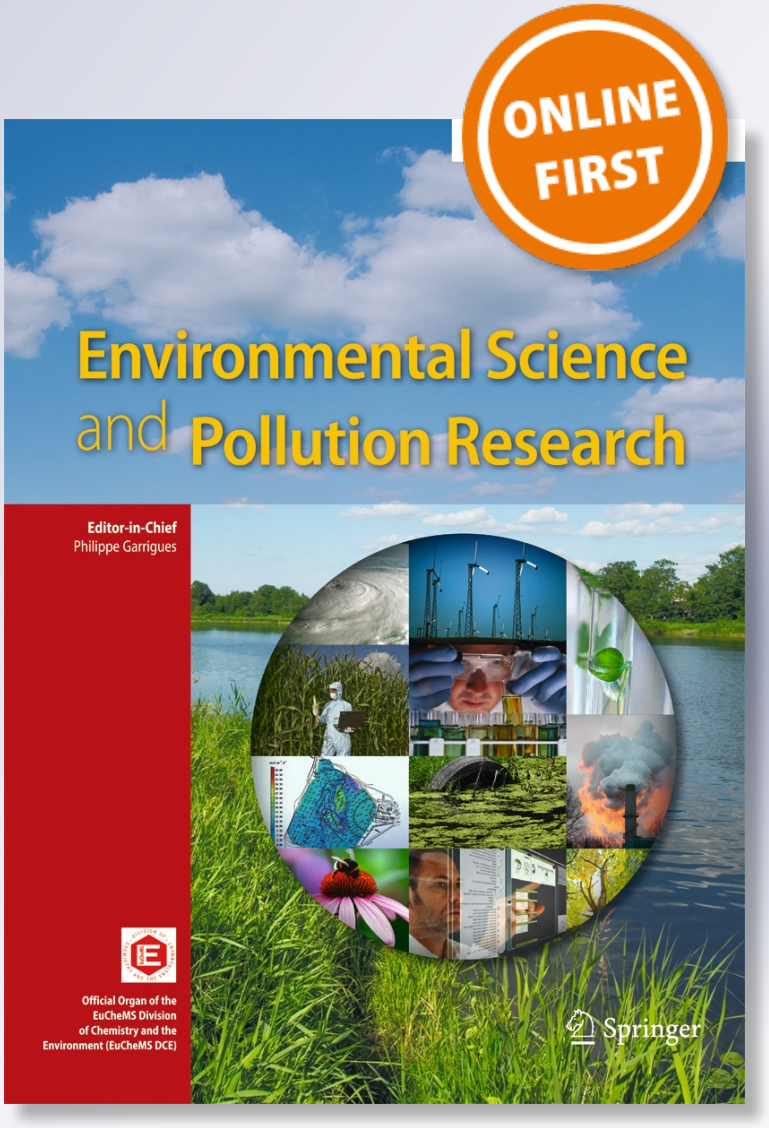
Residues of organochlorine pesticides in surface water of a megacity in central China: seasonal-spatial distribution and fate in Wuhan

Lili Cui, Liangfu Wei & Jun Wang

Environmental Science and Pollution Research

ISSN 0944-1344

Environ Sci Pollut Res
DOI 10.1007/s11356-016-7956-7



Springer

Environ Sci Pollut Res
DOI 10.1007/s11356-016-7956-7

RESEARCH ARTICLE

Residues of organochlorine pesticides in surface water of a megacity in central China: seasonal-spatial distribution and fate in Wuhan

Lili Cui[1,2] · **Liangfu Wei**[1,2] · **Jun Wang**[1,3]

Received: 7 June 2016 / Accepted: 20 October 2016

Abstract Surface water quality closely correlating with human health suffered increasing organochlorine pesticide (OCP) pollution due to the intensive anthropogenic activities in megacities. In the present study, 112 water samples collected from 14 lakes and 11 drinking water source sites in Wuhan were detected for the residues of OCPs in November 2013 and July 2014, respectively. The ΣOCPs ranged from 5.61 to 13.62 ng L^{-1} in summer with the maximum value in Yezhi Lake and 3.18 to 7.73 ng L^{-1} in winter with the highest concentration in Yandong Lake. Except dichlorodiphenyltrichloroethanes (DDTs), OCP concentrations in summer were significantly higher than those in winter mostly due to the non-point source pollution including land runoff in summer. Source apportionment of hexachlorocyclohexanes (HCHs) and DDTs revealed the historical use of technical HCH and lindane and the new input of DDT, respectively. The spatial distribution of OCPs was not uniform in the surface water of Wuhan because of the significant influence of land development and fishery. The risk assessments showed the heptachlor, and heptachlor epoxide in most sampling sites exceeded the threshold set by the European Union, indicating the possible adverse effects for aquatic lives. Negligible non-carcinogenic risks for drinking and bathing as well as carcinogenic risks for bathing were found in the surface water. However, the total carcinogenic risks of all OCPs (∑Rs) caused by drinking in summer were higher than the safe level of 10^{-7} in all sampling sites. It was implied that the surface water in Wuhan was not safe for directly drinking without effective purification.

Keywords Organochlorine pesticides (OCPs) · Lakes · Drinking water sources · Seasonal variation · Non-point source pollution · Risk assessment

Responsible Editor: Hongwen Sun

Electronic supplementary material The online version of this article (doi:10.1007/s11356-016-7956-7) contains supplementary material, which is available to authorized users.

✉ Jun Wang
wangjun@wbgcas.cn

[1] Key Laboratory of Aquatic Botany and Watershed Ecology, Wuhan Botanical Garden, Chinese Academy of Sciences, Wuhan 430074, China

[2] University of Chinese Academy of Sciences, Beijing 100049, China

[3] Sino-Africa Joint Research Center, Chinese Academy of Sciences, Wuhan 430074, China

Introduction

Notorious for the environmental persistence, bioaccumulation in fatty tissues, long-range transport capacity, and toxicity for organisms, organochlorine pesticides (OCPs) such as hexachlorocyclohexanes (HCHs) and dichlorodiphenyltrichloroethanes (DDTs) have been prohibited successively worldwide since the 1970s (Cui et al. 2015; Huang et al. 2013; Oliveira et al. 2016). But until then, the production of HCHs and DDTs came to 14,800,000 and 2,000,000 t, respectively, for the usage of pest control and public health campaigns (Li et al. 2012). In China, large amounts of OCPs have been used and manufactured in order to obtain high yield for sustaining overpopulation. The ban of HCHs and DDTs whose respective consumption accumulated to 4,900,000 and 400,000 t occurred in 1983 (Li et al. 2014). But there is acceptable use—the production of DDT as intermediate continues in small scale due to the manufacture demand of dicofol which is mainly applied in apple and cotton cultivation (Die et al. 2015; Liu et al. 2015a, b; Zhang et al. 2002). Lindane (almost pure γ-HCH) was not weeded out until 2014 with

Published online: 31 October 2016

the specific exemption of medical application for the control of human head lice and scabies (Standing Committee of the National People's Congress 2013). Vast OCP residues were left, and high levels have been detected in various environmental compartments especially the aquatic environment (Zhi et al. 2015; Liu et al. 2013; Yang et al. 2014; Zhao et al. 2010). Surface water is vulnerable to the OCP contamination. Over decades of forbidden use of OCPs and significant advances in controlling point-source pollution mostly triggered by the centralized employment of OCPs in farming, the non-point source pollution such as land runoff was dominated in surface water (Calhoun 2005). And other than that, the atmospheric transportation of OCPs from other regions contributes more or less for the surface water contamination.

Proverbially, the water resources in China are insufficient, while the shortage has been even serious as a consequence of water pollution brought by rapid economic growth and urbanization in recent decades (Hu and Cheng 2013). It is surveyed that more than two thirds of Chinese cities suffer inadequate water supply (Zhang et al. 2014). And particularly, the megacity whose population in the central urban area is 5,000,000–10,000,000 is experiencing much severer water deterioration and shortages (State Council of the People's Republic of China 2014). The toxic pollutants such as OCPs in surface water have been well documented nationwide and pose a tremendous challenge for the urban water conservation.

OCPs in megacities originated from many sources. They were frequently applied around the residential and commercial areas for pest elimination and intensively used in landscaping projects and golf course management in the 1970s, but the application amount was hard to estimated (LeVeen and Willey 1983). Additionally, the urban expansion and land development boom brought about serious losses of arable land and accelerated the release of OCP residues in soil (Juhasz et al. 2016). Along with the special use of OCPs at present, the historical use of OCPs mentioned above from the non-point source runoff appears to be the main contributor of OCP contamination in the surface water of megacities nowadays.

Wuhan as the largest city in Central China covers an area of 8494 km^2 and has a population of almost eight million in the urban area. According to the newest notice about adjustment of city size classification issued by the State Council of the People's Republic of China, Wuhan is classified into the category of megacity (Wuhan Bureau of Statistics 2014). Since Wuhan underwent rapid urbanization after the policy of reform and opening up in the 1980s, the surface water has been increasingly exposed to toxic pollutants especially OCPs. The farmland in Wuhan used to be the main operation place of OCPs has been shrunk at an average rate of 0.18% each year since the 1990s as a result of occupation by urban construction (Yan 2005). And the occupied arable land tended to be the new source of OCPs during the land development, which enhanced the exposure risk of surface water to OCPs. Against this background, few studies referred to the OCP pollution of surface water. An early OCP investigation performed by Tang et al. was in 2005 while the samples were limited to 30 covering the Yangtze River and six lakes (Tang et al. 2008). And yet, the surface water quality of Wuhan exacerbated sharply in the recent one decade. One of the most striking examples was the East Lake. Yang et al. who monitored the OCP concentrations of surface water in the East Lake in 2013 proved that the DDTs and HCHs might induce carcinogenic risks for human consumption (Yang et al. 2014). Therefore, more comprehensive and renewed research is imperative for understanding the current status of OCP contamination in the surface water of Wuhan.

In this study, we analyzed 112 surface water samples collected from 14 lakes and 11 drinking water source sites of Wuhan in winter 2013 and summer 2014 to (1) obtain data of the levels of target OCPs in the sampling area, (2) observe the spatial distribution, (3) reveal the seasonal variation trends of OCPs, (4) trace the possible sources of OCPs for the purpose of future effective environmental management, and (5) assess the potential risk of OCPs in surface water.

Materials and methods

Study area

Owing to abundant water resources, Wuhan was known as the "city of hundreds of lakes" and "river city." All 166 floodplain lakes characterized with low depth were star-studded in the city, and 40 of them were in the urban area (Lv et al. 2011; Wuhan Water Authority 2015). The Yangtze River and its largest tributary the Hanjiang River intersected in the middle of the city, dividing it into three towns (Fig. 1). The major climatic characteristic was humid subtropical monsoon with an average atmospheric temperature of 16.9 °C. The wet season between June and September has the most frequent rainfall whose amount was almost tenfold of that in the dry season (October–January) (Wuhan Bureau of Statistics 2014, 2015). The surface water area we chose in the present study covered 14 major lakes, mostly circled by Wuhan Ring Expressway, and 11 major drinking water source sites located on Yangtze River and Hanjiang River. More details of the sampling area were listed in Table 1.

Sample collection, extraction, and analysis

Overall, 112 surface water samples were collected from 11 drinking water source sites and 14 lakes in November 2013 (dry season) and July 2014 (wet season), respectively. The sampling sites in lakes were equally distributed due to the flat lakebeds. The sample collection in drinking water source sites

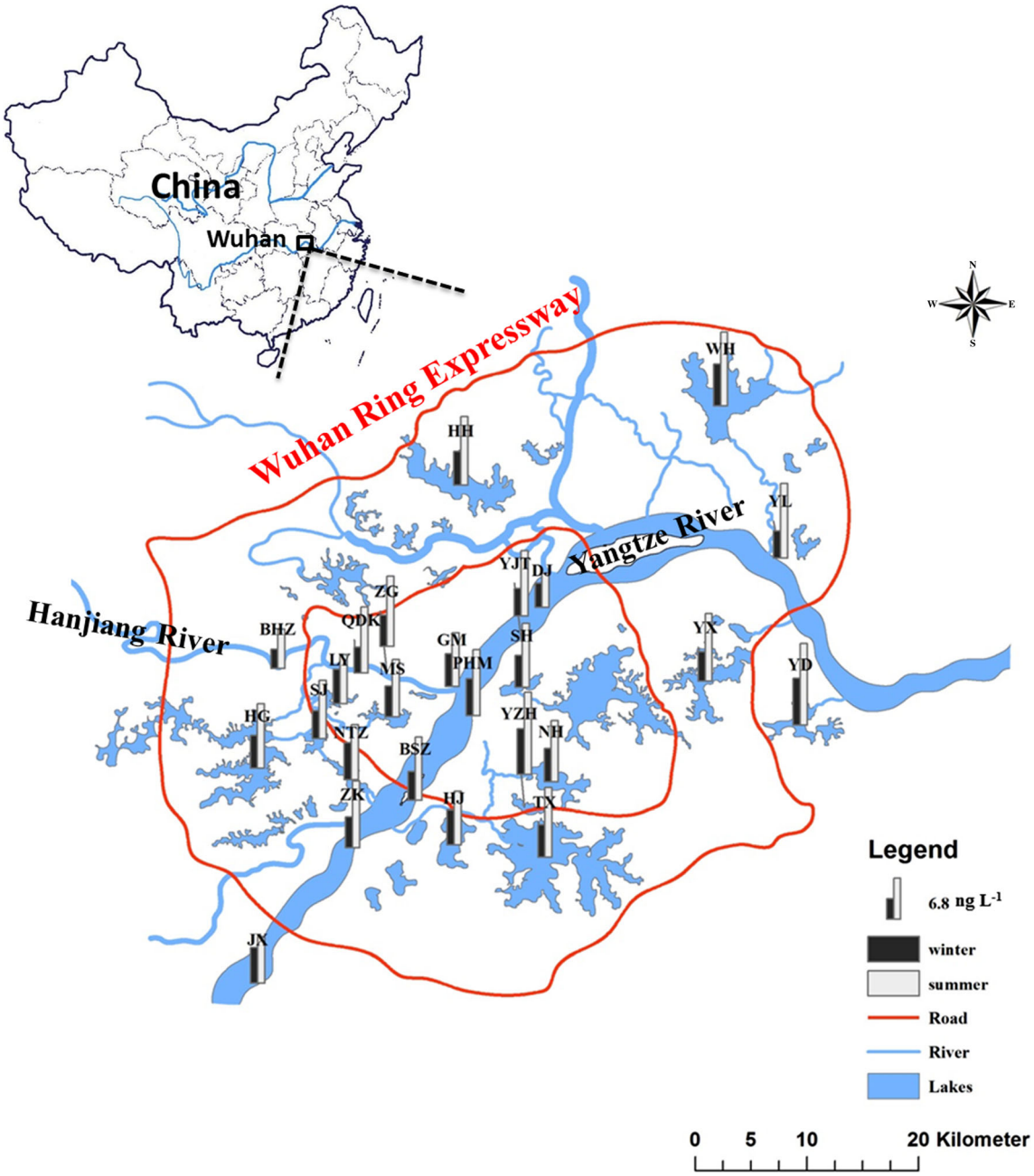


Fig. 1 Mean concentrations and spatial distribution of ∑OCPs in the surface water of Wuhan

was carried out in the range of 100 m near waterworks inlets. Three subsamples from each site were combined. Precleaned hard glass bottles were applied for the storage of water under the condition of 4 °C in a refrigerating chamber. All extraction was accomplished in 3 days.

All the organic solvents employed in sample extraction and analysis were of HPLC grade (Fisher Scientific, USA). Sixteen target OCPs were purchased from AccuStandard Co. (New Haven, CT, USA). In addition to the HCHs and DDTs (including α-HCH, β-HCH, γ-HCH, δ-HCH, *p,p′*-DDE, *p,p′*-DDD, and *p,p′*-DDT) heavily produced and used in China, other OCPs such as heptachlor, aldrin, heptachlor epoxide, endrin aldehyde, endrin, α-endosulfan, β-endosulfan, endosulfan sulfate, and methoxychlor which have been found to be extensively distributed in the sediment and fishes in the lakes of Wuhan were also investigated in the present study (Cui et al. 2015; Tang et al. 2008; Yang et al. 2014; Yun et al. 2014). The OCPs were extracted by solid phase extraction (SPE) method established by Yang et al. (2015). And the brief procedures with minor modifications were elucidated as follows: dichloromethane (5 mL), methanol (5 mL), and water (5 mL) were used to pretreat the BondEluent C18 500 mg cartridges (Agilent, USA) successively. One liter of water sample blended with 100 mL isopropyl alcohol was loaded

Table 1 The information of sampling sites

Lakes						Drinking water sources sites			
Location code	Full name	*n*	Administrative district	Lake area[a] (km^2)	Mean depth[a] (m)	Location code	Full name	*n*	Administrative district
YX	Yanxi Lake	10	Hongshan	14.23	1.9	BSZ	Baishazhou	1	Hongshan
YD	Yandong Lake	9	Hongshan, Jiangxia	9.11	1.2	GM	Guomian	1	Hanyang
SH	Shahu Lake	3	Wuchang	3.08	–	PHM	Pinghumen	1	Wuchang
NH	Nanhu Lake	6	Hongshan	7.67	–	YJT	Yujiatou	1	Hongshan
YZH	Yezhi Lake	3	Hongshan	1.62	–	DJ	Dijiao	1	Jiangan
TX	Tangxun Lake	15	Jiangxia,Hongshan	47.62	2.32	QDK	Qinduankou	1	Hanyang
HJ	Huangjia Lake	3	Hongshan	8.12	–	ZG	Zongguan	1	Qiaokou
MS	Moshui Lake	5	Hanyang	3.64	–	BHZ	Baihezui	1	Dongxihu
LY	Longyang Lake	3	Hanyang	1.68	–	JX	Jiangxia	1	Jiangxia
NTZ	Nantaizi Lake	4	Hanyang, Caidian	3.57	1.7	YL	Yangluo	1	Xinzhou
SJ	Sanjiao Lake	3	Caidian	2.39	2.6	ZK	Zhuankou	1	Caidiant
HG	Houguan Lake	16	Caidian	34	1.14				
WH	Wuhu Lake	12	Xinzhou, Huangpi	27.8	2.13				
HH	Houhu Lake	9	Huangpi	16.1	1.14				

n number of sampling sites

[a] Cited from records of lakes in Wuhan (Wuhan Water Authority 2015)

on the cartridge at an appropriate speed. After that, the cartridges were dried under vacuum for 2 h. Dichloromethane (15 mL) was adopted for eluting the chemicals. Finally, the eluate was dried under a gentle stream of high-purity nitrogen. And residues were redissolved by 100 μL of n-hexane for gas chromatographic (GC) analysis.

The extracted samples were analyzed on an Agilent 7890A gas chromatograph equipped with an electron capture detector (GC-ECD) (Agilent Technologies, Santa Clara, CA, USA). An HP-5 capillary column (30 m × 0.25 mm i.d. × 0.25-μm film thickness, Agilent) was used for the separation of the analytes. The chromatographic conditions referred to the previous research by Yun et al. (2014).

Quality assurance and quality control

Rigorous quality control procedures were implemented for all analysis. During analysis, calibration of instruments with standards was carried out every day. Solvent blanks, matrix blanks, and standard solutions were tested every ten samples throughout the whole procedure to check the status of the instrument and cross-contamination. Each sample was analyzed in triplicate. High, medium, and low levels (1, 5, 10 ng L^{-1}) of spiked water samples were analyzed prior to and during the experiment. Mean recoveries ranged from 88 to 105%. The method detection limits (MDLs) were defined by the concentrations of analytes whose signal-to-noise (S/N) ratios were three and ranged from 0.05 to 0.15 ng L^{-1}. Those samples with concentrations detected less than MDLs were treated as not detected (n.d.). All the reported concentrations were not recovery corrected.

Human health risk assessment

Health risk assessment was used to estimate the possible influence of contaminates for human beings over a specified period (Huang et al. 2014). Individuals could be exposed to OCPs in the surface water through several ways, mainly including drinking and bathing. The carcinogenic and non-carcinogenic risk assessment model recommended by the USEPA was adopted in the present study (USEPA 1989). The calculation formulas of carcinogenic risk (R) and non-carcinogenic risk (HI) were listed as follows:

$$R = SF \times E \qquad Risk < 0.01 \tag{1}$$

$$R = 1 - \exp(-SF \times E) \qquad Risk \geq 0.01 \tag{2}$$

$$HI = E/R_{FD} \tag{3}$$

$$E_{drinking} = C_B \times \frac{IR_w \times EF \times ED}{BW \times AT} \tag{4}$$

$$E_{dermal} = 2 \times 10^{-3} \times k \times C_B \times \sqrt{6 \times t \times TE/\pi} \times \frac{A_{sd} \times EF \times FE \times ED}{BW \times AT \times f} \tag{5}$$

In Eq. ((1)), SF is the slope factor (mg kg^{-1} day^{-1}(body weight))$^{-1}$. The SF values of α-HCH, β-HCH, γ-HCH, ΣHCH, heptachlor, heptachlor epoxide, *p,p′*-DDT, *p,p′*-DDE, *p,p′*-DDD, and ΣDDT were 6.3, 1.8, 1.3, 0.35, 4.5,

9.1, 0.34, 0.34, 0.24, and 0.35, respectively. In Eq. (3), R_{FD} is the oral reference dose (mg kg^{-1} day^{-1} (body weight)). The R_{FD} values of α-HCH, β-HCH, γ-HCH, ΣHCH, heptachlor, heptachlor epoxide, *p,p'*-DDT, ΣDDT, α-endosulfan, and β-endosulfan were 0.0005, 0.0002, 0.0003, 0.0003, 0.0005, 0.000013, 0.0005, 0.0005, 0.006, and 0.006, respectively. In Eqs. ((1)), ((2)), and (3), E is the exposure dose (mg kg^{-1} day^{-1}). In Eqs. (4) and (5), C_B is the pollutant concentration (mg L^{-1}), IR_w is the water intake rate (2 L day^{-1}), EF is the exposure frequency (365 days $year^{-1}$), ED is the exposure duration (30 years), BW is the body weight (the mean value of 60 kg for Chinese), AT is the average exposure time (365 days × 70 years was employed in R value calculation and 365 days × 30 years was employed in HI value calculation), k is the skin permeability parameter (0.001 cm h^{-1}), t is the lag time for each pollutant inside the body (1 h), TE is the bathing time (0.4 h), A_{sd} is the surface area of the body (16,600 cm^2), FE is the bathing frequency (0.3 times day^{-1}), and f is the intestinal absorption ratio (1).

However, the exposure of human was not only to one kind of pollutant. Two or more pollutants inside the body might induce additive and/or interactive effects, and the risk addition should be applied in such situation. The calculation formulas were listed as follows:

$$\sum \mathrm{R} = \mathrm{R}_1 + \mathrm{R}_2 \ldots\ldots \mathrm{R}_n \quad (6)$$

$$\sum \mathrm{HI} = \mathrm{HI}_1 + \mathrm{HI}_2 \ldots\ldots\ldots \mathrm{HI}_n \quad (7)$$

As suggested by USEPA, the acceptable carcinogenic risk of OCPs was between 10^{-7} and 10^{-4}. The maximum limit value of HI was 1.0 (USEPA 1989). So the water was considered to be safe for drinking and bathing unless the Rs and HIs were below 10^{-7} and 1.0, respectively.

Data analysis

Data analysis was conducted by SPSS 19 (IBM, USA). Seasonal variations of OCP levels were revealed by independent-samples t test (two tails). Spatial distribution of OCPs in surface water was illustrated by ArcGIS 10.2 software package (ESRI, USA).

Results and discussion

Levels of OCPs in surface water of Wuhan

Among the 16 target OCPs, HCHs, heptachlor, aldrin, and endosulfan sulfate were frequently detected in all samples in both summer and winter. The detailed concentrations of OCPs were shown in Table S2-1 and 2-2. The ΣOCP concentrations of surface water ranged from 5.61 to 13.62 ng L^{-1} in summer and 3.18 to 7.73 ng L^{-1} in winter (Table 2). The ΣOCP concentrations from 14 lakes were slightly higher than those of drinking water source sites in both summer and winter. The highest ΣOCP concentrations in the lakes were found at YZH (13.62 ng L^{-1}) in summer and YD (7.73 ng L^{-1}) in winter, respectively. For drinking water source sites, YL (12.32 ng L^{-1}) in summer and PHM (6.16 ng L^{-1}) in winter presented the highest ΣOCPs, respectively. The ∑HCHs of surface water was almost twofold of ∑DDTs in summer while the ∑DDTs in winter was five times higher than ∑HCHs. This inverse trend might be interpreted by the different sources of HCHs and DDTs in summer and winter.

The OCP levels in the surface water of Wuhan were compared with analogous studies regarding other locations (Table S1). Tang et al. and Yang et al. investigating smaller scales of the surface water in Wuhan compared to our study indicated similar levels of OCPs in the Yangtze River catchment and nearly twofold OCP contents in the East Lake of those in the present study (Tang et al. 2008; Yang et al. 2014). Except Honghu Lake and natural water along the Yangtze River, more OCPs were detected in a number of rivers and lakes in China than in the surface water of Wuhan (Table S1). The ∑OCPs of 132.4 ng L^{-1} in Chaohu Lake was about ten times higher than those in the surface water of Wuhan, and the ∑OCPs of Honghu Lake and natural water along the Yangtze River were almost half of those in the present study (Liu et al. 2011; Liu et al. 2013; Yuan et al. 2013). By comparison with OCP levels of surface water in other developing countries including Nigeria, India, and Ghana, the ∑OCPs in the present study were one or two orders of magnitudes lower (Agarwal et al. 2015; Ibigbami et al. 2015; Kuranchie et al. 2012). In general, the comparisons suggested that the OCP pollution in the surface water of Wuhan was at a relatively low level.

Compositions and sources of OCPs in the surface water of Wuhan

Obviously, the HCHs and DDTs were dominant among the OCPs, which was constant with the massive amount of usage in history. But the congeners' pattern of HCHs and DDTs varied significantly among the sampling lakes.

The dominant isomers of HCHs were β- and γ-HCH (32 and 29%) in winter and β- and δ-HCH (26 and 36%) in summer. The dominance of β-HCH in both seasons has been found in many other studies (Dai et al. 2011; Hu et al. 2014; Huang et al. 2014; Liu et al. 2011; Liu et al. 2013; Yun et al. 2014). Primarily due to the equatorial bonds of β-HCH between H, C, and Cl at all six positions, the molecular structure is denser and resistant to environmental degradation (Walker et al. 1999). Technical HCH (containing 60–70% α-HCH, 5–12% β-HCH, 10–15% γ-HCH, and 6–10% δ-HCH) and lindane (almost pure γ-HCH) were two typical HCH products ever used or still in use. The ratio of α/γ-HCH was a common

Table 2 Levels of ∑HCHs, ∑DDTs, ∑other OCPs, and ∑OCPs (ng L^{-1}) in the surface water of Wuhan

	Summer				Winter			
	∑HCHs	∑DDTs	∑other OCPs	∑OCPs	∑HCHs	∑DDTs	∑other OCPs	∑OCPs
Lakes								
HJ	3.59 (2.84–4.06)	1.85 (0.01–3.16)	3.42 (2.6–4.16)	8.85 (7.01–10.7)	0.54 (0.41–0.66)	3.31 (2.69–3.85)	1.94 (1.65–2.25)	5.79 (5.27–6.2)
YZH	5.84 (5.22–6.89)	2.63 (2.39–3.05)	5.15 (4.24–6.46)	13.62 (11.91–15.73)	0.39 (0.26–0.52)	5.44 (3.4–7.48)	1.82 (1.41–2.24)	7.65 (5.07–10.24)
SH	3.42 (3.3–3.54)	2.41 (2.37–2.45)	4.73 (3.54–5.93)	10.56 (9.21–11.92)	0.66 (0.45–1.01)	2.70 (2.33–3.17)	1.87 (1.35–2.3)	5.23 (4.99–5.64)
SJ	3.90 (3.29–4.51)	2.51 (2.39–2.63)	3.24 (2.41–4.08)	9.65 (8.33–10.97)	0.44 (0.43–0.45)	2.71 (2.47–2.96)	1.46 (1.13–1.79)	4.61 (4.51–4.71)
YX	4.91 (2.48–8.04)	1.96 (0.03–2.67)	4.32 (2.3–5.91)	11.19 (7.09–14.37)	0.47 (0.29–0.84)	2.67 (2.3–3.07)	1.73 (0.78–4.11)	4.87 (3.6–7.26)
TX	4.67 (2.71–6.21)	2.11 (0.01–2.95)	4.74 (3.14–6.7)	11.52 (6.78–13.86)	0.65 (0.43–1.62)	3.36 (2.8–3.85)	1.38 (0.56–2.22)	5.38 (4.19–6.31)
YD	5.93 (2.93–9.8)	2.40 (0.41–3.01)	5.15 (2.19–9.65)	13.48 (6.01–21)	0.76 (0.6–0.94)	4.31 (4.03–4.76)	2.65 (1.31–6.47)	7.73 (6.31–11.45)
NTZ	3.25 (2.2–4.35)	2.34 (2.31–2.36)	3.57 (1.5–4.91)	9.16 (6.01–11.48)	0.77 (0.67–0.94)	2.55 (2.41–2.78)	2.82 (2.29–3.6)	6.14 (5.43–7.32)
MS	4.00 (2.77–5.85)	2.02 (0.31–2.7)	3.41 (1.64–4.57)	9.43 (7.11–11.29)	0.69 (0.23–1.47)	2.46 (1.75–3.77)	1.95 (1.17–2.35)	5.09 (3.87–6.54)
NH	3.92 (2.1–6.24)	2.49 (2.33–2.72)	3.72 (1.02–5.67)	10.12 (5.45–14.47)	0.54 (0.32–1)	2.78 (2.25–3.42)	2.19 (1.28–3.38)	5.52 (4.26–7.29)
LY	3.04 (1.58–3.87)	2.35 (2.33–2.36)	2.70 (1.1–3.87)	8.10 (5.01–10.1)	0.61 (0.48–0.8)	3.39 (2.54–4.5)	1.70 (1.47–2.09)	5.70 (5.08–6.58)
WH	5.50 (2.11–9.02)	2.39 (0.01–3.74)	4.29 (1.87–5.32)	12.19 (6.39–16.94)	0.62 (0.4–0.9)	3.74 (2.56–5.68)	2.48 (1.47–3.98)	6.84 (5.18–8.4)
HH	4.52 (1.72–10.92)	2.46 (2.3–3.01)	4.39 (3.09–5.57)	11.37 (7.31–17.7)	0.53 (0.15–0.79)	2.90 (2.14–3.5)	2.15 (0.63–6.13)	5.59 (2.92–9.4)
HG	4.30 (2.5–5.9)	2.35 (0.39–3.04)	4.09 (2.15–5.31)	10.75 (5.03–13.33)	0.71 (0.19–2.42)	2.81 (2.36–4.26)	1.93 (0.85–4.19)	5.46 (3.51–8.64)
Mean	4.34 (3.04–5.93)	2.31 (1.85–2.63)	4.07 (2.70–5.15)	10.71 (8.10–13.62)	0.60 (0.39–0.77)	3.22 (2.46–5.44)	2.01 (1.38–2.82)	5.83 (4.61–7.73)
Water sources								
BSZ	4.02	1.95	4.49	10.45	0.26	3.16	1.31	4.73
PHM	4.13	2.47	4.29	10.89	0.58	4.02	1.56	6.16
QDK	4.34	0.31	6.26	10.91	0.21	3.54	0.52	4.27
GM	2.69	1.70	4.47	8.87	0.29	3.26	1.81	5.36
DJ	2.16	2.28	1.16	5.61	0.31	2.67	1.03	4.01
YJT	3.68	2.70	4.42	10.80	0.45	2.61	1.48	4.54
ZG	4.30	2.40	4.90	11.60	0.38	3.62	1.13	5.13
BHZ	3.62	0.37	2.48	6.47	0.39	1.80	0.99	3.18
ZK	4.40	1.63	4.95	10.99	0.49	2.74	1.88	5.11
JX	3.49	0.03	4.13	7.66	0.36	2.44	3.02	5.82
YL	5.60	2.36	4.36	12.32	0.26	3.09	1.16	4.51
Mean	3.86 (2.16–5.60)	1.66 (0.03–2.70)	4.17 (1.16–6.26)	9.69 (5.61–12.32)	0.36 (0.21–0.49)	2.99 (1.80–4.02)	1.45 (0.52–3.02)	4.80 (3.18–6.16)
Means of surface water	4.13 (2.16–5.93)	2.02 (0.03–2.70)	4.11 (1.16–6.26)	10.26 (5.61–13.62)	0.49 (0.21–0.77)	3.12 (1.80–5.44)	1.76 (0.52–3.02)	5.38 (3.18–7.73)

Data was shown as mean and ranges in parenthesis

indicator for determining the sources of HCHs (Kutz et al. 1991). With the ratio less than 1, the use of lindane might be the source. While the ratio was between 3 and 7, HCHs might originate from the input of technical HCH (Jiang et al. 2009). In the present study, the values of α/γ-HCH in winter varied from 0.20 to 2.29 except the undetected α-HCH in the QDK site. And in summer, the ratios of α/γ-HCH were in the range of 0.87 to 2.49 with the exceptions of undetected γ-HCH in the DJ site and an ultra-high value of 79.82 in SH lake. As suggested in Fig. 2, the HCHs in most sites might derive from the historical use of technical HCH and lindane, which was in accordance with some previous studies in the surface water of Wuhan (Tang et al. 2008; Yang et al. 2014). Significant higher values of α/γ-HCH were observed in summer than in winter ($p < 0.05$). Early research by Newland et al. revealed that γ-HCH could be converted into α- and δ-HCH by microorganism (Newland et al. 1969). The more active microorganism in summer might result in more γ-HCH degradation to α-HCH. In addition, photoisomerization of γ-HCH to α-HCH was another possible reason for the higher values of α/γ-HCH in summer (Walker et al. 1999).

Technical DDTs containing 80% *p,p′*-DDT was once applied universally in China. *p,p′*-DDT and its metabolites including *p,p′*-DDD and *p,p′*-DDE were all detected in the majority of water samples. The ratios of (DDD + DDE)/∑DDTs ranged from 0.0081 to 0.13 in winter with the undetected *p,p′*-DDD and *p,p′*-DDE in the QDK site. In summer, the ratios of (DDD + DDE)/∑DDTs were in the range of 0.0039–0.26 except the undetected *p,p′*-DDD and *p,p′*-DDE in the DJ site and *p,p′*-DDT in QDK, BHZ, and JX sites. The ratios less than 1 implied the new input of *p,p′*-DDT in the study area. It has been reported that dicofol contained relatively low DDT residues (<0.1%), but sometimes the improper production technology led to 20% DDTs in some dicofol products (Bosch et al. 2015; Qiu et al. 2005). And the technical DDT was also required in malaria control and mosquito repellency as well as the manufacture of antifouling paint for fishing ships (Cui et al. 2015; Liu et al. 2009). Therefore, even after the DDTs were strictly banned in 1983, the DDT application for dicofol production and public health campaigns as well as boat paint might be the current sources of DDTs in the surface water of Wuhan.

Seasonal variation of OCPs in surface water of Wuhan

Figure 3 illustrated the ∑OCPs of surface water in summer and winter. By the independent-samples *t* test, the differences of the total concentrations of OCPs in winter and summer were significant ($p < 0.05$). It has been widely reported that the non-point source pollution of OCPs including runoff from farmland and the developing land which used to be a farmland was dominated in surface water (Kuranchie et al. 2012; Liu et al. 2015a, b; Wang et al. 2012; Yuan et al. 2013; Yun et al. 2014). The most frequent rainfall accounting for about 70% of the annual precipitation in Wuhan was during June and September. Furthermore, the studied area was mostly low-lying and paved inadequate municipal drainage pipeline. The riparian areas around most lakes and rivers were impermeable and lack of vegetative filter strips. Consequently, the lakes and rivers probably became the main water body receiving the overland runoff, which was supposed to result in the elevated OCP concentrations in summer.

Although the input of overland runoff principally controlled the seasonal variation of OCPs, there were some differences between HCHs and DDTs. In summer, the ∑HCHs and ∑DDTs were 4.13 and 2.02 ng L^{-1}, respectively. But by contrast, the separate levels of ∑HCHs and ∑DDTs were 0.49 and 3.12 ng L^{-1} in winter. The inverse seasonal fluctuation between HCHs and DDTs might be the results of their different physical and chemical properties. On one side, DDTs with

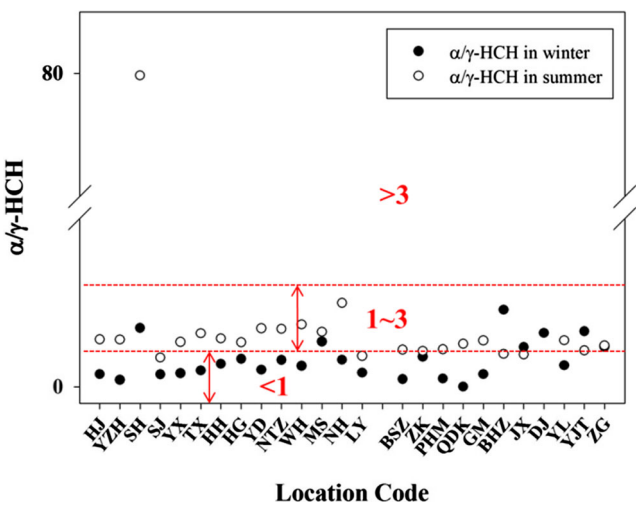


Fig. 2 Source apportionment of HCHs in the surface water of Wuhan

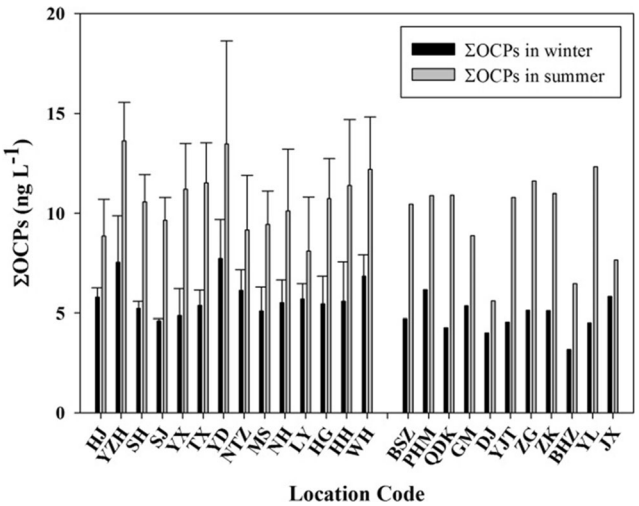


Fig. 3 Seasonal variation of ∑OCPs in the surface water of Wuhan

nearly twofold octanol-water partition coefficients of HCHs tend to more easily portion into organic phase and combine with particulate matter in water (Hale et al. 2010; Shen and Wania 2005). And the water turbulence caused by rainstorm, human recreation, shipping, and fishing occurred more frequently in summer than in winter. Hence, it appeared that the DDTs in water were less in summer than in winter. On the other side, Henry's law constants of DDTs are higher than those of HCHs, which means DDTs are more likely to evaporate into the air (Suntio et al. 1988). Even if there was some input of DDTs in overland runoff, the loss of combination with particulate matter and evaporation into the air might be more than the input.

Spatial distribution of OCPs in surface water of Wuhan

As shown in Table 2, the OCP levels in lakes were slightly higher than those in the drinking water source sites, partly because the rivers were much more fluid than the lakes, which was beneficial for the dilution of pollutant concentration (Zhou et al. 2006). For another, the anthropogenic activities around the drinking water source sites probably causing water pollution were severely restricted.

Among the lakes, YZH and YD showed fairly high OCP levels in both seasons (Fig. 1). YZH located in the Hongshan District with the population density of 3077 persons km^{-2} was affected by intensive anthropogenic activities (Table 1). And unlike many other lakes, YZH was not connected with other lakes or the Yangtze River, so the OCPs in YZH were at a state of low fluidity and easy to accumulate (Wuhan Water Authority 2015). Especially in summer, the input of land runoff evidently enhanced the OCP levels in YZH. As to YD, it was situated at the area near Wuhan Ring Expressway which used to be a farming area and underwent development later. The OCP residues in soils could be released by land development and entered into the lake by surface runoff. Furthermore, YD was also

Table 3 Health risk assessment of OCPs in 14 Lakes of Wuhan

	Lakes							
	Drinking				Bathing			
	Winter		Summer		Winter		Summer	
	Mean	Maximum	Mean	Maximum	Mean	Maximum	Mean	Maximum
Carcinogenic								
α-HCH	1.02E−08	2.25E−08	8.38E−08	1.28E−07	4.42E−11	9.79E−11	3.64E−10	5.56E−10
β-HCH	4.84E−09	6.69E−09	3.07E−08	4.86E−08	2.10E−11	2.91E−11	1.34E−10	2.12E−10
γ-HCH	3.24E−09	4.65E−09	1.09E−08	1.56E−08	1.41E−11	2.02E−11	4.73E−11	6.79E−11
∑HCHs	3.00E−09	3.85E−09	2.17E−08	2.97E−08	1.30E−11	1.68E−11	9.45E−11	1.29E−10
Heptachlor	1.46E−10	3.40E−10	2.85E−10	8.27E−10	6.34E−13	1.48E−12	1.24E−12	3.59E−12
Heptachlor epoxide	2.16E−10	5.49E−10	2.75E−10	9.27E−10	9.38E−13	2.39E−12	1.19E−12	4.03E−12
p,p′-DDE	1.52E−08	2.60E−08	1.05E−08	1.18E−08	6.62E−11	1.13E−10	4.58E−11	5.14E−11
p,p′-DDD	1.61E−08	2.72E−08	1.15E−08	1.32E−08	7.02E−11	1.18E−10	5.02E−11	5.72E−11
p,p′-DDT	4.41E−08	9.20E−08	7.66E−08	8.56E−08	1.92E−10	4.00E−10	3.33E−10	3.72E−10
∑DDTs	1.95E−09	5.21E−09	1.85E−08	5.21E−08	8.49E−12	2.26E−11	8.04E−11	2.26E−10
∑R	9.90E−08	1.63E−07	2.65E−07	3.25E−07	4.30E−10	7.09E−10	1.15E−09	1.41E−09
Non-carcinogenic								
α-HCH	7.52E−06	1.67E−05	6.19E−05	9.46E−05	3.28E−08	7.26E−08	2.70E−07	4.12E−07
β-HCH	3.13E−05	4.33E−05	1.99E−04	3.15E−04	1.36E−07	1.89E−07	8.65E−07	1.37E−06
γ-HCH	1.93E−05	2.78E−05	6.49E−05	9.32E−05	8.43E−08	1.21E−07	2.83E−07	4.06E−07
∑HCHs	6.64E−05	8.55E−05	4.82E−04	6.58E−04	2.90E−07	3.72E−07	2.10E−06	2.87E−06
Heptachlor	2.09E−04	3.56E−04	1.44E−04	1.62E−04	9.09E−07	1.55E−06	6.29E−07	7.05E−07
Heptachlor epoxide	2.15E−04	3.62E−04	1.54E−04	1.75E−04	9.35E−07	1.58E−06	6.69E−07	7.63E−07
p,p′-DDT	4.56E−05	9.52E−05	7.93E−05	8.86E−05	1.99E−07	4.15E−07	3.45E−07	3.86E−07
∑DDTs	3.84E−05	1.02E−04	3.64E−04	1.02E−03	1.67E−07	4.46E−07	1.59E−06	4.46E−06
α-Endosulfan	3.73E−07	8.33E−07	6.58E−07	1.67E−06	1.62E−09	3.63E−09	2.87E−09	7.26E−09
β-Endosulfan	1.23E−07	3.33E−07	9.99E−07	2.22E−06	5.35E−10	1.45E−09	4.35E−09	9.67E−09
∑HI	6.32E−04	8.96E−04	1.55E−03	2.61E−03	2.76E−06	3.91E−06	6.76E−06	1.14E−05

an important aquaculture base in Wuhan. The DDTs in anti-fouling paint for fishing ships were also a noteworthy contributor for the OCP pollution in YD (Wu et al. 2016).

For drinking water source sites, relative high OCP concentrations were detected in YL and PHM in both seasons. As recorded in Table 1, YL was in the Xinzhou District which was a traditional agriculture production area of Wuhan. The consumption of pesticide in Xinzhou was much more than any other district in the present and the past, which consequently gave rise to the high OCP levels in YL (Wuhan Bureau of Statistics 2014). PHM was in the central urban area of Wuhan, and no large-scale farming activities occurred here historically or currently. The most possible source of OCPs in PHM was the transportation of pollutants by river flow from the upper reach.

Risk assessment

Owing to the early recognition of OCPs' toxicity for human and aquatic organisms, different environmental protection departments issued quality standards of surface water in which some OCP concentrations were regulated. In the quality standard from the China State Environmental Protection Administration (MEP P.R. China 2002), the standard limits of DDTs, lindane, and heptachlor epoxide were 1000, 2000, and 20 ng L^{-1}, respectively, which were far higher than the concentrations detected in the present study. The national recommended aquatic life criteria made by the US EPA set the freshwater criterion maximum concentrations of aldrin, α-endosulfan, β-endosulfan, endrin, lindane, heptachlor, heptachlor epoxide, and 4,4′-DDT for 3000, 220, 220, 86, 950, 520, 520, and 1100 ng L^{-1}, respectively (USEPA 2009). The corresponding levels in the surface water of Wuhan were about three to five orders of magnitudes lower. Compared with the maximum allowable concentration (MAC) in environmental quality standards issued by the European Union (10, 40, 0.3 ng L^{-1} for endosulfan, HCH, heptachlor and heptachlor epoxide, respectively), heptachlor and heptachlor epoxide in all sampling sites except BSZ and DJ in winter

Table 4 Health risk assessment of OCPs in 11 drinking water source sites of Wuhan

	Drinking water source sites							
	Drinking				Bathing			
	Winter		Summer		Winter		Summer	
	Mean	Maximum	Mean	Maximum	Mean	Maximum	Mean	Maximum
Carcinogenic								
α-HCH	7.04E−09	1.53E−08	7.98E−08	1.05E−07	3.06E−11	6.66E−11	3.47E−10	4.58E−10
β-HCH	2.95E−09	6.18E−09	2.39E−08	4.32E−08	1.28E−11	2.69E−11	1.04E−10	1.88E−10
γ-HCH	1.88E−09	3.90E−09	1.33E−08	1.67E−08	8.16E−12	1.70E−11	5.79E−11	7.28E−11
∑HCHs	1.81E−09	2.90E−09	1.93E−08	2.80E−08	7.87E−12	1.26E−11	8.39E−11	1.22E−10
Heptachlor	1.86E−10	6.81E−10	4.86E−10	1.46E−09	8.07E−13	2.96E−12	2.11E−12	6.34E−12
Heptachlor epoxide	1.87E−10	4.80E−10	2.46E−10	1.20E−09	8.14E−13	2.09E−12	1.07E−12	5.22E−12
p,p′-DDE	1.41E−08	1.89E−08	7.20E−09	1.18E−08	6.14E−11	8.20E−11	3.13E−11	5.12E−11
p,p′-DDD	1.50E−08	2.01E−08	8.28E−09	1.35E−08	6.52E−11	8.75E−11	3.60E−11	5.88E−11
p,p′-DDT	3.67E−08	8.30E−08	6.93E−08	8.30E−08	1.59E−10	3.61E−10	3.01E−10	3.61E−10
∑DDTs	8.28E−10	5.21E−09	3.67E−09	2.60E−08	3.60E−12	2.26E−11	1.59E−11	1.13E−10
∑R	8.07E−08	1.26E−07	2.25E−07	2.90E−07	3.51E−10	5.48E−10	9.80E−10	1.26E−09
Non-carcinogenic								
α-HCH	5.21E−06	1.13E−05	5.90E−05	7.79E−05	2.27E−08	4.93E−08	2.57E−07	3.40E−07
β-HCH	1.91E−05	4.00E−05	1.55E−04	2.80E−04	8.31E−08	1.74E−07	6.74E−07	1.22E−06
γ-HCH	1.12E−05	2.33E−05	7.95E−05	9.99E−05	4.88E−08	1.02E−07	3.46E−07	4.35E−07
∑HCHs	4.02E−05	6.44E−05	4.28E−04	6.22E−04	1.75E−07	2.81E−07	1.87E−06	2.71E−06
Heptachlor	1.93E−04	2.58E−04	9.86E−05	1.61E−04	8.43E−07	1.13E−06	4.30E−07	7.02E−07
Heptachlor epoxide	1.99E−04	2.68E−04	1.10E−04	1.80E−04	8.69E−07	1.17E−06	4.80E−07	7.84E−07
p,p′-DDT	3.80E−05	8.59E−05	7.17E−05	8.59E−05	1.65E−07	3.74E−07	3.12E−07	3.74E−07
∑DDTs	1.63E−05	1.02E−04	7.22E−05	5.12E−04	7.10E−08	4.46E−07	3.15E−07	2.23E−06
α-Endosulfan	2.37E−07	7.22E−07	5.65E−07	1.72E−06	1.03E−09	3.14E−09	2.46E−09	7.50E−09
β-Endosulfan	1.26E−07	3.33E−07	4.89E−07	2.05E−06	5.50E−10	1.45E−09	2.13E−09	8.95E−09
∑HI	5.23E−04	6.93E−04	1.08E−03	2.02E−03	2.28E−06	3.02E−06	4.68E−06	8.81E−06

exceeded the threshold, indicating the possible adverse effects for aquatic lives (European Union 2013). Take the juvenile zebrafish (*Danio rerio*) as an example, the fishes' growth and gill morphology might be affected by heptachlor (Campagna et al. 2007).

In regard to the human health risk, the carcinogenic and non-carcinogenic risk assessments of drinking and bathing were conducted in the present study. By the way, although the lakes in Wuhan were no longer used for drinking water supply, what we found in the field investigation was that some residents around the lakes got used to drinking the lake water. And bathing near the drinking water source sites often occurred in summer. Therefore, human health risks by the two exposure routes were both assessed in lakes and drinking water source sites. As listed in Tables 3 and 4, ∑HIs in all sampling sites were three to six orders of magnitudes lower than the threshold 1.0, implying the ignorable non-carcinogenic risks by drinking and bathing. However, the carcinogenic risks caused by drinking could not be neglected. The ∑Rs in summer were more than 10^{-7} in all sampling sites and with the mean values of 2.65E−07 and 2.25E−07 in lakes and drinking water source sites, respectively. For individual pollutant, α-HCH in summer posed the highest carcinogenic risk among the target OCPs, with the Rs of YZH, TX, YD, WH, and YL higher than 10^{-7} (Table S4-1). In winter, the carcinogenic risks were much lower. The mean values of ∑Rs were 9.90E−08 and 8.07E−08 in lakes and drinking water source sites, respectively. But the ∑Rs of YZH, SH, HG, YD, WH, ZK, and JX were above 10^{-7}, indicating the unnegligible risk (Table S3-1). The carcinogenic risks for bathing in both seasons were much lower than 10^{-7} in all sampling sites (Table S3-1 and 4-1). Therefore, the surface water in Wuhan was not safe for directly drinking because of the non-negligible carcinogenic risks. Effective purification actions for removing the OCPs in surface water were urgently required before drinking. But bathing in the surface water was safe since the carcinogenic and non-carcinogenic risks were both lower than the thresholds.

Conclusions

The present work reported the systemic data on the contamination status of OCPs in the surface water of Wuhan. A wide occurrence of OCPs has been observed in 14 lakes and 11 drinking water source sites. Compared with other rivers and lakes worldwide, OCP pollution in the surface water of Wuhan was at a relatively low level. Due to the non-point source pollution mainly including land runoff, higher ∑OCP concentrations in summer than in winter were detected, while the different physical and chemical properties of HCHs and DDTs resulted in the inverse seasonal variation trend between them. The source apportionment of HCHs and DDTs showed the historical use of technical HCH as well as lindane and the new input of *p,p′*-DDT, respectively. Drinking water source sites were less contaminated by OCPs compared with lakes. Anthropogenic activities including land development and fishery were the main factors affecting the spatial distribution of OCPs. The heptachlor and heptachlor epoxide in most sampling sites exceeded the threshold set by the European Union, indicating the possible adverse effects for aquatic lives. The total carcinogenic risks of all target OCPs in all sampling sites implied that the surface water in Wuhan was not safe for drinking without purification but suitable for bathing. Non-carcinogenic risks for drinking and bathing could be neglected.

Acknowledgments This project was supported by the Natural Science Key Foundation of Hubei Province of China (No. 2014CFA114), the Funding Project of Sino-Africa Joint Research Center, the Chinese Academy of Sciences (Y623321K01), and the Hundred Talents Program of the Chinese Academy of Sciences (No. Y329671K01).

References

Agarwal A, Prajapati R, Singh O, Raza S, Thakur L (2015) Pesticide residue in water—a challenging task in India. Environ Monit Assess 187:1–21

Bosch C, Grimalt JO, Fernández P (2015) Enantiomeric fraction and isomeric composition to assess sources of DDT residues in soils. Chemosphere 138:40–46

Calhoun Y (2005) Water pollution. Chelsea House Publisher, New York

Campagna AF, Eler MN, Fracácio R, Rodrigues BK, Verani NF (2007) The toxic potential of aldrin and heptachlor on *Danio rerio* juveniles (Cypriniformes, Cyprinidae). Ecotoxicology 16:289–298

Cui L, Ge J, Zhu Y, Yang Y, Wang J (2015) Concentrations, bioaccumulation, and human health risk assessment of organochlorine pesticides and heavy metals in edible fish from Wuhan, China. Environ Sci Pollut Res 22:15866–15879

Dai G, Liu X, Liang G, Han X, Shi L, Cheng D, Gong W (2011) Distribution of organochlorine pesticides (OCPs) and poly chlorinated biphenyls (PCBs) in surface water and sediments from Baiyangdian Lake in North China. J Environ Sci 23:1640–1649

Die Q, Nie Z, Yang Y, Tang Z, Huang Q (2015) Persistent organic pollutant waste in China: a review of past experiences and future challenges. J Mater Cycles Waste Manag 17:434–441

European Union (2013) Directive 2013/39/EU of the European parliament and of the council of 12 August 2013 amending directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy. Off J Eur Union

Hale S, Martin T, Goss K, Arp H, Werner D (2010) Partitioning of organochlorine pesticides from water to polyethylene passive samplers. Environ Pollut 158:2511–2517

Hu Y, Cheng H (2013) Water pollution during China's industrial transition. Environ Dev 8:57–73

Hu Y, Yuan L, Qi S, Liu H, Xing X (2014) Contamination of organochlorine pesticides in water and sediments from a waterbird-inhabited lake, East Central China. Environ Sci Pollut Res 21:9376–9384

Huang Y, Xu Y, Li J, Xu W, Zhang G, Cheng Z, Liu J, Wang Y, Tian C (2013) Organochlorine pesticides in the atmosphere and surface water from the equatorial Indian Ocean: enantiomeric signatures, sources, and fate. Environ Sci Technol 47:13395–13403

Huang T, Guo Q, Tian H, Mao X, Ding Z, Zhang G, Li J, Ma J, Gao H (2014) Assessing spatial distribution, sources, and human health risk

of organochlorine pesticide residues in the soils of arid and semiarid areas of northwest China. Environ Sci Pollut Res 21:6124–6135

Ibigbami O, Aiyesanmi A, Adeyeye E, Adebayo A (2015) Persistent organochlorine pesticide residues in water, sediments and fish samples from Ogbese river. Environ Natur Resour Res 5:28

Jiang Y, Wang X, Jia Y, Wang F, Wu M, Sheng G, Fu J (2009) Occurrence, distribution and possible sources of organochlorine pesticides in agricultural soil of Shanghai, China. J Hazard Mater 170: 989–997

Juhasz AL, Herde P, Smith E (2016) Oral relative bioavailability of dichlorodiphenyltrichloroethane (DDT) in contaminated soil and its prediction using in vitro strategies for exposure refinement. Environ Res 150:482–488

Kuranchie M, Atiemo S, Palm L, Blankson A, Tutu A, Fosu P (2012) Determination of organochlorine pesticide residue in sediment and water from the Densu river basin, Ghana. Chemosphere 86:286–292

Kutz F, Wood P, Bottimore D (1991) Organochlorine pesticides and polychlorinated biphenyls in human adipose tissue. Reviews of environmental contamination and toxicology, Springer, pp 1–82

LeVeen E, Willey W (1983) A political economic analysis of urban pest management. See Frankie Kohler 1983:19–40

Li W, Yang H, Gao Q, Pan H, Yang H (2012) Residues of organochlorine pesticides in water and suspended particulate matter from Xiangshan Bay, East China Sea. B Environ Contam Tox 89:811–815

Li P, Wang Y, Huang W, Yao H, Xue B, Xu Y (2014) Sixty-year sedimentary record of DDTs, HCHs, CHLs and endosulfan from emerging development gulfs: a case study in the Beibu Gulf, South China Sea. B Environ Contam Tox 92:23–29

Liu X, Zhang G, Li J, Yu L, Xu Y, Li X, Kobara Y, Jones K (2009) Seasonal patterns and current sources of DDTs, chlordanes, hexachlorobenzene, and endosulfan in the atmosphere of 37 Chinese cities. Environ Sci Technol 43:1316–1321

Liu C, Yuan G, Yang Z, Yu T, Xia X, Hou Q, Chen L (2011) Levels of organochlorine pesticides in natural water along the Yangtze River, from headstream to estuary, and factors determining these levels. Environ Earth Sci 62:953–960

Liu W, He W, Qin N, Kong X, He Q, Ouyang H, Xu F (2013) 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. Environ Sci Pollut Res 20:2033–2045

Liu L, Bai L, Man C, Liang W, Li F, Meng X (2015a) DDT vertical migration and formation of accumulation layer in pesticide-producing sites. Environ Sci Technol 49:9084–9091

Liu M, Feng J, Kang B, Chen Y, Liu Q, Sun J (2015b) Ecological and health risks of DDTs and HCHs in surface water from the upper reaches of the Huaihe River. Toxicol Environ Chem 97:1124–1136

Lv J, Wu H, Chen M (2011) Effects of nitrogen and phosphorus on phytoplankton composition and biomass in 15 subtropical, urban shallow lakes in Wuhan, China. Limnol Ecol Manag Inland Waters 41:48–56

MEP P.R. China (2002) Environmental quality standards for surface water (GB3838-2002). Ministry of environmental protection of the People's Republic of China

Newland L, Chesters G, Lee G (1969) Degradation of γ-BHC in simulated lake impoundments as affected by aeration. J Water Pollut Control Fed R174-R188

Oliveira A, Cavalcante R, Duaví W, Fernandes G, Nascimento R, Queiroz M, Mendonça K (2016) The legacy of organochlorine pesticide usage in a tropical semi-arid region (Jaguaribe River, Ceará, Brazil): implications of the influence of sediment parameters on occurrence, distribution and fate. Sci Total Environ 542:254–263

Qiu X, Zhu T, Yao B, Hu J, Hu S (2005) Contribution of dicofol to the current DDT pollution in China. Environ Sci Technol 39:4385–4390

Shen L, Wania F (2005) Compilation, evaluation, and selection of physical-chemical property data for organochlorine pesticides. J Chem Eng Data 50:742–768

Standing Committee of the National People's Congress (2013) Amendments of the new nine persistent organic pollutants (POPs) in "Stockholm Convention on POPs". Bulletins of the Standing Committee of the National People's Congress 5:737–745

State Council of the People's Republic of China (2014) Notice about adjustment of city size classification issued by the State Council of the People's Republic of China (http://www.gov.cn/zhengce/content/2014-11/20/content_9225.htm)

Suntio L, Shiu W, Mackay D, Seiber J, Glotfelty D (1988) Critical review of Henry's law constants for pesticides. Reviews of environmental contamination and toxicology, Springer, pp 1–59

Tang Z, Yang Z, Shen Z, Niu J, Cai Y (2008) Residues of organochlorine pesticides in water and suspended particulate matter from the Yangtze River catchment of Wuhan, China. Environ Monit Assess 137:427–439

United States Environment Protection Agency (USEPA) (1989) Risk assessment guidance for superfund. vol I: human health evaluation manual (Part A), EPA/540/1-89/002, Office of emergency and remedial response, Washington, DC (http://www.epa.gov/oswer/riskassessment/ragsa/index.htm)

United States Environment Protection Agency (USEPA) (2009) National recommended water quality criteria. (https://www.epa.gov/wqc/national-recommended-water-quality-criteria-aquatic-life-criteria-table)

Walker K, Vallero D, Lewis R (1999) Factors influencing the distribution of lindane and other hexachlorocyclohexanes in the environment. Environ Sci Technol 33:4373–4378

Wang X, Xu J, Guo C, Zhang Y (2012) Distribution and sources of organochlorine pesticides in Taihu Lake, China. B Environ Contam Tox 89:1235–1239

Wu Y, Wang X, Ya M, Li Y, Hong H (2016) Distributions of organochlorine compounds in sediments from Jiulong River Estuary and adjacent Western Taiwan Strait: Implications of transport, sources and inventories. Environ Pollut (Available online)

Wuhan Bureau of Statistics (2014) Wuhan statistical yearbook—2013. China Statistics Press, Beijing

Wuhan Bureau of Statistics (2015) Wuhan statistical yearbook—2014. China Statistics Press, Beijing

Wuhan Water Authority (2015) Records of lakes in Wuhan. Changjiang Publishing Media and Hubei Fine Arts Publishing House, Wuhan

Yan D (2005) The research of dynamic change of cultivated land in Wuhan. J Huazhong Agricultural University (Social Science Edition) (in Chinese) 5, 6, 19–21

Yang Y, Yun X, Liu M, Jiang Y, Li Q, Wang J (2014) Concentrations, distributions, sources, and risk assessment of organochlorine pesticides in surface water of the East Lake, China. Environ Sci Pollut Res 21:3041–3050

Yang Y, Xie Q, Liu X, Wang J (2015) Occurrence, distribution and risk assessment of polychlorinated biphenyls and polybrominated diphenyl ethers in nine water sources. Ecotox Environ Safety 115: 55–61

Yuan L, Qi S, Wu X, Wu C, Xing X, Gong X (2013) Spatial and temporal variations of organochlorine pesticides (OCPs) in water and sediments from Honghu Lake, China. J Geochem Explor 132:181–187

Yun X, Yang Y, Liu M, Wang J (2014) Distribution and ecological risk assessment of organochlorine pesticides in surface sediments from the East Lake, China. Environ Sci Pollut Res 21:10368–10376

Zhang G, Parker A, House A, Mai B, Li X, Kang Y, Wang Z (2002) Sedimentary records of DDT and HCH in the Pearl River Delta, South China. Environ Sci Technol 36:3671–3677

Zhang W, Wang C, Li Y, Wang P, Wang Q, Wang D (2014) Seeking sustainability: multiobjective evolutionary optimization for urban wastewater reuse in China. Environ Sci Technol 48:1094–1102

Zhao L, Hou H, Zhou Y, Xue N, Li H, Li F (2010) Distribution and ecological risk of polychlorinated biphenyls and organochlorine pesticides in surficial sediments from Haihe River and Haihe Estuary Area, China. Chemosphere 78:1285–1293

Zhi H, Zhao Z, Zhang L (2015) The fate of polycyclic aromatic hydrocarbons (PAHs) and organochlorine pesticides (OCPs) in water from Poyang Lake, the largest freshwater lake in China. Chemosphere 119:1134–1140

Zhou R, Zhu L, Yang K, Chen Y (2006) Distribution of organochlorine pesticides in surface water and sediments from Qiantang River, East China. J Hazard Mater 137:68–75