

1 **ACCEPTED MANUSCRIPT**

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3 **INCREASING RISK OF HEAVY METAL CONTAMINATION IN SILVOFISHERY PONDS**

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ACCEPTED MANUSCRIPT

17 **INCREASING RISK OF HEAVY METAL CONTAMINATION IN SILVOFISHERY**
18 **PONDS**

19
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25 **Abstract**

26 This research aims to observe the concentration of organic matter, Pb and Cd in a
27 silvofishery pond, to study the toxicity level status, to analyze changes in their concentration within
28 a year's period, and to analyze the correlation between the concentration and changes. The research
29 was conducted through field observation and laboratory analysis from May 2016 to July 2017,
30 which included five observation activities. Statistical analysis was conducted by using ANOVA and
31 correlation tests. The results show that the concentration of organic matter, Pb and Cd, was
32 increasing in all five observations. Throughout the research, the ranges of organic matter, Pb and
33 Cd, were recorded at 1.60–3.30 mg/kg, 3.130–8.230 mg/kg, and 1.089–2.820 mg/kg, respectively.
34 In all observations, toxicity level showed that Cd concentration in the sediment had exceeded the
35 standards recommended by US EPA (≤ 1.0 mg/kg) and ANZECC & ARMCANZ (≤ 1.5 mg/kg),
36 while Pb was within the safe range (≤ 21 mg/kg and ≤ 50 mg/kg). The correlation analysis showed
37 that the concentration and accumulation of Pb and Cd were highly related, which indicated the
38 possibility of the same pollutant sources. Recommendations for a better management plan to avoid
39 heavy metal accumulation in silvofishery ponds would include the arrangement of mangrove plants
40 in inlet canals and periodic pruning to hinder heavy metal from returning to the environment
41 through litter fall.
42

43 **Keywords:** aquatic toxicology, bioremediation, cadmium, heavy metal, sediment chemistry,
44 silvofishery
45

46 **INTRODUCTION**

47 Silvofishery has been applied to support aquaculture activities by emphasizing its
48 environmental services (Suwanto et al. 2015). The Silvofishery system integrates mangrove plants
49 into ponds to hinder environmental pollution caused by its effluent (Hastuti and Budihastuti 2016).
50 The existence of mangrove plants is also expected to improve natural primary productivity because
51 of nutrient cycling by mangrove vegetations (Budihastuti et al. 2013).

52 Mangrove plays an important role in controlling aquatic pollutants in the coastal ecosystem.
53 Sediment which contains pollutants such as organic material and heavy metal can be trapped by
54 mangrove rooting (Yunus et al. 2011). Thus, the concentration of pollutants in the aquatic system
55 could be reduced. Mangrove plants are also capable of absorbing pollutants from the environment
56 and accumulating them in certain organs, and this function is known as bioremediation
57 (Pakzadtoochaei 2013). Thus, in addition to controlling pollutant distribution, mangrove also
58 remediates its growing environment.

59 Mangrove roots may trap sediment which contains various types of pollutants. The trapped
60 sediment is then suspended around the tree. Thus, as pollutants accumulate, their concentration
61 below the mangrove stands increases (Kathiresan et al. 2014). However, the absorption of pollutants
62 by mangrove stands would decrease their concentration (Selanno et al. 2015). Thus, the
63 accumulation and shrink rates of pollutants are affected by the balance between the supply of
64 pollutants and their absorption by mangrove plants.

65 Heavy metal is one of the most dangerous types of pollutants. It could be consumed and
66 accumulated in plants' and animals' organs, which causes toxicity (Reis et al. 2010; Asati et al.
67 2016). Thus, food chain is the most important aspect in the distribution of heavy metal toxicity
68 (Hajeb et al. 2014). Contamination of lower level organisms would lead to the accumulation of
69 heavy metal in higher level organisms. Thus, the risk of heavy metal contamination may increase in
70 a polluted environment.

71 Among the available heavy metal contaminants in the aquatic system, Pb and Cd are among
72 the most common pollutants produced from daily activities. Sources of Pb include volcanic
73 explosions, forest fires, lead emissions from industry and transportation, metal processing, and
74 manufacturing (Zhang et al. 2015). Anthropogenic sources of Pb pollution include paints and glazed
75 building materials (Walraven et al. 2016). The main source of Cd in the sediment is industrial and
76 mining wastes (Yu et al. 2010; Donovan et al. 2016). The application of P fertilizer was also
77 considered as an additional source of Cd in agricultural soil, although this is still debatable (Roberts
78 2014).

79 In order to control the environmental safety against pollutant contamination, some countries
80 had developed environmental quality standards. Different countries might apply different standards.
81 US EPA applies a maximum value of 1 mg/kg and 21 ppm respectively for cadmium (Cd) and lead
82 (Pb), while ANZECC & ARMCANZ recommends the value of 1.5mg/kg and 50mg/kg for each
83 element (Hubner et al. 2009).

84 The threat of heavy metal contamination is obvious due to the determined standards
85 applicable from many environmental agencies. However, heavy metal would not available as single
86 element in the environment. Heavy metal is mostly attached to organic matters. The accumulation
87 of heavy metal is related to the accumulation of organic matters (Yang et al. 2010). Therefore, the
88 importance of studying the trend of organic matter accumulation is as important as the heavy metal.

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

94 Silvofishery is mostly applied in degraded ponds (Yunus et al. 2015). Thus, in addition to its
95 poor environmental quality, the pollutant level may also be high. However, the accumulation of
96 pollutants in silvofishery ponds such as organic matter and heavy metal has never aroused any
97 serious concern. This research was conducted to study the concentration of organic matter, Pb and
98 Cd in a silvofishery pond, to study the toxicity level status, to analyze changes in their concentration
99 within a year's period, and to analyze the correlation between the concentrations and changes.

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MATERIALS AND METHODS

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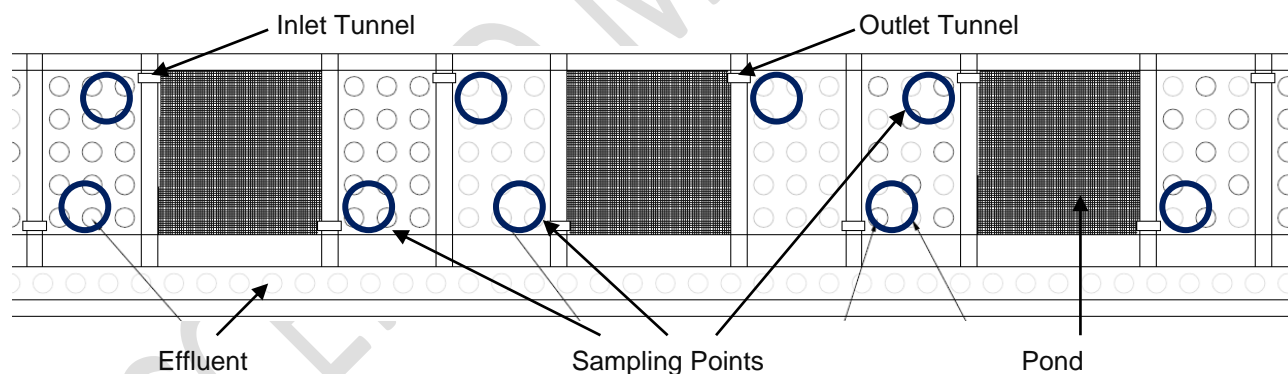
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Figure 1. Schematic of sampling points

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Samples of the sediment were taken from the surface to the depth of 30 cm using scoop and dipper as many as 500 gr. The sediment was easily washed, therefore ekman grab could not be used for sediment sampling. The water was tapped before the sediment was moved into the plastic bag. Laboratory analysis was carried out in Wahana Laboratorium, Semarang. The method used in organic matter analysis was ashing, while heavy metals were analyzed using Atomic Absorption Spectrophotometry (AAS) method. Analysis was carried out for total heavy metal content.

Data processing was conducted to calculate changes in organic matter and heavy metal concentration at each sampling point. Analysis of variance was performed to analyze the mean

125 differences of values pertaining to organic matter and heavy metal taken from different sampling
 126 periods, as well as their changes over those periods. Correlation analysis was performed to analyze
 127 the relation between periodic average organic matter, Pb and Cd concentrations and their changes
 128 during the research using the Pearson correlation.

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RESULTS AND DISCUSSION

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Table 1. Concentration of organic matter, Pb and Cd in the sediment from a silvofishery pond, its level status, and its mean differences in five different observation periods.

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 ^ε -2.409 ^φ	1.118 ^ε -2.440 ^φ	1.460 ^ε -2.820 ^φ	1.510 ^φ -2.710 ^φ	1.583 ^φ -2.772 ^φ
b	Average	1.887 ^φ ±0.353 ^p	1.851 ^φ ±0.344 ^p	2.110 ^φ ±0.339 ^q	2.120 ^φ ±0.336 ^q	2.171 ^φ ±0.258 ^q
c	Average Change		-0.026 ^x	0.259 ^y	0.010 ^x	0.052 ^x

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Annotation: 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 in the same line indicates a significant difference.

Concentration of heavy metals as shown in Table 1 indicated that Pb was still within the safe range. The observations of Pb concentration generated the maximum value of 8.090 mg/kg, which was far below the critical value proposed by US EPA and ANZECC & ARMCAZ. Thus, the silvofishery pond in Mangunharjo Village was safe from Pb contamination. However, the observations of Cd concentration showed that it exceeded the recommended limit. The minimum concentration of Cd in the first to third observations only exceeded the US EPA limit with the expected concentration of ≤ 1 mg/kg. Unfortunately, the fourth and fifth observations showed that

152 the minimum concentration of Cd even exceeded the limit set by ANZECC & ARMCAZ with the
 153 expected concentration of ≤ 1.5 mg/kg.

154 The analysis of variance of organic matter showed that there was no significant difference in
 155 mean values and concentration changes from one observation to another. This indicated that the
 156 management of organic matter concentrations and changes in the silvofishery pond was effective.
 157 However, the analysis of variance of Pb and Cd concentrations and changes showed that there was
 158 a significant difference among different treatments. There were two groups of mean concentration:
 159 the first group which consisted of the first and second observations, and the second group which
 160 consisted of the third to fifth observations. This indicated that there was significant accumulation of
 161 Pb and Cd which took place in the second period. Analysis of concentration changes showed that
 162 there was a significant change which occurred between the second period and another period. The
 163 significant increase in Pb and Cd concentration in the second period indicated the domination of
 164 heavy metal accumulation in certain sampling points.

165 Another important concern raised by these findings is the increasing accumulation of Pb and
 166 Cd. In the first period, the concentration of both heavy metal elements tended to decrease, but in the
 167 second to fourth periods it tended to increase. This indicated the increasing risk of heavy metal
 168 pollution in the silvofishery pond, which might trigger the contamination of the cultivars. The
 169 concentration of Cd alone was sufficient proof that the pond was contaminated. Thus, a more
 170 effective pond management plan should be formulated in order to avoid the danger of heavy metal
 171 pollution and contamination.

172 The accumulation of heavy metal in the sediment is related to the sedimentation process.
 173 The sediment usually carries various substances including organic matter and heavy metal. Thus,
 174 the accumulation of heavy metal might be interrelated with that of any other substances. This
 175 research evaluated the correlation between heavy metal and organic matter, as shown in Table 2.
 176

177 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			----

178 Annotation: * = correlation is significant at the 0.05 level

179 ** = correlation is significant at the 0.01 level

180 Some cells are left blank intentionally due to similar correlation items

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182 Table 2 shows that even though the correlation coefficients were high, organic matter
183 concentration did not correlate with Pb and Cd concentration, as well as with concentration
184 changes. Surprisingly, the concentrations and changes of Pb and Cd correlated with each other. This
185 indicated that the source of organic matter was not only sediment accumulation. Low correlation
186 level between concentration changes indicated the difference in the accumulation rate between the
187 organic matter of Pb and Cd. Fortunately, the high and significant correlation between Pb and Cd
188 concentrations and changes showed that both heavy metal elements came from the same source, i.e.
189 the contaminated sediment.

190 The silvofishery pond in Mangunharjo Village tended to accumulate organic matter and
191 heavy metal elements, which was indicated by the increasing concentration of those substances in
192 the sediment. Mangrove vegetations in the pond provide fine sediment trapping by slowing down
193 the current; thus, the sedimentation rate might increase (Adame et al. 2010). An increase in heavy
194 metal concentration in ponds is highly related to the water sources. Coastal ponds are supplied by
195 both sea and river water. However, river streams are considered as the main source of pollutants in
196 the coastal area since they have become channels for anthropogenic and industrial waste disposal
197 (Kumar et al. 2015).

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

205 The increasing heavy metal concentration in the silvofishery pond indicated that the water
206 sources were polluted. The source could be from the freshwater river stream or the seawater.
207 Semarang rivers, as well as the city's coastal area, have been known to be polluted due to high
208 industrial activities (Hastuti 2015). Thus, there is a severe risk of pollutant contamination in
209 silvofishery ponds. The pollutant might increase due to sediment trapping by mangrove roots in the
210 ponds. Thus, there is a probability that heavy metal concentration would keep increasing.

211 Mangrove has been known as a bioremediator of heavy metal pollutants (Kannan et al.
212 2016). However, its capacity is limited. Mangrove would accumulate heavy metal elements in its
213 various parts, but the elements would be returned to the environment through litter fall and
214 decomposition processes (Martuti et al. 2017). Thus, the accumulation rate of heavy metal
215 pollutants would depend on the supply rate from the sources and the uptake rate by mangrove. The
216 trend of increasing accumulation of pollutants showed that the mangrove uptake rates could not

217 match the pollutant supply rates. This requires the development of more effective management
218 plans to prevent the risk of heavy metal contamination and to reduce the toxicity of the cultivars.

219 The extreme increase of organic matter and heavy metal concentration in the second period
220 (from the 2nd to 3rd observations) showed the high input rate of sediment during that period. This
221 might occur due to frequent rain occurrences which cause pond flooding (Gharbi et al. 2016). Thus,
222 high amount of sediment might be carried from the river stream to ponds. Ponds which generally
223 occupy a single canal as water inlet and outlet would trap incoming water, thus increasing the
224 chance of sediment deposition.

225 The concentration of Cd in the silvofishery pond indicated an insecure environment.
226 Although heavy metal concentration in the sediment is not directly related to water quality, this may
227 affect the availability of dissolved heavy metal in the water (Hassaan et al. 2016). US EPA
228 recommends a maximum concentration of 1 mg/kg of Cd in the sediment, while ANZECC &
229 ARMCANZ's standard is higher at 1.5 mg/kg (Hubner et al. 2009). However, the average
230 concentration of heavy metal exceeded both standards in all observations, which showed the
231 possibility of a polluted environment since the beginning. Therefore, such increase in concentration
232 values should be taken into consideration in the management of silvofishery ponds.

233 This research shows that Pb and Cd were accumulating in the silvofishery pond sediment.
234 However, only Cd exceeded the recommended value, while Pb was still within the safe range. Such
235 condition increases the possibility of heavy metal contamination of cultivated fish (Kumar et al.
236 2011). Any toxic elements in the aquatic system can be easily transported from one organism to
237 another through food chain. Heavy metal pollutants would be absorbed by a low trophic level
238 organism such as phytoplanktons, which are then consumed by zooplanktons and finally by fish
239 (Hassaan et al. 2016). Thus, the risk of contaminated fish consumption by humans increases as well.

240 Heavy metal elements are bioaccumulative in living organisms which include plants,
241 animals, and humans. The toxicity in a plant is usually shown by several symptoms, such as
242 changes in leaf color or abnormal plant growth (Nagajyoti et al. 2010). Unfortunately, mangrove
243 has a high level of tolerance due to its bioaccumulation capability (Kannan et al. 2016). Thus, the
244 heavy metal toxicity in mangrove is usually unnoticeable. The effect of toxicity may expand to
245 other aquatic animals including crabs, shells, and cultured fish. Some animals are known to be
246 tolerant to heavy metal toxicity, such as shells (Shirneshan and Bakhtiari 2012). However, the
247 animals with low tolerance level may show toxicity symptoms, such as decreasing the growth rate
248 and the damage of liver and kidney tissues (Jayakumar et al. 2016).

249 The bioaccumulative capability of aquatic organisms increases the chance of heavy metal to
250 be consumed by humans and accumulated in the body. Consuming a small amount of contaminated
251 fish would not affect the human body in a short period of time. However, the effects of heavy metal

252 toxicity would be noticeable when the concentration is high enough. Various symptoms caused by
253 Cd toxicity include kidney damage, osteoporosis, cardiovascular diseases, hypertension, diabetes,
254 modification of several organs, infertility, and many others (Bernhoft 2013). To avoid Cd toxicity,
255 humans can only take a maximum of 1 µg/kg per day (USEPA 1998).

256 The analysis shows that the concentration and accumulation of Pb and Cd correlated with
257 each other. This indicated that there was a common factor which had caused the accumulation of
258 both elements. Sediment input from contaminated sources was considered as one possible factor.
259 Thus, both heavy metal elements were deposited along with the sedimentation (Zhang et al. 2014).
260 Insignificant correlation between heavy metal and organic matter concentration and accumulation
261 indicated that organic matter might experience further decomposition to nutrient.

262 This research shows that even though silvofishery provides various beneficial services to the
263 environment, some negative impacts might arise after a considerable period of time. Various heavy
264 metal elements might accumulate over time, which contributes to their increasing concentration.
265 After certain periods of time, the accumulation of heavy metal may exceed the safe limit and cause
266 environment toxicity. This leads to environmental hazards, especially for aquaculture activities
267 which are related to human livelihood.

268 In order to maintain the environmental quality and to avoid heavy metal toxicity in
269 silvofishery ponds, more effective management plans and actions should be devised and carried out.
270 Decreasing heavy metal concentration in the pond sediment must be the main objective, especially
271 in Mangunharjo Village. However, the process may take quite a long time, and controlling heavy
272 metal concentration in sediment cannot be done instantly because it is a gradual process. An
273 important thing to do in order to decrease the sediment input into the pond is by separating the
274 mangrove plantation. Mangroves should be planted in the inlet as a canal formation, while dikes
275 need to be constructed around it. Thus, water input could be filtered, and sediment would be
276 deposited before it enters the pond. By doing this, the risk of heavy metal accumulation in the pond
277 would decrease.

278 Another strategy to decrease heavy metal concentration in the ecosystem is by pruning the
279 mangrove leaves and branches. Thus, heavy metal elements which have been absorbed and
280 accumulated in the plant organs, especially its leaves and branches, could be totally removed from
281 the ecosystem by preventing the elements from reentering the ecosystem through litter falls. The
282 pruning also stimulates the development of fresh branches and leaves. Thus, more heavy metal
283 could be absorbed from the ecosystem. In this way, the concentration of heavy metal in the
284 silvofishery pond would gradually decrease. The combination of both techniques may improve the
285 effectiveness of heavy metal removal from the silvofishery pond ecosystem. Thus, the risk of heavy
286 metal toxicity could be prevented.

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CONCLUSION

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