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Levels and distributions of Dechlorane Plus in coastal sediment of North China

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ABSTRACT

Dechlorane Plus (DP) has been determined in surface sediments from three Chinese coastal bays, e.g. Jiaozhou, Sishili and Taozi Bay in North China. DP concentrations ranged from <1.2 to 187pg/g dry weight (dw) (mean: 24.7 pg/g dw) in Jiaozhou Bay, <1.2 to 135 pg/g dw (mean 69.9 pg/g dw) in Sishili Bay and <1.2 to 66.7 pg/g dw (mean: 40.4 pg/g dw) in Taozi Bay, respectively. Additionally, two dechlorinated species were quantified, which accounted for 0.6% to 5.1% in the ∑DP concentration.

The f_{syn} values (*syn*-isomer/(*syn*- + *anti*- isomer)) in sediments from Jiaozhou Bay (mean 0.29) were close to technical DP mixture (0.2 to 0.4), probably indicating a local usage of DP. In contrast, sediments in Sishili and Taozi Bay showed much lower f_{syn} values (mean

0.16).During transportation the DP isomers underwent stereo selective degradation which partly resulted in the enrichment of *anti*-DP in coastal sediment.

Keywords: **Dechlorane Plus, coastal sediment, North China**

Capsule: study for occurrence of Dechlorane Plus in sediment of coastal environment of North China

1. Introduction

Dechlorane Plus (DP, $C_{18}H_{12}Cl_{12}$), the common name of bis(hexachlorocyclopentadieno) cyclooctane, which is used as a substitute for the toxic Declorane (Mirex) has been produced for about four decades (Gauthier et al., 2007; Qiu et al., 2007). As a highly chlorinated flame retardant, DP has been used in plastic roofing material, hardware connectors for computers and for coating electrical wires and cables (Tomy et al., 2008). The annual production of DP was reported as 5000 tons, and it is sold in North American, Europe and Asia (Kang et al., 2010; Wang et al., 2010a). As a result of the wide application, DP has been detected in various environmental compartments, such as air, water, sediment, fish and serum of human being (Hoh et al., 2006; Tomy et al., 2007; Ren et al., 2009; Qi et al., 2010; Wang et al., 2010b). Bioaccumulation and biomagnification were reported for organisms of high tropic levels, which suggest a potential threat for humans (Tomy et al., 2008; Ren et al., 2009).

In China DP has been produced by Anpon Corporation for seven years, with a total volume of 2100 to 7000 tons. This manufacturing plant has been recognized as the main DP source for the surrounding area (Wang et al., 2010b). The DP concentration in soil near the plant was 1490 ± 3580 ng/g dry weight (dw), which is one magnitude higher than the maximum concentration in Lake Ontario (586 ng/g dw) (Sverko et al., 2008; Wang et al., 2010b). The usage and unintended disposal spread DP in non-producing areas. Ren et al. (2008) collected

air samples from 97 Chinese urban and rural sites, and stated that the highest concentration was found in Kunming, a famous tourist city far away from the DP manufactory. In Harbin, an industrial city of Northeast China without DP manufacture, the mean concentration of DP was 0.11 ± 0.05 ng/g dw in urban sediments (Qi et al., 2010). Besides domestic usage, the import of DP or DP containing products (including e-waste) from overseas could also elevate the levels in the Chinese environment. Yu et al. (2010) found that in an e-waste recycling site in Qingyuan, the concentration of DP reached up to 3327 ng/g dw in surface soil.

Sediments have been regarded as one of the major sinks for persistent organic pollutants (POPs), e.g., polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) and brominated flame retardants (BFRs) (Hung et al., 2006; Minh et al., 2007; Guzzella et al., 2008). These pollutants enter the marine sediments through atmospheric deposition, riverine runoff and direct release from human activities on the ocean (Moon et al., 2007b). Both, the *syn*- and *anti*- isomers have a high octanol-water partition coefficient (logK_{ow}) value of 9.3, which is similar to some polybrominated diphenyl ethers (PBDEs) that strongly adsorbed to sediments (Palm et al., 2002; Moon et al., 2007a; Sverko et al., 2008). The OxyChem report employing EPIWIN version 3.12 presented that 66.6% of the DP was distributed in sediments (US, 2008). High concentrations of DP have been detected in sediments from Lake Ontario with an inventory of DP calculated to about 20 tons (Qiu et al., 2007; Sverko et al., 2008). Presently, only few data are available on the distribution of DP in coastal sediments. Our present study focuses on the concentrations, distributions and possible sources of DP in surface sediments of three Chinese bays.

2. Materials and Methods

2.1 Sample Collection

Surface sediments (0-5 cm) were collected in coastal zones of Yellow Sea in April, 2010. The locations of all the 48 samples are shown in Fig. 1. Twenty-five samples were collected

in the Jiaozhou Bay which is surrounded by Qingdao City (35°35'-37°09'N, 119°30'-121°00'E), and four river sediment samples were collected in Dagu River and Qingdao River, discharging into the Jiaozhou Bay. Six samples were collected in Taozi Bay and thirteen in Sishili Bay near Yantai City (36°16′-38°23′N, 119°34′-121°57′E). The sediments were collected by stainless spades and then sealed in polyethylene (PE) bags. They were frozen immediately after sampling at -20 °C until extraction. Prior to extraction, sediments were freeze-dried for three days at -55°C and then ground, homogenized by agate mortar and pestle and wrapped up in pre-cleaned filter papers.

2.2 Extraction and cleanup

All solvents were analytical grade and were re-distilled before using. Filter papers were Soxhlet extracted for 72 h using dichloromethane (DCM). Samples were spiked with 20 ng of PCB 209 as surrogate standard prior to extraction. 20 g of sediments were extracted in a Soxhlet apparatus for 72 h using DCM. Activated copper slices were added to the collection flask to remove elemental sulfur. Extracts were evaporated to 10 mL and the solvent was changed to hexane before further evaporation to <2 mL. A 8 mm i.d. modified column packed with 1 cm anhydrous sodium sulfate, 3 cm 50% sulfuric acid silica, 3 cm neutral silica gel and 3 cm neutral alumina from the top to the bottom was employed for clean-up. The silica was precleaned with acetone and DCM, activated at 180 °C for 12 h and further 3 % deactivated using Millipore water. The neutral alumina was treated in a similar way, activated at 250 °C. Sulfuric acid silica was prepared by adding 50 % (w/w) HPLC grade sulfuric acid (98 %) to the deactivated silica. The anhydrous sodium sulfate was baked at 450 °C for 4h. The fraction was concentrated to 50 µL under a gentle high-purity nitrogen (>99.99 %) stream. 20 ng of BDE-77 was spiked as an internal standards before injection.

2.3 Instrument Analysis

The standards of *syn*-DP, *anti*-DP, and two dechlorinated species, $aCl_{11}DP$ and $aCl_{10}DP$, were obtained from Wellington Laboratories. An Agilent 7890A GC equipped with a 30 m ×

0.25 mm i.d. (0.25 μm film thickness, J&W Scientific) DB-5 fused silica capillary column was connected to a 5975C MSD under a negative chemical ionization (NCI) mode using methane as the reagent gas. The injector temperature was 280 °C employed in splitless mode with 1 μL injection volume. The oven program was as follows: initial 60 °C for 2 min, 30 °C min^{-1} to 180 °C, 2 °C min^{-1} to 280 °C, 30 °C min^{-1} to 300 °C and held for 6 min, and then ramped at 30 °C min^{-1} to 310 °C and held for a final 7 min. MS was operated in single ion monitoring (SIM) mode with ion source, quadrupole and transfer line temperatures held at 150, 150 and 280 °C, respectively. The following ions were monitored: *m/z* 653.8, 617.9 and 583.9 for *syn*– and *anti*-DP; *m/z* 617.8, 583.7 and 547.8 for $aCl_{11}DP$; *m/z* 583.8, 549.9 and 513.8 for $aCl_{10}DP$; *m/z* 79.0 and 81.0 for BDE77; and *m/z* 497.7 and 499.7 for PCB209.

2.4 QA/QC

The criteria for the identification and quantification of target compounds are given as follow: (1) The retention times matched those of the standard compounds within ± 0.05 min. (2) The signal–to–noise (S/N) ratio of all peaks was greater than 5:1. (3) The theoretical isotopic ratios of the qualifier ions were within ± 15% of the standard values. The linear dynamic range of the instrument was between 2 to 25 pg on the column (R^2>0.996) for DP isomers and their dechlorinated species. PCB 209 was spiked in every sample as DP recovery indicator. The mean recovery rate was 96±12%. Concentrations of sediments were not recovery corrected. In three procedure blanks, both DP isomers and their dechlorinated moieties were not detected. Method detection limits (MDLs) were calculated based on the instrumental S/N ratios of 10. They were 0.4 pg/g for *syn*-DP, 0.8 pg/g for *anti*-DP, 0.5 pg/g for $aCl_{10}DP$ and 0.9 pg/g for $aCl_{11}DP$, respectively.

3. Results and Discussion

3.1 Concentrations and spatial distribution of Dechlorane Plus

127 The concentrations of total DP (*syn*- +*anti*-DP) in river and marine sediments are shown in
128 Table 1. DP was detected in all surface sediment samples from Jiaozhou, Taozi and Sishili
129 Bay. Large spatial variations of total DP were observed, with <1.2 to 187 pg/g in Jiaozhou
130 Bay, <1.2 to 66.7 pg/g in Taozi Bay and <1.2 to 135 pg/g in Sishili Bay, respectively. Qi et al.
131 (2010) reported DP in sediments from Songhua River, Northeast China. The concentrations (>
132 4.5 to 160 pg/g dw) were similar to those found in this study. Comparing with data from the
133 sediments near manufactory locations, the concentrations in the three bays were one to three
134 orders of magnitude lower. Wang et al. (2010) reported concentrations ranging from 1.86 to
135 8.0 ng/g dw in the canal sediments near Anpon Corporation in Huai'an, China, and Sverko et
136 al. (2008) reported high concentrations in sediment of Great Lakes ranging from 0.061 to 586
137 ng/g near the OxyChem company. According to Wang et al. (2010), DP was only
138 manufactured by Anpon Corporation in Huai'an, China, which is 300 to 500 km far away
139 from target bays to the south, and there has been no manufactory reported near Jiaozhou,
140 Taozi and Sishili Bay.
141 Significant differences of the DP distributions have been found between Jiaozhou Bay
142 and Taozi and Sishili Bay (student *t*-test, $p<0.05$). In Jiaozhou Bay, the highest concentration
143 (187 pg/g dw) was found at the estuary of Haibo River, where one of Qingdao waste water
144 treatment plants (WWTP) is located nearby. The concentration at this site was one to three
145 orders of magnitudes higher than those at other sites. Guardia et al. (2010) detected DP in
146 sewage sludge from a WWTP in USA ranging from 112 to175 ng/g TOC with the content of
147 the TOC ranging from 7% to 28% (Guardia et al., 2010). Assuming all of the sewage sludge
148 contained 7% TOC, the calculated lowest DP concentration should be 7.8 to 12.3 ng/g dw. It
149 was higher than most of reported concentrations in sediments except Lake Ontario (Canada),
150 which indicates that the WWTP might be a major DP source for adjacent area. The three
151 highest concentrations of DP in the coastal sediments of Jiaozhou Bay were found near the
152 Haibo estuary, which may be influenced by the WWTP, too. In riverine sediments from the

153 Dague River, the DP concentration in the upstream sediment (3.0 pg/g dw) was one order of
154 magnitude lower than that in the downstream sediment (56.9 pg/g dw). Discharge from the
155 Jiaozhou town (36.17° N, 120.00° E) located near Dagu River might be the local source of
156 DP.

157 Concentration of DP in the sediment collected near the Qingdao Harbour, which was the
158 third busiest habour in China, was 31.8 pg/g dw, however, in farther locations, the levels were
159 one order of magnitude lower (4.0 and 2.0 pg/g dw).

160 In Taozi and Sishili Bay, the mean concentrations of DP (33.8 and 64.5 pg/g dw,
161 respectively) were higher than that of Jiaozhou Bay (23.7 pg/g dw). In Sishili Bay, high
162 concentrations were not found along the coastal zone but in the inner bay. Ren et al. (2008)
163 reported significant correlation between the airborne DP concentration and the population
164 over one million in one city. Yantai City has a population of 1 800 000 which is smaller than
165 that of Qingdao City (a population of 2 296 000). The DP concentration in Yantai should be
166 lower than that of Qingdao, which indicated the high levels in the sediments may not mainly
167 come from the local atmospheric deposition. Harbors were also suspected as possible source
168 because of intensive human activities. Yantai Harbor is settled at the west coast of Sishili
169 Bay, but it is smaller than the Qingdao Harbor (southwest of Jiaozhou Bay), which means
170 Yantai Harbor may not contribute much to the DP contamination in Sishili Bay. There was no
171 intensive industrial source reported in this area, either. The only suspected local source is the
172 WWTP settled at the Zhifu Island. This WWTP has limit ability to treat the city domestic and
173 industrial wastewater, and discharges waste water directly into the sea.

174

175 *3.2 Franctional abundances of Dechlorane Plus isomers*

176 The f_{syn} value was calculated as *syn*-DP/(*syn*-DP+*anti*-DP) (Qi et al., 2010). The mean f_{syn}
177 values for Jiaozhou, Taizi and Sishili Bay were 0.29 ± 0.06, 0.18 ± 0.05, and 0.15 ± 0.07,
178 respectively. Wang et al. (2010) reported the f_{syn} for commercial DP mixture to be 0.40 in

179 China which is higher than the DP (f_{syn} = 0.2 to 0.35) produced and widely used in North
180 America(Hoh et al., 2006; Tomy et al., 2007). In Chinese coastal sediments, the values of f_{syn}
181 were all below 0.40, and only 8 out of 48 samples showed ≥ 0.35 f_{syn} values. This indicates
182 definite stereo selective degradation of DP and the *anti*-DP seems more stable than *syn*-DP in
183 the coastal sediments.

184 In the semi-closed Jiaozhou Bay, as shown in Table 1, most of f_{syn} values (0.21 to 0.39,
185 exclude one site with value of 0.11) were close to the technical DP mixture. They were
186 comparable with that of Huai'an canal sediments (0.24 to 0.30) which were close to the
187 manufacture area (Wang et al., 2010b). The DP in this bay should come from the input of the
188 technical mixture from the adjacent region.

189 Both Sverko et al. (2007) and Tomy et al. (2008) found that the f_{syn} in the Lake Ontario
190 was below 0.20, but the concentrations of total DP were much higher than that of the
191 sediments from other places, e. g. Lake Winnepig (30.0±3.2 pg/g dw) and Lake Erie (0.061 to
192 8.62 ng/g dw)(Tomy et al., 2007; Sverko et al., 2008). The same trend was obvious in this
193 study. In Sishili and Taozi Bay, the concentrations were significantly correlated with the f_{syn}
194 values ($r = 0.58$, $n=19$, $p<0.05$), and the higher concentration was accompanied by the lower
195 f_{syn} values. As opposed to Jiaozhou Bay, Sishili and Taozi Bay are more open. Coastal
196 sediments from these bays may not only receive input from land, but also the inner Bohai Sea
197 (Cheng and Gao, 2000). As shown in Fig. 4, the two bays were at the south-east of the Bohai
198 strait. Marine current runs though the narrow strait with a high speed carrying the sediments
199 from the Bohai Sea. When it comes to the Yellow Sea, it slows down at the spacious area. As
200 a result, some suspended sediments settle down, and some bottom sediments stop moving.
201 The arrows in Fig. 4 show the main direction of the sediment movement which supported that
202 Taozi and Sishili Bay receive sediment from Bohai Sea. Sverko et al. (2008) reported f_{syn}
203 below 0.1 in Niagara and Lake Ontario non-depositional sediments. This suggests that the
204 foreign sediment would enrich *anti*–DP and decrease the f_{syn} value. The Bohai Sea is a hot

spot for POPs study. High concentrations of PAHs, PBDEs and PCBs have been found in the sediments (Pan et al., 2010; Zhao et al., 2005; Liu et al., 2009). Though there is no DP data in Bohai Sea reported, high concentration of DP can be supposed as it receives large input of contaminations from surrounding cities. More works should be conducted to figure out the sources of DP in Bohai Sea.

3.3 Dechlorinated species of Dechlorane Plus

Sverko et al. (2008) reported DP decomposition in the environmental compartments. To guarantee the ion fragment clusters truly come from the environmental samples but not the artifices during analysis procedure, a new GC liner was applied before injection. Moreover, one 10 pg/μL quality control standard (including *syn*-DP, *anti*-DP, aCl10DP and aCl11DP) was analyzed after every eight samples to supervise the possible degradation in GC-MS system. The relative deviation of all target compounds in QC standards were within 15% of the original standards in all cases, which means no obvious decomposition of *syn*- and *anti*-DP had happened in the GC-MS system. In all of the 48 samples, 9 samples showed aCl11DP concentration above the MDL, whereas, all the samples showed aCl10DP concentration below the MDL. 7 out of 13 samples were detected aCl11DP in Sishili Bay, and the degradation rate ((aCl10DP+aCl11DP)/(*syn*-DP+*anti*-DP+aCl10DP+aCl11DP)%) ranged from 0.9% to 5.1%. Both photodegradation and aerobic microbial degradation may happen in surface sediments which result in the appearance andd dechlorinated species (Sverko et al. 2008).

3.4 Inventory of DP in the marine sediment

The inventory of DP in sediment in the bays was calculated by the equation as follow:

$$\text{Inventory} = C \times \rho \times A \times D \times a$$

where C (pg/g) is the mean concentration of DP in the bay, ρ (g/cm^3) is the dry density of the sediment. A (cm^2) represents the area of the bay, D (cm/a) is the sedimentation rate and a (a) is years which are interesting in this study.

The parameters for the three bays were presented in Table 3. The sedimentation rate of the three bays were close to 1 cm/a, so it is estimated for five-year inventories. They were 0.33 kg in Jiaozhou Bay, 0.37 kg in Taozi Bay and 0.49 kg in Sishili Bay. The smallest Sishili Bay (130 km^2) stores most of DP (0.49 kg). Qiu et al. (2007) reported the inventory of DP per area (9cm and 16cm depth) was 120 ng/cm^2 in Lake Ontario, and Wang et al. (2010) calculated the 20.3±7.5 cm soil inventory to be 15000 to 110000 ng/cm^2 in Huai'an. The 5 cm depth burdens of DP in per area were 8.5 ng/cm^2 in Jiaozhou Bay, 20.1 ng/cm^2 in Taozi Bay and 37.7 ng/cm^2 in Sishili Bay, respectively. Sediment would store DP for a long time as one of the possible major sink(Wang et al., 2010). With a half-live of 14 years, DP is bioavailable for many aquatic organisms, and it might be harmful especially to the benthic biota (Ismail et al., 2009).

4. Conclusions

DP was detected in all surface sediments from three Chinese bays, but the different distribution patterns and sources were found. In semi-closed Jiaozhou Bay, higher concentrations were detected near the WWTP which was considered as the major DP source. Usage of commercial DP products resulted in the contamination in Jiaozhou Bay. In contrast, high DP concentrations were found in the inner places of Sishili Bay with low f_{syn} . DP in Taozi and Sishili Bay may come from the transportation from Bohai Sea and WWTP at Zhifu Island could elevate DP level in Sishili Bay. The five-year inventories in the three bays indicated Sishili Bay stores more DP than other two bays.

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Figure caption and table title

Figure 1. The sampling sites in Taozi, Sishili and Jiaozhou Bay

Figure 2. Distributions of DP in surface sediments from Jiaozhou Bay

Figure 3. Distributions of DP in surface sediments from Toazi and Sishili Bay

Figure 4. Sketch map of sediment transportation. Five-pointed stars represent some cities with population over one million

Table 1. Concentritions (pg/g dw) of syn-DP, anti-DP, $aCl_{10}DP$, and $aCl_{11}DP$ and the f_{syn} value in surface sediments of Jiaozhou Bay, Taozi Bay and Sishili Bay, North China

Table 2. Comparisons of DP level in China and other area in the world

Table 3. Inventory parameters of Jiaozhou. Taozi and Sishili Bay. China

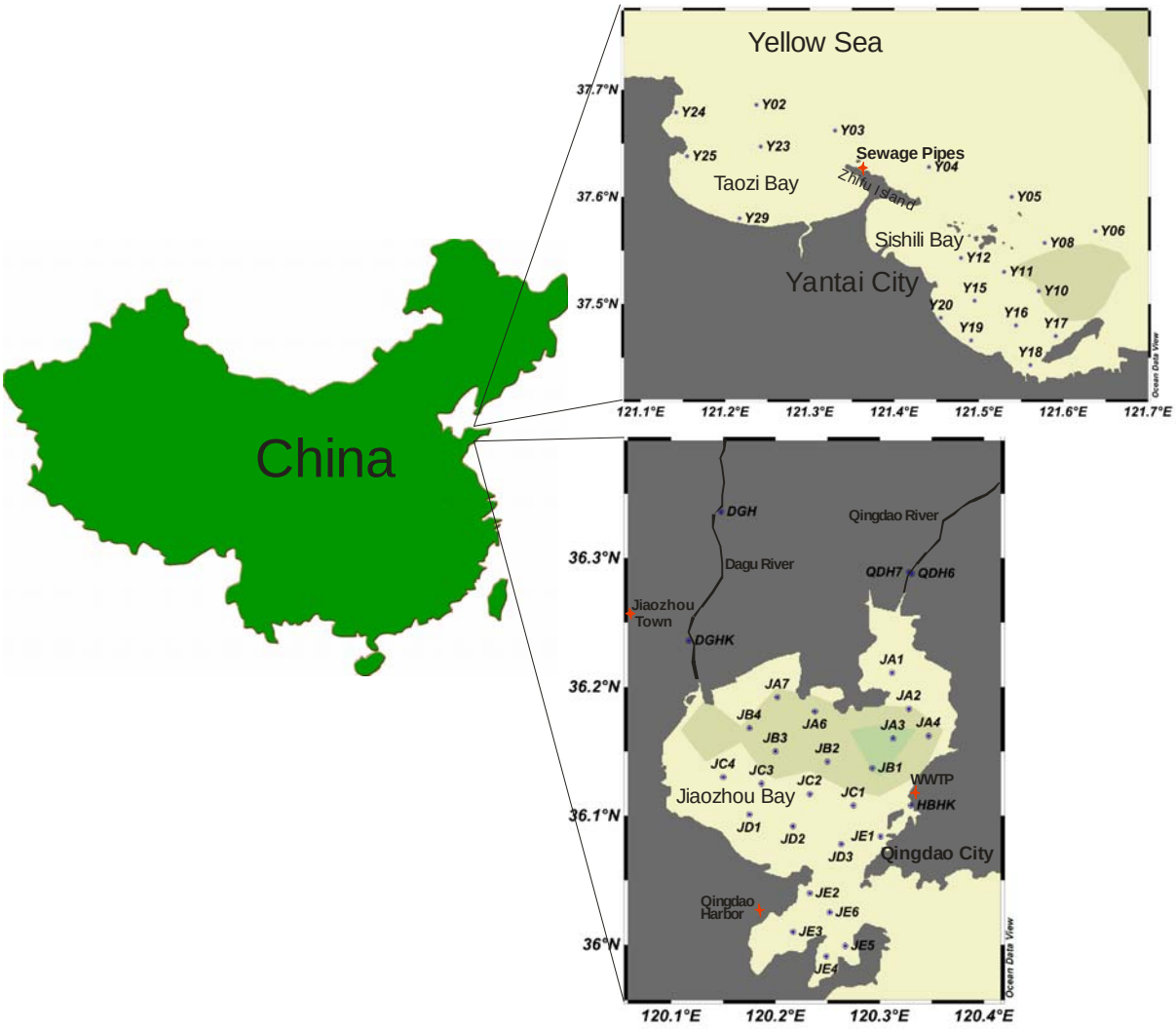

Fig. 1 The sampling sites in Taozi, Sishili and Jiaozhou Bay

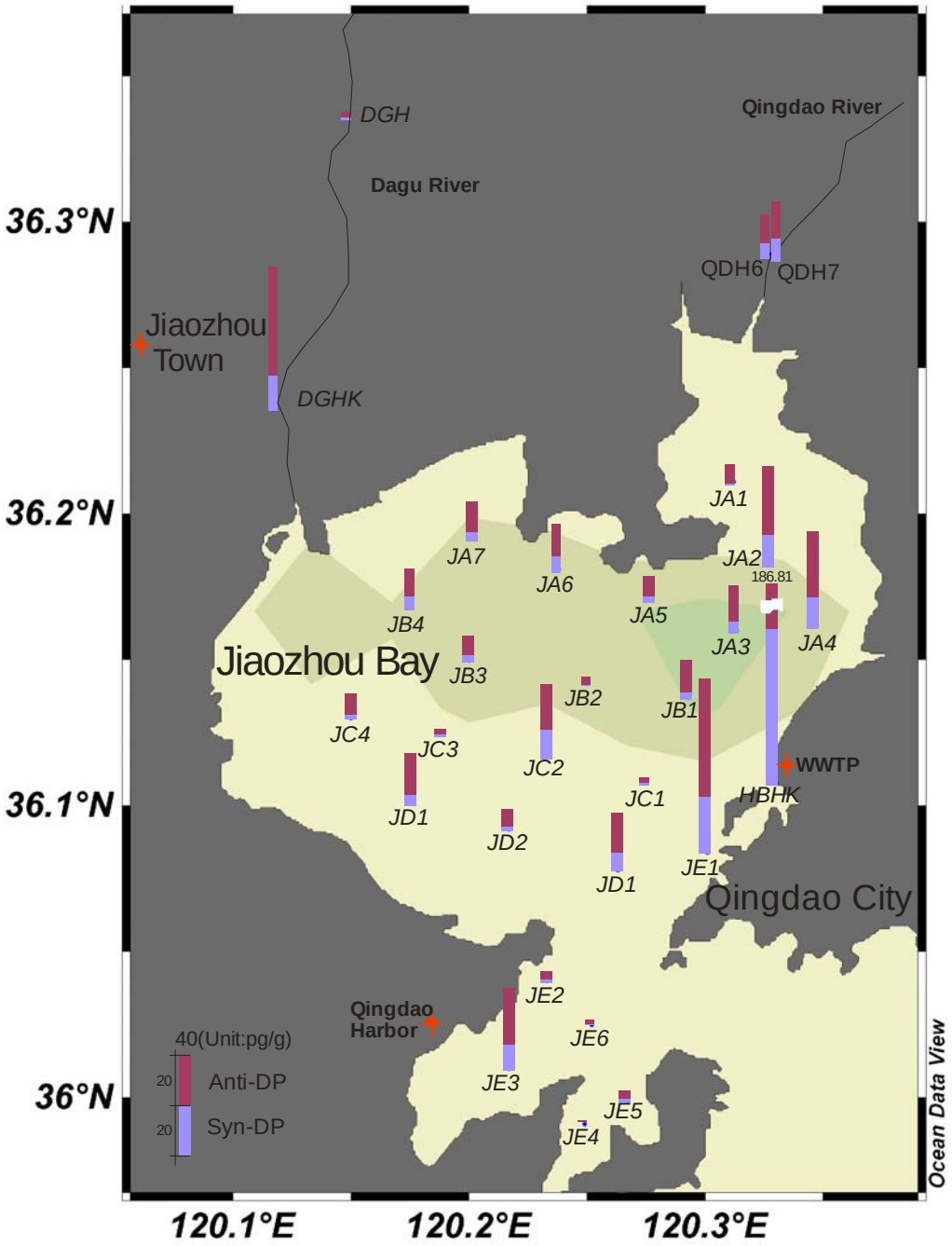

Fig. 2 Distributions of DP in surface sediments from Jiaozhou Bay

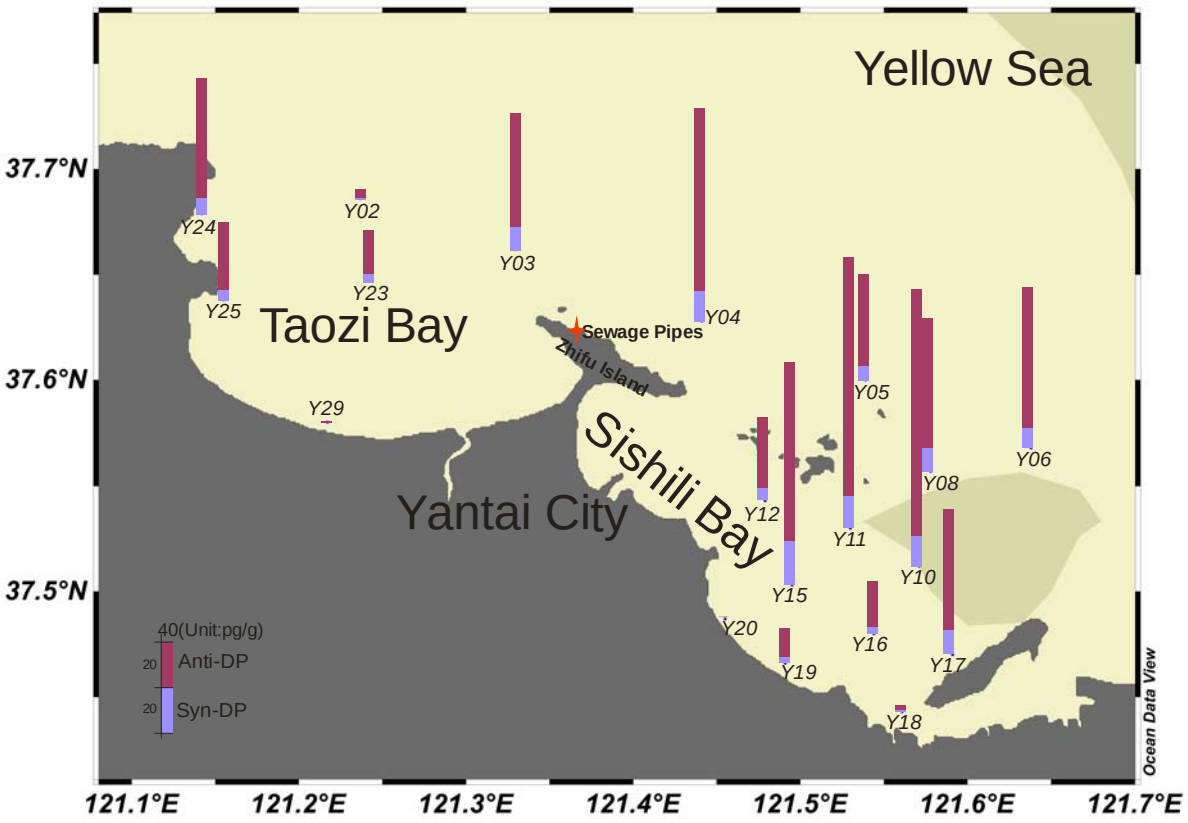

Fig. 3 Distributions of DP in surface sediments from Toazi and Sishili Bay

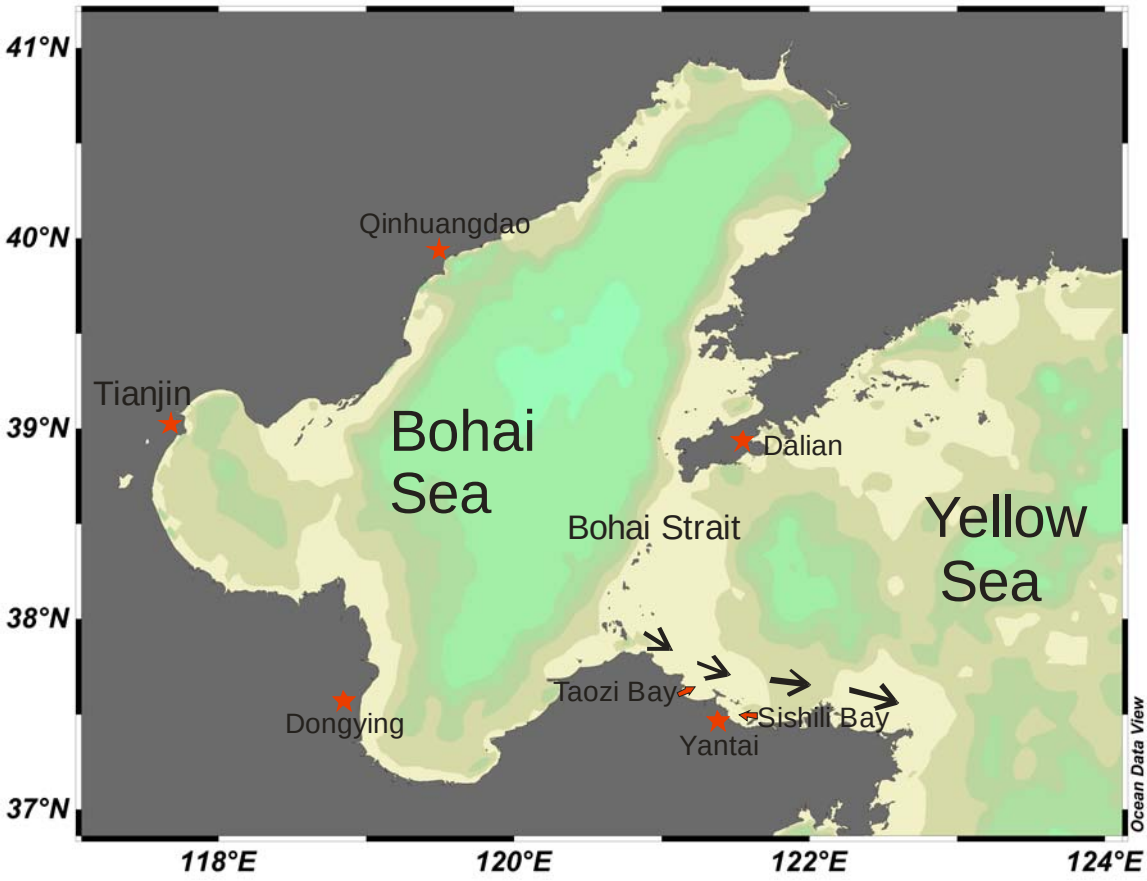

Fig. 4 Sketch map of sediment transportation. Five-pointed stars represent some cities with population over one million