Status of heavy metal in sediment of Saguling Lake, West Java

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Abstract The quality of water in the Saguling lake has been monitored since 1990 by the state agency for environmental control. However, no data on heavy metal in sediment had been reported. Metal pollutants have receives considerable attention due to their persistence, biogeochemical recycling, and environmental risk. The objective of this study was to assess the level contamination of heavy metals (Cd, Cr, Cu, and Pb) in the surface sediment of the Saguling Lake West Java. Surface sediment samples were collected from 10 location of the Saguling Lake on July and November 2015. The concentration of heavy metals in the surface sediment on July and November 2015 decreased in the order of Cu>Cr>Pb>Cd and Cr>Cu>Pb>Cd respectively. Mean metals concentrations (mg/kg) in July 2015 were in the range of: Cd: 10.69-16.65, Cr: 76.67 -138.38, Cu:106.02-229.54, Pb: 23.93-80.17. Mean metals concentrations (mg/kg) in November 2015 were in the range of Cd: 18.64-23.25, Cr: 152.16-197.98, Cu: 63.32-152.53, Pb: 20.31-32.74. Geochemical approaches such as contamination factor, and pollution load index were exploited for the assessment of the contamination and enrichment level of heavy metals in the lake sediment. Contamination factor and pollution load index values indicated that surface sediments around Saguling Lake were polluted with heavy metals. The finding of this study would help in formulating guidelines to control the pollution and suggested for Saguling lake revitalization.

1. Introduction
Lake Saguling was built on The Citarum River. The Citarum Watershed is home to the largest industrial area in West Java Province. Several economic activity are performed along Citarum River, including mining and agriculture. Originally, the Saguling was planned for a single purpose lake to generate electricity. Later, considering the environmental problems of the area, the Saguling was re-planned as a multipurpose lake such as raw water, fisheries, aquaculture, and tourism [1].

Lakes are very important and significant bodies in preserving fresh-water and replenishing underground water, also act as a key role in adjusting local climate and ameliorating environment, consequently, they are considered one of the most versatile ecosystems in the world [2]. Recent years, with the rapid urbanization and intensive development of industry, the pollution of the lake environment with heavy metals has become a worldwide problem because they are indestructible and most of them have toxic effects on organisms [3] Heavy metals enter in lake from natural and anthropogenic sources [4]. Generally, in unaffected environment, most of heavy metals are in very low concentration, and natural geological weathering of rocks and soil, directly exposed to surface waters, is usually the largest natural source [5].

Main anthropogenic sources of heavy metal contamination are mining, disposal of untreated and partially treated effluents contain toxic metals, as well as metal chelates from different industries and
indiscriminate use of fertilizer containing heavy metals [6], [7]. Heavy metals discharged in to aquatic system by natural or anthropogenic sources during their transport are distributed between the aqueous phase and sediments. Because of adsorption, hydrolysis and co-precipitation only a small portion of free metal ions stay dissolved in water and a large quantity of them are deposited in these sediment. Although heavy metals are refractory though natural processes in the environment, they can be chemically altered by organisms and converted into organic complexes, some of which may be more hazardous to animal and human life [8]. While metals settled in sediments may be re-suspended and cause secondary contamination to the water environment, because sediments act both as a sink and a source for metals in the aquatic environment [9].

Over the past decade, a number of indices have been developed to assess heavy metal contamination of sediments. These indices, mainly including sediment quality guide-lines (SQGs), the relative enrichment factors (REF), the pollution load index (PLI) and index of geo-accumulation (I-geo), provide useful information and could be easily communicated to local managers and decision makers [10].

Hence, the main aspects of the present work are to (1) determine the content of heavy metals (Cd, Cr, Cu, and Pb) in Saguling lake surface sediment. (2) Calculate the pollution load index (PLI) in order to assess quality of the sediment.

2. Experimental

2.1. Study area

The Saguling is an artificial lake. The lake was built on the Citarum River, the largest river in West Java Province, in February 1985. The Saguling Lake area is 4,710 Ha the water storage capacity is 730.5 million m³. The Saguling Lake is an embankment Lake on the headwater of Citarum River in West Java, Indonesia. The primary purpose of the lake is hydroelectric power generation but it also provides for water supply and aquaculture. The surrounding area of Saguling lake is hilly, while the river has many tributaries at this location. This makes the shape of Saguling lake very irregular or dendritic, with many extended bays. The lake area was formerly densely populated by farmer population with extensive agricultural lands. The catchment areas of the lake or the upper Citarum River basin are faced with high population pressure. This is because over 50% of the population consists of farmers with a high annual growth rate (2.34% is the national average). The growth of farmer population has caused the decrease of landholding and this condition forced them to extend their agricultural land by forest clearing and utilize marginal lands. As a consequence, there is a prevailing problem of floods accompanied by accelerated soil erosion in rainy seasons. Due to the high population density in the upper catchment of the lake, extensive agricultural land, soil erosion and the presence of industries, the lake water became polluted and eutrophic. The growth of aquatic weeds has been accelerated, with frequent blooms of Microcystis algae. The water is also contaminated by heavy metals, pesticides, etc. An extensive growth of water-hyacinth is maintained by fencing to reduce pollutant contents of water at inlets of the Citarum River [11].

2.2 Sample collection and chemical analysis

Surface sediment samples were collected from ten different sampling in Saguling lake in July and November 2015, sampling location are presented in Table 1. The point chosen because of their classification as having poor water quality according to water monitoring report Indonesia Power as a Saguling lake authority. The sampling locations were recorded (latitudinal and longitudinal position) using hand-held Global Positioning System (GPS). Two stations (1A and 1B) were located in the Citarum River, eight stations (2-9) located in the Saguling lake.

Surface Sediment samples were collected at each station by mixing four to five random samples, whereas composite sediments were collected from the top 10 cm of the surface with a grab sampler. Samples were then placed into polyethylene bags, and refrigerated at 4°C. They were analysed in the laboratory. Each sediment sample was dried at 50°C, the samples were then grounded using pestle and mortar until all particles passed a 200-mesh nylon sieve. This fraction is mainly contained silt and
clay; thus, this fraction may adsorb a higher heavy metal concentration. The extraction of heavy metals were made using conventional digestion procedure of sediment samples, which consists of digesting aliquots of 1 g of dry sediment was weighted into Teflon beakers, to which a mixture of concentrated HNO₃ : HCl (3:1) was added and digested in a water bath during 3 h at 120°C. The concentration of Cd, Cr, Cu, and Pb were determined for each sample using an Inductively Coupled Plasma Optical Emission Spectrometry or ICP-OES.

Table 1. Sampling stations in Saguling lake.

| Station | Location                                | Coordinate       |
|---------|-----------------------------------------|------------------|
| 1A      | Citarum River Nanjung section           | 06°56'29.8" S   |
|         |                                         | 107°32'10.7" E  |
| 1B      | Citarum River section Trash Boom Batujajar | 06°54'58.9" S   |
|         |                                         | 107°28'35.0" E  |
| 2       | Cihaur Village Cipeundeuy               | 06°53'13.5" S   |
|         |                                         | 107°28'32.3" E  |
| 3       | Cimerang                                | 06°53'13.4" S   |
|         |                                         | 107°27'09.0" E  |
| 4       | Cihaur Estuary Maroko Village           | 06°54'13.0" S   |
|         |                                         | 107°25'54.4" E  |
| 5       | Cipatik Estuary                         | 06°56'07.6" S   |
|         |                                         | 107°27'25.5" E  |
| 6       | Ciminyak Estuary-fishing floating located | 06°57'14.6" S   |
|         |                                         | 107°26'03.8" E  |
| 7       | Cijere Estuary                          | 06°56'14.9" S   |
|         |                                         | 107°24'50.8" E  |
| 8       | Cijambu Estuary                         | 06°56'00.4" S   |
|         |                                         | 107°22'22.4" E  |
| 9       | Near Intake Structure                   | 06°54'54.4" S   |
|         |                                         | 107°22'26.3" E  |

3. Result and discussion

3.1. Heavy metals concentration in Saguling lake

The concentration of heavy metals (Cd, Cr, Cu, and Pb) in the sediment of the Saguling lake in July and November 2015 are presented in Table 2. The concentration of heavy metals in the surface sediment on July 2015 decreased in the order of Cu>Cr>Pb>Cd. Cu was the most abundant metal in the sediment, with measured concentrations ranging from 106.02 to 229.54 mg/kg follows by Cr (76.67-138.38 mg/kg), Pb (23.93-80.17 mg/kg), and Cd (10.69-16.65 mg/kg). The highest concentration of Cd, Cr, Cu, Pb found in the station 4, 1A, and 1B respectively, with a value of Cd 16.65 mg/kg, Cr 138.38 mg/kg, Cu 229.54 mg/kg, and Pb 80.17 mg/kg. The average value were determined to be as follows (mg/kg): Cd: 13.54±1.98, Cr: 100.83±21.94, Cu: 165.57±42.57, and Pb: 38.32±16.94. When compared with the standard based on ANZECC ISQG-Low for all heavy metals examined, only Pb concentration accordance sediment quality standard. Three heavy metals Cd, Cr, and Cu did not appropriated sediment quality standard in all sampling locations.

The concentration of heavy metals in the surface sediment on November 2015 decreased in the order of Cr>Cu>Pb>Cd. Cr was the most abundant metal in the sediment, with measured concentrations ranging from 152.16 to 197.98 mg/kg follow by Cu (63.32-152.53 mg/kg), Pb (20.31-32.74 mg/kg), and Cd (18.64-23.25 mg/kg). The highest concentration of Cd, Cr, Cu, Pb found in the station 4, 1A, and 1B respectively, with a value of Cd 23.35 mg/kg, Cr 197.98 mg/kg, Cu 152.53 mg/kg, and Pb 32.74 mg/kg. For all station, the average value were determined to be as follows (mg/kg): Cd: 21.08±4.63, Cr: 174.10±15.86, Cu: 100.22±33.04, and Pb: 26.12±3.78. When compared with the standard based on ANZECC ISQG-Low for all heavy metals examined, only Pb concentration accordance sediment quality standard. Three heavy metals Cd, Cr, and Cu did not appropriated sediment quality standard in all sampling locations.
Table 2. Range concentration of heavy metals in Saguling lake.

| Station | Heavy metal concentration in July 2015 (mg/kg) | Heavy metal concentration in November 2015 (mg/kg) |
|---------|---------------------------------------------|--------------------------------------------------|
|         | Cd  | Cr   | Cu   | Pb  | Cd  | Cr   | Cu   | Pb  |
| 1A      | 15.64 | 113.75 | 225.36 | 80.17 | 23.25 | 197.98 | 63.32 | 23.79 |
| 1B      | 16.65 | 125.09 | 229.54 | 51.61 | 22.92 | 184.39 | 152.53 | 25.79 |
| 2       | 13.27 | 81.00  | 178.66 | 42.06 | 20.92 | 160.10 | 135.35 | 29.11 |
| 3       | 12.86 | 106.18 | 175.55 | 25.69 | 20.62 | 169.78 | 64.96  | 20.31 |
| 4       | 16.05 | 138.38 | 182.88 | 35.00 | 22.90 | 191.02 | 87.25  | 32.74 |
| 5       | 11.31 | 76.67  | 119.93 | 36.90 | 22.00 | 174.25 | 82.92  | 28.92 |
| 6       | 12.88 | 83.94  | 106.02 | 31.65 | 19.15 | 157.97 | 132.80 | 27.90 |
| 7       | 10.69 | 82.74  | 116.70 | 23.93 | 20.61 | 189.56 | 83.04  | 25.18 |
| 8       | 13.36 | 83.74  | 158.95 | 30.15 | 18.64 | 152.16 | 73.36  | 21.18 |
| 9       | 12.70 | 116.83 | 162.09 | 26.10 | 19.81 | 163.75 | 126.71 | 26.27 |
| Mean    | 13.54 | 100.83 | 165.57 | 38.32 | 21.08 | 174.10 | 100.22 | 26.12 |
| Maximum | 16.65 | 138.38 | 229.54 | 80.17 | 23.25 | 197.98 | 152.53 | 32.74 |
| Minimum | 10.69 | 76.67  | 106.02 | 23.93 | 18.64 | 152.16 | 63.32  | 20.31 |
| Standard deviation | 1.98 | 21.94  | 42.57  | 16.94 | 1.63 | 15.86  | 33.04  | 3.78 |
| Standard* | 1.50 | 80.00  | 65.00  | 50.00 | 1.50 | 80.00  | 65.00  | 50.00 |

Total concentration Cd ranged from 10.69-16.65 mg/kg in July and 18.64-23.25 mg/kg in November. Sampling site 1B contained maximum Cd concentration. Higher Cd concentration might be related to industrial activity, atmospheric emission, and deposition of organic and fine grain sediments, leachates from defused Ni-Cd batteries and Cd plated items. The concentration of Cr in sediment ranged from 76.67-138.38 mg/kg in July and 152.16-197.98 mg/kg in November. The sampling point 4 located in Cihaur estuary area contains maximum amount of Cr in July. The sampling point 1A located in Citarum river Nanjung section contain maximum amount of Cr in November. The untreated Cr containing huge leather tanning wastes mix with the lake water might results in theses extreme Cr contamination. The US EPA has classified Pb as a probable human carcinogen. Average Cu content in the sediment was 165.57 and 100.22 mg/kg for July and November respectively. High level of Cu indicates its higher input in these sites, which might be originated from urban and industrial wastes. Lead is a good indicator of traffic related sources or battery recycling plant, and is considered as an indicator of pollution by urban runoff water [13]. A comparison of metal concentration in sediment with reference in our study can indicated that metal accumulation in sediment lake was either by natural and anthropogenic source.

Table 3 presented mean concentration of measured elements in lake sediment samples from different countries along with present study. Comparing with the data from other country, the heavy metals concentration in Saguling lake were found higher. Heavy metals concentration in Saguling Lake must be considered from the government because the lake Saguling functions are related to public health as a source of irrigation water, fisheries, and raw water sources of drinking water for Bandung Basin and surrounding region.
Table 3.
Mean concentration of measured elements in lake sediment samples from different countries along with present study.

| No | Name of the lakes                  | Heavy metals concentrations (mg/kg) | Cd    | Cr   | Cu   | Pb   |
|----|-----------------------------------|-------------------------------------|-------|------|------|------|
| 1  | East lake, Wuhan, China           |                                     |       | -    | 54.8 | 40.3 |
| 2  | Songkhla lake, Southern Thailand  | 0.1-2.4                             | 54.8  | 1.8  | 125  | 8.2  |
| 3  | Laguna lake Philippines           | 0.02-0.09                           |       | 9.7  | 18.7 | 17.23|
| 4  | Manchar lake, Pakistan            | 4.9-9.7                             | 14.7  | 15.6 | 29.7 | 14.6 |
| 5  | Taihu lake, China                 | 0.94                                | 56.2  | 36.7 | 51.8 |
| 6  | Hussainsagar lake, India          |                                     |       | 40   | 40   | 40   |
| 7  | Saguling lake, Indonesia          |                                     |       |      |      |      |
|    | July 2015                         |                                     | 10.69 | 76.67 | 106.02 | 23.93 |
|    | November 2015                     |                                     | 18.64 | 152.16 | 63.32 | 20.31 |

1-6 [14]; 7 present study

3.2. Assessment of sediment contamination in Saguling lake

The level of contamination of lake sediment by given toxic substance (metals) is often expressed in terms of a contamination factor and is calculated as follows [15].

$$\text{CF} = \frac{C_{\text{metal}}}{C_{\text{background}}} \quad (1)$$

Where, CF is the contamination factor, $C_{\text{metal}}$ is the concentration of pollutants in sediment, $C_{\text{background}}$ is the background value for the metals. Background concentration of Cd, Cr, Cu, and Pb was respectively 0.30 mg/kg, 90 mg/kg, 45 mg/kg, and 20 mg/kg [16]. With the help of contamination factor values the intensity of contamination can be inferred from Hakanson, 1980 as follows: low contamination, $\text{CF} < 1$; moderate contamination $1 < \text{CF} < 3$; considerable contamination, $3 < \text{CF} < 6$; and very high contamination, $\text{CF} > 6$. Pollution load index (PLI) is defined as the root of the product of the CF.

Tomlinson, 1980 has developed a simple method based on PLI to assess the degree of pollution by metals in aquatic sediments, PLI was calculated using the equation 2 where $n$ the number of metals. A value of 0 on the PLI would indicated no pollutants; a value of 1 would suggest the presence of baseline levels of pollutants, while value that exceed 1 would indicated a progressive deterioration of the site and lake [17].

$$\text{PLI} = (\text{CF}_1 \times \text{CF}_2 \times \text{CF}_3 \times \ldots \times \text{CF}_n)^{\frac{1}{n}} \quad (2)$$

Contamination factor value calculated is summarized in Table 4 Contamination factor value in Saguling lake. The contamination factor value Cd in July ranged between 35.62 to 55.48 with average value 45.13. The contamination factor value Cd in November ranged between 62.14 to 77.49 with average value 70.27. Based on contamination factor indicated that Saguling surface sediment in July and November catagorized very high contamination by Cd. The contamination factor value Cr in July ranged between 0.85 to 1.54 with mean value 1.12. The contamination factor value Cr in November ranged between 1.69 to 2.20 with mean value 1.93. Based on contamination factor indicated that Saguling surface sediment in July catagorized low to moderate contamination and November catagorized moderate contamination by Cr. The contamination factor value Cu in July ranged between 2.36 to 5.10 with mean value 3.68. The contamination factor value Cu in November ranged between 1.41 to 3.39 with mean value 2.23. Based on contamination factor indicated that Saguling surface sediment in July and November catagorized moderate to considerable contamination by Cu.
The contamination factor value Pb in July ranged between 1.20 to 4.01 with mean value 1.92. The contamination factor value Pb in November ranged between 1.06 to 1.46 with mean value 1.31. Based on contamination factor indicated that Saguling surface sediment in July categorized moderate to considerable contamination and November categorized moderate contamination by Pb. Contamination factor category is showed in Table 5 Contamination factor category in Saguling lake.

### Table 4. Contamination Factor value in Saguling lake.

| Station | July 2015 | November 2015 |
|---------|-----------|---------------|
|         | Cd        | Cr            | Cu    | Pb     | Cd     | Cr  | Cu | Pb |
| 1A      | 52.13     | 1.26          | 5.01  | 4.01   | 77.49  | 2.20 | 1.41 | 1.19 |
| 1B      | 55.48     | 1.39          | 5.10  | 2.58   | 76.41  | 2.05 | 3.39 | 1.29 |
| 2       | 44.22     | 0.90          | 3.97  | 2.10   | 69.73  | 1.78 | 3.01 | 1.46 |
| 3       | 42.87     | 1.18          | 3.90  | 1.28   | 68.73  | 1.89 | 1.44 | 1.02 |
| 4       | 53.48     | 1.54          | 4.06  | 1.75   | 76.33  | 2.12 | 1.94 | 1.64 |
| 5       | 37.68     | 0.85          | 2.67  | 1.84   | 73.33  | 1.94 | 1.84 | 1.45 |
| 6       | 42.93     | 0.93          | 2.36  | 1.58   | 63.83  | 1.76 | 2.95 | 1.39 |
| 7       | 35.62     | 0.92          | 2.59  | 1.20   | 68.69  | 2.11 | 1.85 | 1.26 |
| 8       | 44.52     | 0.93          | 3.53  | 1.51   | 62.14  | 1.69 | 1.63 | 1.06 |
| 9       | 42.32     | 1.30          | 3.60  | 1.31   | 66.04  | 1.82 | 2.82 | 1.31 |
| Mean    | 45.13     | 1.12          | 3.68  | 1.92   | 70.27  | 1.93 | 2.23 | 1.31 |

### Table 5. Contamination Factor category in Saguling lake.

| Station | July 2015 | November 2015 |
|---------|-----------|---------------|
|         | Cd        | Cr            | Cu    | Pb     | Cd     | Cr  | Cu | Pb |
| 1A      | Very High | Moderate      | Considerable | Considerable | Very High | Moderate | Considerable | Moderate |
| 1B      | Very High | Moderate      | Considerable | Moderate   | Very High | Moderate | Considerable | Moderate |
| 2       | Very High | Low           | Considerable | Moderate   | Very High | Moderate | Considerable | Moderate |
| 3       | Very High | Moderate      | Considerable | Moderate   | Very High | Moderate | Considerable | Moderate |
| 4       | Very High | Moderate      | Considerable | Moderate   | Very High | Moderate | Considerable | Moderate |
| 5       | Very High | Low           | Moderate    | Moderate   | Very High | Moderate | Considerable | Moderate |
| 6       | Very High | Low           | Moderate    | Moderate   | Very High | Moderate | Considerable | Moderate |
| 7       | Very High | Low           | Moderate    | Moderate   | Very High | Moderate | Considerable | Moderate |
| 8       | Very High | Low           | Considerable | Moderate   | Very High | Moderate | Considerable | Moderate |
| 9       | Very High | Moderate      | Considerable | Moderate   | Very High | Moderate | Considerable | Moderate |

According to the classification carried out on the basis of CF, bed sediment are very high contaminated with Cd in July and November 2015, low-moderate contaminated with Cr in July 2015 and moderate contaminated with Pb in November 2015. Bed sediment are moderate-considerable contaminated with Cu in July and November 2015. Bed sediment are moderate-considerable contaminated with Pb in July 2015 and moderate contamination in November 2015. Sediment re-suspension can be caused by storm water run off, sediment re-suspension caused by storm water runoff increased its potential for heavy metals release in sediment to water column. During re-suspension of anoxic sediment, the physical and chemical environment surrounding sediment bound metals will change. Bedded sediments is drawn up into the water column which is often fully saturated with dissolved oxygen [18].

PLI represents the number of times by which the metal content in the sediment exceeds the background concentration and give a summative indication of the overall level of heavy metal toxicity in a particular sample. PLI < 1 indicates no sediment pollution, whereas PLI > 1 points to pollution.
The value of PLI obtained in The Saguling sediments in July and November are 4.21 and 4.39 respectively. According to PLI categorized Saguling lake sediment was polluted with heavy metals. Higher concentration of heavy metals may be caused by external distinct anthropogenic source lie agricultural runoff, industrial waste water discharge and atmospheric dry deposition [19]. It also provide valuable information to the policy makers on the pollution levels in the study area.

In the Saguling catchment located 556 industries which are 442 textile industries, 8 pulp and paper industries, 11 tanning industries, 16 rubber and plastic industries, 14 chemical industries 40 electroplating and metals industries, and 25 food and beverage industries. The growth of industries population has caused the decrease of landholding. As a consequence, there is a prevailing problem of floods accompanied by accelerated soil erosion in rainy seasons. Due to the high population density in the upper catchment of the lake, extensive agricultural land, soil erosion and the presence of industries, the lake water became polluted and eutrophic. The water is also contaminated by heavy metals, pesticides, etc. The study suggests that the urban wastewater and effluents emerging from the industries must be monitored periodically for maintaining the standards prescribed by the pollution control board for various industries in the region. Further treatment of industrial effluents particularly from textile mills, electroplating, tanning manufacturing and chemical industries before discharging in the upper catchment is prerequisite for controlling the level of pollution in the Saguling lake.

4. Conclusion
The heavy metals were identified in the Saguling lake surface sediment. The total heavy metals contents in the surface sediment in July and November decrease in order of Cu>Cr>Pb>Cd and Cr>Cu>Pb>Cd respectively. Two pollution indices were used for the environmental assessment of Saguling lake surface sediment are contamination factor (CF) and pollution load index (PLI). Based on PLI value Saguling lake surface sediment was polluted by heavy metals. The possible main source of the metals contamination in the sediment are municipal and industrial wastewater discharge, agriculture runoff, and atmospheric input. A variety of useful tools, methods, and indices were used to evaluate heavy metal pollution in the surface sediment of the Saguling lake. However, heavy metals can transfer from the sediments to other ecosystem components such as underground water, crops an animals and affect human health through the water supply and food chain. Therefore, it is necessary to design an environmental strategy not only manage but also monitor the same with priority.

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