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Aquatic biota as potential biological indicators of the contamination, bioaccumulation and health risks caused by organochlorine pesticides in a large, shallow Chinese lake (Lake Chaohu)

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ABSTRACT

Aquatic biota have long been recognized as bioindicators of the contamination caused by hydrophobic organic contaminants (HOCs) in aquatic environments. The primary purpose of the present study is to identify which species of aquatic biota are the most sensitive to organochlorine pesticides (OCPs) in Lake Chaohu and can therefore serve as indicators of the lake's health and assist in the assessment of OCPs risks to human health. OCP levels in eight species of aquatic biota were measured using GC–MS, and the relationships between the biota and OCP levels in the water and suspended solids were studied. DDTs pose potential human health risks and were the predominant OCP components found in the aquatic biota. DDT had the highest mean bioaccumulation factor (BAF) and biota suspended solids accumulation factor (BSSAF) of all of the studied OCP components. The food web magnification factors (FWMF) for *p*, *p*′-DDT were greater than 1, implying that biomagnification occurred. This finding indicates that DDTs still pose a serious threat to the ecosystem and human health in Lake Chaohu, even though the agricultural application of DDT powder has been officially banned since 1983. There were significant positive relationships between OCPs levels in *Culter erythropterus* and those in both water and suspended solids, as well as between OCPs levels in *Protosalanx hyalocranius* and those in suspended solids. This finding suggests that *C. erythropterus* and *P. hyalocranius* are the most sensitive aquatic biota to OCPs and may serve as the most effective bioindicators for monitoring OCP contamination in the water and suspended solids of Lake Chaohu. *Megalobrama amblycephala*, which contained the highest wet weight mean OCP concentration, is the most sensitive OCP indicator and can be used to assess the human carcinogenic risk of OCPs in Lake Chaohu.

1. Introduction

Aquatic biota are frequently used as biological indicators to monitor the levels of Persistent Organic Pollutants (POPs) in aquatic environments (van der Oost et al., 2003; Lanfranchi et al., 2006; Guo et al., 2008; Zhao et al., 2013; Lacorte et al., 2006; Jaspersa et al., 2013), to analyze POP bioaccumulation (Haruhiko et al., 2003; Arnot and Gobas, 2006; van Leeuwen et al., 2008; Coat et al., 2011) and biomagnification in the food chain (Hu et al., 2010; Zhang et al., 2013; Villa et al., 2011; Li et al., 2008b), and to evaluate human health risks associated with the consumption of contaminated water products (Dong et al., 2006; Cheung et al., 2007; Dennis, 2007; Yu et al., 2011; Zhou et al., 2008; Li et al., 2008a). Organochlorine pesticides (OCPs) are a category of POPs that have aroused widespread concern due to their high carcinogenicity and their persistence, semi-volatility, effects on wildlife and ability to bioaccumulate (Willett et al., 1998; Wu et al., 1999; Zhou et al., 2001). The production and use of sixteen types of OCPs, including dichloro-diphenyl-trichloroethane (DDT), chlordane, mirex, aldrin, dieldrin, endrin, hexachlorobenzene, heptachlor, toxaphene, α-hexachlorocyclohexane (α-HCH), β-HCH, lindane (γ-HCH), chlordecone (kepone), pentachlorobenzene and endosulfan, have been prohibited by the Stockholm Convention on Persistent Organic Pollutants (UNEP, 2001, 2011). In China, the agricultural application of DDT and HCH has been banned since 1983, while the production, transport, use, and import/export of DDT, chlordane, mirex, and hexachlorobenzene (HCB) have been forbidden since May 2009 (MEP, 2009). However, some industrial

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products, such as lindane (which is 99% gamma-HCH) and dicofol (which contains DDT analogs that form as manufacturing impurities), are still used in some regions of China (Qiu et al., 2005), and DDT is also used as a secondary material in antifouling paint for ships (Yu et al., 2011). As a result, OCPs have continued to be detected in water and aquatic biota throughout China in recent years (Li et al., 2007; Guo et al., 2008; Tao et al., 2005, 2008; He et al., 2012; Xu et al., 2013; Ouyang et al., 2012, 2013, 2014). While extensive studies on the occurrence of OCPs in water and aquatic biota have been performed (Hoekstra et al., 2003; Skarphedinsdottir et al., 2010; Wang et al., 2014; Zhou et al., 2008; Goutner et al., 2012; Liu et al., 2012; He et al., 2014), little research into the bioaccumulation and biomagnification of OCPs in relation to suspended solids exists; and little is known about the influence of particulate OCPs other than dissolved OCPs on aquatic biota in freshwater ecosystems (Hendriks et al., 1998; Burkhard and Lukasewycz, 2000; He et al., 2014). Yet, suspended solids may be an important route for the bioaccumulation of OCPs. The biological composition and environmental conditions of aquatic systems may differ from one another, which may result in variations in biotic sensitivity to OCPs from ecosystem to ecosystem. Therefore, it is necessary to identify which species of aquatic biota are the most sensitive bioindicators for monitoring OCP contamination and the associated ecological effects of OCPs in specific aquatic ecosystems.

Lake Chaohu, which is located in the heart of Anhui Province (30°25′28″–31°43′28″ N, 117°16′54″–117°51′46″ E), is the fifth-largest freshwater lake in China and is approximately 760 km^2. In addition to fishing and its use in agricultural irrigation, Lake Chaohu is source of drinking water for the 9.6 million residents in the surrounding areas; and the water quality directly affects the health and safety of the residents. Before OCPs were banned, each environmental medium within Lake Chaohu was contaminated by long-term and extensive agriculture activities (Li et al., 2010; Ouyang et al., 2013). The growing amount of industrial and domestic wastewater discharged into the lake has further exacerbated the situation. Previous studies of Lake Chaohu have shown that OCP contamination remains in various media, including the water, suspended solids, sediment, the air and dust fall (He et al., 2012; Wang et al., 2012; Liu et al., 2013; Ouyang et al., 2012, 2013, 2014). However, limited information has been reported on the relationship between OCPs in aquatic biota and those in the water and suspended solids, as well as on the bioaccumulation, biomagnification and health risks of OCPs. The objectives of this study are to (1) investigate residual levels of OCPs in aquatic biota and identify the relationships between those levels and OCPs in the water and suspended solids; (2) analyze the relationships between OCP bioaccumulation and concentrations in suspended sediments and dissolved in water; (3) assess the potential health risks associated with the consumption of aquatic biota; and (4) identify the species of aquatic biota that are the most sensitive to OCPs and can serve as bioindicators for monitoring the contamination and associated risks of OCPs in the water and suspended solids of Lake Chaohu.

2. Materials and methods

2.1. Sample collection

In January 2011, a fisherman was employed to catch aquatic biota throughout the lake. Different sizes of six species of commonly consumed freshwater fishes, one species of shrimp (*Leander modestus Heller*) and one species of snail (*Cipangopaludina chinensis*) were collected. The six fish species were spotted steed (*Hemibarbus maculates*), carp (*Cyprinus carpio*), topmouth culter (*Culter erythropterus*), blunt snout bream (*Megalobrama amblycephala*), large icefish (*Protosalanx hyalocranius*), and bighead carp (*Aristichthys nobilis*). The fish were caught and stored in polypropylene boxes filled with lake water. To reduce individual differences, the muscles on both dorsal flanks and the chests of three or five of the same fish species were combined into one mixed sample, and a total of three parallel samples were taken for each fish species. After the wet weight was obtained, the samples were freeze-dried (FDU-830, Tokyo Rikakikai Co., Japan), weighed to measure dry weight and ground into a granular powder with a ball mill (MM400, Retsch GmbH, Germany). Amber glass bottles were used to hold the sample powder and sealed into a dryer until analyzed. The physical characteristics of the investigated species are presented in Table S1 of Appendix A.

Water and suspended solids data for Lake Chaohu were collected and published in two previous studies (Liu et al., 2013; Ouyang et al., 2013). Water samples were collected from May 2010 to April 2011, and the distribution of sampling sites is shown in Figure S1. The sampling methods used in those studies are as follows (Liu et al., 2013; Ouyang et al., 2013): One liter aliquot of each total water sample was filtered through a 0.45 μm glass fiber filter (ashed at 450 °C for 4 h) using a peristaltic pump (80EL005; Millipore Co., USA) and a 142 mm diameter filter plate to separate suspended solids out. Before use, the glass fiber filters were dried and weighed to a constant weight for 24 h. After air-drying, the suspended solid samples were stored in aluminum foil in desiccators to maintain a constant weight. The weight difference between the filters before and after filtering established the weight of the suspended solids. A total of three comparable samples for water and suspended solids were produced for each sampling site. Phytoplankton were collected using 10 L or 20 L water samples concentrated to approximately 50 mL using a 400 mesh plankton net (mesh diameter of 37 μm) and held in 100 mL vials to perform a stable N isotope analysis.

2.2. Extraction and cleanup

Two-gram powder samples were weighed into an extraction tube, and a recovery indicator and internal standard were added. After microwave extraction, the extracts were pressure filtered and concentrated to approximately 1 mL by rotary evaporation. 10 mL of ethyl acetate were added to the extracts, which were then re-concentrated to 1 mL. The samples were filtered through a 0.45-μm filter and subsequently transferred to GPC vials. After adding 3 mL of ethyl acetate, the samples were cleaned using a GPC instrument (GPC800+, Lab Tech Ltd., China) with a Bio Beads SX-3 column (300 mm × 20 mm, Bio-Rad Laboratories, Inc. USA). A ratio of 1:1 ethyl acetate/hexane was used at a flow of 5 mL/min. The injection volume was 2 mL. Lipids were also extracted by GPC and collected before the target OCPs. Lipid content was collected from 2 to 10 min and stored in a weighed eggplant-shaped flask. Fractions collected at 10–22 min were the target compounds. These extracts were concentrated to approximately 1 mL by rotary evaporation and then re-concentrated to 1 mL after 10 mL of hexane was added. Subsequently, each concentrate sample was loaded into a silica gel SPE cartridge (6 mL, 500 mg, Supelco Co., USA). These cartridges were conditioned with 10–15 mL of hexane before use. After loading, the cartridge was eluted by hexane (two times, 5 mL per elution) and a mixed solution of dichloromethane and hexane (V:V = 1:1, four times, 5 mL per elution). The extracts were concentrated to 1 mL, pentachloronitrobenzene (PCNB, 100 ng, AccuStandard, Inc.) was added to the sample as an internal standard, and the samples were transferred to vials and sealed for analysis.

The extraction and cleanup methods for water and suspended solids samples are presented in Appendix A and our previous papers (Liu et al., 2013; Ouyang et al., 2013). The algae samples were freeze-dried and sealed into a dryer until analyzed.