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RESEARCH ARTICLE

Assessment of the ecological and human health risks from metals in shrimp aquaculture environments in Central Java, Indonesia

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Abstract

The occurrence and contamination level of seven important toxic metals (Cd, Cu, Co, Cr, Hg, Pb, and Zn) and three additional metals (Al, Fe, and Mn) in the water, sediment, and shrimp muscle in aquaculture areas located in Central Java, Indonesia, were investigated. The results suggest that the majority of metals have higher concentrations in the inlet followed by the outlet and ponds. Cd dissolved in the waters exhibited the highest level in Pekalongan (3.15 ± 0.33 μg L^{-1}). Although Pb was not detected in the water, it was detected in the sediment, and the concentration ranged from 7.6 to 15.40 mg kg^{-1} dw. In general, the heavy metal concentrations in the sediments were found to decrease in the sequence $Al > Fe > Mn > Zn > Cr > Cu > Co > Pb$. Concentrations below the effects range low level based on the Canadian sediment quality guidelines were found for Cr, Cu, Pb, and Zn, whereas moderate sediment pollution (25–75 mg kg^{-1} dw) was observed for Cr (all regions), Cu (except in the Pekalongan region), and Zn (Brebes and Tegal regions) according to the US EPA standard. The status of the waters was evaluated by calculating a pollution index derived mostly from Mn and Zn. The ecological risk (geoaccumulation index (I_{geo}), contamination factor (CF), pollution load index (PLI), and potential ecological risk index (ERI)) determined in the sediments indicated that all studied areas had low to moderate contamination. The concentrations of all metals in shrimp were generally below the maximum limits for seafood, except for Zn (in all stations), Pb, and Cr (Tegal and Pekalongan). The hazard index values for metals indicated that consuming shrimp would not have adverse effects on human health.

Keywords Sustainable aquaculture · *Litopenaeus vannamei* · Food safety · Trace metals · Pollution index

Introduction

World aquaculture production continues to increase, with shrimp farming representing one of the major aquaculture industries. The shrimp market has become the most valuable globally traded seafood commodity and had the highest production value in 2016. In the same year, FAO data classified Indonesia as the third largest aquaculture producer, with

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production reaching 4,950,000 tons, which accounted for 6.18% of the world's aquaculture production (FAO 2018a). In Indonesia, white-legged shrimp, also known as the king prawn (*Litopenaeus vannamei*), is the dominant farmed species, and it generates the highest revenue on a *per kg* basis compared to the other fishery commodities (KKP 2017).

The expansion and intensification of this sector have emphasized the need for continuous monitoring and control of environmental quality, which is essential for ensuring the sustainability of aquaculture and the safety of the product (Paul and Vogl 2013) based on the 14th indicator of the Sustainable Development Goals Agenda 2030, which aimed to ensure the contribution and conduct of fisheries and aquaculture towards food security and nutrition (FAO 2018b). Good water quality is also essential in aquaculture to maintain the health, optimal growth, and survival of the cultured species (Mohanty et al. 2018; Suantika et al. 2018).

Shrimp aquaculture environments face natural and human impacts from urban and suburban areas. In this study, we focused on seven important toxic metals (Cd, Cu, Co, Cr, mercury (Hg), Pb, and Zn) and three additional metals (Al, Fe, and Mn) related to their transportation. These metals can enter aquatic environments through surface runoff and from natural sources, such as geologic weathering and atmospheric inputs, or they can be derived via effluent discharge from multiple anthropogenic sources, including industrial, agricultural (Gu et al. 2014), mining, shrimp farming (Lacerda et al. 2006), and domestic effluents (Ju et al. 2017; Sarkar et al. 2016).

Heavy metal contamination is becoming a major concern in coastal systems, which are often associated with shrimp farming activity (Syakti et al. 2015), due to the ability of heavy metals to persist in the environment (Ju et al. 2017). Heavy metals are also toxic at high concentrations (Sfakianakis et al. 2015) and could pose risks to human health (Fakhri et al. 2018; Garcia-Hernandez et al. 2018; Zhang et al. 2018), which is why exposure must be evaluated.

In the present study, we monitored the occurrence and evaluated the state of contamination of ten metals (Al, Cd, Co, Cr, Cu, Fe, Mn, Pb, Zn, Hg) in three environmental matrices, i.e., water, sediment, and shrimp, from the aquaculture area in Indonesia, which is located on the northern coast of Central Java in the Brebes, Tegal, Pekalongan, and Pemalang regions. Some studies have reported that aquatic organisms tend to take metals up from their direct environment (Núñez-Nogueira et al. 2012; Wang et al. 2013). For water samples, we investigated the occurrence of metals in the environment and evaluated their contamination and/or status in order to determine whether the quality of the aquatic environment is suitable for intended uses, i.e., aquaculture. Our evaluation based on the extent of dissolved metals in water compared acceptable or safe metal levels via direct comparisons and appropriate indexes. Sediment is a primary location for metal accumulation in the environment and is the main source of contamination in the water column due to resuspension and trapping processes associated with salinity or metal dissolution through biotic/abiotic processes. As a valuable commodity, shrimp muscle tissue must be monitored for contaminants, e.g., heavy metals, because these animals live in water and are in contact with sediments. Several studies have documented the ability of shrimp to accumulate these metals in their tissues since many metals have the ability to form complexes with organic substances (Jonathan et al. 2017; Nascimento et al. 2016; Pourang et al. 2004; Ruelas-Inzunza and Páez-Osuna 2004; Qiu 2015; Zhang et al. 2016). These metals do not tend to be excreted and thus may be passed up the food chain to higher organisms, including humans (Soto-Jiménez et al. 2011). Due to its high toxicity, we also evaluated the levels of Hg in the edible muscles of the shrimp. In this study, we investigated the occurrence of metals in the environment and the contamination status and assessed the potential ecological and human health risks.

Materials and methods

Study area

Sampling campaigns were carried out in shrimp farms in four major areas located on the north coast of Central Java, i.e., the Brebes, Tegal, Pekalongan, and Pemalang regions (Fig. 1). These different regions were selected as the target study areas since they are the highest producers of shrimp in Central Java, which is characterized by water sources from different riverine systems that are among the most important in Indonesia. In each selected area, twenty ponds were selected: 4 ponds in Brebes, 4 in Tegal, 5 in Pekalongan, and 7 in Pemalang. The sample number from each site was determined based on the number of shrimp ponds in each area. *Brebes* Regency is a shrimp farming center in Central Java that covers 14 ha of intensive shrimp farming and 403 ha of extensive shrimp farming. However, shrimp production in Brebes has decreased since 2016 due to environmental pressures, such as industrial waste, agricultural fertilizers, onion/shallot farming activities, industrial activities, and settlements (Bayuaji et al. 2018). It is well documented that there are many problems associated with heavy metal contamination in this area (Ismanto 2019; Hartini 2011).

The shrimp farms in *Tegal* have a lowland topography with upstream rivers flowing into the Java Sea. Tegal has four rivers that pass through 16 villages (59.26%). Similar to Brebes, *L. vannamei* shrimp farms have shown declining shrimp production, and the two largest threats to production are the spread of shrimp white feces disease (0.76) and environmental pollution (0.67). *Pemalang* consists of a 363.82-

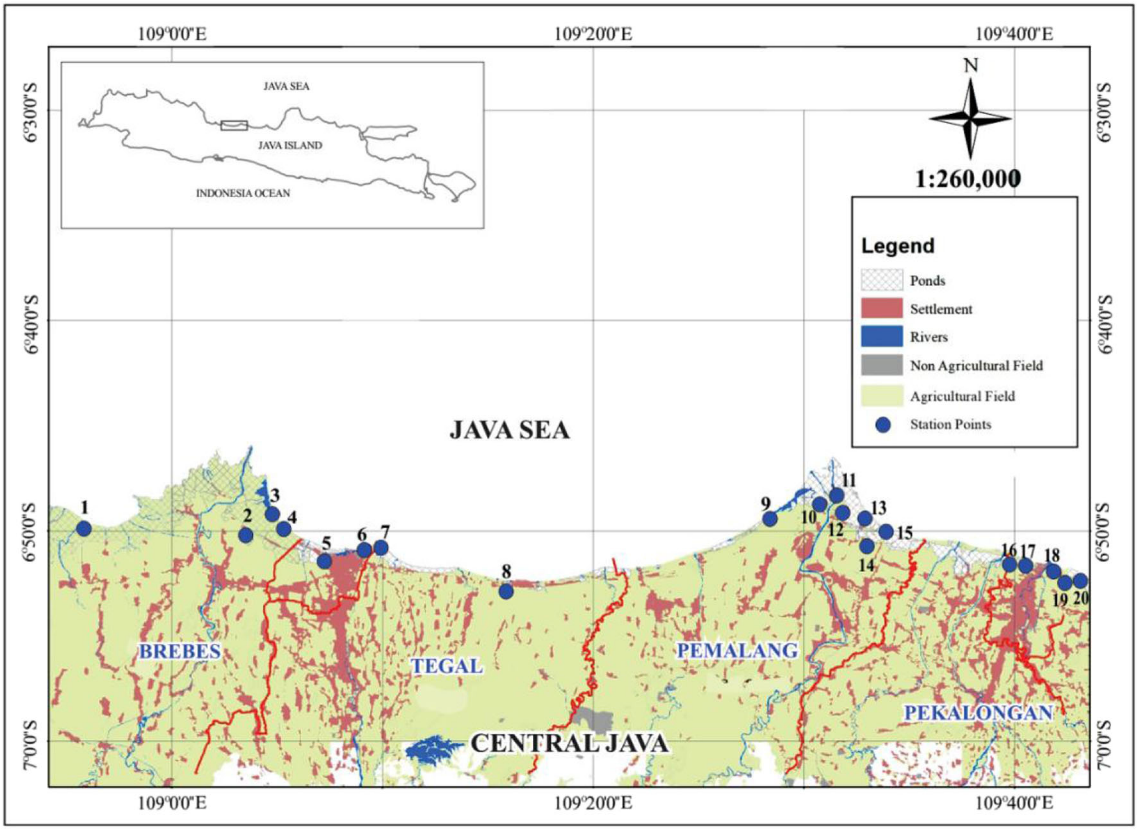


Fig. 1 Study sites. The Brebes (1–4), Tegal (5–8), Pemalang (9–15), and Pekalongan regencies (16–20)

km^2 wetland area and a 751.48-km^2 nonwetland area (BPS 2018c). The population of Pemalang Regency generally lives in coastal areas in city centers, and the number of people living in coastal areas accounts for 57.77% of the total population (Wahyuningrat et al. 2018). The area of farms in this region is 1728 ha and includes milkfish and shrimp farms. Moreover, the fisheries sector is the largest contributor to the gross regional domestic product in this regency (Wahyuningrat et al. 2018). In parallel with the contribution of the shrimp sector to the local economy of the region, there are many problems in local ecosystems caused by anthropogenic activity, including Pb and Cd contamination, which might originate from domestic waste in the upstream areas carried by the Comal River (Puspita et al. 2018; Yulianto et al. 2019) and deterioration of soil and water quality related to mangrove exploitation in this coastal region (Sawitri and Karlina 2006). *Pekalongan* has a coastline along the Java Sea of approximately 6 km stretching from west to east. The coastal area of Pekalongan City has two large river estuaries, namely the Pekalongan River estuary and the Sodetan River estuary (Rachmansyah et al. 2010). In 2017, Pekalongan had a brackish farm area of 379 ha, with 752 aquaculture households in this region (BPS 2018b). More than 1500 small-scale textile industries, namely *batik* industries, have developed rapidly in this region and may represent an important contributor of Cr enrichment in this area (Apriliani and Widiyanto 2018).

Sample collection

Water and sediment samples were collected during the dry season (August 2017), while shrimp samples were collected in both the dry season (simultaneously with water and sediment samples) and wet season (March 2018). In each pond, surface water samples were collected at three different points in the pond, i.e., the inlet (before water enter to the pond), pond (inside the pond), and outlet (after the discharge gates), using polypropylene samplers that were prewashed with 10% nitric acid and then rinsed with distilled water. The water samples from each pond were prepared as composite samples by mixing water from three subpoints at each collection site, and then, they were transferred in acid-washed plastic bottles, placed in an ice box at 4 °C, and transported to the laboratory.

In the laboratory, the samples were filtered using 0.45-μm Whatman mixed cellulose ester (MCE) membrane filters, preserved at 4 °C in 20-mL polyethylene bottles, and acidified with 1% nitric acid (HNO_3) to pH 1.5–2.0 until analysis.

Regarding the sediments from each pond, bulk samples representing the recently deposited material in the shrimp ponds were collected using a 10-L perforated plastic bucket at the pond siphoning system outlet, and they were collected five times with a 15-min interval to generate subsamples. Dewatering was conducted by decantation, and a composite sample for each pond was created by mixing the 5 subsamples in the same quantities. The solid mixed subsamples of sediment/sludge were immediately placed in labeled polyethylene zip bags and stored in a cool box to minimize microbial degradation during transport to the laboratory. The sediment samples were subsequently freeze-dried to a constant weight (approximately 72 h), disaggregated, sieved through 2-mm stainless steel sieves, and then ground using a Retsch RM 200 agate mortar grinder. The sediments were then stored in hermetic plastic vials until analysis.

Shrimp (size 15.02–32.54 g in the dry season and 15.61–31.07 g in the wet season; Electronic Supplementary Material 1, ESM 1) in the intermediate-final stage of the culture cycle (84–110 days of culture (DOC)) were collected using a non-metallic (nylon) net at different points in each pond and then immediately transferred into labeled polyethylene zip bags and stored in a cool box for transport to the laboratory. A composite sample for each pond was created by mixing together the same quantity of each shrimp subsample ($n = 10$). The muscle tissue samples were freeze-dried to a constant weight (approximately 72 h), homogenized with a Retsch RM 1000 ultracentrifugal mill, and stored at − 20 °C until processing for the metal analyses.

Sample digestion and element analysis

The sediment samples were digested using microwave-assisted acid digestion methods: ISO 11466 (1995). Briefly, approximately 0.5 g of the ground freeze-dried sediment sample was digested in a microwave digestion system (Milestone Start D) using a mixture of 7 mL of 37% hydrochloric acid (HCl) (trace metal grade) and 3.5 mL of 65% HNO_3 (trace metal grade) at 200 °C for 40 min. The obtained solutions were transferred into a 50-mL graduated flask, filtered through a 0.45-μm mixed cellulose ester membrane filter, and then stored in high-density polyethylene (HDPE) flasks at 4 °C (Ahmed et al. 2017). The blank and reference material digestion procedures were also conducted as described above.

The shrimp muscles were ground (Retsch ZM 1000 with tungsten blades) to pass through a 0.2-mm titanium sieve. Then, 0.5 g was weighed and placed in a Teflon digestion reactor, to which 3 mL of concentrated HCl and 6 mL of concentrated HNO_3 were added (trace metal grade acids). The samples were then digested in a microwave digestion system (Milestone Start D) and mineralized consecutively at 110 °C for 10 min, 150 °C for 10 min, and 180 °C for 5 min. The supernatant was then transferred into a 25-mL volumetric flask and filtered using a 0.45-μm MCE membrane filter.

Quality assurance and quality control

The method of analysis was validated using Canadian Certified Reference Materials (STSD-3) for the sediments and DORM-4 certified reference material for the shrimp samples (National Research Council of Canada). The accuracy was checked by analyzing the certified reference material under the same conditions. During the analysis, a verification of the stability of the measurement was carried out every 12 to 15 samples through the measurement of a standard or these quality controls (QCs), and the acceptance was carried out for a maximum drift of 5%. The blanks and standard solutions were prepared and included in the measurements to define the background correction and any form of unintended contamination during the sample preparation.

The detection limits ($\mu g\ L^{-1}$) of Al, Cd, Co, Cr, Cu, Fe, Mn, Pb, Zn, and Hg were 3.76, 0.48, 0.99, 6.21, 1.39, 115.36, 0.27, 14.94, 13.38, and 0.45 in the water samples, respectively; 1.43, 0.06, 0.06, 0.21, 0.11, 3.18, 0.12, 9.64, 0.25, and 0.02 in the sediment samples, respectively; and 31.37, 0.11, 0.16, 0.38, 3.93, 3.29, 0.26, 1.10, 1.02, and 0.02 in the shrimp samples, respectively. The limits of quantification ($\mu g\ g^{-1}$) of Al, Cd, Co, Cr, Cu, Fe, Mn, Pb, Zn, and Hg were 12.53, 1.61, 3.31, 20.70, 4,62, 384.53, 0.91, 49.80, 44.60, and 1.43 in the water samples, respectively; 4.77, 0.19, 0.21, 0.71, 0.35, 7.45, 0.39, 11.74, 0.82, and 0.07 in the sediment samples, respectively; and 94.06, 0.36, 0.32, 1.03, 10.50, 10.95, 0.88, 3.68, 3.14, and 0.07 in the shrimp muscles, respectively.

The blank samples containing the acids used for digestion were also processed in the same way. All mineralized samples were then stored at 4 °C in HDPE flasks until analysis. The metal and trace metal concentrations in the water samples and digestion samples (except mercury) were measured using an inductively coupled plasma atomic emission spectrometry (ICP-AES) system (Horiba Jobin Yvon JY2000-2), while mercury was analyzed after reduction and cold vapor generation using flame atomic absorption spectrometry (FAAS) (Thermo Scientific ICE 3000). Quality assurance, quality controls, accuracy, and precision were checked using the standard sediment reference materials (STSD-3, CCRMP-PCMRC, Canada) and fish protein certified reference material (DORM-4, NRC-CNRC, Canada), with accuracies within 100 ± 10%. The low RSD% of the concentration values of each metal did not exceed 10% using the proposed method, thus indicating the method precision.

Occurrence of metals in water bodies and assessment of metal water quality

The most widely used method for calculating contamination and/or pollution indexes is the Nemerow pollution index, which includes a single pollution index for a particular heavy metal (P_i) and a comprehensive pollution index (P_n) (Dadzie et al. 2020; Yan et al. 2016). The single-factor index reflects the contamination degree of every metal, while the P_n reflects the overall effect of all metals on water quality.

The single-factor index method of the Nemerow pollution index (P_i) was calculated using the following equation (Gąsiorek et al. 2017):

$$P_i = \frac{C_i}{S_i}$$

where P_i represents the single-factor index, C_i is the measured concentration of metals in water, and S_i is the standard value for the aquatic body as the organisms' habitat as proposed by different guidelines (ESM 2). The single-factor pollution index is categorized into five grades, where $P_i \leq 1$ represents nonpolluted water, $1 < P_i \leq 2$ signifies slight pollution, $2 < P_i \leq 3$ indicates mild pollution, $3 < P_i \leq 5$ represents moderate pollution, and $P_i \geq 5$ indicates heavy pollution (Dadzie et al. 2020).

The comprehensive Nemerow pollution index reflects the degree of contamination from all the measured metals and is calculated as follows (Brady et al. 2015; Liu et al. 2015; Zhang et al. 2017):

$$P_n = \left[\frac{\max\limits_i P_i^2 + \operatorname{avg}\limits_i P_i^2}{2}\right]^{1/2}$$

where P_n represents the composite index, which indicates the degree of heavy metal pollution in the water; $\operatorname{avg}_{i\epsilon I} P_i^2$ represents the average value of P_i^2; and $\max\limits_{i\epsilon I} P_i^2$ is the maximum value of P_i^2. Liu et al. (2015) categorized the P_n values as follows: $P_n < 0.7$, uncontaminated water; $0.7 \leq P_n \leq 0.7$, under the threat of contamination; and $P_n > 1$, heavily contaminated water.

Assessment of the sediment

Sediment quality guidelines

A comparison of the measured concentrations of various contaminants within the sediments with these guideline values will provide a basic indication of the degree of contamination and likely impact on the ecosystem. In the present study, two sets of sediment quality guidelines (SQGs), i.e., the threshold effects level (TEL), probable effects level (PEL), effects range low (ERL), and effects range median (ERM), from the Canadian sediment quality guidelines were applied to assess the possible risk from metal contamination in the sediment (CCME 2007).

Enrichment factor

The enrichment factor (EF) is a geochemical index used to evaluate a possible source of anthropogenic activities for trace metals using the normalization of metal concentrations by uncontaminated background values (Dickinson et al. 1996). The EF was calculated based on the following equation (Gargouri et al. 2018; Nowrouzi and Pourkhabbaz 2014):

$$\mathrm{EF} = \frac{(C_i/C_{\mathrm{Fe}})_{\mathrm{s}}}{(C_i/C_{\mathrm{Fe}})_{\mathrm{r}}}$$

where EF is the enrichment factor of an element, $(C_i/C_{\mathrm{Fe}})_{\mathrm{s}}$ represents the metal/Fe ratio of the sample concentrations at each sampling point, and $(C_i/C_{\mathrm{Fe}})_{\mathrm{r}}$ is the corresponding element/Fe ratio of the concentrations in the reference sediments. Ideally, the reference sediment values for metals would be estimated from in situ sediments, although in the absence of these data, earth's crust values can sometimes be substituted as shown in Abrahim and Parker (2008). For this study, the average shale concentration of the sedimentary rock in earth's crust (Turekian and Wedepohl 1961) was adopted as the baseline value. In this work, we employed Fe as a conservative element for geochemical normalization of the heavy metal data in the EF calculation because Fe is one of the three conservative elements generally used (Ganugapenta et al. 2018; Pandey et al. 2015). The six contaminants were categorized based on their enrichment factors.

Geoaccumulation index

The geoaccumulation index (I_{geo}) has been widely used to evaluate the degree of metal contamination or pollution in terrestrial, aquatic, and marine environments (Kabir et al. 2011; Muller 1969) and is calculated using the following equation:

$$I_{\mathrm{geo}} = \log_2\left[\frac{C_i}{1.5B_i}\right]$$

where C_i is the measured concentration of the examined metal (i) in the sample and B_i is the background concentration of the corresponding metal (i). In this study, we used the following as the background concentrations: 17 mg kg^{-1} for Co and Pb (McLennan 2001), 35 mg kg^{-1} for Cr, 25 mg kg^{-1} for Cu (Ranjan et al. 2018), 56,000 mg kg^{-1} for Fe (Taylor 1964), 292.2 mg kg^{-1} for Mn (Xavier et al. 2017), and 59.11 mg kg^{-1} for Zn (Yang et al. 2018). A factor of 1.5 was applied to the geochemical background concentration to minimize the possible variations in the background value of a given metal due to terrigenous effects (Lizárraga-Mendiola et al. 2008; Lu et al. 2009).

Contamination factor and pollution load index

The contamination factor (CF) is used to determine the contamination status of a sediment and is the ratio of the concentration of a metal in the sediment sample to that in the background. The CF was calculated using the following equation

(Hakanson 1980):

$$\mathrm{CF} = \frac{C_{\mathrm{metal}}}{C_{\mathrm{background}}}$$

The CF values were categorized as follows: $\mathrm{CF} < 1$, low degree of contamination; $1 \leq \mathrm{CF} < 3$, moderate degree of contamination; $3 \leq \mathrm{CF} \leq 6$, considerable degree of contamination; and $\mathrm{CF} > 6$, very high degree of contamination (Satapathy and Panda 2015).

To estimate the overall pollution status of the samples, the pollution load index (PLI) has also been used. The PLI of a set of n polluting elements is defined as a value calculated from the geometric mean of the respective contamination factors of those elements using the following equation (Qing et al. 2015):

$$\text{PLI station} = \sqrt[n]{\mathrm{CF1} \times \mathrm{CF2} \ldots \times \mathrm{CF}n}$$

where CF represents the contamination factor of a metal and n represents a specific metal's contamination factor. The PLI values were used to classify samples as unpolluted ($\mathrm{PLI} \leq 1$), moderately polluted ($\mathrm{PLI} = 1$–3), highly polluted ($\mathrm{PLI} = 3$–5), or very highly polluted ($\mathrm{PLI} > 5$).

To determine the contamination status in the overall zone studied, we used the following relationship:

$$\text{PLI zone} = \sqrt[n]{\text{PLI station } 1 \times \text{PLI station } 2 \ldots \times \text{PLI station } n}$$

Potential ecological risk index and integrated ecological risk index

The potential ecological risk of each metal was calculated by the Hakanson equation as previously described by Wu et al. (2017)

$$E_{\mathrm{r}}^{i} = T_{\mathrm{r}}^{i} \times C_{\mathrm{f}}^{i}$$

where E_{r}^{i} is the coefficient of potential ecological risk from metal i, T_{r}^{i} is the toxicity coefficient of metal i, and C_{f}^{i} is the accumulation coefficient (contamination factor) of metal i. The toxic response factors used were as follows: $\mathrm{Cu} = \mathrm{Pb} = 5$, $\mathrm{Cr} = 2$, $\mathrm{Zn} = 1$, $\mathrm{Co} = 5$, and $\mathrm{Mn} = 1$ (Hakanson 1980).

To evaluate the overall ecological risk status (integrated potential ecological risk index (RI)) of the sediment, the following equation is widely used:

$$\mathrm{RI} = \sum_{i=1}^{n} E_{\mathrm{r}}^{i}$$

where RI is the integrated potential ecological risk factor for the metal in sediments. The terminology used to describe the ecological risk index is as previously defined (Hakanson 1980).

Occurrence and human health risk assessment of the shrimp

Estimated daily intake

The estimated daily intake (EDI) of the different metals was evaluated based on the tissue concentrations of each metal in the shrimp and the average daily shrimp ingestion rate (Santos et al. 2004). The EDI represents the estimated daily intake of metal through the consumption of shrimp by an adult human (in mg kg^{-1} day^{-1}). The following relationship was used to calculate the EDI (Liu et al. 2018):

$$\mathrm{EDI} = \frac{\mathrm{EF} \times \mathrm{ED} \times \mathrm{IR} \times C_{m}}{\mathrm{WAB} \times \mathrm{TA}}$$

where EF is the exposure frequency (365 days year^{-1}), ED is the exposure duration (70 years, equivalent to the average life span as used by Anandkumar et al. (2018), Liu et al. (2018), and Keshavarzi et al. (2018)), IR is the ingestion rate (g day^{-1}), C_{m} is the concentration of metal in the shrimp (mg kg^{-1} dw), WAB is the average adult body weight (kg), and TA is the average lifetime (70 years × 365 days year^{-1}, i.e., 25,550). The daily consumption rate was set at 20.67 g person^{-1} day^{-1} as per the report of the Statistics Indonesia database for the fish and shrimp rates of consumption (BPS 2016), while the average adult body weight in Asia (including Indonesia) was assumed to be 57.7 kg (Walpole et al. 2012). A value of 4.54 was used as the conversion factor to convert the dry weight to wet weight based on a shrimp moisture content of 78% (Qiu et al. 2017).

Target hazard quotient

The target hazard quotient (THQ) is defined as the maximum tolerable daily intake of a specific metal that does not result in any deleterious health effects. In the present study, we used the THQ to assess the human health risk level due to pollutant exposure from consuming metal-contaminated shrimp. The THQ was calculated as the ratio of the average daily intake of a specific chemical over a lifetime to the oral reference dose (RfD) of the trace metal. This metric is an estimate of the daily oral exposure by a human that does not have an appreciable risk of deleterious effects during a lifetime (USEPA 2002).

The following equation was used to determine the THQ (Anandkumar et al. 2018; Liu et al. 2018):

$$\mathrm{THQ} = \frac{\mathrm{EDI}}{\mathrm{RfD}}$$

Oral RfDs (mg kg^{-1} day^{-1}) of 0.001, 0.14, and 0.3 were used for Cd, Mn, and Zn, respectively, and these values were based on US EPA guidelines (USEPA 2018): 0.03 was used for Co (Finley et al. 2012), 0.003 was used for Cr (Liu et al.

2018; Zhao et al. 2016; Yabanli and Alparslan 2015), 0.02 was used for Cu (Ahmed et al. 2015), 0.004 was used for Pb, and 0.0003 was used for Hg (Yabanli and Alparslan 2015); these values were used to assess the exposure hazards from shrimp consumption. THQ values of < 1 indicate that adverse health effects are unlikely to occur, and THQ values of > 1 indicate that the consumer population has potential health risks.

Hazard index

Since exposure is usually not associated with a single toxicant, the hazard index (HI) was developed from the THQs and expressed as the sum of the hazard quotients (USEPA 2018). The hazard index derived for the shrimp was determined according to the following equation:

$$\mathrm{HI} = \sum_{i=1}^{n} \mathrm{THQ}_i$$

where THQ_i is the target hazard quotient of an individual metal and HI is the total hazard index for all metals. In the present study, HI is the total hazard index for eight metals (Cd, Cr, Co, Cu, Mn, Pb, Zn, Hg). $\mathrm{HI} > 1$ indicates a potential for an adverse effect on human health.

Carcinogenic risk

The carcinogenic risk was estimated as the incremental probability of an individual developing cancer over a lifetime of exposure to a potential carcinogen by employing a target cancer risk value (USEPA 1989). The carcinogenic health risks related to the consumption of seafood were measured based on the target cancer risk (TR), which was calculated as follows:

$$\mathrm{TR} = \mathrm{EDI} \times \mathrm{CSFo} \times 10^{-3}$$

where CSFo is the oral carcinogenic slope factor from the Integrated Risk Information System database. For Cr and Pb, the slope values were 0.5 mg kg^{-1} day^{-1} and 0.0085 mg kg^{-1} day^{-1}, respectively. According to the New York State Department of Health (NYSDOH 2007), the threshold TR values for categorizing risk are as follows: $\mathrm{TR} \leq 10^{-6}$, low risk; 10^{-4} to 10^{-3}, moderate risk; 10^{-3} to 10^{-1}, high risk; and $\geq 10^{-1}$, very high risk.

Statistical analysis

A principal component analysis (PCA) was applied to the results to establish the significance at a probability of 95% for the spatial and temporal variations in the metal concentrations in the water, sediments, and shrimp. The possible relationship between the different variables was estimated by determining the correlation coefficients. A high correlation coefficient implies a strong relationship between variables, and although this value is not a measure of quantitative changes of one variable with respect to the other, it is a measure of the intensity of the association between the two variables. All statistical analyses were performed using R statistical software.

Results and discussion

The selected shrimp farms were originally constructed based on single pond operators in which the operations were modified to include reservoirs to treat water from supply canals (river, coastal, or piped from a well) before use. However, the system had no wastewater treatment system for effluents discharged from the culture ponds during operation and harvest. Generally, the ponds were arranged in rows, with discharge canals on opposite sides. The farms were operated with a pond size of 1000–3000 m^2, a shrimp density of 80–150 m^{-2}, a culture period of 84–110 days, a feeding period for 4–5 times, a feed conversion ratio (FCR) of 1.2–1.8, and a shrimp size of 15.02–32.54 g piece^{-1}. The operation applied a 3–10% water exchange periodically. The technology used for white shrimp culture consists of middle (semi-intensive) and high (intensive) levels. The shrimp farm characteristics and management activities during this study are summarized in ESM 3.

Occurrence of metals in the water and assessment of metal water quality

The concentrations of metals observed in the water samples are displayed in Fig. 2. The metal concentrations revealed that Zn and Fe were the dominant metals in shrimp ponds in Brebes and Tegal, with the concentrations ranging from 49 to 681 μg L^{-1} and from 38 to 169 μg L^{-1} for Zn, respectively, and from 68 to 279 μg L^{-1} and from 35 to 404 μg L^{-1} for Fe, respectively. Mn and Fe were also the dominant metals in Pekalongan and Pemalang, and the concentrations ranged from 82 to 537 μg L^{-1} and from 123 to 2939 μg L^{-1} for Fe, respectively, and from 48.8 to 1428.7μg L^{-1} and from 39.8 to 1336.8 μg L^{-1} for Mn, respectively. In the present study, Pb was not detected in the water samples from any of the studied areas (LOD of 14.94 μg L^{-1}). The Zn concentrations showed similar trends in all regions and increased from the inlets to the ponds and eventually decreased from the ponds to the outlets. These trends indicated the application of metals in aquaculture systems.

One possible source for various metals is from foods enriched by some metals. Katya et al. (2016) investigated the content of trace minerals of the commercial sources of diets applied to *L. vannamei* and found levels of Zn in the

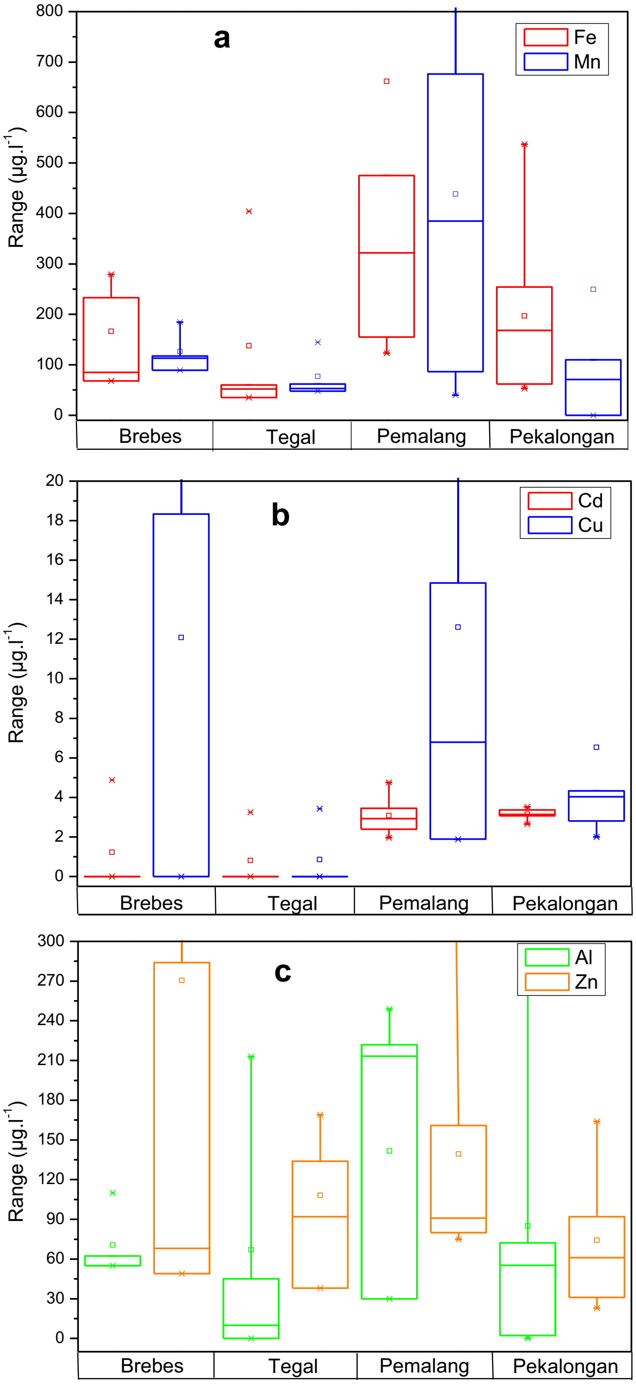


Fig. 2 Metal concentrations (µg L^{-1}) of **a** Fe and Mn, **b** Cd and Cu, and **c** Al and Zn in the surface water of shrimp ponds on the northern coast of Central Java

concentration of 29.3–182 ppm. Another study conducted by León-Cañedo et al. (2017) found that the main source of Cu and Zn metals in a simulated culture system with *L. vannamei* was the diet, which accounted for 91.8% of Cu and 97.0% of Zn. Zn is known as an essential nutrient for many physiological functions, such as growth, development (Watanabe et al. 1997), and immune function (Rink and Kirchner 2000). Similarly, Lin et al. (2013) found that zinc significantly reduced the cumulative mortality of *L. vannamei* against *Vibrio harveyi*, indicating an enhancement of the immune system and hence an improved resistance of the shrimp. Zn intake could reduce Cd toxicity; thus, the presence of zinc in water plays an important role for shrimp (Brzóska and Moniuszko-Jakoniuk 2001).

Mn is ubiquitous in the environment and accounts for approximately 0.1% of earth's crust, and it can also be released into water by discharge from aquaculture activities and industrial activities and the combustion of fossil fuels. Katya et al. (2016) found Mn as one of trace minerals in the commercial diet sources for *L. vannamei*, and the concentration ranged from 25.9 to 132.2 ppm. Mn is essential at low concentrations but becomes lethal at higher concentrations (Srichandan et al. 2016). Municipal wastewater discharges, sewage sludge, and emissions from alloys are additional sources of Mn in the environment (WHO 2004). The operation of some metallurgical industries may contribute to Mn contamination in the water in this study area, since this industry potentially releases Mn into the environment (Rahman et al. 2014). Some metallurgical industries are established in Brebes (BPS 2018a), Tegal (BPS 2018d), and Pekalongan (BPS 2018b).

In the case of Cu, our results showed that Cu concentrations (µg L^{-1}) generally increased from 1.20, 2.84, and 2.63 in the inlets to 1.22, 3.15, and 3.08 in the ponds in Brebes, Pekalongan, and Pemalang, respectively (ESM 4). These results were slightly higher than those of Wang et al. (2014), who found that the Cu concentration ranged from 0.52 ± 0.33 to 2.49 ± 0.73 µg L^{-1} in Maluan Bay, China, which is the habitat of *L. vannamei* (Wang et al. 2014). The concentrations were also lower than the concentrations that may cause mortality and induce adverse shrimp physiological responses. Frías-Espericueta et al. (2008b) determined the survival and possible histological alterations in the gills and hepatopancreas of *L. vannamei* juveniles exposed to copper sulfate and found that the mortality of shrimp juveniles during 96 h of exposure was 50% (96-h LC$_{50}$) at a concentration of 35.12 mg L^{-1}, whereas the absence of pillar cells, loss of regular structure of the cuticular epithelium, and multifocal necrosis were observed after 3 weeks of exposure to a concentration of 1.756 mg L^{-1}. Cu-related histological alterations were also reported by Soegianto et al. (2013) and Qian et al. (2020) in *L. vannamei* juveniles. Soegianto et al. (2013) found that after 15 days of exposure to 0.675 mg L^{-1}, tissue hyperplasia was evident in the filaments, which resulted in a narrowing of the hemolymphatic lacunae. Furthermore, necrosis and loss of hemolymphatic lacunae were observed in the gills of shrimp exposed to 1.325 mg L^{-1} and 2.010 mg L^{-1}. A study by Qian et al. (2020) indicated that Cu concentrations above 0.2 mg L^{-1} can decrease growth performance, Cu^{2+} concentrations above 0.5 mg L^{-1} damaged the hepatopancreas histology, and Cu^{2+} concentrations above 0.5 mg L^{-1} can reduce the hemolymph immunologic function of *L. vannamei*. Our result is also below the concentration of metal mixtures (Cd, Cu, Fe, Mn, Pb, and Zn) that might cause histological and structural damages to the gills, epipodites, hepatopancreas, and midgut of *L. vannamei* juveniles (Frías-Espericueta et al. 2008a).

Boyd and Massaut (1999) reported that some trace metals are present as natural components in feeds, impurities in fertilizers, or active substances in pesticides. Katya et al. (2016) analyzed the content of trace minerals of the traditional inorganic and commercial sources of diets applied to *L. vannamei* and showed that the Cu concentration ranged from 12.8 to 185 ppm. Yuan et al. (2019) reported that Cu in copper sulfate and chelated copper compounds are very effective algicides, which are applied to ponds to reduce the abundance of nuisance aquatic plants. Boyd and Massaut (1999) reported that some trace metals are present as natural components in feeds, impurities in fertilizers, or active substances in pesticides. Katya et al. (2016) analyzed the content of trace minerals of the traditional inorganic and commercial sources of diets applied to *L. vannamei* and showed that the Cu concentration ranged from 12.8 to 185 ppm. Cu was also presented in the chemical additives (lime and chloride) applied to shrimp farms at concentrations ranging from 1.9 to 3.3 μg g^{-1} dw (Lacerda et al. 2006). However, they concluded that aquafeeds are, by far, the largest contributor of Cu to shrimp ponds due to the higher Cu concentrations and the larger amount used. The total Cu load from aquafeeds and other products applied to ponds contributed 194.5 g ha^{-1} $cycle^{-1}$.

It is well documented that in aquaculture systems of *L. vannamei*, dietary Cu (along with zinc) plays an important role as a promoter of growth and immune function (Cheng et al. 2014). Davis et al. (1993) showed that the weight of shrimp increased in response to copper supplementation, indicating a dietary copper requirement for shrimp growth and survival. Furthermore, Yuan et al. (2019) demonstrated that dietary chelated copper is effective at improving oxidation resistance and nonspecific immunity of *L. vannamei*.

To evaluate the metal contamination, the single-factor pollution index (P_i) and the Nemerow pollution index (P_n) were calculated, and the results are shown in Table 1, ESM 5, and ESM 6. A comparison with the grades of the P_i showed that the surface water of shrimp farms was generally contaminated by Mn and Cd. The P_i and P_n have been broadly used to evaluate the heavy metal contamination in water and sediment. For example, Zhong et al. (2015) employed these

Table 1 Single-factor pollution index for six metals in the surface water of shrimp farm

Region	sub-site	min & max	Range of Pi value											
			Cd	status	Cr	status	Cu	status	Mn	status	Pb	status	Zn	status
Brebes	Inlet	min	0.00	np	0.00	np	0.32	np	5.60	hp	0.00	np	0.50	np
		max	4.80	mop	0.00	np	0.89	np	10.07	hp	0.00	np	4.04	mop
	pond	min	0.00	np	0.00	np	0.00	np	0.89	np	0.00	np	0.98	np
		max	4.88	mop	0.00	np	3.75	mop	1.85	sp	0.00	np	13.62	hp
	outlet	min	0.00	np	0.00	np	0.60	np	7.99	hp	0.00	np	0.66	np
		max	4.27	mop	2.02	mp	2.47	mp	34.33	hp	0.00	np	2.84	mp
Tegal	Inlet	min	0.00	np	0.00	np	0.00	np	0.16	np	0.00	np	0.88	np
		max	4.48	mop	0.00	np	0.30	np	5.31	hp	0.00	np	1.30	sp
	pond	min	0.00	np	0.00	np	0.00	np	0.48	np	0.00	np	0.76	np
		max	3.25	mop	0.00	np	0.43	np	1.45	sp	0.00	np	3.38	mop
	outlet	min	0.00	np	0.00	np	0.00	np	0.90	np	0.00	np	0.84	np
		max	3.44	mop	0.00	np	0.20	np	27.30	hp	0.00	np	3.70	mop
Pemalang	Inlet	min	1.56	sp	0.00	np	0.00	np	0.09	np	0.00	np	0.00	np
		max	3.93	mop	0.00	np	2.08	mp	42.19	hp	0.00	np	5.40	hp
	pond	min	1.96	sp	0.00	np	0.24	np	0.40	np	0.00	np	1.50	sp
		max	4.76	mop	0.00	np	5.68	hp	13.37	hp	0.00	np	7.64	hp
	outlet	min	0.00	np	0.00	np	0.00	np	0.57	np	0.00	np	0.44	np
		max	5.09	hp	0.00	np	4.19	mop	23.03	hp	0.00	np	5.74	hp
Pekalongan	Inlet	min	2.20	mp	0.00	np	0.29	np	0.35	np	0.00	np	0.60	np
		max	3.30	mop	0.00	np	0.51	np	2.42	mp	0.00	np	1.48	sp
	pond	min	2.65	mp	0.00	np	0.25	np	0.49	np	0.00	np	0.46	np
		max	3.52	mop	0.00	np	2.44	mp	14.29	hp	0.00	np	3.28	mop
	outlet	min	0.63	np	0.00	np	0.00	np	0.52	np	0.00	np	0.28	np
		max	3.34	mop	0.00	np	0.57	np	5.44	hp	0.00	np	2.10	mp

np nonpollution (green), *sp* slight pollution (pink), *mp* mild pollution (yellow), *mop* moderate pollution (blue), *hp* heavy pollution (red) two risk assessment models to determine the heavy metal contamination in groundwater for drinking purposes, Dadzie et al. (2020) applied these indexes to determine the contamination status of water from lake and rivers, and Zhang et al. (2017) used these indexes to evaluate the heavy metal contamination of marine reserve waters. Furthermore, many authors have used the P_i and P_n indexes to evaluate heavy metal contamination in sediment (Asare-Donkor et al. 2018; Yavar Ashayeri and Keshavarzi 2019; Mirza et al. 2019; Yan et al. 2016). In the present study, we applied P_i and P_n to assess the contamination level of metals in water for shrimp as a brackish aquatic organism, and S_i is the standard value for aquatic organisms.

Kowalska et al. (2018) stated that P_i and P_n can be used to highlight which heavy metal represents the highest threat at the site. In addition, since different heavy metals may have impacts on one site, Yan et al. (2016) stated that the P_n index could provide a reasonable interpretation of the heavy metal contamination at each site as a whole.

With regard to the health of the shrimp ponds, for the shrimp *L. vannamei*, Boyd (2009) and Prapaiwong and Boyd (2014) have proposed safe and acceptable metal levels. Our study showed that metal contamination was within the safe or acceptable levels for the shrimp ponds' carrying capacities. However, Cu and combinations of various metals may represent a hazard to the juvenile shrimp (5–6.5 cm), and a previous study showed that histological effects may be observed at 2.5 μg L^{-1} of Cu (Fig. 2) (Frías-Espericueta et al. 2008a, b); unfortunately, such analyses are beyond the scope of our study.

Occurrence of metals in the sediment

The maximum, mean ± SD, and minimum concentrations (mg kg^{-1} dw) of the metals in the sediments as well as a comparison with the SQGs are provided in Table 2. The metal concentrations in the shrimp pond sediment showed that Cr, Cu, Pb, and Zn were below the ERL based on the Canadian sediment quality guidelines, which were 81, 34, 46.7, and 150 mg kg^{-1} dw for Cr, Cu, Pb, and Zn, respectively. According to the US EPA standard, the sediment was moderately polluted (25–75 mg kg^{-1} dw) by Cr (all regions), Cu

Table 2 Comparison between the sediment quality guidelines (SQGs) and metal concentrations (mg kg^{-1} dry weight) in the shrimp pond sediment

Region	Cr	Cu	Pb	Zn
Brebes				
Mean	55.5 ± 0.8	31.8 ± 4.2	13.9 ± 1.2	93.7 ± 1.6
Min	54.7	27	12.2	91.6
Max	56.4	37.2	15	95.3
Tegal				
Mean	46.6 ± 4.53	51.825 ± 9.11	14.1 ± 1.11	99 ± 4.86
Min	39.9	40	12.7	92.3
Max	49.8	61.3	15.4	103.8
Pekalongan				
Mean	32.48 ± 0.9	21.08 ± 3.1	9.11 ± 2.04	75 ± 8.24
Min	31.2	19.4	7.6	70.8
Max	33.6	26.6	12.7	89.7
Pemalang				
Mean	38.16 ± 6.51	27 ± 3.32	9.84 ± 1.65	76 ± 8.02
Min	28.5	22	8.3	67
Max	48	33.3	12.9	90
Canadian sediment quality guideline				
ERL	81	34	46.7	150
ERM	370	270	218	410
TEL	52.3	18.7	30.2	124
PEL	160	108	112	271
US EPA sediment quality guideline				
Not polluted	< 25	< 25	< 40	< 90
Moderately polluted	25–75	25–50	40–60	90–200
Heavily polluted	> 75	> 50	> 60	> 200

(except in the Pekalongan region), and Zn (Brebes and Tegal regions).

Enrichment factor

The EF is generally employed to explain the contribution of metals other than those of lithogenic origin and to assess the degree of anthropogenic influence. The EFs of the metals in the sediments of the study area are displayed in ESM 7. The highest EF value was recorded for manganese in all regions (indicating *considerable enrichment* for Pemalang), while no enrichment of Pb was observed in any region. The EF value of Zn was from 1 to 2, indicating a *moderate* enrichment by this element. The EF value can also be used to differentiate between crustal and noncrustal sources of trace metals. Metals are considered to originate from the crust when the EF is between 0.5 and 1.5 and from an anthropogenic source when the EF is more than 1.5 (Zhang and Liu 2002). The EF value for the Cr in Brebes was above 1.5, suggesting an anthropogenic source of Cr in this region, while in the other three regions, Cr came from natural sources. The EF values of Pb and Cu were lower than 1.5 in all studied areas, indicating that these metals originated from the crust. In contrast, Mn was considered to have originated from anthropogenic sources in all regions except Pekalongan.

Geoaccumulation index

According to Muller (1969), the metal contamination levels were classified into seven levels: uncontaminated ($I_{geo} \leq 0$), uncontaminated to moderately contaminated ($0 < I_{geo} \leq 1$), moderately contaminated ($1 < I_{geo} \leq 2$), moderately to heavily contaminated ($2 < I_{geo} \leq 3$), heavily contaminated ($3 < I_{geo} \leq 4$), heavily to extremely contaminated ($4 < I_{geo} \leq 5$), and extremely contaminated ($I_{geo} = 5$) in terms of I_{geo}.

As shown in Table 3, almost all metals had I_{geo} index values of less than 0, which suggested that the sediments in the study area were not polluted by these metals. There was no contamination by Mn in Pekalongan, the sediments in Brebes and Tegal were uncontaminated to moderately contaminated, and those in Pemalang were moderately contaminated.

An I_{geo} value of less than 0 indicates a lithogenic origin, while a positive I_{geo} index indicates the enrichment of metals of nonlithogenic origin. The results suggest that Mn was generally enriched by anthropogenic sources while the other metals were of lithogenic origin.

Contamination factor and pollution load index

Based on the CF value (ESM 8), Fe and Pb showed a low degree of contamination in all regions and Mn contaminated the sediment in all regions (moderate degree of contamination in Brebes, Tegal, and Pekalongan and considerable degree of contamination in Pemalang). The PLI value of Pekalongan was less than 0, indicating a low degree of contamination, while that of Brebes, Tegal, and Pekalongan indicated a moderate degree of contamination, with values of 1.11 ± 0.08. The PLI indicated a moderate degree of contamination of the study zone (Fig. 3).

Potential ecological risk index

The potential ecological risk index for metal was proposed by Hakanson (1980) to evaluate the potential ecological risk posed by the metals in sediments based on the toxicity of heavy metals and the response of the environment. ESM 9 shows the value of the ecological risk factor (E_r^i) and the RI of the sediment. Generally, the highest RI value among the trace metals in the sediments was that of Cu followed by that of Pb, although both still indicated low potential ecological risk.

Occurrence of metals in the shrimp

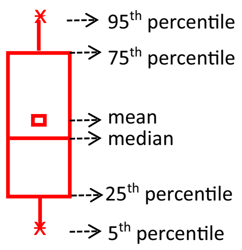
Generally, there were significant differences ($p > 0.05$) in the heavy metal concentrations (dry weight) in the whole body tissue of shrimp of different sizes. The pattern of metal

Table 3 Geoaccumulation index (I_{geo}) for the metal concentrations at the 20 stations

Region	The geo-accumulation index (I-geo)					
	Co	Cr	Cu	Mn	Pb	Zn
Brebes	-0.76±0.03	-0.73±0.19	-0.24±0.19	0.74±0.26	-0.88±0.13	-0.18±0.02
Tegal	-0.77±0.014	-0.04±0.27	0.45±0.027	0.14±0.48	-0.86±0.01	-0.11±0.07
Pekalongan	-0.6±0.016	-1,33±0.019	-0.84±0.019	-0.25±0.57	-1.51±0.29	-0.51±0.15
Pemalang	-0.29 ±0.17	-0.97 ±0.17	-0.48 ±0.17	1.64 ±0.32	-1.39 ±0.23	-0.49 ±0.15

(🟩 $I_{geo} \leq 0$), class 0 (uncontaminated); 🟨 ($0 < I_{geo} \leq 1$), class 1 (uncontaminated to moderately contaminated)

Note:



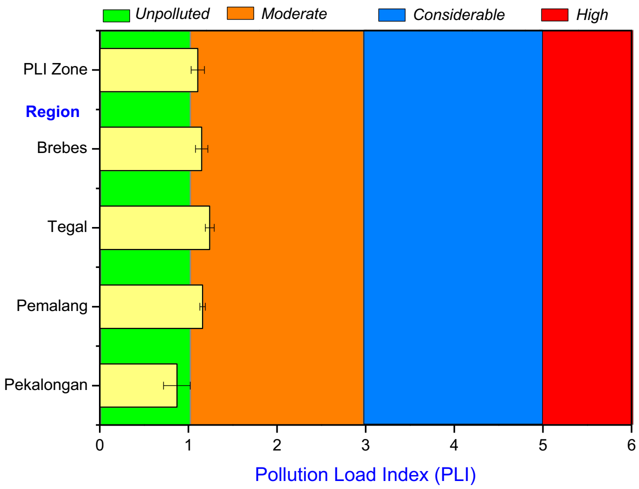


Fig. 3 Pollution load index (PLI) of the metals in the sediments of the studied area

accumulation in the shrimp muscle tended to be in the same decreasing order in Tegal and Pekalongan, in which Al and Fe were the most accumulated and Co was the least accumulated. Wu and Chen (2005) showed that Cd mostly accumulated in the hepatopancreas followed by the gills and muscle. Overall, the descending order of the concentrations of the metals analyzed in this study during the wet season was as follows:

Brebes: Al > Zn > Fe > Cu > Mn > Cr > Hg

Tegal: Al > Fe > Zn > Cu > Mn > Cr > Pb > Hg > Cd > Co

Pekalongan: Al > Fe > Zn > Cu > Mn > Cr > Pb > Hg > Co > Cd

Pemalang: Al > Fe > Zn > Cu > Mn > Cr > Hg > Pb > Cd > Co

A similar tendency was observed for metal accumulation during the dry season as follows:

Brebes: Al > Zn > Fe > Cu > Mn > Cr > Hg > Cd > Co > Pb

Tegal: Al > Fe > Zn > Cu > Mn > Pb > Cr > Hg > Co > Cd

Pekalongan: Al > Fe > Zn > Cu > Mn > Pb > Cr > Hg > Co > Cd

Pemalang: Al > Fe > Mn > Zn > Cu > Cr > Hg > Pb > Cd > Co

Generally, the metal concentrations in the shrimp in the dry season were slightly higher than those in the wet season as shown in Pekalongan (ESM 10).

Among the ten metals, Al presented the highest level in all regions, followed by Fe and Zn. A similar tendency was observed in the wet and dry seasons. Wu and Yang (2011) found that Fe and Zn were the two most concentrated metals in the muscle tissue of wild *L. vannamei*, and the order of concentration was Fe > Zn > Cu > Cr > Mn.

In the present study, the Cu concentration (4.08 ± 2.98 mg kg^{-1} dw) was relatively similar to that reported for the muscle tissues of *L. vannamei* by Lacerda et al. (2009), which ranged from 23.2 to 63.4 µg g^{-1} dw. Previously, Páez-Osuna and Ruiz-Fernández (1995) reported Cu concentrations in the muscle tissue of this species in natural populations that varied from 22.7 to 27.5 µg g^{-1} dw. The metal concentration in *L. vannamei* was compared with that of samples from different countries as shown in Table 4.

Cu, Fe, Mn, and Zn are essential oligo-elements for *L. vannamei* (Frías-Espericueta et al. 2009), although high concentrations may affect metabolism. Dietary Cu impacts the composition of gut microbiota and subsequently affects the immune response and growth. For instance, the dietary Cu required to meet the growth and immune functions of *L. vannamei* has been estimated at approximately 30 mg kg^{-1} (Ayisi et al. 2017). At the higher level of 64 mg kg^{-1}, the survival of *L. vannamei* has been shown to be slightly depressed as reported by Davis et al. (1993).

The average concentrations (µg g^{-1} dw) of Cu in the studied shrimp muscle in the Brebes, Tegal, Pekalongan, and Pemalang regions were 26.9 ± 8.72, 27.3 ± 2.6, 25.6 ± 4.8, and 36.5 ± 5.3 during the wet season, respectively, and 42.6 ± 2.6, 37.3 ± 6.9, 31.1 ± 5.05, and 31.6 ± 6.5 during the dry season, respectively.

Wu and Chen (2004) found that Zn affects the gill function of this shrimp species. Frías-Espericueta et al. (2009) studied the effect of exposure to a mixture of Cd-Zn on *L. vannamei* and showed an antagonistic effect on this species, which aligned with the results of Barata et al. (2006), who emphasized that there is a high probability of competition for protein binding sites because Zn, Cu, and Cd are inducers of metallothionein synthesis. Thus, the combined effects of Cd and Zn may be expected to be antagonistic.

Concerning Hg, the average concentrations (µg g^{-1} dw) of Hg in shrimp ranged from 0.16 ± 0.04 (Pemalang) to 1.35 ± 0.89 (Brebes). The Hg source may have originated from the drainage basin (industrial, agricultural, and domestic effluents), where Hg is trapped and accumulates in a mangrove tidal creek, which may be a potential source of contamination in adjacent aquaculture sites (Costa et al. 2013), or the source may have originated from the shrimp farms because of the large amounts of feed applied during breeding. Lacerda et al. (2011) estimated an EF from intensive shrimp farming of 83.5 mg Hg ha^{-1} cycle^{-1}, which is comparable to the Hg emissions from urban wastewaters (200 mg ha^{-1}) and solid waste disposal (400 mg ha^{-1} year^{-1}) from cities located in the NE Brazil.

Table 4 Comparison of the mean concentrations (mg kg^{-1} dw) of trace metals reported for shrimp worldwide (muscle)

Location	Species	Al	Cd	Co	Cu	Cr	Fe	Mn	Pb	Zn	Hg	Ni	Rb	Ref
Mexico	*L. vannamei*		0.77 ± 0.01	1.18 ± 0.5	27.5 ± 2.4	0.85 ± 0.14	53.9 ± 9–0	4–54 ± 3.84		70.4 ± 4.9		0.81 ± 0.10		1
Mexico	*L. vannamei*										0.194 ± 0.509			2
Mexico	*L. vannamei*				19.2 ± 0.8		37 ± 9	3.0 ± 1.7		56 ± 6				3
India	*Penaeus monodon*				77 ± 11		297 ± 8	15.2 ± 3.3	0.147 ± 0.10	58 ± 21				4
Malaysia	*L. vannamei*		0.1 ± 0.04	1.5 ± 0.20	51.3 ± 10.12	3.8 ± 0.13		31.7 ± 1.77	3	72.4 ± 0.14		2.2 ± 0.35	4.5 ± 0.04	5
Bangladesh	*Parapenaeopsis sculptilis*		0.713 ± 0.06		5.049 ± 0.07	< 0.08	18.713 ± 2.63	< 0.2	0.69 ± 1.56	13.5 ± 0.43	< 0.03			6
NW Mexico	*L. vannamei*										0.20 ± 0.01			7
Malaysia	*Penaeus vannamei*		0.001 ± 0.001		6.806 ± 0.33	0.023 ± 0.015			0.108 ± 0.03	12.67 ± 0.2	0.04 ± 0.003			8
China	*P. vannamei*		n.d.	n.d.	24.26 ± 8.36	20.86 ± 5.27	61.35 ± 30.76	5.33 ± 2.50	n.d.	171.6 ± 118.7				9
China	Shrimp		0.001–0.32		2.28–28.13	0.18–2.14				9.58–19.16	0.001–0.018	0.001–0.25		10
California	*L. vannamei*		0.14 ± 0.02		18.4 ± 2.99				0.56 ± 0.04	13.0 ± 1.28				11
Indonesia	*L. vannamei*	200.82 ± 171.7	0.09 ± 0.06	0.12 ± 0.1	4.08 ± 2.98	32.48 ± 8.7	133.58 ± 86.48	31.93 ± 50.37	4.49 ± 8.96	67.49 ± 5.52	0.36 ± 0.43			Present study

References: 1, Páez-Osuna and Ruiz-Fernández (1995); 2, Delgado-Alvarez et al. (2015); 3, Páez-Osuna and Tron-Mayen (1996); 4, Mohapatra et al. (2007); 5, Anandkumar et al. (2017); 6, Baki et al. (2018); 7, Ruelas-Inzunza and Páez-Osuna (2004); 8, Mok et al. (2012); 9, Wu and Yang (2011); 10, Liu et al. (2019); 11, Jara-Marini et al. (2009)

Estimated daily intake

To evaluate whether the metal levels found in the shrimp samples were safe for consumers, a comparison was performed with their tolerable intake. The calculated daily intake of the metals through the consumption of shrimp was below the acceptable daily intake suggested by the joint FAO/WHO Expert Committee on Food Additives and ATSDR for almost all samples with the exception of Cr during the dry season in Tegal and Pemalang (ESM 11). Based on this result, it can be recommended that the consumption of average amounts of the analyzed shrimps does not pose any health risk for shrimp consumers.

Based on the Indonesia Statistics Center database, the consumption rate of fresh fish and fish products in Central Java was 20.67 g person^{-1} day^{-1} (BPS 2016). By considering the mean body weight of Asian adults as 57.7 kg (Walpole et al. 2012), the daily intake of metals was calculated. A linear relationship is observed based on the weight applied to estimate the EDI and THQ values in Taiwan by Ju et al. (2017), which was 57.3 kg for female adults aged 19–65 years old.

Target hazard quotient and hazard index

The THQ and HI were calculated to assess the potential risks due to metals from the consumption of shrimp raised in aquaculture ponds. In both the wet season and dry season, the THQ was less than 1, indicating that there was no obvious health risk from the intake of the individual metals. The hazard index value of below 1 also suggested no potential adverse effect from the combined metals from shrimp consumption throughout the year. However, the HI value tended to be higher in the dry season than the wet season except for Brebes (ESM 12). Mercury had a significant influence on the HI value in this region.

Pb had the highest THQ value during the dry season in Pekalongan, followed by Cr. It is well known that Pekalongan is one of the batik industrial cities in Indonesia (Rukayah et al. 2015) and that batik production has become the dominant activity among the local inhabitants (Hadiyanto et al. 2018). This industry produces wastewater that contains heavy metals, such as lead, chromium, and copper as a result of the production process (Puspita et al. 2011; Rashidi et al. 2012; Setiyono and Gustaman 2017). Synthetic dyes containing Pb and Cr are widely used during the coloring process of batiks to increase the bonding strength between the dyes and fabrics (Hadiyanto et al. 2018).

Madusari et al. (2016) detected the toxic metals Pb and Cd in the water column and *Chanos chanos* from a pond in this region. It was assumed that those metals came from the riverine system as the water source for the aquaculture system. Anandriyo Suryo and Indah (2013) reported that the rivers in Pekalongan have been polluted. Because of the limitation of actual wastewater treatment plant facilities, they estimated that the wastewater in Pekalongan that cannot be treated amounted to 3600 m^3 per day from 632 home batik industries in 2011. As the result, wastewater entered into the riverine system. Furthermore, the local societies and batik entrepreneurs lack an awareness of the riverine environments, which leads to worse river pollution.

Target cancer risk

Chromium in the white-legged shrimp tissue is the primary risk for human health (Wu and Yang 2011). Cr showed a moderate cancer risk (TR = 10^{-4}) for the exposed population in all regions, in both the wet season and dry season (ESM 13). While Pb indicated a low cancer risk during the wet season in all regions, the risk increased from low to moderate during the dry season in Tegal and Pekalongan as shown by the value of 10^{-5} (between 10^{-6} and 10^{-4}) stipulated by the New York State Department of Health (NYSDOH 2007).

Predominant metals in different environmental matrices

The PCA results correlating the extent of the metals in water are presented in Fig. 4. The cumulative variance contribution of PC1 was 44%. PC1 had high loadings and strong correlations with Al and Fe ($R = 0.79$) (Fig. 4a). The Brebes, Pekalongan, and Pemalang districts were correlated with Al and Fe concentrations (ESM 14a). This result confirmed the important contribution of the riverine system to the major metal contaminants rather than anthropogenic inputs, such as the precipitation of aerosol particles released by industrial activities (Ye et al. 2017) and the use of fertilizer in agriculture in the upstream areas (Comero et al. 2014). PC2 accounted for 22.1% of the variations and had high loadings of Zn and Mn, although a low correlation was observed between them ($R = 0.12$) (Fig. 4a). The most prominent metals at the Tegal district stations were Cu and Cd, which were moderately correlated ($R = 0.46$). In this district, which has the densest suburban population, the presence of Cu and Cd can be associated with industrial activities, i.e., fishing, farming, and shipyard activity (De Witte et al. 2016).

In the sediment, we observed that PC1 accounted for 44.1% of the variance and had high loadings on Cu and Cr as well as Pb and Zn, while PC2 elucidated 18% of the total variance and had a high loading of Mn (Fig. 4). The occurrence of Zn was correlated with Cu and Pb (both $R = 0.73$), and Cr was correlated with Al ($R = 0.73$) and Pb ($R = 0.65$) (ESM 14b). Zn and Cr were correlated, with $R = 0.60$. Al showed a strong correlation with Pb ($R = 0.55$) and Zn ($R = 0.50$), while Fe and Mn were also strongly correlated with Co ($R = 0.56$) (ESM 14b). Figure 4 b shows that the sediment from Pekalongan and Tegal was characterized by high

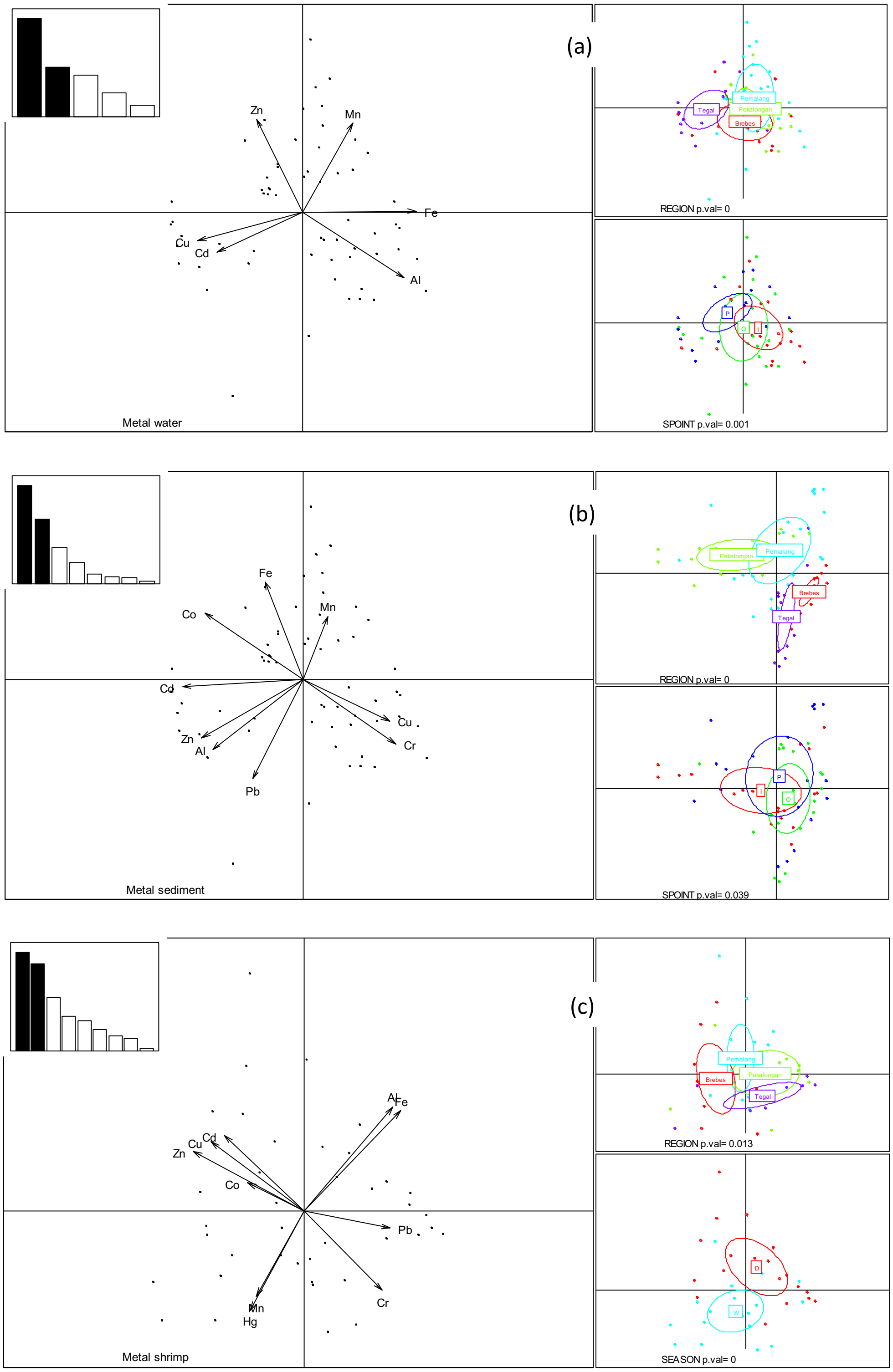


Fig. 4 Principal component analysis of the metals in **a** water, **b** sediment, and **c** shrimp

concentrations of Cu, Cr, Pb, and Zn. In fact, Pekalongan is the area known for batik textiles (Rukayah et al. 2015), which are processed with Cr dye; thus, Cr ends up as a byproduct and an environmental contaminant in the riverine system. As shown in Fig. 4 a, b, and c, Al, Cd, Co, and Fe were associated with the second component, which may indicate that the sources of those metals in the sediment were similar and likely from natural sources. This finding was confirmed by the use of several indexes, with the EF, I_{geo}, and CF showing that the Al, Cd, Co, and Fe concentrations in the sediment at some sampling sites were lower than the background concentrations.

Concerning the shrimp, PC1 accounted for 27.86% of the total variance and PC2 accounted for 24.7%. Figure 4 c shows that the Al concentration was highly correlated with the Fe ($R = 0.94$), Mn ($R = 0.64$), and Zn ($R = 0.54$) concentrations. The points in the figure represent the soil samples ($n = 40$). Many points were close, which indicates that these samples had less variability but were adequately discriminated. Figure 4 c also shows that the Cd concentrations had the same higher magnitude as the eigenvalues of Cu, Co, and Zn, which formed a group that was not attributed to a specific site. Figure 4 c and ESM 14c show that the Fe concentrations were highly correlated with the Mn ($R = 0.66$), Pb ($R = 0.54$), and Zn ($R = 0.53$) concentrations, whereas the Mn and Hg concentrations were not correlated despite sharing the same eigenvalues. ESM 12 shows that Pb and Cr were the most influential to the THQ, especially in Tegal.

In the present study, the metal environmental status of *L. vannamei* in aquaculture systems in Indonesia has been described for the first time. This study used a comprehensive pollution index and demonstrated that aquaculture pond water is probably contaminated by Mn and Zn from natural sources. Regarding the sediment, our findings from various indexes (I_{geo}, CF, and PLI) showed that the sediments were not polluted by the studied metals and were considered to present a low ecological risk. Despite the occurrence of many heavy metals in shrimp, Zn remained a source of concern because it was found in excess in the *L. vannamei* muscle.

Conclusions

Here, we investigated the ecological and human health risks of heavy metals in the water and sediment in aquaculture environments. The results showed that the occurrence of heavy metals in water was dependent on the site. The results suggested that riverine systems contributed to higher concentrations of Fe and Mn. The nonessential element Cd was observed in the shrimp ponds as a trace element in all regions, with the highest level found in Pekalongan ($3.15 \pm 0.33\ \mu g\ L^{-1}$). From the water assessment, the greatest potential ecological hazard was Mn based on the P_n value. Concerning the sediment, Cr, Cu, Zn, Co, and Pb showed similar spatial variation trends with decreasing concentrations. Their high-value area was located in Brebes, and the lowest value occurred in Pekalongan. The risk assessment for sediment using the SQG results suggested that the ecological risk of heavy metals at the studied sites was low. However, the EF, I_{geo}, CF, and PLI values for Mn showed a medium-high environmental risk. In shrimp, the accumulation of Al, Fe, Zn, Cu, and Mn showed similar trends at all stations and low values of Hg, Pb, Cd, and Co were observed. The findings of this research also revealed that the lack of risk associated with consumption if we assumed that protein daily intake per capita was approximately 20 g person^{-1} day^{-1} for all studied heavy metals. Furthermore, the THQ and HI were higher in the dry season, and our findings demonstrated that Pb and Cr influenced the THQ while Hg significantly influenced the HI value. This research provided an adequate overview of the heavy metal contamination in different environmental matrices, although further study is necessary to design the conceptual framework model for heavy metal contamination in an aquaculture field to better mitigate the hazards and risks in sustainable aquaculture.

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