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Identification of Trace Organic Pollutants in Drinking Water and the Associated Human Health Risks in Jiangsu Province, China

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Abstract Organochlorine pesticides (OCPs), polycyclic aromatic hydrocarbons (PAHs), dibutyl phthalate (DNBP) and di-2-ethylhexyl phthalate (DEHP) were all detectable in surface water in Jiangsu Province, China. Concentrations of OCPs ranged from 5.13 to 8.15 ng/L. PAHs were found ranging from 14.7 to 24.5 ng/L. Concentrations of DNBP and DEHP ranged from 16 to 5,857.5 ng/L and 556 to 15,670.7 ng/L, respectively. Greater than 70 % of chemicals were removed in water treatment processes. The carcinogenic risks posed by trace organic pollutants through tap water ingestion were lower than 10^{-6}, and the non-carcinogenic risks were less than 10^{-5}.

Keywords Surface water · Tap water · Organic pollutants · Health risk

Jiangsu Province is one of the most developed provinces in China. Large amounts of organic chemicals in industrial wastewater are discharged into the surface water (Xing et al. 2005). Jiangsu Province has also had rice fields for hundreds of years. In these fields, organochlorine pesticides (OCPs) were widely used in the 1960s (Li et al. 2001; Qu et al. 2011). Organic pollutants have been detected in the water sources of this region (Shi et al. 2010). Previous studies have indicated that filtration and coagulation of water have limited effectiveness in removing organic pollutants (Choi et al. 2006; Kim et al. 2007). The residues of organic pollutants were still measurable in tap water.

Trace organic pollutants in water may pose potential health risks to the public (Chowdhury and Hall 2010; Shi et al. 2012). Nevertheless, little attention has been given to the risk posed by these pollutants (Wu et al. 2010). The areas that were selected for study include Binhu (BH), Chanshu (CS), Wojin (WJ), Pexian (PX) and Dafen (DF) (Fig. 1). BH is located in the northeastern region near Taihu Lake, and the lake is a primary water source for this region. Domestic and industrial waste water, primarily from the city of Wuxi, is discharged into the lake. CS is located in the southeastern region near Taihu Lake and is most notable for its petrochemical and electrical industries. WJ is located in a highly urbanized region near Yangtze River. PX and DF are much less industrialized, and their primary water source is ground water. The two primary objectives of this research are to detect the environmental concentrations of PAHs, OCPs, DNBP and DEHP in surface water and tap water in Jiangsu Province and to assess the potential health risk posed by these pollutants.

Materials and Methods

To detect the concentrations of the trace organic pollutants, surface water near water treatment plants and related tap water were collected from BH, CS and WJ in October 2010. For the locations in PX and DF, which obtain source water from groundwater, only tap water was collected. A volume of 4 L of water was collected in brown glass bottles that had been rinsed with high-purity acetone (TEDIA), methanol (TEDIA) and the water samples. Samples were stored at 4°C prior to extraction and were analyzed within 24 h of collection to

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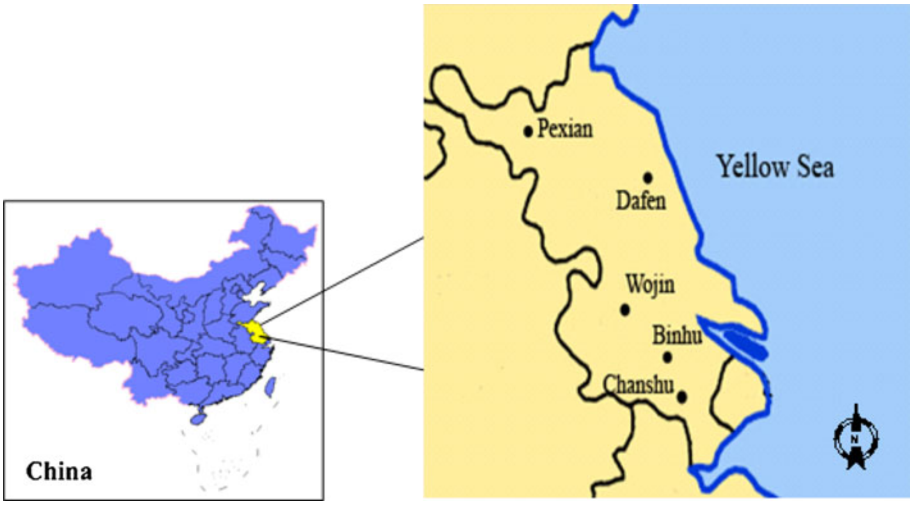

Fig. 1 Map of sampling points in Pexian, Dafen, Wojin, Binhu and Chanshu

minimize alteration of sample constituents and to avoid the need to preserve or stabilize samples further. OCP and PAH compounds were obtained from Sigma-Aldrich (Poole, Dorset, UK), and the purity was 99.5 %. DNBP and DEHP were obtained from Labor Dr. Ehrenstorfer-Schäfers (Augsburg, Germany), and the purity was 99.0 %.

Detailed descriptions of the extraction and fractionation procedures have been presented elsewhere (Kuster et al. 2010). Briefly, water samples were passed through solid-phase extraction cartridges (500 mg Oasis HLB cartridge; Waters, Milford, MA, USA) and eluted with 10 ml of hexane and dichloromethane (1:1) followed by 8 ml of methanol. Extracts were concentrated by rotary evaporation. Chemicals were analyzed using a Thermo DSQ single quadrupole GC/MS (San Jose, CA, USA). The detection limits of PAHs, OCPs, DNBP and DEHP for this method were 0.1, 0.01, 1 and 1 ng/L, respectively. The average rates of recovery of individual PAHs ranged from 62.4 to 113.2 %. Recoveries of OCPs, DNBP and DEHP were between 87 and 103 %.

Ingestion of tap water would be the primary pathway through which residents are exposed to organic pollutants. To identify potential health risks posed by the ingestion of trace organic pollutants, guidelines for exposure assessment from USEPA were applied in this study. The exposure dose was calculated as follows (USEPA 1989).

$$\mathrm{CDI} = \frac{\mathrm{C} \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}}$$

where CDI reprensts the chronic daily dose (mg/kg-day); C the chemical concentration (mg/L); IR the water ingestion rate (L/day); EF the exposure frequency (day/year); ED the exposure duration (year); BW the body weight (kg) and AT the averaging time (day).

The lifetime carcinogenic risk posed by ingestion of tap water was calculated according to the following equation (USEPA 1989):

$$\mathrm{R} = \mathrm{CDI} \times \mathrm{SF}$$

where R is the probability of an individual developing cancer over a lifetime; SF is the carcinogenicity slope factor (kg-day/mg).

The noncarcinogenic risk is usually characterized by risk quotient (RQ), which is defined as the quotient of the chronic daily dose divided by the reference dose of a specific chemical. It was calculated using the following equation (USEPA 1989):

$$\mathrm{RQ} = \frac{\mathrm{CDI}}{\mathrm{RfD}}$$

where RfD is the reference dose (mg/kg-day).

For a mixture of contaminants with a similar mode of action, the risk quotient of the mixture was characterized as the sum of RQs.

The ingestion rate of drinking water (IR) and the body weight (BW) were calculated according to values reported in the literature (Bennett et al. 1999; Zhao and Li 2009). The carcinogenicity slope factors (SF) and reference dose (RfD) were obtained from the Integrated Risk Information System of the EPA. For carcinogenicity risk, 10^{-6} (one in million) was taken as an acceptable cancer risk criterion (US EPA 1992). As for noncarcinogenic effect, an HQ value greater than one indicated that the potential for an adverse health effect existed and that further investigation was needed.

Results and Discussion

Most of the OCPs that were detected were found in surface water samples ranged from 5.13 to 8.15 ng/L (Table 1). HCHs were the predominant contaminants in surface water at sites in CS and BH, with levels ranging from 3.87 to

Table 1 Concentrations of primary contaminants (ng/L)

Chemicals	Locations							
	WJ-Sur	WJ-Tap	CS-Sur	CS-Tap	BH-Sur	BH-Tap	PX-Tap	DF-Tap
α-HCH	0.35	N.D	N.D	N.D	0.2	0.01	N.D	N.D
β-HCH	0.62	N.D	0.05	0.01	1.03	0.01	N.D	N.D
γ-HCH	2.54	0.01	4.22	0.01	5.28	0.02	N.D	N.D
δ-HCH	0.36	N.D	0.05	0.01	0.23	0.11	N.D	N.D
p, p′-DDE	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D
o, p′-DDT	2.38	0.01	0.11	0.02	0.05	0.01	N.D	0.01
p, p′-DDD	0.09	0.01	0.63	0.02	0.11	0.02	N.D	0.05
p, p′-DDT	1.81	0.02	0.07	0.01	0.08	0.01	N.D	0.01
Naphthalene	6.5	N.D	1.5	1.0	0.8	0.1	N.D	0.8
Acenaphthylene	1.5	N.D	0.8	0.7	1.0	N.D	N.D	1.2
Acenaphthene	1.2	N.D	2.2	1.0	1.8	0.1	0.1	2.1
Fluorene	4.7	N.D	4.7	0.8	2.8	0.4	N.D	1.7
Phenanthrene	6.5	N.D	6.6	0.3	2.5	0.4	N.D	1.5
Anthracene	1.6	N.D	1.0	0.3	0.5	0.1	N.D	1.0
Fluoranthene	1.9	N.D	2.3	0.1	0.7	0.2	N.D	0.6
Pyrene	0.5	N.D	0.7	0.1	0.4	0.2	N.D	0.5
Benz[a]anthracene	N.D	N.D	0.1	N.D	0.1	0.1	N.D	0.1
Chrysene	0.1	N.D	0.5	N.D	0.7	0.3	N.D	0.2
Benzo[b]fluoranthene	N.D	N.D	0.2	N.D	1.5	1.2	N.D	0.1
Benzo[k]fluoranthene	N.D	N.D	N.D	N.D	0.5	0.3	N.D	0.1
Benzo[a]pyrene	N.D	N.D	0.1	N.D	0.6	0.4	N.D	N.D
Indeno[1,2,3-c,d]pyrene	N.D	N.D	N.D	N.D	N.D	N.D	N.D	0.1
Dibenz[a,h]anthracene	N.D	N.D	N.D	N.D	N.D	N.D	N.D	0.1
Benzo[g,h,i]perylene	N.D	N.D	0.1	N.D	0.8	0.4	N.D	0.1
DNBP	66	37	5,857.5	82	16	10	29	22
DEHP	642	55	15,670.7	97	556	46	61	40

N.D not detectable

6.74 ng/L being observed. Generally, γ-HCH contributed more than 65% of the total HCHs in all the surface water samples and was generally 1–2 orders of magnitude higher than other HCHs. Importantly, γ-HCH was produced and used extensively in the past decades in this region for pesticides in rice fields. The ratios of α-HCH to γ-HCH in water sources were generally less than 1, which indicated the use of lindane, rather than technical mixtures. Although γ-HCH was banned in China in 2009, it could still be detected in various locations (Li et al. 2001). The highest concentration of HCHs was found in the surface water from BH, which is a region that consists largely of farmland.

DDTs showed a large variation in concentrations in various species and among different locations. Concentrations of total DDTs in the sampling locations ranged from 0.24 to 4.28 ng/g, with o, p′-DDT and p, p′-DDT being the predominant species. The highest concentration of DDTs was detected in WJ surface water. Although widespread application and the production of DDTs have been banned in China since the 1980s, the long half-life of the class of chemicals still warrants continuous environmental monitoring. Moreover, DDTs account for 3 % of the pesticide dicofol, which is still in use. The use and production of dicofol may cause the introduction of new DDTs into the environment.

The total concentration of DDTs and HCHs in surface water samples from WJ, CZ and BH was lower than the national environmental quality limit for surface water (1,000 ng/L for DDTs and 5,000 ng/L for HCHs) that was regulated by the Ministry of Environmental Protection of China.

The OCP concentrations in tap water samples from WJ, CS and BH were much lower than those in the surface water. More than 70 % of OCPs were removed by water treatment processes, including filtration, coagulation, chlorination and ozonation. The results indicate that the water treatment techniques used in treatment plants were effective in reducing the detected contaminants. However, certain contaminants were still detectable in tap water, and further treatment should be performed to minimize the potential for exposure.

PAHs were found in all surface water samples, ranging from 14.7 to 24.5 ng/L. Flu and Phe were dominant. The highest concentration of PAHs was detected in WJ. PAHs levels in this study were lower than previously reported levels in surface water from Zhejiang Province, China (70.3–1,844.4 ng/L) (Guo et al. 2007). PAHs were also detectable in some tap water samples except at sites in WJ. Concentrations of PAHs ranged from 0.1 to 10.2 ng/L. The total concentration of seven potentially carcinogenic PAHs (B[a]A, Chr, B[b]F, B[k]F, B[a]P, Ind, DBA) ranged from below the detection limit to 3.4 ng/L. Elevated concentrations of PAHs were observed in tap water from DF. Moderately high concentrations of PAHs were found in tap water from CS and BH. However, none of the tap water or source water samples had PAH levels that exceeded the national limits.

The relative contributions of PAHs with different numbers of rings varied among different surface water samples. Three -ring PAHs, such as Acy, Ace, Phe, Flu and Ant, contributed over 50 % to the total PAHs concentrations. These findings indicate that oil combustion is likely to be the main source of these contaminants because other studies have found that oil-fired power generation stations were characterized by high levels of three-ring PAHs, especially Phe (Masclet et al. 1986).

Concentrations of DNBP and DEHP in surface water ranged from 16 to 5,857.5 ng/L and 556 to 15,670.7 ng/L, respectively. The highest concentrations were found in CS. Moderate levels of DNBP and DEHP (ranging up to 82 and 97 ng/L) were also detected in tap water. Previous studies indicated that treated water from water treatment plants in Beijing contained up to 28.9 μg/L of DNBP and DEHP. The concentrations of DNBP found in treated drinking water in Chongqing (17.3 μg/L) and Hangzhou (76 μg/L) were also higher than those in this study (Cai et al. 2003; Zhao et al. 2010).

Because organic pollutants were detected at various sites in Jiangsu Province, it is necessary to assess health risks associated with ingestion of drinking water. Carcinogenic and noncarcinogenic risk assessments were conducted for various trace organic pollutants that were detected in tap water. The estimated carcinogenic risks were presented in Table 2. The risks in all of the locations were less than 10^{-6}. The overall carcinogenic risks were found to be the highest in BH (8.75×10^{-8}). We also estimated the health risks for children. The children's carcinogenic risks were somewhat lower than the risk for adults and ranged from 0 to 2.71×10^{-8} for male children and 0 to 2.65×10^{-8} for female children. The highest risk was also found in BH. PAHs contributed approximately 97 % of the total carcinogenic risks in BH for male children. Among the seven potentially carcinogenic PAHs, B[a]P contributed approximately 73 % to the total risk (Fig. 2). The estimated noncarcinogenic risks for adults and children were both less than 10^{-5}.

In conclusion, concentrations of OCPs, PAHs, DNBP and DEHP were measured in surface water and tap water in five cities in Jiangsu Province, China. OCPs were detectable in all of the surface water samples. γ-HCH contributed more than 65 % of the total HCHs and its concentrations were generally 1–2 orders of magnitude higher than those of the other HCHs. Concentrations of total DDTs in samples from the various locations ranged from 0.24 to

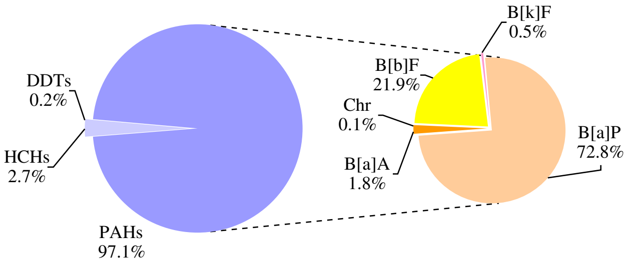

Fig. 2 Analysis of individual chemical contribution to carcinogenic risks in BH (male children)

Table 2 Human health risk assessment results

Locations	Carcinogenic risk				Noncarcinogenic risk			
	Male		Female		Male		Female	
	Child	Lifetime	Child	Lifetime	Child	Lifetime	Child	Lifetime
WJ-Tap	1.50E−10	4.85E−10	1.47E−10	5.13E−10	2.48E−6	1.60E−6	2.42E−6	1.69E−6
CS-Tap	2.65E−10	8.56E−10	2.59E−10	9.05E−10	4.96E−6	3.21E−6	4.85E−6	3.39E−6
BH-Tap	2.71E−8	8.75E−8	2.65E−8	9.26E−8	3.89E−6	2.52E−6	3.81E−6	2.66E−6
PX-Tap	0	0	0	0	5.63E−8	3.64E−8	5.51E−8	3.85E−8
DF-Tap	6.57E−9	2.12E−8	6.43E−9	2.25E−8	5.82E−6	3.77E−6	5.70E−6	3.98E−6

4.28 ng/g, with o, p′-DDT and p, p′-DDT being the predominant species. Concentrations of PAHs measured in surface water varied among sampling locations. Three-ring PAHs were predominant, which indicated that the pollution may have derived from oil-fired power generation stations. Concentrations of DNBP and DEHP in surface water ranged from 16 to 5,857.5 ng/L and 556 to 15,670.7 ng/L, respectively, with the highest concentration being found in CS. Concentrations of the contaminants in tap water were much lower than those in the surface water. More than 70 % of the chemicals that were detected were removed in water treatment processes. This study also estimated the associated carcinogenic and noncarcinogenic human health risks from ingestion of tap water. The total carcinogenic risks were found to be highest in BH (8.75×10^{-8}).

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