Distribution of heavy metals (Ag, Hg, Cd) concentration and the pollution status in the western part of Segara Anakan Lagoon, Indonesia

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Abstract. Segara Anakan lagoon has continuously gained pressures as a result of human activities. Unsustainable land use and forest destruction in the upper river areas have brought about erosion bringing also heavy metals into the lagoon. This study investigated the distribution of heavy metals (Ag, Hg, and Cd) concentration and determined the pollution status in the western part of Segara Anakan Lagoon, Cilacap, Central Java, Indonesia. The study was conducted for six months, from June to November 2016, with a sampling interval of once a month. The study was located in six stations spread over from the rivers to the estuary area. The distribution of Ag content tended to be stable at every station (except in August). Low Ag content was found in June, July, September, and October, while the highest was in November. The higher Ag content was found in the estuary area. The distribution of Hg varied and mostly increased toward the estuary area. Low Hg content was found in November, while the highest was in October. The distribution of Cd tended to be stable at every station (except in July). Low Cd content was found in June, while the highest was found in August-October. A higher concentration of Cd was obtained in the channel area up to the confluence area of several rivers. According to the pollution index, the western part of Segara Anakan Lagoon was moderately polluted.

1. Introduction
Segara Anakan is located between 7°35’ and 7°50’ S and 108°45’ and 109°03’ E on the border of West Java and Central Java Provinces, the southern part of Java Island [1]. It covers approximately 24,000 hectares in total area, comprising waters, mangrove forests, and mud land formed by sedimentation. Segara Anakan region is a wetland area, covered mainly by 26 types of mangrove plants as a place for spawning, feeding, and nursery for more than 45 types of marine fish. Not only for settled fish, such as ringtail-cardinal fish (Apogon aureus), shrimp, crabs, lobster, mud clam (shell), red grouper, squid, white pomfret, barramundi, large head hairtail fish, stingray, cuttlefish, swamp eel, shark, and other marine biotas but also 17 types of unsettled/migratory marine biota such as eel (Anguilla sp) [2, 3].

However, the Segara Anakan has continuously gained pressures year by year as a result of human activities. Unsustainable land use in urban areas and forest destruction in the upper river areas have brought about high erosion levels, culminated in the lagoon. According to Segara Anakan Management
Office (BPKSA), the Segara Anakan covered approximately 6,400 Ha in 1900, but it has been shrinking due to sedimentation so that in 2008 the total area was only 750 hectares [1]. Erosion in the rivers flowing into Segara Anakan Lagoon has contributed mud material and wastes as much as 1,000,000 m³ per year, ultimately deposited in the lagoon. About 75% were materials brought by the flow of Citanduy River, while the rest was derived from materials deposited by other rivers. Accordingly, total sediment in the lagoon from 1994 to 2010 was more than 5,000,000 m³ [3].

The western part of Segara Anakan Lagoon is a wetland area with a lagoon type inhabited by a mangrove’s ecosystem. Unlike the eastern part, the primary activities that attract attention in this area are agriculture, settlement, and forestry. The problems in the western part of Segara Anakan Lagoon were diverse than in the eastern part, namely sedimentation and water siltation, rapid population growth, and over-exploitation of fish resources. The danger of erosion carried mud and waste materials into the lagoon, which also carried harmful materials such as heavy metals (Ag, Hg, and Cd). Heavy metals will be difficult to degrade, and they will even be absorbed in the organism body [2,4,5,6]. This study aimed to investigate the distribution of heavy metals concentration (Ag, Hg, and Cd) and determine the pollution status in the western part of Segara Anakan Lagoon, Cilacap, Central Java, Indonesia.

2. Methodology

2.1. Time and Location
This study was carried out for six months, or six months, from June to November 2016, with a sampling interval of once a month. The sample was taken from six stations in the western part of Segara Anakan Lagoon, Cilacap, Central Java, Indonesia, distributed from the rivers to the estuary area. Station 1 is the estuary of the western part (also called West Plawangan), Station 3 is the Citanduy River, Station 3 is the Cibeureum River, and Stations 2, 4, 5, and 6 are the main water body of the western part. Sample analysis activities were conducted at the Laboratory of Productivity and Aquatic Environment, Department of Aquatic Resources Management, IPB University (figure 1).

2.2. Methods and Parameters Observed
Water samples were taken from the studied area, placed in bottles, and then preserved according to the APHA standards method [7]. Heavy metal concentration was determined by the Atomic Absorption Spectrophotometry (AAS), and other water quality parameters refer to the APHA standards method [7].

![Figure 1](image_url)  
**Figure 1.** Study site in the western part of Segara Anakan Lagoon, Cilacap, Central Java Province, Indonesia.

2.3. Data Analysis

2.3.1. Analysis of Heavy Metals.
Analysis of heavy metals was performed by the AAS method based on Lambert-Beer Law. Heavy metals concentration was obtained with the following formula:
Heavy metals concentration (mg/L) = $\frac{\text{AAS concentration} \times \text{determination vol.}}{\text{dry weight}}$ \hspace{1cm} (1)

2.3.2. **Cluster Analysis.**
Cluster analysis is a method used to identify a homogeneous group known as a cluster. The principal objective is to combine the objects with similarities into groups or clusters [10], using the Minitab software version 16.

The Canberra Index is used to determine the grouping based on the physical and chemical similarities of the waters. The parameters used to identify the cluster were heavy metals (Ag, Hg, and Cd), salinity, pH, and DO. Determination of habitat grouping using the following formula [11]:

$$C = 100\% \times \left(1 - \frac{1}{n} \sum_{i=1}^{n} \frac{|Y_{i1} - Y_{i2}|}{Y_{i1} + Y_{i2}}\right)$$ \hspace{1cm} (2)

Note:
- $C$ = Canberra similarity index
- $n$ = number of parameters compared
- $Y_{i1}$ = The value of parameter $i$ at location 1
- $Y_{i2}$ = The value of parameter $i$ at location 2

2.3.3. **Spatial Analysis.**
The distribution of heavy metals (Ag, Hg, and Cd) in the western part of Segara Anakan Lagoon are revealed both spatially and temporally, processed with ArcGIS ver. 10.1, developed by ESRI (Environment Science & Research Institute). This application's functions show spatial data, creating layered maps, and performing the basic spatial analysis [12].

2.3.4. **Determination of Water Pollution Status.**
The Pollution Index (PI) is used to determine the water pollution status (Decree of Minister of EnvironmentNumber 115 of 2003 about the Guidelines for Determining the Water Quality Status) [13]. The quality standard used in this study is the seawater quality standard for marine biota (Decree of the State Minister of the Environment Number 51 of 2004 about seawater quality standard (appendix 3) [8].

The PI value was obtained from the following equation:

$$P_{ij} = \sqrt{\frac{(C_i)^2}{(L_{ij})_M^2} + \frac{(C_i)^2}{(L_{ij})_R^2}}$$ \hspace{1cm} (3)

Note:
- $P_{ij}$ = pollution index based on its designation ($j$)
- $C_i$ = concentration of water quality parameter measured according to the quality standards based on its designation ($j$)
- $L_{ij}$ = concentration of water quality parameters according to the quality standards based on its designation ($j$)
- $(C_i/L_{ij})_M$ = maximum value of $C_i/L_{ij}$
- $(C_i/L_{ij})_R$ = average value of $C_i/L_{ij}$

Then, the PI value was evaluated using the classification of pollution levels (Decree of Minister of Environment Number 115 of 2003) [13], presented in Table 1.

**Table 1. Classification of pollution levels based on PI value.**

| Score   | Criteria                  |
|---------|---------------------------|
| $0 \leq \text{PI} \leq 1,0$ | Meet the quality standard (good condition) |
| $1,0 \leq \text{PI} \leq 5,0$ | Lightly polluted          |
| $5,0 \leq \text{PI} \leq 10,0$ | Moderately polluted       |
| $\text{PI} \geq 10,0$ | Heavily polluted          |
3. Result and Discussion

3.1. Water Quality Parameters Conditions
Most of water quality data in the western part of Segara Anakan Lagoon were not according to the quality standards [8]. It can be seen that pH and turbidity values were not according to the quality standard in all stations, while the temperature, DO, and TSS were not in most areas. Only salinity and ammonia have an excellent value and according to the quality standard in all areas (table 2 and 3).

Table 2. The average water quality in the western part of Segara Anakan Lagoon spatially.

| Station | Temperature (°C) | pH  | DO (mg.L⁻¹) | TDS (mg.L⁻¹) | TSS (mg.L⁻¹) | Turbidity (NTU) | Salinity (psu) | Ammonia (mg.L⁻¹) |
|---------|------------------|-----|-------------|--------------|--------------|----------------|---------------|------------------|
| 1       | 27.42            | 6.75| 4.35        | 8,719        | 47.67        | 68.07          | 15.83         | 0.13             |
| 2       | 27.83            | 6.79| 4.41        | 5,378        | 137.83       | 138.52         | 12.17         | 0.09             |
| 3       | 27.50            | 6.08| 5.00        | 428          | 296.50       | 301.60         | 1.00          | 0.08             |
| 4       | 28.08            | 6.67| 4.23        | 5,922        | 115.00       | 108.03         | 11.17         | 0.11             |
| 5       | 29.00            | 6.75| 4.70        | 3,400        | 69.00        | 55.86          | 8.50          | 0.08             |
| 6       | 28.33            | 6.67| 4.06        | 3,606        | 114.17       | 89.42          | 10.50         | 0.09             |
| QS      | 28-32            | 7-8.5| 5.00 | -           | 80.00        | 5.00           | 34.00         | 0.30             |

Table 3. The average water quality in the western part of Segara Anakan Lagoon temporally.

| Month | Temperature (°C) | pH  | DO (mg.L⁻¹) | TDS (mg.L⁻¹) | TSS (mg.L⁻¹) | Salinity (psu) | Ammonia (mg.L⁻¹) |
|-------|------------------|-----|-------------|--------------|--------------|---------------|------------------|
| Jun   | 30.83            | 6.75| 4.53        | 8,663        | 30.67        | 12.33         | 0.01             |
| Jul   | 28.67            | 7.50| 3.59        | 242          | 195.50       | 21.00         | 0.13             |
| Agt   | 27.83            | 6.83| 4.76        | 15,896       | 23.50        | 19.50         | 0.03             |
| Sep   | 26.33            | 6.63| 4.76        | 2,008        | 114.83       | 4.00          | 0.07             |
| Oct   | 27.00            | 6.00| 5.11        | 488          | 87.67        | 2.33          | 0.17             |
| Nov   | 27.50            | 6.00| 4.00        | 157          | 328.00       | 0.00          | 0.17             |
| QS    | 28-32            | 7-8.5| 5.00 | -           | 80.00        | 34.00         | 0.30             |

3.2. Station Clustering
Sampling stations clustering were depicted in a dendrogram (figure 2). All stations were grouped into one zone, except for Station 1, which formed its own. It was assumed that the value of heavy metals and other water quality parameters at Station 2, 3, 4, 5, and 6 (zone 1) tended to be the same, whereas, in Station 1 (zone 2), it was different.
3.3. Distribution of Heavy Metals (Ag, Hg, and Cd)

Heavy metals have high conductivity and density of more than 5 kg.m⁻³ [14]. The high content of heavy metals in waters may lead to contamination, accumulation, and even pollution towards its environment, such as the biota, water, and sediment [16]. Heavy metals in waters can be found in two forms that are dissolved (ions in complex heavy metals or heavy metals that bind to organic/inorganic compounds) and undissolved (particles and complex metal compounds which are absorbed in suspended substances) [17]. The solubility of heavy metals may be lower or higher, depending on the condition of the waters [18]. At high salinity, most heavy metals tended to accumulate in sediment, the reason for which is still unknown [19]. Temperature indirectly gave an essential effect in metal form specification. Temperature is an important factor that regulates the metal shape between two different phases that indirectly affect the kinetic reaction of that metal [20]. The temperature increasing might trigger the rise of dissolved toxic compounds, most of which were bioactive [21].

Silver (Ag) metal is a kind of heavy metal that tended to be rare. Naturally, Ag did not derive from the sea but land. Ag is a contaminant in numerous mining and smelting wastes, closely related to many wastes’ disposal cases in the sea. Ag in the sea is a crucial indicator regarding anthropogenic influence as its chemical characteristics tend to be stable in dissolved conditions and easily spread. Dissolved Ag was widely spread around the anthropogenic sources [22].

Based on figure 3, in June, July, September, and October, the concentration of Ag was below the determination method’s detection limit (<0.005 mg.L⁻¹) in all areas, while in August, it seems to increase towards the estuary area with a value from ≥0.005–0.009 mg.L⁻¹ to ≥0.009–0.013 mg.L⁻¹. In November, the Ag concentration was reached the highest value in all areas (≥0.013–0.017 mg.L⁻¹).
The spatial distribution of Ag did not vary in each month of observation, except in August 2016. The tendency for higher concentrations in the estuary in August 2016 was related to the pH and salinity. The pH decreased towards the estuary while the salinity increased (table 3). The pH has a close relationship with the solubility characteristics of heavy metals. Free ions of heavy metals were released into the water column in a low pH, so the dissolved heavy metals in water would be higher than those suspended or accumulated in the sediment [23]. Salinity also largely influenced the Ag chemical behaviours. The thermodynamic model shows that complex chlorine compounds (AgCl\(^0\), AgCl\(^2-\), AgCl\(^2+\), AgCl\(^3-\)) would become dominant in salinated waters [24]. Ag (I) was very strongly bonded with Cl, and it was hydrolysed into AgCl\(^2-\) (66%) and AgCl\(^-\) (26%) as Ag compounds in inorganic form (dissolved) and available for the aquatic organism. The increase of dissolved Ag concentration could become an indicator of waste pollution and could be utilized to track the wastewater discharge point source [25]. Furthermore, a significant increase in Ag concentration in November 2016 was thought to be due to the increase in runoff. Based on the rainfall prediction, the highest rainfall during the study period occurred in November [26].

Based on ANZECC & ARMCANZ (2000) [9], the Ag concentration in the western part of Segara Anakan Lagoon was still below the quality standard (0.003 mg.L\(^{-1}\)) except in August and November 2016. Ag chemical characteristics significantly influenced the toxicity levels, not only in the sea but also at the estuary. Ag was easily accumulated in invertebrates. Ag was one of three highly toxic elements (together with Cu and Hg) in invertebrates and algae in the estuary and sea areas based on the bioassay test. However, several organisms in both estuary and sea were sensitive to very low Ag concentration (under bioassay conditions) [27]. The range of Ag toxicity values of <1-14 µL\(^{-1}\) is low for a trace element toxicity value. Ag in the dissolved form also became a trigger for significant physiological damage to the population, such as premature and decreased larval production and decreased glycogen storage for reproductive needs [27]. In several coastal areas, Ag was found in high concentration and could influence the planktonic compositions and other trophic levels [28].

Mercury is a unique metal and very toxic in the form of methylmercury. The methylmercury was created in a water ecosystem when methylate bacteria were in the organic forms due to the reaction between water and sediment in low pH conditions [29]. Methylmercury can be dissolved well in the water, pass biota membrane, and persistent in fatty tissue biota. Methylmercury can also be biomagnified in the food chain. The amount of Hg in the highest level of the organism in the food chain may reach a thousand or million times higher than in the water and sediment [30].

The distribution of Hg concentration in the western part of Segara Anakan Lagoon from June to November 2016 can be seen in figure 4. This figure shows that the distribution of Hg concentration varied in each month of the study period and tends to be higher in the estuary area except in November 2016. Hg concentration in July and September 2016 was homogeneous in all areas amounted to ≥0.001–0.003 mg.L\(^{-1}\). In October 2016, the Hg concentration reached the highest value of ≥0.003–0.005 mg.L\(^{-1}\) spread over Stations 1, 4, 5, and 6. Nevertheless, the Hg concentration in November 2016 was dropped and even had a very low value in the estuary area.
Figure 4. Distribution of Hg concentration in the western part of Segara Anakan Lagoon.

The distribution of Hg in the western part of Segara Anakan Lagoon tends to be higher in the estuary area due to increased dissolved oxygen (table 3). In waters where oxygen was well dissolved, dissolved Hg (Hg^{2+}) became dominant [31]. Moreover, a previous study on Segara Anakan Lagoon revealed that the increase of heavy metals in the form of dissolved Hg occurred in high salinity conditions [32]. In November 2016, Hg concentration went down towards the estuary area because there was a freshwater flood at the western part of Segara Anakan Lagoon, proven by the salinity value of 0 psu in all areas (table 4).

Based on the Decree of the State Minister of the Environment Number 51 of 2004 [8], the most measured Hg concentration was above the quality standard (0.001 mg.L^{-1}), which was harmful to the aquatic biota. Previous research in the estuary of Donan River that connected to the eastern part of Segara Anakan Lagoon also shows a similar result [33]. Mercury is well known to attack the physiological system, leading to the death of biota. In humans, Hg can increase death risk due to coronary heart disease, cardiovascular disease, and fast development of carotid atherosclerosis [34]. Moreover, mercury has a very toxic characteristic due to its high volatility, so that it can be easily spread throughout the region [35]. Every Hg compound had a distinct character in toxicity, distribution, accumulation, and retention period in the body [36]. In addition, water temperature, pH, hardness, and the existence of other metals affect the toxicity of mercury on fishes. When the water temperature was higher than a fish body, the metabolic rate of fish will increase so that Hg would be accumulated more in the fish body in the dry season [30].

Cadmium (Cd) is found in a tiny amount and was not dissolved in water. Cadmium content in the earth's crust was approximately 0.2 mg.kg^{-1}. Cadmium is a kind of toxic heavy metal to human beings. Cd metal has a higher level of toxicity and solubility in freshwater because Cd in estuary and seawater meets the chloride (Cl) to become an undissolvable molecule [37]. Deposition of heavy metals in water occurred because there was an anion carbonate, hydroxyl, and chloride. Heavy metals had the property to bind organic matter easily and deposited at the bottom of waters, mixed with sediment [14].

The distribution of Cd concentration in the western part of Segara Anakan Lagoon from June to November 2016 can be seen in figure 5. Cd concentration tended to be stable at every station, except in July 2016. In July, the distribution of Cd concentration varies with a range of <0.001–0.006 mg.L^{-1} with a tendency of lower values in the estuary area. Salinity influenced the availability of Cd metals in waters, Cd would be increased in line with the rise of salinity, but the influence size depended on sediment
oxidized [38, 39]. The temporal distribution of Cd concentration increased from June to October 2016. Cd concentration in June 2016 was below the determination method's detection limit of <0.001 mg.L\(^{-1}\), while in the following months, it increases until it reached the highest value of ≥0.003–0.006 mg.L\(^{-1}\). However, in November 2016, the Cd content decreased in all areas to ≥0.001–0.003 mg.L\(^{-1}\). Cd concentration would drop in line with the rise of TSS because TSS bonded Cd in the waters column that makes a dissolved Cd become dropped [40]. This study result showed that the decrease of Cd concentration was also followed by the rise of TSS concentration and vice versa (Table 4).

Based on the Decree of the State Minister of the Environment Number 51 of 2004 [8], Cd concentration was above the quality standard (0.001 mg.L\(^{-1}\)), so it was dangerous for aquatic biota. Previous research in the estuary of Donan River that connected to the eastern part of Segara Anakan Lagoon also shows a similar result [33]. Cd metal could kill aquatic biota in a particular condition, for example, the crustaceans would die within 24 to 504 hours when exposed to Cd metal or compounds with a concentration of 0.005-0.150 mg.L\(^{-1}\) [41]. Cd could create direct or indirect impacts on the population or individuals in the ecosystem. Cadmium could cause bone deformity and kidney disorders in fish, indirectly giving a lethal effect [30].

3.4. Pollution Index (PI)
Pollution in waters occurred when wastes or other substances entered the water body and caused changes in the physics, chemistry, and biology of the waters [42]. The pollution status of the western part of Segara Anakan Lagoon was determined using a Pollution Index (PI). Figure 6 represents the average value of PI in each sampling station at the western part of Segara Anakan Lagoon. The PI values ranging from 5.99 to 7.89, the western part of Segara Anakan Lagoon has been moderately polluted.

The pollution index value at Station 3 was higher than other stations. It was thought to be influenced by tidal. The tidal types in Segara Anakan Lagoon are mixed tides, which tend to be diurnal tides (two high tides and one low tide) [43]. These two high tides were thought to have pushed the material back toward the Citanduy River (Station 3). In addition, the Citanduy River also gets many pollutants from non-point sources on the mainland, generally from agricultural and animal husbandry activities. Runoff that brings soil material from agricultural land increases heavy metals such as Ag, Hg, and Cd in waters. Liquid waste from chicken and cattle farms along the Citanduy River also worsens the water quality [44]. Heavy metals in these activities are generally used for fertilizers and pesticides.
4. Conclusion
Ag concentration tended to be stable at every station in each month of observation (except in August). Low concentration was found in June, July, September, and October, and the highest concentration was in November. Relatively high Ag concentration was found in the estuary. The distribution of Hg varied and generally increased toward the estuary. The lowest Hg concentration was in November, while a relatively high concentration was in October. The distribution of Cd tended to be stable at every station (except in July). Low concentration was in June, whereas the highest occurred in August - October. High concentrations of cadmium were in the channel area up to the confluence area of several rivers. Based on the pollution index, the western part of Segara Anakan Lagoon was moderately polluted.

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