

Occurrence and distribution of organochlorine pesticides (DDT and HCH) in sediments from the middle and lower reaches of the Yellow River, China

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Abstract Sediments used in this study were selected from 23 stations in the middle and lower reaches of the Yellow River and its tributaries in November 2005. The levels and distribution patterns of selected organochlorine pesticides (OCPs = *p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, α -, β -, γ -, and δ -HCH) in samples were investigated by analysis of gas chromatography coupled with micro-electronic capture detector. Concentration of OCPs in the sediments from the Yellow River ranged from 0.05 to 5.03 ng g⁻¹ (mean, 1.02 ng g⁻¹) for \sum DDT, 0.09–12.89 ng g⁻¹

(mean, 1.08 ng g⁻¹) for \sum HCH. The concentration distribution of \sum DDT and \sum HCH varied significantly with different sampling station, indicating their different contamination sources. Composition analyses demonstrated that residues of DDTs in sediments came from the previous inputs of organochlorine pesticides, while β -HCH and γ -HCH significantly dominated in the sediments for HCHs.

Keywords Organochlorine pesticides · DDT · HCH · Sediment · The Yellow River

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Introduction

Persistent organic pollutants especially organochlorine pesticides have become widespread pollutants in the environment and now represent a global contamination problem because of the large production and usage, ubiquity, their higher bioaccumulation potential, persistence in the environment, and high toxicity to non-target organisms (Loganathan and Kannan 1994; Jones and de Voogt 1999). Several studies have reported that the contamination of OCPs in the sediments (Pandit et al. 2002; Zhang et al. 2003; Yang et al. 2005a, b) from Asian coastal areas and several rivers indicates the presence of their major emission sources in these regions. China is the second largest country producing and using

organochlorine pesticides in the world. Technical 1,2,3,4,5,6-hexachlorocyclohexanes (HCHs) and 2,2-bis(*p*-chlorophenyl)-1,1,1-trichloroethane (DDT) have been widely used from 1950 to 1983 in China. The production of HCHs and DDT in this country were about 4.9 million and 0.4 million tons, accounted for 33% and 20% of the total world production, respectively (Hua and Shan 1996). These compounds were used on a large scale in agricultural practices. Subsequent runoff of the compounds in the waterways, atmospheric deposition, and other means have resulted in their accumulation in freshwater, estuarine, and marine environment in China (Hong et al. 1995; Wu et al. 1999; Yuan et al. 2001). Although most of the developed countries have banned or restricted the production and usage of the DDTs and HCHs during 1970s and 1980s, for China in 1983, some OCPs such as DDT, HCH, and lindane are still used in some countries around the tropical and subtropical belts for agricultural and medicinal purposes (de Brito et al. 2002). In China, the original sources of DDT, the use of Dicofol, which contains DDT as impurities, is still allowed.

The Yellow River, located in the northern China, is the second longest river with a length of about 5,464 km, and it flows through nine of 32 provinces in the north of the country and drains a catchment area of about 795,000 km² (including isolated inflow area 42,000 km²). The rapid urbanization and industrialization have taken place in the last few decades along the river, especially in the middle and lower reaches of the river. Meanwhile, the drainage area of the middle and lower reaches have become the main area in agriculture production, and about several important cities obtained their drinking water and industrial water from the river.

The middle and lower reaches of the Yellow River has received an increasingly large volume of organic wastewater and sewage and had been polluted by organic chemicals since the end of twentieth century. Although some investigations of pollution assessment have been carried out in the area, most of those have focused on a limited number of sites and detection for polycyclic aromatic hydrocarbons, nitrobenzenes, and others

species (Gao et al. 2001; Liu et al. 2004). Given that there was almost no previous data on OCPs levels in the Yellow River and its tributaries and a priori evaluation of possible pollution, it is of great significance to the water quality and aquatic ecological environment to research on the residue levels of OCPs in the river.

This paper presents the study of organochlorine pesticides (DDTs and HCHs) in sediments from the middle and lower reaches of the Yellow River. The purpose of the research was to assess the contamination levels of OCPs (DDTs and HCHs) in this area to analyze their distribution and composition for distinguishing the previous and current inputs of the formerly used organochlorine pesticides and deducing the possible sources of OCPs, and the monitoring data in these selected sediments is indispensable for the risk assessment of OCPs in these sites.

Materials and methods

Study area and sampling

In November 2005, samples were collected from 23 stations in mid- and downstream sections of the Yellow River (From Fenglingdu Bridge to confluence with Jindi river). This area is located between 34°25' N to 36°07' N and 110°17' E to 110°06' E with a length of 177 km. Global positioning system was used to locate the sampling stations. Among these stations, 12 are from the mainstream and eight are from the tributaries (Yiluo River, Si River, Xinmang River, Qin River, and Jindi River). The wastewater discharge locations of Luoyang Petrochemical Company (station 4) and Shouyangshan Power Plant (station 9) are also included. Locations of sampling stations are shown in Fig. 1.

About 2 kg of samples were collected from the surficial sediments (top, 0–5cm) with a stainless steel grab and then placed into polytetrafluoroethylene bags. Two or more distributed samples were collected in each sampling points according to its width. All the samples were immediately transferred to the laboratory and keep at –20°C in

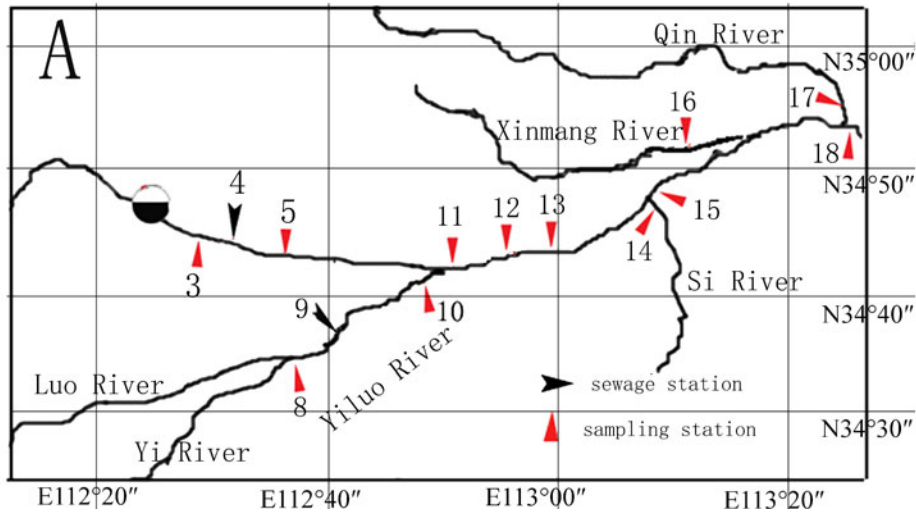
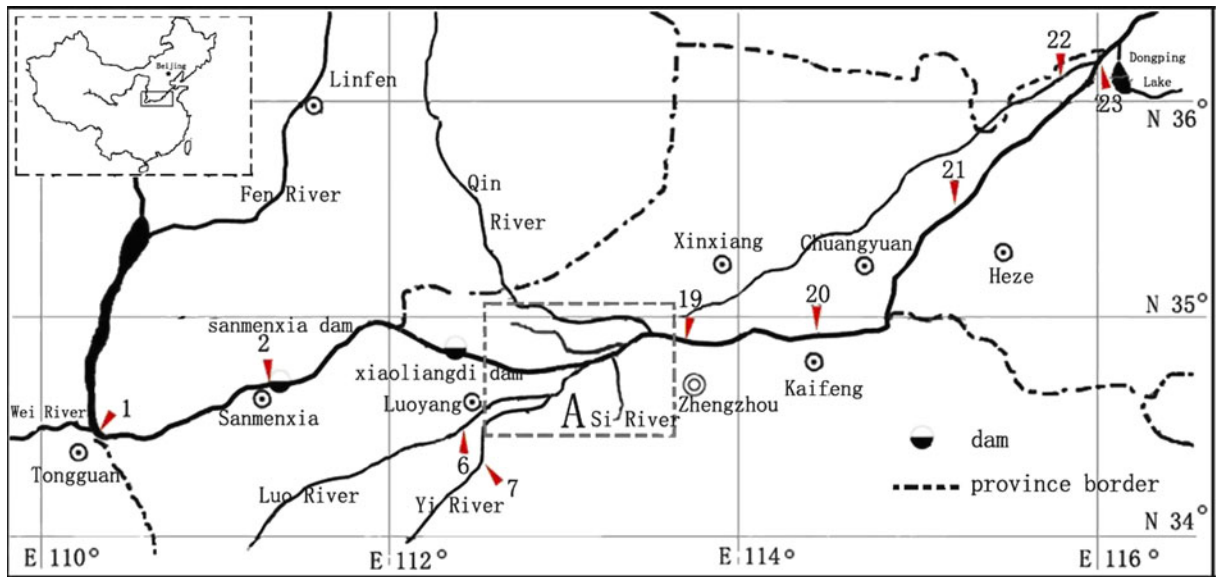


Fig. 1 Map of sampling stations of the middle and lower reaches of the Yellow River (Source: Distribution of polycyclic aromatic hydrocarbons (PAHs) in Henan Reach

of the Yellow River, Middle China; *Ecotoxicology and Environmental Safety*, Volume 72, Issue 5, July 2009, Pages 1614–1624)

the refrigerator. Prior to analysis, sediment samples were freeze-dried and homogenized.

Reagents and standards

All glassware and apparatus were baked at 450°C or rinsed with redistilled hexane and dichloro-

methane (DCM) prior to use. All solvents including DCM and hexane were redistilled in a full-glass distilling appliance before use. Neutral silica gel and alumina (80–100 mesh) were treated with DCM by Soxhlet extraction for 72 h, dried at 60°C, activated at 180°C and 250°C for 12 h, respectively, and then deactivated with 3% (w/w)

of deionized water. Anhydrous sodium sulfate (Na_2SO_4) was baked at 450°C for 4 h. The standard organochlorine pesticide solution, including *p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDE, and *p,p'*-DDD and α -, β -, γ -, and δ -HCH, was purchased from the Accustandard Co. and diluted to the desired concentration. The recovery surrogate and internal standard for determining the OCPs were decachlorobiphenyl (PCB209) and pentachloronitrobenzene (PCNB).

Extraction procedure

Approximately 35 g of freeze-dried sample was extracted for 48 h with DCM in a Soxhlet apparatus. Before extracted, each sediment sample was spiked with 20 ng of decachlorobiphenyl (PCB209) as surrogate. Activated copper granules were added to the collection flask to remove elemental sulfur. The extract was subsequently concentrated on a rotary evaporator at below 30°C and solvent-exchanged to hexane and further reduced to approximately 0.5 ml under a gentle nitrogen stream. This was further purified with a glass column (25 cm length \times 9 mm i.d.) loaded with 3 g of alumina, 3 g of silica gel, and 4 g of H_2SO_4 /silica from bottom to top in turn. On the top of the glass column, 1 cm of anhydrous Na_2SO_4 was added on the top to removed water. The OCPs was eluted with 35 ml of hexane/DCM (1:1 v/v). The effluents were reduced finally to a volume of 0.5 ml under a gentle stream of pure nitrogen, and a known amount of internal standard, PCNB, was added prior to gas chromatography (GC) analysis.

Analytical procedure

Total organic carbon (TOC) was determined by a Vario EL III elemental analyzer (Heraeus, Germany) on freeze-dried powdered samples. The carbonate fraction was eliminated through an HCl treatment in silver capsules. The grain size was measured using Fritsch A22 and Malvern Mastersizer laser grain-size analyzers. About 2–5 g of disaggregated sample are placed in a beaker. Ten milliliters of dispersant [$(\text{NaPO}_3)_6$] (density,

0.05 N) is added to the solution. The sample solution is ultrasonicated for 10 min. The sample solution is transferred to the sample container of the laser particle size analyzer for measurement. For the samples including larger than 2 mm grain size sediments, the sample should be sieved using water, dried, and weighed. The results finally were combined together. The identification and quantification of OCPs in the extracts were accomplished by A HP6890 plus GC with a micro-cell election capture detector. A chrompack capillary column (50 cm length \times 0.25 mm i.d. \times 0.25 μm film thickness) was used for separating the OCPs. One microliter of each sample was injected in the GC system with a splitless model. Oven temperature increased from 60°C (held for 2 min) to 200°C at the rate of $6^\circ\text{C}/\text{min}$, followed to 210°C at the rate of $1^\circ\text{C}/\text{min}$, and then programmed to 290°C (held for 30 min) at $10^\circ\text{C}/\text{min}$. The temperatures of injector and detector were 250°C and 290°C , respectively. Data acquisition and processing was controlled by HP Chemstation software. High pure nitrogen was used as both the carrier gas and make-up gas with a flow rate of 1.0 and 33.3 ml/min, respectively.

Prior to sample analysis, relevant standards were analyzed to check column performance, peak height, and resolution. For every set of samples to be analysed, a solvent blank, a standard mixture, and a procedural blank were run in sequence to check for contamination, peak identification, and quantification. Peak identification was conducted by the accurate retention time of each standard ($\pm 1\%$). All the results for OCPs in the sediment samples were reported on a dry-weight basis.

Quality control and quality assurance

All analytical procedures were monitored using strict quality assurance and control measures. The method blanks showed no detected target compounds. Three field duplicates showed quite uniform OCPs concentrations with a standard deviation of 5–8%. For every set of nine samples, a spiked sample and a procedural blank containing all chemicals were run to check for interference

and cross-contamination. The residue levels of OCPs were quantitatively determined by the internal standard method using peak area. The correlation coefficients (*r*) of calibration curves of HCHs and DDTs were all higher than 0.998. The limits of detection (LOD) for OCPs were described as three times signal versus noise value (S/N). The recoveries of surrogate standard and LOD of target compounds ranged from 76% to 105%, 0.08 to 1.3 ng, respectively, for all the compounds studied. Reported values are recovery corrected.

Results and discussion

Sediment particle size and content of TOC distribution

The distribution of particle size and content of TOC in the sediments from all sampling stations are given in Table 1. For all sampling samples, there were significant differences in the particle size and TOC distribution. The sediments from the Yellow River were dominated by high sand and slit content. The total ratio of sand and slit approximately accounts to 90%. The sediments have low clay content with average ratio of 10%. The sand carried by the Yellow River is more than any other river in the world.

For TOC content, the results show that there is a lower level TOC content in sediments with a range of 0.8–20.24% and mean of 5.95%. For the sediments from stations 2, 9, 14, 20, and 22, higher TOC content and much more clay content were determined because these sampling stations are located near cities, villages, and industrial area and had been influenced by human activities.

Contamination levels of OCPs

The contamination levels of DDTs (*p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDE, and *p,p'*-DDD) and HCHs (α -, β -, γ -, and δ -HCH) in the sediments from the middle and lower reaches of the Yellow River

Table 1 Particle size distribution of the sediments from the middle and lower reaches of the Yellow River (%)

Sampling station	TOC (mg g ⁻¹)	Sand 2–0.063 mm	Slit 63–4 μm	Clay <4 μm
1	1.14	35.733	58.775	5.492
2	20.00	23.713	60.511	15.776
3	1.19	80.339	15.747	3.914
4	14.13	47.132	38.043	14.825
5	1.14	81.813	15.136	3.052
6	5.89	58.014	34.738	7.249
7	5.02	66.015	26.165	7.820
8	5.18	56.254	32.202	11.544
9	8.29	9.017	70.069	20.914
10	2.18	22.909	60.068	17.023
11	0.80	88.019	9.909	2.072
12	1.02	84.221	13.325	2.454
13	0.89	95.736	3.293	0.971
14	11.35	5.208	72.235	22.557
15	1.78	52.007	35.823	12.171
16	16.76	49.279	39.706	11.015
17	2.21	46.576	43.349	10.075
18	8.74	26.973	64.342	8.685
19	1.00	75.330	19.902	4.768
20	20.24	42.039	40.373	17.588
21	1.86	22.303	71.844	5.853
22	5.14	4.803	65.833	29.364
23	0.87	65.802	30.061	4.137
Average	5.95	49.53	40.06	10.41

were shown in Table 2. The concentration of \sum DDT ranged from 0.05 ng g⁻¹ at station 12 to 5.03 ng g⁻¹ at station 22. An obvious higher trend of concentrations at sample stations from the tributaries was observed. The levels of OCP contamination in sediments in this study were compared with those in other riverine, estuary, and coastal regions, such as the Yangtze River, the East China Sea in China, Kaveri River in India, etc (Table 3). About 20 years ago, DDTs in sediments were not detected; but now, it raised in the range of 0.05–5.03 ng g⁻¹. Among the Chinese large rivers, the levels of \sum DDT in the sediments from the Yellow River ranked a little higher, with which the residue levels in the sediments from East China Sea was commensurate (Hong et al. 1999; Wu et al. 1999; Liu et al. 2003; Zhang et al. 2003; Yang et al. 2005a, b). When compared with other regions over the world, the concentrations of \sum DDT were higher than those from the Indian

Table 2 Concentration of selected OCPs and TOC contents in the sediments from the middle and lower reaches of the Yellow River (ng g^{-1} d.w.)

Sampling station	α -HCH	β -HCH	γ -HCH	δ -HCH	Σ HCH	<i>p-p'</i> -DDT	<i>o-p'</i> -DDT	<i>p-p'</i> -DDE	<i>p-p'</i> -DDD	Σ DDT
1	n.d.	0.03	0.06	n.d.	0.09	0.02	0.02	0.04	0.05	0.13
2	0.11	0.42	0.28	0.09	0.90	0.21	0.43	0.78	0.42	1.84
3	0.02	0.08	0.10	0.01	0.21	0.01	0.02	0.04	0.04	0.11
4	n.d.	n.d.	n.d.	n.d.	0.00	0.25	0.39	0.96	0.61	2.21
5	0.01	0.04	0.13	0.02	0.19	0.01	0.01	0.02	0.02	0.06
6	0.03	0.13	0.12	0.04	0.33	0.07	0.07	0.28	0.15	0.56
7	0.03	0.14	0.11	0.03	0.31	0.58	0.04	0.51	0.28	1.40
8	0.05	0.17	0.13	0.03	0.38	0.21	0.14	0.41	0.18	0.94
9	0.37	0.79	0.22	0.17	1.55	0.06	0.05	1.30	0.72	2.13
10	0.12	0.59	0.29	0.06	1.07	0.31	0.08	0.59	0.35	1.33
11	0.02	0.06	0.12	0.02	0.22	0.01	0.02	0.03	0.02	0.08
12	0.02	0.05	0.08	0.01	0.15	0.01	0.01	0.02	0.02	0.05
13	0.02	0.06	0.12	0.02	0.23	0.01	0.01	0.02	0.02	0.06
14	0.18	0.78	0.37	0.08	1.41	0.39	0.30	0.72	0.33	1.74
15	0.03	0.16	0.09	0.02	0.28	0.14	0.25	0.20	0.15	0.74
16	1.52	8.94	2.43	n.d.	12.89	0.45	0.68	1.25	1.11	3.48
17	0.03	0.10	0.14	0.02	0.30	0.07	0.05	0.11	0.08	0.30
18	0.02	0.14	0.08	0.02	0.26	0.05	0.04	0.10	0.07	0.26
19	0.02	0.09	0.12	0.03	0.25	0.02	0.03	0.05	0.03	0.13
20	0.07	0.41	0.39	n.d.	0.87	0.03	0.03	0.13	0.07	0.26
21	0.03	0.10	0.11	0.02	0.26	0.10	0.13	0.18	0.12	0.53
22	0.24	1.10	0.89	0.26	2.48	0.57	0.12	1.88	2.45	5.03
23	0.02	0.06	0.13	0.01	0.21	0.01	0.01	0.07	0.05	0.15

Kaveri River (Rajendran and Surbramanian 1999) and much less than those from Taiwanese Keelung River, Australian Parramata River, and Matanido River in Solomon Islands (Iwata et al. 1994).

Concentrations of Σ HCH in sediments from the Yellow River ranged from 0.09 ng g^{-1} at station 1 to 12.89 ng g^{-1} at station 16. Our data showed that the HCH residues in sediments dramatically increased almost more twice times than

Table 3 Comparison of Σ DDT and Σ HCH (ng g^{-1}) in surface sediment from other Chinese rivers and world rivers, estuary, and coastal areas

Locations	Year	Σ DDT	Σ HCH	References
The Yellow River	2005	0.05–5.03	0.09–12.89	This study
The Yellow River	1984	n.d.	1.00–5.00	Wu et al. (1999)
Yangtze Estuary	2001	n.d.–0.6		Liu et al. (2003)
Pear river	1996–1997	1.36–8.99	0.28–1.23	Hong et al. (1999)
Minjiang River	1997	1.6–13.1	3–16.2	Zhang et al. (2003)
Haihe River	2003	0.32–30.18	1.88–18.76	Yang et al. (2005a, b)
The East China Sea	2002	n.d.–6.04	n.d.–1.45	Yang et al. (2005a, b)
Kaveri river, India	1998	0.69–4.85	4.35–158.4	Rajendran and Surbramanian (1999)
Keelung river, Taiwan	1997–1998	0.53–11.4	0.99–14.5	Iwata et al. (1994)
Parramata river, Australia	1990	26	7.7	Iwata et al. (1994)
Matanido River, Solomon Islands	1990	750	140	Iwata et al. (1994)

those in 1984. The concentrations of \sum HCH in most sample stations were less than \sum DDT. For the concentration of \sum HCH, some regions in China, such as Minjiang River and Haihe River, showed relatively higher concentrations (Zhang et al. 2003; Yang et al. 2005a, b). However, compared to other rivers in the world, the levels of HCH contamination were much lower than those from the Indian Kaveri River (Rajendran and Surbramanian 1999) and Matanido River in Solomon Islands (Iwata et al. 1994) but higher than from the Taiwanese Keelung River and Australian Parramata River (Iwata et al. 1994).

The pollution of OCPs was much influenced by TOC in the sediments and soils. TOC in sediments is affected by autochthonous and lo-cochthonous organic input, as well as by post-depositional preservation of organic matter. The sediments from middle and lower reaches of the Yellow River, which carried the maximum sediment concentration over the world, were mainly composed of sand and slit. The data analysis showed that the concentrations of \sum DDT and \sum HCH had a similar and relatively well correlation with TOC contents. The correlation coefficients were, respectively, 0.45 and 0.43 ($p < 0.05$) between DDT and HCH with TOC contents, which is possibly due to the higher content of clay and silt. When it comes to clay content, DDTs in sediments was correlated well with clay ratio content ($r = 0.77$), while HCHs showed no obvious

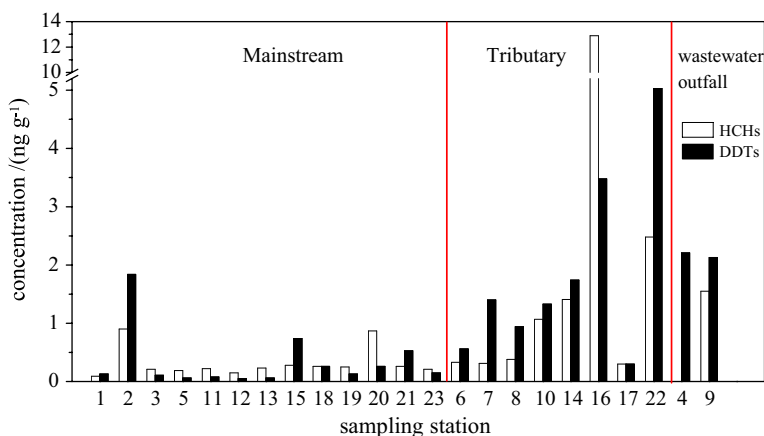
correlation ($r = 0.22$), which could indicate that DDTs affixed to clay easier than HCHs.

Spatial distribution of OCPs

The spatial distribution of DDTs and HCHs concentration in all samples sediments is shown in Fig. 2.

According to Fig. 2, we could find that the distribution of DDTs and HCHs in the sediments from the middle and lower reaches of the Yellow River varied much among all sampling stations. The highest concentrations of DDTs and HCHs were located at stations 22 and 16 from the tributaries, respectively. In general, the higher concentration levels of DDTs and HCHs were detected in the sediments of the tributaries compared with those in the sediments of the mainstream. Samples from the mainstream had a higher concentration of HCHs, while samples from the tributaries had a higher concentration of DDTs. The residue levels of OCPs from station 2 was higher than other mainstream stations; for the Sanmenxia reservoir, it received a great amount of pollutants including some industrial wastewater from chemical plant and agriculture pollutants during the surface runoff chronically. In the mainstream, the residues of OCPs in sediments were significantly lower with ranges of 0.09–0.87 ng g⁻¹ for HCHs and 0.05–1.84 ng g⁻¹ for DDTs, respectively. There were logical reasons. First, the sediments

Fig. 2 The spatial distribution of DDTs and HCHs in sediments from the middle and lower reaches of the Yellow River



from the Yellow River comprised much lower content of clay and TOC, which OCPs usually have strong affinity of. Clay and TOC in sediments play an important role to determine the levels of OCPs. Second, the sediments in the Yellow River were stirred easily due to the enormous sand content in the Yellow River, which carry the largest quantum of sand in the world. The sediments from river banks or at the bottom of the river several decades when OCPs had not been used during that time have been turn up again.

The chief tributaries along the river in the drainage area are Yiluo River, Si River, Qin River Xinmang River, and Jindi River. The concentrations of \sum DDT in tributaries followed the sequence: Jindi River > Xinmang River > Si River > Yiluo River > Qin River. The contamination levels of HCHs in tributaries followed a slight different sequence: Xinmang River > Jindi River > Si River > Yiluo River > Qin River. The spaciousness area of the cropland distributed along the drainages of Jindi River and Xiangmang River and a large amount of OCPs probably had been used in the 1970s and the beginning of the 1980s. The residues of OCPs have been accumulated in the sediments of the river. Therefore, the sample sediments from stations close to the tributaries of Yiluo River, Si River, and Xinmang River, such as stations 7, 8, 10, 14, 16, and 22, were

measured with higher concentrations. Except for the agro-contaminants, the Yiluo River mainly received wastewater from Hennan Kaipu Chemical Co. Ltd. of China, Shouyangshan Power Plant, and large numbers of rural industries; the samples from these two rivers have fewer residues of DDTs and HCHs. Stations 4 and 9 have a little higher contamination levels than most stations because the sample stations are closed to the industrial contaminative resources, such as the Luoyang Petrochemical Company and Shouyangshan Power Plant, and the high contaminative sediments near industrial resources enriched the OCPs deposit in the sediments.

Compositions analyses

Composition analysis of the HCH isomers and DDT congeners remaining in the environment can indicated the different pollution resources (Iwata et al. 1995). DDTs have a long persistence in the environment. Commercial-grade DDT generally is composed of 75% *p,p'*-DDT, 15% *o,p'*-DDT, 5% *p,p'*-DDE, <0.5% *p,p'*-DDD, <0.5% *o,p'*-DDD, <0.5% *o,p'*-DDE, and <0.5% unidentified compounds (WHO 1979). DDT can be degraded to different metabolites according to its ambience. DDT can be biodegraded to DDE under aerobic condition and to DDD under anaerobic condition

Fig. 3 Composition of DDTs in sediments from the middle and lower reaches of the Yellow River

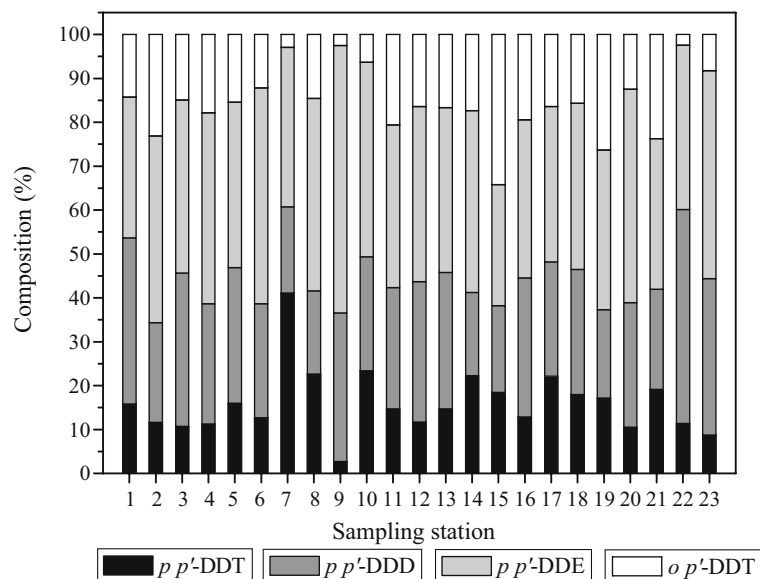
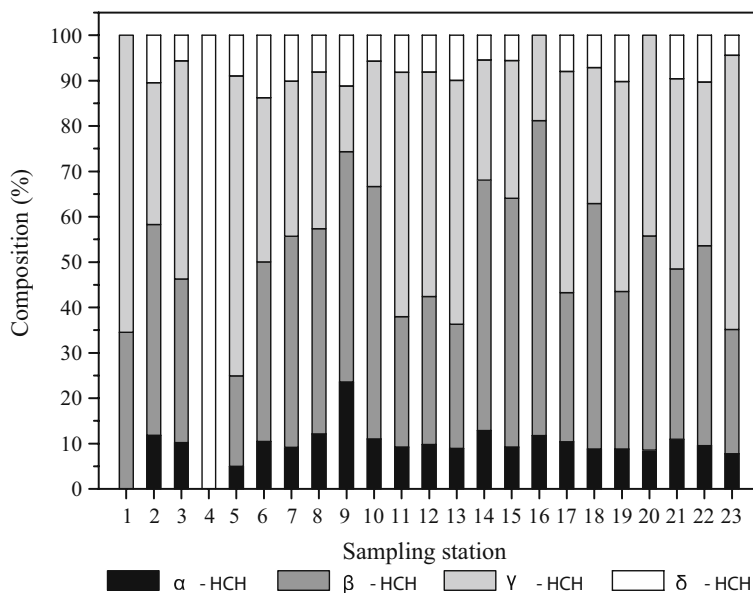


Fig. 4 Composition of HCHs in sediments from the middle and lower reaches of the Yellow River



(Doong et al. 2002). Comparing the concentrations of DDT and its metabolites, it can be inferred whether DDT's input are recent or not. The ratio of DDE and DDD to \sum DDT >0.5 can indicate long-term weathering (Hong et al. 1999). The composition of DDTs in this study is shown in Fig. 3; DDD and DDE occupied the predominant percentage and its average percentages were 28.25% and 40.9%, respectively. Here, the ratio of $(\text{DDD}+\text{DDE})/\sum\text{DDT}$ ranged from 0.47 to 0.95 with a mean ratio of 0.83, which suggested that the degradation of DDTs occurred significantly after the official ban of DDT and HCH in 1983.

Industrial HCHs and Lindane are the principal sources of HCH in China and had been used as a broad spectrum pesticide for agricultural purpose. The typical industrial HCHs generally contains 55–80% of α-HCH, 5–14% of β-HCH, 8–15% of γ-HCH, and 2–16% of δ-HCH, respectively (Lee et al. 2001), while Lindane consists of 99% γ-HCH. The physicochemical properties of these HCH isomers differed from each other. β-HCH has the lowest water solubility and vapor pressure, which is the most stable and relatively resistant to microbial degradation (Ramesh et al. 1991), and it also should have been reported that α-HCH can be converted to β-HCH in the proper conditions (Wu et al. 1997; Walker 1999). The higher percentage of α-HCH in some environmental sam-

ples indicated the recent use of industrial HCH. β-HCH dominating in sediments from the river and estuary environment after a long-term migration and transformation have been reported in many reports (Wu et al. 1999; Lee et al. 2001; Doong et al. 2002). In this study, a higher percentage of β-HCH and γ-HCH was recorded (Fig. 4). The average composition of HCH isomers measured in the sediment samples from the Yellow River are 9.57% for α-HCH, 40.03% for β-HCH, 39.05% for γ-HCH, and 7.00% for δ-HCH.

Conclusion

The residue levels of selected OCPs (DDT and HCH) in the sediments from the middle and lower reaches of the Yellow River and its tributaries were investigated in this study. The concentration and distribution of \sum DDT and \sum HCH varied significantly with different sampling stations, indicating their different contamination sources. Yiluo River, Xinmang River, and Jindi River, three of the main branches of the Yellow River in middle and lower reaches, were seriously contaminated by DDT and HCH. Compared with the concentrations reported from other rivers and estuaries in China and abroad, the content of DDT was lower than most rivers except Yangtze

Estuary and Indian Kaveri River, while the concentration of HCH was higher in Pear River, the East China Sea, Taiwanese Keelung River, and Australian Parramata River but much lower than Matanido River in Solomon Islands and Indian Kaveri River. \sum DDT and \sum HCH in the sediments had a similar and relatively well correlation with TOC contents, with correlation coefficients of 0.45 and 0.43 ($p < 0.05$), respectively. Composition analyses and the residue levels of DDT and HCH indicated that great changes have occurred due to transformation and degradation of the contaminants and an extensive use of DDT and HCH, especially Lindane, in the past in the drainage area of the middle and lower reaches of the Yellow River.

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