Levels and Mass Burden of DDTs in Sediments from Fishing Harbors: The Importance of DDT-Containing Antifouling Paint to the Coastal Environment of China

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DDT remains an important type of persistent organic pollutant (POP) in the environment of China. One of the current applications of DDT in China has been through antifouling paint for fishing ships as an active component. It has been estimated that approximately 5000 t of DDT was released into the Chinese coastal environment during the last two decades. Therefore, sediments in coastal fishing harbors of China may be the important sinks of DDT. In this study, DDT and its metabolites in 58 sediment samples from nine typical fishing harbors along the coastal line of China were characterized to assess their accumulation levels, sediment burdens, and potential ecological risks. The concentrations of DDTs ranged from 9 to 7350 ng/g dry weight, which were generally 1−2 orders of magnitude higher than those of the adjacent estuarine/marine sediments. The high concentrations of DDT coupled with the lower concentrations of HCH and TOC clearly indicated a strong local DDT input, i.e., DDT-containing antifouling paint, within the fishing harbors. A significant correlation between the total DDT concentrations and p,p′-DDT concentrations further confirmed the existence of fresh DDT input. The overall burden of DDTs within the upper 10 cm sediment layer in the fishing harbors of the Pearl River Delta, southern China, was estimated to be 1.0−5.7 t, which was several times higher than the DDT accumulation in the surface sediment of the Pearl River estuary. The concentrations of DDTs in the fishing harbor sediments significantly exceeded the sediment quality guidelines on the basis of adverse biological effects. The absence or low concentrations of *p,p′*-DDD in aquatic organisms and human may imply that either *p,p′*-DDD may be less bioaccumulated by fish and human, or is biotransformed to other metabolites. A national ban of DDT as an additive to antifouling paint was implemented in 2009 in China; however, the legacy high DDT burden in the coastal fishing harbors needs further monitoring and proper management.

Introduction

DDT is still one of the major persistent organic pollutants (POPs) in China (*1, 2*). For example, the long-term Mussel Watch program in Asia found that the mussel samples from coastal China had the highest concentrations of DDTs in Asia (*3*); while a recent monitoring campaign of POPs in the atmosphere across Asia, using passive sampling techniques, also reported that the DDT concentrations in the atmosphere over the eastern coastal zone of China, in particular in the Pearl River Delta (PRD) of South China, were the highest in East Asia (*4*). In addition, the DDT residues in human breast milk from Guangzhou and Hong Kong within the PRD were also among the highest in the world (*5*).

DDT was widely used in China as a pesticide from the 1950s until the production of DDT was officially banned in 1983. Over more than 30 years, the total production of DDTs in China was estimated to be 0.4 million metric tons (MT), accounting for 20% of the total world production (*6*). Although the agricultural use of technical DDT has been prohibited, DDT is still allowed to be used for hygiene purposes, such as malaria control, in Asian developing countries, including China, upon the 5-year exemption of the Stockholm Convention (*7*). Qiu et al. (*8, 9*) revealed that dicofol, a new pesticide, contains high DDT content at an average 20% in the Chinese formula. However, this seems inadequate to explain why such high concentrations of DDTs mostly occurred in the atmosphere of the coastal regions, provided that dicofol is used both in inland and coastal regions of the country. Recently, high DDT levels were found in the atmosphere of the PRD, concurring with the fishing suspension season (June−August) in the northern South China Sea, and, for the first time, the seasonal use of DDT-containing antifouling paint was identified to be an important source of DDTs in the coastal zone of China (*10*−*13*).

The average annual production of DDT is estimated to have been 4519 MT during 2000−2003 in China, and there are no DDT imports from other countries. About 4% of total DDT production was used as an additive for the production of antifouling paint. It is estimated that about 150−300 MT of DDT per annum enters the environment through antifouling paint usage in China, corresponding to roughly 5% by weight in the antifouling paint products. Most of the products are used in small to medium size fishing boats (*14*). Therefore, sediments in fishing harbors along the Chinese coastline are expected to be heavily contaminated by DDTs from the use of antifouling paint.

Here we report the levels, inventory, and potential ecological risk of DDTs in the sediments of fishing harbors along the coastline of China. The results prove that a large

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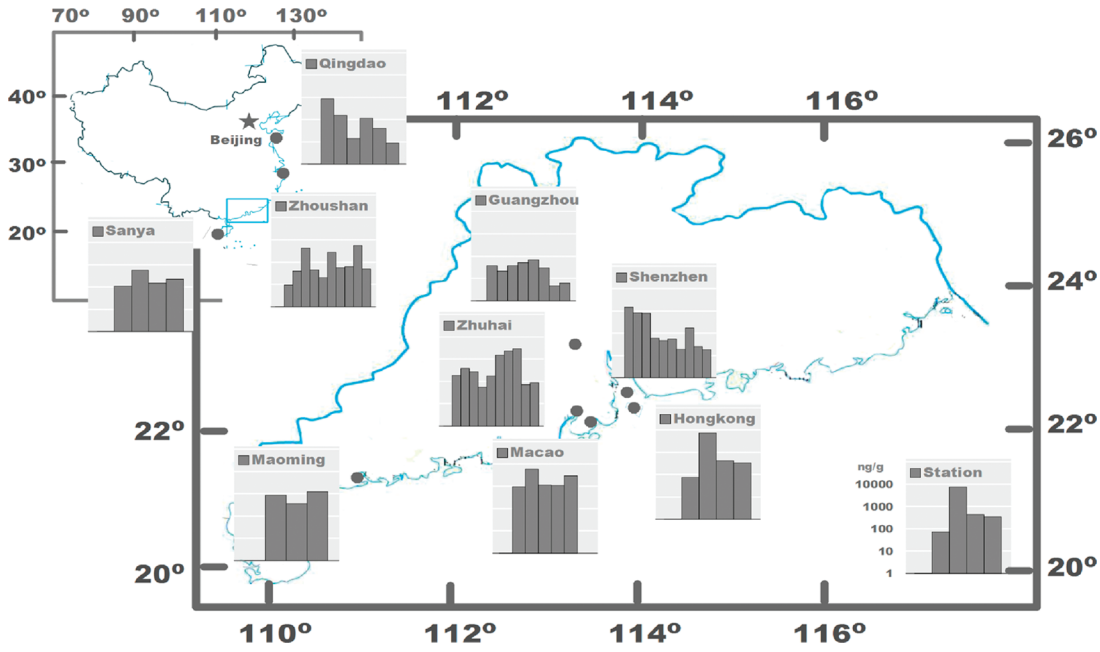

FIGURE 1. Sampling locations and concentration levels of DDTs in sediment from the fishing harbors in China.

burden of DDTs exits in the fishing harbor sediments, denoting that the phasing out of DDT-containing antifouling paint is of urgent need for the protection of the coastal environment and human health.

Materials and Methods

Sediment Sampling. Nine fishing harbors were selected in this survey, with 5 stations located in the PRD, e.g., Guangzhou (GZ), Zhuhai (ZH), Shenzhen (SZ), Macao (MC), and Hong Kong (HK). Detailed information about the sampling stations is provided in Figure 1 and Table S1. A total of 58 sediment samples were collected from August 2007 to November 2007, using a Kajak corer (KC Denmark Co., Denmark). At each site, the upper 10 cm sediment was collected, sealed in polytetrafluoroethylene (PTFE) bags, immediately transferred to the laboratory, and stored frozen until analysis.

Analysis. In the laboratory, sediment samples were freeze-dried for 24 h, pulverized, and sieved through 80-mesh stainless steel. About 10 g of the subsamples were Soxhlet-extracted for 48 h with dichloromethane (DCM) at a rate of 4−6 cycles/h. A mixture of 2,4,5,6-tetrachloro-*m*-xylene (TC*m*X) and decachlorobiphenyl (PCB209) was added to each of the samples as surrogate standards prior to extraction. Activated copper granules were added to the collection flask to remove elemental sulfur. The extract was concentrated and solvent-exchanged to hexane and purified on an 8 mm i.d. alumina/silica column packed, from the bottom to top, with neutral alumina (6 cm, 3% deactivated), neutral silica gel (10 cm, 3% deactivated), 50% (on a weight basis) sulfuric acid silica (10 cm), and anhydrous sodium sulfate. Alumina, silica gel, and anhydrous sodium sulfate were Soxhlet-extracted for 48 h with DCM, and then baked for 12 h at 250, 180, and 450 °C, respectively, before use. The column was eluted with 50 mL of dichloromethane/hexane (1:1) to yield the organochlorine pesticide (OCP) fraction. The fraction was solvent-exchanged to *n*-hexane and concentrated to 0.5 mL under a gentle nitrogen stream. A known quantity of pentachloronitrobenzene (PCNB) was added as an internal standard prior to gas chromatography-electronic capture detector (GC-ECD) analysis.

Six major isomers and metabolites of DDT, i.e., *p,p′*-DDE, *o,p′*-DDE, *p,p′*-DDD, *o,p′*-DDD, *p,p′*-DDT, *o,p′*-DDT, as well as α-, *β*-, *γ*-, and *σ*-HCH were measured by GC-ECD (Agilent-6890 GC system, Hewlett-Packard, USA). A CP-Sil 8 CB capillary column (50 m, 0.25 mm, 0.25 μm; DB-5MS, Agilent, USA) was used, with helium as carrier gas at 1.2 mL min^{-1} under constant flow mode. Helium was filtered with moisture, hydrocarbon, and oxygen traps before entering the GC-ECD system. The oven temperature began at 60 °C for 1 min and increased to 290 °C (10 min hold time) at a rate of 4 °C min^{-1}. Split/splitless injection of a 1 μL sample was performed with a 5 min solvent delay time. Injector temperature was 250 °C. The inlet degradation of DDT was checked daily and controlled within 15%, using a pure *p,p′*-DDT stardard. The original GC-ECD traces of selected sediment samples are provided in the Supporting Information (see Figure S1).

Quality Assurance/Quality Control (QA/QC). All data were subject to strict quality control procedures, including the analysis of blanks and spiked samples. Six method blanks (solvent), 6 duplicate samples, and 6 spiked blanks (standards spiked into solvent) were analyzed. The limit of detection (LOD) for each compound, estimated as three times response of signal-to-noise of 3:1 in blank sample, was 0.17 ng/g for *p,p′*-DDE, 0.87 ng/g for *o,p′*-DDE, 0.10 ng/g for *p,p′*-DDD, 0.60 ng/g for *o,p′*-DDD, 0.17 ng/g for *p,p′*-DDT, 0.67 ng/g for *o,p′*-DDT, 0.04 ng/g for α-HCH, 0.06 ng/g for *β*-HCH, 0.04 ng/g for *γ*-HCH, and 0.06 ng/g for *σ*-HCH. The average surrogate recoveries were 75.4 ± 10.7% and 98.9 ± 10.0% for TC*m*X and PCB209, respectively. The reported concentrations of OCPs were not corrected for surrogate recoveries.

Results and Discussion

Levels and Sources of DDTs in the Harbor Sediments. Table 1 illustrates the concentration ranges of individual DDTs in the surface sediments from the fishing harbors. The concentrations of DDTs ranged from 9 to 7350 ng/g, which were generally 1−2 orders of magnitude higher than those from adjacent estuarine/marine systems (Table S3), and much higher than those reported in other places of the world (Table S3), except for several harbor sediments, e.g., the Inner Harbor of Macao (1630 ng/g, ref *15*), Xiamen harbor (4.5−311 ng/g, ref *16*) in southeast China, and Alexandria Harbor in Egypt (<0.25 to 885 ng/g, ref *17*).

Sediment is the important sink for hydrophobic organic compounds (HOCs) in coastal environments. In addition to DDTs, HCHs (another mostly widespread organochlorine pesticide) were also measured in the sediment samples (see

TABLE 1. Concentrations (ng/g Dry Weight) of DDTs in Surface Sediments of Fishing Harbors in China

city	city code	*o,p′*-DDE	*p,p′*-DDE	*o,p′*-DDD	*p,p′*-DDD	*o,p′*-DDT	*p,p′*-DDT	DDTs
Sanya	SY	0.13−1.0	11−130	15−87	54−270	3.7−27	12−46	108−561
Maoming	MM	0.30−1.3	20−98	50−210	190−830	34−140	67−180	361−1250
Macao	MC	1.5−4.1	170−420	130−760	520−2900	29−530	65−1200	967−5810
Zhuhai	ZH	0.11−2.5	9.3−280	9.7−520	32−2000	0.53−130	2.2−310	55−3040
Hong Kong	HK	0.20−3.0	20−300	8.7−650	34−2600	4.3−1500	9.4−2300	76−7350
Shenzhen	SZ	0.17−2.0	2.0−220	2.6−270	5.4−940	0.10−21	0.36−37	17−1460
Guangzhou	GZ	N.D.[a]−0.13	3.2−13	2.4−6.7	9.5−27	3.0−12	3.6−10	21−68
Zhoushan	ZS	N.D.−0.36	1.6−91	1.3−40	3.5−140	0.58−110	2.2−200	9.0−563
Qingdao	QD	N.D.−0.49	1.0−46	1.0−110	3.9−450	0.47−73	2.0−160	9.0−839

[a] N.D. = Not detectable.

Table S2 for detailed results). The results showed that the total HCH concentrations (the summation of α-, β-, γ-, and σ-HCH isomers) ranged from N.D. to 12 ng/g, which were very similar to the reported concentrations in sediments from the adjacent coastal area and river estuary, a sharp contrast to the high concentrations of DDTs in the harbor sediments. The overall ratios of DDTs/HCHs in the fishing harbor sediments (mean 450) were much higher than those of the adjacent coastal/estuarine sediments (Table S3). Spearman's rank correlation coefficient (RS) is significant to the ratio of DDTs/HCHs and concentrations of DDTs in sediment ($R^2 = 0.59$ $p < 0.01$). This reflects that, unlike HCHs, there is input of DDTs within the fishing harbors.

The total DDT concentrations were also normalized to the TOC contents in sediment, a key factor influencing the accumulation of HOCs in sediment. The TOC content of all the fishing harbor sediments ranged from 0.12 to 2.6% (see Table S2), well within the range for estuarine/marine sediments in the world. In the studied fishing harbors of the Pearl River Delta, the TOC normalized concentrations of DDTs were 1.5−321 μg/g TOC, with a mean value of 58 μg/g TOC, while much lower concentrations of 0.11−13 μg/g TOC were observed for the Pearl River estuary sediments (*15, 16*). For HCHs, the TOC normalized concentrations were 0.03−0.70 g/g TOC, and HCH is significantly correlated to the concentrations of TOCs in sediment ($R^2 = 0.37$ $p < 0.01$). The above evidence shows that unlike HCHs, TOC was not the key factor responsible for the accumulation of DDTs in the harbor sediments, further pointing to a strong local input in the fishing harbor.

The most possible DDT source in the fishing harbors is the release of DDT from fishing boats with DDT-containing antifouling paint. China is the only country that continues using DDT-based antifouling paint on its ships. Along China's 18,000 mile coastline around 300,000 fishing vessels work and use about 10,000 MT of antifouling paint, of which approximately half (5000 MT) is DDT-based (*14*). In fact, a previous investigation of DDT in commercially available antifouling paints in the PRD reported a high DDT residue of 525−2360 μg/g in the products (*11*), although much lower than the earlier estimation reporting 5% DDT in antifouling paints leading to the release of 250 MT of DDT into the marine environment of China each year (*14*). In addition, the heavy metal Cu, which is also an active component in antifouling paints, showed good correlation with DDTs in these fishing harbor sediments ($R^2 = 0.52$, $p < 0.01$) (Figure S2). Hence it is clearly implied that the enrichment of DDTs within the fishing harbors was related to fresh input from DDT.

Compositions of DDTs. It was widely reported that DDT is likely to be dehydrochlorinated to DDE under aerobic conditions, whereas in an anaerobic setting DDD is the degradation product (*18*). The ratios of DDD/DDE can thus be used as an indicator of redox condition in sediment. Almost all of the sediments from the fishing harbors in this study showed DDD/DDE ratios ranging from 1.1 to 11.8 (mean 4.5), except for two samples with <1 values in the sediments SZ10 (0.60) and QD6 (0.58). The dominance of DDD over DDE in the sediments indicated a reductive dechlorination of DDT to DDD under anaerobic conditions in the fishing harbor sediments.

DDD and DDE dominated the compositions of DDTs in most of the harbor sediment samples, and the sum of DDD and DDE represented about 40−98%, with an average value of 81%, of the total DDTs. Strong correlation was found between the sum of DDD and DDE and DDTs in the fishing harbor sediments ($R^2 = 0.92$ $p < 0.01$) (Figure S3). Similarly, it should be pointed out that the sediments with higher total DDT concentrations generally showed higher proportions of DDT; and the correlation coefficient between total DDT concentrations and DDT concentration was 0.78 ($p < 0.01$; Figure S3). This clearly indicates that the enrichment of DDTs within the fishing harbor sediments was related to the fresh input of DDT and its degradation products. Furthermore, good correlation between DDD and DDE was observed in the fishing harbor sediments. This suggests a strong DDT original source in the fishing harbors.

Distribution of DDTs within Individual Harbor. Sediments with high concentrations of DDTs were generally located at sheltered locations and/or mooring areas within the fishing harbors. The concentrations of DDTs decreased sharply, by 1 or 2 orders of magnitude, from the harbor centers to the outside channel sites.

For example, in Zhuhai fishing harbor, those sites with total DDT concentrations higher than 1000 ng/g were located at the mooring area in the right side (Figure 2) of the harbor. The concentrations of DDTs were lower in the temporary mooring area and ship channel (170−380 ng/g dw, (refer to Figure 2). The concentrations dropped to 71−88 ng/g at the two outside sites. In Shenzhen fishing harbor, the total DDT concentrations gradually declined from 1500 ng/g at the main mooring area to 18 ng/g dw at the harbor mouth (Figure 2).

However, in Maocao Inner Harbor, with very intensive boat mooring activities and poor water mixing/circulation, a narrow range of total DDT concentrations (970−5800 ng/g) was observed. Figure 2 shows that higher proportions of DDT generally coincided with higher total DDT concentrations in sediments from the mooring areas, despite the dominance of DDD and DDE.

The decline in the concentrations of DDTs from fishing harbor sediment to the outer harbors is possibly due to the higher rate of DDT contamination in the fishing harbor sediments on account of the DDT related antifouling paints used in the fishing boats. Outer harbors being away from the source of contamination showed lower DDT levels. Furthermore the outer harbor sediments are more open to higher water mixing or dilution due to closer proximity to the mainstream river or open sea.

Sediment Burden of DDTs. Based on the results obtained from this study, we attempted to assess the sediment burden of DDTs in the fishing harbor sediments in the PRD region.

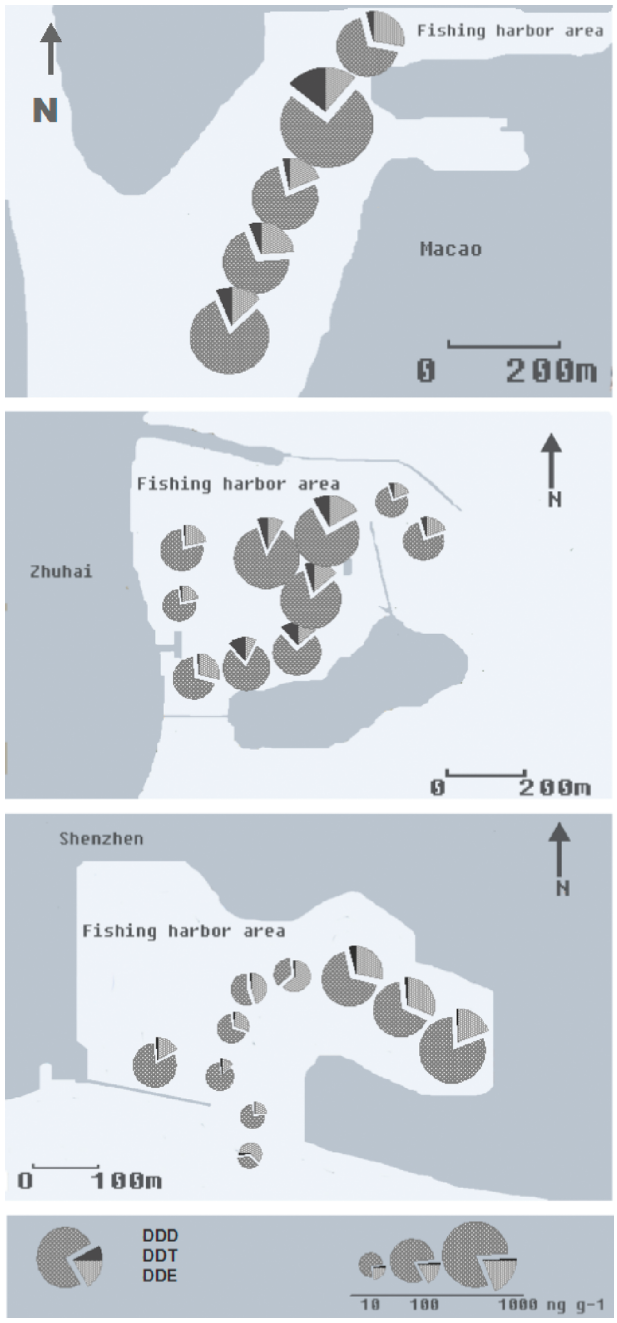

FIGURE 2. Spatial distribution of DDTs in sediment from Macao, Zhuhai, and Shenzhen harbors in the Pearl River Delta.

The method assumed that the concentration of DDTs and the distance from the central harbor showed a good exponential correlation due to mixing and dilution in fishing harbor areas. Concentration data of 35 sediment samples from the PRD region were used.

The mean value of DDTs is 480 ng/g (median 87 ng/g) after excluding the three abnormally high values as obtained from the box plot (Figure S4).

The mass inventory (I) was calculated according to the following formula

$$I = CAdp$$

where C is the concentration of DDTs in sediment, A is the total water area of fishing harbors in the PRD region (80 km^2) (*13*), d is the assumed sediment density of 1.5 g/cm^3, and p is sediment thickness (in this study it is 10 cm).

In the box plot the median (87 ng/g dw) and mean (480 ng/g dw) values reflecting the lower limit and the upper limit, respectively (Figure S4), were adopted to deduce the conservative estimation of total DDT. The calculated inventories ranged 1.0−5.7 t for total DDTs in fishing harbor sediments of the PRD. In a previous study using a similar approach, it was calculated that 0.4 and 1.4 t of total OCPs were accumulated in the regional sediments (0−5 cm) of the Pearl River Estuary and the northern South China Sea (*19*). Clearly, the mass inventories of DDTs in fishing harbor sediment greatly surpass the environmental loadings of the regional sediments, highlighting the importance of DDT-containing antifouling paint to the DDT contamination along the coastal line of China.

Ecological Risk Assessment. Although the area of each fishing harbor is generally less than 1 km^2, the overall water area of fishing harbors in the coastal zone still represents a considerable proportion of the total water area. For example, the water area for all fishing harbors in the Pearl River estuary reaches 84 km^2, which is approximately 1/25 of the total water area of the estuary (2100 km^2). This denotes the importance of an ecological risk assessment of high DDT contents in the harbor sediments.

The DDT concentrations in the fishing harbor sediments were compared with the sediment quality criteria to assess the potential ecological risk (see Table S4). All the individual species of DDTs from the sediments were above the corresponding ER-L values. The maximum *p,p′*-DDD concentration was 140 times higher than its ER-M value, and *p,p′*-DDT exceeded by maximum 330 times of the quality criteria specified by the U.S. Environmental Protection Agency (*20*) and by Canadian Council of Minister of the Environment (*21*). For the total DDT concentrations, 65% samples were above the ER-M value, with most of the exceptions in the offshore harbors of Zhoushan and Qingdao.

Implication to Food Chain Transfer. As mentioned above, *p,p′*-DDD accounted for a large proportion of *p,p′*-DDTs, and an average of 64% of DDTs in the fishing harbors presented as *p,p′*-DDD. The sediment burden of *p,p′*-DDD was calculated to be in the range of 0.6−2.8 t in the fishing harbor sediments in the Pearl River Delta. The question may arise whether the *p,p′*-DDD can be bioaccumulated and transferred in the coastal food chain and, eventually, reach humans.

It was reported that the estimated dietary intakes (EDIs) of DDTs via fish consumption in China were higher than those in many other countries (*22, 23*). High DDT concentrations were observed in dermersal fishes from the Pearl River estuary (*13*), and the concentrations of DDTs in human breast milk in Guangzhou and Hong Kong within the PRD were among the highest in the world (*24*). However, in all the fish samples and human breast samples, *p,p′*-DDE and *p,p′*-DDT were identified to be the dominant DDT isomer (*24*). It was widely reported that, in organisms located at the upper trophic levels of a food web, such as birds, whales, and humans, *p,p′*-DDE was the most predominant DDT isomer accounting for more than 90% of the total *p,p′*-DDTs (*25*−*28*). This could be attributed to the greater accumulation rates for *p,p′*-DDE by organisms, and to the metabolization of *p,p′*-DDT to *p,p′*-DDE by organisms (*27, 29, 30*).

It is well-known that significant proportions of *p,p′*-DDD to *p,p′*-DDTs were observed in littoral sediment (*15*) and even in benthic species, such as mussels and shellfish (*31, 32*), but little information is available about the bioaccumulation and metabolization of *p,p′*-DDD in the food chain. The absence or very low concentrations of *p,p′*-DDD in aquatic organisms and humans may imply that either *p,p′*-DDD may be less bioaccumulated by fish and humans, or it is, more likely, biotransformed to other metabolites, such that it cannot be tracked down in the food chain in its original form.

Implications to Coastal Environment Management. The high concentrations of DDTs in the fishing harbor sediments implied that a considerable amount of DDTs may be accumulated from the use of DDT-containing antifouling paint. Both natural hydrological processes, such as water mixing/exchange and stormwater flushing, and anthropogenic activities, in particular dredging activities, can result in potential disperse of DDTs from fishing harbors to regional waters. For instance, large-scale silt removing and dredging projects have been carried out at regular intervals in harbors of the Pearl River Delta, and some 350,000 m^3 of silt were dredged and abandoned in the nearby coastal region between February and March, 2007 in Zhuhai fishing harbor (*33*).

China officially banned the application of DDTs as an additive in antifouling paints in May 2009. However, the legacy high DDT burden in the fishing harbor sediments needs careful monitoring and proper management in the future.

Acknowledgments

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Supporting Information Available

Three tables showing relevant information for the sampling stations, the concentrations of compounds at each sampling station, including *o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDT, *p,p′*-DDT, α-HCH, *β*-HCH, *γ*-HCH, *σ*-HCH, the total organic carbon content, comparison of DDTs and HCHs in surface sediments from other estuarine and marine environments in the world, risk assessment guidelines and results. Four figures depicting the collected GC-ECD chromatogram of sediment samples with higher DDT concentrations, correlation between DDTs and Cu, correlation between DDTs and DDT, correlation between DDTs and sum DDD and DDE, correlation between DDD and DDE, and a box plot of DDT concentrations in sediment from the fishing harbors in the PRD. This material is available free of charge via the Internet at http://pubs.acs.org.

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