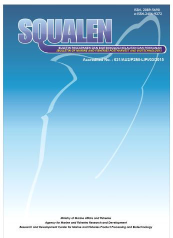

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NON-TARGET SCREENING METHOD FOR THE IDENTIFICATION OF PERSISTENT AND EMERGING ORGANIC CONTAMINANTS IN SEAFOOD AND SEDIMENT FROM JAKARTA BAY

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Abstract

Identification of persistent and emerging organic contaminants in green mussels (*Perna viridis*), various fishes, banana shrimp (*Penaeus merguiensis*) and sediment samples from Jakarta Bay has been employed. A non-target GC-MS screening approach has identified more than 60 individual organic compounds from the whole fractions either non-polar, semi-polar or acidic-polar compounds. The substances comprised as persistent organic pollutants (POPs) including DDT (dichlorodiphenyl trichloroethane) and its metabolites as well as high molecular weight polycyclic aromatic hydrocarbons (HMW-PAHs). Noteworthy, a number of emerging contaminants detected in the present study have never been reported previously either from the same location or from Asian waters. They include some priority contaminants of non-persistence halogens and emission of technical products, such as di-*iso*-propylnaphthalenes (DIPNs) dichlorobenzene (DCB), dichlorodiphenyl chloroethene (DDMU) and phenylmethoxynaphthalene (PMN). In general, the concentration order of the priority organic contaminants was sediment > green mussel > fishes > shrimp. Further analysis based on the spatial distribution, individual concentrations and bioavailability suggested that some contaminants are applicable as molecular marker for the assessment of anthropogenic emission in Jakarta Bay, i.e. DIPNs, linear alkylbenzenes (LABs), phenylmethoxynaphthalene (PMN), PAHs, dichlorobenzene, DDT and its metabolites.

Keywords: non-target screening, persistent organic pollutants, emerging contaminants, seafood, Jakarta Bay

1. Introduction

Jakarta Bay is an essential coastal ecosystem as it is situated in the Capital City Jakarta with more than 10 Million inhabitants (BPS, 2014). The bay provides several functions, such as for maritime transportation, supply natural resources (mainly seafood), as recreational facilities, and support waste regulating services. Importantly, the bay is also exposed to high pollution load derived either from up-land regions through discharging rivers into the bay or from direct coastal activities. The terrestrial contamination is estimated to contribute up to 80 % of the pollution in Jakarta Bay (Anon., 2007). Those contaminants are delivered from various sources, such as domestic disposal, industrial waste, transportation, oil spills, etc. (Uneputty & Evans, 1997). Hence, the burden of the contaminants potentially increases significantly in accordance with the population growth and the development of industrial activities.

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Jakarta city is currently connected to its suburb regions i.e. Bogor, Depok, Tangerang and Bekasi by which the region known as Jabodetabek or Jakarta Metropolitan Area (JMA). There is huge amount of domestic waste polluting Jakarta Bay resulted from 28.3 Million population of JMA. Additionally, more than 20 thousand industries are situated in Jabodetabek and potentially contribute to pollute Jakarta Bay, either from by-products of combustion and solid or liquid disposal. Further on, many hospitals in Jabodetabek are supposed contribute on organic waste disposal, as well as pesticide/herbicide residues from agricultural effluent in suburb regions (Dsikowitzky et al., in draft; Dwiyitno et al., in draft). At least 161 ton/day of solid waste and 6.5 Million ton/day of liquid waste are discharged into Jakarta Bay (Takarina & Adiwibowo, 2011; BPLHD, 2014). On a longer period, accumulation of contaminants could damage the aquatic ecosystem and also produce harmful effects on the environment and human health.

Since Jakarta Bay represents an essential ecosystem, in particular regarding fishing and marine/aquaculture activities for the Jakarta city and surrounding regions, the impact of organic contamination into this bay has to be thoroughly in concern. Studies on the organic pollutants have been reported from Jakarta Bay which was focused on the priority pollutants such as PAHs, DDT, polychlorinated biphenyls (PCBs), tributhyltin (TBT), polybrominated diphenyl ethers (PBDEs) and hexachlorohexanes/HCHs (William et al., 2000; Monirith et al., 2003; Sudaryanto et al., 2007). However, all of those studies were conducted based on either single or selected target approach. To date, one of the biggest issues associated with analytical environmental contaminant is the limitation of target analysis which only focuses on selected contaminants. Selected or targeted compound monitoring approach, with mass spectrometry (MS) for example, using selected ion monitoring (SIM) mode would only assess a limited number of analytes, whereas other potentially harmful or non-target analytes might not be detected (Ibanez et al., 2008). For that reason, non-target approach has been implemented to screen a wide range of known or unknown contaminants and identify the transformation products that were not performed in traditional selected analytical method (Hernandez et al., 2012). Non target screening has been showing an alternative approach for the identification of organic contaminants in water and sediment samples in developed countries (Hernandez et al., 2007; Grigoriadou et al., 2008; Schwarzbauer et al., 2010; Schymanski et al., 2014). Different instruments have demonstrated powerful tools for non-target screening analysis, such as LC-TOF-MS (Diaz et al., 2012), LC-MS (Hird et al., 2014), and GC-MS (Shaikh et al., 2014).

Investigation of organic contaminants in aquatic biota by using non-target screening has not been reported yet. Through bio-magnification pathway via aquatic food web, the contaminants potentially accumulate in the aquatic organism including seafood at which the concentration will increase as the longer of exposure. Notably, studies on the emerging pollutants such as emission of pharmaceutical and personal care products (PPCPs), hormones and other technical/industrial products like DIPNs, phenylmethoxy- naphthalene in Indonesia and Asian regions are very limited or even none (Suzuki et al., 2007; Terasaki et al., 2008; Dsikowitzky et al., 2014). This research was aimed to study the application of non-target screening using GC/MS approach to investigate the presence and distribution of persistent and emerging organic contaminants related to seafood and sediment compartments in Jakarta Bay. The result of this study is important to evaluate contamination profile, determine the priority contaminants, as well as the safety status of seafood from Jakarta Bay.

2. Material and Methods

2.1. Study Site and Sample Collection

The study was conducted in the coastal system of Jakarta Bay. The bay stretches from 106°20' to 107°03' east longitude, and from 5°10' to 6°10' south latitude, and is administratively within three provinces, i.e. Banten, the DKI of Jakarta, and West Java Provinces. The bay receives water from 13 main rivers, some of which flowing through the Jakarta city and 6 main rivers with in the suburb regions. The regions are strongly influenced by typical tropical climate. The northwest monsoon (November-March) brings rainfall into Jakarta metropolitan area, while the Southeast Monsoon (May-September) coincides with the dry season. This nature results in significant variation of precipitation rate between dry and rainy season i.e. 2-118 mm/month (dry season) and 158-275 mm/month during rainy season of 2013, which was higher than that of 2012 (0-79 mm/month in dry season and 47-259 mm/month during rainy season). Unlikely, average temperature in the region was relatively similar during 2012 and 2013 i.e. 27.7-30.8 °C and 27.3-29.1 °C respectively (BPLHD Jakarta, 2012; 2013).

A number of 10 different biological samples have been directly collected from local fisherman in Jakarta Bay during October 2012, May and October 2013 in order to represent the dry and rainy season contamination. They include 6 species of demersal and 2 pelagic fishes, 1 shrimp species (banana shrimp/

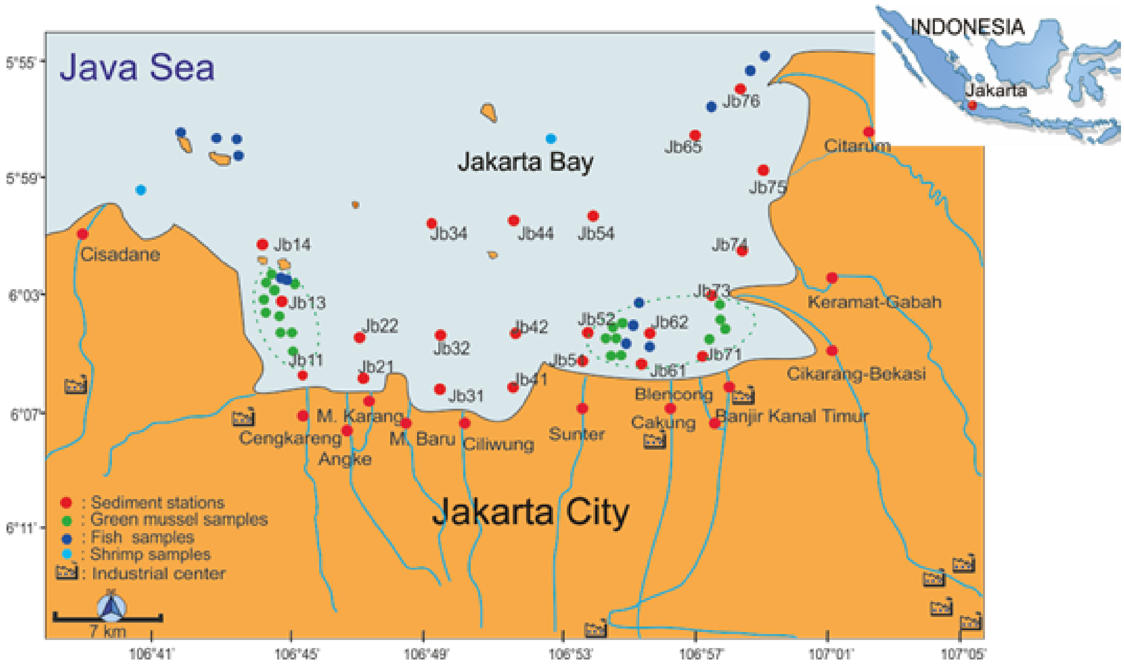

Figure 1.Schematic sketch of the study area and sampling location.

P. maerguiensis) as well as green mussel (*P. viridis*) from 11 different stations. In order to determine contaminant level in the environment compartment, surface sediment was collected from 22 stations in Jakarta Bay with a stainless steel *Van Veen Grab* (20 x 20 cm^2). During the sampling campaign, all samples were conserved with ice and after reach the laboratory, all samples were stored at -18 °C until chemical analysis. Sampling points of biological samples and surface sediment are performed in Figure 1, while characteristic of biological samples is presented in Table 1.

2.2. Extraction and Fractionation

A non-target screening approach was applied based on the method described by Schwarzbauer *et al*. (2000). Sediment extraction was performed with a high-speed dispersion tool (*Ultra-Thurax Turrax*T$_{25}$ basic, IKA Labortechnich) using mixture of acetone and *n*-hexane, while fractionation was conducted based on the method explained by Franke *et al.* (2007) into 7 fractions (Figure 2). After separation of the phases, the organic extracts were added 50 µl of surrogate standard containing *d34*-hexadecane (6.00 ng/µl), *4*-fluoroacetophenone (7.17 ng/µl) and decafluorobenzophenone (6.96 ng/µl). After concentrated, aliquot was dehydrated with sodium sulphate anhydrate (Na_2SO_4) and eluted with the similar solvent. On the other hand, the polar fraction was further derivatized with diazomethane prior to fractionation as performed in fraction 1-5. Prior to GC and GC/MS-analysis, all extracts were concentrated to approximately 50 µl (fraction 1-5) and ± 200 µl (fraction 6-7). All chemicals were purchased from Sigma-Aldrich, Germany.

Dialysis method using EC-*Exsposmeter* clean-up semipermeable membrane (*ExposMeter* sampling technologies, Sweden) was employed for extracting biological samples as described by Dwiyitno et al., in draft. After extracted with dichlorometane (DCM) for 72 hours, supernatant was concentrated and fractioned as performed in sediment sample. One microliter of sample aliquot was injected in to a Trace GC-MS system (*ThermoQuest,* Germany) linked to *HRGC*-5160 Mega Series (Carlo Erba, Italy) equipped with a *ZB-XLB* silica capillary column (30mx0.25mm ID, 0.25 µm film thickness; *Zebron-Phenomenex,* Germany). The oven temperature was held at 60 °C for 3 min then increased at 3 °C/min to 310 °C and held for 20 min with injection temperature of 270 °C. Helium was used as carrier gas at 40 cm/s velocity. Mass spectrometer was operated in full-scan mode at a resolution of 1000 with a source temperature of 200 °C. Electron impact ionization mode (EI$^+$, 70 eV) was operated at a scanning mode from 35 to 700 amu at a rate of 1 s/scan. The schematic flow of the analysis method is summarized in Figure 2.

2.3. Identification of organic contaminants

Individual compound was identified by comparison of the EI-Mass spectra and specific ion fragment (Table 2) with those of mass spectral libraries (*NIST98*

Table 1. Characteristic of investigated fish, shrimp and mussel species from Jakarta Bay

No	Common name/ *Scientific name/* Local name	Habitat/ Feeding habit	Average of weight±S.D. (g)	Average of length±S.D. (cm) *)	Sampling period
1	Green mussel/ *Perna viridis/* Kerang hijau	Pillar/ filter feeder	16.0 ± 6.5 12.4±2.1 16.4±6.4	6.8 ± 0.8 6.3±1.2 7.0±1.0	October 2012, May 2013, October 2013
2	Mackerel/ *Rastrelliger kanagurta/* Kembung	Pelagic/ omnivore	149.4±13.1 87.2±12.8 88.5±20.0	20.7±0.6 16.3±0.75 16.3±1.1	
3	Slender shad/ *Ilisha elongata/* Mata belo	Pelagic/ carnivore	103.2±15.2	113.5±7.1	October 2012
4	*White emperor/* *Lethrinus lentjan/* *Lencam*	Demersal/ carnivore	108.3±30.4	15.7±1.9	
5	Streaked spinefoot/ *Siganus javus/* Baronang	Demersal/ herbivore	183.1±32.3	16.8±1.0	
6	Johnii snapper/ *Lutjanus johnii/* Ikan tanda	Demersal/ carnivore	103.2±15.2 118.5±33.4	14.4±0.9 15.6±0.9	October 2012, May 2013
7	Croaker/ *Argyrosomus amoyensis/* Gulamah	Demersal/ omnivore	204.6±37.3 183.9±58.9	21.0±1.2 21.1±2.1	
8	Banana shrimp/ *Penaeus maerguiensis/* Udang putih	Demersal/ omnivore	26.2±6.2 25.1±7.9	13.8±2.6 12.8±2.8	
9	White-spotted rabbitfish/ *Siganus canaliculatus/* Baronang susu	Demersal/ herbivore	62.1±7.2 91.7±15.8	12.8±0.7 14.4±1.3	May 2013, October 2013
10	Sea catfish/ *Netuma thalassina/* Manyung	Demersal/ carnivore	467.9±67.8 588.6±246.4	29.9±1.7 33.4±5.5	

*)Fish length is measured as standard length

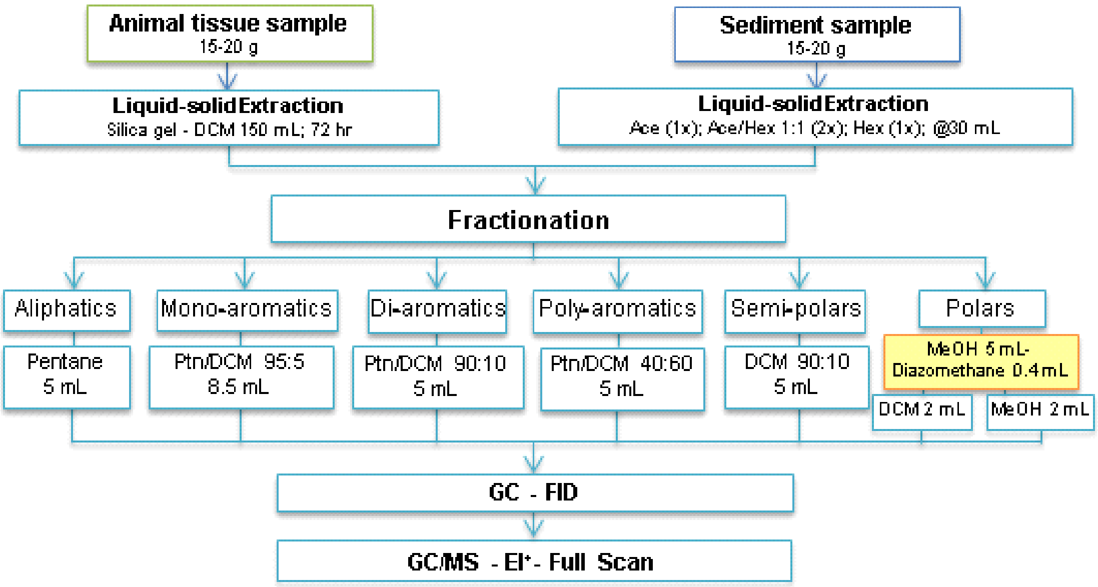

Figure 2. Schematic flow of non-target screening approach (Schwarzbauer et al., 2010; Dwiyitno et al., in draft) (DCM: dichloromethane; Ace: Acetone; Hex: Hexane; Ptn: Pentane; MeOH: Methanol).

and *Wiley* 4th Ed.), as well as by integrating the gas chromatographic retention time or scan number of authentic reference materials. Quantitation was performed by integrating ion chromatograms on selected m/z and calculated by *Xcalibur* based on the comparison of sample spectral and response factors (RF) of reference materials. Contaminant concentration was calculated as follow:

$$\text{Concentration}\left[\frac{\text{ng}}{\text{g}}\right]=\frac{\text{Area of sample}}{\text{Area of surrogate standard}}\times\text{Response factor}\times\frac{\text{Surrogate standard (ng)}}{\text{Sample weight (g)}}$$

2.4. Determination of Dry Weight and Total Organic Matter

Organic contaminant concentrations in the animal tissue samples were given based on fat content, while the contaminant concentrations in sediment samples were presented based on total organic matter content. Dry weights were determined by drying of ± 2 g sample aliquots at 105 °C for 18 hours to constant weight (Heiri et al., 2001). Total organic matter contents were further calculated by determining the loss of ignition (LOI) at 550 °C using *Nabertherm* furnace (LT 15/13) for 3 hours.

2.5. Determination of fat content

Total fat content was quantified based on gravimetric method using *soxhlet* system (AOAC, 2002). Approximately 2.0 g of fresh seafood tissue was weighed and placed in an extraction thimble and placed in the soxhlet apparatus. Fat was extracted through repeated washing/percolation with 150 mL petroleum ether for ± 6 hours. After extraction, the fat extract collected in the extraction pot was oven dried and weighed in order to compare to the weight of the initial sample.

2. Result and Discussion

Non target GC/MS screening analysis applied on green mussels, fish species, shrimp, and sediment samples of Jakarta Bay revealed a wide range accumulation of lipophilic organic substances. Table 2 represents the summary of more than 60 individual organic contaminants identified from the whole fractions, whereas concentrations of selected priority compounds are given in Table 3. In general, sediment samples accumulate more various with the higher concentration of organic contaminants in comparison to biological samples (green mussel, fishes and shrimp).

The solid-liquid extraction method on biological samples and sediment produced seven fractions representing the non-polar, semi-polar and acidic relatively polar compounds respectively. However, the majority of contaminants were detected in fractions 2-5, while minor contaminants were also identified in fraction 1, 6 and 7. This indicates that the majority of organic contaminants were semi polar substances, such as halogenated compounds (fractions 2-4), aromatic compounds and technical products (fractions 2-5), as presented in Figure 2. Fraction 1 consisted of relatively non-polar compounds, while methylated of relatively polar compounds were eluted in fractions 6 and 7. In contrast, low volatile polar compounds, such as organotins, are typically more sensitively and selectively detected using liquid chromatography instead of gas chromatography (Zou & Schreiber, 2012).

Spatial distribution showed that contaminant concentration in green mussel from eastern part of Jakarta Bay was relatively higher than that from the western part. This difference seems correspond to the contaminant concentration in the environment compartment as presented in sediment sample (Figure 4-7). Additionally, contaminant concentration in river sediment was higher than that of coastal sediment, indicating the contaminant origin were predominantly from terrestrial emission. This fact was also supported the fact that contaminant concentration in coastal sediment apparently reflected the river sediment concentration.

Due to hydrophobic property, lipophilic compounds tend to be separated from the aqueous phase for the further adsorption into colloidal material or organic matter or sediment by van der Waals force (Walker, 1987). Hydrophobicity of organic compound is commonly indicated by coefficient partition of octanol-water (K_{ow}). Persistency of organic compounds in aquatic ecosystem is also affected by their chemical stability to the chemical degradation such as photochemical and microbial degradation (Ricking & Schwarzbauer, 2012). Environmental behavior like pH, temperature, oxygen availability may also contribute in the nature of contaminant accumulation (Kim et al., 2003).

The relatively less variety and lower concentration of organic contaminants in fish and shrimp species compared to that of green mussel in the present study suggested the different uptake and metabolism nature, as explained earlier by Dwiyitno et al. (in draft). Earlier studies showed that occurrence of elimination rate of xenobiotic in mussel species tends to be lower than that of fish species (D'Adamo et al., 1997; Zhao et al., 2014). With regard to shrimp species, relatively low detoxification rate was performed on the bioaccumulation study of PCBs on *Palaemonetes*

Table 2. Organic compounds identified in sediment and biological samples of Jakarta Bay

No	Organic Compound	Ion Fragment	Scan Nr. (c.a.)	Green mussels	Fishes	Shrimp	Sediment
A	**HALOGENATED COMPOUNDS**			3	5	1	7
1	Bis(4-chlorophenyl)-1,1,1-trichloroethane (DDT)	235, 237, 354	4860-5175	-	√	-	√
2	Bis(4-chlorophenyl)-1,1-dichloroethane (DDD)	235, 237, 320	3860-4744	-	√	-	-
3	Bis(4-chlorophenyl)-1,1-dichloroethene (DDE)	246, 248, 318	4450-4690	√	√	√	√
4	Bis(4-chlorophenyl)-1-chloroethylene (DDMU)	212, 282, 284	4510	√	√	-	√
5	Dichlorobenzenes	146, 148	370	√	√	-	√
6	Trichloromethylbenzyl-acetate	149, 107	2570	-	-	-	√
7	Methyltriclosan	304, 254	4420	-	-	-	√
8	Chlorpyrifos	316, 199	4100	-	-	-	√
B	**POLYCYCLIC AROMATIC HYDROCARBONS (PAHs)**			27	19	5	33
9	Naphthalene	128	1000	√	√	√	√
10	C1 to C4-Naphthalenes	142, 156, 170, 184	1510-3300	√	√	√	√
11	Biphenyl	154, 76	1880-1920	√	√		√
12	C1 to C4- Biphenyls	168, 182, 196, 210	2050-3200	√	√	√	√
13	Acenaphtylene	152, 176	2180	√	√	-	√
14	Acenaphthene	154, 153	2270	√	√	-	√
15	Fluorene	166, 165	1690-2770	√	√	-	√
16	C1 and C2-Fluorenes	180, 194	3080-3530	√	√	-	√
17	Phenanthrene	178	3350	√	√	√	√
18	Anthracene	178	3450	√	√	√	√
19	C1 and C2-Anthracene/-Phenanthrene	192, 206	3680-4100	√	√	-	√
20	Fluoranthene	202	4200	√	√	-	√
21	Pyrene	202	4460	√	√	-	√
22	C1- Fluoranthene/Pyrene	216, 215	4740-4890	√	-	-	√
23	Chrysene/Benz(a)anthracene/Triphenylene	228	5320-5420	√	-	-	√
24	C1-Chrysene/Benz(a)anthracen	242	4590	√	-	-	√
25	Benzaceanthrylene/-acephenanthrylene/-fluoranthene	252	5570	√	-	-	√
26	Benz(a)pyrene/Benz(e)pyrene/Benz(k)fluoranthene	252	6490	√	-	-	√
27	Perylene	252	6580	√	-	-	√
28	Dibenzo(a)Anthracene	278	7100	-	-	-	√
29	Indeno(1,2,3-cd)pyrene	276, 138	7240	-	-	-	√
30	Benzo(ghi)perylene	276, 138	7250	-	-	-	√

Table 2. Organic compounds (continued)

No	**Organic Compound**	**Ion Fragment**	**Scan Nr. (c.a.)**	**Green mussels**	**Fishes**	**Shrimp**	**Sediment**
31	Tetraline	132, 104	920	√	-	-	√
32	C1 to C3-Tetralines or C1 to C4-Indanes	146, 160, 174 188, 202	1500-2400	√	√		√
33	Decaline	138, 109	520	-	-	-	√
34	C5-Decalines or C6-Octahydroindane	166-208	1970-2045		√	-	√
35	Tetrahydroreten (THR)	238, 223	4290	√	-	-	√
36	Dibenzothiophene	184, 139	3280	√	√		√
37	C1 and C2-Dibenzothiophene	198, 212	3530-3660	√	√		√
38	C1-Dihydronaphthalene	172, 157	1760-1830	-	-	-	√
39	Dibenzofuran	169, 139	2490	√	√	-	√
40	C1 and C2-Dibenzofurans	182, 181	2880	√	√	-	√
41	Hydroxy-3-methyl-1,4-naphtoquinone	188, 131	2800-2940	-	-	-	√
C	**ALKYLBENZENES**			2	2	2	2
42	C2 to C6-Benzenes	106, 120, 134, 148, 162	10-940	√	√	√	√
43	Linear alkylbenzenes (C_{10} to C_{13} -LABs)	218, 232, 246, 260	2320-3500	√	√	√	√
D	**STEROIDS**			-	-	-	2
44	Coprostanol	388	6540	-	-	-	√
45	Cholesterol	386	6840	-	-	-	√
E	**ISOPRENOIDS**			2	2	-	2
46	Limonene	136, 93	410	√	√	-	√
47	Cadalene	198, 183	3000	√	√	-	√
F	**TECHNICAL PRODUCTS**			9	3	3	15
48	Di-*iso* -propylnaphthalenes (DIPNs)	212, 197	2880-3200	√	√	√	√
49	Phenylmethoxynaphthalen	234, 91	4500	√	√	√	√
50	N,N,-Diethyl-*m* -toluamide (DEET)	191, 119	2590	-	-	-	√
51	Tributylphosphate	99	2860	√	-	-	√
52	Phenyl-terphenyl	306	6560-6820	√	-	-	√
53	Terphenyl	230	3750-6820	√	-	-	√
54	Phenyl naphthalene	203, 179	3690	√	-	-	-
55	Aminoacetophenone	135,12	1750	-	-	-	√
56	Trichloroethylphenyl	149, 107	2570	-	-	-	√
57	Formado acetophenone	163, 135	2750	-	-	-	√
59	Trimethyl-1,3-pentandi-	71	2620	√	–	–	√
60	Diphenylether	170, 141	1980	–	–	–	√
61	Isopropylbiphenyl	200	3470	√	–	–	–
62	Diisopropylbiphenyl	238, 223	3870	–	–	–	√
63	Phenyl trimethylindane	236, 221	3110-3200	√	√	√	√
64	Indole	90, 117	1690	–	–	–	√
	Total			**44**	**31**	**11**	**59**

√ : detectable; - : non detectable (LOQ: 0.5 ng)

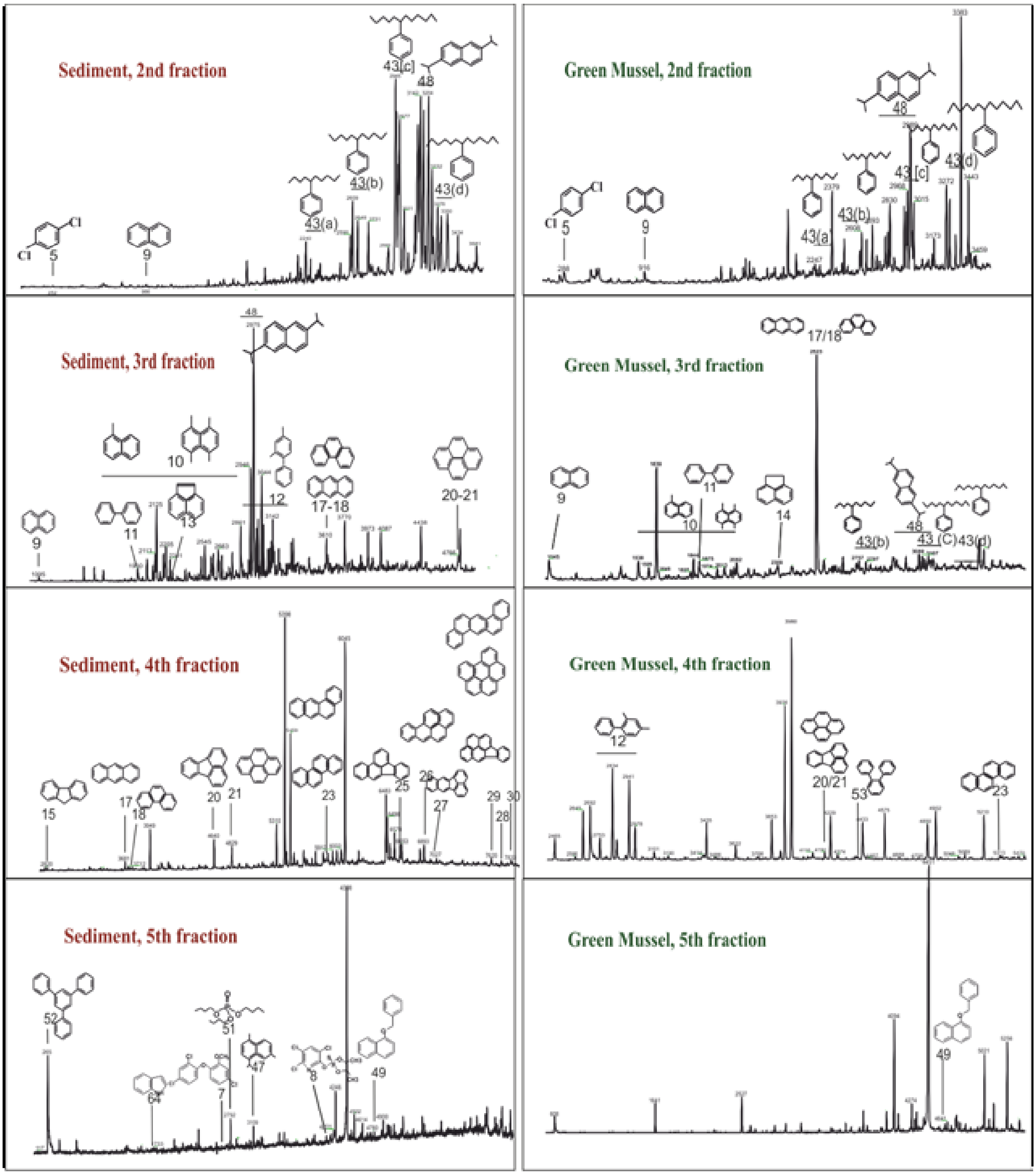

Figure 3. Comparison of total ion chromatogram (TIC) of different fractions between sediment and green mussel samples (numbers of identified compounds correspond to the sample numbers in Table 2).

varians shrimp conducted by Grilo et al. (2014). Noteworthy, concentration of contaminants in the environment is crucial factor affecting their bioaccumulation into aquatic animal. In Jakarta Bay, green mussel is cultivated in the relatively contaminated environment near the coast of western (Dadap and Kamal) and eastern part (Muara Tawar and Cilincing) of the bay (Figure 1).

Basically, aquatic animal accumulates xenobiotics through different routes i.e. direct uptake from water phase via epidermis organs or gill (bioconcentration) and via gastrointestinal tract from ingestion of either suspended particulate matter or from consumption of contaminated feed or known as bioaccumulation and biomagnification (Pierard *et al.*, 1996). Further on, bioavailability of the contaminant in the environment

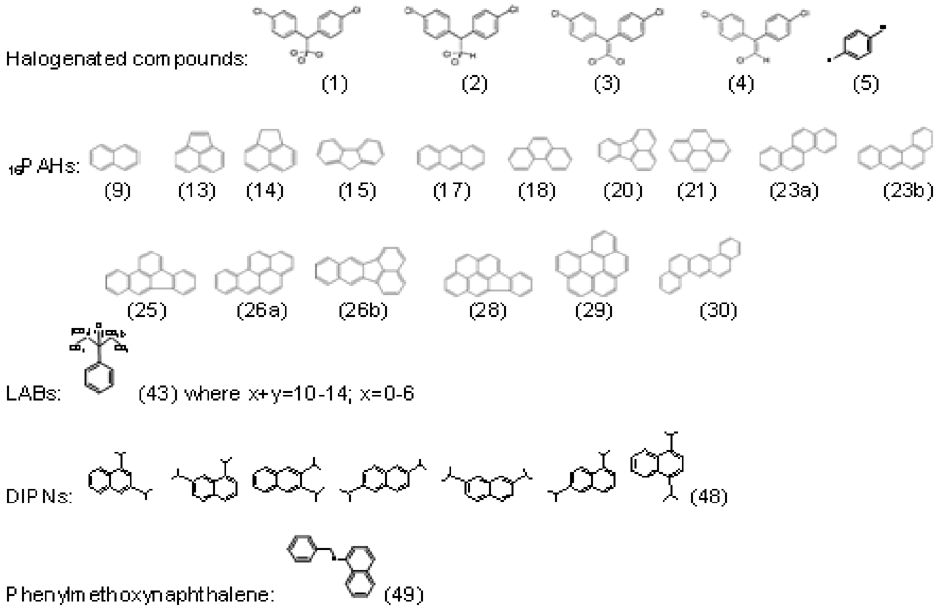

Figure 3. Chemical structures of important contaminants identified in sediment, green mussel and fish samples (the numbers correspond to the sample numbers in Table 2)

Table 3. Geometric mean and (min-max) concentration of priority contaminants in Jakarta Bay

No	Organic Compound	Mussels (ng/g Fat)		Fish (ng/g Fat)	Shrimp (ng/g Fat)	Jakarta Bay Sediment (ng/g TOC)	River Sediment (ng/g TOC)
		West	East				
1	Dichlorodiphenyl-trichloretane (DDX)	90 (n.d.-220)	80*)	90 (n.d. -470)	75*)	30 (n.d.-80)	1,04 (n.d.-1,560)
2	Dichlorobenzene (DCB)	280 (110-950)	280 (n.d.-1,100)	40 (n.d.-530)	n.d	n.d.	150 (n.d.-750)
3	Polycyclic aromatic hydrocarbons (Σ_{16}PAHs)	750 (330-2,750)	1,22 (850-2,990)	60 (n.d.-280)	350 (n.d.-350)	940 (230-5,070)	4,17 (340-15,800)
4	Linear alkylbenzenes (C_{10}-C_{13} LABs)	7,59 (2,400-50,600)	50,21 (12,900-136,700)	5,79 (n.d.-28,400)	1,1 (1,000-1,200)	32,96 (210-170,000)	65,1 (1,400-6,900,000)
5	Di-*iso*-propyl naphthalenes (DIPNs)	990 (240-3,700)	2,93 (990-21,870)	350 (20 – 14,380)	700 (370-1,320)	910 (80-32,00)	5,13 (240-100,000)
6	Phenylmethoxy-naphthalene (PMN)	200 (n.d. – 500)	1,95 (n.d. - 3,560)	220 (n.d. – 400)	270 (n.d. – 270)	240 (n.d.-1,600)	740 (n.d. -3,300)

n.d. : non detectable (LOQ: 0.5 ng); DDX is sum of DDT, DDE, DDD and DDMU; *) Only single sample was contaminated.

and elimination and biotransformation or detoxification rate in the organism would also contribute in the bioaccumulation of contaminant in aquatic animals (Franke et al., 1994). As filter feeder sedentary species, not only from diffusion of dissolved water, but also green mussel tends to accumulate contaminant from particulate matter, whereas fish accumulate contaminant predominantly from food web (van der Oost et al., 2003).

3.1. Persistent Organic Pollutants (POPs)

Persistent organic pollutants (POPs) is defined as organic compounds that resist mainly to photolytic, biological and chemical degradation IPCS (1995). POPs are often halogenated and characterized by low water solubility and in contrast high lipid solubility, leading to their bioaccumulation in fatty tissues. They are group of semi-volatile compounds, enabling them to move long distances in the atmosphere before deposition occurs.

DDT metabolites and high molecular weight (HMW) PAHs were among persistent contaminants detected in biological samples and sediment samples from Jakarta Bay. HMW-PAHs consisting of at least four benzene rings are known as persistent pollutants and classified as carcinogenic and mutagenic substances to aquatic biota and human health (ATSDR, 1999). They include benzo[*a*]pyrene (BaP), benzo[*a*] antracene (BaA), chrysene (Chr), benzo[*b*] fluoranthene (BbF), benzo[*j*]fluoranthene (BjF), benzo[*k*] fluoranthene (BkF), dibenzo[*a,h*] anthracene (DBahA), indeno[1,2,3-*cd*] pyrene (IP), and benzo[*ghi*] perylene (BghiP). Figure 3 represents molecular structures of important organic contaminants detected in the present study as they are spatially distributed in different compartments at relatively significant concentration.

Four DDT metabolites were detected in sediment and biological samples of Jakarta Bay, i.e. DDT, DDD, DDE and DDMU. All of these DDT metabolites, furthermore known as DDX, appeared at relatively low concentration in fish sample and river sediment, whereas DDT and DDD were neither detected in green mussel nor in sediment samples. DDE and DDD are the main metabolite products of DDT in the environment, by which DDE is known either more persistent or more toxic than the parent compound DDT (Walker, 1987). Furthermore, microorganism activity, gastrointestinal metabolism, oxygen level or environmental catalyzer (such as KOH, NaOH and other strong alkalis) could promote the dechlorination or dehydrochlorination transformation of DDT compounds. Ratio of DDT metabolites over parent compound is often applies to identify their emission behavior (Schwarzbauer et al., 2003). The presence of *p,p*'-DDE in sediment and in animal samples in the present study supported that this metabolite is the main degradation product of DDT which is also in line with the earlier investigations (Monirith et al., 2000; Boonyatumanond et al., 2002; Sudaryanto et al., 2005; Bayen et al., 2003).

Comparable DDT metabolites concentrations (DDT, DDE and DDD) in sediment samples from Jakarta Bay were reported earlier by Munawir (2013). Concentrations of DDT metabolites in *P. viridis* in the present study were also comparable to those earlier reported by Monirith et al. (2003) from the same location or from other South East Asian coastal. DDT concentrations in fish in the present study were comparable to those earlier investigation at the same location (Sudaryanto et al., 2007). As presented in Table 3, concentration of DDX in mussels and selected fish in this study was far below the FAO/WHO (2000) maximum residue limit (MRL) of 1,000 ng/g fat. Similarly, the concentration in sediment was below the threshold of 6,000 ng/g dw weight (US DI, 1998).

With regard to PAHs contamination, the concentrations in some sediment and mussel samples from Jakarta Bay have exceeded the threshold. A global threshold limit of PAHs in food product has been regulated such as 2 ng/g ww (BaP) or equivalent to 12 ng/g ww of total BaP, BaA, BbF

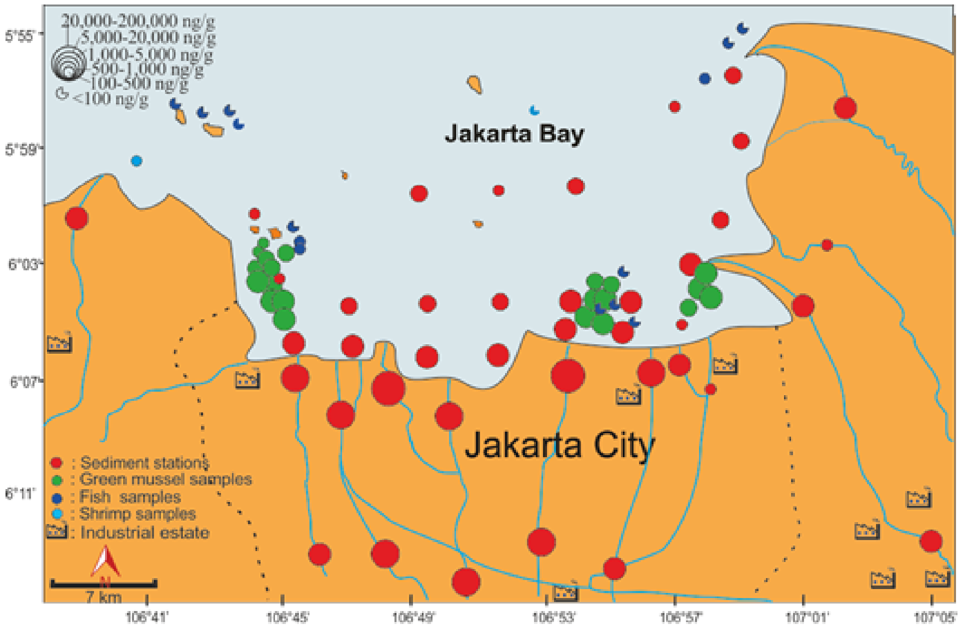

Figure 4. Spatial distribution of PAHs contaminant in Jakarta Bay and JMA rivers sediment (ng/g TOC) and in biota samples (ng/g fat).

and Chr or 200 ng/g of total 8 PAHs mentioned previously (EC, 2006). However, those long chains PAHs were not detected in fish and shrimp samples from the study area. The lower bioavailability of high molecular weight (HMW) PAHs could be related to their stronger attachment to sediment particles compared to the low molecular weight PAHs (Baumard *et al*., 1999). In general, accumulation of PAHs in green mussel was similar to that of sediment samples containing low molecular weight (LMW) and HMW-PAHs, indicating ingestion of particulate matter from contaminated waters was predominantly accumulation route into green mussel. In contrast, fish and shrimp predominantly accumulated LMW-PAHs and only partly accumulate pyrene and fluoranthene of HMW supported their bioaccumulation rate was predominantly via aquatic food chain (D'Adamo et al., 1997; Zhao et al., 2014). Incomplete combustion is believed as the main source of PAHs compounds in the environment. Important releases of PAHs include fossil fuel combustion, natural gas for industrial or domestic purposes, forest fire, and the materials used for industrial manufacturing (Maliszewska-Kordybach, 1999; WHO, 2000).

HMW–PAHs have been linked to carcinogenic properties as they could promote active metabolite which can bind to DNA (Neff, 1976). Due to the availability of toxicological data, BaP has been used as the chemical index to calculate toxicity equivalent values (TEQ) of PAHs contamination. Concentrations of PAHs in Jakarta Bay sediments in this study were the highest so far reported from Asian coastal regions (Takada et al., 1992; Isobe et al., 2004). In the present study, PAH concentrations in mussels were much higher than those reported earlier from the same location or from other South East Asian bays (Tsutsumi et al., 2002, Isobe et al., 2007).

3.2. Emerging Organic Contaminants

A number of emerging organic compounds have been detected in sediment and biological samples from Jakarta Bay. The contaminants were dominated by products for technical applications, low molecular weight (LMW) PAHs and halogenated compounds (Table 1). Further on, some contaminants that present in the majority of the samples, potentially harm aquatic environment or human health and possibly applicable as marker for identification of anthropogenic emission were assessed. They include LABs, DIPNs, dichlorobenzene, LMW-PAHs and PMN.

3.2.1. Linear alkylbenzenes (LABs)

Linear alkyl benzenes are manufactured in the common surfactant formulation of synthetic detergent, known as linear alkyl benzenes sulfonate/LAS (Eganhouse *et al*., 1983). These compounds belong to a group of secondary phenyl alkane (C_6H_5-C_nH_{2n+1}, n= 10-14). However, as the result of incomplete sulfonation, minor constituent of LABs may remain in the surfactant and are carried over to the detergent. Furthermore, they often appear in municipal and domestic wastes and some would be discharged into riverine system including marine environment. Currently, LABs concentration and profile in sediment, water and aquatic organisms have been widely utilized as molecular tools to assess quality and impact of urban sewage released to the environment (Ishiwatri et al., 1983).

Concentrations of LABs in Jakarta Bay sediments and green mussel samples in the present study was remarkable high and found the highest so far detected in Asian coastal regions (Takada et al., 1992; Isobe et al., 2004). The concentrations in sediment were higher than those from the same location (Rinawati et al., 2012) or from Asian coastal (in Mumbai coast India and in Manila Bay) reported by Isobe et al. (2004). However, no previous reports about LABs concentrations in fish and shrimp species sampled in Indonesian coastal waters or in other Asian regions are available, but the concentration is higher than that in white croaker (*Genyonemus lineatus*) collected near a large wastewater outfall in southern California (Phillips et al., 2001). So far, Asia Pacific regions being the largest market for LABs and account for 46% of total market value. LAS plays as the leading application for LABs (97%), followed by laundry powder production and cleaner application. One of Indonesian company (Unggul Indah Kiat) is among top ten global LABs producers, which are globally dominated by China's companies (Grand View Research, 2015)

3.2.2. Di-iso-propylnaphthalenes (DIPNs)

Various industrial applications use DIPNs, such as carbonless paper, while small amount DIPNs are also applied as heat carrier, solvent for pesticides and for other applications (Suzuki et al., 2007). Contamination of DIPN in sediments of Jakarta Bay in the present study, was the highest concentration reported in coastal sediments (Franke et al., 2007; Terasaki et al., 2012). Additionally, DIPNs concentrations in selected fish species from Jakarta Bay were comparable to reported values of fish from Himeji Coast and Shizuoka coastal waters Japan, which concentrations were 1,2 and 9,0 ng/g respectively (Suzuki et al., 2012; Terasaki et al., 2012). To our knowledge, DIPNs contamination in mussel and shrimp samples has not been reported previously.

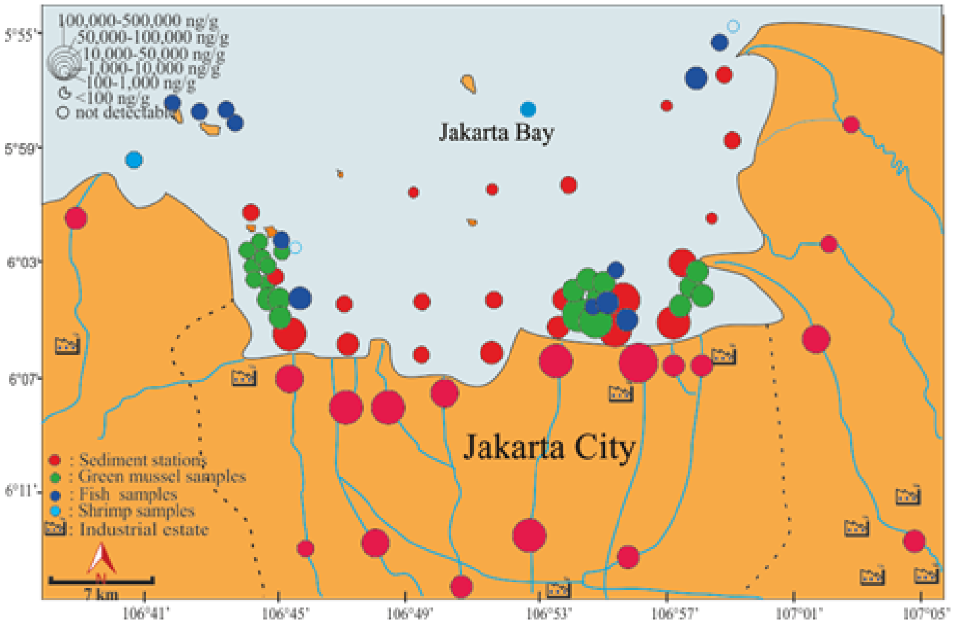

Figure 5. Spatial distribution of LABs contaminant in Jakarta Bay and JMA rivers sediment (ng/g TOC) and in biota samples (ng/g fat).

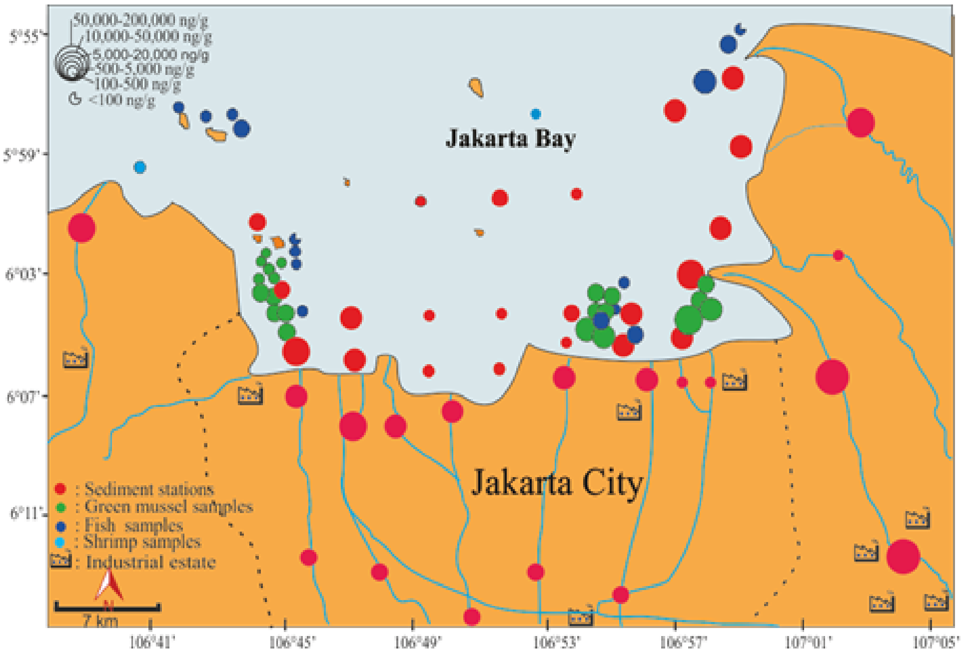

Figure 6. Spatial distribution of DIPNs contaminant in Jakarta Bay and JMA rivers sediment (ng/g TOC) and in biota samples (ng/g fat).

According to Stockholm Convention, DIPNs is classified as not easily biodegradable compound with T_{50} in water of ± 2 months. Further toxicity analysis showed LD_{50} on yellowtail (*Seriola quinqueradiata*) was approx. 2 mL/kg which corresponds to high aquatic toxicity according to the Globally Harmonized System. Long-term toxicity test showed the NOEC study with *Daphnia* was 13 µg/L. Bioassay analysis on rat indicated an increase of liver weight, disturbance of lipid metabolism in the liver and serum and disturbance of glucose metabolism. However no acceptable exposure limits levels are available (UNEP, 2014).

Recently, the Food and Environment Research Agency UK has categorized DIPNs as common contaminants migrating from food contact materials, beside heavy metal, phthalates and polychlorinated

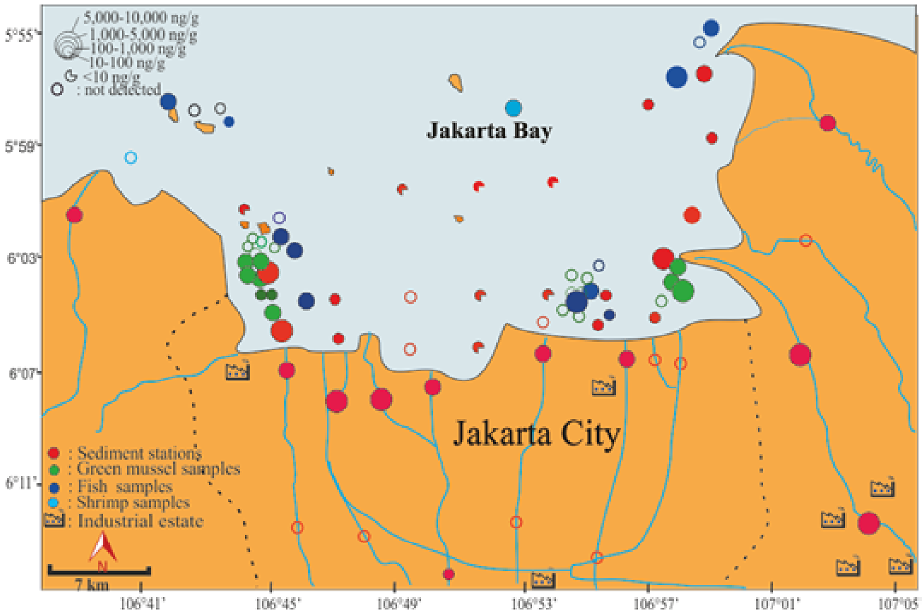

Figure 7. Spatial distribution of PMN contaminant in Jakarta Bay and JMA rivers sediment (ng/g TOC) and in biota samples (ng/g fat).

biphenyl. On a study conducted in the UK detected DIPNs concentration in most samples of retail food packaging at up to 44 mg/kg. Jamnicki et al. (2012) reported that deinking flotation of recycled paper could reduce DIPNs content 50% from 18.2 ng/dm^2 to 9.2 ng/dm^2. Migration simulation test of DIPNs contaminated paper packaging on butter showed migration rate of 74% compared to initial concentration at 22°C (Zülch & Piringer, 2010).

3.2.3. Phenylmethoxynaphthalene

Phenylmethoxynaphthalene (PMN), also known as benzyl naphtyl ether, is classified as irritating substance to the skin and eyes, and as either very toxic to aquatic life with long term effects or may cause long-lasting harmful effects to aquatic life. Acute toxicity test on rainbow trout showed LD$_{50}$ and NOEL value of PMN was >0.69 mg/L. Earlier studies showed that the occurrence of PMN in aquatic environments is related to paper industry emissions (Terasaki et al., 2008 & 2012). Textile manufacturing may also contribute in the emission of PMN contamination into environment (Bridgen et al., 2012), which is used as dye solvent.

In the present study, PMN contamination was detected at relatively similar concentrations in different animal tissue samples at concentrations of 10 to 210 ng/g dw in green mussel and from <10 to 50 ng/g dw in fish samples. PMN concentration in sediments was affected by the season (p-value < 0.05), but this was not observed for animal tissue samples. PMN concentrations in selected fish species in the present study are in line to that reported values for fish samples from Shizuoka coastal waters, which were 3-66 ng/g ww (Terasaki et al., 2012).

3.2.4. Dichlorobenzenes (DCB)

Chlorinated benzenes are frequently detected in riverine and estuarine sediment (Kronimus & Schwarzbauer, 2007). In the present study, DCB were detected in nearly all mussel samples and in some fish species as well as in some river sediment (Table 2). In contrast, DCB was neither appears in coastal sediment nor in shrimp samples. DCBs are either used as product for a large variety of industrial syntheses such as disinfectants, pesticides, deodorizer, air freshener and moth repellent (Baurskens et al., 1994).

The absence of DCB in coastal sediment samples compared to that in green mussel and fish samples suggested this compound is less persistent or more biodegradable in sediment, due to relatively low log K$_{ow}$ (3.43-3.63) compared to other compounds (e.g. DIPNs: 6.08). The relatively high concentration of dichlorobenzene in mussel indicated either the higher uptake from dissolve water phase or the lower clearance rate of this compound in mussel rather than in fish species, which was also reported in hexachlorocyclohexane (HCH) by Bute et al. (1991). Noteworthy, identified DCB in riverine sediment suggested their high concentration or relatively close to the point of source.

Based on the animal study, chlorinated benzenes, especially 1,4-DCB is classified as carcinogenic category 3 compound by EU. Threshold value of NOAEL oral for liver cancer in mice was reported as 300 mg/kg BW per day (EC, 2010). For the aquatic organism, DCB is categorized as very toxic with LC_{50} to fish 1.12-14.2 ppm as well as relatively persistent as the biodegradation rate in surface water and sediment are 4,6%/day and 0,2%/day respectively (US EPA, 1998; EC, 2004). This compound is the main formula in the production of air fresheners, moth repellents and toilet block, besides also used in syntheses of pesticides, resins and dyestuff (Euro Chlor, 1999). In general, these products contain more than 95% of DCB. The use of DCB has been prohibited in Sweden and several federal government of USA (EC, 2010).

4. Conclusions

The present study demonstrated the usefulness of a non-target GC/MS screening method for assessing persistent and emerging organic contaminants in aquatic environment and seafood from Jakarta Bay. Most of the identified contaminants have not been reported previously either from the same location or from South East Asia region, such as DIPNs, DCB, DDMU and PMN. Spatial distribution of the organic contaminants showed that eastern part of Jakarta Bay was contaminated more intensively, mainly by industrial emission, compared to that of western part of the bay, which was predominantly contaminated by municipal discharges. This different contaminant intensity was also reflected on the accumulation of the biota, mainly green mussel.

Among the different compartments, sediment accumulated organic contaminants more variety and more concentrated (particularly river sediment), followed by green mussel, fishes and shrimp. Bioavailability of the compounds (such as due to the polarity, persistency and concentration) as well as the behavior of the compartments (uptake and elimination rate) may undergo the different accumulation rate into the different compartments. Mussels for example are known as sedentary filter feeder that accumulates contaminant mainly from soluble fraction or particulate matters whereas fish predominantly accumulates contaminants from food web through bio-magnification.

Since lipophilic organic compounds are typically accumulated in lipid containing tissue of the animal, it would be worthy to investigate contaminant concentration in different fatty organs. Investigation of low volatilized polar compounds using liquid chromatographic approach is also important to get the whole overview of the organic contaminants.

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