

# Distribution, residence time, autotrophic production, and heterotrophic removal of DOP in the Mirs Bay, northern South China Sea

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## Abstract

The importance of dissolved organic phosphorus (DOP) as a potential nutrient source for primary producers in marine systems has been recognized for up to eight decades, but currently, the understanding of the biogeochemistry of DOP is in its infancy. In the present study, monthly data between 2000 and 2014 were used to analyze the temporal and spatial distributions of DOP in the Mirs Bay, the northern South China Sea. The DOP residence time ( $T_{DOP}$ ) was also investigated using a simple regression analysis in combination with chlorophyll *a* (Chl *a*) measurements while excess DOP ( $\Delta DOP$ ), produced by the biogeochemical processes of autotrophic production and heterotrophic removal, was determined using a two-component mixing mass-balance model in combination with salinity measurements. The results showed that the DOP concentration was  $(0.017 \pm 0.010)$  mg/L higher in the surface-water compared with the bottom-water and higher in the inner Tolo Harbour and waters adjacent to Shaotoujiao compared with the main zone of the bay. Although seasonal changes and annual variability in the DOP were small, the surface DOP concentration was higher in the wet season (April–September) than in the dry season (October–March) due to the impacts of seaward discharges and atmospheric deposition into the bay. Measurement and regression results showed that the DOP release rate from phytoplankton production was about  $1.83$  (g P)/(g Chl *a*) and the  $T_{DOP}$  was about 7 d, which implied that the DOP cycle in the bay was rapid. The  $\Delta DOP$  was calculated from the model to be about  $0.000$  mg/L in the main zone of the bay and about  $0.002$  mg/L in the inner Tolo Harbour and waters adjacent to Shaotoujiao, suggesting that the autotrophic production of DOP was almost balanced by the heterotrophic removal in the main zone of the bay and dominated in the inner Tolo Harbour and waters adjacent to Shaotoujiao. In conclusion, the Mirs Bay is very productive and fairly heterotrophic.

**Key words:** dissolved organic phosphorus, temporal and spatial distributions, residence time, autotrophic production, heterotrophic removal, Mirs Bay, northern South China Sea

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## 1 Introduction

Phosphorus, a macronutrient, is essential for all marine organisms, and it plays an important role in the support of phytoplankton production and at times in the limitation of productivity in various open oceans and coastal marine environments (Ruttenberg and Dyrman, 2005). Phosphorus limitation in coastal waters can cause biodiversity losses (Glibert et al., 2005) and in the euphotic zone, it can cause a decrease of phytoplankton productivity (PP) or “biological pump” efficiency, which influences the global climate through the sequestration of atmospheric carbon dioxide to the deep ocean (Paytan and McLaughlin, 2007).

Although dissolved inorganic phosphorus (DIP) is considered the most available form of phosphorus to microorganisms, phytoplankton and autotrophic bacteria utilize dissolved

organic phosphorus (DOP) to fulfill at least part of their phosphorus demand (Björkman and Karl, 2003). In marine environments, heterotrophic bacteria are responsible for hydrolyzing DOP to DIP. Phytoplankton and autotrophic bacteria, which take up DIP from solution for their metabolic needs, also hydrolyze DOP via alkaline phosphatases when their demand for phosphorus is not satisfied by DIP (Cotner and Biddanda, 2002; Azam et al., 1983). Direct secretion and/or excretion by living marine organisms release DOP to seawater (Cotner and Biddanda, 2002; Ward and Bronk, 2001). Viral and dead cell lysis releases cellular DOP to seawater too (Anderson and Zeutschel, 1970).

The importance of DOP as a potential nutrient source for primary producers in marine systems has been recognized for up to eight decades (Redfield et al., 1937; Jackson and Williams, 1985; Bentzen and Taylor, 1992). Increasing attention has been

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paid to studies of DOP in global oceans and coastal waters (Clark et al., 1999; Karl and Tien, 1997; Monaghan and Ruttenberg, 1999); however, historically, the emphasis on DOP has not been comparable to the emphasis on DIP for both large oceanographic survey programs and smaller-scale studies (Ruttenberg and Dyhrman, 2005). Currently, the understanding of the biogeochemistry of DOP, such as its autotrophic production and heterotrophic removal, is in its infancy. The understanding of bioavailability and biogeochemical behaviors of DOP in marine environments remains ambiguous and hinders the comprehension of marine phosphorus cycles.

Even though the Mirs Bay, located in the northern South China Sea (NSCS), is not rich in nutrients, it maintains high PP (Li et al., 2014). A previous study showed that DIP concentration is low in the bay, especially in the surface-water, during the summer (Zhou et al., 2011). During times of DIP depletion, to support the production of phytoplankton, DOP becomes the most important nutrient source in the bay. The aim of the present study was to determine the intensity of DOP variability, the rate of DOP turnover, and the scale of the imbalance between the autotrophic production and heterotrophic removal of DOP.

The Environmental Protection Department (EPD), the Government of the Hong Kong Special Administrative Region has monitored ecological and environmental parameters in the coastal waters around Hong Kong every one to two months since 1986. Monthly monitoring data from the water quality monitoring program of the EPD for the period of 2000–2014 were chosen for use in the present study. Data derived from a subset of the DIP and the total dissolved phosphorus (TDP) concentration series were used to analyze the temporal and spatial distributions of DOP concentration in the bay. In addition, a simple regression analysis and elemental stoichiometry ratios of the biogeochemistry were used in combination with chlorophyll *a* (Chl *a*) measurements to estimate the DOP residence time ( $T_{DOP}$ ) in the bay. A two-component mixing mass-balance model (Lee and Krothe, 2001; Li et al., 2013a, 2014) was used in combination with salinity (*S*) measurements to determine the imbalance of DOP resulting from the synchronous autotrophic production and heterotrophic removal processes in the Mirs Bay.

## 2 Materials and methods

### 2.1 Study area and monitoring sites

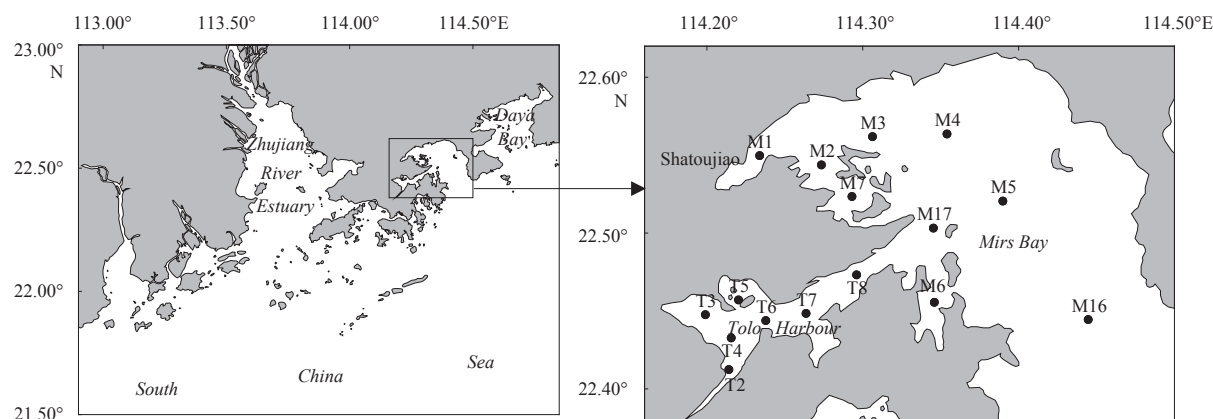
The Mirs Bay, a semi-enclosed bay located in the NSCS, is embedded in the terrene about 22 km north-northwestwards,

and neighbored with the Zhujiang (Pearl) River Estuary westward and with the Daya Bay eastward. It covers about 382 km<sup>2</sup> and has a coastal length of about 210 km (including the coasts of Hong Kong and Shenzhen). It is fed by a few small and short streams and surrounded by hills and low mountains. The water-depth in the bay is generally greater than 16 m, being shallower to the northeast and deeper to the southwest, with a maximum depth of 22 m to the southwest and in the Chimen Channel (Li et al., 2014).

The Mirs Bay has a northerly subtropical climate and experiences a wet season for half the year during the summer period (April–September) and a dry season during the winter period (October–March). The catchment area of the bay is about 440 km<sup>2</sup>, and it has an annual total rainfall rate of about 2 398.5 mm/a and an annual total evaporation rate of about 1 227.3 mm/a (Hong Kong Observatory, 2015). The difference in total monthly rainfall between the wet and dry seasons was about 290.0 mm/month. For comparison, the difference in the total monthly evaporation between the wet and dry seasons was about 35.0 mm/month, which is much less than the rainfall. The annual total combined wet and dry deposition of inorganic phosphorus (IP) and organic phosphorus (OP) from the atmosphere near the bay was 10.4 and 11.0 kg/(km<sup>2</sup>-a), respectively (Chen et al., 2014). Between 2000 and 2014, in the streams draining into the bay, with *S* near zero, the average DIP and DOP concentrations were 0.089 and 0.080 mg/L, respectively. The annual total wastewater discharge from the cities of Hong Kong and Shenzhen into the catchment was about 40×10<sup>6</sup> m<sup>3</sup>/a, and the DIP and DOP concentrations in the wastewater were about 2.17 and 0.907 mg/L, respectively (Cooperative Research Group of Hong Kong and Shenzhen, unpublished data). The annual volume of freshwater (FW) discharged into the bay from the rainfall in the catchment, accounting for evaporation and total wastewater discharge, was 995×10<sup>6</sup> m<sup>3</sup>/a.

Using these presumptions and the DIP and DOP concentrations in the streams and the wastewater, and the annual deposition of IP and OP (all the deposited IP and OP were considered as DIP and DOP, respectively, in the waters), the average DIP and DOP concentrations in the FW discharged into the bay were calculated as 0.136 and 0.081 mg/L, respectively. The observed sedimentation rate in the bay was about 4.62 mm/a (Guan et al., 1995).

Sixteen monitoring sites, representing different geographical zones, were selected from the comprehensive sampling program of the EPD (Fig. 1). Seven sites (T2–T8) were representative of the



**Fig. 1.** Water quality monitoring sites in the Mirs Bay, northern South China Sea.

Tolo Harbour & Chimen Channel zone, one site (M1) represented the waters adjacent to Shatoujiao, and eight sites (M2–M7, M16 and M17) represented the main zone of the bay.

## 2.2 Sample collection and storage

A computer-controlled Rosette<sup>®</sup> water sampler linked to a SEACAT 19+ conductivity-temperature-depth (CTD) profiler was used to collect water samples. The samples were collected from three depths: the surface (1 m below the sea surface), middle (half the water depth) and bottom (1 m above the seabed). When the water depth was  $\geq 4$  m and  $< 6$  m, the middle sample was omitted. When the water depth was  $< 4$  m, only the surface sample was collected. Before use, all the glass-sampling containers were rinsed with hot, dilute HCl and then rinsed several times with reagent water. For Chl *a*, 500-mL samples were concentrated by vacuum filtration (maximum  $0.3 \times 10^5$  Pa) as soon as possible after collection through a membrane filter (0.45- $\mu$ m porosity and 47-mm diameter). The concentrated samples, on the filters, were placed in airtight plastic bags and stored frozen and in the dark during transport to the laboratory for analyses. For nutrients (including DIP and DOP), the samples were filtered immediately after collection through 0.45- $\mu$ m membrane filters. Before use, the membrane filters were cleaned by soaking them in distilled water for at least 24 h. Subsamples were collected in PET bottles, immediately preserved in an upright position at  $-20^\circ\text{C}$  in a freezer during transport to the laboratory for analysis.

## 2.3 Temperature, salinity, pH, dissolved oxygen and Secchi disc depth measurement

A SEACAT 19+ CTD profiler, equipped with an instrumental SBE 18 pH sensor and a SBE23Y DO sensor, was used to measure temperature (*T*), *S*, pH and dissolved oxygen (DO) in the field. Secchi disc depths were measured with a Secchi disc depth plate according to the instructions of the manufacturer.

## 2.4 Chl *a* measurement

Chl *a* concentrations were determined using a spectrophotometer in the laboratory. The pigments were extracted from the plankton concentrates by shaking samples in 10-mL 90% aqueous acetone at  $4^\circ\text{C}$  for 24 h. The particles were then separated centrifugally, and the optical density (absorbance) of the 90% acetone extract was determined at light wavelengths of 664 and 665 nm before and after acidification, respectively. The  $\rho[\text{Chl } a]$  ( $\text{mg}/\text{m}^3$ ) was calculated using the following equation:

$$\rho[\text{Chl } a] = 26.7(664_b - 665_a) \times V_1 / (V_2 \times W), \quad (1)$$

where  $V_1$  is the volume of the extract in L,  $V_2$  is the volume of the sample in  $\text{m}^3$ ,  $W$  is the light-path width or length of the cuvette in cm,  $664_b$  and  $665_a$  are the optical densities of the 90% acetone extract before and after acidification, respectively. The value 26.7 is the absorbance correction (APHA, 1998a).

## 2.5 DIP and DOP measurement

The subsamples were defrosted at room temperature and the DIP or TDP was determined with a flow-injection auto analyzer following the standard operating procedure of the laboratory based on APHA (1998b). Subsamples, before measuring the TDP, were digested with sulfuric acid following the digestion procedure of the laboratory. The DOP was calculated from the equation,  $\rho[\text{DOP}] = \rho[\text{TDP}] - \rho[\text{DIP}]$ .

## 2.6 Quality control and uncertainty

*T*, *S*, pH, DO and Secchi disc depth had been measured by the EPD. Chl *a*, DIP and TDP had been measured by the Government Laboratory (GL) of Hong Kong. Both the EPD and the GL hold regular in-house training classes for various grades of staff. The EPD carry out regular metrological verification or calibration using contrast tests for the instrumental thermistor, the electrical conductivity sensor, the pH electrodemetic sensor, the DO membrane electrode sensor on the CTD, and the Secchi disc depth plate. The GL carry out regular metrological verification or calibration for all the test instruments.

Data uncertainty was determined by comparing duplicate analyses of the samples. At least 10% of all samples were analyzed in duplicate. Uncertainties or reporting limits for the *T*, *S*, Secchi disc depth, DO, Chl *a*, DIP and TDP were  $0.1^\circ\text{C}$ , 0.1, 0.1 m,  $0.1 \text{ mg}/\text{L}$ ,  $0.2 \text{ mg}/\text{m}^3$ ,  $0.002 \text{ mg}/\text{L}$  and  $0.02 \text{ mg}/\text{L}$ , respectively (EPD, 2018).

## 2.7 Data treatment

Using all the measurements recorded between 2000 and 2014, the DOP concentrations, in the water column (including the surface, middle, and bottom levels) for the various sites, were averaged per month. The January, April, July and October averages were chosen to represent winter, spring, summer and autumn, respectively, to analyze the seasonal variability of DOP. The Surfer<sup>®</sup> software, Version 8.0C, was used to construct contour plots of the horizons. The surface, middle and bottom DOP concentrations from all the sites were averaged for each month to analyze the intra-annual variability of DOP. The DOP concentrations from all the sites were averaged for each of the cruises to analyze the inter-annual variability of DOP. In addition, during the regression analyses, data points, in which any party in a pair of data was under its uncertainty or reporting limit, were discarded, and a filter was set up to exclude some scattered data points to exclude accidental experimental errors. The exclusion criteria for scattered points were that the distance to the regressive line was nearly the farthest and the final regressive correlation coefficient ( $R^2$ ) nearly hit the maximum; however, the number of excluded scattered points was controlled to less than 1%.

## 3 Results and discussion

### 3.1 Variability of the hydrographical and biogeochemical settings

A northeastern monsoon prevails in winter and a southwestern monsoon prevails in summer over the NSCS coastal waters; the hydrographical and biogeochemical settings in the Mirs Bay are strongly influenced by the monsoons. Surface- and bottom-waters are well mixed during the dry season, whereas strong density stratification occurs in the water column during the wet season (Li et al., 2014). Therefore, the *T*, *S*, pH, DO, Chl *a* and DIP concentrations do not differ very much throughout the water column during the dry season, but they differ greatly during the wet season. The *T*, *S*, pH, DO, Chl *a* and DIP concentrations averaged for the surface- and bottom-waters at various sites during the dry and wet seasons between 2000 and 2014 are listed in Table 1.

The average surface *T* ( $21.0^\circ\text{C}$ ) was slightly higher than the average bottom *T* ( $20.6^\circ\text{C}$ ) during the dry season, and during the wet season, the average surface *T* ( $27.2^\circ\text{C}$ ) was much higher than the average bottom *T* ( $24.9^\circ\text{C}$ ). In contrast, the average surface *S* (32.0) was slightly lower than the average bottom *S* (32.4) during the dry season, and the average surface *S* (30.5) much lower than the average bottom *S* (32.6) during the wet season. The pH and

**Table 1.** *T*, *S*, pH, DO, Chl *a* and DIP concentrations averaged for surface- and bottom-waters at various sites for the dry and wet seasons<sup>1)</sup> in the Mirs Bay between 2000 and 2014

Period	Site	<i>T</i> /°C		<i>S</i>		pH		DO/mg·L <sup>-1</sup>		Chl <i>a</i> /mg·m <sup>-3</sup>		DIP/mg·L <sup>-1</sup>	
		Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom
Dry season	M1	21.2	20.8	31.9	32.3	8.1	8.1	7.1	7.3	8.4	5.6	0.011	0.010
	M2	21.2	20.6	32.3	32.5	8.1	8.1	7.1	7.2	3.8	3.1	0.007	0.008
	M3	20.9	20.5	32.5	32.6	8.1	8.1	7.1	7.2	2.8	2.4	0.007	0.008
	M4	20.8	20.4	32.5	32.7	8.1	8.1	7.0	7.1	2.4	2.6	0.008	0.008
	M5	20.7	20.4	32.5	32.7	8.1	8.1	7.2	7.1	2.2	2.3	0.007	0.008
	M6	20.5	20.3	32.5	32.5	8.0	8.1	7.0	7.2	2.9	2.6	0.007	0.007
	M7	21.0	20.5	32.4	32.5	8.1	8.1	6.8	7.1	3.1	3.1	0.007	0.007
	M16	20.8	20.4	32.6	32.8	8.0	8.1	7.2	7.1	1.8	1.8	0.008	0.009
	M17	20.7	20.4	32.4	32.6	8.1	8.1	6.9	7.1	2.9	2.3	0.007	0.008
	T2	21.0	20.8	30.8	31.4	8.0	8.0	6.7	7.0	9.0	7.0	0.007	0.008
	T3	21.3	20.9	31.3	32.0	8.1	8.1	7.2	6.9	9.7	5.8	0.006	0.006
	T4	21.1	20.8	31.2	32.0	8.1	8.0	7.1	6.6	7.4	5.0	0.005	0.006
	T5	21.3	21.1	31.6	31.9	8.1	8.0	6.6	7.0	4.8	4.8	0.006	0.006
	T6	21.0	20.6	31.6	32.4	8.1	8.0	7.0	6.4	5.5	3.0	0.005	0.007
	T7	21.0	20.5	31.8	32.4	8.1	8.1	6.9	6.5	4.0	3.4	0.005	0.007
	T8	20.8	20.4	32.1	32.6	8.1	8.1	6.8	6.6	3.1	2.2	0.006	0.009
	Average		21.0	20.6	32.0	32.4	8.1	8.1	7.0	7.0	4.6	3.6	0.007
Wet season	M1	27.5	26.1	30.3	31.8	8.3	8.1	7.1	5.9	12.0	5.4	0.009	0.010
	M2	27.3	25.1	30.9	32.6	8.3	8.1	6.7	4.9	4.6	2.7	0.005	0.011
	M3	27.2	24.0	31.2	33.2	8.2	8.0	6.6	4.5	3.7	2.0	0.006	0.013
	M4	26.9	23.8	31.5	33.4	8.2	8.0	6.4	4.8	2.3	2.0	0.006	0.011
	M5	26.9	23.6	31.4	33.6	8.2	8.0	6.5	4.7	2.1	2.0	0.006	0.011
	M6	26.7	24.6	31.2	32.8	8.1	8.0	6.4	5.1	2.8	2.7	0.005	0.010
	M7	27.3	24.8	31.0	32.8	8.2	8.1	6.5	4.7	3.8	3.1	0.005	0.013
	M16	26.8	23.3	31.6	33.9	8.2	8.0	6.7	5.0	1.6	1.4	0.005	0.011
	M17	26.9	23.7	31.2	33.4	8.2	8.0	6.5	4.5	3.3	1.8	0.005	0.013
	T2	27.3	26.8	29.0	30.8	8.2	8.1	6.9	6.6	11.4	6.9	0.007	0.007
	T3	27.6	25.9	29.2	31.9	8.3	8.1	7.4	5.5	12.8	4.7	0.007	0.008
	T4	27.4	25.6	29.3	32.1	8.3	8.0	7.3	5.0	8.5	4.3	0.006	0.010
	T5	27.9	27.0	29.6	31.1	8.2	8.1	6.7	6.6	7.2	6.1	0.006	0.007
	T6	27.4	24.8	29.9	32.7	8.2	8.0	6.9	4.1	7.4	2.7	0.006	0.013
	T7	27.1	24.9	30.4	32.6	8.2	8.0	6.8	4.8	5.5	3.7	0.006	0.011
	T8	27.1	23.8	30.7	33.3	8.2	7.9	6.7	3.5	5.1	1.7	0.006	0.017
	Average		27.2	24.9	30.5	32.6	8.2	8.0	6.8	5.0	5.9	3.3	0.006

Note: <sup>1)</sup> The dry season is October–March and the wet season is April–September.

DO concentration were about 8.1 and 7.0 mg/L, respectively, and did not show much difference between the surface- and the bottom-waters during the dry season; however, during the wet season, they were about 8.2 and 6.8 mg/L from the surface-water and about 8.0 and 5.0 mg/L from the bottom-water, respectively. The average of surface Chl *a* concentration was 4.6 mg/m<sup>3</sup> in the dry season and 5.9 mg/m<sup>3</sup> in the wet season, while the average of bottom Chl *a* concentration was 3.6 mg/m<sup>3</sup> in the dry season and 3.3 mg/m<sup>3</sup> in the wet season. The surface and bottom DIP concentrations were about 0.007 and 0.008 mg/L in the dry season and about 0.006 and 0.011 mg/L in the wet season, respectively.

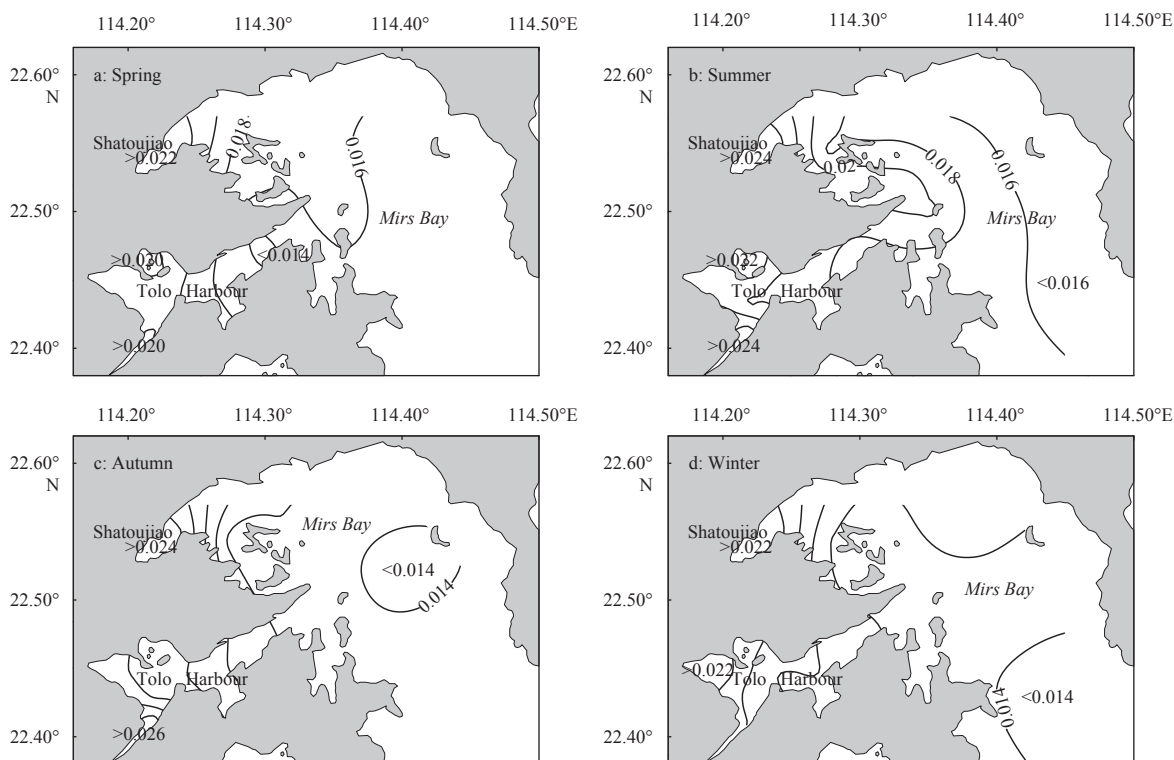
Table 1 shows that the surface *S* was lower and the Chl *a* concentration was higher in the inner Tolo Harbour and the waters adjacent to Shatoujiao than the main zone of the bay for both the dry and wet seasons. A similar DIP gradient was also observed in the bay, which implied an impact from the seaward discharges from the cities of Hong Kong and Shenzhen.

During the wet season, the bottom concentration of DIP was higher than the surface concentration, because autotrophic production mainly occurred in the upper water column and the DIP

was consumed in the surface-water. In addition, density stratification occurred in the water column and limited the exchange of DIP between the surface- and bottom-waters in the bay, and the southwestern monsoon, over the NSCS coastal waters, drives the coastal continental shelf bottom water (CCSBW) into the bay during summer (Li et al., 2014). Based on the measurements in the same period at the three sites (M13–M15) located in the coastal waters adjacent to the bay (Li et al., 2014), the CCSBW is characterized by low *T*, high *S* and high levels of inorganic nutrients. The CCSBW brings bottom-water with high levels of DIP into the bay during summer.

### 3.2 Temporal and spatial distributions of DOP concentrations

In the bay, the DOP concentration was (0.017±0.010) mg/L between 2000 and 2014. During spring, summer, autumn and winter, it ranged from 0.014–0.023, 0.015–0.025, 0.014–0.027, and 0.013–0.023 mg/L with an average of 0.017, 0.019, 0.018, and 0.017 mg/L, respectively. Figure 2 illustrates the horizontal contour plots of DOP concentration averaged for spring, summer, autumn, and winter over the period studied. Figure 2 shows that



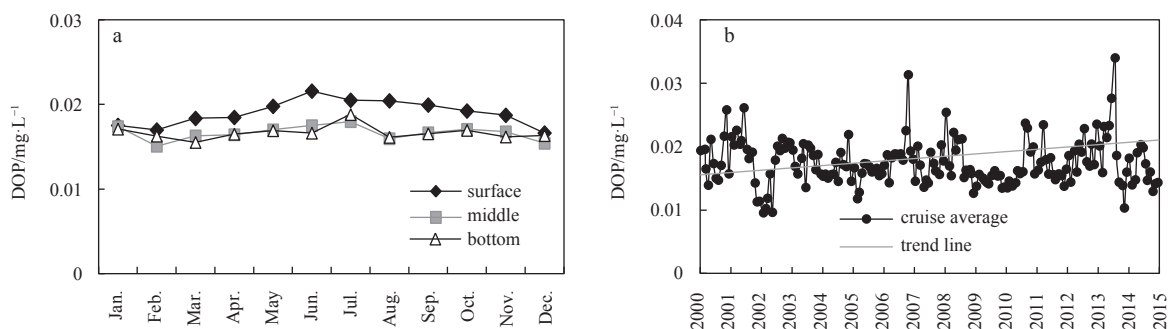
**Fig. 2.** Horizontal distribution of the average DOP concentrations (mg/L) in the Mirs Bay between 2000 and 2014. a. Spring, b. summer, c. autumn, and d. winter.

the DOP concentration had the same horizontal distribution trends for all the various seasons; it was higher in the inner Tolo Harbour and the waters adjacent to Shatoujiao than the main zone of the bay with small seasonal changes.

Figure 3 illustrates the intra- and inter-annual variabilities of the monthly and cruise averaged DOP concentrations in the bay between 2000 and 2014. As shown in Fig. 3a, the monthly averaged DOP concentration was higher from the surface-water than from the bottom-water; it varied with a similar annual periodicity, but the intra-annual variability was not big for the surface-, middle- and bottom-waters. The maximum DOP concentration, about 0.019 mg/L, occurred in June and the minimum, about 0.016 mg/L, occurred in February. As illustrated in Fig. 3b, the DOP concentration wavered around 0.017 mg/L with some pulse signals at times. It almost showed a trend of no change throughout the whole period studied.

Summarized from the above, the DOP concentration was

higher in the inner Tolo Harbour and the waters adjacent to Shatoujiao than in the main zone of the bay. The TDP and DIP concentrations exhibited the same pattern (Zhou et al., 2011, 2012) mainly due to the influence of the seaward discharges from the cities of Shenzhen and Hong Kong and the limitations of the natural environmental conditions around the Tolo Harbour and the waters adjacent to Shatoujiao. Both the Tolo Harbour and the waters adjacent to Shatoujiao are semi-enclosed with poor hydrodynamic conditions and weak water-exchange capability. This makes it difficult for the seaward discharges to spread outwards, and high levels of nutrients remain in both zones all year round, especially in the inner Tolo Harbour where red tides are often driven and spread to other places in the bay (Leng and Jiang, 2004). The DOP concentration was higher from the surface-water compared to the bottom-water because (1) the DOP concentration in the FW discharged into the bay was much higher than the DOP concentration in the bay water (BW), (2) the auto-

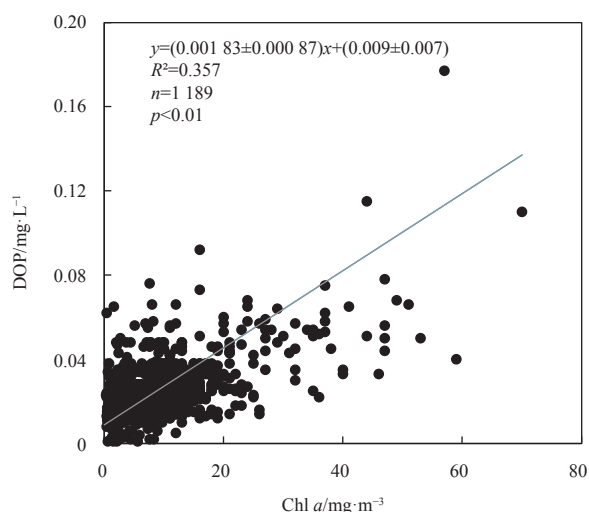


**Fig. 3.** Variabilities of DOP concentration in the Mirs Bay between 2000 and 2014. a. The intra-variability of the monthly average and b. inter-annual variability of the cruise average.

trophic production mainly occurred in the upper water column, and (3) strong density stratification occurred in the water column during summertime (Li et al., 2014), which blocked vertical mixing of the upper and lower waters. The surface DOP concentration started increasing in March and hit an annual maximum in June (Fig. 3a), which indicates that the DOP was accumulating in the surface-water during this period. After its maximum, the surface DOP concentration maintained at a high level of about 0.020 mg/L until September and then decreased to the minimum in December; it then remained at a low level of about 0.017 mg/L until February in the next year, which revealed that the DOP was reduced in the surface-water during the other periods. The seasonal changes and annual variability of the DOP concentration were small, which implied that the ecosystem was stable and the bay had a high capacity for self-purification. With regard to the inter-annual variability, the DOP concentration almost maintained a trend of no change while the dissolved organic nitrogen (DON) concentration exhibited an increasing trend (Li et al., 2013a). This implied that the terrestrial DON input had increased but the terrestrial DOP input was almost unchanged between 2000 and 2014. Jin and Liu (2013) also showed that nitrogen input has increased but phosphorus input decreased in the coastal waters of the China seas over the past few decades.

### 3.3 Correlation of DOP with Chl *a*

The concentration of Chl *a* is a measure of the phytoplankton standing crop. Growth and proliferation of phytoplankton directly or indirectly cause the release of DOP (Orrett and Karl, 1987); in general, this share of DOP is quickly utilized by plankton. Regression of the surface DOP with the Chl *a* in the bay between 2000 and 2014 showed that the DOP closely correlated with the Chl *a* with a linear correlative coefficient ( $R^2$ ) of 0.36 (Fig. 4), which reflected that the *in-situ* production of phytoplankton was a major source of DOP in the bay. As illustrated in Fig. 4, the regressive line slope  $\times 1\,000$  was about 1.83 (g P)/(g Chl *a*), which represented the DOP release rate in the phytoplankton production process; this was proportional to the Chl *a* concentration in the bay. The linear intercept on the  $x$ -axis was 0.009 mg/L, which represented the share of DOP that was refractory or less utilized by the plankton, accounting for about 53% of the total DOP. This implied that only half of the DOP pool was rapidly util-



**Fig. 4.** Regression of the surface DOP with chlorophyll *a* (Chl *a*) in the Mirs Bay between 2000 and 2014.

ized and cycled in the bay, although there is no direct evidence on hands to support this. Phosphate has become an ultimately limiting factor for harmful algal blooms in the bay (Zhou et al., 2011).

There was a close, positive linear correlation between the DOP and Chl *a*, which reflected that the DOP is closely correlated with the phytoplankton standing crop. The DON is also closely correlated with the phytoplankton standing crop in the bay (Li et al., 2013a). Hu and Smith (1998) showed that the release of DON is highly correlated with PP. Based on the particulate organic matter (POM) and Chl *a* measurements, the release rate of the particulate organic carbon (POC) during the process of phytoplankton production was 42.6 (g C)/(g Chl *a*) in the bay, from which the release rate of the particulate organic phosphorus (POP) was calculated as about 1.04 (g P)/(g Chl *a*), according to the Redfield ratio (Redfield et al., 1963). Comparing the release rate of DOP, about 1.83 (g P)/(g Chl *a*), to the release rate of POP, the release rate of DOP was about 1.8 times higher than for POP during the phytoplankton production process.

### 3.4 Residence time of DOP ( $T_{DOP}$ )

Given that the release rate for DOP was about 1.8 times higher than for POP during the phytoplankton production process, the  $T_{DOP}$  was estimated using the same method used to calculate the residence time of the POM (Liu et al., 1995; Li et al., 2013b). A simple formula from Cadée and Hegeman (1974) was used to calculate the PP (g/(m<sup>2</sup>·d), calculated by carbon):

$$PP = \rho [\text{Chl } a] \times A \times E \times L / 2, \quad (2)$$

where  $A$  is the assimilation coefficient, using 3.7 (mg C)/((mg Chl *a*)·h) from Ryther and Yentsch (1957);  $E$  is the euphotic depth (m), using three times the Secchi disc depth (Yamaguchi and Ichimura, 1972); and  $L$  is the length of daytime, using 12 h as the average. The integrated DOP (IDOP) was then integrally calculated:

$$IDOP = \sum_{i=1}^{n-1} (DOP_i + DOP_{i+1})(D_{i+1} - D_i) / 2, \quad (3)$$

where  $i=1, 2, \dots, n-1$ ;  $D_i$  is the  $i$ th sampling depth from the surface to the bottom at a site; and  $DOP_i$  is the DOP concentration at the  $i$ th sampling depth.  $T_{DOP}$  was then obtained from the following equation:

$$T_{DOP} = IDOP / (1.8PP \times 31 / 1\,272), \quad (4)$$

where 31 is the total atomic weight of phosphorus and 1 272 is the total atomic weight of carbon from the molecular formula for phytoplankton by Redfield et al. (1963). The results obtained using Eq. (4) are listed in Table 2.

The  $T_{DOP}$  ranged from 2.5–13.4 d with an average of 7.2 d and was proportional to the water depth (Table 2). This pattern was identical to those for the residence times of DON and POM in the bay (Li et al., 2013a, b). In the shallow water of the inner Tolo Harbour and the waters adjacent to Shatoujiao, where the seaward discharges from the cities of Hong Kong and Shenzhen usually occur, the PP was high and the IDOP was low, resulting in the short  $T_{DOP}$ . Therefore, it was concluded that in the Mirs Bay, the DOP is rapidly cycled.

The DOP residence times, estimated above, presume that the autochthonous production is the sole input for the DOP, besides

**Table 2.** Average residence times for DOP at various sites in the Mirs Bay between 2000 and 2014

Site	Water depth/m	IDOP/g·m <sup>-2</sup>	PP/g·m <sup>-2</sup> ·d <sup>-1</sup>	T <sub>DOP</sub> /d
M1	6	0.135	1.126	2.8
M2	11	0.181	0.720	5.9
M3	16	0.249	0.643	9.1
M4	18	0.279	0.512	12.7
M5	20	0.290	0.506	13.4
M6	12	0.182	0.569	7.5
M7	13	0.201	0.657	7.2
M16	17	0.237	0.447	12.4
M17	22	0.340	0.712	11.2
T2	4	0.093	0.878	2.5
T3	7	0.150	1.346	2.6
T4	8	0.156	1.106	3.3
T5	4	0.081	0.524	3.6
T6	12	0.218	1.100	4.8
T7	11	0.193	0.812	5.6
T8	22	0.358	0.797	10.5

the assumption of a steady state system. However, riverine export, atmospheric deposition, and sewage discharge are other sources of DOP in the bay. To determine how much they influence the estimated results, if the mean PP is 0.778 g/(m<sup>2</sup>·d) from Eq. (2), then the total autochthonous production of DOP can be calculated as 4 860 (t P)/a using the areal extent of the bay, the DOP release rate in the bay, and the Redfield ratios. Applying the mean DOP concentration in the stream FW and using the total rainfall and total evaporation for the catchment area, the total riverine export of DOP can be estimated at about 41.4 (t P)/a, which accounts for about 0.85% of the total autochthonous DOP production. Based on the annual total wastewater discharge and the mean DOP concentration in the wastewater, the total sewage discharge of DOP can be calculated as about 36.3 (t P)/a, which accounts for about 0.75% of the total autochthonous DOP production. Based on the total wet and dry deposition of OP and the areal extent of the bay, the total atmospheric deposition of DOP can be estimated at about 4.3 (t P)/a, which accounts for about 0.09% of the total autochthonous DOP production. In addition, the sediment-water interface might be another source of DOP in the shallow waters. The sedimentary organic matter (SOM) release rate in the lower Zhujiang Estuary has been observed to be about 1.15 mmol/(m<sup>2</sup>·d) (calculated by NH<sub>3</sub>) (Pan et al., 2002). Assuming that the SOM release rate is the same as the Zhujiang River Estuary, the total SOM release from the sediments of the bay, using the areal extent of the bay, can be estimated at about 273 (t P)/a, which accounts for about 5.61% of the total autochthonous DOP production. However, He et al. (2010) showed that most of the SOM, released from sediments at the sediment-seawater interface, is in the form of inorganic matter. Therefore, the DOP residence time estimated above should have a confidence level greater than 95%.

### 3.5 Autotrophic production and heterotrophic removal of DOP

The two competing biogeochemical processes, autotrophic production and heterotrophic removal, for organic matter are known to exist globally in upper open oceans and coastal waters. Heterotrophic bacteria selectively remove organic matter from solution for their metabolic needs, and autotrophic bacteria and phytoplankton synthetically produce and, ultimately, release organic matter to solution (Anderson and Zeutschel, 1970; Cotner

and Biddanda, 2002; Ward and Bronk, 2001). In Section 3.3, the DOP released from the autotrophic production was proportional to the Chl *a* concentration in the field. The release rate was found to be about 1.83 (g P)/(g Chl *a*) from the regression of DOP with Chl *a*. To determine the rate of heterotrophic removal of DOP, it was necessary to quantitatively determine the amount of excess DOP ( $\Delta$ DOP) in the autotrophic production and the heterotrophic removal processes in the bay as follows. Water in the bay can be considered as a mixture of FW, including the rainwater drained into the bay from the catchment areas, and seawater (SW), intruded into the bay from the open sea. The FW and SW fractions of the water in the bay were estimated by a two-component mixing mass balance model (Lee and Krothe, 2001; Li et al., 2013a, 2014) in combination with the *S* measurements. Based on the theory of mass balance, the following equations can be derived:

$$w[\text{FW}] + w[\text{SW}] = 1, \quad (5)$$

$$S_{\text{BW}} = w[\text{FW}]S_{\text{FW}} + w[\text{SW}]S_{\text{SW}}, \quad (6)$$

where  $w[\text{FW}]$  and  $w[\text{SW}]$  are the FW and SW fractions, respectively; and  $S_{\text{FW}}$ ,  $S_{\text{SW}}$ , and  $S_{\text{BW}}$  are the *S* of the FW, SW, and BW, respectively. On the assumption that  $S_{\text{FW}} \approx 0$ , the following equations are derived from Eqs (5) and (6):

$$w[\text{FW}] = 1 - S_{\text{BW}}/S_{\text{SW}}, \quad (7)$$

$$w[\text{SW}] = S_{\text{BW}}/S_{\text{SW}}. \quad (8)$$

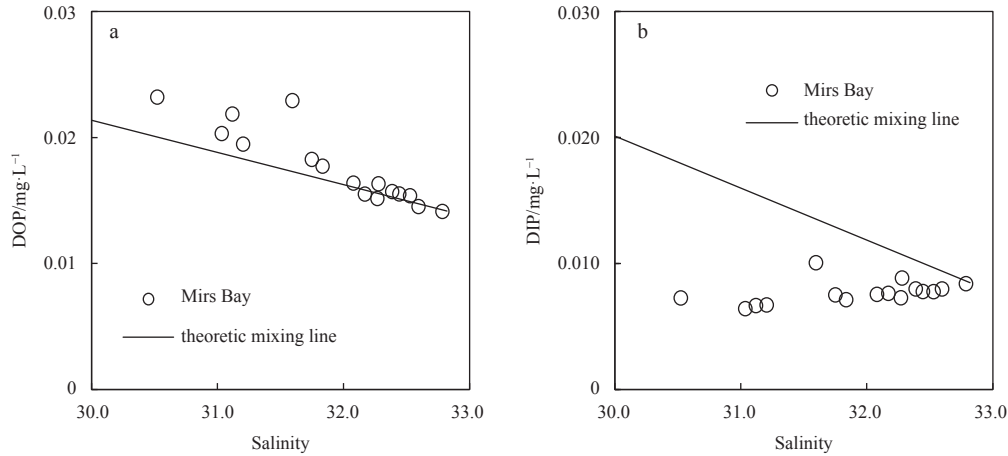
Then the  $\Delta$ DOP resulting from the autotrophic production and the heterotrophic removal processes can be determined by the following equation:

$$\begin{aligned} \Delta\text{DOP} &= \rho[\text{DOP}_{\text{BW}}] - (w[\text{FW}] \times \rho[\text{DOP}_{\text{FW}}] + w[\text{SW}] \times \rho[\text{DOP}_{\text{SW}}]) \\ &= \rho[\text{DOP}_{\text{BW}}] - (1 - S_{\text{BW}}/S_{\text{SW}}) \times \rho[\text{DOP}_{\text{FW}}] - \\ &\quad S_{\text{BW}}/S_{\text{SW}} \times \rho[\text{DOP}_{\text{SW}}], \end{aligned} \quad (9)$$

where  $\rho[\text{DOP}_{\text{FW}}]$ ,  $\rho[\text{DOP}_{\text{SW}}]$  and  $\rho[\text{DOP}_{\text{BW}}]$  are the DOP concentrations in the FW, SW and BW, respectively.

The *S* and DOP concentration were about 32.8 and 0.014 mg/L, respectively, in the SW adjacent to the bay, during the period between 2000 and 2014. The DOP concentration was about 0.081 mg/L in the FW discharged into the bay for the same period. On the assumption that the *S* of the total rainwater and wastewater discharged into the bay was near zero, a ligature for the data points of *S*-DOP in the SW and FW could be regarded as theoretical mixing line of *S* and DOP concentration from the SW and FW while the autotrophic production is totally balanced by the heterotrophic removal in the bay. Figure 5a illustrates the *S* and DOP concentrations averaged at various sites in the bay between 2000 and 2014 with the theoretical mixing line. As shown in Fig. 5a, the DOP concentration decreased as the *S* increased, which implied the dilution of DOP in the bay by SW from the open sea. The average  $\Delta$ DOP, at various sites in the bay, calculated from Eq. (9) are listed in Table 3. The  $\Delta$ DOP ranged from 0.000 to 0.006 mg/L with an average of 0.002 mg/L. The data points of *S*-DOP in the main zone of the bay were around the theoretical mixing line with slightly positive or negative deviations. The absolute values were even less than the DOP measurement uncertainty, which suggested that the autotrophic production was nearly balanced





**Fig. 5.** Correlation of the salinity with DOP and DIP concentrations averaged at various sites in the Mirs Bay between 2000 and 2014. a. Salinity with DOP and b. salinity with DIP.

**Table 3.** Average  $\Delta$ DOP and  $\Delta$ DIP at various sites in the Mirs Bay between 2000 and 2014

Site	Water depth/m	$\Delta$ DOP/mg·L <sup>-1</sup>	$\Delta$ DIP/mg·L <sup>-1</sup>
M1	6	0.006	-0.003
M2	11	0.001	-0.004
M3	16	0.001	-0.002
M4	18	0.001	-0.002
M5	20	-0.000	-0.001
M6	12	-0.000	-0.003
M7	13	0.000	-0.003
M16	17	-0.000	0.000
M17	22	0.001	-0.002
T2	4	0.004	-0.010
T3	7	0.004	-0.008
T4	8	0.002	-0.008
T5	4	0.003	-0.009
T6	12	0.002	-0.005
T7	11	0.002	-0.005
T8	22	0.001	-0.002

by the heterotrophic removal of the DOP. Those in the inner Tolo Harbour and the waters adjacent to Shaotoujiao were above the mixing line with small positive deviations, suggesting that autotrophic production dominated. Although the  $\Delta$ DOP was nearly equal to the DOP measurement uncertainty in the inner Tolo Harbour, the autotrophic production of DOP was statically significant and representative because it was derived from 15-year long-term observations.

The relative standard deviation for the S at various sites in the bay ranged from  $\pm 3.7\%$  to  $\pm 7.9\%$  with an average of  $\pm 4.9\%$ , and for the DOP, it was from  $\pm 38.2\%$  to  $\pm 81.4\%$  with an average of  $\pm 50.6\%$ . The absolute values for the relative  $\Delta$ DOP were much less than the relative standard deviations for the DOP, but they were comparable to the relative standard deviations for the S. Comparison of the relative  $\Delta$ DOP and the relative standard deviations for the S showed that the mixing line covered Sites M2–M7 and M16–M17 in the main bay and Sites T6–T8 in the inner bay. The relative measurement uncertainty was about 0.3% for the S and about 11.3% for the DOP. Therefore, the  $\Delta$ DOP error was mainly from the DOP measurements, and the  $\Delta$ DOP should be representative and statistically significant. Overall, the autotrophic pro-

duction of DOP was slightly dominant in the bay, which suggested that some excess DOP had been transported out of the bay to the open sea by water exchange.

The DIP concentration was  $(0.008 \pm 0.006)$  mg/L, which accounted for 32.0% of the total TDP, and the DOP concentration was  $(0.017 \pm 0.010)$  mg/L, which accounted for 68.0% of the total TDP in the bay over the period studied. As shown in Sections 3.1 and 3.2, the DIP concentration from the surface-water was lower than from the bottom-water, while the DOP concentration from the surface-water was higher than from the bottom-water. To better understand the relationship between DOP and DIP, the DIP was treated in the same way as the DOP to investigate the impact of the autotrophic production and heterotrophic removal processes on the DIP. The DIP concentrations of about 0.009 mg/L in the SW and 0.136 mg/L in the FW, over the period studied, were used to replace the DOP as used in the previous section. Therefore, a theoretical mixing line of the S and DIP concentration from the SW and FW could be found while the impact of the autotrophic production and the heterotrophic removal on the DIP in the bay was disregarded. Figure 5b illustrates the S and DIP concentrations averaged at various sites in the bay between 2000 and 2014 with the theoretical mixing line. As shown in Fig. 5b, all of the data points for S-DIP were under the mixing line and had negative deviations. The DIP replaced the DOP in Eq. (9), and the following equation was derived:

$$\begin{aligned} \Delta \text{DIP} &= \rho[\text{DIP}_{\text{BW}}] - (w[\text{FW}] \times \rho[\text{DIP}_{\text{FW}}] + w[\text{SW}] \times \rho[\text{DIP}_{\text{SW}}]) \\ &= \rho[\text{DIP}_{\text{BW}}] - (1 - S_{\text{BW}}/S_{\text{SW}}) \times \rho[\text{DIP}_{\text{FW}}] - \\ &\quad S_{\text{BW}}/S_{\text{SW}} \times \rho[\text{DIP}_{\text{SW}}], \end{aligned} \quad (10)$$

where  $\rho[\text{DIP}_{\text{FW}}]$ ,  $\rho[\text{DIP}_{\text{SW}}]$  and  $\rho[\text{DIP}_{\text{BW}}]$  are the DIP concentrations in the FW, SW and BW, respectively. The average  $\Delta$ DIP, at various sites across the bay, calculated from Eq. (10) are listed in Table 3. The  $\Delta$ DIP ranged from  $-0.010$  to  $0.000$  mg/L with an average of  $-0.004$  mg/L. There was a depletion of DIP that was much larger than the total excess of DOP in the bay. This revealed that the removal of DIP was greater than the release of DIP, which suggested that most of the depletion was achieved by DIP converting to POP, which sank to the bottom and was buried or transported to the open sea by water exchange.

The relative standard deviation for the DIP ranged from



$\pm 55.0\%$  to  $\pm 125.7\%$  with an average of  $\pm 74.4\%$ . The absolute values for the relative  $\Delta$ DIP were much larger than those for the relative  $\Delta$ DOP, which implied that the DIP was much more variable than the DOP. The absolute value for the relative  $\Delta$ DIP was less than the relative standard deviation for the DIP at each site in the main bay; and it was also less at some sites and bigger at the other sites in the inner bay. The mixing line covered Sites M4–M5, M16 and T8. The relative measurement uncertainty for the DIP was about 26.3% and about 0.3% for the S; therefore, the  $\Delta$ DIP error was mainly from the DIP measurements, and therefore, the  $\Delta$ DIP should also have great statistical significance. Overall, the biological assimilation of DIP was strongly dominant in the bay. This also supported the suggestion of dominative autotrophic production in the bay. Most of the DIP depletion should have been replenished from the open sea through water exchange; however, it was clear that the water exchange brought DIP from the open sea into the bay and removed DOP from the bay to the open sea at the same time. This implies that the Mirs Bay is a sink for atmospheric carbon dioxide.

SOM was found to be about 1.07% of the total sediment (by wet weight) based on the measurements of the surface sediments at 14 sites in the bay (Li et al., 2012) between 2000 and 2014. According to the method developed by Xu et al. (2017), about 75.8% of the SOM was from an aquatic source in the bay. Therefore, using the areal extent of the bay, the sedimentation rate, the sediment moisture content and specific gravity in the bay, and the Redfield ratios, 75.8% of the total sedimentary organic phosphorus (SOP) was estimated at 225 t/a, which accounted for about 4.63% of the total autochthonous DOP production. This share of the SOP, from the autochthonous POP, that had sunk to the bottom and was buried, was more than the total DOP input from the riverine export, wastewater discharge, and atmospheric deposition. This supports the above suggestion that most of the DIP depletion was the result of conversion to POP. Although the DIP removal processes also include physicochemical scavenging due to particle reactivity of phosphate, it could be considered that the removal of DIP by scavenging was small compared to the removal of DIP by biological assimilation, and its impact could be ignored. In conclusion, the Mirs Bay is a very productive and, at the same time, fairly heterotrophic bay.

#### 4 Conclusions

The monthly water quality monitoring data from the EPD, between 2000 and 2014, showed that the concentration of DOP was high in the inner Tolo Harbour and the waters adjacent to Shaotoujiao all year round. This implies that these two zones suffer from the heavy impact of seaward discharges from the cities of Hong Kong and Shenzhen. The surface DOP concentration increased between March and June and decreased during the rest of the year, which suggested that the DOP was accumulating in the surface-water during this period but not during the rest of the year. The seasonal changes and annual variability in the concentration of DOP were small, which implied that the ecosystem had a high capacity for self-purification and was stable within the bay.

The measurements showed that there was a close, positive linear correlation between the surface DOP and Chl *a*, reflecting that the DOP concentration was closely correlated with the phytoplankton standing crop in the bay. The DOP release rate was about 1.83 (g P)/(g Chl *a*), which was about 1.8 times higher than for the POP, and in combination with the IDOP, the  $T_{DOP}$  was estimated as short, about 7 d. Therefore, it was concluded that the DOP was rapidly cycled in the bay.

On the basis of the observed data during the period studied, the  $\Delta$ DOP from the autotrophic production and the heterotrophic removal processes was determined using a two-component mixing mass balance model to be about 0.000 mg/L in the main zone of the bay and about 0.003 mg/L in the inner Tolo Harbour and the waters adjacent to Shaotoujiao. This suggested that the autotrophic production of DOP was nearly balanced by the heterotrophic removal in the main zone of the bay and dominated in the inner Tolo Harbour and the waters adjacent to Shaotoujiao. In conclusion, the Mirs Bay is a very productive and, at the same time, fairly heterotrophic bay.

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