



Distribution and bioaccumulation of organochlorine pesticides in surface sediments and benthic organisms from Taihu Lake, China

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ABSTRACT

The spatial distribution and bioaccumulation of organochlorine pesticides (OCPs) in surface sediments and benthic organisms from Taihu Lake were studied. OCPs were detected in all sediment samples with total concentrations ranging from 4.22 to 461 ng g⁻¹ dry weight (dw). The ratios of certain metabolites to their parent compounds indicated there are still new inputs of parent DDT (dichlorodiphenyltrichloroethane) to Taihu Lake, while the highest residues of HCHs (hexachlorocyclohexanes) mainly came from earlier usage and fresh γ -HCH (lindane). No positive correlation was found between the distribution of OCPs and organic matter contents in sediments. Concentrations of OCPs and lipids in typical large benthic organisms, *Bellamyia aeruginosa* (*B. aeruginosa*) and *Corbicula fluminea* (*C. fluminea*), increased with body weight. HCHs, DDTs, chlordanes and heptachlors were the dominant compounds detected in organisms and *C. fluminea* accumulated much more OCP than *B. aeruginosa*. Higher values of biota-sediment accumulation factor (BSAF) were detected in *C. fluminea*, which was both affected by biological characteristics of the organisms and physicochemical properties of the compounds.

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1. Introduction

Organochlorine pesticides (OCPs) were the first artificial pesticides, and most are among the substances restricted or banned globally under the Stockholm Convention on Persistent Organic Pollutants (UNEP/UNDP, 2001) due to their high lipophilicity, persistence, long-range transport and biological effects in the environment. In aquatic systems, OCPs are easily absorbed on particulate organic matter due to their high octanol–water partition coefficient (K_{ow}) and accumulate in sediment, which makes the sediment a potential contamination source. Moreover, these pollutants can be bioaccumulated in sediment-dwelling organisms and then transferred to higher trophic levels, which may threaten the aquatic ecosystem and food safety. Despite a ban and restriction on usage of some OCPs in developed countries during the 1970s and 1980s, some developing countries are still using them for agricultural and public purposes because of their low cost and versatility in controlling insects (Iwata et al., 1994; Tanabe et al., 1994; Monirith et al., 2003). Most studies documented OCP levels in waters, sediments and mussels from the marine environment, and little information is available on OCP contamination in freshwater samples (Nakata et al., 2005).

China is one of the largest producers and consumers of pesticides in the world, and technical HCHs and DDTs were the most widely used pesticides in China during the 1950s. Total production of HCHs and DDTs were 4.9 and 0.4 million tons, which accounted for 33% and 20% of total world production, respectively. Although they have been prohibited in China since 1983, there are still residues in various environmental matrices (Liu et al., 2001; Mai et al., 2001; Yang et al., 2005). Taihu Lake, the third largest freshwater lake in China, is situated in the lower reaches of the Yangtze River with an area of 2338 km² and a mean depth of 1.9 m. With the development of industry, agriculture and increasing population, it has suffered from overloading inputs of nitrogen and phosphorus, and most scientists have focused on nutrients cycling and eutrophication. Fewer studies have focused on the distribution and bioaccumulation of OCPs in sediments and benthic organisms living in sediments in Taihu Lake (Yuan et al., 2003; Qiao et al., 2004). The sediment component of aquatic ecosystems can deposit pesticides and many researchers have pointed out that physicochemical characteristics of the sediments, such as the organic matter and grain size, might play an important role in the fate of OCPs in aquatic sediments (Sarkar and Sen Gupta, 1991; Miglioranza et al., 2003; Bakan and Ariman, 2004). However, there are still no consistent results concluded about the relationship between OCPs and physicochemical characteristics of sediments.

Benthic organisms are good biomarkers for pollution because of their long life-history and relative stable residence, and thus they can provide more precise prediction of the bioaccumulation of

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organic pollutants. Bioaccumulation of persistent contaminants may be controlled by a myriad of physiological and environmental factors. Lipids are often correlated with concentrations of lipophilic contaminants, such as organochlorines, but the assumption of a relationship between the lipid and organochlorine contents has been challenged (Jackson et al., 2001; Davis et al., 2002). Understanding the mechanisms of bioaccumulation in aquatic organisms is still critical to predicting potential toxicity to human health.

The objective of this work is to characterize OCP residues in surface sediments in Taihu Lake. By statistical analysis, the potential relationship between OCPs and physicochemical parameters in sediments was evaluated. *Bellamyia aeruginosa* as a kind of gasteropoda and *Corbicula fluminea* as a kind of mollusc, because of their abundance, ubiquity, and long life span, were collected from Taihu Lake to study on the bioaccumulation of OCPs in sediment-dwelling organisms and potential affecting factors, which could provide information on the status of bioaccumulation in higher trophic organisms.

2. Materials and methods

2.1. Chemicals and reagents

A mixture of standard solution containing α , β , γ , δ -HCH, heptachlor, heptachlor epoxide, α -chlordane, γ -chlordane, aldrin, endrin, dieldrin, endosulfan I, endosulfan II, endrin aldehyde, endosulfan sulphate, endrin ketone, p,p'-DDE, p,p'-DDD, p,p'-DDT, and methoxychlor was obtained from SupelCo (Bellefonte, Pennsylvania, USA). The internal standards 2,4,5,6-tetrachloro-*m*-xylene (TCMX) and decachlorobiphenyl (DCB) were also obtained from SupelCo. All solvents (dichloromethane, acetone, *n*-hexane and methyl alcohol) used for sample processing and analyses were analytical grade and purchased from Sinopharm Chemical Reagent Co. Ltd. (Shanghai, China) and redistilled by glass apparatus before use. The silica gel (80–100 mesh) and alumina (120–200 mesh) were 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). Through the analysis, deionized water was used (Millipore Milli-Q system, Massachusetts, USA).

2.2. Study area and sampling

Details of sampling sites can be identified in Fig. 1. 20 sites throughout the lake were sampled to analyze the contents of OCPs in the surface sediments. Throughout the survey, a global positioning system (GPS) was used to locate the sampling sites. All surface sediment samples (5 cm) were collected with a Peterson grab sampler and homogenized on site, then immediately stored at 4 °C in polyethylene bags. *B. aeruginosa* and *C. fluminea* were collected using a Peterson grab sampler at site s11, situated in East Taihu Lake, and then stored as described for sediment samples. All samples, sediments and *B. aeruginosa* and *C. fluminea*, were collected in May, 2006 and kept at 4 °C during transportation to the lab. The organisms were washed with distilled water and classified into five groups according to weight (Table 1). All samples were stored in a deep-freezer at –20 °C until extraction.

2.3. Sample preparation and chemical analysis

2.3.1. Surface sediments

Physical–chemical analysis: Sediment water content was determined by oven drying of about 20 g wet sediment for 12 h (or to constant weight) at 105 °C. Dried sediments were heated at

550 °C for about 6 h to determine organic matter, which was expressed as loss of ignition (LOI, %). Silt and clay analyses of sediment were analyzed with a laser particle size analyzer (Malvern Mastersizer 2000, Malvern Town, England). The oxidation–reduction potential (ORP or E_h) of sediment samples was detected by E_h sensor (Thermo ORIN, Massachusetts, USA) *in situ*.

OCPs analysis: Surface sediments were freeze-dried and ground in mortar to above 100 mesh. 25 g of homogenized sediments, mixed with 10 g anhydrous sodium sulphate (baked at 550 °C for 6 h) were extracted in a Soxhlet apparatus for 48 h with 250 mL dichloromethane. Copper was added into the samples to remove sulphur. The solution was first dried by anhydrous sodium sulphate and then evaporated nearly to dryness by a rotary evaporator (Buchi R-200, Flawil, Switzerland) and redissolved in 10 mL *n*-hexane to remove dichloromethane. The hexane extracts continued to be concentrated to approximately 1 mL and were further cleaned up and fractioned by a silica gel-alumina (2:1) power column with 15 mL *n*-hexane, 70 mL *n*-hexane/dichloromethane (7:3, V/V) mixture and 30 mL methyl alcohol respectively. The *n*-hexane/dichloromethane elution was collected and concentrated to 1 mL under a gentle nitrogen stream. The quantitative internal standards (TCMX and DCB) were mixed with each sample prior to instrument analysis.

2.3.2. Benthic organisms

B. aeruginosa and *C. fluminea* specimens were defrosted, and the flesh of each size grade were extracted and freeze-dried for OCP analysis. 5 g of the homogeneous samples were extracted in the same way as the surface sediments with a Soxhlet apparatus. Lipid content was determined gravimetrically from an aliquot of the extract according to Kidd et al. (2001), while the others were cleaned by concentrated sulphuric acid (Sakellarides et al., 2006) for a first lipid purification until the organic layer remained colorless and further cleaned with a silica gel-alumina (2:1) power column as the sediment samples.

2.3.3. Identification and quantification of OCPs

The purified sample extracts, both sediments and organisms, were quantitatively analyzed using a HP-6890 gas chromatograph (Agilent, Palo Alto, California, USA), equipped with a ^{63}Ni electron capture detector (ECD) and a DB-5MS fused silica capillary column (60 m \times 0.32 mm \times 0.25 μm , J&W Scientific, Folsom, California, USA). Helium was the carrier gas (1.5 mL/min), and nitrogen was the make-up gas (60 mL/min). The pressure was set at 13.4 Psi. The temperatures of the injector and detector were kept at 250 °C and 320 °C, respectively. The oven temperature was programmed from 60 to 170 °C (2 min hold) at the rate of 25 °C/min to 250 °C (4 min hold) at the rate of 3 °C/min to 280 °C (1 min hold) at the rate of 4 °C/min and finally to 300 °C at the rate of 20 °C/min. The injection volume was 1.0 μL splitlessly. Identification of 20 OCPs was confirmed using gas chromatography–mass spectrometry (GC–MS, Thermo Finnigan DSQII, Massachusetts, USA) equipped with a HP-5 capillary column (30 m \times 0.32 mm \times 0.25 μm).

During analysis, blank tests and recovery of OCPs was performed. 20 g of quartz, which had been baked at 450 °C for 6 h, were mixed with 80 ng standards of OCPs and then extracted in a Soxhlet extractor. The recoveries of 20 OCPs ranged from 63% to 115%, and the concentrations were not corrected for recovery efficiency. The method detection limits (MDLs) (Doong et al., 2002) were determined as the concentration of analytes in a sample giving a peak with a single-to-noise (S/N) of 3 and ranged from 0.01 to 1.55 ng g^{–1}. All results were expressed on a dry weight basis, and those samples with concentrations detected less than MDLs were treated as not detected (ND).

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