

Water–gas exchange of organochlorine pesticides at Lake Chaohu, a large Chinese lake

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Abstract Organochlorine pesticides (OCPs), a potential threat to ecosystems and human health, are still widely residual in the environment. The residual levels of OCPs in the water and gas phase were monitored in Lake Chaohu, a large Chinese lake, from March 2010 to February 2011. Nineteen types of OCPs were detected in the water with a total concentration of 7.27 ± 3.32 ng/l. Aldrin, DDTs and HCHs were the major OCPs in the water, accounting for 38.3 %, 28.9 % and 23.6 % of the total, respectively. The highest mean concentration (12.32 ng/l) in the water was found in September, while the lowest (1.74 ng/l) was found in November. Twenty types of gaseous OCPs were detected in the atmosphere with a total concentration of 542.0 ± 636.5 pg/m³. Endosulfan, DDTs and chlordane were the major gaseous OCPs in the atmosphere, accounting for 48.9 %, 22.5 % and 14.4 % of the total, respectively. The mean concentration of gaseous OCPs was significantly higher in summer than in winter. *o,p'*-DDE was the main metabolite of DDT in both the water and gas phase. Of the HCHs, 52.3 % existed as β -HCH in the water, while α -HCH (37.9 %) and γ -HCH (30.9 %) were dominant isomers in the gas phase. The average fluxes were -21.11 , -3.30 , -152.41 , -35.50 and -1314.15 ng/(m²day) for α -HCH, γ -HCH, HCB, DDT and DDE, respectively. The water–gas exchanges of the five types of OCPs indicate that water was

the main potential source of gaseous OCPs in the atmosphere. A sensitivity analysis indicated that the water-gas flux of α -HCH, γ -HCH and DDT is more vulnerable than that of HCB and DDE to the variation of the parameters. The possible source of the HCHs in the water was from the historical usage of lindane; however, that in the air was mainly from the recent usage of lindane. The technical DDT and dicofol might be the source of DDTs in the water and air.

Keywords OCPs · Water · Gas phase · Water–gas exchange · Lake Chaohu

Introduction

Organochlorine pesticides (OCPs) are typical persistent organic pollutants (POPs) in the environment. Their stable construction and strong resistance cause OCPs to have a low degradation rate in the natural environment, and their semi-volatility gives them a high migration rate and brief interaction with environmental media, which allow OCPs to remain unchanged in environment processes (Liu 2005a; Colborn et al. 1996; UNEP 2004). In addition, the degradation products of OCPs may still be toxic, such as DDD and DDE, the degradation product of DDT. Thus, once emitted into the environment, OCPs will persist in water, soil, atmosphere and organisms (Carrera et al. 2001; Aaron et al. 2001; de Joode et al. 2001). Although OCPs have been disallowed in production and use since the 1970s and 1980s, numerous surveys have reported that OCPs can still be detected and have even shown an upward trend in terms of concentration and harmfulness in some regions (Cotham and Bidleman 1991; Turgut 2003; Zhao et al. 2010).

Water is easily contaminated by OCPs. The highest residual levels of HCHs (more than 1,000 pg/l) have been found in areas at approximately 40°N latitude, such as the

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Bering Sea, the Alaska Bay and the North Pacific. The concentration of α -HCH increased from 810 pg/l in the South China Sea to 1,200 pg/l in the Bering Sea with the rise of latitude (Chernyak et al. 1995), while the concentration of HCHs in the Kucuk Menderes River was only 187–337 ng/l (Turgut 2003). The residual level of DDTs between 1995 and 1996 was 1.97–6.77 ng/l in the Ebro River, Spain (Fernandez et al. 1999), and ND to 4578.0 ng/l in the Gomti River, India (Singh et al. 2005). Numerous studies have also been performed in China, with the results showing that the residual levels of OCPs differ significantly among regions. It was reported that the residual concentrations of HCHs and DDTs were 1.10–7.50 and 4.45–78.87 ng/l in the Huaihe River (Yu et al. 2004), 5.8–20.6 and 0.52–1.13 ng/l in the Pearl River (Yang et al. 2004), and 5.9–16.2 and 3.10 ng/l in the Guanting Reservoir, Beijing (Wang et al. 2003).

OCPs also reside in the atmosphere. HCHs and DDTs have even been detected in the polar regions, where no OCPs were ever used, due to long-term transport. Approximately 97 tons of α -HCH and 13 tons of β -HCH were imported to the North Pole (Cotham and Bidleman 1991). The atmospheric concentrations of DDTs measured from Shanghai, China, to the Arctic Ocean during the Third China Arctic Research Expedition in 2008 by Wu et al. (2011) amounted to $36 \pm 31 \text{ pg/m}^3$, ranging from 2.0 to 110 pg/m^3 . The atmospheric concentrations of α -HCH and γ -HCH in North America were 1.5–170 and 5–400 pg/m^3 (Shen et al. 2004).

Volatilization and dissolution are two major processes that affect the distribution of OCPs between the water

and gas phase. Volatilization is influenced by temperature, water flow velocity, and the content of, for example, suspended matter, sediment, organic matter and dissolved organic matter. High organic matter content will restrain volatility rates, while high temperature and water flow velocity will promote them (Liu 2005b). In addition, OCPs with low solubility tend to volatilize, and OCPs with high solubility tends to remain. In general, it is easier for HCHs to volatilize compared with DDTs because of their higher Henry's Law Constant and vapor pressure, and a high temperature results in more volatilization. Thus, the volatility rate of HCHs, especially α -HCH, is particularly high in tropical regions (Kannan et al. 1995). Gaseous OCPs may also enter water through dry and wet deposition.

Lake Chaohu, in central Anhui Province, is the fifth largest freshwater lake in China, covering an area of 760 km^2 . As one of the primary food supply areas, large amounts of OCPs were used in agricultural activities in the area around Lake Chaohu before they were banned. These OCPs are still residual in the environment, leading to a potential threat to ecosystems and human health. In addition to aquaculture and agricultural irrigation, Lake Chaohu provides drinking water for 9.6 million residents and determines their security. The aims of this paper are to measure the concentrations of OCPs in the water and gas phase to determine their residual levels in Lake Chaohu and to assess the water–gas exchange of OCPs for a better understanding of their migration.

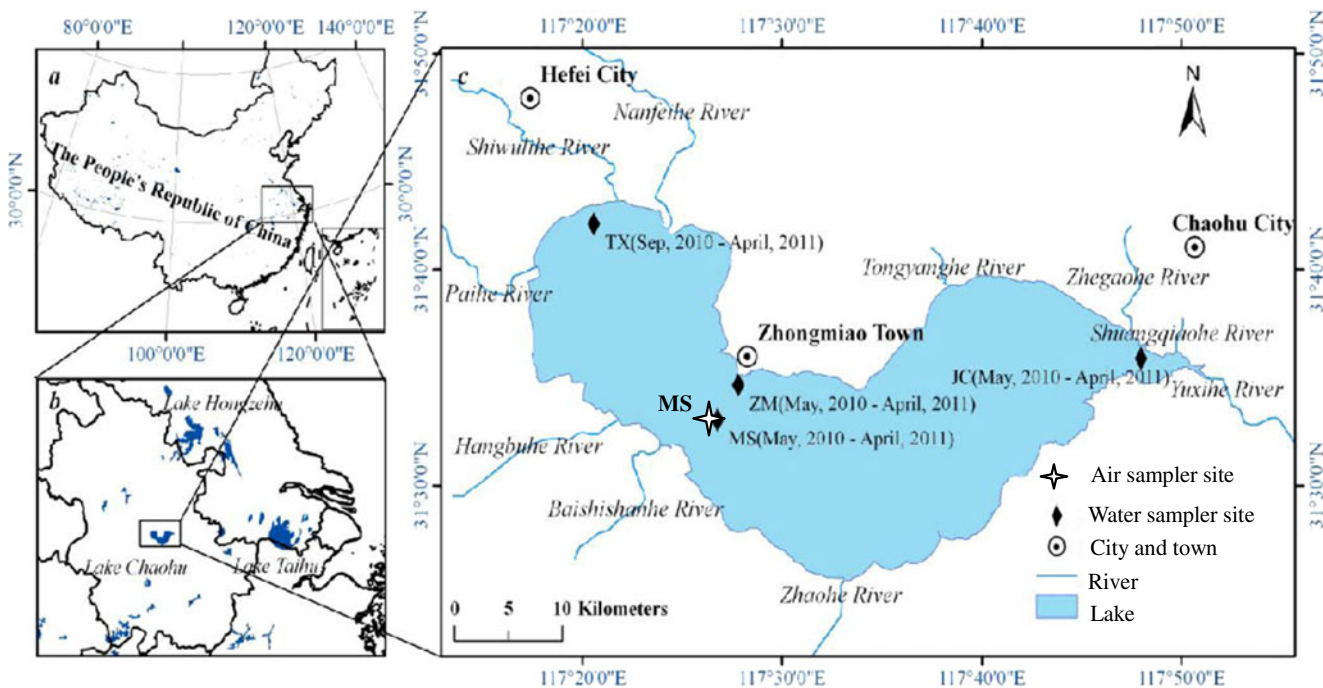


Fig. 1 Geographical locations of a Lake Chaohu in China, b Lake Chaohu in East China and c the sampling sites

Materials and methods

Sample collections

The gaseous sample collection was conducted on Mushan Island (MS) in Lake Chaohu. Four water sample collections were made: ZM (200 m south of the Zhongmiao Temple), MS (200 m east of Mushan Island), JC (water intake section near an automatic monitoring station), and TX (150 m south of the water intake section in Tangxi). The sampling sites are marked in Fig. 1. Both water and gaseous samples were collected from March 2010 to February 2011.

During water sampling, 10 l of water (0–0.3 m below the surface) was collected in clean brown glass jar for the following filtering and extracting processes. For gaseous sample collection, Polyurethane foam plugs (PUF, 60×100 mm) were used in a high-volume sampler (PM10-PUF-300) to collect gaseous OCPs, referring to Method TO-13A provided by the USEPA (1999). The PUF was Soxhlet extracted successively with acetone, dichloromethane and *n*-hexane for 8 h each before sampling and packed in an aluminum foil after sampling.

Sample preparation

The water samples were prepared by filtration and enrichment. One liter of collected water was filtered by a 0.45- μ m glass fiber with a recovery indicator PCNB (100 ng) through a 142-mm filter plate propelled by a peristaltic pump (80EL005; Millipore Co., USA). Then, OCPs in the water were enriched by solid phase extraction using octadecylsilane SPE cartridges (SPE, C18, 6 ml, 500 mg; Supelco, Co., USA). The cartridges were activated following the order of 6 ml DCM, 6 ml methanol and 6 ml ultrapure water before use. During the activation and extraction, the liquid level remained tangent to the filter to keep the C18 wet. After extraction, the cartridge was dried using a vacuum pump for approximately 1 min.

In the laboratory, OCPs were eluted from the SPE cartridge for measurement. The SPE cartridge was connected to an anhydrous sodium sulfate (5 g) cartridge and eluted by DCM (three times, 6 ml per elution). The extracts were concentrated to approximately 1 ml by a vacuum rotary evaporator (Eyela N-1100; Tokyo Rikakikai Co., Japan) and then concentrated to 1 ml again after adding 10 ml *n*-hexane. 2,4,5,6-Tetrachloro-*m*-xylene (TCMX) was added to the samples as an internal standard, and the samples were transferred to vials and sealed for analysis.

After sampling, the PUF with 100 ng recovery indicator (1-bromo-2-nitrobenzene) was Soxhlet extracted with 100 ml of a 1:1 mixture of *n*-hexane and acetone for 24 h. The extracts were concentrated to 1 ml by rotary evaporation first and then concentrated to 1 ml again after adding

10 ml of *n*-hexane. The sample was then transferred to a silica/alumina chromatography column for cleanup. The initial solution eluted by 20 ml of *n*-hexane was discarded, and the subsequent eluate was collected, while 50 ml of a 1:1 mixture of *n*-hexane and DCM was used to elute the OCPs. The eluate was concentrated to 1 ml by rotary evaporation first and then concentrated to 1 ml again after adding 10 ml of *n*-hexane. PCNB was added to the sample as an internal standard. The samples were concentrated to 10 μ l with flowing nitrogen, transferred to microvolume inserts, and sealed for analysis. Three procedural blank samples were analyzed for solvent and glassware contamination, and the results indicated that contamination could be ignored.

Sample analyses and quality control

The samples were analyzed using an Agilent 7890A/5975C gas chromatography and mass spectrometer detector and a HP-5MS fused silica capillary column (30 m×0.25 mm×0.25 μ m; Agilent Co., USA). Helium was used as a carrier gas at a flow rate of 1 ml/min. The samples (1 μ l) were

Table 1 The method recoveries and the instrument detection limits

	Recoveries (%)		Method detection limits		Instrument detection limits (ng/ml)
	Water	Gas phase	Water (ng/l)	Gas (pg/m ³)	
HCB	54.0	69.6	0.01	0.01	0.05
α -HCH	100.4	79.5	0.05	0.06	0.5
β -HCH	99.1	98.8	0.05	0.05	0.5
γ -HCH	88.7	89.6	0.06	0.05	0.5
δ -HCH	90.3	89.4	0.06	0.05	0.5
Heptachlor	68.0	121.4	0.1	0.04	0.5
Aldrin	71.6	95.1	0.07	0.05	0.5
Isodrin	74.7	93.3	0.03	0.02	0.2
γ -Chlordane	66.6	94.6	0.01	0.08	0.5
α -Chlordane	72.9	109.9	0.01	0.05	1
Endosulfan I	89.4	67.4	0.06	0.07	0.5
Endosulfan II	95.7	90.0	0.05	0.05	0.5
Endrin	107.4	162.1	0.47	0.3	5
<i>o,p'</i> -DDE	87.1	115.7	0.02	0.08	1
<i>p,p'</i> -DDE	60.4	113.2	0.01	0.04	0.5
<i>o,p'</i> -DDD	80.1	113	0.06	0.04	0.5
<i>p,p'</i> -DDD	84.2	64.8	0.59	0.7	5
<i>o,p'</i> -DDT	66.4	116.8	0.12	0.4	5
<i>p,p'</i> -DDT	84.6	135.8	0.01	0.03	0.5
Mirex	73.5	97.8	0.07	0.09	1

injected by an autosampler under a splitless mode at a temperature of 220 °C. The oven temperature program was as follows: 50 °C for 2 min, 10 °C/min for 150 °C, 3 °C/min for 240 °C, 240 °C for 5 min, 10 °C/min for 300 °C and 300 °C for 5 min. The ion source temperature of the mass spectrometer was 200 °C, the transfer line temperature was 250 °C, and the quadrupole temperature was 150 °C. The compounds were quantified by the selected ion mode and a calibration curve with an internal standard.

There were two parallel samples in each sampling site, and the values from the parallel samples were averaged to get one value per sample. The samples, method blanks and procedure blanks were prepared in the same manner. The method recoveries and the instrument detection limits were measured before sample analysis, and the results are shown in Table 1. The method recovery for water samples was between 54 % and 107.4 %, and that for gaseous samples was between 64.8 % and 162.1 %. The instrument detection limits varied from 0.01 to 5 ng/ml.

Results and discussions

Residual levels and components of the OCPs in the water and gas phase

The residual levels of the OCPs in the water and gas phase are shown in Table 2 and Fig. 2. The components of the OCPs in the water and gas phase are shown in Figs. 3 and 4, respectively.

Nineteen OCP types were detected in the water at Lake Chaohu, including HCHs (α -, β -, γ , δ -HCH), DDTs (*o,p'*-, *p,p'*-DDE, DDT, DDD), HCB, heptachlor, aldrin, isodrin, chlordane (α -, γ -chlordane), endosulfan (endosulfan I, II) and mirex, with a total concentration of 7.27±3.32 ng/l. Figure 3 shows aldrin (2.79±2.14 ng/l), DDTs (2.10±1.71 ng/l) and HCHs (1.72±1.29 ng/l) were the major OCPs in the water, accounting for 38.3 %, 28.9 % and 23.6 % of the total, respectively. The proportion of other OCPs was only 9.2 %. *o,p'*-DDE was the main metabolite of DDT, and 52.3 % of the HCHs existed as β -HCH in the water.

Table 2 Residual levels of OCPs in the water and gas phase at Lake Chaohu

OCPs	Water (ng/l)				Gas phase (pg/m ³)			
	A-mean	G-mean	SD	Detection ratio	A-mean	G-mean	SD	Detection ratio
HCB	0.17	0.15	0.07	100 %	9.7	4.8	13.7	90 %
α -HCH	0.46	0.33	0.46	100 %	17.9	15.9	15.2	100 %
β -HCH	0.90	0.80	0.48	100 %	10.7	6.0	17.4	90 %
γ -HCH	0.28	0.20	0.31	100 %	14.0	9.9	14.0	100 %
δ -HCH	0.08	0.05	0.10	100 %	3.3	1.9	5.5	90 %
HCHs	1.72	1.43	1.29	100 %	45.9	38.4	43.2	100 %
Heptachlor	0.16	0.09	0.19	90 %	3.6	4.6	3.5	70 %
Aldrin	2.79	2.16	2.14	100 %	14.8	7.0	31.9	60 %
Isodrin	0.17	0.12	0.17	100 %	2.0	2.0	2.0	80 %
γ -Chlordane	0.01	0.01	0.02	50 %	3.8	3.1	3.7	80 %
α -Chlordane	ND	ND	ND	0 %	73.5	8.8	153.8	100 %
Chlordane	0.01	0.01	0.02	60 %	77.3	12.7	154.1	100 %
Endosulfan I	0.02	0.02	0.03	70 %	216.4	58.7	272.2	100 %
Endosulfan II	0.10	0.02	0.29	70 %	46.1	18.5	71.6	80 %
Endosulfan	0.12	0.04	0.28	90 %	262.5	66.0	338.9	100 %
Endrin	0.03	0.05	0.05	50 %	1.4	5.6	2.6	30 %
<i>o,p'</i> -DDE	1.64	0.40	1.82	90 %	78.2	20.1	101.5	90 %
<i>p,p'</i> -DDE	0.02	0.02	0.04	40 %	26.3	19.0	28.5	80 %
<i>o,p'</i> -DDD	0.03	0.06	0.04	40 %	2.4	4.4	4.2	50 %
<i>p,p'</i> -DDD	0.06	0.07	0.11	40 %	1.5	2.0	1.9	70 %
<i>o,p'</i> -DDT	0.14	0.06	0.39	50 %	7.7	5.2	8.1	80 %
<i>p,p'</i> -DDT	0.22	0.20	0.22	80 %	6.9	4.1	9.4	80 %
DDTs	2.10	1.23	1.71	100 %	123.1	44.8	145.7	100 %
Mirex	0.01	0.02	0.02	20 %	1.7	1.9	1.7	80 %
OCPs	7.27	6.28	3.32	100 %	542.0	222.1	636.5	100 %

A-mean arithmetic mean, *G-mean* geometric mean, *SD* standard deviation

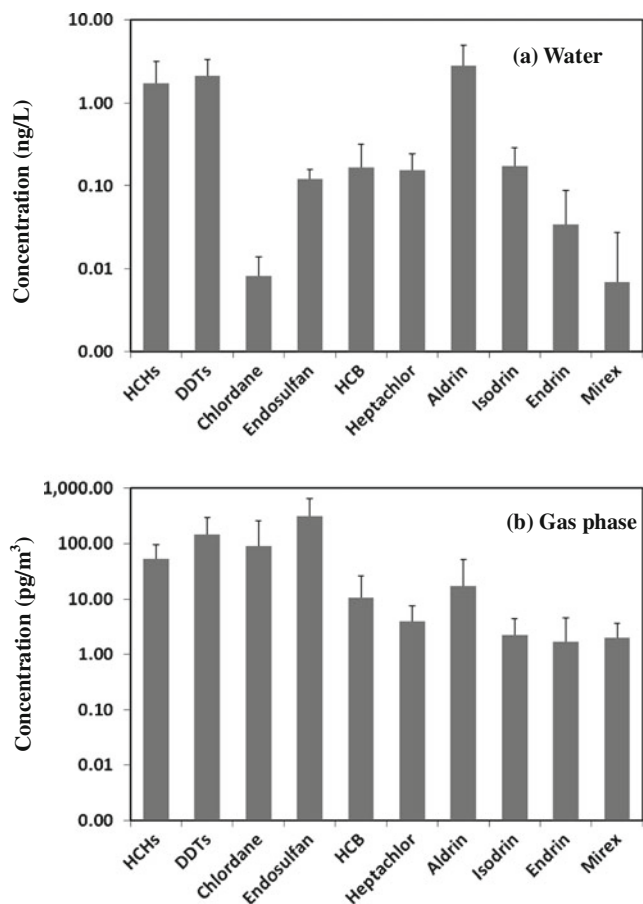


Fig. 2 Distributions of OCPs in the water (a) and gas phase (b) at Lake Chaohu

The residual level of the HCHs in the water was lower than that in the Yangtze River catchment of Wuhan (5.18 ± 2.59 ng/l) (Tang et al. 2008), the Qiantang River in Zhejiang (33.07 ± 14.64 ng/l) (Zhou et al. 2008) and the Jiulong River in Fujian (71.1 ± 85.5 ng/l) (Maskaoui et al. 2005), and it was higher than that in Lake Co Ngoin in Tibet (0.3 ng/l) (Zhang et al. 2003), and similar to that in Lake Baiyangdian (2.1 ± 0.8 ng/l) (Hu et al. 2010). The residual level of DDTs was relatively low compared with the Guanting Reservoir in Beijing ($3.71\text{--}16.03$ ng/l) (Wan et al. 2009), the Huangpu River ($3.83\text{--}20.90$ ng/l) (Xia et al. 2006) and Soozhow Creek ($17\text{--}99$ ng/l) (Jiang et al. 2000). The residual level of Aldrin was lower than that in the Pearl River artery estuary in the low flow season (4.17 ± 3.07 ng/l) (Yang et al. 2004), the karst subterranean river in Liuzhou (9.22 ± 1.90 ng/l) (Wei et al. 2011) and the Kucuk Menderes River in Turkey ($17\text{--}1790$ ng/l) (Turgut 2003); it was higher than that in the Xiangjiang River catchment of Changsha ($0.22\text{--}0.51$ ng/l) (Tian et al. 2010) and the Yangtze River catchment of Wuhan (1.88 ng/l) (Zhi et al. 2008); and it was similar to that in the Guanting Reservoir in Beijing (2.26 ± 2.84 ng/l) (Kang et al. 2003).

Twenty types of gaseous OCPs were detected in the atmosphere at Lake Chaohu, including HCHs (α -, β -, γ -, δ -HCH), DDTs (*o,p'*-, *p,p'*-DDE, DDT, DDD), HCB, heptachlor, aldrin, isodrin, endrin, chlordane (α -, γ -chlordane), endosulfan (endosulfan I,II) and mirex, with a total concentration of 542.0 ± 636.5 pg/m³. Endosulfan (262.5 ± 338.9 pg/m³), DDTs (123.1 ± 145.7 pg/m³) and chlordane (45.9 ± 43.2 pg/m³) were the major gaseous OCPs in the atmosphere, accounting for 48.9 %, 22.5 % and 14.4 % of the total, respectively. *o,p'*-DDE was the main metabolite of DDT in the gas phase, and α -HCH (37.9 %) and γ -HCH (30.9 %) were the dominant HCHs isomers in the gas phase.

The residual levels of HCHs and DDTs in the gas phase at Lake Chaohu were lower than those in other regions, such as Guangdong (HCHs 0.655 ng/m³, DDTs 1.458 ng/m³), Hong Kong (HCHs 0.161 ng/m³, DDTs 0.444 ng/m³), Anhui (HCHs 0.185 ng/m³, DDTs 0.297 ng/m³), Hebei (HCHs 0.117 ng/m³, DDTs 0.336 ng/m³), Jiangsu (HCHs 0.0948 ng/m³, DDTs 0.771 ng/m³), Seoul in South Korea (HCHs 0.262 ng/m³, DDTs 0.033 ng/m³), Japan (HCHs 0.124 ng/m³, DDTs 0.011 ng/m³), Alabama in the USA (HCHs 0.168 ng/m³, DDTs 0.011 ng/m³), Mexico (HCHs 0.103 ng/m³, DDTs 0.574 ng/m³) and Belize in America (DDTs 1.159 ng/m³). These levels were even 2 orders of magnitude lower than those in India (HCHs 0.91–35.57 ng/m³) (Murayama et al. 2003; Jantunen et al. 2000; Alegria et al. 2000, 2006; Shao et al. 2007).

Temporal distributions of OCPs in the water and gas phase

The temporal distributions of the OCPs in the water and gas phase are shown in Tables 3 and 4 and Fig. 5.

The concentration of the total OCPs in the water at Lake Chaohu varied by month. The highest mean concentration

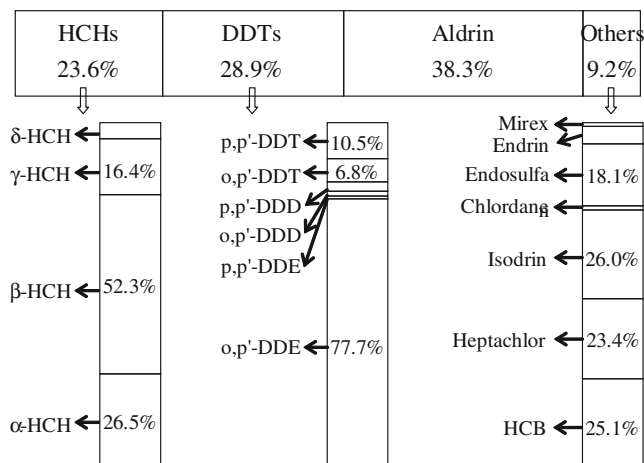
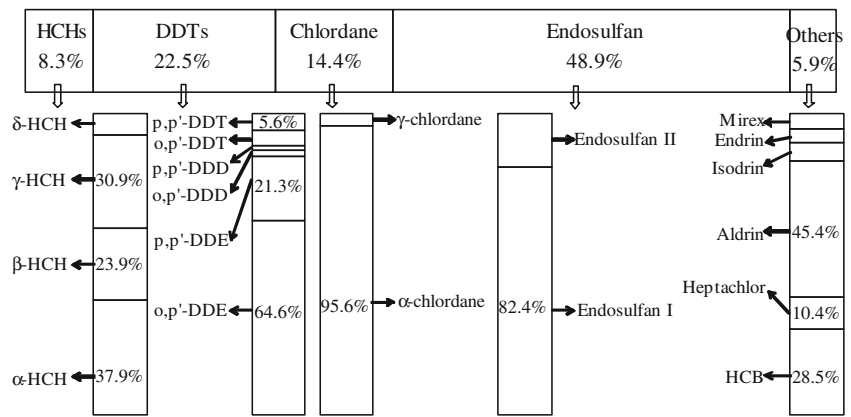


Fig. 3 The OCP components in the water at Lake Chaohu

Fig. 4 The gaseous OCP components in the atmosphere at Lake Chaohu



(12.32 ng/l) was found in September, while the lowest (1.74 ng/l) was found in November. Aldrin, the main type of OCPs in the water, showed a similar distribution to the total OCPs, and it reached a peak in September. The HCHs concentrations in the water leveled off between May and October, decreased in November and December, and recovered and peaked in January and February. The DDTs concentrations were relatively high from May to August and

relatively low between November and February. For both HCHs and DDTs, the concentration level was higher in spring and summer than that in autumn. In winter, however, the HCHs concentration reached a peak, while the DDTs reached the lowest point. This discrepancy may be due to the change of water volume and the use of related pesticides, for instance HCHs and DDTs (see the section “Possible sources of DDTs and HCHs” for details). During June and

Table 3 Temporal distribution of OCPs in the water at Lake Chaohu (ng/l)

	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb
HCB	0.10	0.17	0.20	0.17	0.16	0.21	0.06	0.07	0.25	0.29
α-HCH	0.44	0.27	0.35	0.16	0.32	0.35	0.14	0.14	0.69	1.69
β-HCH	0.66	0.62	0.70	0.86	1.12	0.71	0.43	0.45	1.59	1.86
γ-HCH	0.23	0.22	0.26	0.16	0.19	0.12	0.07	0.07	0.37	1.13
δ-HCH	0.11	0.07	0.07	0.01	0.04	0.05	0.02	0.03	0.08	0.34
HCHs	1.44	1.17	1.38	1.19	1.67	1.23	0.66	0.68	2.72	5.01
Heptachlor	ND	0.01	0.02	0.23	0.08	0.07	0.07	0.08	0.46	0.55
Aldrin	2.09	1.93	3.36	2.28	8.23	2.78	0.38	1.05	2.18	3.60
Isodrin	0.05	0.48	0.18	0.51	0.05	0.14	0.06	0.04	0.13	0.09
γ-Chlordane	ND	ND	0.06	ND	ND	0.01	0.01	ND	ND	ND
α-Chlordane	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Chlordane	0.00	ND	0.06	ND	ND	0.01	0.01	ND	ND	ND
Endosulfan I	ND	ND	ND	ND	0.02	0.03	0.03	0.02	0.08	0.03
Endosulfan II	0.91	ND	ND	0.01	0.05	0.01	0.01	0.01	ND	ND
Endosulfan	0.91	ND	ND	0.01	0.06	0.05	0.03	0.02	0.09	0.03
Endrin	0.13	0.02	0.11	ND	0.02	0.06	ND	ND	ND	ND
o,p'-DDE	3.61	2.43	4.37	3.80	2.01	0.03	0.02	0.03	0.07	ND
p,p'-DDE	ND	0.05	ND	ND	ND	0.12	0.01	ND	ND	ND
o,p'-DDD	0.03	0.13	ND	ND	ND	0.07	ND	ND	0.06	ND
p,p'-DDD	ND	0.27	ND	0.01	ND	0.27	0.03	ND	ND	ND
o,p'-DDT	ND	0.07	ND	0.05	ND	1.26	0.05	0.01	ND	ND
p,p'-DDT	ND	ND	0.11	0.38	0.04	0.72	0.35	0.25	0.11	0.27
DDTs	3.64	2.94	4.48	4.23	2.05	2.47	0.46	0.28	0.24	0.27
Mirex	ND	ND	0.06	ND	ND	ND	0.01	ND	ND	ND
OCPs	8.36	6.72	9.84	8.60	12.32	7.01	1.74	2.23	6.08	9.83

August, the water flowing into Lake Chaohu is more than that flowing out, resulting in increased water volume. This increase in water may have reduced the residual level of the OCPs by dilution or increased the level by bringing in new pollutants. The use of OCPs around the lake may result in increased residual levels in spring and summer, when there are more agricultural activities. Other industrial products containing HCHs or DDTs may also lead to an irregular seasonal variation. The distributions of the other OCPs in the water showed no distinct trend.

The temporal distribution of total gaseous OCPs is unimodal, with a significantly higher mean concentration in summer than in winter, which may be due to the high temperature in summer, which promotes volatilization from the water to the atmosphere. The HCHs (except in July), DDTs and endosulfan present a similar distribution to that of the total OCPs. Chlordane was detected in all 10 months, but the concentrations in September, October and November were 2 orders of magnitude greater than those in the other months.

Water–gas exchange of OCPs

The water–gas exchange of OCPs depends on the fugacity of OCPs in the water and gas phase. The fugacity gradient, f_w/f_g , was calculated using the following formula:

$$f_w/f_g = \frac{C_w H}{C_g R T}$$

where f_w and f_g are the water fugacity and gas fugacity, C_w and C_g are the concentration in the water and gas phase (ng/m^3), H is the Henry's law constant ($\text{Pa}/(\text{m}^3 \text{mol})$), R is the gas constant ($8.314 \text{ Pa}/(\text{m}^3 \text{molK})$), and T is the temperature of the water–gas interface (K). In general, $f_w/f_g > 1$ indicates a volatilization from the water to gas phase, and $f_w/f_g < 1$ indicates a deposition from the gas phase to water. f_w/f_g is approximately 1.0 for a relatively balanced water–gas system.

The f_w/f_g of the five OCP types was calculated based on the parameter values in Table 5. Table 6 illustrates the f_w/f_g values, which indicate the transfer direction of those OCPs. α -HCH (except in November and

Table 4 Temporal distribution of OCPs in the gas phase at Lake Chaohu (pg/m^3)

OCPs	March	April	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb
HCB	2.0	7.3	1.2	1.0	1.1	33.3	2.3	9.6	4.3	41.7	12.7	ND
α -HCH	12.6	ND	7.9	18.5	6.8	58.9	25.2	22.3	17.1	26.3	7.3	12.2
β -HCH	1.2	ND	17.3	59.7	ND	27.0	6.8	5.6	3.0	1.8	2.4	4.0
γ -HCH	2.8	ND	9.3	29.5	27.9	45.9	18.2	14.6	9.0	4.2	2.3	4.3
δ -HCH	0.9	ND	1.5	2.1	0.2	19.6	5.3	5.8	2.5	ND	0.8	0.6
HCHs	17.6	ND	36.0	109.9	34.9	151.3	55.6	48.3	31.6	32.2	12.8	21.1
Heptachlor	ND	4.4	ND	ND	1.5	6.7	4.3	7.8	2.1	8.2	8.6	ND
Aldrin	6.8	ND	ND	101.0	59.7	ND	0.9	ND	1.7	4.8	2.8	ND
Isodrin	ND	1.5	3.0	5.1	1.9	6.0	0.7	1.4	0.7	3.5	ND	ND
γ -Chlordane	3.9	0.6	8.8	10.3	8.4	5.8	1.9	3.0	0.9	1.6	ND	ND
α -Chlordane	1.7	ND	3.8	5.3	4.6	504.9	245.6	97.7	0.4	0.7	3.6	14.1
Chlordane	5.6	0.6	12.6	15.6	13.0	510.7	247.5	100.7	1.3	2.3	3.6	14.1
Endosulfan I	13.2	ND	168.8	443.4	673.4	739.6	360.3	141.0	22.0	32.4	1.2	1.3
Endosulfan II	0.8	ND	37.4	98.0	229.5	127.3	42.5	13.1	2.5	2.2	ND	ND
Endosulfan	14.0	ND	206.2	541.5	902.9	866.9	402.8	154.1	24.6	34.6	1.2	1.3
Endrin	ND	ND	6.9	5.1	5.1	ND	ND	ND	ND	ND	ND	ND
<i>o,p'</i> -DDE	4.4	1.1	71.6	176.3	283.3	236.4	111.3	44.9	3.4	5.5	ND	0.6
<i>p,p'</i> -DDE	3.1	4.7	51.6	72.8	57.4	71.9	14.4	19.9	9.5	10.3	ND	ND
<i>o,p'</i> -DDD	ND	ND	ND	ND	14.0	6.5	1.3	ND	ND	ND	4.2	3.4
<i>p,p'</i> -DDD	ND	ND	1.9	4.7	3.4	5.4	1.2	1.1	ND	ND	0.7	ND
<i>o,p'</i> -DDT	15.7	1.0	13.7	18.5	21.2	13.5	3.2	3.2	1.3	1.0	ND	ND
<i>p,p'</i> -DDT	1.4	0.8	16.4	23.8	25.4	6.3	2.2	2.7	1.5	2.3	ND	ND
DDTs	24.5	7.6	155.2	296.1	404.6	340.1	133.5	71.8	15.6	19.1	4.9	4.0
Mirex	ND	ND	3.8	4.0	2.9	3.4	3.3	1.8	0.5	0.4	ND	ND
OCPs	70.4	21.3	424.9	1079.2	1427.6	1918.5	850.9	395.5	82.4	146.8	46.6	40.4

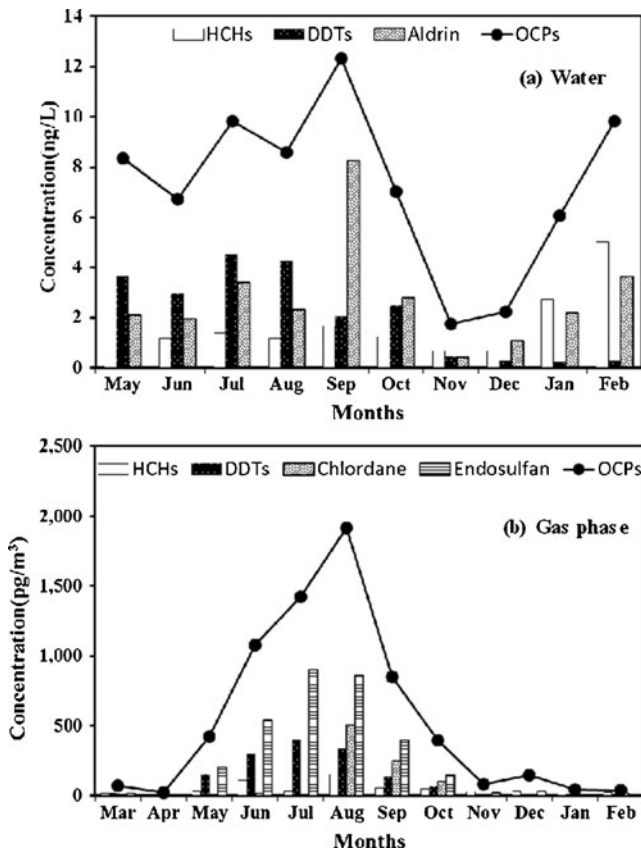


Fig. 5 Temporal distributions of OCPs in the water (a) and gas phase (b) at Lake Chaohu

December), γ -HCH (except in August, October, November and December), DDT (except in June) and DDE showed a significant volatilization trend from the water to the gas phase.

Having been determined by f_w/f_g , the net water–gas flux was calculated using the following formula:

$$F_g = K_g \left(C_g - \frac{C_w H}{RT} \right)$$

$$K_g = \frac{1}{\frac{1}{k_a} + \frac{H}{RTk_w}}$$

where K_a and K_w are the air-side transport velocity and water-side transport velocity, respectively. The parameter values are listed in Table 5, where T (K) is the mean temperature of Hefei between 1950 and 2000, and k_a and k_w are values after temperature correction.

Figure 6 illustrates the calculated F_g of the five types of OCPs. The black lines represent the mean fluxes in each month. Compared with water–gas fluxes, the gas–water fluxes were insignificant. The average fluxes were -21.11 , -3.30 , -152.41 , -35.50 and $-1,314.15$ $\text{ng}/(\text{m}^2 \text{day})$ for α -HCH, γ -HCH, HCB, DDT and DDE, respectively. Monte Carlo simulation was used to calculate the uncertainties of the net fluxes. In the present study, 5,000 times iterations were conducted. The uncertainties were demonstrated by grey areas in Fig. 6, displaying the range between the 25th and 75th percentiles. The majority of the grey areas were below zero, indicating the exchanges of α -HCH, γ -HCH, HCB, DDT and DDE were mostly from the water to the atmosphere, which suggested that the water was the main potential source of gaseous OCPs in the atmosphere.

A parameter sensitivity analysis was conducted to find out the most sensitive parameter that affects F_g . In this study, all the six parameters (except the gas constant R)

Table 5 Parameter values for calculating the water–gas exchange

		May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb
α -HCH	T (K)	294.5	298.2	301.3	300.8	296.1	290.4	283.9	278.2	275.8	277.7
	H (Pa/(m ³ mol))	0.64	0.86	1.11	1.06	0.73	0.46	0.28	0.18	0.14	0.17
	k_a (m/day)	402	402	402	402	402	402	402	402	402	402
γ -HCH	k_w (m/day)	0.94	1.01	1.08	1.07	0.97	0.85	0.72	0.60	0.55	0.59
	H (Pa/(m ³ mol))	0.30	0.39	0.48	0.46	0.34	0.23	0.15	0.10	0.09	0.10
	k_a (m/day)	402	402	402	402	402	402	402	402	402	402
HCB	k_w (m/day)	0.94	1.01	1.08	1.07	0.97	0.85	0.72	0.60	0.55	0.59
	H (Pa/(m ³ mol))	38.81	49.32	60.30	58.38	43.05	29.75	19.53	13.50	11.55	13.07
	k_a (m/day)	508	508	508	508	508	508	508	508	508	508
DDT	k_w (m/day)	1.21	1.31	1.39	1.38	1.25	1.10	0.92	0.77	0.70	0.75
	H (Pa/(m ³ mol))	0.72	1.01	1.35	1.29	0.83	0.49	0.27	0.16	0.13	0.15
	k_a (m/day)	356	356	356	356	356	356	356	356	356	356
DDE	k_w (m/day)	0.84	0.91	0.97	0.96	0.87	0.76	0.64	0.53	0.48	0.52
	H (Pa/(m ³ mol))	29.03	41.75	56.60	53.89	33.97	19.41	10.25	5.86	4.63	5.58
	k_a (m/day)	365	365	365	365	365	365	365	365	365	365
		0.84	0.91	0.97	0.96	0.87	0.76	0.64	0.53	0.48	0.52

Table 6 f_w/f_g values at different times

f_w/f_g	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb
α -HCH	14.56	5.06	22.81	1.15	3.77	2.99	0.97	0.41	5.77	10.20
γ -HCH	3.03	1.17	1.79	0.64	1.44	0.78	0.49	0.72	6.31	11.38
HCB	1320.89	3381.85	4376.69	119.17	1216.51	269.54	115.45	9.80	99.15	–
DDT	0	0.67	1.27	11.20	2.50	68.11	16.34	5.45	–	–
DDE	347.41	167.66	289.81	265.60	220.65	18.61	10.10	4.81	–	0

were assumed to change $\pm 10\%$. The sensitivity of parameter X ($S(x)$) is defined as follows,

$$S(x) = ABS\left(\frac{(F_{g1} - F_{g2})/F_{g0}}{(X_1 - X_2)/X_0}\right) = ABS\left(\frac{F_{g1} - F_{g2}}{0.2 \times F_{g0}}\right)$$

where F_{g1} , F_{g2} and F_{g0} are F_g values when the parameter X was multiplied by 0.9, 1.1 and 1.0, respectively. X_1 , X_2 and X_0 are parameter values after multiplied by 0.9, 1.1 and 1.0, respectively.

As shown in Fig. 7, F_g of α -HCH, γ -HCH and DDT are highly sensitive to C_w , C_g , T and H , while F_g of HCB and

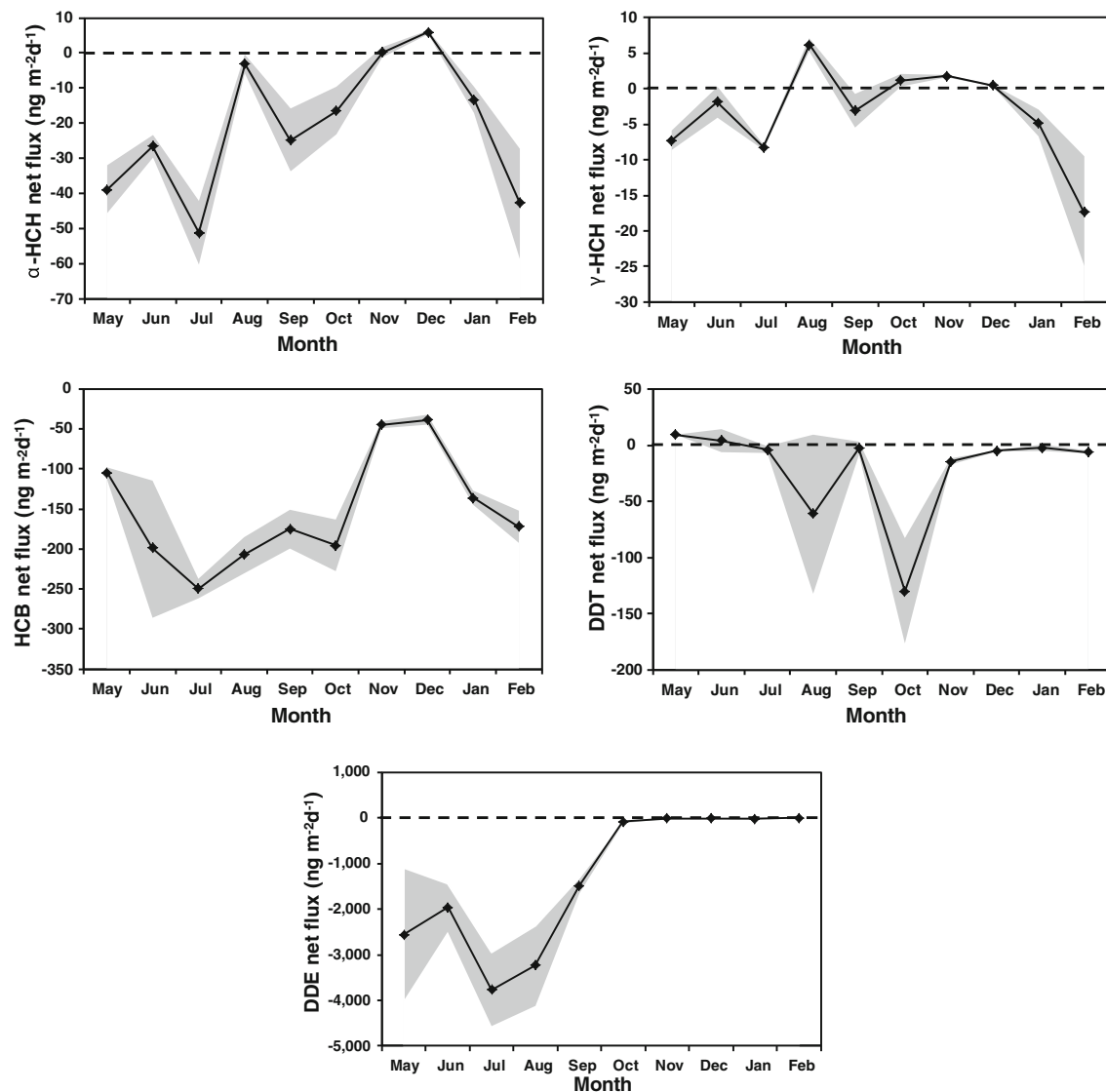


Fig. 6 Temporal distributions of α -HCH, γ -HCH, HCB, DDT and DDE net fluxes from May 2010 to February 2011. *Black line* represents mean flux. *Grey area* represents flux range between 25th and 75th percentile

DDE are more sensitive to C_w and K_w . $\sum S(x)$ of α -HCH, γ -HCH, HCB, DDT and DDE between May 2010 and February 2011 were 3.9–137.4, 4.3–27.0, 2.2–2.9, 2.2–18.4, and 2.2–3.8, respectively. The variations and discrepancies of $\sum S(x)$ imply that time and species are significant factors that have influences on the sensitive of F_g . The sensitivity analysis indicated that the F_g of α -HCH, γ -HCH and DDT are more vulnerable than that of HCB and DDE to the variation of the parameters.

Possible sources of DDTs and HCHs

The possible sources of DDTs and HCHs can be identified by looking at the isomer ratios of these pesticides (Iwata et

al. 1993, 1995; Willett et al. 1998; Qiu et al. 2004; Ouyang et al. 2012). For the case of HCHs, the ratios of α -/ γ -HCH and β -/(α + γ)-HCH can be used to gauge the age and source of the HCHs residues. Two types of HCHs products, technical HCH and lindane, have been used around the world: technical HCH consists of 60–70 % α -HCH, 5–12 % β -HCH and 10–15 % γ -HCH, and lindane contains more than 99 % γ -HCH (Qiu et al. 2004). The α -/ γ -HCH ratio for technical HCH ranges from 3 to 7, while that for lindane is less than 0.1. Therefore, high ratio of α -/ γ -HCH indicates the input of technical HCHs, while low ratio points to the dominant use of lindane (Willett et al. 1998). If the α -/ γ -HCH ratio is greater than 7, the HCHs may come from atmospheric input (Iwata et al. 1993, 1995), since α -HCH

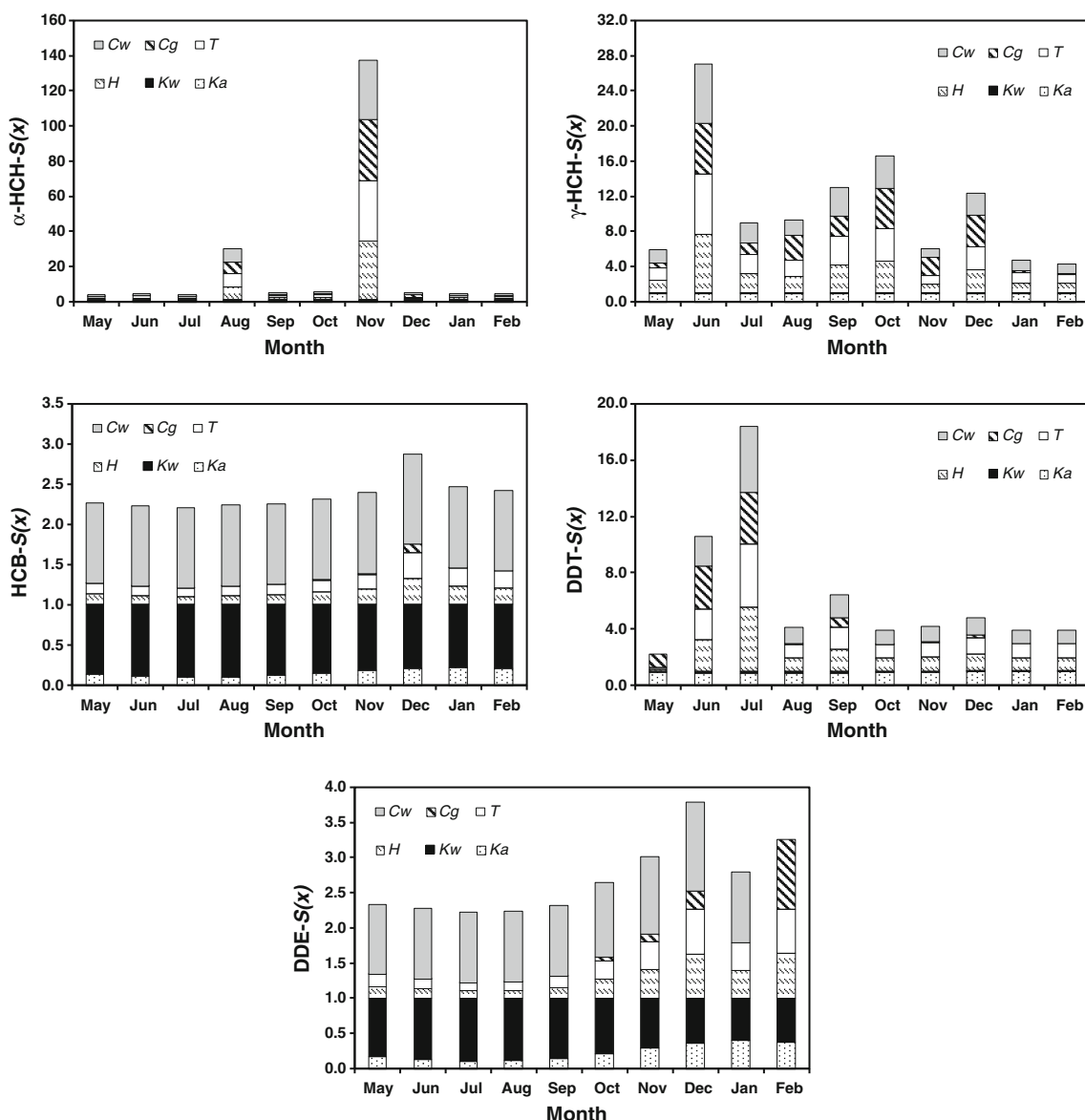


Fig. 7 Sensitivity analysis of F_g to the input parameters C_w , C_g , T , H , K_w and K_a

with a high vapor pressure tends to be residual in the atmosphere and can be transported for long distances. Compared with other three HCH isomers, the β -HCH is the most stable isomer with the lowest biodegradation in the environment. Therefore, the β -/(α + γ)-HCH ratio can be used to gauge the age of the HCHs residues. A high β -/(α + γ)-HCH ratio indicates a historical usage of technical HCH or lindane (Willett et al. 1998). When the β -/(α + γ)-HCH ratio is less than 0.5, the source of the HCHs may be the recent usage of lindane or atmospheric transport (Wang et al. 2012; Ouyang et al. 2012).

For the case of DDTs, proportion of DDT congeners can be used to gauge the age and source of the DDT residues. The ratio of DDT/(DDE+DDD) ≥ 1 indicates the new input of DDT, while the ratio < 1 means the history application (Wang et al. 2012; Ouyang et al. 2012). A high proportion of the parent compound, *p,p'*-DDT indicates a fresh signature while a low proportion implies an old residue (Harner et al. 2000). There are no definitive guidelines as to what constitutes a high and low ratio, but comparisons can be made among the regions studied. Ratios easy to understand, difficult to average, therefore, fractions are preferred to ratios for statistics (Harner et al. 2000). A high proportion of the parent compound, *p,p'*-DDT, thus higher F(DDT_O) (F(DDT_O) = *p,p'*-DDT/(*p,p'*-DDT + *o,p'*-DDT)) values indicates a fresh signature of technical DDT, while a low proportion implies an old technical DDT residue (Harner et al. 2000). The F(DDT_O) is lower for residues transported from China, due to the prevalence of *o,p'*-DDT in "dirty dicofol." The World Health Organization gives the composition of technical DDT as follows: 77 % *p,p'*-DDT, 15 % *o,p'*-DDT, 4 % *p,p'*-DDE, and the rest other residues (WHO 1989). In Chinese dicofol, the average contents of *o,p'*-DDT, *p,p'*-Cl-DDT, *o,p'*-DDE, and *p,p'*-DDT

were 114, 69, 44, and 17 g/kg, respectively (Qiu et al. 2004). So, the F(DDT_O) for technical DDT are 0.84 and 0.95, respectively, while the F(DDT_O) for Chinese dicofol is 0.13.

The ratios of α -/ γ -HCH and β -/(α + γ)-HCH as well as DDT/(DDE+DDD) and F(DDT_O) can be calculated for the water from Table 3, and for the air from Table 4, and the results are tabulated in Table 7. The ratios of α -/ γ -HCH and β -/(α + γ)-HCH for the water were 1.74 and 1.61, ranging from 1.23 to 2.0 and from 0.66 to 2.69, respectively. This means that the possible source of the HCHs in the water was from the historical usage of lindane. However, the possible source of the HCHs in the air was mainly from the recent usage of lindane, as indicated by the ratios of α -/ γ -HCH and β -/(α + γ)-HCH for the air in Table 7. The α -/ γ -HCH ratios for the air ranged from 0.24 to 1.96 during May to November, indicating the source of lindane. The higher α -/ γ -HCH ratios in December, January and March indicated the input of technical HCHs or the effects of atmospheric α -HCH from northern China in the Winter season. The β -/(α + γ)-HCH ratios in the air in March and during August to February were less than 0.3, which indicates the recent usage of lindane or atmospheric α -HCH (Wang et al. 2012; Ouyang et al. 2012).

The F(DDT_O)s in the air during April to December ranged from 0.32 to 0.70, with the average value of 0.50, which were lower than those of technical DDT, and higher than those of dicofol (Table 7). This seems to imply that the technical DDT and dicofol might be the source of DDTs in the air. The DDT/(DDE+DDD) ratios in the air during April to December were much lower than 1.0, indicating the source of historical DDT. In March, the recent input of dicofol was the major source of DDTs in the air, as indicated by the lowest F(DDT_O) value (0.08) and highest DDT/(DDE + DDD) ratio (2.28) (Table 7). However, technical DDT was

Table 7 The isomer ratios of HCHs and DDTs in the water and air at Lake Chaohu

	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb
Air												
α -/ γ -HCH	4.50	/	0.85	0.63	0.24	1.28	1.38	1.53	1.90	6.26	3.17	2.84
β -/(α + γ)-HCH	0.08	/	1.01	1.24	/	0.26	0.16	0.15	0.11	0.06	0.25	0.24
DDT/(DDE+DDD)	2.28	0.31	0.24	0.17	0.13	0.06	0.04	0.09	0.22	0.21	/	/
<i>o,p'</i> -/ <i>p,p'</i> -DDT	11.21	1.25	0.84	0.78	0.83	2.14	1.45	1.19	0.87	0.43	/	/
F(DDT _O)	0.08	0.44	0.54	0.56	0.55	0.32	0.41	0.46	0.54	0.70	/	/
F(DDT _E)	0.31	0.15	0.24	0.25	0.31	0.08	0.13	0.12	0.14	0.18	/	/
Water												
α -/ γ -HCH	/	/	1.91	1.23	1.35	1.00	1.68	2.92	2.00	2.00	1.86	1.50
β -/(α + γ)-HCH	/	/	0.99	1.27	1.15	2.69	2.20	1.51	2.05	2.14	1.50	0.66
DDT/(DDE+DDD)	/	/		0.02	0.03	0.11	0.02	4.04	6.67	8.67	0.85	/
<i>o,p'</i> -/ <i>p,p'</i> -DDT	/	/	/	/	/	0.13	/	1.75	0.14	0.04	/	/
F(DDT _O)	/	/	/	/	/	0.88	/	0.36	0.88	0.96	/	/
F(DDT _E)	/	/	/	/	/	/	/	0.86	0.97		/	/

F(DDT_O) = *p,p'*-DDT/(*p,p'*-DDT + *o,p'*-DDT); F(DDT_E) = *p,p'*-DDT/(*p,p'*-DDT + *p,p'*-DDE)

the major source of DDTs in the water in August, October and December, as indicated by the $F(\text{DDT}_o)$ values of 0.88, 0.88 and 0.96, respectively. The $F(\text{DDT}_o)$ in the water in October (0.36) was nearby that of dicofol, indicating dicofol might be the major source of DDTs. The $\text{DDT}/(\text{DDE}+\text{DDD})$ ratio indicated the recent input of dicofol to the water in October, and the recent input of technical DDT in November and December, as indicated by the ratio values of 4.04, 6.67 and 8.67, respectively (Table 7).

Conclusions

Nineteen types of OCPs were detected in the water, with a total concentration of 7.27 ± 3.32 ng/l. Aldrin (38.3 %), DDTs (28.9 %) and HCHs (23.6 %) were the major OCPs in the water. Twenty types of OCPs were detected in the gas phase, with a total concentration of 542.0 ± 636.5 pg/m³. Endosulfan (48.9 %), DDTs (22.5 %) and chlordane (14.4 %) were the major OCPs in the gas phase. The temporal distribution of the total OCPs in the water at Lake Chaohu varied according to month. The highest mean concentration (12.32 ng/l) was found in September, while the lowest (1.74 ng/l) was found in November. The temporal distribution of the total gaseous OCPs was unimodal, with a significantly higher mean concentration in summer than in winter. *o,p'*-DDE was the main metabolite of DDT in both the water and gas phase. Of the HCHs, 52.3 % existed as β -HCH in the water, while α -HCH (37.9 %) and γ -HCH (30.9 %) were the dominant forms in the gas phase. The average fluxes were -21.11 , -3.30 , -152.41 , -35.50 and $-1,314.15$ ng/(m² day) for α -HCH, γ -HCH, HCB, DDT and DDE, respectively. Compared with water–gas fluxes, the gas–water fluxes were insignificant. The exchanges of α -HCH, γ -HCH, HCB, DDT and DDE were mostly from the water to the atmosphere, which suggested that the water was the main potential source of gaseous OCPs in the atmosphere. The F_g of α -HCH, γ -HCH and DDT are highly sensitive to C_w , C_g , T and H , while F_g of HCB and DDE are more sensitive to C_w and K_w . The sensitivity analysis indicated that the F_g of α -HCH, γ -HCH and DDT are more vulnerable than that of HCB and DDE to the variation of the parameters. The possible source of the HCHs in the water is form the historical usage of lindane; however, that in the air is mainly from the recent usage of lindane. The technical DDT and dicofol might be the source of DDTs in the water and air.

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