Distribution and Source of Sedimentary Polycyclic Aromatic Hydrocarbon (PAHs) in River Sediment of Jakarta

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ABSTRACT

In this study, the distribution and source identification of sedimentary PAHs from 13 rivers running through Jakarta City were investigated. Freeze-dried sediment samples were extracted by pressurized fluid extraction and purified by two-step of column chromatography. PAHs were identified and quantified by gas chromatography-mass spectrometry (GC-MS). High concentrations of PAHs, ranging from 1992 to 17635 ng/g-dw, were observed at all sampling locations. Ratios of alkylated PAHs to parent PAHs exhibited both petrogenic and pyrogenic signatures with predominantly petrogenic inputs. High hopanne concentrations (4238-40375 ng/g dry sediment) supported the petrogenic input to Jakarta's rivers. The high concentration of PAHs is indicator for organic micropollutant in the aquatic urban environment in Jakarta that may have the potential to cause adverse effect to the environment.

Keywords: *polycyclic aromatic hydrocarbons; organic micropollutant; water pollution*

ABSTRAK

Dalam penelitian ini, distribusi dan identifikasi sumber PAH sedimen yang diperoleh dari 13 sungai yang melewati kota Jakarta telah dianalisis. Sedimen yang telah dikeringbekukan diekstraksi mengunakan teknik pressurized fluid extraction, dan dimurnikan melalui dua tahap kolom kromatografi. Ekstrak yang diperoleh diidentifikasi dan ditentukan konsentrasinya menggunakan kromatografi gas-spektrometri massa. Konsentrasi PAH yang tinggi mulai dari 1992 sampai 17635 ng/g-dw ditemukan pada hampir semua lokasi sampling. Rasio antara PAH alkil dan PAH induk menunjukkan sumber PAH yang berasal dari petrogenik dan pirogenik, dengan sumber PAH petrogenik yang lebih dominan. Konsentrasi hopan yang tinggi (4238-40375 ng/g) mendukung dugaan adanya sumber petrogenik ke sungai-sungai yang ada di Jakarta. Konsentrasi PAH yang tinggi dapat digunakan sebagai indikator adanya polutan organik di lingkungan perairan Jakarta yang mempunyai potensi untuk memberikan pengaruh yang berbahaya pada lingkungan.

Kata Kunci: *polisiklik aromatik hidrokarbon; mikropolutan organik; pencemaran air*

INTRODUCTION

PAHs (polycyclic aromatic hydrocarbons) constitute a class of persistent organic pollutants that are widely distributed in every environmental medium, including the atmosphere, water, soil, sediment and organisms. The United States Environmental Protection Agency (USEPA) has listed 16 PAHs as priority pollutants because they exhibit mutagenic, carcinogenic and persistent [1]. PAHs also have the ability to affect the endocrine system of humans and animals [2] and increase lung cancer risk from the inhalation system exposure to PAHs [3-4].

PAHs are introduced to the environment through atmospheric transport and deposited into the river sediments or directly discharged from leakage of crude oil and refined product, urban runoff which contains street dust, industrial effluent and municipal waste water. Rivers is one of the main contributor sources of PAHs to sea [5-6]. Because of their hydrophobic nature, PAHs have a tendency to associate with particulate matter in the aquatic environment, with underlying sediment as their ultimate sink. The composition of the organic pollutants found in sediment reflects the relative contributions from different sources, both natural and anthropogenic [7].

Natural sources of PAHs include volcanic eruption, forest fires, and biological precursors during early diagenesis. However, anthropogenic sources of PAHs are of the greatest concern because they are the primary sources of PAHs in the environment. Anthropogenic sources of PAHs include the combustion

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of organic matter from wood or fossil fuels as well as biomass. Forest fires, vehicle engines combustion by gasoline or diesel fuel, industrial operations and garbage incineration are classified as pyrogenic sources [8]. The other sources are petrogenic, including crude oil and petroleum products such as kerosene, gasoline, diesel fuel, lubricating oil and asphalt. Climate conditions and socio-economic circumstances could be some of the factors affecting the distribution of PAHs. Both petrogenic and pyrogenic PAHs were recorded in many studies [8-11]. Studies of these compounds have been conducted in many countries, but there is only limited data from Indonesia, although industrialization, urbanization, and motorizations have been increasing significantly since several decades ago. Data on PAHs as one of organic micropollutant are essential for the effective control of input pollutant and as the basis for action plans to reduce them.

There is a lack of available information on the occurrence of PAH contamination in Jakarta's rivers. This study presents a comprehensive survey of PAHs in the main river sediments in Jakarta, including the Cakung River, the Sunter River, the Ciliwung River, the Barat Banjir Canal, the Angke River, the Kamal River, the Cengkareng Drain and the Moonkaveret River. The objectives of this study were to investigate the pollution level and distribution of PAHs and to assess the possible sources of PAHs in Jakarta rivers. To obtain more information on petrogenic PAHs, hopanes, a group of hydrocarbons, were included as target pollutant. The pentacyclic triterpenes are ubiquitous components in crude oil and petroleum products. Hopanes consist of a range of homologs from C_{27} to C_{35} with various stereoisomers. The compositions of members of this compound class vary depending on origin, whether presence of plant-derived materials, and the maturity of the petroleum stages. These characteristics, as well as their resistance to environmental alteration, provide a fingerprint for the identification of the sources of oils [8]. This study could reveal the level and inputs of PAHs as well as the effectiveness of countermeasures or policies created to address the problems of organic micropollutant.

EXPERIMENTAL SECTION

Materials

Surface sediment samples were collected at 13 locations in Jakarta's rivers (JKSE 1, JKSE 2, JKSE 3, JKSE 4, JKSE 5, JKSE 6, JKSE 7, JKSE 8, JKSE 9, JKSE 10, JKSE 13, JKSE 14 and JKSE 25) from September–October, 2010. Sampling location maps are available in Fig. 1. All samples were collected using an Ekman dredge. The sediments (top 2 cm) were taken with a stainless steel scoop and kept in a stainless steel container. The samples were stored at -30 °C and freeze-dried before analysis.

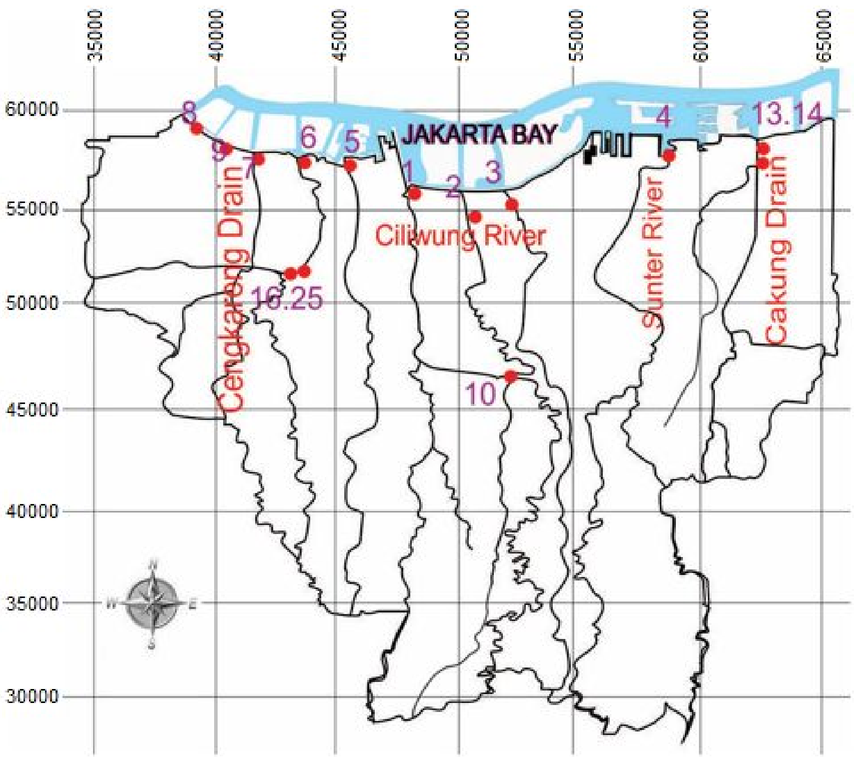

Fig 1. Sampling location in Jakarta river sediment

Instrumentation

All samples were extracted using Dionex ASE 200 (Accelerated Solvent Extraction) and identified by Gas Chromatography-Mass Spectrometry (GC-MS) using a 30 fused silica capillary column (HP-5MS) installed in a GC (HP 5890) interface with a Hewlett Packard 5972 A quadrupole mass-selective detector (SIM Mode).

Procedure

All samples were analyzed for PAHs and hopanes. Details of the extraction and purification are described elsewhere [8]. Briefly, 1 to 2 g of freeze-dried sediment was extracted by pressure fluid extraction in a Dionex ASE 200 accelerated solvent extractor using a 3:1 mixture of dichloromethane:acetone (v/v). The extract solutions were spiked with PAH and LAB surrogates. The PAH surrogates consisted of deuterated PAHs (i.e., anthracene-d10, p-terphenyl-d14, benz[a]anthracene-d12). The extracts were purified by two-step silica gel column chromatography and analyzed by GC/MS.

It was monitored 15 parent PAH species in selected ion monitoring (SIM) mode at *m/z* = 178 (phenanthrene [Phe], anthracene [Anth]), 190 (4H-cyclopenta[*def*] phenanthrene [CPP]), 202 (fluoranthene [Fluo], pyrene [Pyr]), 228 (benz[*a*]anthracene [BaA], chrysene [Chry]), 252 (benzo[*b*]fluoranthene [BbF], benzo[*j*]fluoranthene + benzo[*k*]fluoranthene [BF], benzo[*e*]pyrene [BeP], benzo [*a*]pyrene [BaP], perylene [Pery]), 276 (indeno[1,2,3-*cd*]pyrene [IndPy], benzo[*ghi*]perylene [BghiP]), and 300 (coronene [Cor]). It was also monitored 12 methylated PAHs in SIM mode at *m/z* = 192 (3-, 2-, 9-, and 1-methylphenanthrene [3-, 2-, 9-, 1-MP] in order of elution), 216 (3 peaks of methylpyrenes [MPy] or methyl-Fluo), and 242 (5 peaks of methylchrysenes [MC] or methyl-BaA). The details of identification and quantification have been described previously [3]. The sum of the concentrations of 14 of the parent PAH species (i.e., Phe, Anth, CPP, Fluo, Pyr, BaA, Chry, BbF, BF, BeP, BaP, IndPy, BghiP, Cor) is expressed as Σ14PAH. As this research focuses on anthropogenic activities, we excluded perylene from the sum. Perylene is derived from biological precursors during early diagenesis [6]. The sum of all 26 PAH species except perylene is written as Σ26PAH. The sum of the 14 parent PAHs and the 4 MPs is written as Σ18PAH. The ratio of the sum of MPs to Phe is written as MP/P.

Hopanes were monitored in SIM mode at *m/z* = 191. As analytical standards, we used 17α(H)-22,29,30-trisnorhopane (Tm), 17α(H),21α(H)-norhopane (C_{29}17α), 17β(H),21α(H)-norhopane (C_{29}17β), 17α(H),21α(H)-hopane (C_{30}17α), 17β(H),21α(H)-hopane (C_{30}17β), 18α-oleanane, 18β-oleanane, and 17α(H),21β(H)-homo hopane (C_{31}17α). Individual hopanes were quantified by comparing the integrated peak area of the selected ion with the peak area of the internal injection standard (IISTD: 17β,21(H)β-hopane).

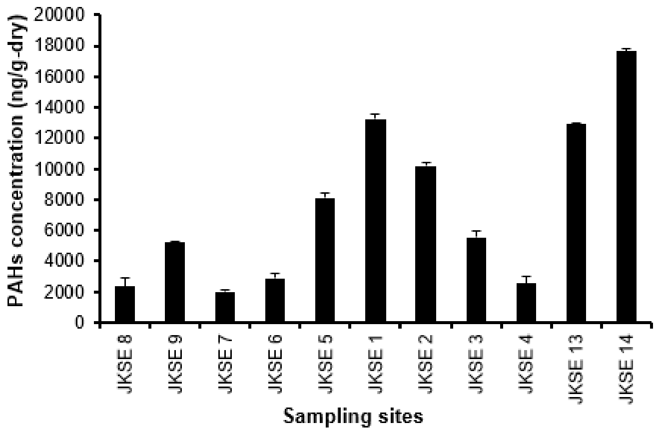

Fig 2. PAHs concentration in Jakarta river sediment

RESULT AND DISCUSSION

Level and Distribution of PAHs

The number of sediment sample collected is 14 samples, however we only analyzed 11 samples sample. Sediment samples from location 16, 25, 10 were not analyzed due to condition of sample containing only sand material indicating undetectable level of PAH. The total PAH concentrations in sediment collected from Jakarta's rivers are shown in Fig. 2. The total PAH concentrations describe the sum of all PAH compounds (Σ26PAH). Perylene was excluded because this research focuses on anthropogenic activities only, and perylene has been discussed as a natural PAH. The spatial distribution was observed at different sampling locations and ranged from 1,992 to 17,635 ng/g dry sediment with a mean concentration of 7147 ng/g. In general, the PAH concentrations in the sampling sites in the eastern region (JKSE 13 and JKSE 14 except JKSE 4) and central region (JKSE 5, JKSE 1, JKSE 2 and JKSE 3) were higher than those in the western region (JKSE 25, JKSE 8, JKSE 7, JKSE 6, except JKSE 9).

JKSE 13 and 14 are situated downstream of the Cakung Drain, which flows through the largest industrial area and a densely populated area in Jakarta. Various industries, such as chemical, plastic, rubber, and metal industries, with small, medium and large capacities are located in the industrial area, which covers approximately 1,000 ha. JKSE 4 is located at the mouth of the Sunter River. The PAH concentrations at this site were low, even though this river is also heavily polluted. The low PAH concentrations were most

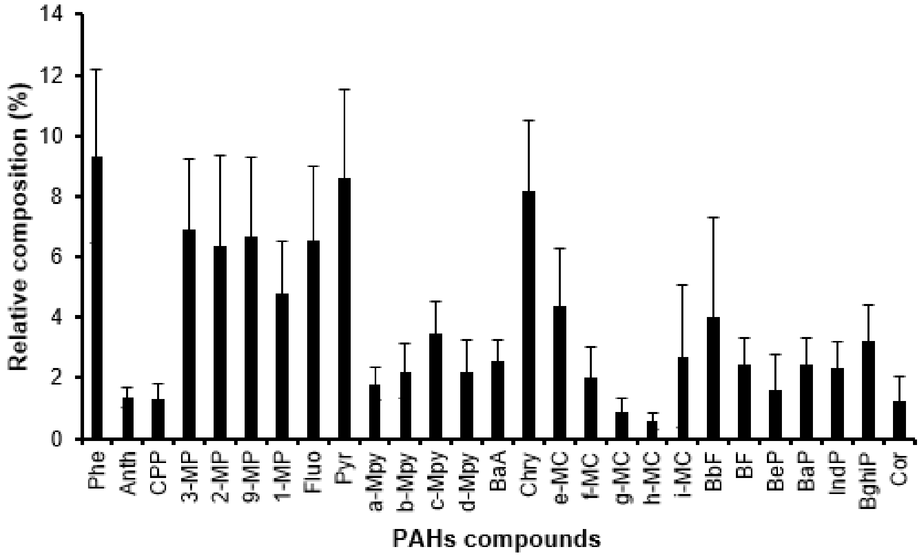

Fig 3. PAHs profile in Jakarta river sediment

likely due to the strong river current. The central region of the sampling sites (JKSE 5, JKSE 1, JKSE 2 and JKSE 3), which also contain high PAH concentrations, is located downstream of the Ciliwung River. The high total PAH concentrations in this region are most likely due to their location, which is a heavily populated, urbanized, and industrialized area situated in the middle stream and downstream of the Ciliwung River, where a greater potential exists for loading of PAHs from urban activities into the river. This river is one of the main rivers flowing through Jakarta. There are several rivers passing through Jakarta City, but this river is the largest, representing the main waterway in the city. Locations JKSE 25, JKSE 8, JKSE 7, and JKSE 6 are located in rivers running through areas that are less densely populated, less commercial and less industrial, and a correspondingly lower potential for PAH loading from this region is observed. PAH concentrations at location JKSE 9 were the highest among the sampling sites in the western region, probably due to the location of the sampling site near industrial areas.

The level of sedimentary PAHs can be classified into four categories: (1) low, 0-100 ng/g; (2) moderate, 100-1,000 ng/g; (3) high, 1,000-5,000 ng/g; (4) very high, >5,000 ng/g) [12]. The contamination level of PAHs in Jakarta riverine sediments fell within the range of high levels (1,000-5,000 ng/g) to very high levels (>5,000 ng/g). The total concentration of PAHs from this study was higher than that of most rivers in Malaysia [13-15], Luan River estuary, China [16] and Yangtze River Estuary, China [17]. The highest total PAHs concentration in Jakarta's river was lower than that Bharalu River, India [18] and Scarpe Rivers, Northern France [19].

Sources of PAHs

Fig. 3 displays the profile of PAHs and presents the composition of the PAH compounds. The high concentrations of low molecular weight (LMW) and alkyl PAHs have been attributed to petrogenic sources of PAH contaminants, including crude oil and its refined products such as gasoline, diesel fuel and lubricating oil. The PAH profile of Jakarta riverine sediments is similar to that of petrogenic sources with relatively abundant alkyl PAHs. This profile demonstrates that there was a heavy impact of petrogenic sources. The PAH profile of Jakarta riverine sediments contrasts with that of industrial countries which showed a severe depletion in alkylated PAHs [7,10,20]. Petrogenic sources of PAHs normally consist of alkylated PAHs and lower molecular weight (LMW) PAHs with severe depletion of higher molecular weight (HMW) PAHs, while pyrogenic sources are abundant in HMW PAHs [8,11]. Although the PAH profile of the Jakarta riverine sediments had abundant alkyl and LMW PAHs, a considerable amount of HMW PAHs was detected, indicating the input of both petrogenic and pyrogenic sources.

To express the abundance of alkyl PAHs more quantitatively, the ratio of alkyl PAHs to parent PAHs was calculated. The ratio of the sum of MPs to Phe is written as MP/P. The ratio of (MPy + methyl-Fluo) to (Pyr + Fluo) is written as MPy/Py. The ratio of (MC + methyl-BaA) to (Chry + BaA) is written as MC/C. Lastly, the ratio of the sum of all methyl-PAH species to (Phe + Pyr + Fluo + Chry + BaA) is written as MPAH/PAH. A previous analysis of potential source materials determined the thresholds of MP/P, MPy/Py, MC/C,

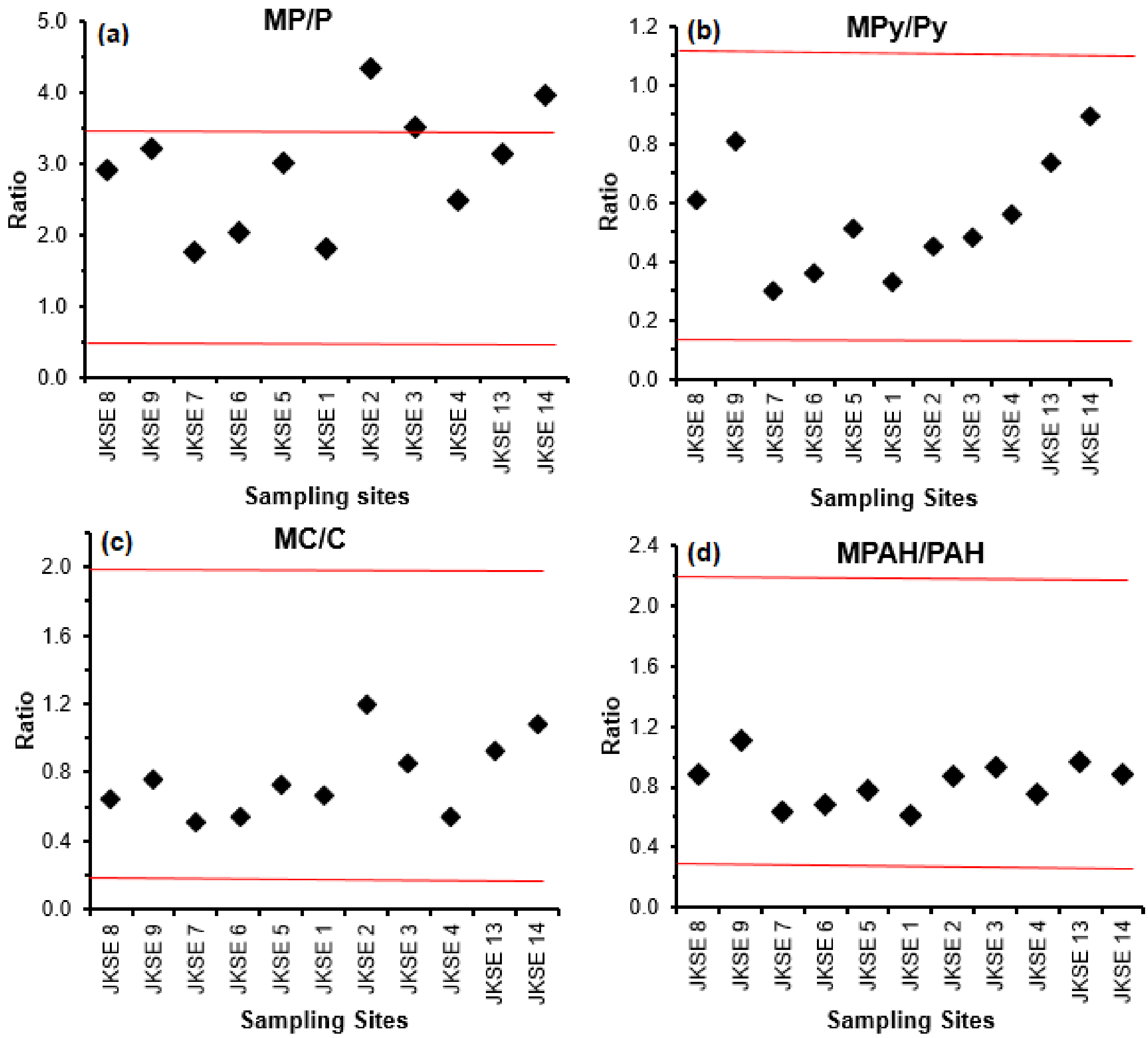

Fig 4. The ratio of alkyl PAHs to parent PAHs in Jakarta river sediment: (a) MP/P, (b) (MPy/Py), (c) MC/C, (d) MPAH/PAH

and MPAH/PAH for exclusively pyrogenic and petrogenic signatures [8]. When the ratio is lower than the pyrogenic threshold, an exclusively combustion origin is indicated; when the ratio is higher than the petrogenic threshold, an exclusively petrogenic origin is indicated. The threshold values for MP/P (exclusively pyrogenic – exclusively petrogenic) are as follows: MP/P, 0.5–3.5; MPy/Py, 0.15–1.5; MC/C, 0.2–2.0; and MPAHs/PAHs, 0.3–2.2. Values between the two thresholds indicate a mixture of pyrogenic and petrogenic contributions.

The MP/P ratios in all samples of the riverine sediments (1.8-4.4) were greater than 0.5, as a minimum threshold (Fig. 4a). Locations JKSE 14 and JKSE 2 had MP/P ratios greater than 3.5 (maximum threshold for exclusively petrogenic), and locations JKSE 13, JKSE 3, JKSE 5, JKSE 9, and JKSE 8 showed MP/P ratios near the threshold of the petrogenic signature, indicating a heavy impact of the petrogenic signature. The other locations showed MP/P ratios in the range of a mixture of petrogenic and pyrogenic sources, suggesting both petrogenic and pyrogenic contributions. The MPy/Py ratios showed that all sampling sites had within a range of 0.3–0.9, which were larger than the minimum threshold of 0.15 and lower than the maximum threshold of 1.5. This profile indicates that the sediments in this region were also impacted by a mixture of petrogenic and pyrogenic sources (Fig. 4b). All samples had MC/C ratios ranging from 0.5 to 1.2, indicating a mixed input of petrogenic and pyrogenic sources (Fig. 4c). Locations JKSE 13, 14 and 2 have higher ratio that that of others location, supported petrogenic input there. The MPAH/PAH ratios for this region ranged between 0.5–1.1 (Fig. 4d), which were

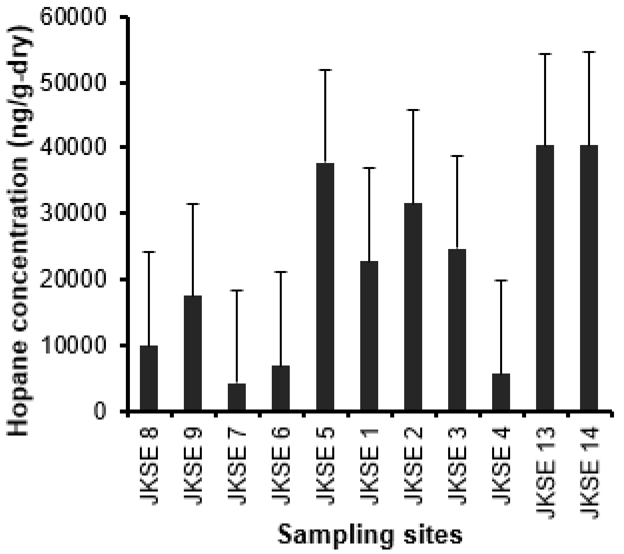

Fig 5. Hopanes concentration in Jakarta river sediment

higher than the minimum threshold of 0.3 and lower than the maximum threshold of 2.2. Overall, the high ratios of alkyl PAHs to parent PAHs exhibited both petrogenic and pyrogenic contributors with predominant petrogenic inputs in some locations. This result also demonstrates that there were no exclusively pyrogenic sources. The petrogenic input were attributed to spill oil and fuel, while the pyrogenic signature may be related to extensive use of fossil fuel with combustion from motor vehicles in Jakarta City. Motor vehicles, the numbers of which have increased significantly during the last two decades, were identified as the main agent of pollution in Jakarta.

The hopane concentrations in Jakarta riverine sediments ranged from 4,238 to 40,375 ng/g (Fig. 5). The highest hopane concentration at location JKSE 13 and JKSE 14 supported the predominance of petrogenic inputs, suggesting that some local petrogenic sources, such as fuel and oil from fishing boats or illegal oil disposal, might contribute in this area. High hopane concentrations were also detected at JKSE 5, JKSE 1, JKSE 2 and JKSE 3, confirming the previous analysis indicating that those locations demonstrated a large input of petrogenic sources. Hopane concentrations in other locations (JKSE 25, JKSE 9, JKSE 7, JKSE 6, and JKSE 4) were lower than those in the previous area. Overall, the high hopane concentration in most sampling locations supported the petrogenic input to the riverine sediments of Jakarta.

CONCLUSION

High concentrations of PAHs were observed at all sampling locations in sediments from 13 rivers that run through Jakarta City. Ratios of alkylated PAHs to parent PAHs exhibited both petrogenic and pyrogenic signatures with predominantly petrogenic inputs while high hopane concentrations supported the petrogenic input to Jakarta's rivers. The high concentration of PAHs is one of indicator function for high organic pollutant in the aquatic urban environment in Jakarta.

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