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Sedimentary record of hydrophobic organic compounds in relation to regional economic development: A study of Taihu Lake, East China

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PAHs, DDTs and PBDEs are still increasing in the Taihu Lake sediment.

A B S T R A C T

Sediment cores taken from Taihu Lake, East China were analyzed for polycyclic aromatic hydrocarbons (PAHs), organochlorine pesticides (OCPs) and polybrominated biphenyl ethers (PBDEs). The results showed a general sharp increase of HCH, DDT, PAH and PBDE concentrations in the surface layers, corresponding to a sedimentation time of 1980 and 1990 onward in the Meiliang Bay and Xukou Bay, respectively. The source of PAHs has largely transferred from petrogenic to pyrogenic origin, and good relationships were observed between sediment PAH concentrations and the regional gross domestic product. The sharp increase of DDTs in recent years may be related to the mobilization and migration of these chemicals from surface soil to lake sediment, as a result of enhanced soil run-off due to large scale land transform, as well as the contribution of current usage of dicofol and DDT-containing anti-fouling paints.

1. Introduction

Lake sediments are recognized as a major sink for hydrophobic organic compounds (HOCs), such as polycyclic aromatic hydrocarbons (PAHs), organochlorine pesticides (OCPs) and polybrominated biphenyl ethers (PBDEs). These and other chemicals may enter aquatic systems through direct/indirect discharge, surface run-off and atmospheric deposition. Contaminated sediments can directly affect bottom-dwelling organisms and represent a continuing source for toxic substances in aquatic environments that may affect wildlife and humans *via* food chains (Kannan et al., 2005). Therefore, an understanding of the time trends of HOC accumulation in sediment is vital if we are to assess the current status of surface water quality and to enact management strategies. It is possible, however, to construct the chronological pollutant trends through the investigation of sediment cores, and also provides a useful way to elucidate the relationship between regional economic development and environmental pollution.

The use of sediment cores to reconstruct historical input of contaminants has been well documented in numerous studies (Hites et al., 1997; Hites and Zhu, 2005; Mai et al., 2005; Van Metre et al., 1997). On a global scale, sedimentary PAH displayed decreasing trends in the 1970s and 1980s (Hites et al., 1997; Simcik et al., 1996; Yamashita et al., 2000), owing to the transition from home heating with coal to the use of oil and natural gas, as well as the improved combustion efficiencies in power generation. However, sedimentary PAHs have actually increased in some fast developing regions, due to rapid urbanization and increasing demand for energy consumption (Lima et al., 2003; Liu et al., 2005; Van Metre et al., 2000).

OCPs are a class of persistent organic pollutants (POPs) that have aroused global concern due to their negative effect on human and wildlife. To date, OCPs application has been restricted or banned in most countries. Earlier studies on OCPs in dated sediment cores from a number of locations revealed a gradual decrease in overall concentrations in the 1970s and 1980s (Chen et al., 2002; Van Metre et al., 1997; Wong et al., 1995), obtained sedimentary profiles similar to their production and usage history. In those regions with large scale land transform, however, the accumulation flux of DDTs showed abnormal peaks around the 1990s, and wash-out DDTs from soil was considered to be a possible cause. Specific examples include the North Africa wetlands (Peters et al., 2001), the Pearl River Delta in China (Zhang et al., 2002) and the Mississippi River Delta in the

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United States (Santschi et al., 2001). PBDEs are a group of brominated compounds that widely used as flame-retardants in furniture and consumer products. The concentration of PBDEs in sediments have increased from the 1960s or 1970s to the 1990s in Europe, North America and Asia (Binelli et al., 2007; Chen et al., 2007; Hites and Zhu, 2005; Sakai et al., 2002; Song et al., 2005; Zegers et al., 2003). Although the use of penta- and octa-BDEs has been restricted in the European Union and in certain parts of the United States (Renner, 2004), a vast number of products containing PBDEs still remain in use. The distribution pattern, potential sources and fate of these pollutants in the environment are still of great concern.

The Yangtze River Delta is one of the most important economic engines of China, characteristic of fast urbanization and industrialization. The Taihu Lake basin, which is the heart of the Yangtze River Delta, is also an important grain production base in China and has a record of the highest pesticide application in the country. The basin covers only 0.4% of the total territory of the country, but contributes 3.1% of the national food production and 10% of the gross domestic product (GDP). With rapid economic development and population increase, man-made pollutants derived from household, agricultural and industrial activities have been discharged into the water, resulting in serious pollution and eutrophication in Taihu Lake.

In this study, two lacustrine sediment cores collected from Taihu Lake were analyzed for PAHs, OCPs and PBDEs. The regional HOCs pollution history was reconstructed based on the core analyse, and the relationship between HOCs pollution and regional economic development indexes was further investigated.

2. Experiment section

2.1. Site descriptions and sampling

Located in the southern part of the Yangtze River Delta, Taihu Lake (Fig. 1) is the third largest freshwater lake in China. The lake has a catchment area of 2340 km^2 and an average water depth of 1.9 m. Sediment cores were collected in Meiliang (ML) Bay, a typical algae zone, and Xukou (XK) Bay, a typical macrophyte zone. A plastic static gravity corer (8 cm i.d.) was employed to minimize the disturbance of the surface sediment layer. The core samples were sectioned at 1-cm intervals and immediately stored at −20 °C until analysis.

2.2. Sediment dating

Sedimentation rates were calculated from the unsupported ^{210}Pb activity and used to determine the year of deposition of each sediment layer. The procedure of sediment dating was described in detail elsewhere (Zhang et al., 2002). In summary, the ^{210}Pb activity concentrations in sediment samples were determined by analysis of the α-radioactivity of its decay product ^{210}Po, on the assumption that both are at equilibrium. The Po was extracted, purified, and self-plated onto Ag discs at 75–80 °C in 0.5 mol/L HCl, with ^{209}Po used as yield monitor and tracer in quantification. Counting was done by computerized multi-channel α-spectrometry with Au–Si surface barrier detectors. Supported ^{210}Pb was obtained by indirectly determining the α-activity concentration of the supporting parent ^{226}Ra, which was carried by co-precipitated BaSO$_4$. In this study, dates were calculated using a constant rate of supply (CRS) dating model, obtained an average sedimentation rate of 0.21 cm/yr and 0.31 cm/yr in the XK core and ML core, respectively.
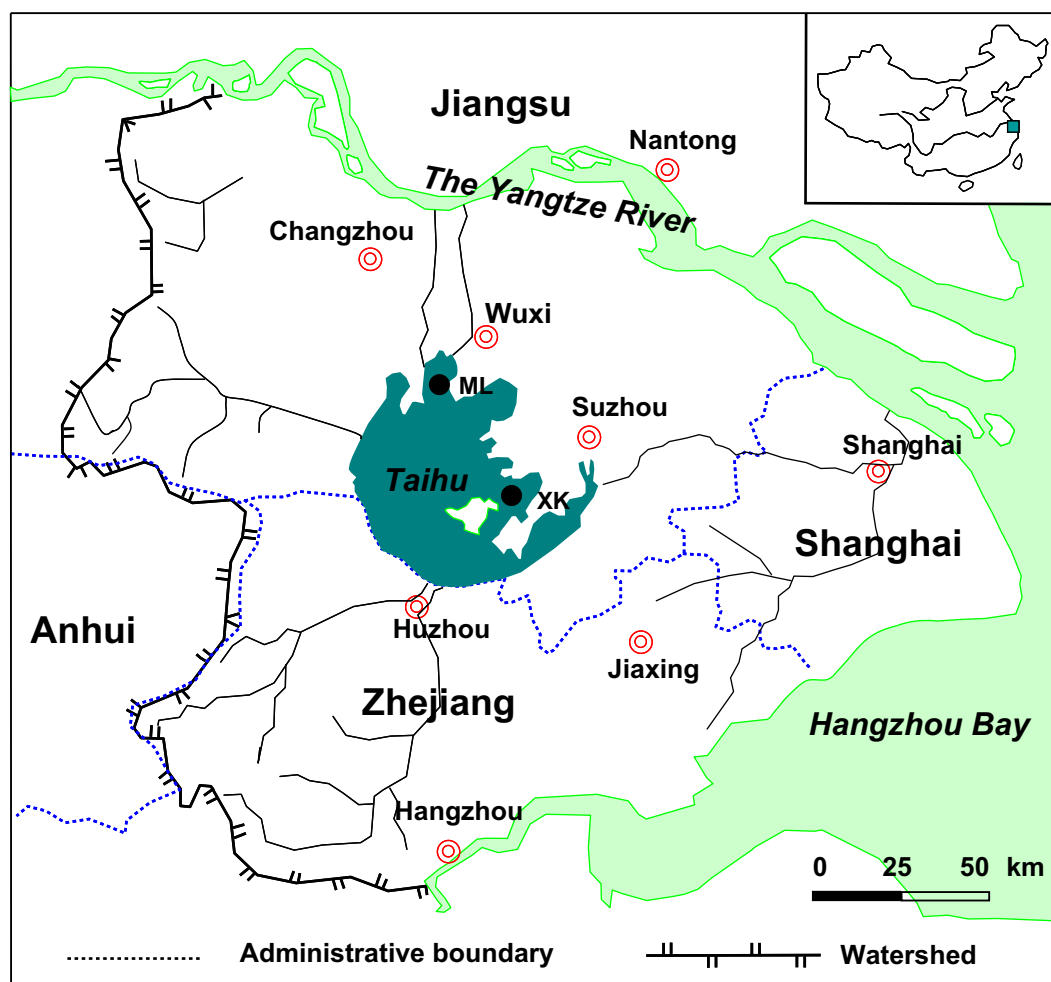

Fig. 1. Sketch map of Taihu Lake region with the sampling locations.

2.3. Sample extraction and cleanup

The procedures for the extraction and purification of PAHs, OCPs, and PBDEs are described in detail elsewhere (Liu et al., 2005; Mai et al., 2005; Zhang et al., 2002). In brief, sediment samples were freeze-dried, ground, homogenized, and extracted with acetone/hexane (1:1 v/v) using an accelerated solvent extraction (ASE) system with surrogates spiked (naphthalene-d8, acenaphthene-d10, phenanthrene-d10, pyrene-d10, chrysene-d12 and perylene-d12 for PAHs, TCmX, PCB 209, ^{13}C-PCB 141 for HCHs, DDTs and PBDEs). Activated copper granules were added to the collection flask to remove elemental sulfur. Concentrated extracts were loaded on a 10-mm i.d. silica/alumina column, packed from the bottom to top with neutral alumina (6 cm, 3% deactivated), neutral silica gel (2 cm, 3% deactivated), and anhydrous sodium sulfate (1 cm, 450 °C baked), and eluted with 60 ml of hexane/dichloromethane (6:4 v/v). The final volume was reduced to 200 µl under a gentle nitrogen stream. A known quantity of internal standard (hexamethybenzene) was added to all the samples prior to GC–MSD analysis.

After PAHs analysis, samples were further cleaned-up using a mixed silica gel column (from the bottom to top: 3 cm neutral alumina gel, 5 cm neutral silica gel, 6 cm 50% sulfuric acid silica gel, 2 cm anhydrous sodium sulfate). OCPs and PBDEs mixture were eluted with 50 mL of hexane. The extracts were concentrated to 0.2 ml under a gentle N$_2$ stream, and a known amount of internal standard, penta-chloronitribenzene (PCNB) for OCPs, ^{13}C-PCB 208 for PBDEs was added to prior to GC–ECD and GC–NICI-MS analysis.

2.4. Instrument analysis

PAHs were quantified on an HP-5971 GC–MSD system operating at 70 eV under scan mode. Analytics were separated with a DB5-MS column (30 m × 0.25 mm diameter, 0.25 µm film thickness). The GC temperature program was as follows: initially at 80 °C for 5 min, increased to 290 °C at 3 °C/min, and held for 30 min. A 1 µL sample was injected into the splitless injector with a 5 min solvent delay. Peak confirmation and quantification was performed on a DOS-based HP ChemStation system. The following 16 PAHs were selected as target compounds in the analysis: naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flo), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chry), benzo[b]-fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo-[a]pyrene (BaP), dibenz[a,h]-anthracene (DiA), indeno[1,2,3-c,d]pyrene (InP) and benzo[g,h,i]perylene (BghiP).

HCHs and DDTs were measured using an HP-6890 gas chromatograph (ECD) and an HP5-MS silica fused capillary column (30 m × 0.25 mm diameter, 0.25 µm film thickness). The oven temperature began at 60 °C (held 1 min) and increased to 290 °C at 4 °C/min (held 25 min). A 1 µL sample was injected into the splitless injector with 1 min solvent delay time. The inlet degradation of DDT was checked daily and controlled within 15%.

Polybrominated biphenyl analysis was performed with a Shimadzu model 2010 gas chromatograph (GC) coupled with a model QP2010 mass spectrometer (MS) (Shimadzu, Japan) using negative chemical ionization (NCI) under SIM mode. Ten PBDEs were measured in this study, including congeners 28, 47, 66, 100, 99, 154, 153, 138, 183, and 209. A DB-5MS (30 m × 0.25 mm i.d., 0.25 µm film thickness) capillary column was used for the determination of PBDE congeners, except for BDE-209. The column temperature was initiated at 110 °C (held for 1 min) and increased to 180 °C at 8 °C/min (held for 1 min), 240 °C at 2 °C/min (held for 5 min), 280 °C at 2 °C/min (held for 25 min), and 290 °C at 5 °C/min (held for 15 min). For BDE-209, a CP-Sil 13 CB (12.5 m × 0.25 mm i.d., 0.2 mm film thickness) capillary column was used. The oven temperature was programmed from 110 to 300 °C at a rate of 8 °C/min (held for 20 min) and the high-pressure splitless injection mode was used with a solvent delay time of 1 min.

2.5. Quality control and assurance

The instruments were calibrated daily with calibration standards. Method blanks (solvent), spiked blanks (standards spiked into the solvent) and sample duplicates were routinely analyzed. The surrogate recoveries for HCHs and DDTs were in the range of 81.6–112%, with a mean value of 95%. The mean recovery for PAHs was 112%, ranging from 70% to 140%. The surrogate recoveries for PBDEs were in the range of 72–115%, with a mean value of 90%. Reported concentrations were corrected for surrogate recoveries. The levels of p,p′-DDT, phenanthrene, BDE-47 and BDE-99 in procedure blanks were well below 5% of the levels found in the sediment samples, and the difference between duplicates of all chemicals was <15%.

3. Results and discussion

3.1. Surface sediment

The surface slice (top 2 cm of the core) represents the current status of sediment contamination. Chemicals in this layer are more prone to re-suspension and represent a continuing source for toxic substances in aquatic environments. The surface sediment concentrations of

$\sum$PAHs, HCHs (sum of α-, β-, γ- and δ-HCH), DDTs (sum of p,p′-DDT, p,p′-DDD, p,p′-DDE and o,p′-DDT), $\sum_9$PBDEs and BDE-209 were 1180, 6.3, 6.8, 0.3, and 25 ng/g in the ML core, and 530, 1.8, 2.5, 0.2 and 5.3 ng/g in the XK core.

The PAH levels in Taihu Lake were close to those found in the Bohai Sea and the Yellow Sea in China (877–5730 ng/g) (Ma et al., 2001), and in river sediments, such as the Yalujiang River (68–1500 ng/g) (Wu et al., 2003) and the Minjiang River Estuary (112–877 ng/g) (Zhang et al., 2004). However, these levels were lower than those reported in highly urbanized and industrialized regions, such as the Pearl River (1434–10,811 ng/g), the Macau Inner Harbor (294–12, 714 ng/g) (Mai et al., 2002), the Michigan Inland Lakes (50–16,900 ng/g) (Kannan et al., 2005), the Kiel Harbor (3–30, 000 ng/g) (Baumard et al., 1999), and the Ýzmit Bay, Turkey (250–25, 000 ng/g) (Tolun et al., 2001). For HCHs and DDTs, the residue levels found in this study were well within the worldwide ranges. The levels of $\sum_9$PBDEs in Taihu Lake were comparable to those reported in river/sea sediments in China, such as the Yangtze River Delta (n.d–0.55 ng/g) (Chen et al., 2006), the South China Sea (0.04–94.7 ng/g) (Mai et al., 2005), and nearshore sediments of Qingdao (0.1–5.5 ng/g) (Yang et al., 2004). These levels were also close to those found in the Ebro river basin (NE Spain) (n.d–20.9 ng/g) (Lacorte et al., 2006), but were higher than those of the Danube Delta (<0.1 ng/g) (Covaci et al., 2006). The levels of BDE-209 in Taihu Lake fell in the low end of the global range, as extremely high concentrations of BDE-209 have been reported in sediments around the world, such as 11, 600 ng/g in Japanese rivers (Watanabe et al., 1995), 7100 ng/g in Swedish rivers (Sellström et al., 1988), and 7340 ng/g in the Pearl River Delta, South China (Mai et al., 2005).

3.2. Time trend of PAHs and sources

The temporal trends of PAH concentrations in the sediment cores are shown in Fig. 2. $\sum$PAH concentrations ranged from 180 to 1600 ng/g and from 88 to 610 ng/g in the ML Bay and XK Bay, respectively. In the core profiles, PAH concentrations began to dramatically increase in the 1980s in the ML Bay and 1990s in the XK Bay. The differences between the cores may be explained by possible different hydrological conditions at the sediment sites, as well as by the local economic development patterns. Located in the northern part of Taihu Lake, ML Bay serves as the main water supply for Wuxi city and has received large amounts of municipal sewage and industrial waste water since the 1980s. In contrast, XK Bay, downstream of the lake, is less developed and therefore contains less contaminants than that of the ML bay.

PAHs, produced mainly from the incomplete combustion of fossil fuels, are good indicator of anthropogenic activities (Lima et al., 2003; Liu et al., 2005; Van Metre et al., 2000). In the last two decades, the Taihu Lake region has undergone rapid economic
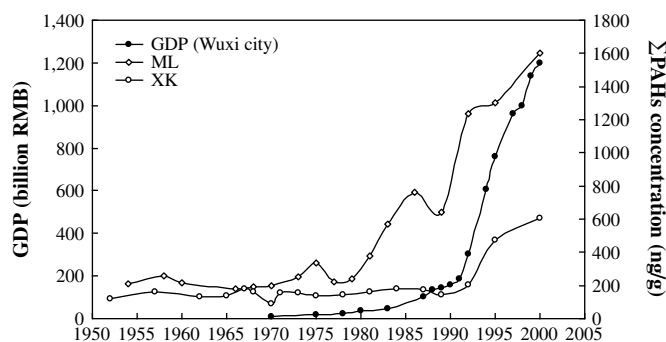

Fig. 2. Time trends of PAH concentrations in the sediment cores versus GDP.

development, and the Suzhou-Wuxi-Changzhou city region in southern Jiangsu is one of the most developed regions around the lake, with flourishing economy and the highest level of urbanization. Taking Wuxi city as an example, the increasing rate of GDP was 13.5% during the 1980s, and reached 22.7% between 1990 and 1994 (Wuxi Municipal Statistical Bureau, 2000). This was much higher than the country's average. Coincident with the rapid economic development in this region, PAH concentrations showed sharp increase in the ML Bay sediments. As shown in Fig. 2, the total PAH concentrations increased from 310 ng/g in 1980 to 1600 ng/g in 2000, at a rate of 65 ng/year over the past 20 years. The results clearly elucidated the effect of human activity on the aquatic environment.

There are two types of anthropogenic sources of PAHs: petrogenic and pyrogenic. Petrogenic PAHs are typically low molecular weight (LMW) PAHs, whereas high molecular weight (HMW) PAHs are generated mainly through combustion processes (Hwang et al., 2003; Yunker et al., 2002). Fig. 3 shows the changes in PAH compositions through time. Phenanthrene appeared to be the dominant compound in both core sediments deposited before the mid-1970s. After this time, the proportion of HMW PAHs increased, while that of LMW compounds decreased. The PAH composition profiles in both cores indicates a general shift of PAH sources from petrogenic to pyrogenic origins in recent years, coincides with the rapid increase in energy consumption and diesel emission in the Taihu Lake region.

3.3. Time trends of DDTs and HCHs

The vertical trends of HCHs and DDTs in the sediment cores are presented in Fig. 4. HCH concentrations ranged from 0.4 to 6.0 ng/g and from 0.4 to 2.7 ng/g in the ML Bay and XK Bay, respectively. For DDT, it was 1.5–12 ng/g and 0.4–9.0 ng/g, respectively. In the core profile, a steady increase in HCH levels was observed since the 1950s, followed by the first peak in the 1970s. Since then, HCHs and DDTs concentrations showed sharp increase in the ML Bay and reached a maximum in the early 1980s. In the years that followed, HCH and DDT concentrations either decreased or remained constant. Since the late 1980s, HCH and DDT levels showed sharp increase in both cores, and reached the overall maximum.

It is interesting to note the increasing trends or sharp rebound of HCH/DDT after their production ban imposed in China in 1983. DDT is degraded to DDD under anaerobic conditions and to DDE in aerobic environments (Hitch and Day, 1992). The ratio of DDE/DDD and (DDD + DDE)/DDT has been used to trace the environment of decomposition and the new input of DDT (Lee et al., 2001; Rapaport et al., 1985). Fig. 4 shows a sharp increase of (DDE + DDD)/DDT ratio between the late 1970s and early 1990s, concurrent with the increasing trend of DDT concentrations. These results may suggest that DDT deposited after the production ban was more likely to be "weathered" DDT.

Historically the Taihu Lake region was an important grain-growing region of China, and agriculture activity played a dominant role in basic land use patterns. However, since the 1980s, the region has undergone rapid economic development and population growth, which led to a widespread land conversion from agriculture land to urban areas. There were two peak periods of arable land loss, which were round about 1985 and 1993 (Yang, 2001), coincident with the HCH and DDT concentration peaks in the 1980s and 1990s. We suggest that the sharp increase of HCH and DDT in
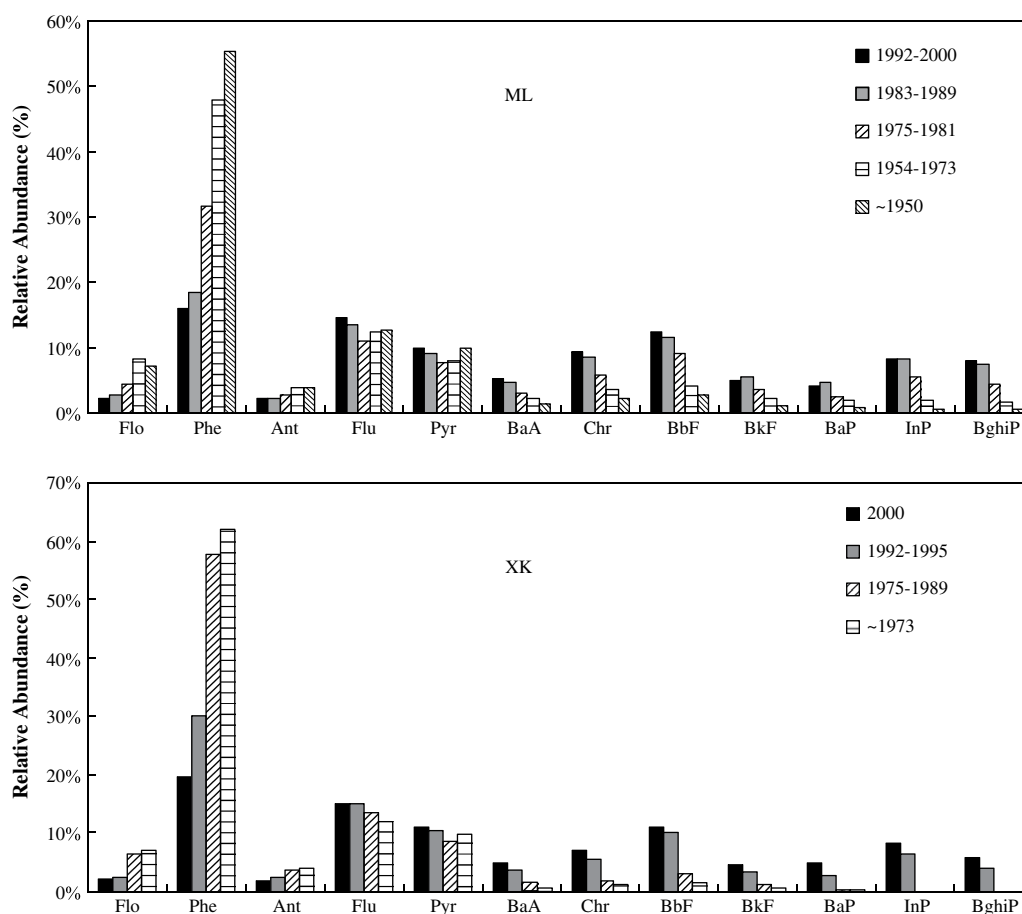

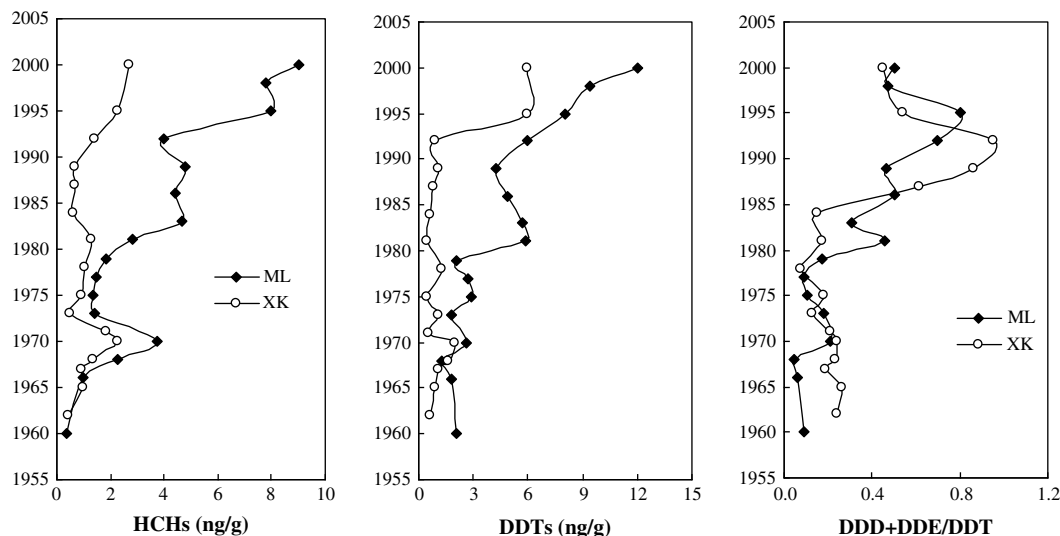**Fig. 3.** Temporal variations of PAH compositions in the core sediments.

Fig. 4. Sedimentary HCH and DDT concentrations and the ratios of (DDD + DDE)/DDT.

the surface layer sediments may be related to the mobilization and migration of these chemicals from surface soil to the sediment pool, as a result of enhanced soil run-off due to large scale land transform.

It is worthwhile to note the decreasing trend of (DDE + DDD)/ DDT ratios in recent years, which may indicate new inputs of DDT. Although the production and usage of DDT has been officially banned in China since 1983, DDT is still allowed to be used for dicofol production, anti-fouling paint, malaria control and mosquito repellents. Between 1988 and 2002, 6000–7000 t of DDT was produced annually in China (Qiu et al., 2005). The majority of this was used for dicofol production, and 150–300 t of DDT per annum was used for anti-fouling paint (Wang et al., 2007a). Recent studies have indicated that DDTs levels in the air around Taihu Lake were still high, and the current usage of dicofol was suggested to be the possible source (Qiu et al., 2004, 2005). Fishery is one of the important economic functions of Taihu Lake, and there are thousands of ships traveling on the canals and rivers every day. During the fish suspension seasons, large amounts of DDTs accompanied with anti-fouling paints usage would be introduced to the aquatic environment. Based on the above evidence, we suggest that the usage of dicofol and

anti-fouling paint for fishing ship are the current DDT sources in the Taihu Lake region.

3.4. Congener pattern and time trend of PBDEs

Of the 10 PBDE congeners measured in this study, BDE-209 was the most abundant compound, making up to 86–99% of the total PBDEs. For the other non-deca congeners, the sediment pattern largely resembles the commercial penta mixture, with BDE 47 and BDE 99 dominant, followed by BDE 28 and BDE 66. In both of the commercial products, DE-71 and 70-5DE, BDE-99 is more abundant than BDE-47 (Hale et al., 2008; LaGuardia et al., 2006). And most studies reported a higher contribution of BDE-99 than BDE-47 in sediments from the Columbia River and Ebro river (Lacorte et al., 2006; Rayne et al., 2003). In the Great Lake (Hites and Zhu, 2005; Song et al., 2005) and Taihu Lake sediments, however, BDE-47 was more abundant than BDE-99. Such differences might result from the use of different types of commercial penta-BDE products, or PBDE congener fractionation/partition in various environment, as BDE-47 is more volatile than BDE-99, and is relatively stable when exposed to sunlight (Eriksson et al., 2004). And the formation of BDE-47 during the photolysis of highly brominated congeners would be another possibility (Bezares-Cruz et al., 2004; Söderström et al., 2004).

Historical trends of PBDEs in various environmental and human tissues have been reported throughout the world. Due to differences in the timelines of production and use of technical PBDE, PBDEs concentrations have been decreasing or leveling off in recent decades in Europe (Law et al., 2006), while still increasing in certain North America and Asia environment (Hites and Zhu, 2005; Song et al., 2005; Wang et al., 2007b). The PBDE concentrations in the ML and XK cores are illustrated in Fig. 5 as a function of deposition year. Similar to the core profiles of PAHs, $\sum$PBDE and BDE-209 concentrations showed sharp increase in the surface layer sediments, corresponding to a sedimentation time of 1980 in the ML Bay and 1990 in the XK Bay. This is consistent with the rapid economic development and increasing demand for PBDEs in the Taihu Lake region. To our knowledge, bans or restrictions of PBDEs have not been commenced in China. The increasing trend of both $\sum$PBDEs and BDE-209 found in this study may suggest that penta- and octa-BDE mixtures are still in use in the Taihu Lake region, and there is no sign of use replacement by deca-BDE formula.
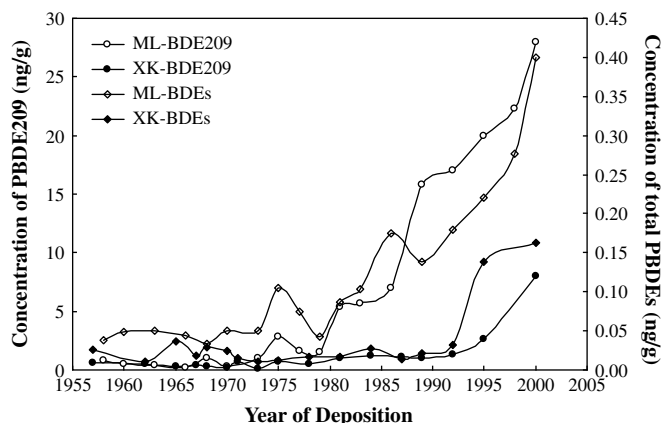

Fig. 5. Time trend of $\sum_9$PBDE and BDE-209 concentrations in the sediments.

4. Conclusion

In Taihu Lake, East China, sedimentary PAH, HCH, DDT and PBDE concentrations began to dramatically increase in the 1980s in the ML Bay and 1990s in the XK Bay. Good relationships were observed between the sediment HOC concentrations and the regional economic indexes such as gross domestic product and cultivated land loss. The source of PAHs has largely transferred from petrogenic to pyrogenic origin. The sharp increase of DDT after the production ban may be related to the mobilization and migration of these chemicals from surface soil to lake sediment, as a result of enhanced soil run-off due to large scale land transform, as well as the contribution of current usage of dicofol and DDT-containing anti-fouling paints. There has been no sign of use replacement of PBDEs by other flame-retardants in the Taihu Lake region.

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