100-YEARS SEDIMENTARY RECORD OF POLYCHLORINATED BIPHENYLS IN URBAN LAKE SEDIMENTS OF WUHAN, CENTRAL CHINA

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Abstract

The concentrations and depositional fluxes of polychlorinated biphenyls (PCBs) were investigated in a dated sediment core collected from Donghu Lake, central China. The concentrations of ΣPCB in the sediment core varied from 1.3 to 43.1 $ng \cdot g^{-1}$. The profile of ΣPCB concentrations closely reflected the changes of production and usage of PCBs in this region, and the profile of ΣPCB fluxes were similar to that of ΣPCB concentrations except for a distinct decrease of ΣPCB fluxes after around 1998, which was probably attributed to the Yangtze River catastrophic flood of 1998 in China. This result suggested that the floods and heavy rains washing these compounds from land to the lake may be an important source of PCBs in the lake. Sedimentary profiles for PCB homologues presented the concentrations decrease in the following order: penta~hexa->tetra->tri->hepta-PCBs. The relative abundances of tri-PCBs and tetra-PCBs in the core accounted for more than 80% of the total PCBs before the first commercial use of PCBs, suggesting post-depositional mobilization of less chlorinated PCB homologues in the sediment core.

Keywords: PCBs, Sources, Composition, Sediment, Sedimentary record, Depositional fluxes

Introduction

Polychlorinated biphenyls (PCBs) are of great global concern due to their bioaccumulation, persistence, and impact on ecosystems. Leakages from such equipment, however, have recently been reported. Recently, pollution incidents involving PCBs have resulted in severe consequences in some regions of China[1-3]. Typically, lake systems have historically been in close proximity to industrial facilities and the recipients of anthropogenic waste, which likely results in a significant sink for PCB contamination in lake sediments. Furthermore, it has been evidenced that release of PCBs from aquatic sediments to overlying water and PCB volatilization from water bodies brought on significant losses of PCBs at temperatures and the losses were congener dependent[4]. It is therefore essential to evaluate the regional status of PCB contamination in subtropical lake environment for understanding the global context. Lake sediments are widely used to reconstruct temporal profiles of environmental contaminant inputs. Undisturbed sediment cores thus provide insight into local and global trends of past and present use of a wide range of substances. The objective of the present study was to determine the spatial distributions of PCBs in sediments, to identify their possible sources, and to assess the ensuing environmental risks in the middle reaches of the Yangtze River.

Materials and methods

A sediment core was collected in the central of Donghu Lake on September 2006 with a stainless steel static gravity corer (8 cm i.d.). The water depth at this sampling site was 4 m and the whole core was 52 cm long. The core was divided at 2 cm intervals immediately after collection using a stainless steel blade. These freeze-dried sediment samples were ground, homogenized, and stored at -20 °C prior to analysis. The dating of sediment core followed the method described by Kim et al.[5]. The sediment core was analyzed for ^{210}Pb, ^{226}Ra and ^{137}Cs by direct gamma assay using a high purity germanium detector (BE 5025, CANBERRA). Radiometric dates were calculated from the ^{210}Pb records based on the constant initial concentration model and corrected where appropriate using the 1963 and 1986 depth determined from the ^{137}Cs stratigraphic records as reference levels. The average depositional rate is determined to be 0.56 cm·yr^{-1}, which is in agreement with that reported by Yang et al.[6]. The subsample of sediment was frozen-dried and sieved with a 200-mesh stainless steel sieve. The sample was treated with 1.6% (v/v) hydrochloric acid to removed inorganic carbon and oven-dried again at 60 °C. Organic carbon was measured on a Liqui total organic carbon (TOC) (Elementar, Germany) at 950 °C.

Results and discussion

Figure 1 shows that the ΣPCB concentrations range from 1.0 to 43.1 ng·g^{-1} dry weight, with the mean value 12.2 ng·g^{-1}, and the top 4 cm layers had the highest ΣPCB concentrations. A high ΣPCB concentration in the core was observed in 1932, compared to the fact that PCBs were firstly manufactured commercially in 1929 and PCB-containing transformers and capacitors were imported to China, suggesting a time lag of a few years between production and incorporation into the lake sediments. Subsequently, the relatively constant ΣPCB concentrations were observed between mid-1930s and mid-1960s, but a slightly peak of ΣPCB concentration (8.1 ng·g^{-1}) presented around 1958 which could reflect the increase of PCB-containing by-products relating to the combustion or industrial processes during the specific historical period in China. In the concentration profile, a sharp increase of ΣPCB concentrations was commenced in 1965 and peaked again in 1976 (11.92 ng·g^{-1}). The increasing trend of PCBs was parallel to the fact that technical PCBs produced firstly in 1965 and after that much of PCB emission occurred discretionarily as a result of disorder of production and management for historical reasons in China (1966-1976). A decrease of PCB concentrations in the late-1970s (1976-1980) was evident, probably parallel to the first decline in the industrial production of technical PCBs in China around 1974[7]. The PCB concentrations started to rise again after early-1980s, and finally escalated to the maximum value at the surface layer as a great amount of PCB-containing electric equipments had been used in China until the

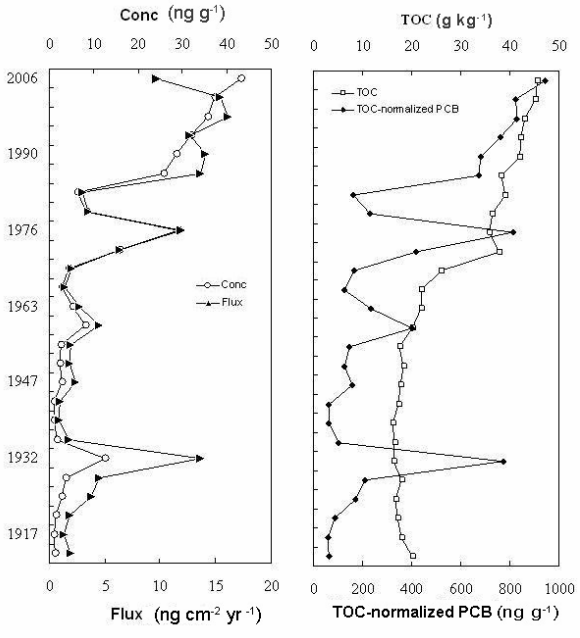

Figure 1 Variations of total PCB concentrations, PCB fluxes, TOC contents, and TOC-normalized PCBs in the sediments

late 1990s[7]. Decreases of PCB residues in the last two decades were observed from several compartments of the aquatic ecosystem in several recent studies[8-10]. However, this pattern was not registered in the sediment core analyzed in this study, which probably suggested that PCB-containing electric equipments are still in use or the releasing of industrial wastewater and domestic sewage containing PCBs exists in this region. The increasing trend has previously been observed for PCBs in sediment profiles elsewhere, such as the Pearl River Delta and Taihu Lake in China[1,11].

ΣPCB fluxes in the sediment core shown in Figure 1 varied from 0.92 to 16.16 ng·cm^{-2}·yr^{-1}. An exception was a distinct decrease of ΣPCB fluxes in the sediment core after around 1998, probably attributed to the Yangtze River catastrophic flood of 1998 in China. Relatively high ΣPCB fluxes after mid-1980s were observed in the core as a result of the large-scale process of land use and rapidly urban development began with the initiation of the "Reform and Open" in China in 1978. Another interesting feature of the sediment core was the presence of PCBs in deep sediment sections, dated prior to their industrial synthesis[1,4]. In this study, ΣPCB fluxes reached averaging 5.92 ng·cm^{-2}·yr^{-1}, within the range of PCB fluxes in the Pearl River Delta (4.6-16.2 ng·cm^{-2}·yr^{-1})[1]. However, the ΣPCB deposition fluxes were higher than those reported for the sediments from Nansihu Lake[9], Northern Great Lakes[8] and some remote Chilean Andean Lakes[12]. An alternative explanation for this is the inputs of PCBs come from the industrial wastewater and domestic sewage as well as the liberated PCBs from the large-scale process of land transformation[1,13]. A significant positive correlation (r=0.874, n=26, p<0.01) between TOC contents and ΣPCB concentrations (see Figure 1) also indicated that TOC can play an important role for transport and redistribution of PCBs in sediments.

Sedimentary profiles for PCB homologues (Figure 2) show that PCB congeners with ⩾8 chlorines were below detection in most sections of the core. The concentrations of penta~hexa-PCBs in the range of 0.01-26.09 ng·g^{-1}, averaging 5.90 ng·g^{-1}, were the highest. The concentrations decreased in the following order: penta~hexa->tetra->tri->hepta-PCBs. Exceptionally, The penta~hexa-PCBs and tri-PCBs depart most noticeably from this trend with a gradual increase toward the surface layer of the sediment after 1990, reflecting the

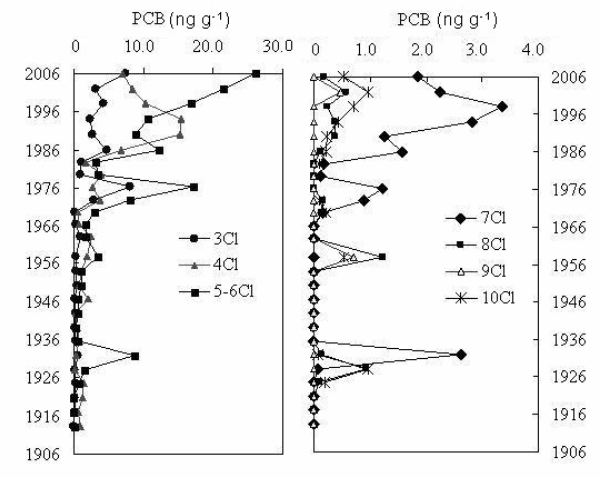

Figure 2 Historical profiles of PCB homologues in the sediment core

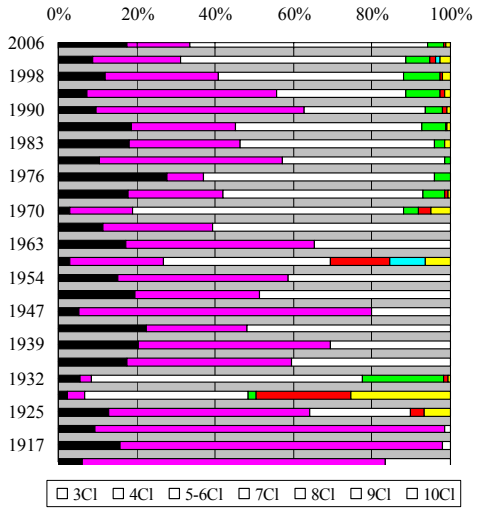

Figure 3 Vertical variations of relative abundance of PCB homologues in the sediment core

contribution of the low to moderate chlorinated PCBs in the growth of ΣPCB in recent years[3,14]. A similar observation in both soils and sediments in China led to speculations that it may be related to some source process. Contrarily, an apparent decrease for the highly chlorinated PCBs (7 to 10 chlorinated homologues) was observed after around 1998, which combined with the sharp decrease of ΣPCB fluxes in the sediment core (Figure 1), probably further suggesting that the floods and heavy rains washing these compounds from land to the lake are at present the main source of PCBs for Donghu Lake, but not a direct consequence of human activity in this region. Furthermore, it also further evidenced that higher chlorinated PCBs congeners commonly accumulated near source of PCBs[13,15], whereas lower chlorinated PCBs may be transported over longer distances.

Figure 3 displays vertical variations of relative abundance of PCB homologues in ΣPCB in the sediment core. The relative abundances of tri-PCBs and tetra-PCBs in the core accounted for more than 80% of the total PCBs in sediments predating the first commercial use of PCBs (Figure 3), indicating that less chlorinated homologue groups were preferably subject to post-depositional mobilization[1,4,16]. In the sediments deposited after mid-1930s, no apparent abundance changes for PCBs homologues were observed with depth except for a slight increase of 7-Cl homologues after 1970, suggesting that the higher chlorinated PCBs were the least affected by post-depositional solution phase mobility.

Acknowledgements

The research was supported by the National Basic Research Program of P.R. China (973 Project, 2003CB415204). We gratefully acknowledge project 40701166 supported by National Natural Science Foundation of China.

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