Squalen Bull. of Mar. and Fish. Postharvest and Biotech. 12 (3) 2017, 85-97

www.bbp4b.litbang.kkp.go.id/squalen-bulletin

Squalen Bulletin of Marine and Fisheries Postharvest and Biotechnology

ISSN: 2089-5690
e-ISSN: 2406-9272

POTENTIAL RISK OF ORGANIC CONTAMINANTS TO THE COASTAL POPULATION THROUGH SEAFOOD CONSUMPTION FROM JAKARTA BAY

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Article history:
Received: 10 September 2017; Revised: 27 November 2017; Accepted: 3 December 2017

Abstract

A comprehensive study on exposure assessment of the priority organic contaminants via seafood consumption has been conducted to the coastal population of Jakarta Bay. Seafood is essential food source in Indonesia and also important income for the majority of coastal populations. A number of 152 respondents from 4 districts surounding the bay were interviewed to record their frequency and pattern on seafood consumption. In the same time, 13 seafood species were collected directly from Jakarta Bay during the dry and wet seasons for the assessment of organic contaminants. A non-target GC/MS screening identified more than 40 organic contaminants in which 6 of them are potentially considered as priority contaminants including 3 groups of carcinogenic contaminants i.e. dichlorodiphenyl-trichlorethane (DDT) and its metabolites (DDXs), dichlorobenzenes (DCB) and carcinogenic PAHs (PAH_4). Further exposure analysis suggested cumulative health risk of these contaminants was less than official minimal risk level (MRL) and therefore categorized safe for the corresponding population. However, attention must be paid since additional exposure of either from the different food category or other exposure route may contribute to significantly elevate the health risk on the population as well as potential exposure of emerging contaminants.

Keywords: exposure assessment, organic contaminant, coastal population, seafood, Jakarta Bay

1. Introduction

Organic contaminant in marine water is a threat to aquatic environment, with acute and chronic toxicity effect to aquatic organism, as well as harmful to human health (Jiang, Lee, & Fang, 2014). Jakarta Bay produces significantly seafood for local community in the regions of Jakarta, Bogor, Depok, Tangerang and Bekasi or better known as megapolitan Jabodetabek. More than 200,000 Tons of seafood is landed annually in Jakarta Fishing Port (BPS, 2014). Out of them, approximately 36,300 Tons are contributed from Jakarta Bay, composed of 35,000 Ton from capture fishery, and 2,300 Tons from marine and coastal culture. Nevertheless, the bay receives high load of contaminants mainly from the terrestrial discharges, either domestic waste, industrial disposal or agricultural outflow (Nur, Fazi, Wirjoatmodjo, & Han, 2001; Rinawati et al., 2012). The wastes are then delivered into Jakarta Bay through 13 main rivers across the regions (Dzikowitsky et al., 2014). The load of contaminants potentially elevates in line the rapid grow of the megapolitan region.

A number of contaminants related to environment and seafood from Jakarta Bay have been investigated previously. They include inorganic contaminants or heavy metals (Rumengan et al., 2008; Williams, Rees,

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DOI: http://dx.doi.org/10.15578/squalen.v12i3.289.

& Setiapermana, 2000) and organic contaminants such as polycyclic aromatic hydrocarbons (PAHs), 2,2-bis(chlorophenyl)-1,1,1-trichlorethane and its metabolites (DDXs), polychlorinated biphenyls (PCBs), tributyltin (TBT), polybrominateddiphenyl ethers (PBDEs) and hexachlorocyclohexanes (HCHs), (Monirith et al., 2003; Sudaryanto et al., 2007). Additionally, assessment on the exposure of harmful contaminants has become global concern due to their potential adverse effect to the environment, including health risk to human being (Kiljunen et al., 2007).

Accumulation of harmful contaminants in seafood species over the maximum residue limit leads to toxicological effect via human exposure route (Ding, Ni, & Zeng, 2013; FAO/WHO, 2006). Persistent organic pollutants (POPs), such as organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polychlorinated dibenzodioxin (PCDD), polychlorinated dibenzofuran (PCDF) and carcinogenic PAHs, have been proved to harmful wildlife and humans, due to their carcinogenic, mutagenic, endocrine disruption, or immunotoxic properties (UNEP, 2001). Many countries, including Indonesia, have derived their national threshold on maximum contaminant limit (ML) and tolerable daily intake (TDI) or allowable daily intake (ADI) of certain contaminants. FAO and WHO (2000),for example, define ML of DDT and its metabolites in meat, poultry and fish as 7 ppm (per fat) with ADI of 0.01 mg/kgBW/day, which is adopted in Indonesian as national standard No. SNI 01-6366-2000 (BSN, 2000). Nevertheless, exposure assessment on chemical contaminants via foodstuff consumption from Indonesia is very rare. These limited studies include exposure assessment of heavy metals in seafood (Agusa, Kunito, & Sudaryanto, 2007), organochlorines and PBDE in human breast milk (Sudaryanto et al., 2008), and organochlorine in selected food products (Shoiful, Fujita, Watanabe, & Honda, 2013).

The present study was aimed to examine the possible exposure of various organic contaminants via seafood diet from Jakarta Bay. The dietary exposure assessment combines data of consumption pattern of selected population in the northern area of Jakarta city with data of contaminant concentration in different seafood groups and species from Jakarta Bay, including pelagic and benthic species, as well as molluscs and crustaceans. Food frequency survey was applied to estimate the seafood consumption in selected population. Further on, the result would reveal the level of human health risk in the study area associated with corresponding contaminants.

2. Material and Methods

2.1. Survey on Food Consumption Frequency

In order to assess exposure of organic contaminants in the study area, a number of 152 respondents were selected based on stratified random and purposive sampling in 4 districts directly around Jakarta Bay i.e. Cilincing, Penjaringan, Teluk Naga and Untung Jawa (Figure 1). Dietary pattern of the respondents was then assessed through a direct interview to record their consumption history of the last 7 days using food consumption frequency survey/ FFQ (FAO/WHO, 2006). The number of respondent was calculated by Eq. (1) based on Cochran (1977) formula:

$$N = \frac{Z_{\alpha/2}^{2} p.q}{E^{2}} \quad (1)$$

where N is the sample size; $Z_{\alpha/2}$ is the abscissa of the normal curve that cuts off an area a at the tails (1-a equals the desired confidence level was 95%). A number of 150 respondents were calculated based on the desired precision level (E) of 8%, which is in the range of acceptable level (5-10%) as mentioned by Bartlett, Kotrlik, and Chadwick (2001). Furthermore, p is the estimated proportion of an attribute that is present in the population (0.5), and q is 1-p. Respondents were then selected based on the proportional gender and represent two age groups, i.e. 15-39 years old as younger adult and 40-70 years old for older adult (Norimah et al., 2008).

2.2. Determination of Contaminants

Gas chromatography-mass spectrophotometry (GC/MS) was employed to determine organic contaminant concentration in selected seafood of economically important species collected directly from Jakarta Bay. The seafood samples included 3 pelagic fishes, 6 benthic species, 2 crustaceans (banana shrimp and blue swim crab) and green mussel (*Perna viridis*) from different sites as illustrated in Figure 1. At least 10 individual fish and shrimp species were collected directly from the local fisherman, while at least 50 individual of green mussel were harvested directly from each culture sites in Kamal, Kalibaru and Cilincing. The detail of sample extraction, fractionation as well as contaminant identification and quantification following the procedure explained elsewhere (Dwiyitno et al., 2016).

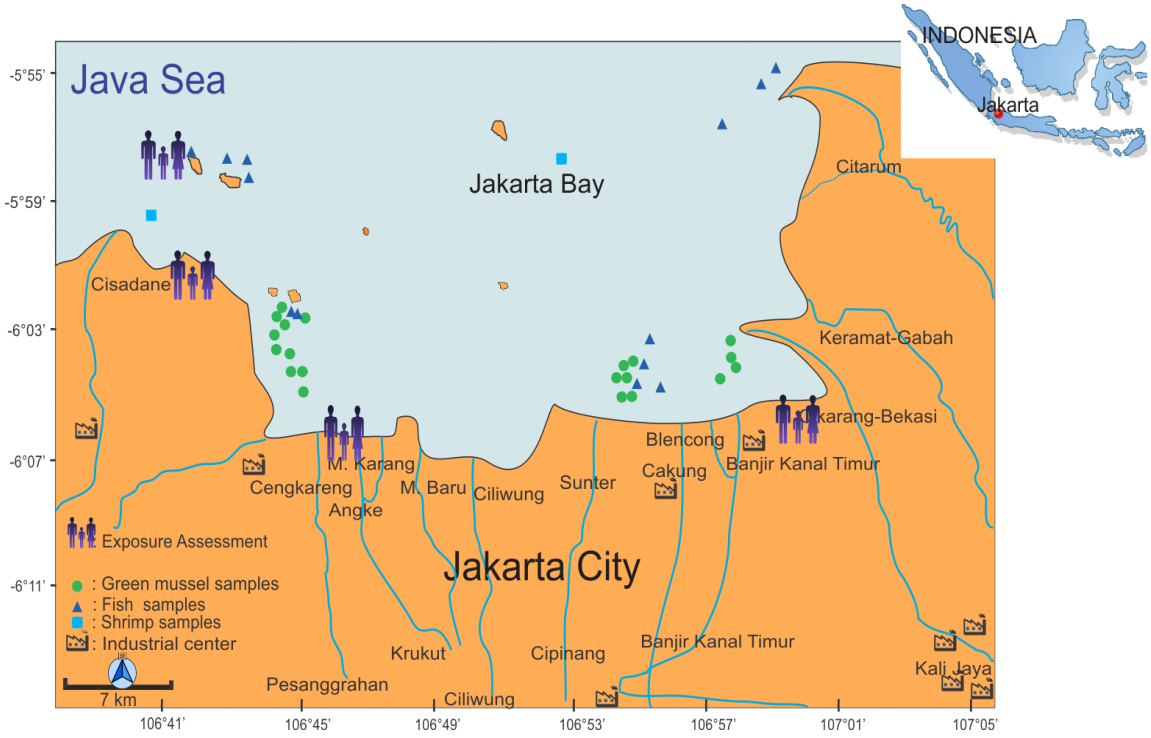

Figure 1. Sampling site of seafood samples and food consumption survey.

2.3. Extraction and Quantification of Organic Contaminants

A non-target screening approach using EC-*Exsposmeter* clean-up semipermeable membrane (*ExposMeter* sampling technologies, Sweden) was employed for extracting seafood species (Dwiyitno et al., 2016). After extracted with dichlorometane (DCM) for 72 hours, supernatant was concentrated and fractioned as performed by Franke et al. (2007). After separation of the phases, the organic extracts were added 50 µl of surrogate standard containing *d34*-hexadecane (6.00 ng/µl), *4*-fluoroacetophenone (7.17 ng/µl) and decafluorobenzophenone (6.96 ng/µl). One microliter of sample aliquot was injected in to a Trace GC-MS system (*ThermoQuest,* Germany) linked to HRGC-5160 Mega Series (Carlo Erba, Italy) equipped with a *ZB-XLB* silica capillary column (30 m x 0.25 mm ID, 0.25 µm film thickness; *Zebron-Phenomenex,* Germany). The oven temperature was held at 60 °C for 3 min then increased at 3 °C/min to 310 °C and held for 20 min with injection temperature of 270 °C. Mass spectrometer was operated in full-scan mode at a resolution of 1000 with a source temperature of 200 °C. Electron impact ionization mode (EI^+, 70 eV) was operated at a source temperature of 200 °C, scanning from 35 to 700 amu at a rate of 1 s/scan.

Individual compound was identified by comparison of the EI-Mass spectrum and specific ion fragment with those of mass spectral libraries (*NIST98* and *Wiley* 4th Ed.), as well as by integrating the gas chromatographic retention time or scan number those of authentic reference materials. Quantitation was performed by integrating ion chromatograms on selected m/z. Quantification of selected contaminants was calculated by *Xcalibur* based on the comparison of sample spectral and response factors of reference materials.

2.4. Fat Content

Total fat content was quantified based on gravimetric method using common *Soxhlet* method (AOAC, 2010). Fat was extracted through repeated washing/percolation with 150 mL petroleum ether for approximately 6 hours. After extraction, the fat extract collected in the extraction pot was oven dried to constant weight and weighed in order to measure the weight difference compared to the initial weight.

2.5. Data Processing and Analysis

Data of FFQ survey was tabulated and presented by descriptive statistic as mean or median $\pm$ S.D.; 95%-tile and minimum-maximum values. Non parametric test was employed to statistically differentiate among parameter using SPSS ver.16. Variables of data input included consumption frequency per week, seafood consumption per day, information on respondent's body weight and kind of

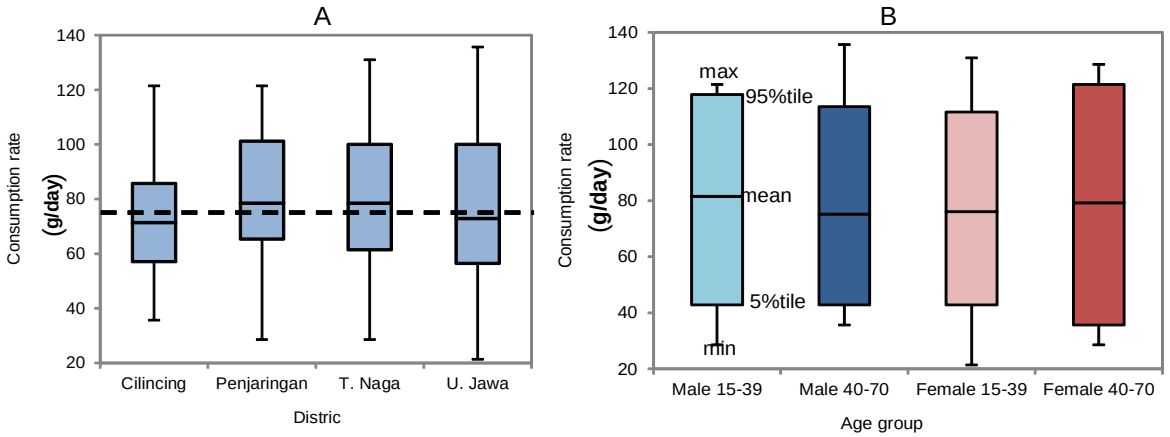

Figure 2. Profile of seafood consumption in the study area (A) and seafood consumption of the household in gender and age group (B).

seafood consumed. Estimated daily intake (EDI; µg/kg BW/day) of contaminant was calculated by multiplying the contaminant concentration in seafood with corresponding seafood consumption rate according to Eq. (2):

$$EDI_j = \sum_{i=1}^{n}(C_i .IR_{ij})/BW_j] \quad (2)$$

where C is concentration of contaminant in seafood species (ng/g wet weight or ww); IR is intake rate (g/day); BW is body weight of individual respondent (kg); *i* and *j* indicate seafood of the i^{th} and j^{th} sub-group, respectively.

Concentration of contaminant in seafood was calculated from the result of GC/MS quantification on selected species (Table 1). For other species that the samples were not represented, concentration was estimated from the similar species based on the similar habitat and feeding habit, assumed has the similar accumulation behavior. Concentration of each contaminant was then compared to that of officially maximum residue limit (ML) set by FAO/WHO or other official agencies. Risk characterization was calculated based on the potential risk (hazard index) on the Lifetime Average Daily Dose (LADD) by the Eq (3):

$$\text{Lifetime Average Daily Dose (µg/kg/day)} = \frac{(C \times IR \times EF \times ED)}{(BW \times AT)} \quad (3)$$

where C is concentration of contaminant in each seafood sample (µg/g); IR is intake rate (g/day); EF is exposure frequency (365 day/year); ED is exposure duration (70 year); BW is body weight (g); AT is exposure averag in time (day). Potential risk (hazard index) is determined for estimating risk characterization on human (FAO/WHO, 2011). Hazard index (potential risk) was calculated based on the LADD multiplying carcinogenic factor or slope factor (q*) of corresponding contaminant as in Eq. (4):

$$\text{Hazard index} = LADD \times q^* \quad (4)$$

where hazard index is level of carcinogenic risk; LADD is average daily dose; and q* is carcinogenic factor (mg/kg BW/day), e.g. for DDT is 3.4×10^{-1}. Hazard index (risk) over minimal risk level (MRL) indicates high risk of cancer with the possibility to injure one people per one million population.

3. Results and Discussion

3.1. Consumption Survey

With regard to food frequency survey, 42 species of seafood consumed by respondents in the study area were identified. They included 14 pelagic and 21 demersal fishes, 2 species of crustacean (banana shrimp and crab) and 2 groups of mollusk (squid and green mussel). In total, the average of seafood consumption in the study area was 77.7±24.2 g per person per day (g/capita/day) as presented in Figure 2 and 3. Population of Penjaringan and Teluk Naga Districts consumed seafood slightly higher than those in Untung Jawa and Cilincing (Figure 2A). Fish consumption in the study area was slightly lower than that of Jakarta metropolitan consumption data (84 g/person/day) or the national rate (90 g/cap/day), as reported by MMAF (2013). However, the consumption level in the present study was comparable to that reported by FAO (2012) which was 79.2 g/cap/day.

In Indonesia, fish consumption contributes to approximately 57% of total animal protein consumption (BPS, 2011). Comparison of seafood consumption based on gender and age group in the study area

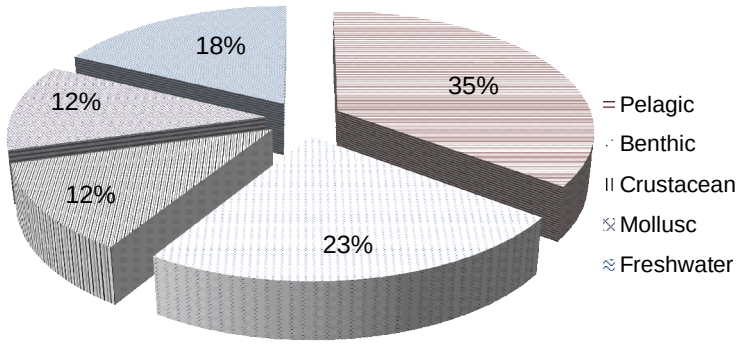

Figure 3. Contribution of seafood group on the contaminant exposure.

showed that both male and female respondents consumed seafood at relatively equal amount, likewise between respondents of different age group (Figure 2B). Concerning the frequency of seafood consumption per month, there was equal frequency between male and female respondents in the study area i.e. 19.17±2.17 and 15.96±2.73 days, respectively with minimum-maximum range of 8-20 days/month.

Figure 3 shows that fish species contributed significantly to the seafood consumption in the study area as compared to crustacean and mollusc. Among the fish group, pelagic species mainly mackerel, eastern little tuna, and milk fish were consumed predominantly by respondents, while relatively significant amount of benthic species were also consumed in the study area i.e. pomfret, snapper, and jack trevally. Likewise, green mussel, crab, and banana shrimp were consumed by respondent in the study area. Top seafood species consumed by respondents in the study area were performed in Figure 4. The relatively higher consumption rate of each seafood species by eater only respondent (Figure 4B), indicating that the concerning species was not consumed by all respondents. Figure 4 also explains that mackerel, little tuna and milkfish are relatively consumed by predominantly respondents. All of these species represent the major seafood species captured in Jakarta Bay. There was also minor contribution of fresh water species to the seafood consumption in the study area, but were assumed not exposed significantly to contamination in Jakarta Bay. The seafood consumption data collected in the present study represent the consumption of seafood originated from Jakarta Bay since the majority of respondents knew well that the seafood was captured in Jakarta Bay (65%), while 5% not from Jakarta Bay and the rest of 30% was not sure the origin of the seafood.

3.2. Concentration of Organic Contaminant

Non-target screening approach using GC/MS identified more than 40 organic contaminants, including persistent pollutants and emerging

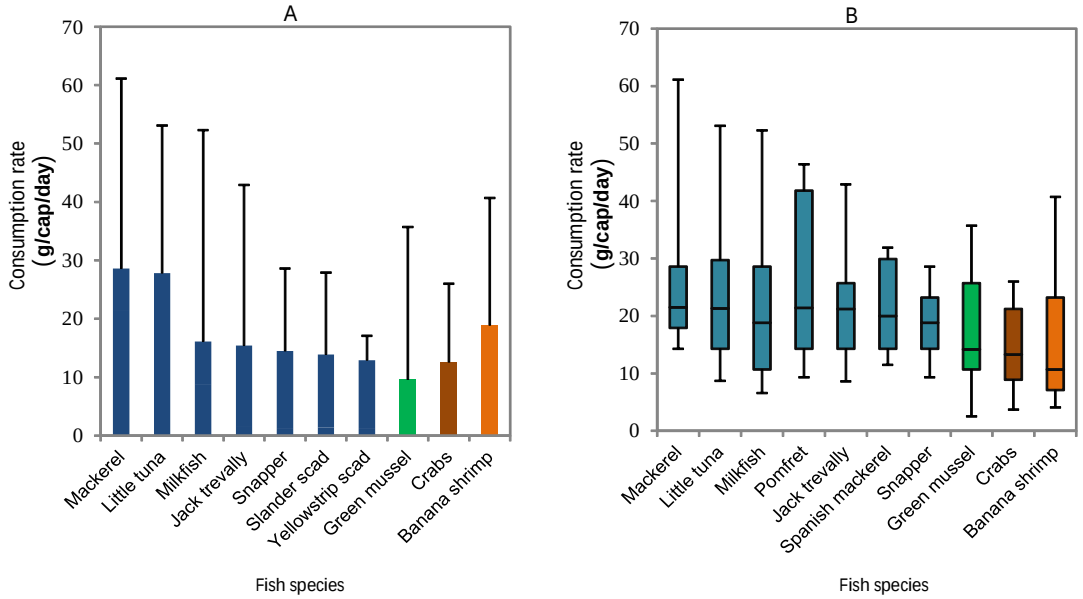

Figure 4. Top seafood consumption based on species (A) Consumption by total respondents; (B) Consumption by eater only.

Table 1. Concentration of selected organic contaminants in seafood samples from Jakarta Bay

No	Seafood Species	Organic Contaminant (µg/kg ww)							Fat (%ww)
		DDXs[a)]	DCB	PAH_{16}	PAH_4	DIPNs	LABs	PMN	
1	Green mussel (*Perna viridis*)	n.d.-7	n.d.-26	3-87	n.d.-7	10-231	n.d.-12,300	n.d.-75	2.6
2	Slender shad (*Lisha elongata*)	1	n.d.	10	n.d.	10	235	n.d.	3.6
3	Mackerel (*Rastrelliger kanagurta*)	n.d.	n.d.-5	4-7	n.d.	3-37	50-971	n.d.-27	6.3
4	Spanish mackerel (*Scomberomorus commerson*)	5	n.d.	24	1	3	5	9	6.7
5	Milkfish (*Chanos chanos*)	4	n.d.	10	3	22	26	12	2.7
6	Croaker (*Argyrosomus amoyensis*)	n.d.-1	n.d.	3-10	n.d.	2-14	n.d.-57	n.d.-6	1.3
7	White emperor (*Lethrinus lentjan*)	n.d.-1	n.d.	1-12	n.d.-1	3-4	n.d.-54	n.d.-11	1.3
8	Mahogany snapper (*Lutjanus johnii*)	n.d.-1	n.d.-5	n.d.-12	n.d.	3	n.d.-21	n.d.-7	2.7
9	Rabbitfish (*Siganus javus*)	n.d.-10	n.d.-5	8-12	n.d.	3-7	65-617	n.d.	1.1
10	Sea catfish (*Netuma thalassina*)	n.d.-1	n.d.-5	2-6	n.d.	14-143	45-194	n.d.-2	3.8
11	Mullet (*Mugil seheli*)	5	n.d.	8	2	21	22	n.d.	1
12	Banana shrimp (*Penaeus maerguiensis*)	n.d.-1	n.d.	n.d.-5	n.d.	3-13	8-12	n.d.-5	3.2
13	Blue crab (*Callinectes sapidus*)	1	7	15	1	2	10	13	3.2
	ML*[b)] *(µg/kg ww)	***500[c)]***	***75/600[d)] (in water)***		***12/ 30[e)]***	***n.a.***	***n.a.***	***n.a.***	

Note:

[a)] DDXs is sum of DDT, DDE and DDD; [b)]ML: maximum residue limit; [c)]FAO/WHO (2000);[d)] US-EPA (2009) for 1.4 and 1.2-DCB; [e)]EC No.835/2011(PAH_4: sum of BaP, BaA, Chr, BbF; 12µg/kg for fish and 30 µg/kg for mollusc); n.a.: not applicable; n.d.: not detected (LOQ:1.0 µg/kg ww)

contaminants in selected seafood species as reported elsewhere (Dwiyitno et al., 2015). In the present study, 6 priority contaminants were selected to generate dietary intake on the population around Jakarta Bay. The contaminant were selected due to their category as either persistent or emerging contaminants. They include DDXs, DCB, PAHs, DIPNs (di-iso-prophyl naphthalenes), LABs (linear alkyl benzenes) and PMN (phenyl methoxy naphthalene) as presented in Table 1. These compounds were selected as priority organic contaminants in Jakarta Bay due to their potential toxicity, persistent properties in the environment, widely distribution in various seafood species and relatively high concentration compared to those in similar ecological system (Wilson & Jones-Lepp, 2013).

Green mussel species in the study area predominantly accumulated organic contaminant than that of fish species and crustacean. This could be affected by their different feeding behavior and living habitat (Baumard et al., 1999). Known as filter-feeder species, green mussel from Jakarta Bay accumulate LABs, DIPNs, PAHs, and PMN predominantly than fish species (Dwiyitno et al., 2016). PAHs, LABs and PCBs contaminants in sediment of Jakarta Bay have also been reported earlier by Rinawati et al. (2012). Among fish species collected in the present study, milkfish, mackerel, spanish mackerel, and mullet

predominantly accumulate organic contaminant than other species. This is likely related to their relatively high fat content (Table 1). As reported previously, accumulation of lipophilic compound could be attributed to several factors including fat content, tropic level, feeding habit, and migration pattern (Stange & Klungsoyr, 1997; Ueno et al., 2003).

To date, no official regulation or threshold is established for DIPNs, PMN, and LABs. DIPNs is widely used to replace PCBs and mainly applied in carbonless paper production (Suzuki, Matsumura, Moriguchi, & Nakano, 2007). Earlier studies showed that emission of PMN is related to textile manufacturer and paper industry (Terasaki, Jozuka, & Makino, 2012; Brigden, Labunska, House, Santillo, & Johnston, 2012). Consequently, this may contribute to the dietary exposure of the contaminants to the consumer from green mussel intake. LABs is potential indicator for domestic discharges as a side product of detergent synthesis which is massively used in urban community (Takada & Eganhouse,1998).

In the present study, contaminant level of DDXs, DCB and PAHs in seafood samples was lower than that of official threshold or maximum level (ML). PAHs contaminant was presented as total of 16 priority PAHs, known as the most significant congeners based on the toxicity, information available and commonly reported worldwide (ATSDR, 2002; EC, 2006). Furthermore, total carcinogenic PAHs (PAH_4) was calculated based on the sum of benzo[a]pyrene (BaP), benz[a]anthracene (BaA), benzo[b]fluoranthene (BbF) and chrysene (Chr), which are known as suitable indicator for PAH contaminant in food (EC, 2011). It was identified that maximum concentration of PAH_4 in green mussel was 7µg/kg and 3 µg/kg for fish species (milkfish) which were below the threshold of 30 and 12 µg/kg respectively (EC, 2011). PAH_2 (BaP and Chr) and PAH_8 (BaP, BaA, BbFl, BkFl, BghiP, Chr, DbahA, and IcP) are common PAHs indicators for foods, though PAH_4 is more acceptable (EFSA, 2008).

Different accumulation pattern appeared on organochlorine compounds between mussel and fish or crustacean. DDT metabolites were predominantly accumulated in fish species rather than in green mussel. In contrast, green mussel accumulated more DCB than fish species. This fact indicates DCB is less persistent or more biodegradable than DDT due to relatively low of log K_{ow} (3.43-3.63) with higher uptake from dissolve water or the lower clearance rate in mussel than in fish species as reported by Bute Fox, and Zauke (1991).

Since the earlier studies showed highly genotoxic and possible carcinogenic to human, the use of DDT and its metabolites (including DDT, DDE and DDD) has been banned in many countries since 1970s (ATSDR, 2002). Further on, DDT is categorized as persistent organic pollutants (POPs) according to Stockholm Convention in 2001 (UNEP, 2001). Technical DDT generally contains 65-80 % *p,p'*-DDT, 15-21 % *o,p'*-DDT and up to 4 % *p,p'*-DDD (ATSDR, 2002). Accumulation of DDXs in seafood species from Jakarta Bay in the present study was comparable to that reported earlier from the same location (Monirith et al., 2003; Sudaryanto et al., 2007).

DCB was primarily produced as mothballs and deodorant blocks. Recently, other applications also use DCB to produce herbicides, fumigant, insulating material and dielectric properties (ATSDR, 2006). Practically, major exposure route of DCB are through inhalation, oral and dermal routes. Since minor oral exposure of DCB is likely occurred, no official guideline of maximum residue limit for DCB per food product is established and therefore the DCB contamination in seafood in the present study could not be compared to any official MRL. So far, US EPA (2009) recommends a maximum contaminant level (ML) of 1,4-DCB and 1,2-DCB in drinking water as 75 µg/L and 600 µg/L respectively. Further toxicological evaluation on animal concluded that *para*-DCB causes a high evidence of liver tumors and therefore classified as carcinogenic to human (IARC, 1987).

With regard to PAHs contaminant, naphthalene (Na), phenanthrene (Ph) and fluoranthene (Fl) were detected in all green mussel samples, but fluoranthene was not detected in some fish species (Dwiyitno et al., 2015). This indicates a different feeding behavior between green mussel and fish and crustacean (Dwiyitno et al., 2016). This fact was also supported by the absence of high molecular weight (HMW) PAHs such as BaP, benzo[*e*]pyrene (BeP), BaA, Chr, B*b*F, benzo[*j*]fluoranthene(BjF), benzo[*k*]fluoranthene (BkF), dibenzo[*a,h*]antracene (DahA), indeno[1,2,3-*cd*]pyrene (InP), and benzo[*ghi*]perylene (BghiP) in fish and crustaceans, in contrast with green mussel. HMW group of PAHs containing four fused rings or more are regarded as potentially carcinogenic and genotoxic compounds where BaP is the most carcinogenic congener and commonly used as indicator for PAH contaminations (FAO/WHO, 1991; EC, 2011). Earlier study showed that PAH_4 was also detected in green mussel sample from the same location (Isobe et al., 2007).

Occurrence of carcinogenic PAHs, including BaP, was detected particularly in fish samples suggesting that PAH or BaP are less metabolized in certain fish species such as spanish mackerel, milkfish, white emperor and mullet or more accumulation due to the relatively high concentration. This fact indicates the

Table 2. Comparison of organic contaminant exposure via seafood consumption to TDI

Contaminant	Dietary intake (ng/kg BW/d)		Daily intake (ng/day)	TDI (ng/kg BW)	Mean Intake vs TDI (%)	95% tile Intake vs TDI (%)
	Mean (Min-Max)	95% tile				
DDXs	0.04 (0.003-0.2)	0.1	2	500 [a)] $1x10^4$ [b)]	0.008 0.0004	0.02 0.001
DCB	0.05 (0.001-1.6)	0.1	2	$1x10^5$ [a)] $1.07x10^5$ [c)]	0.00005	0.01
PAH_{16}	0.02 (0.03-0.6)	0.32	8	$1x10^5$ [d)]	0.00002	0.01
PAH_4	0.02 (0.005-0.08)	0.04	0.8	$1x10^4$ [d)]	0.0002	0.002
DIPNs	0.2 (0.01-1.3)	0.51	*11*	*n.a.*	*n.a.*	*n.a.*
LABs	6.1 (0.08-74.0)	20	*339*	*n.a.*	*n.a.*	*n.a.*
PMN	0.2 (0.02-2.0)	0.45	*10*	*n.a.*	*n.a.*	*n.a.*

Note:
[a)] RIVM (2001); [b)] FAO/WHO (2000); [c)]WHO (2003);[d)]JECFA (2006);n.a.: no available data

important of assessing PAHs contaminant in fresh fish, which is in contrast with earlier opinion by EC (2011) stated that PAHs contaminants is predominantly in processed products. Relatively low PAHs accumulation in fish species compared to that of green mussel suggests a more efficient PAHs detoxification or elimination in fish species as also reported by D'adamo, Pelosi, Trotta, and Sansone, (1997) and Baumard et al. (1998). Total PAHs and carcinogenic PAHs concentration in fish species in the present study was slightly lower than that of various fish species from Mumbai India (Dhananjaya & Muralidharan, 2012; Nwaichi & Ntorgbo, 2016).

3.3. Dietary Exposure of the Priority Contaminants

As presented in Table 1, 3 groups of potentially carcinogenic contaminants were accumulated in fish and shellfish from Jakarta Bay and therefore local consumer are potentially exposed to those contaminants, i.e. DDXs, PAHs and DCB. Toxicological studies of these contaminants indicated human healt risk related to different types of cancer and other adverse outcomes (ATSDR, 2002; BfR, 2010; FSA, 2015).

Table 2 shows the dietary intake of priority organic contaminants via seafood consumption from Jakarta Bay. As reflected from the contribution ratio, the predominant organic contaminants related to seafood samples in Jakarta Bay were LABs, DIPNs and PMN. Since toxicological studies of these compounds are very limited, these contaminants were consequently not determined for the risk analysis. However, this group could be suggested as emerging contaminants and possible indicator of anthropogenic emission in the study area. It has to be concerned also that DIPNs and PMN may toxic or cause long-lasting harmful effects to aquatic life (Brigden et al., 2012; Scarlet et al., 2011; Terasaki et al., 2012).

In comparison to official guideline, dietary exposure of DDXs, DCB and PAH_4 in the present study indicates lower than pertinent threshold (TDI). FAO/WHO (2000) recommends a 10 µg/kg BW as maximum limit of daily intake for DDXs. However, different countries have set their own limits, possibly due to different toxicological test (e.g. NOAEL, LOAEL), sensitive variation among population group and the lack of data on human experiment. In the present study, DDT contaminant in green mussel and the majority of fish samples were contributed mainly by *p,p'*-DDE metabolite. This result is in line with the earlier studies that reported *p,p'*-DDE in fish sample from several location in Indonesia (Shoiful et al., 2013; Sudaryanto et al., 2007). DDE is dehydrochlorinated metabolite of DDT in aerobic environment catalyzed by glutatione-*S*-transferase. DDE is known as the most toxic and primarily DDT metabolite, supported by relatively high

K_{ow} and persistency in the environment compared to other DDT metabolites. The K_{ow} of *p,p'*-DDE, *p,p'*-DDT and *p,p'*-DDD are 7.0; 6.9 and 6.2 respectively (de Bruijn, Busser, Seinen, & Hermens,1989) with half-life (T_{50}) of 250, 28 and 24 days in *Columbia livia* respectively. The *p,p'*-DDE was also detected as the most abundant DDT congener in river sediment sample from the eastern part of Jakarta Bay (Munawir, 2005). Similarly, studies in the human revealed *p,p'*-DDE was predominantly detected in the blood plasma compared to any other DDT congeners (Waliszewski, Aguirre, Infanzon, & Siliceo, 2002; Zamir et al., 2009). Animal study in mice demonstrated that DDT, DDE and DDD could pose cancer (primarily in the liver), may alter reproduction and development of hormones or affect calcium metabolism (ATSDR, 2002).

According to WHO (2003), $1.07x10^5$ ng/kg BW/day is suggested as TDI of 1,4-DCB based on LOAEL test of 150 mg/kg BW, while TDI for *ortho-para* DCB is 429 µg/kg BW/day. Additionally, RIVM (2001) adopted this level to define TDI level of *1,4-* and *1,2-*DCB as $1.0x10^5$ ng/kg BW/day and $4.3x10^5$ ng/kg BW/day respectively. With refer to this value, DCB intake via seafood consumption from Jakarta Bay was below TDI as presented in Table 2. The different TDI level between *ortho-para* and *para-para* DCB attributed to their toxicological properties since *p,p'*- DCB is categorized as possible carcinogenic compound (2B), but not for *o,p'*- DCB (ATSDR, 2006).

Dietary intake of PAH_4 was estimated based on the sum of BaP, BaA, BbF and Chr, presented as toxic equivalence quotient or *TEQ*-B[a]P. Since individual PAH exhibits different potency of carcinogenic activity, a relative carcinogenic potency of selected PAHs to BaP has been established (FSA, 2015). Concerning PAH_4, the carcinogenic factors of BaA, BbF and Chr are 0.1; 0.1 and 0.001 relative to BaP respectively. In the present study, exposure of PAH_4 was predominantly contributed by BaA and BbF of green mussel as well as certain pelagic and benthic fish intakes such as milk fish, Spanish mackerel and mullet. Although InP and DbA appeared in certain samples as carcinogenic PAHs, their contribution seems not significant as also explained earlier (FSA, 2015). Dietary intake of PAHs in the present study was comparable to that of various fish species in Mumbai India (1.8-10.7 ng/kg BW/day), Korea (13.8-16.7 ng/kg BW/day), Kuwait (231 ng/day) and Spain (627-712 ng/day) as reported previously (Dhananjaya & Muralidharan, 2012; Falco et al., 2005; Saeed, Al-Yakoob, Al-Hashash, & Al-Bahloul, 1995).

3.4. Potential Health Risk

Persistent pollutants such as organochlorines may accumulate in human body for long term period and any health effect could arise thereafter. In order to determine the possible long term risk of such contaminant to individual health risk, potential risk could be estimated by comparing hazard index (see Eq. 4) and official MRL of corresponding contaminant. Generally, LADD was calculated based on 70 year human exposure and the availability information of individual exposure (US EPA, 2005). In the present study, long term exposure was generated from the real information of estimated individual exposure of

Table 3. Potential risk (ng/kg BW/day) and risk level of carcinogenic contaminant DDXs, DCB and PAH_4

Contaminant		Male Respondent		Female Respondent		MRL*)
		15-39 yr	40-70 yr	15-39 yr	40-70 yr	(ng/kg BW/d)
DDXs	Mean	0.012	0.0075	0.0068	0.0044	10 a)
	95%-tile	0.023	0.023	0.015	0.0089	
	RL (%)*	***2.3^A***	***2.3^A***	***1.5^{AB}***	***0.9^B***	
DCB	Mean	0.022	0.032	0.016	0.016	7.000 b)
	95%-tile	0.061	0.076	0.04	0.063	
	RL (%)*	***0.0006^A***	***0.00076^A***	***0.0004^A***	***0.0004^{4A}***	
PAH_4	Mean	0.0072	0.0044	0.0052	0.0042	10 c)
	95%-tile	0.016	0.01	0.016	0.01	
	RL (%)*	***0.2^A***	***0.1^A***	***0.2^A***	***0.1^A***	

Note:

a)RIVM (2001); b)ATSDR (2006); c) US-EPA (2001); *) MRL: minimal risk level;

**) RL: risk level; the same capital letters in the same row notify non- statistically different means ($p<0.05$)

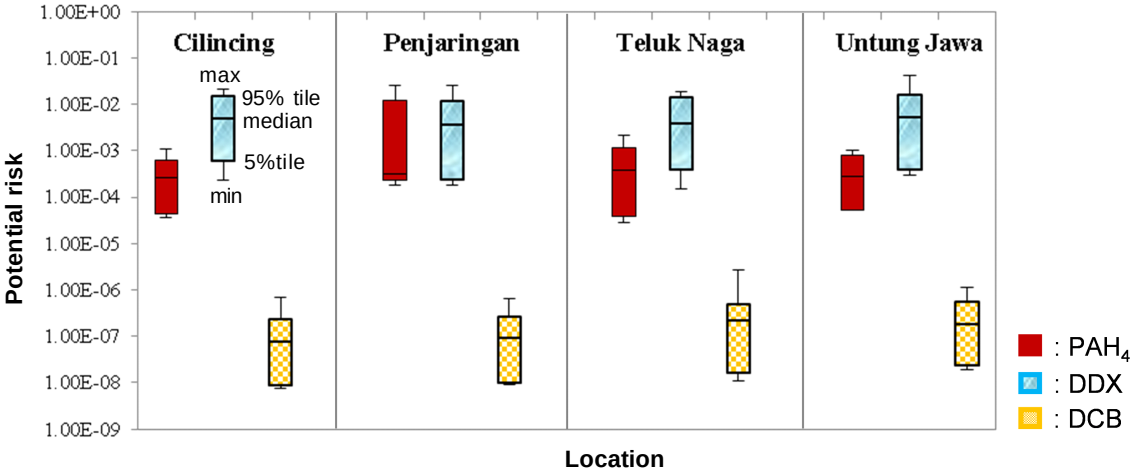

Figure 5. Potential risk level of each contaminant on the long term exposure in the study area.

carcinogenic contaminants DDT, DCB and PAH_4. Exposure factors (slope factors) of 0.34 and 0.73 were used to estimate potential risk of DDT and PAH_4 respectively (RIVM, 2001). Further on, MRL established by international agencies (e.g. RIVM, US EPA, ATSDR) were used to estimate average risk of individual contaminant. A level of 10^{-5} was selected as moderate MRL for DDT and PAH_4 contaminant as suggested by RIVM (2001) and US-EPA (2005) respectively, while MRL for DCB was $7x10^{-2}$ (ATSDR, 2006). This level corresponds to a cancer risk of one in 100,000 populations.

As presented in Table 3, average of total health risk from the ingestion of DDT, DCB and PAH_4 in the study area was well below the threshold of moderately and serious risk limit suggested by RIVM (2001), ATSDR (2002; 2006) as indicated by the risk level (RL) below 1 or 100 %. There was significant difference ($p<0.05$) between female respondent of 40-70 year old to the other population group on the total risk of organic contaminant. This difference is likely affected by the variation on exposure period of this population group that lead to be exposed by the contaminants shorter than the younger group (Ding et al., 2013). In the current study, respondent group of 15-39 year old would be exposed to the contaminants for approximately. 61.33 years while for the 40-70 year group was 48.18 years. The relatively higher seafood consumption of male 15-39 year old population (Figure 1) may also contribute to the higher contaminant exposure and consequently posed the higher risk to this population group. DDXs demonstrated as the predominant contributor on the total health risk of organic contaminants which support the earlier study that seafood is the main contributor of organochlorine exposure via oral in Indonesia (Shoiful et al., 2013).

In general, DDXs contributed predominantly in the exposure of organic contaminants in the study area, except in Penjaringan by which PAH_4 was also contributed equally (Figure 5). Compared to other studies, the average lifetime expose of cancer risk induced mainly by carcinogenic DDXs and PAH_4 via seafood consumption in Jakarta Bay is higher to that from fish consumption in Mumbai India ($2.37x10^{-7}$–$1.43x10^{-6}$) conducted by Dhananjaya and Muralidharan (2012). With regard to exposure of concerned organic contaminant in the population by ingestion of seafood from Jakarta Bay, attention should be paid to the additional contribution of harmful contaminants either from other compounds, different exposure route or contribution of other food categories.

Besides contaminated by organic contaminants, food products, including fish, are also potentially threatened by either inorganic or organo-metallic contaminants such as various heavy metals,tributyltin and PCBs (Skibniewska, 2003; Overmire et al., 2009; Copat et al., 2013). Earlier studies reported that exposure of heavy metals become concern as potential contaminant for aquatic biota in Jakarta Bay (Agusa et al., 2007) and thereby must be taken into concern by the authority.

4. Conclusions

The present study known as the first report to the exposure of different organic contaminants via seafood intake from South East Asian region. Three groups of priority organic contaminants associated with health risk to the population from consumption of seafood from Jakarta Bay were DDXs, DCB and PAHs. With refer to the seafood diet profile, DDXs and PAH_4 predominantly expose the population in the study area.

In general the daily intake of these contaminants was still below the TDI limit, revealed maximum risk level 0.02 % of TDI. Based on the 95th percentile lifetime cancer risk, male respondentshowed the higher risk of long term exposure of DDXs contaminant (2.3%) compared to female group (0.9-1.5 %). In order to prevent 50 % chronic risk of cancer, this population is limited to consume seafood from Jakarta Bay not exceed than 1,700 g/day, which is relatively high compare to their diet rate. However, possible exposure of other emerging contaminants such as DIPNs, PMN, and LABs has to be concerned. Since the greater exposure of harmful contaminants may be occured than this estimation, further information from the other possible contaminant sources, non-food pathway or other food category, is valuable ini conducting a comphrehensive risk assessment.

5. Acknowledgement

This study was supported by the German Federal Ministry of Education and Research (BMBF, Grant No.03F0641E) under the Indonesian-Germany SPICE-III project and the Agency for Marine and Fisheries Research, Indonesia. The authors acknowledge the German Academic Exchange Service (DAAD) for the financial support.

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