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Research Article

Mercury pollution in the soil and river water of the Ratai watershed by artisanal and small-scale gold mining activities in Pesawaran District, Lampung, Indonesia

Slamet Budi Yuwono[1*], Irwan Sukri Banuwa[2], Nano Suryono[1], Hiroaki Somura[3], Dermiyati[2]

[1] Department of Forestry, University of Lampung, Jl. Sumantri Brojonegoro No. 1, Bandar Lampung, Indonesia
[2] Department of Soil Science, University of Lampung, Jl. Sumantri Brojonegoro No. 1, Bandar Lampung, Indonesia
[3] Graduate School of Environmental and Life Science, Okayama University, 3-1-1 Tsushima-naka, Kita-ku, Okayama, Japan

[*]corresponding author: sbyuwono_unila@yahoo.com

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Abstract

The high risk of mercury pollution in the Ratai watershed due to artisanal and small-scale gold mine activities in Pesawaran District, Lampung, Indonesia, was evaluated. Studies are needed to improve the understanding of the effect of heavy metal pollution, especially mercury (Hg), in soil and river water along the watershed because of erosion. The Universal Soil Loss Equation (USLE) integrated with Geographic Information Systems (GIS) model was used to analyze the transport of mercury (Hg) from nonpoint source pollution loads to the Way Ratai River using rainfall-based erosion. Soils and river water samplings were conducted in 2020. Biophysical conditions, the land cover, and the rainfall data of the Ratai watershed were also taken into account. The results indicated that Hg concentrations in the soil and the river water were high, ranging from 0.26-28.9 mg L^{-1} and from 0.08-14.1 mg L^{-1}, respectively. The reason for the high Hg contents in the soils and the river waters was the high erosion rate in the watershed. As the study area was characterized by high rainfall erosivity and low to high soil erodibility, the erosion-caused Hg contamination in soil and water can be significant if no conservation strategies are developed.

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Introduction

Processing of rocks containing gold (ore) into gold that has very high economic value generally uses the mercury (Hg) amalgamation method, especially for artisanal and small-scale gold mining (ASGM). This type of mining occurs in most nations in the tropics (Villas-Boas et al., 2001; Donkor et al., 2005; Odumo et al., 2014), including Indonesia (Damayanti and Lutfie, 2009; Arifin et al., 2020). However, handling Hg in the field is not optimal due to lacking knowledge, capital, and equipment owned by small-scale miners. On the other hand, the miners generally process ore into gold without using a wastewater treatment plant so that the processing waste (tailings) will be dumped into the ground and carried by rainfall, erosion, and surface runoff, which will eventually enter the river. Esdaile and Chalker (2018) stated that mercury-dependent ASGM could be a source of mercury pollution more than any other human activities. According to Fernández-Martínez et al. (2005), from mining activities, surface runoff or infiltrated water brings metallurgical wastes or Hg pollutants into the environment. So, ASGM by Hg amalgamation causes water resource contamination by heavy metals (Donkor et al., 2005).

Further, mining ore and placer gold deposits have 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 mercury pollution in soils, surface waters, and sediments in areas affected by Hg mining operations in Asturias, Spain is vast. Testing the Hg contents due to ASGM activities in Indonesia, especially Hg in soils (Damayanti and Lutfie, 2009; Mirdat et al., 2013) and water (Yulis, 2018; Arifin et al., 2020) have been carried out. Most of the results show that ASGM causes the mercury content in the soil and water to exceed the mercury quality standard that has been set. However, research on the effect of erosion using USLE models on mercury pollution-dependent ASGM in soils and surface waters is still lacking.

The ASGM activities in Pesawaran District, Lampung, Indonesia**,** are estimated to have been running for more than ten years, so it is estimated that the lands and river waters around the processing site of ore to gold using the amalgamation method have experienced Hg pollution. As we know, Hg pollution is hazardous 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 contaminations in soils, water, sediments, and biota are needed to be concerned because it causes toxicity, persistency, and accumulation in food chains.

Accumulation of Hg in soil and water can occur due to erosion. In nature, the two leading causes of erosion are wind and water. In wet tropical climates such as Indonesia, water is the leading cause of erosion, while the wind does not have a significant effect. In the event of erosion, soil or parts from one place/location are eroded and transported and then deposited in another. Soil erosion by water occurs through three main processes: the detachment of soil (as particles or aggregates) from the soil mass, the movement of loose material, and the deposition (Weil and Brady, 2017; FAO, 2019; Gachene et al., 2019). The considerable surface runoff will increase the amount of erosion so that it will carry Hg to a distant stream, deposit in the soil, the river water, and even reach the sea; it will affect the life of biota in the ocean.

Soil erosion processes occur due to rain and runoff, which are influenced by various factors, including rainfall (intensity, diameter, duration, and amount of rain), soil characteristics (physical properties), land cover, slope steepness, and slope length (FAO, 2019). These factors work simultaneously in influencing erosion. Loss of soil will only occur if the two processes above are carried out. Without a destructive process and soil particles, erosion will not occur; erosion will be minimal without the transportation process. The quantity of Hg that enters soils and water because of erosion is uncertain due to a lack of sufficient data (Panagos et al., 2021). USLE-based soil erosion modelling has been widely used elsewhere (Pandey et al., 2007; Kinnell, 2010; Mahapatra et al., 2018; Borrelli et al., 2021). Soil erosion models can estimate the level of erosion and predict the occurrence of heavy metal pollution due to erosion which can affect health and the environment.

This research aimed to study the direct impact of ASGM activities on Hg accumulation in soil and river water based on the estimation of soil erosion using USLE and GIS, especially in the case of Ratai Watershed Pesawaran District, Lampung, Indonesia.

Materials and Methods

Mapping the research location and the drainage system

The research was conducted in Bunut Seberang Village, Teluk Ratai Subdistrict, Pesawaran District, Lampung Province, Indonesia, from September to December 2020 (Figure 1). The research began with mapping the location of the excavation of gold rock and the location of processing ore into gold. Followed by mapping the drainage system (map of the river network) to estimate the direction of the surface flow to the nearest river from the excavation location and the processing of ore into gold location.

Soil sampling

Soil samples were taken at six locations that were not expected to be affected by mining activities and were suspected of being affected by the upstream, middle, and downstream of the Ratai watershed (the coordinates of sampling locations are shown in Table 3). Samplings of river water were in the upstream part that was not affected by mining activities and in the downstream part that was suspected of being affected by mining activities.

The estimation of erosion

The analysis approach was based on the watershed, where the biophysical conditions, the land cover of the watershed, and the rainfall data significantly affect soil erosion that affects the amount of Hg in the soil and river water. Erosion for each land unit was estimated using the Universal Soil Loss Equation (USLE) model (Wischmeier and Smith, 1978) as follows:

A = R.K.L.S.C.P

In SI units, the average annual soil erosion rate (A) in t ha^{-1} yr^{-1}, the rainfall-runoff erosivity factor (R) in MJ mm h^{-1} ha^{-1} yr^{-1}, the soil erodibility factor (K) in t ha h ha^{-1} MJ^{-1} mm^{-1}; while the slope length and steepness factor (LS), the land cover and management factor (C), the soil conservation or prevention practices factor (P) are dimensionless. The impact of the R factors captures the energy and amount of sediment yield.

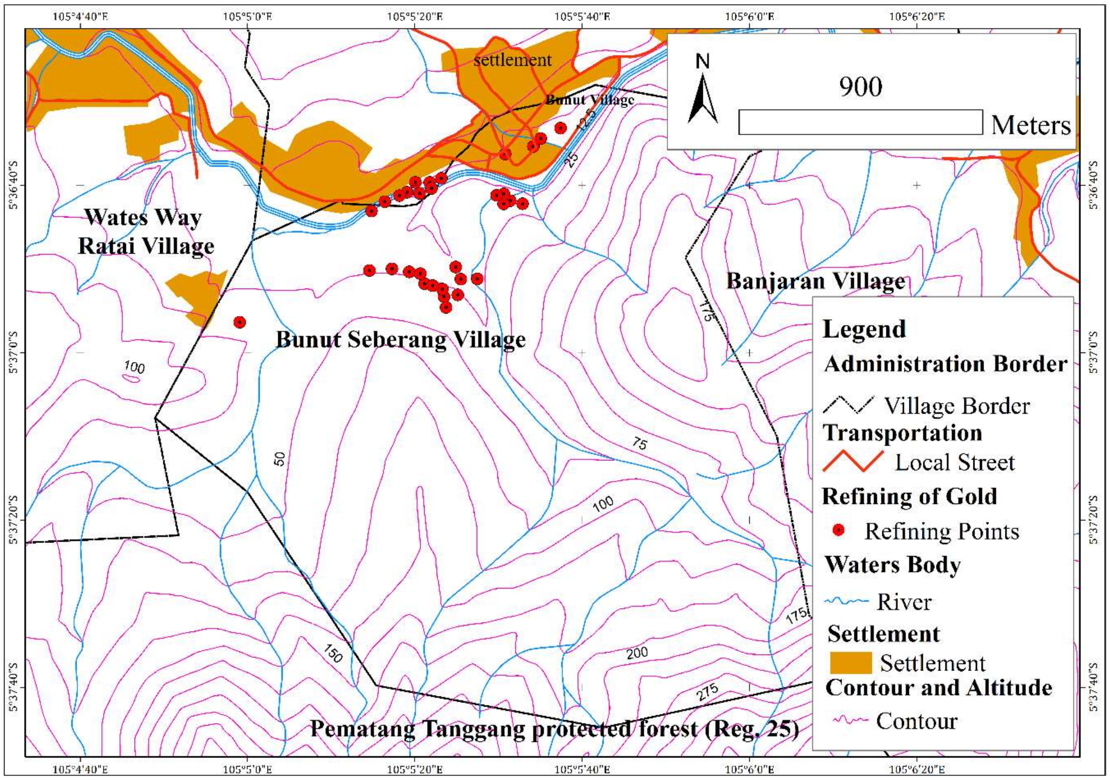

Figure 1. Research location.

USLE-type models have successfully predicted long-term average annual soil loss reasonably well at some geographic locations (Kinnell, 2010; Borrelli et al., 2021). Process-oriented models require an application of the used equations at a given spatial scale, ranging from plot to basin and at the event temporal scale (Alewell et al., 2019). USLE and GIS could be integrated for mapping the spatial distribution of soil erosion from the entire watershed (Bekele and Gemi, 2021).

Rain erosivity factor (R)

Wischmeier and Smith (1978) used EI30 as an index of rain erosivity; because total rain energy (E) and maximum intensity for 30 minutes (I30) annually showed a very close relationship with the amount of eroded soil. The formula calculated the kinetic energy of rain in USLE: E = 210 + 89 log I. In Indonesia, the daily rainfall data for calculating EI were not widely available, so scientists usually used the EI formula developed by Bols (1978). According to Bols (1978), R factor was the sum of monthly rainfall erosion index values based on the following equation:

$$R = \sum_{i=1}^{12} (EI30)i$$

Lenvain (1975) proposed the formula to calculate EI30 value as follows:

$$EI30 = 2.34\ R^{1.98}$$

The total rainfall at the study location is 1,699.5 mm yr^{-1}.

Soil erodibility factor (K)

Soil erodibility, the rate of erosion per rain erosion index for soil that was allowed from standard small plots of 22 m long, was located on a 9% slope without plants. Soil erosion sensitivity was strongly influenced by soil organic matter, soil texture, permeability, and stability of soil structure. Soil erodibility was calculated using the formula (Wischmeier and Smith, 1978):

$$100K = \{1.292\ (2.1\ M^{1.44}\ (10^{-4})(12 - a) + 3.25\ (b - 2) + 2.5\ (c - 3)\}$$

where:

K = Soil erodibility (dimensionless)
M = Soil texture class (% fine sand + % dust) (100 - % clayey)
A = Organic material (%)
B = Soil structure code
C = Soil profile permeability code

Slope length and steepness (LS)

Slope length factor (L) was the ratio between the amount of erosion on a particular slope length and soil erosion with a slope length of 22 m in identical conditions. Meanwhile, the slope steepness factor (S) was the ratio between the amount of soil erosion at a particular slope and soil erosion at a slope of 9% with identical conditions. LS factors could also be calculated directly (combined) according to the formula (Wischmeier and Smith, 1978):

$$LS = \sqrt{X(0{,}0138 + 0{,}00965S + 0{,}00138S^2)}$$

where:

LS	=	Slope length and steepness factor
X	=	Length of slope (m)
S	=	Slope steepness (%)

The topographical conditions of the research site were mostly 61% in the steep category, 19% wavy, and 20% flat.

Vegetations factors and management (C)

The determination of the C factor for various plants, such as mixed cropping, coffee, etc., was based on various previous studies (Wischmeier and Smith, 1978).

Conservation action factor (P)

The conservation action factor was also determined based on various previous studies (Wischmeier and Smith, 1978).

Surface runoff volume

Estimating surface runoff volume in a watershed used a rainfall-runoff relationship model, namely the USDA- Soil Conservation Services (SCS) method. The amount of runoff volume (Q) depends on the rainfall (P) and the volume of storage available to hold water (S). The equation used is:

$$Q = \frac{(P - 0.2S)^2}{P + 0.8S}$$

where:

Q	=	Total surface flow (mm)
P	=	Rainfall (mm)
S	=	Maximum potential water retention (mm)

Based on the empirical equation, the value of S is estimated using the equation:

$$S = \frac{25400}{CN} - 254$$

where:

S	=	Maximum potential water retention (mm)
CN	=	Curve number (runoff curve number)

The runoff curve number depended on the soil properties, soil use, hydrological conditions, and previous water conditions. CN values were determined based on soil type, land use, infiltration, and soil hydrological conditions (previous groundwater conditions).

The Erosion Index

The erosion index was the ratio between actual erosion and tolerable soil loss. Based on data on land cover, soil type, topography, and rainfall, using the USLE method, the amount of erosion could be estimated. The magnitude of the erosion index described the erosion hazard level.

$$\text{Surface Run Coefficient (SRC)} = \frac{\text{Total DRO a year (764.81 mm)}}{\text{Total rainfall a year (1699.5 mm)}} \text{ x } 100\% = 45\%$$

The estimation of the surface runoff volume at the research location using the SCS (Soil Conservation Services-USDA) method was 45%, called the surface runoff coefficient (SRC). From the total rainfall that falls, 45% of rainfall would become surface runoff and enter the river flow. According to the Minister of Forestry of the Republic of Indonesia (2014), the surface runoff coefficient of 45% is categorized as high. Excessive runoff volume could potentially cause flooding downstream. Indonesian climate is generally divided into wet and dry climates. From the research data, November till March was wet months, while July till October was dry months. Oldeman's climate classification was to make climate types based on the number of wet months and dry months**.** The wet month category was the month with rainfall >200mm and the dry month with rainfall <100 mm. The annual rainfall that accumulates in a short period (December-February) causes the soil to be unable to accommodate all the volume of rainwater. As a result, most of the rainwater became surface runoff; this was exacerbated by the increasing conversion of forest functions to other uses such as agriculture, housing, industry, and rice fields, which could cause considerable flooding in the downstream area. Furthermore, it could be said that the significant surface runoff would also cause excessive erosion, which would directly reduce soil fertility. Decreasing soil fertility would cause less vegetation to grow properly, so the land cover would decrease. Therefore, recharging water reserves in the upstream area would reduce and result in drought during the dry season.

Mercury (Hg) analysis method

Mercury (Hg) contents in the soils and river water were analyzed using Cold Vapor Atomic Absorption Spectroscopy (CV-AAS) or a mercury analyzer. The analysis used US EPA SW-846-7470A method. In principle, Hg^{2+} ions were reduced by Sn^{2+} to Hg atoms, and then these atoms were analyzed quantitatively with a cold vapor-atomic absorption spectrophotometer at a wavelength of 253.7 nm. Preparation of a calibration curve in the range of 1 g Hg L^{-1}-20 g Hg L^{-1} by inserting 100 mL of a working standard Hg solution at levels (1, 2, 4, 8, 10, 15 and 20) g Hg L^{-1} into each 250 mL Erlenmeyer each. Then add 5 mL of concentrated H_2SO_4 and 2.5 mL of concentrated HNO_3 into each Erlenmeyer, then add 15 mL of $KMnO_4$ solution and wait up to 15 minutes. If the purple color disappears, add more $KMnO_4$ until the purple color does not disappear. After that, add 8 mL of $K_2S_2O_8$ and heat it in a water bath for 2 hours at 95 ºC, then cool it to room temperature. If the temperature of the solution has cooled, then add enough NaCl hydroxylamine solution to reduce the excess $KMnO_4$ and add 5 mL of $SnCl_2$. Immediately measure the level of Hg in the solution using a cold steam AAS that has been optimized according to the instructions of the tool (Hadi and Aisah, 2015).

Data analysis

A Geographic Information System (GIS) was integrated with the USLE (Universal Soil Loss Equation) model in the identification of rainfall-based erosion and the transport of mercury (Hg) from nonpoint source pollution loads to the Way Ratai river. ArcGIS version 10.3 was used. Data on soil types, land cover conditions, and topography were analyzed descriptively to the extent that mining impacts would pollute soils and river water.

Results and Discussion

Soil type and land coverage

The total area of the watershed as the basis for analysis was 2,667.01 ha. The soil type in the research location was divided into two parts, namely, Inceptisols and Ultisols. Inceptisols dominate the watershed area of as much as 85.07% (2,268.8 ha), and Ultisols cover an area of 14.93% (398.17 ha); the soil type map is presented in Figure 2. Inceptisols are young soils with a high level of fertility. Meanwhile, Ultisols are old soils that have undergone advanced leaching levels so that they have low base saturation and low pH with argillic or kandic horizons (Subardja et al., 2014). Based on data presented in Table 1, the soil pH at the study site ranged from 5.28 to 6.46, including the slightly acidic category. Ignatavičius et al. (2022) stated that pH is one of the factors that affect the release of Hg into the environment. Hg uptake is affected by soil pH and decreases with acid pH. On the other hand, the contents of N, P, and K in the soil are in the very low category; total N ranged from 0.01% to 0.15%, available P ranged from 3.54 ppm to 18.48 ppm, and exchangeable K ranged from 0.24 me 100 g^{-1} to 1.05 me 100 g^{-1}.

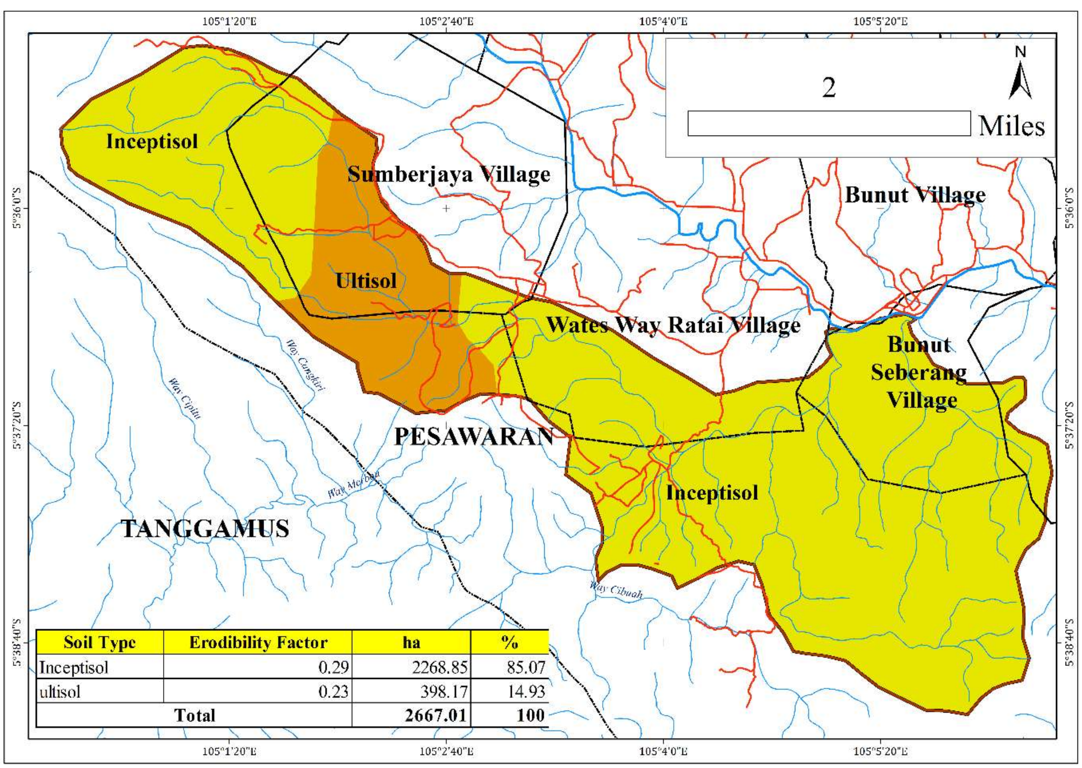

Figure 2. Soil types at the research location.

Table 1. Soil chemical and physical properties of site location.

Location	pH	Total N (%)	Available P (ppm)	Exchangeable K (me $100g^{-1}$)	Soil Texture		
					Sand (%)	Silt (%)	Clay (%)
1	6.46	0.01	7.30	0.47	68.13	17.83	14.04
2	6.04	0.03	18.48	0.89	67.64	16.01	16.35
3	5.64	0.02	4.55	0.39	82.77	7.43	9.80
4	5.85	0.15	3.54	1.05	36.99	42.72	20.29
5	5.86	0.01	5.06	0.24	82.70	7.46	9.84
6	5.28	0.03	6.57	0.50	70.05	11.70	18.25

Source: Soil Science Laboratory, University of Lampung (2020).

Low nutrients indicate low soil fertility due to the high level of leaching that occurs. Nutrients of N, P, and K are utilized by plants and microbes, and some are lost because they are leached into the soil and carried away by surface runoff. According to FAO (2019), the loss of surface material causes a decrease in soil nutrients. Nutrient loss could be prevented by implementing effective land management policies (Yustika et al., 2019). Table 1 also shows that the sand content in the study area ranged from about 37% to 70%, silt about 7% to 43%, clay about 10% to 20%. The high sand content indicates that the study area has undergone further leaching. The higher the sand content, the lower the ability of the soil to bind ions and the higher the leaching, which will cause Hg to be eroded and contaminate the environment. The low clay content at the study site causes Hg not to be adsorbed by the soil. Ignatavičius et al. (2022) stated that Hg sorption capacity is influenced by the amount and quality of clay. The land cover conditions are presented in Figure 3. The research location was dominated by forest cover of 81.78%, followed by dry land agriculture of 15.30%, settlement of 1.8%, and rice fields of 1.17%. It can be seen that the research areas were mostly covered by forest. According to Torri and Poesen (2014), with increasing vegetation density, resistance by the soil to concentrated flow erosion increase runoff discharge during a rainfall event decreased. Generally, sheet and rill erosion was reduced by 50% at about 20%, 75% at about 30 to 35%, and 90% at about 60% vegetation covers (Gyssels et al., 2005). In addition, the soil's physical properties, the topographic soil conditions, and the rainfall will also affect the amount of erosion. For this reason, the topography and rainfall conditions of the research location must be considered. Topographic conditions and rainfall of the research location are presented in Figure 4 and Table 2, respectively.

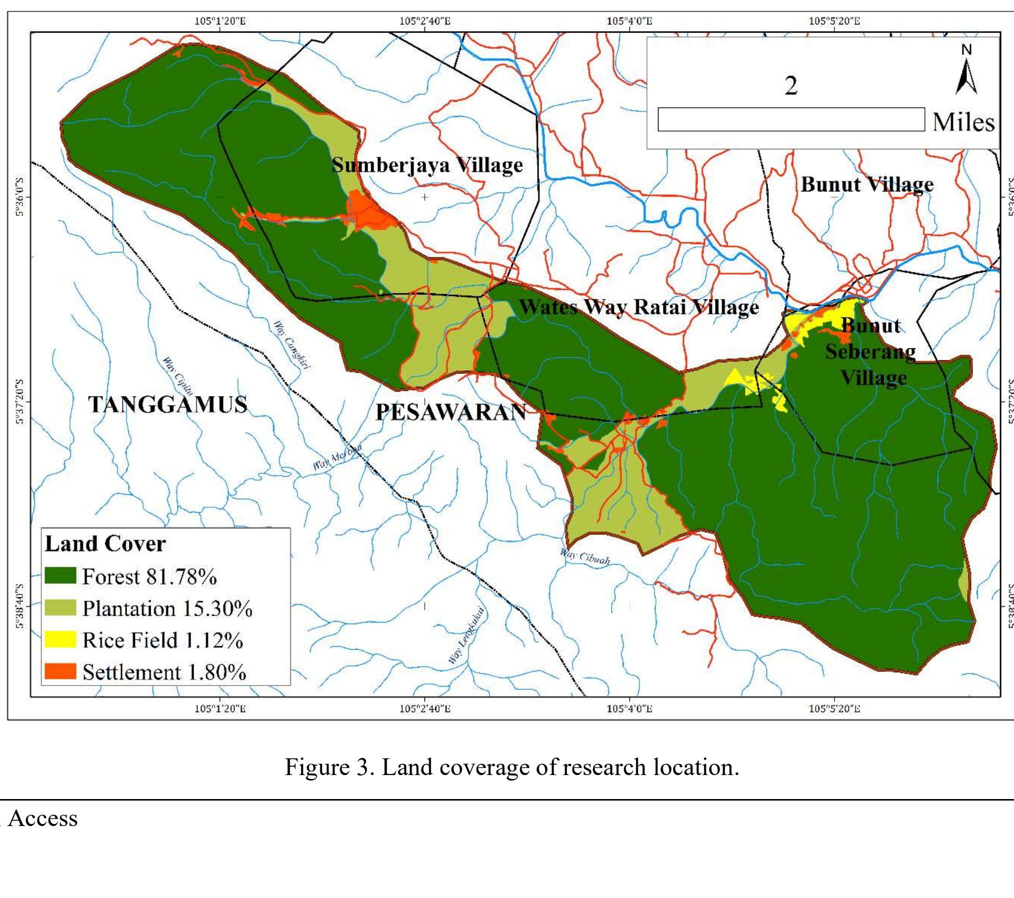

Figure 3. Land coverage of research location.

In general, the erosion index in the research location was in the very low to the low category, as much as 86%, while in the medium to the very high category was only 14%. The erosion index is presented in Figure 5. The surface runoff is determined mainly by the surface runoff coefficient, where surface runoff results from the response of forest cover, topography, and soil type to falling rainfall.

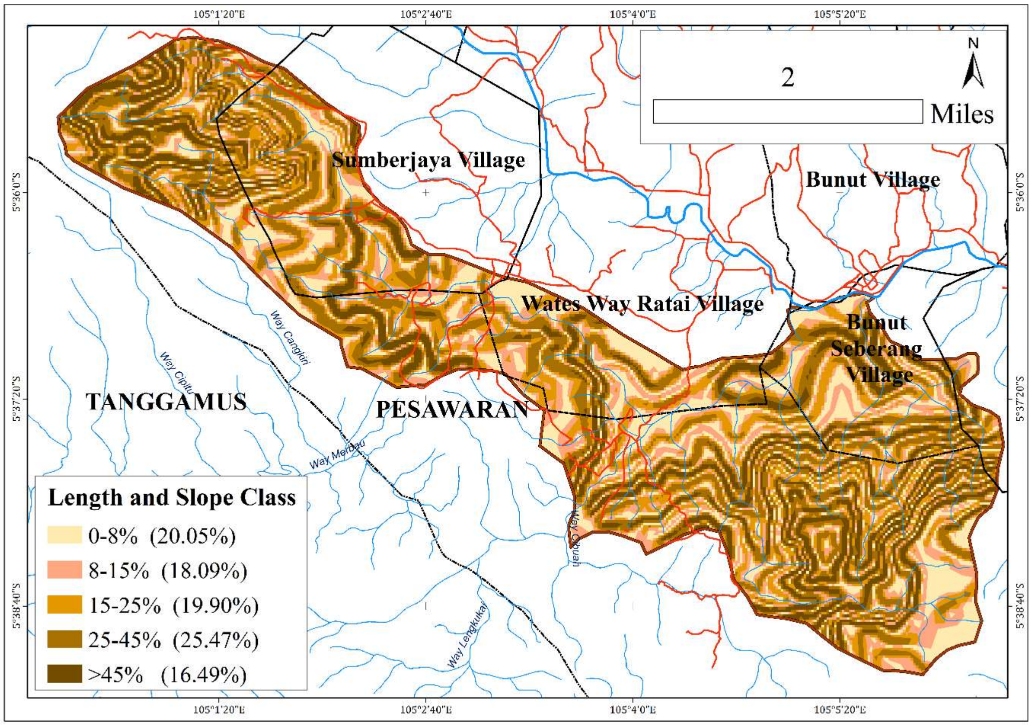

Figure 4. Topographic condition of research location.

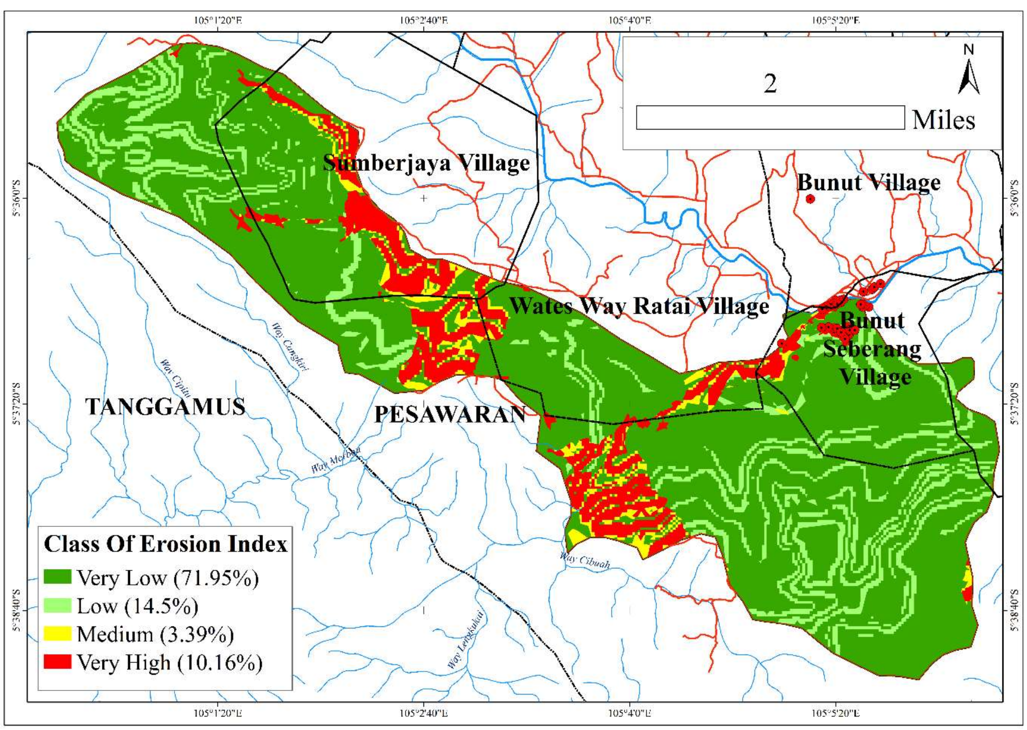

Figure 5. Erosion index of site location.

Soil erosion was affected by wind, rainfall, and associated runoff processes, the vulnerability of soil to erosion, and the characteristics of land cover and management (Aksoy and Kavvas, 2005; Panagos et al., 2015). The results of the surface runoff in the location sites are presented in Table 2.

Table 2. The surface runoff volume from the research area.

Month	Rainfall (mm)	Direct Runoff (DRO) (mm)
January	124	143.54
February	202	74.19
March	205	115.53
April	167	85.77
May	150	119.18
June	105	59.96
July	0	0
August	15	23.26
September	139	13.46
October	65	30.18
November	286	64.40
December	255	35.35
Total	1699.5	764.81

During wet months (from January to June), rainfall ranged from 105 mm to 205 mm (with an average of 158.83 mm). While, direct runoff (DRO) ranged from 59.96 mm to 143.54 mm (with an average of 99.70 mm). DRO was calculated by US Soil Conservation Service (SCS) method as described previously. From these data, Surface Run-off Coefficient (SRC) a year, which is the ratio between total DRO and total rainfall a year, was calculated, and the value was 0.45. It means that 45% of the total rainfalls a year would become surface runoff and flow to the river. SRC 0.45 is included as a high category. FAO (2019) stated that rainfall and runoff affected the rate of water erosion at a site because both detach and transport the eroded soils.

Mercury (Hg) contents in the soils and river water

The water and soil sampling locations are presented in Figure 6, while the mercury contents in the soils and river water are shown in Table 3. The results showed that the mercury contents in all samples from soil and river water were very high from the acceptable range. Hg content in soils ranged from 0.26 to 28.9 mg L^{-1}, and in the river water ranged from 0.08 to 14.1 mg L^{-1}. The mercury contents in the soils and water at all sampling points have exceeded the quality standard. Actually, the quality standard for mercury in soil and water should not exceed 0.005 mg L^{-1} based on Indonesian Government Regulation No. 82 year 2001 concerning water quality and pollution control (President of the Republic of Indonesia, 2001). The reasons why Hg contents in soils and river waters are high; because, firstly, there is no proper waste management; secondly, the distance from the ore-to-gold refining or processing site is close to the river flow. In this study, the distances of the soil samples from the processing site are between 30-663 m, while the water samples are between 30 m and 1,161 m. Martin et al. (2021) stated that most of the released Hg was deposited within 15 km of the source.

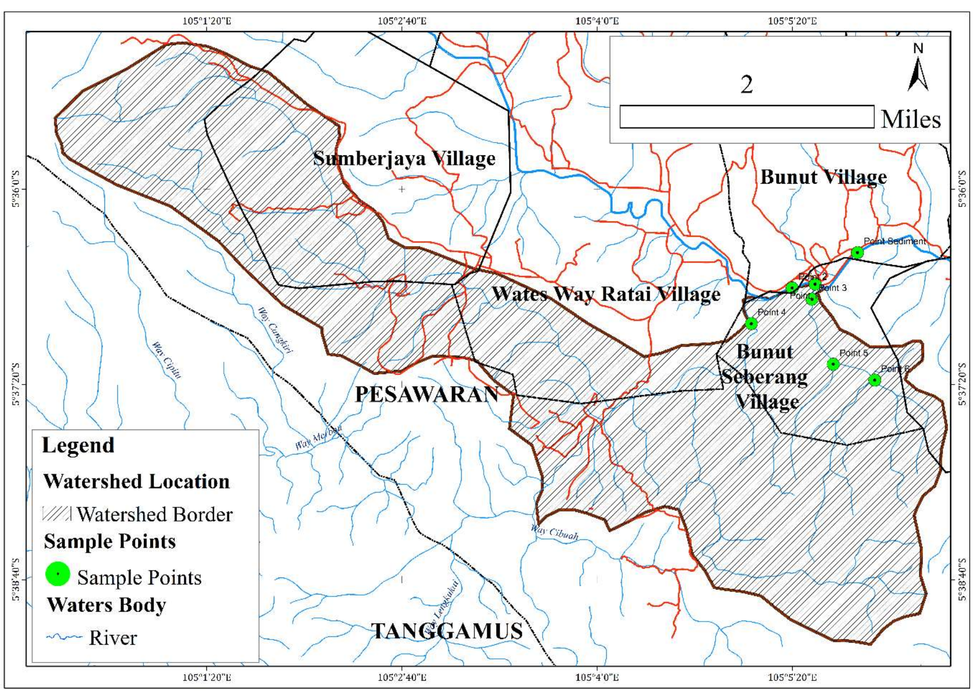

Figure 6. The soil and water sample's point location.

Table 3. The mercury (Hg) content in the soil and river water samples.

Sampling Point	Coordinates of soil location		Coordinates of water location		Hg content ($mg\ L^{-1}$)		Analysis Method
	X UTM	Y UTM	X UTM	Y UTM	Soil	Water	
1	510135	9379816	510131	9379803	3.84	9.6	US EPA SW-846-7470A
2	509845	9379768	509844	9379771	5.75	14.1	US EPA SW-846-7470A
3	510094	9379626	510092	9379626	21.8	0.08	US EPA SW-846-7470A
4	509330	9379319	509331	9379319	28.9	0.88	US EPA SW-846-7470A
5	510363	9378808	510363	9378809	0.26	0.08	US EPA SW-846-7470A
6	510886	9378609	510886	9378609	0.91	0.14	US EPA SW-846-7470A

Mercury contents in the soils and the river water were strongly influenced by erosion and surface runoff at the study site ($p<0.05$). Water erosion depends on rainfall, which is the source of rain splash detachment, while runoff is generated during the rainfall event. Moreover, from the research data, it can be said that the study area was characterized by high rainfall erosivity. The erosion that occurred at the study site was also high. The erosion index (EI) in the research area ranged from very low to high. Therefore, potential erosion ranged from weak to very strong. Although, more than 80% of the research area was covered by forest, which was in the outstanding category. However, the slope with a steepness of more than 25% was about 42%, categorized as steep, resulting in high surface flow and causing high erosion.

The surface flow or runoff brought Hg particles into the soils and river water. Consequently, high erosion will be related to the polluted Hg in the environment. Therefore, small-scale gold mining activities without permission have brought the soil and water bodies (rivers) into the severely polluted category. Likely, ASGM activities in Pesawaran District caused Hg pollution in soils and surface waters of the Ratai watershed. Saturday (2018) reviewed that environmental pollution due to Hg contamination occurred as shown by Hg concentration found in soils, sediments, and water samples. Besides, a high Hg content in the soil and river water is dangerous for health. Hg can be transformed by bacteria into methylmercury, and it then bioaccumulates in fish, shellfish, and other high-level predators, thus posing a human health risk (Habiba et al., 2017). Mercury may have toxic effects on the nervous, digestive, immune systems, lungs, kidneys, skin, and eyes.

There was no correlation between the sampling distance and Hg contents in the soils and river water. It is likely because the ore-to-gold processing is not carried out continuously over time, depending on the availability of ore material from the mining area. However, there was a tendency that the shorter the sampling distance from the ore to the gold processing site, the higher the Hg content in soils and water samples. Similarly, Odumo et al. (2014) stated the closer the distance from mining areas, the higher the mercury levels**.**

Conclusion

Mercury (Hg) pollution in the soil and river water of the Ratai watershed by ASGM activities in Pesawaran District, Lampung, Indonesia, is high. The Hg contents in the soil and river water samples have exceeded the quality standard for Indonesia's Hg pollution criteria. Moreover, Hg pollution in the Ratai watershed by ASGM activities is related to high erosion because of high surface runoff and steep slopes in the research area.

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