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

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8 Muhammad Yudhistira Azis^{1,2}, Anne Piram², Laurence Asia², Buchari Buchari¹, Pierre
9 Doumenq², Agung Dhamar Syakti^{3,4,*}
10

11 ¹Analytical Chemistry Division, Department of Chemistry – Faculty of Mathematics and
12 Natural Sciences, Institut Teknologi Bandung, Jl Ganesha No 10, Bandung 40132, Indonesia

13
14 ².Aix Marseille Univ, CNRS, LCE, Marseille, France

15
16 ³Environmental Science Department – Raja Ali Haji Maritime University, Jl. Politeknik
17 Senggarang-Tanjungpinang, Riau Islands Province, 29100, Indonesia

18
19 ⁴Center for Maritime Biosciences Studies – Institute for Sciences and Community Service-
20 Jenderal Soedirman University, Kampus Karangwangkal, Jl. dr. Suparno, Purwokerto 53123,
21 Indonesia
22
23
24
25
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27
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31

32 * Corresponding author. Tel./Fax: +62 771 4500089/+62 771 4500090;

33 E-mail address: agungsyakti@chemist.com (A.D. Syakti)
34

ABSTRACT

Organic pollutants (PAHs, PCBs, and organochlorine pesticides (OCPs) from sediments and Asian green mussels (*Perna viridis*) in Jakarta Bay were studied using GC-MS/MS. The PAHs concentrations in the sediments ranged from 186.64 to 915.71 $\mu\text{g.kg}^{-1}$ per sediment dry weight (dw), while the PCBs and the OCPs concentrations ranged from 3 to 117 $\mu\text{g.kg}^{-1}$ and 3 to 17.99 $\mu\text{g.kg}^{-1}$ per sediment dw, respectively. For the *P. viridis*, 16 types of PAHs concentrations ranged from 591 to 997 $\mu\text{g.kg}^{-1}$ per dw of mussels, while the OCPs concentrations varied from 5 to 6 $\mu\text{g.kg}^{-1}$ per dw of mussel. The concentration of PAHs in mussels were higher than sediments, meanwhile the organic pollutant contains chlorine (PCBs and OCPs) were potentially found in sediments. The reclamation, urban and mangrove areas (station G and H) in this study found more highly risk of than other sampling stations of sediments and mussels. The According to the environmental risk assessment parameters (TEL, ERM, and ERL) from the sediment quality guidelines, the stations has various potential ecological risks, with Station G and H being most contaminated, while Station A being least polluted. Based on l'AFSSA (Agence Francaise de Sécurité Sanitaire des Aliments, 2003) *P. viridis* is shown to be highly contaminated in polluted stations (except station A) that were not recommended for consumption ($> 200 \mu\text{g.kg}^{-1}$ tissue dw). The selected PAHs ratios, including Ant/ $\Sigma 178$, Fl/ $\Sigma 202$, BaAnt/ $\Sigma 228$, and IPyr/ $\Sigma 276$, potentially showed that the sources of the hydrocarbons in sediments were generally biogenic and terrigenous than for mussels were petroleum source.

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

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
71 depth of 15 m and covers about 514 km², with a coastline of about 72 km. The bay receives
72 highly polluted water from the Jakarta metropolitan area which is heavily impacted by the
73 human activities in and around the city as well as in the bay itself. Solid and liquid wastes
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
96 al., 2011; Murgarella et al., 2016), known to be capable of accumulating hydrophobic
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

101 pollutants (Dsikowitzky et al., 2011; Weis, 2013). Bivalves can also absorb or ingest
102 pollutants from the food chain, which results in bioaccumulation (Liu and Kuch, 2005;
103 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 (Galvani et al., 2011). These
136 aromatic hydrocarbons were also have many derivatives such as nitro-PAHs (NPAHs), hydroxy-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.

168

169 Material and Methods

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

174

175 **Study Area**

176 There are eight sampling stations for the surface sediments (Station A-H) and five
177 sampling stations for the green mussels (*P. viridis*) (Station A, C, D, E, and F) that are located
178 in Jakarta Bay, Indonesia (**Figure 1**). The GPS locations of the sampling sites and sample
179 characterization are shown in **Table 1**. Stations A and B are close to industrial activities,
180 while stations C, D, E, and F are close to Banten Province and touristic sites in *Kepulauan*
181 *Seribu*. Station G is close to reclamation activities, and station H is close to urban activities
182 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*)**

196 The green mussel (*P. viridis*) samples were harvested at five sampling sites (50-150 cm
197 depth) which were "wild" sites (Stations C, D, and F) and aquaculture facilities (Stations A
198 and E) (Figure 1). The "wild" sites means, the green mussels move and life in randomly
199 habitat (such as refinery tank oil, under ships). The mussels were immediately placed into an
200 aluminum container and stored in a cool box at low temperature (-4°C). The length of the
201 mussels ranged from 4 to 11 cm, and the diameter was approximately 5 cm. The cockles of
202 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
215 acetonitrile (ACN), are of SupraSolv for GC, HPLC and residue analysis $\geq 99.9\%$ grade
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 (naphthalene- d_8 , acenaphthylene- d_8 , acenaphthene- d_8 , fluorene- d_{10} ,
219 phenanthrene- d_{10} , anthracene- d_{10} , fluoranthene- d_{12} , pyrene- d_{12} , benzo(a)anthracene- d_{12} ,
220 chrysene- d_{12} , benzo(b) fluoranthene- d_{12} , benzo(k) fluoranthene- d_{12} ; benzo(a) pyrene- d_{12} ,
221 indeno (123cdi) pyrene- d_{12} ; benzo(ghi) perylene- d_{12} ; and dibenzo(ah) anthracene- d_{14}), pp'-
222 DDT- d_4 , pp'-DDE- d_4 , PCB-101- d_3 (CIL CLUZEAU, Andover, MA, USA), and [$^2\text{H}_{40}$]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 (naphthalene, 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

237 purchased from VWR International and Merck (Darmsdadt, Germany) for the purification
238 method 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
245 before extraction (PAH mix 9-deuterated 10 mg. L^{-1} and nonadecane- d_{40} 15 mg. L^{-1}). The
246 extract was then concentrated to approximately 2 mL with a rotary evaporator (Heidolph,
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
255 were evaporated on a rotary evaporator and then under a gentle stream of N_2 before being
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.

271

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
279 QuEChERS extraction tube. Ten μL of each internal deuterated standard (10 mg. L^{-1}) was
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).

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

298

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

300 Both the F1 and F2 fractions from the Soxtherm methods and the extracts from the ASE
301 and QuEChERS methods were analyzed using GC-TQMS system (gas chromatography-triple
302 quadrupole mass spectrometry; Agilent 7000, USA). The analysis parameters were evaluated
303 using the TQ-Quantitative and TQ-Qualitative analysis in the GC/MS software manufacturer
304 (Agilent Technologies, 2010). The chromatographic conditions were as follows: splitless

305 injection (30 s), DB5 MS column (30 m x 0.25 mm i.d. x 0.25 μm film thickness), GC oven
306 was temperature programmed from 80°C (held for 2 minutes) to 300°C at 12°C.min⁻¹ and
307 then held for 5 minutes. The carrier gas was operated in a constant mode at 3 mL.min⁻¹, and
308 the PSS injector was programmed from 80°C (0.1 minutes) to 250°C (200°C.min⁻¹), held for
309 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**

321 The procedures for all data are subject to quality verification and control procedures. All
322 concentration data are based on dry weight (dw), and their analyzes had replicated. The
323 matrix samples are evaluated with proper samples to determine the extraction yield and the %
324 recovery, which are in the range of 80 to 120% for all investigated molecular targets. The
325 detection limit is evaluated for 16 PAHs, which are 0.05 $\mu\text{g.kg}^{-1}$ dw, while for PCBs and
326 OCPs in the real samples, they are 0.5 $\mu\text{g.kg}^{-1}$ dw and the quantification limit is slightly < 500
327 $\mu\text{g.kg}^{-1}$ dw.

328

329 **Results and Discussion**

330

331 **General Observations**

332 The gravimetric data are shown in **Table 2**. The concentration of extractable organic
333 matter (EOM) ($n = 3$ replicats) ranged from 980 to 2380 mg.kg^{-1} sed. dry weight (dw). The
334 lowest and highest values were < 1000 and >2000 mg.kg^{-1} sed. dw at Stations F and H,
335 respectively. The total hydrocarbon content (THC) varied significantly from 680 (Station F)
336 to 1510 mg.kg^{-1} (Station E) sed. dw. The proportion of THC in the EOM ranged between 53%
337 (Station H) and 75% (Station A and C). The n-alkane fractions in Jakarta Bay had been
338 described in previous study (Azis et al., 2016). The fraction 1 masses were ranged from 680-

339 1,440 sed.dry weight mg.kg^{-1} sed dry weight with the total n-alkanes ($n\text{-C}_{13}\text{-nC}_{33}$)
340 concentrations ranged from 480 to 1,934 $\mu\text{g.kg}^{-1}$ sediment 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 et al., 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 $\mu\text{g.kg}^{-1}$ dw. The total PAHs content was relatively high in the sediments
349 from Stations C, D, E, G, and H (428.19 to 915.71 $\mu\text{g.kg}^{-1}$ dw) comparing with station A, B
350 and F (below 300 $\mu\text{g.kg}^{-1}$ dw). The industrial activities in A and B stations obtained a few
351 PAHs, because most of activities from electrical industry and home industry of automotive.
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 $\mu\text{g.kg}^{-1}$ 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 $\mu\text{g.kg}^{-1}$ 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 activities (transportation, tourist and fisheries cultivation area), The
362 PAHs range concentration levels from this research were the same order of magnitude as
363 previously reported in the bay of Jakarta (257 to 1511 $\mu\text{g.kg}^{-1}$ dw, Rinawati et al., 2012), (<
364 10 to 550 $\mu\text{g.kg}^{-1}$ dw; Williams et al., 2000) and the last study was found from river sediment
365 in Jakarta, the PAHs concentrations ranged 1992 to 17635 ng.g^{-1} dw (Rinawati et al. 2017)
366 dw. Several investigation of PAHs concentration from previous studies proven that Jakarta
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
369 to 25,000 $\mu\text{g.kg}^{-1}$ dw; Dsikowitzky et al., 2011), the Gulf of Fos in the Mediterranean (27 to
370 2700 $\mu\text{g.kg}^{-1}$ dw; Mille et al., 2007), and Tokyo Bay (1372 to 1615 $\mu\text{g.kg}^{-1}$ dw; Rinawati et
371 al., 2012), Qinhuai river and Xuanwu lake (796.2 ng.g^{-1} to 10.470 ng.g^{-1} dw; Zhao et al.,
372 2017), Bintan island Indonesia (13.492 ng.g^{-1} ; Yogaswara et al., 2019). Meanwhile, PAHs

373 range in sediment from several sites of Jakarta bay were still higher than other sites such as in
374 Bizerte Lagoon (2-538 $\mu\text{g.kg}^{-1}$ dw; Louiz et al., 2008), in Euphrates river, Iraq (0.197-80.006
375 ng.g^{-1} dw; Al Saad et al., 2016),. Those comparison with other sites could shown the
376 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/ Σ 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).

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

400 **Figure 2** showed the use of the triple diagnostic ratio Fl/ Σ 202 vs. Ant/ Σ 178, Fl/ Σ 202 vs
401 IndP/ Σ 276 and Fl/ Σ 202 vs. B[a]A/ Σ 228 to differentiate the source of PAHs. Interestingly,
402 plotting Fl/ Σ 202 vs Ant/ Σ 178 as well as B[a]A/ Σ 228 indicated both a mixed source and a
403 petroleum origin. Confirmation was obtained using the correlation of the Fl/ Σ 202 vs.
404 IP/ Σ 276, showing petroleum combustion for the majority of the sediments. Plotting Fl/ Σ 202
405 vs. Ant/ Σ 178 and Fl/ Σ 202 vs. B[a]A/ Σ 228 identified that the PAHs in the sediment were

406 from both petroleum and combustion sources, while the IndP/Σ 276 and Fl/Σ202 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 Makkongpai 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**

430 The totals of the PCBs in the sediments ranged from 3 to 117 µg.kg⁻¹ sed. dw The highest
431 PCB concentrations (101 and 116 µg.kg⁻¹ sed dw) corresponding to highly polluted sediments
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
434 11 µg.kg⁻¹ sed. dw. Mangrove sites can expect to affect the speciation and bioavailability of
435 pollutants which associated with observed toxic effects in mangrove ecosystem species
436 (Dzikowitzky 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
444 Bay since 2011, which were found to be 2-9 $\mu\text{g.kg}^{-1}$ dw, and those in Tokyo Bay, which were
445 found to be 37-47 $\mu\text{g.kg}^{-1}$ dw (Rinawati et al., 2012). Additionally, the levels in Jakarta Bay
446 were higher than those in other locations around the world, such as the Hauveane River in
447 France, the levels were 27.2-36.4 $\mu\text{g.kg}^{-1}$ dw (Kanzari et al., 2014), although they were lower
448 than those in Mazaleh Lake (125-330 $\mu\text{g.kg}^{-1}$ dw), Mariout Lake (4-304 $\mu\text{g.kg}^{-1}$ dw) and the
449 Nile River (53-1500 $\mu\text{g.kg}^{-1}$ dw) in Egypt (Yamashita et al., 2000). On the other hand, PCB
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
452 surface sediments (pp'-DDE, hexachlorobenzene and pp'-DDT + pp'-DDD) (**Fig 4**). Only
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 $\mu\text{g.kg}^{-1}$
458 sed. dw . A previous study in Jakarta Bay (Williams et al., 2000) reported that the pp'-DDE
459 had the highest concentration among the organochlorines (1-7 $\mu\text{g.kg}^{-1}$ dw), while other areas
460 had lower levels than those in Jakarta Bay, such as the Hauveane River in France (0.06-5.48
461 $\mu\text{g.kg}^{-1}$ dw; Kanzari et al., 2014) and Bothnian Bay of the Baltic Sea (0.48-1.1 $\mu\text{g.kg}^{-1}$ dw;
462 Strandberg et al., 2000). We observed that the highest concentration levels of OCPs in the
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
466 islands, and *Kepulauan Seribu*) had lower concentrations than pp'-DDE. The pp'DDE
467 concentrations, which was found in the mussel shell dry mass with a range of 5-6 $\mu\text{g.kg}^{-1}$
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**

472 The ecological risk assessment of sediment uses several references, such as the sediment
473 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).
484 Specifically, for the PAHs, the published SQGs for the protection of sediment-dwelling
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
492 are higher than recommended by the TEL ($870 \mu\text{g}\cdot\text{kg}^{-1} \text{ dw}$) but lower than the ERL value,
493 which clearly indicated contamination for severe PAHs (4-5 rings, such fluoranthene, pyrene,
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
498 $\mu\text{g}\cdot\text{kg}^{-1} \text{ dw}$), but they were still lower than the TEL (threshold effect Level; $180 \mu\text{g}\cdot\text{kg}^{-1} \text{ dw}$).
499 In addition, 6 stations (Station A, B, C, D, E, and F) were lower than the median range. The
500 NOAA guidelines defined assessments for $\Sigma\text{pp}'\text{-DDE}$ concentrations and have four
501 parameters, the probable effect level (PEL) ($6.75 \mu\text{g}\cdot\text{kg}^{-1} \text{ dw}$), effect range low (ERL; 2.0
502 $\mu\text{g}\cdot\text{kg}^{-1} \text{ dw}$), threshold effect level (TEL; $1.42 \mu\text{g}\cdot\text{kg}^{-1} \text{ dw}$), and effect range median (ERM).
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,
518 dibenzo(a,h)antracen). The limit for the consumption PAHs contaminated seafood was 0.2
519 mg.kg⁻¹ tissue dw or 200 µg.kg⁻¹ tissue dw. This threshold applied to crustaceans and
520 mollusks, which were exposed to contaminants. Most of the green mussels in Jakarta Bay
521 located in polluted stations that were not recommended for consumption (> 200 µg.kg⁻¹ tissue
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
526 915.71 µg.kg⁻¹ sed. dw. The concentration of 7 PCBs ranged from 2.85-116.49 µg.kg⁻¹ sed.
527 dw, and the total OCPs ranged from 2.97-17.99 µg.kg⁻¹ sed. dw. In the mussels, the
528 concentration of 16 PAHs ranged from 591 µg.kg⁻¹ to 997 µg.kg⁻¹ mussel dw, and the OCPs
529 concentrations varied from 5 to 6 µg.kg⁻¹ mussel dw. The selected PAHs ratios, including
530 Ant/Σ178, Fl/Σ202, BaAnt/Σ228, and IPyr/Σ276, potentially showed that the sources of the
531 hydrocarbons in sediments were generally biogenic and terrigenous. The ecotoxicological
532 evaluation results, based on the international sediment quality guidelines (SQGs), showed that
533 stations have environmental risks between low and medium-low, except for Stations G and H,
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.

542

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549

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792 **List of Tables**793 **Table 1. GPS coordinates of the sampling locations**

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

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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 %
A	1060	700	100	7	800	88	12	75
B	1870	1280	60	21	1340	96	4	72
C	1630	1120	110	10	1230	91	9	75
D	1150	680	120	6	800	85	15	70
E	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
H	2240	990	200	5	1190	83	17	53

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

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

Ratios of PAHs	St A	St B	St C	St D	St E	St F	St G	St H
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
Σ PAHs	210.81	192.04	407.90	671.41	428.19	186.64	480.56	915.70
(An/ Σ 178)	0.24	0.19	0.21	0.17	0.23	0.15	0.13	0.17
(Fluo/ Σ 202)	0.33	0.33	0.37	0.30	0.38	0.36	0.46	0.46
(BzA/ Σ 228)	0.16	0.33	0.19	0.44	0.13	0.15	0.19	0.25
(IndP/ Σ 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
Σ C1 Phe/Phe	0.99	0.66	1.02	0.77	0.71	0.61	1.04	1.21

801 Sediments (St A-St H), Mussels, *Perna viridis* (PVA-PVF), HMW = High Molecular Weight
 802 (Fluor+Pyr+Bz(a)anth+Chry+IndP+Bper+Dibenzoper); LMW = Low Molecular Weight (LMW = Naph+Acy+Fluo+Ace+Phe+Anth); Naph
 803 = Naphthalene; Acy = Acenaphthylene; Ace = Acenaphthene; Fluo = Flurene; Phe=Phenanthrene; Anth=Anthracene; Fluor = Fluoranthene;
 804 = Pyrene; BzA = Benzo(a) anthracene; Chrys = Chrysene;BzbF =Benzo (b)Fluoranthene; BzkF= Benzo(k)Fluoranthene;Bzapyr =
 805 Benzo(a)pyrene; IndP = Indeno(123cdi)pyrene; Bper = Benzo(ghi)Perylene; C1Phe = Methyl Phenanthrene; (Mille et al., 2007; Asia, 2012;
 806 Kanzari et al., 2014) , nd = not detected, LOD = Limit of Detection
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809 **Table 4.** Classification of the PAHs from sampling stations according to different criteria

Ratios	Pyrolytic origin		Mixed origin		Petrogenic origin	
	Sediment	Mussel	Sediment	Mussel	Sediment	Mussel
An/Σ178	> 0.1 A,B,C,D, E,F,G,H	>0.1 A,C,D,E, F -	1	-	< 0.1	-
Phe/An	< 10 A,B,C,D, E,F,G,H	< 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, plant, coal combustion)	> 0.5 (wood, plant, coal combustion)	0.2-0.5 (fuel combustion)	0.2-0.5 (fuel combustion)	< 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	

810 HMW = High Molecular Weight (Fluor+Pyr+Bz(a)anth+Chry+IndP+Bper+Dibenzoper); LMW = Low
811 Molecular Weight (LMW = Naph+Acy+Fluo+Ace+Phe+Anth); Naph = Naphthalene; Acy = Acenaphthylene;
812 Ace = Acenaphthene; Fluo = Flurene; Phe=Phenanthrene; Anth=Anthracene; Fluor = Fluoranthene; Pyr = Pyrene;
813 BzA = Benzo(a) anthracene; Chrys = Chrysene;BzbF =Benzo (b)Fluoranthene; 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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827 **Table 5.** Index parameter of priority the sum of 16 PAHs (USEPA) in the green mussels,
 828 *Perna viridis*, from Jakarta Bay, Indonesia ($\mu\text{g}\cdot\text{kg}^{-1}$ dw \pm SD).

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

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

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836 **List of Figures**

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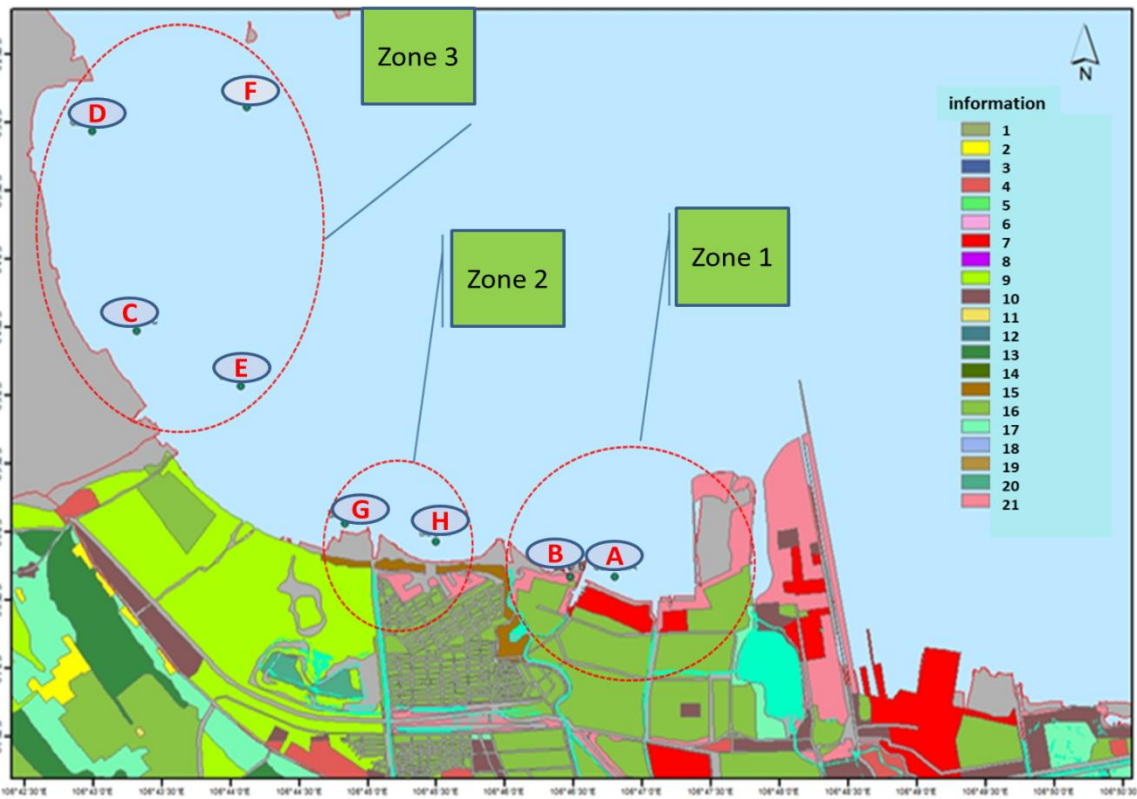


Fig 1. Sampling sites of the sediments and the mussels, *Perna viridis*, from Jakarta Bay, Indonesia; the majority of the stations were situated near Jakarta Bay. 1= Garage, 2 = Processing industry, 3 = Health service; 4 = Government service; 5 = Education service; 6= Workshop service; 6= Industrial 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

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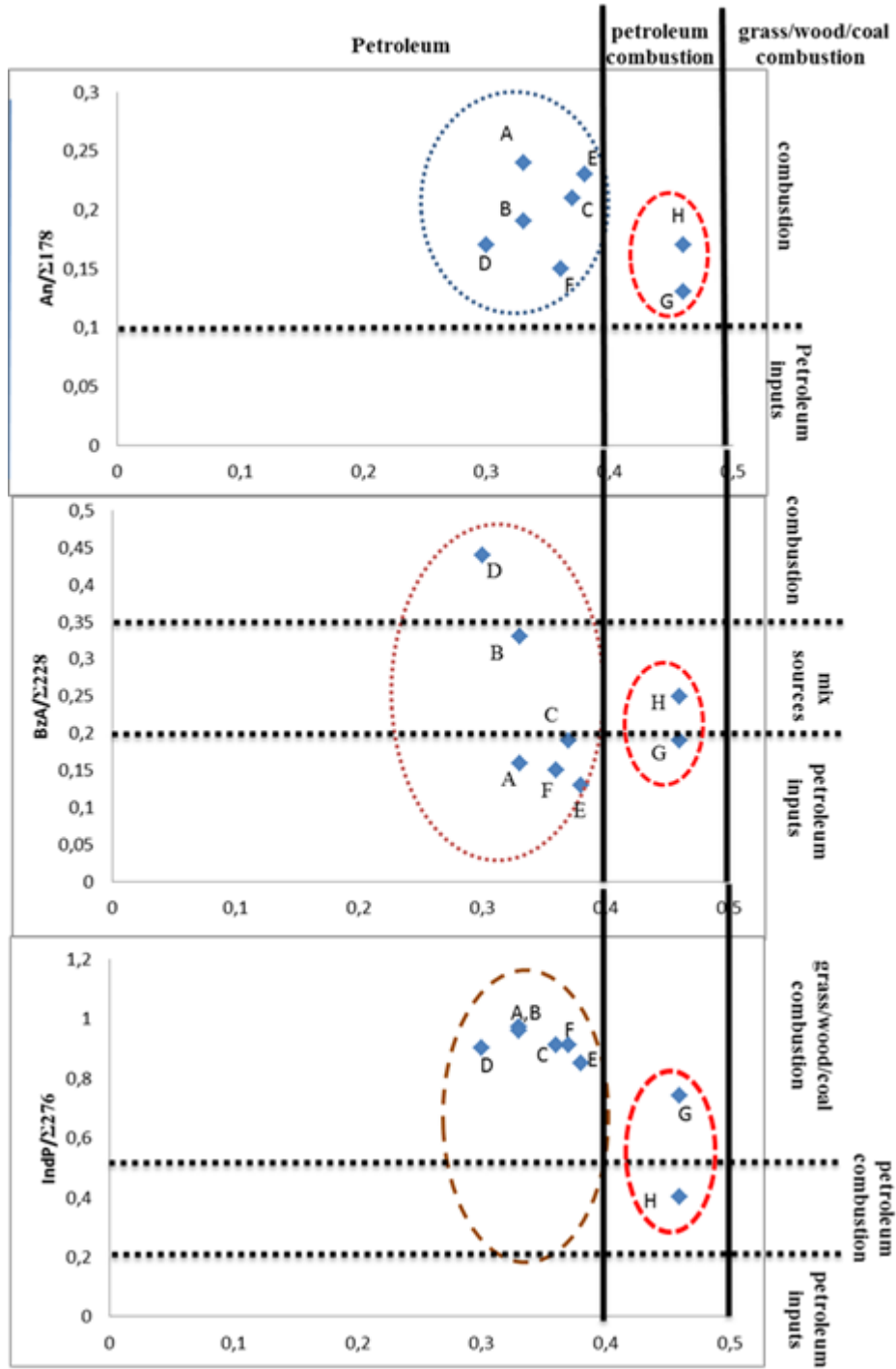


Fig 2. Cross plots of the isomeric ratios Fl/Σ202 vs. An/Σ178 vs (star shapes), B[a]A/Σ228 (square shapes) and IPyr/Σ276 (triangle shapes) in the sediment. Literature boundary ratios were derived from Yunker et al. (2002).

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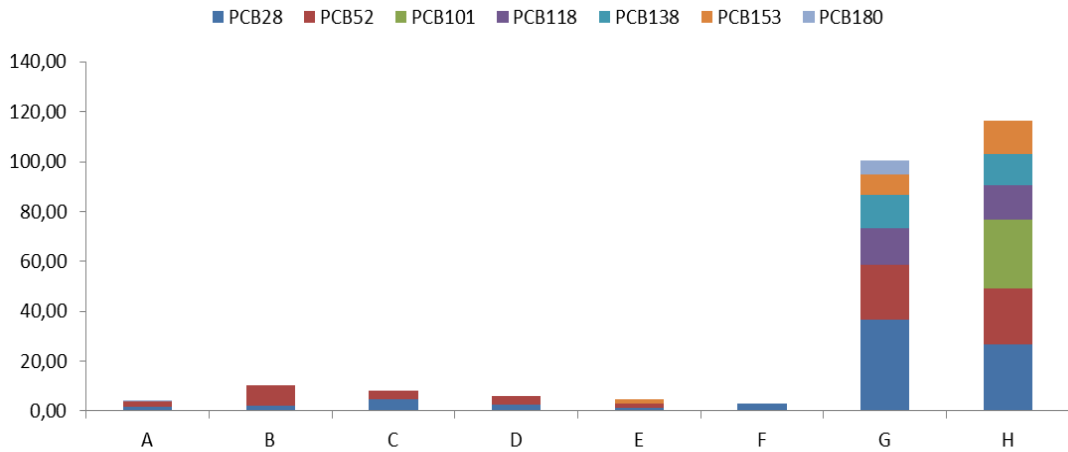


Fig 3. Concentration of PCBs ($\mu\text{g.kg}^{-1}$ sediment dw) at station A-H (Jakarta Bay, Indonesia).

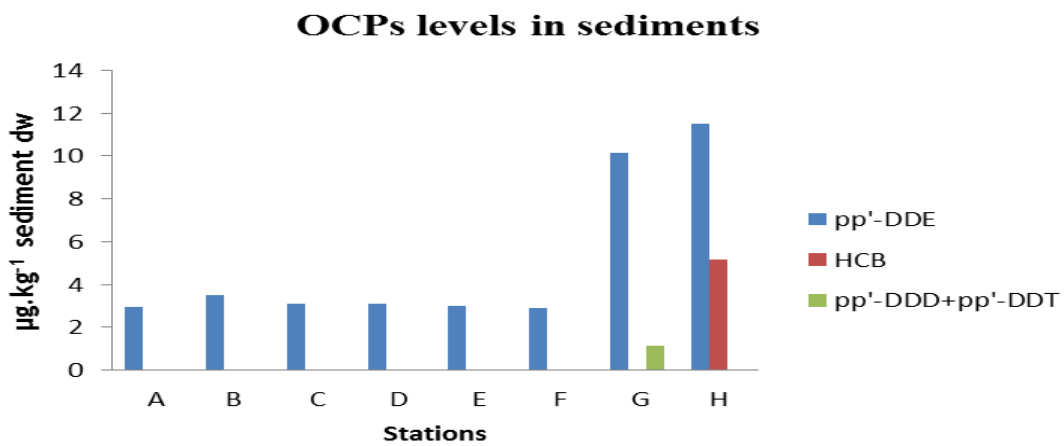


Fig 4. Concentration of OCs ($\mu\text{g.kg}^{-1}$ dw) in the sediments and mussels from Jakarta Bay, Indonesia.

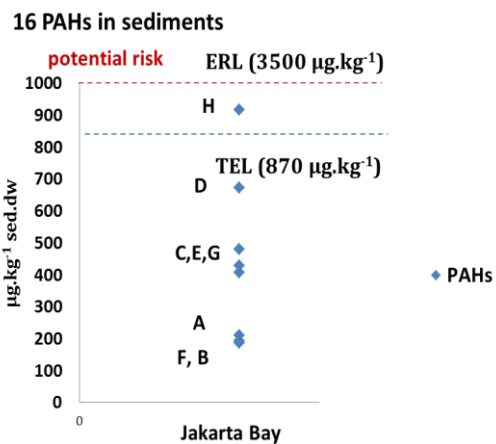
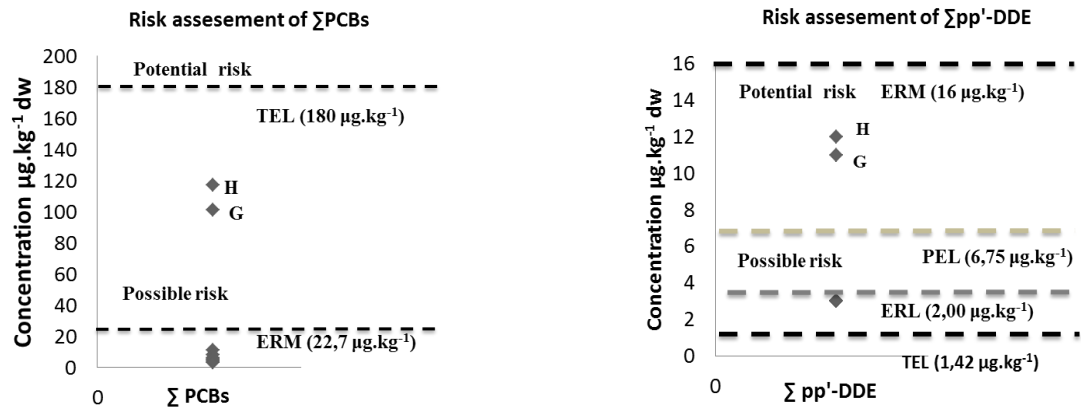


Fig 5. Assessment risk of the Σ 16 PAHs ($\mu\text{g.kg}^{-1}$ sed. dw) in sediment.

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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)

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

Compounds	Quantification transition			Confirmation transition		
	Precursor ion (amu)	Product ion (amu)	Collision Energy (V)	Precursor ion (amu)	Product ion (amu)	Collision energy (V)
Window 1 (11,5 to 15,6 minute)						
Aldrin	298,0	→ 263,0	8	263,0	→ 191,0	30
Hexachlorobenzen	283,9	→ 248,8	25	283,9	→ 213,9	35
Heptachlore	271,1	→ 237,0	10			
PCB 28	256,0	→ 186,0	26	258,0	→ 186,0	26
PCB 52	289,9	→ 220,0	28	291,9	→ 222,0	28
Window 2 (15,6 to 25 minute)						
Chlordecone	271,7	→ 236,8	10	236,8	→ 143,0	15
Dieldrin	262,8	→ 193,0	30	262,8	→ 191,0	15
Endosulfan 1	240,9	→ 205,9	10	240,9	→ 169,9	20
Heptachlor epoxy	352,7	→ 262,9	20	352,7	→ 316,9	20
ppDDD	237,0	→ 165,0	20	235,0	→ 199,1	8
pp'DDD d4	242,9	→ 173,1	20			
pp'DDE	248,0	→ 176,0	30	246,0	→ 211,0	20
pp'DDT	237,0	→ 165,0	20	235,0	→ 199,1	20
pp'DDE d4	253,9	→ 184,2	30			
PCB 101	323,9	→ 253,9	28	325,9	→ 255,9	28
PCB 101 d3	328,7	→ 258,9	20			
PCB 118	323,9	→ 253,9	28	325,9	→ 255,9	28
PCB 138 and 153	357,0	→ 287,9	28	359,8	→ 289,9	28
PCB 156 d3	363,0	→ 293,0	20			
PCB 180	393,8	→ 323,9	30	395,8	→ 325,9	30

1000 **Table 6.** SIM (selected ion monitoring) mode for the PAHs

Target compounds (standard marquer)	Qualification m/z (amu)	Quantification m/z (amu) (standard marquer)
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 / Pyrene (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 / Benzo(g,h,i)pérylene (d_{12}/d_{12})	276/ 274	276 (288)
Dibenzo(a,h)anthracene (d_{14})	274	278 (292)

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