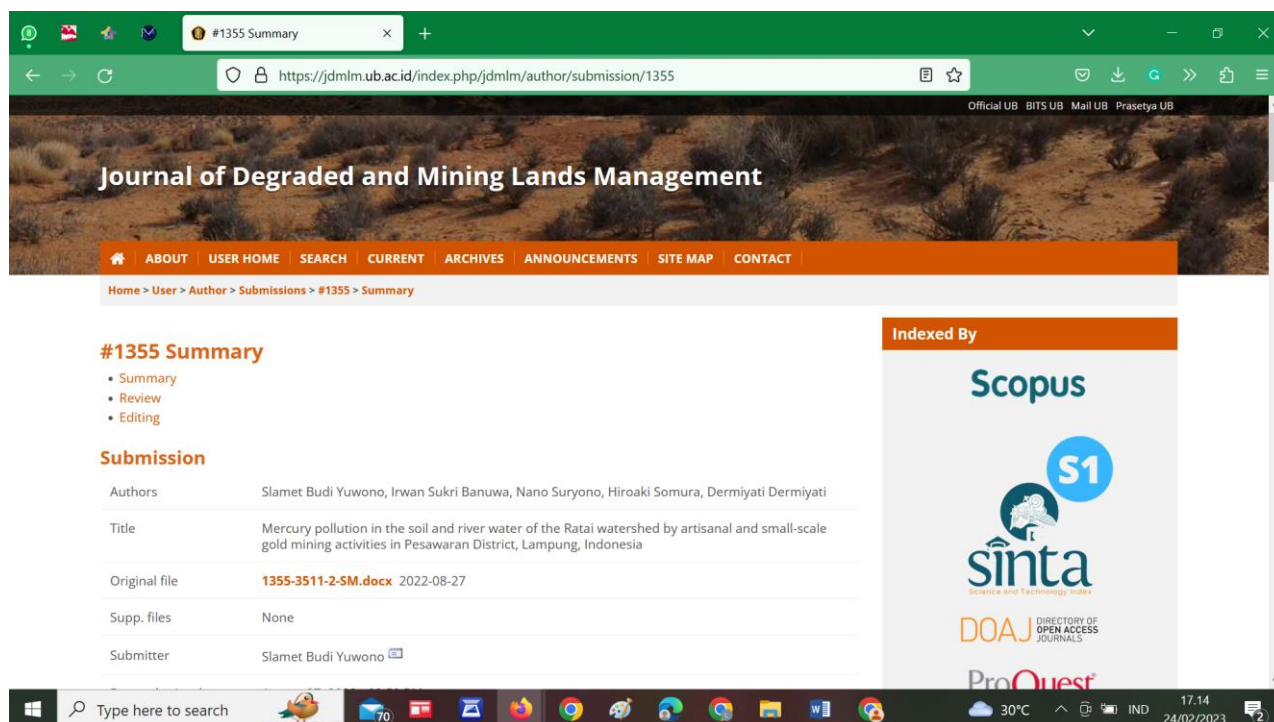


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Resumably it can be processed further. Looking forward to hearing from you soon.

Salam hormat,



Slamet Budi Yuwono

Research Article

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

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Abstract

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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 high risk of mercury pollution in the Ratai watershed due to Artisanal Small-Scale Gold Mine activities in Pesawaran District, Lampung, Indonesia, was evaluated. The Universal Soil Loss Equation (USLE) integrated with Geographic Information Systems (GIS) model is 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 concentration in the soil and the river water ranged from 0.26 – 28.9 mg L⁻¹ and from 0.08 – 14.1 mg L⁻¹, respectively. The Hg contents are high and above the quality standard for mercury in soils and water based on Indonesian Government Regulation Number 82 of the year 2001, which should not exceed 0.005 mg L⁻¹. The reason for the high Hg contents in the soils and the river waters was due to high erosion rate in the watershed. As the study area was characterized by high rainfall erosivity and low to high soil erodability, 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 small-scale gold mining (ASGM). This type of mining occurs in most nations in the tropics (Villas-Boas et al., 2001), including Indonesia, because it is becoming a vital source of income for the communities (ILO, 1999). However, handling Hg in the field is not optimal due to limited 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 run-off, which will eventually enter the river. Esdaile and Chalker (2018) stated that Mercury-dependent ASGM is the largest source of mercury pollution on Earth and causes more mercury pollution than any other human activities. According to Fernández-Martínez et al. (2005), from mining activities, pollutants can reach into the mine water system and ultimately into the environment when the mine/metallurgical wastes enter contact with surface run-off or infiltrated water. Donkor et al. (2005) stated that ASGM by Hg amalgamation is the primary source of water resource contamination with heavy metals.

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,

2002). Fernández-Martínez et al. (2005) stated that mercury pollution in soils, surface waters, and sediments in areas affected by Hg mining operations in Asturias, Spain is vast. However, data about the mercury pollution-dependent ASGM in soils and surface waters due to erosion in Indonesia are 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 water 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) and the environment (Donkor et al., 2005). Therefore, the contamination of soils, sediments, water, and biota by Hg has become a primary concern because of their toxicity, persistence, and accumulation in food chains.

Accumulation of Hg in soil and water can occur due to erosion. Erosion is moving or transporting soil or parts of soil from one place to another by natural media. In nature, two leading causes are active in this process: 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 (Arsyad, 2006). The erosion process occurs through crushing, transportation, and deposition (Meyer et al., 1991; Weil and Brady, 2017). 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). A considerable surface run-off 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 so that it will affect the life of biota in the ocean.

Soil erosion processes occur due to rain (rain) and run-off, which are influenced by various factors, including rainfall (intensity, diameter, duration, and amount of rain), soil characteristics (physical properties), land cover, slope steepness, slope length (Wischmeier and Smith, 1978). 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 amount of erosion can be measured directly in the field using small plots or predicted using models. Erosion prediction models that are commonly used today are parametric. A parametric model to predict the erosion of a plot of land has been developed by Weischmeier and Smith (1978), known as The Universal Soil Loss Equation (USLE). USLE allows planners to estimate the average erosion rate of a given soil on a steep slope with a specific rainfall pattern for each type of cropping and management action (soil conservation action) that may be taken or is being used. The equations used to classify the

various physical and management parameters that affect the rate of erosion into six main variables whose values for each site can be expressed numerically. To predict the erosion in the research area, The USLE is integrated with Geographic Information System (GIS).

The research aimed to study the direct impact of ASGM activities on Hg accumulation in soil and river water based on the estimation of the soil erosion with associated nonpoint source pollution loads, 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 District, Pesawaran Regency, Lampung Province, Indonesia, from September to December 2020 (Figure 1).

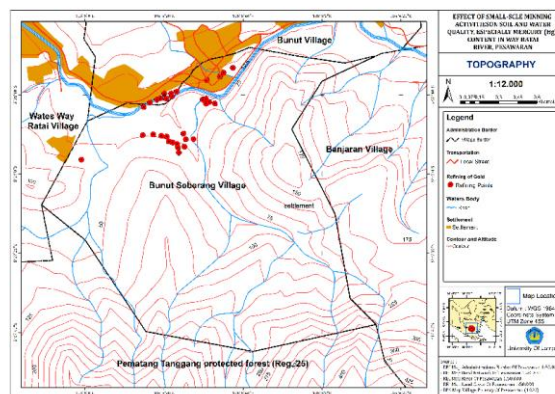


Figure 1. Research Site Location

The research implementation was begun by 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. 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 in this approach, the biophysical conditions, the land cover of the watershed, and the rainfall data would significantly affect the soil erosion that would affect the amount of Hg in the soil and river water.

Erosion estimation for each land unit was calculated using the Universal of Soil Loss Equation (USLE) model (Wischmeier and Smith, 1978). The determination of the value of the factors in the USLE model can be calculated using existing formulas:

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

Where: A ($\text{Mg ha}^{-1} \text{ yr}^{-1}$) is the annual average soil erosion, R ($\text{MJ mm h}^{-1} \text{ ha}^{-1} \text{ yr}^{-1}$) is the rainfall-runoff erosivity factor, K ($\text{Mg h MJ}^{-1} \text{ mm}^{-1}$) is the soil erodibility factor, L (dimensionless) is the slope length factor, S (dimensionless) is the slope steepness factor, C (dimensionless) is the land cover and management factor, P (dimensionless) is the soil conservation or prevention practices factor. The impact of the factors R capturing the energy and amount of sediment yield.

USLE-type models are designed to predict long-term average annual soil loss; they have successfully predicted event soil losses reasonably well at some geographic locations (Kinnell, 2010). The choice of a soil erosion prediction tool depends on the spatial and temporal scale of the intended model application, as the question of scale is crucial in choosing the right modeling approach. 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). Integrating USLE and GIS is an effective tool for mapping the spatial distribution of soil erosion from the entire watershed (Bekele and Gemi, 2021).

Rain erosivity factor (R). Rain erosion is the number of rain erosion index units which is the multiplication of total rain energy (E) with a maximum rainfall intensity of 30 minutes (I30) annually. Wischmeier and Smith (1978) use EI30 as an index of rain erosivity; because the product of rain energy (E) and maximum intensity for 30 minutes (I30) shows a very close relationship with the amount of eroded soil. The formula calculates the kinetic energy of rain in USLE: $E = 210 + 89 \log I$. In Indonesia, the daily rainfall data for calculating EI are not widely available, so usually use the EI formula developed by Bols (1978).

According to Bols (1978), the rain erosivity factor (R) is the sum of the monthly rainfall erosion index values and is calculated based on the equation:

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

The value of EI30 is calculated using the following formula proposed by Lenvain (1975 in Bols, 1978):

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

Soil erodibility factor (K). Soil erodibility, the rate of erosion per rain erosion index for a soil that is allowed from standard small plots of 22 m long, is

located on a 9% slope without plants. Soil erosion sensitivity is strongly influenced by soil texture, soil organic matter content, permeability, and stability of soil structure. Soil erodibility is calculated using the Wischmeier and Smith formula (1978):

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

Where:

K = Erodibility of soil (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 slope (LS). Slope length factor, namely the ratio between the amount of erosion on a particular slope length and soil erosion with a slope length of 22 m and in identical conditions.

Meanwhile, the slope factor is the ratio between the amount of soil erosion at a particular slope and soil erosion at a slope of 9% with identical conditions. Factors for the slope length and slope can also be calculated directly (combined) according to the formula (Wischmeier and Smith, 1978):

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

LS = Slope length and slope factor

X = Length of slope (m)

S = slope steepness (%)

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

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

Estimating surface run-off volume in a watershed can use a rainfall-runoff relationship model, namely the U.S. method Soil Conservation Services (SCS). The amount of run-off volume (Q) depends on the rainfall (P) and the volume of storage available to hold water (S).

The equation used is:

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

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:

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

CN

S = Maximum potential water retention (mm)

CN = Curve number (run-off curve number)

The run-off curve number depends on the properties of the soil, soil use, hydrological conditions, and previous water conditions. CN values are determined based on soil type, land use, infiltration, and soil hydrological conditions (previous groundwater conditions).

Mercury (Hg) analysis method

Mercury (Hg) contents in the soils and river water were analyzed using EPA's method, which is Method 7470a Mercury in Liquid Waste (Manual Cold-Vapor Technique) (U.S. EPA 1994). Method 7470 is a cold-vapor atomic absorption procedure approved for determining mercury concentration in mobility-procedure extracts, aqueous wastes, and ground waters. (Method 7470 can also be used for analyzing particular solid and sludge-type wastes; however, Method 7471 is usually the method of choice for these waste types). All samples must be subjected to an appropriate dissolution step prior to analysis. The Hg concentration was measured using Atomic Absorption Spectroscopy (AAS).

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. Further, data on Hg contents in soil and river water were also correlated with the distance of Hg pollutants from the nearest purification (nonpoint source pollution).

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 soil dominated

the watershed area as much as 85.07% (2,268.8 ha), and Ultisols soil covered an area of 14.93% (398.17 ha); the soil type map is presented in Figure 2. Ultisols are categorized as soil orders susceptible to surface soil loss through erosion. Ultisols have soil horizon Fragic or Duric Layers cemented by iron, aluminum, or silica (Larson and Pierce, 1994; Pennock, 1997).

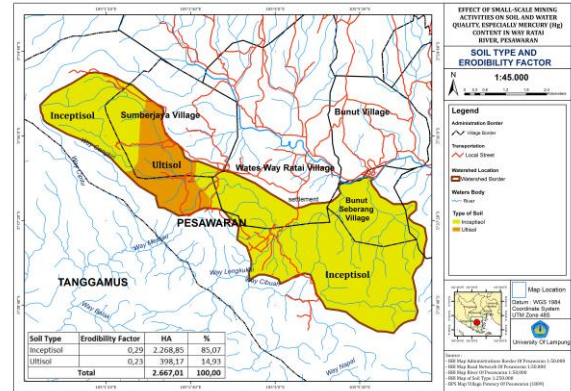


Figure 2. The Soil Type at Research Location

Soil chemical and physical properties data are presented in Table 1. Soil conditions were described by the mineral and soil chemical content, based on the criteria for soil nutrient status by Soil Research Institute (1983). Generally, soil pH ranged from 5.28 to 6.64, including the slightly acidic category. The total soil nitrogen (N) content ranged from 0.01-0.15%, including the very low to low category. The available soil phosphorus (P) content ranged from 3.54-15.48 mg L⁻¹, including the very low to low category. At the same time, the soil exchangeable potassium content ranged from 0.24 to 1.05 me 100g⁻¹, including medium to very high category. The soils in the study area had low soil fertility, primarily N and P, due to the leaching of nutrients by surface run-off. According to FAO (2019), the loss of surface material leads to a decrease in the nutrient-supplying power of the soil. Yustika et al. (2019) stated that nutrient loss could be prevented by implementing effective land management policies.

The physical condition of the soil related to the erosion process is the soil texture. In general, the soil texture in the research location included sandy soil sensitive to erosion. The loss of surface material leads to decreased nutrient-holding power, most pronounced in sandy soils (FAO, 2019).

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

Location	pH	Total N (%)	Available P (ppm)	Exc. K (me 100g ⁻¹)	Soil Texture		
					Sand (%)	Loam (%)	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	82.7	7.46	9.84
6	5.28	0.03	6.57	0.50	70.05	11.70	18.25

*Soil Science Laboratory, University of Lampung (2020)

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 are mostly covered by forest. Torri and Poesen (2014) mentioned that with increasing vegetation density, there is an increase in resistance by the soil to concentrated flow erosion and a decrease in run-off discharge during a rainfall event. Generally, sheet and rill erosion is reduced by 50 percent at vegetation covers of about 20 percent, 75 percent at covers of about 30 to 35 percent, and 90 percent at covers of about 60 percent (Gyssels et al., 2005).

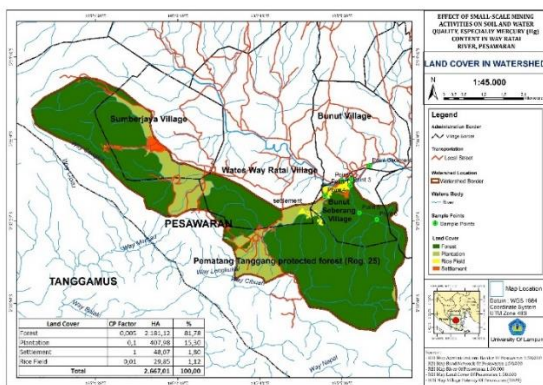


Figure 3. Land coverage of research location

The amount of erosion that occurs in the area is strongly influenced by the land cover conditions. 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 Figures 4 and 5, respectively. The total rainfall at the study location was 1699.5 mm yr⁻¹, while the topographical conditions were mostly 61% in the steep category, 19% wavy, and 20% flat.

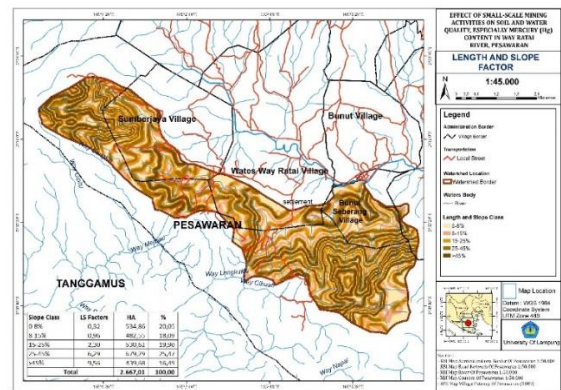


Figure 4. Topographic condition of research location

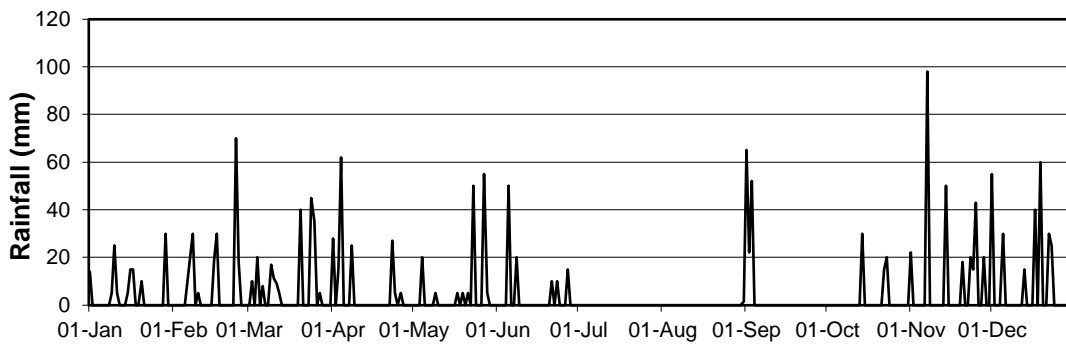


Figure 5. Monthly Rainfall condition in the year 2018.

The erosion index is 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 can be estimated. The magnitude of the erosion index describes the erosion hazard level. In general, the erosion index in the research location is in the very low to a low category, as much as 86%, while in the medium to a very high category is only 14%. The erosion index is presented in Figure 6.

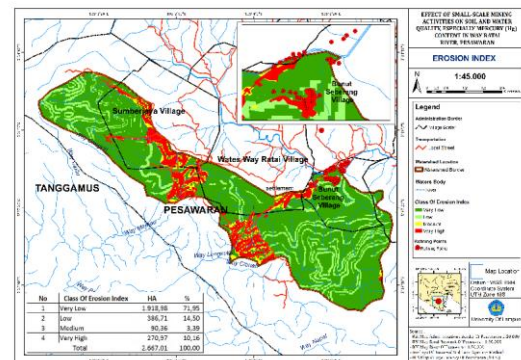


Figure 6. Erosion index of site location.

The surface run-off is determined mainly by the surface run-off coefficient, where surface run-off

results from the response of forest cover, topography, and soil type to falling rainfall. Soil erosion is affected by wind, rainfall, and associated run-off processes, the vulnerability of soil to erosion, and the characteristics of land cover and management (David, 1988; Aksoy and Kavvas, 2005; Panagos et al., 2015).

Table 2. The volume of run-off by the U.S. method Soil Conservation Services (SCS method).

Month	Rainfall (mm)	Direct Run Off (DRO) (mm)
-------	---------------	---------------------------

Jan	124	143.54
Feb	202	74.19
Mar	205	115.53
Apr	167	85.77
May	150	119.18
Jun	105	59.96
Jul	0	0
Aug	15	23.26
Sep	139	13.46
Oct	65	30.18
Nov	286	64.4
Dec	255	35.35
Total	1699.5	764.81

$$\text{Surface run coefficient (SRC)} = \frac{\text{Total DRO a year (764.81 mm)}}{\text{Total rainfall a year (1699.5 mm)}} \times 100\% = 45\%$$

The estimation of the surface run-off volume at the research location using the SCS (Soil Conservation Services-USDA) method is 45%, called the Surface Run-off Coefficient (SRO). The total rainfall that falls, 45% of rainfall would become surface run-off and enter the river flow. The results of calculating the surface run-off using the SCS method are presented in Table 2. The rate of water erosion occurring at a site depends on the rainfall itself (the source of rain splash detachment) and the run-off generated during the rainfall event, which both detaches and transports the eroded soil. Water added to the soil surface can either infiltrate the soil or flow along the soil surface as run-off (assuming a slight slope is present). The proportion of water that infiltrates the soil depends on the nature of the precipitation event (such as rainfall intensity, drop size, and snowmelt rates); the slope of the surface (generally, the higher the slope, the lower the percentage of water that infiltrates); and the infiltration rate of the soil (FAO, 2019).

According to the Minister of Forestry of Republic Indonesia (2014), the surface run-off coefficient of 45 percent is categorized as high. Excessive run-off volume can potentially cause flooding downstream. Indonesian climate is generally divided into wet and dry climates. From the research data, November till March is wet months, while July till October is dry months. Oldeman's climate classification is to make climate types based on the number of wet months and dry months. The wet month category is the month with rainfall >200mm and the dry month with rainfall <100 mm. According to Irianto (2003), 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 becomes surface run-off; this is exacerbated by the increasing conversion of forest functions to other uses such as agriculture, housing, industry, and rice fields, which can cause considerable flooding in the downstream area. Furthermore, it is said that the significant surface run-off will also cause excessive erosion, which will directly reduce soil fertility. Decreasing soil fertility will cause less vegetation to grow properly, so the land cover will decrease. Therefore, recharging water reserves in the upstream area will reduce and result in drought during the dry season.

Mercury (Hg) contents in the soils and river water

The location of water and soil sampling is presented in Figure 7, while the analysis results of the mercury contents in the soils and the river water are presented in Table 3. The results show the existence of pollution in soils and surface waters in the areas affected by Hg mining. Mercury contents in the soils and the river water were strongly influenced by erosion and surface run-off at the study site. Data 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. Based on Indonesian Government Regulation No. 82 the year 2001 (President of Republic Indonesia, 2001) concerning water quality and pollution control, the quality standard for mercury in soil and water is not exceeded 0.005 mg L⁻¹. While the mercury contents in the soils and water at all sampling points have exceeded the quality standard. Small-scale gold mining activities without a permission have brought the soil and water bodies (rivers) with the severely polluted category. 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 sample from purification points are between 30 m - 663 m, while the water samples from the purification point are between 30 m - 1161 m.

Besides, a high Hg content in the soil and river water should be considered because it will be dangerous for health. Hg can be transformed by bacteria into methylmercury and then bioaccumulates in fish, shellfish, and other high-level predators, thus posing a human health risk (Turner and Southworth 1999). Mercury may have toxic effects on the nervous, digestive, immune systems, lungs, kidneys, skin, and eyes.

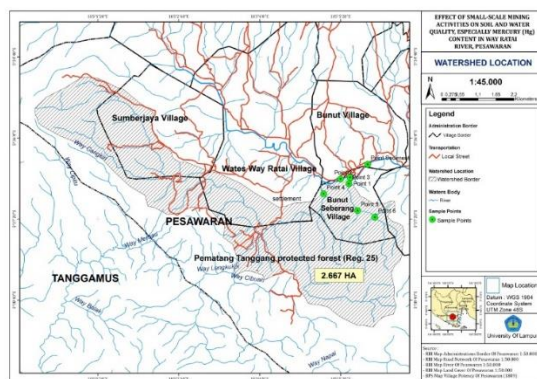


Figure 7. The soil and water samples point location

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

Location Number	Hg Content (mg L ⁻¹)		Analysis Method
	Soil	Water	
1	3.84	9.6	US EPA SW-846-7470A
2	5.75	14.1	US EPA SW-846-7470A
3	21.8	0.08	US EPA SW-846-7470A
4	28.9	0.88	US EPA SW-846-7470A
5	0.26	0.08	US EPA SW-846-7470A
6	0.91	0.14	US EPA SW-846-7470A

Table 4. Correlation between distance to the nearest purification and Hg content in the soil and water.

Distance to the nearest purification	r value*	
	Hg content in the soil	Hg content in water
Soil sample	-0.488ns	-0.572ns
Water sample	-0.545ns	-0.545ns

*ns: no significance

There is no correlation between the distance of soil samples from the nearest purification and Hg contents in the soils and river water. Similarly, there is no correlation between the distance of water samples from the nearest purification and Hg contents in the soil and river water (Table 4). The correlation between the distances of purification points and the concentrations of Hg is not significant. It is likely because the purifications from ore to gold are not carried out continuously over time, depending on the availability of ore material from the mining area. However, there is a tendency that the shorter the distance of soils and water samples from the nearest purification, the higher the Hg content in soils and water samples. Human-induced water erosion like Hg mining-dependent ASGM could lead to higher sediment inputs into stream channels and increased sedimentation into reservoirs along the stream channels.

Conclusion

Mercury (Hg) pollution in the soil and river water of the Ratai watershed by ASGM activities in Pesawaran District, Lampung, Indonesia, was high. The Hg contents in the soil and river water samples have exceeded the quality standard for Indonesia's Hg pollution criteria.

The surface run-off coefficient is 45 percent, including the high category, which means the study area is characterized by high rainfall erosivity. The erosion that occurs at the study site is also high. The erosion index in the research area ranged from very low to high. Therefore, potential erosion ranged from weak to very strong. Although, more than 80 percent of the research areas were covered by forest, which is in the outstanding category. However, the slope steepness was more than 40 percent, categorized as steeply, resulting in high surface flow and caused high erosion.

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