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Distribution and sources of organochlorine pesticides in sediments of the coastal East China Sea

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ABSTRACT

Forty-two surface sediment samples collected from the southern coastal East China Sea (ECS) were analyzed for organochlorine pesticides (OCPs). The results showed high concentrations of DDTs and parent DDT fractions in the nearshore area, suggesting that there is still a fresh input of these species from non-point sources, probably due to the recent usage of relevant products. However, for HCHs, the high concentrations with elevated β -HCH fraction were focused at the local major river estuaries, indicating that soils-released HCH input was mainly from the major river runoffs. The distribution and composition of OCPs in the coastal ECS could be more prominently controlled by the direct regional inputs rather than the general natural sorption process on particulate organic carbon. DDTs and chlordane are more important for their potential ecotoxicological risk on neighboring benthic community in the nearshore region and the local major river estuaries.

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1. Introduction

Organochlorine pesticides (OCPs) are a class of ubiquitous environmental persistent organic pollutants, and could pose a risk by causing adverse effects to human health and environment (Long et al., 1995). It was estimated that the worldwide total pesticides consumption mainly including dichlorodiphenyltrichloroethanes (DDTs), hexachlorocyclohexanes (HCHs) and Toxaphene was about 20 million tons until 2004 (Lohmann et al., 2007). In the meantime, a total of 4.9 million tons of HCHs and 0.4 million tons of DDTs had been produced in China, equivalent to one third and one fifth of the total world production, respectively (Wei et al., 2007). Although OCPs were restricted or banned for use in the early 1980s in China, these organic chemicals can still be routinely detected in various environmental compartments (i.e. in air, water, soil, sediments and organisms) (Hu et al., 2004; Doong et al., 2008).

The coastal East China Sea (ECS) is adjacent to the Zhejiang and Fujian provinces (Fig. 1). The previous surveys of pesticide consumption per unit area from 1985 to 1991 indicated that Zhejiang and Fujian ranked top one and top three, respectively (Wang et al., 2005). The consumption of pesticides in Zhejiang was about 9.96 kg km⁻² which was 3.7 times higher than the national average

level (2.71 kg km⁻²) in 1996 (Wang et al., 2005). In recent years, DDTs and chlordane are still being largely used for the special purposes (e.g. killing termites, and controlling diseases) in the two provinces, and the application of chlordane in Zhejiang province ranked 1st in China (Hu et al., 2004). Consequently, the high level of OCP contaminations was detected in sediments from rivers and bays within these two provinces (Zhou et al., 2006; Gong et al., 2007; Wang et al., 2009; Yang et al., 2010). As a significant sink for those persistent organic pollutants, the latest occurrences and potential risks of these land-based OCP species in the sediments of the coastal ECS should be concerned; however, this dataset of the region is still very limited. The objectives of this work are to examine the recent occurrence level, distribution and sources of OCPs, and to assess the potential biological risks of these OCPs in this area.

2. Materials and methods

2.1. Sampling

The sampling sites are shown in Fig. 1. Most samples were located in the southern inner shelf mud area of the ECS. The surface sediment samples were collected during the cruise conducted by Science 1 of Institute of Oceanology, Chinese Academy of Science in 2007. Surface sediment samples (0–3 cm) were collected using a stainless steel box corer. Sediment samples were wrapped in aluminum foil and stored at -20 °C until analysis.



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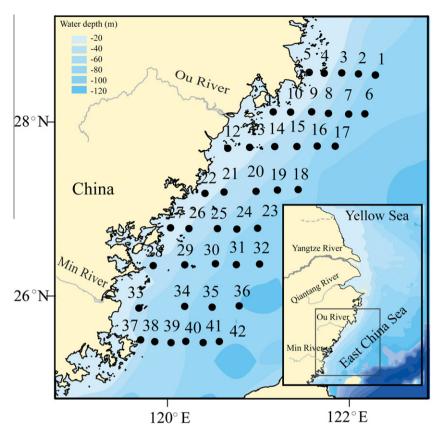


Fig. 1. Study area and locations of sampling sites in the coastal ECS.

2.2. Analysis of OCPs

A 100 ng mL⁻¹ standard solution of OCPs, including HCHs (α -HCH, β -HCH, γ -HCH, δ -HCH), DDTs (p,p'-DDT; o,p'-DDT; p,p'-DDD and p,p'-DDE), trans-chlordane (TC), cis-chlordane (CC), hexachlo-robenzene (HCB), α -endosulfan, β -endosulfan, heptachlor, aldrin, dieldrin, endrin and merix was purchased from AccuStandard Inc. (USA) and diluted to the desired concentrations. All solvents were of analytical grade and redistilled to remove impurities prior to use.

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 extracted in a Soxhlet extractor for 48 h with dichloromethane (DCM) at a rate of 4-6 cycles/h. A mixture of 2,4,5,6-tetrachloro-*m*-xylene (TcmX) 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 (3 cm, 3% deactivated), neutral silica gel (3 cm, 3% deactivated), 50% (on a weight basis) sulfuric acid silica (2 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 before use, respectively. The column was eluted with 50 mL of dichloromethane/hexane (1:1) to vield 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.

OCPs 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 μ m; 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 μ L sample was performed with a 5 min solvent delay time. Injector temperature was 250 °C.

2.3. Quality assurance/quality control (QA/QC)

The levels of OCPs were quantitatively determined by external reference standard method. The target compounds of the sample were identified quantitatively on the basis of their respective retention times and peak areas compared with mixture of standard solution. Before analysis, relevant standards were run to check quality parameters, such as extraction efficiency as indicated by recovery of the standard material, and limits of detection (LOD) were defined as three times the signal-to-noise ratio (S/N). The recoveries of standard material and LOD of target compounds ranged in 81–103% and 0.04–0.31 ng g^{-1} , respectively, for all the compounds studied. Method blanks (solvent) and duplicate samples were analyzed. The target compounds were not detected in the method blanks. The relative standard deviation of replicate analyses was <8%. The average surrogate recoveries were 68 ± 13% and 95 ± 9% for TCmX and PCB209, respectively. All results for sediment samples in this study were reported on a dry-weight basis.

2.4. Analysis of total organic carbon

Dried samples were treated with 4 M hydrochloric acid to remove carbonate and dried overnight at 60 °C. The carbonate-free samples were then analyzed for total organic carbon (TOC) with a Vario EL-III Elemental Analyzer and average values were reported. Replicate analysis of one sample (n = 6) gave a precision of ±0.02 wt.% for TOC.

3. Results and discussion

3.1. Occurrence levels of OCPs

The detection frequencies for α -HCH, β -HCH, γ -HCH, δ -HCH, p,p'-DDT, o,p'-DDT, p,p'-DDD, p,p'-DDE and HCB in our samples (n = 42) were up or close to 100%, followed by TC (88%) and CC (58%), suggesting a wide occurrence of these compounds in this region. However, α -endosulfan, β -endosulfan, heptachlor, aldrin, dieldrin, endrin and merix were only detected in few sediment samples, therefore, these OCP compounds were not discussed in this work.

Concentrations of measured OCPs in sediments ranged from 2.4 to 7.2 ng g⁻¹ (Table 1). In general, DDTs and HCHs are the two most widely used OCP species and thereby with the most readily detection in the sediments. The level of DDTs $(1.2-5.6 \text{ ng g}^{-1}, \text{ with a mean of } 2.9 \pm 1.0 \text{ ng g}^{-1})$ was higher than that of HCHs $(0.2-1.6 \text{ ng g}^{-1}, \text{ with a mean value of } 0.58 \pm 0.27 \text{ ng g}^{-1})$. Compared with other marine regions in China, the DDT and HCH levels in the coastal ECS were similar to those detected in Northern South China Sea (Chen et al., 2006), Bohai Sea in North China (Hu et al., 2009); but slightly higher than those detected in the Yellow Sea

Table 1 Concentrations (ng g^{-1}) of OCP species and TOC in surface sediments of the coastal ECS.

(Liu et al., 2008). As shown in Table 2, the DDTs' level in the coastal ECS was significantly lower than those detected in the coastal environments of Korea (Hong et al., 2006), Turkey (Bakan and Ariman, 2004), Vietnam (Hong et al., 2008) and France (Wafo et al., 2006). The HCH concentrations were lower than those in Turkey (Bakan and Ariman, 2004) and Singapore (Wurl and Obbard, 2005), but comparable to those found in the coastal environments of Korea (Hong et al., 2006), Vietnam (Hong et al., 2008), and Caspian Sea (DeMora et al., 2004). The concentrations of HCB were 0.13-0.59 ng g^{-1} with a mean value of 0.37 ± 0.10 ng g^{-1} which were lower than those in Bohai Bay, North China $(9.1-1300 \text{ ng g}^{-1})$ (Hu et al., 2005), and Pear River Estuary, South China Sea (1.1- 30 ng g^{-1}) (Fung et al., 2005). The concentration of sum of TC and CC (TC + CC) varied from 0.2 to 0.67 ng g^{-1} with a mean value of 0.34 ± 0.11 ng g⁻¹. These values were lower than those detected in the Daya Bay, South China Sea $(0.39-1.88 \text{ ng g}^{-1})$ (Wang et al., 2008), and Taihu Lake. East China (with a mean of 5.13 $ng g^{-1}$) (Zhao et al., 2010b).

3.2. Distribution of OCPs

Distribution of DDTs, TC + CC, HCHs and HCB are shown in Fig. 2. For DDTs, the high concentrations were observed in the nearshore area with a significant decrease gradient extending seaward, suggesting the impact of the proximity to the non-point

Sites	HCHs ng g^{-1}	DDTs ng g^{-1}	$\rm HCB~ng~g^{-1}$	TC + CC ng g^{-1}	TOC (%)	DDE + DDD/DDTs	DDE/DDD	β-HCH/HCH
1	0.56	2.06	0.40	0.25	0.75	0.83	1.61	0.45
2	0.62	2.42	0.45	0.20	0.58	0.76	0.59	0.46
3	0.46	2.48	0.50	0.34	0.69	0.91	1.57	0.33
4	0.52	3.70	0.47	0.41	0.68	0.83	1.81	0.39
5	0.49	4.25	0.36	0.40	0.54	0.79	1.99	0.44
6	0.57	2.47	0.38	0.37	0.75	0.81	2.04	0.45
7	0.75	2.25	0.55	0.27	0.69	0.71	1.50	0.46
8	0.62	2.77	0.35	0.22	0.67	0.70	1.57	0.36
9	0.45	2.17	0.36	0.22	0.54	0.74	1.36	0.46
10	0.60	3.29	0.30	0.27	0.59	0.66	1.15	0.64
11	1.64	4.77	0.33	0.50	0.59	0.81	1.52	0.52
12	0.66	4.31	0.45	0.46	0.62	0.73	1.61	0.37
13	0.61	3.10	0.38	0.30	0.53	0.70	1.49	0.49
14	0.75	3.57	0.33	0.28	0.63	0.67	1.40	0.62
15	0.49	3.49	0.44	0.27	0.62	0.66	1.29	0.41
16	0.45	1.79	0.43	0.21	0.66	0.67	1.95	0.42
17	0.43	1.74	0.40	0.25	0.66	0.93	1.79	0.41
18	0.44	1.29	0.39	0.27	0.71	0.74	0.90	0.31
19	0.52	2.02	0.45	0.29	0.74	0.77	1.38	0.35
20	0.50	2.38	0.43	0.26	0.73	0.79	1.74	0.39
21	0.88	4.76	0.59	0.51	0.64	0.73	1.64	0.33
22	0.51	5.67	0.33	0.45	0.64	0.67	1.45	0.38
23	0.81	3.00	0.38	0.34	0.71	0.79	1.67	0.54
24	0.53	2.96	0.42	0.36	0.63	0.76	1.76	0.40
25	0.45	2.90	0.42	0.32	0.62	0.72	1.19	0.47
26	0.57	3.41	0.31	0.21	0.78	0.74	1.50	0.49
27	0.53	4.96	0.35	0.48	0.71	0.49	1.96	0.39
28	0.22	2.48	0.14	0.36	0.71	0.87	1.84	0.17
29	0.50	2.44	0.50	0.38	0.71	0.78	1.87	0.31
30	0.38	1.64	0.38	0.33	0.71	0.80	2.00	0.37
31	0.27	1.77	0.43	0.36	0.72	0.77	1.82	0.35
32	0.41	1.98	0.36	0.34	0.57	0.81	2.09	0.35
33	0.48	4.81	0.33	0.67	0.72	0.67	1.19	0.38
34	1.47	4.51	0.32	0.41	0.68	0.76	1.76	0.57
35	1.03	3.31	0.36	0.40	0.76	0.78	1.87	0.47
36	0.52	3.09	0.57	0.64	0.49	0.79	1.60	0.30
37	0.20	3.10	0.18	0.18	0.83	0.72	1.56	0.52
38	0.65	3.34	0.14	0.21	0.51	0.55	0.85	0.42
39	0.83	3.30	0.28	0.29	0.68	0.67	1.36	0.52
40	0.47	2.27	0.39	0.34	0.81	0.70	1.95	0.28
41	0.44	2.68	0.42	0.32	0.66	0.67	1.45	0.43
42	0.35	1.85	0.29	0.25	0.65	0.76	1.61	0.50

Table	2

Comparison of HCHs and DDTs concentrations in surface sediments of the coastal ECS and other coastal areas in the world (ng g⁻¹, dry wt.).

Locations	Year	HCHs	DDTs	TOC/%	References
Coastal ECS	2007	0.1-1.6	1.2-5.6	0.49-0.83	This study
Northern South China Sea	2002	0.04-2.48	0.08-1.38	0.06-1.02	Chen et al. (2006)
Bohai Sea	2006	0.2-3.6	0.24-5.7	0.04-0.69	Hu et al. (2009)
Coastal area of Korea	1997-2002	N.D5.46	0.006-135 ^b	-	Hong et al. (2006)
Black Sea region of Turkey	1998	N.D37 ^a	N.D71 ^b	2.43-12.8	Bakan and Ariman (2004)
Coastal marine sediments, Vietnam	2008	N.D1.00	0.31-274	0.34-6.38	Hong et al. (2008)
Marseille, France	2002	-	2.02-254.8 ^b	0.15-5.49	Wafo et al. (2006)
Singapore coastal sea	2003	3.3-46.2	2.2-11.9 ^b	-	Wurl and Obbard (2005)
Caspian Sea	2000-2001	0.01-3.5	0.01-13.4 ^c	0.08-2.8	DeMora et al. (2004)

Year = Sample collected time.

- = Date is not available.

N.D. = Not detectable.

^a HCHs = α -HCH + β -HCH + γ -HCH.

^b DDTs = p,p'-DDD + p,p'-DDE + p,p'-DDT.

^c DDTs = p,p'-DDD + o,p'-DDD + p,p'-DDE + o,p'-DDE + p,p'-DDT + o,p'-DDT.

sources of the coast. TC + CC had the same distribution as DDTs, implying that they have the similar sources.

Unlike the DDTs and TC + CC, the HCHs' distribution did not exhibit a decreased trend from the nearshore towards the offshore, and the high concentrations of HCHs were mainly focused at the Min River Estuary in Fujian province and Ou River Estuary in Zhejiang province, suggesting direct HCH inputs from the two major river runoffs in the region. The high concentrations of HCB were detected in the offshore area between the 20-m and 50-m isobaths which was the center of the mud area in the coastal ECS. The main source of sediments in the inner shelf mud is from the Yangtze River (the fourth largest river in world in term of sediment discharge) into the sea, and about 30% Yangtze River-derived sediments are deposited there (Liu et al., 2007). HCB is used principally as an intermediate to produce pentachlorophenol (PCP) and Na-PCP in closed systems (Hu et al., 2004). It is likely that HCB is released into the environment by wastewater from the plants. Several studies revealed that HCB production in some chemical plants induced elevated HCB levels in the surrounding river environment in China, such as the Nanpaiwu River (with maximum of 141,400 ng g⁻¹)

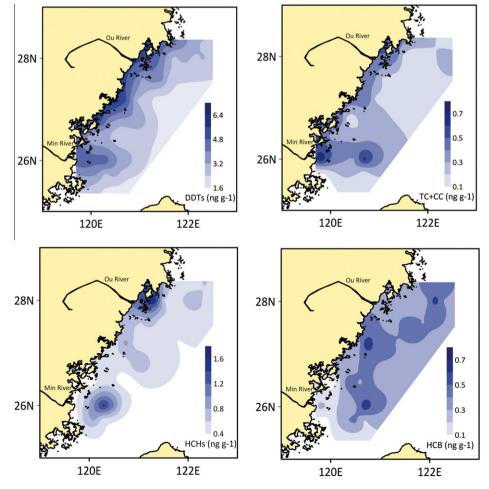


Fig. 2. Distribution of DDTs, TC + CC, HCHs, and HCB in sediments of the coastal ECS.

(Hu et al., 2005), Haihe River (n.d.–835 ng g^{-1} with a mean of 173 ng g^{-1}) (Zhao et al., 2010a). The Yangtze River Delta is one of two important HCB production bases in China in history (Hu et al., 2004); therefore, HCB in sediments of the offshore mud area could be more probably attributed to the Yangtze River outflows.

In the aquatic environment, the total organic carbon (TOC) in sediments was an important factor for controlling the distribution of OCP compounds. Generally, high total concentrations of OCPs are accompanied by the high TOC since OCP compounds are usually removed from the water column and adsorbed on the particulate matters due to their high affinity for organic matter (Mai et al., 2002). However, this correlation was not achieved in this work. This inconsistence was also reported in other coastal areas (Mai et al., 2002; Tan et al., 2009). It was noted that there was a relatively narrow range of TOC content (0.49-0.83%), while coupled with a wide scope of the OCP specie levels (Fig. 3). It was exhibited that there was a unique distributional pattern for the various OCPs species within the regime of the coastal ECS, indicating that the OCP residues feature in the coastal ECS may be more prominently controlled by the direct continuous regional inputs rather than the natural sorption process on particulate organic carbon.

3.3. Sources of OCPs

DDT can be biodegraded under aerobic condition to DDE and under anaerobic condition to DDD, thus the relative proportion of the parent DDT compound and its metabolites DDD and DDE can be used for assessing possible pollution sources (Hitch and Day, 1992). In this study, the ratios of (DDE + DDD)/DDTs were in the range of 0.49–0.93 (Table 1), indicating that these metabolites occurred significantly in the coastal ECS. DDE/DDD >1 was observed in most sites, suggesting the aerobic condition of the degradation processes, and the DDT residues in agricultural soils could be thus regarded as the important source (Table 1). However, as shown in Fig. 4, higher DDT concentration with elevated DDT/ (DDE + DDD) values were found in the nearshore area and they decreased seaward together with the distance from the coast, indicating that there was still a fresh DDT input along the coast. The fresh sources of DDT include technical DDT and dicofol. The technical DDT contains less o,p'-DDT (about 15%) than p,p'-DDT (about 85%), while dicofol has a reverse pattern (Qiu et al., 2005). The o,p'-DDT/p,p'-DDT ratios were 1.0-2.2 in sediment samples, indicating the mixed DDT-typed inputs. Dicofol is used as insecticide on many crops, fruit trees and tea trees after the ban of technical DDT in 1980s in China (Qiu et al., 2005). Technical DDT was still used as an additive for the production of antifouling paints used in fishing ships (Lin et al., 2009). Technical chlordane is composed of mixture with abundant components TC and CC, which is the current main source of TC and CC in the environment (Hu et al., 2004). In general, the ratio of CC/TC ranged between 1.2 and 1.3 in the technical chlordane products (Li et al., 2006), while this ratio would gradually decrease in environment because the degradation of CC might be faster than TC (Hung et al., 2005). As shown in Fig. 4, the ratios of CC/TC were found close to 1.2 in the nearshore area, and decreased with the distance from the coast, indicating the fresh land-based sources along the coast. Technical chlordane was actually still used as a termiticide which was widely used in Southeast China (Hu et al., 2004).

Commercial HCHs were produced and used mainly in two forms, technical HCH containing about 55–70% α -HCH, 5–14% β -HCH, 10–18% γ -HCH, 6–10% δ -HCH and minor proportions of other isomers, and a purified γ -isomer of lindane (γ -HCH > 99%)

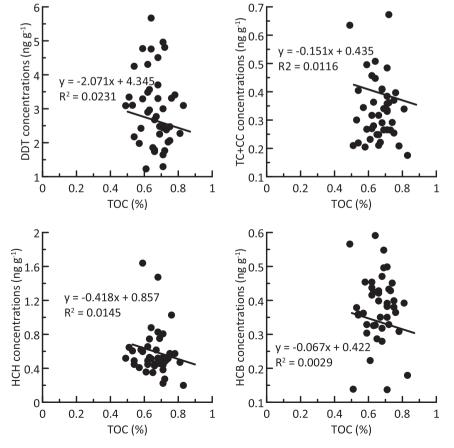


Fig. 3. Correlations between concentrations of DDTs, TC + CC, HCHs and HCB and TOC in sediment samples of the coastal ECS.

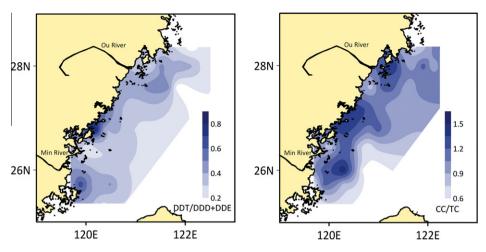


Fig. 4. Distribution of ratios of DDT/DDD + DDE and CC/TC in sediments of the coastal ECS.

(Li et al., 1998). However, compared with its original components, β -HCH was the most abundant one among the four isomers of HCHs (with a mean percentage of 41%) in this study (Table 1), suggesting that there was no evidence of recent technical HCH input (Li et al., 1998). The high concentrations of HCHs with elevated high β -HCH fraction were observed in the Min River Estuary and Ou River Estuary. This was similar with those detected in the bank soil and estuarine sediment samples from the Min River (Zhang et al., 2011), suggesting that the HCHs with high concentrations in the estuaries were mainly derived from the weathered HCHs preserved in the agricultural soils via local major river runoffs.

3.4. Potential ecotoxicological risks of OCPs

Potential environmental risk of OCPs residues was evaluated by two widely used sediment quality guidelines (SQG), i.e. the effects range-low value (ERL) and effects range-median value (ERM) guidelines (Long et al., 1995), specified by the USEPA (1997), as well as the threshold effects level (TEL) and probable effects level (PEL) guidelines from Canadian Council of Ministers of the Environment (CCME, 2002). Both of them were applied to evaluate the possible ecotoxicological risks of OCPs in the study. Taking into account the low level of OCP concentrations in this study, ERL and TEL were only used to assess the potential ecotoxicological impacts of these measured OCPs. As shown in Table 3, the concentrations of p,p'-DDD, o,p'-DDT and γ -HCH were all below the ERL value and TEL value, suggesting that the pernicious effects would be rarely observed for those compounds. The ERL for p,p'-DDE, *p*,*p*'-DDT and chlordanes were exceeded at 9% (n = 4), 16% (n = 7) and 7% (n = 3) of total sites in the study area (mostly observed in the nearshore areas and Min River Estuary as noted above), respectively. For DDTs, >90% (n = 41) of these investigated samples were higher than ERL value, indicating an intermediate ranking of

Table 3

Assessments of potential biological risks of selected OCPs in surface sediments of the coastal ECS using two sediment quality guidelines.

	ER-L	Above ER-L	TEL	Above TEL
p,p'-DDE	2.2	4	2.07	6
p,p'-DDD	2	0	1.22	5
o,p'-DDT	1	0	1.19	0
p,p'-DDT	1	7	1.19	4
DDTs	1.58	41	3.89	8
r-HCH	-	-	0.32	4
Chlordane	0.5	3	2.26	0

sediment toxicity within the neighboring benthic community by the exposure of these OCP species. On the whole, DDTs and chlordanes were the species of OCPs with more ecotoxicological concern in the coastal ECS, especially in the nearshore area and Min River Estuary, largely due to the recent use and extensive historical application in their adjacent land.

4. Conclusions

There was a wide occurrence of HCHs, DDT, HCB and chlordanes in the southern coastal ECS. The inputs of DDT and chlordane could be attributed to the recent use of relevant products and the extensive historical application in adjacent area, while HCHs are dominantly from historical application which preserved in agricultural soils released via major river runoffs into the sea. The contribution of HCB is likely from the Yangtze River outflows. The regime of coastal ECS is an important sink for these OCP species over a long-time and regional scale. DDTs and chlordanes are two more concerned OCP species for the ecotoxicological risk in the southern coastal ECS. This work found that the combination of distribution and composition of individual OCP could be useful to trace the sources and environmental fate of OCPs.

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