The residues, distribution, and partition of organochlorine pesticides in the water, suspended solids, and sediments from a large Chinese lake (Lake Chaohu) during the high water level period

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Abstract The levels of organochlorine pesticides (OCPs) in the water, suspended solids, and sediments from Lake Chaohu during the high water level period were measured by a solid-phase extraction gas chromatograph–electron capture detector. The spatial distributions of the three phases and the water/suspended solids and sediment/water partition coefficients were analyzed. The results showed the following: (1) The mean contents of OCPs in the water, suspended solids, and sediments were 132.4 ± 432.1 ng/L, 188.1 ± 286.7 ng/g dry weight (dw), and 13.7 ± 9.8 ng/g dw, respectively. The dominant OCP components were isodrin (85.1 %) for the water, DDTs (64.4 %) for the suspended solids, and both isodrin (48.5 %) and DDTs (31.8 %) for the sediments. (2) β-HCH was the primary isomer of HCHs in the water and sediments, and the proportions were 61.7 and 41.3 %; γ-HCH was the primary isomer in the suspended solids, accounting for 49.3 %; p,p'-DDT was the dominant content of DDTs in the water and suspended solids, whereas p,p'-DDD was the main metabolite of DDTs in the sediments. (3) The concentrations of contaminants in the water from the western lake were greater than those from the eastern lake, but the concentrations in the suspended solids from the western lake were less than those from the eastern lake. (4) There was no significant correlation between the water–suspended solids partition coefficient K_d and the n-octanol–water partition coefficient K_{ow}, and between the sediment–water organic-C weighted sorption coefficients K_{oc} and K_{ow}.

Keywords OCPs · Residue levels · Spatial distribution · Partition coefficient · Water · Suspended solids · Sediments · Lake Chaohu

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Introduction

Organochlorine pesticides (OCPs), which are typical persistent organic pollutants, were widely used in China. Due to their high toxicity, persistence, semi-volatility, and bioconcentration, OCPs are still detectable in various environmental media, such as water, sediment, soil, and air (Carrera et al. 2001; Fisk et al. 2001; De Joode et al. 2001), and pose threats to the ecosystem and human health through the food chain (Helaleh et al. 2005; Gustin 2005; Cheng et al. 2006). Large quantities of OCPs have been produced and used in China. Although the production and usage of some OCPs (such as DDTs) have been banned, the residues of OCPs are still detectable in various environmental media, and they even exhibit a trend of increasing concentration and

harmfulness in some regions (Yu et al. 2002; Cotham and Bidleman 1991; Turgut 2003; Zhao et al. 2010).

The principal ways that organochlorine pesticides enter water systems include the following: wastewater discharge from pesticide factories, pesticide droplets or dust falling into the water during the process of application, pesticide residues in environmental media entering into the water via surface runoff, and washing of pesticide containers and agricultural equipment (Liu 1998). Because of their hydrophobicity and lipophilicity, OCPs are insoluble in water; therefore, they are adsorbed on the surface of suspended solids and sediments. At the same time, several physical and chemical processes that include volatilization, migration, photolysis, hydrolysis, aquatic animal metabolism, absorption, and enrichment also occur (Zhou et al. 2006; Yang et al. 2010). The OCPs in sediments can reenter the water by desorption and resuspension, causing secondary pollution. They can also adversely affect humans and other organisms by enrichment and magnification through the food chain (Nelson and Hites 1980; Steinheimer et al. 1981; Gobas and Maclean 2003; Zhao et al. 2009). Therefore, to understand the pollution status and migration transformation characteristics of OCPs in the environment, it is important to study the residue levels, distribution and partition of OCPs in the water, suspended solids, and sediments.

Lake Chaohu, located in the center of Anhui Province in southeast China (Fig. 1), is the fifth largest freshwater lake in China, with a water area of approximately 760 km^2. Lake Chaohu is also an important source of drinking water and industrial and agricultural water for the 9.6 million residents in the surrounding areas; its water quality directly affects the health and social and economic development of the residents (Xu et al. 1999; Xu et al. 2001). Chaohu Basin is located in the north–south air flow intersection and northern subtropical humid monsoon climate zone, with an annual average temperature of 15–16 °C. Chaohu Basin is a primary area for cotton, tea, vegetable, and fruit plantations, but it is also suitable for the growth of termites, leading to problems with termite damage and schistosomiasis. To address the need for agricultural pest control, support human health, and prevent epidemics, OCPs (such as DDTs, hexachlorocyclohexanes (HCHs), chlordane, and endosulfan) were used extensively. Studies have shown that the residues and risks of OCPs in the water and sediment were relatively high in Lake Chaohu (Zhang et al. 2010; He et al. 2012); however, the residues, distribution, and partition of OCPs in the water, suspended solids, and sediments in Lake Chaohu still remain to be studied. The objectives of this study are to measure the concentrations of OCPs in the three phases, study the migration transformation characteristics, and provide a theoretical basis for a comprehensive

understanding of OCP pollution and prevention in the Chaohu water system.

Materials and methods

Sample collection

Samples were collected from Lake Chaohu in October 2009. The distribution of sampling sites, which were located in the water source protection areas and the rivers that feed the lake from Guishan to the east of Zhangjiahu, is shown in Fig. 1. The river sites B6, C1, C2, and C4 are located in the Shuangqiao estuary, Small Zhegao River, Big Zhegao River, and the drain of the thermal power station. The sites A6, B2, B3, B4, B5, and B7 were in the eastern water source area; A1, A2, A3, and A4 were in the eastern lake (excluding the water source area, similarly hereinafter), and D1 was in the western lake.

Twenty liters of water was collected from each sampling site. After shaking and mixing, a 1-L aliquot of each water sample was filtered through a glass fiber filter (ashed at 450 °C for 4 h), using a peristaltic pump (80EL005; Millipore Co., USA) and a filter plate with a diameter of 142 mm to separate the suspended solids. Pentachloronitrobenzene (PCNB, 100 ng) was added to the water as an HCH recovery indicator.

The suspended solids were collected by filtering the water samples. Before use, the glass fiber filters were ashed at 450 °C for 4 h and weighed after drying to constant weight for 24 h. After air-drying, the suspended solid samples were stored in aluminum foil in desiccators to maintain a constant weight. The weight difference of the filter before and after filtering was the quality of suspended solids.

The surface sediment samples from 14 sampling sites (excluding B7) were collected using a grab sampler and stored in sealed bags. The samples were frozen at −20 °C in a refrigerator until analysis. After the sediment samples were air-dried and benthonic organisms such as plant tissues and shells were removed, the samples were ground with a mortar and pestle into a granular powder and sieved through a 200 mesh sieve.

Sample extraction and cleanup

A solid-phase extraction (SPE) system was used to extract the filtered water samples. Before extraction, the octadecyl-silane SPE cartridges (SPE, C18, 6 ml, 500 mg, Supelco, Co., USA) were first washed with 10 ml dichloromethane and conditioned with 6 ml methanol and 6 ml ultrapure water, and the cartridges were not dried before loading the samples. After activation, the water samples were loaded using a large volume sampler (Supelco Co.) that was
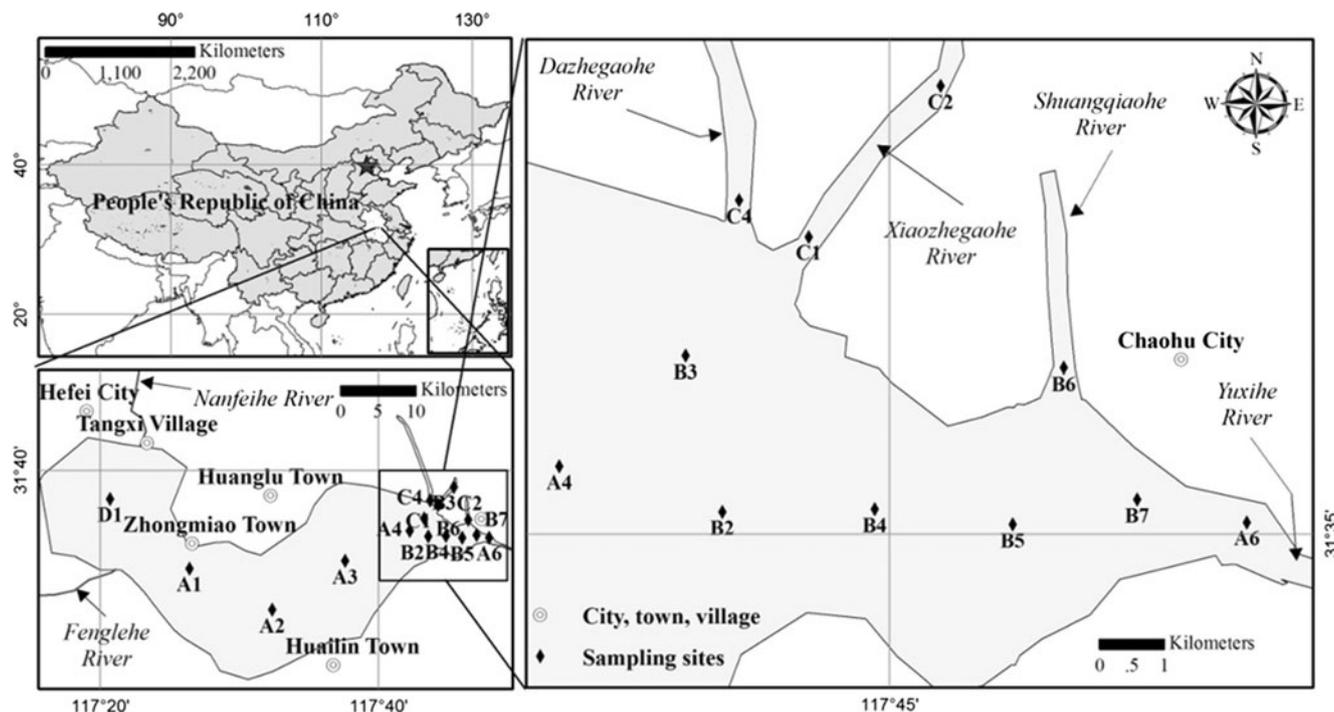
Fig. 1 The location of Lake Chaohu and the distribution of sampling sites

connected to the SPE vacuum manifold (Supelco Co.), and the cartridges were dried by vacuum pump after the extraction step. The SPE cartridges were sealed and delivered back to the laboratory prior to elution and purification. Each cartridge was connected to an anhydrous sodium sulfate (5 g) cartridge and eluted using dichloromethane (DCM, three times, 6 ml per elution). The extracts were concentrated to approximately 1 ml with a vacuum rotary evaporator (R-201; Shanghai Shen Sheng Technology Co., Ltd., Shanghai, China). The solvent was changed to hexane, and the samples were again concentrated to approximately 1 ml. 2, 4, 5, 6-Tetrachloro-*m*-xylene was added to the samples as an internal standard. The samples were transferred to vials and sealed until analysis.

The weighed filters were cut into pieces with stainless steel scissors and put into microwave extraction tubes. A 25-ml volume of mixed solvent (mixed by hexane and acetone, $V/V=1:1$) and 50 ng 1-bromo-2-nitrobenzene (recovery indicator) were added to the tubes. The extraction step using a microwave digestion/extraction system in 1,200 W was based on the following procedure: ramp to 100 °C over the course of 10 min and hold at 100 °C for another 10 min.

The extracts were concentrated to approximately 1 ml by rotary evaporation and then reconcentrated to 1 ml after adding 10 ml of *n*-hexane. The extracts were subsequently transferred to silica/alumina (lower layer/upper layer, $V/V=1:1$) chromatography columns for cleanup, and 50 ml of mixed solvent (mixed with DCM and *n*-hexane, $V/V=2:3$) was used to elute the OCPs. The eluate was first

concentrated to 1 ml by rotary evaporation and then reconcentrated to 1 ml after adding 10 ml of *n*-hexane. PCNB was added to the sample as an internal standard, and the samples were transferred to vials and sealed for analysis.

After they were sieved and mixed, 8-g sediment samples were weighed, and their extraction and cleanup procedures were the same as those of the suspended solids.

Sample analysis and quality assurance

The water samples were analyzed using an Agilent 6890 gas chromatograph equipped with a 63Ni microelectronic capture detector and an HP-5 column (30 m×0.32 mm× 0.25 μm, Agilent Co., USA). The samples were injected using an autosampler in splitless mode with a venting time of 0.75 min. The oven temperature was programmed to increase from 50 to 150 °C at 10 °C/min, increase to 240 ° C at 3 °C/min, and finally to maintain 240 °C for 15 min. Nitrogen was used as both the carrier (1 ml/min) and make-up gas (60 ml/min). The injector and detector temperatures were 220 and 280 °C, respectively.

The suspended solid and sediment samples were analyzed using an Agilent 6890-5976C gas chromatograph and mass spectrometer detector and a HP-5MS fused silica capillary column (30 m×0.25 mm×0.25 μm, Agilent Co., USA). Helium was used as the carrier gas at a flow rate of 1 ml/min. Samples (1 μl) were injected by the autosampler under a splitless mode at a temperature of 220 °C. The column temperature program was as follows: 50 °C for

2 min, 10 °C/min to 150 °C, 3 °C/min for 240 °C, 240 °C for 5 min, 10 °C/min for 300 °C, and 300 °C for 5 min. The ion source temperature of the mass spectrometer was 200 °C, the temperature of the transfer line was 250 °C, and the temperature of the quadrupole was 150 °C. The compounds were quantified in the selected ion mode, and the calibration curve was quantified with the internal standard.

It is necessary to add the quality control/quality assurance procedure, which includes the method blank, standard blank, matrix adding standard, parallel samples, and standard reference materials, to control sample analysis during the procedure. The recovery rate of the entire procedure was controlled by the recovery indicator (PCNB and 1-bromo-2-nitrobenzene). The recoveries of the OCPs in the water ranged from 41.4 to 107.4 %, and the detection limits of the method ranged from 0.01 to 1.30 ng/L. Due to the drawbacks of the SPE method, the recoveries of o,p'-DDE and p,p'-DDE were low (Sibali et al. 2009), but the SPE method is still well suited to the preparation of a large quantity of water samples in the field because of its carrying convenience, ease of operation, and high throughput. The recoveries of suspended solid samples ranged from 76.3 to 142.4 %, and the detection limits of the method ranged from 0.02 to 2.59 ng/g. The recoveries of sediment samples ranged from 86.73 to 128.47 %, and the detection limits of the method ranged from 0.01 to 0.67 ng/g.

Other analyses

The sediment samples were divided into two parts to determine total carbon (TC) and inorganic carbon (IC) using a total organic carbon (TOC) analyzer (TOC-5000A; Shimadzu Corp., Japan) with a solid sampler (SSM-5000A) to obtain TOC levels. Analytically pure glucose was used to measure the TC standard curve, and analytically pure sodium carbonate was used to measure the IC standard curve.

Results and discussion

Residual levels and compositions of OCPs in the water, suspended solids, and sediment

Residual levels of OCPs in the water, suspended solids, and sediment

Table 1 indicates that 24 types of OCPs were detected in the water with a mean concentration of 132.4±432.1 ng/L. The levels of isodrin (112.7±402.4 ng/L) were the highest and account for 85.1 % of total OCPs. β-HCH (1.2±2.1 ng/L) was the dominant HCH isomer, followed by γ-HCH (0.4± 0.8 ng/L) and α-HCH (0.4±0.8 ng/L). The dominant DDT

metabolites were in the water p,p'-DDT (3.1±2.3 ng/L) and o,p'-DDT (1.0±1.1 ng/L). Methoxychlor, mirex, and DDMU were the primary pollutants in the remaining 14 OCPs.

Fifteen types of OCPs were detected in the suspended solid samples, with a mean concentration of 188.1± 286.7 ng/g. DDTs (121.2±165.3 ng/g) and HCHs (13.1± 11.8 ng/g) were the primary OCPs, which accounted for 64.4 and 7.0 %, respectively. The dominant DDT metabolites in the suspended solids were p,p'-DDT (72.8± 98.3 ng/g) and o,p'-DDT (31.7±46.1 ng/g), which accounted for 60.1 and 26.1 % of total DXs, respectively. In the four isomers of HCH, γ-HCH (6.5±7.2 ng/g) was dominant, followed by α-HCH (2.6±2.4 ng/g), β-HCH (2.3±3.3 ng/g), and δ-HCH (1.8±2.2 ng/g). Isodrin and HCB were the dominant pollutants in the remaining OCPs.

Thirteen types of OCPs were detected in the sediments from Lake Chaohu, with a mean concentration of 13.7± 9.8 ng/g, including the four isomers of HCHs (α-HCH, β-HCH, γ-HCH, and δ-HCH), two types of DDTs and three metabolites of DDTs (o,p'-DDT, p,p'-DDT, p,p'-DDE, o,p'-DDD, and p,p'-DDD), HCB, isodrin, dieldrin, and endrin. Isodrin (6.6±6.3 ng/g) and DDTs (4.4±7.9 ng/g) were the dominant OCPs and accounted for 48.5 and 31.8 %. The p,p'-DDD (2.4±7.3 ng/g) and p,p'-DDE (1.2±0.9 ng/g) were two dominant DDT metabolites. The total concentration of the four HCH isomers was only 0.8±0.6 ng/g. Endrin (0.8± 1.7 ng/g) was the dominant pollution of the remaining OCPs, at a level that was slightly greater than those of HCHs.

Table 2 shows that the concentrations of HCHs and DDTs in the water, suspended solids, and sediments from Lake Chaohu were comparable to other lakes and rivers around the world. In the water, the residual levels of HCHs in Lake Chaohu were similar to those in the Nanjing section of the Yangtze River and Huaihe River and slightly greater than those in Lake Small Baiyangdian and Lake Baikal in Russia, but they were lower than those in the Beijing Guanting Reservoir and Ebro River in Spain and much lower than those in the Gomti River in India and Kucuk Menderes River in Turkey. The levels of DDTs were similar to those in the Beijing Guanting Reservoir and greater than those in Lake Small Baiyangdian, the Nanjing section of the Yangtze River, the Haihe Plain, and the Ebro River, but they were much lower than those in the Huaihe River and the Gomti River. Data on the OCP residues in the suspended solids were too few to demonstrate that the levels of HCHs were comparable to those in Lake Small Baiyangdian and the Nanjing section of the Yangtze River. The levels of DDTs were greater than those in the Nanjing section of the Yangtze River and comparable to those in the Haihe Plain. In the sediments, the concentrations of HCHs in Lake Chaohu were comparable to those of the Huaihe

Table 1 Residual levels of OCPs in the water, suspended solids, and sediment from Lake Chaohu

Pesticides	Water (ng/L)		Suspended solids (ng/g, dw)		Sediments (ng/g, dw)	
	Range	Mean±SD	Range	Mean±SD	Range	Mean±SD
a-HCH	ND–3.2	0.4±0.8	0.7–9.7	2.6±2.4	0.1–0.6	0.2±0.2
b-HCH	0.4–8.6	1.2±2.1	ND–12.5	2.3±3.3	0.1–0.9	0.3±0.3
r-HCH	ND–3.0	0.4±0.8	1.3–30.1	6.5±7.2	0–0.3	0.1±0.1
d-HCH	ND–0.2	0.1±0.1	ND–7.8	1.8±2.2	ND–0.2	0.1±0.1
HCHs	0.4–14.7	2.0±3.6	2.3–47.6	13.1±11.8	0.2–1.8	0.8±0.6
o,p'-DDE	ND–2.7	0.8±0.8	ND	ND	ND	ND
p,p'-DDE	ND–1.3	0.2±0.3	1.7–48.4	11.1±13.4	0.2–3.3	1.2±0.9
o,p'-DDD	ND–2.2	0.8±0.7	ND	ND	0.1–1	0.2±0.2
p,p'-DDD	ND–1.4	0.2±0.5	ND–32.1	5.6±8.8	ND–27.8	2.4±7.3
o,p'-DDT	ND–3.5	1.0±1.1	3.1–180.2	31.7±46.1	ND–0.3	0.1±0.1
p,p'-DDT	0.5–10.1	3.1±2.3	6.8–396.4	72.8±98.3	0.1–0.9	0.4±0.3
DDTs	0.5–18.4	5.9±4.3	12.9–657	121.2±165.3	0.3–31	4.4±7.9
HCB	ND–0.8	0.1±0.2	2.6–49	11.4±12.6	ND–0.2	0.03±0.1
Isodrin	0.8–1,565.6	112.7±402.4	2.4–90.7	18.2±23.5	0.1–22.5	6.6±6.3
Dieldrin	ND–2.1	0.7±0.5	ND–3.7	0.3±1	ND–1.8	0.3±0.6
Endrin	ND–4.2	0.4±1.1	ND–4.7	0.3±1.2	ND–6.1	0.9±1.7
Heptachlor	ND–0.8	0.2±0.3	ND	ND	0.9–36.9	ND
Aldrin	ND–0.9	0.3±0.3	ND–17.3	3.8±5.1	ND	ND
hep-epo-A	ND–0.6	0.1±0.2	ND	ND	ND	ND
hep-epo-B	ND–1.3	0.3±0.4	ND	ND	ND	ND
r-Chlordane	ND–2.1	0.4±0.6	ND–7.8	0.6±2	ND	ND
DDMU	ND–24.3	3.4±6.2	ND	ND	ND	ND
a-Chlordane	ND–0.6	0.2±0.2	ND	ND	ND	ND
Endosulfan	ND–3.7	0.9±1.0	ND–32.1	4.9±8.3	ND	ND
Methoxychlor	ND–18.2	1.3±4.7	ND–42.8	2.9±11.1	ND	ND
Mirex	ND–17.4	3.6±4.6	ND–48.0	6.5±13.2	ND	ND
OCPs	1.6–1,678.6	132.4±432.1	18.6–1,046.8	188.1±286.7	0.9–36.9	13.7±9.8

ND not detected

River, Lake Small Baiyangdian, the Bahlui River in Romania, and Ulsan Bay in Korea, greater than the Ebro River and Lake Baikal, but two orders of magnitude lower than the Gomti River. The concentrations of DDTs were comparable with the Suzhou River and the Bahlui River, greater than those in Lake Small Baiyangdian, the Haihe Plain, the Ebro River, Lake Baikal, and Ulsan Bay, but they were lower than those in the Gomti River.

Compositions of OCPs in the water, suspended solids, and sediments

The relative composition characteristics of OCPs in the water, suspended solids, and sediments from Lake Chaohu are indicated in Figs. 2a, b, and c. Figure 2a shows that isodrin, mirex, DDMU, and HCHs were the dominant OCPs in the water from Lake Chaohu, and they accounted for 85.1, 4.5, 2.8, 2.6, and 1.5 %, respectively. In the suspended solids, DDTs, isodrin, HCHs, HCB, and mirex were the

dominant OCPs, and they accounted for 66.1, 10.0, 7.2, 6.2, and 3.6 %, respectively. The dominant OCPs in the sediments were significantly different from those in the suspended solids, which were heptachlor, isodrin, DDTs, endrin, and HCHs, with percentages of 51.4, 24.9, 16.4, 3.2, and 2.8 %, respectively.

Figure 2b indicates that β-HCH was the major isomer of HCHs in the water and accounted for 59.6 %, followed by γ-HCH, α-HCH, and δ-HCH at 19.7, 17.2, and 3.9 %, respectively. In the suspended solids, γ-HCH was the major isomer and accounted for 49.3 %. The concentrations of α-HCH, β-HCH, and δ-HCH were similar and accounted for 19.8, 17.6, and 13.3 %, respectively. In the sediments, β-HCH and α-HCH were the dominant isomers, accounting for 41.9 and 30.3 %, respectively, and the concentrations of γ-HCH and δ-HCH were relatively lower and accounted for 16.2 and 11.5 %, respectively.

Figure 2c illustrates that the relative content of DDTs was approximately 70.0 % in the water and that 51.6 % was

Table 2 Comparisons of HCH and DDT contents in the water, suspended solids, and sediments from Lake Chaohu with those from other regions worldwide

Sampling area	HCHs			DDTs			References
	Water (ng/L)	SS (ng/g, dw)	Sediment (ng/g, dw)	Water (ng/L)	SS (ng/g, dw)	Sediment (ng/g, dw)	
This study	0.4–14.7	2.3–47.6	0.2–1.8	0.5–18.4	12.9–657.0	0.3–31.0	
Lake Small Baiyangdian	0.5–6.8	ND–104.4	0.2–3.1	N.D.	60.5–320.0	0.03–1.4	Wang et al. (2012b)
Suzhou River	17–90		ND–13.4	17–99		8.5–28.5	Hu et al. (2005)
Nanjing section of Yangtze River	9.27–10.51	11.3–21.87		1.57–1.79	14.63–20.72		Jiang et al. (2000)
Beijing Guanting Reservoir	3.9–38.9		1.1–7.7	3.7–16.0		3.0–10.5	Wan et al. (2009)
Haihe Plain				0.3±0.7	423.1±577.9	7.1±7.6	Wang et al. (2012a)
Huai He River	1.1–7.6			4.5–78.9			Yu et al. (2004)
Huai He River			0.9–1.9			1.4–4.1	Huang et al. (2008)
Ebro River, Spain	0.2–28.6		0.001–0.03	2.0–6.8		0.9–9.0	Fernandez et al. (1999)
Gomti River, India	0.02–4,846.0		0.1–1,650.0	ND–4,578.0		ND–509.0	Singh et al. (2005)
Kucuk Menderes River, Turkey	187–337			ND–120			Turgut (2003)
Bahlui River, Romania			0.4–3			0.18–18	Neamtu et al. (2009)
Lake Baikal, Russia	0.06–0.96		0.02–0.12	ND–0.02		0.014–2.7	Iwata et al. (1995)
Ulsan Bay, Korea			0.02–4.55			0.02–4.9	Khim et al. (2001)

derived from *p,p'*-DDT and 17.2 % from *o,p'*-DDT; the relative contents of *o,p'*-DDE (12.7 %) and *o,p'*-DDD (12.8 %) were similar; low levels of *p,p'*-DDE (2.8 %) and *p,p'*-DDD (2.9 %) were also observed. There were several

differences between the composition of DDTs in the suspended solids and in the sediments. The relative content of DDTs in the suspended solids was 86.2 %, and *p,p'*-DDT and *o,p'*-DDT were dominant, accounting

Fig. 2 Compositions of OCPs, HCHs, and DDTs in the water (**a**), suspended solids (SS) (**b**), and sediments (**c**) from Lake Chaohu
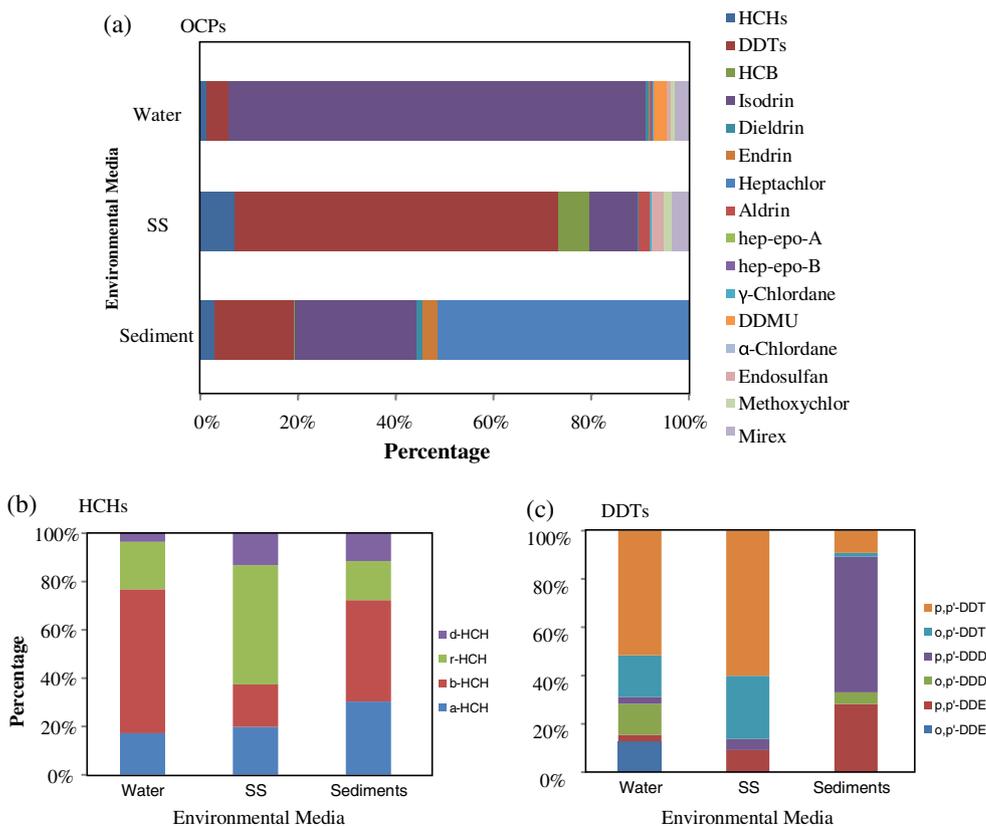
for 60.1 and 26.1 %, respectively, followed by p,p'-DDE (9.2 %) and p,p'-DDD (4.6 %); the levels of the other two metabolites were below the detection limit. In the sediments, the dominant metabolites were p,p'-DDD and p,p'-DDE, which accounted for 56.2 and 28.2 %, respectively. The relative contents of p,p'-DDT, o,p'-DDT, and o,p'-DDD were 9.3, 1.6, and 4.9 %, respectively, while o,p'-DDE was not detectable.

Spatial distributions of OCPs residues in the water, suspended solids, and sediment

The site-to-site distributions of OCP residues in the water, suspended solids, and sediments are presented in Fig.3a, b, and c and in Tables S1, S2, and S3 in the supporting information (SI), respectively. Table 3 shows that the spatial distributions of OCPs in different areas including the river and lake, the western and eastern lake (WL, EL), as well as the eastern water source lake area (EWSA) and eastern lake area (ELA) excluding EWSA.

Figure 3a and Table S1 in the SI reveal that the highest content of OCPs in the water was found at site B6 (Shuangqiaohe River), followed by those at sites C2 (the upstream of Xiaozhegaohe River) and C4 (Dazhegaohe River). In the suspended solids, the

highest content of OCPs was found at site C1 (downstream of Xiaozhegaohe River), followed by those at sites C4 and B6 (Fig.3b and Table S2 in the SI). However, the contents of OCPs in the sediments at sites C1 and B6 were lower, while those at sites B2 in the EWSA, A1 in the ELA, and D1 in the WL were higher (Fig.3c and Table S3 in the SI).

Table 3 shows that the contents of total OCPs, HCHs, DDTs, and other OCPs in the water and suspended solids were higher in the river than those in the lake. This might indicate that the OCPs in the lake were from the inputs of the inflowing rivers. For the lake water, the contents of total OCPs, HCHs, DDTs, and other OCPs were higher in the WL than those in the EL; however, the converse spatial distribution trends in the contents were found for the suspended solids. This may be derived from the different source of OCPs for the water and for the suspended solids. The OCP residues in the water might be primarily from the urban and industrial wastewater, while those in the suspended solids mainly from the agricultural runoff. The large amount of the urban and industrial wastewater from Hefei City, the capital of Anhui Province, might be the cause for the higher OCP contents in the water in the WL than those in the EL; while the effects of
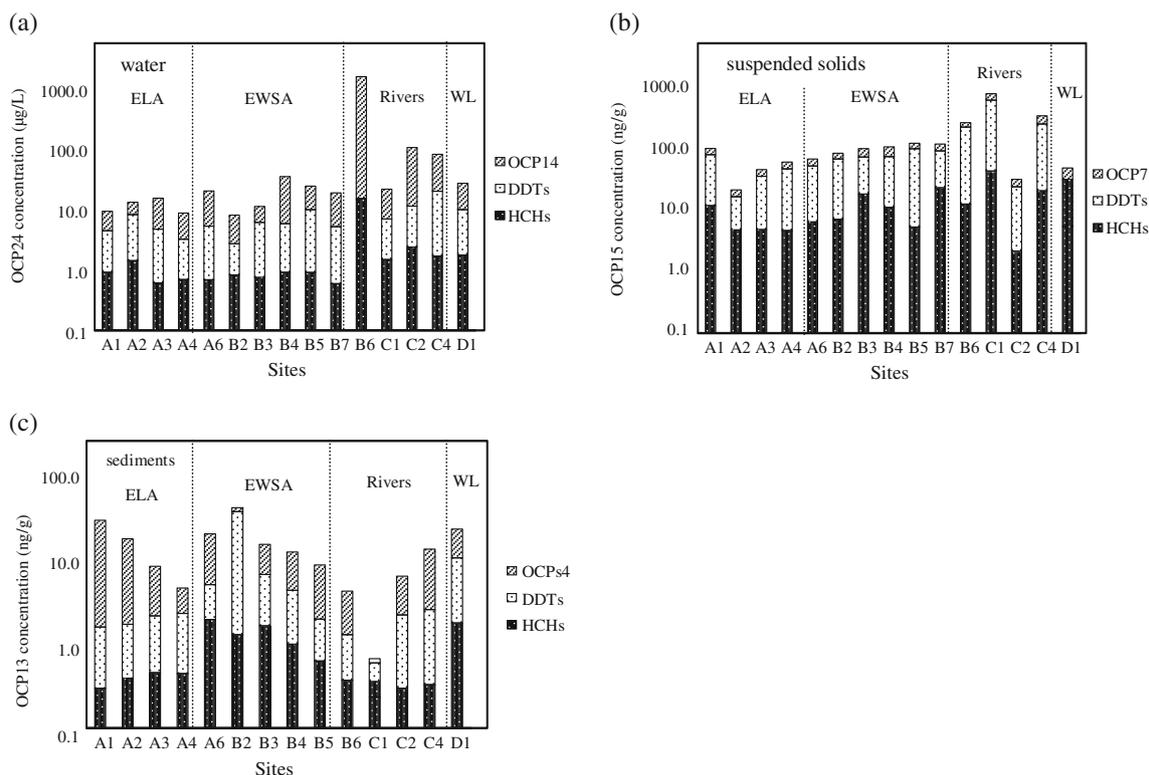

Fig. 3 The site-by-site distributions of OCPs in the water (a), suspended solids (b), and the sediments (c) from Lake Chaohu (*ELA* eastern lake area, *EWSA* eastern water source area, *WL* western lake)

Table 3 Spatial distributions of typical OCP compounds in the water, suspended solids, and sediments from Lake Chaohu

Environment media	Area	Contents (ng/L for water, ng/g dw for suspended solids and sediments)					
		OCPs	HCHs	DDTs	Isodrin	HCB	Other OCPs
Water	WL	27.2	1.8	8.2	0.8	ND	16.5
	ELA	11.7±3.2	0.9±0.4	4.3±1.8	4.0±3.2	0.04±0.09	2.4±1.3
	EWSA	19.7±9.7	0.8±0.1	5.1±2.3	2.9±1.2	0.06±0.04	10.9±9.0
	EL	16.5±8.5	0.8±0.2	4.8±2.1	3.3±2.1	0.05±0.06	7.5±8.1
	Lake	17.5±8.7	0.9±0.4	5.1±2.2	3.1±2.2	0.05±0.06	8.3±8.1
	River	448.4±757.7	5.1±6.5	8.3±7.6	414.1±768.5	0.28±0.39	20.6±23.8
Suspended solids	WL	53.6	5.7	29.1	6.0	4.59	8.2
	ELA	63.6±37.7	7.0±3.9	42.5±26.5	7.5±5.6	6.02±3.33	0.5±1.1
	EWSA	112±23.7	12.9±8.0	72.4±18.6	12.3±6.9	7.10±1.65	7.3±4.5
	EL	92.6±37.6	10.5±7.1	60.4±25.8	10.4±6.6	6.67±2.35	4.6±4.9
	Lake	89.1±37.5	10.1±6.9	57.6±26.2	10.0±6.4	6.48±2.31	4.9±4.8
	River	407.0±362.7	21.5±19.3	296.2±263.6	40.8±39.0	25.09±19.66	23.4±24
Sediments	WL	20.2	1.7	7.7	10.8	0.09	ND
	ELA	13.2±9.5	0.4±0.1	1.4±0.2	10±9.8	0.06±0.09	1.4±1.0
	EWSA	17.1±11.0	1.2±0.5	8.5±12.6	5.3±4.4	0.01±0.01	1.8±2.5
	EL	15.4±9.9	0.8±0.6	5.4±9.7	7.4±7.2	0.03±0.06	1.8±1.9
	Lake	15.9±9.5	0.9±0.6	5.6±9.2	7.7±6.9	0.04±0.06	25.2±1.9
	River	5.5±4.7	0.3±0.1	1.2±0.8	4.0±4.0	0.01±0.01	ND

WL included site D1; ELA included sites A1, A2, A3, and A4; EWSA included sites A6, B1, B2, B3, B4, B5, and B7; EL included both ELA and EWSA sites; Lake included all lake sites; River included B6, C1, C2, and C4. The detection limits of HCB in the water, suspended solids, and sediments were 0.01 ng/L, 0.003 ng/g, and 0.001 ng/g, so two decimal places were retained

Chaohu City might produce higher OCP contents in the water in the ESWL than those in the ELA (Table 3). The agricultural runoff from the farmland distributing on the shore of southwestern lake was mainly inflowing into the eastern lake, which contributes about 60 % of total volume of water into Lake Chaohu. This might be contributing to the higher contents of OCPs in the suspended solids in the EL than those in the WL.

For the sediments in the river and lake, compared with the water and suspended solids, the converse spatial distribution trends in the contents of total OCPs, HCHs, DDTs, and other OCPs were found, which showed that the contents were higher in the lake sediments than those in the river sediments. For the lake sediments, the contents of total OCPs, HCHs, DDTs, and other OCPs were higher in the WL than those in the EL, which showed the same spatial trends with those in the lake water. Unlike the water and suspended solids that are affected by the source of OCPs at short term, the sediments are affected by the deposition of suspended solids at long term. The spatial distributions of OCPs in the sediments would be mainly attributed to the sources of OCPs, the particle size and its organic carbon content, as well as the hydrodynamic conditions in the deposition environment. The fine particulate matter with a large specific surface area and high organic carbon content has strong ability for OCP adsorption. Compared with the inflowing rivers, the lake is characterized by the finer particulates and weaker hydrodynamic conditions. This might be the reason for the higher OCP contents in the lake sediments than those in the river sediments. The higher OCP contents in the western lake sediments would be derived from the continuous inputs of OCPs from the three most polluted inflowing rivers, Nanfeihe, Shiwulihe, and Paihe, receiving large amounts of the OCP-containing domestic effluents and industrial wastewater from Hefei City, the capital of Anhui Province. However, the OCP contents in the eastern lake sediments would be mainly affected by the particles from agricultural runoff. The OCP contents in the agricultural runoff-deduced particles may be higher than those in the wastewater-containing particles, but the contributions of the agricultural runoff-deduced particles to the sediments are not continuous, which are controlled by the rainfall in the lake catchments. No statistic correlations were found for the contents of OCPs and organic carbon in the water, suspended solids, and sediments, which indicated that the organic carbon contents might not be the governing factors for the spatial distribution of OCPs.

The partition of OCPs in the water, suspended solids, and sediments and the correlation with K_{ow}

The partition of OCPs between the water and suspended solids and the correlation with K_{ow}

The organic pollutants in the water can be assigned to the suspended solids by dissolution and achieve partition equilibrium after a period of time; meanwhile, they are adsorbed by the suspended solids, reducing the concentration of pollutants in the water. The adsorption process can reduce the residual levels and the toxicity of pollutants in the water (Means 1995). The water–suspended solid partition coefficient K_d of OCPs was calculated by the following equation:

$$K_d = \frac{C_w}{C_{ss}} \quad (1)$$

where K_d is the partition coefficient (in liter per gram), and C_w (in nanogram per liter) and C_{ss} (in nanogram per gram) are the concentrations of OCPs in the water and the suspended solids. Due to the lack of data on total organic carbon in suspended solids, K_{oc} cannot be calculated.

The results of K_d are indicated in Table 4, and all of the values of K_d (except isodrin) were less than 1, which shows that the OCPs were located in the suspended solids.

Various factors affected the pollutant adsorption by suspended solids. Among the most important were the physical and chemical properties of the pollutants (such solubility and K_{ow}) (Karickhoff et al. 1979; Heemken et al. 2000). In this study, there was no correlation between $\log K_d$ and $\log K_{ow}$ ($r=0.034$, $p=0.916$, $N=12$). During the high water level period, the particles in Lake Chaohu were derived

from surface runoff, and the partition of OCPs between water and suspended solids did not achieve partition equilibrium (Gao et al. 2001). Please see the next section for more explanations.

The partition of OCPs between the water and sediments and the correlation with K_{ow}

The difference in OCP residues between the water and the sediments resulted from the adsorption of OCPs by sediment organic matter (Li et al. 1998). The sediment–water organic-C weighted sorption coefficient (K_{oc}) was calculated by the following equation:

$$K_{oc} = \frac{K_d}{TOC} \quad (2)$$

$$K_d = \frac{C_{sed}}{C_w} \quad (3)$$

where K_d is the partition coefficient calculated by the ratio of the concentration of OCPs in the sediment (C_{sed}, in nanogram per gram) to the concentration in the water (C_w, in nanogram per liter); TOC is the relative content of total organic carbon in the sediments. The results are shown in Table 5. The values of $\log K_{oc}$ in this study were all greater than those in the references, and there was no correlation between $\log K_{oc}$ and $\log K_{ow}$ ($r=0.121$, $p=0.707$, $N=12$).

Our previous study on the partitioning of HCHs and DDTs between water and sediments in Lake Small Baiyangdian also found that the measured $\log K_{oc}$ values were greater than those in the references (Wang et al. 2012b). One possible explanation for these differences is that previously published $\log K_{oc}^*$ values were calculated based on soil–water partitioning. Li et al. (1995) noted that one difference between sediments and soil is the extent of water saturation. With higher polarity, the dissolution of organics in water leads to a lower polarity of organic matter in sediments than in soil. Because organic matter polarity has a substantial effect on organic carbon partitioning of non-ionic pesticides in sediments and soil, a lower polarity relates directly to a higher K_{oc} value (Li et al. 1995). Furthermore, the $\log K_{oc}^*$ was measured in a laboratory after dynamic sorption and desorption reached chemical equilibrium conditions, which cannot be guaranteed in the field. From the observed $\log K_{oc}$ values, it is likely that the partitioning of OCPs was not in equilibrium between the solid and aqueous phase in Lake Chaohu, and therefore, a portion of OCPs may partition into the aqueous phase (Tan et al. 2009).

The partitioning of OCPs between water and solid phase, and their movement across the water/sediment interface is primarily dependent on the sorption–desorption process

Table 4 The water–suspended solid partition coefficient of OCPs (K_d)

	SD	Media	Max	Min	Mean	GeoMean
a-HCH	0.29	0.04	1.14	0.00	0.16	0.08
b-HCH	0.71	0.26	2.25	0.04	0.57	0.31
r-HCH	0.25	0.01	0.73	0.00	0.12	0.05
d-HCH	0.18	0.05	0.59	0.00	0.13	0.13
HCHs	0.34	0.11	1.08	0.02	0.24	0.13
p,p′-DDE	0.03	0.00	0.11	0.00	0.02	0.04
p,p′-DDD	0.10	0.00	0.30	0.00	0.04	0.17
o,p′-DDT	0.18	0.03	0.62	0.00	0.11	0.07
p,p′-DDT	0.13	0.07	0.43	0.00	0.11	0.06
DDTs	0.15	0.07	0.53	0.00	0.13	0.06
HCB	0.03	0.00	0.11	0.00	0.02	0.03
Aldrin	0.24	0.05	0.71	0.00	0.14	0.06
Isodrin	24.67	0.32	95.27	0.04	7.97	0.47
Endosulfan	0.75	0.22	2.08	0.00	0.52	0.33
Mriex	0.17	0.03	0.39	0.00	0.13	0.14
OCPs	1.45	0.21	5.24	0.02	0.74	0.24

Table 5 The sediment–water partition coefficients of OCPs

Pollutant	K_d	K_{oc}	LogK_{oc}	LogK_{ow}*	LogK_{oc}*
a-HCH	4,118.62	784,246.67	5.39	3.80	3.25
b-HCH	541.80	100,164.84	4.60	3.78	3.36
r-HCH	1,291.18	256,255.61	4.79	3.76	3.00
d-HCH	1,376.55	219,375.13	5.11	4.15	3.82
HCHs	815.98	162,517.06	4.83		
HCB	117.92	26,088.11	4.13	5.73	3.99
Isodrin	3,092.86	364,696.58	4.94	-	
Dieldrin	473.80	61,370.32	5.39	8.78	
Endrin	1,781.45	663,680.71	5.65	8.78	
p,p'-DDE	9,609.63	1,203,359.26	5.76	5.96	4.82
o,p'-DDD	397.95	59,370.21	4.48	6.20	
p,p'-DDD	1,763.33	180,483.67	5.26		
o,p'-DDT	47.25	4,399.62	3.84	6.91	
p,p'-DDT	193.85	31,175.89	4.27		5.31
DDTs	1,648.63	280,571.33	4.83		
OCPs	756.59	97,286.97	4.76		

LogK_{ow} and logK_{oc}* were the values in the references (Shen and Wania 2005; Sabljić et al. 1995)

governed by their solubility in water and their affinity to the solid phase (Tao et al. 1999). The sorption of an organic chemical in the soil or sediment is usually described by an organic carbon normalized sorption coefficient (K_{oc}). During the past decades, quite a number of experimental K_{oc} have been reported; however, the amount of experimental data available for K_{oc} is still much less than that for K_{ow} (n-octanol/water partition coefficient), while the experimental error associated with experimental K_{oc} is higher than that with measured K_{ow} (Tao et al. 1999). Since 1970s, there have been numerous efforts attempting to estimate K_{oc} based on molecular structure or other parameters, due to the difficulty in the experimental measurements of K_{oc}, especially for the strong hydrophobic compounds with logK_{oc}>4 (Piao et al. 1999; Tao et al. 1999). Through a review of the various alternative methods, Gawlik et al. found that the statistic models for the relationship between the K_{oc} and K_{ow} known as logK_{oc} vs. logK_{ow} were the most commonly used approach to describe the soil/sediment sorption of organic chemicals (Gawlik et al. 1997). However, the different logK_{ow}–logK_{oc} models have large uncertainties, particularly for the compounds with logK_{ow}>4 (Kanazawa 1989; Sabljić et al. 1995; Gawlik et al. 1997; Piao et al. 1999). In fact, the K_{ow} only describes the partitioning of compounds in the pure organic phase and aqueous phase; however, the K_{oc} involves the partitioning of compounds in the complex heterogeneous soil/sediment medium. The adsorption behavior of OCPs might have large differences in the n-octanol phase and in the organic matter of the soil/sediment. The former is relatively simple, while the latter is more complex. This might be the fundamental reason why the relationships between logK_{ow} and logK_{oc} based on the experimental data are with such large

uncertainties. Therefore, it is not surprising that there is no relationship between the K_{ow} values from the experimental data and the K_{oc} values from the calculations based on the measured data in the actual environment for OCPs.

Except for the effects of the physicochemical parameters, such as K_{ow}, there are some other factors affecting the partitioning of organic chemicals in water and sediment, for instance, the contents of particle and dissolved organic matter in the water (Turner et al. 1999; Krauss and Wilcke 2001), and the features and sources of particulate matters, the content and structure of organic matters, and the black carbon content in the sediments (Isaacon and Frinj 1984; Johnson and Amy 1995; Perminova et al. 1999; Johnson et al. 2001; Leppanen and Kukkonen 2000). There are a variety of natural colloids in sediments, such as clay minerals, organic matter, iron, manganese, aluminum hydrous oxides, and silica colloid and so on, which play an extremely important role in the adsorption and accumulation of organic pollutants (Yu et al. 2005). The more complex adsorption mechanism was reported by many investigators, such as the nonlinear adsorption process and the phenomenon of slow adsorption and desorption (Song et al. 2002; Accardi-Dey and Gschwend 2002; Ghosh et al. 2001; Talley et al. 2002). As typical hydrophobic and lipophilic organic contaminants, OCPs are supposed to be closely related to the organic matter in sediments. Many studies have proved that the TOC contents in the sediments were one of the most important governing factors for the residues and distributions of OCPs which would be mainly attributed to the adsorption of organic matter (e.g., Xing and Pignatello 1998; Upal and Jeffrey 2001; Song et al. 2002; Accardi-Dey and Gschwend 2002; Talley et al. 2002). However, the present and other studies found that there were no correlations

between the contents of OCPs and TOC in the sediments (Wu et al. 2004; Yang et al. 2005; Zhao et al. 2008; Wang et al. 2009, 2012a, b, c). This would provide the additional supporting information for explicating the large uncertainties of $\log K_{ow}$ vs. $\log K_{oc}$ relationships.

The prevention of OCPs' eco-risks in Lake Chaohu

In the Chinese Environmental Quality Standards (EQSs) for Surface Water (GB3838-2002), only four types of OCPs including HCB, DDT, lindane, and heptachlor epoxide were suggested to set up the EQSs for the centralized drinking water source area. The EQS values are 0.05, 0.001, 0.002, and 0.0002 mg/L for HCB, DDT, lindane, and heptachlor epoxide, respectively. In the present study, the residual levels of these four types of OCPs were much lower than their EQS values. Our previous study on the ecological and health risks of DDTs and HCHs in the water from Lake Chaohu found that both carcinogenic and noncarcinogenic risks of DDTs and HCHs associated with the water use were very low (He et al. 2012). However, the MOS10 (margin of safety) values suggested that the lake was facing the potential ecological risks of p,p'-DDT to invertebrates, especially to shrimps and crabs, and the potentially affected fractions of p,p'-DDT to shrimps and crabs calculated by the species sensitive distribution models were between 0.07 and 0.52 % (He et al. 2012). The more serious problem was the recent input of pesticides containing DDT in the water samples from all studied four inflow rivers and from lake sites A2, B2, and B3, as indicated by the DDT/(DDD+DDE) ratios being greater than one (He et al. 2012).

Our previous study on the ecological risks of OCPs in the surface sediments from Lake Chaohu based on the Consensus-Based Sediment Quality Guidelines proposed by MacDonald et al. (2000) showed that the DDT and its metabolites in the surface sediments from some lake areas posed significant ecological risks to benthic organisms (Wang et al. 2012c). The residual levels of DDE and ΣDDTs in lake site D1 and the ΣDDT level in lake site B2 were between their TECs (consensus-based threshold concentrations) and PECs (consensus-based probable effect concentrations), which indicated that there were possible adverse effects of DDE and ΣDDTs to the benthos. However, the DDD level in lake site B2 was higher than its PEC, which meant that the significant adverse effects of DDD to the benthos usually occurred (Wang et al. 2012c).

Special attention should be paid to prevent the pollution and eco-risks of DDT-related components for the environmental department of Lake Chaohu. The agricultural use of DDT has been forbidden since 1983 in China; however, DDT use in China is not completely banned. DDT, as the auxiliary material of an antifouling paint, is still in use. Ten thousand tons of antifouling paints are used in fishing vessels each year in China, and about 250 tons of DDT is used in antifouling paints (He et al. 2012). In Lake Chaohu, there are about 3,500 fishing vessels, and most of them anchor in the eastern lakes, especially at Shuangqiaohe River, Zhegaohe River, and Chaohu City Dock. DDT's distribution might result from the ships' distribution. The development and use of new antifouling paints without DDTs are necessary to reduce the eco-risks of DDT-related components in Lake Chaohu.

Conclusion

The mean concentration of total OCPs in the water in Lake Chaohu was 132.4±432.1 ng/L during the high water level period. Isodrin and DDTs were the dominant pollutants and accounted for 85.1 and 4.5 %. In the suspended solids, the concentration of total OCPs was 188.1±286.7 ng/g. DDTs, isodrin, and HCHs were the dominant pollutants and accounted for 64.4, 9.7, and 7.0 %, respectively. In the sediments, the concentration of total OCPs was 13.7± 9.8 ng/g. Isodrin and DDTs were the dominant pollutants and accounted for 48.5 and 31.8 %, respectively. β-HCH was the dominant HCH isomer in the water and sediments, while γ-HCH was the dominant isomer in the suspended solids. p,p'-DDT was the major pollutant in the water and suspended solids, while p,p'-DDD was the main metabolite in the sediments. The mean levels of OCPs, HCHs, and DDTs in the water and suspended solids were all greater than those in the lake. In the water, the levels of the pollutants in the western lake were greater than those in the eastern lake, while data on the suspended solids reveal the opposite trend. The residual levels of total OCPs, HCHs, and DDTs in the sediments from the lake were greater than those from the river. There was no significant correlation between $\log K_d$ and $\log K_{ow}$ or between the sediment–water organic-C weighted sorption coefficients $\log K_{oc}$ and $\log K_{ow}$.

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