Sedimentary record of polycyclic aromatic hydrocarbons in Lake Erhai, Southwest China

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Abstract

The temporal distribution of polycyclic aromatic hydrocarbons (PAHs) was investigated in a sediment core from Lake Erhai in Southwest China using gas-chromatography/mass spectrometry (GC/MS) method. The total organic carbon (TOC) normalized total PAHs concentrations (sum of US Environmental Protection Agency proposed 16 priority PAHs) ranged from 31.9 to 269 μg/g dry weight (dw), and were characterized by a slowly increasing stage in the deeper sediments and a sharp increasing stage in the upper sediments. The PAHs in the sediments were dominated by low molecular weight (LMW) PAHs, suggesting that the primary source of PAHs was low- and moderate temperature combustion processes. However, both the significant increase in high molecular weight (HMW) PAHs in the upper sediments and the vertical profile of diagnostic ratios pointed out a change in the sources of PAHs from low-temperature combustion to high-temperature combustion. The ecotoxicological assessment based on consensus-based sediment quality guidelines implied that potential adverse biological impacts were possible for benzo(ghi)perylenelene and most LMW PAHs. In addition, the total BaP equivalent quotient of seven carcinogenic polycyclic aromatic hydrocarbons (BaA, CHr, BbF, BkF, BaP, DBA and INP) was 106.1 ng/g, according to the toxic equivalency factors. Although there was no great biological impact associated with the HMW PAHs, great attention should be paid to these PAH components based on their rapid increase in the upper sediments.

Introduction

Polycyclic aromatic hydrocarbons (PAHs) in sediment have various origins, including diagenic sources, petrogenic sources and pyrogenic sources (Gallon et al., 2005). PAHs can be introduced to aquatic environments through atmospheric deposition, riverine and municipal runoff, accidental oil spills, etc. As a result, PAHs are ubiquitous in aquatic ecosystems (Fernández et al., 2000; Usenko et al., 2007). PAHs in sediment are not necessarily trapped permanently, because they can be recycled via chemical and biological processes (Zeng and Venkatesan, 1999; Hu et al., 2010). Accordingly, sediment acts as an important sink of PAHs, as well as a secondary source. Due to their toxicity, persistence and long-range transport properties, PAHs are one of the most concerned contaminants worldwide (Xu et al., 2006; Wilcke, 2007; Lang et al., 2008).

PAHs have a variety of well documented toxic effects,

including immunotoxicity, genotoxicity, carcinogenicity and reproductive toxicity (Sverdrup et al., 2002). Sediment toxicity tests can provide direct evidence of exposure to contaminants, but these approaches involve great technical and logistic efforts. As an alternative, ecological risk assessment can provide a framework for evaluating the toxic effects of sedimentary pollutants in aquatic environments. Numerous sediment quality guidelines (SQGs) for freshwater ecosystems have been developed and summarized by MacDonald et al. (2000). Nevertheless, each approach has certain advantages and limitations that influence their application to sediment quality assessment. To reach an agreement among these published SQGs, consensus-based SQGs that include a threshold effect concentration (TEC) and a probable effect concentration (PEC) have been developed and shown to be reliable for assessing sediment quality conditions in freshwater ecosystems (MacDonald et al., 2000).

Lake Erhai (100°5′E–100°17′E, 25°35′N–25°58′N) is the largest fault lake in Southwest China. The lake has

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a surface area of 249 km^2 and a drainage area to surface area ratio of 11.2. The length and the maximum width of Lake Erhai are 43 and 8.4 km, respectively. In addition, the maximum depth of Lake Erhai is 21.5 m, with a corresponding volume of 2.88×10^9 m^3 and a water residence time of 2.75 years (Wu et al., 1997; Wan et al., 2003). The lake plays an important role in the local socio-economic activities (Wu et al., 2001; Guo et al., 2001). However, the rapid economic development, rapid urbanization and industrialization that have occurred in its catchments within the last three decades have had adverse environmental impacts. Moreover, the city of Dali, which is located at the southern end of the lake and well-known for its natural scenery and ethnic customs, has become one of the most popular tourist destinations in Southwest China. The rapid tourist development could further accelerate the deterioration of the environmental quality in the basin. Unfortunately, based on the authors' knowledge, very few data, including PAHs in sediment, were associated with the environmental concerns in Lake Erhai.

Based on the above concerns, this study was conducted to investigate the temporal changes in PAHs in a sediment core from Lake Erhai, and to evaluate potential ecotoxicological impacts. Possible PAH sources were also identified. The results from the present study provide critical information regarding the increasing PAHs emission and source change in China, as well as the environmental changes occurred in Lake Erhai.

1 Materials and methods

1.1 Sampling

Sediment core was collected from Lake Erhai in May of 2006. The sampling site (25°49′N, 100°11′E) was the lowest part of the lake, which is far from any point-pollution sources and subject to little sediment redistribution or anthropogenic processes (Fig. 1). The sediment core was collected using a gravity corer with a diameter of 6 cm. The entire core, which was 65 cm long, was sectioned at 2 cm intervals. Upon collection, the cores were wrapped in pre-cleaned aluminum foil and then transported with ice
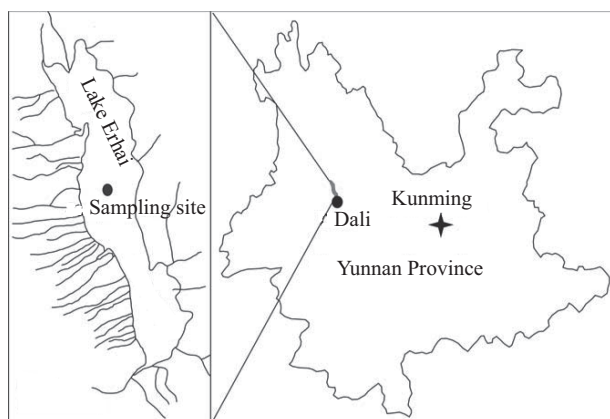

Fig. 1 Map of the sampling sites.

to the laboratory, where they were stored at –20°C until further treatment.

1.2 Extraction and instrumental analyses

The detailed sample extraction procedures have been reported elsewhere (Mai et al., 2002). Briefly, an aliquot of freeze-dried, homogenized sample (approximately 5 g dry weight) was spiked with a deuterated PAHs mixture (naphthalene-d$_8$, acenaphthene-d$_{10}$, phenanthrene-d$_{10}$, chrysene-d$_{12}$, and perylenelene-d$_{12}$) as a recovery surrogate, and then Soxhlet extracted with a 200 mL mixture of hexane and acetone (1:1, V/V) for 48 hr. Approximately 2 g of activated copper were added for desulphurization. The extracts were then filtered, concentrated, and solvent-exchanged with hexane. The cleanup and fractionation was conducted on an alumina/silica gel chromatography column with 70 mL dichloromethane/hexane (3:7, V/V) and finally concentrated to 200 μL under a gentle flow of nitrogen. The internal standards, 2-fluorobiphenyl and P-terphenyl-d$_{14}$, were added prior to instrumental analysis.

Instrumental analysis was conducted using an Agilent 6890 GC system equipped with an Agilent 5975B MSD (Agilent Technologies, Inc., Wilmington DE, USA) operating in selective ion monitoring mode. A DB-5 fused silica capillary column (60 m length × 0.25 mm i.d. × 0.25 μm film thickness) was used for chromatographic separation. Splitless injection of 1.0 μL sample was conducted using an auto-sampler. The GC oven temperature was programmed to increase from 90 to 180°C at a rate of 10°C/min, then to 220°C at a rate of 2°C/min, and finally to 290°C at a rate of 8°C/min, where it was held for 30 min. Ultrahigh purity helium (99.999%) was employed as the carrier gas at a constant flow rate of 1 mL/min. The injector and detector temperatures were 280 and 300°C, respectively. Mass spectra were acquired in electron ionization (EI) mode with an impact voltage of 70 eV. The quantified compounds included naphthalene (Nap), acenaphthylene (Ace), acenaphthene (Ac), fluorene (Flu), phenanthrene (Phen), anthracene (Ant), fluoranthene (Fluo), pyrene (Pyr), benzo(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbP), benzo(k)fluoranthene (BkP), benzo(a)pyrene (BaP), indeno(1,2,3-cd)pyrene (INP), dibenzo(ah)anthrathene (DBA), benzo(ghi)perylenelene (BghiP) and perylenelene (perylene).

1.3 Quality control and quality assurance

The surrogate recoveries for all samples were 58.2% ± 6.3% for naphthalene-d$_8$, 78.6% ± 6.0% for acenaphthene-d$_{10}$, 83.3% ± 5.2% for phenanthrene-d$_{10}$, 77.6% ± 3.4% for chrysene-d$_{12}$, and 63.5% ± 2.1% for perylenelene-d$_{12}$. For method quality control, a procedural blank (solvent), a triplicate spiked blank (standards spiked into solvent), and triplicate spiked matrices (standards spiked into matrices) were analyzed for each batch of samples. The mean recoveries of the sixteen target PAHs ranged from 75.8% ± 4.9% to 114.3% ± 8.2% in triplicate spiked blanks and from 60.8% ± 1.1% to 120.9% ±9.1% in triplicate spiked matrices. The instrument was calibrated daily using standard solution and the daily calibrations were < 15% for

all target analyses. In addition, only trace levels of targets were detected in blanks, and these were subtracted from those in the samples. The detection limits were 0.01–0.02 ng/g dry weight for PAHs (defined as $S/N > 3$). All results were expressed on a dry weight basis and corrected for surrogate recoveries.

2 Results and discussion

2.1 Temporal trends of PAHs in sediment cores

At the same sampling site, the sedimentation rate of Lake Erhai sediment cores has been determined in detail using both ^{210}Pb and ^{137}Cs, and the results of both methods were similar, being 0.21 and 0.20 cm/yr, respectively (Zhang et al., 1993). In the present study, the sedimentation rate of 0.21 cm/yr was used to reconstruct the temporal trends in the PAHs in the sediment core, and only the top 20 cm of the core were analyzed. Because TOC can probably influence the vertical profile of PAHs in sediment, the TOC normalized concentrations were used, and the results are shown in Fig. 2. The TOC normalized total PAHs (TPAHs, sum of US Environmental Protection Agency proposed 16 priority PAHs) concentrations ranged from 31.9 to 269 μg/g, and were increased roughly from the deeper segments to the surface. Overall, the vertical profile of the TPAHs was clearly characterized by two different stages: a slowly increasing stage from segment 18–20 cm to segment 8–10 cm (approximately 1918 to 1965), and a sharply increasing stage from segment 6–8 cm to the top sediment (approximately 1976 to the present). Throughout the core, there was an unexpected decrease in PAHs from segment 8–10 cm to segment 6–8 cm (approximately 1966 to 1975), with decreasing TOC normalized TPAHs concentration decreasing from 91.4 to 65.4 μg/g. In China, a political movement called Cultural Revolution happened in the right period (1966–1976), greatly damaged social-economic activities, resulting in many manufacturing plants either stopping or greatly reducing their production, especially during the later period
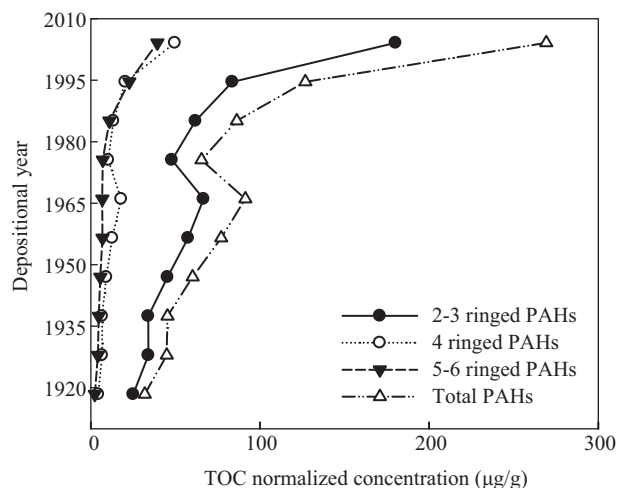
(Guo et al., 2006). This is reflected well in the sediment core. Similar trends were also observed in a core from the East China Sea (Guo et al., 2006) and a core from the Pearl River Delta, South China (Liu et al., 2005b). As for the grouped PAHs (Fig. 2), the temporal trends of 2–3 ringed PAHs and 4 ringed PAHs were very similar to those of TPAHs, but the concentrations of 5–6 ringed PAHs were quite flat in the deeper sediments and sharply increased in the upper sediments. The relatively high concentration of low to middle ringed PAHs in segment 8–10 cm (ca. 1966) was likely due to the destruction of large quantities of forest for steel-making that occurred during this period. The extensive wood combustion processes resulted in large amounts of low to middle ringed PAHs being emitted to the environment, without a visible increase in 5–6 ringed PAHs. After the "Reform and Open Policy" was implemented in 1978, China entered a period of rapid social-economic growth characterized by rapid population growth and large-scale industrialization and urbanization. At the same time, the demand for energy from fossil fuels increased drastically (Guo et al., 2006). The significant increase in the TOC normalized concentration of TPAHs (including 5–6 ringed PAHs) from 65.4 to 269 μg/g in the upper sediments likely reflects the socio-economic development that has occurred during the past three decades.

2.2 Composition and source identification of PAHs

The relative abundance of individual PAH species in different time intervals is shown in Fig. 3. Throughout the core, the PAHs in the sediments were clearly dominated by Phen and Flu, which accounted for 38.9% ± 3.5% and 18.1% ± 2.2% of the TPAHs, respectively. Phen and Flu are primarily produced by low and moderate temperature combustion processes such as the burning of biomass and domestic coal (Harrison et al., 1996; Mai et al., 2003). The significantly high abundance of Phen and Flu suggested that the PAHs input to the sediments were primarily from low and moderate temperature combustion. Nevertheless, the relative abundances of high molecular weight PAHs (including BbF, BkF, BaP, DBA, INP and BghiP) were significantly increased in the upper sediments. BbF is a product of high-temperature combustion (Mai et al.,
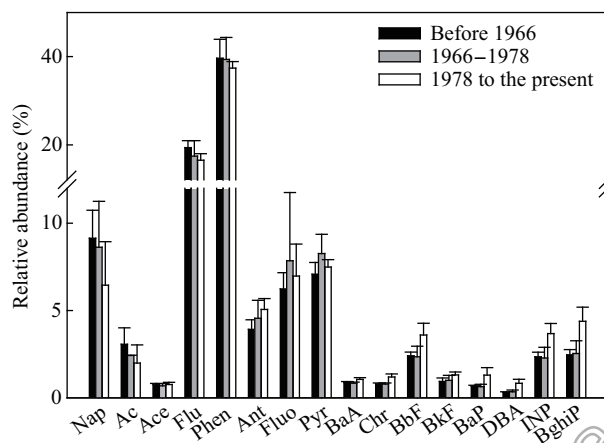

Fig. 2 Vertical profiles of TPAHs and grouped PAHs in the sediment core.

Fig. 3 Relative abundance of individual PAHs in different time intervals.

2003), and INP and BghiP are tracers of vehicular exhaust (Miguel and Pereira, 1989; Harrison et al., 1996). The increasing abundance of these PAH species implied that PAHs related to high-temperature combustion processes such as industrial coal combustion, vehicular emission and gas-fired cooking (Miguel and Pereira, 1989; Harrison et al., 1996) contributed more portion to the total PAHs emission in recent years.

PAHs in sediments from various emission sources are very complicated; therefore, it is not easy to determine their origins. Diagnostic ratios (typically isomers) of PAHs can likely provide some useful information (Liu et al., 2005a; Brändli et al., 2007; Peng et al., 2008). For example, an INP/INP+BghiP ratio greater than 0.5 could imply a source of combustion of coal and biomass, while an INP/INP+BghiP ratio between 0.2 and 0.5 suggests that liquid fossil fuel combustion is the origin (Guo et al., 2006). Conversely, ratios of BaA/BaA+CHr and INP/INP+BghiP less than 0.2 would suggest a petrogenic origin (Katsoyiannis et al., 2007). The depth profiles of BaA/BaA+CHr and INP/INP+BghiP are shown in Fig. 4. Throughout the core, the ratios of BaA/BaA+CHr and INP/INP+BghiP ranged from 0.44 to 0.54 (mean: 0.50) and 0.43 to 0.50 (mean: 0.48), respectively. These findings clearly suggested a pyrogenic source of PAHs. The INP/INP+BghiP values were very close to 0.50, indicating a mixture source of biomass burning and fossil fuel combustion. Evaluation of the depth profile revealed that the INP/INP+BghiP values were quite flat in deeper sediments, but obviously decreased in the upper sediments (from segment 6–8 cm to the top sediment, corresponding to the last three decades). A similar trend was also observed for BaA/BaA+CHr. These findings suggested a

source change from coal and biomass burning to petroleum combustion, which was in agreement with the increasing abundance of high molecular weight PAHs in the upper sediments, and the recent energy structure change in China (Guo et al., 2006).

Apart from the 16 priority PAHs proposed by the US EPA, significantly high levels of perylene (9.5–24.4 ng/g) were also observed in the sediment core. The variation in the concentration of perylene in the sediment core differed completely from that of the other PAHs (Fig. 5). Similar to other PAH species, perylene in sediment has various origins, such as the combustion of biomass and fossil fuels and in situ diagenesis of organic matter in anoxic sediments (Venkatesan, 1988; Baumard et al., 1998). Although the major source of perylene appears to be natural in situ formation in subaqueous sediments, i.e., anaerobic diagenesis of organic matter, the precursor material remains enigmatic (Silliman et al., 2001). Nevertheless, the natural source is often dominant in its parent PAHs (Gschwend et al., 1983), while the pyrogenic origin of perylene accounts for a very small portion of its parent PAHs (Luo et al., 2006). This facilitates us to differentiate the different sources of perylene. The ratio of perylene to the sum of 5 ringed PAHs (perylene/$\sum$5R-PAHs) has been used to differentiate the sources of perylene (Baumard et al., 1998). If perylene/$\sum$5R-PAHs is less than 0.1, a pyrolytic source of perylene is suggested, otherwise a diagenetic source is suggested. In the present study, the perylene/$\sum$5R-PAHs ratios were between 0.54 and 0.89, which is much higher than 0.1, obviously pointing to a predominant diagenetic source, i.e., diagenesis of organic matter.
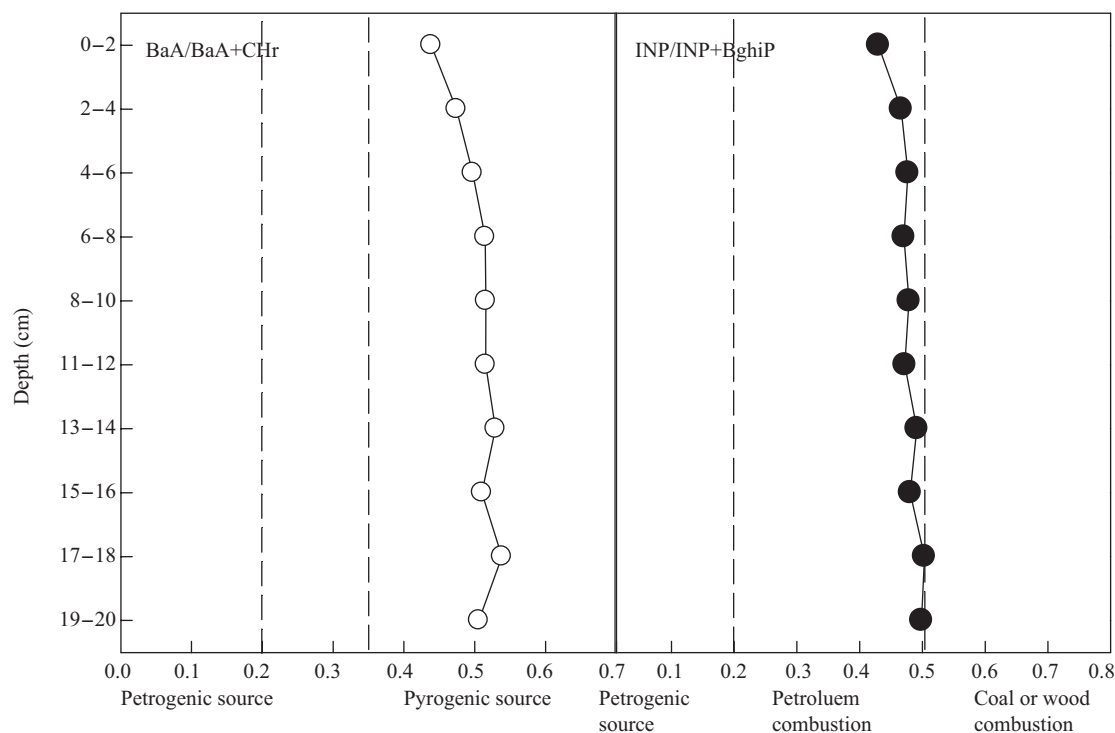

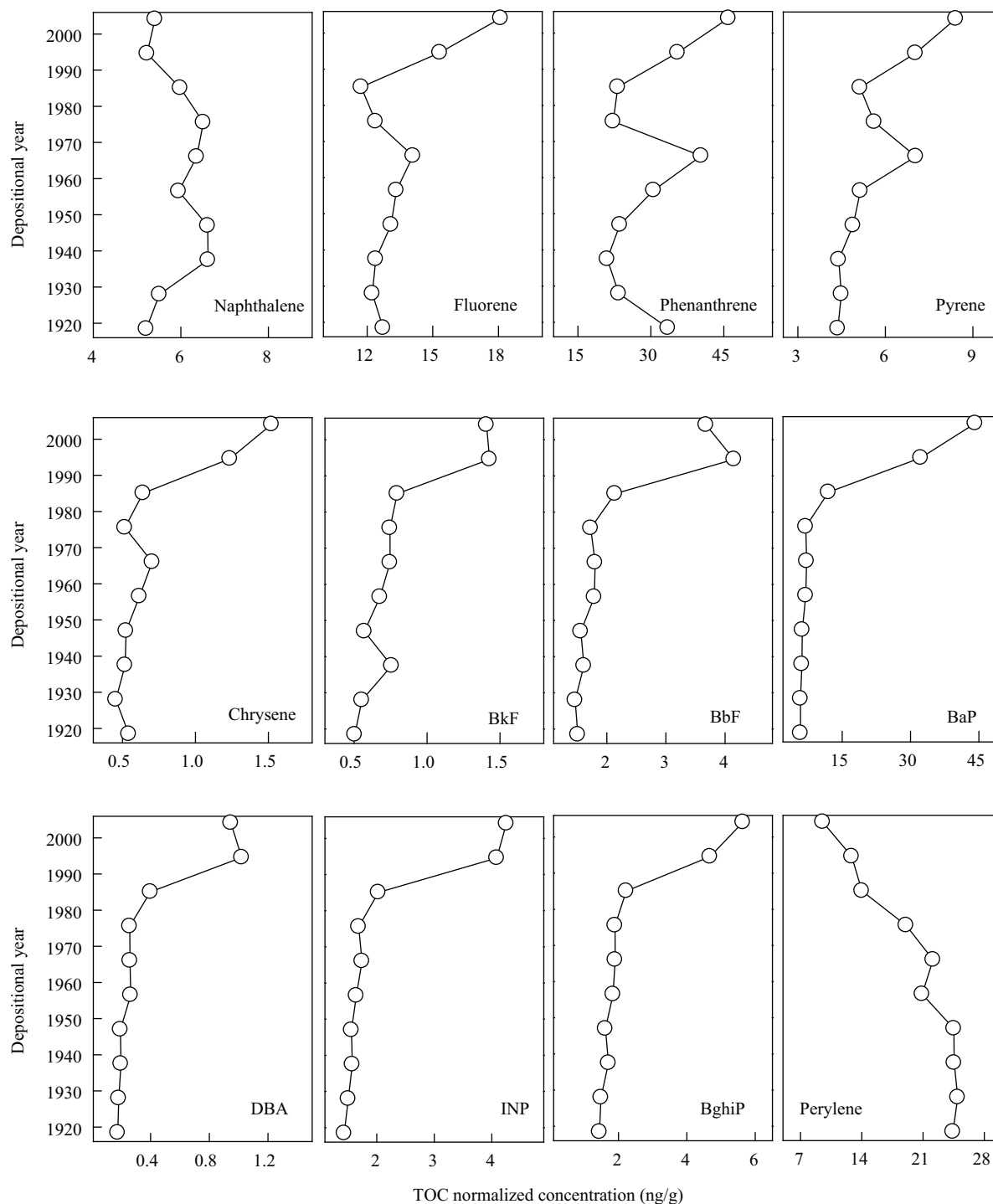**Fig. 4** Diagnostic ratios of PAHs in lake sediments.

Fig. 5 Vertical profiles of selected PAHs in sediment core from Lake Erhai.

2.3 Potential toxicological significance of PAHs

Several sediment quality guidelines have been developed to assess the potential biological effects of the contaminants, each of which has certain advantages and limitations that influence their application in the sediment quality assessment process (MacDonald et al., 2000). To reach an agreement among these published SQGs, consensus-based SQGs that included a threshold effect concentration (TEC) and a probable effect concentration (PEC) (MacDonald et al., 2000) were used in the present study. Because the consensus-based SQGs cannot provide benchmark concentrations for all 16 EPA PAH priority

pollutants, the threshold and probable effect levels from the Canadian Sediment Quality Guidelines (CCME, 2010) for Ac, Ace, and DBA, and the low and severe effects levels proposed by Persaud et al. (1993) for BkP, BghiP, and INP, as an alternative, were used for the assessment.

The measured concentrations of PAHs in the surface sediment were used for comparison with the TEC and PEC value, to derive the hazard ratios (HR_TEC and HR_PEC, representing the hazard ratio of the measured concentration of individual PAH species to the TEC and PEC values, respectively). If HR_TEC is less than 1, rare adverse biological effects are expected, while occasional

or frequent adverse biological effects could occur when HR_TEC > 1 or HR_PEC > 1. The results of the assessment are shown in Fig. 6. The HR_TEC of most low molecular weight PAHs (including Nap, Ac, Ace, Flu, Phen, Ant, and Pyr) and BghiP, as well as the TPAHs were greater than 1, suggesting occasional adverse biological effects could exist. In addition, the HR_PEC of both Flu and Phen were also greater than 1. Hence, Flu and Phen should receive greater attention due to their frequent adverse biological effects. In the present study, the sampling site was selected to intentionally avoid any point-pollution sources and was likely representative of the lowest part of PAHs contamination in Lake Erhai. Hence, further investigation of the entire lake is strongly recommended.

Another assessment focusing on the seven carcinogenic PAHs (BaA, CHr, BbP, BkP, BaP, DBA and INP) based on the toxic equivalency factors (TEFs) was also conducted. Among the seven carcinogenic PAHs, BaP is the only component for which there is sufficient toxicological data available for derivation of a carcinogenic factor (Peters et al., 1999). For other PAH components, TEFs were used for quantification. According to the US EPA (1993), the TEFs for BaP, BaA, CHr, BbP, BkP, INP and DBA are 1, 0.1, 0.001, 0.1, 0.01, 0.1, and 1, respectively (Table 1). The total BaP equivalent quotient (TEQ, ng/g) for seven carcinogenic PAH components are calculated using the following equation:

$$\text{TEQ} = \sum C_i \times \text{TEF}_i$$

where, C_i (ng/g) is the surface concentration of individual PAH and TEF_i is the corresponding toxic equivalency factor. The TEQ value for the surficial sediment from Lake Erhai was 106.1 ng/g (Table 1). The contribution of each PAH component to the TEQ decreased in the following order: BaP (41.6%) > DBA (29.5%) > INP (13.2%) > BbP (11.5%) > BaA (3.7%) > BkP (0.44%) > CHr (0.05%). The TEQ in this study was much lower than those of surface sediments (ranging from 94 to 856 ng/g, with a mean value of 407 ng/g from Meiliang Bay in the northern part of Lake Taihu in Eastern China (Qiao et al., 2006) and surface sediments (334.3 ng/g) from Lake Hongfeng in Southwest China (Guo et al., in press) , but comparable to those of the bottom sediments (92 ± 94 ng/g) from Guba Pechenga, Barents Sea in Russia (Savinov et al., 2003).

3 Conclusions

Temporal trends in polycyclic aromatic hydrocarbons in the sediment core from Lake Erhai were investigated in this study and their possible sources and potential toxicological significance were also identified. The concentrations of TPAHs in the sediment core ranged from 31.9 to 269 μg/g and were characterized by a slow increase in deeper sediments and a sharp increase in the upper sediments. These findings likely reflect the local socioeconomic development that occurred during the depositional time interval. Throughout the core, PAHs were dominated by low molecular weight components, especially Phen and Flu. These findings suggest that the dominant source of PAHs was low and moderate temperature combustion processes such as biomass burning and domestic coal burning. However, the temporal trends in high molecular weight PAHs and the BaA/BaA+CHr and INP/INP+BghiP ratios strongly suggested a change in energy usage from low-temperature combustion to high-temperature combustion.

The biological assessment conducted based on the consensus-based SQGs implied that potential adverse biological impacts are likely in response to BghiP and most low molecular weight PAHs (including Nap, Ac, Ace, Flu, and Phen), especially Flu, and Phen. As with the seven carcinogenic PAH components, the toxicological evaluation suggested that the TEQ of these PAH components was 106.1 ng/g. Although little biological impact is associated with high molecular weight PAHs, greater attention should be paid to these PAH species based on their rapid increase in the upper segments.
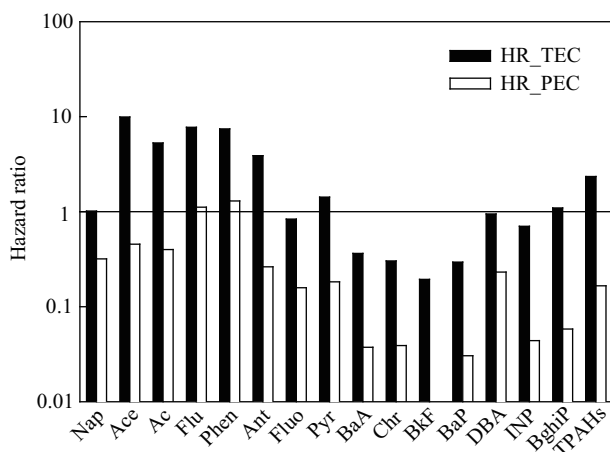

Fig. 6 Risk assessments of PAHs in surface sediment from Lake Erhai HR_TEC and HR_PEC denote the ratio of the measured concentration of individual PAHs to the TEC and PEC values, respectively.

Table 1 Toxicological assessment of the seven carcinogenic PAHs in surface sediment

Compound	TEF	BaP equivalent quotient (ng/g)	Percentage to TEQ (%)
BaA	0.1	3.9	3.70
CHr	0.001	0.1	0.10
BbF	0.1	12.1	11.5
BkF	0.01	0.5	0.40
BaP	1	44.2	41.6
DBA	1	31.3	29.5
INP	0.1	14.0	13.2

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