



Characterization, ecological and health risks of DDTs and HCHs in water from a large shallow Chinese lake

Wei He, Ning Qin, Qi-Shuang He, Yan Wang, Xiang-Zhen Kong, Fu-Liu Xu *

Ministry of Education Laboratory for Earth Surface Process, College of Urban & Environmental Sciences, Peking University, Beijing 100871, China

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ABSTRACT

The levels of dichloro diphenyl trichloroethanes (DDTs) and hexachlorocyclohexanes (HCHs) in water from Lake Chaohu were measured. The residues, possible sources and potential ecological and health risks of these compounds were analyzed. The results show that the contents of total DDTs and HCHs in the water varied from 1.52 to 21.79 and from 1.58 to 31.66 ng L⁻¹, respectively, which were higher than those in other Chinese lakes. The main sources of HCHs and DDTs were lindane and technical DDT, respectively. The o,p'-DDT/p,p'-DDT ratios indicated the new illegal inputs of DDT in all studied inflow rivers and some lake areas. The MOS10 (margin of safety) values suggested that the Lake was facing a potential ecological risks from p,p'-DDT, whereas the risk of γ -HCH was small. Both carcinogenic and noncarcinogenic risks of DDTs and HCHs associated with the water use were very low.

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1. Introduction

Organochlorine pesticides (OCPs) have been detected in vegetables (Tao et al., 2005), animal muscle tissue (Blanes et al., 2009), human breast milk (Furst et al., 1994) and environmental media (Gewurtz et al., 2007; Li et al., 2007; Tao et al., 2005, 2008) due to their wide usage in the past, very slow degradation in the environment or in organisms and bioaccumulation (Zhang et al., 2005). The persistence and semi-volatility of these substances also result in their long distance transportation (Geisz et al., 2008). Dichloro diphenyl trichloroethanes (DDTs), including o, p'-DDT, p, p'-DDD, o, p'-DDD, o, p'-DDE and p, p'-DDE have been proven to be potential endocrine disruptive chemicals (EDCs) and highly carcinogenic xenobiotic substances that increase the risk of human liver, pancreatic and breast cancers (Snedeker, 2001; Turusov et al., 2002). Among four isomers of hexachlorocyclohexanes (HCHs, including α -HCH, β -HCH, γ -HCH and δ -HCH), α -HCH has been found to cause human neurological disorders, gastrointestinal discomfort and liver and kidney damage; β -HCH is the most persistent isomer in the natural environment, and the bioaccumulation and chronic toxicity of this substance have been demonstrated to have serious consequences (SCPOPs, 2009a). Despite its easy degradation, slow-acting toxicity effects and low carcinogenic risk, lindane (γ -HCH) is associated with some degree of neurotoxicity and is still widely used in some countries. Therefore, α -HCH, β -HCH and lindane (γ -HCH) were included in

the *Nine New Controlled POPs List* during the *Stockholm Convention on Persistent Organic Pollutants* (SCPOPs, 2009b).

The use of OCPs has been banned since 1983 in China, but their production and use prior to this for more than 30 years has caused 14 million hectares of arable land to become polluted (Feng et al., 2003). Additionally, OCP residues in the soil have been transported into water or groundwater by surface runoff, underground runoff, infiltration, eluviation and desorption, resulting in water pollution, drinking water safety problems and ecological risks to the aquatic environment (Cao et al., 2004). It has been found that DDT and HCH pesticide residues are widely distributed in freshwater bodies such as ponds, rivers, lakes, reservoirs and estuaries (Dua et al., 1996; Janiot et al., 1994; Maskaoui et al., 2005; Tao et al., 2007; Yamashita et al., 2000; Zhou et al., 2008).

Lake Chaohu, a shallow freshwater lake, located in southeastern China, is one of the five largest freshwater lakes in China. It was well-known for its scenic beauty and richness in aquatic biodiversity before the 1950s. However, the lake and some of its inflowing rivers have been suffering from serious eutrophication and heavy metal pollution in the last several decades, which are caused by population growth, economic development and the lack of pollution monitoring and risk management (Li et al., 2010; Xu et al., 1999, 2001; Zhang et al., 2007). Phthalate esters (PAEs) and polycyclic aromatic hydrocarbons (PAHs) have been detected in Lake Chaohu, and it has been found that the northwest part of the lake has been heavily polluted by domestic and industrial wastewater (Yang et al., 2009). Xu et al. (2001) suggested that pesticide contamination was also one of the most important chemical stresses to the health of ecosystem in Lake Chaohu. The objectives of the present study were: (1) to investigate the residual levels of DDT

* Corresponding author. Tel./fax: +86 10 62751177.

E-mail address: xufl@urban.pku.edu.cn (F.-L. Xu).

and HCH pesticides in surface waters from Lake Chaohu during the high water period; (2) to identify the potential sources of DDTs and HCHs in the surface water; (3) to evaluate the ecological and health risks of DDTs and HCHs in the surface water; (4) to provide scientific supporting information for control of the lake POPs pollution.

2. Methods

2.1. Measurement of DDTs and HCHs Contents in Water

2.1.1. Reagent and materials

Analytical grade n-hexane and dichloromethane (DCM) (Beijing Reagent Company, Beijing, China) were purified by distillation. An organochlorine pesticide stock standard mixture was prepared by diluting a commercial mixed standard (Accustandard Inc., New Haven, Connecticut, USA) with n-hexane, and the working standard solution was prepared by diluting the stock standard in n-hexane. We used pentachloronitrobenzene (PCNB) (Accustandard Inc., New Haven, Connecticut, USA) as an internal standard (IS), respectively. An SPE device and C18 SPE cartridges (ENVI-18, 6 mL, 500 mg) were purchased from Supelco Co., Bellefonte, Pennsylvania, USA, and the methanol (pesticide grade) used for activation of the SPE column was purchased from Tedia Co. Inc., Fairfield, Ohio, USA. Granular anhydrous sodium sulfate (Sinopharm Chemical Reagent Co., Ltd, Shanghai, China) was heated at 650 °C in a furnace for 6 hours and stored in a sealed desiccator until used. All glassware was cleaned in an ultrasonic cleaner (KQ-500B, Kunshan Ultrasonic Instrument, Kunshan, China) and was heated at 400 °C for 6 h.

2.1.2. Sample collection and pretreatment

Fifteen surface water samples were collected from Lake Chaohu from August 8th to 9th, 2009. The distributions of the sampling sites are presented in Fig. 1. Sample sites A1, A2, A3 and A4 are located in eastern Lake Chaohu (EL), excluding the drinking-water source area (WS), whereas sites A6, B2, B3, B4, B5 and B7 are located in the drinking-water source area. Samples B6, C1, C2 and C4 were collected from three inflowing rivers (R), whereas D1 was collected from the center of western Lake Chaohu (CW). Twenty liters of water were collected from each sampling site. After shaking and mixing, an aliquot of one liter of each collected water sample was filtered through a 0.45- μm glass fiber filter (burned at 450 °C for 4 h) using a filtration

device consisting of a peristaltic pump (80EL005, Millipore Co., USA) and a filter plate with a diameter of 142 mm.

2.1.3. Sample extraction

The pretreated water samples were passed through octadecyl silane (ODS) C18 filler in the SPE cartridge, with the filler first being cleaned using 10 mL (5 times, 2 mL per time) of DCM, then activated by 6 mL of methyl alcohol (3 times, 2 mL per time), and finally washed using 10 mL of ultrapure water. During activation and ultrapure water washing, the liquid level should be tangent to the filler to keep the C18 wet. After enrichment of the target compound, 10 mL of DCM (2 times, 3 mL per time, 1 time, 4 mL per time) was used to elute the C18 SPE cartridge, which was jointed to an anhydrous sodium sulfate (5 g) cartridge. The extracts were concentrated to approximately 1 mL in an eggplant-shaped flask using a vacuum rotary evaporator (R-201, Shanghai Shen Sheng Technology Co., Ltd., Shanghai, China) at a temperature below 38 °C. PCNB was then added to the flask as an IS. The samples were sealed in vials and stored at -20 °C before analysis.

2.1.4. Sample analysis and quality assurance

Samples were analyzed using an Agilent gas chromatograph 6890 equipped with a Nickel 63 electron capture detector (μECD) and an HP-5 column (30 m \times 0.32 mm inside diameter, 0.25 μm film thickness). The samples were injected by auto-sampling at 50 °C in splitless mode, with a venting time of 0.75 min. The oven temperature was programmed to first increase from 50 °C to 150 °C at 10 °C min^{-1} and then to increase to 240 °C at 3 °C min^{-1} , where the temperature was maintained for 15 min. Nitrogen was used as both the carrier (1 mL min^{-1}) and makeup gas (60 mL min^{-1}). The injector and detector temperatures were 220 °C and 280 °C, respectively.

A mixed working standard with a concentration of 20 ng L^{-1} was used for six repeats of calibration. The recoveries of o, p'-DDT, p, p'-DDT, o, p'-DDD, p, p'-DDD, p, p'-DDE, p, p'-DDE, α -HCH, β -HCH, γ -HCH and δ -HCH by this method were 47.3%, 82.4%, 89.7%, 89.3%, 50.8%, 74.0%, 107.3%, 107.0%, 92.7% and 94.2%, respectively. Although the recoveries of o, p'-DDT and p, p'-DDE were not as good as other chemicals, which was also proved by other study (Sibali et al., 2009), it was a highly recommended method of accumulation and extract of pollutants in the wild because of its portability, easy operation, and more capacity of samples. The detection limits for the water samples were 0.5 ng L^{-1} for p, p'-DDT and o, p'-DDE, 0.3 ng L^{-1} for p, p'-DDD,

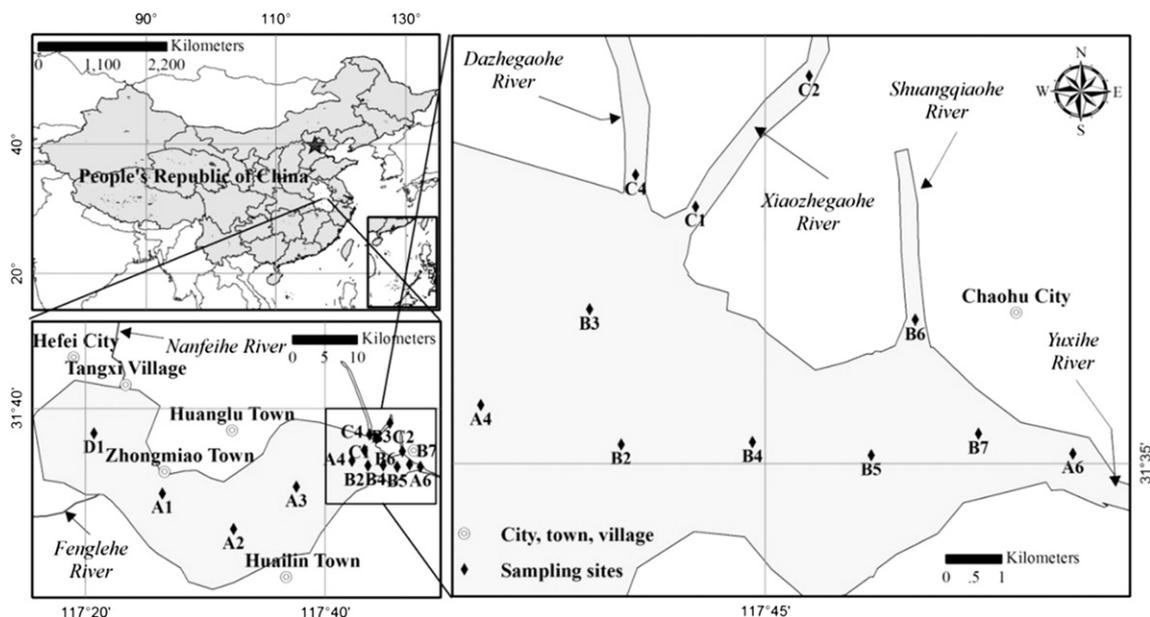


Fig. 1. Location of Lake Chaohu and water sampling sites.

0.1 ng L⁻¹ for p, p'-DDE, 0.05 ng L⁻¹ for α-HCH, γ-HCH, δ-HCH and o, p'-DDD and 0.01 ng L⁻¹ for β-HCH and o, p'-DDT. Two procedural blank samples were run to check for contamination by solvents and glassware. All samples were extracted and analyzed in duplicate.

2.2. Ecological Risk Assessment of DDTs and HCHs in Water

A species sensitivity distribution (SSD) was applied to evaluate the ecological risks of p,p'-DDT and γ-HCH for different species. An SSD can be used to describe the cumulative distribution of eco-toxicity data and exposure data (Posthuma et al., 2002; Wang et al., 2009). Toxicity data were collected from the EPA ECOTOX database (<http://cfpub.epa.gov/ecotox/>), using search criteria including the type of freshwater medium, endpoint of LC50, exposure duration of < 10 days and test location of the laboratory. The species considered in the distribution consisted of algae, vertebrates and invertebrates. The vertebrates included fish and amphibians, whereas the invertebrates were comprised of shrimp, crabs and other small crustaceans. According to the estimated trophic levels of fishes (data from FISHBASE, <http://www.fishbase.net/>), fishes were divided into three types: fishes II, fishes III and fishes IV. Fishes II, fishes III and fishes IV are composed of fishes for which the trophic level values were 2.0–2.9, 3.0–3.9 and 4.0–4.9, respectively. The acute and chronic ratio (ACR) was employed to convert data from acute data (LC50) to chronic data, and the ACR recommended by United States Environment Protection Agency (USEPA) was 10 (USEPA, 1991). The BurrI/OZ tool designed by Australia's Commonwealth Scientific and Industrial Research Organization (CSIRO) was employed to calculate relevant parameters of the SSD (Table 1) (Kurt-Karakus et al., 2006). The potentially affected fraction (PAF), which is a measure of the toxic risk probability of p,p'-DDT and γ-HCH for species could be obtained using the BurrIII (Eq. (1)), ReWeibull (Eq. (2)) and Repareto (Eq. (3)) equations, as follows:

$$PAF(x) = 1/[1 + (b/x^c)]^k \quad (1)$$

$$PAF(x) = \exp(-b/x^c) \quad (2)$$

$$PAF(x) = (x/b)^k \quad (3)$$

where x is contamination concentration (ng L⁻¹), and b , c , k are three parameters.

Table 1
Species sensitivity distribution parameters of species and study areas calculated by BurrI/OZ.

Species and areas	p, p'-DDT				γ-HCH			
	<i>m</i>	<i>b</i>	<i>c</i>	<i>k</i>	<i>n</i>	<i>b</i>	<i>c</i>	<i>K</i>
All species	456	21.00	0.55	7.69	453	1.40 × 10 ³	0.67	3.78
Algae	12	1.00 × 10 ⁷	–	0.72	13	9.55 × 10 ¹⁵	3.06	–
Vertebrates	364	83.51	0.65	–	370	2.79 × 10 ³	0.76	3.04
Invertebrates	107	0.60	0.48	18.97	70	94.14	0.57	–
Fishes	351	102.23	0.69	–	358	3.25 × 10 ³	0.80	2.76
Fishes II	63	215.73	0.72	–	104	6.38 × 10 ³	0.69	2.38
Fishes III	203	70.73	0.62	–	202	9.97 × 10 ³	0.99	1.54
Fishes IV	85	285.90	1.28	2.65	52	2.71 × 10 ⁸	2.41	–
Amphibians	13	2.19 × 10 ⁵	1.32	0.63	12	6.78 × 10 ⁵	4.44	0.45
Shrimps & crabs	81	7.10	0.56	6.67	48	630.63	0.86	–
Other crustaceans	26	25.44	0.46	–	22	1.47 × 10 ⁴	0.50	1.90
WB	15	3.92	2.01	–	14	0.91	2.45	0.54
WL	11	3.58	19.47	0.11	10	0.68	–	1.63
EL	4	3.36	3.48	–	3	0.65	–	0.84
WS	6	3.69	23.93	0.15	6	0.39	7.97	9.85
R	4	10.19	–	1.03	4	2.14	2.32	–

Notes: m and n are number of toxicity and exposure data of p, p'-DDT and γ-HCH, respectively. WB – all the water bodies including all the sampling sites, WL – the whole lake including all the lake sampling sites, EL – the eastern lake including A1, A2, A3 and A4, WS – the drinking water source area including A6, B2, B3, B4, B5 and B7, R – the inflow rivers including B6, C1, C2 and C4.

2.3. Health risk assessment of DDTs and HCHs in water

The chemicals' carcinogenicity is the endpoint of health risk assessment. According to various toxicity data, there are three main carcinogenic risk assessment methods: (1) unit risk calculation on basis of epidemiological investigation data (Bertoldi et al., 2012); (2) carcinogenic risk assessment according to animal toxicity experiment data (OECD, 1993); (3) carcinogenic risk assessment based on genotoxicity testing (Mason et al., 1990). The human epidemiological investigation data is the most effective toxicity data in the procedure of the carcinogen risk assessment, but the acquisition of these data is often very difficult. So the animal experimental data is the most direct data in the human carcinogenic risk assessment. However, animal experiments are more expensive. Genotoxicity method is relatively simple, rapid, efficient and economical method of evaluating mutagenicity of chemicals, but it cannot detect all carcinogens. To our knowledge, carcinogenic effects include not only damage of DNA, but also other carcinogenic mechanism. Therefore, the experimental data of animal toxicity is still the main data source of the human health risk assessment. To obtain human intake dose, animals' high doses should be extrapolated to low dose, and their low doses should be extrapolated to the human dose. The above methods can be used to illustrate the carcinogenic risk of chemicals and their potential hazards to human being. However, these methods are still based on chemicals' properties. To objectively reflect the health risk of pollutants to the local population, we should consider both toxicity data and exposure data. Chemicals' carcinogenic risk and non-carcinogenic risk assessment methodology recommended by the U.S. EPA is the one that combines toxicity data and exposure data. According to different pathways (air, drinking water, skin, etc.), different calculation methods are included to calculate the health risks of chemicals within one year (USEPA, 1989a). The risk level is different in different countries. U.S. EPA set 10⁻⁴ as the maximum acceptable level of risk, and environmental protection departments in Sweden, Netherlands and United Kingdom set 10⁻⁶ as their maximum acceptable level of risk (Ni et al., 2010).

In this paper, a health risk assessment (HRA) model proposed by the USEPA (1989a) was used to assess the risks of DDTs and HCHs for human health (Hamidin et al., 2008). The carcinogenic and non-carcinogenic risks of α-HCH, β-HCH, γ-HCH, total HCH, p, p'-DDT, p, p'-DDE, p, p'-DDD and total DDT for human health were evaluated.

Noncarcinogenic risk (HI) equation is as follows:

$$HI = E/RfD \quad (4)$$

and carcinogenic risk (R) is as follows:

$$R = SF \cdot E \quad R < 0.01 \\ R = 1 - \exp(-SF \cdot E) \quad R \geq 0.01 \quad (5)$$

where RfD is the reference dose (mg kg⁻¹ day⁻¹); SF is the slope factor ([mg kg⁻¹ day⁻¹]⁻¹); and E is the exposure level (or intake) (mg kg⁻¹ day⁻¹), which was calculated using two exposure approaches as follows:

$$E_1 = (C \cdot IR_w \cdot EF \cdot ED)/BW/AT \quad (6)$$

$$E_2 = (6\tau \cdot TE/\pi)^{0.5} \cdot (C \cdot k \cdot A_{sb} \cdot EF \cdot FE \cdot ED)/500/BW/AT/f \quad (7)$$

where E_1 is exposure to water for drinking, and E_2 is exposure to water for bathing. C is the concentration of HCHs and DDTs in the water (mg L⁻¹); IR_w is the water intake rate (a value of 2 L day⁻¹ recommended by USEPA); EF is the exposure frequency (a value of 365 days year⁻¹ was employed because of the daily need of drinking water); ED is the exposure duration (a value of 30 years recommended by USEPA); BW is average of body weight (60 kg for Chinese people); AT is average time (70 years × 365 days year⁻¹); the non-carcinogenic

equation employs $ED \times 365 \text{ days year}^{-1}$; A_{sb} is body surface area ($16,600 \text{ cm}^2$); FE is bathing frequency ($0.3 \text{ times day}^{-1}$); TE is bathing time (0.4 h); f is the intestinal absorption ratio (1); k is a skin permeability parameter (0.001 cm h^{-1}); and τ is lag time for each pollutant in the body (assuming a value of 1 h) (Wan et al., 2009). The SF values of α -HCH, β -HCH, γ -HCH, total HCH, p, p'-DDT, p, p'-DDE, p, p'-DDD and total DDT were 6.3, 1.8, 1.3, 0.35, 0.34, 0.34, 0.24 and 0.35, respectively. The RfD values of γ -HCH and total HCH, p, p'-DDT and total DDT were 0.0004, 0.0004, 0.0005 and 0.0005, respectively (USEPA, 1989b, 2010).

3. Results and discussion

3.1. The concentrations of DDT and HCH in the water

On the basis of a Kolmogorov–Smirnov test (S–K test) of the directly-measured and log-transformed data, a log-normal distribution pattern was identified for α -HCH, β -HCH and γ -HCH, whereas a normal distribution pattern was found for DDTs and δ -HCH. Due to these distribution patterns, the median and range were employed to describe the statistical character of the residue levels. It was reported that the residues of total DDT and total HCH in the surface water from Lake Chaohu were 16.57 ng L^{-1} (1.93 – 53.46 ng L^{-1}) and 5.56 ng L^{-1} (1.19 – 64.01 ng L^{-1}) in December, 2008 (Zhang et al., 2010). Our measurements (Table 2) showed that the residues of total DDT and total HCH were 9.31 ng L^{-1} (1.52 – 21.79 ng L^{-1}) and 2.03 ng L^{-1} (1.58 – 31.66 ng L^{-1}), respectively, and the concentrations of total DDT and total HCH in summer, 2009 were half those found in winter, 2008. Degradation of DDTs and HCHs by oxidation, hydrolyzation, photolysis and microorganisms might be the main reason for this (Devipriya and Yesodharan, 2005; Motosugi and Soda, 1983). However, the water level might be another significant for this finding, as the average monthly water levels in December, 2008 and August, 2009 were 8.96 m and 9.28 m, respectively. The rise of the water level resulted in a $2.26 \times 10^8 \text{ m}^3$ increase of lake water storage, which is 9.3% greater than the water storage in December, 2008. There are some other reasons should not be neglected, for instance, (1) when the climate condition or the physical and chemical conditions of the lake mediums varies, the velocity of the sediment releasing the DDT into water also changes. (2) Quantity of water inflowing into the lake changes with seasons, which may lead to the different content.

The residues of p,p'-DDT and o,p'-DDT were 2.86 ng L^{-1} (1.11 – 10.19 ng L^{-1}) and 1.95 ng L^{-1} (not detected (ND)– 4.13 ng L^{-1}), respectively (Table 2); the concentrations of p,p'-DDD and o,p'-DDD were 1.40 ng L^{-1} (ND– 2.99 ng L^{-1}) and 2.35 ng L^{-1} (ND– 3.45 ng L^{-1}), respectively; DDE was less than DDT and DDD; and the concentrations of p,p'-DDE and o,p'-DDE were 0.61 ng L^{-1} (ND– 1.62 ng L^{-1}) and

1.15 ng L^{-1} (ND– 3.58 ng L^{-1}), respectively. Among the four isomers of HCHs (Table 2), the β -HCH concentration was the highest, with a value of 0.83 ng L^{-1} (0.64 – 17.78 ng L^{-1}), and the levels of α -HCH, γ -HCH and δ -HCH were 0.33 ng L^{-1} (0.22 – 6.86 ng L^{-1}), 0.60 ng L^{-1} (ND– 6.84 ng L^{-1}) and 0.28 ng L^{-1} (0.13 – 0.34 ng L^{-1}), respectively.

The total DDT and total HCH in the different lake areas is shown in Table 2. The average concentrations of total DDT and total HCH for the inflowing rivers were $11.62 \pm 8.52 \text{ ng L}^{-1}$ and $10.14 \pm 14.35 \text{ ng L}^{-1}$, respectively, whereas those for the lake area were $9.39 \pm 2.83 \text{ ng L}^{-1}$ and $2.00 \pm 0.31 \text{ ng L}^{-1}$. The concentrations of total DDT and total HCH in the center of the western lake were 7.61 ng L^{-1} and 2.25 ng L^{-1} , respectively, which was much lower than the results reported by Zhang et al. (2010). The average concentrations of total DDT and total HCH in the eastern lake were $7.78 \pm 2.45 \text{ ng L}^{-1}$ and $1.92 \pm 0.43 \text{ ng L}^{-1}$, respectively, whereas those in the eastern water source were $10.75 \pm 2.74 \text{ ng L}^{-1}$ and $1.95 \pm 0.07 \text{ ng L}^{-1}$. The results showed a high-east and low-west distribution pattern for total DDT and a high-west and low-east pattern for total HCH.

The reasons might be that: (1) although agricultural use of DDT has been forbidden since 1983, DDT in China is not completely banned. DDT, as the auxiliary material of a marine anti-fouling paint is still in use. 10000 tons of anti-fouling paints are used in fishing vessels each year in China, and about 250 tons of DDT is used in anti-fouling paints. From 1950s to 2005, approximately 1.1 million tons of DDT was used to produce marine anti-fouling paints. In Lake Chaohu, there are 3500 fishing vessels, 2500 of which anchor in the eastern lakes, especially at Shuangqiaohe River, Zhegaohe River, and Chaohu City Dock. DDT's distribution might result from ships' distribution. (2) Farmland in Lake Chaohu Basin mainly distributes on the shore of southwestern lake. Rivers, flowing through this area, carry 60% of total volume of water into the Lake Chaohu. More usage of HCH in agriculture and more inflow of HCH to the lake might be the main reason that the content of HCH in the western part of the lake is higher than that in the eastern part of the lake.

3.2. The composition of DDT and HCH in the water

The composition of DDT, its metabolites and HCH isomers (Fig. 2) determined in this study indicated that the fraction of p,p'-DDT and o,p'-DDT in the lake area (40–44%) was lower than that in the inflowing rivers (54%), whereas the fractions of DDT metabolites were higher than in the inflowing rivers. The proportions of DDD and DDE in the eastern water source area were both higher than in other areas. β -HCH in the lake area was the predominant HCH isomer present, exhibiting a proportion of 41–55% of the total HCH isomers present. The proportion of γ -HCH in the lake area (16–29%) was lower than in the inflowing rivers (37%), whereas the δ -HCH proportion in the

Table 2
Residues of DDT with its metabolites and HCH isomers in Lake Chaohu and its inflow rivers, ng L^{-1} (August, 2009).

	WB			EL		WS		R		CW	
	Median	Min	Max	Mean	SD	Mean	SD	Mean	SD	Mean	SD
p,p'-DDT	2.86	1.11	10.19	1.82	0.85	2.88	0.74	5.11	3.89	2.06	
p,p'-DDE	0.61	ND	1.62	0.68	0.34	0.62	0.33	0.91	0.68	ND	
p,p'-DDD	1.40	ND	2.99	1.26	0.66	1.65	0.61	1.67	1.25	1.62	
o,p'-DDT	1.95	ND	4.13	2.12	1.21	2.16	0.76	2.70	1.77	1.29	
o,p'-DDE	1.15	ND	3.58	2.12	0.83	1.61	1.17	0.80	0.49	1.06	
o,p'-DDD	2.35	ND	3.45	1.52	0.78	3.06	1.62	2.62	1.40	1.57	
DDTs	9.31	1.52	21.79	7.78	2.45	10.76	2.73	11.62	8.52	7.61	
α -HCH	0.33	0.22	6.86	0.28	0.06	0.35	0.11	2.04	3.21	0.57	
β -HCH	0.83	0.64	17.78	0.94	0.31	0.79	0.03	5.25	8.36	1.38	
γ -HCH	0.60	ND	6.84	0.56	0.29	0.56	0.09	2.64	2.80	0.42	
δ -HCH	0.28	0.13	0.34	0.28	0.02	0.26	0.07	0.21	0.09	0.19	
HCHs	2.03	1.58	31.66	1.92	0.43	1.95	0.07	10.14	14.35	2.55	

Notes: SD – standard deviation, ND – not detected. CW – the centre of the western lake. The WB, EL, WS and R are the same with those in Table 1.

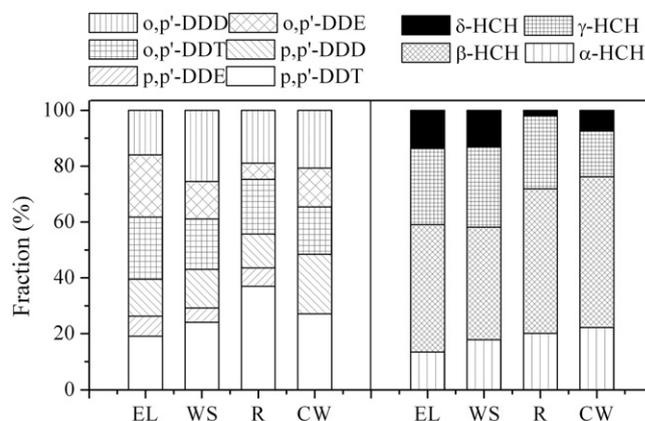


Fig. 2. Compositions of DDT with its metabolites and HCH isomers in various water bodies of Lake Chaohu. The meaning of EL, WS, R and CW are the same with those in Table 1.

lake area (7–14%) was higher than in the inflowing rivers (4%). This may suggest that γ -HCH in the lake area was derived from the inflowing rivers.

The DDT and HCH residues in Chinese lakes are generally less than in other developing countries, such as Lake Burullus (DDTs, $208.5 \pm 159.7 \text{ ng L}^{-1}$, HCHs, $467.0 \pm 277.8 \text{ ng L}^{-1}$) in Egypt (Said et al., 2008), Lake Paranoa (DDTs, $1\text{--}52 \text{ ng L}^{-1}$) in Brazil (Caldas et al., 1999) and Lake Hanoi (DDTs, $31.7 \pm 60.4 \text{ ng L}^{-1}$, HCHs, $5.07 \pm 6.88 \text{ ng L}^{-1}$) in Vietnam (Hung and Thiemann, 2002). However, the concentrations of DDTs and HCHs in Lake Chaohu were still higher than those of other Chinese lakes, such as Lake Baiyangdian (DDTs, $2.1 \pm 0.8 \text{ ng L}^{-1}$, HCHs, $2.4 \pm 0.6 \text{ ng L}^{-1}$) (Hu et al., 2010), Lake Taihu (DDTs, $> 1.02 \text{ ng L}^{-1}$, HCHs, $> 0.04 \text{ ng L}^{-1}$) (Na et al., 2006), Lake Co Ngoin (DDTs, 1.81 ng L^{-1} , HCHs, 0.30 ng L^{-1}) and Lake Yamzho Yumeo (DDTs, 3.83 ng L^{-1} , HCHs, 0.27 ng L^{-1}) (Zhang et al., 2003).

3.3. Source identification of DDTs and HCHs in the water

DDT sources can be identified on the basis of the composition of DDTs and their metabolic products. DDTs produced from technology-manufacturing sources contain 15% o,p'-DDT and 85% p,p'-DDT, whereas 1 kg dicofol contains approximately 114 grams of o,p'-DDT and 17 grams of p,p'-DDT. Therefore, the o,p'-DDT/p,p'-DDT ratio for technical DDT is approximately 0.2, and that for dicofol is approximately 7. Thus, in the case of an o,p'-DDT/p,p'-DDT ≥ 7 , dicofol can be assumed to be the main DDT source, whereas an o,p'-DDT/p,p'-DDT ratio of approximately 0.2 indicates a technical DDT source. The portion of DDT metabolic products is indicative of new inputs or historical usage. DDT (including o,p'-DDT and p,p'-DDT) can be degraded to DDD (o,p'-DDD and p,p'-DDD) and DDE (o,p'-DDE and p,p'-DDE). If the DDT/(DDD + DDE) ratio is greater than 1, it indicates that the DDT in a given sample represents a new input, whereas if this ratio is not greater than 1, the DDT is due to historical input. (Hitch and Day, 1992). A DDD/DDE > 1 implies that DDD is the main metabolite present, which is indicative of anaerobic metabolic conditions; whereas a DDD/DDE < 1 indicates that DDE is the main metabolite present, which is indicative of aerobic metabolic conditions (Hitch and Day, 1992). In the present study, the o,p'-DDT/p,p'-DDT ratio was found to range from 0.40 to 2.18 (Table 3), indicating that technical DDT was the main DDT source. The DDT/(DDD + DDE) ratios for all river samples (B6, C1, C2 and C4), B2 and B3 in the eastern water source area, and A2 in eastern lake area were greater than 1, suggesting that there was recent input of pesticides containing DDT. The DDT/(DDD + DDE) ratios for other sites were less than 1, implying historical inputs of DDT as the main DDT source. The sites D1, in the center of the western lake area, A1 and B4, in the eastern lake area and B5 and B7, in the eastern water source area were under anaerobic conditions, as indicated by their ratios of DDD/DDE > 1 . The sites A3, in the eastern lake area, and A6, in the eastern water source area, were under aerobic conditions, as indicated by their ratios of DDD/DDE < 1 .

Technical HCH consists of 60–70% α -HCH, 5–12% β -HCH and 10–15% γ -HCH, with a α -/ γ -HCH ratio of approximately 4–7 and a β -/($\alpha + \gamma$)-HCH ratio of approximately 0.06–0.17. For lindane, which includes γ -HCH as its major component, the α -/ γ -HCH ratio is less than 0.1, and the β -/($\alpha + \gamma$)-HCH ratio is less than 0.06. Because of the high vapor pressure, α -HCH and γ -HCH are the main isomers in the gaseous phase and could be transported over long distances in air. Therefore, the α -/ γ -HCH ratio in the gaseous phase is greater than that for industrial sources of HCH, i.e., α -/ γ -HCH > 7 , which can be used to identify the atmospheric source of HCHs (Iwata et al., 1993, 1995). Because it exhibits more stable chemical and physical properties than other HCH isomers, β -HCH is the dominant isomer in water environments. Therefore, the β -/($\alpha + \gamma$)-HCH ratio can be used to identify whether a water environment is polluted by historical inputs of HCHs. When the β -/($\alpha + \gamma$)-HCH ratio is less than 0.06, it indicates that new inputs of lindane or technical HCH, or dry-wet α -HCH

Table 3

Ratios used in source identification of DDT with its metabolites and HCH isomers in Lake Chaohu and its inflow rivers.

	o,p'/p, p'-DDT	DDT/ (DDD + DDE)	DDD/DDE	α -/ γ -HCH	β -/($\alpha + \gamma$)-HCH
A1	–	0.60	1.36	–	4.58
A2	0.87	1.29	1.69	0.52	1.34
A3	0.94	0.41	0.79	0.67	0.80
A4	2.18	0.99	0.38	0.39	0.92
A6	0.61	0.50	0.91	0.58	0.99
B2	1.15	1.21	12.64	0.41	0.87
B3	0.95	2.07	0.60	0.46	0.93
B4	0.42	0.83	2.86	1.11	0.82
B5	1.05	0.91	1.65	0.96	0.82
B6	–	2.80	0.00	1.00	1.30
B7	0.55	0.74	3.19	0.43	0.81
C1	0.40	1.04	1.91	0.31	0.57
C2	0.45	1.49	2.61	0.30	0.76
C4	0.41	1.92	3.61	0.45	0.57
CW	0.63	0.79	3.01	1.36	1.39
WB	0.63	0.99	1.39	0.49	0.87
	(0.4–2.18)	(0.41–2.8)	(0–12.64)	(0.3–1.36)	(0.57–4.58)
EL	1.33 ± 0.74	0.82 ± 0.40	1.06 ± 0.58	0.52 ± 0.14	1.91 ± 1.80
WS	0.79 ± 0.30	1.04 ± 0.55	3.64 ± 4.53	0.66 ± 0.30	0.87 ± 0.07
R	0.42 ± 0.03	1.81 ± 0.75	2.03 ± 1.52	0.52 ± 0.33	0.80 ± 0.34

Notes: CW is the same with that in Table 2. WB, EL, WS and R are the same with those in Table 1. The value for WB is shown as median (minimum-maximum), and the value for EL WS and R are shown as mean \pm standard deviation.

deposition exist. However, a higher β -/($\alpha + \gamma$)-HCH ratio indicates that there has been a large amount of historical lindane or technical HCH input (Willett et al., 1998). In present study, the α -/ γ -HCH ratios of all the of samples were less than 1.5, whereas the β -/($\alpha + \gamma$)-HCH ratios of all samples were greater than 0.4, indicating historical usage of lindane (Table 3).

In different sampling sites, there are several factors such as climate condition, water depth, sediment resuspension, microorganism species and activity and runoff from the rivers influencing the water mixing effect and pollutants' distribution. Although the parameters in different sampling sites are different, we still can obtain statistic information of pollutant source for a specific area if there are enough sampling sites.

3.4. Ecological risk of p, p'-DDT and γ -HCH in the Water

The potentially affected fractions (PAFs) of p,p'-DDT and γ -HCH for species in different lake areas were calculated and are presented in Table 4. The ecological risk of p,p'-DDT for invertebrates (e.g., shrimps & crabs, 0.07–0.52%) was much greater than for vertebrates (e.g., fishes, 3.3×10^{-28} – 3.1×10^{-13} %), but among all of the investigated vertebrates, the risk for amphibians (0.006–0.014%) was much greater than that for other vertebrates. The ecological risk probability of p,p'-DDT for all of the species in the distribution varied from 5.5×10^{-4} % to 0.014%. The species sensitivity to p, p'-DDT was ranked as: shrimps & crabs $>$ amphibians $>$ algae $>$ other crustaceans $>$ fishes IV $>$ fishes III $>$ fishes II. We found that the risk of p,p'-DDT to fishes increased with increasing trophic level. The risk of p,p'-DDT to water bodies was ranked as: inflowing rivers (R) $>$ drinking-water source area (WS) $>$ core of the western lake (CW) $>$ eastern lake (EL). Contrary to our findings for p,p'-DDT, the ecological risk of γ -HCH for invertebrates (e.g., shrimps & crabs, $0.0\text{--}4.4 \times 10^{-117}$ %) was much smaller than that for vertebrates (such as fish, 2.5×10^{-7} – 1.5×10^{-5} %), whereas the risk for other crustaceans (0.004–0.025%) was the highest among all the investigated species. The species sensitivity to γ -HCH was ranked as: other crustaceans $>$ fishes III $>$ fishes II $>$ amphibians $>$ shrimps & crabs $>$ fishes IV = algae. We also found that the risk of γ -HCH to fishes at the third trophic level was greater than the second trophic level. The ecological risk probability of γ -HCH for all the species varied from

Table 4
PAF and MOS10 of p,p'-DDT's and γ -HCH's in different areas of Lake Chaohu.

	p, p'-DDT						γ -HCH					
	WB	WL	EL	WS	R	CW	WB	WL	EL	WS	R	CW
<i>PAF (%)</i>												
All species	3.2×10^{-3}	1.4×10^{-3}	5.5×10^{-4}	2.5×10^{-3}	1.4×10^{-2}	8.3×10^{-4}	1.3×10^{-6}	1.9×10^{-7}	2.6×10^{-7}	2.5×10^{-7}	1.2×10^{-5}	1.2×10^{-7}
Algae	2.0×10^{-3}	1.7×10^{-3}	1.4×10^{-3}	1.9×10^{-3}	2.9×10^{-3}	1.5×10^{-3}	0.0	0.0	0.0	0.0	0.0	0.0
Vertebrates	5.1×10^{-16}	3.6×10^{-19}	2.3×10^{-23}	5.6×10^{-17}	2.5×10^{-11}	2.0×10^{-21}	1.4×10^{-6}	2.3×10^{-7}	3.1×10^{-7}	3.0×10^{-7}	1.1×10^{-5}	1.6×10^{-7}
Invertebrates	8.4×10^{-2}	3.9×10^{-2}	1.5×10^{-2}	6.6×10^{-2}	3.0×10^{-1}	2.3×10^{-2}	4.1×10^{-38}	1.6×10^{-59}	3.0×10^{-55}	1.1×10^{-55}	2.7×10^{-22}	5.4×10^{-66}
Fishes	5.4×10^{-19}	6.2×10^{-23}	3.3×10^{-28}	3.4×10^{-20}	3.1×10^{-13}	9.1×10^{-26}	2.0×10^{-6}	3.8×10^{-7}	5.0×10^{-7}	4.8×10^{-7}	1.5×10^{-5}	2.5×10^{-7}
Fishes II	8.9×10^{-40}	3.2×10^{-48}	1.3×10^{-59}	2.4×10^{-42}	1.3×10^{-27}	2.5×10^{-54}	6.0×10^{-5}	1.7×10^{-5}	2.1×10^{-5}	2.0×10^{-5}	2.6×10^{-5}	1.3×10^{-5}
Fishes III	9.0×10^{-14}	2.1×10^{-16}	6.9×10^{-20}	1.4×10^{-14}	7.9×10^{-10}	2.7×10^{-18}	9.4×10^{-5}	2.9×10^{-5}	3.6×10^{-5}	3.5×10^{-5}	3.7×10^{-4}	2.2×10^{-5}
Fishes IV	2.2×10^{-5}	9.3×10^{-6}	3.5×10^{-6}	1.7×10^{-5}	1.2×10^{-4}	5.4×10^{-6}	0.0	0.0	0.0	0.0	0.0	0.0
Amphibians	9.5×10^{-3}	7.6×10^{-3}	6.0×10^{-3}	8.8×10^{-3}	1.4×10^{-2}	6.7×10^{-3}	2.5×10^{-10}	5.4×10^{-11}	7.0×10^{-11}	6.8×10^{-11}	1.5×10^{-9}	3.8×10^{-11}
Shrimps and crabs	1.8×10^{-1}	1.0×10^{-1}	4.9×10^{-2}	1.5×10^{-1}	5.2×10^{-1}	6.8×10^{-2}	2.1×10^{-257}	0.0	0.0	0.0	4.4×10^{-117}	0.0
Other crustaceans	2.9×10^{-5}	4.3×10^{-6}	4.0×10^{-7}	1.6×10^{-5}	5.9×10^{-4}	1.2×10^{-6}	1.1×10^{-2}	5.2×10^{-3}	5.9×10^{-3}	5.8×10^{-3}	2.5×10^{-2}	4.4×10^{-3}
<i>MOS10</i>												
All species	23.6	40.8	52.7	38.7	14.6		1.1×10^3	2.7×10^3	3.0×10^3	2.6×10^3	0.5×10^3	
Algae	4.1×10^5	7.2×10^5	9.3×10^5	6.8×10^5	2.6×10^5		7.5×10^4	1.9×10^5	2.1×10^5	1.9×10^5	3.5×10^4	
Vertebrates	42.1	73.0	94.3	69.3	26.1		1.4×10^3	3.6×10^3	3.9×10^3	3.4×10^3	0.6×10^3	
Invertebrates	7.1	12.3	15.9	11.7	4.4		0.4×10^3	1.0×10^3	1.1×10^3	1.0×10^3	0.2×10^3	
Fishes	41.6	72.1	93.1	68.4	25.8		1.4×10^3	3.5×10^3	3.8×10^3	3.4×10^3	0.6×10^3	
Fishes II	89.1	154.2	199.2	146.4	55.3		1.9×10^3	4.8×10^3	5.2×10^3	4.6×10^3	0.9×10^3	
Fishes III	39.9	69.1	89.2	65.5	24.7		1.7×10^3	4.3×10^3	4.6×10^3	4.1×10^3	0.8×10^3	
Fishes IV	36.7	63.6	82.2	60.4	22.8		1.3×10^3	3.4×10^3	3.7×10^3	3.3×10^3	0.6×10^3	
Amphibians	2.3×10^3	4.0×10^3	5.2×10^3	3.8×10^3	1.4×10^3		1.3×10^5	3.2×10^5	3.5×10^5	3.1×10^5	5.9×10^5	
Shrimps and crabs	5.8	10.0	12.9	9.5	3.6		0.4×10^3	1.0×10^3	1.0×10^3	1.0×10^3	0.2×10^3	
Other crustaceans	31.0	53.7	69.3	50.9	19.2		1.6×10^3	4.0×10^3	4.4×10^3	3.9×10^3	0.7×10^3	

Notes: WB, WL, EL, WS and R are the same with those in Table 1, and CW is the same with that in Table 2.

$1.2 \times 10^{-7}\%$ to $1.2 \times 10^{-5}\%$. The risk of γ -HCH to the water bodies was ranked as: R > EL > WS > CW.

Based on the SSD model, the margin of safety (MOS10) was used to illustrate the risk of contaminants to aquatic organisms in Lake Chaohu (Zolezzi et al., 2005). This was calculated by dividing the 10th percentile for SSD of eco-toxicity data by the 90th percentile for the cumulative distribution of exposure data. When the study area included all of the investigated water bodies, the MOS10 of p,p'-DDT for shrimps and crabs was below 1, indicating that p,p'-DDT posed a potential ecological risk to these species (Table 4). The orders of magnitude of the MOS10 of algae and amphibians were three and five. This suggested that the risk to these groups could be neglected. The risk of p,p'-DDT for invertebrates (MOS10, 4.4–15.9) was higher than for vertebrates (MOS10, 26.1–94.3). We also found that fishes at higher trophic levels exhibited a smaller MOS10. The MOS10 for species in rivers was lower than in lakes, indicating that the aquatic species in rivers were in danger. The MOS10 of γ -HCH for all of the species was much greater than 1 in the various water bodies investigated, meaning that γ -HCH did not pose a potential ecological risk for the species there. Thus, Lake Chaohu was found to be facing a potential threat from p, p'-DDT, whereas γ -HCH was not a potential pollutant in the Lake Chaohu ecosystem.

3.5. Human health risk of p, p'-DDT and γ -HCH in the Water

As we all know, supplying clean drinking water is one of the most important ecological service function in fresh water ecosystem. The proposed method is widely used to assess the carcinogenic risk or noncarcinogenic risk for human health when drinking or bathing (Cothorn et al., 1986; Ni et al., 2010; Zeng et al., 1998). The high risk indicates that the water is being contaminated seriously and the ecosystem is losing her ecological function. Eastern part of Lake Chaohu is mainly used to supply to citizens in Chaohu City for drinking and bathing. Therefore, it is necessary to evaluate whether this area is suitable as drinking water source.

The results of our human health risk assessment using the USEPA (1989a) method (Eqs. (4)–(7)) are shown in Table 5. The data in the table illustrate the human health risk of DDTs and HCHs for the residents using water from the drinking water source in eastern Lake Chaohu for drinking or bathing. According to the cancer statistics and risk management and remediation goals, the acceptable carcinogenic risk of chemical pollutants ranges from one per ten million (10^{-7}) to one per ten thousand (10^{-4}), and the acceptable noncarcinogenic hazard index is 1.0 (USEPA, 1989a). This indicates that water can be safely used when carcinogenic risks are below 10^{-7} and noncarcinogenic hazard indices are less than 1.0. In the present study, both the carcinogenic risks and noncarcinogenic hazard indices of DDTs and HCHs were much lower than the threshold values, indicating that the water from the drinking water source in Lake Chaohu was very safe for drinking and bathing. The levels of DDT and its metabolic products, as well as of the different isomers of HCHs detected in samples from Lake Chaohu did not pose carcinogenic risks, and the

Table 5
Human health risk of DDTs and HCHs pesticides in Lake Chaohu.

Substances	Drinking	Bathing
<i>Carcinogenic risk (10^{-9})</i>		
p,p'-DDT	11.9	0.0518
p,p'-DDD	0.07	0.000305
p,p'-DDE	1.73	0.00753
DDTs	39.6	0.172
α -HCH	46.4	0.202
β -HCH	31.5	0.137
γ -HCH	14.7	0.0639
HCHs	14.5	0.0631
<i>Non-carcinogenic risk (10^{-6})</i>		
p,p'-DDT	16.3	0.0711
DDTs	52.8	0.23
γ -HCH	8.78	0.0382
HCHs	32.2	0.14

risks of p,p'-DDT (1.19×10^{-8}) and α -HCH (4.64×10^{-8}) were much greater than those of the other investigated substances.

4. Conclusion

The concentrations of DDTs and HCHs in Lake Chaohu were lower than those in other developing countries, but higher than those in other Chinese lakes. HCHs and DDTs in Lake Chaohu originated from the historical usage of lindane and technical DDT, respectively. The residue of p,p'-DDT in the lake water posed high potential ecological risks and little human-health risks, while γ -HCH did little risks to both the lake ecosystem and human health.

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