Environmental Science and Pollution Research (2022) 29:33427–33442
https://doi.org/10.1007/s11356-022-18497-4

RESEARCH ARTICLE

One-century sedimentary record, sources, and ecological risk of polycyclic aromatic hydrocarbons in Dianchi Lake, China

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Received: 12 October 2021 / Accepted: 30 December 2021 / Published online: 14 January 2022

Abstract
In this study, the sedimentary records, sources, and ecological risks of polycyclic aromatic hydrocarbons (PAHs) in Dianchi Lake were analyzed. The concentrations of ΣPAH_{16} in the sediments of Dianchi Lake ranged from 368 to 990 ng/g, with an average value of 572 ng/g, peaking in 1988. Economic development, rapid population growth, and rapid growth of coal consumption have a greater impact on the HMW (high molecular weight) PAHs than on the LMW (low molecular weight) PAHs in the sedimentary environment. The results of the diagnostic ratios and PCA (principal component analysis) model show that the main sources of PAHs were coal and biomass combustion, as well as the fossil fuel combustion in individual years. The risk assessment results showed that the PAH concentrations in the sediment were within a safe range. In the past 100 years of sediment pore water, other 2–3 ring LMW PAHs were within a safe range (except for Phe, which reached chronic toxic pollution levels in some years). With an increase in industrialization and urbanization, the burning of fossil fuels such as coal and petroleum has increased, and some of the 4–6 ring HMW PAHs have reached chronic toxicity or even acute toxicity in the sediment pore water.

Keywords Polycyclic aromatic hydrocarbon · Source apportionment · Risk assessment · Century sedimentary record · Dianchi Lake

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are persistent organic pollutants (POPs) that exist in different environmental media (Gregg et al. 2015; Lu et al. 2012; Sandro et al. 2018; Walker et al. 2005). PAHs can originate from natural processes such as forest fires (Freeman and Catell 1990; Ma et al. 2020), volcanic eruptions (Kim et al. 2003; Morillo et al. 2007; Ma et al. 2018), and diagenesis of organic matter in oxygen-deficient sediments (Baumard et al. 1998; Van Metre et al. 2000). However, human activities such as garbage incineration (Mastral and Callen 2000), fossil fuel combustion (Blumer and Youngblood 1975; Ma et al. 2021b), and wood burning for household heating and cooking (Lima et al. 2005; Zhang et al. 2007) are generally considered to be the main sources of PAHs entering the environment (Viguri et al. 2002).

Lake sediment is an important source and sink of pollutants (Li et al. 2021). PAHs enter the aquatic ecosystem through various processes, such as urban and agricultural runoff, automobile exhaust emissions, and fossil fuel leakage (Rinawati et al. 2012; Wang et al. 2021). Owing to the low solubility and

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strong hydrophobicity of polycyclic aromatic hydrocarbons (Boehm and Farrington 1984; Mouhri et al. 2008), they are easily combined with particles and eventually accumulate in lake sediments (Donahue et al. 2006; Gogou et al. 2000; Liu et al. 2007). However, as the main source and sink of PAHs, sediments may also cause secondary pollution to the aquatic ecosystem, such as Dianchi Lake through pore water transportation, posing a substantial threat to animals, plants, and humans (Rockne et al. 2002; Tao and Liu 2019; Tao 2021). PAHs have attracted worldwide attention because of their potential carcinogenicity, mutagenicity, and teratogenicity, as well as their persistence in the environment and possible health risks (Han et al. 2019; Jia et al. 2021; Meyer et al. 2011).

Dianchi Lake is the largest freshwater lake in Yunnan Province, China. Due to its low replenishment coefficient and long lake water retention period, pollutants are concentrated in the lake (He et al. 2015). Since the reform and opening up of China, especially after the 1990s, rapid urbanization and industrialization have promoted an increase in human activities, such as industrial and agricultural development, deforestation, and tourism (Gu et al. 2017; Liu et al. 2008; Zeng and Wu 2009; Zhang et al. 2015). These activities have led to a sharp increase in the discharge of pollutants, a decline in water quality, and serious threats to Dianchi Lake and its ecological environment, making Dianchi Lake one of the most polluted lakes in China (Li et al. 2003b). Examining the sediments in Dianchi Lake and providing a risk assessment of PAHs in the sediments and pore water can provide a scientific basis for pollution control and risk management of PAHs in this lake.

Although some studies have investigated the deposition records and sources of PAHs in Chinese plateau lakes (Guo et al. 2010; Yang et al. 2016; Zhao et al. 2014), information on persistent organic pollutants in lakes in the western plateau is still limited. Dianchi Lake is an important source of water for industrial development and human use in Kunming (Ma et al. 2021a); the quality of its water is directly related to the development of the surrounding industries, and it also affects people's health. Therefore, it is necessary to evaluate the concentration levels, sources, and ecological risks of PAHs in Dianchi Lake. The main objectives of this study were to: (1) study the depositional records of polycyclic aromatic hydrocarbons in the sediments of Dianchi Lake and their relationship with human activities, (2) explore the source of PAHs in Dianchi Lake, and (3) assess the ecological risk of PAHs in sediments and pore water from 1860 to 2014.

Materials and methods

Sampling and experimental analysis

Dianchi Lake (24°40′–25°03′ N, 102°37′–102°48′ E) is located on the Yunnan-Guizhou Plateau in China, southwest of Kunming (Fig. 1). The Dianchi Lake basin covers an area of 2,920 km^2 and has an average depth of 4.7 m (Du et al. 2011). It is approximately 40-km long from north to south and is 12.5-km wide (Gu et al. 2017; Ma et al. 2020). Sediment cores (102.67E, 24.69 N) were collected in July 2014 from the eastern part of Dianchi Lake using a gravity sampler with an internal diameter of 8.3 cm (Fig. 1). The sediment core (length: 39 cm) was cut into 1 cm segments, and each section was sealed in polygon bags at -4 °C and transported to the laboratory, where they were stored at -50 °C until further analysis.

Sediment core dating

The dating of each sediment core was based on the activity of ^{210}Pb. Briefly, the activity of ^{210}Pb and ^{226}Ra in the samples was measured using an Ortec HPGe GWL series, well-type, coaxial, low background, intrinsic germanium detector. The activities of ^{210}Pb and ^{226}Ra were determined from the gamma emissions at 46.5 or 295 keV, and 352 keV, respectively. These were emitted in gamma rays by the daughter isotope (^{214}Pb), which was stored for three weeks prior to dating in sealed containers to enable radioactive equilibration. Unsupported ^{210}Pb (^{210}Pbex) was calculated as the difference between the measured total ^{210}Pb at 46.5 keV and an estimate of the supported ^{210}Pb activity determined by the parent nuclide at 351 keV [^{210}Pbex = ^{210}Pb$_{tot}$-214Pb] (Huang et al. 2018; Ma et al. 2018).

Microwave extraction and analysis by GC–MS

A detailed description of the extraction and cleanup methods for the samples is provided in previous studies (Ma et al. 2020, 2021b; Zhang et al. 2017), and the experimental procedure is briefly explained in this study. First, 2 g of each sample was weighed, and then 25 mL hexane/acetone (1:1, v/v) solution was mixed with each sample and subjected to microwave extraction. Next, the extract was centrifuged at 3000 rpm for 15 min and repeated three times, after which 20 mL of hexane/acetone (1:1, v/v) solution was added. Third, the concentrated extract was reduced to 1 mL by rotary evaporation. Fourth, the extracts were purified by a chromatography column (1.5 g of 100–200 alumina mesh and 1.5 g of 80–100 silica gel mesh, 1 g of sodium sulfate) with a 50-mL solution of hexane/acetone (1:1, v/v). Finally, the extracts were reduced to 1 mL by rotary evaporation, and 1 mL of the extract was protected from light by amber glassware (Ma et al. 2020).

Shimadzu QP2010plus gas chromatography-mass spectrometry (GC–MS) was used to determine the PAH

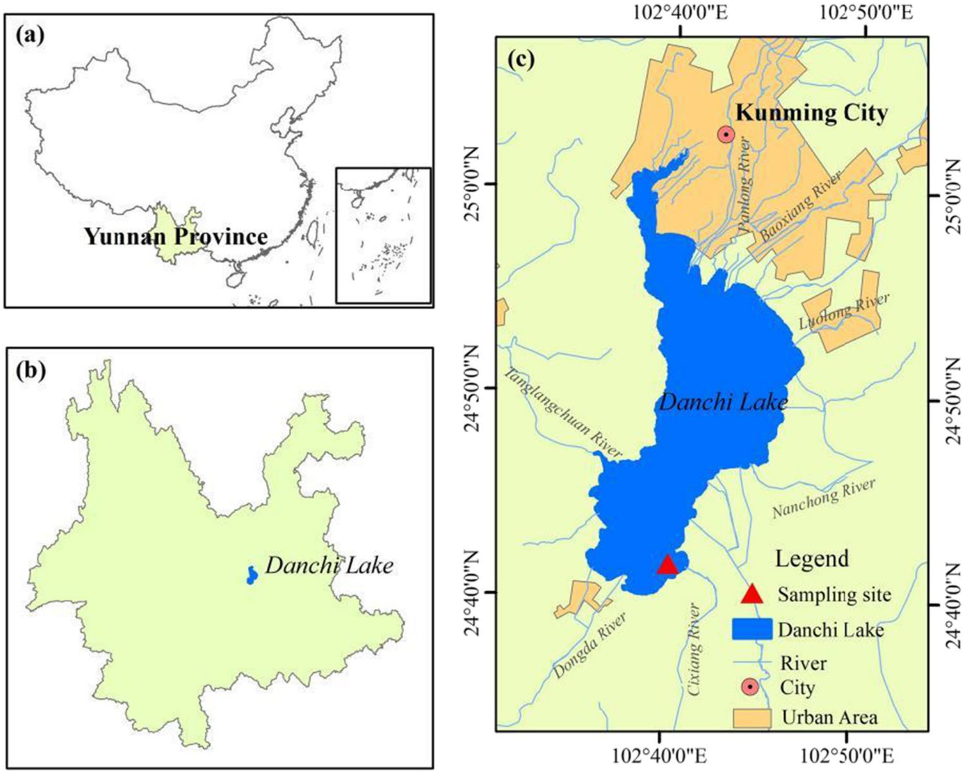


Fig. 1 Location of the sampling site in Dianchi Lake

concentrations. The PAHs were separated at a set temperature in a silica capillary column (HP-5MS; diameter, 30 m×0.25 mm; film thickness, 0.25 µm). Helium was used as the carrier gas (99.999%) at a constant pressure of 20.06 psi. Approximately 1 µL of each sample was added using the splitless injection method. The injector temperature, detector temperature, and initial oven temperature were 250 °C, 280 °C, and 90 °C, respectively. The initial oven temperature was first increased to 160 °C at a rate of 20 °C/min, then increased to 200 °C at a rate of 6 °C/min, maintained for 1 min, increased to 230 °C at 2 °C/min, maintained for 2 min, and finally increased to 280 °C at a rate of 20 °C/min and maintained for 2 min. The mass spectrum was scanned in electron ionization mode (70 eV) (from 45 to 600) and then scanned in the selected ion monitoring mode. Sixteen PAHs were classified based on retention times and m/z values, quantified according to an internal standard peak area calibration, and the GC–MS was auto-tuned via perfluorotributylamine (Ma et al. 2020). Sixteen types of PAHs were analyzed in this study (Table S6). The methods of quality assurance and quality control for the PAH analysis have been described in detail in previous studies (S1) (Ma et al. 2018).

Calculation of PAH concentrations in pore water

The relative distribution of PAHs in the solid and liquid phases in the sedimentary environment can be used to predict their bioavailability, environmental changes, behavior, and toxic effects (Bucheli and Gustafsson 2000; Han et al. 2015). The concentration of ΣPAH16 in pore water reflects PAH pollution in lake water during different periods (Dueri et al. 2008).

The solid-water distribution coefficient (K_d) in sediments is usually estimated using the equilibrium distribution model of TOC and the solution. The calculation formula is as follows:

$$K_d = K_{TOC} \cdot f_{toc} \qquad (1)$$

where K_{TOC} represents the normalized distribution coefficient of TOC [(mol/kg organic carbon)/(mol/L solution)] and f_{toc} represents the ratio of TOC in solids (mass of organic carbon/mass of total solids). The formula for calculating the concentration of PAHs (C_w) in pore water is as follows:

$$C_w = C_s / K_d \quad (2)$$

where C_w represents the concentration of PAHs in the pore water and C_s represents the concentration of PAHs in the sediment. This model has been widely used in field samples (Hawthorne et al. 2010; Li et al. 2019; Sun et al. 2003). The octanol–water partition coefficient used in this study (that is, the normalized carbon partition coefficient) and the PAH toxicity values used for risk assessment are shown in Tables S1 and S3.

Statistical analysis

The sampling map of the Dianchi sedimentary column was constructed using ArcGIS 10.0. Correlation analysis was performed using the SPSS 20.0. Data processing and deposition records of economic parameters and PAH were performed using Microsoft Excel 2013 and OriginPro 9.0. Principal component analysis (PCA) was used for source apportionment. Socioeconomic data were obtained from the Yunnan Statistical Yearbook (2015). Because of the limitations of historical statistics, we only collected total population, GDP (gross domestic product), rural population, and urban population data from 1971 to 2014, and coal consumption data from 1975 to 2014.

Results

Historical sedimentary records of PAHs and their correlation with human activities

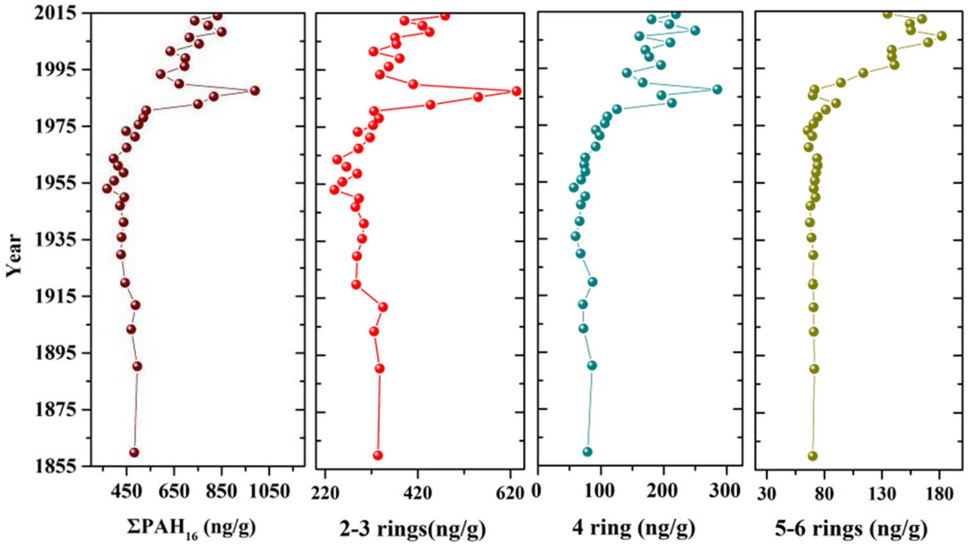
The vertical distributions of ΣPAH_{16} and 2–6 ring PAH concentrations in the sediments are shown in Fig. 2. The concentrations of ΣPAH_{16} in the sediments of Dianchi Lake range from 368 to 990 ng/g, with an average value of 572 ng/g (Fig. 2 and Table S2). In the profile (Fig. 2), the slight change at the bottom of the sediments before the mid-1970s may reflect background PAH values. The PAH content in sediments increased sharply from 1975 to 1988, peaking at 990 ng/g in 1988. Figure 3 shows a high correlation between ΣPAH_{16} and GDP ($R^2 = 0.89$, Fig. 3a) and ΣPAH_{16} and coal consumption ($R^2 = 0.76$, Fig. 3b) between 1975 and 1988. The rapidly increasing concentration of ΣPAH_{16} was consistent with the dramatic increase in GDP and coal consumption.

After 1988, as GDP and coal consumption continued to increase, the PAH concentrations showed a fluctuating downward trend. However, there was a high correlation between ΣPAH_{16} and coal% ($R^2 = 0.41$, Fig. 3c) from 1989 to 2014. As the proportion of coal decreased, the concentration of ΣPAH_{16} also fluctuated and decreased.

The concentrations of 2–3, 4, and 5–6 ring PAHs in the sediments were 240–634, 57–285, and 66–182 ng/g, respectively (Figs. 2, 4). Table 1 shows that 2–3 ring PAHs were significantly correlated with the total population, GDP, and urban population from 1971 to 2014, with correlation coefficients of 0.357, 0.368, and 0.392, respectively (with $P < 0.05$).

Furthermore, 4 ring PAHs were significantly correlated with the total population, GDP, and urban population from 1971 to 2014, with correlation coefficients of 0.439, 0.450, and 0.474, respectively, with $P < 0.01$ (Table 1). The correlation coefficient between 4 ring PAHs and coal consumption was 0.344 ($P < 0.01$) between 1975 and 2014 (Table 1). This indicates that population, GDP, and coal consumption are the main factors that affect 4 ring PAHs, which is consistent with previous research results (Guo et al. 2007; Hafner et al. 2005; Karina et al. 2014; Liu et al. 2012; Ma et al. 2021b;

Fig. 2 Historical trend of ΣPAH_{16} and 2–6 ring PAH concentrations in sediments of Dianchi Lake



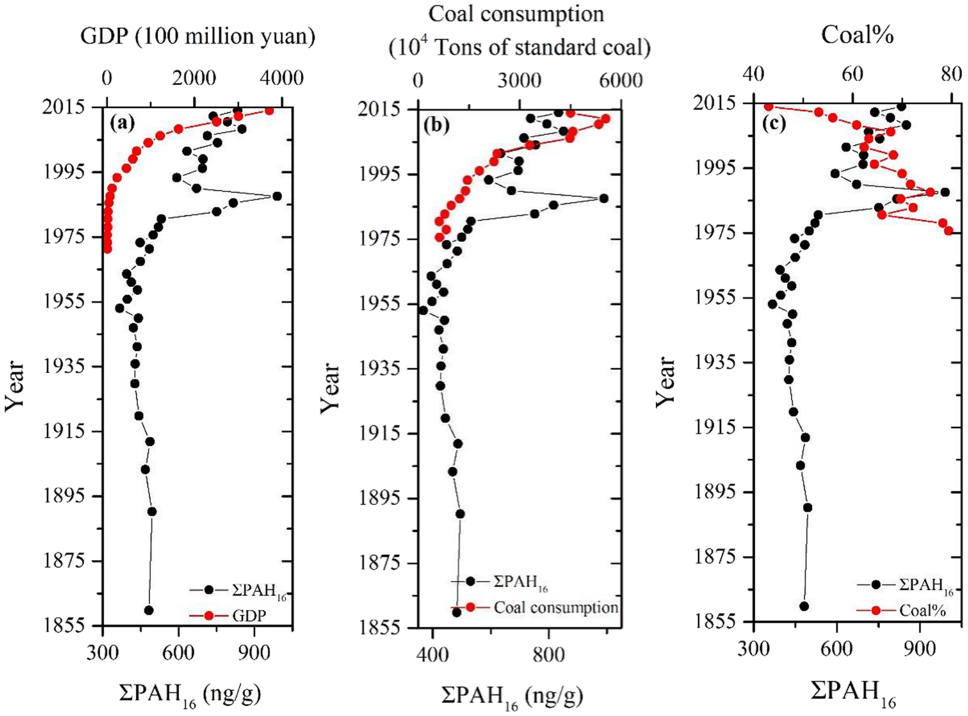
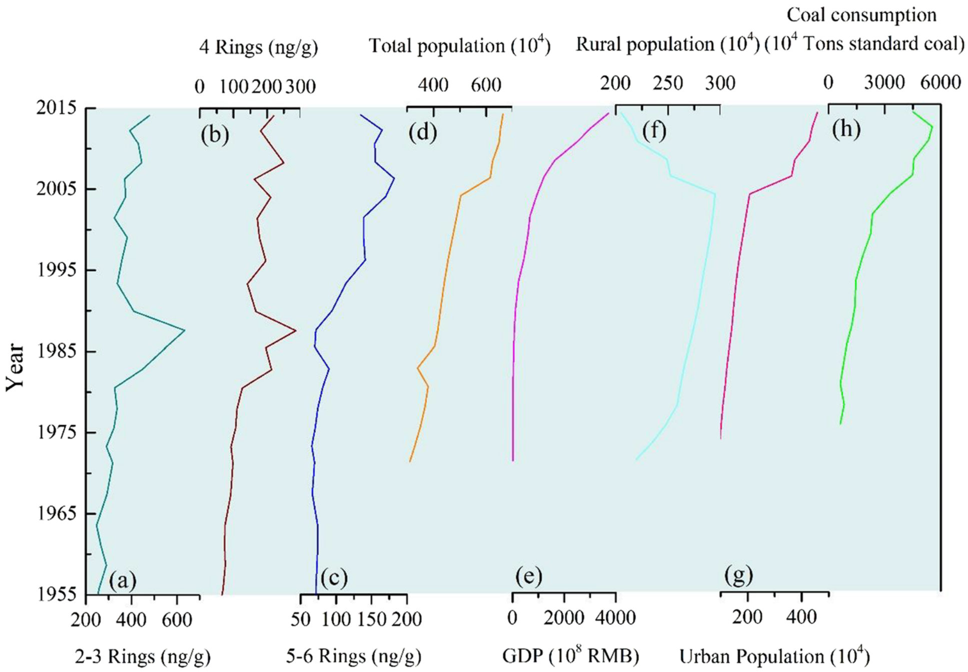


Fig. 3 Comparison of ΣPAH_{16} concentrations and economic parameters in the Dianchi Lake sediment core. **a** ΣPAH_{16} and GDP (gross domestic product) ($\Sigma PAH_{16} = 0.12 * GDP$ -48, $R^2 = 0.89$ (1975–1988)). **b** ΣPAH_{16} and coal consumption ($\Sigma PAH_{16} = 1.02 * coal + 142$, $R^2 = 0.76$ (1975–1988)). c ΣPAH_{16} and coal% ($\Sigma PAH_{16} =$ —$0.07 * coal\% + 114$, $R^2 = 0.41$ (1975–1988))

Fig. 4 Comparison of the concentrations of 2–6 ring PAHs and economic parameters to identify trends in the data: **a** 2–3 ring PAH concentrations, **b** 4 ring PAH concentrations, **c** 5–6 ring PAH concentrations, **d** total population, **e** gross domestic product (GDP), **f** rural population, **g** urban population, and **h** coal consumption



Zhang and Tao 2009). However, after 1988, with an increase in population, GDP, and coal consumption, the 2–3 and 4 ring PAHs decreased to a certain extent (Figs. 2 and 4).

Before 2005, the concentration of 5–6 ring PAHs continued to increase, but after 2005, the concentration of PAHs declined. Table 1 shows that 5–6 ring PAHs were

Table 1 Correlation coefficients between PAHs and human activities

	Total population	GDP	Rural Population	Urban population	Coal
2–3 rings	0.357*	0.368*	0.029	0.392*	0.191
4 rings	0.439**	0.450**	0.135	0.474**	0.344**
5–6 rings	0.661**	0.673**	0.193	0.696**	0.632**

**Correlation is significant at the 0.01 level (2-tailed). *Correlation is significant at the 0.05 level (2-tailed)

also significantly correlated with the total population, GDP, urban population, and coal consumption.

According to the correlation coefficients between PAHs and human activities, the correlation coefficients between population, GDP, and coal consumption and 2–3 ring LMW PAHs are smaller than the correlation coefficients with 4–6 ring HMW PAHs (Table 1). This indicates that from 1971 to 2014, population, GDP, and coal combustion had a greater impact on HMW PAHs than on LMW PAHs.

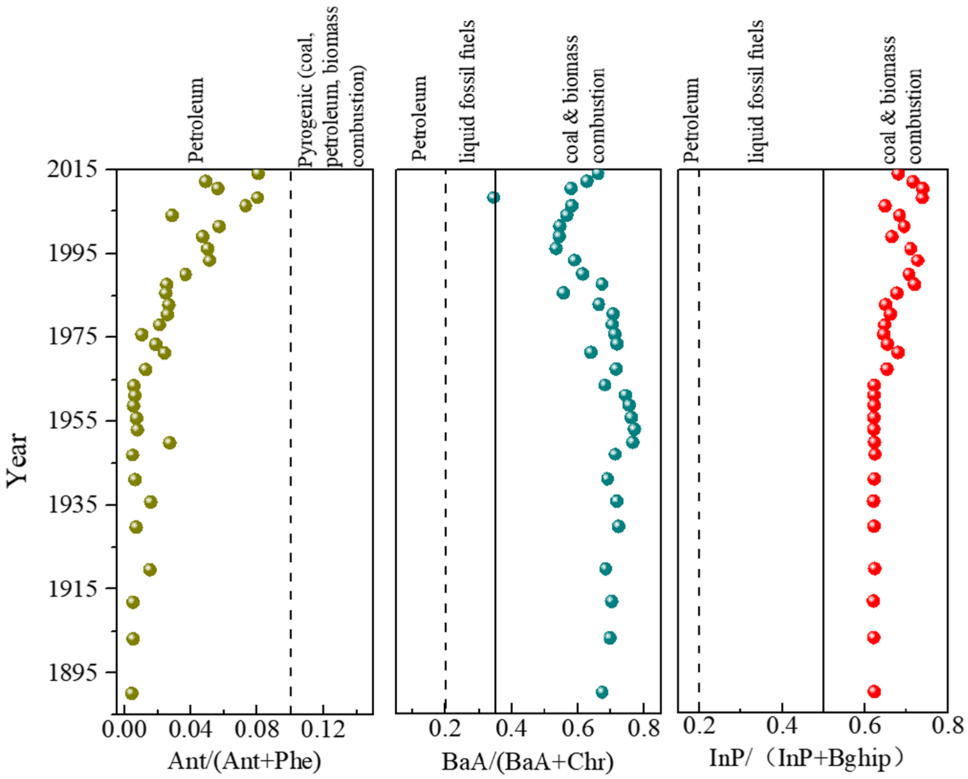
Source apportionment and identification of PAHs

Diagnostic ratios of PAHs

The diagnostic ratios of Ant/(Ant + Phe), BaA/(BaA + Chr), and InP/(InP + BghiP) were used to analyze the possible sources of PAHs in Dianchi Lake (Colombo et al. 2006; Guo et al. 2010; Han et al. 2021; Yim et al. 2005). Figure 5 shows that the ratio of BaA/(BaA + Chr) is mostly > 0.35, except for one year, which is between 0.2 and 0.35. This shows that PAHs in Dianchi Lake are mainly derived from coal and biomass combustion, and fossil fuel combustion occurs during several years. The ratios of InP/(InP + BghiP) were all > 0.5 (Fig. 5), which indicates that coal and biomass combustion are the main sources of PAHs in Dianchi Lake. Therefore, the ratios of BaA/(BaA + Chr) and InP/(InP + BghiP) suggest that coal and biomass combustion mainly contribute to the PAH concentrations in Dianchi Lake and fossil fuel combustion sources exist in individual years.

However, the ratios of Ant/(Ant + Phe) were all < 0.1, which suggest that the PAHs mainly originated from petroleum sources (Fig. 5). This difference in interpretation between Ant/(Ant + Phe), which suggests a petroleum source, and the ratios BaA/(BaA + Chr) and InP/(InP + BghiP), which suggest a coal and combustion source, has been observed in previous studies (Yan et al. 2005, 2006). This discrepancy is caused by differences in the environmental behavior of the Ant and Phe isomers (Hwang et al. 2003; Yan et al. 2005).

Fig. 5 The ratios Ant/(Ant + Phe), BaA/(BaA + Chr), and InP/(InP + BghiP) of PAHs in Dianchi Lake



Previous studies have shown that Ant has a higher photolytic ability than Phe (Ma et al. 2021b; Sanders et al. 1993). Therefore, the applicability of Ant/(Ant + Phe) in diagnosing PAH sources is questionable. This suggests that the ratios of BaA/(BaA + Chr) and InP/(InP + BghiP) are more reliable and show that the PAHs in Dianchi Lake are mainly derived from coal and biomass combustion, as well as fossil fuel combustion sources in individual years.

Principal component analysis for source apportionment

We used principal component analysis (PCA) to further analyze the source of PAHs in Dianchi Lake. The concentrations of the individual PAHs at each depth were used as the 16 variables in the PCA. Based on the loading of all 16 PAHs, three principal components (PC1, PC2, and PC3) were extracted (Fig. 6 and Table 2). PC1, PC2, and PC3 accounted for 53.14%, 14.87%, and 11.68% of the total variance, respectively (Tab. S4).

PC1 accounted for 53.14% of the total variance, of which Acy, Flo, Ant, Flu, Pyr, BaA, Chr, and BKF had the highest loading values (Tables 2 and S4). Acy and Flo are generally believed to be characteristic indicators of wood burning (Ravindra et al. 2008; Zhang et al. 2013), and Ant, Flu, Pry, BaA, Chr, and BKF are molecular indicators of coal burning (Duval and Friedlander 1981; Harrison et al. 1996; Li et al. 2003a; Wang et al. 2009a), indicating that PC1 may represent a source of wood and coal combustion.

PC2, which explained 14.87% of the total variance, was mainly composed of BbF, DBA, IcdP, and Bghip (Fig. 6 and Table 2). Previous studies have shown that BbF and BghiP are indicators of gasoline engine emissions (Chen et al. 2011; Motelay-Massei et al. 2007; Qian et al. 2016; Li et al. 2003a; Sofowote et al. 2008), and Inp and DBA have been identified as indicators of diesel emissions (Fang and Chang 2004; Li and Kamens 1993; Liu et al. 2017; Wang et al. 2009b, 2016; Yunker et al. 2002). Therefore, PC2 represents gasoline and diesel emissions originating from vehicle emissions.

PC3 accounted for 11.68% of the total variance, which was highly correlated with Phe (Fig. 6 and Table 2). Phe is a typical marker of coal combustion (Cao et al. 2016; Ramdahl 1983; Ravindra et al. 2008). Therefore, PC3 was identified as an indicator of coal combustion. According to the analyses above, PAHs in Dianchi Lake were mainly derived from the combustion of coal and wood, followed by gasoline and diesel emissions originating from vehicle emissions. This result is consistent with the results of the diagnostic ratio method.

Table 2 PCA analysis of PAHs in the sediments of Dianchi Lake

	Component		
	PC1	PC2	PC3
Nap	0.492	-0.087	0.342
Acy	**0.822**	0.113	0.217
Ace	0.000	-0.560	-0.455
Flo	**0.711**	0.492	-0.061
Phe	0.519	-0.089	**0.773**
Ant	**0.895**	0.096	0.027
Flu	**0.803**	-0.020	0.346
Pyr	**0.824**	-0.131	0.355
BaA	**0.910**	0.284	-0.247
Chr	**0.758**	0.336	-0.289
BbF	0.563	**0.547**	-0.273
BKF	**0.834**	0.213	-0.463
BaP	**0.662**	-0.137	-0.279
DBA	-0.762	**0.577**	0.176
IcdP	-0.434	**0.791**	0.149
Bghip	-0.825	**0.503**	0.129

Note: bold values are the high load values in each principal component and the main positive contributing factors of each principal component

Ecological risk assessment of PAH contamination

In this study, a risk assessment including seven PAHs with potential human carcinogenicity, namely, BaA, Chr, BbF, BkF, BaP, DBA, and InP, was conducted. The toxicity of BaP was

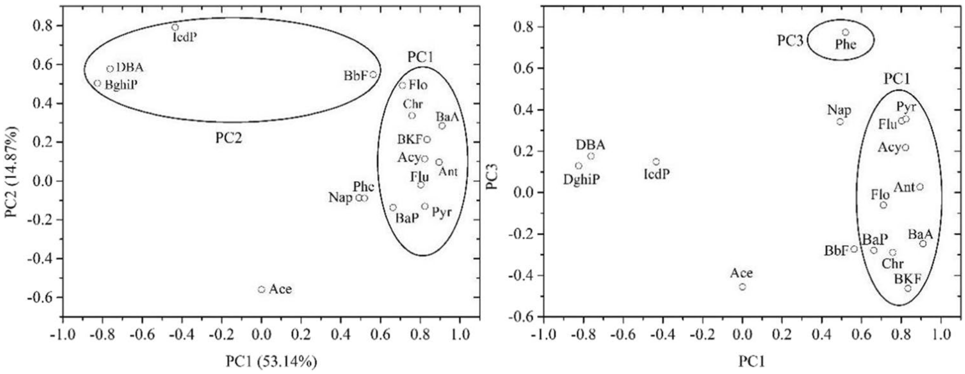


Fig. 6 Two-dimensional plot of the principal component loading of 16 PAHs from the Dianchi sediment core

used as the standard to quantify the toxic equivalent TEQ_{BaP} of the six other carcinogenic PAHs (Table S5) (Han et al. 2021; Qiao et al. 2006; Tsai et al. 2004). The concentration of TEQ_{BaP} in the sediments of Dianchi Lake was calculated and is shown in Table 3. Toxicity equivalent concentration ranges of BaA, Chr, BbF, BkF, BaP, InP, and DBA were 0.72–3.20 ng/g, 0.03–0.55 ng/g, 1.68–0.48 ng/g, 8.85–1.10 ng/g, 65.64–5.25 ng/g, 14.08–8.47 ng/g, and 1.64–1.04 ng/g, respectively, and the average values were 1.52 ng/g, 0.10 ng/g, 1.05 ng/g, 3.39 ng/g, 17.23 ng/g, 11.35 ng/g, and 1.42 ng/g, respectively. The toxicity equivalent concentration range of ∑7PAHs was 22.38–87.79 ng/g, and the average value was 36.05 ng/g. Compared with the global average TEQ_{BaP} concentration (804.94 ng/g) in sediments (Li et al. 2014; Sprovieri et al. 2007), the TEQ_{BaP} concentration in Dianchi Lake sediments was much lower than this toxicity level. The PAH concentrations in the sediments were all within a safe range (Liu et al. 2010; Li et al. 2019).

Pore water occurs as groundwater in the pores between sediment grains. Pore water can interact with sediments, and PAHs can be released or adsorbed from or onto the sediment. Therefore, the concentration of ΣPAH_{16} in sediment pore water can reflect the pollution of PAHs in lake water from different time periods to a certain extent (Arp et al. 2009; Chiou et al. 1981; Lücker et al. 2003; Yong et al. 2009). In this study, the risk threshold of PAHs in lake water was used to assess the risk of PAHs in the sediment pore water of Dianchi Lake (Neff et al. 2005). As shown in Fig. 7, the pollution level of 2–3 ring LMW PAHs has been within the safe range over the past 100 years. Only the concentration of Phe in some years has reached the chronic toxicity pollution level (55 μg/L).

Among the 4–6 ring HMW PAHs, most of the concentrations of Flu, Chr, and BbF were within the safe range before the 1980s. After the 1980s, their concentrations increased and reached chronic toxicity levels of 11, 2.2, and 2.9 ug/L, respectively. Most of the concentrations of Pyr, BaA, BkF, BaP, and BghiP reached their chronic toxicity levels of 12, 2.0, 1.7, 1.5, and 0.49 ug/L, respectively. Furthermore, the concentrations of BkF and BaP on the surface reached acute toxicity levels of 8.6 ug/L and 7.6 ug/L, respectively. DBA and InP are the two most polluting compounds. Over the past 100 years, all the samples had DBA and InP concentrations over acute toxicity levels of 1.3 and 0.64 ug/l, respectively.

Table 3 Toxicity equivalent concentrations of PAHs in the sediments of Dianchi Lake (TEQ_{Bap}) (ng/g)

	TEQ_{BaP}		
	Mix	Min	Mean
BaA	0.72	3.20	1.52
Chr	0.03	0.55	0.10
BbF	1.68	0.48	1.05
BkF	8.85	1.10	3.39
BaP	65.64	5.25	17.23
InP	14.08	8.47	11.35
DBA	1.64	1.04	1.42
∑7PAHs	22.38	87.79	36.05

Discussion

Different economic development models cause different growth characteristics of PAHs

With the founding of New China, especially with the reforms and opening policies implemented in 1978 (Ma et al. 2018), energy consumption caused by urbanization and industrialization increased rapidly. This may be the main reason for the rapid increase in PAH concentrations after 1975. However, the concentrations of PAHs fluctuated and declined after 1988. This may be because coal and biomass combustion are the main sources of PAH emissions in Yunnan Province (Xu et al. 2006), and while the proportion of total coal consumption has declined, the proportion of cleaner energy (oil and natural gas) has increased (Ma et al. 2020; Yunnan Statistical Yearbook 2015); therefore, PAH emissions were relatively reduced.

The population expansion brought about by rapid economic development has led to an increase in fuel for household cooking and basic heating (Guo et al. 2012; Ma et al. 2020), which may be the main reason for the increase in 2–3 ring PAH pollutants. Simultaneously, with economic development and population increase, industrialization and urbanization have led to an increase in fossil fuel combustion and a significant increase in motor vehicle emissions, leading to an increase in 4 and 5–6 ring HMW-PAH emissions (Ma et al. 2020, 2021b).

Previous studies have shown that household energy usage in Yunnan Province is dominated by coal and biomass, as biomass combustion is used for cooking and basic heating purposes (Ma et al. 2020; Zhang et al. 2007). With the rapid development of the society and economy, people's lifestyles have undergone significant changes, especially through substituting coal and biomass combustion for household needs with cleaner energy sources (Han et al. 2016; Liu et al. 2012; Tian and Chen 2017). In addition, the proportion of coal consumption in Yunnan has declined, while the proportion of cleaner energy (oil and natural gas consumption) has increased (Ma et al. 2020). These may be the main reasons for the decline in 2–3 and 4 ring PAHs. Furthermore, in recent years, improvements made to the vehicle emission standards in China (Ma et al. 2020; Tang et al. 2015) may have led to a decrease in the concentration of 5–6 ring PAHs.

Comparing PAH deposition records from lake sediments in other parts of China (Fig. 8), the concentrations of PAHs in the southern Dianchi Lake were higher than those in

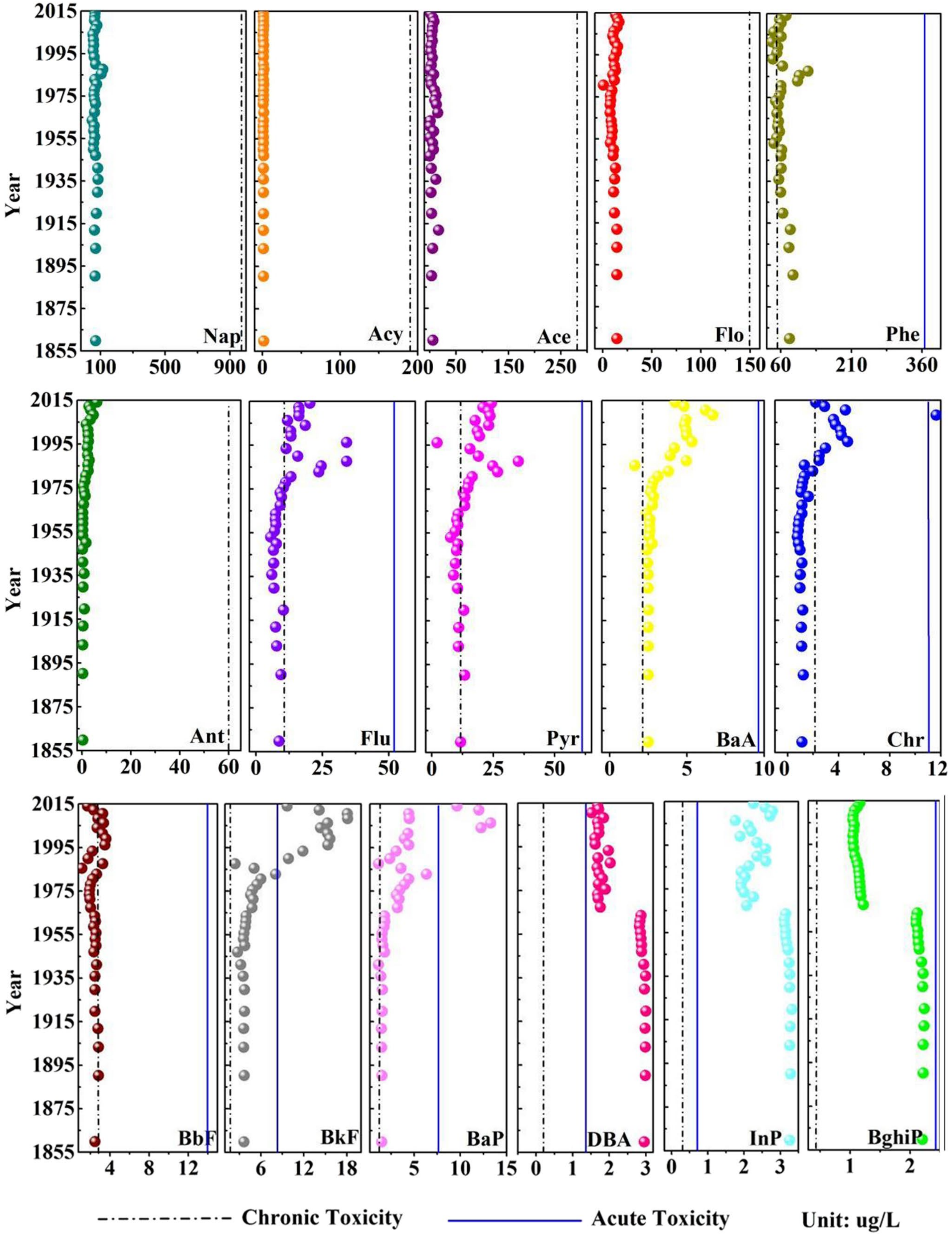
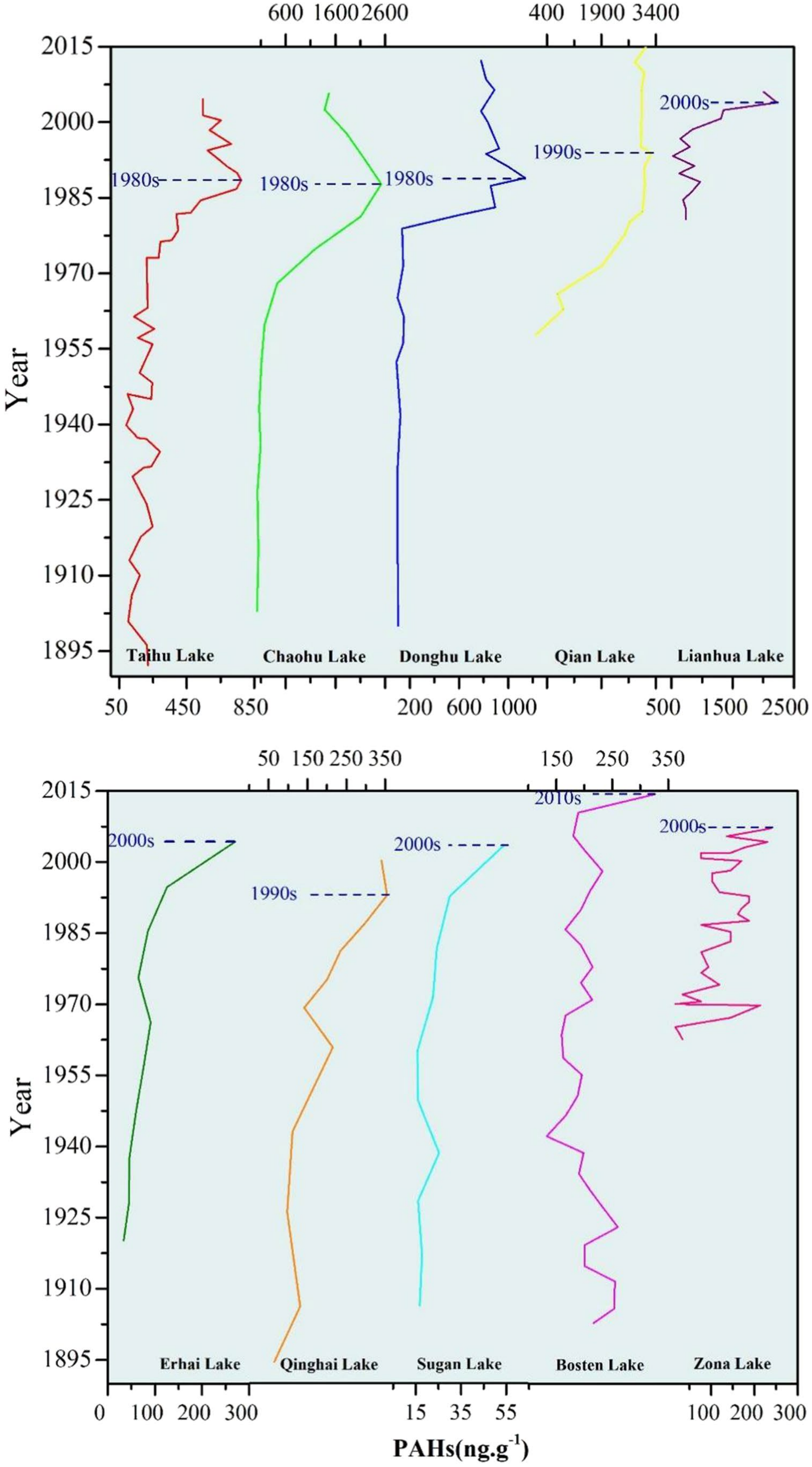


Fig. 7 Risk assessment of 16 PAHs in Dianchi Lake pore water

Fig. 8 Sediment records of PAHs in different lakes from China. Note: The above figures are referenced and modified from the literature (Zhang 2018; Li et al. 2016; Shanying Li 2016; Sun and Zang 2013; Guo et al. 2011; Wang et al. 2010; Guo et al. 2010; Yang et al. 2016)



western China and remote lakes on the Qinghai-Tibet Plateau, such as Erhai Lake (Guo et al. 2011), Qinghai Lake (Wang et al. 2010), Sugan Lake (Guo et al. 2010), Bosten Lake (Guo et al. 2010), and Cona Lake (Yang et al. 2016). However, they were lower than that of Chaohu Lake (Li et al. 2016), Donghu Lake, and Qianhu Lake (Li 2016) in the middle and lower reaches of the Yangtze River and lower than that of Lianhua Lake in northeast China (Sun and Zang 2013). The PAH concentrations in the southern part of Dianchi Lake is equivalent to that in Taihu Lake (Zhang 2018).

The peak times of PAHs in lake sediments from different regions of China era also different. The lakes in the middle and lower reaches of the Yangtze River have PAH peaks earlier, mainly between the 1980s and the 1990s. In contrast, Lianhua Lake in the northeastern region has a PAH peak in the 2000s. In western China (northwest and southwest) and remote areas of the Qinghai-Tibet Plateau, the peaks also appeared later, mainly between the 1990s and the 2000s. The PAH peak in the middle and lower reaches of the Yangtze River appeared earlier, which may be due to the rapid development of urbanization and industrialization in the region. Dianchi Lake is located near Kunming, the capital city of Yunnan Province, and its industrial development occurred relatively early, so the peak also appeared earlier. The late occurrence of PAH peaks in western China and remote areas of the Qinghai-Tibet Plateau may be due to their slower local economic and social development.

Compared with the PAH records from lake sediments in developed countries in Europe and the USA, the peak timing of PAH concentrations in Dianchi Lake was significantly later, and the concentration levels were also significantly lower than that of developed countries (Fig. 9). The PAH concentrations in lakes in developed countries gradually increased from around 1880, which may be consistent with the beginning of the industrial revolution (Furlong et al. 1987; Fernández et al. 2000). Pollution levels usually peaked in the 1950s and the 1980s (Kannan et al. 2005; Fernández et al. 2000). By the 1970s and 1980s, the PAH concentrations showed a declining trend, which may be related to the adjustment of the energy structure in developed countries in recent decades (Kannan et al. 2005), where coal is being replaced with cleaner fuels such as oil and natural gas (Gschwend and Hites 1981; Gevao et al. 1998).

Developed countries usually completed industrialization and urbanization before the 1980s. However, China was still in the developing stage at this time, especially after reforms and opening policies were implemented in 1978, at which point the rapid development of industrialization and urbanization began. Therefore, the different patterns in PAH concentrations as recorded in lake sediments between China and developed countries are consistent with their different development histories of industrialization and urbanization.

Energy utilization structure affects the source composition and pollution level of PAHs

The southern part of Dianchi Lake is mainly located in a rural and mountainous area. Biomass and coal combustion are the main energy sources for household cooking and heating in this area. Some studies have shown that the household energy utilization structure in Yunnan Province is dominated by coal and biomass (Xu et al. 2006; Zhang et al. 2007). In this study, the main sources of PAHs in the sediments of southern Dianchi Lake were coal combustion and biomass combustion, which also demonstrated the accuracy of the source analysis results.

According to the risk assessment of PAHs, most of the PAH concentrations in the sediments and pore water were within a safe range. However, the concentration of some 4–6 ring PAHs in the pore water reached a chronic toxicity pollution level, and some even reached the acute toxicity pollution level. This may be related to an increase in fossil fuel burning, such as coal and oil, caused by the development of industrialization and urbanization since the reform and opening up (Li et al. 2018; Ma et al. 2021b; Nemr et al. 2007; Wang et al. 2016).

Dianchi Lake is the largest freshwater lake in Yunnan Province and is an important source of water that supports human life and economic activities in Kunming. The ecological environment of Dianchi Lake has a greater impact on local residents. Therefore, it is necessary for the government to take more effective measures and increase investment to help control the environmental problems in Dianchi Lake. For example, central heating in residential areas can replace small coal stove heating. Briquettes should be selected for industrial use so that the coal can be fully burned. Clean energy should be developed, and natural gas should replace the use of coal and oil. In cities, the emissions of automobile exhaust are strictly controlled, and devices are installed to treat automobile exhaust. Implementing these measures for pollution control may reduce the input of PAHs to some degree in the lake.

Conclusion

The concentration of ΣPAH_{16} in the sediments of Dianchi Lake ranged from 368 to 990 ng/g, with an average value of 572 ng/g and a maximum value in 1988. The concentrations of 2–3, 4, and 5–6 ring PAHs in the sediments were 240–634, 57–285, and 66–182 ng/g, respectively. Population and GDP were the main influencing factors of 2–6 ring PAHs, and coal consumption was the main

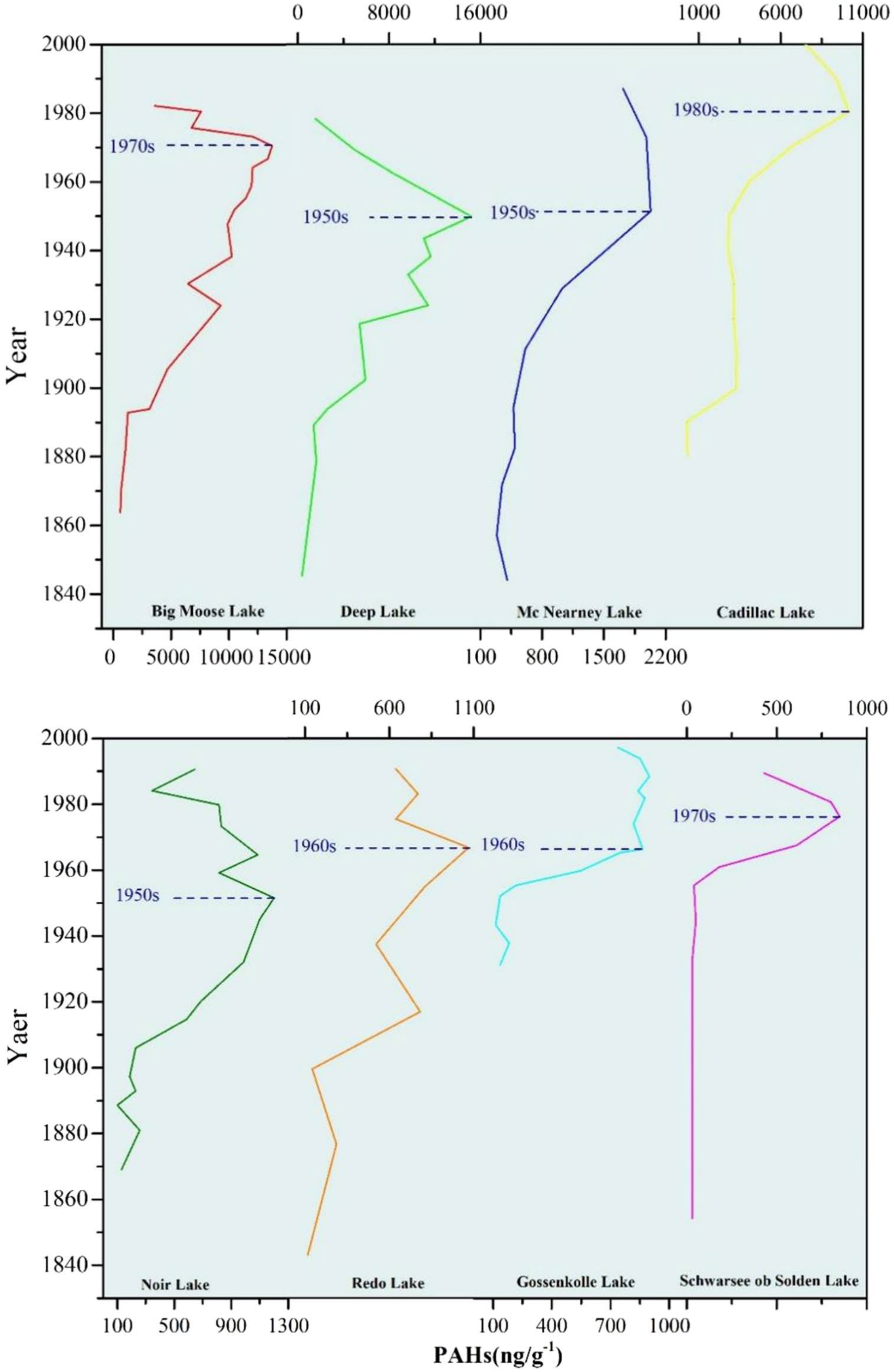


Fig. 9 Sediment records of PAHs in different lakes from different countries. Note: The above figures are referenced and modified from the literature (Furlong et al. 1987; Kannan et al. 2005; Fernández et al. 2000)

influencing factor for the 4–6 ring HMW PAHs. Moreover, population, GDP, and coal combustion have a greater impact on HMW PAHs than on LMW PAHs. The results of the diagnostic ratios and PCA model show that the main sources of PAHs were coal and biomass combustion, as well as fossil fuel combustion in a few individual years. PAH concentrations in the sediment were within a safe range. In the sediment pore water, 2–3 ring LMW PAHs were within a safe range for the past 100 years, except for Phe, which reached chronic toxicity pollution levels in some years. With the development of industrialization and urbanization, the burning of fossil fuels such as coal and petroleum has increased, and some 4–6 ring HMW PAHs have reached chronic toxicity or even acute toxicity in the sediment pore water.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s11356-022-18497-4.

Acknowledgements We sincerely thank Yaoxin Liu, Congcong Zhou, and Shuyan Cheng for their contributions to the experiments.

Author contributions X.H.M., T.H., and C.C.H. designed the experiments. X.H.M. and S.D.L. carried out the experiments and performed the analyses. X.H.M., H.Y., T.H., and C.C.H. contributed substantially to interpreting the results and writing the paper.

Funding This work was funded by the Postdoctoral Science Foundation of China [Grant numbers 2021M703299], the National Natural Science Foundation of China [Grant numbers 41773097, 41673108 and 41971286], the Youth Top Talent funded by Nanjing Normal University, and a project funded by the Priority Academic Program Development of Jiangsu Higher Education Institutions.

Data availability The research data are available on request: huangchangchun_aaa@163.com.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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