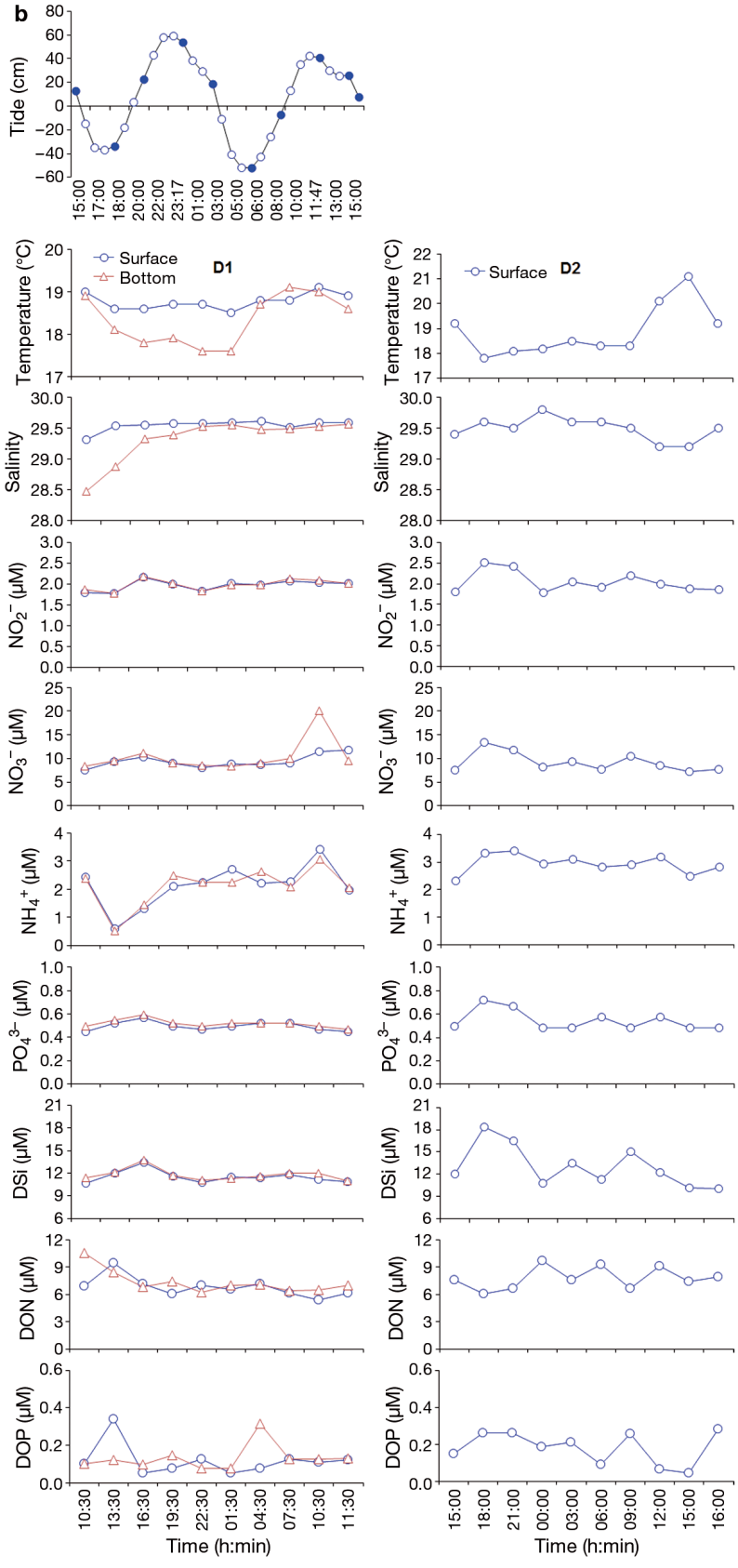


Fig. 7. Concentrations of nutrients (µM) at: (a) the anchor station in April 2013; (b; next page) the anchor station in October 2013. The water depth (m) in April 2013 and tide heights (cm) in October 2013 are provided, and the filled circles represent the nutrient sampling times. DSi: dissolved silicate, DON (DOP): dissolved organic nitrogen (phosphorus)



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periods were mainly from May in one year to November in the following year (approximately 500 d). Hence, the total DIN and phosphate excreted by scallops and oysters in SGB amounted to 70.9×10^6 and 4.19×10^6 mol yr $^{-1}$, respectively. Nutrients are removed from the bay as a consequence of bivalve harvest. The dry weight nitrogen content of the soft tissue and shell of *C. gigas* is 8.19 and 0.12 % (Zhou et al. 2002b), respectively, while the phosphorus content is 0.379 and 62.1×10^{-4} % (Zhou et al. 2002b), respectively. The dry weight nitrogen and phosphorus content of the soft tissue of *C. farreri* is 12.36 and 0.839 % (Zhou et al. 2002b), respectively, and in the shell is 0.09 and 62.1×10^{-4} %, respectively. Therefore, in total the harvest of *C. farreri* and *C. gigas* removes 304 t of nitrogen and 16.7 t of phosphorus from the bay.

Saccharina japonica and *Gracilaria lemaneiformis* are the main algae cultivated in SGB. The weight of individual kelp plants at seeding is 1.2 g, and the cultivation area and density are 3331 ha and 12 ind. m^{-2}, respectively (Nunes et al. 2003). The dry weight:wet weight ratio of kelp is 1:10 (Tang et al. 2013). Hence, the dry weight of kelp at seeding is 48 t, while 87 040 t of dried kelp are produced annually in the bay (data from Rongcheng Fishery Technology Extension Station). The dry weight nitrogen and phosphorus content of kelp is 1.63 and 0.38 % (Zhou et al. 2002b), respectively. Hence, 1419 t of nitrogen and 331 t of phosphorus are removed from the bay as a consequence of kelp harvest. Similarly, 25 410 t wet weight of *G. lemaneiformis* are produced annually in the bay (data from Rongcheng Fishery Technology Extension Station). Therefore, 41.4 t of nitrogen and 9.66 t of phosphorus are removed from the bay as a consequence of *G. lemaneiformis* harvesting.

The nutrient transport fluxes from rivers and groundwater into SGB were determined from surveys undertaken during the period 2012 to 2014. The nutrient concentrations in rainwater were based on measurements at Qianliyan Island, in the western Yellow Sea (Han et al. 2013). Benthic fluxes in SGB were based on surveys undertaken during the same period

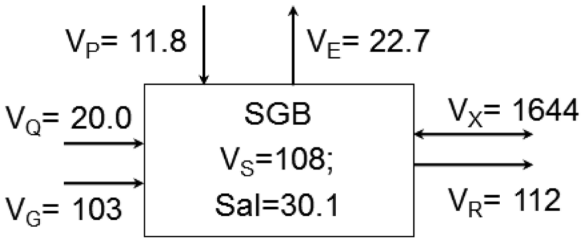

Fig. 8. Water and salt budgets for Sanggou Bay (SGB). Units: water volume, 10^7 m^3; water and salt fluxes, 10^7 m^3 and 10^7 psu m^3 mo^{-1}, respectively. V_Q, V_P, V_E, V_G, V_S, V_R, and V_X are the mean flow rate of river water, precipitation, evaporation, groundwater, the volume of the system of interest, the residual flow, and the mixing flow between the system of interest and the adjacent system, respectively. For comparison, salinity of the adjacent system = 32.23

(Ning et al. 2016, this Theme Section). For the nutrient budget, estimates of DSi removed through kelp and bivalve harvesting were not included, as no data were available.

The nutrient budgets showed that SGB behaved as a source of PO_4^{3-} and as a sink of DSi and DIN (Table 3). The model results indicated that PO_4^{3-} was mainly derived from bivalve excretion, which accounted for 65% of total influx, while benthic flux contributed 16% of total influx. Bivalve excretion may be an important source of PO_4^{3-} when phytoplankton growth is phophorus-limited in the bay. The DSi load in the bay was mainly from river input and benthic flux, which contributed 47 and 34% of total influx (Table 3), respectively. Groundwater was the major source of DIN entering SGB, accounting for 41% of total influx. In addition, bivalve excretion accounted for 19% of total DIN influx. DIN and PO_4^{3-} were mainly removed through kelp harvesting, which represented up to 64 and 81% of total outflux, respectively. The results show that aquaculture activities play an important role in nutrient cycling in SGB.

DISCUSSION

Nutrient transport in rivers

Nutrient levels in rivers varied widely (Table 2). The DIN concentrations in the rivers fell between those for polluted waters (110 µM) and severely polluted waters (350 µm) (Smith et al. 2003), except for the Bahe river. The DIN concentrations in the studied rivers were also higher than in most other small to medium-sized rivers in temperate China (Liu et al. 2009), and high relative to major Chinese rivers including the Yellow, Yangtze, and Pearl rivers (Liu et al. 2009). The extremely high DIN concentrations resulted in the high DIN:PO_4^{3-} ratios in these rivers.

The DIN loading to streams is directly related to the extent of agriculture in the catchment (Heggie & Savage 2009). The high NO_3^- concentrations, which dominated the DIN in rivers, is primarily attributable to anthropogenic nutrient sources, particularly to washout of fertilizers not used by target plants (Bellos et al. 2004). Rivers in the study area flow through villages and Rongcheng City, then discharge directly into SGB. Untreated industrial and domestic sewage is also discharged directly into rivers. The drainage

Table 3. Nutrient budgets for Sanggou Bay, China. V_RC_R: residual nutrient transport out of the system of interest (Eq. 1); V_XC_X: mixing exchange flux of nutrients (Eq. 2); influx (outflux): total nutrient flux into (out of) the system of interest. Δ (=∑outflux – ∑influx) is the non-conservative flux of nutrients. Negative and positive signs of Δ indicate that the system is a sink or a source, respectively. DIP (DIN): dissolved inorganic phosphorus (nitrogen), DSi: dissolved silicate (units: 10^6 mol)

	DIP	DSi	DIN	Reference
River input (V_QC_Q)	0.29	22.4	83.2	Present study
Atmospheric deposition (V_PC_P)	0.41	0.87	14.6	Han et al. (2003)
Groundwater discharge (V_GC_G)	0.55	8.27	155	Wang et al. (2014)
Benthic fluxes	1.05	16.3	57.8	Ning et al. (2016)
Bivalve excretion	4.19		70.9	Zhou et al. (2002a,b)
Influx	**6.49**	**47.8**	**382**	
Kelp harvest	–10.7		–101	Zhou et al. (2002a,b)
Gracilaria lemaneiformis harvest	–0.32		–2.96	Zhou et al. (2002a,b)
Bivalve harvest	–1.19		–21.7	Zhou et al. (2002a,b), Zhang et al. (2013)
Residual flow (V_RC_R)	–0.38	–8.26	–7.31	Present study
Mixing exchange (V_XC_X)	–0.65	–16.1	–26.2	Present study
Outflux	**13.2**	**24.4**	**159**	
ΔY (=∑outflux – ∑influx)	**6.71**	**–23.4**	**223**	

areas of the Yatouhe, Sanggouhe, and Shilihe rivers are small (<30 km^2) and are therefore readily affected by human activities. We conclude that the high NO_3^- concentrations in rivers are derived from agriculture, urban, and industrial wastewater in their drainage basins, as well as surface runoff from Rongcheng City.

The concentrations of PO_4^{3-} in the Bahe and Guhe rivers were between those for pristine (0.5 µM) and clean (1.4 µM) water, and apparently lower than in the Shilihe and Sanggouhe rivers (Table 2). The high PO_4^{3-} concentration (up to 6.02 µM) in the Sanggouhe, and industrial and domestic sewage, might be the most important sources of PO_4^{3-} to water bodies. DSi is little affected by human activities (Jennerjahn et al. 2009) and mainly originates from natural sources. The high DSi levels in rivers adjacent to SGB may be related to the underlying rock types and weathering rates.

Rain events can result in nutrient inputs derived from hinterland areas. Approximately 73.3% of annual precipitation occurs during summer (June to September), and the annual rainfall in Rongcheng City is 819.6 mm. River discharges can be enhanced by rainfall, and weathering rates are affected by precipitation and temperature (Liu et al. 2011), which can lead to higher nutrient values during the wet seasons. High nutrient concentrations (especially dissolved silicate) but low salinities were found in the bay (Fig. 4), suggesting that rainfall might be an important factor affecting nutrient supply to SGB in summer.

Nutrient fluxes from the bay to the Yellow Sea

In this study, nutrient budgets were developed to provide an overview of nutrient cycles under the impact of aquaculture activities. Despite some uncertainties, the nutrient budgets indicated that large quantities of nitrogen and silicate would probably be buried in the sediment or transformed into other forms in the bay (Table 3). Seaweeds can absorb large amounts of nutrients from the water column, resulting in the removal of these nutrients from the system when the plants are harvested (Schneider et al. 2005). The budgets indicated that a large proportion of DIN and DIP were removed during seaweed and bivalve harvesting (Table 3), demonstrating that aquaculture activities are a significant sink for nutrients in the bay.

Based on the budgets, nutrient fluxes from SGB to the Yellow Sea were estimated as the sum of the net residual flux ($V_R C_R$) and mixing flux ($V_X C_X$) (Table 3). With the exception of DIN, nutrient fluxes to the Yellow Sea were 1.1 to 3.6 times the riverine input ($F_{model} = VC_Q$), indicating that nutrient cycling in the bay (including regeneration, aquaculture effluents) may magnify the riverine fluxes, especially bivalve excretion, which contributed to 65% of the total DIP influx. Additionally, the molar ratios of DIN:PO_4^{3-} and DSi:DIN were approximately 49 and 0.2 in all external nutrient inputs to the studied system, respectively, while the corresponding flux ratios in the output waters to the Yellow Sea were approximately 35 and 0.7. These ratios deviated significantly from the Redfield ratio, indicating that aquaculture activities have significantly influenced nutrient cycling in the bay.

Wang et al. (2014) estimated that approximately 4.76×10^7 mol mo^{-1} of DIN and 5.58×10^6 mol mo^{-1} of PO_4^{3-} are input from fertilizer and feed, based on protein data of shellfish and kelp in the bay during summer being used to construct a mass balance. Based on their data, fertilizer and feed would be the major source of nutrients in the bay. By visiting local farming households, we confirmed that fertilizers were used; however, fertilizer and feed are only used in fish farming during summer in SGB, thus the amounts might be far below the estimated values. If fertilizer and feed for fish farming were taken into account, the uncertainty might rise. Hence, nutrient input from feed was ignored in the box model. Furthermore, aquaculture effluents were not taken into account. Consequently, more studies on nutrient cycling in relation to aquaculture activities in SGB are needed to improve our understanding of the nutrient sink or source function of the bay.

Effects of aquaculture activities on nutrient biogeochemical cycles

The nutrient concentrations varied significantly among seasons in SGB. The dissolved inorganic nutrient levels in SGB in summer were quite low compared with other seasons; they increased from summer to autumn and reached the highest values in October (Figs. 4 & 5), indicating a shift from consumption to autumn accumulation. These seasonal variations corresponded with aquaculture activities in the bay, and this was confirmed by statistical analysis. Zhang et al. (2012) reported that nutrient biogeochemical processes and cycles were significantly affected by intensive kelp and bivalve aquaculture activities in SGB. Shi et al. (2011a) also reported that

Saccharina japonica assimilates substantial nutrients in spring. During the growth period of kelp from November to May, the NO_3^- and PO_4^{3-} concentrations decreased rapidly because of assimilation by kelp (Fig. 6). Nitrogen removed through kelp harvesting accounted for 64% of total outflux (Table 3). Kelp was a net sink for nutrients during winter and spring, and competed with phytoplankton for nutrient utilization during kelp seeding; as a consequence, phytoplankton growth was restrained. Following the kelp harvest in late May, phytoplankton could grow fast because of adequate solar radiation and temperature. As a result, the dissolved inorganic nutrient concentrations continued to decrease (Figs. 4–6).

Shellfish aquaculture generally commences in May, during the period when kelp is harvested. Bivalves in turn become another source of nutrients through excretion. During early summer, the bivalves are in the early growth stage, and produce only low levels of nutrients. The dissolved nutrients released through bivalve excretion have the potential to stimulate phytoplankton production at local scales and promote the risk of harmful algal blooms (Pietros & Rice 2003, Buschmann et al. 2008). The highest concentrations of chlorophyll *a* have been reported in summer (Hao et al. 2012). The dissolved nutrients in aquaculture effluents, coupled with high solar radiation, result in high phytoplankton production in summer (Shpigel 2005). At this time, *Gracilaria lemaneiformis* replaces kelp, and is cultivated from June to October in SGB; because it can use available nitrogen efficiently (Buschmann et al. 2008), it absorbs nutrients from seawater and probably reduces the nutrient levels in summer. This probably leads to the nutrient levels dropping rapidly to the lowest level in summer (Fig. 6).

In September, the bivalves are in active growth stages and generate large quantities of metabolic byproducts. The maximum metabolic rates for oysters are recorded in July and August (Mao et al. 2006), and lead to high nutrient concentrations in seawater (Fig. 5). Bivalves filter phytoplankton larger than 3 µm in size, thereby reducing their biomass in the water column (Newell, 2004). Phytoplankton growth is also limited by the level of solar radiation (Shi et al. 2011b). Thus, as nutrient utilization by phytoplankton decreased, the dissolved inorganic nutrient concentrations increased as a result, and increased to a greater extent in regions where bivalve monoculture occurred. Based on the nutrient budget in our study, phosphorus released from bivalve excretion could account for 65% of total influx to SGB. Hence, from June to October, prior to kelp seeding, bivalves and fish excretion may constitute an important nutrient source in SGB, leading to increased nutrient levels. Particulate waste material (feces or pseudofeces) from bivalves and phytoplankton are consumed by bivalves, and the nutrients involved may be removed through bivalve harvesting (Shpigel 2005, Troell et al. 2009). As top-down grazers, bivalves filter phytoplankton, which results in a reduction in the nutrient turnover time and speeds up nutrient cycling.

Nutrients can be produced indirectly via remineralization and subsequent release from enriched sediments (Forrest et al. 2009). Nutrient release from sediment is also a common phenomenon occurring beneath bivalve farms in SGB (Cai et al. 2004, Sun et al. 2010). The nutrient budgets also show that benthic flux is another important source of nutrients in SGB, especially for DIP and DSi (Table 3), and that this is significantly affected by aquaculture activities in the bay (Ning et al. 2016). Based on studies of other bivalve culture systems and natural or restored oyster reefs, it is evident that benthic fluxes are determined by processes involving filter feeding and excretion of dissolved nutrients, as well as biodeposition and sediment remineralization of nutrients (Newell 2004, Forrest et al. 2009). The TDN in SGB was dominated by DON in both summer and winter (Figs. 4 & 5), as observed in landbased aquaculture (Jackson et al. 2003, Herbeck et al. 2013). Burford & Williams (2001) reported that most of the dissolved nitrogen leaching from feed and shrimp feces was in organic rather than in inorganic forms. Hence, DON leaching from feces or pseudofeces might be an important source of DON in the bivalve cultivation regions in SGB (Fig. 6). Furthermore, increased sedimentation of organic matter from feces and pseudofeces underneath mussel farms can have significant ecosystem effects on the biogeochemical cycles of nitrogen and phosphorus (Stadmark & Conley 2011).

Biogeochemical cycling of DSi can be affected by diatom dissolution, sediment resuspension, and terrigenous input. In our study, the average concentrations of DSi increased by 9.0 µM from July to October, and decreased rapidly from 14.2 to 4.76 µM in January. Phytoplankton abundance was tightly controlled by filter feeding of oysters (Hyun et al. 2013), so the high metabolic rates of oysters may result in a reduction of diatom biomass, leading to high levels of DSi in autumn. In addition, as the water depth in SGB is ≤20 m, sediment resuspension and diatom dissolution might be important sources of DSi during the summer to autumn period. The dissolution of diatom frustules depends on a variety of factors, including microbial activity (Olli et al. 2008). Bacteria can

attack the organic matrix protecting the diatom frustule, exposing biogenic silica, and substantially increase the dissolution rate (Bidle & Azam 1999). The maximum biomass in SGB occurred in autumn (Chen 2001), and diatoms dominated in the bay in summer. Consequently, dissolution of diatom frustules may be an important source of DSi in the bay.

Although the aquaculture area and quantities of effluents released in SGB were high (Table 3), nutrient levels in the bay were not significantly elevated compared with other bays used for aquaculture, including Jiaozhou Bay (Liu et al. 2007) and Sishili Bay (Zhou et al. 2002b). This is attributed to the fact that nutrients released from shellfish are taken up by seaweeds during their growth periods. Large-scale kelp cultivation plays an important role in keeping nutrients at low levels and maintaining relatively good water quality.

Effects of physical factors on nutrient changes

The marine IMTA culture system used in SGB is suspended aquaculture. Water exchange between SGB and the Yellow Sea could be hindered by kelp (*S. japonica*), especially during kelp harvesting (Zeng et al. 2015). Our depth study showed that nutrien changes over the tidal cycle generally closely followed changes in water depth at Stn D2 (Fig. 7), indicating that water exchange is greater at Stn D1 (in the northern mouth of SGB), and weaker at Stn D2. Furthermore, in April 2013, the nutrients were well mixed at Stn D1, while at Stn D2, the nutrient concentrations were higher in bottom water than in the surface water (Fig. 7). This indicates that the current was affected by the aquaculture facilities and kelp at Stn D2, which may have led to higher nutrient concentrations in the bottom water than in the surface water. These results are consistent with the *in situ* measurements of Zeng et al. (2015), which showed that the vertical tidal flux at the northern entrance of SGB was much larger than at the southern entrance. In addition, the current structure in SGB has been significantly changed by the presence of aquaculture activities (Shi et al. 2011a). The tidal current in the surface layer is only half that in the middle layer when kelp is at its maximum length (Shi et al. 2011a). As a result, particulate matter and nutrients in bottom waters are constrained from entering the upper water layers because of the influence of aquaculture facilities and species (Wei et al. 2010).

The current flow generally tends to decrease in suspended aquaculture areas because of the extra drag caused by the presence of aquaculture facilities. In SGB, bivalves and fish are grown in cages, nets, or other containers hung from floats or rafts. Based on a 3-dimensional physical–biological coupled aquaculture model (Shi et al. 2011a), the average current flow speed can be reduced by approximately 63% by aquaculture facilities and cultured species. Moreover, Grant & Bacher (2001) reported a 20% reduction in current speed in the main navigation channel in SGB, and a 54% reduction in the middle of the culture area because of the effects of suspended aquaculture. Nutrients are likely to be retained in the bay because of the weaker current in the bivalve culture areas. The nutrient budgets showed that bivalve excretion was an important source of nutrients (Table 3). Large quantities of nutrients could accumulate in the west of the bay, and red tides have occurred in SGB in recent years (Zhang et al. 2012). The effects of consequent shading and competition pressure from the increased algae biomass on the valuable habitats involved may negatively affect the seagrass meadows in the southwest of the bay, and the production of bivalves may be reduced. To conserve the natural services provided by the bay, aquaculture effluents should be treated before they are released into natural water bodies.

Water exchange can also cause differences in nutrient species inside and outside SGB. Wei et al. (2010) observed that the flow speed declined by approximately 70% from the mouth to the southwestern part of the bay, and the outflow was slowed by the increased aquaculture activities and infrastructure (Fan & Wei 2010). Thus, movement of nutrients from the southwest of the bay to the open sea may be impeded, which was suggested by the high concentrations of nutrients found in this part of the bay in summer and autumn (Fig. 4).

Long-term trends of nutrients in SGB

Fig. 9 shows compiled data for DIN, DSi, and PO_4^{3-} in SGB, based on historical data and our observations (Song et al. 1996, P. Sun et al. 2007, S. Sun et al. 2010, Zhang et al. 2010, 2012, this study), reflecting the long-term variations for the period 1983 to 2014. No trends in the PO_4^{3-} concentrations were evident because of the high variability in this parameter (Fig. 9). In contrast, the DIN concentrations increased over time and were significantly higher in 2003 to 2011 than in previous years (Fig. 9). Prior to the 1980s, kelp was the main aquaculture species, and the DIN concentration was low in the bay (Fang et al. 1996a,

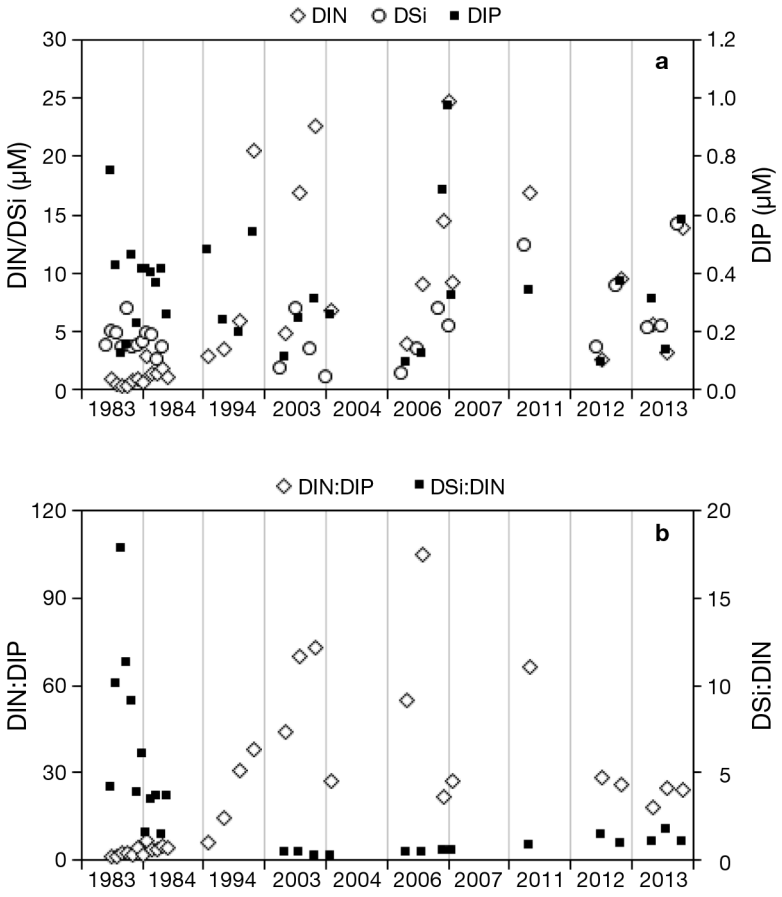

Fig. 9. Long-term changes in (a) the concentrations of dissolved inorganic nitrogen (DIN), dissolved inorganic phosphorus (DIP), and dissolved silicate (DSi) concentrations, and (b) the DIN:PO_4^{3-} and DSi:DIN ratios for the period 1983 to 2013

Ning et al. 2016). Polyculture was introduced into the bay for economic reasons (Fang et al. 1996a), and its rapid development may have been responsible for increasing levels of nutrients in the bay, and resulted in long-term alterations to the nutrient conditions (Shi et al. 2011a, Zhang et al. 2012). In SGB, nutrient-rich aquaculture effluents are released into the natural water body without prior treatment. The high concentrations of nitrogen in aquaculture effluents mainly originate from excess feed or from excretion from the farmed animals (Burford & Williams 2001).

As a result of the increased nitrogen levels, the DIN:DIP ratios in SGB shifted from severe nitrogen limitation in 1983 to the ecologically desirable Redfield ratio (16) in summer 1994, and continued to increase until summer 2006, when the DIN:PO_4^{3-} ratio reached 105; phytoplankton growth is now limited by phosphorus in summer. The increase in the DIN:PO_4^{3-} ratios in SGB is a common phenomenon observed in long-term studies of estuarine and coastal areas affected by human activities, and also in semi-closed bays used for aquaculture, including Chesapeake Bay in the US (Tango et al. 2005), and Jiaozhou (Shen 2002, Sun et al. 2011) and Daya Bays (Wang et al. 2009) in China. Turner et al. (1998) reported that the risk of harmful algal blooms increases with shifts in the DSi:DIN ratio to values <1, when phytoplankton becomes dominated by non-diatom species. Molar ratios of DSi:DIN in SGB changed from 1.4–18 in 1983 to <1 during the 2003 to 2011 period. Red tides were observed in April 2011 (Zhang et al. 2012), and were apparent in small areas in 2013. In addition, an increase in the DIN concentration will lower the DSi:DIN ratio, and could change ecosystem structure of the bay (Billen & Garnier 2007).

Because of its combination of environmental, economic, and social benefits (Allsopp et al. 2008, Nobre et al. 2010), IMTA has been gaining recognition as a sustainable approach to aquaculture, and the water quality in SGB has remained in good condition compared with other bays affected by aquaculture activities. Environmental management strategies will need to include both reduction of nutrient pollution and monitoring of the relative abundance of nutrients. The ecological and economic health of SGB should be tightly monitored to ensure a rapid response to critical changes.

CONCLUSION

We have reported on the nutrient dynamics of SGB, which represents a typical watershed for IMTA. The results of our investigation show that aquaculture activities play an important role in nutrient cycling in SGB. Nutrients showed considerable seasonal variation in the bay, and nutrient composition and distribution were also affected by the cultured species in the bay. The nutrient budgets showed that SGB behaved as a source of PO_4^{3-} and as a sink of DSi and DIN. The model results indicated that PO_4^{3-} was mainly derived from bivalve excretion. Bivalve excretion may be an important source of PO_4^{3-} when phytoplankton growth is phosphorus-limited in the bay. Seaweed and bivalve harvesting play an important role in removing DIN and PO_4^{3-} from the bay. Under the combined effects of natural processes and aquaculture activities, nutrient biogeochemistry in the bay has been affected.

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