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Current status and historical variations of DDT-related contaminants in the sediments of Lake Chaohu in China and their influencing factors☆

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ABSTRACT

The temporal-spatial distributions of DDT-related contaminants (DDXs), including DDT (dichlorodiphenyltrichloroethane), DDE (dichlorodiphenyldichloroethylene) and DDD (dichlorodiphenyldichloroethane), in the sediments of Lake Chaohu and their influencing factors were studied. p,p-DDE and p,p-DDD were found to be the two dominant components of DDXs in both surface and core sediments. The parent DDT compounds were still detectable in sediment cores after the late 1930s. Historical usage of technical DDT was identified as the primary source of DDXs in sediments, as indicated by DDT/(DDD + DDE) ratios of less than one. The residual levels of DDXs were higher in the surface and core sediments in the western lake area than in other lake areas, which might be due to the combined inflow effects of municipal sewage, industrial wastewater and agricultural runoff. The DDX residues in the sediment cores reached peak values in the late 1970s or early 1980s. There were significant positive relationships between DDX residues in sediment cores with annual DDT production and with fine particulate sizes (<4.5 μm). The relationship between the DDXs and TOC in sediment was complex, as indicated by the significant differences among the surface and core sediments. The algae-derived organic matter significantly influenced the amount of residue, composition and distribution of DDXs in the sediments. The DDD/DDE ratios responded well to the anaerobic conditions in the sediments that were caused by algal blooms after the late 1970s in the western lake area. This suggests that the algae-derived organic matter was an important factor and served as a biomarker of eutrophication and also affected the DDX residues and lifecycle in the lake ecosystem.

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1. Introduction

Dichlorodiphenyltrichloroethane (DDT) was first synthesized in 1874; it was first used as pesticide in 1937 (Apeti et al., 2010) and for epidemic prevention in the US Army during the Second World War (Kannan et al., 1995). Since the mid-1940s, the production of DDT rapidly increased until the 1960s, when DDT and its metabolites (including DDE (dichlorodiphenyldichloroethylene) and DDD (dichlorodiphenyldichloroethane)) were found to resist degradation in the environment and to have negative impacts on non-target organisms (Saxena and Siddiqui, 1980). During the 1970–1980s, many countries began to regulate the productions and uses of DDT. DDXs, including o, p′-DDT, p, p′-DDT, o, p′-DDD, p, p′-DDD, o, p′-DDE and p, p′-DDE, have been proven to be seriously harmful to ecosystems and to human health (Snedeker, 2001; Wetterauer et al., 2012; Mrema et al., 2013). In 2001, DDT was listed in the Stockholm Convention on Persistent Organic Pollutants as one of the 12 initial POPs (UNEP, 2001). Meanwhile, some procedures have been undertaken to prohibit the productions and use of DDXs (SCPOPs, 2005). However, although their productions and use have been globally regulated for years, there are still detectable levels of DDXs in various media worldwide (e.g., Satcher, 2015; Li et al., 2015; Najam and Alam, 2015; Bouwman et al., 2015; Aamir et al., 2016; Gerber et al., 2016; Booij et al., 2016).

China started to produce technical DDT in the 1950s and became

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the second largest DDT manufacturer in the world by the time it was banned in 1983 (Wang et al., 2013). From the 1950s to 1982, over 0.45 million tons of DDT was produced, accounting for 20% of the total world production during that time (Wang et al., 2013). In China, DDT was not officially banned for agricultural usage until 1983, and small scale DDT applications are still allowed for public health purposes, as an additive of marine anti-fouling paint, and as the primary raw material of dicofol, which is a pesticide for controlling a variety of mites and annihilating mite eggs in agriculture (He et al., 2015). It is estimated that 10,000 tons of anti-fouling paints are used in fishing ships in China each year, and approximately 250 tons of DDT are used in anti-fouling paints (He et al., 2012). Although the production and use of DDT have been regulated for more than 30 years in China, DDXs are still detectable in different environmental media, such as soil, water, dustfall, sediment and ambient air (Li et al., 2006; Zhou et al., 2008; Wu et al., 2005; Liu et al., 2012; Yang et al., 2008). Among the various environment media, sediments are the primary sinks of DDXs, since they are easily adsorbed onto particulate matter due to their high K_{OW} (i.e., octanol–water partition coefficient) or K_{OC} (carbon normalized partitioning coefficient) and transported into the water-sediment system in surface runoff and dry-wet deposition (Cao et al., 2007; He et al., 2012, 2016). Lake sediment cores are high-resolution information databases that record the history of such contaminants as DDXs (Zhang et al., 2010). However, information regarding DDXs in sediment cores from shallow lakes is still limited (Barakat et al., 2013; Li et al., 2015) compared with studies of the historical records of DDXs in sediment cores from marine and estuarine environments (Qiu et al., 2009).

Lake Chaohu, with an average depth of 3 m and a water area of approximately 760 km^2, is located in the center of Anhui Province in southeastern China (Fig. 1). As the fifth largest freshwater lake in China, it was well known for its scenic beauty and rich aquatic biodiversity before the 1950s (Xu, 1997). However, the lake has since suffered from serious eutrophication, with frequent algal blooms in spring and summer, and it has been one of the most eutrophic lakes in China since the early 1980s due to increasing pressures from rapid population growth and economic development in the drainage area (Xu et al., 1999; Jiang et al., 2014). Our previous studies from recent years have shown that there are still high DDX residues in the water, sediment, suspended particulate matter, ambient air, dust fall, and aquatic biota (He et al., 2012; Liu et al., 2012, 2013; Ouyang et al., 2012, 2013, 2014; He et al., 2014, 2015; Liu et al., 2016). This could be attributed to the extensive historical DDT usage in the Lake Chaohu basin, which is one of the primary farming areas of China, before it was banned in 1983 (Ouyang et al., 2013). Furthermore, new inputs of technical DDT in Lake Chaohu have been found from the source apportionment of DDXs in the water, dust fall and suspended particulate matter, which may be a result of the recent usage of dicofol and marine paints (Ouyang et al., 2013, 2014; He et al., 2015). Recently, a study on the historical deposition behaviors of organochlorine pesticides (OCPs) in Lake Chaohu sediments was conducted (Li et al., 2015). It was determined that DDT contamination derived from the historical use of technical DDTs and that the DDX concentrations were highly significantly related to the total organic carbon (TOC) content, sediment grain sizes, nutrient content and heavy metal content (Li et al., 2015). However, the effects of algae-derived organic matter in the sediments on the residual levels and composition of DDXs remains unknown. Previous studies have been undertaken to determine the relationship between algae-derived organic matter and typical organic pollutants in different media. For example, algae-derived organic matter was found to have a significant influence on low-molecular polycyclic aromatic hydrocarbons (PAHs) (Wu et al., 2012). As for DDXs, their water solubility could be enhanced by the existence of algae-derived organic matter (Ma et al., 2012). However, limited information is available about the relationships between sedimentary records of DDXs and algae-derived organic matter in the sediments of shallow eutrophic lakes. This is very important information for understanding the historical accumulation of DDXs in the sediments of Lake Chaohu and their association with intensive algal blooms.

The objectives of this study were as follows: (1) to investigate the residual levels and distributions of DDXs in the surface and core sediments in Lake Chaohu; (2) to identify the composition and potential sources of DDXs in the surface and core sediments; and

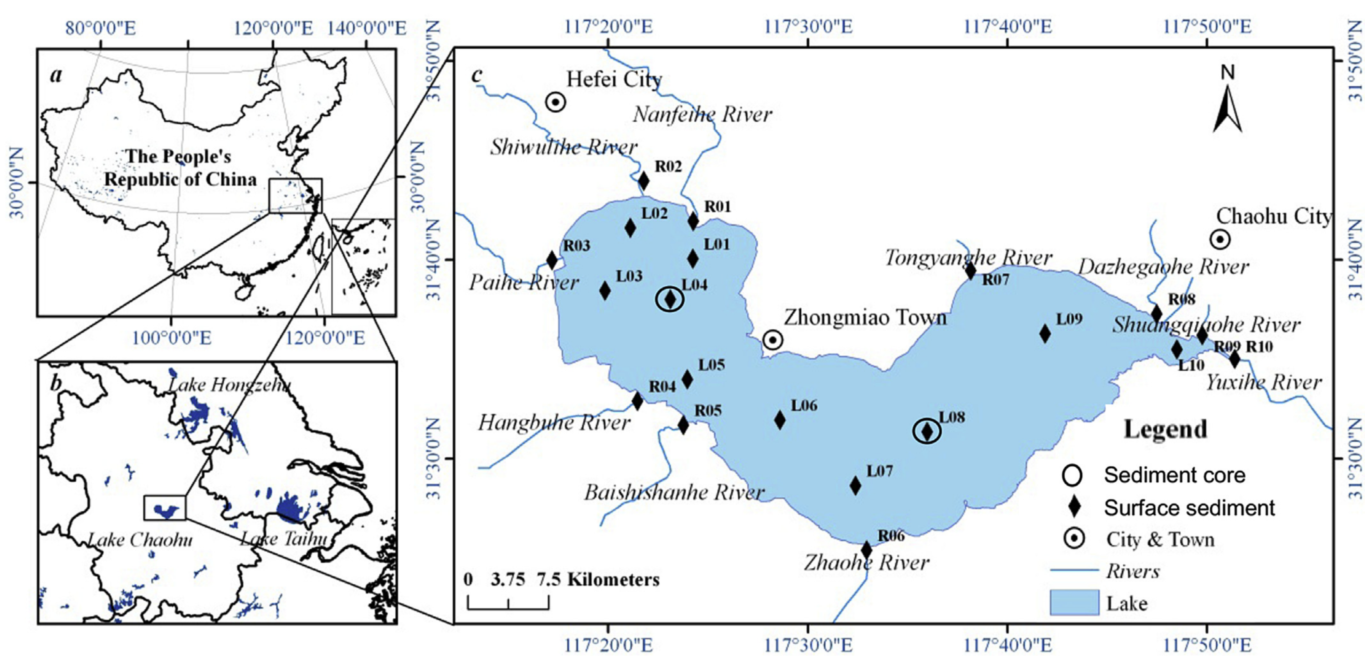

Fig. 1. Geographic locations of (a) Lake Chaohu, (b) Lake Chaohu in East China and (c) the sediment sampling sites in Lake Chaohu.

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