Environmental Monitoring and Assessment (2006) **123:** 351–370
DOI: 10.1007/s10661-006-9202-0

THE STATUS OF PESTICIDE RESIDUES IN THE DRINKING WATER SOURCES IN MEILIANGWAN BAY, TAIHU LAKE OF CHINA

TA NA, ZHOU FANG, GAO ZHANQI, ZHONG MING and SUN CHENG*
State Key Laboratory of Pollution Control and Resouce Reuse, School of Environment,
Nanjing University, Nanjing 210093, China
*(*author for correspondence, e-mail: chengsun@public1.ptt.js.cn)*

(Received 27 October 2005; accepted 16 January 2006)

Abstract. The study was carried out to assess the levels of pesticide residues in the water of Meiliangwan Bay, Taihu Lake of China. The most commonly employed organochlorine pesticides (OCPs), organophosphorus pesticides (OPPs) and herbicide atrazine were analyzed. The water samples were collected seasonally from Meiliangwan Bay within a period of one year. The pesticides were analyzed by gas chromatography (GC) with μECD or NPD after solid-phase extraction (SPE), which was confirmed by GC with an ion trap mass spectrometry (MS). The mean concentrations were 1.98 ng/l for lindane, 0.378 ng/l for heptachlor epoxide, 0.367 ng/l for p,p′-DDE, 0.496 ng/l for p,p′-DDD, 1.06 ng/l for p,p′-DDT and 51.6 ng/l for dichlorvos, 39.0 ng/l for demeton, 346 ng/l for dimethoate, 4.12 ng/l for methyl parathion, 11.6 ng/l for malathion, 2.17 ng/l for parathion and 217 ng/l for atrazine. Generally, low concentrations of OCPs were found, whereas the concentrations of the OPPs and atrazine in the water of Taihu Lake were relatively high. Heptachlor epoxide and lindane were the two most commonly encountered OCPs while dichlorvos, demeton and dimethoate were found to have much higher concentrations and occurrences than other OPPs.

Keywords: Taihu Lake, pesticide, solid phase extraction, gas chromatography-mass spectrometry, drinking water sources

1. Introduction

Taihu Lake is a typical shallow freshwater lake with an area of 2338 km^2 and an average depth of 1.9 m (Qiu *et al.*, 2004), which is one of the five largest lakes in China. The Meiliangwan Bay locates in the north of Taihu Lake and provides drinking water supply for Wuxi City. Its water quality has been influenced by the discharge of industrial waste water, domestic sewage and agricultural pollutants from Taihu Lake Area (TLA) which is one of the most developed regions and densely populated area in China. However, the water pollution management has not kept pace with the rapid development of the economy and the intensive use of water sources, as a result the water quality of Taihu Lake has been deteriorated seriously (Zhu *et al.*, 2002). The economic loss caused by the water pollution is approximately 1.2 billion US dollar per year (Wang *et al.*, 2003), which has great influence on sustainable development of TLA. The water contamination has threatened safety of the drinking water supply of millions of people living in Wuxi City.

One of the main reasons of the water quality deterioration may be due to the pesticide application in this region. The applied pesticides may be released to the aquatic environment via soil percolation, air drift or surface run-off, etc., polluting the natural water and affecting abundance diversity of non-target species as well as producing complex effects on the ecosystems (Rovedattil *et al.*, 2001).

TLA is one of the most developed agriculture areas with high-yield and high-intensity pesticide application in China (Wang *et al.*, 2005). The pesticides have been detected in various environmental matrices of this area such as soil, air, sediment, fish and eggs of night heron (Feng *et al.*, 2003; Gong *et al.*, 2001). Recently, the pesticide residues in Taihu Lake are of great concern in China. However, up to now, few systematic researches have been reported regarding the OCPs, OPPs and atrazine pollutions in Taihu Lake.

In order to understand the present pollution status of the pesticides in the water of Taihu Lake and to recommend future rationalization of controlling, reducing and eliminating releases of the pesticides in TLA, the pesticides in water of Meiliangwan Bay, Taihu Lake were investigated seasonally within a period of one year. Results of this study should be of value not only to pollution control of pesticides in TLA, but also to guarantee the safety of drinking water.

2. Materials and Methods

2.1. MATERIALS

OCPs standard was a mixed solution (2000 μg/l) in toluene:hexane (50:50) including 17 organochlorine pesticide components, but only five of them were listed here, which were investigated in this study: lindane(99.8%), p,p′-DDD(99.4%), p,p′-DDE(99.6%), p,p′-DDT(98.9%), heptachlor epoxide (99.9%), OPPs were as following: dichlorvos (99.6%), trichlorfon (97.8%), demeton O&S, (30.9%O, 60.1%S), dimethoate (99.9%), methyl parathion (99.8%), malathion (97.3%), parathion (99.0%), atrazine (99.0%), decachlorobiphenyl (99.0%, internal standard) and pentachloroanisole (97%, internal standards). The pesticide standards and internal standards mentioned above were purchased from Supelco, Bellefonte, PA, USA.

The HPLC grade solvents (*n*-hexane, dichloromethane, isopropanol and methanol) used in the analysis were purchased from Tedia, Fairfield, OH, USA. Anhydrous sodium sulfate (analytical grade) was purchased from Nanjing Chemical Factory.

2.2. SAMPLE COLLECTION

The water samples under the surface of 0.5 m were collected from 10 different sampling sites in Meiliangwan Bay by the aid of Global Position System. Figure 1

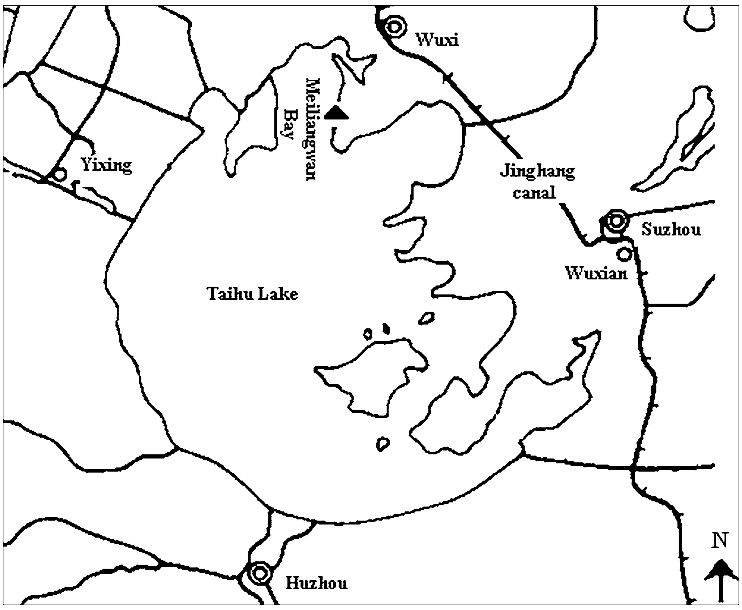

Figure 1. Map of Taihu Lake and the location of Meiliangwan Bay.

shows the map of Taihu Lake and the location of Meiliangwan Bay. The water samples were collected three times within one year period which covered the normal season (Nov. 2003), the dry season (Jan. 2004) and the rainy season (Aug. 2004).

Before sampling, glass bottles were washed with detergent, tap water, distilled water and the sample water finally. The glass bottle was filled to the top with the sample water to eliminate air bubbles. The water samples were acidied to pH 2.0 firstly with sulfuric acid to inhibit biological activity and then was carried to the laboratory and stored at 4 °C, followed by extraction within 48 h.

2.3. SAMPLE PREPARATION

The water samples were prefiltered through 0.45 μm filter to eliminate particulate matters and were adjusted to pH 6.0–7.0 with sodium hydroxide solution. The Supelclean ENVI$^{\text{TM}}$-18 SPE tube contains 1 mg C18 bonded silica with average particle size 56.0 microns and total volume is 6 ml which was purchased from Supelco, Bellefonte, PA, USA and was used for the sample preparation. Before enrichment, the SPE tubes were conditioned with 10 ml of methanol and then 10 ml of distilled water and stored in distilled water till collection of pesticides from water samples. 1 L water samples were mixed well and trapped through the SPE tubes with a flow rate of 5 ml/min under vacuum. The SPE tubes enriched pesticides were eluted by 5 ml different solvents twice respectively for different target pesticides, and then the collected eluent was evaporated in an evaporator.

This solution was further concentrated by a gentle stream of N_2 to 100 μl. The mixture of *n*-hexane- isopropanol (20:1, v/v), the mixture of dichloromethane – isopropanol (9:1, v/v) and methanol were optimized to elute OCPs, OPPs and atrazine, respectively.

2.4. ANALYTICAL PROCEDURE

A known amount of internal standard was added to the above final extracts for analysis of OCPs and atrazine or not added internal standard for analysis of OPPs, and then the extract was transferred to a glass micro vial. Decachlorobiphenyl and pentachloroanisole were applied as internal standards for analysis of OCPs and atrazine, respectively. However, without finding a suitable internal standard, the external standard method was applied for of OPPs. 1.0 μl of the final extracts was automatically injected into Agilent 6890 GC with different detectors and columns for different pesticides analysis. μECD was chosen as a detector for analysis of OCPs and atrazine, NPD was chosen as a detector for analysis of OPPs. GC columns, detectors, oven temperature programs and various gas flow rate were listed in the Table I.

Analysis of the final extracts was also conducted by a Thermo Finnigan Trace GC interfaced with a PolarisQ ion trap mass spectrometry (GC-MS). The extracts were automatically injected into GC-MS with DB-5 fused-silica capillary column (30 m × 0.25 mm i.d., 0.25 μm film thickness). The instrument was operated with the Xcalibur software from Finnigan. The conditions of the analysis were as follows: injection volume was 1.0 μl with splitless mode, injector temperature was set at 250 °C and transfer line temperature was at 200 °C. The carrier gas was helium with flow rate of 1.5 ml/min at constant flow with vacuum compensation. Oven temperature program of GC-MS was as same as of GC for the corresponding pesticides. The MS was operated with a 70-eV electron impact (EI) mode and the scan range is 50–650 unit. The selected ion monitoring (SIM) mode was chosen to confirm the target pesticides by typical ions for each pesticide. The retention time, the selected ions and the monitoring time range for each pesticide were shown in Table II.

3. Results and Discussion

More than one hundred water samples from Meiliangwan Bay of Taihu Lake were analyzed from Nov. 2003 to Aug. 2004 for 14 different kinds of organic pesticides and their metabolites. Representative GC chromatograms of standard solutions and water samples are shown in Figures 2–4. The concentration of pesticide in different water seasons and the data summary were presented in Tables III–VI respectively. Additionally, the mean fortified recovery ranges and

TABLE I
GC columns and detectors and conditions

Conditions	Organochlorine	Organophosphorus	Atrazine
Column	DB-1 60 m × 0.25 mm i.d. × 0.25 μm	DB-17 30 m × 0.25 mm i.d. × 0.25 μm	DB-1 60 m × 0.25 mm i.d. × 0.25 μm
Detector	ECD	NPD	ECD
Oven temperature program	$60\,^\circ\mathrm{C} \xrightarrow{25^\circ\mathrm{C/min}} 170\,^\circ\mathrm{C}$ $\xrightarrow{4^\circ\mathrm{C/min}} 190\,^\circ\mathrm{C}$ $\xrightarrow{8^\circ\mathrm{C/min}} 230\,^\circ\mathrm{C}$ (4 min) $\xrightarrow{1^\circ\mathrm{C/min}} 240\,^\circ\mathrm{C}$ $\xrightarrow{25^\circ\mathrm{C/min}} 300\,^\circ\mathrm{C}$ (7 min)	$60\,^\circ\mathrm{C} \xrightarrow{20^\circ\mathrm{C/min}} 100\,^\circ\mathrm{C}$ $\xrightarrow{15^\circ\mathrm{C/min}} 190\,^\circ\mathrm{C}$ $\xrightarrow{4^\circ\mathrm{C/min}} 240\,^\circ\mathrm{C}$ (2 min)	$60\,^\circ\mathrm{C} \xrightarrow{10^\circ\mathrm{C/min}} 200\,^\circ\mathrm{C}$ (2 min) $\xrightarrow{4^\circ\mathrm{C/min}} 240\,^\circ\mathrm{C}$ $\xrightarrow{10^\circ\mathrm{C/min}} 280\,^\circ\mathrm{C}$ (2 min)
Injector temperature	250 °C	220 °C	250 °C
Detector temperature	320 °C	320 °C	300 °C
Carrier gas flow	1.6 ml/min	4 ml/min	1.1 ml/min
Make-up gas flow	60 ml/min	100 ml/min	60 ml/min

TABLE II
The retention time, selected ions and monitoring time range

Pesticide	Retention time (min)	Selected ions	Time range (min)
Lindane	9.08	183,219	7.50–10.00
Heptachlor epoxide	12.96	353,355	12.50–13.50
p,p′-DDE	14.49	246,279	14.20–14.80
p,p′-DDD	15.66	235,237	15.50–15.90
p,p′-DDT	16.84	165,239,272	16.50–18.80
Dichlorvos	5.35	185,109,187	5.00–8.00
Demeton-o	8.41	88,171,60	8.00–9.50
Dementon-s	9.45	88,171,60	9.50–11.00
Dimethoate	9.56	125,93,87	9.50–11.00
Methyl-parathion	11.36	263,125,109	11.00–12.00
Malathion	12.26	173,127,99	12.00–12.50
Parathion	12.63	291,109,97	12.50–13.00
Atrazine	11.77	217,215,200	11.50–12.00

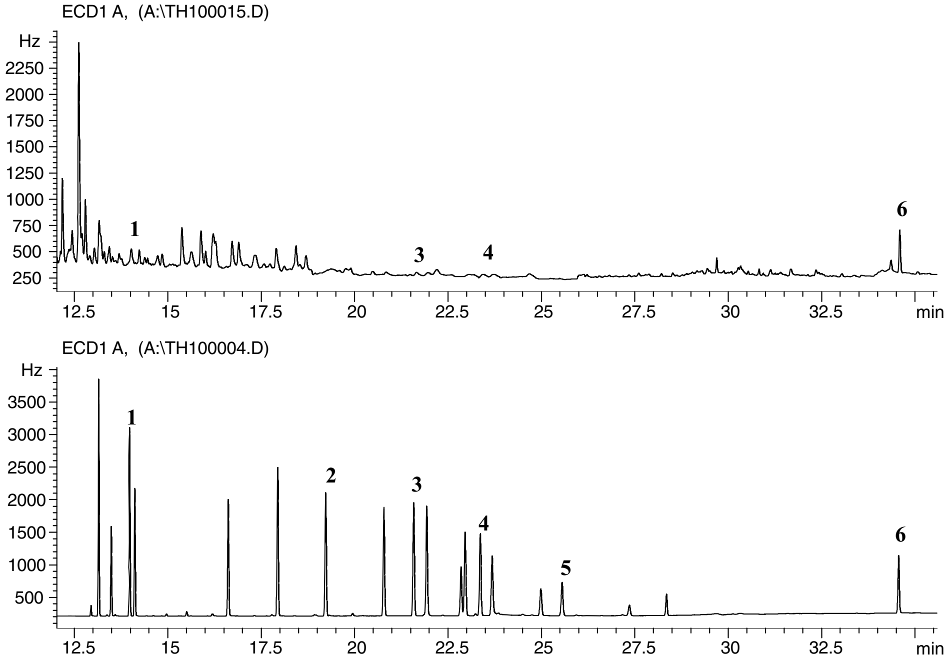

Figure 2. GC-ECD chromatogram of OCPs standard solution and that in water sample: 1: Lindane; 2: Heptachlor epoxide; 3: p,p′-DDE; 4: p,p′-DDD; 5: p,p′-DDT; 6: decachlorobiphenyl (internal standard).

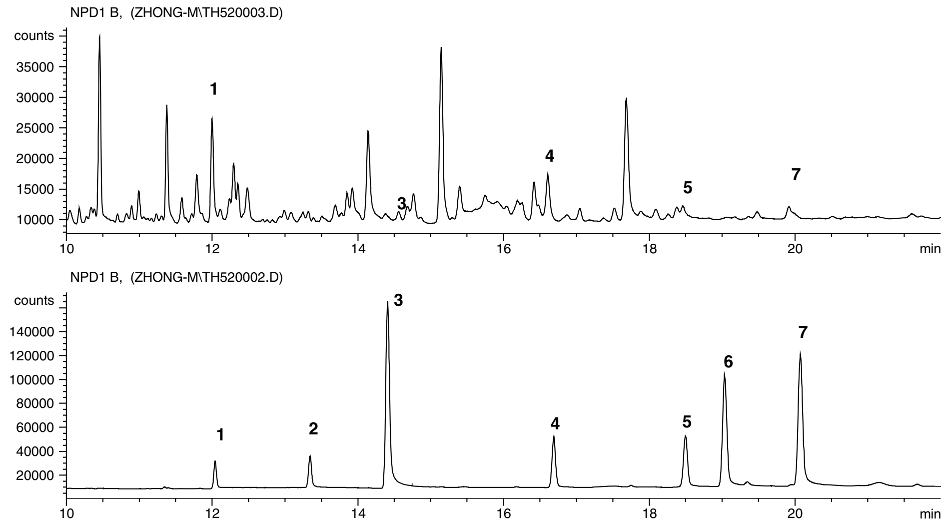

Figure 3. GC-NPD chromatogram of OPPs standard solution and that in water sample: 1: Dichlorvos and trichlofon; 2: Demeton-o; 3: Dementon-s; 4: Dimethoate; 5: Methyl-parathion; 6: Malathion; 7: Parathion.

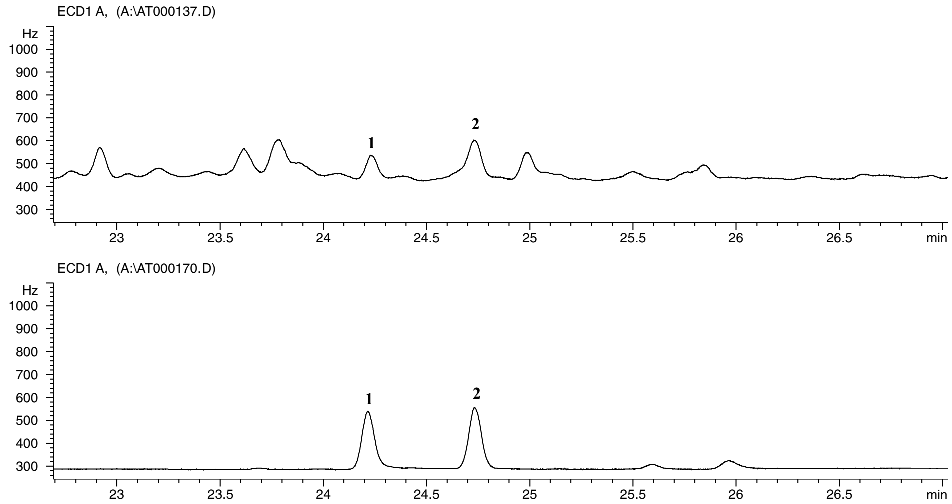

Figure 4. GC-ECD chromatogram of atrazine standard solution and that in water sample: 1: atrazine; 2: pentachloroanisole (internal standard).

detection limits of the pesticides were also conducted and presented in Table VII. Spiked recoveries were conducted at three fortified levels for repeating four times and the detection limits were calculated by using a signal-to-noise (S/N) ratio of 3.

TABLE III
Concentration of pesticides detected in water of Meiliangwan Bay, Taihu Lake(ng/l) (Nov. 2003)

Sampling site	Lindane	Heptachlor epoxide	p,p′-DDE	p,p′-DDD	p,p′-DDT	Dichlorvos	Demeton	Dimethoate	Methylparathion	Malathion	Parathion	Atrazine
1	1.25	0.149	0.280	0.086	0.394	33.8	35.8	ND	ND	7.06	ND	278
2	1.94	0.294	0.185	ND	ND	25.3	27.9	273	ND	9.47	ND	161
3	4.61	0.101	0.241	ND	0.246	72.2	28.4	255	ND	4.01	ND	394
4	2.64	0.263	0.103	ND	0.293	79.5	23.2	248	ND	ND	ND	273
5	1.54	0.205	0.407	0.066	0.416	ND	14.5	172	ND	ND	1.32	504
6	1.21	0.239	0.334	0.296	ND	30.2	17.6	479	ND	7.78	ND	60.0
7	1.07	0.177	2.40	0.153	ND	33.4	17.1	384	ND	10.5	2.67	25.8
8	1.50	0.592	0.177	0.019	0.329	27.8	8.14	507	ND	5.45	3.07	108
9	0.810	0.693	0.818	0.425	ND	1.88	14.9	ND	ND	4.23	ND	98.2
10	0.852	0.183	0.381	0.065	ND	5.63	15.5	ND	ND	4.70	4.78	614

ND: not detected.

TABLE IV
Concentration of pesticides detected in water of Meiliangwan Bay, Taihu Lake(ng/l)(Jan. 2004)

Sampling site	Lindane	Heptachlor epoxide	p,p′-DDE	p,p′-DDD	p,p′-DDT	Dichlorvos	Demeton	Dimethoate	Methylparathion	Malathion	Parathion	Atrazine
1	3.11	ND	0.942	2.12	3.88	25.3	17.0	214	ND	30.5	ND	254
2	5.03	ND	0.575	1.17	3.15	17.9	27.6	173	ND	15.5	ND	294
3	4.77	0.847	0.635	1.43	4.27	41.6	31.1	265	ND	29.7	7.86	423
4	4.92	0.770	0.512	0.974	ND	30.9	28.4	328	ND	42.6	8.35	128
5	6.09	1.09	0.482	1.64	ND	18.8	34.5	192	ND	23.8	5.90	238
6	1.31	0.673	0.289	2.34	3.91	46.6	25.8	210	ND	23.2	ND	224
7	1.15	0.485	0.223	1.24	ND	21.3	33.3	193	ND	35.2	7.01	166
8	2.26	0.382	0.214	0.816	3.58	79.0	15.5	43.9	ND	27.6	ND	157
9	1.35	0.620	0.645	0.525	5.27	50.0	18.6	34.8	ND	ND	ND	385
10	1.74	ND	0.614	1.35	6.67	19.8	15.2	22.0	ND	18.8	2.83	502

ND: not detected.

TABLE V

Concentration of pesticides detected in water of Meiliangwan Bay, Taihu Lake(ng/l)(Aug. 2004)

Sampling site	Lindane	Heptachlor epoxide	p,p′-DDE	p,p′-DDD	p,p′-DDT	Dichlorvos	Demeton	Dimethoate	Methylparathion	Malathion	Parathion	Atrazine
1	1.13	0.352	0.089	ND	ND	35.8	106	716	9.96	7.21	ND	115
2	0.610	0.835	ND	ND	ND	189	38.3	662	25.0	6.81	5.71	253
3	0.352	0.101	0.122	ND	ND	80.0	96.9	760	13.4	ND	ND	63.1
4	1.99	0.119	0.049	0.174	ND	44.8	68.7	713	ND	ND	ND	193
5	1.40	0.510	ND	ND	ND	106	41.9	472	ND	ND	7.92	107
6	0.885	0.329	ND	ND	ND	14.3	118	478	12.1	ND	ND	31.2
7	1.29	0.515	ND	ND	ND	234	66.5	692	51.1	17.8	7.62	190
8	0.685	0.210	ND	ND	ND	22.6	69.9	918	11.8	7.50	ND	260
9	1.03	0.188	ND	ND	ND	155	75.3	644	ND	ND	ND	89.4
10	0.810	0.407	ND	ND	ND	69.6	37.8	320	ND	7.32	ND	21.6

ND: not detected.

TABLE VI
Mean concentration (ng/l) and occurrence (%) of pesticides in water of Meiliangwan Bay, Taihu Lake

Pesticide	Lindane	Heptachlor epoxide	p,p′-DDE	p,p′-DDD	p,p′-DDT	Dichlorvos	Demeton	Dimethoate	Methyl-parathion	Malathion	Parathion	Atrazine
Mean value (Nov. 2003)	1.74	0.290	0.533	0.11	0.168	31.0	20.3	232	0.00	5.32	1.18	252
Occurrence (Nov. 2003)	100	100	100	70	50	90	100	70	0	80	40	100
Mean value (Jan. 2004)	3.17	0.487	0.542	1.36	3.07	35.1	24.7	168	0.00	24.7	3.20	277
Occurrence (Jan. 2004)	100	70	100	100	70	100	100	100	0	90	50	100
Mean value (Aug. 2004)	1.02	0.357	0.026	0.017	0.00	95.1	71.9	638	12.3	4.66	2.13	132
Occurrence (Aug. 2004)	100	100	30	10	0	100	100	100	60	50	30	100
Total mean concentration	1.98	0.378	0.367	0.496	1.06	51.6	39.0	346	4.12	11.6	2.17	217
Total mean occurrence	100	90	78	60	40	98	100	90	20	73	40	100

TABLE VII
Mean spiked recoveries and detection limits

Pesticide	Mean recoveries (%)	Detection limits (ng/l)
Lindane	100.5–116.0	0.05
Heptachlor epoxide	63.0–79.6	0.04
p,p′-DDE	30.5–36.9	0.07
p,p′-DDD	62.0–75.0	0.09
p,p′-DDT	91.0–122.3	0.2
Dichlorvos	65.6–88.2	1.0
Demeton-o	59.4–85.1	1.0
Dementon-s	60.7–72.6	1.0
Dimethoate	58.0–85.0	5.0
Methyl-parathion	76.1–86.9	3.0
Malathion	68.9–76.8	2.0
Parathion	75.9–94.6	2.0
Atrazine	75.6–97.8	8.0

3.1. OCPs RESIDUES

In this study, the determined OCPs were lindane, heptachlor epoxide, p,p′-DDT, p,p′-DDE and p,p′-DDD. Their mean concentrations in the water of Meiliangwan Bay, Taihu Lake were 1.98, 0.378, 1.06, 0.367 and 0.496 ng/l, and their mean occurrences were 100, 90, 40, 78 and 60, respectively. All the concentrations of OCPs were much lower than the maximum allowable limits of drinking water source standards of China which are in μg/l level (Table VIII).

It was observed that lindane had higher occurrences and concentrations than other OCPs and was detected in all the water samples. Lindane, a commercial name of γ-HCH, is the biological active isomer of technical HCH. Unlike β-HCH, γ-HCH is not bioaccumulated in human being (Gao *et al.*, 1995). In the past 30 years, as one of the predominant OCPs, technical HCH has played an important role in agriculture of China and the total production of technical HCH was around 4 million tons from 1952 to 1984 (Qiu *et al.*, 2004). Nevertheless, with the improving recognition of its properties such as resistance to degradation, susceptibility to large-scale transport, extreme lipophilicity and high toxicity, the use and production of technical HCH was forbidden by the government of China in 1983. However, due to its longer half-life time, it may still remain in aquatic systems for years. The previous use of technical HCH may be a source of lindane in the water of Meiliangwan Bay. Moreover, another source of lindane may come from a small amount of illegal use of lindane which is still produced mainly for export purpose in China. In 1993, the total output of lindane was over 2000 tons

TABLE VIII
Maximum allowable limits of drinking water source standards of China (GB3838–2002)

Pesticide	Concentration limit (μg/l)
Lindane	2
Heptachlor epoxide	0.2
p,p′-DDE	1
p,p′-DDD	–
p,p′-DDT	–
Dichlorvos	50
Trichlorfon	50
Demeton	30
Dimethoate	80
Methyl-parathion	2
Malathion	50
Parathion	3
Atrazine	3

—: not regulated.

and still 10% of it was employed in China and 90% of lindane was exported to other countries (Cai *et al.*, 1995).

Generally, the mean concentration of lindane was found to be much lower in comparison with the previous work (Jiang *et al.*, 1988) which reported a decline trend of HCH concentrations in Taihu Lake from 1980 to 1988. The concentration of lindane in 1980 was 255 ng/l, but was dramatically decreased to 6 ng/l in 1988. In our study, the mean concentration of Lindane followed this decreasing trend was 1.98 ng/l. This indicates that although a large amount of technical HCH has been applied in China, the ban of its use over twenty years has significantly reduced its residues in the water of Taihu Lake.

The mean concentration of p,p′-DDT was higher than heptachlor epoxide but slightly lower than that of lindane. As one of the common pesticides, p,p′-DDT was broadly applied in the past in China. From 1951 to 1983, the total production of DDT reached 0.27 million tons (Qiu *et al.*, 2004). In addition, DDT is still in production as raw material for synthesis of dicofol which is an acaricide used in corn, fruit tree and cotton to mite control. In dicofol, there still remain relatively higher percents of DDT and an intermediate, 2, 2, 2, 2-tetrachloro-1, 1-bis (4-chloropheyl) ethane, that can easily converted to DDE (Huang *et al.*, 2000). Therefore, the impure dicofol can increase the environmental residues of DDT and its metobolites. It is necessary to conduct further research to confirm whether dicofol is a source of DDT and its metabolites in the water of Taihu Lake. It is also apparent that the occurrence of p,p′-DDT was lower than that of p,p′-DDE and p,p′-DDD which are two metobolites of p,p′-DDT, this may be the reason that p,p′-DDE and p,p′-DDD are more stable than p,p′-DDT.

Heptachlor epoxide was found in higher frequency, which was detected in almost all the water samples with lower concentration. That may be due to a continuous use of heptachlor epoxide with a small quantity in the TLA.

Additionally, an obvious temporal trend was observed from the above analytical results. The detected maximum concentration was 6.09 ng/l for lindane, 1.09 ng/l for heptachlor epoxide, 2.40 ng/l for p,p′-DDE, 2.34 ng/l for p,p′-DDD and 6.67 ng/l for p,p′-DDT. This did not occur in the rainy season (in Aug. 2004) but in the dry season (Jan. 2004) with the exception of the highest concentration of p,p′-DDE which was found in the normal season (Nov. 2003). It is indicated that the OCPs residues might be concentrated in the water of Taihu Lake in the dry season. In the dry season, the water depth was 0.5–1.0 m lower than that in the rainy season. We suggest that the concentration and dilution effect may play a crucial role in temporal trend of concentration of OCPs. As a result, the concentration of OCPs in the rainy season was lower than that in the dry season, because of the dilution effect.

Our data was also compared with the previous studies available at Jiulong River, Macau coastal water, Xiamen Harbour, Guangting Reservoir and Yongdinghe River (China), Manzala Lake (Egypt), (Table IX). From the comparison, it is apparent that our data and literature data show similar levels of OCPs.

3.2. OPPS RESIDUES

The determined OPPs were dichlovos, trichlofon, demeton-o, demeton-s, dimethoate, methyl-parathion, malathion, and parathion. Because trichlofon can be easily transformed to dichlovos, and demeton-o and demeton-s is a pair of isomers, thus, their total concentrations were determined, respectively.

In the present study, far higher concentrations of OPPs were observed than that of OCPs. The mean concentration of dichlovos, demeton, dimethoate, methyl-parathion, malathion, and parathion were 51.6, 39.0, 346, 4.12, 11.6, 2.17 ng/l, respectively. The corresponding occurrences were 98%, 100%, 90%, 20%, 73%, 40%, respectively. Due to their efficiency and inexpensiveness, OPPs have been widely used in agriculture for crop protection and fruit tree treatment as substitute for OCPs. The OPPs have occupied the dominant position in production and use in China since the use of OCPs such as DDT and HCH were banned in 1983. There are over 20 kinds of OPPs in production and it was reported that, 0.11 million tons of OPPs were used in 1989, occupying 52.8% of the total usage of pesticides and 77% of the total usage of insecticide in China (Pan *et al.*, 1997).

It is obvious that dichlorvos, demeton and dimethoate had higher occurrence and concentration than other OPPs, because of a large amount of application loads.

TABLE IX
Comparison of OCPs residues in Meiliangwan Bay and in other natural water systems (ng/l)

Water body	Lindane	Heptachlor epoxide	p,p′-DDE	p,p′-DDD	p,p′-DDT	Reference
Jiulong River	0.26–7.84	–	0.06–40.9	ND-12.6	ND-9.73	Zhang *et al*., 2001
Macau coastal water	5.33	0.39	1.49	10.13	13.11	Yang *et al*., 2004
Xiamen Harbour	0.10–0.40	ND	0.01–0.58	ND-0.32	0.76–1.56	Zhang *et al*., 2000
GuangtingReservoir Yongdinghe River	ND-18.2	ND-5.76	–	–	ND-15.2	Kang *et al*., 2003
Manzala lake, Egypt	ND-20.6	–	ND-38.3	–	ND-44.6	Abouarab *et al*., 1995
Meiliangwan Bay	0.610–6.09	ND-1.09	ND-2.40	ND-2.34	ND-6.67	This study

—: not investigated.

It was reported that in 2002, over 10 thousand tons of dichlorvos, trichlofon and dimethoate were employed in China, respectively, which were far more than other OPPs usage (Hua *et al*., 1999). Furthermore, because of the acute toxicity for human being and other endotherm, parathion and methyl parathion were gradually reduced in use. In 2004, parathion and methyl parathion was decreased by 22.51% and 14.52% in usage compared to in 2003 (Shao *et al*., 2004). In Jan. 2004, the registration of five highly toxic OPPs including parathion and methyl parathion was stopped and from Jan. 2007, their use will be completely forbidden in China (Tang *et al*., 2004). This can give a good explanation of the lower concentration and occurrence of them.

Another conclusion drawn from the data was a significant difference in concentration between the rainy and the dry seasons. The highest concentration of those OPPs were 234 ng/l for dichlorvos 118 ng/l for demeton, 918 ng/l for dimethoate, 51.1 ng/l for methyl-parathion, 17.8 ng/l for malathion and 42.6 ng/l for parathion. Except for parathion, the highest concentration of OPPs appeared in the rainy season (in Aug. 2004). This can be explained by the special hydrology of Taihu Lake which follows a certain pattern. During the rainy season, the water flows from the tributaries of the west, southwest, northwest side of the lake and flows out from the tributaries of east side of the lake, but takes a reverse direction during the dry season (Zou *et al*., 1996). Accordingly, during the rainy season, the pesticide contamination is caused mainly by the contribution from the tributaries on the west side of the lake, which can directly flow into the water of Meiliangwan Bay and cause a higher concentration. However, the reverse water flow during the dry season minimized the contribution from the tributaries on the west of lake, but increased the contribution from the east side of the lake. The transportation process of pesticide to Meiliangwan Bay from the east side of the lake might take several months. During such a long-time transportation, the pesticide might be deposited in sediment and degraded. As a result, the concentration of OPPs in the rainy season was higher than that in the dry season. In general, the source of those OPPs principally originated from the agricultural fields in TLA, depending on the run-off and leaching process of the OPPs residues that entered the Meiliangwan Bay.

Our results are also compared with a few other studies conducted in Jiulong River Estuary and Yangtze River (Table X). It is worth paying special attention to the fact that much higher OPPs pollutions exist in the water of Meiliangwan Bay, Taihu Lake.

Acting on acetylcholinesterase activity, the OPPs are more toxic than OCPs, and it has been believed that OPPs degrade faster. However, it has been proved that they may persist in the aquatic system for a long time, sometimes more than half year (Lartiges *et al*., 1995). It is suggested that the exposure to longer period and lower concentration residues of OPPs in the water of Taihu Lake will bring adverse effect on eco-system and can threaten the safety of the drinking water source.

TABLE X

Comparison of OPPs residues in Meiliangwan Bay and in other natural water systems (ng/l)

Water body	Dichlorvos	Demeton	Dimethoate	Methyl- parathion	Malathion	Parathion	Reference
Jiulong River Estuary	6.67–49.8	–	6.39–32.6	ND-14.5	ND-51.6	ND-8.95	Zhang *et al.*, 2002
Yangtse River	1.59–6.68	3.05–4.42	14.5–61.3	ND-3.33	ND-5.16	1.93–2.90	Zhou *et al.*, 2005
Meiliangwan Bay	ND-234	8.14–118	ND-918	ND-51.1	ND-42.6	ND-8.35	This study

—-: not investigated.

3.3. ATRAZINE RESIDUES

Atrazine is used worldwide as a selective pre- and post emergence herbicide for the control of widespread weeds in many agricultural crops like corn, wheat, maize and barley. Atrazine inhibits the growth of target weeds by interfering with the normal function of photosynthesis (Graymore *et al.*, 2001). Since 1980s, atrazine has been used in China (Helena *et al.*, 2005) and the amount of usage reached 2835 tons in 2000 with a 20% increment annually (Hua *et al.*, 1999).

In our study, atrazine was detected in all the water samples with considerably high concentration. The mean concentration level of atrazine was 217 ng/l and the highest was 614 ng/l in Nov. 2003 that was under the maximum allowable limits of drinking water source standards of China. It was reported that atrazine persists under cool, dry conditions in a stable pH environment and its metebolites were also toxic and persistent as atrazine (Graymore *et al.*, 2001). However, the effects of long-term exposure to atrazine at lower concentration on human being is unknown yet, which requires further study.

4. Conclusion

The pollution status of OCPs, OPPs and atrazine were studied in the surface water of Taihu Lake. In general, the contamination follows a decreasing order of OPPs > atrazine >>OCPs. The mean concentrations of OCPs were 1.98, 0.378, 0.357, 0.496, 1.06 for lindane, heptachlor epoxide, p,p′-DDE, p,p′-DDD, p,p′-DDT, respectively. The mean concentrations of OPPs were 51.6, 39.0, 346, 4.12, 11.6, 2.17 and 217 ng/l for dichlorvos demeton, dimethoate, methyl-parthion, malathion, parathion and atrazine, respectively. The highest concentration levels of the detected pesticides were far below the maximum allowable limits of drinking water sources standards of China. Moreover, the presence of detected pesticides was attributed to the intense agricultural activity of TLA. The effects of multi-contamination from pesticide exposure on humans via drinking water are uncertain, and additional work is needed to reduce this uncertainty.

Acknowledgments

This study was supported by Chinese National Programs for High Technology Research and Development 863 project (NO. 2002AA601011-03).

References

Abou-arab, A. A. K., Gomaa, M. N. E. and Badawy, A.: 1995, ‘Distribution of organochlorine pesticides in Egyptian aquatic system’, *Food Chemistry* **54**, 141–146.

Cai, D. J., Liu, Y. K. and Tao, S. M.: 1995, 'Protection the pollution of uneffective isomers of HCH', *Pesticide Science and Administration (in Chinese)*. **60**(4), 31–32.

Feng, K., Yu, B. Y. and Ge, D. M.: 2003, 'Organochlorine pesticide (DDT and HCH) residues in the Taihu Lake Region and its movement in soil – water system I. Field survey of DDT and HCH residues in ecosystem of the region', *Chemosphere* **50**, 683–687.

Gao, X. F. and Zhang, G. G.: 1995, 'Utilization of by-products of lindane', *Research of Environmental Science (in Chinese)* **8**(1), 3–4.

Gong, Z. M., Dong, Y. H. and An, Q.: 2001, 'Organochlorine residues in eggs of night heron breeding in yuantouzhu,wuxi and their function as bioindicater', *Environmental Sceince (in Chinese)* **22**(2), 110–114.

Graymore, M., Stagnitti, F. and Allinson, G.: 2001, 'Impacts of atrazine in aquatic ecosystems', *Environ. Int.* **26**, 483–495.

Helena, P. and Lucija, Z. K.: 2005, 'Evaluation of photolysis and hydrolysis of atrazine and its first degradation products in the presence of humic acids', *Environmental Pollution* **133**, 517–529.

Hua, X. M. and Shan, Z. J.: 1999, 'Production, usage and environmental influence factors of pesticides in China', *Science and Technology of Anhui (in Chinese)* **6**, 6–10.

Huang, X. Z., Ji, Y. and Ye, J. M.: 2000, 'Analysis of dicofol technical material and its impurties with GC–MS', *Pesticide Science and Administration (in Chinese)*. **5**, 9–12.

Jiang, X. F., Chen, S. J. and Wang, X. F.: 1988, 'Determination of HCH in water of Taihu Lake', *Environmental Science and Technology (in Chinese)*. 5–7.

Kang, Y. H., Liu, P. B. and Wang, Z. J.: 2003, 'Persistent organochlorinated pesticides in water from Guanting Reservoir and Yongdinghe River, Beijing', *Journal of Lake Sciences (in Chinese)*. **15**(2), 125–132.

Lartiges, S. B. and Garrigues, P. P.: 1995, 'Degradation kinetics of organophosphorus pesticides in different waters under various environmental conditions', *Environ. Sci. Techno.* **29**, 1246–1254.

Pan, Y. H., Wei, A. X. and Zhao, G. D.: 1997, 'Analysis of trace organophosphorus pesticides in water by high performance liquid chromatography/mass spectrometry', *Advances in Environmental Science (in Chinese)* **1**, 32–40.

Qiu, X. H., Zhu, T. and Li, J.: 2004, 'Organochlorine pesticides in the air around the taihu lake, China', *Environ. Sci. Technol.* **38**, 1368–1374.

Rovedattil, M. G., Castan, P. M. and Topalia, M. L.: 2001, 'Monitoring of organochlorine and organophosphorus pesticides in the water of the Reconquista River (Buenosaires, argentina)', *Water Res.* **35**(14), 3457–3461.

Shao, Z. R. and Su, F.: 2004, 'The prospect for pesticide usage in 2002 and 2003', *Pesticide Science and Administration (in Chinese)* **25**(2), 33–35.

Tang, Y. F., Wang, Y. X. and Cai, H. S.: 2004, 'Application and pollution of organophosphorous pesticides', *Journal of Wuhan Institute of Chemistry and Technology (in Chinese)* **26**(1), 11–14.

Wang, T. Y., Lu, Y. L. and Zhang, H.: 2005, 'Contamination of persistent organic pollutants (POPs) and relevant management in China', *Environ. Int.* **31**, 813–821.

Wang, H., Wang, C. X. and Wu, W. Z.: 2003, 'Persistent organic pollutants in water and surface sediments of taihu lake, China and risk assessment', *Chemosphere* **50**, 557–562.

Yang, Q. S., Mei, B. X. and Luo, X. J.: 2004, 'Vertical distribution of organochlorine pesticides (OCPs) in Macau coastal waters', *Acta Scientiae Circumstaniae (in Chinese)* **24**(3), 428–434.

Zhang, Z. L., Chen, W. Q. and Khalid, M.: 2001, 'Evaluation and fate of the organic chlorine pesticides at the waters in Jiulong River Estuary', *Environmental Science (in Chinese)*. **22**(3), 88–92.

Zhang, Z. L., Hong, H. S. and Khald, M.: 2000, 'Study on organochlorine pesticide and PCBs at surface water in Xiamen Harbour', *Marine Environmental Science (in Chinese)* **19**(3), 48–51.

Zhang, Z. L., Yu, G. and Hong, H. S.: 2002, 'Environmental behavior and risk assessment of organophosphorus pesticides at water from Jiulong River Estuary', *Environmental Science (in Chinese)* **23**, 73–78.

Zhou, F., Sun, C. and Zhong, M.: 2005, 'Determination of of organophospheorous pesticides by gas chromatography in drinking water sources', *Environmental Pollution and Protection (in Chinese)* **27**(2), 151–153.

Zhu, X. and Zhang, Y. W.: 2002, 'The status of water quality and its protection policy in Meiliangwan Bay', *Water Source Protection (in Chinese)* **4**, 28–30.

Zou, H. X., Sheng, G.Y. and Sun, C.: 1996, 'Distribution of organic contaminants in Lake Taihu', *Water Res.* **30**(9), 2003–2008.