INCREASING RISK OF HEAVY METAL CONTAMINATION IN SILVOFISHERY PONDS

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Received 18 January 2018/Accepted 17 April 2020

ABSTRACT

This research aimed to determine the concentration of organic matter, Pb and Cd found in a silvofishery pond, to assess the toxicity level, to analyze the changes in their concentration within a year's period and to analyze the correlation between concentration and changes. The research was conducted by doing five field observation activities and laboratory analysis from May 2016 to July 2017. Data were analyzed using ANOVA and correlation tests. The concentration of Pb and Cd in the organic matter, was increasing in all five observations. Throughout the research, the concentration level ranged at 1.60 – 3.30 mg/kg for organic matter, 3.130 – 8.230 mg/kg for Pb, and 1.089 – 2.820 mg/kg for Cd. In all observations, the toxicity level of Cd concentration in the sediment exceeded the standards recommended by US EPA (≤ 1.0 mg/kg) and ANZECC & ARMCANZ (≤ 1.5 mg/kg), while Pb concentration was within the safe range (≤ 21 mg/kg and ≤ 50 mg/kg, respectively). Moreover, the concentration and accumulation of Pb and Cd were strongly correlated, which indicated the possibility of having the same pollutant sources. Therefore, a better management plan is recommended to avoid heavy metal accumulation in silvofishery ponds, a plan that would include the arrangement of mangrove plants in inlet canals and their periodic pruning to prevent the heavy metal from returning to the environment through the litter fall.

Keywords: aquatic toxicology, bioremediation, cadmium, heavy metal, sediment chemistry, silvofishery

INTRODUCTION

Silvofishery has supported the aquaculture activities through its emphasis on environmental services (Suwarto *et al.* 2015). The Silvofishery system integrates mangrove plants into ponds to minimize environmental pollution caused by its effluent (Hastuti & Budihastuti 2016). The mangrove ecosystems has also improved the natural primary productivity because of nutrient cycling (Budihastuti *et al.* 2013).

Mangrove plays an important role in controlling the pollutants in the coastal ecosystem. Its root ecosystem can trap the organic matter sediments which contain the heavy metal pollutants (Yunus *et al.* 2011). Thus, reducing the concentration of pollutants in the aquatic system. Mangrove plants can also act as bio-remediators that are capable of absorbing pollutants from the environment and accumulating these in its body parts (Pakzadtoochaei 2013). Thus, mangroves have dual functions of controlling pollutant distribution, and remediating its growing environment.

Mangrove roots trap the polluted sediments and these sediments are then suspended around the tree. Thus, as pollutants accumulate, their concentration below the mangrove stands increases (Kathiresan *et al.* 2014). However, by absorbing the pollutant, the mangroves would decrease its concentration in the sediments (Selanno *et al.* 2015). Thus, the accumulation and diminishing rates of pollutants are affected by the balance between the deposition of pollutants and the absorption capacity of mangrove plants.

Heavy metal is one of the most dangerous types of pollutants that could be consumed and accumulated in plants and animals organs,

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resulting in toxicity (Reis *et al.* 2010; Asati *et al.* 2016). Thus, food chain is the most important aspect in the accumulation of heavy metal toxicity (Hajeb *et al.* 2014). Contamination of lower level organisms would lead to the accumulation of heavy metal in higher level organisms. Obviously, the risk of heavy metal contamination may increase in a polluted environment.

Among the heavy metal contaminants, Pb and Cd are the most common aquatic pollutants produced from daily activities. Sources of Pb include volcanic explosions, forest fires, lead emissions from industry and transportation, metal processing, and manufacturing (Zhang *et al.* 2015). Anthropogenic sources of Pb pollution include paints and glazed building materials (Walraven *et al.* 2016). The main sources of Cd in the sediment are the industrial and mining wastes (Yu *et al.* 2010; Donovan *et al.* 2016). Although still debatable, the application of P fertilizer was also considered as an additional source of Cd in the agricultural soil (Roberts 2014).

In order to secure environmental safety against pollutant contamination, some countries had developed environmental quality standards which differ in their applications. US EPA applies a maximum value of 1 mg/kg for cadmium (Cd) and 21 ppm for lead (Pb), while ANZECC & ARMCANZ recommends 1.5 mg/kg for Cd and 50mg/kg for Pb (Hubner *et al.* 2009).

In view of the environmental standards imposed by many agencies, the threat of heavy metal contamination is still very evident. Moreover, heavy metal does not appear as single element but is mostly attached to organic matter in the environment. The accumulation of heavy metal is related to the accumulation of organic matter (Yang *et al.* 2010). Hence, studies on the trend of organic matter and heavy metal accumulation are equally important.

Silvofishery has been considered as a "good management system" applied in conservative aquaculture (Jonell & Henriksson 2014). In most cases, however, the real application is different from the proposed model. For instance, plantations are set in the middle of ponds without any separating dikes which are supposed to prevent direct contact between the pool and the mangrove. Thus, the risk of pollutant contamination to the livestock cultivars might increase.

Silvofishery is mostly applied in degraded ponds (Yunus *et al.* 2015), making the heavy metal accumulation an additional risk to its already poor environmental quality. However, little concern has been directed towards organic matter and heavy metal accumulation in silvofishery. Hence, this research was conducted to study the concentration of Pb and Cd in a silvofishery pond, and its toxicity level, to analyze the changes in their concentration level within a year's period, and to analyze the correlation between these concentrations and changes.

MATERIALS AND METHODS

The research was conducted from May 2016 to July 2017 in a silvofishery pond in Mangunharjo Village, Tugu District, Semarang City, Central Java, Indonesia. Twenty-seven sampling points were selected and distributed throughout the pond area (Fig. 1). Data were collected during the five sampling periods in May, July, and September 2016 and in May and July 2017. Generally, the weather was bright with warm temperature during the sampling period. Since the sampling was carried out during the dry season, the fresh water dominated the supply to the silvofishery pond. Data collection was carried out to determine the concentration of organic matter, lead (Pb) and cadmium (Cd) in the sediment across a one-year period. Samples obtained from the sediment accumulated below the mangrove stands were then analyzed in the laboratory.

As much as 500 g sediment samples were taken from the surface to a depth of 30 cm using scoop and dipper. The sediment was readily washed. Therefore, Ekman grab could not be used for the sediment sampling. The water was filtered before the sediment was moved into the plastic bag. Laboratory analysis was done at the Wahana Laboratorium, Semarang. The organic matter was analyzed using the ashing method, while total heavy metal content was analyzed using Atomic Absorption Spectrophotometry (AAS) method.

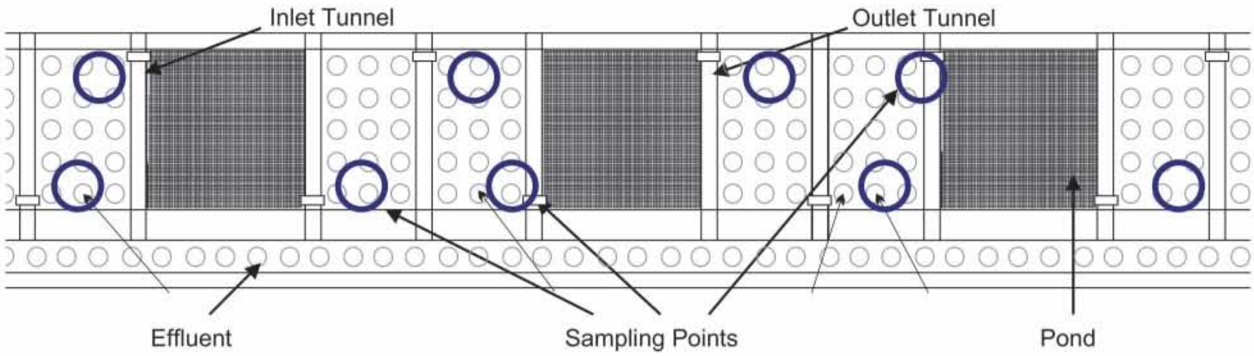

Figure 1 Schematic diagram of sampling points

Changes in organic matter and heavy metal concentration at each sampling point were also computed and analyzed. Analysis of variance was performed on the mean values of organic matter and heavy metal taken from different sampling periods, as well as their changes over those periods. Pearson Correlation analysis was performed to determine the relation between periodic average of organic matter, Pb and Cd concentrations and their changes during the study period.

RESULTS AND DISCUSSION

Variations existed in the concentration of organic matter, Pb and Cd in the silvofishery pond over the five different sampling periods. Data showed that there were increases in average concentrations and decreases in range, indicating a trend towards stabilization. The concentration of organic matter, Pb and Cd decreased from the first to the second observation. Among the sampling periods, the second period showed the most significant increase in the concentration of Pb and Cd (Table 1).

Concentration of heavy metals indicated that Pb was still within the safe range (Table 1). The maximum Pb concentration (8.090 mg/kg), was far below the critical value proposed by US EPA (21 mg/kg) and ANZECC & ARMCAZ (50 mg/kg). Thus, the silvofishery pond in Mangunharjo Village was safe from Pb contamination. However, the Cd concentration has exceeded the recommended limit. The minimum concentration of Cd in the first (1.089 mg/kg) to third (1.460 mg/kg) observations exceeded the US EPA limit of $\leq$ 1 mg/kg. In the fourth and fifth observations, the minimum concentration of Cd even exceeded the limit set by ANZECC & ARMCAZ with the expected concentration of $\leq$ 1.5 mg/kg.

Table 1 Concentration level of organic matter, Pb and Cd in the silvofishery pond sediments

No.	Parameters	Observation Period				
		I	II	III	IV	V
1.	Organic Matter (%)					
	a Range	1.89-3.30	1.71-3.12	1.60-2.70	1.65-2.62	1.90-2.83
	b Average	2.23 ± 0.28^{p}	2.21 ± 0.31^{p}	2.27 ± 0.27^{p}	2.29 ± 0.25^{p}	2.38 ± 0.20^{p}
	c Average change		-0.04^{x}	0.05^{x}	0.02^{x}	0.09^{x}
2.	Pb (mg/kg)					
	a Range	3.130-8.230	4.120-7.230	4.353-8.090	4.501-7.930	5.130-7.449
	b Average	5.178 ± 1.258^{p}	5.234 ± 0.747^{p}	6.028 ± 0.868^{q}	6.041 ± 0.759^{q}	6.138 ± 0.627^{q}
	c Average change		-0.044^{x}	0.795^{y}	0.012^{x}	0.098^{x}
3.	Cd (mg/kg)					
	a Range	1.089$^{\varepsilon}$-2.409$^{\phi}$	1.118$^{\varepsilon}$-2.440$^{\phi}$	1.460$^{\varepsilon}$-2.820$^{\phi}$	1.510$^{\phi}$-2.710$^{\phi}$	1.583$^{\phi}$-2.772$^{\phi}$
	b Average	1.887$^{\phi}$ ± 0.353^{p}	1.851$^{\phi}$ ± 0.344^{p}	2.110$^{\phi}$ ± 0.339^{q}	2.120$^{\phi}$ ± 0.336^{q}	2.171$^{\phi}$ ± 0.258^{q}
	c Average change		-0.026^{x}	0.259^{y}	0.010^{x}	0.052^{x}

Notes: ε = exceeded the standard value of US EPA; ϕ = exceeded the standard value of US EPA and ANZECC & ARMCAZ; $^{p, q, x, y}$ = Different letter across the same row indicates significant differences.

The analysis of variance (ANOVA) of organic matter did not show any significant difference among the mean values and concentration changes of the five sampling periods. This indicated that the management of organic matter concentrations and changes in the silvofishery pond was effective. However, the ANOVA of Pb and Cd concentrations and changes showed that there was a significant difference among the different sampling periods. There were two groups of mean concentration: the first group which consisted of the first and second observations, and the second group which consisted of the third to fifth observations. This indicated that a significant increase in Pb and Cd accumulation happened in the second period. Analysis of the concentration changes showed that a significant change occurred between the second period and the other periods. The significant increase in Pb and Cd concentration in the second period indicated the dominance of heavy metal accumulation in certain sampling periods.

Another important concern manifested by these findings is the increasing accumulation of Pb and Cd. In the first period, the concentration of both heavy metal elements was relatively low, but in the second to fourth periods it tended to increase. This indicated the increasing risk of heavy metal pollution in the silvofishery pond, which might trigger the contamination of the livestock cultivars. The concentration of Cd alone was sufficient proof that the pond was contaminated. Thus, a more effective pond management plan should be formulated in order to avoid the danger of continuous deposition of heavy metal pollution and subsequent contamination.

Heavy metal accumulation in the sediment is related to the sedimentation process. The sediment carries various substances including organic matter and heavy metal. Thus, the accumulation of heavy metal might be interrelated with that of any other substances. This research also evaluated the correlation between heavy metal and organic matter (Table 2).

Although the correlation coefficients were high, the organic matter concentration was not related to Pb and Cd concentration, as well as with concentration changes (Table 2). However, the concentrations and changes of Pb and Cd correlated with each other. This indicated that the source of organic matter was not only sediment accumulation. Low correlation level between concentration changes indicated the difference in the accumulation rate between the organic matter of Pb and Cd. The high and significant correlation between Pb and Cd concentrations and changes showed that both heavy metal elements came from the same source, i.e., the contaminated sediment.

The silvofishery pond in Mangunharjo Village has accumulated organic matter and heavy metal elements, as indicated by the increasing concentration of these substances in the sediment. Mangrove vegetations in the pond provide fine sediment trapping by slowing down the current; thus, resulting in an increase in the sedimentation rate (Adame *et al.* 2010). An increase in heavy metal concentration in ponds is highly related to the water sources. Coastal ponds are supplied by both sea and river water. However, river streams are considered as the main source of pollutants in the coastal area since these are the channels of anthropogenic and industrial waste disposal (Kumar *et al.* 2015).

Table 2 Correlation among OM, Pb, and Cd concentration and changes in a silvofishery pond

		Correlation (Sig.)		
		OM	Pb	Cd
Average concentration	OM	----	0.770 (0.128)	0.810 (0.097)
	Pb		----	0.998 (0.000)**
	Cd			----
Average concentration change	OM	----	0.394 (0.606)	0.484 (0.516)
	Pb		----	0.995 (0.005)**
	Cd			----

Notes: * = correlation is significant at the 0.05 level; ** = correlation is significant at the 0.01 level; some cells are left blank intentionally due to similar correlation items

Various inland activities produce heavy metal contaminated wastes which then enter the aquatic system (Zhang *et al.* 2010). When the freshwater flows to the estuaries, the heavy metal is dispersed throughout the surrounding ecosystem (Ruzhong *et al.* 2010), such as the ocean and ponds. Most heavy metal pollutants are bound to fine sediments (Zhang *et al.* 2014). When the water enters vegetated ecosystem, the flow would slow down, increasing the chance of sedimentation (Turgut *et al.* 2015). Thus, the accumulation of heavy metal occurs along with sediment deposition.

The increasing heavy metal concentration in the silvofishery pond indicated that the water sources were polluted. The source could be from the freshwater stream or the seawater. The Semarang rivers, as well as the city's coastal areas, have been known to be polluted due to high industrial activities (Hastuti 2015). Thus, there is a severe risk of pollutant contamination in the silvofishery ponds. Moreover, the heavy metal concentration is expected to continually increase due to sediment trapping by mangrove roots in the ponds.

Although the mangroves are known as bioremediators of heavy metal pollutants *(Kannan et al.* 2016), their capacity is limited. The mangrove trees could accumulate heavy metal elements in their various parts but the elements would be returned to the environment through litter fall and the decomposition processes (Martuti *et al.* 2017). Thus, the accumulation rate of heavy metal pollutants would depend on the supply rate from the sources and the uptake rate by the mangroves. The increasing trend of pollutants accumulation showed that the mangrove uptake rates could not match the pollutant supply rates. This requires the development of more effective management plans to prevent the increasing risk of heavy metal contamination and to reduce the toxicity level of the livestock cultivars.

The extreme increase of organic matter and heavy metal concentration in the second period (from the 2^{nd} to 3^{rd} observations) manifested the high input rate of sediment during that period. This might have occurred due to frequent rains that flooded the pond (Gharbi *et al.* 2016). and carried high amount of sediments from the river stream to the ponds. The ponds which generally occupy a single canal as water inlet and outlet would trap incoming water, thus increasing the chance of sediment deposition.

Cd concentration in the silvofishery pond indicated an unsafe environment. Although heavy metal concentration in the sediment is not directly related to water quality, this may affect the availability of dissolved heavy metal in the water (Hassaan *et al.* 2016). US EPA recommends a maximum concentration of 1 mg/kg of Cd in the sediment, while ANZECC & ARMCANZ's standard is higher at 1.5 mg/kg (Hubner *et al.* 2009). However, in all observations, the average concentration of heavy metal exceeded both standards, indicating a high possibility of a polluted environment since the beginning. Therefore, such increases in the concentration rates need thorough consideration in the management of silvofishery ponds.

This research showed that Pb and Cd were accumulating in the silvofishery pond sediment. However, only Cd exceeded the recommended value, while Pb was still within the safe range. Such condition increases the alarming possibility of heavy metal contamination of cultivated fish (Kumar *et al.* 2011). Any toxic elements in the aquatic system can be easily transported from one organism to another through the food chain. Heavy metal pollutants would be absorbed by a low trophic level organism such as phytoplanktons, which are then consumed by zooplanktons and finally by fish (Hassaan *et al.* 2016). Thus, the risk of contaminated fish consumed by humans increases as well.

Through the food chain phenomena, heavy metal elements will bioaccumulate in living organisms; plants, animals, and humans. The toxicity in a plant is usually shown by several symptoms, such as changes in leaf color or abnormal plant growth (Nagajyoti *et al.* 2010). Unfortunately, toxicity in mangrove is usually unnoticeable as they have a high level of tolerance due to their bioaccumulative capability (Kannan *et al.* 2016). Thus, the effect of toxicity may expand to other aquatic animals including crabs, shells, and cultured fishes. Some animals are also known to be tolerant to heavy metal toxicity, such as shells (Shirneshan & Bakhtiari 2012). However, the animals with low tolerance level may show toxicity symptoms, such as declining growth rate and damaged liver and kidney tissues (Jayakumar *et al.* 2016).

The bioaccumulative capability of aquatic organisms increases the chance of heavy metal consumption and subsequent accumulation in the human body. Consuming a small amount of contaminated fish would not affect the human body in a short period of time. However, the effects of heavy metal toxicity would be noticeable when the concentration is high enough. Various symptoms caused by Cd toxicity include kidney damage, osteoporosis, cardiovascular diseases, hypertension, diabetes, modification of several organs, infertility, and many others (Bernhoft 2013). To avoid Cd toxicity, humans can only take a maximum of 1 μg/kg per day (USEPA 1998).

The analysis shows that the concentration and accumulation of Pb and Cd correlated with each other indicating that a common factor had caused the accumulation of both elements. Sediment input from contaminated sources was considered as one possible factor. Thus, both heavy metal elements were deposited along with the sediments (Zhang *et al.* 2014). The insignificant correlation between the heavy metal and organic matter concentration and accumulation indicated that organic matter had further decomposed to nutrients.

This research shows that even though silvofishery provides various beneficial environmental services, some negative impacts might arise after a considerable period of time. Various heavy metal elements might accumulate over time, thereby resulting in their increasing concentration. After certain periods of time, the accumulated heavy metal may exceed the safe limit and cause environmental toxicity leading to environmental hazards, especially for aquaculture activities related to human livelihood.

In order to maintain environmental quality by avoiding heavy metal toxicity in silvofishery ponds, a more effective management action plan is urgently needed. Decreasing the heavy metal concentration in the pond sediments must be the main objective, especially in Mangunharjo Village. However, the process may take a long time, and controlling the heavy metal concentration in sediments is a gradual process and cannot be done instantly. An important management intervention to decrease the sediment input from the pond is by separating the mangrove plantation. Dikes need to be constructed around it, mangroves should be planted in the inlet as a canal formation, thereby filtering the water input and sediment deposition before entering the pond. In so doing, the risk of heavy metal accumulation in the pond is decreased.

Another strategy to decrease heavy metal concentration in the ecosystem is by pruning the mangrove branches and leaves. Thus, heavy metals absorbed and accumulated in the plant organs, particularly the leaves and branches, could be totally removed and prevented from reentering the ecosystem through litter falls. The pruning also stimulates the development of fresh branches and leaves, thereby providing more space for heavy metal absorption in the ecosystem. In this way, the concentration of heavy metal in the silvofishery pond would gradually decrease. The combination of both techniques may improve the effectiveness of heavy metal removal from the silvofishery pond ecosystem, thus, minimizing the risk of heavy metal toxicity.

CONCLUSION

The silvofishery pond has accumulated organic matter rich with heavy metallic elements Pb and Cd, indicating an increased risk of environmental toxicity. The pond was contaminated with Cd whose concentration has exceeded the recommended level by both US EPA (≤ 1.0 mg/kg) and ANZECC & ARMCANZ (≤ 1.5 mg/kg). This excessive concentration might affect the aquatic organisms in the ecosystem. The increasing accumulation rate during the last periods of observation indicated an imbalance between pollutant supply and mangrove uptake rates. The concentration and accumulation rates of Pb and Cd show a high degree of correlation, indicating a possibility that both metallic elements come from the same sources. The recommendations for better management plans and actions include changing the pond setting by planting mangroves in the inlet canals, developing dikes to avoid direct rapid water flow from the mangroves to the pond, and pruning the mangrove leaves and branches to decrease heavy metal recycling. Such pruning would also promote the development of fresh leaves and branches which can increase heavy metal absorption.

ACKNOWLEDGEMENT

This research was supported by the PNBP DIPA University of Diponegoro funding source batch 2017 No. 275-066/UN7.5.1/PG/2017.

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