



# Sources and Health Risk of Organochlorine Pesticides in Surface Water from Buerhatong River and Hunchun River in Northeast China

Liu Qiang · Zheng Jinping · Guo Zhongling · Fan Chunnan

Received: 6 May 2021 / Accepted: 25 August 2021 / Published online: 29 September 2021  
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**Abstract** Organochlorine pesticides (OCPs) were widely used in agriculture and disease control, which causes potential harm to eco-environment and human health, and the OCP pollution is closely related to past or current human activities. A total of 27 surface water samples were collected from two rivers with different human interference intensity, and 13 OCPs were analyzed. The study results showed that the mean concentrations of OCPs in Buerhatong River (BR) and Hunchun River (HR) were 780.45 ng L<sup>-1</sup> and 900.69 ng L<sup>-1</sup>, respectively. On the basis of environmental quality standards of China, the concentrations of DDTs, heptachlor epoxide, and  $\gamma$ -HCH were lower than standard values, but the maximum concentration of  $\gamma$ -HCH in HR (1923.26 ng L<sup>-1</sup>) is close to the standard limit, showing a potential risk to the aquatic environment. And there are significant differences in residues of OCPs between two tributaries due to the different human activities. DDTs and HCHs are the main components of OCPs, and the DDTs in BR were mostly derived from historical use residues, whereas HR might have recent DDT use input. And BR had a good water environment with high dissolved oxygen, but there are some anaerobic sections in HR. The HCHs in BR are mainly the historical residues, except for one site, which might

have lindane input in the environment, and there also might be recent imports of lindane in the HR basin. The health risk assessment results showed that there was no adverse health effects for non-carcinogenic risk. But carcinogenic risk caused by dermal contact for adults had moderate risk in two sites of the studied rivers. The study of the impact of human activities on the residues of organochlorines in surface water is of great significance to the prevention and control of organochlorine pesticide pollution in the water environment and the protection of the ecological environment and the health of the population, especially for adults.

**Keywords** Human activities · Health risk · Sources · Organochlorine pesticides · Organochlorine pesticides · River

## 1 Introduction

OCPs are the persistent organic pollutants (POPs) and have persistence, high toxicity, refractory, and bioaccumulativity, which can be long-term remained in the environmental mediums at global scale (Ravindran et al., 2016). Because of their high pesticide efficiency and low cost, OCPs were widely used in agriculture, industry, and for malaria control (Buah-Kwofie & Humphries, 2017; Luz et al., 2014). Therefore, a large amount of OCPs have been remained in the environmental media, which can enter the human body by

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consuming contaminated water and aquatic products (Zhou et al., 2008b). In the process of applying OCPs, the OCPs can enter the soil or atmosphere by spray drift, and the residual OCPs in the soil will volatilize into the atmosphere and then fall out into the rivers through dry and wet deposition (Agarwal et al., 1987; Kaushik, 1989, 1991; Kaushik et al., 1987, 1991). OCPs can cause a variety of diseases, such as cancer, reproductive defects, endocrine, and immunological toxicities, so they have been widely banned all over the world (Mrema et al., 2013). But because OCPs are harmful and resistant to degradation and have low water solubility, so they have persistence in the environment and still have a high detection rate in different environment media (Antary et al., 2020; Chakraborty et al., 2019; Fernandes et al., 2020; Tang et al., 2020; Tham et al., 2019). OCP residues in the environment media have a significant impact on the ecological environment and human health. The study results of Ravi River and its three northern tributaries in Pakistan showed that all the studied streams for endosulfan ( $\alpha$ -endosulfan) and endrin had a high ecological risk, and the carcinogenic risk assessment indicated that the water of study area is possessing carcinogenic risk to local population and is unsafe for human bathing purposes (Baqar et al., 2018).

In China, OCPs are also ubiquitous and have been produced in a large amount, such as dichloro-diphenyl-trichloroethanes (DDTs), hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB), and mirex (Hua, 1996). Approximately 4.5 million tons of technical HCHs and 0.27 million tons of DDTs were produced in China after 1952 until being banned in 1983 (Liu et al., 2016). And about 11,400 t of lindane was still reportedly being produced after 1983 (Li et al., 2001), and DDTs have been continuously produced for approximately 20 years owing to export demands, malaria control, and dicofol production (Yang et al., 2008). OCPs may enter the river via agricultural runoff, direct applications, spray drift, aerial spraying, and erosion (Chakraborty et al., 2016), so the OCP pollution in estuaries and rivers also has attracted general attention in China (Luo et al., 2004; Tang et al., 2008; Wang et al., 2010; Wei et al., 2014). Moreover, OCPs have long-distance migration ability through the food chain and even can reach high latitudes through atmospheric deposition (dry and wet) (Ribes et al., 2002). Previous research showed that OCPs could be detected in Tuotuo River (the origin of Yangtze River),

but OCPs had never been used in this area because of its 4,540-m height above sea level (Liu et al., 2011).

Buerhatong River and Hunchun River are the main tributaries of Tumen River, which have an important role in the sustainable development of the Tumen River basin. Tumen River basin is an important area for China's open development along the border and an important gateway to Northeast Asia. There are significant differences in soil land use patterns in the two river basins, and the proportion of agricultural land and urban land in the Buerhatong River basin is significantly higher than Hunchun River. Although the two rivers are in the Tumen River basin, the degree of human disturbance in history shows significant differences. And affected by natural conditions, industrial structure, economic level, etc., the ecological environment of BR and HR has been contaminated to varying degrees. Therefore, the research on the pollution and risks of organochlorine pesticide residues in rivers under different disturbance intensity of human activities will provide important theoretical and data support for the in-depth exploration of the impact of human activities on the distribution of organochlorine pollution. The objectives of the current study were to firstly investigate the residue concentrations and distribution pattern of 13 OCPs ( $\alpha$ -HCH,  $\beta$ -HCH,  $\gamma$ -HCH,  $\delta$ -HCH, PP'-DDE, PP'-DDD, OP'-DDT, PP'-DDT, HCB, heptachlor, heptachlor epoxide, dieldrin, and endrin) in the surface water of BR and HR; to identify the sources of OCPs and indicate the effect of human activities on the residues of OCPs in the study area; and to assess the non-carcinogenic risk and carcinogenic risk of OCPs for children and adults through different exposure pathways.

## 2 Materials and Methods

### 2.1 Site Description

The Buerhatong River and Hunchun River are the main tributaries of the Tumen River basin. The BR flows about 242 km and originates in Haerba mountain ridge with an approximately tributary area of 7141km<sup>2</sup>, and HR, about 200 km long, flowing from Pan mountain ridge to Tumen River. Climate of the study area is continental monsoon climate with an annual average temperature of 2.6–5.4°C, and annual rainfall varied from 400 to 800 mm mostly distributed

in high flow period (June to September). The area proportion of different land use types in the different watersheds of Tumen River is listed in Table 1 (Li et al., 2013). The population density in BR basin is high, and the industry and agriculture are relatively developed, and the industry is dominated by coal, food, tobacco, textile and clothing, electricity, wood processing, etc. The HR basin is less affected by the human activities relative to Buerhatong River.

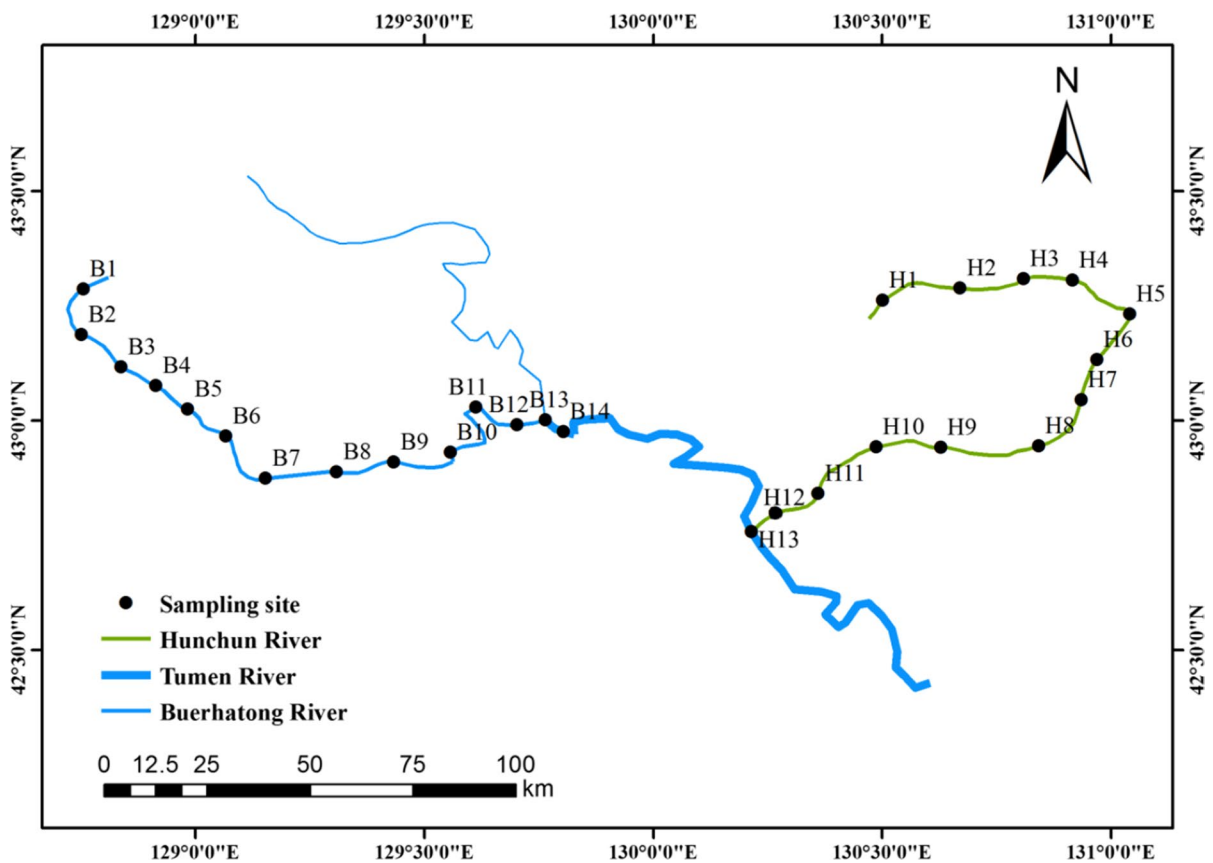
### 2.2 Sampling

Sampling sites along BR and HR were selected equably to cover the research area. Fourteen and

thirteen surface water samples were collected with pre-cleaned 1-L dark glass bottles using cylinder samplers from Buerhatong River and Hunchun River, respectively, and the sampling locations were marked as B1 to B14 for Buerhatong River and H1 to H13 for Hunchun River, respectively (Fig. 1), and the distance between two adjacent sampling points is about 10–15 km. Water samples were collected within 1 day, and the pH of samples were adjusted to <2 with hydrochloric acid solution after sampling and stored at 4 °C. The extraction was completed within 7 days, and the analysis was completed within 40 days.

**Table 1** Area proportion of different land use types in the different watersheds

River	Farmland	Woodland	Grassland	Canal	Cities and towns	Wetland	Other
BR (%)	23.33	70.38	3.27	0.26	2.34	0.21	0.21
HR (%)	13.05	83.72	1.67	0.10	0.85	0.49	0.12



**Fig. 1** Sampling sites of surface water in BR and HR

### 2.3 Chemical Analysis

The chemical analysis method of this study mainly refers to the environmental protection standard of the People's Republic of China (HJ 921–2017). First, 10 mL of methanol and 20.0 μL standard solution of substitute (tetrachloro-*m*-xylene, decachlorobiphenyl) were added in 200.0 mL of water sample and mixed well. In turn, 5 mL of ethyl acetate, 5 mL of methanol, and 10 mL of water were used to activate the solid-phase extraction cartridge at a flow rate of about 5 mL/min. The water sample was passed through the solid-phase extraction cartridge at a flow rate of 10 mL/min. After loading, the solid-phase extraction cartridge was rinsed with 10 mL of water and the cartridge was drained. Then the solid-phase extraction cartridge was eluted with 2.5 mL of ethyl acetate and 5 mL of dichloromethane in sequence, with a flow rate of about 5 mL/min, and the eluent was collected into a concentration tube. The eluent was passed through the drying column, and the concentration tube was washed with a small amount of dichloromethane 2–3 times, and the washing solution was dehydrated through the drying column. All the dehydrated eluate was collected into a concentration tube and was concentrated to about 3 mL. Then purification process was proceed, Flory diatomaceous earth was infiltrated with 8 mL of *n*-hexane, the extract was transferred to the small column before the liquid level disappeared, the concentration tube was washed with 1–2 mL of *n*-hexane, the washing solution was put on the column together (note: always keep a liquid surface above the packing), then the column was eluted with 10 mL of acetone/*n*-hexane, and all the eluent was collected. The eluent was concentrated to less than 1 mL, 5.0 μL of internal standard use solution was added, was diluted to 1.0 mL with *n*-hexane, was mixed well, and was transferred to the autosampler vial for testing. Analytical reagents and distilled water that met national standards were used for residue analysis.

### 2.4 Quantitative Analysis and Quality Control

A Shimadzu GC-2010Plus equipped with a micro-cell electron capture detector (ECD) was used for OCP quantification in the current study, and the capillary column used was Rxi-5 ms (30 cm × 0.25 mm × 0.25 μm), which is a low

polarity 5% diphenyl/95% dimethyl polysiloxane stationary phase. The inlet temperature is 250 °C, and the injection is splitless. The GC column temperature was programmed as follows: initially at 80 °C (equilibrium time 1 min), increased to 150 °C at the rate of 20 °C min<sup>-1</sup> and held for 2 min, before reaching at 300 °C at the rate of 5 °C min<sup>-1</sup>, and then held for 5 min. The column flow rate is 1.0 mL/min. While analyzing the sample, take the same volume of pure water, prepare a blank sample according to the preparation of the sample, and measure the blank sample with the same instrument analysis conditions as the calibration curve. The relative standard deviation was 1.50–5.54%, and the surrogate recovery in this method was 80–110%, and the method detection limits (MDLs) ranged from 0.022 to 0.069 ng L<sup>-1</sup> (HJ 921–2017).

### 2.5 Statistical Analysis

The analysis of variance for different OCP components in surface water of BR and HR was conducted using the software of SPSS version 21, and Origin Pro 8.0 was used for graph making.

The hazard quotient (*HQ*) is used to assess non-carcinogenic risks exposure to OCPs in surface water. The exposure dose through the pathways of water ingestion and dermal contact were calculated with Eqs. (1) and (2) (Chen et al., 2020; Kerr et al., 1998):

$$ADD_{ing} = C \times \frac{IngR \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (1)$$

$$ADD_{der} = C \times \frac{SA \times k \times ET \times EF \times ED}{BW \times AT} 10^{-9} \quad (2)$$

where *C* is the arithmetic mean for OCP concentrations of water. Exposure was expressed in terms of a daily dose (mg/kg/day) and calculated separately for each OCP. *ADD<sub>ing</sub>* (mg/kg/day) is daily exposure amount of OCPs through ingestion, and *ADD<sub>der</sub>* (mg/kg/day) is daily exposure amount of OCPs through dermal contact. The exposure factors for these models are shown in Table 2.

After *ADD<sub>ing</sub>* and *ADD<sub>der</sub>* were calculated, a hazard quotient (*HQ*) based on non-cancer toxic risk can then be calculated by dividing daily dose to a specific reference dose (*RfD*):

**Table 2** Exposure parameters for dose models

Parameter	Definition	Unit	Value		Reference
			Children	Adult	
IngR	Oral intake rate	L/day	0.12	0.12	Chen et al. (2020)
EF	Exposure frequency	Days/year	40	40	Chen et al. (2020)
ED	Exposure duration	Year	7	43	Chen et al. (2020)
BW	Body weight	kg	18	60	Li et al. (2014)
SA	Exposed skin area	cm <sup>2</sup>	2800	5700	USEPA (2001)
ET	Daily exposure time	h/day	1	1	Chen et al. (2020)
AT	Average time	Days	26,280	26,280	Chen et al. (2020)
	k	RfD <sub>ing</sub>	RfD <sub>der</sub>	SF <sub>ing</sub>	SF <sub>der</sub>
α-HCH	7.98	8.00E-03	8.00E-03	6.30E+00	6.47E+00
β-HCH	7.98	/	/	1.80E+00	1.99E+00
γ-HCH	7.98	3.00E-04	2.98E-04	1.30E+00	1.99E+00
δ-HCH	7.98	/	/	/	/
PP'-DDE	78.73	3.00E-04	3.00E-04	3.40E-01	4.25E-01
PP'-DDD	63.70	3.00E-05	3.00E-05	2.40E-01	3.00E-01
OP'-DDT	165.66	/	/	/	/
PP'-DDT	197.45	5.00E-04	5.00E-04	3.40E-01	4.25E-01
HCB	98.36	8.00E-04	8.00E-04	1.60E+00	3.20E+00
Heptachlor	31.62	5.00E-04	4.00E-04	4.50E+00	5.63E+00
Heptachlor epoxide	3.56	1.30E-05	1.30E-05	9.10E+00	9.10E+00
Dieldrin	15.22	5.00E-05	5.00E-05	1.60E+00	16.00E+00
Endrin	15.22	3.00E-04	3.00E-04	/	/

k is the dermal permeation constant of each OCP, cm/h; RfD is the non-carcinogenic reference dose for the specific exposure pathways (ingestion and dermal contact) of the OCP, g/(kg·day); SF is the cancer slope factor for the specific exposure pathways (ingestion and dermal contact) of OCP, kg·day/mg

$$HQ = \frac{ADD}{RfD} \tag{3}$$

where *HQ* is the hazard quotient, indicating the non-carcinogenic risk of single contamination; the reference dose (*RfD*) is an estimate of maximum permissible risk to a human population through daily exposure during a lifetime. When  $HQ \leq 1$  indicates no adverse health effects and  $HQ > 1$  indicates likely adverse health effects, and the hazard index (*HI*) was equal to the sum of *HQs* and was used to estimate the health risk, *HI* values of  $\leq 1$  indicate no adverse health effects, and *HI* values  $> 1$  indicate possible adverse health effects (USEPA, 2001). Toxicological characteristics of the investigated OCPs used for health risk assessments are presented in Table 2.

For carcinogens, the carcinogenic health risk can be calculated by multiplying ADD by cancer slope factor (SF):

$$CR_{ing} = ADD_{ing} \times SF_i \tag{4}$$

$$CR_{der} = ADD_{der} \times SF_i \tag{5}$$

where  $CR_{ing}$  is the carcinogenic risk for the exposure pathway of ingestion and  $CR_{der}$  is the carcinogenic risk for the exposure pathway of dermal contact.  $SF_i$  is the corresponding slope factor for the specific exposure pathway of the *i*th OCP. For carcinogenic risk (*CR*), risks surpassing  $1 \times 10^{-1}$  are viewed as very high risk, risk values between  $1 \times 10^{-3}$  and  $1 \times 10^{-1}$  are considered to pose high risk, risks ranging from  $1 \times 10^{-4}$  to  $1 \times 10^{-3}$  represent moderate risk, values between  $1 \times 10^{-6}$  and  $1 \times 10^{-4}$  indicate low risk, and values smaller than  $1 \times 10^{-6}$  indicate very low risk (Qu et al., 2015).

### 3 Results and Discussion

#### 3.1 Content and Distribution Characteristics of Organochlorine Pesticides in BR and HR

Residues of 13 OCPs were detected in surface water along BR and HR, and the concentration and

**Table 3** Concentrations and detection rate of OCPs in BR and HR

OCPs	BR			HR		
	Mean (ng L <sup>-1</sup> )	Range (ng L <sup>-1</sup> )	Detection rate (%)	Mean (ng L <sup>-1</sup> )	Range (ng L <sup>-1</sup> )	Detection rate (%)
$\alpha$ -HCH	1.00	0.0–7.92	28.6%	0.00	0.0–0.0	0.0%
$\beta$ -HCH	387.78	0.0–1455.87	92.9%	4.29	0.0–55.82	7.7%
$\gamma$ -HCH	303.91	0.0–695.64	92.9%	334.59	0.0–1923.26	92.3%
$\delta$ -HCH	17.18	0.0–75.62	92.9%	449.66	0.0–1443.19	92.3%
$\sum$ HCHs	709.87	67.70–1977.73	100.0%	788.54	0–3366.45	92.3%
PP'-DDE	16.64	0.0–50.36	71.4%	0.45	0.0–4.69	15.4%
PP'-DDD	0.08	0.0–1.08	7.1%	2.35	0.0–17.10	23.1%
OP'-DDT	20.97	0.0–153.25	71.4%	84.91	12.92–163.42	100.0%
PP'-DDT	1.56	0.0–13.4	14.3%	0.00	0.0–0.0	0.0%
$\sum$ DDTs	39.25	1.08–153.25	100.0%	87.72	12.92–180.52	100.0%
HCB	12.76	0.48–65.40	100.0%	4.90	0.0–38.58	61.5%
Heptachlor	10.59	0.0–32.87	85.7%	3.72	0.0–18.23	46.2%
Heptachlor epoxide	5.36	0.0–37.15	21.4%	1.17	0.0–8.93	15.4%
Dieldrin	0.00	0.0–0.0	0.0%	2.00	0.0–9.42	38.5%
Endrin	2.62	0.0–6.82	50.0%	12.65	0.0–63.78	38.5%
$\sum$ OCPs	780.45	111.56–2211.49	100.0%	900.69	12.92–3521.54	100.0%

Detection rate means the proportion of samples above the detection limit to the total number of samples. Technical Specifications for Surface Water and Wastewater Monitoring (HJ/T91-2002) stipulates that when the measurement result is above the detection limit (or minimum detection concentration), the actual measurement result shall be reported, and when it is lower than the detection limit of the method, report the detection limit of the method used and count it as 0 when counting the total pollution

detection rate of OCPs are shown in Table 3. The current study has revealed that a significant number of pesticide residues were detected in the BR and HR, a total of 12 OCPs were detected in surface water of BR, and the detection rates of  $\beta$ -HCH,  $\gamma$ -HCH,  $\delta$ -HCH, and HCB are over 90%, of which HCB achieved 100%, but Dieldrin had not been detected. And except for  $\alpha$ -HCH and PP'-DDT, 11 OCPs were also detected in Hunchun River, and  $\gamma$ -HCH (92.3%),  $\delta$ -HCH (92.3%), and OP'-DDT (100%) have high detection rate. The concentration of  $\sum$ OCPs in BR ranged from 111.56 to 2211.49 ng L<sup>-1</sup> with the mean concentration of 780.45 ng L<sup>-1</sup> and varied from 12.92 to 3521.54 ng L<sup>-1</sup> in HR with the mean concentration of 900.69 ng L<sup>-1</sup>. Among all the sampling sites, the B9 station and H6 station had the highest  $\sum$ OCPs.

By comparing and analyzing the residual characteristics of OCPs in different rivers, it is found that there are significant differences in OCP pollution levels in many rivers at home and abroad, but the pollution of HCHs and DDTs is prominent. In the current study, the HCHs and DDTs in the two rivers are also the hot pollutants. In China, technical HCH

and DDT were widely used in agriculture before they were banned in 1983 (Gan et al., 2002; Tao et al., 2005), and the other OCPs, such as HCB, heptachlor, heptachlor epoxide, dieldrin, and endrin, have also been produced and applied in China in the past. But during nearly 40 years, research shows that many rivers in China still have the OCP residues, which are most likely caused by the historical residues and the continued use of OCPs in other fields. For example, DDT is still allowed to be used for controlling the reproduction of mosquitoes and prevent diseases such as malaria (World Health Organization, WHO). OCPs were monitored along the Yangtze River from headstream to estuary, and the results showed that the concentrations of  $\sum$ HCHs and  $\sum$ DDTs ranging from 0.11 to 13.68 and from 0 to 23.31 ng L<sup>-1</sup> (Liu et al., 2011), respectively, were lower than the current study. Moreover, the mean concentration of  $\sum$ HCHs and  $\sum$ DDTs in BR and HR was also higher than Pearl River (Tang et al., 2018; Yang et al., 2004), Huaihe River (Feng et al., 2011; Wang et al., 2009), Peacock River in Xinjiang (Chen et al., 2011), Qiantang River (Zhou et al., 2008a), and Shayi River (Bai et al.,

2018). But the OCPs in the current study were also lower than the other rivers, such as the concentrations of  $\sum$ DDTs and dieldrin in the surface river from BR and HR were lower than Tonghui River in Beijing (Zhang et al., 2004),  $\sum$ HCHs, heptachlor, dieldrin, and endrin were significantly lower than the surface water from Lagos lagoon complex in Nigeria (Adeboyejo et al., 2011), and the  $\sum$ DDT content was also lower than river Yamuna in Haryana and Delhi, India (Kaushik et al., 2008).

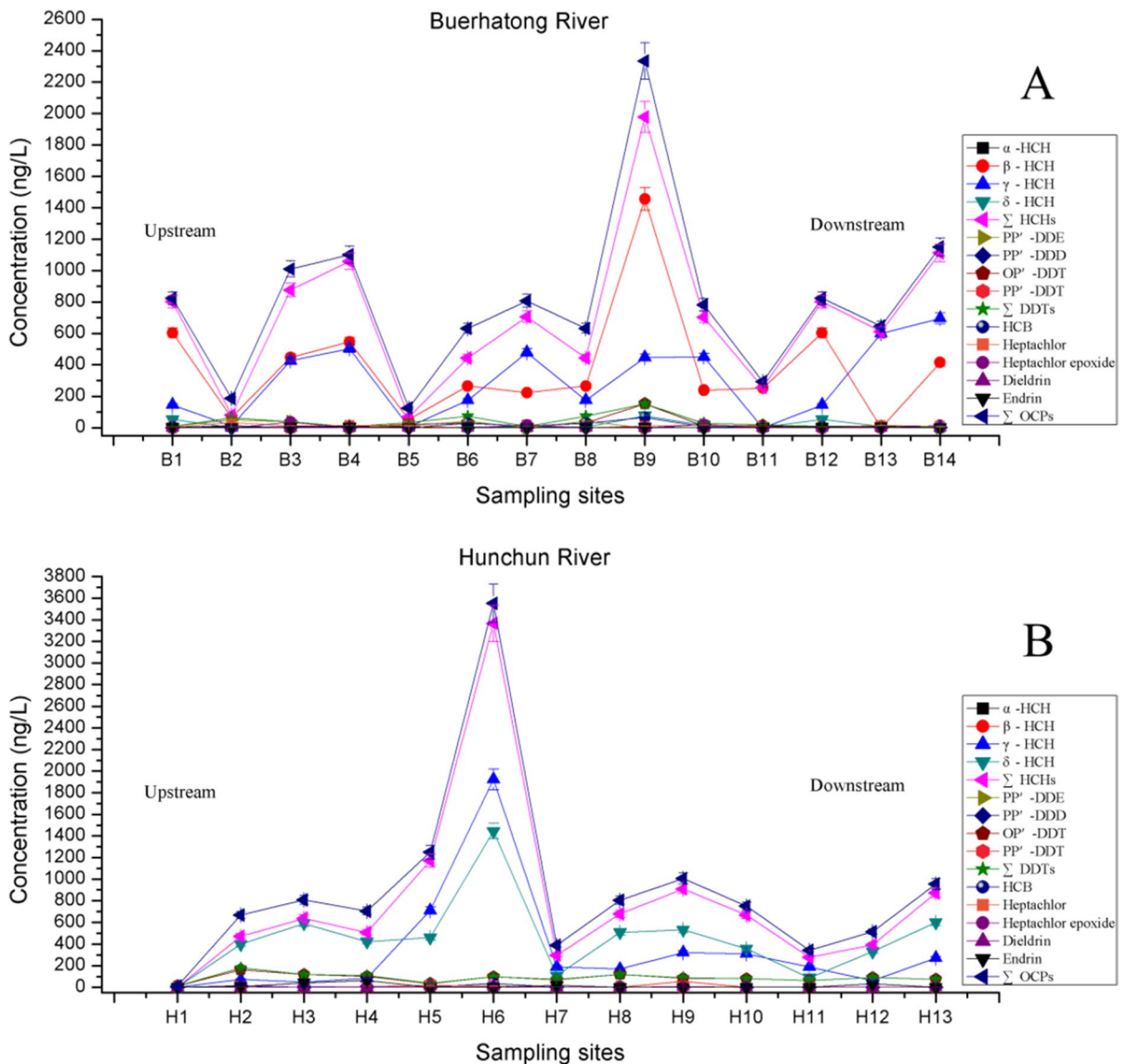
Affected by the way and degree of disturbance of human activities, there are significant differences in the residues of OCPs in different sections of the river from upstream to downstream, and the high residues were mainly present in the middle reaches in the current study (Fig. 2A and B). For BR, the maximum residual concentrations of  $\beta$ -HCH,  $\delta$ -HCH,  $\sum$ HCHs, OP'-DDT,  $\sum$ DDTs, HCB, and  $\sum$ OCPs were present in the sampling site of B9, and the maximum concentrations of PP'-DDE and heptachlor were in B2,  $\gamma$ -HCH and PP'-DDD were in B14, and  $\alpha$ -HCH, heptachlor epoxide, and endrin were present in B5, B3, and B13, respectively, except for PP'-DDT and dieldrin which had no residues (Fig. 2A). For HR, the sampling site of H6 had the maximum residual concentrations for  $\gamma$ -HCH,  $\delta$ -HCH,  $\sum$ HCHs, HCB, and  $\sum$ OCPs, and H2 had the maximum concentrations for PP'-DDD, OP'-DDT, and  $\sum$ DDTs, and H5 had the maximum concentrations for heptachlor, heptachlor epoxide, and dieldrin; the high residues of  $\beta$ -HCH, PP'-DDE, and endrin were present in H9, H7, and H4, respectively, except for  $\alpha$ -HCH and PP'-DDT which had no residues (Fig. 2B). So the sites of B9 (the entrance of BR into Yanji city) and H6 (near the village of Pear ditch) are the pollution hotspot for OCPs in BR and HR, respectively, indicating that the regional environment of B9 and H6 is easy to cause the accumulation of OCPs and is not conducive to the degradation, and it might be caused by the specific physical and chemical properties of river sediments in this area, which could provide anaerobic conditions where the metabolites may be different. But in order to ensure the safety of water use for local residents, effective control measures should be taken against high-pollution areas to improve the quality of the water environment.

According to Chinese guideline (GB 3838–2002), the concentration of DDTs should be less than 1000 ng L<sup>-1</sup> (same to the critical value for DDT in

USEPA guideline), and the concentration of lindane and heptachlor epoxide should be less than 2000 ng L<sup>-1</sup> and 200 ng L<sup>-1</sup>, respectively. The maximum concentrations of DDTs for BR (153.25 ng L<sup>-1</sup>) and HR (180.52 ng L<sup>-1</sup>) are significantly lower than the value of standard, and the maximum concentrations of heptachlor epoxide for BR (37.15 ng L<sup>-1</sup>) and HR (8.93 ng L<sup>-1</sup>) are also significantly less than the value of 200 ng L<sup>-1</sup>. Meanwhile, the average concentrations of  $\gamma$ -HCH (account for 99% of lindane (Li et al., 2018) for BR (303.91 ng L<sup>-1</sup>) and HR (334.59 ng L<sup>-1</sup>) are significantly lower than 2000 ng L<sup>-1</sup>, so the quality of surface water of BR and HR are reasonably good. But compared to other regions (Bai et al., 2018; Chen et al., 2011; Feng et al., 2011; Liu et al., 2011; Tang et al., 2018; Wang et al., 2009; Yang et al., 2004; Zhou et al., 2008a), the values of  $\sum$ HCHs are relatively high for both rivers, and the maximum concentration of  $\gamma$ -HCH in HR (1923.26 ng L<sup>-1</sup>) is close to the standard limit, which is a potential risk to the aquatic environment and human health.

### 3.2 Effect of Human Activities on the OCP Pollution

OCPs are a class of synthetic insecticides with a broad spectrum of insecticides, low toxicity, and long residual effect. However, because they pose a serious threat to the ecological environment and human health, they are now banned by many countries, including China (Gan et al., 2002; Tao et al., 2005). The OCP pollution is closely related to history or current human activities, and OCP residues in the current environment may come from historical residues or new imports. In the current study, BR and HR had different disturbed intensities by human activities, and the proportion of land use area for farmland, grassland, canal, cities, and town in BR basin is significantly higher than HR (Table 1), indicating that BR basin had strong human being activities compared to HR. The results show that the concentrations of  $\alpha$ -HCH,  $\delta$ -HCH, PP'-DDE, OP'-DDT,  $\sum$ DDTs, heptachlor, and dieldrin are significantly different between the two rivers (Fig. 3). BR had higher residue levels of  $\alpha$ -HCH,  $\beta$ -HCH, PP'-DDE, and heptachlor than HR, but the concentrations of  $\delta$ -HCH and OP'-DDT were lower, indicating that although the historical activities of human beings have a greater interference with the river environment, the degree of change in the residual amount of OCPs between



**Fig. 2** Spatial distribution of OCPs in surface water of BR and HR (B1-B14 and H1-H13 are the sampling sites for BR and HR, respectively)

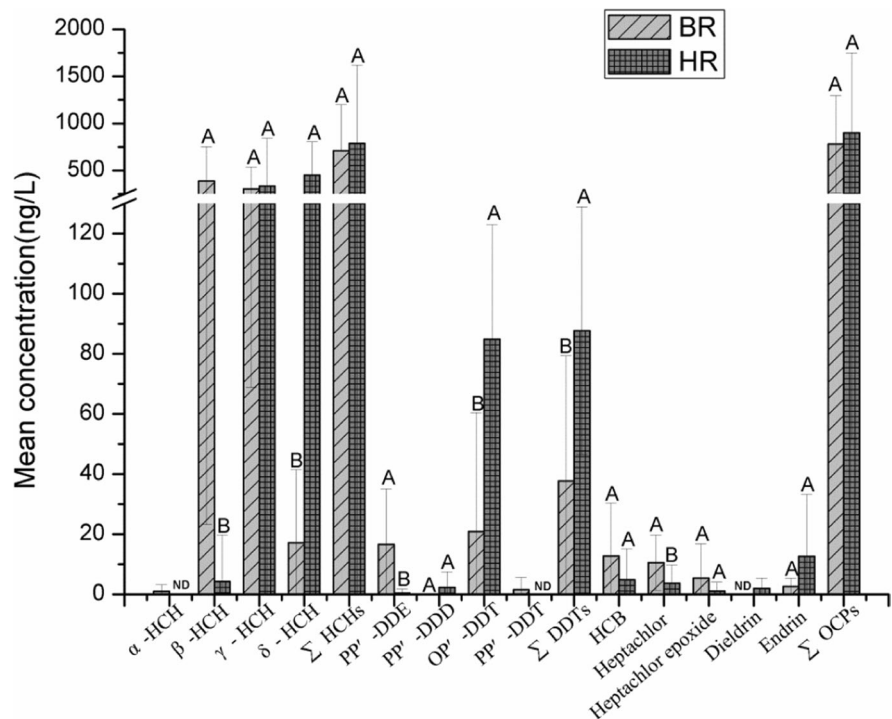
different rivers will be significantly different that may be due to the environmental conditions of dissolved oxygen, temperature, altitude, later input, etc. (Chakraborty et al., 2016; Hitch & Day, 1992; Liu et al., 2011). In order to further analyze the reasons of OCP residue in BR and HR, the sources of the main OCP residues were analyzed.

Among the OCPs, DDTs and HCHs are the most widely used OCPs with high content and detection rate (Bai et al., 2018; Chen et al., 2011; Feng et al.,

2011; Wang et al., 2009; Zhou et al., 2008a), and in the current study, the proportion of DDTs and HCHs in OCPs averaged 95.7%. DDTs are mainly derived from the direct use of industry and the use of dicofol. Due to the different isomer composition of DDT in industrial DDT and dicofol, the source of DDT can be analyzed according to the content characteristics of OP'-DDT and PP'-DDT remaining in the environment (Jaward et al., 2005). In addition, DDTs will degrade into different products of DDD and



**Fig. 3** Differences in the concentrations of OCPs between BR and HR

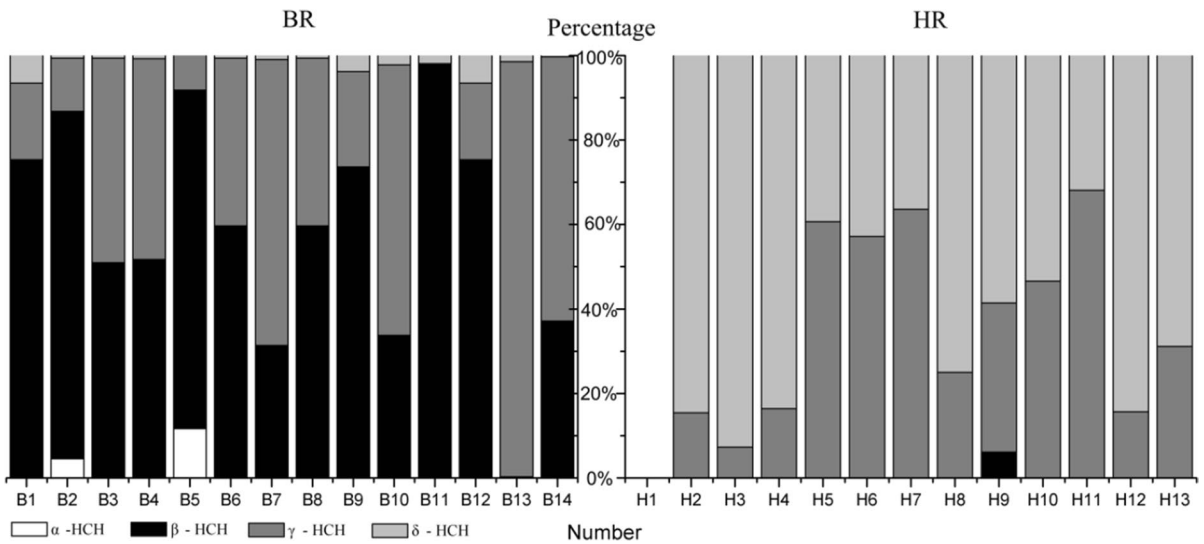
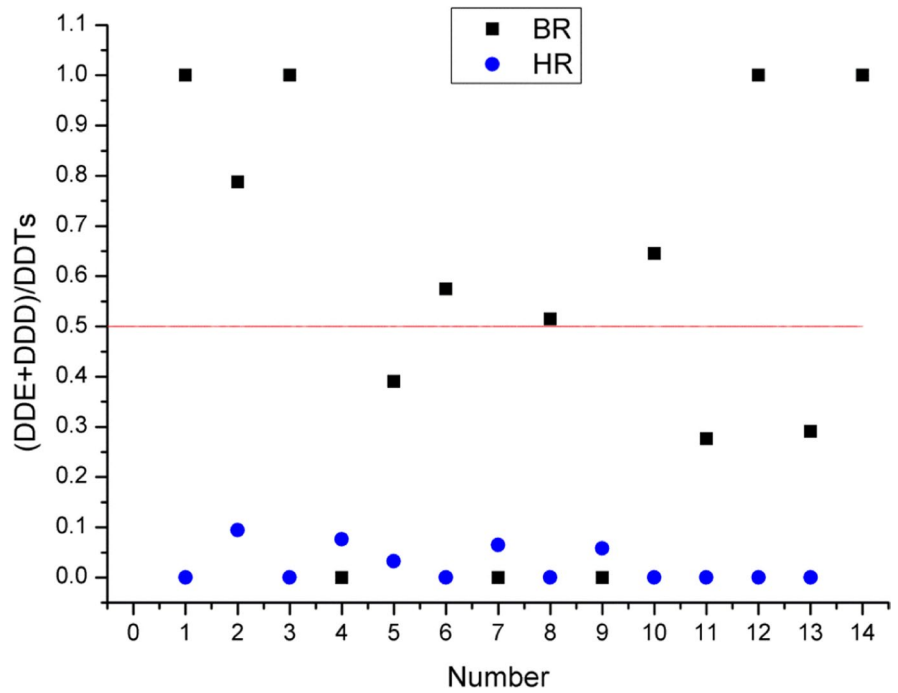


DDE after entering the environment, so the ratio of (DDE+DDD)/DDTs is commonly used to track the degree of degradation of DDTs and used to determine the source of DDTs in the environment (historical use residues or recently used DDTs input), and (DDE+DDD)/DDTs > 0.5 indicates that it is mainly derived from historical use residues; on the contrary, (DDE+DDD)/DDTs < 0.5 indicates that there are recent DDT use input; meanwhile, it can also be used to trace the degradation conditions by analyzing the ratio characteristics between DDD and DDE (Hitch & Day, 1992; Hong et al., 1999). The values of (DDE+DDD)/DDTs for BR and HR are presented in Fig. 4. The ratios of (DDE+DDD)/DDTs in 57.14% of the sampling sites of BR were higher than 0.5, indicating that the DDTs were mainly derived from historical use residues in these area, whereas the ratios of HR were all lower than 0.5, indicating that there were recent DDT use input (Hitch & Day, 1992). The detection rate of DDE in surface water of BR reached at 71.4%, and only one site had DDD, showing that BR had a good water environment with high dissolved oxygen. However, compared with DDE, DDD had a higher detection rate for HR, showing that there are some anaerobic sections in the river, which causes DDTs to be mainly anaerobic degradation. Therefore,

in order to improve the water environment, the water quality should be regulated for these anaerobic sections, which can promote the degradation of OCPs.

Compositional differences of HCHs also can indicate different contamination sources. Technical HCHs contain 60–70%  $\alpha$ -HCH, 5–12%  $\beta$ -HCH, 10–12%  $\gamma$ -HCH, and 6–10%  $\delta$ -HCH, and lindane contains more than 99% of  $\gamma$ -HCH (Li et al., 2018). Technical HCH application is prohibited, but lindane has been widely used until 2019 in China (Ministry of Ecology and Environment et al. 2019). The  $\beta$ -HCH is the most stable isomer of HCHs, and  $\alpha$ -HCH and  $\gamma$ -HCH in the environment can be converted to  $\beta$ -HCH; therefore, if there is no new HCH input, the proportion of  $\beta$ -HCH isomer will gradually increase (Walker et al., 1999). Figure 5 shows the composition distribution of the isomers of HCHs in BR and HR. It can be seen from the figure that the  $\beta$ -HCH component of most sampling points in BR is the highest, and the average proportion of  $\beta$ -HCH is 58%, which is much higher than the initial proportion value (5–12%) of  $\beta$ -HCH in industrial HCH, indicating that the HCHs in the study area have been degraded for a long period of time, mainly based on historical residues. However, the composition of the isomer  $\gamma$ -HCH in the water

**Fig. 4** Ratios of (DDE+DDD)/DDTs for BR and HR



**Fig. 5** Composition distribution of the isomers of HCHs in BR and HR (B1-B14 and H1-H13 are the sampling sites for BR and HR, respectively)

of B13 is the highest, reaching 98%, indicating that there is lindane input in the environment. Meanwhile, the detection rates of  $\alpha$ -HCH and  $\beta$ -HCH in HR are low, but  $\gamma$ -HCH and  $\delta$ -HCH all have a high detection rate, indicating that there may be

recent imports of lindane in the basin, and an in-depth investigation should be conducted to protect ecological safety of HR basin. Moreover, due to the less proportion of  $\delta$ -HCH in technical HCHs, the  $\delta$ -HCH in surface water of HR may derive from the

**Table 4** Non-carcinogenic hazard and carcinogenic risks for each OCP in river water

Items	Non-carcinogenic risk (%)				Carcinogenic risk (%)				
	$HQ \leq 1$	$HQ > 1$	$HI \leq 1$	$HI > 1$	Class I	Class II	Class III	Class IV	Class V
Burhatong River									
Children <sub>ing</sub>	100	0	100	0	0	0	0	0	100
Children <sub>der</sub>	100	0	100	0	0	0	0	100	0
Adult <sub>ing</sub>	100	0	100	0	0	0	0	0	100
Adult <sub>der</sub>	100	0	100	0	0	0	7.1	92.9	0
Hunchun River									
Children <sub>ing</sub>	100	0	100	0	0	0	0	0	100
Children <sub>der</sub>	100	0	100	0	0	0	0	92.3	7.7
Adult <sub>ing</sub>	100	0	100	0	0	0	0	0	100
Adult <sub>der</sub>	100	0	100	0	0	0	7.7	84.6	7.7

$HQ \leq 1$  and  $HI \leq 1$  indicate no adverse health effects, respectively; and  $HQ > 1$  and  $HI > 1$  indicate possible adverse health effects, respectively; class I means very high risk, class II means high risk, class III means moderate risk, class IV means low risk, and class V means very low risk

interconversion between different components of HCHs, and the transformation mechanism needs to be further studied.

### 3.3 Health Risk Assessment

The OCPs contained in river water could enter human bodies and endanger human’s health through dermal contact and mistaken oral intake (Chen et al., 2020). In the current study, dermal contact was the main exposure pathway for the OCPs, and ingestion contributed the lower exposure doses through drinking raw water or accidental ingestion. Values of hazard quotient ( $HQ$ ), hazard index ( $HI$ ), and carcinogenic risk ( $CR$ ) for the studied elements in surface water from BR and HR are listed in Table 4. The  $HQ$  and  $HI$  of OCPs through two pathways were all below 1 in two rivers, indicating no adverse health effects for non-carcinogenic risk. The health risk assessment results showed that the carcinogenic risks for dermal contact and mistaken oral intake were generally low, except for adults, whose  $CR$  caused by dermal contact was moderate risk in one sampling site for BR and HR, respectively. Adults had the highest carcinogenic risk in both exposure pathways, because the adults have larger body weight, exposed skin area, and exposure duration than children, which would make them have the highest exposure doses (Chen et al., 2020). Besides that, the carcinogenic and non-carcinogenic risks caused by dermal contact were much higher than that of ingestion. Thus, the carcinogenic and non-carcinogenic risks of OCPs in rivers to the adults should

be of great concern in the current study area, especially for the exposure pathway of dermal contact.

### 4 Conclusions

The current study exposed residual levels of 13 OCPs in the surface water of two main tributaries of Tumen River, Northeast China. In the research area, the concentration of  $\sum$ OCPs in the surface water of BR ranged from 111.56 to 2211.49 ng L<sup>-1</sup> with the mean concentration of 780.45 ng L<sup>-1</sup> and varied from 12.92 to 3551.50 ng L<sup>-1</sup> in HR with the mean concentration of 900.69 ng L<sup>-1</sup>. According to Chinese guideline (GB 3838–2002), the maximum concentrations of DDTs and heptachlor epoxide for BR and HR are significantly lower than the value of standard, and the average concentrations of  $\gamma$ -HCH are significantly lower than 2000 ng L<sup>-1</sup>, so the quality of surface water of BR and HR are reasonably good. But compared to other regions, the values of  $\sum$ HCHs are relatively high for both rivers, and the maximum concentration of  $\gamma$ -HCH in HR (1923.26 ng L<sup>-1</sup>) is close to the standard limit, which is a potential risk to the aquatic environment and human health. Moreover, the entrance of BR into Yanji city and the site near the village of Pear ditch are the pollution hotspot for OCPs in BR and HR, respectively. Although BR and HR are all the main tributaries of the Tumen River, there are significant differences in residues of OCPs due to the different intensity of human activities. BR had higher residue levels of  $\alpha$ -HCH, PP'-DDE, and heptachlor than HR, but the concentrations

of  $\delta$ -HCH, OP'-DDT,  $\Sigma$ DDTs, and dieldrin were lower, indicating that although the historical activities of human beings have a greater interference with the river environment, the degree of change in the residual amount of OCPs between different rivers will be significantly different due to the environmental conditions of dissolved oxygen, later input, etc. Among the OCPs in the current study, DDTs and HCHs have high content and detection rate, and the proportion of DDTs and HCHs in OCPs averaged 95.7%. The source analysis results showed that DDTs in BR were mostly derived from historical use residues, whereas HR might have recent DDT use input. And BR had a good water environment with high dissolved oxygen, but there are some anaerobic sections in HR, which causes a relatively poor degradation environment for HR compared with BR. The HCHs in BR have been degraded for a long period of time, mainly based on historical residues, except for the site of B13, which might have lindane input in the environment. Meanwhile, there also might be recent imports of lindane and pesticides containing  $\delta$ -HCH in the HR basin. In order to further protect the eco-environmental health of the study area, relevant environmental protection departments should conduct in-depth investigations on the OCP pollution in hotspots, especially the problems of poor degradation conditions or the possibility of recent OCP use input, and take immediate steps in reducing the potential contamination of the study area. The OCPs contained in river water could enter human bodies and endanger human's health through dermal contact and ingestion. The health risk assessment results showed that the *HQ* and *HI* of OCPs through two pathways were all below 1 in two rivers, indicating no adverse health effects for non-carcinogenic risk. But carcinogenic risk caused by dermal contact for adults had moderate risk in two sampling sites in the current study. The adults had the higher non-carcinogenic risk and carcinogenic risk in both exposure pathways than children. Thus, the carcinogenic and non-carcinogenic risks of OCPs in rivers to the adults should be of great concern in the current study area, especially for the exposure pathway of dermal contact.

**Acknowledgements** The authors acknowledge the support of the Education Department of Jilin Province (Study on Habitat Characteristics and Survival Strategies of Three

Early Spring Plants in Summer Green Broad-leaved Forest, JJKH20180350KJ), Scientific Investigation Report Project of Three Lakes National Nature Reserve of Songhua River of Jilin (ZCGJ-2020-FW006), and the Doctoral Scientific Research Foundation of Beihua University (Characteristics of Heavy Metal Pollution and Accumulation in Vegetable Soils in the Suburbs of Typical Industrial Cities).

**Data Availability** The data that support the findings of this study are not openly available due to the requirements of support project and are available from the corresponding author upon reasonable request.

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