1 **Spatial correlation analysis of polycyclic aromatic hydrocarbons (PAHs) and**
2 **organochlorine pesticides (OCPs) in sediments between Taihu Lake and its**
3 **tributary rivers**

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10 **Abstract**

11 The residues of polycyclic aromatic hydrocarbons (PAHs) and organochlorine
12 pesticides (OCPs) in surface sediments from Taihu Lake basin (THB) and Taihu Lake
13 body (THL) were investigated. Higher concentrations of both PAHs and OCPs were
14 observed for THB than THL. The concentrations of PAHs ranged from 12.1 to 2281.1
15 ng g⁻¹ dw for THB and from 11.4 to 209.9 ng g⁻¹ dw for THL, while OCPs ranged
16 from 16.3 to 96.9 ng g⁻¹ dw and from 16.8 to 61.9 ng g⁻¹ dw for THB and THL,
17 respectively. Spatial distribution of PAHs and OCPs showed a high correspondence
18 with the land use of THB and surrounding anthropogenic activity. Additionally, the
19 Kriging interpolation plots demonstrated that the major upper reaches were more
20 polluted than the lower reaches, indicating the transport of pollutants with the water

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flow direction. The organic matter contents were responsible for OCP distribution other than PAHs due to the biodegradation capacity difference of chemicals. Similar compositions of pollutants were observed with 3- and 4-ringed PAHs accounting for a total of 78.3% for THB and 85.8% for THL, respectively. HCHs and DDTs were predominant OCPs, which contributed to 31.8% and 21.7% for THB, and 33.6% and 21.9% for THL, respectively. The isomeric and parent substance/metabolite ratios implied fresh inputs of DDTs and chlordanes, while HCHs and endosulfans were mainly from old usage. PAH source identification performed by diagnostic ratios demonstrated the mixed sources of petrogenic and pyrogenic ones dominated by grass, wood and coal combustion. Furthermore, the hazard quotient (HQ) based on the consensus-based sediment quality guidelines (SQGs) was used to evaluate the ecological risks of sediments. Although no frequently adverse effects were observed, potential ecological risks induced by Ant, BaA, γ -HCH, dieldrin, p,p'-DDT and chlordanes should also be paid attention to considering the continuous inputs of such pollutants.

Keywords

Hazard quotient, PAHs, OCPs, spatial distribution, source identification

1 Introduction

Polycyclic aromatic hydrocarbons (PAHs) are highly toxic organic pollutants containing typically two to eight benzene-member rings (Zhao et al., 2016; Heywood

et al., 2006). They are ubiquitous derived primarily during the pyrolysis of organic materials typical of some processes used in the iron and steel industry, heating and power generation, petroleum refining and the release of petroleum products, among which 16 congeners were defined as the priority pollutants by USEPA due to their highly mutagenic and carcinogenic toxicity and have been widely used to evaluate the pollution of PAHs in environment (Feng et al., 2012; Wang et al., 2015). Organochlorine pesticides (OCPs) containing at least one covalently bonded atom of chlorine were once widely produced and used globally with a long history (Zhao et al., 2009). Although the production and use of OCPs have been banned after the mid-1970s, they are still found in almost all the environmental matrices such as air, water, snow, soil and biota due to their long half-life. Both PAHs and OCPs are of great concern around the world for their persistence, impact on non-target organisms, and bioaccumulation in the tissues of animals as well as humans via the food chain (Nakata et al., 2002; Tang et al., 2007; Ogbeide et al., 2016). More explicit, four or more rings PAHs, hexachlorocyclohexanes (HCHs) and dichlorodiphenyltrichloroethanes (DDTs) may induce a number of adverse effects, such as immunotoxicity, genotoxicity, carcinogenicity, and reproductive toxicity. Consequently, they are considered as endocrine-disrupting chemicals with great potential risk to human health (Sverdrup et al., 2002; Qiao et al., 2006).

Due to the low water solubility and high n-octanol/water partition coefficient (K_{ow}) values, PAHs and OCPs have strong affinities for suspended particulate matter and are subsequently accumulated into stream, river, ocean and marine sediments. PAHs and

OCPs can be absorbed by suspended materials from water and then deposited to become a part of the bottom sediments (Yang et al., 2005; Barhomumi et al., 2014). Thus, sediments can serve as sinks of PAHs and OCPs. Moreover, PAHs and OCPs that were historically deposited can be remobilized upward in sediments and re-suspended into the aquatic environment and, resulting in a long term contamination of aquatic environments. The transfer of PAHs and OCPs from sediments to benthic organisms, through interaction between water and sediment, is now considered as a major route of exposure for many species (Zoumis et al., 2001). Therefore, as a sink of pollutants in aquatic environments, the residues of PAHs and OCPs in sediments can provide valuable records of pollution and denote environmental risks. An investigation into the occurrence, sources and ecological risks of PAHs and OCPs in aquatic sediments can contribute to a better understating of the status of these pollutants in sediments and can facilitate the maintenance of the aquatic ecosystem health and ecological safety.

Moreover, it is known that surface runoff picks up and carries natural and anthropogenic pollutants as the water flow moves, which eventually transports these pollutants into receiving waters (Chen et al., 2004). Due to their persistence, PAHs and OCPs could move long distances in surface runoff, which could be originated far away from the receiving waters. The freshwater lakes, especially those large shallow lakes which are very susceptible to wind-generated sediment resuspension, are the major water supplies for the public and suffering from a large amount of pollutant inputs due to the surrounding anthropogenic activities. It has been certified that the

universe eutrophication status of such freshwater lakes are mainly induced by the nutrient inputs from point or no-point sources around (Wang et al., 2004; Qin et al., 2007). However, most previous studies focused on the fate of PAHs and OCPs in lake or river sediments were conducted separately without considering the potential interactions between the surrounding basin and the lake body. These former studies ignored the ecosystem integrity of lakes and their surrounding tributary rivers as well as the internal interaction between these two compartments, which will induce the one-sidedness of related investigation and quality assessment of lakes. It is necessary to elucidate the pollution status of PAHs and OCPs in both freshwater lakes and its surrounding tributary rivers not only from the understanding of the biogeochemical processes of such pollutants but also from the lake protection perspective.

Subsequently, the objective of this study was to elucidate the residues of PAHs and OCPs in surface sediments from both tributary rivers and lake body, and then the spatial correlations between upper reaches and lower reaches of the specific freshwater lake could be analyzed, which would provide a comprehensive view point to study on the transport of these hydrophobic pollutants. Additionally, source identifications based on typical chemical indexes were performed to demonstrate the origins of such pollutants. Moreover, potential ecological risks of sediments induced by target pollutants were also evaluated, which were helpful for establishing field monitoring and pollution control plans.

2 Materials and methods

2.1 Study area and Sampling

The Taihu Lake Basin (30°7'19" to 32°14'56"N and 119°3'1" to 121°54'26"E) is situated in the Yangtze River Delta in eastern China and covers an area of approximately 36900 km² including a water area of approximately 6134 km², of which rivers and lakes comprise around 50%. The basin is covered naturally by subtropical evergreen broadleaf forest, and there are various topographical types, including hilly areas toward the west, and low-lying flood plain in the northern and eastern parts occupying 83% of the basin. There are more than 200 rivers distributed in the basin, and 172 rivers of channels are connected to Taihu Lake (Zhang et al., 2014). The dense river network comprises 7% of the total drainage area, with a total tributary length of around 120000 km (Huang et al., 2015). The Taihu Lake Basin is one of the most industrialized areas in China. With a population of more than 37 million (2.7% of the total population of China), and as a significant industrial complex, this area contributes 11% to China's gross domestic product (GDP). The plain river network is the main wastewater discharge region of Southern Jiangsu province, and thus, the freshwater ecosystem of the basin is suffering from severe pollution induced by nutrients and kinds of toxic chemicals from point or non-point sources. Although there have been many studies performing the residues, sources and bioaccumulation of PAHs and OCPs in Taihu Lake, little information can be found referring to the fate of such pollutants in rivers surrounding Taihu Lake, not to

mention the investigation about possible interactions of pollutants between lakes and rivers.

According to the spatial distribution of major rivers around Taihu Lake, the Taihu Lake Basin (THB) was divided into 7 different districts such as the West Lake Area, the Yangchengdianmao Area, the Wuchengxiyu Area, the Hangjia Lake Area, the Puxi Area, the Pudong Area, the Zhexi Area, of which the West Lake Area, the Zhexi Area and the Wuchengxiyu Area are defined as the upper reaches of Taihu Lake, while the others are belong to the lower reaches. In addition, the Taihu Lake body (THL) was divided into the Meiliang Bay, the Center Lake, the Western Coastal Lake, the Gonghu Bay, the Zhushan Bay, the East Lake and the Eastern Coastal Lake. The major river catchment areas were designed as sampling sites, and then a total of 81 river sediments from THB as well as 31 lake sediments from THL were used to investigate the residues of PAHs and OCPs. The detailed sampling sites can be found in Fig. 1. The sediments were collected with a stainless grab sampler, and the surface sediments (0-5 cm) were homogenized from triplicate samples and immediately stored in a freezer (-20°C) before pretreatment.

Fig. 1.

2.2 Chemicals

A mixture of standard solution of OCPs containing α -, β -, γ -, δ -HCH, heptachlor, heptachlor epoxide, α -chlordane, γ -chlordane, aldrin, endrin, dieldrin, endosulfan I, endosulfan II, endrin aldehyde, endosulfan sulfate, endrin ketone, p,p'-DDE,

p,p'-DDD, p,p'-DDT, and methoxychlor was defined as the target pesticides. The 16 priority PAH compounds included naphthalene (NaP), acenaphthylene (Any), acenaphthene (Ana), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenz[ah]anthracene (DahA), benzo[ghi]perylene (BghiP), and indeno[1,2,3-cd]pyrene (InP) were analyzed in the present study. Both standard solutions were purchased from Supleco (USA). The stock standard solutions were dissolved in the suggested solvents with methanol/dichloromethane (1:1, v/v) for PAHs and n-hexane for OCPs, respectively. The working solutions were stored at 4°C and prepared daily with suitable dilutions before use.

All solvents used for pretreatment and instrumental analysis were chromatographic grade. The copper powder and anhydrous sodium sulfate were Soxhlet treated for 48 h using dichloromethane and then kept in n-hexane before use. The silica gel (80-100 mesh) and alumina (120-200 mesh) used for purification were also extracted for 48 h in a Soxhlet apparatus, activated in the oven at 180°C and 250°C for 12 h, respectively, and then deactivated with distilled water at a ratio of 3% (m/m).

2.3 Extraction and quantitative determination

The detailed procedures of extraction and purification were performed as described in Zhao et al. (2009, 2013). Ten grams of sediments were mixed with anhydrous sodium sulfate and were then extracted with dichloromethane by an accelerated

solvent extraction system (ASE, Dionex ASE-100, USA). The activated copper granules were also added to remove the potential interference of the element sulfur. The extraction was purified by a silica gel-alumina (2:1) column, and the target elution was collected and concentrated for the following instrumental analysis.

The quantification of OCPs and PAHs was completed as suggested by Zhao et al. (2009, 2013, 2016). An Agilent 7890 gas chromatograph equipped with a ^{63}Ni μ -electron capture detector (GC- μ ECD, Agilent 7890, USA) and gas chromatograph-mass spectrometry (GC-MS, Agilent 7890-5975C, USA) were used together to identify and determine the concentrations of OCPs in sediments. As for PAHs, their concentrations were determined using a high performance liquid chromatography (HPLC) coupled with a diode-array detector (DAD) and a series-wound fluorescence detector (FLD) (HPLC-DAD-FLD, Agilent 1200, USA).

All data were subject to strict quality assurance and quality control procedures, which were assessed previously for all of the target compounds (Zhao et al., 2016). The concentrations of OCPs and PAHs were determined by the external standard method on the basis of peak area and were expressed on a dry weight basis (ng g^{-1} dry weight, ng g^{-1} dw). The method detection limits (MDLs) were defined as the concentration of target compounds that yielded a peak signal-to-noise ratio (S/N) of 3, which were in the range of 0.01-2.81 ng g^{-1} dw for OCPs and 0.03-3.57 ng g^{-1} dw for PAHs, respectively. The observed concentrations less than the MDLs were defined as not detectable (nd). The spiked recoveries of the sample matrix ranged from 68% to 112% for OCPs and from 76% to 103% for PAHs. The listed data here were not

corrected for the recovery efficiency. In addition, the standard solution was added every 15 samples to recalibrate the retention time of compounds, and a procedure blank was run to assess the cross contamination for each set of the samples.

2.4 Total organic matter determination

For analysis of total organic matter contents in sediments, the subsample of sediments was freeze-dried as abovementioned and then sieved with a 100 mesh stainless-steel sieve. Dried sediments were heated at 550°C for 6 h to determine the organic matter contents gravimetrically, which was expressed as loss of ignition (LOI, %) to indicate the concentrations of total organic matter.

2.5 Statistical analysis

The concentrations and spatial distribution of OCPs and PAHs in sediments were mapped out by ARCGIS 9.3 software (ESRI, USA) using Kriging interpolation analysis. The concentration difference of OCPs and PAHs in various lake areas were calculated as the arithmetic mean values of sampling sites, and the box plots were then completed by Origin Pro 8.5 (Origin Lab, USA). The correlation analysis between compound concentrations and LOI in sediments as well as the composition correlations between the upper-, lower-districts and the lake body area were performed using SPSS 16.0 software for windows (SPSS Inc., USA). The Spearman's rank correlation test was conducted to test the strength of associations between parameters, and the statistical significance was considered when $p < 0.05$ (two-tailed

210 tests).

211 **3 Results and discussion**

212 *3.1 Residual levels of PAHs and OCPs*

213 The residual concentrations of PAHs and OCPs are listed in Table 1 and Table 2,
214 respectively. PAHs ranged from 12.1 to 2281.1 ng g⁻¹ dw with the arithmetic mean
215 value of 348.7±399.4 ng g⁻¹ dw for sediments from THB, and the highest residual
216 level was found out at the Wuchengxiyu Area while the lowest value was observed at
217 the Hangjia Lake Area. As for the samples from THL, PAHs were detected in all
218 sediments with the concentrations ranging from 11.4 to 209.9 ng g⁻¹ dw (63.6±40.9 ng
219 g⁻¹ dw). The highest and the lowest values were found out at the Eastern Coastal Lake
220 and the Center Lake, respectively. Among PAHs, a higher concentration of 3-ringed
221 PAHs (sum of Any, Ana, Flu, Phe, and Ant) was observed at THB than the other
222 congeners, which were detected at the value of 2.7-1230.3 ng g⁻¹ dw (136.2±197.4 ng
223 g⁻¹ dw). As for the higher molecular weight (HMW) PAH congeners, the observation
224 levels of 4-ringed (sum of Flt, Pyr, BaA, and Chr), 5-ringed (sum of BbF, BkF, BaP,
225 and DahA) and 6-ringed congeners (sum of BghiP and InP) were detected in the range
226 of 7.6-713.5 ng g⁻¹ dw (33.1±151.7 ng g⁻¹ dw), 0.03-329.9 ng g⁻¹ dw (39.5±69.0 ng g⁻¹
227 dw) and 0.03-227.1 ng g⁻¹ dw (20.7±39.2 ng g⁻¹ dw), respectively. For THL samples,
228 4-ringed PAHs showed the highest residual levels of 3.1-137.7 ng g⁻¹ dw (24.9±25.1
229 ng g⁻¹ dw) among PAHs, followed by 3-ringed ones (4.0-91.0 ng g⁻¹ dw (25.1±17.0 ng
230 g⁻¹ dw)), whereas the observation values ranged from 0.4 to 20.4 ng g⁻¹ dw (6.4±4.7

ng g⁻¹ dw) for 5-ringed PAHs and from 0.1 to 11.9 ng g⁻¹ dw (4.0±3.1 ng g⁻¹ dw) for 6-ringed PAHs, respectively. As for the individual PAHs, Flt was detected at higher residues than other congeners for THB. DahA was detected at the lowest residual level of nd-8.8 ng g⁻¹ dw (0.6±1.4 ng g⁻¹ dw). Flt was also the primary PAH congener detected in sediments from THL, followed by Ana and Any. The coefficients of variation (CV) for both PAH individuals and total PAHs were high, implying the significant spatial differences among the study area.

Table 1.

OCPs were detected in all sediments with the concentrations ranging from 16.3 to 96.9 ng g⁻¹ dw (38.5±19.2 ng g⁻¹ dw) for THB and from 16.8 to 61.9 ng g⁻¹ dw (27.5±10.5 ng g⁻¹ dw) for THL. The highest concentration of OCPs was detected at the Huxi Area, and the highest OCP residue was observed at the Meiliang Bay of THL. Among OCPs, HCHs (α -, β -, γ -, and δ -HCH included) and DDTs (sum of p,p'-DDD, p,p'-DDE, and p,p'-DDT) were predominant compounds for both THB and THL. HCHs were detected in the range of 3.3-64.5 ng g⁻¹ dw with a mean value of 13.0±11.9 ng g⁻¹ dw for THB and 3.6-27.8 ng g⁻¹ dw with a mean value of 10.1±7.4 ng g⁻¹ dw for THL. As for DDTs, both the highest concentration of 55.9 ng g⁻¹ dw and the lowest concentration of 0.2 ng g⁻¹ dw were observed at the Huxi Area of THB. The Meiliang Bay showed the highest residue of DDTs (16.5 ng g⁻¹ dw) and the Center Lake was found out to be the lowest polluted region by DDTs (1.2 ng g⁻¹ dw). Chlordanes (sum of heptachlor, heptachlor epoxide, α - and γ -chlordane), aldrins (sum of dieldrin, aldrin, endrin, endrin ketone, and endrin aldehyde) and endosulfans (sum

of α -, β -endosulfan, and endosulfan sulfate) in sediments from THB were also observed with large variations in residues ranging from 2.0 to 11.6 ng g⁻¹ dw (3.4±1.6 ng g⁻¹ dw), from 2.6 to 32.3 ng g⁻¹ dw (6.9±5.5 ng g⁻¹ dw), from 0.7 to 8.0 ng g⁻¹ dw (1.5±1.2 ng g⁻¹ dw), respectively. Chlordanes, aldrins and endosulfans were also 100% detected in sediments from THL at the concentrations of 2.0-14.4 ng g⁻¹ dw (2.9±2.6 ng g⁻¹ dw), 1.1-19.5 ng g⁻¹ dw (3.8±3.2 ng g⁻¹ dw) and 0.7-3.2 ng g⁻¹ dw (1.2±0.4 ng g⁻¹ dw), respectively. For both THB and THL samples, δ -HCH and p,p'-DDT were observed at much higher concentrations than other compounds. The relative lower CV values of OCPs in THL than THB indicated the potential water mixing action during the transport of OCPs.

Table 2.

The residual levels of HCHs and DDTs in surface sediments from THB were higher than the Huaihe River upper reaches (Sun et al. 2010), which were detected at the values of 1.95-11.05 ng g⁻¹ dw for HCHs and 4.07-23.89 ng g⁻¹ dw for DDTs, respectively. HCHs were found at similar residual levels as the two tributaries of the Chenab River of Pakistan (1.19-33.3 ng g⁻¹ dw), however, DDTs were observed at much lower concentrations comparing with the Chenab River (5.84-89.8 ng g⁻¹ dw) (Mahmood et al., 2014). When compared with the marine and adjacent riverine sediments of North Bohai Sea (HCHs: nd-1964.97 ng g⁻¹ dw; DDTs: nd-86.46 ng g⁻¹ dw), rather lower concentrations of both HCHs and DDTs were detected (Hu et al., 2010). The same phenomenon could also be easily found for the major tributary of the Ganges River of India (Gomti river) with much higher concentration of total OCPs

being found (0.92-813.59 ng g⁻¹ dw) (Malik et al., 2009) than THB. As for PAHs, they were found at similar concentrations with the sediment samples from the upper reaches of the Huaihe River, China (85.7-935.2 ng g⁻¹ dw) (Feng et al., 2012) and riverine sediments from Thailand (263±174 ng g⁻¹ dw) (Boonyatumanond et al., 2006), and were much higher than the residues in sediments from the urban stretch of the River Tiber, Italy (157.8-271.6 ng g⁻¹ dw) (Patrolecco et al., 2010). In addition, PAHs in Bohai Sea (140.6-300.7 ng g⁻¹ dw) and Daya Bay (42.5-158.2 ng g⁻¹ dw) were much lower than the observed residues in THB (Hu et al., 2010; Yan et al., 2009). As for the most developed area of China, the Pearl River Delta (138-6973 ng g⁻¹ dw) (Luo et al., 2008), PAHs in sediments from THB were much lower comparatively.

3.2 Spatial correlation of PAHs and OCPs

The spatial distribution of PAHs and OCPs can be concluded from the Kriging interpolation plots demonstrated by Fig. 2. For OCPs, the Wuchengxiyu Area showed the highest residue of OCPs (19.2-85.8 ng g⁻¹ dw with a mean value of 45.8±23.1 ng g⁻¹ dw) among THB region, followed by the Yangchengdianmao Area (22.0-80.7 ng g⁻¹ dw (42.8±17.4 ng g⁻¹ dw)) and the West Lake Area (20.6-96.9 ng g⁻¹ dw (41.5±22.6 ng g⁻¹ dw)). The Hangjia Lake Area and the Zhexi Area were observed at similar residual levels, which were in the range of 16.3-73.9 ng g⁻¹ dw (38.0±17.6 ng g⁻¹ dw) and 18.5-79.9 ng g⁻¹ dw (31.1±18.0 ng g⁻¹ dw), respectively. Among THB, the least polluted area by OCPs would be the Pudong Area (19.2-49.4 ng g⁻¹ dw (28.6±10.5 ng g⁻¹ dw)) and the Puxi Area (20.5-38.1 ng g⁻¹ dw (27.2±9.5 ng g⁻¹ dw)).

As for PAHs, the highest residual level was also obtained at the Wuchengxiyu Area (93.9-2281.1 ng g⁻¹ dw (686.4±734.4 ng g⁻¹ dw)), followed by the West Lake Area (37.8-1403.7 ng g⁻¹ dw (393.4±392.4 ng g⁻¹ dw)). The Puxi Area and the Pudong Area were observed at much lower PAHs than other THB areas, which were in the range of 164.6-171.0 ng g⁻¹ dw (168.5±3.4 ng g⁻¹ dw) and 24.5-392.9 ng g⁻¹ dw (184.9±121.6 ng g⁻¹ dw), respectively.

Therefore, the major upper reaches (the Huxi Area and the Wuchengxiyu Area) showed higher residues of PAHs and OCPs than the lower reaches. Furthermore, similar distribution was observed for both PAHs and OCPs, which was consistent with the land uses of THB. It could be easily inferred that the Wuchengxiyu Area contained two big cities of Wuxi and Changzhou, which was mainly comprised by the cultivated land and the built-up land (Zhang et al., 2014). It was reported that the total GDP of this area was the highest among THB with the human density of 1543 people/km² (Gao, 2002). Additionally, the decrease rate of the cultivated land and the increase rate of the built-up land were the highest among different districts, and the annual loss rate of the cultivated land was 3.4%, which thus induced the heavy burden of PAHs and OCPs due to the accompanied anthropogenic activity increase. While for the Huxi Area, although it was predominantly composed of the cultivated land and forest land, the northern part of this area was mainly polluted by the Yixing city. The Zhexi Area was mainly covered by the forest, which was responsible for the lowest residual levels of both PAHs and OCPs. The built-up land accounted for the most part of the land use at the Yangchengdianmao Area, the Hangjia Lake Area, the Puxi Area and the Pudong

Area (Zhang et al., 2014). This land use distribution more or less decided the spatial distribution of PAHs and OCPs and showed that anthropogenic activity would be responsible for the release of such chemicals to THB.

For THL, the Meiliang Bay and the Gonghu Bay were detected to be the most polluted area with the concentrations of OCPs being in the range of 18.0-61.9 ng g⁻¹ dw (32.9±13.5 ng g⁻¹ dw) and 26.0-39.4 ng g⁻¹ dw (32.7±9.5 ng g⁻¹ dw), respectively, which were consistent with the eutrophic situation induced by heavy inputs of nutrients from surrounding upper areas (Qin et al., 2007; Xu et al., 2010), documenting the synchronous input of both nutrients and toxic hydrophobic pollutants to Taihu Lake. OCPs in sediments from the Zhushan Bay, the East Taihu Lake and the Eastern Coastal Lake ranged from 21.4 to 38.8 ng g⁻¹ dw (30.1±12.3 ng g⁻¹ dw), from 22.7 to 45.5 ng g⁻¹ dw (30.5±13.0 ng g⁻¹ dw), from 16.8 to 49.4 ng g⁻¹ dw (26.5±13.2 ng g⁻¹ dw), respectively, while the lowest value of OCPs was detected at the Center Lake (18.4-23.6 ng g⁻¹ dw (21.1±2.2 ng g⁻¹ dw)). While for PAHs, the Zhushan Bay was found out to be the most polluted area in the range of 91.0-158.3 ng g⁻¹ dw (124.7±47.6 ng g⁻¹ dw), followed by the Eastern Coastal Lake (25.7-209.9 ng g⁻¹ dw (72.9±77.9 ng g⁻¹ dw)) and the Meiliang Bay (35.1-121.4 ng g⁻¹ dw (71.2±29.6 ng g⁻¹ dw)). The Center Lake was also the least polluted area by PAHs at the residual level of 11.4-75.3 ng g⁻¹ dw (43.1±21.2 ng g⁻¹ dw).

Fig. 2.

Since sediments with high organic matter are more likely to absorb lipophilic persistent pollutants than those with lower organic matter values (Hung et al., 2007),

the potential correlations between total organic matter (LOI, %) and pollutant concentrations were also analyzed in the present study. LOI contents in surface sediments from THB varied significantly as well as in sediments from THL. LOI ranged from 4.31% to 25.79% with the geometric mean value of $13.04 \pm 4.15\%$ in sediments from THB, and the contents were found in the range of 2.04-6.19% ($3.50 \pm 0.93\%$) for sediments from THL. The much higher concentrations of LOI detected in sediments from THB than THL demonstrated the origin of organic matter in lake body from terrestrial sources, which could also be concluded from the high ratios of C/N (higher than 45) in sediments (Zhao et al., 2009). The predominant contribution of terrestrial inputs of organic matter to the estuary regions (70-80%) would finally induce the higher LOI contents in basin regions than in lake body as implied by the stable isotope analysis (Wu et al., 2006). Significant positive correlation between total OCPs and LOI ($r=0.242$, $p<0.05$) was observed (Fig. 3). Additionally, significant positive correlations between OCPs and LOI were also detected for HCHs ($r=0.294$, $p<0.01$), chlordanes ($r=0.290$, $p<0.01$) and endosulfans ($r=0.357$, $p<0.01$), which demonstrated that LOI played a key role in determining OCP residues in sediments from both THB and THL. While for PAHs, no significant positive or negative correlations between different congeners and LOI contents in sediments were detected due to their higher biodegradation capacity than OCPs (Haritash and Kaushik, 2009), which could also be indicated from the negative correlation between DDTs and LOI because of the biodegradation of p,p'-DDT.

Fig. 3.

Moreover, the spatial correlation analysis of PAH and OCP residues between the upper reaches and the lake body as well as between the lake body and the lower reaches were also performed to elucidate the spatial correlations of such pollutants. As shown in Fig. 4, extremely significant positive correlations of both PAH and OCP compositions were observed between the upper reaches and the lake body (PAHs: $r=0.748-0.920$, $p<0.01$; OCPs: $r=0.736-0.986$, $p<0.01$), documenting a major exogenous input of these pollutants into Taihu Lake through surrounding upper rivers. In addition, extremely significant positive correlations were also detected between the lake body and the lower reaches (PAHs: $r=0.746-0.754$, $p<0.01$; OCPs: $r=0.932-0.956$, $p<0.01$), which thus further indicated the transport of PAHs and OCPs along the water flow direction accounting for the sources of these pollutants accumulated in sediments from the lower reaches of Taihu Lake as suggested by our previous study conducted in lake body (Zhao et al., 2009).

Fig. 4.

3.3 Sources of PAHs and OCPs

In terms of the number of fused rings present in the chemical structure of PAHs, similar compositions with 3- and 4-ringed PAHs being the predominant congeners among PAHs were observed in sediments for both THB and THL. For THB, 3- and 4-ringed PAHs accounted for 6.1-87.2% ($38.1\pm16.9\%$) and 7.5-85.1% ($39.6\pm16.7\%$) of total PAHs, respectively. Two-ringed PAH (Nap) contributed 0.2-51.3% ($7.5\pm7.6\%$) to PAHs observed in sediments from THB, and then the percentage contributions of

HMW PAHs such as 5- and 6-ringed compounds ranged from 0.07 to 44.7% (9.8±9.1%) and from 0.04 to 22.2% (5.1±4.6%), respectively. As for THL, 3- and 4-ringed PAHs were also found to be the major contributors, accounting for 14.0-57.5% (40.2±12.1%) and 9.1-65.6% (36.2±12.8%) of PAHs, respectively. It was observed that 2-, 5- and 6-ringed hydrocarbons accounted for 0.7-25.4% (5.5±5.4%), 1.3-43.2% (11.4±9.2%) and 0.2-21.7% (6.8±4.5%) of PAHs in THL sediments.

Among OCPs, HCHs were the dominant compounds, accounting for 5.1-80.7% (31.8±15.7%) for THB and 15.1-59.6% (33.6±12.1%) for THL, respectively. DDTs were also found out to be the primary pesticides in sediments from both areas, which contributed 0.5-65.9% (21.7±12.4%) (THB) and 5.1-33.5% (21.9±7.2%) (THL) to total OCPs. Aldrins, chlordanes and endosulfans were also detected in all sediments from THB with the contribution ranging from 5.4 to 56.2% with a mean value of 17.8±7.7%, from 3.1 to 17.8% (9.8±3.8%) and from 1.2 to 15.1% (4.0±2.0%), respectively. While for THL, aldrins, chlordanes and endosulfans accounted for 3.9-31.5% (13.3 ±5.6%), 4.3-57.1% (11.6±10.7%) and 2.0-13.4% (4.8±2.1%) of observed OCPs. Therefore, similar compositions of OCP individuals demonstrated the synchronous pollution sources for THB and THL, which could also be implied from the significant positive correlations of individual pollutants in sediments between surrounding rivers and the lake body as documented by aforementioned analysis.

Isomeric and parent substance/metabolite ratios have been widely used to identify past and recent sources of pollutants in soils and sediments (Manz et al., 2001). Composition differences of HCH isomers, DDT congeners and other individual

compounds in the environment could indicate different contamination sources of OCPs (Liu et al., 2009; Lin et al., 2012). In the present study (Fig. 5), a higher contribution of β -HCH to HCHs for both THB (2.3-60.2% ($19.2\pm9.9\%$)) and THL (6.0-56.9% ($22.4\pm10.8\%$)) than other HCH isomers demonstrated the aged sources of HCHs due to the resistance to microbial degradation of β -HCH. Additionally, α - and γ -HCH can be transformed into β -HCH in the environment, which may finally lead to an obvious accumulation of β -HCH than other isomers (Li et al., 2006). The ratio of α -/ γ -HCH is relatively stable with a value of 4.64-5.83 for the technical HCH mixtures and nearly zero for lindane, which might increase during their degradation process in the environment. Therefore, the rather low ratios of α -/ γ -HCH ranging from 0.23 to 5.75 (0.96 ± 0.71) for THB and from 0.23 to 1.20 (0.89 ± 0.31) for THL both indicated the predominant use of lindane around this area. As for DDTs, 60.5% of samples from THB showed the ratios of p,p'-DDD/p,p'-DDE were less than 1.0, documenting the aerobic conditions of p,p'-DDT in surface sediments because of the oxidation status in surface sediments induced by strong wind-wave disturbance occurred for both the rivers and the lake body (Zhu et al., 2007; Qin et al., 2007). P,p'-DDT was the primary compound among DDTs, which could be concluded from the results of 95.1% of sediments from THB and 100% from THL showing the ratios of p,p'-DDT/DDTs larger than 0.5. Therefore, new inputs of parent DDT compound could be implied, which might be attributed to the illegal inputs of DDT or usage of dicofol containing DDT. An investigation showed that the input of DDT from dicofol in China between 1988 and 2002 was 8770 tons and there are more than sixty

registered manufacturers in China producing dicofol at present (Qiu et al., 2005), which are then responsible for the fresh sources of DDT in environment. Technical chlordane is generally used as insecticide, herbicide and termiticide, and was reported to be still used in China against termites (Hu et al., 2007). It was found out that only 14.8% of samples from THB and 35.4% from THL showed the ratios of γ -/ α -chlordane >1.0, documenting fresh inputs of chlordanes. As for endosulfans, the ratios of α -/ β -endosulfan were all less than 2.33 and then implied lack of endosulfan inputs surrounding Taihu Lake.

Fig. 5.

In general, pyrogenic and petrogenic sources are two major origins of anthropogenic PAHs in the environment (Heywood et al., 2006). Pyrogenic PAHs are formed as trace contaminants by the incomplete combustion of organic materials, such as wood, fossil fuels, asphalt, and industrial waste. Crude and refined petroleum contain petrogenic PAHs, and are also important sources of PAHs. Diagnostic ratios of PAHs have been considered to be good indicators of routes for transport to environmental media and the sources of pollution, since several markers are widely used to apportion the origin of PAHs presented in different environmental media (Yunker et al., 2002; Bucheli et al., 2004). The ratio of Ant/(Ant+Phe) is one of the most frequently used in distinguishing between combustion and petroleum sources. The ratio <0.1 is taken as an indication of petroleum while a ratio >0.1 indicates a dominance of combustion. In addition, in order to assess sources of PAHs more accurately, the ratio of Flt/(Flt+Pyr) is used as Flt and Pyr isomer pair degrade

449 photolytically at comparable rates (Yunker et al., 2002; Bortey-Sam et al., 2014).
450 Yunker et al. (2002) suggested that a Flt/(Flt+Pyr) ratio <0.4 implies petroleum,
451 $0.4-0.5$ implies petroleum (liquid fossil fuel, vehicle and crude oil) combustion,
452 and >0.5 implies combustion of coal, grass and wood. Moreover, BaP/BghiP and
453 InP/(InP+BghiP) may also characterize the nature of potential PAH emission sources
454 that BaP/BghiP <0.6 and InP/(InP+BghiP) <0.2 are indications of petroleum and
455 petrogenic sources. When BaP/BghiP was between 0.6 and 0.9 and InP/(InP+BghiP)
456 was between 0.2 and 0.5 , the PAHs usually come from mobile sources associated with
457 vehicles. When BaP/BghiP >0.9 and InP/(InP+BghiP) >0.5 , it strongly implies the
458 contribution of coal combustion (Bucheli et al., 2004; Liu et al., 2009). In the present
459 study (Fig. 6), 91.3% of sediment samples from THB were found to have the
460 Flt/(Flt+Pyr) ratios >0.5 , indicating a predominant source of petrogenic processes,
461 while fossil fuel combustion, grass, wood and coal combustion only accounted for 6.2%
462 and 2.5% of the PAH emissions, respectively. Furthermore, a larger proportion of
463 samples (88.9%) showed ratios of Ant/(Ant+Phe) >0.1 , documenting a predominant
464 contribution of combustion. In addition, according to the ratio of InP/(InP+BghiP),
465 28.4% of samples were below 0.2 , and 27.2% were in the range of $0.2-0.5$, and then
466 the rest (44.4%) were found to have the ratios >0.5 , which implied a combination of
467 pyrogenic and petrogenic sources. An equal distribution of petrogenic sources, fossil
468 fuel combustion, biomass and coal combustion could also be implied from the ratio of
469 BaP/BghiP with 59.3% of samples being found to have the ratio <0.6 and 28.4% of
470 samples showing the ratio >0.9 . Similar PAH sources for both THB and THL could be

concluded from the similar compositions, which seemed to be mixed sources of pyrogenic and petrogenic ones dominated by grass, wood and coal combustion-related.

Fig. 6.

3.4 Ecological risk assessment of PAHs and OCPs

For evaluating the possibility of occurrence of adverse ecological effects from the exposure to chlorinated organic compounds in sediments, a Screening Level Ecological Risk Assessment (SLERA) was performed according to the framework of the USEPA (1992, 1997, 1998). The protection of benthic species was selected as the assessment endpoint. For this purpose, the risk was estimated by comparing the maximum concentration of each compound with data of Sediment Quality Guidelines (SQGs, toxicity reference values) in form of hazard quotient (HQ) according to the following equation as listed by Khairy et al. (2009, 2012):

$$HQ = \frac{\text{maximum concentration of the pollutant in sediments } (\mu\text{g/kg dw})}{\text{sediment quality guideline of the pollutant } (\mu\text{g/kg dw})}$$

The consensus-based SQGs were used for calculation the geometric mean of the different reported SQGs according to MacDonald et al. (2000). Furthermore, the consensus-based SQGs were divided into two groups including the threshold effect concentrations (TEC) below which harmful effects are rarely observed and the probable effect concentrations (PEC) above which harmful effects are frequently observed, which thus two kinds of HQ can then be obtained accordingly by the following equations:

$$\text{TEC HQ} = \frac{\text{maximum concentration of the pollutant in sediments } (\mu\text{g/kg dw})}{\text{TEC } (\mu\text{g/kg dw})}$$

$$\text{PEC HQ} = \frac{\text{maximum concentration of the pollutant in sediments } (\mu\text{g/kg dw})}{\text{PEC } (\mu\text{g/kg dw})}$$

According to these equations, adverse ecological effects are only considered when TEC HQ>1, while effects on sediments dwelling organisms are likely to be frequently observed when PEC HQ>1.

The TEC HQ and PEC HQ values for individual PAH congeners are listed in Fig. 7 (A) and OCP risks are shown in Fig. 7 (B). According to TEC HQ, the West Lake Area showed higher ecological risks than other regions with Ant and BaA being the responsible ones. The highest TEC HQ of Ant was observed at the West Lake Area (6.37) followed by the Wuchengxiyu Area (4.50), the Hangjia Lake Area (2.45), and the Yangchengdianmao Area (1.55), while for other districts of THB, the values were all less than 1. BaA was also detected to be the predominant toxicant with the highest TEC HQ of 3.4 being found at the Hangjia Lake Area. The West Lake Area, the Wuchengxiyu Area, the Yangchengdianmao Area and the Zhexi Area were also detected with TEC HQ>1, thus documenting potential adverse ecological effects induced by BaA. As for the total PAHs, only the Wuchengxiyu Area showed higher values of 1.42 than other regions (0.11-0.87). In addition, the TEC-HQ values for both individual congeners and PAHs in THL were all less than 1, which indicated lower ecological risks than THB. As for PEC HQ of PAHs, all the values were less than 1, further demonstrating the adverse ecological effects are unlikely to occur by these substances.

Among OCP individuals, γ -HCH, dieldrin and p,p'-DDT showed higher TEC HQ

(>1) than p,p'-DDE, endrin and p,p'-DDD. The highest value of TEC HQ for γ -HCH was found at the Hangjia Lake Area (2.30), followed by the Yangchengdianmao Area (1.96), the West Lake Area (1.85), the Wuchengxiyu Area (1.28) and the Zhexi Area (1.08), while for the Puxi Area and the Pudong Area, TEC HQ was only detected at the value of 0.70 and 0.97, respectively. As for THL, much lower TEC HQ values were observed comparing with THB. The highest TEC HQ was found at the Meiliang Bay (2.09), and then the values ranged from 0.37 to 0.78 for other lake areas of Taihu Lake with the lowest ecological risk being found at the Center Lake. For heptachlor, the possible ecological risk might be only detected at the Wuchengxiyu Area (1.39) and the Hangjia Lake Area (1.33) of THB, and the Center Lake (2.12) and the Western Coastal Area (2.55) of THL. When it comes to chlordanes, obvious adverse ecological effects could be found at sampling sites distributed around THB except for the Pudong Area. The higher values of TEC HQ than 1 for chlordanes were also detected at the Meiliang Bay (1.34), the Western Coastal Lake (4.46) and the Center Lake (3.16). P,p'-DDT was the primary factor inducing ecological risk for both THB and THL. The TEC HQ for p,p'-DDT were in the range of 1.28-13.33 except for the Puxi Area, and the West Lake Area would be easily to induce adverse ecological effects by p,p'-DDT at present. For THL, TEC HQ values were all higher than 1, and the highest TEC HQ for p,p'-DDT was detected at the Eastern Coastal Lake (3.91), followed by the Meiliang Bay (1.76). Similarly, THL showed lower ecological risks by OCPs than THB due to the lower TEC HQ as demonstrated by PAHs. However, the PEC HQ values were all less than 1, implying the adverse ecological effects were not

frequently detected around this area.

Therefore, the residues of PAHs and OCPs in sediments would pose adverse effects to benthic organisms mainly induced by Ant, BaA, γ -HCH, dieldrin, p,p'-DDT and chlordanes, which were not frequently observed. Furthermore, THB showed higher ecological risks than THL and should be paid much more attention considering the continuous inputs of pollutants to the lake body from basins.

Fig. 7.

4 Conclusions

Both PAHs and OCPs were ubiquitous organic pollutants around Taihu Lake. The residues of PAHs were higher than OCPs due to the application status, and then higher concentrations of both pollutants were detected in sediment samples from THB than THL. The well correspondence of spatial distribution with the land use of THB implied the predominant contribution of anthropogenic activities around for PAH and OCP residues. The major upper reaches were more polluted than the lower reaches, moreover, significant spatial correlations among the upper reaches, the lake body and the lower reaches were observed, which thus indicated the high synchronous pollution of PAHs and OCPs due to the water flow direction and the demand of future monitoring on the potential interaction between rivers and lakes comprehensively. In addition, the spatial distribution of OCPs were mainly manipulated by the total organic matter contents other than PAHs attributed to the physicochemical properties such as the biodegradation capacity of individual chemicals. Similar compositions of

PAHs with 3- and 4-ringed congeners being the predominant ones were observed for both THB and THL, which showed the combined sources of petrogenic and pyrogenic ones dominated by grass, wood and coal combustion. HCHs and DDTs were found to be the major pesticides of OCPs. HCHs mainly originated from old usage, while DDTs showed fresh inputs around Taihu Lake due to the dicofol and antifouling paint usage. The ecological risk assessment based on the TEC HQ demonstrated potential adverse effects were observed due to the pollution of Ant, BaA, γ -HCH, dieldrin, p,p'-DDT and chlordanes. Although such adverse effects to benthic organisms not frequently occurred according to the low PEC HQ values, further monitoring and risk assessment should be conducted due to the continuous inputs of these pollutants.

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732

Table 1 The concentrations of PAH individuals and total PAHs in surface sediments from THB and THL.

Chemicals	THB				THL			
	Range	Mean ^a	SD ^b	CV ^c	Range	Mean	SD	CV
	(ng g ⁻¹ dw)				(ng g ⁻¹ dw)			
Naphthalene	0.7-110.2	19.2	21.9	1.1	0.4-14.3	3.3	3.6	1.1
Acnaphthylene	1.6-690.7	42.2	101.2	2.4	0.7-21.0	6.7	4.0	0.6
Acenaphthene	0.09-233.4	48.4	50.4	1.0	nd-23.9	7.7	6.1	0.8
Fluorene	nd ^d -43.7	10.6	11.2	1.1	0.01-1.8	0.5	0.5	1.0
Phenanthrene	0.2-77.3	7.2	12.9	1.8	0.3-16.7	4.8	4.4	0.9
Anthracene	0.06-364.6	30.8	55.7	1.8	0.5-37.3	5.5	7.0	1.3
Fluoranthene	1.1-627.2	86.0	113.8	1.3	0.3-104.9	17.2	19.6	1.1
Pyrene	0.07-335.8	15.4	39.6	2.6	0.2-28.7	5.3	5.5	1.0
Benzo[a]anthracene	0.2-372.2	28.4	51.5	1.8	0.2-7.4	2.1	1.7	0.8
Chrysene	nd-52.7	4.7	7.6	1.6	0.03-1.5	0.5	0.5	1.0
Benzo[b]fluoranthene	0.08-199.3	20.0	31.2	1.6	0.4-13.2	4.3	3.6	0.8
Benzo[k]fluoranthene	0.02-213.9	12.9	29.1	2.3	0.1-6.3	1.6	1.3	0.8
Benzo[a]pyrene	nd-272.5	7.8	34.3	4.4	0.02-4.9	0.5	0.9	1.9
Dibenzo[a,h]anthracene	nd-8.8	0.6	1.4	2.6	0.02-3.1	0.6	1.0	1.5
Benzo[g,h,i]perylene	0.02-181.2	6.9	20.5	3.0	0.02-11.1	2.3	2.5	1.1
Indeno[1,2,3-c,d]pyrene	0.8-225.4	17.7	37.7	2.1	0.8-8.5	2.3	1.8	0.8
PAHs ^e	12.1-2281.1	348.7	399.4	1.1	11.4-209.9	63.6	40.9	0.6

^a: the arithmetic mean values of specific PAH congeners among different sites located in the THB area.

^b: the standard deviation of PAH concentrations among different sites located in the THB area.

^c: the coefficient of variation (CV) of specific PAH congeners among different sites located in the THB area.

^d: the concentrations of specific PAH congeners below the method detection limits were defined as not detectable (nd).

^e: the sum of 16 PAH compounds (naphthalene (NaP), acenaphthylene (Any), acenaphthene (Ana), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenz[ah]anthracene (DahA), benzo[ghi]perylene (BghiP), and indeno[1,2,3-cd]pyrene (InP) included).

Table 2 The concentrations of OCP individuals and total OCPs in surface sediments from THB and THL.

Pesticides	THB				THL			
	Range	Mean ^a	SD ^b	CV ^c	Range	Mean	SD	CV
	(ng g ⁻¹ dw)				(ng g ⁻¹ dw)			
α -HCH	0.9-11.0	1.3	1.2	1.0	0.1-1.1	0.9	0.2	0.2
β -HCH	1.0-10.2	1.9	1.5	0.8	1.0-9.1	1.9	1.5	0.8
γ -HCH	0.8-5.4	1.5	0.9	0.6	0.2-4.9	1.2	0.8	0.7
δ -HCH	0.9-59.4	8.4	10.7	1.3	1.1-20.5	6.1	5.8	0.9
Heptachlor	1.5-7.2	2.4	0.8	0.3	1.5-6.4	1.9	1.0	0.5
Aldrin	0.5-24.4	2.5	3.8	1.5	0.4-7.7	1.2	1.7	1.4
Heptachlor epoxide	0.3-3.4	1.2	1.1	0.9	5.2-6.3	5.8	0.8	0.1
γ -Chlordane	0.3-3.2	0.4	0.4	0.9	0.3-1.7	0.4	0.3	0.8
α -Chlordane	0.2-3.7	0.3	0.5	1.6	0.03-0.4	0.3	0.1	0.4
Endosulfan I	0.2-2.2	0.3	0.3	1.2	nd-0.6	0.2	0.1	0.5
p,p'-DDE	0.2-11.4	0.8	1.5	1.9	0.04-0.4	0.2	0.1	0.3
Dieldrin	1.0-6.4	2.0	1.1	0.6	nd-2.5	1.2	0.5	0.4
Endrin	0.5-15.5	1.1	2.0	1.8	0.1-0.5	0.5	0.1	0.2
Endosulfan II	0.4-3.4	0.6	0.4	0.6	0.4-0.5	0.4	0.0	0.1
p,p'-DDD	nd ^d -5.0	0.8	1.1	1.4	0.01-2.0	0.5	0.7	1.5
Endrin aldehyde	0.2-7.3	0.6	0.9	1.6	0.03-0.3	0.2	0.1	0.2
Endosulfan sulfate	0.2-4.3	0.6	0.8	1.2	0.08-2.7	0.6	0.4	0.7
p,p'-DDT	3.3-55.4	7.2	7.5	1.0	0.1-16.3	5.4	2.6	0.5
Endrin ketone	0.6-8.4	0.8	0.9	1.0	0.08-9.1	1.2	2.1	1.7
Methoxychlor	2.7-21.2	5.2	3.6	0.7	0.3-5.1	3.7	0.8	0.2
OCPs ^e	16.3-96.9	38.5	19.2	0.5	16.8-61.9	27.5	10.5	0.4

^a: the arithmetic mean values of specific pesticides among different sites located in the THB area.

^b: the standard deviation of OCP concentrations among different sites located in the THB area.

^c: the coefficient of variation (CV) of specific pesticides among different sites located in the THB area.

^d: the concentrations of specific pesticides below the method detection limits were defined as not detectable (nd).

^e: the sum of 20 OCP compounds (α -, β -, γ -, δ -HCH, heptachlor, heptachlor epoxide, α -chlordane, γ -chlordane, aldrin, endrin, dieldrin, endosulfan I, endosulfan II, endrin aldehyde, endosulfan sulfate, endrin ketone, p,p'-DDE, p,p'-DDD, p,p'-DDT, and methoxychlor included).

Figure captions

Fig. 1. The study area and sampling locations from Taihu Lake basin (THB) and Taihu Lake body (THL).

Fig. 2. The Kriging interpolation plots of concentrations and spatial distributions of PAHs (A) and OCPs (B) in sediments from THB and THL.

Fig. 3. Correlation analysis between OCP compounds and LOI contents in surface sediments from THB and THL.

Fig. 4. Spatial correlation analysis of PAH and OCP compositions between upper reaches (the West Lake Area, the Wuchengxiyu Area, and the Zhexi Area) and lake body (A, B) as well as between lake body and lower reaches (the Yangchengdianmao Area and the Hangjia Lake Area) (C, D).

Fig. 5. Isomeric and parent substance/metabolite ratios of OCPs in surface sediments from THB and THL.

Fig. 6. Diagnostic ratios of PAHs in surface sediments from THB and THL.

Fig. 7. The TEC HQ and PEC HQ values of PAHs (A) and OCPs (B) in sediments from THB and THL.

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