

Organic Pollutants Hazard in Sediments and Green Mussels in Jakarta Bay, Indonesia

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ABSTRACT

37 Organic pollutants (PAHs, PCBs, and organochlorine pesticides (OCPs) from sediments and Asian 38 green mussels (Perna viridis) in Jakarta Bay were studied using GC-MS/MS. The PAHs 39 cocentrations in the sediments ranged from 186.64 to 915.71 µg.kg-1 per sediment dry weight (dw), while the PCBs and the OCPs concentrations ranged from 3 to 117 40 µg.kg-1 and 3 to 17.99 µg.kg-1 per sediment dw, respectively. For the P. viridis, 16 41 types of PAHs concentrations ranged from 591 to 997 µg.kg-1 per dw of mussels, 42 43 while the OCPs concentrations varied from 5 to 6 µg.kg-1 per dw of mussel. The 44 concentration of PAHs in mussels were higher than sediments, meanwhile the 45 organic pollutant contains chlorine (PCBs and OCPs) were potentially found in sediments. The reclamation, urban and mangrove areas (station G and H) in this 46 47 study found more highly risk of than other sampling stations of sediments and 48 mussels. The According to the environmental risk assessment parameters (TEL, ERM, and ERL) 49 from the sediment quality guidelines, the stations has various potential ecological risks, with Station 50 G and H being most contaminated, while Station A being least polluted. Based on l'AFSSA (Agence 51 Francaise de Securité Sanitaire des Aliments, 2003) P. viridis is shown to be highly contaminated 52 in polluted stations (except station A) that were not recommended for consumption (> 200 53 μ g.kg-1 tissue dw). The selected PAHs ratios, including Ant/ Σ 178, Fl/ Σ 202, BaAnt/ Σ 228, and 54 IPyr/5276, potentially showed that the sources of the hydrocarbons in sediments were generally 55 biogenic and terrigenous than for mussels were petroleum source.

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57 Keywords: Persistent organic pollutants, urbanization, risk assessment, anthropogenic
58 pollutants, Asian green mussel

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60 Introduction

Jakarta Bay is a large coastal ecosystem connected to several cluster of small islands, known as the *Kepulauan Seribu* region, and with 13 rivers flowing through the Jakarta megacity area, namely Jabodetabek (Jakarta, Bogor, Depok, Tangerang, and Bekasi) (Williams et al., 2000; Rinawati et al., 2012). According to the statistics service center of the Jakarta Metropolitan Area (JMA), (Badan Pusat Statistik (BPS), 2016), this area is inhabited by more than 10 million people in 2015. Like other large coastal ecosystems, Jakarta Bay 67 supports human welfare and economic systems with four major functions, including life 68 support services, natural resources supply, recreational services, and waste management 69 services. The main ecosystem types in the area (mangroves, seagrass beds, and coral reefs) 70 support the marine life and local communities. Jakarta Bay is a shallow bay with an average depth of 15 m and covers about 514 km², with a coastline of about 72 km. The bay receives 71 72 highly polluted water from the Jakarta metropolitan area which is heavily impacted by the human activities in and around the city as well as in the bay itself. Solid and liquid wastes 73 74 enter the ecosystem from domestic, industrial, and agricultural activities, while the pathogens 75 and nutrients enter from septic tanks and household waste, resulting in massive pollution and 76 eutrophication in the bay waters. In addition, human activities within the bay itself, such as 77 sand and coral mining, petroleum exploration, fishing, and tourism can cause degradation to 78 the bay environment (Williams et al., 2000; Rinawati et al., 2012; Zrafi et al., 2013). There 79 are 13 rivers carrying untreated water and solid waste to Jakarta Bay, resulting in significant 80 environmental degradation and functional loss of coral reefs. The bay is also affected by the 81 degradation of anthropogenic habitats caused by urbanization, climate change-related 82 stressors (global warming, desalination, and hypoxia), and pollution by toxic substances or 83 sedimentation (Rinawati et al., 2012; Wendling et al., 2013).

84 The unsustainable exploitation of natural ecosystems and coastal resources continues to 85 increase due to anthropogenic activities in the aquatic environment. In addition, marine 86 sediments can be considered as the main reservoir of persistent organic pollutants (POPs) 87 (Mandalakis et al., 2014). Sediment has been clearly demonstrated to be a natural recipient of 88 regional marine pollution, which is integrated into the sediment input as a result of deposition, 89 runoff, water mixing, and sedimentation process (Mandalakis et al., 2004). The suspended 90 sediments that enter and leave the estuaries carry sediment-related contaminants. Meanwhile, 91 coastal sediments and related contaminants can be buried temporarily or semi-permanently 92 (Pozo et al., 2013) and maintained undisturbed for long periods of time.

93 This situation leads to anthropogenic impacts on the benthic habitats and species in 94 coastal ecosystems in the marine environment (Wendling et al., 2013; Farhan and Lim, 2012), 95 such as bivalve species (mussels, oysters, and clams). They are filter feeders (Riisgard et al., 2011; Murgarella et al., 2016), known to be capable of accumulating hydrophobic 96 97 pollutants in coastal marine environments through food chain (Yap et al., 2009; Azis et al., 98 2016). As a result, mussel watch as one of bivalve species is widely used for biomonitoring 99 (Farrington et al. 2016). Marine organisms like filter feeder mollusks have a very high 100 ability to accumulate POPs due to a lack of an efficient system for metabolizing these

pollutants (Dsikowitzky et al., 2011; Weis, 2013). Bivalves can also absorb or ingest
pollutants from the food chain, which results in bioaccumulation (Liu and Kuch, 2005;
Galgani et al., 2011).

104 The Asian green mussel, *Perna viridis*, Linnaeus, 1758 (Linnaeus, 1758), is an aquaculture 105 organism that grows fast and serves as a cheap protein source in most Asian coastal countries 106 (NIMPIS 2002., Rajagopal et al., 2006; Azis et al., 2016). It is a major seafood commodity in 107 most Indonesia's coastal markets (DJPB KKP RI., 2011; Ningrum et al., 2016). It is 108 distributed from Persian Bay to the Malay Peninsula and most of the islands of Indonesia, 109 including Sumatra, Java, and Sulawesi, as well as the Philippines (Siddal, 1980). These 110 mussels can live for a long time as non-selective feeders and being tolerant of contaminated 111 areas (Azis et al., 2016; Yap and Azri, 2009). In some eutrophic and human-influenced 112 coastal areas in the western archipelago of Indonesia, such as Jakarta Bay, mussels are present 113 at high densities. For example, in Jakarta Bay, they are harvested by mussel farmers from 114 thousands of bamboo structures, where they live naturally, then sell and consume locally 115 (Yaqin, 2010).

116 The green mussels were first cultivated in Indonesia in the late 1970s. The first attempts 117 at yield optimization were conducted in Jakarta Bay and Banten Bay (Davy and Graham, 118 1982; Huhn et al., 2015). The varying growth rate of *P. viridis* is determined by the quality of 119 the habitat. P. viridis exhibits rapid growth and early maturity. It is highly tolerant of a wide 120 range of water temperatures (11-32°C), salinity (18-33 PSU), turbidity, and pollutants (Lee, 121 1986; Benson et al., 2001; NIMPIS., 2011; Mc Donald, 2012; Mc Donald et al., 2018). In the 122 Jakarta Bay area, the green mussel habitats are affected by high levels of heavy metals and 123 organochlorines (Sudaryanto et al 2005; Jalius et al., 2008; Huhn et al., 2015).

124 The sediments and biotas can act as monitors for the occurrence of hydrophobic organic 125 pollutants such as PAHs, PCBs, and Organochlorine pesticides (OCPs) (Huhn et al., 2015; 126 Asia et al., 2009; Syakti et al., 2013; Kanzari et al., 2014). PAHs, end up in the marine 127 environment from atmospheric deposition and river flows. These compounds have potential 128 mutagenic and carcinogenic effects on humans (Syakti et al., 2013). PAHs may have various 129 origins, including direct deposition of petroleum, natural combustion (e.g., forest fires), 130 anthropogenic combustion through atmospheric deposition (e.g., fuel oil, kerosene, gasoline), 131 volcanic eruptions, and early diagenesis of natural products (Mille et al., 2007). PAHs, either 132 from anthropogenic or biogenic sources (Mille et al., 2007; Asia et al., 2009; Syakti et al., 133 2013), enter the aquatic environment especially from distant transport of particulates (Neff, 134 2002). Mussels can directly assimilate low molecular weight (LMW) PAHs, while heavier molecular 135 weight (HMW) hydrocarbons are congested in the digestive system (Galgani et al., 2011). These 136 aromatic hydrocarbons were alseo have many derivatives such as nitro-PAHs (NPAHs), hydoxy-PAHs 137 (OHPAHs), alkyl-PAHs, oxygenated-PAHs found in particulate matter such as dust (Wei et al., 2015), 138 soil (Wang et al., 2018), PM_{2.5} (Zhang et al., 2019) and ambient air (Nadali et al., 2020). Their 139 toxicities also indicating affect to human health and non-negligible carcinogenic risk to human 140 (Wei et al., 2015; Zhang et al., 2019). Two families of these compounds, PCBs and OCPs, are 141 hydrophobic and biologically accumulated, and have an adverse impact even at low concentrations. 142 They are globally prohibited under the Stockholm Convention on POPs (Strandberg et al., 2000; Pozo 143 et al., 2013; Kanzari et al., 2014). Due to the high octanol-water partition coefficient, PCBs and OCPs 144 tend to bind to suspended particulate matters that are deposited along the coastal sediments.

145 PCBs have been used as commercial industrial materials for electrical equipment (e.g., 146 transformers and capacitors) for over 50 years. But in the other hand, they are considered toxic to 147 the immune and reproductive systems (Pozo et al., 2013; Kanzari et al., 2014). OCPs 148 (Dichlorodiphenyltrichloroethane (DDT), and its analogues (DDD, DDT), hexachlorobenzene, etc.) are 149 toxic to humans, animals, and aquatic life. They have a high nonlethal effect on the immune system 150 and cause reproductive damage (Le Maire et al., 2004). OCPs and PCBs can be absorbed orally, by 151 inhalation, and through the gastrointestinal tract. However, the community continues to use OCPs 152 widely for agricultural pest control in agricultural activities in several developing countries, such as 153 Indonesia, Thailand, Philippines, and Malaysia (Cai and Arataki, 2008; Rinawati et al., 2012; Idowu et 154 al., 2013).

155 Little information is available about the level of PAHs and POPs contamination in the marine 156 sediments and organisms in Indonesia's coastal ecosystems, especially in Jakarta Bay, North Jakarta, 157 Indonesia (Rinawati et al., 2012). Williams et al. (2000), Rinawati et al. (2012), and Azis et al. (2016) 158 published information about several organic pollutant levels in Jakarta Bay, such as n-alkanes, PAHs, 159 PCBs, pp'-DDT, Endosulfan, and pp'-DDD. However, they only analyzed the level of contamination in 160 the sediments of random locations. No studies have yet explored the sources of organic pollutants in 161 this site. Baseline data on the environment are needed to provide an actual point of comparison. If 162 this information is not available, future assessment of the environmental impacts of anthropogenic 163 activities will be difficult to quantify before the impact increases and the larger interventions are 164 required (Mandalakis et al., 2014). Therefore, this study aims to explore qualitatively and 165 quantitatively the extents and sources of organic contaminants, such as PAHs, PCBs, and OCPs, in 166 surface sediments and P. viridis mussels in Jakarta Bay, North Jakarta, Indonesia, using selected ratios 167 or indexes to trace and quantify the compounds.

169 Material and Methods

Sampling was conducted in December 2013 in Jakarta Bay, Indonesia. The preliminary
treatment was carried out in the Analytical Chemistry Laboratory at the Institut Teknologi
Bandung, Indonesia, and POP analysis was carried out in the Environmental Chemistry
Laboratory at the LCE Aix-Marseille University, France.

174

175 Study Area

There are eight sampling stations for the surface sediments (Station A-H) and five sampling stations for the green mussels (*P. viridis*) (Station A, C, D, E, and F) that are located in Jakarta Bay, Indonesia (**Figure 1**). The GPS locations of the sampling sites and sample characterization are shown in **Table 1**. Stations A and B are close to industrial activities, while stations C, D, E, and F are close to Banten Province and touristic sites in *Kepulauan Seribu*. Station G is close to reclamation activities, and station H is close to urban activities and mangrove sites. All sampling sites are approximately 50-200 m from coast.

183

184 Sediment Sampling

185 Quantum GIS mapping software is used to draw maps and visually represent several of 186 the activity around the sampling sites. A GPS (Garmin Etrex Summit HC, Kansas City, USA) 187 is used for the geolocation of different stations. Superficial sediments (4-6 m depth) were 188 collected at 8 stations with specific activities (Figure 1) using a Van Veen stainless steel grab 189 sampler and then immediately placed in an aluminum container and stored in a cool box to 190 minimize microbial degradation during transport to the laboratory. Samples were stored at 191 low temperature (-18°C) before treatment in the laboratory. The wet sediment was frozen dry 192 (Operon, Gimpo city, South Korea) at -50°C and 0,008 bars, sieved with a 200 µm stainless 193 steel sieve and homogenized, before analytical treatment.

194

195 Sampling of The Green Mussels (P. viridis)

The green mussel (P. viridis) samples were harvested at five sampling sites (50-150 cm depth) which were "wild" sites (Stations C, D, and F) and aquaculture facilities (Stations A and E) (Figure 1). The "wild" sites means, the green mussels move and life in randomly habitat (such as refinery tank oil, under ships). The mussels were immediately placed into an aluminum container and stored in a cool box at low temperature (-4°C). The length of the mussels ranged from 4 to 11 cm, and the diameter was approximately 5 cm. The cockles of the mussels were removed from the mussels and then left (at room temperature) under a 203 vacuum hood to dry for 1 h and covered with aluminum foil to avoid air contamination. Their 204 wet mass was then weighed, and then the samples were frozen dry as described above. The 205 Body Condition Index (BCI) is a measure of the physical condition of the mussels and is 206 calculated as dry weight of the soft tissue divided by the mussel dry weight. The higher the 207 BCI, the better the nutritional status and resistance of the mussels to environmental stress 208 (Wang et al. 2011). The dry mass was taken with a microscale (±0.0001) (Perkin-Elmer 209 AD2Z), and the mussels were then cut and manually crushed into powder with a mortar and 210 pestle. The dry tissue powder was then sieved with a 200 µm stainless steel sieve, weighed, 211 and homogenized. It was stored at room temperature before analysis.

212

213 Chemicals

214 All solvents used are *n*-heptane (HEPT), acetone (ACET), dichloromethane (DCM), and acetonitrile (ACN), are of SupraSolv for GC, HPLC and residue analysis \geq 99.9% grade 215 216 (Merck, Darmstadt, Germany). The HCl (37%) is pure grade (Fisher Scientific, 217 Leicestershire, UK). The mixture and individual deuterated internal standards were used such 218 as PAH mix 9 deuterated (napthalene- d_8 , acenaphtylene- d_8 , acenapthene- d_8 , fluorene- d_{10} , 219 phenanthrene- d_{10} , anthracene- d_{10} , fluoranthene- d_{12} , pyrene- d_{12} , benzo(a)anthracene- d_{12} , chrysene- d_{12} , benzo(b) fluoranthene- d_{12} , benzo(k) fluoranthene- d_{12} ; benzo(a) pyrene- d_{12} , 220 221 indeno (123cdi) pyrene-d₁₂; benzo(ghi) perylene-d₁₂; and dibenzo(ah) anthracene-d₁₄), pp'-222 DDT-d₄, pp'-DDE-d₄, PCB-101-d₃ (CIL CLUZEAU, Andover, MA, USA), and [²H₄₀]n-223 nonadecane (Sigma Aldrich, St Louis, MO, USA). 9 individual standard solutions of 224 pesticides are used: pp'-DDT, pp'-DDE, pp'-DDD, HCB, endosulfan-1, aldrin, and dieldrin 225 [CIL CLUZEAU, Andover, MA, USA]; heptachlor and heptachlor epoxide [Sigma-Aldrich, 226 St Louis, MO, USA]. Several standard mixed solutions as mixed solutions were used such as 227 PAH Mix 25 and US EPA 16 PAH (napthalene, acenaphthene, acenaphthylene, fluorene, 228 phenanthrene, anthracene, fluoranthene, pyrene benzo(a)anthracene, chrysene, 229 benzo(b)fluoranthene, benzo(k)fluoranthene; benzo(a) pyrene, indeno (123cdi) pyrene, 230 benzo(ghi)perylene and dibenzo(ah)anthracene, (Dr. Ehrenstorfer Laboratories, Augsburg, 231 Germany)). The PCB mix 3, 7 PCB mix solutions (PCB 28 (2,4,4'-trichlorobiphenyl); PCB 232 52(2,2',5,5'-tetrachlorobiphenyl); PCB 101 (2,2',4,5,5'-pentachlorobiphenyl); PCB 118 233 (2,3',4,4',5-pentachlorobiphenyl); PCB 138 (2,2',3,4,4',5'-hexachlorobiphenyl) PCB-153 234 (2,2',4,4',5,5'-hexachlorobiphenyl); and PCB 180 (2,2',3,4,4',5,5'-heptachlorobiphenyl), (CIL 235 CLUZEAU, Andover, MA, USA) are used. The standard working solutions are diluted with 236 DCM (SGS, France). Alumina, silica gel 60 (200-300 mesh) and Cu powder (> 230 mesh) are

purchased from VWR International and Merck (Darmsdadt, Germany) for the purificationmethod of sediments.

239

240 Extraction and Purification of Sediments

241 Hydrocarbons were extracted from frozen dry sediment samples (10 g) in a cleaned 242 cellulose extraction thimble using a Soxtherm apparatus for 3.5 h with a 150 mL SupraSolv 243 for GC, HPLC, and residue analysis \geq 99.9% grade DCM/HEPT (1:1, v:v) mixture. Then, 500 244 µL of each individual standard of the deuterated hydrocarbons were added to the sample before extraction (PAH mix 9-deuterated 10 mg. L^{-1} and nonadecane-d₄₀ 15 mg. L^{-1}). The 245 extract was then concentrated to approximately 2 mL with a rotary evaporator (Heidolph, 246 247 Laborata 4000 efficient, Krackeler Scientific, USA), and the solvent was evaporated under a 248 gentle nitrogen stream. The extractable organic matter (EOM) was weighed on a microscale 249 (Perkin-Elmer AD2Z) before fractionation. The hydrocarbon fractions were separated with a 250 1.0 x 30 cm borosilicate glass column and applied to half silica (8 g) and half alumina (8 g) 251 (each as bottom and top refill materials in the column). Before use, both sorbents were 252 deactivated with distilled water, which weighed 5% (w:w) of the sorbent. F1 (n-alkane 253 fraction) was eluted with 30 mL HEPT and 20 mL HEPT/DCM (90:10, v:v). Elution with 40 254 mL HEPT/DCM (80:20, v:v) yielded F2 (Polyaromatic hydrocarbon fraction). Both fractions were evaporated on a rotary evaporator and then under a gentle stream of N₂ before being 255 256 weighed on a microscale (Perkin-Elmer AD2Z) for gravimetric analysis. The sum (F1+F2) 257 gave the total hydrocarbon content (THC). The sulfur interference from the extract was 258 removed by completing the extract with active Cu powder (washed with HCl 1:4, v:v) (Mille 259 et al., 2007. Asia et al., 2009; Asia, 2012; Syakti et al., 2013).

260 Several modifications to the extraction methods of Villaverde et al. (2008) and Kanzari et 261 al. (2014) conducted for PCBs and OCPs separation. Those compounds were extracted with 262 ASE (accelerated solvent extraction) using a 33 mL thimble cell with filter fiber on the base 263 of a cell with the following composition: approximately 3 g alumina at the base of cells, then 264 approximately 10 gr sediment and approximately 3 g of Cu powder that had previously been 265 washed with HCl (1:4, v:v). Then, the cell volume was added with Fontainebleau sand. The 266 extraction solvent was HEPT: ACETONE (1:1), and the ASE operating conditions as follows: 267 temperature of 100°C, pressure of 1500 Psi, and 2 extraction cycles. The samples were then 268 evaporated with a TurboVap (Lab Commerce Inc., San Jose, CA) under a nitrogen stream 269 until it reached 0.5 mL. The analysis of the POPs (PCBs and OCs) was carried out through a 270 gas chromatography-triple quadrupole mass spectrometry (GC-TQMS) system.

272 QuEChERS extraction and purification of PAHs and POPs from mussels

273 Several modifications of the Kutty et al. (2011) method were performed in this study. 274 QuEChERS (the quick, easy, cheap, effective, rugged and safe method) is an extraction and 275 cleaning method for marine tissue analysis with GC/MS system (Kutty et al 2011). The 276 method has proven popular for the extraction of pesticides through multi-residue methods and 277 is more sensitive, mainly due to the cleaner product produced, than the Soxtec method 278 (Rashid et al., 2010). As much as 2 g of the freeze-dried mussel tissue was homogenized in a QuEChERS extraction tube. Ten μ L of each internal deuterated standard (10 mg. L⁻¹) was 279 280 added to the samples.

281 The samples were then vortexed for 30 seconds and evaporated for 30 minutes at room 282 temperature. Thirteen mL of deionized water and two ceramic homogenizers were added into 283 a tube containing the dry soft tissue sample of the mussels, and the tube was then vortexed for 284 1 minute. Fifteen mL of extraction solvent (1% acetic acid in acetonitrile) was added, and the 285 tube was vortexed for 1 minute. The tube was then centrifuged for 5 minutes at 2683g and 286 cooled in a freezer at -20°C for 30 minutes. The acetonitrile layer was then transferred into a 287 clean and dry centrifuge tube containing 1 g of anhydrous sodium sulfate. It was shaken by 288 hand for 1 minute, and the tube was then placed in a freezer at -20°C overnight. As much as 289 10 mL of clear extract was pipetted into a clean TurboVap tube, and 20 mL of DCM was 290 added into that tube. The solvent was evaporated to 0.5 mL; next, 10 mL HEPT was added, 291 and the extract was re-evaporated to 0.5 mL (acetonitrile elimination).

In the purification process, 1 g commercial alumina SPE (Solid phase extraction) cartridge was used. The SPE was conditioned with 10 mL of DCM and 20 mL of HEPT. The concentrated extracts were placed into the alumina SPE tube and eluted with 13 mL of DCM:HEPT (dichloromethane: heptane, (40:60)). Then, the extract was evaporated to 0.5 mL, and the purification process was repeated once again and then re-evaporated to 0.5 mL. The extract was analyzed with a GC-TQMS system.

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299 Gas Chromatography - Triple Quadrupole Mass Spectrometry (GC-TQMS)

Both the F1 and F2 fractions from the Soxtherm methods and the extracts from the ASE and QuEChERS methods were analyzed using GC-TQMS system (gas chromatography-triple quadrupole mass spectrometry; Agilent 7000, USA). The analysis parameters were evaluated using the TQ-Quantitative and TQ-Qualitative analysis in the GC/MS software manufacturer (Agilent Technologies, 2010). The chromatographic conditions were as follows: splitless injection (30 s), DB5 MS column (30 m x 0.25 mm i.d. x 0.25 μ m film thickness), GC oven was temperature programmed from 80°C (held for 2 minutes) to 300°C at 12°C.min⁻¹ and then held for 5 minutes. The carrier gas was operated in a constant mode at 3 mL.min^{-1,} and the PSS injector was programmed from 80°C (0.1 minutes) to 250°C (200°C.min⁻¹), held for 10 minutes and then reduced to 60°C (200°C.min⁻¹) to minimize solvent peak.

310 The mass spectrometer was operated in positive electron impact ionization (EI) mode (70 311 eV). The full scan (FS) and selected ion monitoring (SIM) modes were used for hydrocarbon 312 analysis. The PCBs and OCPs analysis were optimally scanned in the product ion mode 313 (PIM) at certain collision energies and then scanned in multiple reaction monitoring (MRM) 314 mode. All molecular targets were compared with molecular weights from a mass spectral 315 library to match the retention time parameters and mass fragmentation profiles against the 316 corresponding the profile of standard solutions. The summarized qualification and 317 quantification ions for the SIM mode are shown in Table 5, and the transition for the MRM 318 mode is shown in Table 6.

319

320 Quality Control and Quality Assurance

The procedures for all data are subject to quality verification and control procedures. All concentration data are based on dry weight (dw), and their analyzes had replicated. The matrix samples are evaluated with proper samples to determine the extraction yield and the % recovery, which are in the range of 80 to 120% for all investigated molecular targets. The detection limit is evaluated for 16 PAHs, which are 0.05 μ g.kg⁻¹ dw, while for PCBs and OCPs in the real samples, they are 0.5 μ g.kg⁻¹ dw and the quantification limit is slightly < 500 μ g.kg⁻¹ dw.

328

329 Results and Discussion

330

331 General Observations

The gravimetric data are shown in **Table 2**. The concentration of extractable organic matter (EOM) (n = 3 replicats) ranged from 980 to 2380 mg.kg⁻¹ sed. dry weight (dw). The lowest and highest values were < 1000 and >2000 mg.kg⁻¹ sed. dw at Stations F and H, respectively. The total hydrocarbon content (THC) varied significantly from 680 (Station F) to 1510 mg.kg⁻¹ (Station E) sed. dw. The proportion of THC in the EOM ranged between 53% (Station H) and 75% (Station A and C). The n-alkane fractions in Jakarta Bay had been described in previous study (Azis et al., 2016). The fraction 1 masses were ranged from 680339 1,440 sed.dry weight mg.kg⁻¹sed dry weight with the total n-alkanes ($n-C_{13}-nC_{33}$) 340 concentrations ranged from 480 to 1,934 µg.kg⁻¹sediment dry weight (dw). Then The ratios of 341 specific n-alkanes index parameters (Mille et al., 2007; Asia et al., 2009; Kanzari et al., 2012; 342 Zrafi et al., 2013; Syakti etal., 2013; Kanzari et al., 2014) showed that the hydrocarbons 343 source in the sediments were generally petrogenic and terrigenous input (in the case of G and 344 H).

345 **PAHs in sediment (including pollution level and sources)**

346 The sum of 16 PAHs listed by US-EPA priority pollutants (U.S. EPA., 1996; Keith L.H., 347 2015; Wise et al., 2015) were quantified in Table 3. The sum of 16 PAHs values ranged from 348 186.64 to 915.70 µg.kg-1 dw. The total PAHs content was relatively high in the sediments from Stations C, D, E, G, and H (428.19 to 915.71 µg.kg-1 dw) comparing with station A, B 349 350 and F (below 300 µg,kg-1 dw). The industrial activites in A and B stations obtained a few 351 PAHs, because most of activities from electrical industry and home industry of otomotive. 352 Station F was from tourist activity, means the kind of plastics most found in that station. 353 Stations C, D, and E were connected to the mangrove areas which were the target of tourist 354 visits by boat, while Stations G and H were close to urban areas. The lowest concentrations 355 occurred at Stations A, B, and F (186.64 to 210.81 µg.kg-1 dw). The last stations were located 356 in industrial area (Stations A and B) and in tourist destination (Station F) in the Kepulauan 357 Seribu region. G and H stations were polluted area reached 915.71 µg.kg-1 dw, due to the 358 downstream through from rivers and canal near urban areas of Jakarta City. Those stations 359 were near the fisheries village and cultivation areas of green mussels, The stations G, H were 360 more polluted than others. Meanwhile, the stations C,D and E were potentially polluted were 361 separated from different activitires (transportatoon, tourist and fisheries cultivation area), The 362 PAHs renge concentration levels from this research were the same order of magnitude as 363 previously reported in the bay of Jakarta (257 to 1511 µg.kg-1 dw, Rinawati et al., 2012), (< 364 10 to 550 µg.kg-1 dw; Williams et al., 2000) and the last study was found from river sediment in Jakarta, the PAHs concentrations ranged 1992 to17635 ng.g⁻¹ dw (Rinawati et al.m 2017) 365 dw. Several investigation of PAHs concentration from previous studies proven that Jakarta 366 367 Bay was potentially increasing of PAHs by time in sediment. The concentration levels in this 368 research were still lower than at other sites such as Segara Anakan Lagoon in Indonesia (200 to 25,000 µg.kg⁻¹ dw; Dsikowitzky et al., 2011), the Gulf of Fos in the Mediterranean (27 to 369 2700 µg.kg⁻¹ dw; Mille et al., 2007), and Tokyo Bay (1372 to 1615 µg.kg⁻¹ dw; Rinawati et 370 al., 2012), Qinhuai river and Xuanwu lake (796.2 ng.g-1 to 10.470 ng.g-1 dw; Zhao et al., 371 372 2017), Bintan island indonesia (13.492 ng.g-1; Yogaswara et al., 2019). Meanwhile, PAHs

range in sediment from several sites of Jakarta bay were still higher than other sites such as in
Bizerte Lagoon (2-538 µg.kg-1 dw; Louiz et al., 2008), in Euphrates river, Iraq (0.197-80.006
ng.g-1 dw; Al Saad et al., 2016),. Those comparison with other sites could shown the
baseline reference and evaluating recent environmental impact of PAHs in sediment.

377 Several ratio indices have been used to indicate the different sources of PAHs in sediment. 378 PAHs isomers with mass to charge ratio, m/z 178 (Ant and Phe) and m/z 202 (Fl and Pyr), 379 have been widely used to differentiate between sources of combustion process and 380 petroleum.

381 The ratio (Fl/Fl+Pyr) value of 0.50 was generally defined as the oil/combustion transition 382 point (Yunker et al., 2002). This limit appeared to be more definitive than 0.10 for Ant/ Σ 178. 383 $Fl/\Sigma 202$ was < 0.50 for petroleum combustion samples, such as gasoline, diesel, fuel oil, and 384 crude oil combustion, and > 0.40 for grass, wood/coal combustion samples (Syakti et al., 385 2013). When the Ant/ Σ 178 ratio was < 0.1, a petroleum combustion was inferred, and values 386 > 0.1 indicated a pyrolytic origin (Budzinsky et al. 1997; Syakti et al., 2013). More 387 accurately, the Ant/ Σ 178 ratio was > 0.1 for oil, coal, and some crude oil samples and below 388 0.1 for lignite, diesel, and fuel oil emissions (Mille et al., 2007; Syakti et al., 2013). The ratio 389 of anthracene/[anthracene+phenanthrene] (An/ Σ 178) more than 0.1 indicated a combustion 390 source, while a ratio under 0.1 indicated a pyrolytic source (Budzinski et al., 1997). The ratio 391 of fluoranthene/[fluoranthene+pyrene] (Fl/ Σ 202) or fluoranthene/pyrene (Fl/Py) were 392 characteristic of a pyrolytic source if < 0.5, and a ratio value > 0.5 potentially indicated a 393 combustion source, the Σ methyl phenanthrene/phenantrene ratio from 0.5 to 1.0 indicated 394 pyrogenic sources, and more than 2 indicated pyrolytic sources (Mille et al., 2007; Asia et al., 395 2009; Asia, 2012; Syakti et al., 2013; Kanzari et al., 2014).

High molecular weight PAHs, such as B[a]A, Chry, IPyr, and B[ghi]P, were generally minor
contributors to refined petroleum products (Wang et al., 2001) and were usually present in a
significant quantities only in products with higher molecular weight fractions, such as asphalt
(Readman et al., 2002; Syakti et al., 2013) and possibly bitumen or coal (Asia, 2012).

Figure 2 showed the use of the triple diagnostic ratio $Fl/\sum 202$ vs. $Ant/\sum 178$, $Fl/\sum 202$ vs IndP/ \sum 276 and $Fl/\sum 202$ vs. $B[a]A/\sum 228$ to differentiate the source of PAHs. Interestingly, plotting $Fl/\sum 202$ vs $Ant/\sum 178$ as well as $B[a]A/\sum 228$ indicated both a mixed source and a petroleum origin. Confirmation was obtained using the correlation of the $Fl/\sum 202$ vs. IP/ $\sum 276$, showing petroleum combustion for the majority of the sediments. Plotting $Fl/\sum 202$ vs. vs. $Ant/\sum 178$ and $Fl/\sum 202$ vs. $B[a]A/\sum 228$ identified that the PAHs in the sediment were 406 from both petroleum and combustion sources, while the $IndP/\Sigma$ 276 and Fl/Σ 202 identified 407 that PAHs were from mixed combustion of grass/coal/wood and petroleum. In **Table 4**, we 408 have distinguished the basis of the different criteria for the PAH origin. It clearly appears that 409 several pyrogenic or petrogenic origins or mixtures of the two can be proposed for the 410 essential and the most frequent sources of PAHs in the sediments of Jakarta Bay, Indonesia.

411

412 PAHs in green mussels, *P.viridis*

413 The PAHs concentrations in green mussels was higher than that in sediment. The levels 414 found in P. viridis were more than 900 µg.kg-1 dw at Station C, D, and F. Meanwhile, each of 415 Station A and E showed lower levels of 591 and 777 µg.kg-1 dw (see Table 5). These values 416 were higher than those reported by Isobe et al. (2007), who reported that 19 PAHs contained 417 in P. viridis ranged from 11 to 1133 µg.kg-1 dw in several locations in South and Southeast 418 Asia. The total PAHs values were still lower than those in previous studies in Asia. 419 Mokkongpai et al. (2010) reported that 16 PAHs ranged from 2330 to 3000 and from 456,80 420 to 3115 µg.kg-1 dw on the Chonburi Coast in Thailand and in the eastern coast of China, 421 respectively.

422 Any dissolved organic material were able to transfer into fatty tissue, particularly that found 423 in organs in closest contact with aqueous fluids, e.g. kidneys. The lower the solubility of an 424 organic compound in water, the greater is its ability to accumulate in fatty tissues and the 425 greater is the potential for toxic effect (Reeve R.N., 2002., NIMPIS 2011). That is the 426 potentially reason why several high molecular weight PAHs, such as Benzo(a)pyrene which 427 have lower solubility were almost not detected or below the limit of detection in sediment.

428

429 **PCBs and OCPs in sediment**

The totals of the PCBs in the sediments ranged from 3 to 117 µg.kg⁻¹ sed. dw The highest 430 PCB concentrations (101 and 116 µg.kg⁻¹ sed dw) corresponding to highly polluted sediments 431 432 were found at stations G and H, which clearly demonstrated the prevalence of pollution in 433 urban areas and mangrove sites. Stations A, B, C, D, E and F were slightly polluted, with 3 to 11 µg.kg⁻¹ sed. dw. Mangrove sites can expect to affect the speciation and bioavailability of 434 435 pollutants which associated with observed toxic effects in mangrove ecosystem species 436 (Dsikowitzky et al., 2011; Bayen S., 2012). In addition the sediment morphology of stations 437 G and H were mud ones, those means, organic material concentrations more abundance than 438 other stations. Moreover, congeners containing 3-5 chlorine atoms (trichloro (CB 28), 439 tetrachloro (CB 52), pentachloro (CB 101 and CB 118)) were identified at all stations. Then,

440 the 6-7 chlorine atoms (hexachloro (CB 138 and CB 153) and heptachloro (CB 180) were 441 only found at station H. The summary of the PCB contamination in surface sediments was 442 shown in Fig 3. Several studies can be compared with the PCB contamination levels in the 443 sediments from Jakarta Bay, which were higher than those at several other locations in Jakarta Bay since 2011, which were found to be 2-9 µg.kg⁻¹ dw, and those in Tokyo Bay, which were 444 found to be 37-47 µg.kg⁻¹ dw (Rinawati et al., 2012). Additionally, the levels in Jakarta Bay 445 were higher than those in other locations around the world, such as the Hauveane River in 446 France, the levels were 27.2-36.4 µg.kg⁻¹ dw (Kanzari et al., 2014), although they were lower 447 than those in Mazaleh Lake (125-330 μ g.kg⁻¹ dw), Mariout Lake (4-304 μ g.kg⁻¹ dw) and the 448 Nile River (53-1500 µg.kg⁻¹dw) in Egypt (Yamashita et al., 2000). On the other hand, PCB 449 450 compounds were not detected in green mussels at any mussel sampling station.

451 Nine OCPs were analyzed, but only three OCP molecular weights were detected in the surface sediments (pp'-DDE, hexachlorobenzene and pp'-DDT + pp'-DDD) (Fig 4). Only 452 453 one organochlorine pesticide (pp'-DDE) was found in the tissues of the green mussel P. 454 viridis. The fact that many of the PCBs and organochlorine compounds were not detected in 455 the sediments or in the green mussels does not mean that these compounds were not present in 456 the samples. A specific protocol adapted for both different matrices could probably be used to 457 detect lower concentrations. The total OCPs in the sediment ranged from 2.93 to 17.99 µg.kg⁻ ¹ sed. dw. A previous study in Jakarta Bay (Williams et al., 2000) reported that the pp'-DDE 458 had the highest concentration among the organochlorines (1-7 μ g.kg⁻¹ dw), while other areas 459 had lower levels than those in Jakarta Bay, such as the Hauveane River in France (0.06-5.48 460 µg.kg⁻¹dw; Kanzari et al., 2014) and Bothnian Bay of the Baltic Sea (0.48-1.1 µg.kg⁻¹dw; 461 Strandberg et al., 2000). We observed that the highest concentration levels of OCPs in the 462 463 sediments were found at stations G and H. These sites indicated that agricultural activities 464 were near urban areas and mangrove sites. Hexachlorobenzene and pp'-DDT/pp'-DDD were 465 found in stations A, B (near industrial and port area); E and F (near tourist destinations, small islands, and Kepulauan Seribu) had lower concentrations than pp'-DDE. The pp'DDE 466 concentrations, which was found in the mussel shell dry mass with a range of 5-6 µg.kg⁻¹ 467 468 mussel dw, was found at all sampling stations (A, C, D, E, F). However, pp'-DDE was the 469 largest contributor at all sampling stations in the sediments and green mussels.

470

471 Evaluation of the Ecotoxicological Risk of PAHs, PCBs and pp'DDE in the Sediments

The ecological risk assessment of sediment uses several references, such as the sediment quality guidelines (SQG). The SQGs, including the sediment quality criteria, sediment quality

474 objectives, and sediment quality standards, have been applied and developed by various 475 federal states and provincial agencies in North America for freshwater and marine ecosystems 476 (Long and MacDonald, 1998; Kanzari et al., 2014). This assessment was developed to 477 monitor remediation quality (Long and MacDonald, 1998). However, the average SQGQs had 478 several important limitations and underlying assumptions that should be understood by 479 sediment quality assessors. This paper provided an overview of the derivation methods and 480 some of the principal advantages, assumptions, and limitations of using this sediment 481 assessment tool. Ideally, the average SQGQs should be included with other measures 482 including toxicity test results and benthic community surveys to provide weighted evidence 483 when assessing the relative quality of contaminated sediments (Long et al., 2006). Specifically, for the PAHs, the published SQGs for the protection of sediment-dwelling 484 485 organisms in freshwater ecosystems were grouped (Long and MacDonald, 1998; Long et al. 486 2006) into two categories, threshold effect levels (TELs) and effect range low values (ERLs) 487 (Figure 5). The ERL guideline value represented the concentrations above for which adverse 488 toxicological effects can be detected, and the TEL represented the concentration below which 489 was expected to have an adverse effect on pollutants in the sediment (Burton et al., 2002). 490 Sediments that have concentrations between the TEL and ERL should be considered to have 491 potential toxicity with possible adverse effects. The contamination level at Stations G and H are higher than recommended by the TEL (870 µg.kg⁻¹ dw) but lower than the ERL value, 492 which clearly indicated contamination for severe PAHs (4-5 rings, such fluoranthene, pyrene, 493 494 benzo(a) pyrene, chrysene, benzo(b) fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene) 495 (Kanzari et al., 2014). However, other stations with lower values than the TEL values rarely 496 contained PAHs contamination in the sediments (Burton et al., 2002). Concerning the total 497 PCB concentrations, Stations G and H were higher than the ERM (effect range median; 22.7 $ug.kg^{-1}$ dw), but they were still lower than the TEL (threshold effect Level: 180 $ug.kg^{-1}$ dw). 498 In addition, 6 stations (Station A, B, C, D, E, and F) were lower than the median range. The 499 500 NOAA guidelines defined assessments for $\Sigma pp'$ -DDE concentrations and have four parameters, the probable effect level (PEL) (6.75 µg.kg⁻¹ dw), effect range low (ERL; 2.0 501 μg.kg⁻¹ dw), threshold effect level (TEL; 1.42 μg.kg⁻¹ dw), and effect range median (ERM). 502 503 Six stations (Stations A, B, C, D, E, and F) were identified between the TEL and ERL. Then, 504 two stations (Stations G and H) were shown to have possible risk to the environment, and the 505 biota caused their values to be higher than the PEL and lower than the ERM. The plotted 506 SQGs of the PCBs and pp'DDE in the sediments were shown in Figure 6. This evaluation 507 confirmed that Station G and H had higher potential risks to the environment and biota 508 compared to other stations. This data can be used as a baseline reference for evaluating the 509 current environmental conditions on this site for efficient environmental management in the 510 future and to improve the quality of the bay.

511

512 Evaluation of the Ecotoxicological Risk of PAHs in Perna viridis

513 Bioaccumulation or biomagnification of organic pollutant and heavy metal in the marine biota 514 such as fish, mussels, mangrove can possible impact on human health. (Kumari et al., 2018; 515 Chowdury, A and Maity, S.K., 2016) According to l'AFSSA (Avis d AFFSA, Agence Francaise de Securité 516 Sanitaire des Aliments, 2003), the risk of contamination was evaluated by the sum of the heavy 517 6 PAHs (benz(a)anthracen, benzo(b+j)fluoranthen, benzo(k)fluoranthen, benzo(a)pyren, dibenzo(a,h)antracen). The limit for the consumption PAHs contaminated seafood was 0.2 518 mg.kg⁻¹ tissue dw or 200 µg.kg⁻¹ tissue dw. This threshold applied to crustaceans and 519 mollusks, which were exposed to contaminants. Most of the green mussels in Jakarta Bay 520 located in polluted stations that were not recommended for consumption (> 200 μ g.kg⁻¹ tissue 521 522 dw), and those at Station A should not be consumed.

523

524 Conclusions

525 The concentration of 16 PAHs in the sediments ranged from 186.64 to 915.71 μ g.kg⁻¹ sed. dw. The concentration of 7 PCBs ranged from 2.85-116.49 μ g.kg⁻¹ sed. 526 dw, and the total OCPs ranged from 2.97-17.99 µg.kg⁻¹ sed. dw. In the mussels, the 527 concentration of 16 PAHs ranged from 591 µg.kg⁻¹ to 997 µg.kg⁻¹ mussel dw, and the OCPs 528 concentrations varied from 5 to 6 µg.kg⁻¹ mussel dw. The selected PAHs ratios, including 529 Ant/ Σ 178, Fl/ Σ 202, BaAnt/ Σ 228, and IPyr/ Σ 276, potentially showed that the sources of the 530 531 hydrocarbons in sediments were generally biogenic and terrigenous. The ecotoxicological 532 evaluation results, based on the international sediment quality guidelines (SQGs), showed that stations have environmental risks between low and medium-low, except for Stations G and H, 533 534 which had higher level of ecological risk for marine organisms. The consumption safety 535 evaluation showed that these mussels had been contaminated, and it was not recommended 536 for human consumption. This data can be used for baseline reference concentrations as part of 537 an integrated watershed management program involving biodiversity, freshwater ecology, and 538 economic activities in the study area. More extensive and comprehensive monitoring of 539 organic pollutants, such as the persistent organics and emerging pollutants, will form a more 540 reliable baseline and can serve as a reference for evaluating compliance with environmental 541 regulations.

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792 List of Tables

	Station		Sampli	Sediment	
Code	GPS Cordinate	Depth	Sediment	Perna viridis (PV)	morphology
St A	S 06 ⁰ 6'19.800'' E 106 ⁰ 46'48.202''	4 m(sed) ± 5-20 cm (PV)	V	\checkmark	Brown, Coarse sand
St B	S06 ⁰ 6'19.800'' E106 ⁰ 46'28,700'	4 m (sed) ± 5-20 cm (PV)	\checkmark	-	Brown, Coarse sand
St C	S 06 ⁰ 4'28.976'' E 106 ⁰ 43'21.201''	5 m(sed) ± 5-20 cm (PV)	\checkmark	\checkmark	Brown, Coarse sand
St D	S 06 ⁰ 3'4.097'' E 106 ⁰ 42'58.914''	60 m(sed) ± 5-20 cm (PV	\checkmark	\checkmark	Brown, fine sand
St E	S 06 ⁰ 4'55.920'' E 106 ⁰ 44'7.300''	5 m(sed) ± 5-20 cm (PV	\checkmark	\checkmark	Black green, fine silt
St F	S 06 ⁰ 2'53.160'' E 106 ⁰ 44'7.300''	5 m(sed) ± 5-20 cm (PV	\checkmark	\checkmark	Black green, silt
St G	S 06 ⁰ 5'56.692'' E 106 ⁰ 44'50.276''	6 m(sed) ± 5-20 cm (PV	\checkmark	-	Black mud, gravel or granules
St H	S 06 ⁰ 6'4.392''	6 m(sed)	\checkmark	-	Black mud, gravel or granules

Table 1. GPS coordinates of the sampling locations

796 **Table 2**. Gravimetric data of the sediment samples from Jakarta Bay, Indonesia

Station	EOM (mg.kg ⁻¹)	F1 (mg.kg ⁻¹)	F2 (mg.kg ⁻¹)	F1/F2	THC (mg.kg ⁻¹)	F1/THC %	F2/THC %	THC/EOM %
А	1060	700	100	7	800	88	12	75
В	1870	1280	60	21	1340	96	4	72
С	1630	1120	110	10	1230	91	9	75
D	1150	680	120	6	800	85	15	70
Е	2380	1440	70	21	1510	95	5	63
F	980	620	60	10	680	91	9	69
G	1683	980	158	6	1139	86	14	68
Н	2240	990	200	5	1190	83	17	53

EOM: extractible organic matter, THC: total hydrocarbons content, F1: Fraction 1 and F2: Fraction 2.

U	110	in Jakarta	Day, maoi	με.κ	5 uw).				
	Ratios of	StA	St B	St C	St D	St E	St F	St G	St H
	PAHs								
	Naph	0	0	212.71	438.42	257.88	0	139.11	261.78
	Ace	3	1.91	1.63	1.56	2.52	2.01	2.37	7.65
	Acen	2	2.50	5.17	3.78	3.46	2.56	12.38	10.58
	Flu	44	2.70	6.30	5.10	3.44	4.03	16.50	26.73
	Phe	132	11.12	15.57	18.52	9.84	17.84	44.29	54.18
	Ant	44	2.52	4.13	3.87	2.96	3.02	6.43	11.02
	Fluo	19.52	18.12	12.95	20.32	9.53	16.05	36.21	47.11
	Pyr	40.48	36.48	22.46	47.68	15.78	29.21	42.36	54.93
	BzA	3.05	5.09	3.69	9.57	1.56	1.48	7.79	23.41
	Chrys	15.97	10.41	15.27	12.28	10.14	8.31	33.10	68.52
	BzbF	10.71	9.60	10.28	11.20	9.69	8.59	17.67	86.60
	BzkF	<LOD	nd	nd	nd	< LOD	nd	< LOD	Nd
	BzaPyr	<LOD	<LOD	< LOD	< LOD	nd	< LOD	< LOD	< LOD
	Dibenzanth	47.05	42.71	43.18	43.35	43.90	42.37	45.12	82.59
	Bper	1.35	2.20	5.00	5.43	8.63	4.67	20.28	108.87
	IndP	47.03	46.69	49.56	50.33	48.85	46.51	56.97	71.75
	\sum PAHs	210.81	192.04	407.90	671.41	428.19	186.64	480.56	915.70
	$(An/\Sigma 178)$	0.24	0.19	0.21	0.17	0.23	0.15	0.13	0.17
	$(Fluo/\Sigma 202)$	0.33	0.33	0.37	0.30	0.38	0.36	0.46	0.46
	$(BzA/\overline{\Sigma}228)$	0.16	033	0.19	0.44	0.13	0.15	0.19	0.25
	$(IndP/\overline{\Sigma}276)$	0.97	0.96	0.91	0.90	0.85	0.91	0.74	0.40
	HMW	85.67	75.35	68.21	100.82	47.53	74.71	160.53	212.19
	LMW	125.15	116.70	126.98	132.17	122.77	111.92	180.92	441.73
	LMW/HMW	1.46	1.55	1.86	1.31	2.58	1.50	1.13	1.95
	Phe/An	3.26	4.41	3.77	4.79	3.32	5.91	6.89	4.92
	Fl/Py	0.48	0.50	0.58	0.43	0.60	0.55	0.85	0.86
	BzA/Chrys	0.19	0.49	0.24	0.78	0.15	0.18	0.24	0.34
	IndP/Bper	20.01	21.22	9.91	9.27	5.66	9.96	2.81	0.66
	∑C1 Phe/Phe	0.99	0.66	1.02	0.77	0.71	0.61	1.04	1.21

799 Table 3. Index parameter of priority the sum of 16 PAHs (USEPA) in the surface sediments from Jakarta Bay, Indonesia ($ug kg^{-1} dw$) 800

Sediments (St A-St H), Mussels, Perna viridis (PVA-PVF), HMW High Molecular Weight = = Naphthalene; Acy = Acenaphtylene; Ace = Acenaphtene; Fluo = Flurene; Phe=Phenanthrene; Anth=Anthracene; Fluor = Fluoranthene; Pyr = Pyrene; BZA = Benzo(a) anthracene; Chrys = Chrysene; BzbF =Benzo (b)Fluoranthene; BzkF= Benzo(k)Fluoranthene; Bzapyr = Benzo(a)pyrene; IndP = Indeno(123cdi)pyrene; Bper = Benzo(ghi)Perylene; C1Phe = Methyl Phenanthrene; (Mille et al., 2007; Asia, 2012; Kanzari et al., 2014), nd = not detected, LOD = Limit of Detection 806 807

Dation	Pyrolytic		Mixed	Mixed origin		Petrogenic origin	
Ratios	Sediment	Sediment Mussel Sediment Mussel		Mussel	Sediment	Mussel	
An/Σ178	> 0.1 A,B,C,D,	>0.1 A,C,D,E,	1	-	< 0.1	-	
Phe/An	 < 10 A,B,C,D, E,F,G,H 	 F - < 10 A,C,D,E, F 		10-15	> 15	> 15	
Fl/Σ202	> 0.5	> 0.5	0.4-0.5 G,H	-	< 0.4 A,B,C,D, E,F	-< 0.4 A,C,D,E, F	
Fl/Py	> 1	> 1	1	1	< 1 A,B,C,D, E,F,G,H	< 1 A,C,D,E, F	
BzA/Chry	> 0.4 B,D	> 0.4 A,C,D		-	< 0.4 A,C,E,F, G,H	< 0.4 E,F	
BzA/Σ228	> 0.35 B,D	> 0.35 A,C,D	0.2-0.35 C,G,H	-	< 0.2 A,E,F	< 0.2 E,F	
IndP/Σ276	> 0.5 (wood,pla nt, coal combustio n) A,B,C,D, E.F.G.H	> 0.5 (wood, plant,coal combustio n) A, C,E	0.2-0.5 (fuel combustio n H	0.2-0.5 (fuel combust ion -	< 0.2	< 0.2 D,F	
LMW/HMW	>1 A,B,C,D, E,F,G,H	>1	1	1	< 1	< 1 A,C,D,E, F	
Phe/Phe +ΣC1Phe	0.5-1 A,B,C,D, E,F,G,H		> 1		>2		

809 **Table 4.** Classification of the PAHs from sampling stations according to different criteria

HMW = High Molecular Weight (Fluor+Pyr+Bz(a)anth+Chry+IndP+Bper+Dibenzoper); LMW = Low 810 810 811 812 813 Molecular Weight (LMW = Naph+Acy+Fluo+Ace+Phe+Anth); Naph = Naphthalene; Acy = Acenaphtylene; Ace = Acenaphtene; Fluo = Flurene; Phe=Phenanthrene; Anth=Anthracene; Fluor = Fluoranthene; Pyr = Pyrene; Benzo(a) anthracene; Chrys = Chrysene;BzbF =Benzo (b)Fluoranthene; BzA = BzkF= 814 Benzo(k)Fluoranthene;Bzapyr = Benzo(a)pyrene; IndP = Indeno(123cdi)pyrene; Bper = Benzo(ghi)Perylene; 815 C1Phe = Methyl Phenanthrene; C2Phe = Ethyl Phenanthrene (Mille et al., 2007; Asia, 2012; Kanzari et al., 816 2014)

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Table 5. Index parameter of priority the sum of 16 PAHs (USEPA) in the green mussels,

Compounds (m/z)	PV A	PV C	PV D	PV E	PV F
Naphthalene (128)	122.63±2.91	128.16±12.03	146.67±5.25	93.43±1.25	81.49±2.63
Acénaphthene (152)	< LOD	< LOD	< LOD	5.55±0.38	5.18±0.26
Fluorene (166)	17.17±1.70	6.99±0.64	5.22±0.33	8.27±0.22	7.78±0.14
Phénanthrene (178)	37.50±7.87	27.13±1.95	10.56±0.01	25.73±0.40	25.84±1.31
Anthracene (178)	11.33±2.04	5.89±0.61	7.09 ± 0.07	6.82±0.25	7.35±0.40
Pyrene (202)	83.47±8.50	145.61±5.46	36.71±0.33	115.06±4.16	117.29±1.97
Fluoranthene (202)	35.96±1.20	41.47±2.56	15.23±1.06	43.93±0.24	32.66±0.92
Chrysene (228)	23.42±0.74	24.39±3.51	6.96±0.21	1.25±0.09	1.30±0.04
Benzo(a)Anthracene (228)	27.43±1.41	22.85±4.17	6.56±0.37	< LOD	< LOD
Benzo(a)pyrene (252)	189.01±20.10	486.89±43.18	755.03±61.26	409.96±3.02	627.84±4.48
Indeno(1,2,3cdi)pyrene	30.16±2.93	18.82±0.75	1.34±0.03	14.64±1.82	< LOD
Dibenzo(ah)Anthracene (278)	13.58±0.91	< LOD	< LOD	10.46±0.13	< LOD
Benzo(ghi)perylene (276)	< LOD	14.17±0.95	4.61±1.05	< LOD	833.10±178.87
ΣΡΑΗ	591.66±50.31	922.37±84.38	995.98±69.97	735.1±11.9	1739.84±190.97
LMW	188.63	168.17	169.54	134.22	127.65
HMW	403.03	754.20	826.44	580.66	1612.19
LMW/HMW	0.46	0.22	0.21	0.23	0.08
Phe/Anth	3.31	4.60	1.49	3.77	3.52
Anth/ $\Sigma 178$	0.23	0.18	0.29	0.21	0.22
Fluo/Σ202	0.30	0.22	0.29	0.28	0.22
Fluo/Pyr	0.43	0.28	0.41	0.38	0.28
$Bz(a)Ant/\Sigma 228$	0.54	0.48	0.48	0	0
Bz(a)Ant/Chry	1.17	0.94	0.94	0	0
IndP/2276	1	0.57	0.22	1	0
pp'-DDE	6.19±0.17	5.64±0.01	5.14±0.85	4.65±0.29	4.61±0.19

Perna viridis, from Jakarta Bay, Indonesia (μ g.kg⁻¹ dw±SD).

Mussels, *Perna viridis* (PVA-PVF), HMW = High Molecular Weight (Fluor+Pyr+Bz(a)anth+Chry+IndP+Bper+Dibenzoper); LMW = Low Molecular Weight (LMW = Naph+Acy+Fluo+Ace+Phe+Anth); Naph = Naphthalene; Acy = Acenaphtylene; Ace = Acenaphtene; Fluo = Flurene; Phe=Phenanthrene; Anth=Anthracene; Fluor = Fluoranthene; Pyr = Pyrene; BzA = Benzo(a) anthracene; Chrys = Chrysene; BzbF = Benzo (b)Fluoranthene; BzkF= Benzo(k)Fluoranthene; Bzapyr = Benzo(a)pyrene; IndP = Indeno(123cdi)pyrene; Bper = Benzo(ghi)Perylene; C1Phe = Methyl Phenanthrene; C2Phe = Ethyl Phenanthrene (Mille et al., 2007; Asia, 2012; Kanzari et al., 2014)



Fig 1. Sampling sites of the sediments and the mussels, *Perna viridis*, from Jakarta Bay, Indonesia; the majority of the stations were situated near Jakarta Bay. 1= Garage, 2 = Processing industry, 3 = Health service; 4 = Government service; 5 = Education service; 6= Workshop service; 6= Industrial Area; 7= Cemetery; 8 = Central Business; 9 = Agency Business; 10 = Market; 11 = Storage; 12 = Fishery; 13 = Agricultural Wetland; 14 = Dry land farming; 15 = Regular housing; 16 = Irregular housing; 17 = Livestock; 18 = Transport infrastructure; 19 = Open space; 20 = Vacant land allocated; 21 = vacant land allocated





petroleum

grass/wood/coal

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Fig 6. Sedimentary guidelines values for effect range median (ERM) and threshold effect level (TEL) in the PCBs and the probable effect level (PEL), effect range low (ERL), threshold effect level (TEL) and effect range median (ERM) values for the PCBs and pp'-DDE in the sediments from Jakarta Bay (Long et al., 1998; Burton et al., 2008)

Table 5. MRM (multiple reaction monitoring) mode for the PCBs and OCPs

Compounds	Qu	Quantification transition			Co	onfirm	ation transi	tion
	Precurs	or	Product	Collision	Precursor ion		Product	Collision
	ion (am	u)	ion	Energy	(amu)	1	ion	energy (V)
			(amu)	(V)			(amu)	
	I	V	Vindow 1 (1	1,5 to 15,6 mi	nute)			
Aldrin	298,0	\rightarrow	263,0	8	263,0	\rightarrow	191,0	30
Hexachlorobenzen	283,9	\rightarrow	248,8	25	283,9	\rightarrow	213,9	35
Heptachlore	271,1	\rightarrow	237,0	10				
PCB 28	256,0	\rightarrow	186,0	26	258,0	\rightarrow	186,0	26
PCB 52	289,9	\rightarrow	220,0	28	291,9	\rightarrow	222,0	28
		,	Window 2 (1	15,6 to 25 min	ute)			
Chlordecone	271,7	\rightarrow	236,8	10	236,8	\rightarrow	143,0	15
Dieldrin	262,8	\rightarrow	193,0	30	262,8	\rightarrow	191,0	15
Endosulfan 1	240,9	\rightarrow	205,9	10	240,9	\rightarrow	169,9	20
Heptachlor epoxy	352,7	\rightarrow	262,9	20	352,7	\rightarrow	316,9	20
ppDDD	237,0	\rightarrow	165,0	20	235,0	\rightarrow	199,1	8
pp'DDD d4	242,9	\rightarrow	173,1	20				
pp'DDE	248,0	\rightarrow	176,0	30	246,0	\rightarrow	211,0	20
pp'DDT	237,0	\rightarrow	165,0	20	235,0	\rightarrow	199,1	20
pp'DDE d4	253,9	\rightarrow	184,2	30				
PCB 101	323,9	\rightarrow	253,9	28	325,9	\rightarrow	255,9	28
PCB 101 d3	328,7	\rightarrow	258,9	20				
PCB 118	323,9	\rightarrow	253,9	28	325,9	\rightarrow	255,9	28
PCB 138 and 153	357,0	\rightarrow	287,9	28	359,8	\rightarrow	289,9	28
PCB 156 d3	363,0		293,0	20				
PCB 180	393,8		323,9	30	395,8	\rightarrow	325,9	30

Target compounds (standard marguer)	Qualification m/z (amu)	Quantification m/z (amu) (standard marguer)
Naphtalene (d_8)	127	128 (136)
Acenaphtylene (d_{10})	-	154 (162)
Acenaphtene (d_{10})	-	152 (160)
Fluorene (d_{10})	-	166 (176)
Phénanthrene / Anthracene (d_{10}/d_{10})	174/ 174	178 (188)
Fluoranthene / Pyrne (d_{10}/d_{10})	200/200	202
Benzo(a) anthracene / Chrysene (d_{12}/d_{12})	226/226	228 (240)
Benzo(b)Fluoranthene	250	
Benzo(k)fluoranthene		252 (264)
Benzo(a)pyrene $(d_{12}/d_{12}/d_{12})$		
Indéno (1,2,3) cd pyrene /	276/274	
Benzo(g,h,i)pérylene (d, d)		276 (288)
(a_{12}/a_{12}) Dibenzo(a,h)anthracene (d_{14})	274	278 (292)

Table 6. SIM (selected ion monitoring) mode for the PAHs