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# Kriging-GIS model for the spatial distribution of seawater heavy metals

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**ABSTRACT**

Development in the seawater region of Bungus Bay - Padang City continues to develop rapidly, this can cause damage to the surrounding water environment due to seawater pollution and high sedimentation. Therefore, research is needed on the analysis of seawater's heavy metal contents and the spatial distribution of contamination values based on the Decree of the Minister of Environment (KEPMEN LH) No. 51/2004. This study aims at elaborating the correlation between the distribution of heavy metals (Cd, Cr+6, Pb, and Cu) in the seawater region of Bungus Bay. The data processed is data on heavy metal content from the results of sample measurements using the AAS, where the points and values of the sample analysis that have been collected using GPS at 3 location points are then imported into GIS using GIS Kriging. The value of heavy metal index for seawater based on the quality standard of KEPMEN LH No/5/2004 shows that a category does not fit, where Cd (0.0013-0.0031 mg/l), Cr+6 (0.008-0.013 mg/l), Pb (0,009-0,017 mg/l), and Cu (0.010-0.014 mg/l). The value of concentrations of these heavy metals generally shows a spatial distribution pattern different in each direction of movement. On one hand, the highest concentrations of this heavy metal are generally located in the Northwest of the bay (Cd, Cr+6, Pb) except Cu which is in the Northern part of the bay, as it is close to the discharge locations and outlet of rainwater from the catchment area. Generally, heavy metal contamination in the seawater region of Bungus Bay comes from the household, industrial, and ship loading and unloading waste. The pollution occurs as the result of the oil from transferring tankers, oil spills from fishing boats that rinse at Bungus PPS, and debris resulted from the plywood industry activity.

**Keywords**: heavy metals, kriging-gis, seawater, bungus bay

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## 1. Introduction

Heavy metals are metal elements whose density exceeds 5 gr/cm$^3$ which are classified as contaminants and are dangerous because they can accumulate in the water column, sediment, and organisms and cannot be degraded naturally [1] Heavy metals that come from nature, such as the result of tectonic, volcanic, upwelling, and input from the atmosphere, while heavy metals that come from human activities such as the results of industrial and mining wastewater discharges[2]. Heavy metals are one of the factors in determining the quality status of water quality, where heavy metals that have exceeded the threshold will cause losses, in addition to having an impact on the ecosystem in polluted waters, and the health of the surrounding community will also be disturbed [3]. In [4, 5], explained that seawater has a great ability to dissolve various substances, both in the form of gases, liquids, and solids. One of the solutes contained in seawater is heavy metal. In [6], added that metals are often considered indicators of anthropogenic effects in the marine environment and have a potential risk to the natural environment. Some authors explain the presence of dissolved heavy metal levels in seawater, sediment, and seashell (geloina coaxans) greatly affect the good and bad conditions of seawater [7]. This is because heavy metals are more resistant towards weathering, either physically, chemically or biologically. Heavy metal contamination in marine waters can damage the biochemical systems that exist in marine biota; it will become a health threat if consumed by humans [8]. Such as 1) Heavy metal Pb is toxic to marine life, in certain types of fish, heavy metal levels of Pb at 0.1 - 0.2 ppm can cause poisoning and can kill fish at levels of

188 ppm [9], Also, heavy metal Pb contamination in the water column can enter the body through food, drink, respiration, skin contact, eye contact, and parenteral[10]; 2) Heavy metal Cr is naturally in valence 3 ($Cr^{3+}$) and valence 6 ($Cr^{6+}$). Heavy metal $Cr^{6+}$ is more toxic than heavy metal $Cr^{3+}$, because of its solubility and high mobility in the environment. Through the food chain, heavy metal $Cr^{6+}$ can be deposited in the body parts of living things which at a certain size can cause poison [11]; 3) Heavy metal Cu that enters the aquatic environment can occur naturally or as a side effect of human activities [12]. Naturally, heavy metal Cu enters the waters from erosion, erosion of rocks, or from the atmosphere brought down by rainwater, while human activities such as industrial activities, copper mining, and the shipbuilding industry along with activities in ports are one of the fast routes to occur. increasing the Cu heavy metal's solubility in water [13, 14], and 4) Heavy metal Cd is a natural non-essential metal which is relatively small in number in the waters but can increase if there is industrial waste disposal or oil leftover from combustion or heating and excessive fertilization. Industries that can produce Cd heavy metal waste are the textile, battery, paint, plastics industry, and others. According to [15], in water, heavy metal Cd can be spread as far as 50 km from the source. Heavy metal Cd has the characteristics of silvery-white like aluminum metal, heat resistance, and corrosion resistance. Research on heavy metal analysis has been carried out, such as heavy metals in the dissolved form [16], in sediments [17], deposited in the bay waters which are densely human activities. Analysis of heavy metals contained in marine organisms such as seashell shows that the heavy metal content in water and sediment correlates with the heavy metal contents contained in seashell bodies [18, 19]. Based on research from [20] regarding the bioaccumulation of heavy metals in green clams in Cirebon sea waters, it shows that the season affects the levels of heavy metal cadmium which accumulates in green clams. Heavy metals in sediments will continue to settle then will enter and accumulate in the body of marine biota which will then enter biota with a higher trophic level through the food chain, this is also called bioaccumulation and biomagnification [21, 22]. In [23], added that heavy metal Pb pollution in waters that are more than the threshold concentration will have an impact on the biota death in these waters. A high concentration of heavy metals is considered severely polluting the aquatic ecosystem due to their ability to penetrate and accumulate in the food chain[24]. Despite their function as important micronutrients for flora and fauna, several types of heavy metals such as Fe, Co, Cu, and Zn are dangerous in high concentrations. In the meantime, other forms of toxic heavy metals such as Cr, Pb, and Cd are considered carcinogenic elements [25]. Geographical information systems (GIS) is a tool of analysis and modeling that functions to integrate the correlation between features of Earth's surface and its environmental effects [26, 27]. It is also employed for spatial analysis and operations at basic levels. That said, it is essential to assess the spatial distribution of some heavy metals and their effects on sediments; it is also to delineate the areas that are contaminated. Limited sampling scale is one of the serious concerns in the measurement of contamination in an area or in water. To solve this problem, the GIS-Kliging method becomes an alternative in delivering data and information. The method is in accordance with the common methods of spatial statistical research in terms of spatial characteristics [28] it also relies on the principle that points with closer space tend to be more similar to each other than those with more distant spaces. Geostatistical and geometric methods involve various spatial algorithms. Recently, the field of spatial modeling and various studies have been employing GIS software in GIS-Kliging interpolation analysis to the contamination zoning of aquatic contaminants [29]. The study aims at determining the spatial correlation between the distribution of heavy metals (Cd, $Cr^{+6}$, Pb, and Cu) in the seawater region of Bungus Bay - Padang City.

## 2. Methods

The research was conducted in the seawater region of Bungus Bay which is administratively located in the Bungus Teluk Kabung Sub-district, Padang City at coordinates 100°22'23" - 100°29'13" East Longitude and 0°59'1" - 1°5'44" South Latitude with a coastline length of ± 21,050 m and a bay length of 5,418 m, and the volume of water in the bay of 223,255,052.2 $m^3$ and has a surface that tends to be rounded with a surface area of 1,384 ha. Sampling was carried out at three points of location, such as 1) In the mangrove area (Southern part) which is a marine biota and mariculture habitat which is very close to port activities, industry, and close to fishing settlements (Dempo Pertamina Port); 2) Waters of the Electric steam power plant port (PLTU - Teluk Sirih) is located at the Bay mouth; and 3) Waters of the Bungus Ocean Fishing Port (PPS). After the sample points are determined, a ground check is carried out using the Global Position System (GPS) to see the correctness of the area information. Measurement of metal content in seawater samples using Atomic Absorption Spectroscopy (AAS) is carried out through the following steps: 1) Sample (25 ml) was taken using a dropping pipette and then the sample was put into a 50 ml beaker glass, then the sample was added with a 2.5

ml $HNO_3$ solution; 2) After that the sample that has been mixed with the $HNO_3$ solution is placed on a hotplate until the sample volume becomes 10-15 ml, then the sample is transferred to a 25 ml volumetric flask; 3) Beaker glass is rinsed using distilled water 3 times, then the water from the rinsing beaker glass is put into the volumetric flask, distilled water is added to the flask until it reaches the 25 ml limit mark, and 4) The final step, the sample is transferred to a plastic bottle (tightly closed) before being put into AAS. The analysis is conducted after the prior steps are carried out; it is done by analyzing the heavy metal contents that are displayed on a computer screen. Where the data processing is data on heavy metal content from the results of sample measurements using the AAS tool. Data processing includes calculating the average value of the heavy metal content concentration of each sample. Heavy metal sampling was carried out during the rainy season consisting of Cd, $Cr^{+6}$, Pb, and Cu which was carried out based on the biophysical requirements of waters in Decree of the Minister of Environment (KEPMEN LH) No. 51/2004 and regulation the Minister of Marine and Fisheries (PERMIT KP) No. 23/2016 and, then performed the scoring with GIS approach. The analysis results samples in the laboratory that have been collected with GPS location points, then imported into GIS and have been interpreted using GIS-Kriging (from Geostatistics) can be expressed as follows: There are n points of observation with values of Z ($x_1$), Z ($x_2$),. .., Z ($x_n$) at the observation point $x_1$, $x_2$, ..., $x_n$ is distributed to $x_o$. The best estimators at $x_o$ are:

$$Z^*(x_0) = \sum_{i=1}^{N} \lambda_i Z\ (x_i)$$

$$i = 1, \ldots, n$$

λ - the kriging weight is determined as minimizing the variance of error σ2 k = var [Z*($x_o$)-Z($x_o$)] checking for bias limits: E [Z*($x_o$)-Z($x_o$)] = 0 (means error = 0). To interpolate by kriging, one must correctly define the variogram (also called the structural function). A variogram is defined as half of the mathematical expectation of a random variable. Thus,

$$\lambda(h) = \frac{1}{2}[Z(x) - Z\ (x + h)]^2$$

Where:

Z ($x$), Z ($x$ + $h$): two known values with a separation distance of $h$

N ($h$): the number of pairs of data points with a distance of $h$

The spatial distribution mapping of heavy metals in the seawater region of Bungus Bay was carried out using the Kriging model in the ArcGIS program (10.6); which is applied to develop predictive maps for measured elements.

## 3. Results and Discussion

The results of measurements of heavy metal content from 3 sample point locations in the Bungus Bay area can be seen in Table 1. From the results of laboratory analysis, it can be seen that most of the sample points contain heavy metals.

Table 1. Laboratory analysis results for testing the content of heavy metals Cd, $Cr^{+6}$, Pb, Cu

| Location | Parameter | Measurement | | | Standard KEPMEN LH |
|---|---|---|---|---|---|
| | | Unit | Result | Quality standards | |
| PPS Bungus | Kadmium (Cd) | mg/l | 0,0031 | 0,001 | No. 51/2004 |
| | Kromium heksavalen ($Cr^{+6}$) | mg/l | 0,011 | 0,005 | |
| | Timbal (Pb) | mg/l | 0,009 | 0,008 | |
| | Tembaga (Cu) | mg/l | 0,014 | 0,008 | |
| Dempo Pertamina | Kadmium (Cd ) | mg/l | 0,0022 | 0,001 | |
| | Kromium heksavalen ($Cr^{+6}$) | mg/l | 0,008 | 0,005 | |
| | Timbal (Pb) | mg/l | 0,011 | 0,008 | |
| | Tembaga (Cu) | mg/l | 0,01 | 0,008 | |

| | | | | |
|---|---|---|---|---|
| PLTU Teluk Sirih | Kadmium (Cd ) | mg/l | 0,0013 | 0,001 |
| | Kromium heksavalen ($Cr^{+6}$) | mg/l | 0,013 | 0,005 |
| | Timbal (Pb) | mg/l | 0,017 | 0,008 |
| | Tembaga (Cu) | mg/l | 0,012 | 0,008 |

Source: Laboratory measurements and analysis.

### 3.1. Heavy metal description results

*Heavy metal Cd*

In the seawater region of Bungus Bay, Cd comes from domestic activities. In the seawater region of Bungus Bay, Cd comes from pesticides, insecticides, domestic waste, coal, and industrial pollution (port activities). The results of Cd analysis at the location point of sample 1 in the waters of PPS Bungus, sample 2 in the waters of Dempo Pertamina port, and sample 3 in the waters of the PLTU Teluk Sirih are in a category that does not fit, i.e 0.0013-0.0031 mg/l. The contents of Cd required in the marine water quality refers to the standard in the KEPMEN LH No. 51 of 2004 for marine biota and mariculture is 0.001 mg/l. Painting of boats or ships using paint containing Cd elements also affects the concentration of Cd heavy metals in the waters, such as ship docking at Bungus PPS. Cd has a very wide distribution in marine waters. There is only one type of heavy metal mineral Cd, namely greenockite (CdS) which is always found together with the mineral sphalerite (ZnS). Heavy metal minerals Cds are very rarely found in marine waters so that in the exploitation of Cd heavy metals, it is usually a byproduct of zinc ore (Zn) smelting and refining events. Zinc ore concentrate contains 0.2 - 0.3% Cd. This means that Zn is the main source of Cd [30].

*Heavy metal $Cr^{+6}$*

Heavy metal $Cr^{+6}$ at the location point of sample 1 in the waters of PPS Bungus, sample 2 in the waters of Dempo Pertamina port, and sample 3 in the waters of the PLTU Teluk Sirih are in a category that does not fit, i.e 0.008-0.013 mg/l. $Cr^{+6}$ content required in the marine water quality standard in KEPMEN LH No. 51 of 2004 for marine biota and mariculture is 0.005 mg/l. The high $Cr^{+6}$ is due to the sampling of seawater conditions which are high tides and large waves. The highest $Cr^{+6}$ was found at sample 3 in the waters of the PLTU Teluk Sirih towards the high seas with a value of 0.013 mg/l. It turns out that the concentration of $Cr^{+6}$ is indeed quite high in the open seas so that due to the large enough wave factor will $Cr^{+6}$ be carried away and will cause the region around in the water of Bungus Bay will be contaminated.

Some of the factors that caused the high $Cr^{+6}$ concentration were due to activities in the seawater region of Bungus Bay. This condition can be seen in the 3 locations of the special ports (PPS Bungus, Dempo Pertamina dan PLTU Teluk Sirih), where at that location there are ships that are anchored that throw their ballast water into the sea and also because the location is not so far from the coast that there is the frequent stirring of water with sediment which contains heavy metal $Cr^{+6}$.

*Heavy metal Pb*

Heavy metal Pb at the location point of sample 1 in the waters of PPS Bungus, sample 2 in the waters of Dempo Pertamina port, and sample 3 in the waters of the PLTU Teluk Sirih is in a category that does not fit, i.e 0.009 - 0.017 mg/l. The Pb content required in the marine water quality standard in the KEPMEN LH No. 51 of 2004 for marine biota and mariculture is 0.008 mg/l. The highest Pb was found at the sample location point 3 in the waters of the PLTU Teluk Sirih towards the high seas with a value of 0.017 mg/l.

The high Pb content at the sample location point 3 in the waters of the PLTU Teluk Sirih is due to the location of the PLTU Teluk Sirih PLTU and also a trap zone for all pollution components through tidal dynamics events. In the downstream part, activities that are estimated to have the potential to produce waste containing Pb are from transportation and port activities. The process of washing and maintaining fishing boats as well as spilt fuel used in transportation activities are thought to have contributed to the Pb concentration in the waters.

Port areas are generally a contributor to the presence of Pb in seawater[30, 31]. Generally, fuel oil gets tetraethyl additives containing Pb to improve the quality of the fuel, especially gasoline as an anti-knocking, corrosion prevention, anti-condensation and colouring agent. According to [32], Pb can be used as a fuel additive and lead pigment in the paint which is the main cause of increased Pb levels in aquatic environments. Pb is also deposited in the estuary area so that it is not carried completely to the high seas. In [33, 34], argue that tides influence the existence of organic material, nutrients, and heavy metal contents in the water. This is what causes Pb levels in estuary sediments to be higher than in the open seas.

*Heavy metal Cu*

Heavy metal Cu at the location of sample 1 in the waters of PPS Bungus, sample 2 in the waters of Dempo Pertamina port, and sample 3 in the waters of the PLTU Teluk Sirih is in a category that does not fit, i.e 0.010-0.014 mg/l. The Cu content required in the marine water quality standard in the KEPMEN LH No. 51 of 2004 for marine biota and mariculture is 0.008 mg/l. The highest Cu was found at sample 3 in PPS Bungus waters with a value of 0.014 mg/l. The Cu concentrations usually occur directly from the atmosphere and due to oil spills from port ships and minerals which are widely contained in the sea itself [35]. Generally, Cu enters the waters as a result of erosion and from the air carried by rainwater. Meanwhile, human activities come from industrial waste. Cu is an essential metal that is useful in the formation of hemocyanin in the blood and enzymatic systems for aquatic animals[32]. However, its high presence in water can be bad for fish, such as inhibiting the oxidation of lactic acid in gills. If the Cu concentration in water bodies is in the 2.5-3.0 ppm range, it will kill the fish in it [12]. Human consumption of Cu-contaminated fish might cause bad effects. Symptoms that arise in humans due to acute poisoning are nausea, vomiting, abdominal pain, hemolysis, metritis, seizures, and finally death. In chronic poisoning, Cu accumulates in the liver and causes hemolysis. Hemolysis occurs due to the accumulation of $H_2O_2$ in red blood cells resulting in oxidation of the cell layer, as a result of which cells can break[32]. For more details, the distribution of heavy metal contamination (Cd, $Cr^{+6}$, Pb, Cu) is based on spatial correlation using GIS-Kliging interpolation analysis in the seawater region of Bungus Bay - Padang City (Fig. 1).

### 3.2. Discussion for environmental conditions

There are heavy metals with economic value such as gold (Au), silver (Ag), copper (Cu), lead (Pb), zinc (Zn), bismuth (Bi), and stibium (Sb). Meanwhile, Mercury (Hg) and arsenic (As) are categorized as dangerous heavy metals. Based on research by[36], it shows that the metal content of sea-floor sediments containing Hg exceeds the threshold according to the sediment quality standards issued by WAC 172-204-320 United States (mailto: bbett461@ecy.wa.gov) of heavy metals that come from the rocks that compose them.

Advancements in the regional development of Bungus Bay coastal area are not matched by conservation efforts of the coastal ecosystem. In addition, some author revealed that mangroves on the coast of Bungus Bay as a protected zone for coastal and river boundaries have decreased in the area from 75.2 ha in 2008 to 73.3 ha in 2014[37]. Sirih cove in the Southern part has experienced a significant decrease in mangrove area since 2008, from 4.11 ha in 1995 to 2.48 ha in 2008 and didn't exist in 2008. In [38], adds that the extinction of the mangrove ecosystem as a protected zone has occurred due to the conversion of land designated for the 10,000 MW National Electrification Project, i.e the construction of a 2 x 112 MW Steam Power Plant (PLTU) in the Southern part of the region of the coastal of Bungus Bay since 2007. The same condition occurred in the mangrove ecosystem in Muaro Sako Labuhan Tarok, where there was a significant reduction in the area from 20.7 ha in 2014 to 10.9 ha in 2015. The decline in the area of the mangrove ecosystem occurred as a result of the conversion of land designated for the development of the palm oil industry by PT. Wira Inno Mas, which is equipped with a dock and water reclamation on an area of 79 ha. The location is close to tourism activities, ports, agriculture, and settlements by converting mangrove ecosystems and the former PT. Singkiong that was there before at that location. The conversion of the function of the mangrove ecosystem area for industrial land, docks, and residential areas on the sandy beach which is economically oriented has changed the environmental hue of the coastal of Bungus Bay as a center for fisheries and tourism into an industrial area. By the regional policies contained in the Padang City Regional Tourism Development Master Plan (RIPPD) 2008 - 2017, the concept of a development strategy in the coastal area of Bungus Bay is prioritized for tourism development [39]. Coastal abrasion is one of the impacts caused by damage to coral reefs. Coral reefs are 50% damaged in Kabung cove and its surroundings, as well as mangrove ecosystem with limited distribution in this area [35]. Sedimentation occurs around river estuaries which are still relatively active, especially during the rainy season. The regional government of Bungus Bay coastal area has applied several measures to cope with environmental changes and disasters that occur as a result of abrasion, floods, and landslides. The government collaborates with the local communities to construct several infrastructure such as embankments. This response includes reviewing spatial planning, placing priority areas, making FADs, and reforesting mangrove forests and protected forests. Generally, heavy metal contamination in the seawater region of Bungus Bay comes from household, industrial, and ship loading and unloading waste. The pollution occurs due to the oil from oil tankers, spills from fishing boats that rinse at Bungus PPS, and plywood industry debris. Moreover, the minimum, maximum, and average values of the contents of analyzed heavy metals are shown in the forecast map Figure for heavy metals Cd, Cr+6, Pb, and Cu as shown in Fig. 1. Concentrations of these heavy metals generally show a different pattern of spatial distribution in each direction of movement. On one hand, the highest concentration of such heavy metal is generally located in the northwest of the bay (Cd, Cr + 6, Pb), except for Cu, which is located in the northern part of the bay, as it is close to the rainwater discharge and outlet from the catchment area. In particular, heavy metal concentrations are also increasing in the area of ship moorings and shipyards. In the bay interior dominated by muddy particles also resides concentrations of heavy metals. The bay is also characterized by organic matters that absorb and accumulate heavy metals in the clay minerals. The research results of [40] explained heavy metal concentrations decreased towards the edge and coastline of the bay for two reasons. Firstly, sand fragments dominate the bay edge. Secondly, active currents and waves at the edges can disturb finer particles; they are further transported and deposited gradually in deeper areas. Moreover, the metals' level might vary based on each metal's mobility, sediment type, and environmental conditions. Further, on the other hand, the variation of these metals at the bay edges and mouth was quite small.

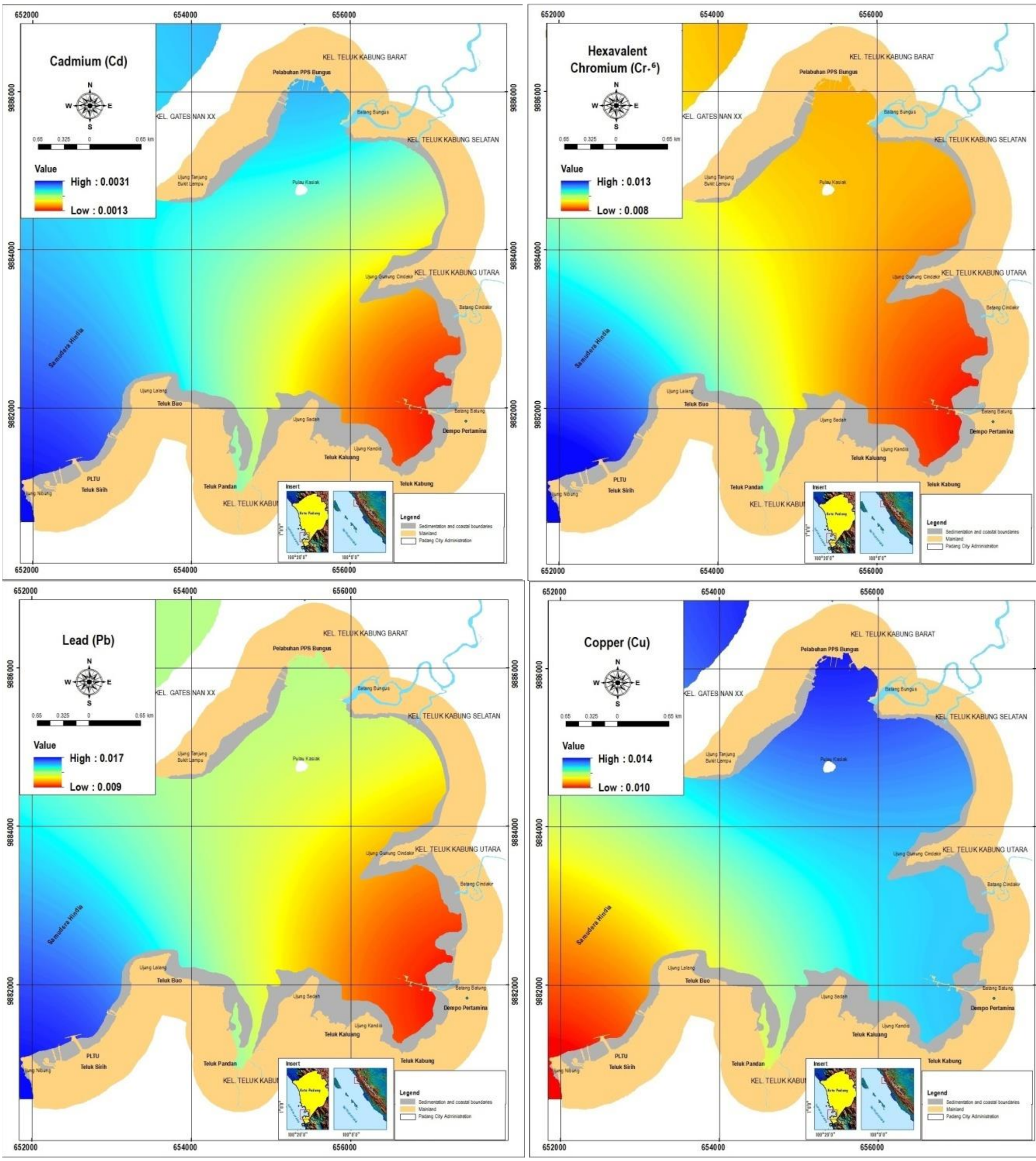


Fig. 1. Distribution map of heavy metals in the waters of Bungus Bay

## 4. Conclusions

The levels of these metals vary across the bay; it depends on the mobility, sediment type, and environmental features. However, the variation of these metals at the bay mouth and was relatively small. The heavy metal in the highest concentrations is generally located in the bay northwest area (Cd, Cr+6, Pb), except Cu, which is located in the northern part of the bay, as it is close to the rainwater discharge and outlet from the catchment area. In particular, heavy metal concentrations are also increasing in the area of ship moorings and shipyards. Moreover, a high concentration of heavy metals is found inside the bay, in which the bay is rich in muddy particles (silt and clay), as well as organic matters that can absorb and accumulate heavy metals in clay minerals. Generally, heavy metal contamination in the Bungus Bay Seawater Region originates from the household, industrial, and ship loading and unloading waste. Further, it is found that pollution occurs as the result of oil from oil tankers, oil spills from fishing boats rinsing at Bungus PPS, and plywood industry debris.

## Conflict of Interest

The authors declare no conflict of interest.

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