



## Seasonal variation and concentration of PAHs in Lake Balaton sediment: A study on molecular weight distribution and sources of pollution

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### ABSTRACT

The temporal and spatial variations of 16 Polycyclic Aromatic Hydrocarbons (PAHs) were examined at multiple sites around Lake Balaton from February 2023 to January 2024. The results indicated that the concentrations of PAHs in sediment were high during the winter months, 448.35 to 619.77 ng/g dry weight, and low during the summer months, 257.21 to 465.49 ng/g dry weight. The concentration of high molecular weight PAHs (HMWPAHs), consisting of 5–6 rings, was greater than that of low molecular weight PAHs (LMWPAHs), which had 2–3 rings. The total incremental lifetime cancer risk (ILCR) for both dermal and ingestion pathways was high for both adults and children during the four seasons, with the highest records as the following: winter > spring > summer > autumn. The ecological effects of the 16 PAHs were negligible except for acenaphthylene (Acy) and fluorene (Fl), which displayed slightly higher concentrations during the autumn and spring, respectively.

### 1. Introduction

PAHs are a class of complex organic chemicals consisting of two or more benzene ring structures (Świt et al., 2023). Scientist worldwide have been more concerned about PAHs compounds due to their difficult degradation, harmful effects on ecosystems, tendency to accumulate in organs, and ability to cause cancer (Zhi et al., 2015). Sixteen PAHs compounds were classified as priority compounds by the United States Environmental Protection Agency (USEPA) due to their potential toxicity (USEPA, 1994). The possible sources of these compounds include but are not limited to traffic emissions, fossil fuel combustion, and biomass pyrolysis. Most water bodies, including lakes, rivers, estuaries, and seas, were found to have a considerable level of PAHs (Yu

et al., 2015). These compounds tend to build up in sediments after reaching their destination. Then, they could be accumulated in the organisms or released back into the water, disrupting the concentration equilibrium between aqueous and solid phases (Huang et al., 2023). Lake sediment has been noticed as the medium for indicating historical human activities, such as industrialization, population increase, agricultural activity, and energy consumption in the last century (Zhang et al., 2024). Ma et al. (2020) demonstrated that changes in the energy structure, namely the occurrence of fossil fuel spills as well as incomplete burning of fossil fuels and biomass, were primary factors that led to elevating the level of PAHs in Dianchi Lake watershed (Ma et al., 2020). The surface sediments could also act as indicators of pollution and can participate in the PAHs contamination as secondary sources (Zhu and Li,

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2014). Thus, it is crucial to examine the potential sources as well as PAHs contributors in sediments to better plan control and management of PAH pollution in the aquatic ecosystem.

Worldwide, lakes cover about half of the total water on the Earth's surface and about 49.8 % of the freshwater (Bhateria and Jain, 2016). In addition to acting as excellent hosts for different species, lakes can also manage floods and intercept pollutants from different land sources. Moreover, the optimal setting of lake ecosystems could lead to significant economic and social benefits for human civilization (Bhateria and Jain, 2016). However, human activities such as urbanization and industrialization lead to the discharge of various contaminants into lakes, deteriorating their ecosystems (Grmasha et al., 2023a; Ambade et al., 2022; Ambade et al., 2020; Ambade, 2016; Al-sareji et al., 2022). When they reach the water body and have the ability to resist degradation, PAHs tend to build up in organisms (Net et al., 2015). As a result, they pass throughout the food chain, causing harmful health consequences for living species when consumed, even at low levels in the environment (Yuan et al., 2017). The presence of organic matter in the sediments allows PAHs to be adsorbed. This is attributed to the positive correlation between the amounts of organic carbon present in the sediments and the partition coefficient ( $K_d$ ) of PAHs (Patrolecco et al., 2010). PAHs could be reintroduced into water from sediment as a consequence of altering or bioturbation in the external environment (Giesy et al., 2016). Hijosa-Valsero et al. (2016) investigated the level of PAHs and pesticides in 53 small lakes in the Mediterranean region

(Hijosa-Valsero et al., 2016). The study determined that the average log  $K_d$  value for compounds in water was 3.61, while for compounds in sediments, it was 4.69. Consequently, sediment is an excellent medium for examining the pollution history of PAHs in aquatic systems. Analyzing the sedimentary records, therefore, supplies vital assistance for implementing environmental mitigation strategies, managing the environment, and controlling contamination (Zhang et al., 2024).

This work is dedicated to investigating the concentration as well as spatial and temporal variation of PAHs in Lake Balaton and provides vital information about the status of one of the biggest lakes in Europe. For two decades, no study has examined the level of PAHs compound in this lake. For instance, Kiss et al. (2001) analyzed atmospheric PAHs concentration in Lake Balaton (Kiss et al., 2001). The study found that PAHs ranged from 4–880  $\text{pg}/\text{m}^3$ , 4–300  $\text{pg}/\text{m}^3$ , 11–1050  $\text{pg}/\text{m}^3$  to 36–5000  $\text{pg}/\text{m}^3$  in spring, summer, autumn as well as winter, respectively. However, this study did not investigate the PAH levels in either Lake Balaton water or its sediment. Bodnár et al. (2004) studied the PAHs level in Lake Balaton sediment (Bodnár et al., 2004). The study found that PAHs average concentration was 132  $\mu\text{g}/\text{kg}$  dry weight for all sites and depths. Considering the time of the above two cited studies, it is therefore crucial to conduct measurements of the current level of these compounds. The objectives of this work are (1) to assess the temporal and spatial variation of PAHs in Lake Balaton after 20 years, (2) to evaluate the toxicity variation of PAHs in the lake sediment (3) to determine the possible source of PAHs, as well as (4) to assess PAHs

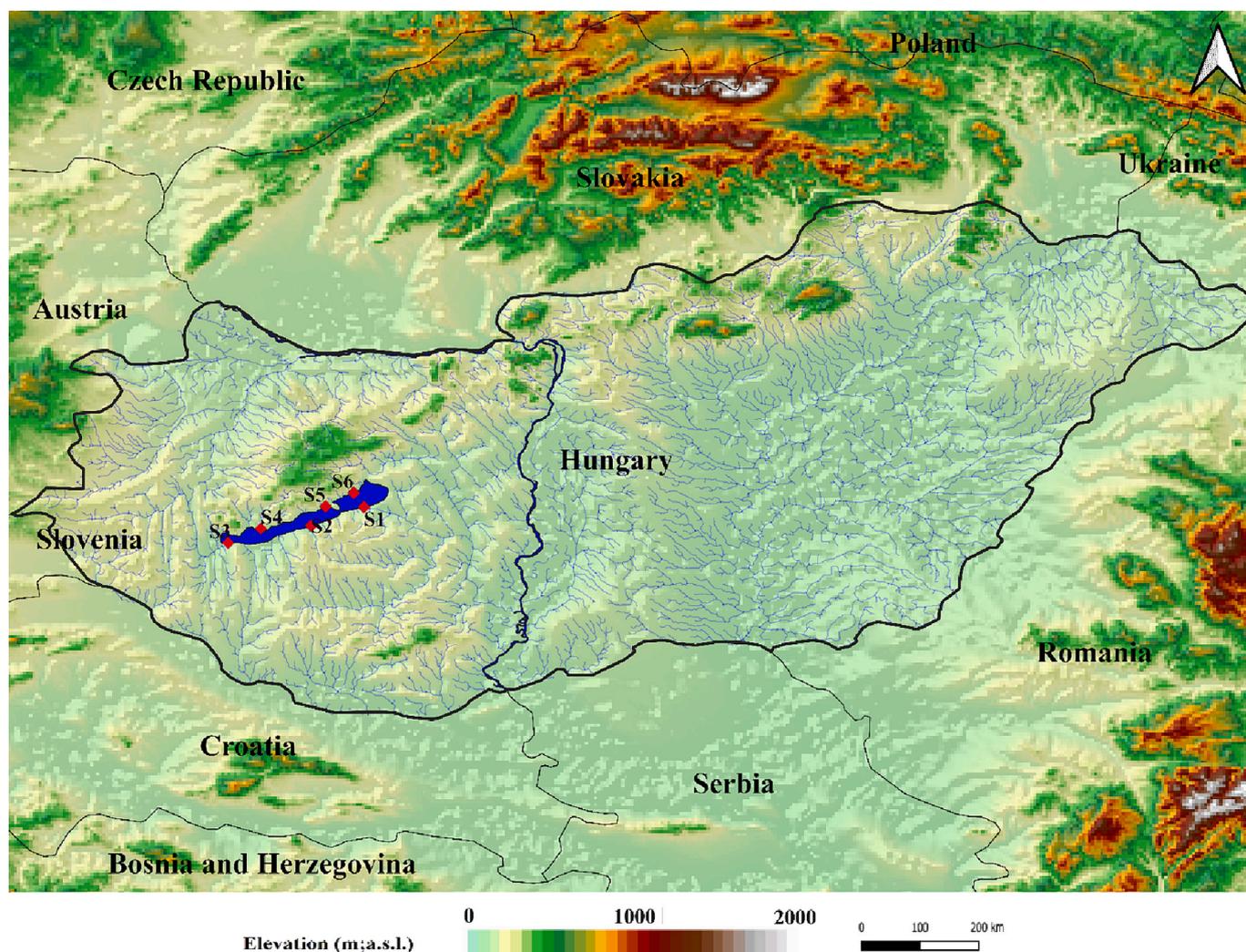


Fig. 1. Lake Balaton location along with the sampling sites.

potential carcinogenicity in Lake Balaton through lifelong cancer risk models.

## 2. Materials and methods

### 2.1. Study area

With two million visitors annually, Lake Balaton in Hungary is considered one of the largest shallow lakes in Central Europe (Svigruha et al., 2023). Fig. 1 illustrates the location of Lake Balaton in Hungary and the sampling site locations. Table S1 shows the coordinates of selected sites. Site 1 (S1) and site 6 (S6) were recreational areas with swimming activities. Other sites were located near the river and other streams inlet as follows: Tetves stream, Zala River, Eger-viz stream, and Örvényes stream for S2, S3, S4 and S5, respectively. The surface area of the lake is 596 km<sup>2</sup> and the mean depth is 3.20 m. The water temperature of the lake ranged between 0 and 29 °C, with an average yearly temperature of 15 °C (Somogyi et al., 2020). The spatial and temporal distribution of the population in this region is uneven. There are around 380,000 people living near the lake, comprising two-thirds of the local populace (Molnar et al., 2021). Moreover, there are around 40 operational wastewater treatment plants (WWTPs) within the watershed of the lake. Among them, Zalaegerszeg WWTP is considered the biggest facility capable of processing 50,000 m<sup>3</sup>/day (Molnar et al., 2021). The city is situated beside the Zala River, the primary outlet of this lake, responsible for almost half of the lake's total water volume.

### 2.2. Chemicals

The solvents utilized in this work were obtained from Fisher Chemical Co. in the US with a minimum purity of 99 %. Sixteen PAHs as the reference standards (QTM PAH-Mix, 2000 g/mL) were obtained from Supelco, Pennsylvania, USA. The 16 priority PAHs are including low molecular weight PAHs (LMWPAHs) such as naphthalene(Nap), acenaphthylene(Acy), acenaphthene(Ace), fluorene(Fl), phenanthrene (Phe), anthracene(Ant), medium molecular weight PAHs (MMWPAHs) such as fluoranthene(Flu), pyrene(Pyr), benzo(a)anthracene(BaA), chrysene (Chr), and high molecular weight PAHs (HMWPAHs) such as benzo(b)fluoranthene(BbF), benzo(k)fluoranthene (BbF), benzo (a) pyrene(BaP), Dibenz(a,h)anthracene (DBA), Benzo(ghi) perylene (BghiP), Indeno (1, 2, 3-cd) pyrene(IND). Table S2 shows the 16 PAH specifications, toxicity equivalent factor (TEF) values, and surrogate. Before each run, the glassware was first cleaned with Heidolph™ ultrasonic cleaners from Fisher Scientific. Subsequently, the glassware was washed with acetone (ACE), n-hexane (HEX), methanol (MeOH), as well as dichloromethane (DCM) to remove any background pollutants. Finally, the glassware was dried at 105 °C before running the experiments. The ENVITM-18 DSK SPE disc solid-phase extraction membrane (47 mm in diameter) and the recovery standards for PAHs and anhydrous sodium sulfate were supplied by Sigma-Aldrich, United States. The silica gel (2–5 mm) and anhydrous sodium sulfate were dried at a temperature of 500 °C in an oven (FI 600-60, Borel) for 4 h to remove any moisture and organics. Afterwards, the materials are taken to a desiccator for preservation until further utilization. MilliQ water with a resistivity of 18.20 MΩ.cm at ambient temperature as well as total organic carbon (TOC) below 5 ppb was used for solutions preparation.

### 2.3. Samples collection, pretreatment and extraction

Monthly sediment samples were collected from February 2023 to January 2024. At a depth of 0–10 cm from sediment, the samples were obtained from the Lake Balaton within six locations around the lake (Fig. 1). The sediment samples were then kept in plastic bags with rapping foil before being sent to the laboratory. They dried at a temperature of 25 °C and then crushed using an agate mortar and sieved (100-mesh). Finally, the samples were stored in brown glass vials for

further use. The sediment samples were extracted by following the processes reported in (Han et al., 2021; Chen et al., 2020). One gram was measured from the frozen sample and added to test tubes. Five millilitres of a mixture containing n-hexane and acetone (1:1) as well as a standard surrogate, was added to the tube. The mixture was vortexed for 1 min and left in an ultrasonic bath for 20 min. The cocktail was centrifuged for 15 min at 2000 rpm to separate the solid and liquid components. The liquid layer was carefully taken by pipette and placed in another tube. A mixture of 1:1 n-hexane and acetone was added to the tube to reach 5 mL. Activated copper was added to remove sulfur. Afterwards, sodium sulfate anhydrous was used to remove water, and the final solution was concentrated by nitrogen to reach 0.5 mL. Gas chromatography-mass spectrometry (GC/MS) was then used in order to conduct the analysis on the solution. A mass selective detector is included in the GC-MS instrument (Agilent 6890 N 5975C, USA). For the purpose of conducting the analysis, a column of HP-5MS measuring 30 m in length, with a diameter of 0.32 mm and a particle size of 0.25 μm, was utilized. Helium was used as a carrier gas at a flow rate of 1.50 ml/min before the selective ion scanning (SIM) approach was employed. The temperature of the injector was fixed at 300 °C. After preheating the oven for 60 s at a temperature of 100° C, the temperature was gradually increased to 300° C at a rate of 8 °C/min. The results of the triplicate tests were found to have a standard deviation that was lower than 5.83 %. The quantification of total organic matter (TOM) was conducted by measuring the weight loss of oven-dried sediments (105 °C, 2 days) after burning at 550 °C 2 h (Liang et al., 2007).

### 2.4. Quality assurance/quality control

Different approaches were considered when running each experiment. These included quality control tests such as blank samples, calibration standards, matrix spike standards, and limits of detection (LOD). To remove background contamination, glassware was regularly cleaned ultrasonically and then washed with n-hexane, acetone, methanol, and dichloromethane and then dried at 105 °C. The dry weight (dw) technique was used for 16 PAHs in sediment samples. LOD was obtained from the signal-to-noise ratio as well as analyte concentration (Wang et al., 2015). A standard solution was added to the sediment sample to recover PAH. No contamination was found in the blanks. The recovery range for the 16 PAHs was found to be between 88.498 % ± 3.62 % and 98.91 % ± 4.27 %. The sediment LOD varied between 0.87 and 1.05 ng/g dry weight. The spiking standard had a recovery ranging from 94.93 % ± 7.43 to 98.32 % ± 5.08 %. The concentration of 16 PAHs was modified for recovery. The accuracy of the analysis was confirmed through measuring samples and references. The resulting standard deviation ranges from 3.65 % to 9.03 %, which is much lower than the acceptable threshold of 25 %. An average value was presented for all measurements. The data was tested via the Shapiro-Wilk test at a significance level of 0.05.

### 2.5. Health risk assessment of soil PAHs

PAHs exposure via three routes, namely ingestion, dermal, and inhalation, could result in adverse effects to humans. Thus, it is important to evaluate the health risks of these routes. The TEF was employed to determine the toxicity equivalent concentration (TEQ<sub>BaP</sub>) for the toxic potency. The TEQ<sub>BaP</sub> value was obtained using Eq. (1) (Nisbet and Lagoy, 1992; Sankar et al., 2023).

$$TEQ_{BaP} = \sum_{i=1}^n (PAH_i \times TEF_i) \quad (1)$$

PAH<sub>i</sub> represents the amount of each PAH, whereas TEF<sub>i</sub> is the toxic equivalency factor for each chemical (as specified in Table S2). The ILCR (Incremental Lifetime Cancer Risk) approach, developed by the United States Environmental Protection Agency (USEPA), was used to assess the health risk posed by PAHs polluted soil to individuals. This method

identifies three potential paths by which humans could get exposed to PAH contamination (USEPA, 2009). The ILCR values were computed using Eqs. (2)–(5) (Ambade et al., 2023).

$$ILCR_{ing} = CS \times EF \times IR_{ing} \times ED \times \left( \sqrt[3]{\frac{BW}{70}} \times CSF_{ing} \right) \times (AT \times BW \times 10^6)^{-1} \quad (2)$$

$$ILCR_{inh} = CS \times EF \times IR_{inh} \times ED \times \left( \sqrt[3]{\frac{BW}{70}} \times CSF_{inh} \right) \times (AT \times BW \times PEF)^{-1} \quad (3)$$

$$ILCR_{der} = SA \times CS \times AF \times EF \times ED \times ABS \left( \sqrt[3]{\frac{BW}{70}} \times CSF_{der} \right) \times (AT \times BW \times 10^6)^{-1} \quad (4)$$

$$\text{Carcinogenic risk(ILCR)} = ILCR_{ing} + ILCR_{inh} + ILCR_{der} \quad (5)$$

The CSF, or carcinogenic slope factor, is a unit of measurement expressed in milligrams per kilogram per day. The United States Environmental Protection Agency (USEPA) provided BaP CSF (Carcinogenic Slope Factor) values of 25, 7.3, and 3.85 (mg/kg<sup>-1</sup> day<sup>-1</sup>)<sup>-1</sup> for the routes of skin contact, ingestion, and inhalation, respectively. CS denotes the cumulative sum of PAH concentrations that have been converted into hazardous equivalents of BaP using the toxic equivalency factor (in ng/g dry weight). Table 1 displays the ILCR parameter values and explanations for the aforementioned equations. Firstly, the calculation of ILCR depends on the identification of PAHs as concentrations equivalent to BaP by using the TEF of each PAH compared to BaP. BaP is the reference chemical compound having a TEF of one. If the ILCR (dimensionless) is below 10<sup>-6</sup>, it is deemed insignificant. Nevertheless, if it exceeds 10<sup>-4</sup>, there is a significant concern. The inhalation component of ILCR was negligible (<10<sup>-6</sup>) and hence was excluded.

Furthermore, an examination was conducted to determine the

**Table 1**  
ILCR model parameters descriptions.

Parameter	Description	Unit	Individual	References
AF	Dermal-adherence-factor	mg/cm <sup>2</sup>	0.07	(USEPA, 2011)
ABS	Dermal-absorption-factor	unitless	0.13	(USEPA (US Environmental Protection Agency), 1989)
AT	Average-time	Days	ED × 365	(Grmasha et al., 2023a; Miao et al., 2023)
BW	Body weight	Kg	70	(Grmasha et al., 2023a; Miao et al., 2023)
ED for sediment	Exposure-duration	Years	30	(Grmasha et al., 2023a; Miao et al., 2023)
EF	Exposure frequency	days/year	350	(Grmasha et al., 2023a; Miao et al., 2023)
IR ingestion for sediment	Ingestion rate	mg/day	100	(USEPA, 2011)
SA	Dermal-surface-area-exposure	cm <sup>2</sup>	5700	(Miao et al., 2023)
ED for water	Exposure-duration	Years	55	(USEPA (US Environmental Protection Agency), 1989)
IR-ingestion rate of water	Ingestion rate	(L/day)	1.5	

correlation between the levels of certain PAHs and the thresholds for Effect Range Low (ERL) as well as Effect Range Median (ERM) values. This work also evaluated the ecological consequences of exposing aquatic species to PAHs present in sediments. The SQGs categories by Long et al. (1995) include exposure intervals that minimal effects range with rare biological effects (<ERL), possible effects range with occasional biological effects (≥ERL and <ERM), as well as probable effects range with frequent biological effects (≥ERL) (Long et al., 1995).

### 3. Results and discussion

#### 3.1. PAHs contamination of sediments

The contamination of 16 PAHs in sediments for four seasons is illustrated in Fig. 2. The overall concentration of the 16 PAHs was the highest in winter, fluctuating between 448.35 and 619.77 ng/g dry weight, followed by spring (279.45 to 595.41 ng/g dry weight), then autumn (300.72 to 491.78 ng/g dry weight), and the least was measured in summer (257.21 to 465.49 ng/g dry weight). The PAHs concentration in lake sediments exhibited spatial variability, reflecting the influence of anthropogenic activities in different areas of the lake. Temporal variability was also seen between and within seasons, which is likely to be attributed to the effect of temperature on PAHs (Maliszewska-Kordybach, 1993). The overall indication of the PAHs status in the lake can be characterized as an HMW PAHs carrier and low LMWPAHs concentration, as seen in Fig. 2. This phenomenon could occur due to the elevated partition coefficients of HMWPAHs, as well as their pronounced hydrophobic nature in aquatic environments (Sheng et al., 2021). Moreover, the reason for reduced LMWPAH levels in sediment is their increased tendency to evaporate compared to HMWPAHs (Miao et al., 2023). PAHs with 4–6 rings had the greatest values in comparison to LMWPAHs. The concentrations of Flu, Pyr, BaA, Chr, BbF, BkF, BaP, DBA, BghiP, and IND ranged from 8.20 to 33.27 ng/g dry weight, 11.11 to 32.22 ng/g dry weight, 15.18 to 37.12 ng/g dry weight, 16.79 to 46.26 ng/g dry weight, 25.22 to 43.84 ng/g dry weight, 29.87 to 34.52 ng/g dry weight, 28.35 to 66.20 ng/g dry weight, 21.31 to 38.30 ng/g dry weight, 16.56 to 38.29 ng/g dry weight, and 19.52 to 57.33 ng/g dry weight, respectively. The LMWPAHs ranges were Ant (9.70–24.43 ng/g dry weight), FI (14.41–19.77 ng/g dry weight), and Phe(16.54–28.92 ng/g dry weight). The amount of Ace, Acy, and Nap found were 12.20–18.41 ng/g dry weight, 14.29–26.79 ng/g dry weight, and 12.99–26.15 ng/g dry weight, respectively. Although the sediment samples were collected over several seasons, no discernible trends in PAH concentration were observed. This suggests that temporal fluctuations do not impact the sediments. Sediment contamination can be categorized into four levels: (A) lightly contaminated (concentrations below 0.1 µg/g), (B) moderately contaminated (concentrations from 0.101 to 1.0 µg/g), (C) heavily contaminated (concentrations from 1.001 to 5.0 µg/g), and (D) very contaminated (concentrations exceeding 5.0 µg/g) (Baumard et al., 1998). Based on this classification, the contamination level in the sediment at Lake Balaton is deemed to be moderate.

The seven carcinogenic CPAHs (BaA, Chr, BbF, BkF, BaP, DBA, and IND) exhibit the greatest amounts of PAHs, ranging from 115.19 ng/g dry weight in spring to 361.41 ng/g dry weight in winter. Fig. 3 demonstrates the presence of CPAHs in sediments throughout the whole year. It can also be noticed from the Figure that the highest recorded values were in S3, which could be explained by the discharge of the Zala River into the lake. This increase was also noticed in S4 as a combination of both Eger-viz stream inlets. Records near other inlets also exhibited an increase in CPAHs. There was also a noticeable amount of CPAHs in S1 and S6, which have been utilized as recreational areas for tourists and locals. The variations in the spatial dispersion of PAH levels in sediments could be ascribed to four contributing factors: (1) untreated wastewater, vehicle emissions, industrial activities, and fuel consumption; (2) diverse weather-related hydrodynamic systems that can cause the

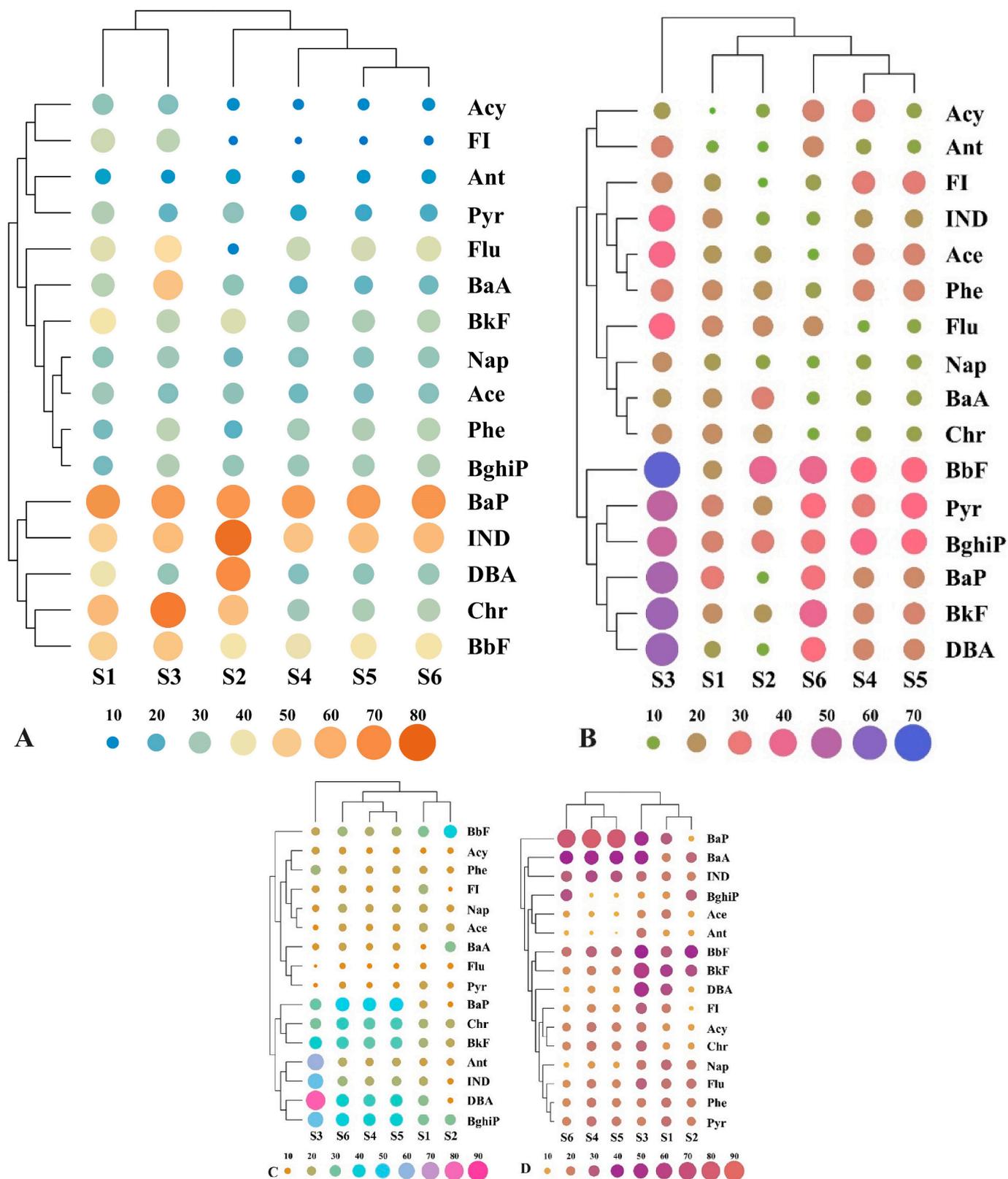


Fig. 2. 16 PAHs concentration (ng/g dry weight) for Lake Balaton in winter (A), spring (B), summer (C), and autumn (D).

stirring and re-suspension of sediments; and (3) alterations in sediment texture resulting from spatial properties of sites (Salmela et al., 2022). The industrial sector is very concerned about the level of PAHs present in wastewater (Sethi et al., 2023). PAHs have the potential to contaminate aquatic bodies if the wastewater effluent is not properly treated

(Xing et al., 2024). PAHs are deemed hazardous chemicals due to their elevated toxicity and long-lasting capacity to contaminate the environment. They possess attributes that have the potential to cause cancer, genetic damage, and mutations, posing a risk to human well-being (Grmasha et al., 2022).

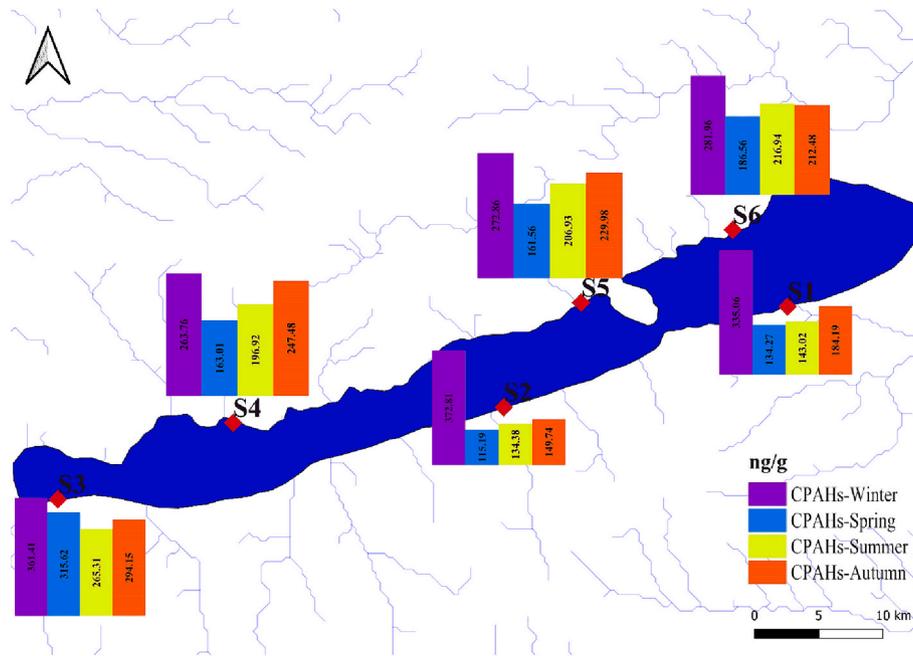


Fig. 3. Carcinogenic PAHs (Seven) concentration in the sampling sites in different seasons.

The reported PAHs levels in sediments of other European lakes in France (Lécrivain et al., 2018), central Italy (Veroli et al., 2010), and Estonia (Kapanen et al., 2013) were not considered exceptionally high. Nevertheless, several lakes in Europe experience the detrimental effects of anthropogenic pollution. For instance, the sediments on the surface of Bergen, located in western Norway, were found to have a total concentration of 16 PAHs of 8161 ng/g dry weight (Andersson et al., 2014). Hijosa-Valsero et al. (2016) studied PAH concentrations in the

sediments of 53 lakes in Spain (Hijosa-Valsero et al., 2016). The sediments had PAH values ranging between 4 and 4286 ng/g dry weight. The PAH pollution in northern Spain was linked to agricultural activities. Lake Pamvotis sediments in Greece have been investigated for PAH levels by Daskalou et al. (2009). The study revealed that PAHs ranged from 34.7 and 1600 µg/kg, with petroleum contamination detected as the main source (Daskalou et al., 2009). Comparing the PAHs results obtained in this study to the abovementioned European lakes, it can

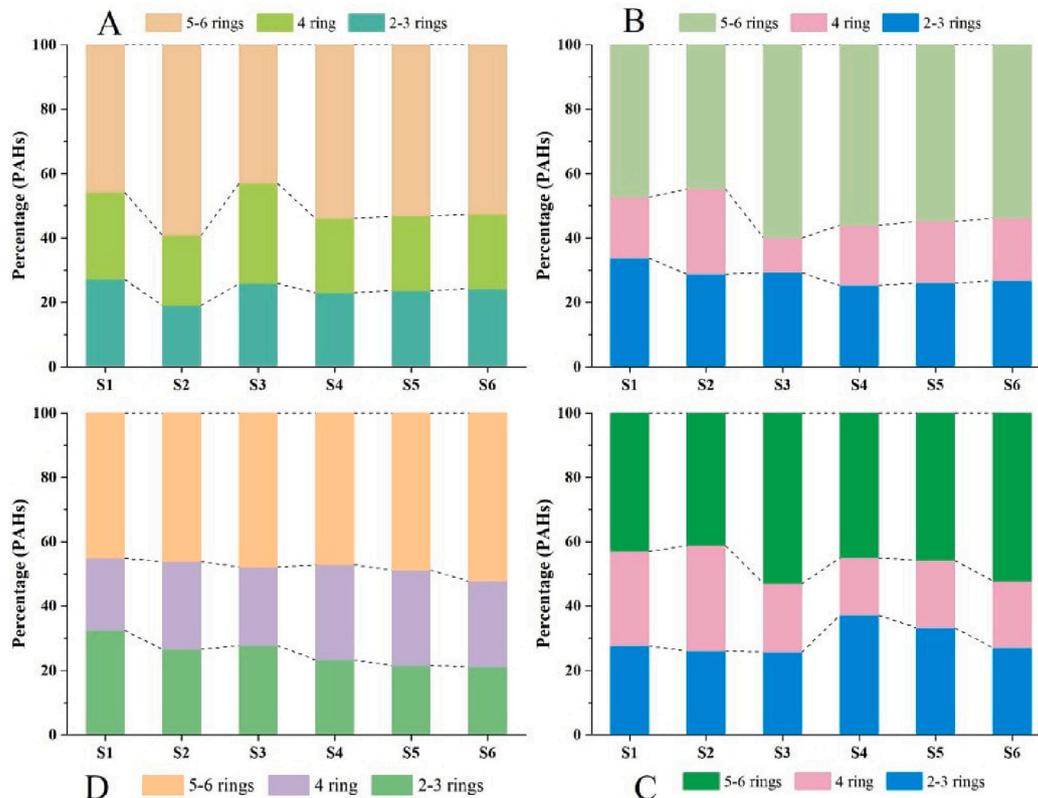


Fig. 4. The percentage of PAHs ring profile for Lake Balaton in winter (A), spring (B), summer (C), and autumn (D).

generally be said that Lake Balaton was not heavily polluted by PAHs compounds.

### 3.2. PAHS ring profile

Examining the composition profiles of PAHs in the sediment samples of Lake Balaton assists in determining their origin. The sediments of Lake Balaton were mostly composed of PAHs with 5–6 rings, as seen in Fig. 4. The spring months had a percentage of 41 % (S2), while the summer months had a percentage of 60 % (S3). The levels of 2-3ring as well as 4-ring PAHs were overall close in all seasons except summer, where there was a 10 % difference (2–3 rings% > 4-ring%). The proportion of high- and medium-molecular-weight PAHs was much higher compared to low-molecular-weight PAHs (Fig. 4). This highlights the elevated level of HMWPAHs and MMWPAHs contents in Lake Balaton. This could be explained by HMWPAHs and MMWPAHs being resistant to degradation and are of a high hydrophobic nature (Cao et al., 2024; Chen et al., 2004). PAHs mostly enter the ecosystem from sources related to petroleum and combustion processes. PAHs sourced from petrogenic are abundant in LMW PAHs, while the reverse is true of pyrogenic sources (Zakaria et al., 2002). The prevalence of MMWPAHs and HMWPAHs in this work indicates that PAHs in sediments likely

originate mostly via pyrogenic sources.

### 3.3. PAH diagnostic ratios

According to Yunker et al. (2002), diagnostic ratio approaches are often employed to assess the potential origins of PAHs (Yunker et al., 2002). The study identified many diagnostic ratios, including Ant/(Ant+Phe), BaA/(BaA + Chr), Flu/(Flu+Pyr), as well as IND/(IND + BghiP). The BaA/(BaA + Chr), IND/(IND + BghiP) and Flu/(Flu+Pyr) ratios indicate the presence of pyrolytic sources, while the Ant/(Ant+Phe) ratio suggested the presence of petrogenic sources. The Ant/(Ant+Phe) ratio below 0.1 suggests that the contamination source is petroleum or combustion byproducts. A Flu/(Flu+Pyr) ratio below 0.4 indicates that the source of combustion is petroleum. Conversely, a Flu/(Flu+Pyr) ratio over 0.5 indicates that the source of combustion is coal, as well as biomass. A ratio between 0.4 and 0.5 suggests the combustion of petroleum-based substances such as kerosene, gasoline, and crude oil, as well as other oil variants (Grmasha et al., 2023b). The graph in Figs. 5 and 6 display the ratios of Ant/(Ant+Phe), Flu/(Flu+Pyr), BaA/(BaA + Chr), as well as IND/(IND + BghiP). The range of IND/(IND + BghiP) values, ranging from 0.26 (spring S6) to 0.86 (autumn S4), suggests that sources were from coal, biomass and petroleum combustion. The BaA/

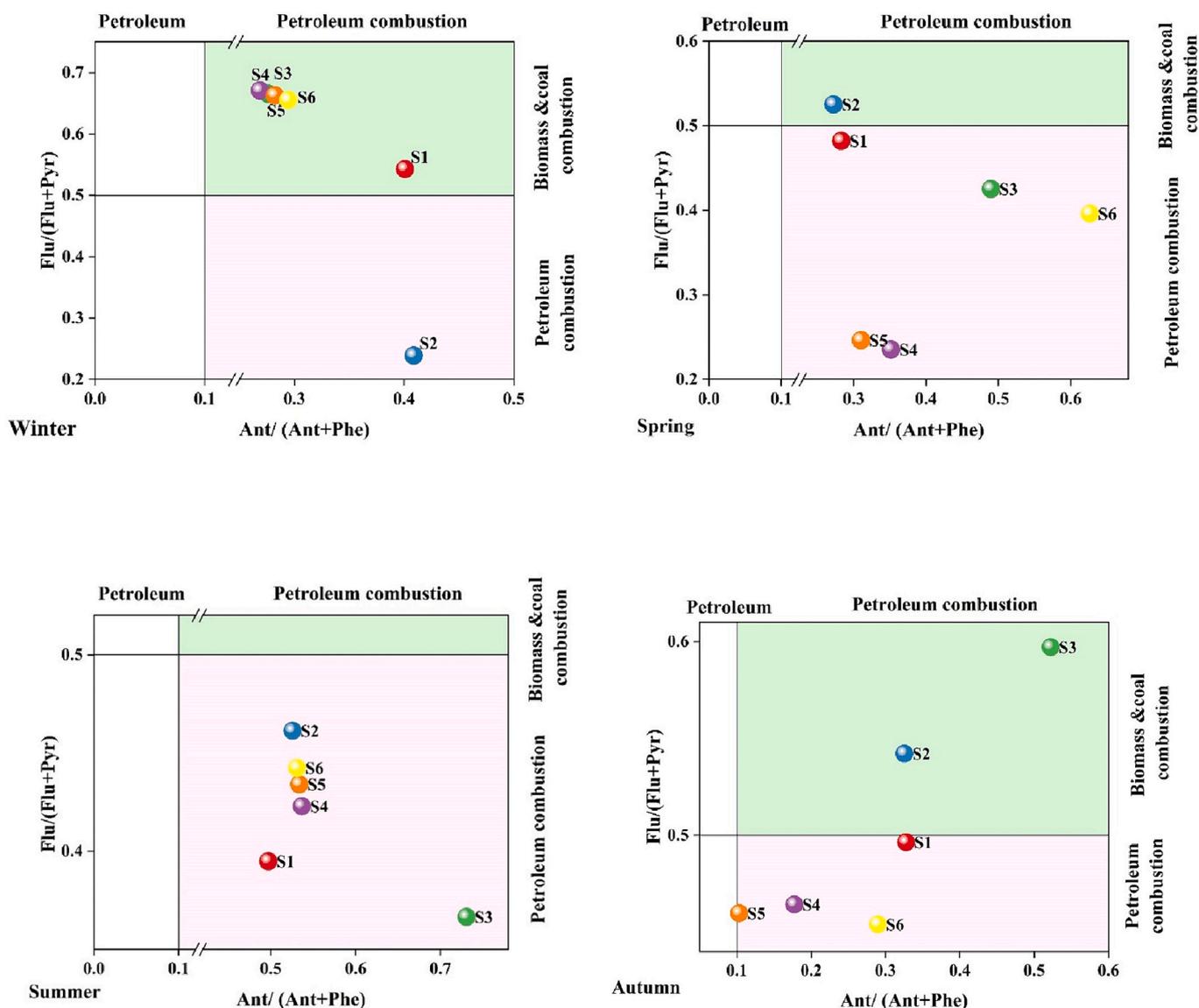


Fig. 5. Diagnostic ratio plots of Ant/(Ant + Phe) vs Flu/(Flu + Pyr) in sediments Lake Balaton for all seasons.

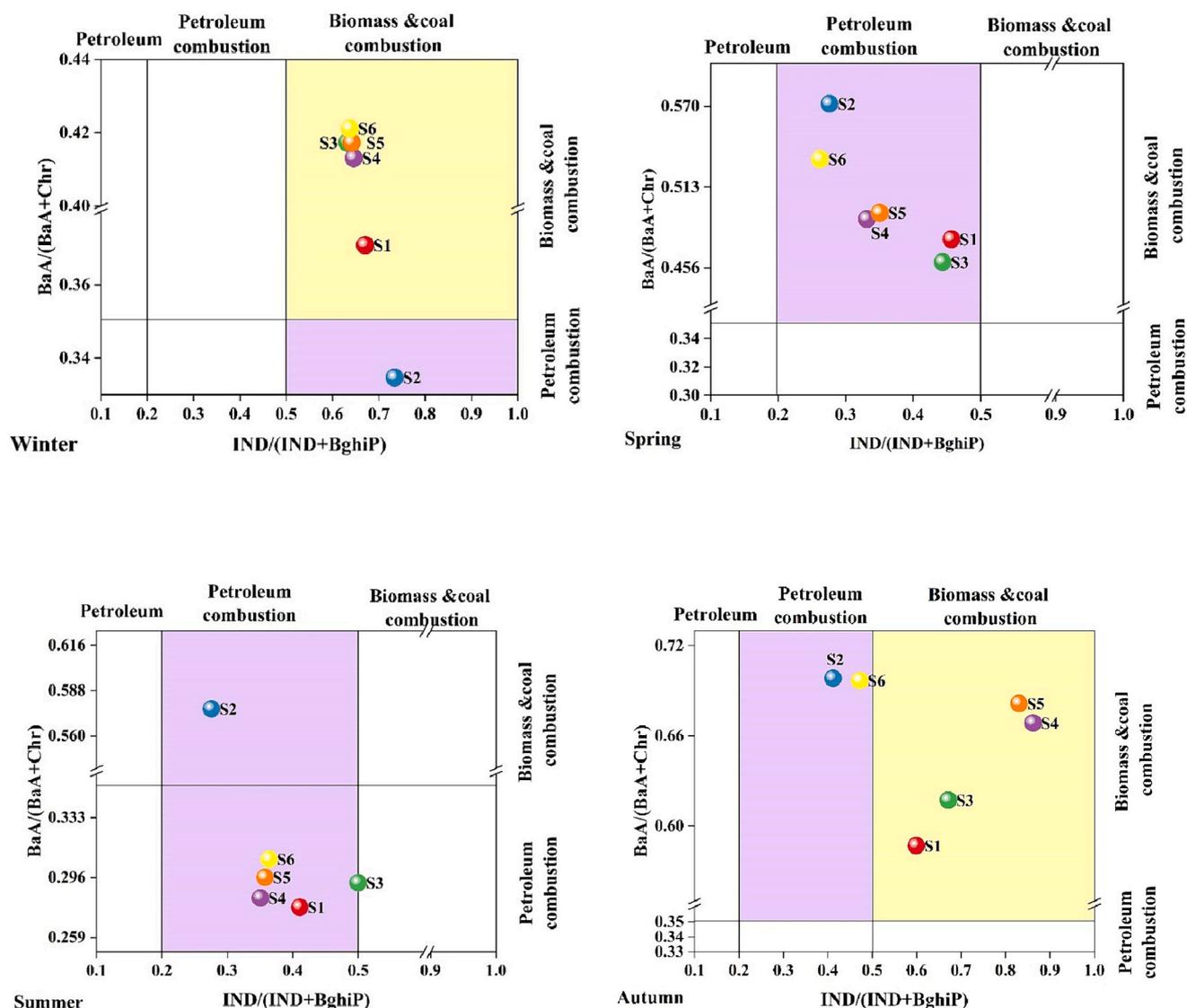


Fig. 6. Diagnostic ratio plots of  $IND/(IND + BghiP)$  vs  $BaA/(BaA + Chr)$  in sediments Lake Balaton for all seasons.

(BaA + Chr) ratios vary between 0.28 (S1, summer) and 0.70 (S6, autumn), whereas the  $Ant/(Ant+Phe)$  values range from 0.1 (S5, autumn) to 0.73 (S3, summer). These values signify the contamination source being combustion. The  $Flu/(Flu+Pyr)$  ranged from 0.24 for S2 (winter) to 0.67 for S3 (winter). Within this range, the assortment comprises a selection of fuel sources, including coal, biomass combustion, and a variety of petroleum products, such as gasoline, kerosene, crude oil, and other petroleum derivatives. The primary sources of sediment contamination in the Lake Balaton basin are the combustion of coal and biomass. Petroleum products also contribute to this contamination, albeit to a lesser degree. Similar patterns are seen in the Liujiang River basin situated in the southwestern region of China (Miao et al., 2023).

Fig. 7 shows the Pearson correlation coefficients of the 16PAHs compounds for Lake Balaton in all seasons. In winter, Ant positively interacts with Pyr, Ace, Bap and BkF. Spring showed a positive interaction between most of the HMWPAHs compounds. For instance, there was a positive correlation between Bap and Pyr, DBA, BkF, BghiP and BbF. The same trend was also noticed in summer with the recoding of a positive link between BghiP and other PAHs compounds like IND, DBA, BkF, Bap and Chr. There was also a positive agreement between BghiP and both Ant, Acy and Phe. In autumn, there was a weak correlation between LMWPAHs and HMWPAHs.

The principal component analysis (PCA) of measured PAHs in the sediment samples is illustrated in Fig. 8 (A). A total of 82.62 % is contributed by two components. The first (Factor 1) attributed 49.67 % of the total variation. There is no significant positive correlation between the PAHs compounds. Among the 16 PAHs, the following were shown to have a slight positive correlation with Factor 1: Nap, Phe, Flu, BbF, BkF, BaP, and IND. According to Wang et al. (2009), the burning of gasoline during engine operation produces significant quantities of BbF, Flu, IND, and BaP (Wang et al., 2009). A study by Zhou et al. (2024) stated that BkF is typically formed by the burning of coal, and IND and BkF may be representative of processes that include the combustion of diesel fuel (Zhou et al., 2024). Phe usually indicates pollution that originates from coke ovens, while Nap is mostly derived from petroleum sources (Chen et al., 2013). Thus, factor 1 is a representation of a source of combustion for fossil fuels. It was shown that factor 2 also had a slight positive correlation with Ant, Chr, DBA, and BghiP, and it explained 32.95 % of the total variation. According to the results of PCA, BghiP is a characteristic tracer of a gasoline engine (Simcik et al., 1999), and Chr is a tracer that originates from the combustion of gasoline (Wang et al., 2009). DBA is a molecular marker for contamination sources of fossil fuel combustion (Wang et al., 2020). Therefore, factor 2 is a representation of a fossil fuel source.

The current work employed regression analysis to examine the

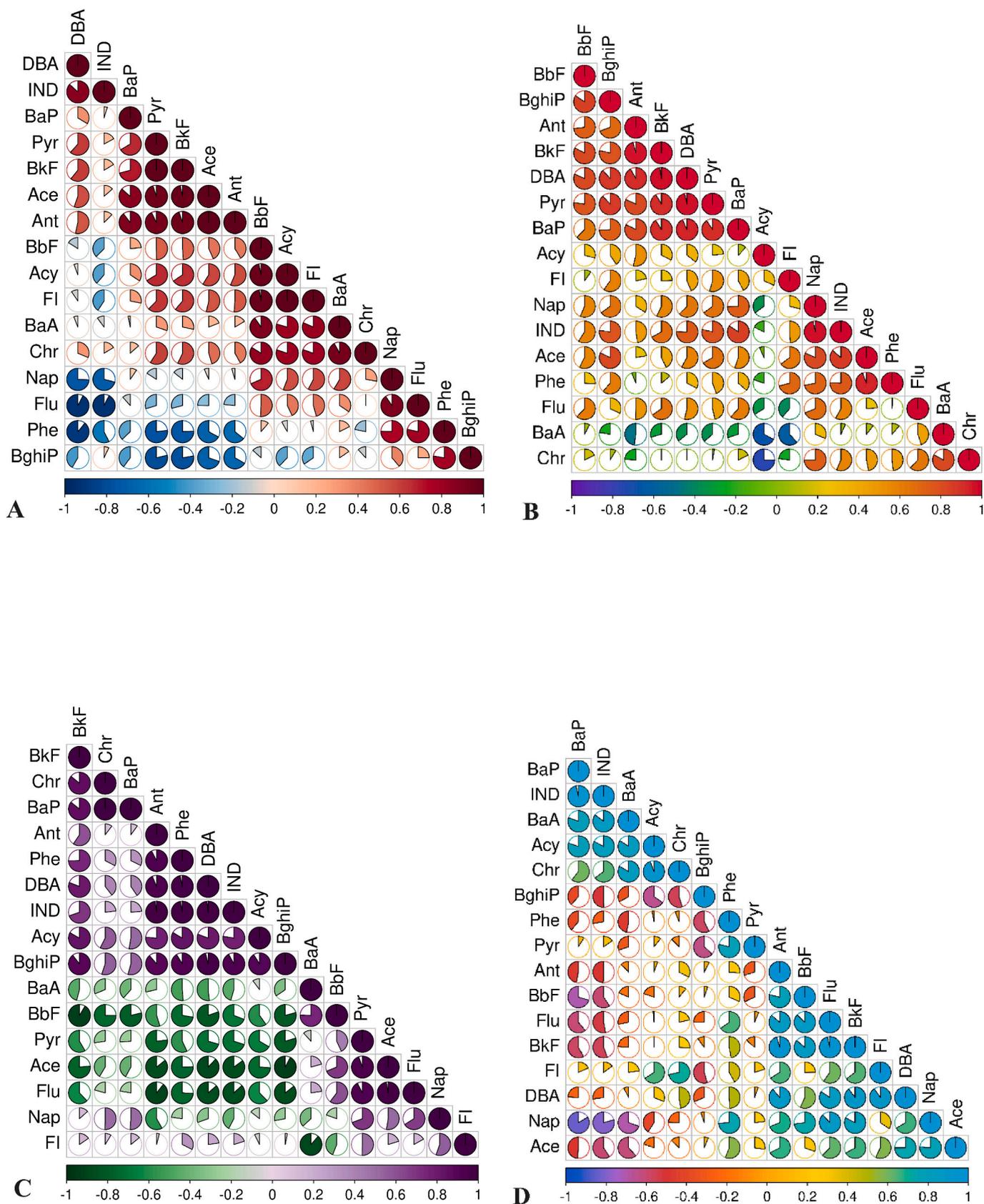


Fig. 7. Pearson correlation coefficients of the 16PAHs compounds for Lake Balaton in winter (A), spring (B), summer (C), and autumn (D).

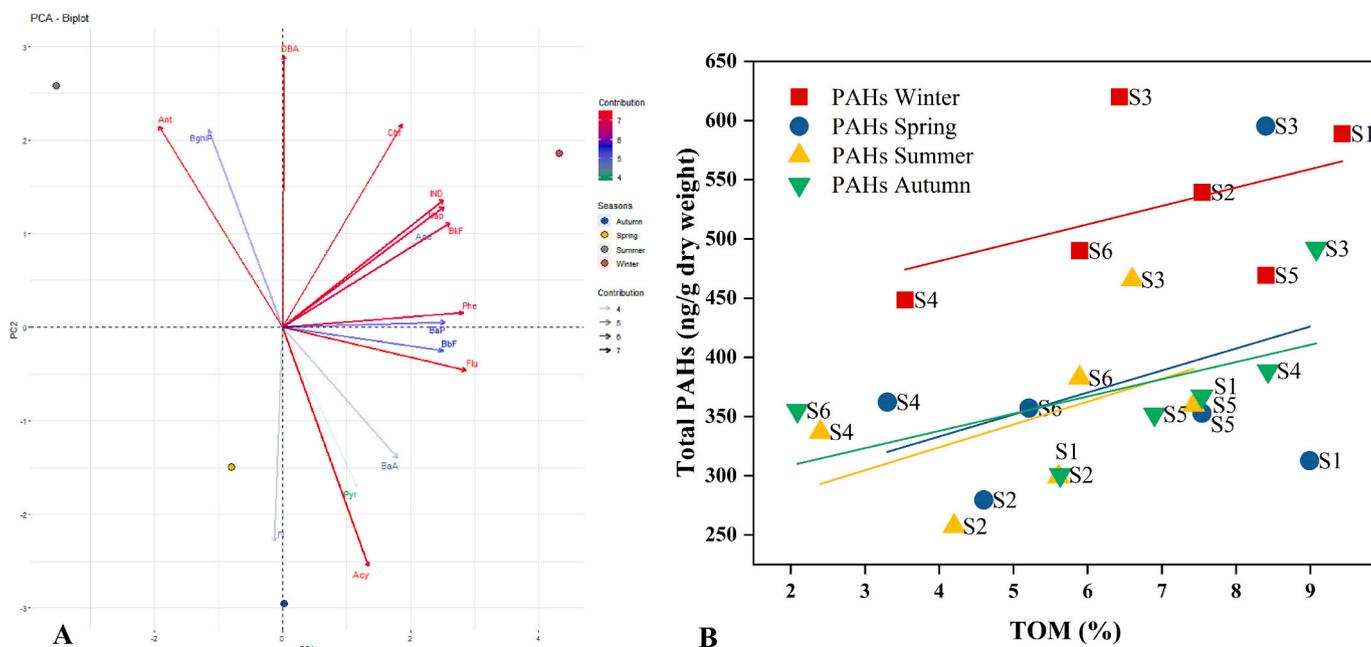


Fig. 8. Principal component analysis for 16 PAHs in Lake Balaton sediment samples (A) and the correlation between total PAH and TOM contents (%) in sediments of Lake Balaton (B).

correlation between the concentration of total PAHs and the percentage of TOM contents (%). Fig. 8 (B) shows the relationship between total PAH and the TOM contents (%). The results of the linear regressions indicate that there is no significant correlation between total PAHs and TOM. A study carried out by Mostafa and colleagues revealed the same findings, indicating a lack of statistically significant link among sediments collected from the western harbor of Alexandria (Mostafa et al., 2003). It was also concluded that there was no correlation between the total PAHs in the sediments of the Hadhramout coastal area in Yemen and the TOM (Mostafa et al., 2009). This correlation indicates that the PAHs concentration and their distribution in sediments are mostly influenced by direct input rather than the specific characteristics of the sediment existing in the neighborhood of Lake Balaton.

### 3.4. ILCR

When evaluating the health hazards to the population, particularly fishermen and swimmers, the only criteria taken into account were the potential harm caused by direct contact with the skin and unintentional consumption of PAHs from the sediments of Lake Balaton. Fig. 9 shows the ILCR in sediments of Lake Balaton for both adults and children. Ingestion exposure has a much greater health hazard compared to

dermal exposure, explaining ingestion is the main route causing the health risk of PAHs. Typically, children are more sensitive to 16 PAHs pollution because of their smaller body weight, which explains the higher ILCR values compared to adults. The ILCR was high at all locations, especially during the winter, surpassing a value of  $10^{-4}$ . The poor solubility of most PAHs compounds in water, which typically have medium to high molecular weights, is likely the cause of this phenomenon. This process facilitates the deposition of chemicals in sediments, hence posing a potential threat to human health. The concentration in most locations is typically below  $10^{-4}$  or somewhat higher, particularly during the autumn months. Overall, the majority of locations exhibited a significant carcinogenic hazard, which poses a challenge to public health, particularly considering the lake’s annual visit by 2 million visitors. Hence, to mitigate the carcinogenic hazard posed by PAHs in the basin, it is essential to decrease the occurrence of hydrophilic activities while also taking proactive measures to battle lake pollution. According to Table 2, the toxicity guidelines with respect to the 16 PAHs range were found to have minimal effect for most PAHs compounds except Acy and FI in autumn and spring, respectively. Acy and FI have values slightly higher than ERL, which could impose a risk to the health of aquatic life in the lake.

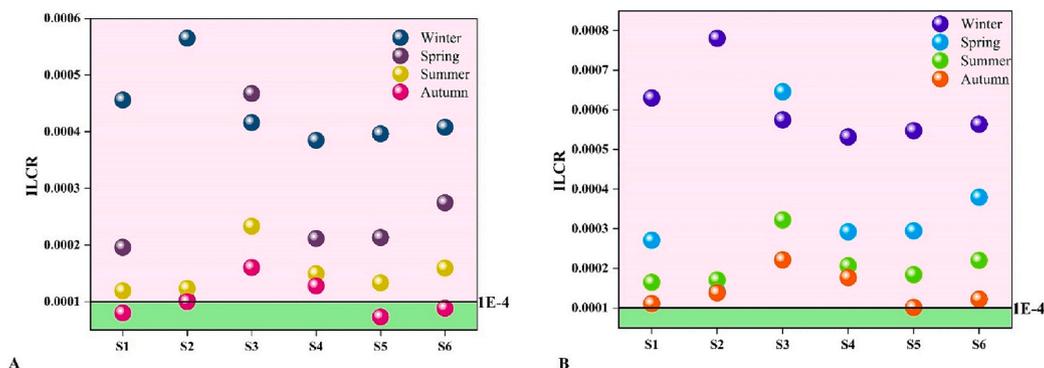


Fig. 9. Incremental Lifetime Cancer Risk values in Lake Balaton for all sites during the seasons (A) adults and (B) children. The green area is below the threshold. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 2

Toxicity guidelines for the 16 PAHs range<sup>a</sup>.

PAH	ERL-ERM	PAH average range (ng/g dry weight)	<ERL	≥ERL and <ERM	≥ERM
Nap	160–2100	14.29–26.79	✓		
Acy	16–500	12.20–18.41	Rest	Autumn	
Ace	44–640	12.99–26.15	✓		
Fl	19–540	14.41–19.77	Rest	Spring	
Phe	240–1500	16.54–28.92	✓		
Ant	600–5100	9.70–28.43	✓		
Flu	85.3–1100	8.20–33.27	✓		
Pyr	665–2500	11.11–32.22	✓		
BaA	261–1600	15.18–37.12	✓		
Chr	384–2800	16.79–46.26	✓		
BbF	320–1880	25.22–43.84	✓		
BkF	280–1620	29.87–34.52	✓		
BaP	430–1600	28.35–66.20	✓		
DBA	63.4–260	21.31–38.30	✓		
BghiP	430–1600	16.56–38.29	✓		
IND	240–	19.52–57.33	✓		

<sup>a</sup> The numbers illustrated in the table are the average of all sites within the season.

#### 4. Conclusion

In this work, the spatial and temporal distribution of 16 priority PAHs in sediments from Lake Balaton were investigated. The  $\Sigma$ 16PAHs in the sediment varied between the lowest 257.21 ng/g dry weight in summer to the highest 619.77 ng/g dry weight in winter. It was shown that the seven carcinogenic CPAHs had high levels of PAHs, with the highest recorded values being in S3 and S4. The PAH levels range from 115.19 ng/g dry weight in the spring to 361.41 ng/g dry weight in the winter. While the sediment samples were gathered throughout the seasons, no noticeable patterns in the PAHs were detected. This implies that temporal fluctuations do not affect the sediments. In accordance with the findings of the TOM analysis with PAHs, the fluctuation in PAH levels that occurs over the seasons may be related to the direct source of PAHs. The main source could be untreated wastewater near the lake. The PAH composition was characterized as HMW with 5–6 rings. PAHs derived from burning coal, biomass, and petroleum were detected at all locations. The elevated ILCR values, particularly over  $10^{-4}$ , indicate that the ingestion of 16 PAHs is the primary pathway by which human health is affected. It is possible that children are especially vulnerable to the pollution caused by PAHs. Therefore, it is crucial to develop mitigation methods in order to effectively manage the pollution of PAHs in the lake.

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#### CRedit authorship contribution statement

**Ruqayah Ali Grmasha:** Writing – review & editing, Writing – original draft, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Osamah J. Al-sareji:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Writing – original draft, Writing – review & editing. **Mónika Meiczinger:** Methodology, Writing – original draft, Writing – review & editing. **Raed A. Al-Juboori:** Investigation, Methodology, Writing – original draft, Writing – review & editing. **Csilla Stenger-Kovács:** Methodology, Writing – review & editing. **Edina Lengyel:** Methodology, Writing – original draft, Writing – review & editing. **Hasan Sh. Majdi:** Conceptualization, Visualization, Writing – original draft, Writing – review & editing. **Rafid Al Khaddar:** Writing – review &

editing, Writing – original draft, Visualization. **Salah Jasim Moham-med:** Writing – review & editing, Writing – original draft, Visualization, Conceptualization. **Khalid S. Hashim:** Writing – review & editing, Writing – original draft, Software.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

Data will be made available on request.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.marpolbul.2024.116333>.

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