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ORIGINAL PAPER

Impact of coal tar pavement on polycyclic hydrocarbon distribution in lacustrine sediments from non-traditional sources

S. Froehner · D. B. de Souza · K. S. Machado · F. Falcão · C. S. Fernandes · T. Bleninger · D. M. Neto

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Abstract The evaluation of potential environmental impacts from polycyclic aromatic hydrocarbon is a subject that requires investigation, especially related to emission sources from highway roads with high traffic. In this paper, the distribution of polycyclic aromatic hydrocarbons was investigated in the surface sediments from a subtropical lake, located in the south of Brazil and away from urban areas. The results showed that all sediments presented such compounds in significant concentration, ranging from 9.50 to 29.88 μg/g. The highest total concentration was found in those sites close to the highway (bridges) and the deepest area of the lake. Moreover, a high concentration of polycyclic aromatic hydrocarbons of high molecular weight, such as benzo(a)anthracene and dibenzo(a,h)anthracene was found. It was laso found that high concentrations might be related to asphalt and coal tar pavement used in the highway construction close to the lake. In addition, the potential capacity of sediment toxicity was compared with values of the interim sediment quality guidelines values. The results indicate that most of the sediment presents concentrations of hydrocarbons above these limits. Only fluoranthene and chrysene were found in concentrations below the guideline values. The concentrations of most of the polycyclic hydrocarbons are higher than the quality guidelines. However, the main concern is associated with the presence of benzo(a)anthracene whose concentration is 416 times higher than the limit establisehed by guidelines. The same observation can be done to traces of benzo(a)-pyrene and dibenz(a,h)anthracene, with concentration values of 13 and 1,993 times higher.

Keywords Asphalt · Organic matter · Polycyclic aromatic hydrocarbons · Surface sediments · Sediment quality

Introduction

Aliphatic hydrocarbons (AHCs) and polycyclic aromatic hydrocarbons (PAHs) are two major classes of compounds present in oil. Thus, such compounds have been used as chemical markers to identify environmental pollution and contamination resulting from crude oil, gasoline, and diesel (Lee et al. 2005; Liu et al. 2008; Savinov et al. 2003). Usually, PAHs can be introduced in the environment by various processes, such as incomplete combustion at higher temperatures of recent and fossil organic matter (pyrolytic origin), slow maturation of organic matter under the geochemical gradient conditions (petrogenic origin) and oil spill accidents, as well as by biogenic processes (Wan et al. 2005). Direct PAHs biosynthesis by organisms, such as bacteria, fungus, and algae have not yet been described sufficiently (Bzdusek et al. 2004; Opuene et al. 2007; Soclo et al. 2000).

In general, the presence of PAHs in the environment is due to anthropogenic inputs, considered as the major source of PAHs. Those inputs are mainly associated with waste discharges from industrialized and urbanized areas, off-shore petroleum hydrocarbon production or petroleum transportation. Each source (pyrolytic, petroleum and diagenetic hydrocarbons) give rise to characteristic PAHs

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S. Froehner (✉) · F. Falcão
Department of Environmental Engineering,
Federal University of Parana, Curitiba, PR 81531-980, Brazil
e-mail: froehner@ufpr.br

D. B. de Souza · K. S. Machado · C. S. Fernandes ·
T. Bleninger · D. M. Neto
Department of Hydraulic and Sanitation,
Federal University of Parana, Curitiba, PR 81531-980, Brazil

patterns. It is, therefore, possible to identify the processes that generated these compounds (Yunker et al. 1999, 2002).

Polycyclic aromatic hydrocarbons ubiquity in the sediments indicates that the accumulation phenomena dominates degradation processes in the sediment matrices. Clearly, some evolution kinetics can be comparable. In addition, chemical behavior comparability was developed to assess various origins of these pollutants based on the molecular indices (Baumard et al. 1998).

It is possible to determine the processes that generated such hydrocarbons in the studied matrices with simultaneous association of various molecular indices. The behavior of PAHS in the environment depends on vapor pressure and solubility in water. Low vapor pressure and its hydrophobic characteristics imply not much mobility when reaching the soil. As a consequence, adsorption becomes an important process, including soil, sediments and particulate matter in the gas phase. Another process is related to degradation that refers to the most common way of reducing PAHs concentrations in the environment. Partially dissolved in water these compounds tend to be absorbed at suspended solids and sediments in the water column what can be justified by the high K_{OC} values (partition coefficient correlated with organic substances). Generally, high concentrations can be observed in rivers and lakes because of these hydrophobic characteristics and rapid adsorption at sediments; especially, in those places were sedimentation is favorable, such as in natural pool areas and stagnation or recirculation zones with lower flow velocities. Once adsorbed to the sediment, they are less susceptible for degradation (Baumard et al. 1998; Heitkamp and Cerniglia 1987; Hinga 2003; Hu et al. 2010).

In such a context, sediments can be considered a pollutant trap and have been proven as an efficient source to identify environmental impacts. It is also important to know the sediments ability to store hydrophobic pollutants besides the levels and sources of these compounds in sediment samples. This is possible because PAHs isomer pairs have similar physical–chemical properties resulting in similar dilutions and distributions in particulate matter and other sources (Khillare et al. 2006; Liu et al. 2009)

The main goal of this research was to investigate the presence of PAHs in surface sediments in Vossoroca Lake, a subtropical lake located in Brazil. Furthermore, the impact of a highway crossing the lake within an environmentally preserved area has been investigated. The impacts of highway include emissions of hydrocarbons by burning fossil fuels and coal tar pavement during highway construction and such impacts are scarcely studied in Brazil. The study was undertaken to determine potential environmental impacts of the highway on the lake considering the toxicity of such compounds, thus making it necessary to know the source of PAHs predominant in sediment. This research has been carried out in the School of Environmental Engineering, Federal University of Parana (Curitiba, Brazil), from September 2010 to February 2011.

Materials and methods

Study area

The Vossoroca Lake is located in the southern area of Sao Jose dos Pinhais city approximately 80 km from Curitiba, the capital of Parana state (south of Brazil). The main portion of the lake is located inside the Environmental Protected Area of Guaratuba (APA). The lake covers an area of 330 ha. The area around the reservoir is predominantly characterized by rural areas for recreation purposes. The upstream part of the lake is strongly influenced by the land that is used for agriculture. The local altitude is 850 m above mean sea level, and the region's climate is classified as humid subtropical with hot summers and winters with frost. The annual rainfall is 1,800–2,000 mm. A main highway crosses the lake via two bridges, linking the southern and southeastern region of Brazil. Although the lake is located in a preserved area, the presence of toxic organic compounds is not discarded as consequence of agricultural activities along the river until reaching the lake.

Sampling campaign

Eight sampling points were established in Vossoroca Lake (Table 1). The sediments were collected with a Van Veen type dredge. The collection campaign was held in October 2010. The samples were packed in plastic bags and kept at about 4 °C until the arrival in the laboratory. In the laboratory, the samples were separated for each analysis for PAHs and aliphatic hydrocarbons.

Table 1 Localization and depth of sampling points in Vossoroca Lake

Site	Latitude (S)	Longitude (W)	Depth (m)
P1 (river)	25°51′01.67″	49°04′36.63″	6.0
P2 (bridge 1NS)	25°50′44.79″	49°04′44.12″	10.5
P3 (bridge 1SN)	25°50′32.73″	49°04′35.35″	10.5
P4 (weathervane)	25°49′56.00″	49°04′25.54″	14.0
P5 (transmission)	25°49′23.28″	49°03′48.23″	16.0
P6 (bridge 2NS)	25°50′29.89″	49°03′12.18″	12.0
P7 (bridge 2SN)	25°50′26.13″	49°03′31.55″	11.5
P8 (float)	25°49′12.56″	49°03′48.12″	17.0

Extraction and analysis of aliphatic hydrocarbons

Polycyclic aromatic hydrocarbons were extracted using 20 g of dried sediment. The compounds were obtained by successive extraction of the sediment with a mixture of dichloromethane (DCM):methanol (2:1) and DCM (2×), by ultra sound for 20 min each. Extracts were combined and rotary evaporated to be dried. The extract was then taken up in 2 mL of DCM. Isolation of compounds was performed in a glass column (1.0 cm i.d. × 8 cm) (effective length) filled with alumina (top) and silica (bottom). The extract was fractionated into two fractions using solvents of increasing polarity: F1 eluted with 18 mL hexane (aliphatic hydrocarbons) and F2 eluted with 20 mL DCM (PAHs). Each fraction was reduced to 1 mL and transferred to small vials (2 mL) and stored frozen until analysis by gas chromatography (GC). The PAHs have been identified and quantified by GC-FID (Agilent 7890), where GC conditions were: injection temperature 270 °C; FID detector temperature 300 °C; carrier gas nitrogen with flow rate 1 mL/min. Temperature program: initially 45 °C, increasing at a rate of 13 °C/min to 150 °C, then at 5 °C/min to 200 °C.

Results and discussion

The strategy to analyze the impact of PAHs distribution on lacustrine surface sediments is based upon interpretation of main results related to molecular weight, toxicity and evaluation from anthropogenic sources. Table 2 shows the PAHs distribution in the sediment samples. The total concentration of PAHs (ΣPAHs) ranged from 9.50 μg/g at site P1 to 29.88 μg/g at sites P5 and P8. Site P1 is located at the location where the river enters the reservoir, and sand dominates the granulometric composition, thus reduced adsorption capacity. The sites P5 and P8 are located in the deepest part of the lake. Most of the suspended matter is transported to this part. Even though the lake is located in a

Table 2 PAHs concentrations in sediments of the Vossoroca Lake in ng/g of dry sediment and molecular ratios of PAHs for defining the source

Compound/ratio	ISQG (ng/g)	P1 (river)	P2 (brigde 1NS)	P3 (brigde 1SN)	P4 (weathervane)	P5 (transmission)	P6 (brigde 2NS)	P7 (bridge 2NS)	P8 (float)
Naphthalene	34.6*	4.69	3.77	3.20	4.64	35.78	74.47	64.44	34.77
Acenaphthylene	5.87*	19.92	26.16	25.10	49.60	50.88	50.42	49.19	50.18
Acenaphthene	6.71*	ND	34.03	30.14	4.38	6.24	11.24	11.84	6.10
Fluorene	21.2	38.79	116.48	106.48	11.36	73.48	7.24	6.74	72.36
Phenanthrene	41.9	21.01	76.91	77.71	62.77	29.03	98.42	96.12	28.01
Anthracene	46.9*	1.47	12.47	11.47	5.00	2.89	68.94	67.90	3.00
Fluoranthene	111	3.14	3.27	2.87	3.17	3.13	3.56	3.18	3.17
Pyrene	53	61.41	65.56	66.55	30.53	45.13	36.51	30.50	45.14
Benzo(a)anthracene	31.7	6,235.22	9,633.52	9,630.02	2,357.11	13,193.38	6,367.79	6,340.02	13,177.22
Chrysene	57.1	ND	17.59	17.80	ND	6.95	6.86	5.09	5.95
Benzo(b)fluoranthene	–	437.66	1,849.52	1,830.52	1,899.42	2,434.19	2,388.10	2,318.10	2,447.19
Benzo(k)fluoranthene	–	392.87	431.76	421.56	231.18	209.37	430.26	431.66	211.01
Benzo(a)pyrene	31.9	186.63	109.97	108.72	421.87	15.57	708.65	712.44	14.27
Indene	–	968.04	4,006.78	4,016.22	3,300.04	4,806.74	3,372.31	3,372.00	4,816.66
Dibenz(a,h)anthracene	6.22*	1,129.28	11,929.10	11,935.19	1,399.41	8,630.69	12,400.24	12,378.04	8,628.88
Benzo(g,h,i)perylene	–	5.91	275.93	277.13	995.67	340.85	1,235.53	1,244.58	342.45
ΣPAHs		9,506.04	28,592.82	28,560.68	10,776.15	29,884.30	27,260.54	27,131.84	29,886.36
Naphthalene/fluoranthene		1.49	1.15	1.11	1.48	11.43	20.92	20.26	10.96
Phenanthrene/anthracene		14.30	6.16	6.77	12.55	10.04	1.42	1.41	9.33
Fluoranthene/pyrene		0.05	0.05	0.04	0.10	0.07	0.10	0.10	0.07
Pyrene/benzo(a)pyrene		0.33	0.60	0.61	0.07	2.89	0.05	0.04	3.16
Benzo(a)pyrene/228		0.82	0.48	0.48	1.84	0.07	3.11	3.12	0.06
Fluoranthene/ (fluoranthene + pyrene)		0.05	0.05	0.04	0.09	0.06	0.09	0.09	0.07
Anthracene/ (anthracene + phenanthrene)		0.06	0.14	0.13	0.07	0.09	0.41	0.42	0.10
LMW/HMW		0.01	0.01	0.01	0.005	0.008	0.01	0.01	0.009

∑*PAHs* total concentration of PAHs, *ISQG* Interim sediment quality guidelines

* Provisional; adoption of marine ISQG developed using the modified NSTP approach

preserved area, the results indicate significant PAHs concentrations (Table 2). The presence of PAHs has been identified as one of the most useful chemical markers for assessing anthropogenic inputs in lakes, rivers, estuarine areas, and in oceans. Liu et al. (2008) found PAHs concentration in river sediments from 0.11 to 1.70 µg/g. In a distinct environment, Fang et al. (2007) observed PAHs in marine sediments ranging from 0.023 to 45.10 µg/g. In addition, Hori et al. (2009) found PAHs in river surface sediments. Nevertheless, its presence of PAHs in water or sediment may not reveal any concern. Its toxicity PAHs depends on a number of factors, including the species of organism, route of exposure, and molecular structure. Generally, PAHs with low molecular weight (LMW) are considered to be acutely toxic and non-carcinogenic to aquatic organisms, whereas, high molecular weight (HMW) are generally considered as not acutely toxic to aquatic organisms, but showing a number of them being carcinogenic (Heitkamp and Cerniglia 1987). The interim sediment quality guidelines (ISQG) were established as a comparative parameter for toxicity capacity of the sediment (CCME 1999). The toxicity, including mortality and sub-lethal effects, occurs at concentrations higher than the ISQGs. Comparisons of found concentrations and ISQG values show that most of the sampled sediment presents PAHs concentrations above the limits. Only fluoranthene and chrysene were found in concentrations below the ISQG limit values. The major concern, however, is related to benzo(a)anthracene which occurred in concentrations 416 times higher than the ISQG value. Similar observations are related to benzo(a)pyrene and dibenz(a,h)anthracene, which occur in concentrations from 13 to 1,993 times higher than the ISQG value, respectively. Although there is a high concentration of these compounds, HMW-PAHs exhibit high values of K_{OC}, thus, they are transferred slowly to the aqueous solution (Froehner et al. 2009). Ecotoxicological studies with marine and freshwater spiked-sediment indicate that toxic levels of PAHs are consistently above than the ISQGs, confirming that the guidelines are reporting concentrations at levels where adverse biological effects will rarely occur.

Higher concentrations of PAHs were generally found at sites close to the highway (P2, P3, P6, P7), probably associated with emissions from cars and trucks or the coal tar pavement. Main anthropogenic sources of PAHs include the incomplete combustion of fossil fuel or wood (pyrogenic sources), and the leakage of oil or petroleum products (petrogenic sources) (Yang et al. 1999; Zakaria et al. 2002). Petrogenic sources, such as fuel oil or refined petroleum products is dominated by LMW-PAHs and had LMW/HMW < 1 (Soclo et al. 2000; Wang et al. 1999). The results showed that PAHs found were predominantly composed by HMW. The ratio LMW/HMW was lower than 1 for all sites investigated, suggesting that PAHs are from petrogenic sources.

The sources are defined by isomer ratios (Sirece et al. 1987; Yunker et al. 2002, 1999). As it was mentioned earlier, one of the objectives was to determine potential environmental impacts of the highway crossing on the lake which is located in an area without industrial activities. Another source of PAHs could be motorized boats of amateur fishery activities. The widely used analysis of molecular compound ratios has been applied to identify the source and origin of the PAHs. The isomer ratios include: naphthalene/fluoranthene, phenanthrene/anthracene, fluoranthene/pyrene, chrysene/benzo(a)anthracene, fluoranthene/(fluoranthene + pyrene), anthracene/(anthracene + phenanthrene), pyrene/benzo(a)pyrene and benzo(a)pyrene/228 (Yunker et al. 2002). Those ratios have been developed for interpreting PAHs compositions and inferring the possible sources (Budzinski et al. 1997). Generally, thermodynamically more stable compounds, such as naphthalene, fluorene, phenanthrene, and chrysene are abundant in petrogenic PAHs, while fluoranthene and pyrene are usually the most abundant compounds is pyrolytic PAHs.

PAHs of molecular mass 178 and 202 are commonly used to distinguish between combustion and petroleum sources (Budzinski et al. 1997; Soclo et al. 2000). Ratios of mass 178 (anthracene/anthracene + phenanthrene) <0.10 usually indicate petroleum while a ratio >0.10 indicates a dominance of combustion products (Budzinski et al. 1997). Applying these rations, the results indicates that most sites relates the PAHs concentrations with petroleum origins. High temperature processes, such as the carbonization of bituminous coal to form creosote (Goyette and Brooks 1998) have PAHs ratios similar to coal tar or coal combustion and generally are indistinguishable from combustion. Accordingly, the anthracene/(anthracene + phenanthrene) ratio limit of 0.10 appears generally applicable under the condition that ratios >0.10 for diesel oil, shale oil, coal and some crude oil samples and <0.10 for lignite, diesel emissions have been reported. Ratios of mass 202, fluoranthene/(fluoranthene + pyrene) of 0.50 are usually defined as the petroleum/combustion transition point (Budzinski et al. 1997). According to this ratio all sites investigated had PAHs from petrogenic source, because all ratios were below 0.50. Ratios of fluoranthene/(fluoranthene + pyrene) above 0.50 have kerosene, grass, most coal and wood combustion sources, while ratios below 0.50 are related to gasoline, diesel, fuel oil and crude oil combustion and emissions from cars and diesel trucks. Thus, the ratio fluoranthene/pyrene greater than 1 implies a pyrolitic origin, while values <1 are attributed to petrogenic sources (Sirece et al. 1987). Ratios of fluoranthene/pyrene were much higher than 1, clearly indicating the petrogenic source of the PAHs in sediments, confirming that petroleum hydrocarbons are the possible main source of PAHs in Vossoroca Lake. In addition, the pyrene/benzo(a)pyrene ratios at different sampling stations ranged from 0.07 to 3.16 proving that most sediment samples were contaminated by petrogenic PAHs.

When entering into the environment, PAHs are widely dispersed through atmospheric and aquatic transport and deposited into the river sediments. A possible source of petrogenic PAHs in sediments is the street dust associated with the leakage of oil from vehicles. However, there is no record of accidental oil spills in the studied area. The street dust can be transported into river sediments via surface runoff due to the high and intense rainfall events. Yang et al. (1999) analyzed the size its distributions for road dust and for engine exhausts of cars and found that road dust was the major contribution of dry deposition flux for a total of 12 PAHs and all individual PAHs. These results infer that the road dust associated with leaked gasoline or other compound derived from petroleum might contribute to LMW-PAHs in the aquatic environment of Vossoroca Lake. However, no conclusive evidence has been found to proof that petrogenic PAHs in sediments from Vossoroca Lake are related to the street dust and associated with leakage of oil from vehicles. Although mixture of source of contamination was already observed elsewhere (Doong and Lin, 2004; Duran and Gonzalez 2009; Wan et al. 2005). Another alternative ratio is benzo(a)anthracene/228, where values up to 0.35 are attributed to combustion products and values lower than 0.20 are related to petrogenic sources (Liu et al. 2009; Soclo et al. 2000). In this study, ratios ranged from 0.005 to 0.35 indicating that the major contribution for PAH in sediments are petrogenic sources.

The fluoranthene/(fluoranthene + pyrene) ratio were below 0.50. Values below 0.50 are associated with gasoline, diesel, fuel oil and crude oil combustion and emissions from cars and diesel trucks, while values above 0.50 are for kerosene, grass, most coal and wood combustion samples and creosote (Yunker et al. 2002; Budzinski et al. 1997). Ratios of benzo(a)anthracene/228 over 0.50 have been taken to indicate combustion while ratios below 0.50 have been attributed to low temperature diagenesis (Soclo et al. 2000). The data summarized in Table 2 suggests that 0.50 is probably too high for the diagenesis/combustion transition, and that BaA/228 ratios lower than 0.20 imply petroleum, and values from 0.20 to 0.35 indicate either petroleum or combustion, and values higher than 0.35 imply combustion. In similar cases, and urban locations a benzo(a)anthracene ratios higher than 0.35 correspond almost always to fluoranthene/(fluoranthene + pyrene) ratios higher than 0.40. However, in remote and light-urban locations fluoranthene/fluoranthene + pyrene ratios again strongly imply combustion, the benzo(a)anthracene/288 ratios are primarily lower than 0.35 and suggest mixed sources. If this can be confirmed, the benzo(a)anthracene/288 ratios may provide a more definitive indicator of vehicle emissions than any other PAHs ratio considered here. Samples from the remote and light-urban regions with benzo(a)anthracene/228 ratios >0.35 usually come from sites close to highways or urban areas (Headley et al. 2002). In particular, recent sediments from Moose Lake (Metre et al. 2009), which has a major transport corridor along its north side, show elevated benzo(a)anthracene.

PAHs of molecular masses 228 and 276 are used less frequently as parent PAHs indicators and few guidelines have been established for their interpretation (Yunker et al. 1999, 2002). The higher weight PAHs are usually found in less percentage in refined petroleum products (Headley et al. 2002; Wang et al. 1999) and generally are present in significant amounts only in higher fractions such as asphalt, and possibly in bitumen or coal. Hence, the large variation in petroleum composition for both benz(a)anthracene, chrysene, indene(1,2,3-cd)pyrene, and benzo(g,h,i)perylene likely is a combination of heterogeneous samples and analytical variability due to low concentrations. Because very low proportions of benzo(a)anthracene or indene(1,2,3-cd)pyrene are rarely observed in combustion samples, a BaA/228 ratio less than about 0.20 likely indicates petroleum. However, there is an intriguing observation: the high concentration of benzo(a) anthracene and dibenzo(a,h)anthracene. According to literature (Mahler et al. 2005), particles in runoff from parking lots with coal tar-based sealcoat and asphalt might account for the majority of stream PAHs loads. Also, lake sediments, inserted in parks with seal coated pavements, presented high concentration of PAHs. In the present case, due the location of the studied lake, crossed by a highway road with intense traffic and the existence of low quality coal tar for pavement can justify the high concentration of PAHs in lake sediments. Furthermore, the high concentration of benzo(a)anthracene, indene(1,2,3-cd)pyrene, and dibenzo(a,h)anthracene could be attributed to coal tar sealcoat and asphalt used in highway pavement. In fact, visual evidence of offsite black flecks in gutters and on sidewalks adjacent to seal coated pavements has been observed. Differences in PAHs assemblages can be investigated by computing ratios of selected PAHs as discussed above, however, the fluoranthene/pyrene and indene (1,2,3-cd)pyrene/benzo(ghi)perylene ratios indicate a coal tar and asphalt contribution (Metre et al. 2009; Mahler et al. 2005). The pavement is basically made up of asphalt and frequently repaired due to high traffic.

Conclusions

The analysis of the PAHs in the upper sediments of the Vossoroca Lake has shown that the total concentration of these compounds and their relative distributions are rather homogeneous within the studied area. Comparisons with other results described in the literature indicates that this area appears to be slightly polluted by PAHs from an ecotoxicological point of view. According to the molecular ratios, most of PAHs present in sediments came from vehicle emissions as a consequence of the highway crossing the lake. The results showed high PAHs concentrations of PAHs with high molecular weight, especially for benzo(a)anthracene, dibenz

(a,h)anthracene, and indene. According to the literature, these compounds can be associated to coal tar pavement under certain circumstances. When considering the location of the studied area, the main source of hydrocarbons are combustion products of petroleum (gasoline and diesel oil), however such products do not have significant occurrence of HMW-PAHs in their composition. It has been concluded that the reservoir is far from being polluted.

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