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Research Article

Heavy metals in water and sediment of Way Ratai River due to small-scale gold mining activities in Pesawaran Regency, Lampung Province (Part I: mercury, cyanide, lead, and arsenic)

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Abstract

The use of mercury and cyanide in the gold separation process at small-scale gold mining in Pesawaran Regency can carry mercury (Hg), cyanide (CN-), lead (Pb), and arsenic (As) as other heavy metals that are also harmful to the environment. This study aimed to determine the concentration and distribution of some heavy metals (Hg, CN-, Pb, As) found in the water and sediment of the Way Ratai River. Water and sediment samples were taken at 5 points along the Way Ratai River. The heavy metal analysis method used a Mercury Analyzer, Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES), Atomic Absorption Spectrophotometry, and X-ray fluorescence. Mapping the distribution of heavy metal concentrations uses ArcGIS with the Inverse Distance Weighting (IDW) method. The results showed that the average concentration of Hg in water was 0.006 ppm, As was 0.023 ppm, and CN- was 0.003 ppm; Pb was not detected in river water. Meanwhile, in the river sediments, the average Hg was 11.83 ppm, As was 449.10 ppm, Pb was 450.88 ppm, and CN- was 0.38 ppm. It can be concluded that the concentration of heavy metals in the sediment of Way Ratai River is in the high category; the heavy metal content in river sediment is higher than that in water. For this reason, it is necessary to manage waste from gold processing activities, such as coagulation and absorption, to reduce the potential for environmental pollution.

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Introduction

Way Ratai River, one of the vital water resources in Pesawaran Regency, Lampung Province, is threatened by heavy metal pollution due to illegal gold ore processing by artisanal and small-scale gold mining (ASGM) in Pesawaran Regency. Gold ore processing in artisanal and small-scale gold mining (ASGM) uses the mercury (Hg) amalgamation method, which is common in tropical countries (Villas-Boas et al., 2001; Donkor et al., 2005; Odumo et al., 2014) including Indonesia. In addition to reducing state revenue, ASGM activities produce heavy metals that pollute the environment and harm health. According to Esdaile and Chalker (2018), ASGM is the largest source of Hg pollution compared to other human activities, where water runoff and infiltration carry Hg into the environment.

From mining activities, surface water runoff or infiltrated water carries metallurgical waste or Hg pollutants into the environment (Fernández-Martínez et al., 2005). Hg waste from the amalgamation process causes water resources to be polluted by heavy metals (Donkor et al., 2005). Mining of placer gold ores and deposits has been identified as one of the most ecologically damaging aspects of the gold mining industry (Tarras-Wahlberg et al., 2001; Tarras-Wahlberg et al., 2002). Fernández Martínez et al. (2005) reported that Hg contamination of soil, surface water, and sediments in areas affected by mining operations in Asturias, Spain, was substantial. Sudarningsih (2020) researched the heavy metal content of Martapura River sediments in South Kalimantan affected by ASGM with the results of Hg (0.092-5.775 ppm), Donkor et al. (2005) stated that research on the presence of mercury content due to ASGM in the sediments of the Pra River, Ghana, measured Hg concentrations ranged from 0.018 to 2.917 mg kg^{-1}. Yuwono et al. (2023) reported mercury's content in soil and river water of the Way Ratai watershed, namely Hg content in soil ranging from 0.26 to 28.9 mg L^{-1} and in river water ranging from 0.08 to 14.1 mg L^{-1}. Furthermore, research by Mulyadi et al. (2020) stated that the concentration of Hg in the sediment of the Tambang Sawah River, which was affected by gold processing activities, from upstream to downstream, was 1.34 ppm, 4.18 ppm, 35.89 ppm, 50.32 ppm, and 114.37 ppm, respectively.

As it is known, Hg pollution is harmful to human health (Jarup, 2003; Björkman et al., 2007; WHO, 2007; Saturday, 2018) and the environment (Donkor et al., 2005; Ignatavičius et al., 2022). Therefore, Hg pollution in soil, water, sediment, and biota must be monitored because it causes toxicity, persistence, and accumulation in the food chain. On the other hand, the amalgamation process, which produces liquid waste, can contain different heavy metals such as Fe, Mn, Pb, and Hg (Maulidyanto and Sari, 2017). According to Government Regulation No. 5 of 2022, gold ore mining using chemicals such as mercury and cyanide can carry other heavy metals such as Zn, Cd, Cr, Pb, Ni, As, and Hg that react due to the amalgamation and cyanidation process.

This study provides a complete analysis by examining the content of mercury (Hg), cyanide (CN-), arsenic (As), lead (Pb), manganese (Mn), ferrum (Fe), cadmium (Cd), copper (Cu), and zinc (Zn) in the water and sediment of the Way Ratai River, which has the potential to exceed safe limits and is harmful to health and the environment. The results of the study are published in two parts: Part one consists of Hg, CN-, As, and Pb; Part two consists of Mn, Fe, Cd, Cu, and Zn. The part one aimed to determine heavy metal (Hg, CN-, Pb, and As) contamination in water and sediment of the Way Ratai River due to small-scale gold processing activities.

Materials and Methods

The study was conducted at a small-scale gold mining processing site in Pesawaran Regency, as seen in Figure 1. Sampling of water and river sediments was carried out at 5 points. Point 1 in the upstream location that has never been or is still slightly polluted is a picture of the initial hue of the environment, which is estimated not or has not been affected by gold processing activities; point 2 and point 3 in locations polluted by gold processing waste, point 4 and point 5 in the downstream river leading to the sea. The heavy metal analyses tested were Hg, As, Pb, and CN-. Water samples were directly tested using Mercury Analyzer MA-3000 with the Reducing Vaporization method (water sample).

Inductively Coupled Plasma Optical Emission Spectrometric (ICP-OES) was used following SNI 6989.82:2018 on How to Test Metals using Inductively Coupled Optical Emission Spectrometric. Testing of cyanide parameters in water and sediment using a Hach Spectrophotometer (test kit) DR3900 with the MU.SS-UJI.91 method (in-house method). Sediment samples were separated from gravel, plants, and other objects, then dried, pulverized, and analyzed using X-Ray Fluorescence and Mercury Analyzer MA-3000 using ASTM D-6722-01 on How to Test Mercury (Hg) by Thermal Decomposition (solid samples), testing sediment cyanide parameters using a Shimadzu UV 1800 Double Beam UV Spectrophotometer using AOAC Method 2012 973.19.

Laboratory testing for the measurement of Hg, CN-, As, and Pb parameters was done at the National Research and Innovation Agency (BRIN), Mining Technology Research Center Lampung Province; testing CN- water sample parameter testing was done at the Lampung Industrial Services Standardization Center (BSPJI) Laboratory; testing CN- sediment sample was done at the Bogor Agricultural University Calibration and Certification Testing Services Laboratory (LJPKSIPB).

The results of the analysis were then compared with the quality standards as stipulated in Government Regulation No. 22 of 2021 Annex VI concerning the Implementation of Environmental Protection and Management and the Japan Environmental Quality Standard (JEQS) 1997, while sediment quality standards referred to the Swedish Environmental Protection Agency (SEPA) 2000, Canadian Council of Ministers of the Environment (CCME) 2002, Food and Agriculture Organization (FAO) 2007, National Oceanic and Atmospheric Administration (NOAA) 1999, and Latvia Environmental Quality Standard (LEQS) 2000. Water and river sediment samples with known absorbance values for each parameter were processed through interpolation and mapped using Geographic Information System (GIS) software based on the level of heavy metal concentration according to the amount using the Inverse Distance Weighted (IDW) method. The aim was to understand the spatial distribution pattern of heavy metals in the study area, interpreted in maps and colors.

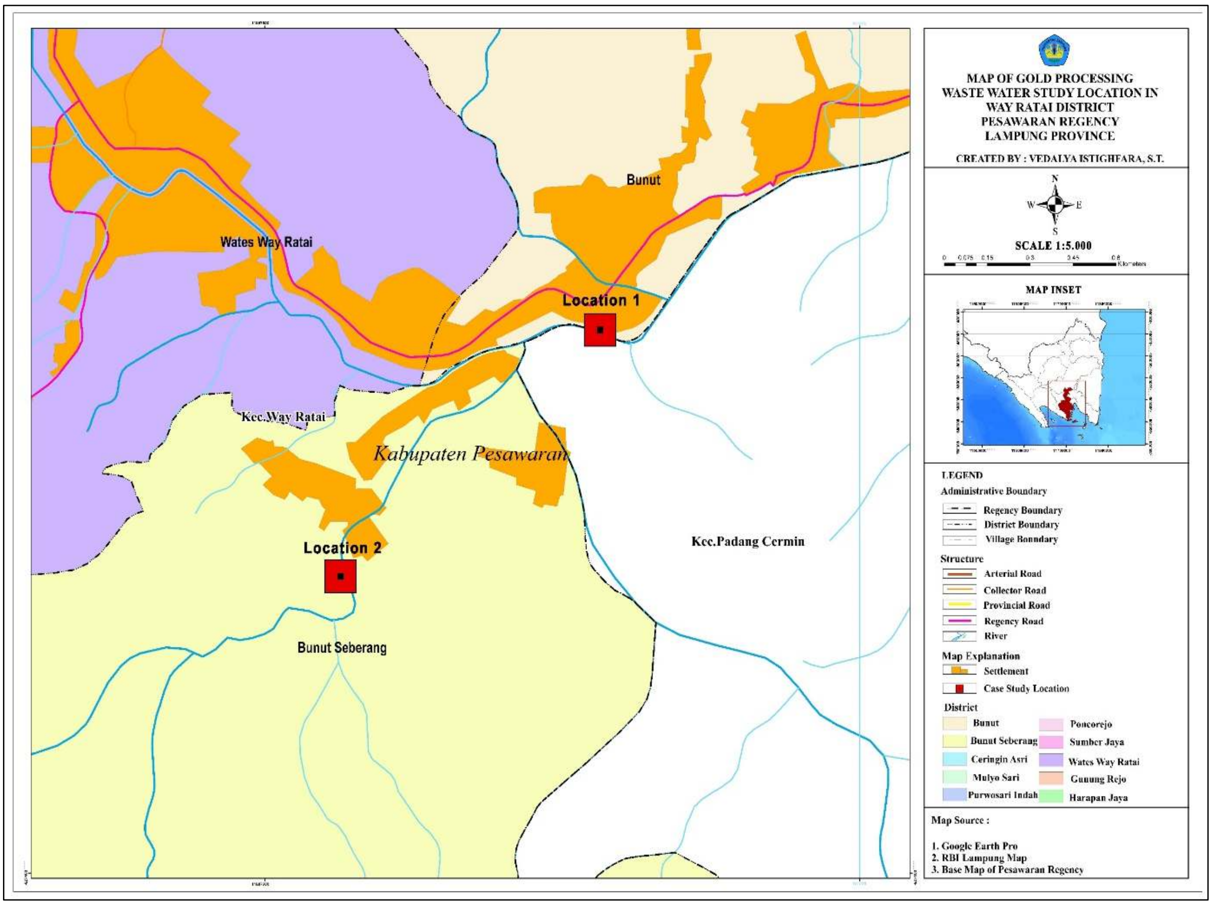

Figure 1. Research location map.

Results and Discussion

Mercury (Hg) content in water and sediment of Way Ratai River

The concentration of Hg in river water was detected at all research points (Figure 2) at point 1, namely (0.0075) ppm, point 2 (0.0097) ppm, point 3 (0.0040) ppm, point 4 (0.0033) ppm, point 5 (0.0035) ppm, where from all points when compared to water quality standards following PP No. 22 of 2021 and Japan Environmental Quality Standard (JEQS) 1997 has exceeded the established quality standards of 0.002 ppm and 0.001 ppm.

Mercury, especially in its methylated form, can accumulate in fish tissues and move up the food chain, posing health risks to both aquatic organisms and humans who consume these fish. This bioaccumulation can be worsened by mercury's resistance to dissolving in water, a quality that keeps it persistent in sediments and aquatic ecosystems for extended periods. Environmental mercury mostly originates from human sources like coal combustion and industrial waste. This has led to increased mercury deposition in water bodies, where it can transform into methylmercury. Methylmercury, in particular, is highly toxic and readily absorbed by fish, which then affects larger predators and humans who consume them. Research from 2023 emphasizes the toxic impacts on the kidneys and liver of fish, demonstrating how mercury exposure causes cellular damage and oxidative stress that can disrupt the physiological functions of these organs (Bijay et al., 2023; Fanny et al., 2023). Thus, in fast-flowing river water, the concentration of Hg will be lower. Research conducted by Muryani et al. (2021) showed that the concentration of Hg in Tajur River and Datar River water due to small-scale gold mining had the lowest value of <0.00007 ppm and the highest of 0.00164 ppm. Compared to the Hg value in the Way Ratai watershed, the concentration is still lower and below the quality standard, according to PP No. 22 of 2021. It shows that the solubility of Hg in water in the Way Ratai River is relatively high. Hg concentration in Way Ratai river sediment (Figure 3) point 1 is (0.926) ppm, point 2

(45.269) ppm, point 3 (3.20) ppm, point 4 (0.16) ppm, point 5 (9.62) ppm, where from all points when compared with sediment quality standards by the Swedish Environment Protection Agency (SEPA) 2000 which is 0.18 ppm and the Canadian Council of Ministers of the Environment (CCME) 2002 which is 0.17 ppm has exceeded the established quality standards.

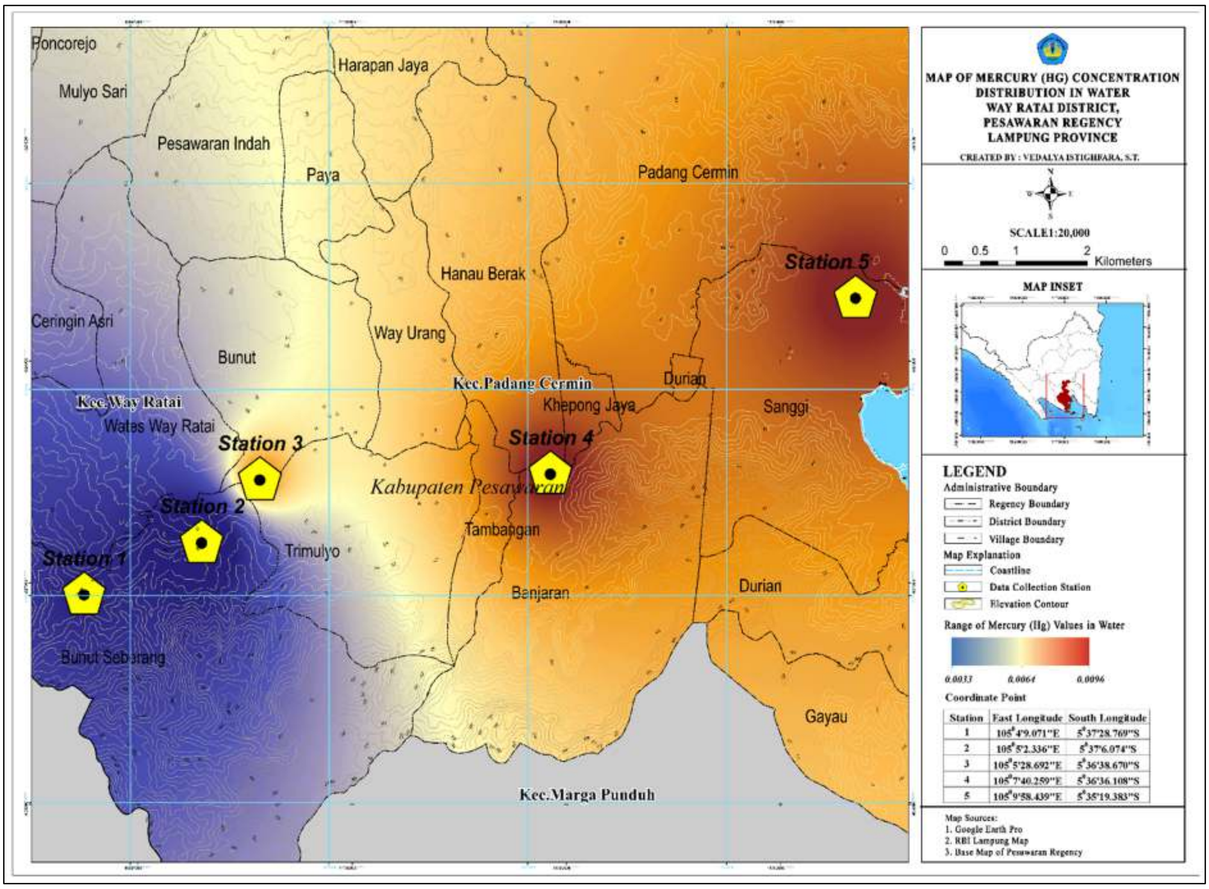

Figure 2. Distribution of Hg in Way Ratai River water.

The results indicated that the Hg content in river sediments is already relatively high due to gold processing activities using mercury that has been going on for a long time around the river. High concentrations of Hg in river sediments were found at points 2, 3, and 5. Points 2 and 3 were directly affected by gold processing activities near the river, while at point 4, the Hg concentration decreased as we moved away from the mining site. However, at point 5 (downstream of the river), the Hg concentration was again high; this is thought to be due to other gold processing around the river flow. The distribution of Hg can also be influenced by water flow or erosion.

Research by Muryani et al. (2021) supports this finding, where Hg concentrations in Tajur and Datar River sediments were higher than in water, reaching 7.74 ppm. The Hg tends to settle in sediments because it is poorly soluble in water and easily bound with dissolved and organic materials. It causes the Hg content in sediments to be higher than in water (Kusuma et al., 2017; Yulis, 2018). Waste from gold processing containing organic matter will react and bind heavy metal cations so that they eventually settle to the bottom of the water and unite with sediments, resulting in a much higher heavy metal content in sediments than in water. The heavier specific gravity of Hg, which is 13.5 times that of water, makes it easier for Hg to settle along with sediments on the riverbed (Kusuma et al., 2017).

According to FAO (2019), rainfall and runoff play a significant role in influencing water erosion rates by loosening soil particles and transporting them. This process leads to the removal of soil layers, impacting soil fertility and structure, which can have negative effects on agricultural productivity and ecosystem health. Rain intensity and runoff volume are key factors in the severity of erosion, as they determine the energy available to displace soil particles and move them across surfaces. Research by Yuwono et al. (2023) in the Way Ratai watershed showed a surface runoff coefficient of 0.45, which means 45% of the annual rainfall becomes surface runoff, contributing to Hg pollution in the river. The high Hg content at point 5 is likely caused by high erosion due to surface runoff and steep slopes, as well as a decrease in elevation from point 1 to point 5 (Table 2).

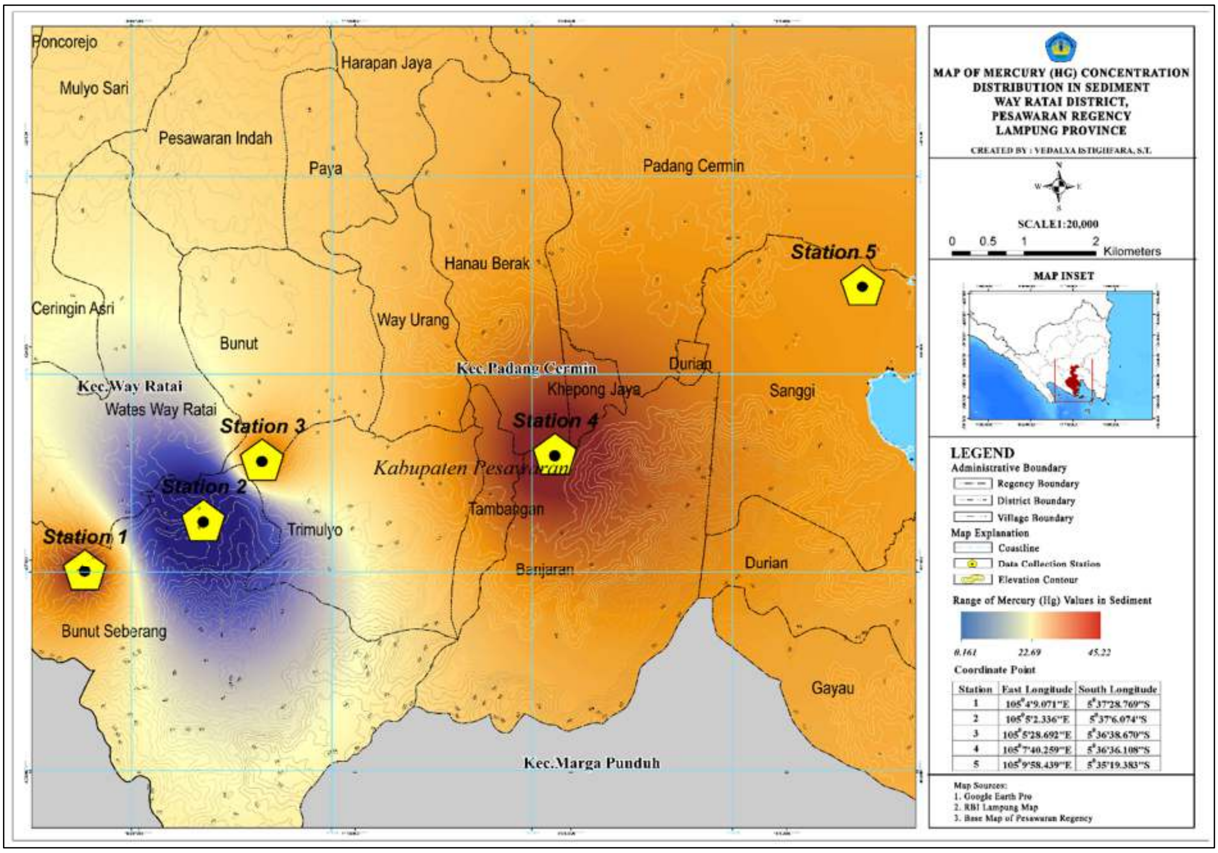

Figure 3. Hg distribution in the sediment of Way Ratai River.

Table 2. Elevation of the research points.

Observation Station	Elevation
Point 1	El. 75
Point 2	El. 52
Point 3	El. 27
Point 4	El. 14
Point 5	El. 9

Arsenic (As) content in water and sediment of Way Ratai River

Figures 4 (water) and 5 (sediment) show the results of measurements of As and its distribution in the water and sediment of the Way Ratai River. 0.01 ppm but are still below the safe limit value according to Appendix VI of PP No. 22 of 2021, which is 0.05 ppm. Arsenic is often found in gold ore, especially in sulfide minerals like arsenopyrite. During the gold extraction process, particularly with methods like cyanidation or ore roasting, arsenic can be released as waste. Based on Figure 4, the As value in river water was detected at all research points. At point 1, it is (0.0234) ppm, point 2 (0.0219) ppm, point 3 (0.0251) ppm, point 4 (0.022) ppm, and point 5 (0.0232) ppm. All five values have exceeded the 1997 Japan Environmental Quality Standard (JEQS). Research by Lasut et al. (2016) on As content in Manado Bay waters close to small-scale gold mining showed concentrations below the quality standards of PP No. 22 Year 2021. Evaluation of the Bailang River and Tondano River showed concentrations of suspended and dissolved As in the Bailang River <0.0005 ppm, and in the Tondano River 0.0012 ppm (suspended As) and 0.0011 ppm (dissolved As), respectively, which are also below the quality standard of 0.005 ppm. It is found as a by-product of smelting metals such as copper and tin. In amalgamation, As is available in the gold-arsen amalgam, it can contaminate the environment if not treated properly. However, this process does not significantly increase as solubility in the water settles more in river sediments (Figure 7).

Based on Figure 5, As concentrations were only detected at points 2 (898.8) ppm and 3 (99.4) ppm. The values at both points exceeded the 2000 Swedish Environment Protection Agency (SEPA) quality standard of 9.79 ppm and the 2002 Canadian Council of Ministers of the Environment (CCME) of 5.9 ppm. Research by Mabuat et al. (2017) showed that the concentration of As in the sediment of the Tondano River ranged from 1,386 ppm to 3,019 ppm, which is still relatively low compared to the Way Ratai River. Kisman (2011) stated that high concentrations of gold (Au) are often followed by high values of As, indicating a relationship between the two in the occurrence process. Contamination can come from natural geological processes and human activities, such as mining, animal husbandry, industry, and agriculture (Patel et al., 2005).

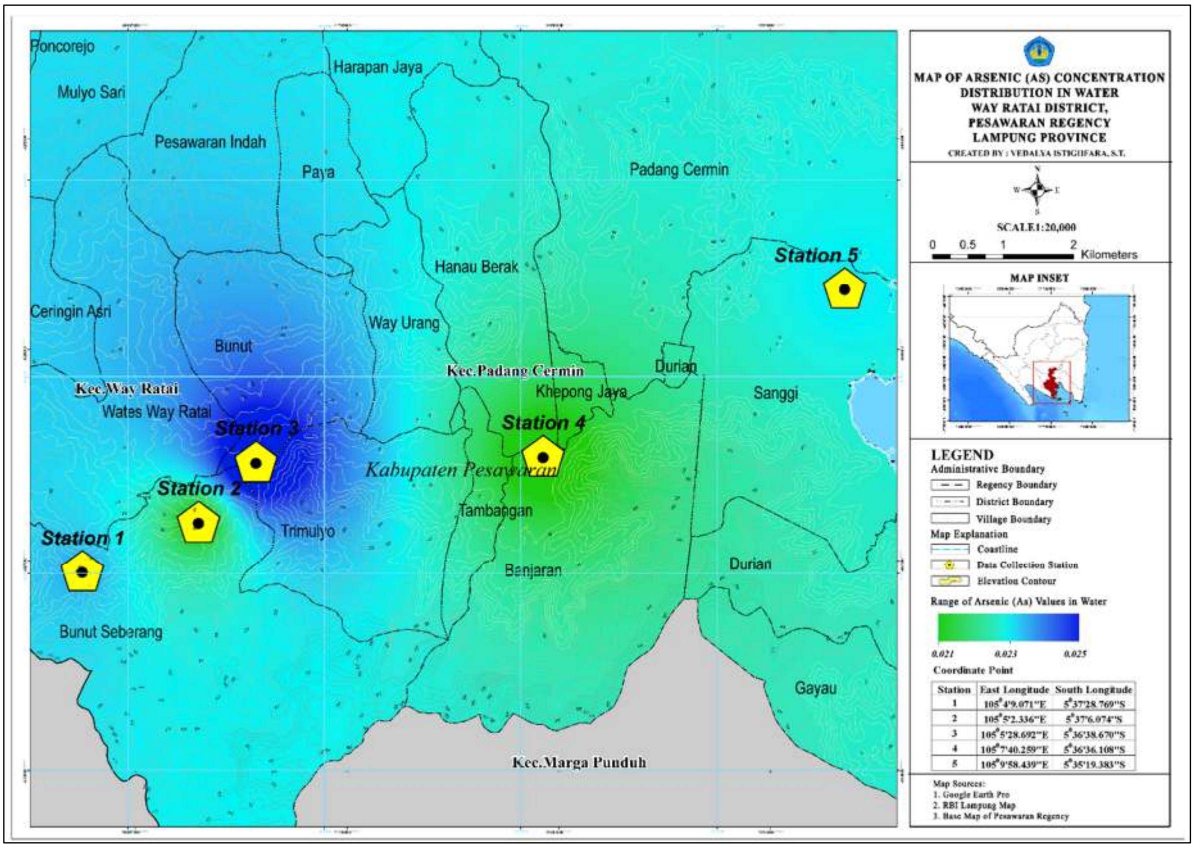

Figure 4. Distribution of As in Way Ratai River water.

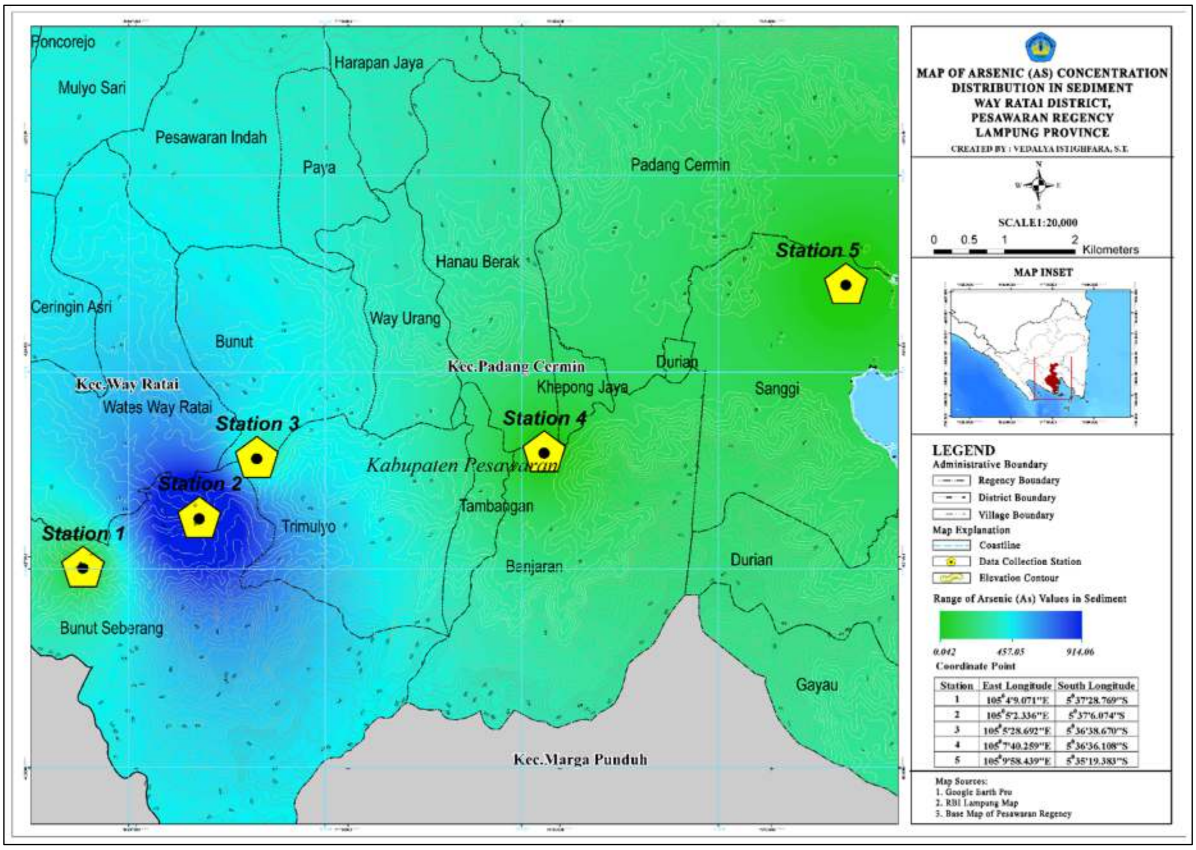

Figure 5. Distribution of As in sediments of Way Ratai River.

Processing of gold ore containing As with tailings disposal in the surrounding environment can increase the As content of streams. Arsenic has chemical properties that allow its ions to bind to minerals or organic materials in river sediment. In the forms of arsenate (As^{5+}) or arsenite (As^{3+}), arsenic can adsorb onto soil or sediment particles, particularly those rich in clay minerals or organic matter. This process is highly influenced by pH and redox potential in the environment, where arsenic more readily precipitates in neutral pH conditions and certain oxidation states. River environments with low oxygen levels and high organic content facilitate the accumulation of arsenic in sediments. High organic content provides many binding sites for arsenic ions, increasing the sediment's capacity to absorb and retain arsenic. Once absorbed, arsenic tends to be stable in sediment, especially in specific conditions (e.g., reducing or anoxic environments). This makes it difficult for arsenic to re-dissolve into the water, so its concentration in sediment remains high over time, particularly in areas near gold-processing waste discharge (Smedley and Kinniburgh, 2002; Alloway, 2013).

Lead (Pb) content in water and sediment of Way Ratai River

Figures 6 (water) and 7 (sediment) show the results of measurements of Pb and its distribution in the water and sediment of the Way Ratai River. Based on Figure 6, Pb concentrations were not detected in river water at all study points or were so small that the test equipment was not read. Laelasari et al. (2017) showed that the Pb content in Ciujung River, Banten, ranged from 0.015-0.022 ppm, which is still below the quality standard of 0.03 ppm (PPRI Number 82 of 2001). Lead is difficult to dissolve in water due to its chemical properties and electron structure, which does not allow strong interactions with water molecules (Fadhila et al., 2022). Lead can also form an oxide layer that protects the metal from reacting with water. Pb can enter waters through crystallization in the air with the help of rain or corrosion of mineral rocks, as well as from human activities, such as industrial and mining waste (Budiastuti et al., 2016). Pb content of the sediment test results (Figure 7) at point 1 is 557.2 ppm, point 2 is 392 ppm, point 3 is 723.2 ppm, point 4 is 285.6 ppm, and point 5 is 296.4) ppm.

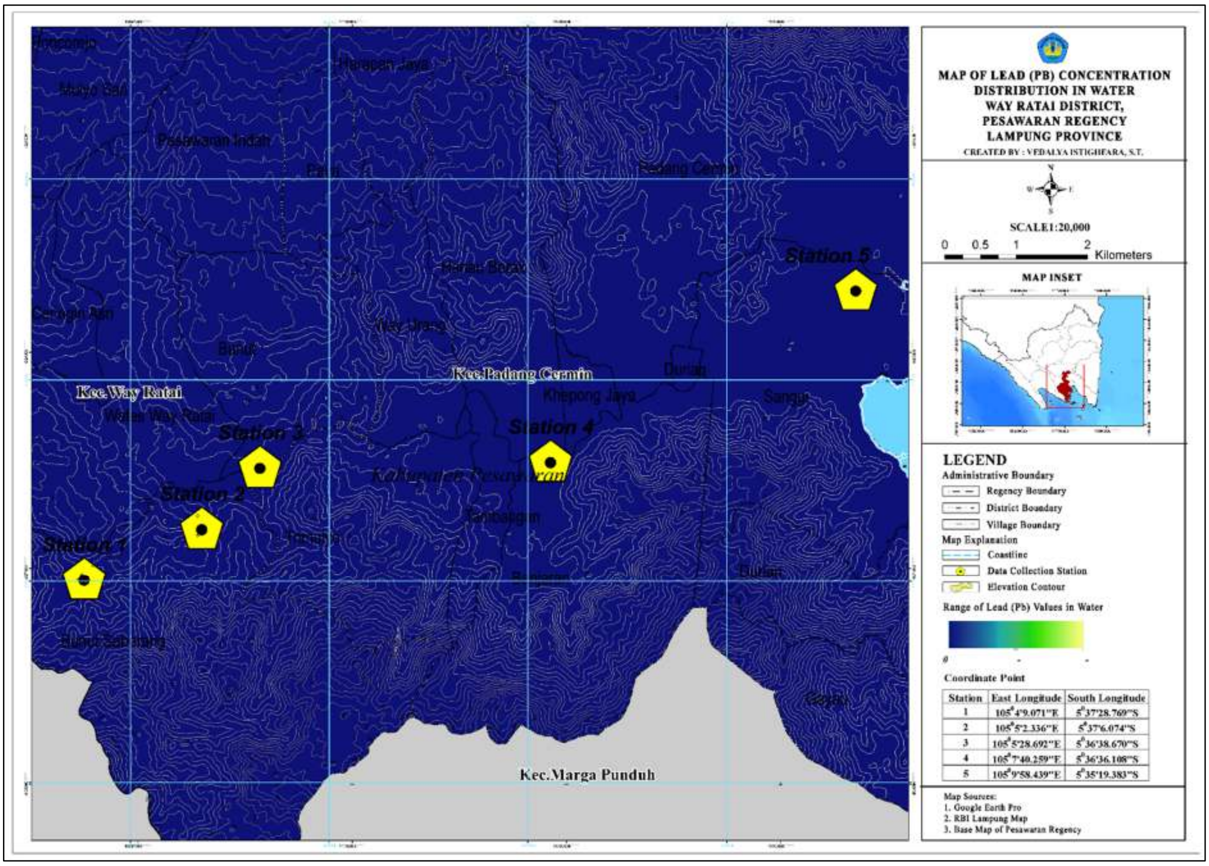

Figure 6. Pb distribution in Way Ratai River water.

The concentration of Pb of the five points has exceeded the quality standard of the Swedish Environment Protection Agency (SEPA) 2000, 35.8 ppm, and the Canadian Council of Ministers of the Environment (CCME) 2002, 35 ppm. The content of Pb values at all research points is accumulated in river sediments because gold processing activities in the Pesawaran Regency have been going on for a long time. From these activities, amalgamation tailings containing Pb are disposed of in the river and downstream along with runoff water. Pb has a high affinity for organic material and minerals in the sediment. When heavy metals like Pb are released into aquatic environments from gold processing activities, they tend to adsorb onto soil or sediment particles. Adsorption occurs because of bonding between Pb ions and the negatively charged surfaces of sediment particles, especially those containing clay minerals or organic matter (Alloway, 2013). River sediment acts as a natural "absorber" for pollutants, including heavy metals like lead. Fine sediment particles, such as clay, have a large surface area and can absorb and retain lead in greater quantities compared to larger particles like sand (Alloway, 2013).

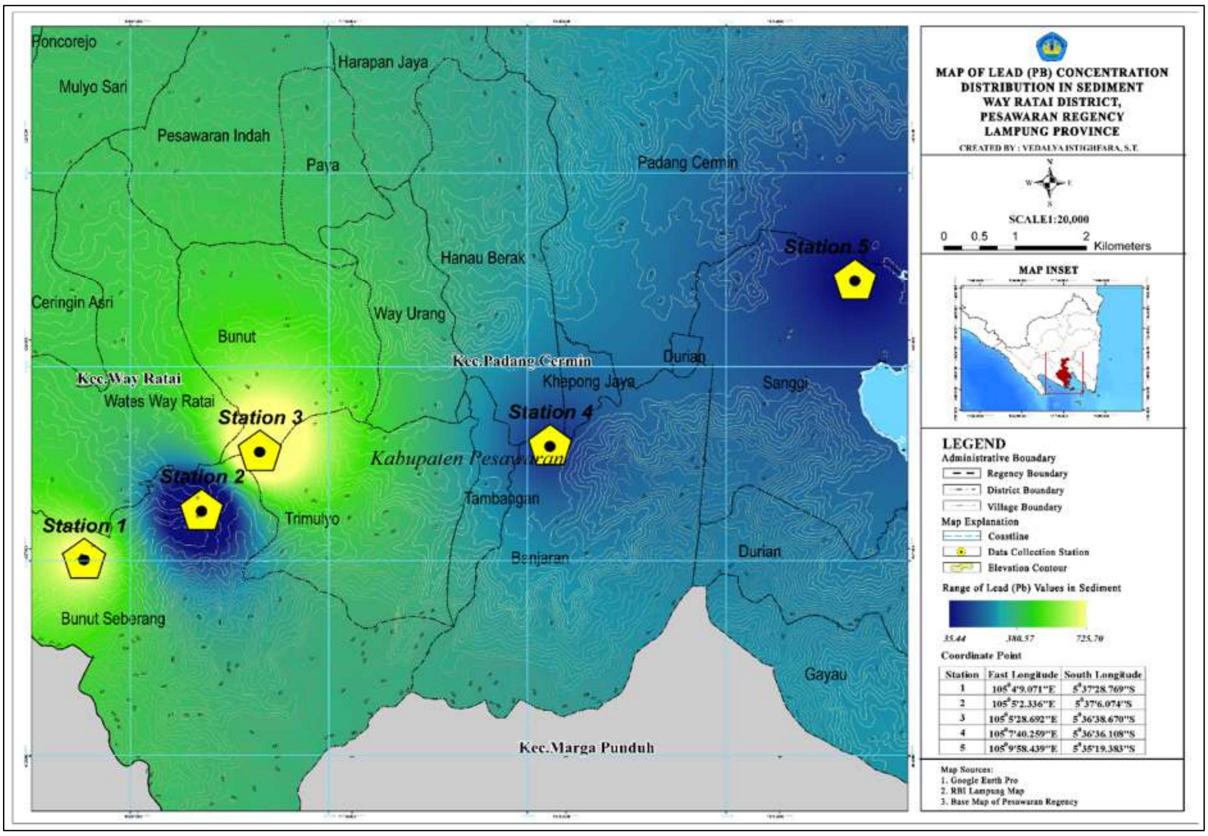

Figure 7. Pb distribution in the sediment of Way Ratai River.

The high Pb content in amalgamation waste is due to the fact that the amalgamation process generally involves using Hg to bind gold metal from the ore. This process can lead to the formation of Pb compounds in the waste due to the reaction between mercury and lead naturally present in the ore. Pb can also be present in gold-bearing rocks due to complex geological processes. The results of the Pb concentration analysis in the Kuin River Banjarmasin sediment by Wibowo et al. (2022) show a very varied concentration between 22.104 ppm and 39.56 ppm. This value exceeds the lowest effect level (LEL) quality limit set in Ontario's Guidelines for the Protection and Management of Aquatic Sediment Quality of 31 mg kg^{-1}. Pb can be present in gold-bearing rocks through hydrothermal processes that simultaneously precipitate metals such as Au and Pb (Indarto et al., 2014). In addition to gold processing activities, high Pb values in river sediments from point 1 to point 5 can be high due to various factors, including industrial, agricultural, and domestic human activities.

Cyanide (CN-) content in water and sediment of Way Ratai River

Figures 8 (water) and 9 (sediment) show the results of measurements taken of CN- and its distribution in the water and sediment of the Way Ratai River. Figure 8 shows the concentration of cyanide in river water at point 1 is (0.001) ppm, point 2 (0.002) ppm, point 3 (0.005) ppm, point 4 (0.002) ppm, and point 5 (0.005) ppm. The five values are still below the quality standard of the Japan Environmental Quality Standard (JEQS) 1997, which is 0.01 ppm, and Appendix VI of PP No. 22 of 2021, which is 0.02 ppm. The cyanide content in water samples is influenced by the distance from the gold mining and processing site; the closer the distance, the higher the content. Point 3 has the highest cyanide concentration because it is the primary location of gold processing using the cyanidation method. Cyanide in small concentrations can be degraded by microbes, such as *Pseudomonas fluorescens* NCIMB 11764, which hydrolyzes cyanide to formic acid and ammonium (Luque-Almagro et al., 2011). Cyanide in CN- ions is not readily soluble in water due to its small size, lack of a large electrical charge, and inability to form strong hydrogen bonds with water molecules (Anggraini and Falahudin, 2021).

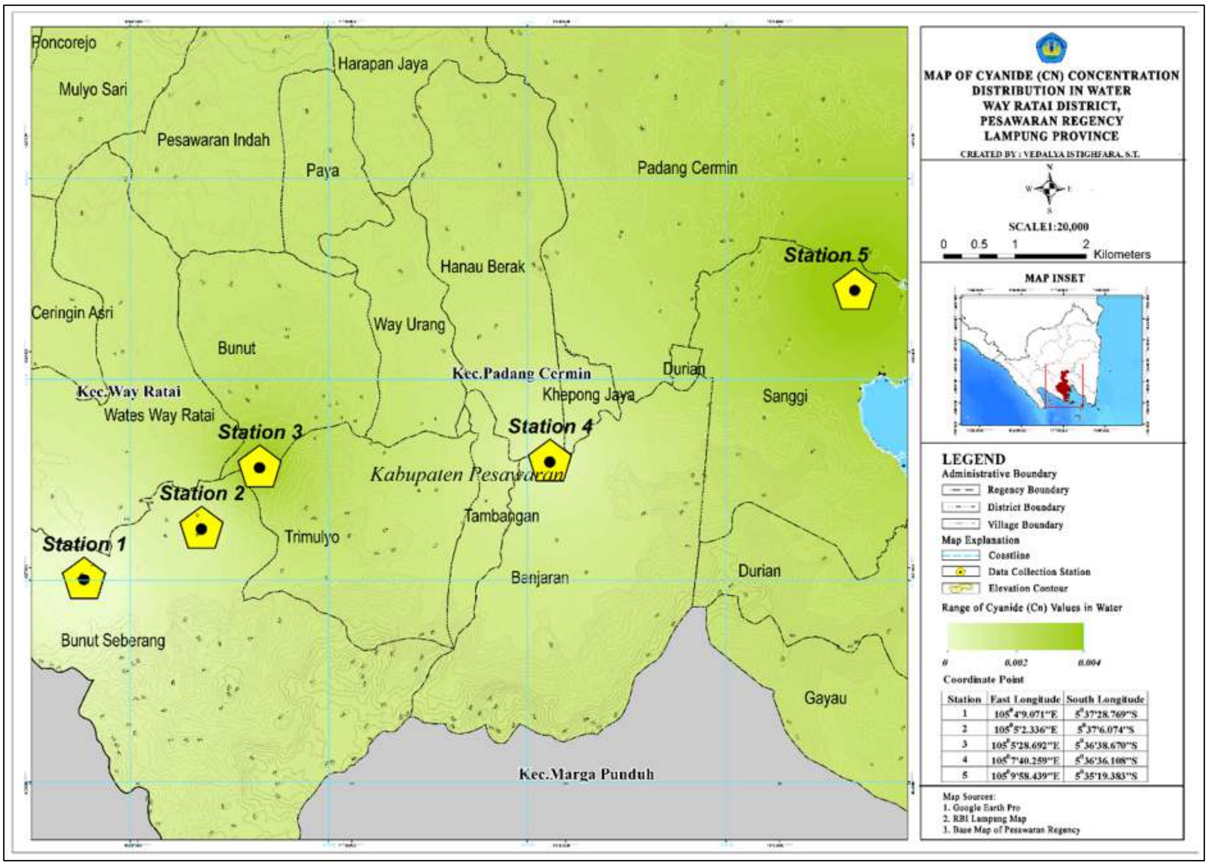

Figure 8. CN- distribution in the water of Way Ratai River.

Based on Figure 9, the cyanide concentration in river sediment at point 1 is (0.41) ppm, point 2 (0.47) ppm, point 3 (0.23) ppm, point 4 (0.5) ppm, and point 5 (0.3). The concentration of CN- at all 5 points is still below the quality standard of Latvia Environmental Quality Standard (LEQS) 2000 and National Oceanic and Atmospheric (NOAA) 1999. The cyanide concentration at point 2 is higher than the other points because this location is close to artisanal gold mining activities.

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Unlike mercury, cyanide in mining tailings can degrade naturally through volatilization, oxidation, photodecomposition, and microbial biodegradation. Biodegradation with a bacterial consortium from gold mine sediments, such as Microbacterium and Brevibacterium, has achieved up to 98% reduction in free cyanide. This method is considered effective as a final treatment step for mine waste (Maria et al., 2023). Cyanide is used in gold processing because of its high extraction success rate, reaching up to 95% (Sabara et al., 2018). Cyanide has direct toxicity effects that are more harmful than mercury, but CN- can decompose or degrade more quickly in the environment. Disposal of cyanide waste without adequate management can contaminate river sediments and have toxic effects on aquatic organisms.

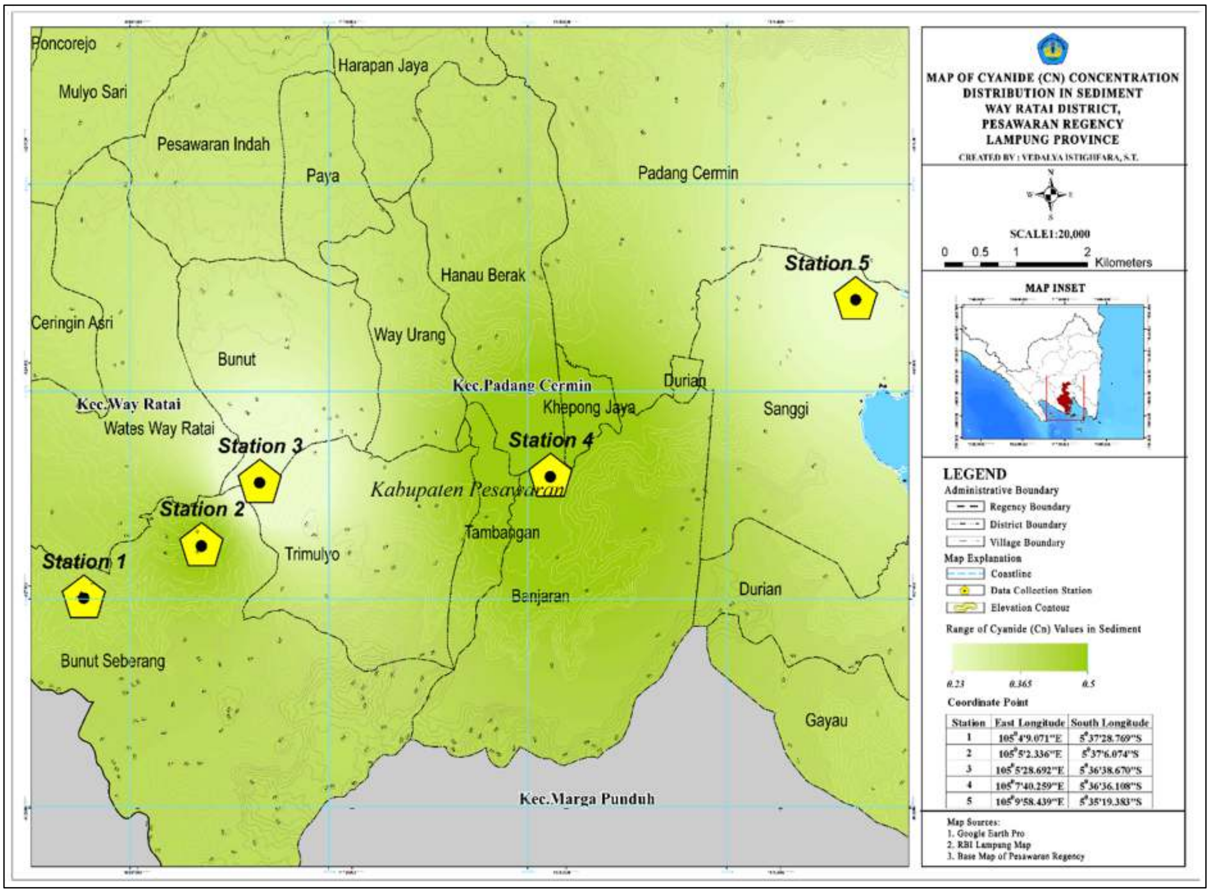

Figure 9. CN- distribution in sediments of Way Ratai River.

Conclusion

The results showed that ASGM waste contains mercury and cyanide from amalgamation and cyanidation processes and other heavy metals such as Pb and As with relatively high concentrations. From the results obtained, the heavy metal content in river sediments is higher than in river water. The higher level of heavy metals in sediments than in water is because heavy metals tend to bind efficiently to organic matter, settle to the bottom of the water, and combine with sediments, which means river sediments can be a significant source of absorption for heavy metals that can have an impact on the environment and living organisms in it. Management of ASGM effluents, such as coagulation and absorption techniques, is essential to reduce potential pollution in the surrounding environment.

Acknowledgments

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