Spatial distribution and heavy metal pollution analysis in the sediments of Garang watershed, Semarang, Central Java, Indonesia

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Abstract. Garang watershed is located in Central Java, Indonesia. Various activities around the Garang watershed cause river pollution, by producing heavy metal waste, which can accumulate in the sediment, being absorbed from the water by organisms. This study was conducted to determine the concentration of heavy metals in the sediments, map the spread in the Garang watershed (Banjir Kanal Barat River, Garang River, Kreo River), and to determine the state of sediment pollution with heavy metals. The sediment samples were collected from 7 stations, with 3 replicates for each station. Samples of sediment were analyzed with Atomic Absorption Spectrophotometry (AAS), then the concentration of metals was mapped according to the sampling location. The sediment pollution index was determined. The results showed that the metal concentrations in sediment varied, from concentrations below the detection limit to tens of thousands of ppb. Lead had the highest concentration, followed by Cr and Cu. Cd and Zn were under the detection limit. The highest lead concentrations were recorded in the Banjir Kanal Barat (BKB) River and the lowest in the Kreo River. The highest Cu concentrations were in the Garang River and the lowest in the Kreo River. The highest concentrations of Cr were in the BKB River and the lowest in the Kreo River. Pb and Cr have similar spatial distribution patterns and differ from the spatial spread of Cu. The sediment pollution index indicated that the sediments of Garang watershed, BKB River, Garang River, and Kreo River had not been contaminated with heavy metals.

Key Words: Banjir Kanal Barat, Kreo, Garang, river, sediment pollution index.

Introduction. Garang watershed stretches from the foot of Ungaran Mountain in Semarang Regency, through 3 sub-districts in Semarang Regency (Bergas, West and East Ungaran), 2 sub-districts in Semarang City (Gunung Pati and West Semarang) to the Java Sea. Garang watershed consists of 4 main rivers, namely the Kreo River, Kripik River, Garang River, and Banjir Kanal Barat (BKB) River.

The rivers of Garang watershed have various functions, such as media transportation, aquaculture, watering farmlands, drinking water resources, and tourism. Various activities around the Garang watershed caused river pollution (Ujianti et al 2018; Haeruddin et al 2019a). Studies conducted by the MMFA (2015) showed that the concentration of metals in the waters from the mouth of Garang watershed has exceeded the water quality standard for marine water and marine biota.

The Government of Semarang and Central Java has been working to control the quality of the water in the Garang watershed through the activities of the Clean Water River Program (PROKASIH), targeting the industries located around the Garang watershed. Marlena (2012) stated that the dominant industries in the target of PROKASIH Garang watershed were the metal industry (50%), food and beverage (20%), pharmaceuticals (20%), and vegetable oils, tiles, and textiles (10%). Various types of metal industry produce heavy metal waste, which can be absorbed from the water by the dissolved particulate materials, and settle in the sediment. Therefore, heavy metals accumulate in the sediment in great concentrations (Bartoli et al 2012; Fu et al 2014). The metals often detected in sediments include iron, manganese, lead, cadmium, zinc, and mercury (US-EPA 2004). The metal concentration in the sediments is usually 3 to 5 times higher than the metal concentration in the water (Bryan & Langston 1992). Therefore, the identification of various metals derived from various sources can be faster by analyzing the sediment than quantifying the metal concentration in the water (Forster & Wittmann 1981).

Sediment can store a wide range of metals in large and consistent quantities. Therefore, the sediment can be used as a reference to determine the status of pollution of dynamic waters, such as rivers and seas (Haeruddin 2006). The spread of metal concentrations in the sediment is influenced by several factors. The highest metal concentrations in the sediment are found in adjacent locations to the source of the contamination, with a high rate of sediment accumulation (ten Brink et al 1997). Some factors that may affect the distribution of pollutants in the sediment are the size of sediment grain, redox state, concentration of organic carbon, and bioturbation (Meador et al 1998). The concentration of heavy metals in the sediments is not only determined by the weathering process of rocks, but also influenced by the concentration of organic matter, the composition of minerals, and the size of mud deposit particles (Togwell 1979). The mobility and bioavailability of metal fractions in the sediment are closely related to the geochemical characteristics of sediment, among others: organic matter, sulfide, Fe-Mn oxide, and redox potential (Zhang et al 2014).

The objective of this research was to determine the concentration of heavy metals, map their distribution in the sediment of the Garang watershed, and to determine the status of sediment pollution based on the Sediment Pollution Index (SPI), according to the Haeruddin method (Haeruddin 2006).

Material and Method. The research was conducted from July to November 2019. Sediment samples were obtained from 7 stations (Figure 1), located on 3 rivers, Banjir Kanal Barat River, Garang River, and Kreo River (Figure 1). At each station, the sediment sample was collected 3 times. Sediment samples were collected using a teflon-plated grab to prevent metal contamination of the grab compound material. Each sediment sample had 1.5 kg. After the sediment sample was removed from the grab, it was placed in a plastic container and labeled.

Moreover, the plastic containers were placed into a cool box filled with ice cubes and blue ice and transported to the Laboratory of Fish Resources and Environment Management. In the laboratory, the sediment samples were placed in a freezer at −10°C. The heavy metals analyzed in this study were: Cd, Cr, Cu, Pb, and Zn. The metal concentrations in the samples were analyzed by using atomic absorption spectrophotometry (AAS) according to APHA (1989) part 3111 C: Extraction/Air-Acetylene Flame Method. The calculated concentration of each metal was expressed in $\mu g\ L^{-1}$ and refers to the appropriate calibration curve.

Metal concentration mapping in the sediment of Garang watersheds was carried out by plotting the coordinates of the sampling location of sediment into the map of the research locations and the concentration of metals in each sample. Mapping was carried out using ArcGIS software.

The SPI is set based on the modification of the arithmetic weighted formula developed by SDD (1976). The highest value of the index is 5 and the lowest value is 0. The meaning of the value is as follows (Haeruddin 2006): 4.01 to 5 - heavily contaminated (highly polluted); 3.01 to 4 - moderately polluted; 2.01 to 3 - lightly polluted; 1.01 to 2 - contaminated; 0 to 1 - not contaminated.

The index value range is obtained from simulated computer results using Microsoft Excel software. The lowest index weight value used in the simulation was 0. Each weighted index is multiplied by the quality rating of sediment from 1 to 5. The obtained index value range is then checked for its spread. If the value of the obtained index is spread normally, the index value is divided into the top percentile of 5 classes, according to the highest number of indexes. The results of the dividing index value of 0-1 are in the

1st to the 20th percentiles range, the 1.01-2 values are in the 21st to the 40th percentiles range, and so on, until the 4.01-5 index value is at the 81st to 100th percentile range.

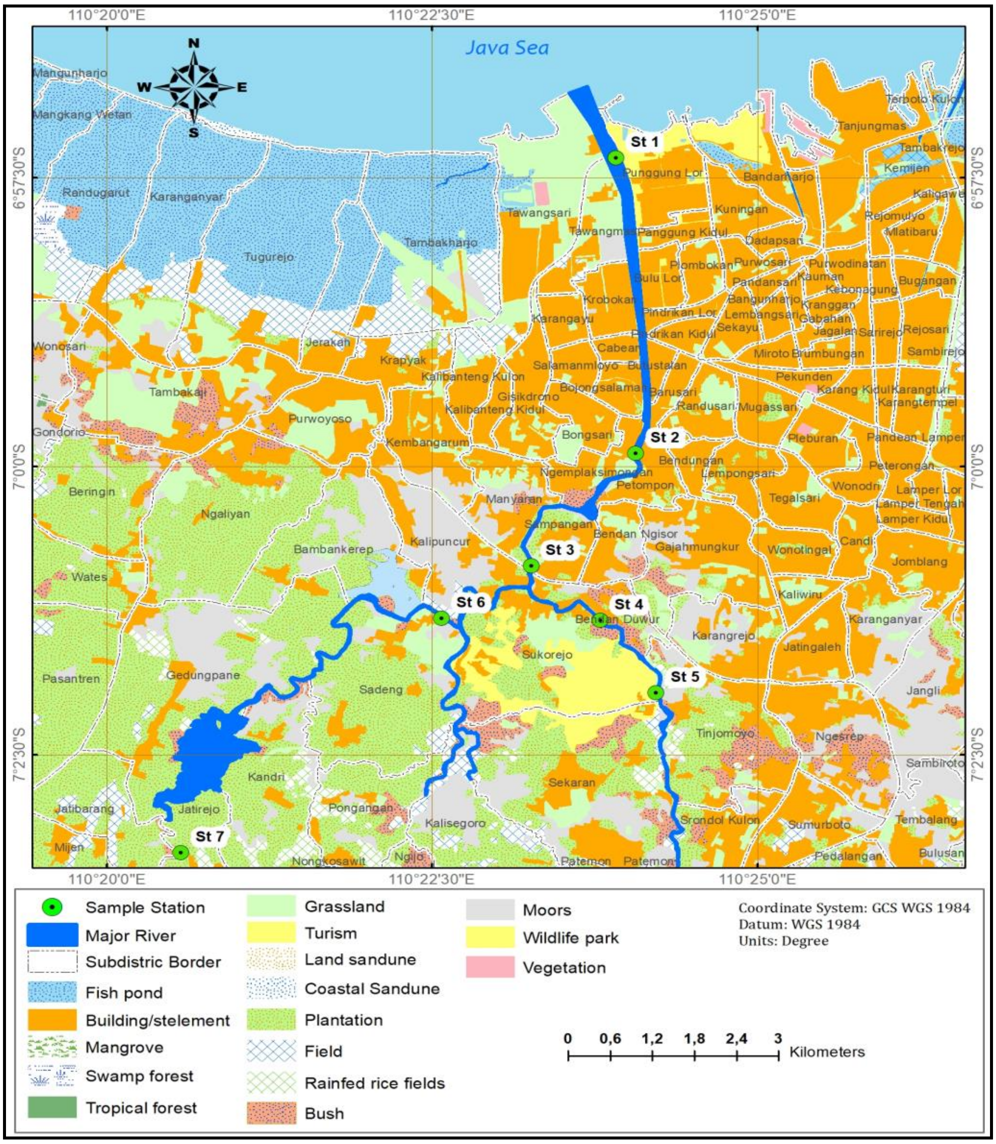

Figure 1. The location of the sediment sampling station.

The value of SPI is calculated using the following formula (Haeruddin 2006):

$$SPI = \frac{(\sum Q_i \times W_i)^2}{5}$$

Where: SPI - Sediment Pollution Index; Q_i – the rating of sediment quality variable i; W_i – the weight of sediment quality variable i; the variables i were: Cd, Pb, Zn, Cu, and Cr.

The SPI is the square of the summation of multiplication between the weights of all sediment quality variables with the rating of all sediment quality variables divided by 5. The weight of the sediment quality variable i is obtained through multiplication between the relative Eigen vector and the relative eigenvalue of the same main component. The relative Eigen vector feature is the result of the relative Eigen vector of the sediment feature on the main component with the sum of all the relative Eigen vector of the selected feature (i. e. the relative Eigen vector of various sediment quality changes that have a load value higher or equal to 0.7). Similarly, the relative Eigenvalue is the result of the selected value of the sediment Eigenvalue with the amount of sediment quality Eigenvalue in one particular key component. The sediment quality variable selected for index counting is sediment quality assessment with an absolute Eigenvalue greater than 0.7, as recommended by Comrey & Lee (1992).

The rating of sediment quality variable is one of the components that need to be known in the determination of the SPI, besides the weight of quality sediment. The quality rating of the sediment is carried out based on the sediment toxicity using the range from the Threshold Effect Level (TEL) to the concentration level that causes the effect - probable effect (PEL). The concentration of sediment variables below the TEL value is rated 1, between TEL and PEL, concentrations are rated 2 to 4, and concentrations above PEL value are rated 5.

The TEL and PEL values are obtained from the sediment quality guidelines from NOAA (1999) through the National Status and Trends (NS&T) Program. TEL and PEL are used to identify 3 ranges of chemical concentrations related to biological effects that are caused by water biota. The TEL can be used as interim sediment quality guidelines (ISQGs) (CCME 2001). The concentration ranges are: lower than TEL - the concentration range rarely poses a detrimental effect to freshwater biota; between TEL and PEL - the concentration range may cause adverse effects on freshwater biota; PEL - the concentration range causes adverse effects on water biota.

Based on the TEL and PEL values for the metal and organic elements of the NOAA (1999) criteria for the rating of metals were compiled (Table 1).

Table 1

Metal rating criteria

SQR	*Pb (ppb)*	*Cd (ppb)*	*Zn (ppb)*	*Ni (ppb)*	*Cr (ppb)*	*Cu (ppb)*
1	<35000	<596	<123100	<18000	<37300	<35700
2	35000<x≤ 56433	596<x≤ 1574	123100<x≤ 187067	18<x≤23967	37300<x≤ 54867	35700<x≤ 89467
3	56433<x≤ 77867	1574<x≤ 2552	187067<x≤ 251033	23967<x≤ 29933	54567<x≤ 72433	89467<x≤ 143233
4	77867<x≤ 99300	2552<x≤ 3530	251033<x≤ 315000	29333<x≤ 35900	72.433<x≤ 90000	143233<x≤ 197000
5	>99300	>3530	>315000	>35900	>90000	>197000

Note: source - NOAA (1999); SQR - sediment quality rating.

Results and Discussion. The metal concentration in sediment samples at each station is presented in Table 2. Pb had the highest concentration in the area of the Garang watersheds, followed by Cu and Cr, while Cd and Zn were not detected (Pb>Cu>Cr>Cd/Zn).

Table 2

Concentrations of different types of metals in the sediment (ppb) of 3 rivers of the Garang watershed

Location	*Limit*	*Cd (ppb)*	*Cr (ppb)*	*Cu (ppb)*	*Pb (ppb)*	*Zn (ppb)*
Banjir Kanal Barat River	Upper	0.005	11400	42700	100600	0.005
	Lower	0.005	0.005	22600	0.005	0.005
	Mean±SD	-	5333.34± 4277.56	32288,89± 7612.56	36866.67± 35400.74	-
Garang River	Upper	0.005	9500	54900	78700	0.005
	Lower	0.005	0.005	22300	0.005	0.005
	Mean±SD	-	4500± 4938.01	33483.33± 11275.71	17300± 30715.34	-
Kreo River	Upper	0.005	5000	28500	41900	0.005
	Lower	0.005	0.005	0.005	0.005	0.005
	Mean±SD	-	1266.67± 2103.96	19533.33± 11658.41	17900± 20471.05	-

Note: SD - standard deviation.

The highest Pb concentration was recorded in the BKB River and the lowest in Kreo River, a situation similar to that of Cr. Pb and Cr have similar spatial distribution patterns and differ from the Cu spatial distribution. The mapping of the metal spatial distribution at various sampling stations is presented in Figure 2.

The results of weighted and rated metal concentrations are presented in Tables 3 to 6.

Table 3
Weights and rating of various types of metals in the sediment of Garang watershed

Principal component	*Eigenvalue*	*Relative Eigenvalue*	*Metal*	*Eigen vector*	*Relative Eigen vector*	*Weight*	*Rating*
1	973180291	0.8997269	Pb	0.979	0.198379	0.178487	1.611
2	92634692	0.0856428	Cu	0.968	0.19615	0.016799	1.167
3	15824633	0.0146302	Cr	0.988	0.200203	0.002929	1
4	0	0	Zn	1	0.202634	0	1
5	0	0	Cd	1	0.202634	0	1
Total	1.082E+09	1		4.935	1		

The calculation result of the SPI is 0.0008, indicating that sediment in the Garang watershed is not contaminated by metals. The result of the calculation of the BKB River sediment pollution index is 0.0012, indicating that BKB River sediment status has been uncontaminated by metals.

Table 4
Weights and rating of various metals in Banjir Kanal Barat River sediment

Principal component	*Eigenvalue*	*Relative Eigenvalue*	*Metal*	*Eigen vector*	*Relative Eigen vector*	*Weight*	*Rating*
1	1266432796	0.9526714	Pb	0.995	0.201417	0.191884	2
2	54868873	0.041275	Cu	0.972	0.196761	0.008121	1.333
3	8047287	0.0060536	Cr	0.973	0.196964	0.001192	1
4	0	0	Zn	1	0.202429	0	1
5	0	0	Cd	1	0.202429	0	1
Total	1329348956	1		4.94	1		

The SPI value of the Kreo River is 0.0007. It indicates that the Kreo River sediment has been polluted, but with an uncontaminated status.

Table 5
The weights and rating of various types of metals in the Kreo River sediment

Principal component	*Eigenvalue*	*Relative Eigenvalue*	*Metal*	*Eigen vector*	*Relative Eigen vector*	*Weight*	*Rating*
1	1062640700	0.970602	Pb	0.941	0.217623	0.211225	1.333
2	20016464	0.018283	Cu	0.72	0.166512	0.003044	1
3	12169718	0.011116	Cr	0.663	0.15333	0.001704	1
4	0	0	Zn	1	0.231267	0	1
5	0	0	Cd	1	0.231267	0	1
Total	1.095E+09	1		4.324	1		

The result of the calculation of the Garang River sediment pollution is 0.0007. This is indicating that the Garang River sediment has been polluted with an uncontaminated status.

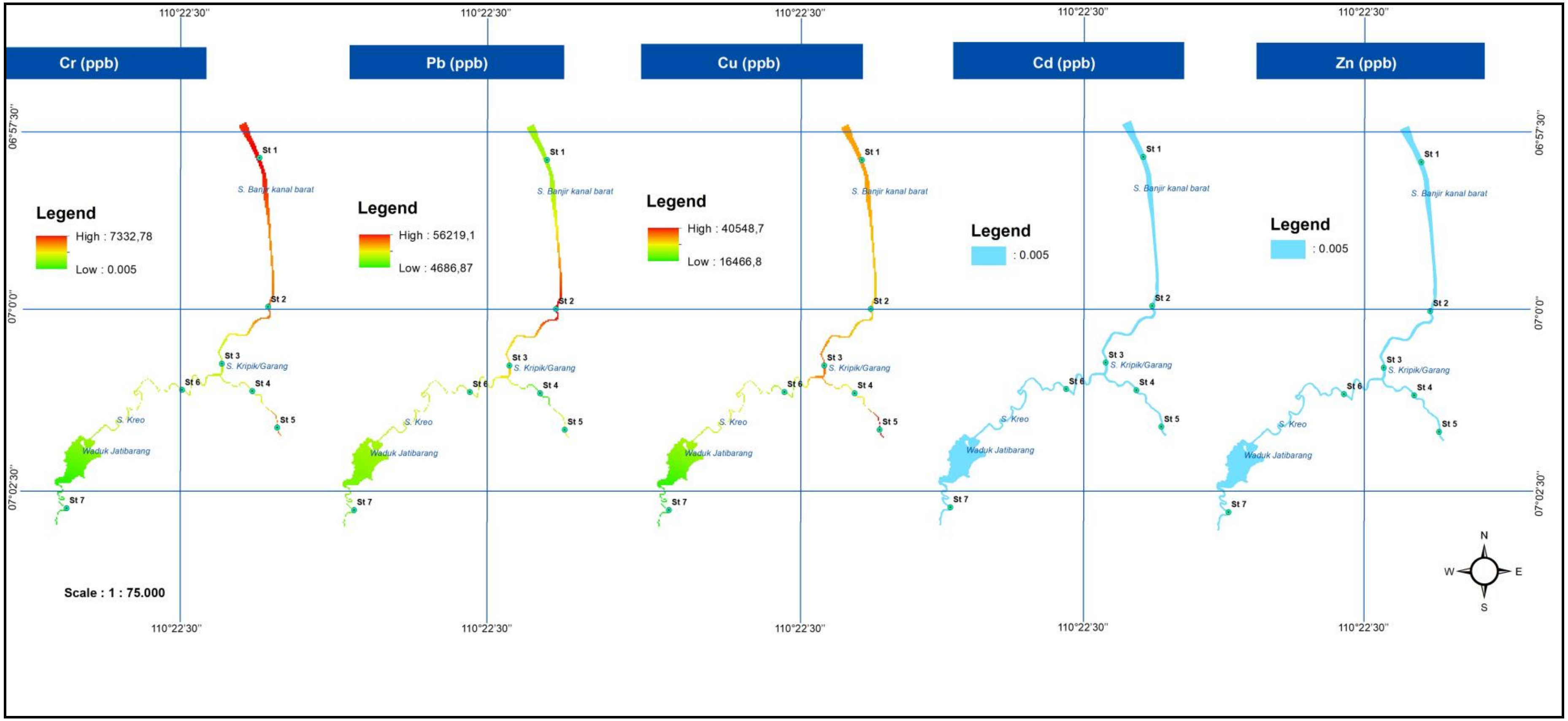

Figure 2. Spatial distribution of various metals in the sediment of the Garang watershed.

Table 6

Weights and rating of different metals in the Garang River sediment

Principal Component	*Eigenvalue*	*Relative Eigenvalue*	*Metal*	*Eigen vector*	*Relative Eigen vector*	*Weight*	*Rating*
1	973180291	0.899727	Pb	0.979	0.198379	0.178487	1.5
2	92634692	0.085643	Cu	0.968	0.19615	0.016799	1.167
3	15824633	0.01463	Cr	0.988	0.200203	0.002929	1
4	0	0	Zn	1	0.202634	0	1
5	0	0	Cd	1	0.202634	0	1
Total	1.082E+09	1		4.935	1		

The measured metal concentration in the sediment is higher than the one measured in the waters. Similar results were obtained by Gawad (2018) in his research in Lake Manzala, Egypt, and Sabbir et al (2018) for Rupsha River, Bangladesh. Bryan & Langston (1992) suggested that the metal concentration in the sediments is usually 3 to 5 times higher than the metal concentration in the water.

Cu concentrations in BKB River water ranged from 0.041 mg L^{-1} to 0.5 mg L^{-1}. This concentration has exceeded the Indonesian Water Quality Standard for Freshwater Biota. The concentrations of Cd and Pb were under the detection limit (Haeruddin et al 2019a). Ujianti et al (2018) also measure the concentrations of Cr (0.007-0.025 mg L^{-1}) and Cu (undetectable - 0.15 mg L^{-1}) in the area. The concentrations of Cu and Cr in this study were lower than in the Brantas River. The average Cu and Cr concentrations in the Brantas River are 49 and 50 ppm, respectively (Mariyanto et al 2019). This is likely due to the higher intensity of activities around Brantas River compared with Garang River.

The pattern of heavy metal spread in the Garang watershed can indicate that the sources of Cr and Pb are the activities from upstream. Cu sources allegedly originate from settlements and weathering rocks around the Garang River. The Kreo River has a low concentration of some metals compared to other rivers, due to less human activity around the lower Kreo River.

Land utilization in the upstream of Garang watershed consists of rice fields, settlements, dryland agriculture (dry fields), plantations, and forests (CBS Semarang City 2018a). According to BBWS Pemali Juana (2015), the biggest land cover upstream of the Garang sub-watershed is for dryland agriculture with an area of 2772.02 Ha (33.11%), followed by settlements with 2259.34 Ha (26.99%), and rice fields with 1706.52 Ha (20.59%). The greater size of settlements indicates a functional shift upstream of the Garang River sub-watershed in Semarang regency (Efendi et al 2012). West Ungaran sub-district has a total land area of 3596.03 Ha, and 2/3 are farmland (CBS Semarang Regency 2018). The central part (Gunung Pati sub-district) is dominated by rice fields, moorings, and yards. Furthermore, the downstream (West Semarang sub-district) is dominated by yards and other uses, especially ponds (CBS Semarang City 2018a, 2018b; Wahyuningtyas et al 2017). In Gunung Pati and West Semarang sub-districts, there are also several licensed industries around the BKB River in the Simongan Industrial Area, including the pharmaceutical industry, steel pipe industry, spinning, and various other industries (Kurniawati & Rengga 2016). Agriculture, industrial and domestic wastes are the main anthropogenic sources of heavy metal in the river ecosystem and they are increasing due to the increase in population (Zhang et al 2014; Feng et al 2017; Wang et al 2017; Goher et al 2019). The heavy metal waste from various types of metal industry can be absorbed from the water by the dissolved particulate materials, settle and accumulate in the sediment in great concentrations (Bartoli et al 2012; Fu et al 2014).

The heavy metals are enriched in the sediments through adsorption, complexation, flocculation, and sedimentation (Forstner & Muller 1973; Wang et al 2016). In the event of a change in environmental conditions, the dynamic equilibrium of the water-sediment interface will be broken, and the heavy metals in the sediment will be transferred, transformed and released to the overlying water, which will lead to the pollution of water (Tao et al 2012; Zhang et al 2012; Islam et al 2015). The migration and transformation of heavy metals occur through the mechanisms of dissolution, and desorption (Duddridge & Wainwright 1981; Lin & Chen 1998). Among a variety of influential factors, pH is one of the main factors, and the effect of pH on the speciation of heavy metals is of great significance to the migration and transformation of metals (Riba et al 2004). The change of pH conditions in the system will have a certain impact on the migration and distribution of heavy metals (Gabler 1997).

Pb enters the watersheds, such as the Garang watershed, from activities like lead melting or refining, fuel combustion containing lead additives, and other metal smelting (WHO 1995). Tetraethyl lead and tetramethyl lead are widely used as additives in fuel, both volatile and difficult to dissolve in water. Trialkyl lead is formed in the environment by a tetraalkyl lead reshuffle. Trialkyl compounds are less volatile, and can easily dissolve in water.

The main natural source of Cu is the erosion of mineral rocks generally happening in the river, while the sources derived from human activities are local, especially waste disposal (Libes 1992). Cu is used for power cords and electroplating, mixed metal production, pipes, photography, anti-body stamped paint, and pesticide formulation. The sources of Cu are mining, metal smelting, refining, and coal-burning industries. Cu enters aquatic environments through natural sources, such as rock weathering or soluble Cu minerals (CCREM 1987).

Almost all hexavalent Cr in the environment comes from human activity. Cr compounds are used in manufacturing ferrochrome, metal plating (electroplating), pigment formation, and tanner. Burning fossil fuels and burning waste are a source of Cr in water and air. The Cr cycle in the environment starts from rocks and continues to soils, water, biota, air, and finally, back to the ground. Nonetheless, most Cr (estimated at 6.7x106 kg per year) is released into rivers, by runoff, piling in the seas (WHO 1988).

Metal concentrations that are generally low in sediment cause the rating value and the weight gained to be small. Low metal concentrations cause low sedimentary toxicity, so that the value of the toxic rank is generally 1. This leads to a small SPI value, less than 1. Thus, the sediment is categorized as not contaminated. This condition has been reported by Haeruddin et al (2019b), who stated that the quality of the sediments of Garang watershed is still in good condition.

Conclusions. The metal concentrations in the measured sediments varied greatly, from concentrations below the detection limit to tens of thousands of ppb. Pb had the highest concentration, followed by Cr and Cu. Cd and Zn concentrations were under the detection limit. The highest Pb concentration was obtained in the BKB River and the lowest in the Kreo River. The highest Cu concentration was in the Garang River and the lowest in the Kreo River, while the highest concentrations of Cr were in the BKB River and the lowest in the Kreo River. Pb and Cr had similar spatial distribution patterns and differed from the spatial spread of Cu. The SPI values indicate that the sediment of Garang watersheds, BKB River, Garang River, and Kreo River, were not contaminated with heavy metals.

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