# Removal and Mass Balance of Phosphorus and Silica in the Turbidity Maximum Zone of the Changjiang Estuary

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**Abstract** The concentration of suspended particulate matter (SPM), sedimentation flux and various forms of phosphorus and silica in turbidity maximum zone (TMZ) in the Changjiang (Yangtze) estuary was studied. Based on the budget of P and Si, their mass balances in the TMZ were calculated. Research results show that the variation in concentration of dissolved inorganic silicon (DISi) was mainly controlled by sea water dilution in the Changjiang estuary, while that of dissolved inorganic phosphorus (DIP) was considerably affected by the buffering of suspended matter and sediment. The sedimentation fluxes of suspended particulate matter and particulate inorganic phosphorus (PIP), total particulate phosphorus (TPP), particulate inorganic silicon (PISi) and biological silicon (BSi) in the TMZ were 238.4 g $m^{-2}$ $d^{-1}$ and 28.3, 43.1, 79.0, 63.0 mg $m^{-2}$ $d^{-1}$, respectively. In addition, a simple method to estimate the ratio of resuspension of sediment in the TMZ was established, indicating that the sediment resuspension in the TMZ influenced significantly the mass balances of P and Si. Particulate-adsorbed P (60.8%) and 35.5% of TPP discharged from the river were filtered and then deposited in the TMZ. The input flux of PIP from the river mouth was 55.9% of that of DIP, being important as biologically available P, while that of PISi was only 3.5% of DISi, showing that particulate-adsorbed Si was much less important than particulate-adsorbed P.

**Keywords** Phosphorus · Silica · Suspended particulate matter · Transport Turbidity maximum zone · Changjiang (Yangtze) estuary

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Turbidity maximum zone (TMZ) of a river estuary is a dynamic section of the estuary directly related to circulation, tide, deposition and scouring (Shen and Pan 2001); it is characterized by high concentration of suspended matter, higher than upstream or downstream of the estuary (Gebhardt et al. 2005). The TMZ plays many important roles during estuarine mixing processes, such as transport and accumulation of suspended particulate matter (SPM), deposition and resuspension of surface sediment, and the formation and development of the estuarine deltaic region. It could also affect the chemical behaviour, removal, and fate of nutrient, organic matter and many heavy metals in the estuary. In an estuarine area, the TMZ acts as a filter between the terrestrial and the marine realm (Gebhardt et al. 2005). The filtering effect reflects almost all the processes in physical, chemical, geological and biological manners in the estuary. Hence, study in biogeochemistry of the TMZ has become a hot topic of marine sciences. Many previous researches have been reported on the regions of the Gironde estuary (Allen et al. 1980), Chesapeake Bay (Fisher et al. 1988), Saint Lawrence estuary (Hamblin 1989), Tamar estuary (Uncles and Stephens 1993), Fly estuary (Wolanski et al. 1995) and Changjiang estuary (Jiufa and Chen 1998; Gebhardt et al. 2005).

The suspended particulate matter in the TMZ is derived mostly from riverine sources and accumulates there, in which it has a longer residence time than in the river upstream (Herman and Heip 1999). High SPM concentration in the TMZ provides an ideal site for physical, chemical and biological interactions between dissolved and particulate species, as well as interaction among particulate species (Gebhardt et al. 2005). Previous studies showed that in the TMZ, P and Si are more easily adsorbed to and released by SPM than other nutrients (Mackenzie and Garrels 1965; Siever and Woodford 1973; Mayer and Gloss 1980; Fox et al. 1986; Froelich 1988; Lebo 1991; Bowes and House 2001). Especially, the release of P from SPM is one of the most abundant sources in marine P cycling (Froelich 1988). Froelich (1988) estimated that on a global scale, fluvial particulates could transport from 1.4 to 14 $\times$ $10^{10}$ mol $a^{-1}$ of reactive phosphate to the sea, some 2–5 times more than that in the dissolved load alone. Therefore, reactive phosphate in fluvial particulate is a potential biologically available source that cannot be ignored. It is known, however, excessive P entering estuary would cause eutrophication in coastal environments. Most SPM carried by rivers would deposit in the TMZ and part of dissolved components including P and Si would deposit later, which is significant for improving the estuarine and coastal environments. Lisitsyn (1995) calculated that in what he calls the "marginal filter", about 93–95% of the suspended and 20–40% of the dissolved riverine materials are deposited worldwide. Thus, it is very important to remove part of nutrient matters from riverine sources via deposition in the TMZ of an estuary.

The Changjiang River is the third largest in length and the fourth largest in sediment load in the world. The influence of the TMZ on transportation and deposition of sediment in Changjiang estuary has become a major issue in research (Milliman et al. 1985; He and Sun 1996; Jiufa and Chen 1998; Shen and Pan 2001; Chen et al. 2004). However, removal and delivery of P and Si in the TMZ in the estuary have not been fully understood. In the past 40 years, due to human activities including the construction of the Three Gorges Dam, eutrophication has become increasingly serious, and occurrence of red tides tended to increase in the Changjiang estuary. For

these reasons, this research was designed for studying the behaviour and removal of various forms of P and Si during estuarine mixing, and determining the sedimentation fluxes of SPM, P, and Si, estimating the mass balances of P and Si in the TMZ and seeking for scientific measures for remediation of eutrophication.

## 1 Study Area and Methods

The Changjiang estuary can be divided into three parts from the mouth seawards: upper, middle and lower estuaries. The upper estuarine is from Datong (the tidal stretch boundary) to Jiangyin (the tidal current boundary), 440 km long. The middle estuarine is from Jiangyin to the beach tip of river mouth, 210 km long, having river run-off and tidal current interacted and the river course branched. The lower estuarine is from the river mouth to the waters of 30–50 m isobath with a large submarine delta where tidal current dominated (Fig. 1). Near Xuliujing in the middle estuarine sector, the Changjiang River is divided into the South Branch and the North Branch, and the run-off and SPM are transported to the ocean mainly through the South Branch (Milliman et al. 1985). A special high SPM content zone stood out in the river mouth-bar area between the middle and lower estuaries, where the TMZ exists all year round. The TMZ was marked not only by high SPM concentration, but also by high wash-load content (Jiufa and Chen 1998). Tides in the estuary are regular semi-diurnal type. Turbidity maximum of the estuary is built in mechanisms of run-off–tidal current interaction and salt–freshwater mixing. The space–time distribution of turbidity maximum changes with the changes in run-off, tidal current and salt–freshwater mixing (Pan et al. 1999). The average run-off to the East China Sea was 29,000 $m^3\ s^{-1}$ or 928.2 $\times\ 10^9\ m^3\ a^{-1}$, of which most are formed in flood season from May to October. The average SPM content and annual total transport were 0.5–1.7 kg $m^{-3}$ and $5 \times 10^8$ t, respectively.

Research cruises were carried out during 29 February–9 March, 24 May–31 May, 30 August–4 September and 3–8 November 2004. Water samples were collected in 23 stations at 0, 5, 10, 20, 30 m and bottom layer (Fig. 1). Salinity (S), SPM, dissolved inorganic phosphorus (DIP), total phosphorus (TP, the sum of dissolved and particulate phosphorus), dissolved inorganic silicon (DISi), chlorophyll-*a* (Chl-*a*) and transparence were measured. Besides, sampling along a transect passing stations 38, 39, 40, 22, 23, 24 and 25 was conducted in May 2004 across the TMZ for determining particulate inorganic phosphorus (PIP), total particulate phosphorus (TPP), particulate inorganic silicon (PISi) and biological silicon (BSi). Sedimentation fluxes of SPM, P and Si were determined using Bloesch multi-tube sediment traps at stations 22 and 23 (water depth ~10 m). Each trap had five columniform sampling tubes (92 cm high and 6.5 cm inner diameter). The sampling system had been fully tested previously (Bloesch and Burns 1980; Zhang et al. 2004). The sediment traps were placed 3–4 m above the bottom for 24 h. After the traps were lifted up, most clear liquid in tubes was carefully removed. The residual water samples and particulate matters were transferred into polyethene bottles and filtered in situ with pre-weighed 0.45 μm Millipore filters. All the filtered membranes were rinsed with deionized water for desalination and stored in an icebox for determining PIP,

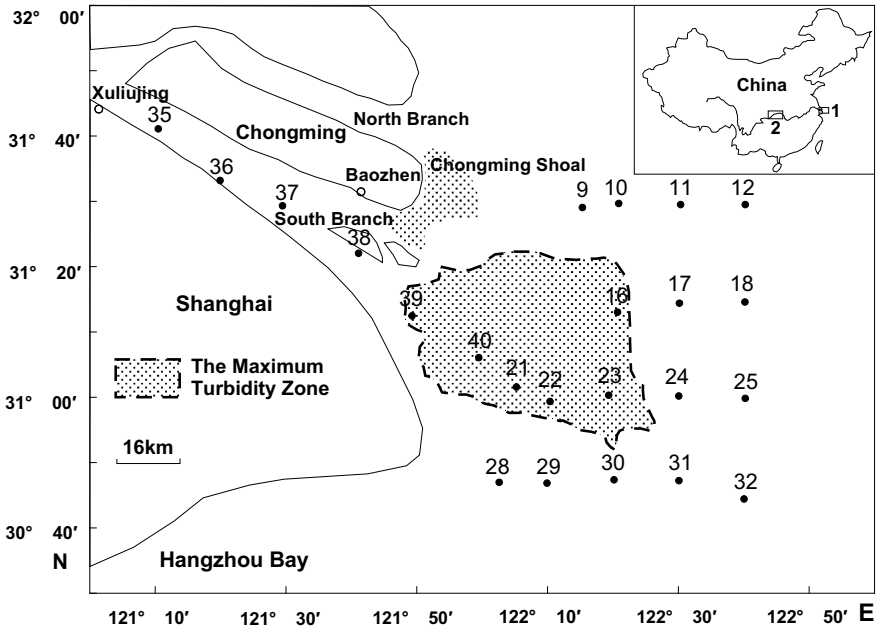


**Fig. 1** Sampling stations in the Changjiang estuary. The shaded area is the TMZ (Shen and Pan 2001); 1: study area; 2: Three Gorges Dam

TPP, PISi and BSi. The filtered membrane for determining particulate matter was dried to a constant weight at 60 °C and stored in a desiccator for further analysis. PIP and PISi were extracted using HCl. The HCl-extracted P was from apatite; laboratory and field studies showed that the phosphorus was released from apatite in natural environment (Smith et al. 1977, 1978; Fox et al. 1985, 1987). The TPP was measured using the method of TP (Koroleff and Grasshoff 1976). The BSi samples were measured as per Treguer and Gueneley (1988). The total inorganic P (TIP) was defined as the sum of DIP and PIP; total inorganic Si (TISi) was the sum of DISi and PISi; and total particulate Si was the sum of PISi and BSi.

## 2 Horizontal Distributions of Salinity, SPM, DIP, TP and DISi

### *2.1 Salinity and SPM*

Horizontal distributions of salinity and SPM in the study area in May are plotted in Fig. 2a and b. In May, the river experienced a season transition from dry to flood. The river-diluted water extended from the river mouth seawards, and salinity gradually increased (Fig. 2a). The salinity was higher in bottom than that in surface. Lower

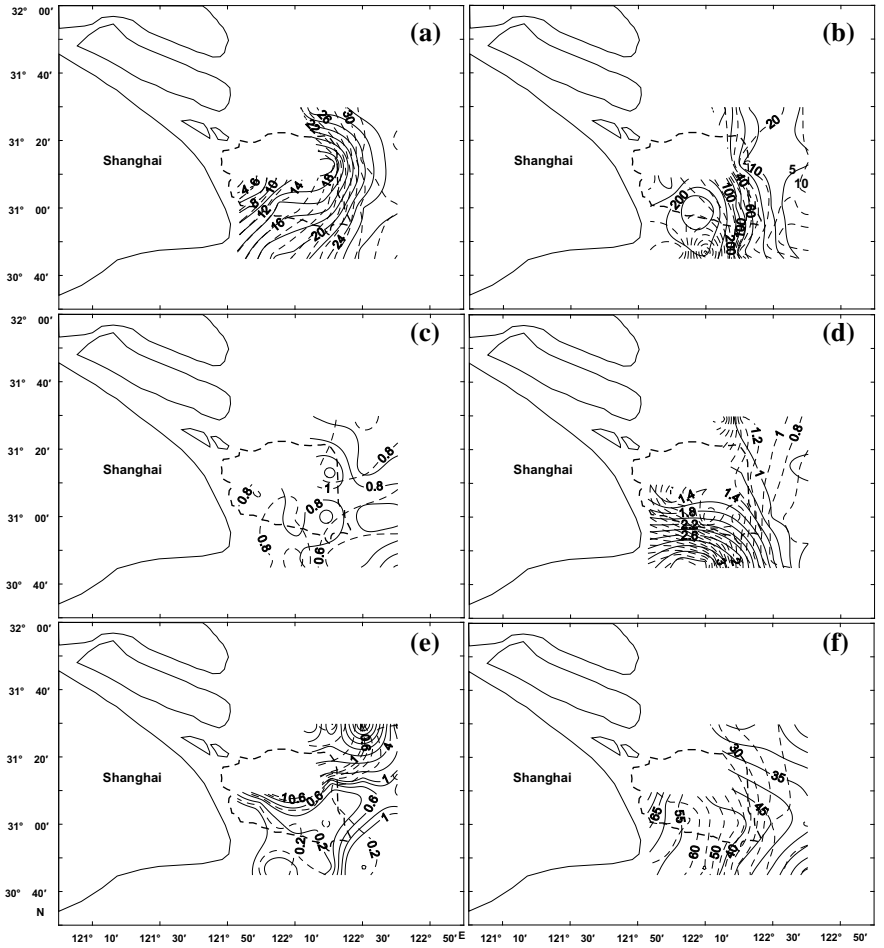


**Fig. 2** Horizontal distributions of S, SPM, DIP, TP, Chl-*a* and DISi; **a** salinity; **b** SPM (mg $L^{-1}$); **c** DIP (μmol $L^{-1}$); **d** TP (μmol $L^{-1}$); **e** Chl-*a* (μg $L^{-1}$); **f** DISi (μmol $L^{-1}$); solid line—surface; dashed line—bottom

salinity was found in most areas of the TMZ, less than 16 and 20 in surface and bottom layer waters, respectively.

SPM contents were 3.3–298.0 and 2.7–628.7 mg $L^{-1}$ in surface and bottom layers with an average of 76.9 ± 95.8 and 122.1 ± 163.0 mg $L^{-1}$, respectively, in the investigation area, and higher in bottom and lower in surface. High SPM content distributed mainly along the river mouth to the south-eastern waters of the mouth and decreased quickly from outer mouth towards the east and north-east (Fig. 2b). The average SPM content in the TMZ was 147.1 ± 120.1 mg $L^{-1}$ in surface layer and 214.8 ± 126.6 mg $L^{-1}$ in bottom layer. High SPM content was found in the west and south of the TMZ, which is consistent with moving direction of the Changjiang diluted water. From stations 21–23 south of the TMZ to station 24 east of the TMZ,

SPM contents in surface and bottom layers decreased from 291.3 to 8.0 mg $L^{-1}$ and from 284.3 to 13.3 mg $L^{-1}$, respectively. Rapid horizontal decrease in SPM content and very high SPM content in bottom layer (Fig. 2b) suggest that the SPM from the Changjiang River deposited mostly in the TMZ. The very high SPM content in bottom layer was probably due to sediment resuspension. A previous study showed that the turbidity maximum was formed by local resuspension of deposits and seabed erosion (Jiufa and Chen 1998). However, the highest SPM content in bottom layer was not inside the TMZ, but at station 29 to the south (Fig. 2b) at 628.7 mg $L^{-1}$, which was three times higher than that in surface layer, because of the proximity to the Hangzhou Bay (Qiantang River mouth, south of the Changjiang mouth) (Chen et al. 2004), in which tide movement was magnificent, such as famous Qiantang Tide there. Sedimentation in the bay mouth caused a large increase in SPM content in waters as flood and ebb tides moving back and forth. A part of SPM turned north-east and invaded the area beyond the Changjiang mouth and formed a locally high SPM concentration area (Yang et al. 1992).

## *2.2 DIP and TP*

The maximum concentration of DIP (1.3 μmol $L^{-1}$) in surface layer occurred outside the mouth at station 16 in the north-eastern part of the TMZ (Fig. 2c). At the station, low transparence (0.6 m) and Chl-*a* content (0.722 μg $L^{-1}$) indicated low biological influence; therefore, the high concentration of DIP was probably related to the P released from suspended matter. In addition, higher DIP concentrations (>0.8 μmol $L^{-1}$) were found in middle part of the investigation area and decreased towards north-east and south-west. DIP concentration was higher in most waters of the TMZ where SPM content was high, transparence was low (0.5–0.8 m) and Chl-*a* content was also low (0.125–0.440 μg $L^{-1}$). It has been shown that transparence was probably a main limiting factor for phytoplankton growth in the TMZ (Shen 1993a; Herman and Heip 1999), shown similarly in the Elbe, Schelde and Gironde estuaries in the western Europe (Muylaert and Sabbe 1999).

The distribution of TP concentration in surface and bottom layers (Fig. 2d) showed similar patterns to those of SPM. High TP concentration was observed mainly in the south-eastern parts outside the mouth. The highest concentration in surface and bottom layers was 3.5 and 4.5 μmol $L^{-1}$, respectively, and decreased towards the east and north-east. In the TMZ, higher TP concentration distributed in the parts west and south of the region, in maximum of 2.0 in surface and 3.1 μmol $L^{-1}$ in bottom layers; and both were >1 μmol $L^{-1}$ seen in other waters of the region. The maximum TP concentration occurred not in the TMZ but in the south of the TMZ. Similar distributions of both TP and SPM in the TMZ suggest that P was likely being adsorbed by suspended particles. TP was mainly composed of particulate P, especially in high TP concentration waters. The average DIP/TP ratio for both surface and bottom layers was 0.30, which was much lower than that of the whole investigation area (0.46). SPM contents were <10 mg $L^{-1}$ in the most areas to the

east of 122 30′ E where TP was mainly composed of DIP; and the average DIP/TP ratio was 0.82. In addition to the Changjiang River input, partial DIP came from dead and decomposed plankton, especially in the north-east part, where Chl-*a* content was very high (Fig. 2e).

### *2.3 DISi*

The distribution of DISi concentration was very different from that of DIP. High concentrations of DISi were found in the river mouth with the maximum inside the mouth. DISi concentration decreased greatly towards the east from the river mouth to the open waters (Fig. 2f). The isoline pattern of DISi was similar to that of the salinity but in opposite magnitude. The average concentration of DISi in surface and bottom layers was 44.2 and 37.0 μmol $L^{-1}$, respectively. The TMZ featured high DISi concentrations with averages of 69.8 μmol $L^{-1}$ in surface layer and 54.1 μmol $L^{-1}$ in bottom layer.

## 3 Variations of P and Si Concentrations Across the TMZ

### *3.1 Variations in P Concentration*

Across the TMZ from stations 38–25 along in May, salinity was from ~0 to 31.73, and concentrations of DIP, PIP, and TPP varied 0.48–1.2 μmol $L^{-1}$, 0.15–0.44, and 0.31–1.5 mg $g^{-1}$, respectively, in surface water, and 0.50–0.78 μmol $L^{-1}$, 0.13–0.35, and 0.24–0.81 mg $g^{-1}$ in bottom. The concentration variation was much smaller in bottom water than in surface water. From station 38 inside the river mouth to station 23 in the TMZ, the DIP concentrations did not decrease with salinity increasing, and the variation showed an opposite pattern to the variations of PIP and TPP concentrations (Fig. 3a and b).

On the east side of the TMZ, SPM contents decreased clearly. Small variation in PIP concentration and increasing in TPP concentration were probably related to the increasing in biological particulate organic P (Fig. 2e), which is consistent with the variation in BSi content (Fig. 3c and d). From the TMZ to the places offshore, the transparence increased from 0.5 to 2.8, which was beneficial to phytoplankton growth. In addition, it could be also proved with positive linear relationship between TPP concentrations and Chl-*a* contents ($r = 0.830$, $p = 0.00044$). The concentration of particulate P increased with SPM decrease in the estuary that was also noticed by Edmond et al. (1985).

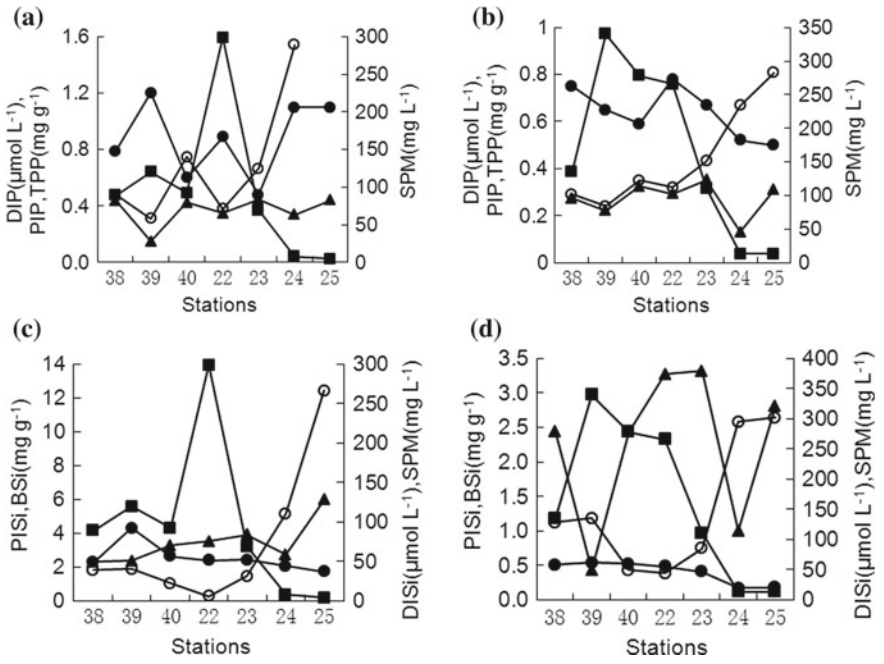


**Fig. 3** Variations in P, Si and SPM contents along the transect: **a** and **c**—surface; **b** and **d**—bottom; **a** and **b**: filled circle—DIP, filled triangle—PIP, open circle—TPP, filled square—SPM; **c** and **d**: filled circle—DISi, filled triangle—PISi, open circle—BSi, filled square—SPM

## *3.2 Variations in Si Concentration*

The variation along the transect in concentration of various forms of Si was much greater than that of P. The concentration ranges of DISi, PISi and BSi were 37.3–92.2 μmol $L^{-1}$, 2.3–6.0 and 0.28–12.4 mg $g^{-1}$ in surface layer; and 19.3–61.4 μmol $L^{-1}$, 0.43–3.3 and 0.39–2.6 mg $g^{-1}$, in bottom layer, respectively. The variation in DISi concentration was generally opposite to that of PISi in the TMZ (Fig. 3c and d). In the area east of the TMZ, with salinity increase, the influence of biological activity increased (Fig. 2e), shown by the variation in BSi content that increased rapidly and reached 12.4 and 2.6 mg $g^{-1}$ in surface and bottom waters, respectively, at station 25. The average concentrations of PISi were higher at $2.7 \pm 0.99$ mg $g^{-1}$ than that of BSi at $1.0 \pm 0.58$ mg $g^{-1}$ in the TMZ, whereas in the area east of the TMZ, the concentrations of the PISi were smaller at $3.0 \pm 2.0$ mg $g^{-1}$ than that of BSi at $6.1 \pm 3.4$ mg $g^{-1}$. Correlation analyses on data of the stations on the transect show clearly positive linear relationships among Chl-*a* content, sea water transparence ($r = 0.979$, $p = 0.00068$) and BSi concentration ($r = 0.827$, $p = 0.00048$).

# 4 Removals of P and Si in the TMZ and Its Adjacent Waters

## *4.1 Removal of P in the TMZ and Its Adjacent Waters*

### 4.1.1 Mixing Process of P in the Estuary

A negative linear relationship was found between DIP and salinity in all the investigation waters (Fig. 4a), showing that from river mouth to open sea, physical mixing controlled DIP concentration to a great extent. However, phytoplankton bloomed in May just before the sampling; it was likely that a part of DIP had been taken up by phytoplankton in upper layer sea water. The biological influence occurred mainly in the area of intermediate salinity north and east to the investigation area and far from the river mouth (Fig. 2e). At stations such as stations 9 and 11, Chl-*a* content in surface water reached as high as 11.659 and 19.515 $\mu g\ L^{-1}$. If we draw a theoretical dilution line in Fig. 4a connecting the average DIP concentrations in river water end-member and sea water end-member at average salinity above 33, one would find that above the data points fall below this line, suggesting that DIP was taken up by phytoplankton. In addition, DIP adsorbed by and released from suspended particles and resuspended sediment could also affect the distribution of DIP in the estuary. For example, at stations 28 and 29 in mild salinity region (Fig. 4a), their data points situate above the theoretical dilution line. Many field observations and laboratory experiments have demonstrated that aquatic particles can release DIP through a desorption reaction (Fox et al. 1986; Froelich 1988; Lebo 1991; Chambers et al. 1995; Suzumura et al. 2000, 2004). Since DIP concentration in the estuary was affected by several processes besides just the dilution mixing, correlation between DIP and salinity was low especially in the TMZ (Fig. 4a), where biological influence was low but suspended particles and sediment resuspension were high.

TP was more negatively correlated with salinity than DIP did (Fig. 4b), suggesting that the distribution of TP in estuarine waters depended mainly on physical mixing, as the distribution of TP (including all forms of P, organic and inorganic, and dissolved and particulate) is irrelevant to biological activity. However, TP distribution in the TMZ was largely affected by both sedimentation of suspended particulate P and constantly released P from resuspended sediment into sea water. If we draw a theoretical dilution line in Fig. 4b with above-mentioned method, it can be also found that some data points (stations 28, 29, etc.) of high concentrations of TP would be positioned above this line, which is consistent to the behaviours of DIP (Fig. 4a).

### 4.1.2 Buffering of Suspended Particulate

Many studies have demonstrated that the interaction of DIP with suspended particulate material was probably one of the most important mechanisms affecting geochemistry of P in estuarine area (Fox et al. 1986; Froelich 1988; Lebo 1991; Chambers

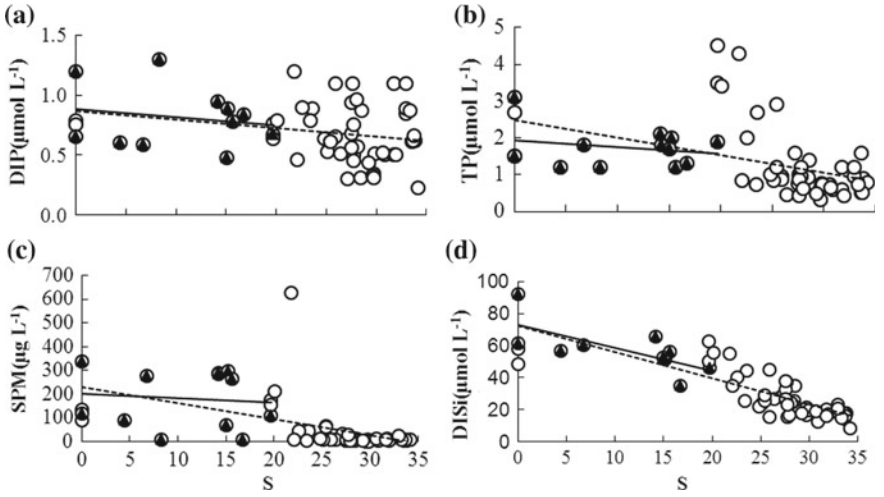


**Fig. 4** Relationships of DIP, TP, DISi and SPM to S, in the Changjiang estuary: open circle—all data; filled triangle—data for the turbidity maximum zone; dashed line—correlative curves for all data; solid line—correlative curves for the turbidity maximum zone; **a**: open circle—DIP = 0.871 − 0.007 S ($r = -0.278$, $p = 0.033$, $n = 59$), filled triangle—DIP = 0.885 − 0.007 S ($r = -0.181$, $p = 0.595$, $n = 11$); **b**: open circle—TP = 2.483 − 0.048 S ($r = -0.473$, $p = 0.00016$, $n = 59$), filled triangle—TP = 1.930 − 0.018 S ($r = -0.225$, $p = 0.483$, $n = 12$); **c**: open circle—SPM = 229.807 − 6.794 S ($r = -0.542$, $p = 7.59 \times 10^{-6}$, $n = 60$), filled triangle—SPM = 201.281 − 1.875 S ($r = -0.752$, $p = 0.102$, $n = 12$); **d**: open circle—DISi = 72.260 − 1.638 S ($r = -0.854$, $p = 1.58 \times 10^{-17}$, $n = 58$), filled triangle—DISi = 73.208 − 1.437 S ($r = -0.702$, $p = 0.024$, $n = 10$)

et al. 1995; Conley et al. 1995; Fang 2000). The variation in DIP concentration was obviously opposite to those of PIP and TPP (Fig. 3a and b), indicating that DIP was adsorbed by or released from SPM. By the buffering of SPM in the TMZ, DIP and PIP concentrations were retained 0.48–1.2 μmol $L^{-1}$ and 0.15–0.44 mg $g^{-1}$ in surface sea water, and 0.59–0.78 μmol $L^{-1}$ and 0.22–0.35 mg $g^{-1}$ in bottom, respectively, which is similar to the result of our previous study (Shen et al. 1992). The general mechanism of DIP buffering involves the equilibration of the dissolved P pool with a reservoir of particle-bound P (Froelich 1988). When the ambient DIP concentration differs from the equilibrium concentration, DIP is either released or adsorbed until equilibrium is achieved (Lebo 1991). Strong disturbance by wind, tide and current would increase the contact between DIP and suspended particulate material; therefore, the DIP equilibrium was closely related to the disturbance.

### 4.1.3 Resuspension of Sediment and the Behaviour of P

In the TMZ, the SPM came mainly from the Changjiang River run-off and resuspension of sediment. The SPM content in the estuarine waters was higher than that of the river run-off, meaning additional SPM from downstream to outside the river

mouth resulted from sediment resuspension (Fig. 4c). The ranges of variation for DIP, PIP and TPP concentrations were notably less in bottom water than those in surface (Fig. 3a and b), reflecting greater buffering effect of suspended matter and sediment to P in bottom water because of near-bottom resuspension.

### 4.1.4 Compositions of P

The average ratio of PIP to TPP was $0.81 \pm 0.17$ inside the river mouth and the TMZ, and $0.22 \pm 0.12$ in the area east of the TMZ, suggesting that inorganic P adsorbed by suspended matter and sediment was a major form of particulate P in the TMZ, while that of DIP/TP was $0.44 \pm 0.19$ within the TMZ and $0.85 \pm 0.18$ beyond the TMZ to the east, indicating that DIP was a main form of P in open sea. The phenomenon that distribution of TPP decreased with change in the seaward direction was also found in other estuaries (Fang 2000).

### 4.1.5 Compared with Other Rivers

TPP concentrations within the salinity gradient ($S < 32$) of the Changjiang estuary ranged from 0.24 to 2.0 mg $g^{-1}$ in average of $0.90 \pm 0.78$ mg $g^{-1}$, which were higher than the P content (0.44–1.10 mg $g^{-1}$) in surface sediment of the TMZ and its upstream (Hou et al. 2006). The average TPP we measured in the Changjiang estuary is close to the total P contents in the fluvial suspended particulate of 0.81 (Sholkovitz et al. 1978) and 1.05 mg $g^{-1}$ (Edmond et al. 1981) in the Amazon River, and 0.99 mg $g^{-1}$ in the Zaire River (van Bennekom et al. 1978), but below the global average of 1.15–1.20 mg $g^{-1}$ (Martin and Meybeck 1979; Froelich 1988). When compared with other urbanized estuaries, the TPP concentration within the salinity gradient of the Changjiang estuary is much lower. For example, the TPP contents in the American Delaware estuary were 1.24–1.55 (Lebo 1991), and 2.70–8.99 mg $g^{-1}$ in northern European Ems and Weser estuaries (Calmano 1981; Salomons and Gerritse 1981; Rehm 1985), possibly indicating locally high content of DIP. The TPP content within the salinity gradient is generally smaller than the total P content in fluvial suspended particulate, possibly due to the part of adsorbed P released from fluvial suspended particulate within the salinity gradient. Lebo (1991) found that the P content of particles within the salinity gradient of the Delaware estuary was much lower than particles from the river.

The PIP concentrations within the salinity gradient in the Changjiang estuary ranged 0.13–0.69 mg $g^{-1}$ in average of $0.33 \pm 0.15$ mg $g^{-1}$, close to the particulate active P concentration of 0.34 mg $g^{-1}$ in the Amazon estuary (Edmond et al. 1981) and 0.31 mg $g^{-1}$ in the Ochlockonee estuary (Kaul and Froelich 1984). All these values are above the global average of 0.16 mg $g^{-1}$ (Froelich 1988). The fraction of PIP in TPP concentration was 36.7% within the salinity gradient in the Changjiang estuary, well above the global one at 15% (Froelich 1988).

### 4.2 Removal of Si in the TMZ and Its Adjacent Waters

DISi concentration was mainly controlled by sea water dilution as shown by the negative linear relationship to the salinity (Fig. 4d). Compared with DIP (Fig. 4a), the relationship between DISi concentration and salinity is stronger (Fig. 4d). However, many data points of DISi distribute in two sides of the theoretical dilution line, suggesting that the behaviour of DISi in the estuary was somewhat non-conservative as it was affected by biological activity and suspended sediments. Biological activity influence on DISi occurred mainly in middle- and high-salinity zones.

Variation in DISi concentration is also opposite to that of PISi in the TMZ (Fig. 3c and d). From stations 39–23, with salinity increase from ~0 to 15.01 in surface layer and from ~0 to 19.71 in bottom layer, the DISi concentration decreased from 92.2 to 52.5 μmol $L^{-1}$ and from 61.4 to 46.4 μmol $L^{-1}$, respectively, and PISi concentration increased from 2.4 to 3.9 mg $g^{-1}$ and from 2.2 to 3.5 mg $g^{-1}$, respectively. Different from DIP, DISi concentrations continuously decreased with salinity due to dilution and adsorption onto particles. Studies have also shown that DISi could be adsorbed by suspended matter and desorbed from suspended matter during estuarine mixing (Mackenzie and Garrels 1965; Fanning and Pilson 1973; Mayer and Gloss 1980; Bowes and House 2001).

Removal of Si and that of P in the studied estuary was similar as both were controlled by sea water dilution. A positive linear relationship between DISi and DIP concentrations existed (through stations 38–25: $r = 0.492$, $p = 0.045$). Additionally, they were affected by the buffering of suspended matter and sediment; therefore, a positive linear relationship between PISi and PIP concentrations was also found (stations 38–25: $r = 0.750$, $p = 0.00052$). During the removal, however, impacting factors of physics, chemistry and biology were considerably different. Compared to the DIP, the DISi concentration was more sea water dilution affected, while DIP concentration was influenced more by suspended matter and sediment.

## 5 Sedimentation Fluxes of P and Si in the TMZ of the Changjiang Estuary

### 5.1 Sedimentation Fluxes of P and Si in the TMZ

Sedimentation flux of particulate matter was calculated by the following equations (Zhang et al. 2004):

$$F = M/HV \tag{1}$$

$$F_c = Mf_c/HV \tag{2}$$

where $F$ is sedimentation flux of particulate matter (g $m^{-2}$ $d^{-1}$); $M$ is the dry weight (g) of particulate matter in the sampling tubes; $V$, the section area ($m^2$) of sampling

tubes; $H$, the sampling time (d); $F_c$ is sedimentation flux of some components in particulate matter (mg $m^{-2}$ $d^{-1}$); and $f_c$ is the content of certain component of particulate matter (mg $g^{-1}$).

The dry weight of particulate matter and its components P and Si contents at stations 22 and 23 are summarized in Table 1. The P and Si contents in sinking particles were less than those in the SPM, indicating probably that part of the P and Si was released from sinking particles during sedimentation. With Formulas (1) and (2), the sedimentation fluxes were calculated and listed in Table 2. Taking the calculated results of stations 22 and 23 as the average value for the TMZ, we obtained the sedimentation fluxes of particulate matter and its components PIP, TPP, PISi, BSi and TPSi in the TMZ (Table 2). PIP flux accounted for 65.7% of TPP, showing that inorganic P dominated the sinking particle P and was consistent with that of SPM. The sedimentation flux of PISi was higher than that of BSi, suggesting that it was related to low phytoplankton quantity. The area of the TMZ was estimated about $20.579 \times 10^8$ $m^2$. Thus, the annual sedimentation fluxes of particulate matter, P and Si were calculated and listed in Table 2.

**Table 1** Dry weight (g) of particulate matter and its components of P and Si contents (mg $g^{-1}$)

| Stations | SPM | PIP | TPP | PISi | BSi | TPSi |
|---|---|---|---|---|---|---|
| 22 | 4.471 | 0.11 | 0.16 | 0.30 | 0.26 | 0.56 |
| 23 | 3.438 | 0.12 | | 0.38 | 0.27 | 0.65 |

**Table 2** Sedimentation fluxes of particulate matter (g $m^{-2}$ $d^{-1}$) and its components of P and Si (mg $m^{-2}$ $d^{-1}$)

| Stations | SPM | PIP | TPP | PISi | BSi | TPSi |
|---|---|---|---|---|---|---|
| 22 | 269.5 | 30.9 | 43.1 | 79.6 | 70.1 | 149.7 |
| 23 | 207.2 | 25.7 | | 78.4 | 55.9 | 134.3 |
| Averages | 238.4 | 28.3 | 43.1 | 79.0 | 63.0 | 142.0 |
| Annual sedimentation (t) | $1.79 \times 10^8$ | $2.126 \times 10^4$ | $3.237 \times 10^4$ | $5.934 \times 10^4$ | $4.732 \times 10^4$ | $10.666 \times 10^4$ |
| Annual resuspension (t) | $1.18 \times 10^8$ | $1.405 \times 10^4$ | $2.140 \times 10^4$ | $3.922 \times 10^4$ | $3.128 \times 10^4$ | $7.050 \times 10^4$ |
| Annual net sedimentation (t) | $0.61 \times 10^8$ | $0.721 \times 10^4$ | $1.097 \times 10^4$ | $2.012 \times 10^4$ | $1.604 \times 10^4$ | $3.616 \times 10^4$ |

## 5.2 Net Sedimentation Fluxes of P and Si in the TMZ

Most suspended particulate matter carried out by Changjiang River deposited in the TMZ. Flocculation and sedimentation are two important mechanisms (Krone 1962; Jiufa and Chen 1998; Shen and Pan 2001) affecting the character and fate of the dissolved and particulate matters from a river (Herman and Heip 1999; Gebhardt et al. 2005).

### 5.2.1 Estimation of the Resuspension Ratio

Due to the influences of wind, tide and current, suspended particulate matter in the TMZ of the Changjiang estuary was frequently disturbed in cycles of suspension, sedimentation, resuspension and resedimentation (Shen and Pan 2001). The suspended particulate matters in the TMZ include those from the river run-off and resuspended sediment. A simple method was used to estimate the ratio of resuspension from sediment of the TMZ. Taking SPM content at station 35 in downstream Xuliujing area as the background value of the river run-off (Chen et al. 2004), a theoretical dilution line between freshwater end-member and sea water end-member would be drawn (Fig. 4c). In case of resuspension or resedimentation, the SPM content would fall over or below this line. As the fraction of combustible particulate matter is very small, only 7.1% in May for our samples, among the total suspended particles in the Changjiang River, biological influence could be ignored. Therefore, the difference in SPM content between the measured in a station and its corresponding point at the salinity line would be equal to the resuspension amount. The resuspension ratio $R$ can be calculated by:

$$R = ((\mathrm{SPM}_1 - \mathrm{SPM}_2)/\mathrm{SPM}_1) \times 100\% \quad (3)$$

where $\mathrm{SPM}_1$ is the SPM content determined at a station in the TMZ, $\mathrm{SPM}_2$ is the corresponding SPM value at the salinity line of theoretical dilution.

### 5.2.2 Net Sedimentation Fluxes of P and Si

Based on the investigation data of suspended matter in February, May, August and November 2004, using Formula (3), the resuspension ratio of particulate matter in bottom water of the TMZ was between 55.7 ± 28.7 and 80.5 ± 10.5% in annual average of 66.1%. Therefore, the annual resuspension amount and net sedimentation of particulate matter and its components of P and Si were calculated (Table 2). It shows that over half of the annual net sedimentation of particulate matter and its components of P and Si came from resuspension of sediment. With the same method, we calculated the resuspension ratio in surface water in the TMZ, which varied between 25.0 ± 40.6 and 73.0 ± 22.2% in annual average of 55.7%, and

smaller than that in bottom layer. Thus, annual resuspension amount of particulate matter and its P and Si components in surface water of the TMZ were calculated to be $1.00 \times 10^8$ t for SPM, $1.186 \times 10^4$, $1.806 \times 10^4$, $3.311 \times 10^4$, $2.641 \times 10^4$ and $5.952 \times 10^4$ t for PIP, TPP, PISI, BSi and TPSi, respectively. It also shows that the resuspension of sediment is one of the main mechanisms forming the TMZ (Jiufa and Chen 1998; Pan et al. 1999). Sedimentation and resuspension of huge amounts of P and Si are important to keep their concentrations at certain ecological levels in the estuarine water.

## 6 The Mass Balances of P and Si in the TMZ

The mass balances of P and Si in the TMZ of the Changjiang estuary can be calculated by:

$$F_{ai} + F_{ri} + F_{si} = F_d + F_{so} + F_{bo} + F_{eo} \tag{4}$$

where $F_{ai}$ is the input flux (kg $s^{-1}$) of atmosphere sedimentation, $F_{ri}$ is the input flux of river (kg $s^{-1}$), $F_{si}$ is the input flux of sediment diffusion to sea water, $F_d$ is the dilution and diffusion flux (kg $s^{-1}$) intercepted in the TMZ, $F_{so}$ is sediment output flux (kg $s^{-1}$), $F_{bo}$ is biological output flux and $F_{eo}$ is the output flux (kg $s^{-1}$) to the East China Sea. The item of $F_{si}$ is not considered due to relatively insignificant and data absence. The Chl-*a* contents in the TMZ were 0.287 mg $L^{-1}$ in May and 0.776 mg $L^{-1}$ in average of February, May, August and November, showing relatively less biological output, and is not considered here. Thus, the above formula becomes:

$$F_{ai} + F_{ri} = F_d + F_{so} + F_{eo} \tag{5}$$

### *6.1 Atmosphere Inputs of P and Si in the TMZ*

The atmospheric sedimentation is considered only in the case of wet sedimentation. Based on observations of 32 times near Baozhen Town (121° 36.5′ E, 31° 30.3′ N) east part of Chongming Island from May 2004 to May 2005, the average contents of DIP and DISi in precipitation were 0.51 and 0.23 μmol $L^{-1}$. For TP content, we used the value of 0.66 μmol $L^{-1}$ observed in the Changjiang River catchment in our previous study. Thus, the precipitation fluxes of P and Si can be computed as follows:

$$F_{ai} = C_a \cdot Q_a \cdot A \cdot f_a \tag{6}$$

where $F_{ai}$ is the average annual precipitation input fluxes (kg $s^{-1}$) of P and Si, $C_a$ is average contents (μmol $L^{-1}$) of P and Si in the precipitation, $Q_a$ is the rainfall (mm) in Shanghai City in 2005, A is area ($m^2$) of the TMZ and $f_a$ is the conversion coefficient

of units (from molar concentration to quality concentration). With Formula (6), the deposition fluxes of DIP, DISi and TP in the TMZ by precipitation were calculated (Table 3), which shows that DIP flux accounted for 77.3% of TP and was the main form of P.

## 6.2 River Inputs of P and Si in the TMZ

The input fluxes of P and Si from the Changjiang River can be calculated with:

$$F_{\mathrm{ri}} = C_{\mathrm{r}} Q_{\mathrm{r}} f_{\mathrm{r}} \tag{7}$$

where $F_{\mathrm{ri}}$ is the average annual input fluxes of P and Si (kg $s^{-1}$) from the river; $C_{\mathrm{r}}$ is the average concentrations of DIP, TP and DISi in five stations within the river mouth (stations 35–39, salinity ~ 0) in February, May, August and November 2004. The values were 0.86, 2.3 and 120.6 μmol $L^{-1}$, respectively. $Q_{\mathrm{r}}$ is the average annual run-off ($m^3$ $s^{-1}$) of the river mouth, and $f_{\mathrm{r}}$ is the conversion coefficient of units. With Formula (7), the fluxes of DIP, DISi and TP to the TMZ from the river were calculated (Table 3). The input fluxes of DIP and DISi in this study are higher than those (0.43 and 63.3 kg $s^{-1}$) annual investigation records during 1985–1986 (Shen 1993b). The input flux of DISi calculated in this study was below 106.5 kg $s^{-1}$ than that reported by Edmond et al. (1985). The input flux of DIP of this study was well below that of the Amazon River in December 1982 (2.35 kg $s^{-1}$) and May 1983 (4.62 kg $s^{-1}$) with corresponding average annual run-offs of the river at 123,668 and 171,232 $m^3$ $s^{-1}$, respectively (Edmond et al. 1981, 1985; Fox et al. 1986; Froelich 1988), which is 4.9 and 6.8 times higher than that of the Changjiang River in this study.

The input fluxes of particulate P and Si could be calculated using the same method. $C_{\mathrm{r}}$ is the concentrations of particulate P and Si in the river mouth water (mg $L^{-1}$). It is the product between the average concentrations of PIP, TPP, PISi and BSi (mg $g^{-1}$) at stations 38 and 39 (salinity ~ 0) in May, August and November 2004 and the average SPM content (mg $L^{-1}$) at station 35 (the background value of SPM from the Changjiang run-off) in February, May, August and November 2004. Afterwards, the input fluxes and annual inputs of PIP, TPP, PISi and BSi were calculated (Table 3). The annual input of PIP from the Changjiang River was less than that of DIP, the former was 55.9% of the latter and cannot be ignored as biologically available P. The annual input of PIP of the Changjiang River was much less than that of the Amazon in 1982 and 1983 ($17.050 \times 10^4$ and $30.690 \times 10^4$ t $a^{-1}$, respectively). The annual input of PIP was 2.3–2.1 times of that of DIP in the Amazon (Edmond et al. 1981, 1985; Fox et al. 1986; Froelich 1988). The ratio of annual input of PIP to DIP in the Ochlockonee River was 0.6 (Froelich 1988) similar to that in the Changjiang River. The difference between TP and TPP + DIP is dissolved organic P (DOP), and the difference between TP and TPP is total dissolved P (TDP). The input fluxes and annual inputs of DOP and TDP were 0.13, 0.81 kg $s^{-1}$ and $0.396 \times 10^4$, $2.537 \times 10^4$ t, respectively. These results show that the input flux of DIP accounted for 81.9% of

**Table 3** Mass balances of P and Si in the TMZ

| | River input | | Precipitation input | | Dilution and diffusion intercepted | | Sediment output | | Output to East China Sea | |
|---|---|---|---|---|---|---|---|---|---|---|
| | kg $s^{-1}$ | $10^4$ t $a^{-1}$ | kg $s^{-1}$ | t $a^{-1}$ | kg $s^{-1}$ | $10^4$ t $a^{-1}$ | kg $s^{-1}$ | $10^4$ t $a^{-1}$ | kg $s^{-1}$ | $10^4$ t $a^{-1}$ |
| TP | 1.79 | 5.631 | 0.0017 | 52.8 | 0.57 | 1.802 | | | 1.22 | 3.834 |
| TPP | 0.98 | 3.094 | | | | | 0.35 | 1.097 | 0.63 | 1.996 |
| TIP | 1.05 | 3.326 | 0.0013 | 40.8 | 0.14 | 0.450 | 0.23 | 0.721 | 0.68 | 2.160 |
| DIP | 0.68 | 2.141 | 0.0013 | 40.8 | 0.14 | 0.450 | | | 0.54 | 1.696 |
| PIP | 0.38 | 1.185 | | | | | 0.23 | 0.721 | 0.14 | 0.464 |
| TISi | 88.89 | 280.326 | 0.00053 | 16.6 | 49.26 | 155.335 | 0.64 | 2.012 | 39.00 | 122.979 |
| DISi | 84.92 | 267.819 | 0.00053 | 16.6 | 49.26 | 155.335 | | | 35.67 | 112.486 |
| PISi | 3.97 | 12.507 | | | | | 0.64 | 2.012 | 3.33 | 10.495 |
| BSi | 3.13 | 9.874 | | | | | 0.51 | 1.604 | 2.62 | 8.269 |
| TPSi | 7.10 | 22.380 | | | | | 1.15 | 3.616 | 5.95 | 18.765 |

TDP, being the main form of dissolved P, and was 64.4% of TIP or 38.0% of TP. The input flux of PIP accounted for 35.6% of TIP or 21.2% of TP. The sum of DIP and PIP is the total active inorganic P (TIP) accounting for 59.2% of TP. The input flux of TPP accounted for 54.9% of TP, and particulate active inorganic P (PIP) was 38.3% of TPP. The input flux of DISi accounted for 95.5% of TISi and PISi was only 4.5%. Among the TPSi flux, PISi and BSi accounted for 55.9 and 44.1%, respectively, and PISi flux was greater than BSi. The annual input of PISi from the Changjiang River mouth was much smaller than DISi, the former was only 3.5% of the latter, which is largely different from the case of P, showing that particulate-adsorbed Si was much less important than particulate-adsorbed P.

## 6.3 Dilution and Diffusion Fluxes of P and Si Intercepted in the TMZ

The statistics for all data of DIP, TP, DISi and salinity in the study area in February, May, August and November 2004 show strong negative linear relationships:

$$\text{DIP}\ (\mu\text{mol L}^{-1}) = 0.942 - 0.009\ \text{S}\ (r = 0.315, n = 251, p = 3.35 \times 10^{-7})$$
$$\text{TP}\ (\mu\text{mol L}^{-1}) = 2.463 - 0.034\ \text{S}\ (r = 0.323, n = 242, p = 2.69 \times 10^{-7})$$
$$\text{DISi}\ (\mu\text{mol L}^{-1}) = 126.7 - 3.226\ \text{S}\ (r = 0.876, n = 252, p = 2.89 \times 10^{-81}).$$

It shows that their behaviours depend mainly on dilution and diffusion of sea water in the estuary, and parts of the P and Si were intercepted in the TMZ during diffusion process. According to the average salinity of 22.82 at the sea water end-member in the TMZ, the average concentrations for DIP, TP and DISi were calculated to be 0.74, 1.68 and 53.1 $\mu$mol $L^{-1}$, respectively, using above equations. Thus, the ratios of the concentrations of DIP, TP and DISi in the freshwater end-member could be thought to be the input flux ratios of them at the river mouth. In other words, 21% of DIP, 32% of TP and 58% of DISi were intercepted in the TMZ. The intercepted and annual intercepted fluxes of DIP, TP and DISi through dilution and diffusion in the TMZ were calculated and listed in Table 3. The maximum interception flux was in DISi. The dilution and diffusion of TP included two parts. Part 1 was dissolved P which was intercepted (0.14 kg $s^{-1}$) mainly through dilution and diffusion, and the concentration decreased with the increase in salinity; Part 2 was particulate P that removed mainly through deposition during dilution and diffusion (Table 3).

## 6.4 Output of P and Si from the TMZ to the East China Sea

According to above calculation results, P and Si amounts in various forms transported from the TMZ to the East China Sea were listed in Table 3. The output flux and annual output of DOP were 0.05 kg $s^{-1}$ and $0.142 \times 10^4$ t and those of TDP were 0.58 kg $s^{-1}$,

$1.838 \times 10^4$ t, respectively. It shows that the output flux of DIP accounted for 92.3% of TDP, dominating dissolved P, and was 78.5% of TIP or 44.3% of TP. The output flux of PIP accounted for 21.5% of TIP or 12.1% of TP, and TIP was 56.4% of TP. The output flux of TPP accounted for 52.6% of TP, and particulate inorganic active phosphorus (PIP) accounted for 23.3% of TPP. Among various forms of Si transported from the TMZ to the East China Sea, the output flux of DISi accounted for 91.5% of TISi, and PISi accounted for 8.5%. For TPSi, the PISi and BSi accounted for 55.9 and 44.1%, respectively, and PISi was more than BSi.

Table 3 shows that during the transportation of P and Si to the East China Sea, the fluxes of PIP, DIP, TIP, DOP, TDP, TPP and TP intercepted in the TMZ accounted for 60.8, 21.0, 35.2, 64.4, 27.8, 35.5 and 32.0% of those from the river mouth, respectively. It shows also that among intercepted P, the ratio of particulate P was greater than that of dissolved P; and the most of particulate-adsorbed inorganic P and over one-third of total particulate P were intercepted in the TMZ via deposition, well reflecting the role of filtration that the TMZ played, which could be important in preventing eutrophication in estuarine waters. In terms of Si, the fluxes of TISi, DISi, PISi, BSi and TPSi intercepted in the TMZ accounted for 56.1, 58.0, 16.1, 16.2 and 16.2% of those from the river mouth, respectively. Different from P, among intercepted Si, the ratio of the dissolved Si was clearly higher than that of the particulate Si. Most of particulate Si was transported to the East China Sea, and among particulate Si, the proportions of PISi and BSi were similar. The difference in existence form between Si and P was probably related to the original ratios at the river mouth. Among the input fluxes of P and Si at the river mouth, the ratio of TDP/TPP was 0.82, and the amount of particulate P was higher than that of dissolved P; the ratio of DISi/TPSi was 11.96, indicating dissolved Si was much higher than that of particulate Si.

Moreover, the ratio of particulate P flux deposited in the TMZ to that of discharged from the river mouth was much higher than that of particulate Si case. Therefore, P can be adsorbed more easily by suspended matter than Si. In the TMZ, adsorbed and deposited P and Si would be released into sea water through sediment resuspension and take part in a new material cycle. Among the particulate P transported from the river mouth into the East China Sea, 39.2% were potential biologically available P. They are important for phytoplankton growth and were also a potential reason of eutrophication in coastal environments.

To sum up, the fluvial suspended particulate matter of the Changjiang River and the resuspension of estuarine sediment play major roles in removing and transporting P and Si. In addition to dilution and diffusion, the removal of DIP and DISi in the estuary includes biological and non-biological aspects. The biological removal of DIP and DISi took place mainly in high-salinity area of the open sea, and the non-biological removal acts in adsorption and release of DIP and DISi by suspended matter mainly in the TMZ (low salinity area). Compared to DIP, DISi was controlled mainly by sea water dilution. Influenced by buffering of suspended matter and sediment resuspension in the TMZ, the DIP concentrations were kept in the levels of 0.48–1.2 $\mu$mol $L^{-1}$.

The fluxes of DIP, TP and DISi from the Changjiang River to the TMZ were estimated at 0.68, 1.79 and 84.92 kg $s^{-1}$, and those of PIP, TPP, PISi and BSi were 0.38, 0.98, 3.97 and 3.13 kg $s^{-1}$, respectively. The input flux of PIP was 55.9% of DIP that is important as biologically available P. Total active inorganic P (TIP) accounted for 59.2% of TP, being a main component of P, while the input flux of PISi was only 3.5% of DISi, showing that particulate-adsorbed Si was much less important than particulate-adsorbed P. Among P and Si transported from the Changjiang mouth to the TMZ, the dissolved component was intercepted mainly during dilution and diffusion, and the particulate component was removed mainly by deposition during dilution and diffusion. In the TMZ, over half of the sedimentation fluxes of particulate matter and bearing P and Si came from sediment resuspension. Most particulate-adsorbed inorganic P and over one-third of the total particulate P from the Changjiang River were intercepted in the TMZ through deposition. The particulate-adsorbed inorganic P accounted for 21.7% of total active inorganic P, which well indicates the role as a filter that the TMZ plays for potential prevention from eutrophication in estuarine waters.

Computation in mass balances was conducted for estimating the output fluxes and annual outputs of various forms of P and Si from the TMZ to the East China Sea, in which total active inorganic P accounted for 56.4% of TP, and PIP was 21.5% of TIP or 12.1% of TP, while PISi was only 8.5% of the TISi. The active inorganic P transported to the East China Sea, which could be a possible reason for eutrophication in coastal environments.

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