Research Article

Sedimentary Record of Polycyclic Aromatic Hydrocarbons from the Shuanglong Catchment, Southwest China

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Polycyclic aromatic hydrocarbons (PAHs) in the dated sediments from the Shuanglong catchment in the southwest of China were measured to characterize source inputs. The PAHs concentrations in the priority controlling list of US EPA (Σ PAHs) ranged within 102.47~563.24 ng g⁻¹, with an average value of 207.18 ng g⁻¹. The 2-3 rings PAHs were predominant, accounting for 73.77% of Σ PAH. According to the classification of pollution levels, Σ PAHs concentrations in the sediments are within the range of moderate pollution level. Both the total and individual PAH concentrations changed with the depth. The profile distribution of PAHs concentration in the sediments suggested that PAHs acted as an effective way to reconstruct the historical trends of socioeconomic changes in the study areas. Results of Ant/(Ant + Phe), Fla/(Fla + Pyr), and BaA/(BaA + Chry) show that petroleum and petrogenic matter are attributed to the potential pollution source in the Shuanglong catchment. The coal combustion and incomplete combustion of gasoline and fossil fuel are dominant. TOC contents had positive correlation with Σ PAHs ($R^2 = 0.72$, $P < 0.001$), 2-3 rings ($R^2 = 0.44$, $P < 0.001$), 4-ring ($R^2 = 0.78$, $P < 0.001$), and 5-6 rings ($R^2 = 0.62$, $P < 0.001$).

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are classical persistent organic pollutants (POPs). They are pervasive in the environmental medias, including atmosphere, soil, sediments, plants, and human bodies [1–4]. They have the characteristics of being carcinogenic, teratogenic, and mutagenic. The deposited PAHs may come from anthropogenic activities (burning processes of wood, coal and grass, incomplete combustion of fossil fuels, vehicle emissions, and leakage of crude oil) as well as natural sources (forest fires, volcanic eruptions, and biosynthesis) [5, 6]. As a result of their low solubility and hydrophobicity, PAHs can be easily absorbed by fine particulates once they get into the aquatic environment and accumulate in the sediments with deposition. Therefore, lake sediments and organic matter (OM) are considered as the main fate of PAHs [7, 8]. As a consequence, PAHs in the

sediments are primarily associated with OM which plays an essential role in affecting the distribution of PAHs [9, 10].

Due to their potential toxicity to living organisms, it is urgent to investigate the distributions and potential risks of PAHs. Plenty of studies have been carried out in the surface water and sediments of seas, oceans [1, 5, 11–16], and atmosphere [17–20], while the food investigations [17, 21] have also been conducted extensively worldwide. However, studies on the small catchment which were affected greatly by the human activities were deficient.

The source and distribution of PAHs are of great environmental significance. Owing to the complicated source of PAHs, difficulties in distinguishing the origins exist. In the past decades, growing attention has been paid to the PAHs environmental behavior. Various tools have been developed to identify the PAHs source [22–27]. Among all the approaches, the Ant/(Ant + Phe), Fla/(Fla + Pyr), BaA/(BaA + Chry), and

FIGURE 1: Location of sampling sites.

IcdP/(IcdP + BghiP) ratios (MDRs) have been applied extensively all over the world [5, 28–31]. Because isomers show substantial intrasource variability and intersource similarity, sources are difficult to be distinguished clearly. Criticism about these indexes exists [32–35]. Even the same sources are not always characterized by the same PAHs emissions profile, and, between different sources, there is significant overlapping in MDRs [36]. They do not respond to know differences in source strength and types between sites (on rural-urban gradients) in atmospheric emission sources [37]. Some researchers [38, 39] commented that use of MDRs for sewage sludge PAHs was unfeasible. Even if different sources-originated PAHs took place during the entire wastewater treatment process.

By dating sediment cores [40, 41] and discriminating the source of PAHs via diagnostic ratios, the time horizons associated with PAH trends, source variation, and quantification of flux into sediments can be determined. Most PAHs corresponded to the development of industrialization and urbanization. Thus, the profile distribution of PAHs in the sediments could elucidate the relationship between the historical trends of PAHs pollution and the socioeconomic development [42–45]. Many studies have focused on the temporal trends of PAHs in the sediments [16, 46]. However, investigations on the PAHs historical variation and effects of human activities in the small catchment turned out to be few.

The Dianchi watershed is located in the Yunnan province, southwest of China. Over the past decades, with the development of economic and society, the population grew rapidly and the chemical fertilizer was applied excessively, resulting in serious environmental problems [47, 48]. Recent studies in the Dianchi watershed focused mainly on Dianchi Lake, especially water eutrophication [49, 50]. However, the PAHs distribution and source identification in the small catchment of the Dianchi watershed are still poorly understood. In order to control the PAHs pollution in the Dianchi watershed, the sedimentary record of PAHs in the small catchment should be focused on. The purpose of this study were as follows: (1) to investigate the pollution level of PAHs in the Shuanglong

catchment; (2) to identify the sources of PAHs; (3) to explore the influence factors on PAHs distribution.

2. Materials and Methods

2.1. Study Area and Sampling. The Shuanglong Reservoir ($24^{\circ}57' \sim 24^{\circ}59' \text{N}$, $102^{\circ}55' \sim 102^{\circ}57' \text{E}$) was built in 1956, belonging to the Dianchi watershed (Figure 1). The Shuanglong catchment is located in the Shuanglong village on the west side of the Kunlun Highway, 70 km away from the Kunming city. The outflow enters from the south to north through the Dongda River into Dianchi Lake. The elevation is 1933.40 m. The rain collecting area is approximately 54 km^2 . The main dam of the reservoir is 18.50 m high, with four auxiliary dams. The total length is 795 m. As the important water source of Jinning County, the capacity of reservoir water storage is 12.16 million m^3 . In the small catchment, land use types include forest land, abandoned land, residential land, and cultivated land. Diversity of soil types such as red soil, purple soil, alluvial soil, limestone soil, and paddy soil exists. Because of the fertile soil, Jinning County has been regarded as the main producing areas of rice, broad beans, wheat, and rapeseed. Being rich in mineral resources and the well-developed mining industry make Jinning County one of the world's four major phosphorus mining zones. In addition, light industry, food, and building materials and other processing industries are developed.

The sampling time was in June, 2012. All the sampling sites were in the southern of the Shuanglong Reservoir (Figure 1). This region is under less disturbance. According to the principle of uniform distributed, three sediment cores (70 cm depth) were collected with self-weight coring equipment. The surface layers (2 cm depth) were taken away to minimize the perturbation of the surface deposited layer. The sediment core was sectioned in 1 cm intervals immediately. The subsamples were put into plastic bags and then transported to the laboratory instantly and frozen at the temperature of -50°C for 72 h.

2.2. Dating. The $^{210}\text{Pb}_{\text{ex}}$ radiometric technique was used with a high-resolution HPGe γ -spectrometry system (GWL-120-15 USA) (for details, see Wang et al., 2013) [51]. The constant CRS model was applied to date the sediment core [52].

2.3. Extraction. Subsamples were lyophilized using a freeze dryer (Eyela FDU-1200, Japan). All freeze-dried samples were ground and passed through the 200-mesh sieve. 200 ± 2 mg of subsample was weighed. A supersonic apparatus was used for lipid extraction with 50 ml of dichloromethane/hexane (1:1, v/v) solution for 1.5 h. Centrifuge for 3 times, moving the upper extraction liquid to the round-bottom flask. The extracts were then concentrated to approximately 2 ml by rotary evaporation. In order to remove the impurities and fractionate, the concentrated extracts went through the anhydrous sodium/silica columns (from bottom to the top, successively filled with 1.5 g anhydrous sodium, 1.0 g activated silica gel and 1.5 g anhydrous sodium) and then eluted with 15 ml mixture of dichloromethane/hexane (1:1, v/v) and 5 ml hexane to obtain the PAH fraction. Then eluent was collected in the bottle and concentrated to 0.5 ml by rotary evaporation.

2.4. Analysis Items. The total organic carbon (TOC) concentrations were determined by a TOC analyzer (CSHCN200, Japan). The subsamples were air-dried and ground via a 200-mesh sieve. Each subsample weighted 200 mg into two containers, respectively. One was calcined at 900°C for the measurement of total carbon (TC) concentration, and the other was added with phosphoric acid at 200°C to measure the inorganic carbon (IC) concentration. Triplicate analyses of every subsample were conducted.

The PAHs identification and quantification were performed through the gas chromatograph-mass spectrometer manufactured by Shimadzu Corporation of Japan (QP2010 ultra, Japan) operating in the electron ionization (EI) mode with a DB-5 fused silica capillary column (30 mm $\times$ 0.25 mm $\times$ 0.25 μm). Splitless injection of 1 μl of the subsamples is executed with an autosampler with a temperature of 250°C , and the high-purity helium (99.999%) is applied as carrier gas. The temperature of GC oven was programmed initially at the thermostatic of 90°C and increased to 180°C at the rate of $12^\circ\text{C min}^{-1}$, keeping this condition for 5 min, and then raised to 280°C at the rate of 5°C min^{-1} , staying for 10 min. The data was collected using a SCAN mode. The qualitative analysis of PAHs is the ratio between relative retention time and the peak intensity of the characteristic fragment ions. The peak area method with 6-point calibration curve was used for quantitative (SIM mode).

3. Results and Discussion

3.1. Chronology. The $^{210}\text{Pb}_{\text{ex}}$ concentrations in the sediment core ranged from 14.31 to 229.35 Bq kg^{-1} . The core covers the periods from 1867 to 2011, 144 years of deposition (Figure 2). The average rate of sedimentation was 0.49 cm year^{-1} , which was similar to the former research in the nearby area [48].

FIGURE 2: Fluctuation of sediment accumulation rate with time by CRS model.

3.2. TOC, TN, and TP Concentrations. The concentrations of TOC, total nitrogen (TN), and total phosphorus (TP) ranged within 0.49~4.35 g kg^{-1} (mean 1.13 g kg^{-1}), 0.50~4.74 g kg^{-1} (mean 1.89 g kg^{-1}), and 0.10~0.55 g kg^{-1} (mean 0.28 g kg^{-1}), respectively (Figure 3). The higher contents of TOC, TN, and TP were detected in the surface layers. Significant negative correlations between depth and TOC ($R = -0.77$, $P < 0.001$), TN ($R = -0.82$, $P < 0.001$), and TP ($R = -0.64$, $P < 0.001$) contents were found. The ratios of carbon to nitrogen (OC/N) were within the range of 0.43~1.20, with a mean value of 0.60, showing that the organic matter (OM) in the sediments was mainly from the phytoplankton and algae.

3.3. Characteristics of the PAHs Distribution. The concentrations of all 16 PAHs ($\sum\text{PAHs}$) in the sediments obtained from the Shuanglong catchment ranged from 102.47 to 563.24 ng g^{-1} , with a mean value of 207.18 ng g^{-1} (Figure 4). Among the 16 PAHs, the concentrations of less molecular weight (2-3 rings) (LPAHs) and high molecular weight (4-, 5-, and 6-rings) (HPAHs) varied from 79.82 to 334.19 ng g^{-1} (mean 152.82 ng g^{-1}) and 21.95 to 304.12 ng g^{-1} (mean 39.65 ng g^{-1}), respectively. The LPAHs were predominant, accounting for 47.42% to 88.84% (mean 73.77%) of the total PAHs. Naphthalene (Nap), in particular, constituted 27.30% to 62.42% (mean 36.27%) of the total PAHs. On the other hand, in the HPAHs, the proportion of 4-ring PAHs ranged from 3.60% to 15.00% (mean 14.47%), and 5- and 6-ring with a contribution of 4.86% to 32.33% (mean 11.76%).

Nap contents mainly indicate the source of crude oil and coal [64]. Fluorene (Flu) and Acenaphthene (Ace) are the dominant PAHs in the coke oven [27]. Phenanthrene (Phe), Fluoranthene (Fla), Pyrene (Pyr), and Anthracene (Ant) all usually acted as indicators for coal combustion [27, 65, 66]. The incomplete burning processes caused by low and medium temperature bring about Phe and Flu [67]. Oil and oil combustion have been regarded as the contributors of Benz[a]anthracene (BaA) and Chrysene (Chry) [66, 68]. Benzo[b]fluoranthene (BbF), Benzo[ghi]perylene (BghiP), and Indeno[1,2,3-cd]pyrene (IcdP) are mainly produced by automobile exhaust [65, 69].

The values of $\sum\text{PAHs}$ show a rising tendency on the whole. From the bottom (70 cm depth, year 1867) to the depth of 16 cm (year 1998), the concentration of $\sum\text{PAHs}$ shows volatile ups and downs. A peak value was obtained in

FIGURE 3: Vertical distribution of TOC, TN, TP, and C/N in the sediment.

the depth of 29 cm (year 1982). Because of the Reform and Open Policy implemented in 1978, China has been going a rapid industrialization and urbanization, accompanying the rapid development of Chinese economy. The concentration of Σ PAHs reached to the peak in the year 1982. The Σ PAHs sediment record during the 1983~1992 (from 28 to 21 cm) arise steadily. This is in accordance with the fact that the growing population in Yunnan and the expanding industries around the lake. In 1998 (16 cm depth), a large scale of forestry engineering began to be implemented by the government in the Yunnan province. Thus, the concentration of Σ PAHs decreased in that year. From the depth of 16 cm (year 1998) to 1 cm (year 2011), it continued to increase. This can attribute to rapid increase in the amount of civilian vehicles and consequent consumption of petrochemical products beginning in the 1990s. As for individual PAH, the temporal trends in the sediment core are shown in Figure 4. The variations of the Nap, Ace, and Acy concentrations are similar to the Σ PAHs. The Nap distribution shows an upward trend in volatility, representing the crude oil and coke oven for main source of PAHs. A positive shift in the Flu, Ace, and Phe values was observed since the 32 cm (year 1978), showing a rising tendency of incomplete combustion. The concentrations of Fla, Pyr, and Ant went up slightly from the bottom to the top, indicating the increased coal combusting input. BaA contents grew rapidly since 12 cm (year 2000). The vertical distribution characteristics of BaA and Chry exhibit the situation of industrial coal and oil burning contributing much more to the recent PAHs contents. BghiP and IcdP were only detected in the 1-2 cm depth (year 2010~2011) of the sediment, suggesting that the automotive emissions have an important impact on the origin of PAHs in recent years. What is more, this was further confirmed by the evidence of BbF concentration. The change of the PAH composition in the cores indicates that, throughout a very long period of time, the PAHs flowing

into Jinning County were dominated by the biomass burning and domestic coal combustion processes. In recent years, the ever increasing vehicular combustion emission has also contributed a portion of the PAH inputs.

3.4. Assessment of PAHs Pollution. Results of this study show relatively low concentration in the Shuanglong catchment. The pollution levels were in moderate. A comparison with other reports on Σ PAHs in the sediments from different areas are listed in Table 1. The PAH concentrations in the sediments found the same extent to Dongjiang catchment [55], Shilianghe catchment [56], and Baoxianghe catchment [57]. Results in this study appeared much lower than those in Shitou Koumen catchment [58], Yongding catchment [59], and Fenhe catchment [60]. And they were less polluted than Xidayang catchment [61] and Feitsui catchment [62]. However, the sampling sites represented a higher contamination than that in Xiaolangdi catchment [53] and Manwan catchment [54].

3.5. Identification of the PAHs Sources. The determination of PAHs sources is to decrease the input of PAHs to the environment. The deposited PAHs may come from natural and anthropogenic activities. The profile distributions are shown in Figure 5. The ratios of Ant/(Phe + Ant) were in the range of 0.03 to 0.64 with an average of 0.56, indicating PAHs were mainly derived from pyrogenic materials. These were further confirmed by the ratios of BaA/(BaA + Chry) ranging from 0.54 to 0.99 with an average of 0.77. It can be concluded that coal and biomass combustion were attributed to the major input of PAHs, which was anastomosed with the historical development in this area. Coal was the predominant energy resource and widely used by most of industries and residents in Jinning County. The discrimination of the

FIGURE 4: Continued.

FIGURE 4: Individual concentration in the sediment.

FIGURE 5: Ant/(Phe + Ant), Fla/(Fla + Pyr), and BaA/(BaA + Chry) ratios in the sediment.

TABLE 1: Comparisons of PAHs in different sediments in China.

Sampling site	Σ PAHs (ng g ⁻¹)	Pollution level ^a	References
Shuanglong Catchment	102.47~563.24	Moderate	This study
Xiaolangdi Catchment	98.10~329.00	Low-moderate	[53]
Manwan Catchment	14.40~137.70	Low-moderate	[54]
Dongjiang Catchment	85.00~658.00	Low-moderate	[55]
Shilianghe Catchment	17.37~839.55	Low-moderate	[56]
Baoxianghe Catchment	162.26~762.24	Moderate	[57]
Shitou Koumen Catchment	1294.51~2755.35	High	[58]
Yongding Catchment	1377.00~2855.00	High	[59]
Fenhe Catchment	539.00~6218.70	Moderate-very high	[60]
Xidayang Catchment	388.81~1205.56	Moderate-high	[61]
Feitsui Catchment	236.00~1197.00	Moderate-high	[62]

^aPollution levels (Baumard et al. 1998), scale of level of contamination: low: 0~100; moderate: 100~1000; high: 1000~5000; very high: >5000 (Baumard et al. 1998) [63].

origins of PAHs was also performed according to the values of Fla/(Fla + Pyr). Fla/(Fla + Pyr) ratio vary from 0.24 to 0.96, showing the mixed inputs of the petroleum and biomass combustion. This deduction was consistent with the former results (Section 3.3). From the depth of 56 cm to 46 cm (years 1934~1949), the Fla/(Fla + Pyr) ratio is within the range of 0.40 to 0.50. The average is 0.45, suggesting that the combustion of liquid fossil fuel and biomass were largely responsible for the input of PAHs. During the 1950s, the ratios of Ant/(Phe + Ant) increased sharply from 0.18 (46 cm depth) to 0.63 (42 cm depth). BaA/(BaA + Chry) ratios were from 0.63 to 0.74 in this period. For Fla/(Fla + Pyr) ratio, a peak value was obtained in the depth of 42 cm (year 1959), showing a strong contribution of coal and grass combustion. The peak-time period in the 1950s might correspond to the rapid development of the quality of life (including economy and population) in the First Five-Year Plan (1951~1955) after the founding of the China, while from the 1970s to 1980s, the values of Ant/(Phe + Ant), Fla/(Fla + Pyr), and BaA/(BaA + Chry) are much more than 0.1, 0.5, and 0.35, respectively. The strong contribution of coal, wood, and grass combustion were deduced. In 1978, the implementation of the "Reform and Open Policy" in China was performed. In particular, during the 2000s, the lower values of Fla/(Fla + Pyr) were found frequently. The liquid fossil fuel was gradually becoming dominant from that time. China's joining the World Trade Organization (WTO) in 2001 may contribute greatly. On the other hand, due to the development of tourism in the surrounding area, the vehicle emission flux is increasing. Thus the incomplete combustion of gasoline and fossil fuel became more and more predominant sources of PAHs year by year. The petrogenic sources may be interpreted to the scour of rainwater. The street dust which potentially contains petrogenic PAHs (spilled fuel, asphalt, and tire-wear materials) may enter into the water body and finally get into the sediments.

3.6. Effects of the Organic Matter Transportation on the PAHs Distribution. With respect to the relationship between the contents of PAHs and TOC, many researches had been conducted. It turned out to be a positive linear relationship between PAHs and TOC contents [1, 62]. The sediments are rich in organic matter (OM), which has a complex pore structure. The pore structure has a fast absorbed outer surface and a slowly absorbed inner surface, making it easily absorb the PAHs. As a result, OM became the storage depot of PAHs. PAHs migrate along with the sediment particles, so the TOC was affected significantly on the distribution of PAHs in the sediment core. The higher contents of TOC corresponded with the higher value of PAHs.

In this study, the relationship between the contents of TOC and PAHs are shown in Figure 6. TOC contents had positive correlation with $\sum$ PAHs ($R^2 = 0.72$, $P < 0.001$), 2-3 rings ($R^2 = 0.44$, $P < 0.001$), 4-ring ($R^2 = 0.78$, $P < 0.001$), and 5-6 rings ($R^2 = 0.62$, $P < 0.001$). HPAHs have the characteristics of being strongly lipophilic and hard-degraded. Therefore, HPAHs may be adsorbed by TOC easily. While LPAHs are relatively more volatile and soluble. Obviously, the

FIGURE 6: Relationship between TOC and PAHs concentrations.

correlation between TOC contents and higher PAHs contents is much higher than that of the lower ones. TOC contents played more contribution in controlling HPAHs than LPAHs.

4. Conclusions

In this study, the PAHs distributions, sources identification, and influence factors analysis in the Shuanglong catchment of the Dianchi watershed were investigated. Based on the results we obtained the following conclusions:

- (1) The $\sum$ PAHs concentrations ranged from 102.47 to 563.24 ng g⁻¹, reflecting a moderate pollution level. The $\sum$ PAHs compositions were dominated by 2-3 rings PAHs. As for individual PAH, Nap, Ace, Flu, BaA, and BaF were main components.
- (2) Based on the molecular indices (Ant/(Phe + Ant), Fla/(Fla + Pyr), and BaA/(BaA + Chry)), it was suggested that the sedimentary PAHs contents in the Shuanglong catchment were dominated by the coal combustion and petrogenic materials.
- (3) Significantly positive correlations between TOC and HPAHs indicated that TOC played a more important role in controlling the distributions of HPAHs than LPAHs in sediments of the Shuanglong catchment.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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