Occurrence and distribution of organochlorine pesticides – lindane, *p,p'*-DDT, and heptachlor epoxide – in surface water of China

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ABSTRACT

Persistent organochlorine pesticides pollutants (OCPs) have been reported to occur at relatively high concentrations in some Chinese waters. In order to map the distribution of organochlorine pesticides in the surface water throughout China, samples were collected from over 600 sites in seven major river basins and three main internal rivers drainage areas during 2003 and 2004. The surface water samples were analyzed for the representative organochlorine pesticides contaminants including lindane (γ -HCH), *p,p'*-DDT and heptachlor epoxide. In general, the most frequently detected compound was lindane, being detected in 83.9% of samples (mean = 31.3 ng/l; range <0.17–860 ng/l), and the highest concentration was present in the Yellow River basin. *p,p'*-DDT was detected in 63.1% of the samples collected (mean = 14.6 ng/l; range <0.14–368 ng/l) with the highest concentration present in the Huaihe River basin. Heptachlor epoxide was detected in only 9.3% of water samples (range <0.11–10 ng/l). Measured concentrations for the three compounds were low and rarely exceed the environment quality standard for surface water of China. Lindane was more frequently detected at much higher concentrations in the rivers of northern China compared with those of southern China. The sites with higher concentration of lindane and *p,p'*-DDT mainly occurred in the Yellow River and Huaihe River basins, so the results of this investigation indicate that the organochlorine pesticide contamination of Yellow River and Huaihe River basins should be of particular concern relative to the other basins. When compared with other regions of the world, it appears that the Chinese surface water is moderately polluted by lindane and *p,p'*-DDT.

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1. Introduction

The continued growth in human population has created a corresponding increase in the demand for the Earth's limited supply of freshwater. Thus, protecting the integrity of our water resources is one of the most essential environmental issues of the 21st century. Recent decades have brought increasing concerns for potential adverse human and ecological health effects resulting from the production, use, and disposal of numerous chemicals that offer improvements in industry, agriculture and so on. Research has shown that many such compounds can enter the environment, disperse, and persist to a greater extent than first anticipated. Over the past few decades, the occurrence of organochlorine pesticides (OCPs) in the environment had been of great concern due to their persistent and long-range transportable nature as well as toxic biological effects (Tanabe et al., 1994; Wania and Mackay, 1999). In

May 2001, the “Stockholm Convention on Persistent Organic Pollutants” (POPs Treaty) that was adopted by the United Nations of Environmental Programme (UNEP) highlighted the need to control the global contamination produced by toxic environmental chemicals. The treaty promotes the global regulations on the production and usage of persistent OCPs, such as aldrin, dieldrin, endrin, heptachlor, chlordanes, hexachlorobenzene, mirex, toxaphene, PCBs and DDTs. While most of the developed countries have already banned or restricted the production and usage of these compounds, some developing countries still use OCPs for agricultural and the public health purpose (Monirith et al., 2003). Several studies have reported the elevated concentrations of OCPs in fishes, mussels and birds collected from Asian countries including India, Vietnam and China (Monirith et al., 2003; Kannan et al., 1995; Kunisue et al., 2003), thereby indicating the presence of significant source of OCPs in these regions. As the world's second largest pesticide manufacturer, it has been estimated that China produced 203,000–381,000 metric tons annually of technical-grade pesticides between 1985 and 1996 (Huang et al., 2000). The magnitude of contamination of these persistent organic pollutants (POPs) in surface water, however, had not been

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Fig. 1. Map of sampling sites distribution.

systematically investigated at the national scale of China. In order to determine the contamination status and the spatial distribution of organochlorine pesticides in China's surface water, lindane, heptachlor epoxide and *p,p'*-DDT were chosen as the representative compounds for analysis. Lindane, heptachlor epoxide and *p,p'*-DDT were widely recognized as endocrine disrupting chemicals, and they had been used widely in China historically, even now, a certain amount of heptachlor epoxide and *p,p'*-DDT and a large amount of lindane are still applied in China, although DDT had been banned to use from 1983. And now, the lindane and *p,p'*-DDT have been widely distributed in Chinese environment with significant accumulation even in remote ecosystems (Guruge and Tanabe, 2001; Iwata et al., 1994), and the average concentrations of OCPs in soils of north China were higher than those in south China (Cai et al., 2008). Adding the persistence and the ecological effects, the three compounds were chosen as representative to characterize the pollution status of organochlorine pesticides in Chinese surface water. In order to illustrate the contamination status of OCPs in Chinese surface water, samples collected from over 600 sites in seven major river basins and three internal rivers drainage areas were analyzed for lindane, heptachlor epoxide and *p,p'*-DDT.

2. Materials and methods

2.1. Sampling and sample pretreatment

Little data were available on the occurrence of lindane, heptachlor epoxide and *p,p'*-DDT at the onset of this investigation. Therefore, the sampling sites were chosen randomly along the individual river basin. The 623 sites sampled during 2003–2004 (Fig. 1), including 217 reservoirs and 406 rivers and lakes. Samples were collected from the seven major river basins, including the Yangtze River basin, the Yellow River basin, the Pearl River basin, the Songhua River basin, the Liaohe River basin, the Haihe River basin and the Huaihe River basin. Samples were also collected from rivers in Southeast, Northwest and Southwest internal rivers drainage areas. The global positioning system (GPS) was used to locate the sampling positions. The sites map distribution is shown in Fig. 1.

All samples were collected by hydrological bureau personnel using consistent protocols and procedures designed to obtain a representative samples using standard

depth and width integrating techniques (Shelton, 1994). At each site, a composite water sample was collected from about 4–6 vertical profiles. Water was passed through a 0.45 μm , baked, glass fiber filter in the field where possible, or else filtration was conducted in the laboratory. Water samples for chemical analysis were stored in precleaned-amber, glass bottles and collected in duplicate. The duplicate samples were used for backup purposes (in case of breakage of the primary sample) and for laboratory replicates. Following collection, samples were immediately chilled by ice and sent to the laboratory.

Aliquots of the sample (5.0 l), which were filtered through a 0.45 μm glass fiber membrane under vacuum and then surrogates (decachlorobiphenyl) were added before extraction, were extracted by solid phase extraction (SPE) following published procedures (Zhou et al., 2002; Zhang et al., 2002a,b). Briefly, the SPE cartridges were first conditioned with 10 ml of methanol followed by 2×5 ml of deionized water. Water samples were passed through the cartridges at a flow rate of 6 ml/min under vacuum and pesticides eluted with 6 ml of dichloromethane, followed by another 6 ml dichloromethane rinse of the surfaces. Residual water was removed by anhydrous Na_2SO_4 , the volume was reduced to dry by rotation evaporator and then add another 15 ml hexane, the new solution volume was reduced by evaporation under nitrogen gas to 0.4 ml and add the internal standard (2,4,5,6-tetrachloro-*m*-xylene), and then adjust to 0.5 ml.

2.2. Chemical analysis

All solvents used for sample processing and analysis (dichloromethane, ethyl acetate, hexane, acetone, methanol) were of HPLC grade (Supelco). Deionized water was prepared using a Milli-Q system (Millipore, Watford). Chemical standards of lindane, heptachlor epoxide and *p,p'*-DDT were obtained from Supelco (purity >99%). Working standards were prepared in hexane and the internal standards 2,4,5,6-tetrachloro-*m*-xylene, were added to each working standard. These solutions were further diluted with hexane to prepare calibration solutions.

Table 1

Recoveries, method detection limits (MDLs), and the relative standard derivation (RSD) of the analytical procedure

Pesticides	Mean recovery (%)	MDLs (ng/l)	RSD (%) ($n=6$)
γ -HCH	105.4	0.17	3.8
<i>p,p'</i> -DDT	97.2	0.14	11.6
Heptachlorine epoxide	116.4	0.11	12.6
Decachlorobiphenyl (surrogate)	86.7	–	–

Table 2The statistical summary of lindane (γ -HCH) concentrations in Chinese surface waters (ng/l)

River basins	Number of samples	Detection rate (%)	Mean concentration	Minimum concentration	90 th percentile	Maximum concentration
Songhuajiang River	40	17.5		<0.17	71.0	80.0
Liaohe River	58	69.0	46.9	<0.17	80.0	120.0
Haihe River	39	92.3	47.8	<0.17	80.0	110.0
Yellow River	50	94.0	75.4	<0.17	100.0	860.0
Yangtze River	150	98.7	12.6	<0.17	28.2	70.0
Huaihe River	39	97.4	41.1	<0.17	80.0	98.0
Pearl River	150	79.3	7.1	<0.17	14.0	48.0
Southeast drainage area rivers	74	97.3	12.9	<0.17	36.0	114.0
Northwest drainage area rivers	18	88.9	57.2	<0.17	80.0	80.0
Southwest drainage area rivers	5	0	0.0	<0.17	0.0	0.0
North China	244	75.4	47.1	<0.17	80.0	860.0
South China	379	89.4	11.4	<0.17	28.0	114.0
Overall detection rate	83.9%					
Overall average concentration	31.3					

An Agilent 6890 GC equipped with a micro-cell electron capture detector (μ ECD) was used for lindane, heptachlor epoxide and *p,p'*-DDT analysis. Peak confirmation was achieved with an Agilent 6890 GC coupled to a model 5973N MS detector in selected ion mode. The capillary column used was HP-5MS (30 m $\times$ 0.25 mm i.d. $\times$ 0.25 μ m film thickness). All the instruments and capillary column were from Agilent company of America. The carrier gases were helium and nitrogen for MS and μ ECD, respectively. The inlet was heated to 250 °C. The GC column temperature was programmed as follows: initially at 60 °C (equilibrium time 1 min), increased to 140 °C at the rate of 10 °C/min, then to 230 °C at 5 °C/min before reaching at 260 °C at the rate of 10 °C/min and then held for 10 min. The temperature of μ ECD was held at 280 °C. The MS temperature was set at 280 °C and the electron impact energy was 70 eV.

2.3. QA/QC procedures

The residue levels of OCPs were quantitatively determined by the internal standard method using peak area. The method detection limits (MDLs) of OCPs were taken to be 3:1 signal versus noise value (S/N). For every set of 10 samples, a procedural blank and a spiked sample with standards were run to check for the interference and cross-contamination. Table 1 illustrates the mean recoveries, MDLs and relative standard derivation (RSD%) of the method. The MDLs ranged from 0.11 ng/l for heptachlor epoxide to 0.17 ng/l for lindane. The spiked recoveries of OCPs using 1 ng of composite standards were in the range of 97.2%–116.4% with RSD% values ranging from 3.8% to

12.6%. These parameters confirmed the suitability of the analytical protocols. A field quality assurance protocol was used to determine the effect, if any, of field equipment and procedures on the concentrations of OCPs in water samples. Field blanks, made from laboratory-grade organic free water, were submitted for about 5% of the sites and analyzed for OCPs. Field blanks were subject to the same sample processing, handling, and equipment as the water samples.

2.4. Statistical analysis

Values for OCPs lower than the method detection limits (<MDL) were substituted with zero prior to statistical analysis. To detect the difference of OCPs concentrations between the north China and south China, the Student *t*-test was employed in this study. Software from Excel Statistics was used in this study.

3. Results and discussion

3.1. Lindane

In all the surface water samples collected, lindane was detected in 83.9% (MDL=0.17 ng/l) of the samples. The concentrations ranged from <0.17 to 860.0 ng/l, with a mean level of 31.3 ng/l (Table 2), and the 90% percentile value is 70 ng/l. For the seven major river basins and three internal rivers drainage areas, the detection rate of

Fig. 2. Distribution of lindane in Chinese surface water.

The statistical summary of *p,p'*-DDT concentrations in Chinese surface waters (ng/l)

River basins	Number of samples	Detection rate (%)	Mean concentration	Minimum concentration	90 th percentile	Maximum concentration
Songhuajiang River	40	5		<0.14	<0.14	50
Liaohe River	58	8.6		<0.14	<0.14	80
Haihe River	39	41		<0.14	60	60
Yellow River	50	26		<0.14	61	130
Yangtze River	150	89.3	13.4	<0.14	34.2	168
Huaihe River	39	71.8	41.3	<0.14	87.6	368
Pearl River	150	79.3	10.1	<0.14	18	184
Southeast drainage area rivers	74	98.6	24.1	<0.14	56	303
Northwest drainage area rivers	18	16.7		<0.14	50	60
Southwest drainage area rivers	5	0		<0.14	<0.14	<0.14
North China	244	27.4	16.3	<0.14	60	368
South China	379	86.0	14.3	<0.14	35.4	303
Overall detection rate	63.1%					
Overall average concentration	14.6					

The high detection frequency of lindane at the national scale should be mainly attributed to historical use of technical HCH widely in China. During the 1950s to 1980s, the production of technical HCH containing 12–14% lindane isomer was about 4.9 million tons and accounted for 33% of the total world production (Zhang et al., 2002c). Although the technical HCH was banned in 1983, the continued mobilization from long-term ‘sinks’, such as soil particles, into river systems may have contributed to present

The lindane pollution in north China river basins is more serious than that in the south China river basins. The much higher concentration sites mainly appeared in the

Fig. 3. Distribution of *p,p'*-DDT in Chinese surface water.

Table 4Comparison of *p,p'*-DDT, lindane, and heptachlor epoxide concentrations in surface water of China with others

	<i>p,p'</i> -DDT (ng/l)	Lindane (ng/l)	Heptachlor epoxide (ng/l)	Reference
Surface water (Czech Republic)	0.036	0.147	–	Vladimír et al. (2007)
Lakes (USA)	54	–	–	Moore et al. (2007)
Water reserve (Spain)	1.5–79.0	1.7–31.6	1.2–86.2	Salvaó et al. (2006)
Gange rivers (India)	ND	ND–290.0	–	Sankaramakrishnan et al. (2005)
Lake Victoria Basin (East Africa)	24–9,985,000	24–148	–	Nyangababo et al. (2005)
Bay of Ohuira (Mexico)	20–3750	20–270	20–230	Osuna-Flores and Riva (2002)
São Paulo state (Brazil)	<5–130	5–390	ND	Sandra et al. (2002)
Northern rivers (Greece)	ND–35	ND–81	ND	Golfonopoulos et al. (2002)
Gangetic alluvial plains (India)	BDL–79,820	BDL–39,510	–	Kunwar et al. (2007)
Ebro river basin (Spain)	<30	<15–129	ND	Claver et al. (2006)
Uluabat Lake (Turkey)	ND–222,000	ND–220,000	ND–205	Nurhayat et al. (2006)
Küçük Menderes River (Turkey)	ND–58	ND–398	ND–316	Turgut (2003)

Yellow River, Huaihe River, Northwest drainage area rivers, Liaohe River and Haihe River of the north China according to Fig. 2. In the previous works, lindane contamination had also been reported in some rivers of north China, such as in the Haihe River (Wang et al., 2003; Yang et al., 2005a,b) and in the Huaihe River (Wang et al., 1999). While in south China, relatively high concentration sites mainly occurred in estuaries of the Yangtze River, Minjiang River and Pearl River (Fig. 2), which was consistent with the previous investigations (Sun et al., 2002; Zhang et al., 2003). Many industries are located in these areas and may be important lindane sources in water.

When compared with other places of the world, the surface water of China is somewhat moderately polluted by lindane. When compared, the concentration of lindane in Chinese surface water may be two or three orders lower than those detected in Gangetic alluvial plains (BDL–39,510 ng/l) in India and Uluabat Lake (ND–220,000 ng/l) in Turkey (Nurhayat et al., 2006; Kunwar et al., 2007), but similar to Bay of Ohuira in Mexico (20–230 ng/l) and surface water of São Paulo state in Brazil (5–390 ng/l) (Osuna-Flores and Riva, 2002; Sandra et al., 2006), and the level of lindane in Chinese surface water was much higher than that in surface water of Czech Republic (0.147 ng/l), and in the water reservoir of Spain (1.7–31.6 ng/l) (Salvaó et al., 2006; Vladimír et al., 2007). The comparison results indicated that the surface water of China is somewhat moderately polluted by lindane.

3.2. *p,p'*-DDT

The test results of *p,p'*-DDT were summarized in Table 3. The detection frequency of *p,p'*-DDT was 63.1% (MDL=0.14 ng/l) for all the consistent water samples collected, the concentrations ranged from <0.14 to 368 ng/l, with a mean value of 14.6 ng/l (Table 3). For the seven major river basins and three main internal rivers drainage areas, the detection rate of *p,p'*-DDT was from 0% (Southwest drainage area) to 98.6% (Southeast drainage area), the second highest detection was the Yangtze River (89.3%), followed by the Pearl River (79.3%) and the Huaihe River (71.8%). The high detection in these rivers was consistent with the previous observations (Zhang et al., 2002b; Yang et al., 2005a,b; Wang et al., 1999; Yang et al., 2005b). The *p,p'*-DDT detection rates of Songhuajiang River, Liaohe River, Haihe River and Yellow River are less than 50%, so the statistical analysis had not been done for these rivers. Among the other basins, the mean concentration of the Pearl River is the lowest (mean=10.1 ng/l) and that in the Huaihe River is the highest (mean=41.3 ng/l), the second highest level occurred in the Southeast internal river drainage area (mean=24.1 ng/l), followed by the Yangtze river (mean=13.4 ng/l). The highest *p,p'*-DDT concentration was detected in the samples from Huaihe River (368 ng/l). These results suggested that the Huaihe River basin was the most seriously polluted by *p,p'*-DDT. The statistical analysis result indicated that there was no significant difference in concentrations between the north China rivers and the south China rivers ($t=1.96$, $p=0.046$).

The high detection frequency of *p,p'*-DDT in the Chinese surface water may be explained by its historic use. As an agricultural country, the DDT was widely used and the total Chinese production of technical DDT was about 0.4 million tons which accounted for 20% of world production (Zhang et al., 2002c) before DDT was banned in 1983. Because of its physicochemical properties, DDT could exist in the environment for a longer period. In addition, the widely applied pesticide dicofol containing 20% *p,p'*-DDT continues to be used on cotton and fruit crops, which may be another factor contributing to the *p,p'*-DDT high detection.

The fact with high *p,p'*-DDT concentration sites are mainly distributed in the Yellow River, and the presence of DDT residues in Huaihe River, Yangtze River, Minjiang River estuary and Pearl river estuary (Fig. 3) indicated that DDT sources still exist in these areas. The Yellow River, Huaihe River and Yangtze River basins are important agricultural areas with an extremely large numbers of agricultural farmlands. Pesticides used yearly on a regular basis, including the pesticide dicofol, which may be a contributing factor for the elevated levels of *p,p'*-DDT concentrations. While for the Minjiang River, Yangtze River and Pearl River estuaries, the three estuaries are important industrial regions and industrial sources probably explain the increased *p,p'*-DDT levels detected at these locations. The results of Minjiang and Pearl River estuaries in this study are similar to those of earlier studies of *p,p'*-DDT contamination in the same estuaries (Zhang et al., 2002b; Zhang et al., 2003; Maskouli et al., 2005; Liu et al., 2003), which suggested that the sources exist around the three estuaries.

According to Table 4, the concentrations of *p,p'*-DDT (<0.14–368 ng/l, with a mean of 31.3 ng/l) in Chinese surface water are two to three orders of magnitude lower than those reported for some of the heavily polluted sites, such as Lake Victoria Basin of East Africa (24–9,985,000 ng/l) (Nyangababo et al., 2005), surface water of Gangetic alluvial plains in India (BDL–79,820 ng/l) (Sankaramakrishnan et al., 2005), and the Uluabat Lake in Turkey (ND–222,000 ng/l) (Nurhayat et al., 2006). While the concentrations of *p,p'*-DDT in Chinese surface water are also similar to São Paulo state in Brazil (<5–130 ng/l). However, the concentrations of *p,p'*-DDT in Chinese surface water are higher than those in some places of the world, such as surface water in Czech Republic (0.036 ng/l) (Vladimír et al., 2007), northern rivers in Greece (ND–35 ng/l) (Golfonopoulos et al., 2003), more results are shown in Table 5. The comparison results indicate that the Chinese surface water is also somewhat moderately polluted by *p,p'*-DDT.

3.3. Heptachlor epoxide

The concentrations of heptachlor epoxide in the surface water samples are presented in Table 5. The overall frequency of detection was quite low, only 9.3% (MDL=0.11 ng/l), and the concentrations range from <0.11 to 10 ng/l, with no statistical mean value at the national scale. For the seven major river basins and three main

Table 5

The statistical summary of heptachlor epoxide concentrations in Chinese surface waters (ng/l)

River basins	Number of samples	Detection rate (%)	Mean concentration	Minimum concentration	90 th percentile	Maximum concentration
Songhuajiang River	40	0		<0.11	<0.11	<0.11
Liaohe River	58	10.3		<0.11	3	10
Haihe River	39	7.7		<0.11	<0.11	10
Yellow River	50	18		<0.11	10	10
Yangtze Yangtze River	150	4.7		<0.11	<0.11	10
Huaihe River	39	20.5		<0.11	6	10
Pearl River	150	6.0		<0.11	<0.11	4
Southeast drainage area rivers	74	17.6		<0.11	2	10
Northwest drainage area rivers	18	16.7		<0.11	10	10
Southwest drainage area rivers	5	0		<0.11	<0.11	<0.11
Overall detection rate	9.3%					
Overall average concentration	–

Fig. 4. Distribution of heptachlor epoxide in Chinese surface water.

internal rivers drainage areas, the detection frequency of heptachlor epoxide was from 0% (Southwest drainage area, Songhuajiang River basin) to 20.5% (Huaihe River). The second highest detection was the Yellow River (18.0%), followed by the Southeast internal rivers (17.6%) and the Northwest internal rivers (16.7%). As the detection rate of heptachlor epoxide was less than 50% of the water samples at any given area, statistical analysis of the data was considered to be inappropriate and mean values are not reported. As illustrated in Fig. 4, samples containing detectable levels of heptachlor epoxide mainly occurred in the Yellow River and Huaihe River.

In general, heptachlor epoxide had limited use as a pesticide and there was limited available information about its historic use in China. From the result of concentration, the conclusion could be drawn that there was a lightly pollution in Chinese surface water because of its low detection frequency and low concentration. According to Table 4, the level of heptachlor epoxide was lower than that in most place of the world, which also indicates that the surface water of China was only lightly polluted by heptachlor epoxide.

Among the three compounds, the rank of detection frequency was lindane (83.9%) > *p,p'*-DDT (63.1%) > heptachlor epoxide (9.3%), and the rank of mean concentration was also lindane (31.3 ng/l) > *p,p'*-DDT (14.6 ng/l) > heptachlor epoxide. From the result, the detection frequency and mean concentration of lindane were all the highest among the three compounds. The concentrations measured of the three compounds in this study included just the dissolved phase. While they are the hydrophobic compound, studies had shown that the concentration should be associated with particulate matter (Venkatesan and Kaplan, 1990). Thus, the concentration and frequency of detection for selected compounds would likely have been reduced because of the sample filtration.

4. Conclusion

The present study reports the contamination status of lindane, *p,p'*-DDT and heptachlor epoxide in Chinese surface water and provided data on the levels of the three organochlorine pesticides. In general, the measured concentrations for the three compounds were low and rarely exceed the environment quality standard for surface water of China. For the three compounds, the lindane and *p,p'*-DDT are the major residue contaminants with higher detection frequency and concentration relative to heptachlor epoxide, and the detection frequency and concentration of lindane in surface water of China are much higher than those of *p,p'*-DDT and heptachlor epoxide. Comparison with other places of the world, the surface water of China was somewhat

moderately contaminated by lindane and *p,p'*-DDT, while the surface water in China was only lightly polluted by the heptachlor epoxide because of its low concentration and detection frequency. In north China, the lindane and *p,p'*-DDT contamination mainly occurred in the Yellow River and Huaihe River basins. While in south China, the lindane and *p,p'*-DDT contamination mainly occurred in the estuaries of Yangtze River, Pearl River and Minjiang River. It is submit that the Yellow River basin, Huaihe River basin and estuaries of Yangtze River, Pearl River and Minjiang River should be given priority in terms of the prevention and control of contamination.

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