Deelaman *et al. J Environ Expo Assess* 2023;2:3
DOI: 10.20517/jeea.2022.29

Journal of Environmental Exposure Assessment

Research Article

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Ecological and health risks of polycyclic aromatic hydrocarbons in the sediment core of Phayao Lake, Thailand

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How to cite this article: Deelaman W, Choochuay C, Pongpiachan S, Han Y. Ecological and health risks of polycyclic aromatic hydrocarbons in the sediment core of Phayao Lake, Thailand. *J Environ Expo Assess* 2023;2:3. https://dx.doi.org/10.20517/jeea.2022.29

Received: 23 Aug 2022 **First Decision:** 29 Sep 2022 **Revised:** 24 Oct 2022 **Accepted:** 24 Nov 2022 **Published:** 1 Jan 2023

Academic Editors: Stuart Harrad, Suwanna Boontanon **Copy Editor:** Ying Han **Production Editor:** Ying Han

Abstract

Polycyclic aromatic hydrocarbons (PAHs) in sediment cores from Phayao Lake were investigated in terms of their concentration, distribution, and potential effects on the environment and human health. The concentration of Σ16PAHs ranged from 77.6 to 1251.1 ng g^{-1}. Additionally, low molecular weight PAHs (two or three fused aromatic rings) were dominant, indicating that the main source of PAHs is the incomplete combustion of organic sources. The toxic equivalent quantities (TEQs) of 16 PAHs varied from 2.67 to 155.09 ng g^{-1}, with an average of 38.29 ± 46.69 ng g^{-1}. Furthermore, the TEQs of 9 known carcinogenic PAHs averaged 20.77 ± 30.63 ng g^{-1}. B[a]P had the highest TEQ, followed by D[a,h]A and B[k]F, with values of 16.93 ± 25.49, 13.89 ± 15.37 and 2.12 ± 3.64, respectively. The $RQ_{(NCs)}$ of ΣPAHs ($RQ_{PAHs(NCs)}$) ranged from 1.65 × 10^{-4} to 6.27 × 10^{-1} with an average value of 1.18 × 10^{0} ± 1.52 × 10^{0}. Moreover, $RQ_{(NCs)}$ was less than 1 for individual PAHs, indicating a minor risk. However, $RQ_{(MPCs)}$ was less than 1 for individual PAHs, indicating a moderate risk. The results showed a significant incremental lifetime cancer risk (ILCR) from sediment-bound PAHs with dermal absorption, followed by ingestion

and inhalation, as the prevalent route of exposure. The measured ILCR values of all PAHs were less than 10^{-6} for both adults and children, which were lower than the baseline value, indicating that each PAH poses a low risk of cancer to humans. This study provides information for control and preventive actions to limit future PAH pollution in Phayao Lake.

Keywords: Polycyclic aromatic hydrocarbons, phayao lake, Thailand, ecological risk assessment, health risk assessment

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are persistent organic pollutants (POPs) with two or more benzene rings in their chemical structure[1-3]. The main origin of PAHs is the incomplete combustion of organic compounds and fossil fuels through human activities, such as transportation and industrial facilities[4-7]. The majority of PAHs are insoluble and long-lasting in the environment. Both direct and indirect exposure of living creatures to PAHs results in accumulation and transmission. According to previous studies, some of the increased cases of cancer may be linked to environmental exposure and/or chemical absorption of PAHs through the food chain[3,8,9]. PAHs are hazardous to health and the environment[10], potentially causing cancer (carcinogen)[11,12] and inducing mutations (mutagen) or embryo defects. Moreover, PAHs have the potential to be teratogenic in both humans and animals[13].

PAHs have the potential to spread and accumulate in lakes and rivers[14,15]. Wind and rivers carry sediments and other particles to lakes. PAHs adsorbed on small solid particles (aerosols) in dust in the atmosphere move from their primary sources and can either fall into lakes or combine with precipitation[16,17]. Water buffers sediments and particles that have settled to the bottom of water bodies from wind and other physical forces. As a result, sediments are maintained in lakes on a very fine scale. PAHs can also leach from the ground, such as wastewater from oil refinery plants, lubricant spills from industrial plants, water from agricultural fields, and water from communities, and then be carried into lakes through adsorption to soil particles[18-21]. Because PAHs are discharged from industrial, municipal, and natural surface run-offs around lakes, lake sediments serve as a significant reservoir of pollutants. The degree of PAH contamination and the main PAH source in lake sediments have been evaluated. In particular, nations that are more developed are more vulnerable to the impacts of PAH pollution arising from different industrial activities. Yuan *et al.* obtained sediment samples from Yangzong Lake, China at a depth of 20 cm for a study on the genesis and history of PAHs[22]. They found that between 1950 and 1990, there were 200.7 to 1913.7 ng g^{-1} of PAHs, the majority of which were produced by the burning of petroleum.

The bioactivity of PAHs has been investigated. For example, benzo[a]pyrene (B[a]P) is physiologically activated by enzymes to produce carcinogenic and mutagenic epoxides[23]. Sayer discovered that ellagic acid, which is abundant in green plants, degrades the diol epoxide form of B[a]P, reducing its potential to induce cancer or mutations[24]. PAHs have been identified in the following environments: lake water[25], marine air, soil/sediment, snow, mosses, lichen, krill, penguin, and skua blood. An exhaustive investigation into the presence of POPs in biotic and abiotic components has been conducted on national and international scales[26]. PAHs in sediment may be volatilized into the atmosphere and adsorbed by aquatic animals as well as aquatic crops through roots and leaves. The accumulation of PAHs in humans and other species is therefore possible if food chains are involved[27]. As a result, monitoring the concentration of PAHs in sediments is critical to understand the course of PAHs through the ecosystem.

Phayao Lake, the largest freshwater lake in northern Thailand, is a significant water supply for human consumption. Furthermore, Phayao Lake supports a large number of aquatic species. Hence, lake sediments commonly contain fossils, including fossils of microscopic creatures that live in the water. The presence or absence of these fossils can reveal information about the water and lake, e.g., lake depth, water temperature, and whether the water is salty or fresh. The sediments in cores, such as sand and clay, can reveal when the lake depth decreases (due to a drier environment) or increases (during times of a wetter climate). All the information assists in determining the settings and climates in which ancestors lived.

Because of the ecological importance and high socioeconomic value of Phayao Lake, investigations of the causes, trends, concentrations, distributions, and toxicity of pollutants in the sediment are required to evaluate the effects of oil exploration, transportation, waste disposal, industrial operations, and pollution in the area. Previous investigations of dated sediment cores from Phayao Lake (covering the past ~150 years) revealed that the greatest flow and composition changes of PAHs follow local industrialization. The concentrations of Σ23PAHs, namely all parent and alkyl PAHs (excluding perylene, which has a predominantly *in situ* biogenic diagenesis origin), low molecular weight (LMW, 2-3 ring)-PAHs, and high molecular weight (HMW, 4 ring)-PAHs, were low and fluctuated minimally until ~1980s. PAH concentrations and fluxes grew until ~1996 and then declined but remained higher than those before ~1980, reflecting the emission history of the Mae Moh power plant[17]. The atmosphere in Chiang-Mai province, located in northern Thailand and close to Phayao Lake, was analyzed during 2017-2018, revealing the presence of 19 PAHs: acenaphthene (Ace), fluorene (Fl), phenanthrene (Phe), anthracene (An), fluoranthene (Fluo), pyrene (Pyr), benzo[a]anthracene (B[a]A), chrysene (Chry), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F), benzo[a]fluoranthene (B[a]F), benzo[e]pyrene (B[e]P), benzo[a]pyrene (B[a]P), perylene (Per), indeno[1,2,3-cd]pyrene (Ind), benzo[g,h,i]perylene (B[g,h,i]P), dibenz[a,h]anthracene (D[a,h]A), coronene (Cor), and dibenzo[a,e]pyrene (D[a,e]P). According to this study, during 2017-2018, the concentration of total PAHs was 2.361 ± 2.154 mg m^{-3}, and the important sources of PAHs in northern Thailand were vehicular exhaust, biomass burning (BB), and diesel emissions[28]. Therefore, there is a scarcity of health risk assessments of Phayao Lake sediments. Han *et al.* determined the concentrations and fluxes of BC, char, soot, and PAHs in aged sediment cores obtained along the lake[17]. Nevertheless, no research has been conducted to assess the ecological and health risks of PAHs. Therefore, in this study, 81 cm long sediment cores from Phayao Lake were analyzed to determine the vertical trends, distributions, profiles, and toxicity equivalent concentrations (TEQs) of PAHs. This is essential for risk evaluations and hazardous action plans used to protect human health and the environment.

EXPERIMENTAL

Sampling and methods

Sampling site

Phayao Lake is the largest lake in northern Thailand and the fourth-largest lake in Thailand. It is centrally located in the city of Phayao, with mountains in the background. It is formed by water flowing from 18 local streams, with an average annual water volume of 29.40 million m^3. It covers 20.5 km^2 of land. Additionally, it serves as a spawning habitat for many different fish species. Phayao Lake's surrounding landscape makes it a natural attraction to visitors[29]. Phayao Lake is exposed to a variety of local and regional pollution as well as transboundary haze pollution, including urban run-off, industrial waste, vehicular emissions, and biomass emissions, such as the cooking of food and burning of agricultural waste[17]. A gravity corer (Uwitec, Austria) was used to obtain sediment cores from the southern part of the lake (N 19°9.8', E 99°53.5'). All pollutants from the upper part of the lake are accumulated at the sample site. Therefore, the collected sediments should contain all PAHs in Phayao Lake that eventually pass through a small stream, as shown in Figure 1. A gravity corer (Uwitec, Austria) was used to obtain sediment core from the southern

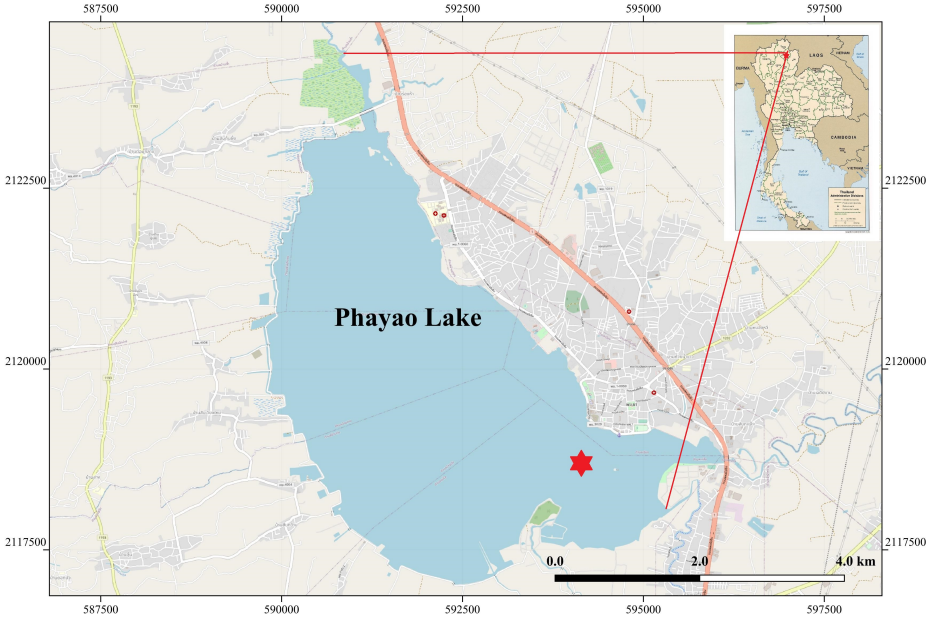

Figure 1. Location of sampling site in Phayao Lake, Thailand.

part of the lake. The length of the core was 81 cm, which was continuously sliced from the top to the bottom to give a total of 25 samples (n = 25). All samples were transported to the laboratory, freeze-dried, and stored there until chemical analysis at a temperature of -20 °C.

Chemical analysis of PAHs

Extraction and analysis of the sediment core (PY) followed the method by Han *et al.*[17]. Briefly, 2 g of sediment was spiked with internal standards (mixture of 7 deuterated PAHs and 2 deuterated OPAHs) and then extracted using an ultrasonic technique with organic solvents. The extracts were then cleaned and fractionated using column chromatography. For identification and quantification of PAHs, the final extract was fed into a gas chromatography coupled to a mass spectrometer (GC/MS, Agilent, Santa Clara, CA, USA). On the dry sediment mass basis, the concentration of each target component was calculated. Quality assurance/quality control (QA/QC) analyses followed the method by Han *et al.*[17]. Briefly, internal standards were used to quantify the component concentrations in the samples, which were then adjusted by subtracting the mean blank values. Spike and recovery studies were used to comprehensively evaluate the analytical method's accuracy and precision.

Ecotoxicological risk and health risk assessment

Toxicity equivalent concentration

In this study, all samples were subjected to health risk assessments for both children and adults using the Toxicity equivalent concentration (TEQ) formula shown in Equation 1. This formula is used to calculate the health risk from exposure to the carcinogen in various substances, which has been compared with the health risk of exposure to the toxin B[a]P[30].

$$TEQ = \sum [C_i \times TEFs] \quad (1)$$

where C_i is the concentration of PAH and TEFs is the toxic equivalency factor used to compare the toxicity of each PAH relative to B[a]P in accordance with the United States Environmental Protection Agency (USEPA) standards[30].

*TEF_s: B[a]P and D[a,h]A was 1; B[a]A, B[b]F, B[k]F, and Ind was 0.1; An, Chry, and B[g,h,i]P was 0.01; and the last group containing Phe, Fluo, and Pyr was 0.001.

Ecological risk assessment of PAHs in sediments

In the environment, PAHs can be found in various ecosystems, particularly in sediments and water[30,31]. Therefore, it is very important to monitor and assess PAHs in the ecosystem, especially in sediments, because they are major sources of many pollutants released into lakes and water resources. The risk of PAHs in the sediment escaping to the environment was evaluated using a risk quotient (RQ). The unfavorable impacts of environmental pollution on ecosystems can be evaluated by measuring the ratio of the concentration of specific pollutants to the corresponding concentrations of pollutants in the medium, as shown in Equation 2[27,30]. Hence, risk assessment using the RQ formula was first applied to evaluate the risk of the sediment to the ecosystem.

$$RQ = \frac{C_{PAHs}}{C_{QV}} \quad (2)$$

where C_{PAHs} and C_{QV} are the concentration of different PAHs and the corresponding level of PAHs in the sediments, respectively.

Second, the negligible (NC) and maximum (MPC) concentrations of PAHs in sediments were calculated using Equations 3 and 4[32]. In this study, we used RQ_{NCs} and RQ_{MPCs} for the calculations, as shown below:

$$RQ_{NCs} = \frac{C_{PAHs}}{C_{QV\,(NCs)}} \quad (3)$$

$$RQ_{MPCs} = \frac{C_{PAHs}}{C_{QV\,(MPCs)}} \quad (4)$$

where $C_{QV(NCs)}$ is the quality value of the NCs for PAHs and $C_{QV(MPCs)}$ is the quality value of the MPCs for PAHs.

Finally, an ecological risk of only nine PAHs, namely naphthalene, anthracene, phenanthrene, fluoranthene, benzo[a]anthracene, chrysene, benzo[k]fluoranthene, benzo[a]pyrene, and benzo[g,h,i]perylene, was evaluated in accordance with USEPA standards for each PAH.

Additionally, the technique by Cao *et al.* was used to determine the ecological risk of PAHs in the ecosystem[33]. The concern that the ecological risk of specific PAHs is disregarded can be addressed with this method. The following definitions apply to $RQ_{\Sigma PAHs}$, $RQ_{\Sigma PAHs(NCs)}$, and $RQ_{\Sigma PAHs(MPCs)}$:

$$RQ_{\sum PAHs} = \sum_{i=1}^{n} RQ_i, \ RQ_i \geq 1 \quad (5)$$

$$RQ_{\sum PAHs\,(NCs)} = \sum_{i=1}^{n} RQ_{i(NCs)}, \ RQ_i \geq 1 \quad (6)$$

$$RQ_{\sum PAHs\,(MPCs)} = \sum_{i=1}^{n} RQ_{i(MPCs)}, \ RQ_i \geq 1 \quad (7)$$

In this study, the ecological risk assessment of certain PAHs was conducted by referring to studies conducted by Cao *et al.*, Wang *et al.*, and Sun *et al.*[33,27,31]. To determine the $RQ_{PAHs(NCs)}$ and $RQ_{PAHs(MPCs)}$ values of individual PAH, the values of $RQ_{(NCs)}$ and $RQ_{(MPCs)}$ that were not less than 1 were added. According to this metric, each PAH may not be of significant concern if $RQ_{(NCs)}$ is less than 1. There is a high chance of contamination from a single PAH if $RQ_{(MPCs)}$ is greater than 1. If $RQ_{(NCs)}$ is larger than 1 and $RQ_{(MPCs)}$ is less than 1, contamination by specific PAHs poses a moderate risk.

Health risk assessment

In accordance with the Human Health Evaluation Manual[34], the incremental lifetime cancer risk (ILCR) was used to quantify the health risk from exposure to environmental PAHs[35]. The population in this study was separated into two groups according to the demarcation between the ages of children and adults. The models used to analyze the ILCRs account for the three major exposure paths: ingestion, skin contact, and inhalation[36]. PAHs are related to the food chain, such as when humans cook aquatic animals for human consumption. Additionally, people in the area can breathe in the pollutants released from the sediment into the air. ILCR was calculated using Equations 8, 9, and 10:

$$ILCR_{Ingestion} = \frac{CS \times \left(CSF_{ingestion} \times \sqrt[3]{\frac{BW}{70}} \right) \times IR_{ingestion} \times EF \times ED}{BW \times AT \times 10^6} \quad (8)$$

$$ILCR_{Dermal} = \frac{CS \times \left(CSF_{Dermal} \times \sqrt[3]{\frac{BW}{70}} \right) \times SA \times AF \times ABS \times EF \times ED}{BW \times AT \times 10^6} \quad (9)$$

$$ILCR_{Inhalation} = \frac{CS \times \left(CSF_{Inhalation} \times \sqrt[3]{\frac{BW}{70}} \right) \times IR_{inhalation} \times EF \times ED}{BW \times AT \times PEF} \quad (10)$$

where CS represents the converted PAH concentration based on TEQ measurements, ABS is the dermal adsorption fraction (0.13 for children and adult)[37], AF is the dermal adherence factor (0.2 mg cm^{-2} h^{-1} for children and 0.07 mg cm^{-2} h^{-1} for adult)[37], AT is the average life span (25,550 days for children and adult)[38], BW is the body weight (15 kg for children and 70 for adult)[34], CSF_{Dermal} is the dermal cancer slope factor of B[a]P (25 kg d mg^{-1} for children and adult)[38], $CSF_{Ingestion}$ is the ingestion cancer slope factor of B[a]P (7.3 kg d mg^{-1} for children and adult)[38], $CSF_{Inhalation}$ is the inhalation cancer slope factor of B[a]P (3.85 kg d mg^{-1} for children and adult)[38], ED is the exposure duration (6 years for children and 20 years for adult)[39], EF is the exposure frequency (350 d $years^{-1}$ for children and adult)[39], $IR_{Ingestion}$ is the ingestion rate (200 mg d^{-1} for children and 100 mg d^{-1} for adult)[37], $IR_{Inhalation}$ is the inhalation rate (10 m^3 d^{-1} for children and 20 m^3 d^{-1} for adult)[38], PEF is the particle emission factor (1.36×10^9 m^3 kg^{-1} for children and adult)[37], and SA is the dermal exposure area (2800 cm^2 for children and 5700 cm^2 for adult)[37].

RESULTS AND DISCUSSION

The concentration and character of PAHs in the sediment core

The concentrations of 16 different PAHs identified in the sediment core from Phayao Lake, namely Nap, Acy, Ace, Flu, Phe, An, Fluo, Pyr, B[a]A, Chry, B[b]F, B[k]F, B[a]P, B[g,h,i]P, Ind, and D[ah]A, and Σ16PAHs were evaluated, as shown in Figure 2. Individual PAHs are presented in the following order, from lowest to highest concentrationas as follows: An < B[a]A < D[a,h]A < Ace < B[g,h,i]P < Ind < Pyr < B[a]P < Chry < B[b]F < Ace < B[k]F < Flu < Fluo < Phe < Nap. The results showed that LMW-PAHs (two or three fused aromatic rings) were dominant in Phayao Lake, indicating that the main source of PAHs is the incomplete combustion of organic sources. The primary sources of PAHs in the environment are pyrogenic, petrogenic, and biological matter, which discharge PAHs into the environment through pyrogenic processes[40].

We also measured the concentrations of 16 PAHs by depth (see Supplementary Table 1), revealing that the concentration of Σ16PAHs was highest at a depth of 34 cm (1251.10 ± 30.55 ng g^{-1}). The PAH levels in the sediment from Phayao Lake at a depth of 34 cm were relatively high compared with those in other samples from other nations (high levels: 1000 to 5000 ng g^{-1})[41]. The depth distribution of PAHs in the sediment core from Phayao Lake showed that LMW-PAHs were dominant at every depth (see Figure 3), indicating that the main source of PAHs is the incomplete combustion of organic sources, including pyrogenic, petrogenic, and biological matter, and thus PAHs are discharged into Phayao Lake through pyrogenic processes[40].

The amounts of various PAHs contained in sediment are categorized as follows: low levels: 0 to 100 ng g^{-1}, moderate levels: 100 to 1000 ng g^{-1}, high levels: 1000 to 5000 ng g^{-1}, and extremely high levels: > 5000 ng g^{-1}[41]. The amounts of Σ16PAHs in world sediment (WS) and Phayao Lake sediment are contrasted in Table 1. The content of all PAHs in WS ranges from 13.7 to 213,000 ng g^{-1}, whereas the concentration of Σ16PAHs in Phayao Lake sediment ranges from 77.6 to 1251.1 ng g^{-1}. Therefore, the levels of PAHs in the sediment from Phayao Lake are moderate compared with those in samples from other countries. Phayao Lake is located in the city of Phayao, which is subjected to a variety of local and regional pollution sources, such as urban run-off, industrial waste, automobile emissions, and biomass emissions, including those from human activities. According to a previous study, in the years after 1980, the levels of LMW-PAHs and HMW-PAHs in Phayao Lake were occasionally high, indicating that BB emissions are a significant source of pollutants released to the environs of Phayao Lake and possibly to most rural parts of Thailand[17]. Additionally, this study demonstrated that elevated local inputs into lake sediments may have contributed to the peak concentration of PAHs.

Ecological Risks of PAHs in the sediment

PAHs that accumulate in sediments might be used by benthic creatures, such as phytoplanktons and zooplanktons, which would then enter the food chain. Therefore, the aquatic ecology may be at risk from PAHs found in sediment samples. An effective approach for assessing the ecological risk of PAHs to organisms and the ecosystem is an ecological risk assessment[52]. The risk quotient (RQ) was developed by Kalf *et al.* to measure the risk of organic compounds[32]. The proposed approach was used to evaluate the ecological risk posed by samples of contaminated silt collected from Phayao Lake.

According to the contamination levels used by Maliszewska-Kordybach[53] to classify soil pollution, soil is considered not contaminated at levels below 200 ng g^{-1}, slightly contaminated at levels between 200 and 600 ng g^{-1}, contaminated at levels between 600 and 1000 ng g^{-1}, and substantially contaminated at levels over 1000 ng g^{-1}. The classification standards (such as 200, 600, and 1000 ng g^{-1}) were created by calculating the risk of PAH exposure to humans and measuring the PAH concentration in European soils. Because the

Table 1. Comparison of polycyclic aromatic hydrocarbons (PAH) concentrations in sediments from different areas in the world

Sampling sites	Nature of sample	Number of PAHs	Concentration (ng g^{-1})	References
Phayao Lake, Thailand	Core sediment	16	77.65-1251.10	This study
Samsun Coast, Turkey	Sediment (~5 cm)	16	71.64-1186.87	[42]
Han River, China	Sediment	16	137.1-1478.4	[43]
Black Sea	Sediment	16	28.47-444.36	[44]
Gorgan Bay, Iran	Surface sediments	16	13.70-23.68	[45]
Escravos River Basin, Nigeria	Surface sediments	16	750-213,000	[46]
Pearl River Estuary, China	Surface sediment	16	73.68-933.25	[47]
Peninsular, Malaysia	Surface sediments (top 5 cm layer)	16	57.7-19,300	[48]
Burrard Inlet, Canada	Sediment	15	20-6840	[49]
Bering Sea	Surface sediments	16	49.84-65.38	[50]
Canadian Basin	Surface sediments	16	27.66-167.48	[50]
Chukchi Sea	Surface sediments	16	52.40-91.25	[50]
Nzhelele River, South Africa	Surface sediments	16	206-13,710	[51]

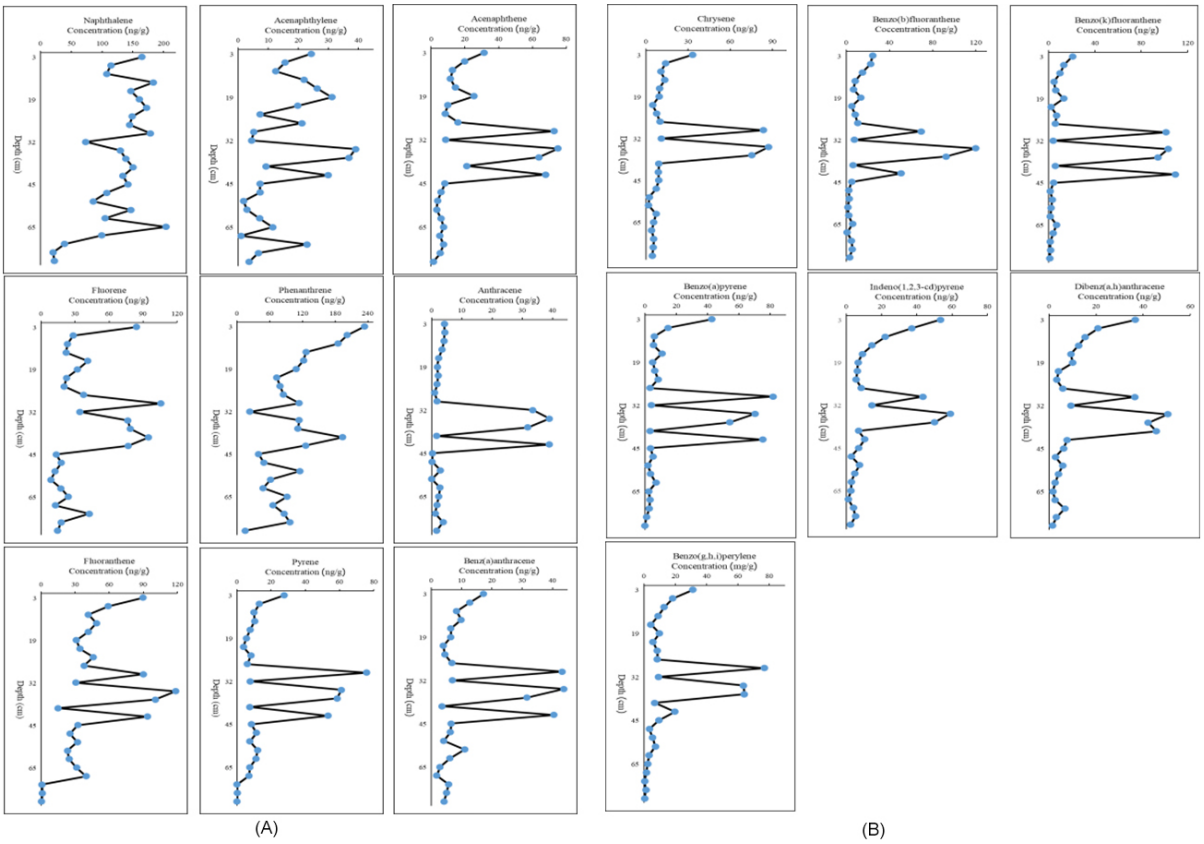

Figure 2. (A) Vertical profiles of the concentrations (ng g^{-1}) of different Polycyclic aromatic hydrocarbons (PAH) congeners and Σ16PAHs in the sediment core collected from Phayao Lake, Thailand; (B) Vertical profiles of the concentrations (ng g^{-1}) of different PAH congeners and Σ16PAHs in the sediment core collected from Phayao Lake, Thailand.

contents of 16 PAHs in the sediment collected from Phayao Lake ranged from 2.67 to 155.09 ng g^{-1}, the sampling location was considered "non-contaminated." Toxic equivalent quantities (TEQs) and risk quotients (RQs) were employed in this investigation to provide more accurate results, as shown in Tables 2 and 3.

Table 2. Toxic equivalent quantities (TEQs) of polycyclic aromatic hydrocarbons (PAHs) in sediment cores collected from Phayao Lake, Thailand

PAHs	TEFs (US-EPA)	Mean	Stdev	Range
Naphthalene	0.001	0.12	0.05	0.02-0.20
Acenaphthylene	0.001	0.02	0.01	ND-0.04
Acenaphthene	0.001	0.02	0.02	ND-0.08
Fluorene	0.001	0.04	0.03	0.01-0.11
Phenanthrene	0.001	0.10	0.06	0.02-0.23
Anthracene	0.01	0.08	0.13	ND-0.39
Fluoranthene	0.001	0.04	0.03	ND-0.12
Pyrene	0.001	0.02	0.02	ND-0.08
Benz[a]anthracene	0.1	1.20	1.29	0.18-4.38
Chrysene	0.001	0.02	0.03	ND-0.09
Benzo[b]fluoranthene	0.1	1.98	3.07	0.09-11.99
Benzo[k]fluoranthene	0.1	2.12	3.64	0.10-10.94
Benzo[a]pyrene	1	16.93	25.49	ND-81.92
Indeno[1,2,3-cd]pyrene	0.1	1.56	1.78	0.11-5.90
Dibenz[a,h]anthracene	1	13.89	15.37	1.49-50.47
Benzo[g,h,i]perylene	0.01	0.15	0.21	0.01-0.77
Σ16PAHs		38.29	46.69	2.67-155.09
Σ9PAHs		20.77	30.63	0.58-97.65

Table 3. Risk classification for different polycyclic aromatic hydrocarbons (PAHs) and ΣPAHs

ΣPAHs			Individual PAHs		
ΣPAHs	**$RQ_{(NCs)}$**	**$RQ_{(MPCs)}$**	**Individual PAHs**	**$RQ_{(NCs)}$**	**$RQ_{(MPCs)}$**
Risk-free	0		Risk-free	0	
Low-risk	≥ 1; < 800	0	Moderate-risk	≥ 1	< 1
Moderate-$risk_1$	≥ 800	0	High-risk		≥ 1
Moderate-$risk_2$	< 800	≥ 1			
High-risk	≥ 800	≥ 1			

Moderate-$risk_1$ and Moderate-$risk_2$ are described about risk classification for the range of ΣPAHs, which already shown it in the Table 3.

To calculate the toxic equivalency factors (TEFs), the toxic equivalent quantities (TEQs) of 16 PAHs were calculated. In Phayao Lake, the TEQs of 16 PAHs varied from 2.67 to 155.09 ng g^{-1}, with an average of 38.29 ± 46.69 ng g^{-1}. Of the 16 PAHs, 9 are known carcinogens, and the TEQs of these 9 PAHs averaged 20.77 ± 30.63 ng g^{-1}. The results showed that B[a]P had the highest TEQ, followed by D[a,h]A and B[k]F, with values of 16.93 ± 25.49, 13.89 ± 15.37, and 2.12 ± 3.64, respectively. The acceptable TEQ value of the 9 carcinogenic PAHs in soil is 600 ng g^{-1}, according to the Canadian soil quality recommendations set by the Council of Canadian Environment Ministers (CCME) for the conservation of the environment and human health[27,50,54]. All the TEQs determined in this research were below the Canadian soil quality guidelines.

If $RQ_{(NCs)}$ < 1 and $RQ_{(MPCs)}$ < 1 for certain PAHs, then the risk may be of minimal concern. However, if $RQ_{(NCs)}$ ≥ 1 and $RQ_{(MPCs)}$ < 1, then the risk is at a moderate level, and it may be essential to exercise control or implement remedial actions. Importantly, $RQ_{(MPCs)}$ ≥ 1 indicates a high-risk situation that necessitates quick action[31,32]. Categorization of the ecological risk of PAHs based on $RQ_{(NCs)}$ and $RQ_{(MPCs)}$ is shown in Table 3. The results of this study showed that $RQ_{(NCs)}$ < 1 and $RQ_{(MPCs)}$ < 1 for individual PAHs, indicating that the risk may be of minimal concern.

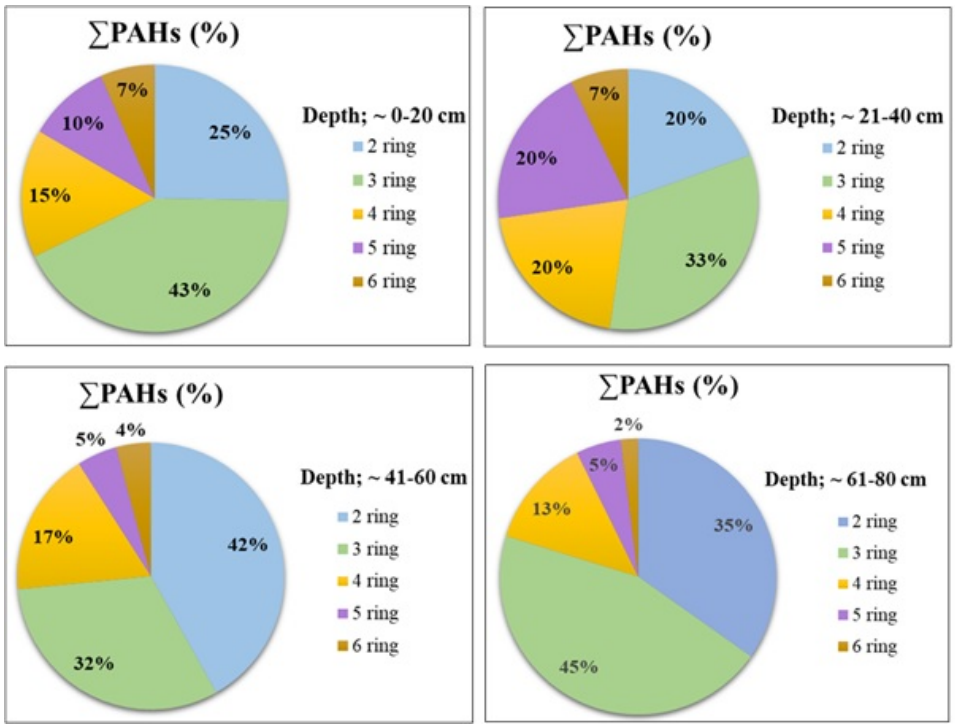

Figure 3. Depth distribution of Polycyclic aromatic hydrocarbons (PAH) in the sediment core from Phayao Lake, Thailand.

Table 4 and Figure 4 show how much each PAH increased the ecological risk of the sediment in Phayao Lake.The $RQ_{(NCs)}$ of ΣPAHs($RQ_{\Sigma PAHs(NCs)}$) ranged from 1.65×10^{-4} to 6.27×10^{-1} with an average value of $1.18 \times 10^{0} \pm 1.52 \times 10^{0}$ (Σ9PAHs), indicating a low risk to the environment, plants, and living organisms. All the values obtained in this study were lower than the values determined using samples from China ($RQ_{\Sigma PAHs(NCs)} > 800$). These samples, which were studied in China, indicated that total PAHs present a significant level of ecological concern owing to petroleum pollution[32]. However, although the hazards are minimal, residents in the northern region should take precautions because Lake Phayao is a significant water source for consumption as well as a tourist attraction. As a result, the findings of this study might provide information for control and preventive actions to limit future PAH pollution in Phayao Lake.

Health risk assessment of PAH contamination

It is well known that PAHs are found in the food chain because humans cook aquatic animals for consumption. Additionally, people in the area can breathe in the pollutants released from the sediment into the air. Therefore, ILCR was used in this study to determine the potential health risks for people exposed to PAHs released from the sediment in Phayao Lake. Investigation of the health risks included three exposure pathways: inhalation ($ILCR_{Inhalation}$), dermal contact ($ILCR_{Dermal}$), and direct ingestion ($ILCR_{Ingestion}$). Children and adults were divided into two age groups. Supplementary Table 2 contains statistical parameters of all ILCRs from sediment in Phayao Lake. The results revealed that the mean values of $ILCR_{inhalation}$ of all samples for children and adults were $7.62 \times 10^{-13} \pm 1.02 \times 10^{-12}$ and $8.48 \times 10^{-12} \pm 1.14 \times 10^{-11}$, respectively. The average values of $ILCR_{Dermal}$ were $2.28 \times 10^{-7} \pm 3.06 \times 10^{-7}$ for children and $1.94 \times 10^{-7} \pm 2.60 \times 10^{-7}$ for adults. Moreover, the mean value of $ILCR_{Ingestion}$ were $1.83 \times 10^{-7} \pm 2.45 \times 10^{-7}$ for children and $1.09 \times 10^{-7} \pm 1.46 \times 10^{-7}$ for adults, which were lower than the reference values, as shown in Figure 5. These findings indicated that dermal contact, followed by ingestion and inhalation, was the primary route of exposure to PAH in the sediment. These results agreed with those of previous studies in that analyses of samples collected from other Asian countries, such as India, Korea, and China, showed large increases in cancer risk as a result of soil-bound

PAHs, with dermal absorption identified as the main route of exposure, followed by ingestion and inhalation[55-57].

Table 4. Statistical parameters of $RQ_{(NCs)}$ and $RQ_{(MPCs)}$ of PAHs in sediments from Phayao Lake, Thailand

PAHs	NCs	MPCs	$RQ_{(NCs)}$			$RQ_{(MPCs)}$		
			Range	Mean	Stdev	Range	Mean	Stdev
Naphthalene	1.4	140	1.49×10^{-2}-1.46×10^{-1}	8.93×10^{-2}	3.44×10^{-2}	1.49×10^{-4}-1.46×10^{-3}	8.93×10^{-4}	3.44×10^{-4}
Acenaphthene	1.2	120	1.14×10^{-3}-6.26×10^{-2}	1.71×10^{-2}	1.93×10^{-2}	1.14×10^{-5}-6.26×10^{-4}	1.71×10^{-4}	1.93×10^{-4}
Phenanthrene	5.1	510	3.05×10^{-3}-4.59×10^{-2}	2.02×10^{-2}	1.08×10^{-2}	3.05×10^{-5}-4.59×10^{-4}	2.02×10^{-4}	1.08×10^{-4}
Fluoranthene	26	2600	1.88×10^{-5}-4.56×10^{-3}	1.68×10^{-3}	1.22×10^{-3}	1.88×10^{-7}-4.56×10^{-5}	1.68×10^{-5}	1.22×10^{-5}
Benz[a]anthracene	3.6	360	5.09×10^{-2}-1.22×10^{0}	3.34×10^{-1}	3.60×10^{-1}	5.09×10^{-4}-1.22×10^{-2}	3.34×10^{-3}	3.60×10^{-3}
Chrysene	107	10700	8.17×10^{-4}-1.81×10^{-5}	1.65×10^{-4}	2.35×10^{-4}	1.81×10^{-7}-8.17×10^{-6}	1.65×10^{-6}	2.35×10^{-6}
Benzo[k]fluoranthene	24	2400	4.03×10^{-3}-4.56×10^{-1}	8.83×10^{-2}	1.52×10^{-1}	4.03×10^{-5}-4.56×10^{-3}	8.83×10^{-4}	1.52×10^{-3}
Benzo[a]pyrene	27	2700	ND-3.03×10^{0}	6.27×10^{-1}	9.44×10^{-1}	ND-3.03×10^{-2}	6.27×10^{-3}	9.44×10^{-3}
Benzo[g,h,i]perylene	75	7500	7.80×10^{-5}-1.02×10^{-2}	2.06×10^{-3}	2.81×10^{-3}	7.80×10^{-7}-1.02×10^{-4}	2.06×10^{-5}	2.81×10^{-5}
Σ9PAHs				1.18×10^{0}	1.52×10^{0}		1.18×10^{-2}	1.52×10^{-2}

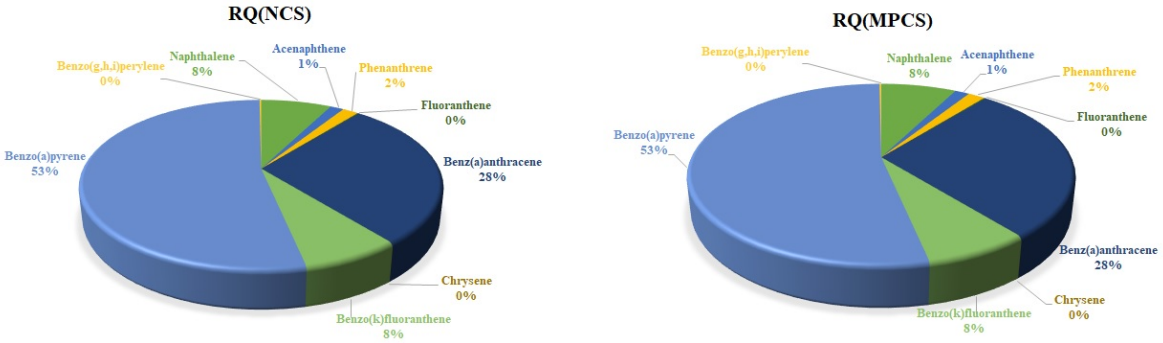

Figure 4. Ecological risks based on $RQ_{(NCs)}$ and $RQ_{(MPCs)}$ for specific Depth distribution of Polycyclic aromatic hydrocarbons (PAH) congeners in sediment samples from Phayao Lake as a percentage contribution.

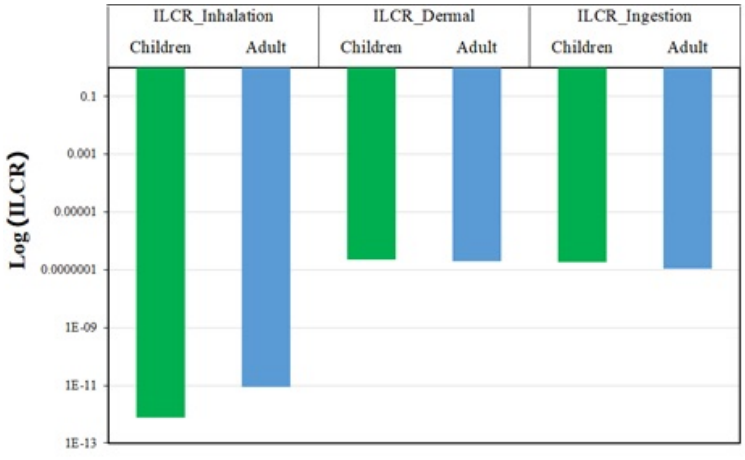

Figure 5. Contributions of different exposure pathways to incremental lifetime cancer risk (ILCR).

The ILCR values of each of the investigated PAHs were less than 10^{-6} for both groups of humans (children and adults), which were lower than the baseline value[38,39], indicating that each PAH poses a minimal risk of cancer to humans. A previous study presented values of the hazard index (HI) for the non-carcinogenic effects of PAHs on humans: 1.68×10^{-6} for adults and 1.10×10^{-5} for children[58]. HI values greater than 1.0 constitute a serious health concern. Nap and Ant has the greatest and least contribution to non-carcinogenic effects on humans, with HI values of 5.87×10^{-7} for Nap and 2.08×10^{-8} for Ant. Therefore, our results showed that the cumulative lifetime cancer risk for both children and adults from exposure to PAHs in sediments from Phayao Lake was less than 10^{-6}, which was below the baseline set for safe levels of PAHs.

CONCLUSIONS

Sediment cores from Phayao Lake were used in this work to examine the vertical trends, distributions, profiles, and hazardous ratios of PAHs. The results showed that LMW-PAHs (two or three fused aromatic rings) were dominant, and the depth distribution of PAHs in sediment cores showed that LMW-PAHs were dominant at every depth, indicating that the main source of PAHs is the incomplete combustion of organic sources, including pyrogenic, petrogenic, and biological matter. Therefore, PAHs are discharged into Phayao Lake through pyrogenic processes. In the present study, RQ(NCs) < 1 and RQ(MPCs) < 1 for individual PAHs, indicating that the risk may be of minimal concern. However, it may be necessary to exert control or take corrective action. $RQ_{(NCs)}$ of ΣPAHs ($RQ_{\Sigma PAHs(NCs)}$) ranged from 1.65×10^{-4} to 6.27×10^{-1} with an average value of $1.18 \times 10^{0} \pm 1.52 \times 10^{0}$ (Σ9PAHs), indicating a low risk to the environment, plants, and living organisms. All the values obtained in this study were lower than the values determined for samples from China ($RQ_{\Sigma PAHs(NCs)} > 800$). These findings demonstrate that total PAHs present a significant level of ecological concern owing to petroleum pollution. Furthermore, the results showed the sediment-bound PAHs with the most prevalent exposure routes in the order of dermal absorption > ingestion > inhalation. The ILCR values of the investigated PAHs were less than 10^{-6} for children and adults, which were lower than the baseline value, indicating that each PAH poses a minimal risk of cancer to humans.

DECLARATIONS

Acknowledgements

The authors acknowledge the Institute of Earth Environment, Chinese Academy of Sciences, Xian, China for supporting this study.

Authors' contributions

Made significant contributions to the study's conception and design as well as the data processing and interpretation: Han Y, Choochuay C

Conceptualization, writing - original draft preparation, review and editing, visualization: Deelaman W, Pongpiachan S

Availability of data and materials

Additional data and information can be made available at request from the corresponding author.

Financial support and sponsorship

The financial support was provided by the NSFC (41625015 and 41991250), the Strategic 398 Priority Research Program of Chinese Academy of Sciences (XDB40000000), and the 399 CAS (QYZDY-SSW-DQC001 and ZDBS-SSW-DQC001).

Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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