Distribution, source identification, and historical trends of organic micropollutants in coastal sediment in Jakarta Bay, Indonesia

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ABSTRACT

We determined concentrations of polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), linear alkylbenzenes (LABs), and hopanes in coastal sediments collected from Jakarta Bay and Tokyo Bay. PAH concentrations in sediments from Jakarta Bay (257–1511 ng/g-dry) were lower than or comparable to those from Tokyo Bay (1372–1615 ng/g-dry). Ratios of alkyl-PAHs to parent PAHs showed a greater contribution of petrogenic inputs in Jakarta Bay than in Tokyo Bay. This difference is consistent with the higher ratio of hopanes to PAHs in Jakarta Bay. LAB concentrations in Jakarta Bay (geometric mean, 1400 ng/g-dry) were higher than those in Tokyo Bay (661 ng/g-dry). The internal to external (I/E) ratios of LABs in Jakarta Bay (0.92–2.88) were lower than those in Tokyo Bay (2.8–4.8), indicating that Jakarta Bay receives untreated or poorly treated sewage. Significant amounts of tetrapropylene-based alkylbenzenes were detected in several locations in Jakarta Bay, suggesting current usage of the non-degradable surfactants alkylbenzene sulfonates that are banned in many countries. The PCB concentration in Jakarta Bay was 1 order of magnitude lower than in Tokyo Bay, suggesting minimal usage of PCBs in industrial or commercial products in Jakarta. Analyses of a sediment core indicate increasing inputs of PAHs, hopanes, and LABs into Jakarta Bay during recent decades.

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1. Introduction

Coastal sediments accumulate both natural and anthropogenic compounds from land-based sources [1]. Huge amounts of domestic wastes, industrial effluent, and urban runoff are carried to coastal areas by rivers. Organic pollutants are important components of these land-derived wastes. Some of these compounds are sorbed to particulate matter [2] and settle in the sediment. The composition of organic compounds found in sediment reflects the relative contributions from different sources, both natural and anthropogenic [3]. Concerns have been raised about organic micropollutants such as polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) owing to their toxicities and persistence. Studies of these compounds have been conducted in many industrialized countries [4–7], but there is only limited data from tropical Asian countries, including Indonesia, although industrialization, urbanization, and motorization have been increasing dramatically. Data on organic micropollutants are essential as the basis for action plans to reduce them.

PAHs are hydrocarbons that contain two or more fused benzene rings and are the foremost widespread class of organic pollutants. PAHs have adverse effects on human health and the environment due to their mutagenic, carcinogenic, and endocrine-disrupting activities [8–10]. PAHs are derived primarily from anthropogenic sources, but also from natural sources. Anthropogenic sources of PAHs can be categorized into two groups. The first group is pyrogenic which is generated during combustion of organic matter such as in biomass burning, forest fires, internal combustion engines, industrial operations, and garbage incineration. Coal tar, which has been proposed as a major source of PAHs in sediments from the eastern and central USA [11], is classified as a pyrogenic source. The second group is petrogenic which is derived from crude oil and petroleum products such as kerosene, gasoline, diesel fuel, lubricating oil and asphalt. Pyrogenic sources are predominant in sediments in many industrialized countries [3,12,13], whereas petrogenic sources are predominant in tropical Asian countries [14–16]. Climate and socio-economic conditions could explain the difference.

Hopanes constitute a group of pentacyclic triterpane hydrocarbons and are ubiquitous components of crude oil and petroleum products. They consist of a range of homologs from C_{27} to C_{35} with various stereoisomers. Their composition depends on the origin

of the crude oil (e.g., oceanic phytoplankton, higher plants) and the maturity stage of the petroleum. This characteristic, as well as their resistance to environmental alteration provide a fingerprint to identify sources of oils [17–20].

PCBs are a class of organic compounds in which 1–10 chlorine atoms are attached to a biphenyl. PCBs used to be commercially produced as complex mixtures of congeners with different degrees of chlorination for a variety of applications, including dielectric fluids in transformers and capacitors, as coolants, and as additives in pesticides, paints, sealants, and adhesives. Their commercial utility was based largely on their chemical and physical properties, including stability, low flammability, and electrical insulating properties [21]. But because of their stability and lipophilic properties, PCBs are persistent after their discharge to the environment and tend to accumulate in organisms, including humans. Although their production and use were banned in the 1970s, PCBs are still detected in the environment.

Linear alkylbenzenes (LABs), with a C_{10}–C_{14} normal alkyl chain, are the raw materials used to produce linear alkylbenzenesulfonates (LASs). LABs are industrially sulfonated to produce LASs, which are anionic surfactants used widely as domestic detergents. Small but significant amounts (1–3%) of unreacted LABs are carried over in detergent and are released to waters through the discharge of domestic wastes. LABs are detected in environments affected by sewage and have been proposed as molecular markers to identify sources of sewage pollution [22–24].

We performed a preliminary survey of organic micropollutants in sediments from tropical Jakarta Bay, Indonesia. Jakarta Bay (05°09′S, 106°50′E), situated north of Jakarta, covers approximately 500 km^2. It is a large coastal ecosystem encompassing several small coral islands that provides many ecosystem functions such as supporting human well-being and economic development. However, this shallow bay receives freshwater runoff from 13 rivers running through Jakarta city, whose population reached 9.5 million in 2010 [25]. The bay thus receives domestic wastes, industrial effluent, urban runoff, shipping wastes and spills with severe pollution potential. For comparison, we also surveyed sediments from Tokyo Bay, Japan, with a population of 26 million in its catchment, which discharges large amounts of treated wastewater into the bay. Majority (~90%) of wastewater generated in the catchment is subjected to secondary treatment before discharge to the bay and adjacent waters. The greater intensity of industrialization and the much more extensive and modern wastewater treatment found in Tokyo Bay make an interesting counterpoint to the setting in Jakarta Bay. The distribution, sources, and behavior of the micropollutants, including markers (LABs and hopanes) targeted in the present study have been well documented in Tokyo Bay [14,16,23,26]. Thus, Tokyo Bay would be a concrete benchmark for the comparison. The results were also compared with those of coastal zone sediments from other regions of the world.

The main objectives of this research were: (1) to investigate the concentrations and distribution of sedimentary PAHs and PCBs in Jakarta Bay in comparison with Tokyo Bay; (2) to characterize the sources of the sedimentary PAHs; and (3) to gauge the magnitude of sewage input by using LABs as molecular markers. To reveal the historical trend, we also analyzed a sediment core.

2. Materials and methods

2.1. Sample collection

We collected surface sediment samples at seven locations in Jakarta Bay (JK1–JK7) in August 2007 and at two locations in Tokyo Bay (TB1, TB2) in December 2009 (Fig. 1, Table S1). All samples comprised the top 2 cm of sediment cakes collected with an Ekman
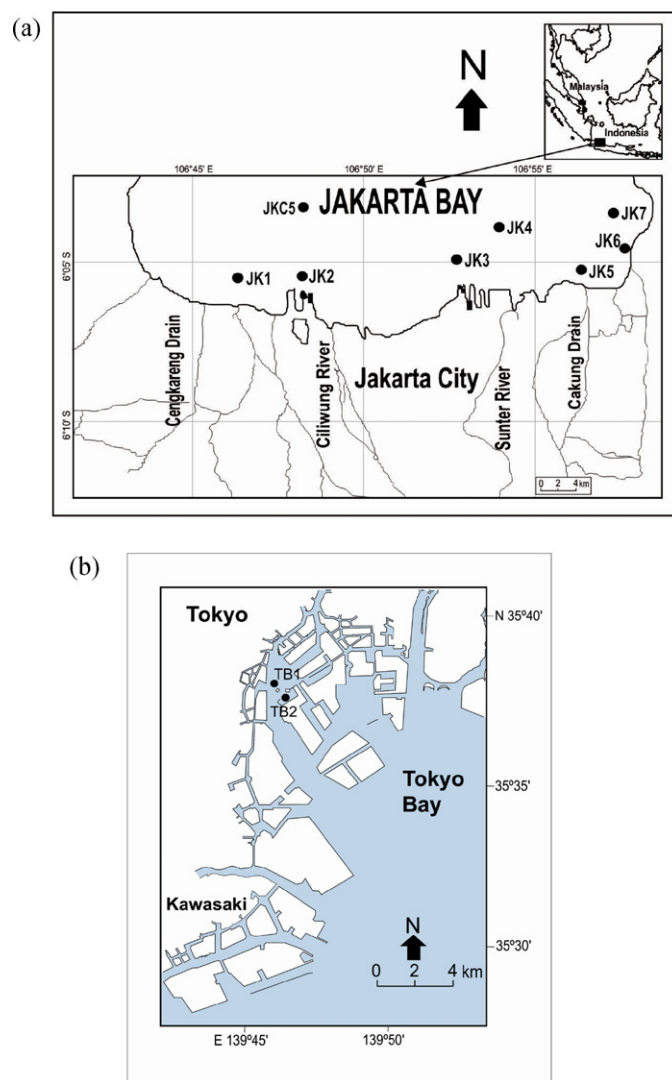
Fig. 1. Sampling locations in (a) Jakarta Bay and (b) Tokyo Bay.

dredge or a Smith–McIntyre sediment sampler. The sediment core was collected further offshore (JKC5) in Jakarta Bay with a gravity corer consisting of an acrylic pipe (11 cm i.d. × 50 cm length) with a stainless steel sediment catcher and weight (20 kg). The sediment core was sliced into 5-cm intervals. The samples were stored at −30 °C and freeze-dried before analysis.

2.2. Analytical procedures

All samples were analyzed for PAHs, hopanes, LABs, and PCBs. Details of the extraction and purification are described elsewhere [14–16]. Briefly, 1–2 g of freeze-dried sediment was extracted by pressure fluid extraction in a Dionex ASE 200 accelerated solvent extractor with a 3:1 mixture of dichloromethane:acetone (v/v). The extract solutions were spiked with PAHs and alkylbenzenes (ABs) surrogates. PAHs surrogates consist of deuterated PAHs (i.e., anthracene-d_{10}, p-terphenyl-d_{14}, benz[a]anthracene-d_{12}). The AB surrogate consists of 1-C_n ABs (n = 8–14). Structures of ABs are expressed as "m-C_n", where "m" means the phenyl substitution position on the alkyl chain and "n" means the number of alkyl carbons. 1-C_n ABs are not detected in commercial detergents or environmental samples, as described in our previous paper [24] and; therefore, were used as the surrogates. The extracts were purified by two-step silica gel column chromatography. Hopanes,

PAHs, and LABs were analyzed by gas chromatograph (GC)–mass spectrometer, and PCBs by GC equipped with an electron capture detector (GC-ECD) [15,26].

We 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]). We 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 were described previously [16]. 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 Σ_{14}PAH. As this research focuses on anthropogenic activities, we excluded perylene from the sum. Perylene is derived from biological precursors during early diagenesis [27,28,12]. The sum of all 26 PAH species except perylene is written as Σ_{26}PAH. The sum of the 14 parent PAHs and the 4 MPs is written as Σ_{18}PAH. 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.

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}$17β), 18α-oleanane, 18β-oleanane, and 17α(H),21β(H)-homohopane (C$_{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).

We quantified LAB congeners by comparing the integrated peak area of the selected ion ($m/z = 91$, 92, 105) with the peak area of the IISTD (biphenyl-d$_{10}$, $m/z = 164$). Synthetic detergents and environmental samples contain 26 m-C$_n$ AB congeners of non-terminal phenyl position (i.e., $m \geq 2$) with a C$_{10}$–C$_{14}$ linear alkyl chain (i.e., $n = 10$–14) [29]. The 26 congeners were measured and the sum of the 26 congeners was termed as ΣLABs. Fifteen peaks of tetrapropylene-based alkylbenzene (TAB) congeners with branched alkyl chain were identified and quantified as described by Tsutsumi et al. [29]. The ratio of the sum of TABs to LABs is expressed as %TABs.

We determined 26 major PCB congeners (CB# 8/5, 18, 28, 52, 44, 66/95, 90/101, 110/77, 149, 106, 118, 132/153, 105, 138/160, 187, 128, 180, 170/190, and 206). They were quantified by comparing the height of the peaks of standard solution as described in our previous papers [17,26].

PAH and alkylbenzene concentrations were recovery-corrected against the surrogate standards (deuterated PAHs and 1-C$_n$ ABs) spiked just after extraction. The extraction efficiency was confirmed through the analysis of marine sediment Standard Reference Material (SRM1941b) from the National Institute of Standards and Technology, USA. PAH concentrations obtained by our methods were >80% of the certified values except for coronene, which was 50%. Ranges of the recoveries of the LABs and PAH surrogates were 82–106% and 77–97%, respectively, throughout the sample analyses. PCB concentrations were not recovery-corrected because of the unavailability of appropriate surrogates for PCB measurement by GC-ECD. The recoveries of the PCBs eluted in the same
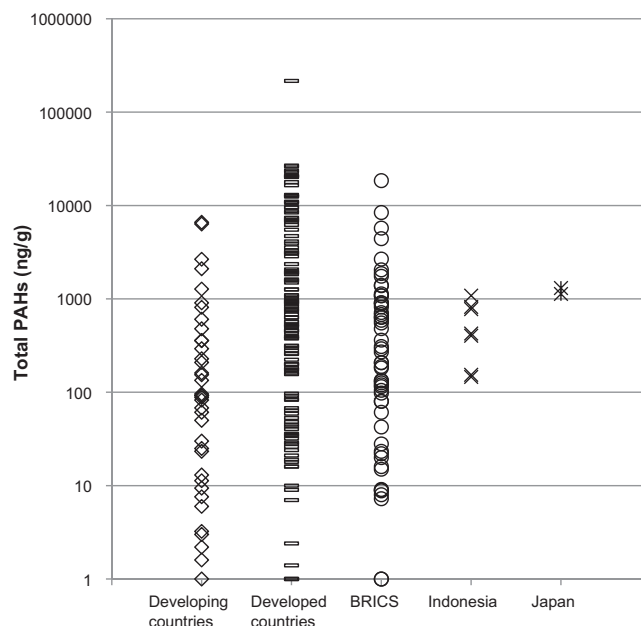

Fig. 2. Comparison of PAH concentrations in Indonesian and Japanese sediments with those in coastal sediments from developing countries, developed countries, and BRICS countries (Brazil, Russia, India, China and South Korea). Data for global comparison were derived from references cited in Table S2 in the Supporting Materials.

fraction as LABs whose recoveries were monitored by using 1-C$_m$ ABs, whose recoveries were satisfactory in all samples. Hopane concentrations were not recovery-corrected owing to minimal possibility of loss during analysis due to their non-volatile, non-polar nature. In advance of analysis, the reproducibility and recovery of all target compounds were tested through four replicate analyses of a sediment sample with and without spiking of standard mixtures. The relative standard deviations of the concentrations were <15% and the recoveries were >83%. Procedural blanks for PAHs, hopane, LABs, and PCB were run with every batch, and analytical values 5 times the values of the blanks were considered significant.

3. Results and discussion

3.1. PAHs

Sites JK2, JK3 and JK5 had higher PAH concentrations; JK1 and JK4 had moderate concentrations; and JK6 and JK7 had lower concentrations (Fig. S1, Table 1). JK1, JK2, and JK5 are close to densely populated areas and business centers; JK3 is near the harbor; whereas JK6 and JK7 are farther from central Jakarta. This distribution is consistent with variations in the amounts of PAHs to urban runoff, sewage discharge, and industrial effluent, and locations of these inputs relative to the sampling sites. The 13 rivers that flow through Jakarta carry a huge amount of urban runoff, industrial effluent, waste oil, and domestic waste. The Ciliwung River, Cengkareng Drain, Sunter River and Cakung Drain are the main polluted rivers that discharge into Jakarta Bay near JK1, JK2, JK3, and JK5 (Fig. 1).

Total PAH concentrations (Σ_{14}PAH) at JK2, JK3 and JK5 were comparable to those in Tokyo Bay (Fig. S1, Table 1). Fig. 2 compares the sedimentary PAH concentrations in Jakarta Bay and Tokyo Bay with those in other coastal zones in developing, developed, and fast-growing countries (Table S2; [10,12,22,30–48,6,49–89]). Most of the values in developing countries range from tens to hundreds of ng/g, whereas those in developed countries range widely over tens of thousands of ng/g. On the other hand, fast-growing

Table 1
PAHs, hopane, LABs and PCBs in Jakarta Bay and Tokyo Bay sediments.

Stations	Σ_{26}PAHs (ng/g-dry)	Σ_{18}PAHs (ng/g-dry)	Σ_{14}PAHs (ng/g-dry)	MP/P	MPy/Py	MC/C	MPAH/PAH	Total hopane (ng/g-dry)	C_{30} hopane/total PAHs	C_{29}/C_{30}	$\Sigma C_{31-35}/C_{30}$	LABs (ng/g-dry)	%TAB	I/E ratio	PCBs (ng/g-dry)
JK1	682	553	426	2.41	0.38	1.75	1.28	2340	0.65	1.73	4.08	1755	NA[a]	1.77	NA[a]
JK2	1319	1182	1086	1.06	0.37	0.60	0.58	2271	0.26	1.57	3.64	235	80	1.44	2
JK3	1017	890	822	1.33	0.40	0.83	0.71	2608	0.42	1.52	3.16	631	NA[a]	2.24	NA[a]
JK4	516	442	403	1.49	0.48	1.00	0.84	1776	0.56	1.50	3.53	349	40	2.88	6
JK5	1511	1252	777	2.51	0.58	0.94	0.79	4148	0.58	1.71	4.90	86745	10	0.92	9
JK6	272	215	154	2.10	0.56	0.91	0.84	1054	0.73	1.57	4.69	2843	2	2.86	3
JK7	257	191	146	2.34	0.90	1.97	1.64	1137	0.98	1.56	3.45	472	NA[a]	2.69	NA[a]
TB1	1615	1454	1308	0.85	0.17	0.54	0.40	2730	0.28	1.38	3.74	1109	7	2.78	47
TB2	1372	1232	1130	0.77	0.19	0.59	0.41	2483	0.29	1.42	3.71	394	10	4.80	37

[a] Not analyzed.

countries (BRICS: Brazil, Russia, India, China, South Korea) show a higher range than developing countries, up to thousands of ng/g. PAH concentrations in Jakarta Bay were comparable to those of fast-growing countries.

PAH concentrations in sediments are controlled by several factors, including intensity of PAH inputs, distance from sources, climate, geography, and type and composition of sedimentary organic matter [15,90]. Sedimentary PAH concentrations in urban waterways, canals, harbors and estuaries which normally have intensive sources of PAHs are often much higher than those in coastal zones (e.g., [91]). The sites in our study were located in coastal zones, and the canals in Tokyo had much higher PAH concentrations (>10,000 ng/g; [16]) than those in the present study. PAH concentrations in urban waterways and canals in Jakarta should be investigated in future studies because of the potential for high concentrations in these environmental settings.

The PAH profile of Jakarta Bay sediment bears a petrogenic signature with relatively abundant alkyl-PAHs (Fig. S2). In contrast, the PAH profile of Tokyo Bay sediment shows great depletion in alkylated PAHs, suggesting a greater contribution of pyrogenic sources, as in other industrialized countries [3,7,41,92,93]. Normally, petrogenic sources consist of alkylated and lighter PAHs, with a depletion of heavier PAHs, while pyrogenic sources are abundant in heavier PAHs [14,60]. In Jakarta Bay, even though the PAH profile has abundant alkylated and lighter PAHs, a considerable amount of heavier PAHs is also present, indicating the input of both petrogenic and pyrogenic sources.

To quantitatively express the abundance of alkyl-PAHs, we calculated the ratios of alkyl-PAHs to parent PAHs (Table 1). A previous analysis of potential source materials determined the thresholds of MP/P, MPy/Py, MC/C, and MPAH/PAH for exclusively pyrogenic and petrogenic signatures [16]. When the ratio is lower than the pyrogenic threshold, it indicates an exclusive combustion origin; and when it is higher than the petrogenic threshold, it indicates an exclusive petrogenic origin. The ranges of threshold values (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.

MP/P ratios in all samples from Jakarta Bay (1.06–2.41) lie between 0.5 and 3.5, indicating both pyrogenic and petrogenic sources (Fig. 3, Table 1). These values are much higher than those recorded in Tokyo Bay, indicating more petrogenic input in Jakarta Bay. The other three ratios showed the same pattern (Fig. 3). These results indicate heavier petrogenic inputs of all homologs in Jakarta Bay than in Tokyo Bay.

The ratios showed spatial variation in Jakarta Bay. MP/P at JK1 and JK7 (2.41 and 2.34) were near the petrogenic threshold values, suggesting predominantly petrogenic inputs in these locations. On the other hand, all ratios were lower at JK2, JK3 and JK4 than at other locations in Jakarta Bay, suggesting more pyrogenic inputs. JK2 and JK3 are near to a gas- and coal-fired power plant that supplies most of Jakarta's electricity. JK4 is farther offshore and therefore might be influenced by atmospherically transported combustion-derived PAHs.

Overall, Jakarta Bay was affected by both petrogenic and pyrogenic sources, with a greater contribution from petrogenic sources. These petrogenic inputs might be related to the extensive use of fossil fuels to support the increasing growth of industry and motor vehicles in Jakarta. Identifying specific sources of PAHs in this area will be important for efficient environmental management in the future.

The ratio of C_{30} hopane to total parent PAHs is used to indicate petrogenic inputs related to automobiles or petroleum products; a ratio of >0.25 indicates the contribution of petrogenic inputs [16]. Ratios in Jakarta Bay ranged from 0.26 to 0.98 (Table 1), indicating
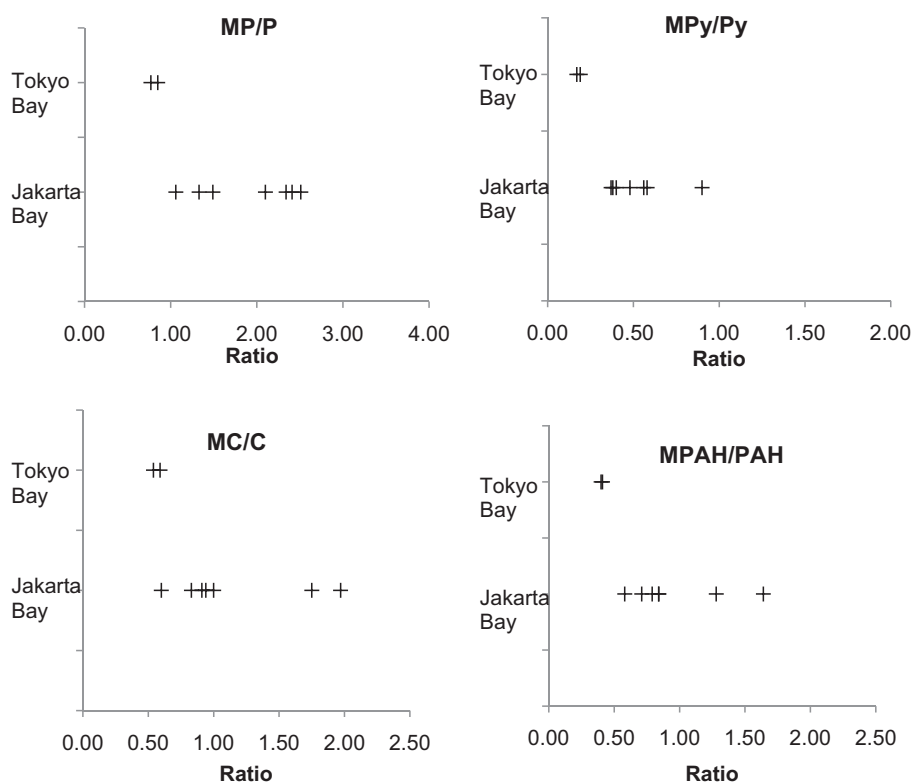
Fig. 3. Ratios of alkyl PAHs to parent PAHs in Jakarta Bay and Tokyo Bay sediments.

widespread inputs of petrogenic PAHs into these areas. On the other hand, ratios in Tokyo Bay (0.28 at TB1, 0.29 at TB2) were close to the threshold, suggesting mainly pyrogenic sources. Hopane concentrations at JK1, JK2, JK3, and JK5 were comparable to or higher than those in Tokyo Bay (Fig. S3, Table 1). The hopane profiles of Jakarta Bay sediments were relatively uniform across all locations and were similar to those of Tokyo Bay (Fig. S4). They were characterized by the predominance of $17\alpha,21\beta(H)$-C_{29} hopane ($C_{29}17\alpha$) over $17\alpha,21\beta(H)$-C_{30} hopane ($C_{30}17\alpha$) and by a step progression from C_{31} to C_{35} homohopanes. The C_{29}/C_{30} and C_{31}–C_{35}/C_{30} ratios in Jakarta Bay (1.50–1.73 and 3.16–4.69, respectively) were very similar to those in Middle Eastern crude oils [14]. The same profiles observed in Malaysian sediments and lubricating oils used in Malaysia indicated the contribution of used crankcase oil to Malaysian waters [14]. Similar hopane profiles observed in Jakarta Bay suggest the contribution of lubricating oil imported from the Middle East. More detailed data from potential source materials will help to narrow down the origin.

3.2. LABs

LAB concentrations in Jakarta Bay ranged very widely (Fig. 4, Table 1). The extremely high concentration at JK5 (86,745 ng/g-dry) indicates intensive input of sewage. JK5 is located near the mouth of a river that passes through a densely populated area where many residents discharge their domestic waste directly into the river. This river also flows through a densely populated area favored by many small businesses dealing in recycling and second-hand products and relying heavily on detergents in their work. These anthropogenic activities could contribute to the high LAB concentration. JK1 and JK6 also had high concentrations of LABs. On the other hand, JK7 had a low concentration, which may reflect a smaller population in the catchment. The distribution of LABs is controlled by lateral transport of sewage particles and dilution by natural particles in addition to the intensity of sewage inputs [22,94]. JK2 is located

off the mouth of the Ciliwung River, in whose catchment frequent intense rainfall supplies large amounts of eroded soil to the bay; thus, dilution at JK2 could be more efficient than at other locations. The lower concentration at JK4 could be explained by its longer distance from the coast and lesser influence of the lateral transport of land-based materials.

On average, LAB concentrations in Jakarta Bay sediments (geometric mean, 1400 ng/g-dry) were higher than those in Tokyo Bay sediments (661 ng/g-dry). The result is consistent with a previous report [94,95] of high concentrations of LABs in the water and sediment of a river passing through the Jakarta metropolitan area. The higher concentrations of sedimentary LABs in Jakarta Bay indicate larger inputs of sewage. In fact, central Jakarta has only one sewage treatment plant, which covers limited areas and only 1.9% of the population. Most of Jakarta is not served by sewage treatment, and domestic wastewater is dumped directly into the rivers or sea. The isomeric compositions of LABs discussed below confirm this result.
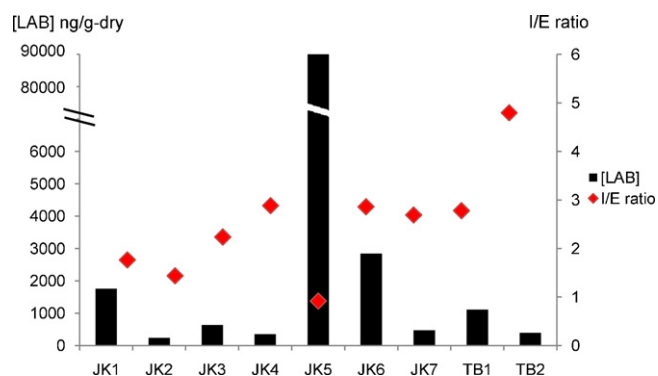

Fig. 4. LAB concentrations and I/E ratio in Jakarta Bay (JK1–JK7) and Tokyo Bay (TB1, TB2) sediments.
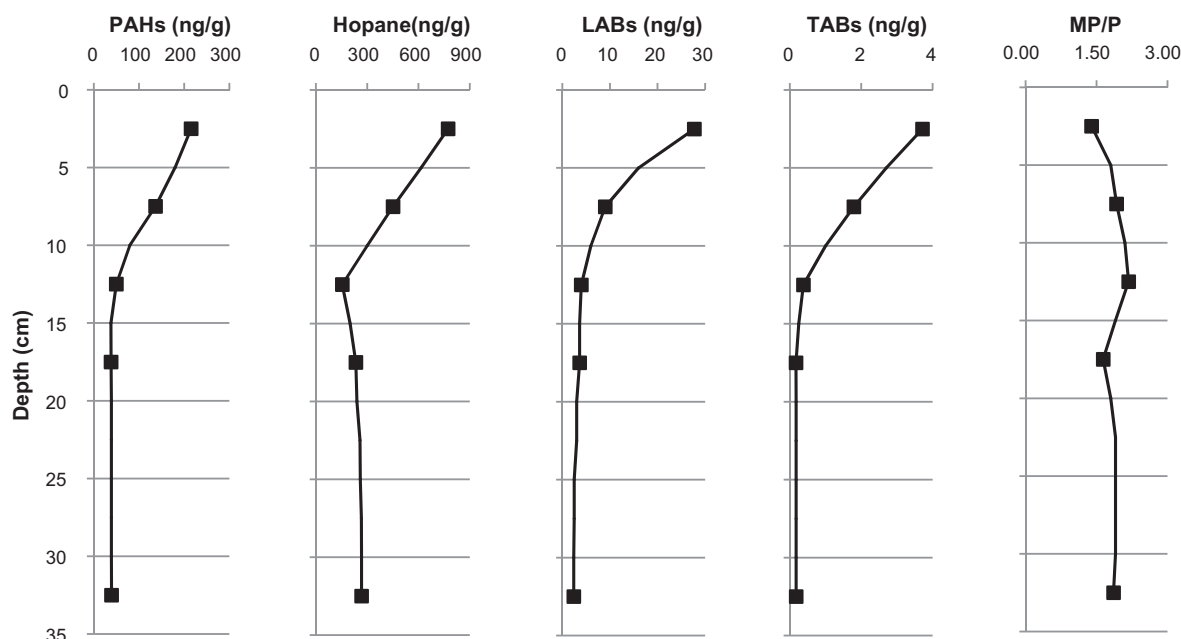
Fig. 5. Vertical profiles of PAHs, hopanes, LABs, TABs, and MP/P ratio.

Degradation by aerobic microbes systematically changes the isomer composition of LABs [96]. External isomers, with a phenyl substitution position near the terminal end of the alkyl chain, such as $2\text{-}C_{12}$ and $3\text{-}C_{12}$, are more biodegradable than internal isomers, with a substitution around the center of the alkyl chain, such as $5\text{-}C_{12}$ and $6\text{-}C_{12}$. The isomer composition can be quantitatively expressed as the internal-to-external isomer (I/E) ratio: $[5\text{-}C_{12} + 6\text{-}C_{12}]/[2\text{-}C_{12} + 3\text{-}C_{12} + 4\text{-}C_{12}]$. Because external isomers are more degradable, a higher I/E ratio indicates greater biodegradation of LABs under aerobic conditions. Conversely, a lower I/E ratio means either fresh inputs of LABs or deposition in anaerobic conditions, which depress microbial degradation. The I/E ratios of LABs in Jakarta Bay were the same or lower than those in Tokyo Bay, indicating less sewage treatment in Jakarta (Fig. 4, Table 1). JK5 had the lowest I/E ratio (0.92), which is in the range of untreated sewage [94], on account of intensive inputs of untreated sewage along the river. The large inputs of sewage, industrial effluent and oil into the river made the depositional environment more anaerobic, depressing the activity of aerobic microbes and keeping the I/E ratio low. On the other hand, JK3, JK4, JK6 and JK7 had higher I/E ratios, comparable to that at TB1 in Tokyo Bay. This does not imply the input of treated sewage, but most probably, microbial degradation during transport through the water column, facilitated by the warm waters [29]. The high concentrations of LABs with lower I/E ratios show that Jakarta Bay is heavily affected by untreated sewage. Sewage contains various hazardous components which can endanger the aquatic environment and humans [97]. The high concentrations of LABs coincide with high concentrations of *Escherichia coli* and coliform bacteria observed in that area [98]. Adequate sewage treatment is urgently needed in Jakarta.

Significant amounts of TABs were detected in Jakarta Bay sediments, and accounted for up to 80% of total LABs (Table 1). In contrast, TABs in Tokyo Bay were less abundant, and accounted for <10% of total LABs. TABs are feedstock used in the production of branched alkylbenzene sulfonate (ABS) surfactants. ABSs were used in detergents but were replaced by LASs in many countries in the 1960s owing to their poor biodegradability in the environment. The first industrial usage of TAB-based detergents in Indonesia began in the early 1970s, but the local government banned detergents containing ABSs in 1977. Yet owing to the lower price of TAB-based detergents and ineffective enforcement, some producers continued to use TABs for detergent production after the ban. The high proportion of TABs in Jakarta Bay suggests current usage of ABSs in synthetic detergents in Jakarta. Isobe [94] detected high concentrations of TABs at several locations in canal and river sediments collected in 2000 in Jakarta and confirmed that commercial detergents available in Jakarta in 2000 contained ABSs. In addition, vertical mixing and remobilization of older sediments may contribute to the TABs we identified in the surface sediment. TABs in Tokyo Bay sediment were attributed to such mixing [99]. However, the proportion of TABs to LABs in Jakarta Bay was much greater than in Tokyo Bay, indicating current or more recent usage of ABS detergents in Jakarta. Further study to apportion the contributions of sediment mixing and current usage is important to quantifying the legacy pollution of regulated chemicals.

3.3. PCBs

Although the use of PCBs was banned in the 1970s, PCBs were still detected in most of the sampling sites (Fig. S5, Table 1). Although concentrations were lower than those in the UK [100], Italy [6], and the USA [101], they were higher than those in Vietnam [102] and comparable to those in Puerto Rico [60] and southern China [103]. The total PCB concentrations in Jakarta Bay were 1 order of magnitude lower than those in Tokyo Bay (Fig. S5). This difference indicates minimal usage of PCBs in industrial or commercial products in Jakarta, and in contrast, very large usage before the 1970s in Japan. This result is concomitant with the spatial pattern of PCB concentrations in plastic pellets [104,105], which were higher in industrialized countries such as the USA, Western Europe, and Japan, but lower in developing countries such as in Southeast Asia and southern Africa. In addition, PCB concentrations in mussels from Jakarta Bay were lower than those in mussels in developed countries such as Japan [106,107]. Many industrialized countries started using PCBs in the 1930s and greatly increased their use during their economic growth phase in the 1960s and early 1970s, whereas many developing countries such as Indonesia did not make large-scale use of PCBs during their economic growth phase, because PCB usage was banned by then.

3.4. Vertical profiles of PAHs, hopanes, and LABs in core

LAB concentrations increased from 4 ng/g in the deeper part of the core (10–15 cm) to 28 ng/g toward the surface (Fig. 5). The deeper part corresponded to no or limited usage of LABs in the catchment in the past. The biggest detergent factory in Indonesia was established in the early 1970s, and was quickly followed by others. The more recent increase in LAB concentrations can be explained by large-scale production and usage of LAS-type detergents in Jakarta, consistent with dramatic population growth since 1960 (~3 million) to 2000 (~9 million). TAB concentrations started to increase slightly deeper (10–15 cm) as can been seen from Fig. 5, consistent with earlier use of ABSs than of LASs. The timing of rapid population growth and industrial development in Jakarta and the depth profile of organic micropollutants in the core (Fig. 5) suggest that the upper 15 cm of the core represents about the past 40 years (early 1970s to 2007).

PAHs showed consistent low concentrations deeper than 10–15 cm and increases of 1 order of magnitude toward the surface, suggesting a recent (i.e., 1980s to the present) increase in their inputs. Throughout the sediment core, MP/P > 1, indicating a greater input of petrogenic PAHs (Fig. 5). The increase in PAH concentration and the contribution of petrogenic sources are consistent with the increase in hopane concentration toward the surface. These increases may be related to increases in the input of traffic-related petroleum in the catchment. According to the Jakarta Regional Police, the number of motorcycles registered increased by 60% from 1.5 million in 1998 to 2.4 million in 2002, and the number of private cars from about 1 million to 1.4 million in the same period [25].

The concentrations of all compounds in the core increased toward the surface. This trend highlights the importance of time-series monitoring of these pollutants and of effective regulation of the inputs of these pollutants into Jakarta Bay. For the effective control of the pollutant inputs, more detailed source identification of the pollutants is necessary.

3.5. Conclusion

Our results clearly demonstrate that the pollution status of PAHs and LABs in Jakarta Bay is comparable to or worse than that in Tokyo Bay. The results show that Jakarta Bay receives a lot of petrogenic inputs and untreated sewage. Current or recent usage of ABSs (non-degradable detergents) was suggested. Adverse effects of these pollutants on the aquatic environment are of concern (e.g., [108]). For effective control of pollutant inputs, more detailed source identification of the pollutants is necessary. Levels of sedimentary PCBs in Jakarta Bay were much lower than those in Tokyo Bay, probably owing to minimal usage of PCBs during the economic growth period in Indonesia. Increases in the concentrations of PAHs, hopanes, and LABs imply the increased inputs of these pollutants into Jakarta Bay for several decades. This study presents a preliminary benchmark of the pollution status of Jakarta Bay. More extensive and comprehensive monitoring of organic micropollutants will establish a more concrete benchmark, which will be essential to evaluating the effectiveness of regulations.

Acknowledgements

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jhazmat.2012.03.023.

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