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Emerging Pollutants from Industrial Emission and Their Health Hazards in Indonesian Seafood: A Review

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Abstract

Emerging pollutants (EPs) or contaminants of emerging concern (CECs) have become global awareness since few decades ago, including in Indonesia. Intensive usage of industrial compounds has led to the massive emission to the environment and therefore their potential adverse effects may endanger aquatic organisms and human health. Based on the available literatures, organotins and flame retardants are two main groups of EPs from industrial emission identified in Indonesian seafood, as well as in those worldwide. However, concentration of both EPs group detected in Indonesian seafood is relatively low than that in developed countries and the majority of South East Asian countries. Toxicological studies revealed that EPs emitted from industrial activities have to be concern, as most of the EPs attributed to endocrine disrupting chemicals. Nevertheless, study on the exposure assessment of EPs in Indonesia is very limited. Even though, the concentrations of EPs in Indonesian seafood produce exposure below tolerable daily intake (TDI) to the local consumer, the long term exposures have to be aware due to their possible continues emission and elevating concentration such as shown by flame retardants. Governmental regulation, monitoring programs, and laboratory approach are among issues to be concern in addressing EPs in Indonesia, especially from industrial emission.

Keywords: emerging pollutants, industrial emission, organotins, flame retardants, aryl hydrocarbons

List of Abbreviations:
BCIPHIPP: Bis(1-chloro-2-propyl)-1-hydroxy-2-propyl phosphate; BDE: Bromo diphenyl ethane; BFRs: Brominated flame retardants; BHA: butylated hydroxyl anisole; BHT: Buylated hydroxyl toluene; BMDL: Benchmark dose confidence limit; BTBPE: Bis-tri-bromo phenoxy ethane; BTs: Butyltins; CECs: Contaminants of emerging concern; CRM: Certified reference material; DBDPE: Decabromo diphenyl ethane; DBT: Dibutyltin; DDT: Dichloro diphenyl trichloroethane; DIPNs: Di-isoprophyl naphthalenes; DOT: Dioctyltin; DPHP: Diphenyl phosphate; EPs: Emerging pollutants; HBCD: Hexabromocyclododecane; IMO: International Maritime Organization; LOEL: Lowest observed effect level; MBT: Monobutyltin; MOE: Margin of exposure; MRL: Maximum residue limit; NOEC: No observed effect concentration; OTs: Organotins; PBDE: Polybrominated diphenyl ether; PCBs: Polychlorinated biphenyls; PFRs: Organophosphate flame retardants; PMN: Phenylmethoxy naphthalene; PnTs: Phenyltins; POPs: Persistence organic pollutants; PPCPs: Pharmaceutical and personal care products; TARL: Tolerable average residue level; TBBPA: Tetrabromobisphenyl A; TBT: Tributyltin; TDI: Tolerable daily intake; TOC:Total organic carbon; TPhT: Triphenyltin; TXIB: Trimethyl pentanediol diisobutirate

Introduction

Emerging pollutants (EPs) or contaminants of emerging concern (CECs) have become global awareness since few decades ago. The concern was highlighted by a 1962's book of *Silent Spring* that criticizes the impact of widespread usage of dichlorodiphenyltrichloroethane (DDT) for eradicating mosquitoes and other pests on bird's species (Carson, 2002). Due to their toxicological harmful to the environment and human health, DDT was finally banned in 1970s followed by other chlorinated pesticides, later on known as persistence organic pollutants (POPs). Through Stockholm Convention, those chemicals are currently replaced by more environmentally friendly compounds such organophosphate and carbamate pesticides (UNEP, 2014).

United States Environmental Protection Agency/ US-EPA (2019) defined CECs as naturally occurring, manufactured or man-made chemicals or materials which have now been discovered or are suspected present in various environmental compartments and whose toxicity or persistence are likely to significantly alter the metabolism of any living organism. Similarly, European Commission uses EPs to define substances that have the potential to enter the environment and causes adverse ecological and human health effects, but are still largely unregulated and whose fate and potential effects are poorly understood (EC, 2018).

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However, scientists pointed different categories of emerging contaminants, i.e. contaminants which are very recently produced and contaminants which have been established in the environment but much concern is only recently (Cantwell & Katz, 2017; Sauvé & Desrosiers, 2014). EPs consists of wide range anthropogenic contaminants, including pesticides, cosmetics, pharmaceuticals, and personal care products (PPCPs), and other compounds which are associated with modern daily use (Thomaidis, Asimakopoulos, & Bletsou, 2012).

Some Indonesian waters have been known to receive high pollution exposure derived either from up-land regions or direct from coastal activities. Those contaminants are delivered from various sources, such as domestic disposal, industrial waste, transportation, oil spills, etc. (Takarina & Adiwibowo, 2010; Arifin, Puspitasari, & Miyazaki, 2012; Dwiyitno et al, 2015 & 2016; Dzikowitsky et al, 2016). Terrestrial pollution load has been shown in Jakarta Bay, as 39% of river water from Jakarta City was categorized as highly polluted and 44% was moderately polluted (BPLHD Jakarta, 2018). As the main river discard to Jakarta Bay, 54% of Citarum water ini West Java was also highly polluted (BPLHD Jakarta, 2018). Hence, the burden of the contaminants potentially increases significantly in accordance with the population growth and the development of industrial activities.

Studies on POPs and other priority contaminants from Indonesian waters have been reported. Nevertheless, studies on the EPs such as emission of pharmaceutical and PPCPs, pesticides, hormones, biological metabolites, perfluoroalkylated surfactants, nanoparticles, and other technical or industrial products in Indonesia waters are very limited (Dsikowitzky et al., 2014; 2016; Dwiyitno et al., 2016; Suzuki, Matsumura, Moriguchi, & Nakano, 2007; Syakti et al., 2013). Through bio-magnification pathway via aquatic food web, the contaminants potentially accumulate in the aquatic organisms including seafood. The present article would be the first comprehensive review concerning present studies on EPs, especially from industrial emission in Indonesian seafood and their potential impact to food safety issues compared to that of worldwide. Data of contaminant concentration in seafood from certain area in Indonesia are then used to estimate the exposure levels in concerned area.

Emerging Pollutants in Seafood

Studies on the occurrence of EPs in Indonesia was dominated from around Java Island waters, as the most populated and concentrated island in the country. Additionally, Jakarta Bay is the most investigated ecosystem for anthropogenic pollutants since is located in the most populated industrilized city of Indonesia, Jakarta. A number of studies on EPs have been conducted in Jakarta Bay, including EPs emitted from industrial activities in seafood species. Based on earlier literatures, industrial EPs could be classified as organotins (OTs), flame retardants, and aryl hydrocarbons (Table 1). Other locations become concern of EPs in seafood sample are Medan, Jambi, Lampung, Cirebon, Surabaya, and Makassar (Dwiyitno et al., 2016; Ilyas et al., 2013; Isobe et al., 2004; Isobe, Ogawa, Ramu, Sudaryanto, & Tanabe, 2012; Monirith et al., 2003; Nakata et al., 2012; Sudaryanto et al., 2007; Sudaryanto, 2002; Tsutsumi et al., 2002) as illustrated in Figure 1. Noteworthy, industrial contaminants associated with PAHs, pharmaceuticals, and pesticides are excluded from this review since they are commonly categorized in different pollutant groups such as domestic waste, pharmaceutical, and Personal Care Products (PPCPs).

Figure 1. Distribution of studies on EPs from industrial emission in Indonesian seafood (illustrated based on literatures presented in Table 1)

Table 1. Emerging pollutants from industrial emission in seafood from Indonesian water and worldwide

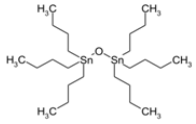
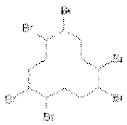
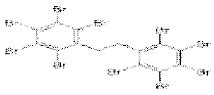
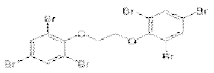
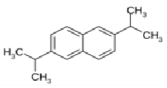
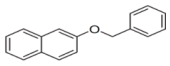
Group and Common use	Compound	Location	Species (year)	Concentration	Reference
1.Organotins Catalyst, stabilizer, preservatives, biocides	Butyltins /BTs 	- Belawan, Medan	Green mussel (1998)	5.0 ng/g ww	Sudaryanto (2002)
		- Kl Tungkal, Jambi		3.7 ng/g ww	
		- T. Harun, Lampung		9.9 ng/g ww	
		- T. Lada, Banten		6.8 ng/g ww	
		- Kamal, Jakarta		27 ng/g ww	
		- Ancol, Jakarta		58 ng/g ww	
		- Bondet, Cirebon		7.2 ng/g ww	
		- Genjeran, Surabaya		45 ng/g ww	
		- Maros, Makassar		6.1 ng/g ww	
		- Kamal, Jakarta	Fish (1998)	21-84 ng/g ww	
		- T. Lada, Banten		4.2-18 ng/g ww	
		- Bondet, Cirebon		3.3-25 ng/g ww	
		Jakarta Bay (TBT)	Green mussel (2003)	14 ng/g ww	Midorikawa et al. (2004)
		Malaysia	Green mussel (1998)	3.5-730 ng/g ww	Sudaryanto et al. (2002)
			Fish (1997-1998)	17-390 ng/g dw	Sudaryanto Takahashi, Iwata, Tanabe, Ismail (2004)
		Philippines	Green mussel (1998)	<1-787 ng/g ww	Prudente (2008)
			Fish (1998)	220 ng/g ww	
		Thailand	Green mussel (1998)	3-680 ng/g ww	Kan-atireklap, Sanguansi, Tabucanon, & Hungspreugs (1997)
		Vietnam	Green mussel (1998)	2.1-64 ng/g ww	Sudaryanto et al. (2002)
		Cambodia	Green mussel (1998)	2.4-88 ng/g ww	
		Hongkong	Green mussel (1999)	16-330 ng/g ww	
		India	Green mussel (1998)	<1-570 ng/g ww	
		Korea	Blue mussel (1997-1999)	17-1200 ng/g ww	Hong et al. (2002)
		Japan	Blue mussel (1989)	20-240 ng/g ww	Harino et al. (2000)
			Fish (1996)	11-182 ng/g ww	
		USA	Blue mussel (1989-1990)	2-276 ng/g ww	Short & Sharp (1989)
		Taiwan	Fish (2006)	Nd-953.7 ng/g ww	Chen et al. (2008)
2.Flame retardants Polymer additive of electronics, building materials and textile products	a. Polybrominated diphenyl ethers/ PBDEs 	Jakarta Bay	Green mussel (2007)	11±1.4 ng/g lw	Sudaryanto, Isobe & Tanabe (2012)
			Fish (2007)	9.1-23 ng/g lw	
		Lampung	Cultured Snapper (2004)	2.6-6.2 ng/g lw	Sudaryanto (2019)
			Wild Snapper (2004)	<1-7.3 ng/g lw	
		Benowo, Surabaya	Nile tilapia (2008)	6.6-11 ng/g dw	Ilyas et al. (2013)
		Jakarta Bay	Green mussel (2003)	17-34 ng/g lw	Isobe et al. (2012)
		Cambodia	Green mussel (2004)	2.3-5.3 ng/g lw	
		China	Green mussel (2004)	10-75 ng/g lw	
		Hongkong	Green mussel (2004)	25-130 ng/g lw	
		India	Green mussel (2004)	56-640 ng/g lw	
		Japan	Gren mussel (2003-2008)	18 ng/g lw	
			Blue mussel (2003-2008)	49-89 ng/g lw	
		Korea	Blue mussel (2005)	6.7-440 ng/g lw	
		Malaysia	Green mussel (2004)	0.5-10 ng/g lw	
		Philippines	Green mussel (2004)	69-140 ng/g lw	
		Vietnam	Green mussel (2004)	0.7-5.4 ng/g lw	
		China	Fish (2009)	8.6-74.3 ng/g lw	Yu et al. (2012)
		USA	Fish (2000-2003)	38-125 ng/g ww	US-EPA (2013)
-Polymer additive of polystyrene foam, thermal isolation	b. Hexabromo cyclododecanes/ HBCDs 	Jakarta Bay	Green mussel (2003)	1.7-2.4 ng/g lw	Isobe et al. (2012)
		Cambodia	Green mussel (2004)	8.8-9.7 ng/g lw	
		China	Green mussel (2004)	2-21 ng/g lw	
		Hongkong	Green mussel (2004)	51 ng/g lw	
		India	Green mussel (2004)	0.3-16 ng/g lw	
		Japan	Green mussel (2003-2008)	10-1,400 ng/g lw	
			Blue mussel (2003-2008)	16-980 ng/g lw	
		Korea	Blue mussel (2005)	6-500 ng/g lw	
		Malaysia	Green mussel (2004)	2.2-15 ng/g lw	
		Philippines	Green mussel (2004)	6.6-17 ng/g lw	
		Vietnam	Green mussel (2004)	<0.01-140 ng/g lw	
		Jakarta Bay, Indonesia	Green mussel (2007)	4.1±1.4 ng/g lw	Sudaryanto et al. (2012)
			Fish (2007)	<1-2.8 ng/g lw	
		Benowo, Surabaya	Nile tilapia (2008)	1.6-3.3 ng/g dw	Ilyas et al. (2013)

Table 1. Emerging pollutants from industrial emission in seafood from Indonesian water and worldwide (continued)

Group and Common use	Compound	Location (year)	Species	Concentration	Reference
2. Flame retardants -Polymer additive of electronic circuits	c. Decabromo diphenyl ethane/ DBDPE 	Jakarta Bay (2003)	Green mussel	<0.3-1.2 ng/g lw	
		Cambodia (2004)	Green mussel	<0.3 ng/g lw	
		China (2004)	Green mussel	<0.3-2.2 ng/g lw	
		Hongkong (2004)	Green mussel	<0.3-2.3 ng/g lw	
		India	Green mussel	<0.3 ng/g lw	
		Japan (2003-2008)	-Green mussel	<0.3-5.7 ng/g lw	Isobe et al. (2012)
			-Blue mussel	<0.3-22 ng/g lw	
		Korea (2005)	Blue mussel	<0.3-6.6 ng/g lw	
		Malaysia (2004)	Green mussel	<0.3-1.4 ng/g lw	
		Philippines (2004)	Green mussel	<0.3 ng/g lw	
		Vietnam (2004)	Green mussel	<0.3-1.1 ng/g lw	
	d. Bis-tri-bromo phenoxyethane/ BTBPE 	Jakarta Bay (2003)	Green mussel	<0.1-0.8 ng/g lw	
		Cambodia (2004)	Green mussel	<0.1 ng/g lw	
		China (2004)	Green mussel	<0.1-0.7 ng/g lw	
		Hongkong (2004)	Green mussel	<0.1-4.3 ng/g lw	
		India	Green mussel	<0.1 ng/g lw	
		Japan (2003-2008)	-Green mussel	<0.1 ng/g lw	Isobe et al. (2012)
			-Blue mussel	<0.1 ng/g lw	
		Korea (2005)	Blue mussel	<0.1-13 ng/g lw	
		Malaysia (2004)	Green mussel	<0.1-2.1 ng/g lw	
		Philippines (2004)	Green mussel	<0.1 ng/g lw	
		Vietnam (2004)	Green mussel	<0.1-0.6 ng/g lw	
3. Aromatic sensitizers -PCBs substitute -Thermal paper -Solvent for dye	Di-isoprophyl naphthalenes/DIPNs 	Jakarta Bay (2012-2013)	- Green mussel	40-231 ng/g w w	Dwiyitno et al. (2016; 2017)
			- Fish	2-143 ng/g w w	
			- Shrimp	3-13 ng/g w w	
			- Crab	2 ng/g w w	
		Japan (2005-2006)	Fish	9-1,000 ng/g w w	Terasaki et al. (2008)
	Phenylmethoxy naphthalene /PMN 	Jakarta Bay (2012-2013)	- Green mussel	Nd -75 ng/g w w	
			- Fish	Nd-27 ng/g w w	Dwiyitno et al. (2016; 2017)
			- Shrimp	Nd-5 ng/g w w	
			- Crab	13 ng/g w w	
		Japan (2005-2006)	Fish	3-66 ng/g w w	Terasaki et al. (2008)

Note: nd: not detected; ww: wet weight; lw: lipid weight; dw: dry weight

Organotins

A number of studies have reported organotin chemicals especially butyltins (BTs) accumulation in green mussel and fish samples from different locations. Based on a study conducted by Sudaryanto (2002), the highest concentration of total BTs in green mussel was found in Jakarta Bay (58 ng/g), followed by that from Surabaya Bay (45 ng/g). Similar trend was found in fish species as performed by the most concentrated fish was from Jakarta Bay (84 ng/g), followed by that from Cirebon Bay (25 ng/g). This fact is in accordance with extensive use of organotins in the last 40 years, especially in industrial applications either as polyurethane and silicone catalyst, plastic stabilizer, or preservative. Other application of BTs is as biocide, either agricultural pesticide or antifouling paint in boat vessel and other marine equipment (Bennett, 1996; Hoch, 2001). Commercially, total BTs consists of mono-BT, di-BT, and tri-BT with tri-BT presents predominantly. Relatively higher concentration of BTs in seafood from Jakarta Bay and Surabaya, compare to those from other Indonesia waters could indicate the accumulation of BTs emission from industrial activities and/or leaching from marine vessels in those areas.

In comparison to other regions of the world, BTs contamination in Indonesian seafood is much lower than that from Malaysia, Thailand, Hongkong, Taiwan, Korea, Japan, and USA (Chen, Huang, Ho, Chang, & Liu, 2008; Harino, Fukushima, & Kawai, 2000; Hong, Takahashi, Min, & Tanabe, 2002; Prudente, 2008; Short & Sharp, 1989; Sudaryanto, 2002) as presented in Table 1. The BTs concentration in Indonesian seafood is comparable to those from Cambodia and Vietnam, eventhough the use of organotin has been banned by International Maritime Organization (IMO). Since Indonesia has not established regulation concerning this compound and therefore, possible emission of this harmful chemical should be taken into account.

Data on BTs pollutants reported in the present article are predominantly conducted before 2000. However, few current studies showed that the accumulation of BTs in marine ecosystem still occur in Indonesian waters. Razak (2005), for example, reported elevation of tributyltin (TBT) concentration in Jakarta Bay sediment based on 2003 sampling campaign (500-600 ng/g ww) compared to that in 1998 (190 ng/g ww). Furthermore, Undap (2016) reported BTs accumulation in Bitung Bay of 667 ng/g dw, elevated in comparison to that from the earlier study of 425 ng/g ww (Undap et al., 2013). Sediment BTs at concentration of 88-593 ng/g dw was also reported recently from Patani, Thailand (Hajisamoh, Nur, Siddique, & Shakya, 2018). This fact suggests that BTs are still used in the present time. In general, BTs concentration in mussel is relatively higher than that of fish species, possible associated with their different feeding behavior.

Flame Retardants

Flame retardants are contaminant typically present from industrial emission. There are three main recognized groups of flame retardants i.e. polybrominateddiphenyl ether (PBDE), hexa-bromocyclododecane (HBCD), and tetra-bromobisphenyl A (TBBPA). PBDE has been widely applied as polymer additive of electronics and building materials and also textile products, while HBCD is the main additive of polystyrene foam and thermal insulation. Due to their toxicity and persistence in the environment, PBDEs have been banned by the Stockholm convention in 2013, while HBCDs joined the list of restricted chemicals/POPs (ECC, 2016; Fayiga & Ipinmoroti, 2017; Poma, Binelli, Volta, Roscioli, & Guzzella, 2014). A number of flame retardants are reported to contaminate seafood species in Indonesian waters and other regions, i.e. PBDEs, HBCDs, bis-tri-bromo phenoxy ethane (BTBPE), and decabromo diphenyl ethane (DBDPE) as presented in Table 1.

Based on the investigation in seafood species, PBDEs was identified as the most concentrated flame retardants, followed by HBCDs, DBDPE, and BTBPE, respectively. Relatively low concentration of DBDPE and BTBPE contaminants in seafood might be associated with the relatively minor and limited use of this compound since they are lately replaced by TBBPA as new flame retardant applied for electronic circuits. Investigations on flame retardants contamination in Indonesian seafood are relatively less, at which Jakarta Bay is the most interesting environment for the study (Sudaryanto et al., 2012). Other studies conducted in Surabaya (Ilyas et al., 2013) and Lampung (Sudaryanto, 2019) revealed flame retardants concentration was much lower than that of Jakarta Bay. In comparison to other regions, concentration of flame retardants in Indonesian seafood is comparable to that from Cambodia, Malaysia, and Vietnam, but much lower than that from China, Hongkong, Korea, Japan, India, and USA (Table 1). As performed in BTs contaminants, concentrations of flame retardant in mussel are also relatively higher than that of fish species.

Aryl Hydrocarbons

Aryl hydrocarbons are reported as relatively new EPs and very limited identified in aquatic ecosystem. These contaminants are first reported by Terasaki, Fukazawa, Tani, & Makino, M. (2008) as typical organic pollutants from paper industries in Himeji Coast and Shizouka Coast, Japan. They include di-isoprophyl naphthalenes (DIPNs), phenylmethoxy naphthalene (PMN),dimethyldiphenylmethane, dimethyl-phenylethane, bismethyl-phenoxyethane, m-terphenyl, chloromethyl phenoxymethyl phenoxyethane, and benzylbiphenyl. Due to global ban of PCBs usage, DIPNs are mainly produced to replace various use of PCBs in industrial applications, as well as stabilizer of carbonless/thermal-paper, heat carrier and pesticide solvent (Suzuki et al., 2007). There is no official data of DIPNs production globally, but Japan has produced them since 1970s with a production rate of approx. 6,000 ton/year (Suzuki, Matsumura, Nakano, & Imaishi, 2012).

Contamination of DIPNs in seafood samples from Indonesian water was so far only reported by Dwiyitno et al. (2016). The samples include green mussel, fish, banana shrimp, and blue crab from Jakarta Bay. Based on their study, DIPNs concentration in fish species are relatively low (2-143 ng/g ww fish tissue) compared to that from Himeji Coast, Japan (9-1,000 ng/g ww) as presented in Table 1. The relatively high concentration of DIPNs in fish from Himeji Coast could be attributed to the high load of DIPNs emission in this area since not only paper industries are located, but also paper recyclers. Earlier study reported that paper recycling could reduce 50% DIPNs concentration in thermal paper (Jamnicki, Lozo, Rutar, & Barušiæ, 2012). Study in Jakarta Bay also reported that green mussel accumulates double concentrations of DIPNs compared to fish species, due to mussel is known as filter feeding species. Lower concentration of DIPNs contaminant was also found shrimp and crab samples (Dwiyitno et al., 2016). However, no regulation concerning either maximum residue limit (MRL) or TDI

of DIPNs is established and therefore the health risk could not be estimated.

The only study on the accumulation of PMN contaminant in Indonesian seafood was also reported by Dwiyitno et al. (2016) from Jakarta Bay ecosystem. They found that PMN concentration in fish samples was relatively low compared to that from Shizouka Coast, Japan as reported by Terasaki et al. (2008). This fact could be attributed to the different pollution loads between those areas as could be seen from the contamination level of PMN in sediment sample in Shizouka was much higher (3,700 ng/g ww) than that in Jakarta Bay (1,600 ng/g TOC) as reported by Terasaki et al. (2008) and Dwiyitno et al. (2016). Similar to DIPNs contaminant, concentration of PMN in green mussel was relatively higher than that in fish, crab, and shrimp. Even though m-terphenyl was not detected in seafood from Jakarta Bay, this compound is present in water and sediment samples as reported by Dwiyitno et al. (2015) that indicates contamination occurrence at lower concentration and/or less persistence than DIPNs and PMN. This could be seen from the lower octanol/water coefficient (log K_{ow}) of *m*-terphenyl (5.52) compared to that of DIPNs (6.08).

Plasticizers and antioxidants are among other industrial emission frequently contaminating aquatic ecosystem (Debonde, Cossu-Leguille, & Hartemann, 2011). Dsikowitzky et al. (2016) reported trimethyl pentanediol diisobutirate (TXIB), bisphenol-A, triacetine, and triethylcitrate are plasticizer compounds frequently identified in surface water of Jakarta Bay. Triethylcitrate is also employed as food additive. TXIB was detected at relatively high concentrations (up to 6000 ng/L). Additionally, bisphenol-A is also commonly applied as plasticizer and Dsikowitzky et al. (2016) detected in Jakarta river water at concentration exceeds the EC_{50} (13.9 ng/L). Antioxidants were not detected, but the derivative of buylated hydroxytoluene/BHT, known as 3,5-di-*tert.*-butyl-4-hydroxybenzoic acid and 3,5-di-*tert.*-butyl-4-hydroxybenzaldehyde were detected in nearly all Jakarta River water (Dsikowitzky et al., 2016). BHT is an antioxidant commonly used in packaging materials, adhesives, cosmetics, and PPCPs. Other antioxidant commonly used in various products is butylated hydroxyanisole (BHA). However, neither plasticizers nor antioxidants are detected in seafood species from Jakarta Bay. They might be used in less amounts or more rapidly degraded in water compartment.

Health Hazard of the Priority Pollutants

Organotins

BTs compound may contaminate aquatic ecosystem through leaching from the antifouling paints used in boat, ship, and other marine equipment, from the PVC products disposal, as well as from the runoff of farming fields. In the environment, organotins have bioaccumulative potential and the predominantly compound of TBT may degrade into derivative products MBT and DBT with the similar toxicity properties (WHO-IPCS, 1999). TBT, DBT, and MBT have been detected in different marine species including fish, mollusks, crustaceans, and cephalopods in different regions. Organotin compounds could pose harmful effects on a variety of aquatic organisms, even at low concentrations and are also suspected to disrupt human endocrine. Their toxicological effect may cause abnormalities in male reproductive systems and disrupt the critical function of human immune cells (ATSDR, 2005).

Recent findings have shown biologically significant effect of organotins in human blood samples from the USA, and Finland, as well as in human liver from Poland (Barbosa, Ferrão, & Graceli, 2018). Several investigations have revealed that seafood is the primary source of human exposure to organotin compounds (de Araújo et al., 2018). This could be attributed to TBT leach from antifouling paints from commercial vessels and/or due to the persistence of TBT in sediments. Since the majority of fish in Indonesia are captured from the sea, it becomes necessary to assess the levels of organotin compounds in seafood at the markets in order to ascertain their potential health risks to the public. Through the ingestion of polluted seafood, organotin compounds may enter the human body.

Sudaryanto (2002) has estimated contamination level of organotin from Indonesian seafood based on tolerable average residue level (TARL) approach. Based on the BTs tolerable daily intake (TDI) value of 250 ng/kg bw/day (EFSA, 2004), and to BTs concentration in green mussel from Sudaryanto (2002) which are 3.7-64 ng/g for green mussel and 3.3-84 ng/g for fish species, the daily intake of BTs to individual 60 kg Indonesian consumer is estimated between 154 and 2,662 ng/day BTs through green mussel consumption and 137-3,494 ng/day BTs via fish. This estimated Indonesian daily intake of BTs through seafood is relatively low in comparison to that of other country such as Japan (3,000-100,000 ng/person/day), Thailand (228-45,714 ng/person/day), and Philippines (2,361-68,312 ng/person/day) as compiled by Sudaryanto et al. (2002). As a comparison, if the BTs exposure around Jakarta Bay is calculated based on the more recent consumption data reported by Dwiyitno et al. (2017) i.e. 2.5-25 g/person/day for green mussel and 6.6-53 g/person/day for fish species, the BTs exposure for consumer in this area is estimated as 9.25-1,600 ng/person/day from green mussel consumption and 21.78-4,460 ng/person/day

from fish. These number are still below the TDI of 15,000 ng/person/day. TBT concentration in blue mussel (*Mytilus galloprovincialis*) and fish (*Tapes* spp) over the TARL has been reported from other region such as from the Lagoon of Venice, Italy (Zanon, Rado, Centanni, Zharova, & Pavoni, 2009).

Flame Retardants

Flame retardants, such as brominated flame retardants (BFRs) may be released into the aquatic environment during their production emission or the disposal of plastics and electronic wastes that contain these compounds. BFRs such as PBDEs and HBCDs have gained public attention as EPs due to their characteristics such as persistency, bioaccumulative properties, and possible harmful effects to wildlife and human. Toxicological studies have shown that PBDEs and HBCDs could disrupt thyroid hormones and endocrine, change the neurobehavioral, affect to fetal development, and possible cause cancer in animal study (Gill, Chu, Ryan, & Feeley, 2004; Hallgren & Darnerud, 2002). The wide use of flame retardants in various daily products has resulted in their accumulation at all environmental compartments (Covaci et al., 2006; Hites, 2004). As these compounds are relatively lipophilic and persistent (K_{ow}: 5.8-7.0), they can easily accumulate and biomagnify through food webs. For that reason deca-and hexa-bromodiphenyl ether are recently categorized as new POPs according to Stockholm convention on POPs in June 2017 (UNEP, 2017). Therefore, their bioaccumulation and biomagnification in aquatic ecosystems could generate the risk in higher trophic level organisms including human.

The commercial PBDE products are predominantly composed of penta-, octa-, and deca-bromodiphenyl ether products. Each product may elicit different toxicological properties. Generally, the penta-BDEs seem to cause toxicity at the comparably lowest dose, while much higher doses are required for effects of the deca-BDEs. The critical effects of penta-BDEs include neurobehavioral disorder (from 0.6 mg/kg body weight) and at higher dose will alter thyroid hormone levels in rats and mice study. Exposure of 2 mg/kg body weight octa-BDEs showed fetal toxicity/ teratogenicity in rats and rabbits, while 80 mg/kg body weight of deca-BDEs revealed thyroid, liver, and kidney morphology disorder in adult animals (Hendriks & Westerink, 2015). Since the official threshold/TDI for BFR compounds has not been established, data of animal studies are used to estimate their BMDL (benchmark dose confidence limit) for calculating margin of exposure (MOE).

US-EPA (1995) and EFSA (2011) have suggested BMDL of BFRs compounds, such as BDE47 (309 µg/kg bw/day), BDE99 (12 µg/kg bw/day), BDE153 (83 µg/kg bw/day), BDE209 (1700 µg/kg bw/day), HBCDs (790 µg/kg bw/day), and TBBPA (16000 µg/kg bw/day). Studies show only deca-BDEs link to carcinogenic effects at very high levels and therefore IARC (1990) revealed deca-BDEs not classifiable as carcinogenic to humans. Based on the animal studies, a critical effect of HBCD was seen in liver and on thyroid hormones (LOEL 100 mg/kg bw/day). This value can also be furthermore used to estimate its BMDL. However, behavioral effects of HBCD on infantile were observed at concentration of 0.9 mg/kg body weight and a sensitive behavior effects endpoint may also present as for other BFRs.

Based on the BFRs concentration in green mussel and fish from Jakarta Bay reported by Sudaryanto et al. (2012), MOE of these contaminants to consumer around Jakarta Bay could be estimated. Using dietary record in 4 districts around Jakarta Bay of other study (Dwiyitno et al., 2017), i.e. 25 g/person/day for green mussel and 53 g/person/day for fish species, the MOE of BFRs for consumer in this area is estimated as 9.5×10^4 and 6.8×10^4 for PBDE99 from green mussel and from fish species, respectively and 3.1×10^4 and 1.3×10^4 for HBCD from green mussel and from fish consumption, respectively. These MOE values are practically much higher than the MOE threshold of PBDE (2.5) and HBCDs (100), indicating consumer is still safe from BFRs exposure through seafood consumption. MOEs of BFRs contaminant over the threshold have been reported from the minority seafood consumer in Portugal (BDE99<2.5) as reported by Aznar-Alemany et al. (2016).

Aryl Hydrocarbons

Since the structural properties of aryl hydrocarbons are analog to PAHs and biphenyls (Table 1), they are considered accumulative and toxic in the environment (Terasaki et al., 2008). According to Stockholm Convention, DIPNs is classified as slow biodegradable compound with T_{50} in water of 2 months. Further toxicity analysis showed LD_{50} on yellowtail (*Seriola quinqueradiata*) was approximately 2 ml/kg which corresponds to highly aquatic toxicity according to the Globally Harmonized System. Long-term toxicity test showed the No Observed Effect Concentration (NOEC) study with *Daphnia* is 13 µg/l. Further bioassay test on rat indicated an increase of liver weight, disturbance of lipid metabolism in the liver, and serum and disturbance of glucose metabolism. Even though earlier studies showed that DIPNs, for example, exhibit toxicity to aquatic animals (*S.*

quinqueradiata and *Daphnia*) with LD_{50} and NOEC 2 ml/kg and 13 μg/l, respectively, those test were not sufficient to establish the exposure limit. However, sub-chronic exposure on rat showed a lethal dose of 100 μg/kg bw when directly administered into stomach (UN 2011; UNEP 2014). Unfortunately, no acceptable exposure limits levels are available (UNEP, 2014).

In contrast with DIPN, a bioassay study of PMN in rat has been conducted by NICNAS (1999). Based on an oral test, PMN showed a Lowest Observed Effect Level (LOEL) concentration for hepatic toxicity of 300 mg/kg bw/day. At this concentration, animal test showed a liver hypertrophy (increase of liver weight). Other study revealed that liver hypertrophy is among common response on stress of rat and in long term may induce hepato-carcinogenesis (Hall et al., 2012). For this reason, MOE for PMN contaminant for consumer around Jakarta Bay could be estimated based on consumption rate reported by Dwiyitno et al. (2017). Based on the LOEL value of PMN 300 mg/kg bw/day with correction factor of 100 and the estimation of seafood consumption 25 g/person/day for green mussel and 53 g/person/day for fish species, MOE of PMN is estimated of 230,760 for green mussel consumption and 142,852 for fish consumption. This value is categorized as fairly safe since the risk threshold for MOE of PMN is 10,000 (EFSA, 2005).

Future Concern

With regard to the adverse effects of EPs, global concerns have been undertaken in order to initiate mitigation, especially to protect the aquatic environment. Different countries have banned the use of TBTs since the mid-1980s (Konstantinou & Albanis, 2004). Organotin-based antifouling paints have been prohibited in France for the use on boats less than 25 m long since 1982, followed by North America, UK, Australia, New Zealand, Hong Kong, and most European countries a few years later (Champ, 2000). A global ban on the application of TBT-based paints was supported by IMO since 1 January 2003, and total prohibition by 1 January 2008 (IMO, 2001). In Europe, Decision No.2455/2001/EC amended directive 2000/60/EC on water policy to agree 11 hazardous chemicals, including TBTs as subject to prohibit the emissions and discharges into aquatic environment. Furthermore, decision No. 415/2004/EC prohibits the usage of organotin compounds on ships.

In national level, the Royal Decree 995/2000 of the Spain regulated the level of organotins less than 20 ng/L in waste discharges to continental. Most developed countries, such as USA, UK, Canada, France, New Zealand, Australia, and the European Union have adopted the restrictions on the use of TBT containing paints (Birchenough et al., 2002). In 1998, the United States introduced Organotin Antifouling Paint Control Act to the federal level a leaching rate of 4 μg/cm^2/day. Additionally, the use compounds as food additive has been regulated by the Food and Drug Administration (ATSDR, 2005). The use of organotin compounds in Taiwan as antifouling agents was restricted to boats under 25 m in length since 2003. However, BT compounds were still identified in water and sediments (Zanon et al., 2009).

In Indonesia, organotins, flame retardants, and aryl hydrocarbons specifically emitted from industrial activities as mentioned in this article have not been-included specifically in the regulation of waste management of hazardous and toxic chemicals (Indonesian Government, 2004). Since there is no official regulation, it is likely that organotins still continue to be used as effective biocides and/or used as wood preservatives. Since BTs are rapidly absorbed by organic materials such as bacteria and plankton, once released to the environment, these compounds are readily accumulated into filter-feeding species and higher organisms such as seafood as well as adsorbed onto suspended particles (Harino et al., 2008).

It has to be noted that recent findings concerning organotins contaminant is not only limited to BTs, but also phenyltins (PhTs) and dioctyltin (DOT). The PhTs, especially triphenyltin (TPhT) has become concern as environment contaminant since it poses harmful effect similar to TBTs as endocrine disrupting compounds either to aquatic organism or human. Concerning the exposure of organotins, WHO has regulated a TDI of 0.25 μg/day/kg bw for TBT contaminant, while TDI for TPhT is 0.5 μg/day/kg bw (WHO-IPCS, 1999). In contrast, European set a TDI for total organotins (2 OTs) of 0.25 μg/day/kg bw as the sum of BTs, TPhT and DOT (EFSA, 2004). Therefore regulation concerning the use of TPhT needs to be addressed in order to protect the harmful effects to the environment.

It has been proved that commercial formulation of flame retardants is dominated by three degrees bromination PBDEs i.e., penta-BDE, octa-BDE, and deca-BDE. Commercial market of PBDEs in 2001 indicated that Asian countries consumed as the second (24,650 tons or 37%) after Americas (33,100 tons or 49%), while Europe was the lowest (8,360 tons or 12%). However, due to toxicological reason, organophosphate flame retardants (PFRs) have recently replaced PBDEs. Consequently, global concerns about PFR contamination and its impact on aquatic environment and human health have increased. A recent study on human exposure suggested that seven out of eleven target PFR metabolites, including diphenyl phosphate (DPHP)

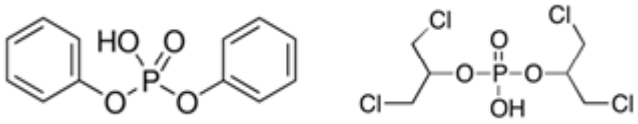


Figure 2. Molecular structure of diphenyl phosphate/DPHP (left) and bis(1-chloro-2-propyl)-1-hydroxy-2-propyl phosphate/BCIPHIPP (right)

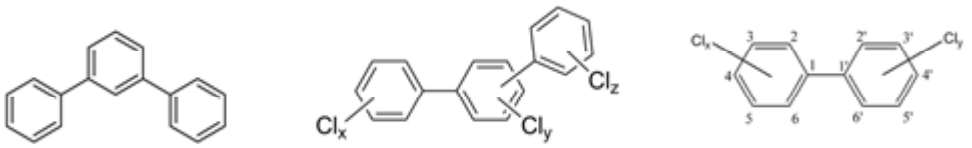


Figure 3. Molecular structure of m-Terphenyl (left), polychlorinated terphenyls (middle), and PCBs (right)

and bis(1-chloro-2-propyl)-1-hydroxy-2-propyl phosphate/BCIPHIPP (Figure 2), were frequently detected in human urine and also the most frequently detected metabolite in both serum and hair (Xu et al., 2019).

Based on limited studies in Japan and Indonesia, some compounds from aryl hydrocarbons group showed significant concentration in aquatic environment, in particular DIPNs and PMN. However, global concern to these contaminants seems very minor, hence no regulatory body addresses this compound. Specifically to terphenyl, which was also detected in Japan Coast and Jakarta Bay (Terasaki et al., 2008; Dwiyitno et al., 2016), the occurrence to develop complex compound such as polychlorinated terphenyls/PCTs (when chlorine is present) must be monitored. This is due to PCTs are classified as polyhalogenated compounds previously used in similar applications as PCBs (Figure 3), but the industrial production was closed in the 1970s. PCTs and PCBs have similar chemical and physico-chemical properties, as well as the toxicological behavior.

Restrictions of OTs and other pollutants will not immediately remove their residue in the environment. TBTs and their degradation products for example, are retained in the aquatic compartments where they persist. Illegal use of this compound and other prohibited chemicals may also be occured if not supported by law enforcement. For that reason a continued monitoring program needs to be established either in the aquatic ecosystem or potential points of pollution sources in the terrestrial. In Indonesia, Ministry of Environment is responsible for the coordination of monitoring environmental pollution, including industrial emission. Related ministries and local environmental agencies may contribute for concerning ecosystem, such as Ministry of Marine Affairs and Fisheries could be in charge for monitoring aquatic ecosystem in order to assure the safe seafood. However, monitoring management has not been established comprehensively due to some limitation such as human resource expertise, supporting facilities, budget, and community awareness (Wahjono & Satmoko, 2006; Yudo, 2016).

Laboratory approach is another issue concerning EPs analysis. Since target compounds are present at relatively minor levels (ppb), their quantification requires highly sensitive equipments. The possible interference content from the environment, such as salt content, may cause difficulties in the determination step (Brunori, Ipolyi, Massanisso, & Morabito, 2005). An accurate characterization of concentration levels of organotin, particularly TBT and its derivates requires sample preparation and chemical analysis, based-on the analyte (e.g. content of water and lipid). Every analytical step (e.g. extraction, separation, derivatization, and detection) potentially can affect the accuracy and precision of the final results (Diez, Jover, Albaiges, & Bayona, 2006).

In the case of flame retardants, most of the studies reported the concentration of total PBDEs. Practically, commercial PBDEs may present in more than 50 possible congeners and each congener has different physico-chemical properties, accumulation pattern, and toxicological pathways. For this reason, identification of each PBDE congener will be beneficial to estimate the more accurate adverse level on the expose assessment which consequently needs more efforts in the laboratory analysis, such as fractionation/ derivatization step, the availability of concerning certified reference materials (CRM) and advanced laboratory equipments.

The limited study of the contamination level of aryl hydrocarbons maybe caused by the rarity of supporting toxicological studies of these compounds. Additionally, due to limited toxicological data, regulation on TDI threshold of flame retardant is not

available yet. Therefore, it is suggested to conduct bioassay studies either in aquatic organisms or mammalian species to ascertain the safety level of this contaminant group and the possible adverse effects to the aquatic environment and human health.

Conclusion

Information of EPs in aquatic environment and seafood from Indonesia, in particular industrial emission is very limited. Based on the literatures, the studies were predominantly focused on organotin compounds, followed by flame retardants. However, the rare studies of organotin contaminants have been conducted in recent years. In general, areas of interest for those studies are situated around Java Island, followed by Sumatera. In comparison to EPs in seafood from other countries, EPs concentrations from industrial emission detected in Indonesian seafood samples are relatively lower than those in developed countries, and the majority of South East Asian countries, but comparable to those in Vietnam and Cambodia. Aryl hydrocarbons contamination is the least investigated subject globally, including in Indonesia. In general, the concentrations are still below the residual limit to pose adverse effect on human health.

Toxicological studies suggested that EPs emitted from industrial activities has to be concern. All the contaminants basically pose adverse effect either to aquatic organism or human health, with most of the EPs attributed to endocrine disrupting compounds. Nevertheless, studies on the exposure assessment of EPs in Indonesia are also quite rare. Some compounds, such as flame retardants and aryl hydrocarbons need further toxicological study to support threshold of safety level (TDI) for exposure assessment. Regulation issue, monitoring program, and laboratory approach are among future focus to be concern in addressing EPs in Indonesia, especially from industrial emission.

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