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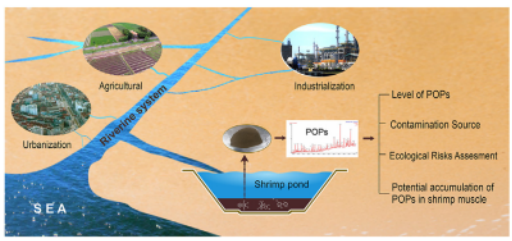
HIGHLIGHTS

- The persistent organic pollutants were detected in the shrimp ponds.
- The occurrence patterns of the POPs in sediment were site dependent.
- The riverine system contributed to the higher concentrations of POPs.
- POPs may from historical inputs rather than a recent application.
- A risk assessment using several indices indicated potential adverse effects.

GRAPHICAL ABSTRACT



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ABSTRACT

Organochlorinated pesticides (OCPs) and Polychlorinated biphenyls (PCBs) in the surface sediments from shrimp ponds in four regions of the northern part of the Central Java coast (namely Brebes, Tegal, Pemalang, and Pekalongan) were investigated. The highest concentration of $\sum$ OCPs was found in Brebes Regency, ranging from 68.1 ± 3.4 to 168.1 ± 9.8 μg kg^{-1} dw. As indicated by the DDT ratio and chlordane ratio, the value suggested that those compounds may mainly originate from historical inputs rather than a recent application. The concentrations of Σ 7 indicator PCBs were determined, with the concentration ranged from 1.2 ± 0.7 μg kg^{-1} dw (Pekalongan) to 2.2 ± 0.4 μg kg^{-1} dw (Tegal). The most toxic PCB congener, PCB 118, was detected in all studied regions, with the highest proportion found in Tegal. Source analysis indicated that PCBs in the sediments mainly originated from Aroclor 1254 and Aroclor 1248. Compared to sediment quality guidelines (SQGs), some OCPs were found with concentrations which potentially posed an adverse effect. Our findings suggested that more attention should be paid to ensure sustainable shrimp culture facing such a risk of the OCPs and PCBs.

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1. Introduction

Indonesia is the world's third-largest rice producer, and Java is the main island for farming activities (USEPA, 2017; FAO, 2004). In Indonesian agricultural areas, including Java, pesticides have been massively used to kill and control organisms occurring on agricultural products and crops (Kartini et al., 2019), potentially resulted in a rapid increase in inputs of large numbers of pesticides into the environment (Glinski et al., 2018).

Furthermore, since around 13% of the Indonesian people live on Central Java with a very high population density, i.e. 1052 people/km^2, there is a dramatic impact of human activities on the environment through different anthropic activities. The growing population of Central Java (more than 34 million in 2018) have led to an increasing input of wastewater, mainly from industry, agriculture and households into aquatic system (BPS, 2019; Dsikowitzky et al., 2011). Furthermore, in line with the strong growth of the country, Indonesian coastal regions are developing very quickly due to commercial, agricultural, and industrial activities. Industrial and urban activities were previously identified as the possible sources of environmental contamination by PCBs. In addition, due to inadequate management, the contamination by anthropogenic pollutants on terrestrial and aquatic ecosystems might occur with a widespread and/or diffuse pollution (Sumon et al., 2018).

Previous authors stated that OCPs and PCBs, being mainly of anthropogenic origin, are transported to aquatic bodies by wastewater discharges (CCME and Environment, 1999), spray drift (Sumon et al., 2016), rain runoff, rivers and streams (Rabiet et al., 2010), and atmospheric deposition (Richards et al., 2016). These substances have become issues of most concern because of their bioaccumulative properties, persistency characteristic (Stockholm, 2011), and potential toxic effects to wildlife and human (González-Mille et al., 2010; Islam et al., 2018; Zhou et al., 2020).

In general, POPs are usually hydrophobic (i.e. "water-hating") and lipophilic (i.e. "fat-loving"). In aquatic systems, due to the latter properties, POPs bind strongly to solids, and more especially to organic matter (Ashraf, 2017), hence evading the aqueous compartment. Therefore, through sedimentation processes, sediments have become the primary reservoir of POPs (Ren et al., 2018). On the other hand, as the most important aquaculture commodities in Indonesia, white pacific shrimp (*Litopenaeus vannamei*) is considered as a benthic organism with a feeding behavior that easily accumulate chemicals from sediment (Martínez-Córdova and Peña-Messina, 2005; Varadharajan and Pushparajan, 2013). Some studies have shown that exposure to chlorinated pesticides caused protein decrease in exposed shrimp, increase in the larval respiratory rate, decrease of glycogen synthesis and nucleic acid content, suggesting drastic metabolic changes (Galindo Reyes et al., 1996). Moreover, OCPs can be retained in living organisms and indirectly threaten human health via ingestion because of their biomagnification capability through aquatic food chains to higher trophic (Ashraf, 2017; Ross and Birnbaum, 2003; Song et al., 2019).

Based on the Minister of Agriculture of the Republic of Indonesia Decree No. 434.1/Kpts/TP.270/7/2001; Minister of Agriculture of the Republic of Indonesia Decree No. 39/Permentan/SR.330/7/2015 and under Indonesia's ratification of the Stockholm Convention on POPs Convention in 2009, the use of these compounds are currently banned or severely restricted in Indonesia. As these chemicals persist in the environment and can remain for a very long time in sediments (Gavrilescu, 2005; Plimmer, 1992), their monitoring must be undertaken. It is important to provide the anthropogenic pollution status associated with their potential risk to the ecosystem, human health, and also the "remediation effects" of adequate management policy. In this study, as sediment is considered as the key matrix in the pollution monitoring programs and as a final sink for many particle-bound chemicals (Martinez et al., 2010), we monitored the presence of OCPs and PCBs in the sediment from 12 different aquaculture systems in the Northern part of Central Java Coast, Indonesia which is considered as the main shrimp producer.

2. Materials and methods

2.1. Reagents and chemicals

Pesticide 8081 standard mix containing 22 compounds, i.e., Aldrin, alpha-Hexachlorocyclohexane (α-BHC), gamma-Hexachlorocyclohexane (γ-BHC), beta-Hexachlorocyclohexane (β-BHC), delta-Hexachlorocyclohexane (δ-BHC), α-Chlordane, γ-Chlordane, Dichlorodiphenyldichloroethane (4,4′-DDD), Dichlorodiphenyldichloroethylene (4,4′-DDE), Dichlorodiphenyltrichloroethane (4,4′-DDT), Decachlorobiphenyl, Dieldrin, α-Endosulfan, β-Endosulfan, Endosulfan Sulfate, Endrin, Endrin Aldehyde, Endrin Ketone, Heptachlor, Heptachlor exo-epoxide, Methoxychlor, and 2,4,5,6-Tetrachloro-*m*-xylene was obtained from Sigma-Aldrich (St. Louis, MO, USA). While the individual pesticides (Chlorfenvinphos, Chlorpyriphos-ethyl, Alachlor, and Metolachlor) were obtained from LGC Standards GmbH D-46485 Wesel. PCB standards were purchased from Dr. Ehrenstorfer Laboratories (Augsburg, Germany). Basic alumina was purchased from VWR (Radnor, PA, USA), copper powder was purchased from Merck (Pessac, France), and Mega Bond Elut Florisil cartridges (1 g, 6 mL) were obtained from Agilent Technologies (USA). SupraSolv grade solvents, including dichloromethane (DCM), n-hexane (HEX) and acetone (ACE) were provided by Merck, Darmstadt, Germany. Hydrochloric acid (35%), nitric acid (69%) and phosphoric acid (85%) of pure grades were provided by Fisher Scientific (Marseille, France). Analytical grades of potassium dichromate ($K_2Cr_2O_7$> 99%) sulfuric acid (H_2SO_4, 95–98%), ferrous sulfate heptahydrate ($FeSO_4 \cdot 7H_2O$> 99%) were bought from Merck, Darmstadt, Germany.

2.2. Study area

The sediment samples were collected from 12 stations corresponding to shrimp farms located in four regencies of the northern part of Central Java, namely Brebes, Tegal, Pemalang, and Pekalongan (see Fig. 1; Supplementary material, SM, Table S1). Besides, those regions were selected as they are among the highest shrimp producer regions in Central Java, and close to agricultural and/or urbanized areas.

2.3. Sample collection

In each pond, superficial sediment (0–5 cm) samples were collected in three points, thereby each sample was a composite sample, made up with three sub-samples. Then, the sediment was mixed in an aluminum tray, immediately placed in polyethylene bags and stored in a cool box with gel ice before being transferred to a laboratory. Upon arrival in the laboratory, sediment samples were subsequently freeze-dried over a period of a 72 h, sieved through stainless steel sieves for a particle fraction of less than 2 mm size, and lightly ground in an agate mortar for homogenization. Then the samples were stored at 4 °C prior to analysis.

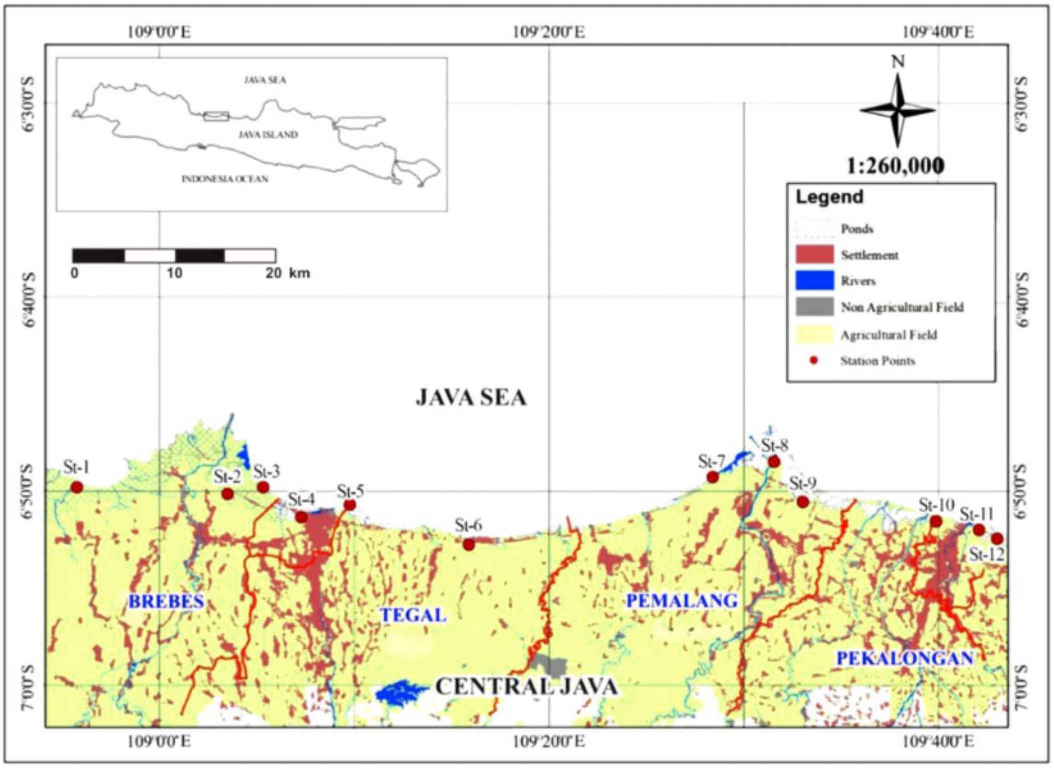

Fig. 1. Map of the Northern Coast of Central Java showing sampling stations (indicated with red circles) and major types of land cover (source: http://portal.ina-sdi.or.id/). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

2.4. Sample extraction

Approximately 10 g sediment samples, 3 g of activated copper particles (>230 mesh), and 4 g of activated alumina were mixed and then transferred to a 33 mL extraction cell of an accelerated solvent extraction system (ASE 350: Thermo Scientific, Waltham, MA). Surrogate deuterated standards (4,4′-DDE-D_8 and PCB 156-D_3) were added, then samples were equilibrated at room temperature for 1 h before extraction. The POPs in the sediment samples were then extracted with (HEX/ACE, 1:1 v/v) using a Thermo Scientific™ Dionex™-Accelerated Solvent Extractor ASE 350 with the following conditions: 100 °C, 1500 psi, 5 min static time and 60% flush volume (Kanzari et al., 2012). The resulting extracts were then evaporated to about 2 mL with a rotary evaporator (Heidolph, Laborata 4000 efficient, Krackeler Scientific, Pessac, France), and the extract was further submitted to fractionation and clean-up.

The procedure used for purification and fractionation for OCPs and PCBs analyses was based on USEPA 3620C methods. Briefly, the concentrated extracts were purified and fractionated into fraction 1 and fraction 2 using MegaBond Elut Florisil Solid Phase Extraction (SPE) cartridges (1 g, 6 mL). PCBs and some OCPs were eluted in "fraction 1" using 2.5 mL of 100% HEX (fraction 1), while the other OCPs was eluted in "fraction 2" using 5 mL of HEX/ACE (80/20 v/v). The eluent was collected in a 2 mL amber vial and then concentrated to about 1 mL under a gentle stream of nitrogen. After that, the concentrated eluent was transferred to a GC vial containing 8 μL of PCB 116 D_5 as internal standards for GC-MS analysis.

2.5. GC-MS analysis

The sediment were analyzed for 22 individual organochlorine pesticides and transformation products: Aldrin, α-Endosulfan, 4.4′-DDD, 4.4′-DDE, 4,4′-DDT, Dieldrin, Endrin, α-HCH, β-HCH, γ-HCH, Heptaclor, Heptachlor Epoxide, δ-HCH, Alachlor, Metolachlor, Chlorpyriphos-ethyl, Chlorfenvinphos, γ-chlordane, α-chlordane, Endrin Aldehyde, Endosulfan Sulfate, and Endrin Ketone. We also analyzed the seven indicator PCB congeners: 2,4,4-Trichlorobiphenyl (PCB 28), 2,2,5,5-Tetrachlorobiphenyl (PCB52), 2,2,4,5,5-Pentachlorobiphenyl (PCB101), 2,2,4,4,5-Pentachlorobiphenyl (PCB118), 2,2,4,4,5,5-Hexachlorobiphenyl (PCB153), 2,2,3,4,4,5-Hexachlorobiphenyl (PCB138), 2,2,3,4,4,5,5-Pentachlorobiphenyl (PCB180). Identification and quantification of OCPs and PCBs were performed in both SIFI mode (full scan) and selected ions recording (SIR) mode data collection running simultaneously. Retention times and characteristic ions of studied compounds are displayed in Table S2. The gas chromatography (PerkinElmer AutoSystems XL, California, USA), equipped with a Restek Rxi-XLB (30 m × 0.25 mm i. d. × 0.25 μm film thickness, Restek Corporation) capillary column, and coupled with a Clarus 600C mass spectrometer (MS) was used for determining the concentrations of the POPs in the sediment samples. The GC oven temperature was programed as follows: initial temperature 70 °C (2 min), then raised to 175 °C (10 °C min^{-1}) and held isothermally for 4 min, then raised to 320 °C (5 °C min^{-1}) and finally held for 1 min. The injector was set from 50 °C (0.1 min isothermal) to 250 °C (200 °C min^{-1}) held for 10 min. MS was run in the electron ionization (EI) mode (70 eV) at a source temperature of 300 °C. The samples were injected in the splitless mode, and a high-purity helium was used as the carrier gas with a constant flow of 1 mL min^{-1}.

2.6. Sediment grain size and organic carbon determination

All samples were analyzed both for grain size and organic matter. Sediment grain size was determined "manually" by wet sieving through a series of ISO 3310-1 stainless steel sieves (Retsch France): sand (between 0.250 mm and 2 mm), fine sand (between 0.063 mm and 0.250 mm), and mud or silt and clay (<0.063 mm). Each fraction was dried, weighed, and classified according to Folk's classification. Total Organic Carbon (TOC) content was determined based on the Walkley-Black chromic acid wet oxidation method. In brief, oxidizable matter in the sediment was oxidized by 1 N $K_2Cr_2O_7$ solution. The reaction is helped by the heat generated when two volumes of H_2SO_4 are mixed with one volume of $K_2Cr_2O_7$. The remaining $K_2Cr_2O_7$ is titrated with $FeSO_4$. The titer of the solution is inversely related to the amount of C present in the sediment sample (Wang et al., 2019; Zhao et al., 2016).

2.7. Quality assurance/quality control (QA/QC)

All analytical procedures were performed under strict quality assurance and control procedures. In order to control any contamination during chemical analysis, laboratory blanks were included in every set of three samples and treated in the same manner as the samples. In all the blank samples, the values were lower than the detection value. A certified reference material (CRM) CNS391 "PAHs, PCBs and Pesticides on freshwater sediment" (Sigma-Aldrich, Molsheim, France) was used to evaluate the method performance. Each sample was processed in triplicate.

Calibration curves of individual target OCP and PCB exhibited a very good linear response ($r^2 > 0.999$) for quantification by GC-MS. Detection limits (LODs) defined as 3 times baseline noise was comprised between 0.04 ng g^{-1} and 0.71 ng g^{-1} for OCPs and 0.02–0.13 ng g^{-1} for PCBs. Recovery rates of 82%–94% were obtained for target compounds. Concentrations of OCPs and PCBs were determined based on standard surrogate of 4,4-DDE D_8 and PCB156 D_3 (100 pg μL^{-1}). Internal standard solutions (40 pg μL^{-1}) of PCB116-D_5 were injected to samples before GC/MS analysis to calculate surrogate recoveries and target compounds. The results for OCPs and PCBs analysis are expressed in μg kg^{-1} dry weight (dw).

2.8. Data analysis

The statistical analyses were performed using R statistics package. We performed statistical analyses using nonparametric tests i.e. Kruskal-Wallis test to highlight differences in the concentration of the OCPs occurrences among regions. Characterization of PCBs homolog were performed with Principal Component Analysis (PCA). In addition to the statistical analyses, diagnostic ratios were used to distinguish the sources of emission and pollution input history, i.e. HCH ratio; cis-chlordane/trans-chlordane ratio; p, p′-(DDE + DDD)/p, p′-DDT ratio; and $\sum$ DDTs/$\sum$ PCBs.

The $\sum$ HCHs was defined as the sum of α-HCH, β-HCH, γ-HCH and δ-HCH. The source of HCH was evaluated using the ratios of the HCH isomer composition, as previously described by (Tan et al., 2009) and (Qiu et al., 2019). High α-HCH/β-HCH ratios usually indicate current sources of technical HCH, while low ratios suggest historical sources (Qiu et al., 2019). The values of α-HCH/γ-HCH in environmental samples can be used to monitor whether a source was technical HCH or lindane. The ratio of α-HCH to γ-HCH concentrations between 3 and 7 (Mitra et al., 2019) or between 4 and 7 (Tang et al., 2020) indicated the possible source of technical HCHs, while the ratio <4 indicates the use of lindane. Cis-chlordane/trans-chlordane ratio is evaluated based on (Alam et al., 2014). The value of cis-chlordane to trans-chlordane ratios <0.79 indicates the recent uses of commercial chlordane, while the value > 0.79 suggests historical use of commercial chlordane. Then, DDT ratio was used to investigate the source of DDT, and calculated according to the previous works (Neves et al., 2018; Tran et al., 2019):

$$DDTs\ ratios = \frac{p,p' - \mathrm{DDT}}{p,p' - \mathrm{DDE} + p,p' - \mathrm{DDD}}$$

Briefly, a DDTs ratio <1 indicates a past input while a DDTs ratio> 1 indicates modern/recent input (<5 years).

3. Results and discussion

3.1. Organic carbon and sediment grain size

In terms of OCPs distribution in sediments, factors such as sediment texture, organic carbon, land use and seasonal variation have been found to exert influence (Duodu et al., 2017). TOC concentration is considered as an important factor in relation to the distribution of organic contaminants (CCME and Environment, 1999). In the present study, we found a positive correlation between concentrations of OCPs and TOC ($R^2 = 0.77$). A similar tendency was reported by (Huo et al., 2017) in Lake Chaohu sediments, China and (Lv et al., 2020) in 14 typical intertidal zones of China. The average of TOC in the studied sediments of the shrimp ponds (1.9 ± 1.8%) was in medium level, that is lower to those coastal waters around intensive shrimp ponds in Lampung province, Indonesia (ranged between 0.30 and 9.85%, average 3.09%) and much lower to those from the 5-year-old shrimp culture ponds in the Shanyutan wetland of Min River estuary, South-east China (22.4 ± 2.0%) (Gao et al., 2019); comparable to those Cua Hoi Estuary (average 1.72%) and Han River Estuary (average 1.78%) in coastal area of central Vietnam (Tham et al., 2019).

Site 3 (Brebes) presents the highest of TOC (5.9 ± 1.7%), while site 3 in Pekalongan is the site with the lowest TOC (0.3 ± 0.01%). A study by (Gao et al., 2019) shown that shrimp aquaculture significantly increased sediment TOC as a result of the decomposition of a large number of residual feeds and excrement. A similar result reported by (Gao et al., 2018) in their previous work in the 3-year-reclamation culture ponds in Coastal Zone of Southeastern China. In addition, Yang et al. (2017) found that CH_4 emission fluxes in shrimp ponds were clearly high during the mid and later part of the aquaculture season and were directly proportional with TOC level (16.2 ± 0.6%).

Other factors also play an important role on the distribution and concentration of POPs in sediments, such as sediment grain size (Barhoumi et al., 2014). Our results demonstrated that there was a weak positive relationship between mud and OCPs level in the surface sediment samples ($R^2 = 0.51$). Similarly, we found a weak positive relationship between clay (mud and silt) and sediment organic carbon contents in shrimp ponds ($R^2 = 0.56$), which is consistent with the previous results obtained by Bergamaschi et al. (1997), Flemming and Delafontaine (2000), and Lee et al. (2019). In general, sediment properties such as grain size and composition are known as important environmental factors to determine the distributions and fate of organic matters in the shallow water system (Serrano et al., 2016). For example, muddy sediment mainly composed of silt and clay retains more organic matter compared to coarse-grained sediments (e.g. sand), due to a greater adsorption capacity of fine-grained particles by earning a larger surface area (Burdige, 2007; Keil and Hedges, 1993).

3.2. The occurrence and sources of organochlorine pesticides (OCPs)

Concentrations of OCPs are presented in Table 1, showing that despite the ban of aldrin, dieldrin, heptachlore, cis-chlordane, endosulfan, DDT, and α-HCH in Indonesia (based on the Decree of Indonesian Minister of Farming Letter No. 434.1/Kpts/TP.270/7/2001), we still found those compounds in all regencies and in most sediment samples.

Of the 20 substances, trans-chlordane, cis-chlordane, aldrin, dieldrin, 4,4′ DDD, γ HCH, and metolachlor were detected in ≥50% of the samples, whereas trans-chlordane and cis-chlordane were identified to be the most dominant of OCPs and quantified in 83% and 67% of the sediment samples, respectively (Fig. 2).

Among the selected regions, Brebes presented the highest total OCPs concentrations, ranging from 68.1 ± 3.4 to 168.1 ± 9.8 μg kg^{-1} dw. Central Java is the main province of onion production in Indonesia (BPS, 2019), with Brebes Regency as the central production area, contributing to about 66% of Central Java production in 2015–2016 (BPS, 2016). Because onions are very susceptible to pests, there is a widespread use of pesticides in this region to prevent and/or eradicate the pest, herb or fungi. That is why Brebes is considered as the regency with the largest pesticide usage both in Indonesia and in South-east Asia (Suhartono, 2018). In their previous work (Suhartono et al., 2012), found that farmers in this area spray the onions about 2–4 times a week in the dry season and even every day in the rainy season. 38 different commercial brands of pesticides (23 types of insecticides, 12 types of fungicides and 3 types of herbicides) are used in onion production. Moreover, in 2017 about 1300 of 3200 pesticide brands registered by the Ministry of Agriculture, Republic of Indonesia are marketed in Brebes Regency (Suhartono, 2018).

Pemalang area is ranked in the second highest for contamination among the four regencies, with total OCPs concentrations ranging between 45.8 and 118.1 μg kg^{-1} dw. In Central Java, Pemalang is one of the nine main Java paddy producer centers with an agricultural area of 34,457.49 Ha (Pemalang Regency, 2019). There are 14 pesticides used in Pemalang, and the dose of the use of pesticides in rice farming areas is the highest in Central Java with a frequency of use of 2–3 times each week (Ardiwinata and Nursyamsi, 2012; Bantarwati et al., 2013; Kurniasih et al., 2013). Through various channels, e.g. riverine run-off and atmospheric deposition, these compounds may find their ways into shrimp farming areas where they become deposited and trapped on surface sediment.

3.3. Chlordanes (trans- and cis-chlordane), heptachlor and heptalochlor epoxyde

In aquatic environment, chlordane is highly hydrophobic, and strongly associated with the organic carbon, clay and silt (Hirano et al., 2007). Chlordane is a ubiquitous and persistent pesticide, which was widely used for both agricultural and residential applications since its introduction in the 1940s (Ouyang et al., 2005). Although the use of chlordane was banned in the 1980s, it is still commonly detected in the aquatic system (Alam et al., 2014; Hirano et al., 2007).

With regard to the distribution of compounds belonging to the Chlordane family, heptachlore had the major contribution (63.3% of ΣChlordanes), followed by trans-chlordane (24.4%), heptachlor epoxide (7.3%), and cis-chlordane (4.9%). Heptachlor, typically originates from technical chlordane, is also used as an insecticide (Baek et al., 2011). In order to evaluate the source of chlordane, we calculated the cis-chlordane/trans-chlordane ratios, based on work done by (Alam et al., 2014). The ratio was higher than 0.79, suggesting that detected chlordane was historical and weathered

Table 1
Concentrations range of OCPs and PCBs (μg kg-1 dw) in surface sediments of shrimp ponds of the northern coast of Central Java.

Chemical Family	Compounds	Range of concentration (μg kg^{-1})			
		Brebes	Tegal	Pemalang	Pekalongan
Drins	Aldrin	nd - 4.9 ± 0.3	nd - 3.4 ± 0.4	nd - 26.5 ± 1.4	nd - 0.5 ± 0.1
	Dieldrin	nd - 10.7 ± 0.4	6.1 ± 0.5–13.2 ± 0.5	nd - 2.4 ± 0.3	nd
	Endrin	nd - 8.8 ± 1.2	nd - 36.2 ± 1.2	nd - 13.7 ± 0.6	nd - 4.7 ± 0.8
	Endrin Aldehyde	nd	nd	nd	nd
	Endrin Ketone	0.31 ± 0.1–0.8 ± 0.1	nd	nd - 9.7 ± 0.3	nd - 0.1 ± 0.1
Chlordanes	Heptachlore	nd - 20.7 ± 0.9	nd	nd	nd
	Heptalochlor epoxyde	nd - 1.8 ± 0.3	nd - 0.6 ± 0.1	nd	nd - 0.4 ± 0.1
	Cis-Chlordane	nd - 0.2 ± 0.1	0.2 ± 0.1–0.6 ± 0.1	nd - 0.4 ± 0.1	nd - 0.1 ± 0.1
	Trans-Chlordane	nd - 0.1 ± 0.1	0.1 ± 0.1–0.1 ± 0.1	0.03 ± 0.01–0.1 ± 0.01	nd - 10 ± 0.9
Endosulfan	Endosulfan I	nd - 31.7 ± 1.6	nd	nd - 39.9 ± 1.2	nd- 39.9 ± 3.4
	Endosulfan Sulfate	nd - 4.7 ± 0.7	nd	nd	nd - 1.2 ± 0.3
DDT	4,4′ DDD	nd - 6.7 ± 0.5	nd - 3.1 ± 0.3	nd	nd - 6.7 ± 0.8
	4,4′ DDE	nd - 11.2 ± 0.2	nd - 10.1 ± 0.4	nd	nd
	4,4′ DDT	nd - 2.8 ± 0.3	nd	nd	nd - 0.3 ± 0.1
HCHs	α HCH	nd - 23.1 ± 1.3	nd	nd - 11.2 ± 0.8	nd - 9.4 ± 1.2
	β HCH	3.9 ± 0.2–70.5 ± 1.7	nd	nd - 4.0 ± 0.4	nd - 5.7 ± 0.7
	γ HCH	nd - 7.1 ± 0.9	nd - 7.1 ± 0.4	nd - 7.8 ± 0.9	nd - 3.8 ± 0.6
	δ HCH	nd - 28.3 ± 1.3	nd	nd - 16.8 ± 0.7	nd - 10.5 ± 1.0
Chloroacetanilide	Alachlore	nd - 24.8 ± 1.1	nd - 1.2 ± 0.2	nd - 5.9 ± 0.5	nd
	Metolachlor	nd - 6.5 ± 0.8	nd - 7.1 ± 0.5	nd - 2.8 ± 0.3	nd - 0.9 ± 0.1
Σ Total OCPs		68.1 ± 3.4–168.1 ± 9.8	31.1 ± 6.4–48.4 ± 5.9	45.8 ± 3.5–118.1 ± 6.8	7.8 ± 2.6–51.5 ± 4.1
PCBs	PCB 28	nd - 0.1 ± 0.05	nd	nd	nd
	PCB 52	0.1 ± 0.1–1.9 ± 0.2	nd - 0.1 ± 0.04	0.1 ± 0.02–3 ± 0.2	0.1 ± 0.02–0.5 ± 0.1
	PCB 101	0.1 ± 0.1–0.1 ± 0.1	nd - 0.1 ± 0.03	nd - 0.18 ± 0.04	0.1 ± 0.1–0.3 ± 0.1
	PCB 118	nd - 0.2 ± 0.1	0.2 ± 0.1–2.2 ± 0.3	nd - 0.9 ± 0.1	nd - 0.9 ± 0.2
	PCB 138	nd - 0.2 ± 0.1	nd - 0.1 ± 0.04	0.1 ± 0.02–0.2 ± 0.1	0.1 ± 0.1–0.2 ± 0.1
	PCB 153	nd - 0.2 ± 0.1	nd - 0.14 ± 0.03	nd - 0.2 ± 0.03	0.1 ± 0.1–0.2 ± 0.1
	PCB 180	0.1 ± 0.04–0.2 ± 0.05	0.1 ± 0.03–0.26 ± 0.1	0.1 ± 0.1–0.1 ± 0.05	0.1 ± 0.04–0.1 ± 0.02
Σ Total 7 PCBs		0.7 ± 0.1–2.2 ± 0.3	0.7 ± 0.1–3.6 ± 0.4	0.8 ± 0.1–3.3 ± 0.5	0.6 ± 0.1–2.2 ± 0.2

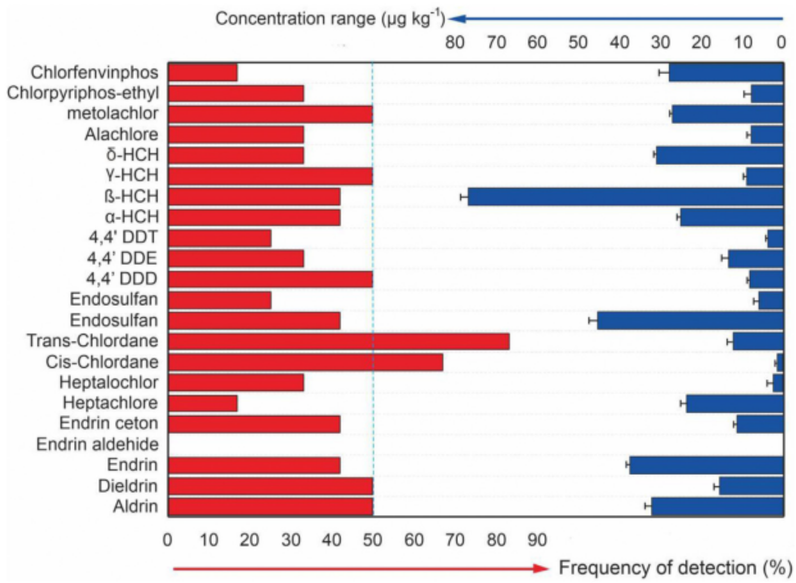

Fig. 2. The occurrence of OCPs in surface sediments of shrimp pond.

residues from commercial sources.

3.4. DDTs

According to Mueller et al. (2011), DDT was widely used for crop and livestock protection from the 1950s, whereas in residential, commercial and industrial area, DDT was applied for mosquito control. In the present study, overall, concentrations of $\sum$ DDTs in the northern part of the Central Java coast were lower than those reported from sediment of shrimp mariculture farm in Hongkong, from 9.95 to 44.4 µg kg-1 with an average of 23.9 µg kg-1 (Wang et al., 2014), but higher than those from sediment of Jincheng Bay mariculture area, China, in a concentration range from 4.1 to 6.7 µg kg-1 (Hu et al., 2015).

Brebes region, which is essentially an agricultural area, presents the highest concentration in DDT, with values ranging between 4.2 ± 0.4–16.3 ± 1.1 µg kg^{-1} dw with an alarming average of about 10.0 ± 6.05 µg kg^{-1} dw. However, these alarming concentrations were lower than those found in agriculture and shellfish farming area of Babitonga Bay, Southern Brazil. In this area, the DDTs were the predominant OCP group, with concentrations ranging from below detection limits to 122 µg kg^{-1} dw (Rizzi et al., 2017).

4,4′-DDE is the main metabolite of DDT detected in sediment samples in Brebes (below detection limit - 11.2 ± 0.2 µg kg-1 dw) and Tegal (below detection limit - 10.1 ± 0.4 µg kg-1 dw). These values are lower than those detected in sediment samples taken in the surrounding areas of shrimp ponds in Jiquilisco Bay, El Salvador, ranging from 3.8 µg kg^{-1} to 30.9 µg kg^{-1} (Nomen et al., 2012). 4,4′ DDE is strongly adsorbed to sediment and is very persistent in aquatic environment. DDT isomers can biodegrade under aerobic conditions to DDE and anaerobic conditions to p,p'-DDD in the environment (Bopp et al., 1982; ATSDR, 2002; Hu et al., 2010; Zhang et al., 2014). Therefore, when DDD/DDE <1, DDT undergoes aerobic biodegradation, and when DDD/DDE> 1, DDT undergoes anaerobic biodegradation (Liu et al., 2008). In sediment samples from Brebes and Tegal, the ratio of 4,4′-DDE, to 4,4′-DDD was smaller than 1, thereby suggesting mainly aerobic degradation.

DDTs ratio was previously used as an indicator of the residence time of DDT in the environment (Olisah et al., 2019). The values of DDT ratios in surface sediments fewer than 1 in Brebes region (Table 2) suggests that the residues are mainly originated from historical usage rather than a recent application. A similar result was recently found in the aquaculture area in Cau Hai lagoon, Vietnam (Tran et al., 2019). It is not possible to calculate the DDT/DDE ratio in Tegal region since the parent pesticide 4,4′-DDT was not detected, but the presence of 4,4-DDE and 4,4′-DDD suggests a lack of recent inputs of the pesticide.

3.5. HCHs (α-, β-, and γ-HCH)

HCHs is present in all samples, whatever the region, with average concentrations of 4.1 ± 3.7 (Pekalongan) to a concerning value of 54.7 ± 18.5 µg kg^{-1} dw found in Brebes. Globally, the composition of HCHs congener profiles in surface sediments was, β-HCH 33.2 ± 7.2%, α-HCH 27.6 ± 3.2%, γ-HCH 13.3 ± 1.1% and δ-HCH 25.9 ± 3.5%. The predominance of the β-isomer can be explained by the environmental transformation of α-HCH and γ-HCH into β-HCH (Maldonado and Bayona, 2002). Moreover, among HCHs isomers, β-isomer is the most persistent in the environment and tends to accumulate in sediments (Doong et al., 2002). Since the β-HCH isomer is relatively resistant with respect to microbial degradation

Table 2
Diagnostic ratios of DDTs, HCHs, and chlordanes in shrimp pond sediments.

Region	Brebes	Tegal	Pemalang	Pekalongan
α-HCH to β-HCH	0.6	NA	2.8	1.7
α-HCH to γ-HCH	4.1	NA	2.7	1.7
β-HCH/HCHs	0.5	NA	0.1	0.2
γ-HCH/HCHs	0.1	1	0.1	0.2
% β-HCH	48.0	0.0	10.9	18.2
% γ-HCH	6.8	100.0	11.3	17.7
CC/TC	3.5	5.0	6.8	7.3
DDT/(DDE + DDD)	0.1	0.0	0.0	0.0

NA: Not Applicable.

and has the lowest volatility, predominance of β-HCH in sediment samples is probably indicative of local contamination by technical HCH (Li, 1999) and/or indicates a historical usage in industry and agriculture (Doong et al., 2002).

Brebes is the region with the highest level of HCHs. It is worth since, beside Pemalang, Brebes is one of the main paddies and onion centers of production in Central Java. It is interesting to note that Zhang et al. (2018) reported that paddy fields are one of possible HCHs source in the Southeast of China. Besides, Wang et al. (2014), who studied for aquaculture-derived OCPs contamination in coastal sediments, showed that aquaculture could cause significant enrichment in OCPs. In mariculture surface sediments (0–5 cm), concentrations of ∑ HCHs and ∑ DDTs were approximately 1.3 and 7.7-fold greater, respectively, than those detected in reference sites. The authors hypothesized that major sources for these enrichments was the use of OCPs contaminated fish feeds.

Around the world, HCH is available in two formulations: technical HCHs and lindane. Technical HCHs contained 60–70% α-HCH, 5–12% β-HCH, 10–15% γ-HCH and 6–10% δ-HCH (α/γ-HCH ranging 3–7), while lindane consists of almost pure γ-HCH (>99%) (Barhoumi et al., 2019; Walker et al., 1999). Concerning the present work, the sources of HCH were evaluated using the isomeric ratios described in material and methods section. The ratio of α-HCH/γ-HCH in Brebes was 4.1, suggesting technical HCHs as the possible source, which is consistent with the percentage of β-HCH (more than 5–14%). This latter result suggested the contamination of shrimp pond sediments by technical HCH. If there were no fresh inputs of technical HCH, β-HCH would be predominant in most sediments. Last, in Brebes, β-HCH was predominant (47%), indicating there is no fresh inputs of technical HCH (Tang et al., 2020). In contrast, the ratio of α-/γ-HCH in Pekalongan and Pemalang was below 4, with α-HCH as predominant isomer. This latter result suggested the use of pure lindane for an agricultural purpose instead of technical HCHs (Tang et al., 2020). It is worthy to note that the production and use of lindane for agricultural purpose has been banned in Indonesia since 2009. These finding results highlight the long-term persistence of these compounds in sediment matrices.

3.6. The occurrence of polychlorinated biphenyl (PCBs)

Based on the recommendations of the International Council for the Exploration of the Sea (ICES) (Webster et al., 2013) and (OSPAR, 2001), this study focused on the set of seven indicator PCB congeners (PCBs 28, 52, 101, 118, 138, 153, and 180) which are used as marker congeners for PCB monitoring and regulation in the USA and Europe. Results for the sum of 7 congeners and individual PCB concentrations (μg kg^{-1} dw) in superficial sediments from shrimp ponds in the northern coast of Central Java are displayed in the bottom of Table 1. Total PCBs concentrations in the different ponds ranged from 0.6 ± 0.1 to 3.6 ± 0.4 μg kg^{-1}, with a grand average of 1.7 ± 1.1 μg kg^{-1}. This concentration of total PCBs was slightly lower than those reported worldwide in other aquaculture sediments, e.g. the concentration of PCBs in surface sediment of aquaculture areas in New Brunswick, Canada (1.07–10.4 ng g^{-1}) (Sather et al., 2006), mariculture area along the coast of Hong Kong and mainland China (5.10–11.0 ng g^{-1}, mean 7.96 ng g^{-1}) (Wang et al., 2011), Xiangshan Bay, China (15.2 μg kg^{-1}) (Lin et al., 2020). Finally, in the present work, the PCB levels in mariculture surface sediments of the shrimp farms were relatively low.

Fig. 3 presents the frequency of detection of the 7 PCBs indicators and PCBs homolog. The PCB homolog distribution indicated that the relative composition of the PCB congeners was dominated by tetra-Cl PCB in Brebes and Pemalang, representing 54%–62% of the total PCBs. Zhang et al. (2008) reported that such homolog was widely present in atmosphere for higher volatility. Furthermore, Totten et al. (2004) and Gioia et al. (2008) demonstrated that PCB concentration levels in the lower atmosphere near the water are sustained by tetra-Cl PCBs volatilized from the upper water layer. Dechlorination of highly chlorinated congeners with anaerobic microbes occurs in the sediments also the possible factor which contributes to the high proportion of light PCBs (Boyle et al., 1992), as reported by Zhong et al. (2020) in the eastern Gaizhou Beach and the coastal area adjacent to Erjie ditch, which is the inputs of PCBs from atmospheric depositions and microbial degradation.

Tegal and Pekalongan have a similar compositional pattern with predominant penta-PCBs (CB101 and CB118) in the shrimp pond sediments. The presence of penta-Cl PCBs represented the commercial PCBs derived from erosion of contaminated soils transported to the aquatic system by surface runoff (Zhong et al., 2020). Those PCB homologs are predominant in the Aroclor 1254 mixtures with an average chlorine content of 54% (IARC and WHO, 2016; USEPA, 2017). The similar pattern was found in Shuangtaizi Estuary, China (Yuan et al., 2015).

A PCA was used to obtain further information on PCB compositions and sources by comparing samples and commercial mixtures (Aroclor 1242, 1248, 1254, 1260). Homolog percent concentrations were used (Frignani et al., 2007) (BMI; GeoChem Metrix. Inc.; U.S.Navy SPAWAR; USEPA, 2017), with a standardization of concentrations was first applied. The score plot obtained from the first two principal components, which together explain 82.4% of the total variance is shown in Fig. 4.

Pemalang and Brebes are grouped around Aroclor 1248 (where tetra-Cl was the most abundant components), while Tegal and Pekalongan showed similarity with Aroclor 1254 (with a significant contribution of penta-Cl). Aroclor 1254 was the technical mixture with the widest range of industrial applications and was the most widely used. Aroclor 1254 was used in electrical equipment like capacitors and transformers, but also as hydraulic fluids, plasticizers, adhesives, wax and pesticide extenders, inks, cutting oils, metal coatings, sealants and caulking compounds (ATSDR, 2000; Wischkaemper et al., 2013).

Tegal is the regency with many metal manufacturing industries, as well as metal smelting industry. Fitriyani et al. (2017) reported that in 2011 there was serious hazardous waste pollution coming from the metal smelting industry in Tegal. The metallurgy industry and smelting process was considered the main sources of dioxin-like PCBs in Korea (Yu et al., 2006) and Portugal (Antunes et al., 2012). Limestone burning industry (cement production) by utilization of used oil in Tegal also reported as one of hazardous waste source in Tegal (DLH and Tegal, 2014). Susanto et al. (2016) characterized used oil on lime production plant, and they found that Aroclor 1254 and Aroclor 1248 were existed. Atmospheric emissions during fuel combustion might also generate PCBs since in the burning process, the used oil was heated on a preheater at 46–70 °C and then injected to the burning oven at 850–960 °C (Susanto et al., 2016). Thus, both of used oil and fuel combustion were assumed to be the other sources of PCBs in Tegal, as reported by Liu et al. (2018) who found that thermal processes (incineration) associated to unintentionally produced PCBs in China.

3.7. Comparison with sediment quality guidelines

The concentrations of targeted compounds were then compared to threshold concentrations from sediment-quality guidelines (SQG) to evaluate the possible ecological risks associated with OCPs (Table 3). The SQG adopted were constituted by Threshold-Effects Level (TEL), Probable-Effects Level (PEL), Effects Range-Low (ERL) and Effects Range-Medium (ERM). The TEL represents the

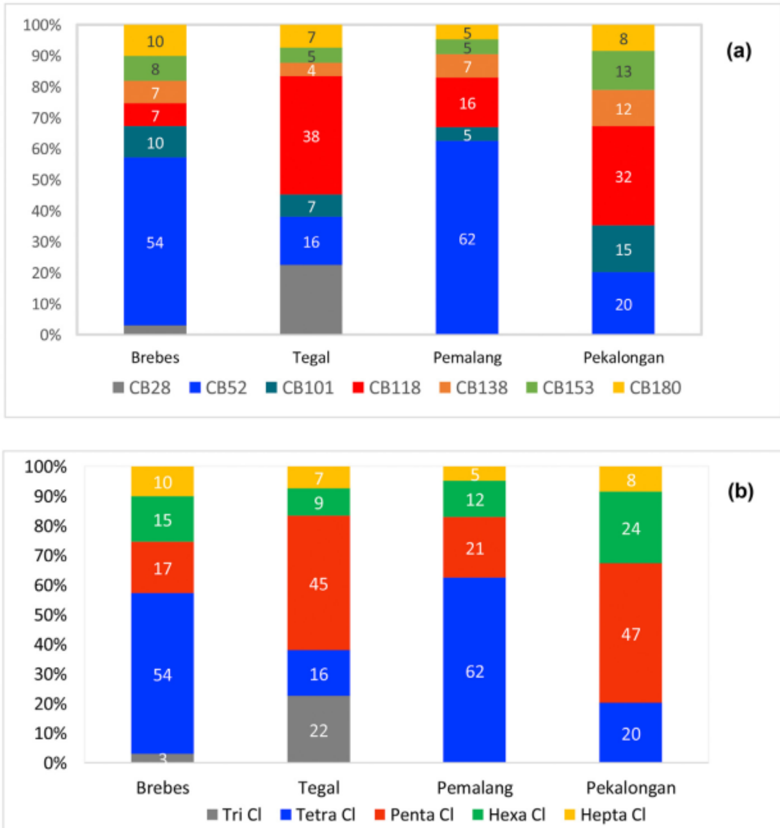

Fig. 3. Proportion (%) of (a) 7 PCBs congeners (CB28, CB52, CB101, CB118, CB138, CB153, and CB180) and (b) PCB homolog (tri-, tetra-, penta-, hexa-, and septa-Cl in sediment samples.

concentration below which adverse biological effects are expected to occur rarely. The PEL represents the level above which adverse effects are expected to occur frequently. The ERL is indicative of concentrations below which adverse effects rarely occur and ERM defines the level above which adverse effects are expected to occur frequently (CCME and Environment, 1999; Long and MacDonald, 1998; Long et al., 1995; Macdonald et al., 1996; NOAA and Administration, 2008).

In Brebes, the area with the highest concentration of POPs (i.e. dieldrin, endrin, 4,4′-DDD and 4,4′-DDT) have the potential to occasionally cause ecological impacts as their average concentrations were greater than TEL, but below PEL. Whereas heptalochlor epoxyde, 4,4′-DDE, and γ-HCH present in the concentration above PEL, indicate these substances could cause adverse ecological impacts. Σ chlordane presents in the concentration above both PEL and ERM in Brebes, suggesting potential ecological risk.

HCH was widely used in agriculture to control crop pests and mainly on rice paddies (Li, 1999). In this study, the ∑ HCHs was defined as the sum of α-HCH, β-HCH, γ-HCH and δ-HCH. As shown in Table 1, the highest concentrations of ∑ HCHs occurred in Brebes Regency, ranged from 34.4 ± 2.3–70.5 ± 6.4 μg kg-1 with a mean of 54.7 ± 18.4 μg kg-1. those values were greater than those concentrations of ∑ HCHs in surface sediments under mariculture facilities in Hongkong, ranged from 5.6 to 20.4 ng g^{-1} with a mean of 13.2 ng g^{-1} (Wang et al., 2014) and Jincheng Bay mariculture area, China with the concentration of total HCHs range from 5.52 to 9.43 μg kg-1 (Hu et al., 2015).

The use of technical HCH had been reduced over the world since technical HCH was substituted by lindane (over 90% is γ-HCH). Among HCHs, γ-HCH (lindane) was found in at least one station of each region, at levels from below detection limits to 7.8 μg kg-1, with a mean concentration (μg kg-1) of 3.7 ± 3.5; 4.1 ± 3.7; 1.8 ± 1.9; 2.6 ± 4.5 respectively for Brebes, Tegal, Pekalongan, and Pemalang. Those values exceeded thresholds of 0.99 μg kg-1 set by various institutions (CCME and Environment, 1999; FDEP, 1994; Long and MacDonald, 1998; Long et al., 1995; Macdonald et al., 1996; NOAA and Administration, 2008). In the light of the findings, contaminations of lindane is still a worrying problem in some aquaculture areas in the northern coast of Central Java.

Concerning DDT, the concentrations of ∑ DDTs from 12 stations ranged from below detection limit to 16.3 ± 1.7 μg kg1, with the highest concentration found again in Brebes Regency, ranged from 4.2 ± 0.9–16.3 ± 1.7 μg kg-1 and a mean of 10 ± 6.1 μg kg-1. Those values were significantly higher than TEL value of 3.89 μg kg-1 and lower than PEL value of 51.7 μg kg-1 set by the Canadian Council of Ministers of the Environment (CCME and Environment, 1999) and Florida Department of Environmental Protection (FDEP, 1994). It indicates that DDT concentration in Brebes Regency has the potential to cause adverse biological effects.

Regarding PCBs, compared to the sediment quality guidelines

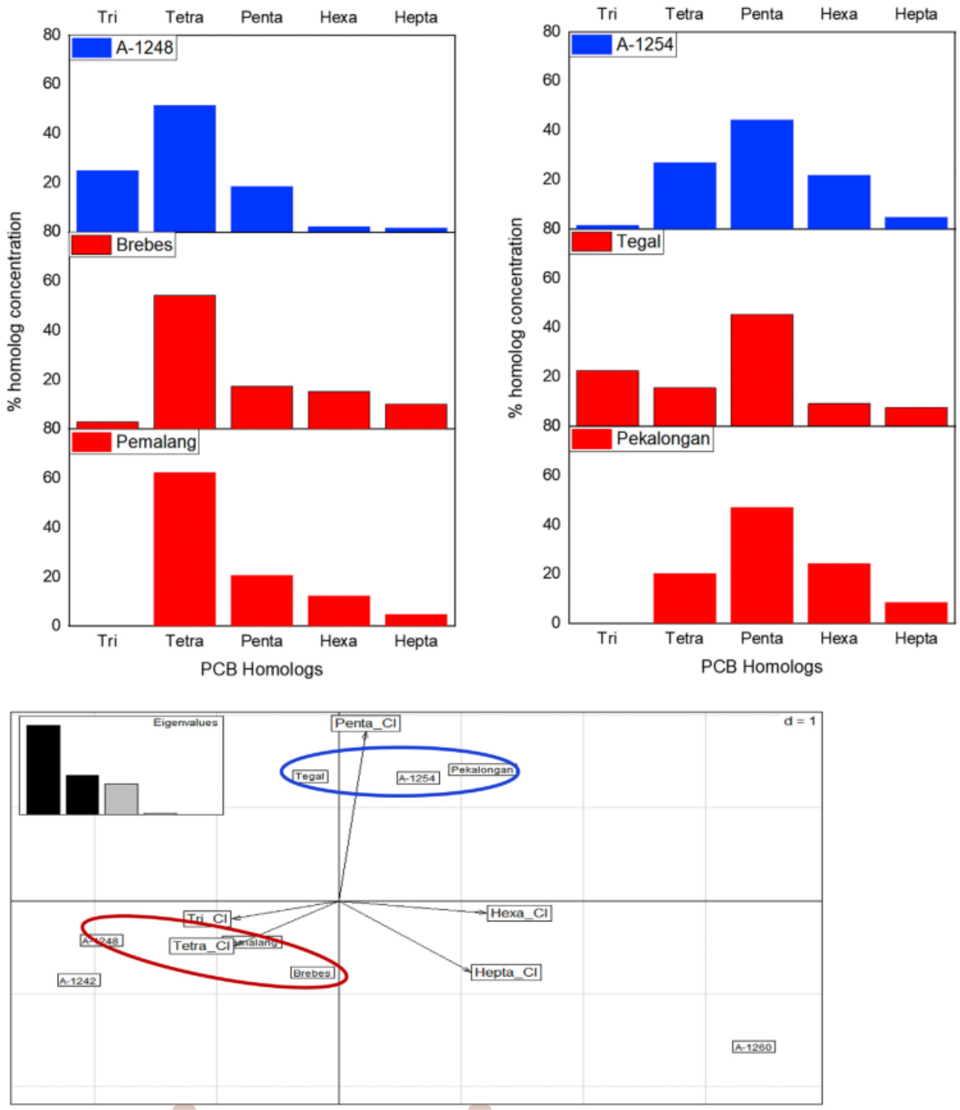

Fig. 4. Comparison of PCB compositions of sediments with those of commercial mixtures.

Table 3
Sediment Quality Guidelines (SQGs) (CCME and Environment, 1999) and the average concentrations of OCPs in surface sediments.

Compounds	SQGs (μg kg^{-1} dw)				Average of Concentration (μg kg^{-1} dw)			
	TEL	PEL	ERL	ERM	Brebes	Tegal	Pekalongan	Pemalang
Dieldrine	0.71	4.3	0.02	8	3.60 ± 6.11	6.43 ± 6.59	n.d.	1.42 ± 1.26
Endrine	2.67	62.4	n.a.	n.a.	2.92 ± 5.07	19.83 ± 18.26	1.59 ± 2.68	4.57 ± 7.92
Heptalochlor epoxyde	0.6	2.74	n.a.	n.a.	8.96 ± 10.6	n.d.	n.d.	n.d.
Σ chlordane	2.26	4.79	0.5	6	9.72 ± 11.79	0.58 ± 0.64	3.59 ± 5.99	0.27 ± 0.21
4,4′ DDD	1.22	7.81	2	20	3.36 ± 3.36	1.22 ± 1.62	0.33 ± 0.32	n.d.
4,4′ DDE	2.07	3.74	2.2	27	5.12 ± 5.63	5.93 ± 5.26	n.d.	n.d.
4,4′ DDT	1.19	4.77	1	7	1.52 ± 1.40	n.d.	0.11 ± 0.19	n.d.
Σ DDT	3.89	51.7	1.58	46.1	10.0 ± 6.05	7.15 ± 5.67	0.44 ± 0.18	n.d.
γ HCH	0.32	0.99	n.a.	n.a.	3.7 ± 3.53	4.09 ± 3.67	1.84 ± 1.89	2.6 ± 4.5
Σ of 7 PCBs	21.6	22.7	189	180	3.9 ± 0.8	6.7 ± 1.4	6.5 ± 1.3	3.7 ± 0.8

TEL (Threshold-Effects Level); PEL (Probable-Effects Level); ERL (Effects Range-Low); ERM (Effects Range-Median); n.a. (No available); n.d. (Not detected).

from Canada (CCME and Environment, 1999), the concentration of ∑7 PCBs were below the concentration which potentially caused an ecological risk. Nevertheless, since the contaminants associated with sediments can accumulate in aquatic organisms (Voorspoels et al., 2004; Zhao et al., 2009), it is essential to monitor POPs level and their potential accumulation in the aquatic organism as well as in the aquatic environment.

Guo et al. (2008) and Liu et al. (2016) evaluate the contamination of OCPs in marine shrimp and the result affirmed that marine shrimp could accumulate the OCPs. Furthermore, Zhang et al. (2014) in their work on bioaccumulation of PCBs in aquatic biota (fish and shrimp) demonstrated a strong bioaccumulation of some OCPs by fish and shrimp. They found biota-sediment accumulation factors (BSAF) values for shrimp of about 0.12 for HCHs, 0.26 for heptachlor, 0.4 for drins (dieldrin and endrin), and 3.4 for p,p'-DDD, and 1.9 for DDTs. Such BSAF values, associated with concentrations found in this study suggest potential accumulated DDTs value of 19.0 μg kg^{-1} dw and HCHs value of 6.84 μg kg^{-1} dw in shrimp in Brebes. By using the conversion factor of 2.8 to convert dry weight to fresh weight (FAO and Statistics, 2004), those values correspond to 6.79 μg kg^{-1} wet weight (ww) of DDTs and 2.44 μg kg^{-1} ww of HCHs.

Jaikanlaya et al. (2009) detected the presence of PCBs in shrimp from the Eastern Coast of Thailand, and they highlighted the levels of PCBs in shrimp was three times higher than that in mussels and oysters. Previously, Tatem (1986) demonstrated that shrimp has an ability in the accumulation of PCBs (specifically Aroclor 1254) from sediment, with biota-sediment accumulation factors (BSAF) value ranged from 0.11 to 0.90 for Aroclor 1242 and 0.20 to 2.40 for Aroclor 1254. McLeese et al. (1980) also reported BSAF value for shrimp exposed to PCB mixed with either a muddy or sandy sediment, ranged from 3.6 to 10.9. A recent research by Lin et al. (2020) showed that the BSAF value of the ∑7 PCBs for shrimp was 8.84 in Xiangshan Bay, China, indicating that the organism was readily subjected to bioaccumulation of PCBs from sediments. Compared to the result of ∑7 PCBs concentration found in this study, the estimated concentration in shrimp will be 19.8 ± 12.7 μg kg^{-1} and 19.3 ± 11.5 μg kg^{-1} in Tegal and Pemalang, respectively. A worrying concentration found for the most toxic of 7 PCBs, i.e. PCB118, in terms of BASF value, since Lin et al. (2020) demonstrated that PCB 118 was more accumulated in organisms, with the value of 111.1. In addition, PCB 118 was detected in all regions, and among PCBs, it presented as the highest detected PCB level in Tegal.

Considering results of this study, it should be noticed since seafood being the one of main source of food for many people in Central Java. Thus, given the high amount of seafood (e.g. shrimp) consumption (20.67 g/person/day) (BPS, 2016), it is essential to monitor the levels of POPs and other dioxin like compounds both in sediments and shrimps and thus validate or invalidate literature BASF for OCPs and PCBs in *L. vannamei*. Thereby, the possible risks for humans of PCBs and related compounds via consumption of local seafood can be determined. In addition, we found that CB118, a mono-ortho-chlorinated PCBs (MO-PCBs), was the second-highest level of PCBs. From a toxicological point of view, most attention is being paid to those PCB congeners that produce a similar toxicity to that of 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD), and they are commonly known as dioxin-like PCBs (dl-PCBs). Considerable dioxin-like toxicity is also attributed to PCB 118, with toxic equivalency factors (TEFs) of 0.00003 set by WHO (Van den Berg et al., 2006).

4. Conclusion

Brebes Regency presents the highest total OCPs concentrations, followed by Pemalang, Tegal, and Pekalongan Regencies. We assumed that paddy and onion production in Brebes maybe the main possible apportionment of pesticides which are intensively used in this region to mitigate the pest, herb or fungi. For other regencies, intensive urban used of pesticides may cause the occurrence of the OCPs. Concerning PCBs, our data showed in different patterns for ∑ 7 PCBs where the highest ∑ 7 PCBs presented in Tegal and Pemalang, followed by Brebes and Pekalongan. The most toxic PCB congener, PCB 118, was detected in all four studied regions (Brebes, Tegal, Pemalang and Pekalongan), with the highest proportion found in Tegal. The congener distribution patterns in these samples indicate the dominance of light chlorinated homologs (tetra- and penta-Cl PCBs). Based on homolog composition analysis, we hypothesized that the presence of PCBs in these regions derived from Aroclor 1248 and Aroclor 1254. In summary, surface sediment contamination in aquaculture system by OCPs and PCBs appeared to potentially cause an adverse effect to the living biota when referencing the sediment quality guidelines (SQGs) approach. Furthermore, we determined the potential accumulated of OCPs and PCBs in the shrimp muscle with the value of about 19 μg kg^{-1} dw for ΣDDTs and 6.8 μg kg^{-1} dw of ΣHCHs in Brebes. Concerning PCBs, we found the highest potential accumulation of Σ 7 PCBs in shrimp muscle of 19.8 μg kg^{-1} dw and 19.3 μg kg^{-1} dw in Tegal and Pemalang, respectively. Even though the results revealed a risk of accumulation, potential level of POPs in shrimp muscle were below the maximum residue limits set by FDA (FDA, 2020).

In the light of above, it should be underlined the importance of continuous and effective POPs monitoring activities. It appears necessary to measure the levels of these pollutants in both sediment and shrimps. This will enable the validation or invalidation of the biota-sediment accumulation factors (BSAF) of the literature. This will finally allow an objective risk assessment for human health for shrimp consumers in turn to take appropriate preventive measures. This will be done soon in a further work.

Credit author statement

Nuning Vita Hidayati: Investigation, Writing - original draft. Laurence Asia: Methodology on sampling and Formal analysis. Imen Khabouchi: Methodology on sampling and Formal analysis. Franck Torre: statistical Formal analysis Conceptualization. Ita Widowati: Supervision. Agus Sabdono: Supervision. Pierre Doumenq: Conceptualization, writing-rewriting, Validation. Agung Dhamar Syakti: Conceptualization, Supervision, writing-reviewing and editing, corresponding authors.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2020.128372.

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