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HEAVY METALS CONTAMINATION LEVEL AND WATER QUALITY PARAMETER CONDITIONS IN JATILUHUR RESERVOIR, WEST JAVA, INDONESIA

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ABSTRACT

Waste pollution into the Citarum River, the main water source of Jatiluhur Reservoir, was dominated by the manufacturing industry such as textile, chemical, metal and pharmaceutical. In general, the manufacturing industry is the most common contributor to heavy metal waste, which will cause various health problems. Therefore, it is essential to conduct studies on heavy metal contamination and water quality parameters conditions in the Jatiluhur Reservoir. The study aimed to provide a reference regarding the current condition of the heavy metal contamination level in both sediment and water of the Jatiluhur Reservoir, as well as to compare the levels of other water quality parameters against the standard of environmental quality. Heavy metals contents, such as Cu, Zn, Hg, Pb and Cd, were determined using X-Ray Fluorescence (XRF) Spectrometry method (for sediment) and Atomic Absorption Spectrometry (AAS) method (for water). Water quality parameters were analyzed by using methods developed by the Indonesian National Standard (SNI). The data obtained were compared to the Canadian Sediment Quality Guidelines (for heavy metal in sediment) and water quality standards from the Government of the Republic of Indonesia Regulation Number 82 of 2001 (Class 3) (for water quality parameters). Based on this study, Jatiluhur Reservoir is divided into three zones i.e., the inlet area, main inundation area and outlet area. Within the sedimentary layer, the mercury (Hg) was found to be accumulated throughout the Jatiluhur Reservoir area, exceeding the maximum limit, while Cu accumulated in the inlet area, exceeding the minimum limit. The other heavy metals (Zn, Pb and Cd) were found to be exceeding the minimum limit at some locations, but more results were below the minimum limit. The high concentration of heavy metals in the sediment was due to household and/or industrial wastes. Although all heavy metals were not detected in water, the presence of heavy metals in sediments could potentially dissolve into the water by means of upwelling. If this happens, the heavy metals can be excessively contained in water, resulting in a harmful habitat for aquatic biota. Water containing heavy metals will also be harmful for human. In general, water quality parameters in Jatiluhur Reservoir meet the standard for water quality. Only ammonia, however, was higher than the standard for sensitive fish life due to massive aquaculture activities in this reservoir. Considering the conditions of heavy metal contamination levels in both sediment and water, the biota that is most likely to be exposed to heavy metal is benthic organisms, because the organisms live at the bottom of the waters. The priority for further attention and countermeasure in improving the sediment and water quality of the Jatiluhur Reservoir was toward Hg, Cu, and Pb and ammonia.

Keywords: heavy metal, reservoir, sediment, water

INTRODUCTION

Jatiluhur Reservoir was the largest reservoir in Indonesia having an area of 8,300 ha. Jatiluhur Reservoir was built in 1957. The reservoir is the first multipurpose reservoir in Indonesia with potential available water of 12.9 billion m^3/year. The Jatiluhur Reservoir was built by blocking the Citarum River. The river passes through various anthropogenic activities and many cities in the West Java Province. Among functions of the Jatiluhur Reservoir are as hydropower, as a source of irrigation for 242,000 ha of surrounding rice fields, as a source of drinking water, as a place for inland fisheries (fish farming in floating net cages (KJA)), as flood control, and as tourism and water sports facilities. The types of fish commonly being

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cultivated in the floating net cages (KJA) in the Jatiluhur Reservoir include common carp, nile tilapia, red devils and catfish (*Pangasius* spp.) (Suprian & Salami 2011).

In the past few years, fishes in the Jatiluhur Reservoir seem to have been contaminated and are not suitable for consumption (Republika.co.id 2009). Likewise, in 2018, a report from Anggoro (2018) stated that there was an appeal for not consuming any kinds of fish from this reservoir. The contamination is allegedly due to the influence of various pollutions from the Citarum River as the main water source of the Jatiluhur Reservoir. The contamination may have also occurred due to the wastes of fish feed from the many KJAs in the reservoir. Industrial activities are the most common contributor of heavy metal waste because heavy metals are used in industrial activities as raw materials, additives and catalysts (Hutagalung *et al.* 1997 in Vigers *et al.* 1996). Heavy metal contamination in waters causes various health problems for aquatic biota and humans, such as problems in the nervous system, respiratory system, liver function, kidney function and growth of bones (Sanusi 1985). Heavy metals are categorized as harmful pollutants because it cannot be destroyed (non-degradable) by living organisms, so they will settle at the bottom of the waters and accumulate (Rochyatun & Rozak 2007).

Heavy metals can bond with organic compounds to form complex compounds that eventually settle at the bottom of the water (accumulate in sediments) (Marchand *et al.* 2006). On the other hand, sediments were an inseparable part of aquatic ecosystems that can provide habitat, feeding grounds, spawning grounds, and nurseries for various aquatic organisms. Contaminated sediments can reduce or eliminate the aquatic organisms that have important values for ecology, commercial or recreational uses (US EPA 2001). Fatoki and Mathabatha (2001) stated that sediment has a function as a metal container which can release the metal into the water through natural and anthropogenic processes. Heavy metals deposited in sediments can cause changes in water quality and transfer toxic chemicals to aquatic organisms (Permanawati *et al.* 2013).

Physical and chemical water quality parameters such as dissolved oxygen (DO), pH, total organic content, temperature and dissolved ion affect the life of aquatic organisms (Effendi 2003). Water quality parameters is influenced by its catchment area which is related to human activities (Wiwoho 2005; Asrini 2017).

A study conducted by Garno (2002) showed that the Jatiluhur Reservoir was hypertrophic (very nutrient-rich) and phytoplankton bloom can occur any time. The hypertrophic condition was largely influenced by the aquaculture activities in this reservoir and the influx of organic matters from settlements. Poor water quality conditions can disrupt aquatic life in the reservoir and reduce the diversity of aquatic biota. A study in Cirata Reservoir carried out by Komarawidjaja *et al.* (2005) showed that there was a growth disturbance in common carp which was thought to be closely related to water quality, especially with the high concentration of chlorophyll-*a* and total N in the waters. Poor water quality of the Jatiluhur Reservoir will also have an adverse impact on humans due to the function of the reservoir as a source of drinking water and as tourism and water sports facilities.

Based on the issues above, the study of heavy metal contamination and water quality parameter conditions in the Jatiluhur Reservoir should be conducted due to the serious polluted condition in the reservoir. This study aimed to obtain data on current condition of the heavy metal contamination level that has occurred in both sediment and water, as well as to compare the levels of other water quality parameters against the standard of environment quality.

MATERIALS AND METHODS

Location and Time

The research was conducted in Jatiluhur Reservoir in February 2019. Water and sediment samples were taken from 6 (six) locations in the reservoir, consisting of 3 (three) zones i.e., the inlet area, main inundation area, and outlet area (Fig. 1). The six locations were: 1. Citarum River inlet; 2) Jamaras; 3) Cilalawi River inlet; 4) KJA zone 5; 5) Pasir Kole and 6) DAM (outlet).

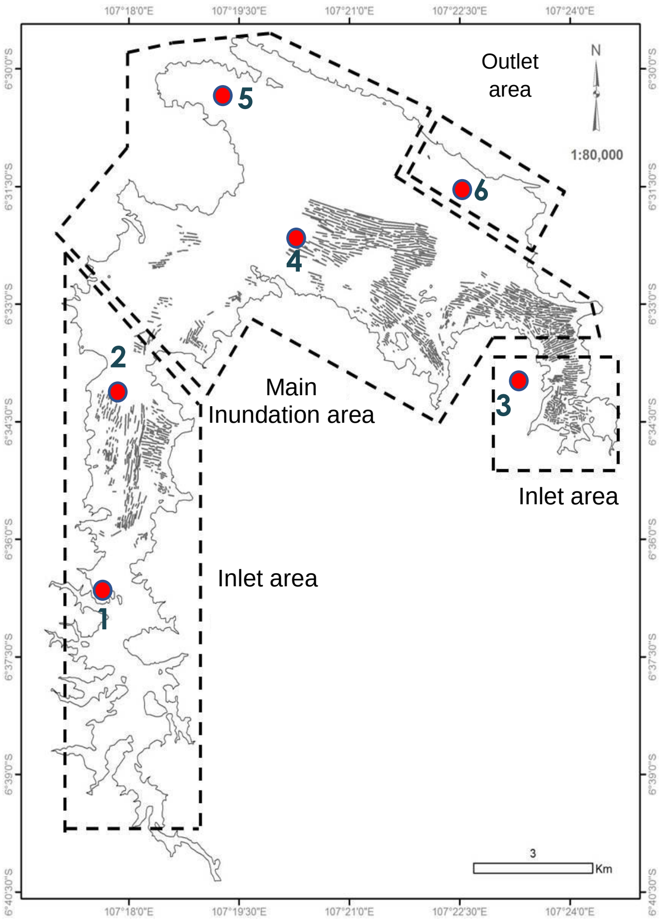

Figure 1 Map of sampling locations in Jatiluhur Reservoir

Methods

The water sample was taken using Van Dorn Water Sampler, then analyzed in the Global QA Laboratory using the methods from the Indonesian National Standard (SNI). Sediment sampling was conducted by using an Ekman Grab. The sediment samples obtained were then analyzed in the Central Nuclear Material Technology Laboratory, BATAN using the X-Ray Fluorescence Spectrometry (XRF) method to find out the heavy metals (Cu, Zn, Hg, Pb and Cd) contents. The XRF Spectrometry method is an application of radioisotopes used as an analytical method for detecting heavy metal content, especially in solid substances such as sediment. This method is still not commonly used in aquatic ecological studies that mostly use the AAS method. Methods used for analyzing the water and sediment samples are presented in Table 1.

Data Analyses

Data of heavy metal contents (in water and sediment) and water quality parameters obtained from this study were compared to the quality standard. The sediment parameters (heavy metal) were compared to the Canadian Sediment Quality Guidelines for the Protection of Aquatic Life on Freshwater set out by the Canadian Council of the Ministry of Environment (CCME). Reference to heavy metal quality standards according to CCME standard was presented in Table 2. Data on water quality parameters were compared to water quality standards from the Government Regulation of the Republic of Indonesia Number 82 of 2001 (Class 3, for the cultivation of freshwater fish, animal husbandry, water to irrigate crops, and or other purposes that require the same water as these uses).

Table 1 Parameters and methods used for analyzing water and sediment samples

Parameter	Unit	Method
A. Water		
Physical Properties:		
Temperature[a]	°C	5.4/IK/GQA/WQ/002
Total Dissolved Solid, TDS[a]	mg/L	SNI 06-6989.27-2005
Total Suspended Solid, TSS[a]	mg/L	SNI 06-6989.3-2004
Chemical Properties:		
pH[a]	-	SNI 06-6989.11-2004
Biological Oxygen Demand, BOD_5[a]	mg/L	SNI 6989.72:2009
Chemical Oxygen Demand, COD[a]	mg/L	SNI 6989.2:2009
Dissolved Oxygen, DO[a]	mg/L	SNI 06-6989.14-2004
Total Phosphate as P[a]	mg/L	5.4-IK-GQA-WQ-062
Nitrogen, Nitrate as N (NO_3-N)[a]	mg/L	5.4-IK-GQA-WQ-043
Ammonia, NH_3-N[a]	mg/L	SNI 06-6989.30-2005
Copper, Cu[a]	mg/L	SNI 6989.6:2009
Zinc, Zn[a]	mg/L	SNI 6989.7-2009
Mercury, Hg[a]	mg/L	SNI 6989.78:2009
Lead, Pb[a]	mg/L	SNI 6989.46:2009
Cadmium, Cd[a]	mg/L	SNI 06-6989.38-2005
B. Sediment		
Copper, Cu	µg/g	XRF Spectrometry
Zinc, Zn	µg/g	XRF Spectrometry
Mercury, Hg	µg/g	XRF Spectrometry
Lead, Pb	µg/g	XRF Spectrometry
Cadmium, Cd	µg/g	XRF Spectrometry

Note: [a] = Accredited by the National Accreditation Committee (KAN).

Table 1 Quality standards for heavy metals in sediments (for freshwater) based on CCME (2001)

Heavy metal	Concentration (mg/kg dry wt)	
	ISQG[a]	PEL[b]
Cu	35.7	197.0
Zn	123.0	315.0
Hg	0.170	0.486
Pb	35.0	91.3
Cd	0.6	3.5

Notes: a = ISQG (Interim Sediment Quality Guidelines): the value of the minimum limit, i.e., the limit of metal concentrations that has the low possibility to cause a negative biological effect; b = PEL (Probable Effect Level): the value of the maximum limit, i.e., the limit of metal concentrations having a greater possibility to cause a negative biological effect.

RESULTS AND DISCUSSION

Heavy Metal Contaminations

Our study showed that all heavy metals (Cu, Zn, Hg, Pb, and Cd) accumulated in the sediment layer of the Jatiluhur Reservoir and had exceeded the minimum limit (ISQG). Mercury (Hg) concentration exceeded the maximum limit (PEL) throughout the Jatiluhur Reservoir area. Copper (Cu) tended to accumulate in the inlet area, with concentration exceeding the minimum limit. The concentration of Zinc (Zn) and lead (Pb) exceeded the minimum limit at some locations (such as sampling point 5), however, the concentration of Cadmium was below the detection limit of the measuring tools (Table 3). Cadmium (Cd) seems to be relatively the safest metal found in the sediment samples compared to the other heavy metals.

Table 3 Results of laboratory analysis for heavy metal in sediment samples

Station	Test	Element (mg/kg)				
		Cu	Zn	Hg	Pb	Cd
1 Citarum River inlet	#1	39.78	105.16	2.1	29.4	<0.3
	#2	40.34	96.32	2.5	29.7	<0.3
	#3	35.71	93.51	1.8	28.6	<0.3
2 Jamaras	#1	36.91	113.59	1	26.8	<0.3
	#2	38.42	106.6	<0.7	26.8	<0.3
	#3	41.46	133.2	2.5	28.8	<0.3
3 Cilalawi River inlet	#1	35.79	129.82	<0.7	34.8	<0.3
	#2	84.12	103.31	2.4	31.9	<0.3
	#3	73.73	92.06	3.2	30.7	<0.8
4 KJA zone 5	#1	22.3	114.48	1.1	30.2	<0.8
	#2	12.94	104.27	2.5	25.9	<0.3
	#3	19.01	113.03	3.8	34	<0.3
5 Pasir Kole	#1	18.69	139.94	2.2	40	<0.3
	#2	23.57	143.16	1.5	36.3	<0.3
	#3	26.28	147.98	3.2	40.1	<0.3
6 DAM (outlet)	#1	20.13	89.41	<0.7	35.3	<0.3
	#2	74.13	103.39	3.6	32.5	<0.8
	#3	27	97.69	<0.7	31	<0.3
Quality Standard[a]		35.7	123	0.170	35.0	0.6
		197.0	315	0.486	91.3	3.5

Notes: a = Based on the Canadian Sediment Quality Guidelines for the Protection of Aquatic Life on Freshwater (CCME 2001).

exceed the ISQG (minimum limit)

exceed the PEL (maximum limit)

Mercury (Hg) was among the most toxic metals for aquatic biota. Mercury can be released from anthropogenic activities, such as from the antifouling paint for the hull of ships, slimicides used in the lumber and paper industries, pesticides and seed dressings in agriculture and pharmaceuticals (Garcia-Rico *et al.* 2006). Mercury has many benefits, but it is very toxic and has a high level of bioaccumulation capabilities (Garcia-Rico *et al.* 2006; Palar 2012). Mercury concentration exceeding the safe limit endangers the life of aquatic biota, either directly or indirectly (Palar 2012). Therefore, the high concentration of mercury in the Jatiluhur Reservoir should be handled immediately, especially when there is an upwelling process in the reservoir.

Our study showed that the concentration of Cu in sediments was much higher and exceeded the minimum limit in inlet areas, whereas the concentration of Zn and Pb was high in the main inundation areas. The spatial difference of the heavy metals' concentrations might be due to the heavy influx from the river into the reservoir, which was already contaminated by various anthropogenic activities. Cu is commonly used in insecticides, fungicides, brass alloy materials for household appliances, machine parts, as well as in water purification or as a food additive (Palar 2012). Household wastes in the form of metabolic waste and corrosion of pipes in residential areas usually also contain Cu (Connell & Miller 2006). The main inundation area has the longest water retention time compared to the other areas and is also much affected by the KJA aquaculture activities (marked with black lines in Fig. 1). Household wastes, water flow from urban areas and phosphate utilization (PO_4), contributed significantly to the entry of Pb metal into the waters (Connell & Miller 2006; Harteman 2011). Sources of Zn, Cu and Pb are chemical fertilizers, household wastes such as corrosion of pipes and detergent, as well as industrial wastes such as battery materials, cosmetics, plastics, rubber, soap, paint and ink, television tubes and fluorescent lamps, deodorants and chemicals for wood preservation (Connell & Miller 2006).

Table 4 Results of laboratory analysis results for heavy metal in water samples

Station	Element (mg/L)				
	Cu	Zn	Hg	Pb	Cd
1. Citarum River inlet	< 0.006	< 0.004	< 0.00009	< 0.0002	< 0.00004
2. Jamaras	< 0.006	< 0.004	< 0.00009	< 0.0002	< 0.00004
3. Cilalawi River inlet	< 0.006	< 0.004	< 0.00009	< 0.0002	< 0.00004
4. KJA zone 5	< 0.006	< 0.004	< 0.00009	< 0.0002	< 0.00004
5. Pasir Kole	< 0.006	< 0.004	< 0.00009	< 0.0002	< 0.00004
6. DAM (outlet)	< 0.006	< 0.004	< 0.00009	< 0.0002	< 0.00004
Quality Standard[a]	0.020	0.050	0.00200	0.0300	0.01000

Notes: a = Based on Government Regulation of the Republic of Indonesia Number 82 of 2001 (Class 3).

The concentrations of heavy metals in water in this study were very low, below the detection limit of the tool (Table 4), which indicated that the concentrations of heavy metals (Cu, Zn, Hg, Pb, and Cd) in the water of the Jatiluhur Reservoir were still within the safe limits. This fact was in agreement with a study in Jatiluhur Reservoir in 2011 conducted by Suprian and Salami (2011), especially for mercury concentration which was below the quality standard of the Government of the Republic of Indonesia Number 82 of 2001 (Class 3). Despite the results that all heavy metals in the water samples were below the safe limit, however, heavy metals in sediments could potentially dissolve into the water. Suprian and Salami (2011) stated that although the value of mercury in water is below the safe limit, however, the existence of mercury has to be monitored because it is difficult to eliminate mercury from the water. According to Fatoki and Mathabatha (2001) and Permanawati *et al.* 2013, sediment can release heavy metal into the water through natural and anthropogenic processes that cause changes in water quality and transfer of toxic chemicals to aquatic organisms. In reservoirs, the most extreme natural process that allows this to happen is upwelling.

Palar (2012) stated that the 0.01 ppm concentration of Cu in water is deadly for phytoplankton, Pb concentration of 2.75-49 ppm is deadly for crustaceans, while Pb concentration of 188 ppm is deadly for fish, and Cd concentration of 0.0028-4.6 ppm is deadly for Oligochaeta. Mercury concentration of ≥ 0.16 ppm is able to reduce the survival and growth rates of fish caused by the increase of stress and organ damage, whereas mercury concentration of ≥ 3 ppm causes mass mortality in common carp (Nirmala *et al.* 2012; Tyas *et al.* 2013). Lethal toxicity test by Prayogo *et al.* (2016) showed that 96 hours mercury exposure with concentration of 0.396 ppm in water had a 50% lethal effect on nilem carp (*Osteochilus hasselti*). If the heavy metals content in the sediment are released into the water, then the concentration of heavy metals can be excessive in the water, causing adverse impact to humans and aquatic biota.

Considering the conditions of heavy metal contamination levels in both sediment and water, the biota that is most likely to be exposed to heavy metal is benthic organisms, because benthic organisms live at the bottom of the waters (Reynolds 2012). Heavy metals can cause a decrease in species richness of benthic organisms and a change in species composition of benthic macroinvertebrate communities (Qu *et al.* 2010). Cu concentrations of 70-90 mg/kg, Zn concentrations of ±350 mg/kg and Pb concentrations of 30-40 mg/kg caused the decrease of Polychaeta and Molluscs biodiversity at the bottom of Jakarta Bay, especially in industrial areas (Takarina & Adiwibowo 2011). The same occurrences might happen in the Jatiluhur Reservoir, especially for Cu and Pb which concentrations have reached high level of contamination. Qu *et al.* (2010) stated that even though contamination by heavy metals is low in the sampling area, the adverse impact on benthic organisms are significant, suggesting that the chronic effects of long-term exposure to heavy metals in aquatic communities could be serious. Thus, it is essential to focus our attention and efforts to handle contamination of Hg, Cu, and Pb in the Jatiluhur Reservoir.

Water Quality Parameter Conditions

In general, water quality parameters in our study met the quality standards (QS) based on the Government of the Republic of Indonesia Regulation No. 82 of 2001 (Class 3). Based on direct measurements on-site for all sampling locations, the temperature ranged from 26.5 °C up to 27.1 °C (QS ± 3), Dissolved Oxygen (DO) concentrations ranged from 4.2 mg/L up to 4.5 mg/L (QS min. 3 mg L), and the water pH ranged from 6.37 up to 7.17 (QS 6-9). Results of the laboratory analysis on the other parameters of water quality are presented in Figure 2.

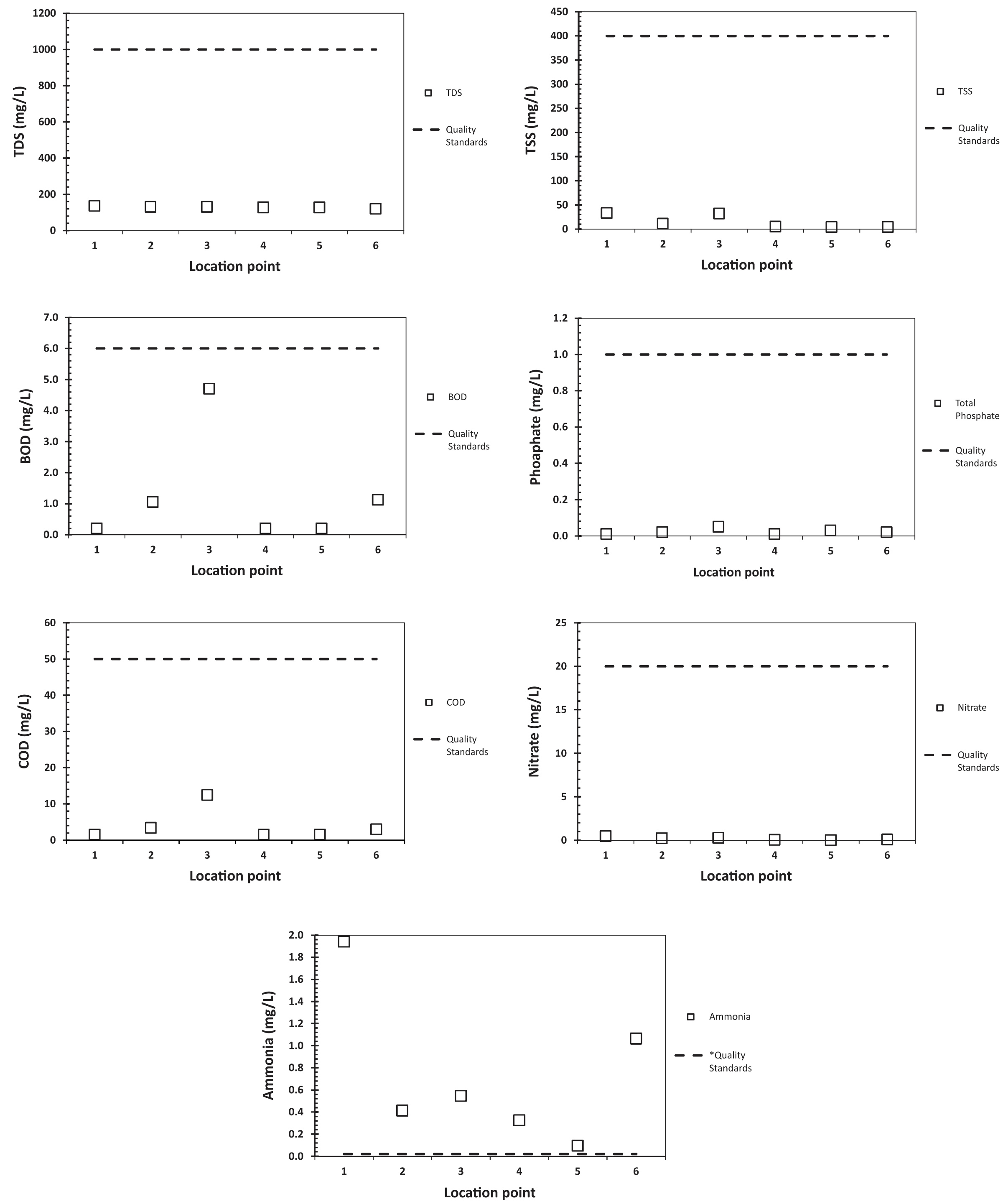

Figure 2 Values of several water quality parameters measured in the Jatiluhur Reservoir
Note: Quality Standards based on the Government of the Republic of Indonesia Regulation No. 82 of 2001 (Class 3), except for ammonia which was not available.

Results of our study showed that TDS concentrations ranged from 120 mg/L up to 136 mg/L (QS 1,000 mg/L), TSS concentrations ranged from 4 mg/L up to 33 mg/L (QS 400 mg/L), BOD concentrations ranged from 0.2 mg/L up to 4.7 mg/L (QS 6 mg/L), COD concentrations ranged from 2 mg/L up to 12 mg/L (QS 50 mg/L), total phosphate concentrations ranged from 0.01 mg/L up to 0.10 mg/L (QS 1 mg/L), nitrate concentrations ranged from 0.01 mg/L up to 0.47 mg/L (QS 20 mg/L) and ammonia concentrations ranged from 0.095 mg/L up to 1.940 mg/L (QS 0.02 mg/L).

Of the ten observed water quality parameters (not including heavy metals concentrations), only ammonia that did not meet the quality standards of water quality parameters. Ammonia is a nitrogen compound that changes to NH_4 ions at low pH conditions. Ammonia can also come from domestic and industrial wastes (Marganof 2007).

The high content of ammonia in the reservoir was presumably due to the fish feed and fish fecal wastes as a result of aquaculture activities in the reservoir. Fish emit 80-90% ammonia (N-inorganic) through the osmoregulation process, while feces and urine account for 10-20% of total nitrogen (Rakocy *et al.* 1992 in Sumoharjo 2010). Ammonia in the reservoir can come from organic and inorganic nitrogen sources found in soil and water or from the decomposition of organic matters by microbes and fungi. Ammonia also comes from the denitrification process during the decomposition of wastes by microbes under anaerobic conditions (Effendi 2003). Commonly, the concentration of Ammonia in the pond should not exceed 0.05 mg/L. According to SNI 6139:2009 (National Standardization Agency 2009), the ammonia value resulting from the nile tilapia aquaculture activity in calm water ponds should not exceed 0.02 mg/L. Ammonia concentrations of 0.02-0.07 mg/L have been shown to inhibit growth and cause tissue damage in several fish species. The toxicity threshold value for ammonia is highly dependent on the type of species, size, fine solids, surface-active compounds, metals and nitrates (Colt 2006).

Based on Anas *et al.* (2017), the water quality status of the Jatiluhur Reservoir is classified as moderate. The moderate status was resulted from the STORET calculation, stated in the Decree of the Minister of Environment No. 115 of 2003) which was caused by high concentrations of BOD, COD and ammonia. The main contributor to the high concentrations of organic matter in the Jatiluhur Reservoir presumably are the number of operating KJA in the reservoir. Based on the Regent of Purwakarta Regency Decree No. 6 of 2000, the optimal number of KJA to operate in the Jatiluhur Reservoir is 2,100 plots. Meanwhile, in 2015 there were 18,038 KJA in the reservoir, which exceeded the carrying capacity of the waters (Astuti *et al.* 2016). A recent study conducted by Fitri *et al.* (2016) stated that in 2014 the number of intensive KJA in the Jatiluhur Reservoir was already excessive, amounting to 23,000 cages. According to their study, the optimal number of KJA was 19,401 plots. Moreover, Fitri *et al.* (2016) stated that the difficulties faced by the Jatiluhur Reservoir related to the KJA problem were caused by multi-parties' management having different perspectives leading to inconsistent decision making. Harmonious perspectives and visions of all managing parties are needed to maximize the productivity of the KJA without sacrificing environmental quality.

CONCLUSION

The Jatiluhur Reservoir has experienced the accumulation of Cu, Zn, Pb, and Cd in the sediment layer with exceeding the minimum limit (ISQG) and Hg with exceeding the maximum limit (PEL). Spatially, Hg concentration was high in the entire Jatiluhur Reservoir area, Cu was found to accumulate in the inlet area, whereas Zn and Pb were relatively high in the main inundation areas. Cadmium (Cd) seemed to be relatively the safest metal in sediment compared to the other heavy metals. Heavy metal concentration in the waters of Jatiluhur Reservoir was below the detection limit of the tool. The other water quality parameters also met the standard of water quality. Only ammonia did not meet the quality standards for the life of sensitive fish (such as Nile tilapia). The high concentration of heavy metals in the sediment of the reservoir was due to household and/or industrial wastes, while the high concentration of ammonia in water was due to fecal materials from the aquaculture activities. Benthic organisms may have been affected by the high concentration of heavy metals in the sediment of the reservoir. Based on the high level of heavy metal concentrations in the Jatiluhur Reservoir, the priority for further attention and countermeasure in the reservoir was toward Hg, Cu, Pb and ammonia. Further studies are recommended to manage the water quality of the Jatiluhur Reservoir.

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