ISSN 2411-3336; e-ISSN 2541-9404

JOURNAL OF MINING INSTITUTE

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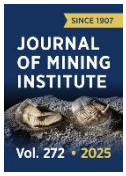
Journal homepage: pmi.spmi.ru



Research article *Geotechnical Engineering and Engineering Geology*

Mercury contamination of sediments at Indonesia Ciujung watershed: contribution of artisanal small-scale gold mining

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***How to cite this article*:** Willy C. Nugraha, Huiho Jeong, Phan Dinh Quang, Randy Novirsa, Tomohiro Komorita, Yasuhiro Ishibashi, Jun Kobayashi, Koji Arizono, Asep Bayu Dani Nandiyanto. Mercury contamination of sediments at Indonesia Ciujung watershed: contribution of artisanal small-scale gold mining. Journal of Mining Institute. 2025. Vol. 272. N 16339, p. 59-71.

Abstract

Artisanal small-scale gold mining is the largest artificial source of mercury pollution and has been a very common problem recently. This study examined the contribution of Hg release due to artisanal small-scale gold mining in contaminating the sediment of the watershed in Indonesia. Different from other methods, we measured Hg pollution using the stable isotopes of carbon and nitrogen. This report is the first time analytical reports in Indonesia since most papers focus primarily on only detecting heavy metals in river water and sediments, neglecting deep research on Hg pollution and resources. The findings of this study indicate that artisanal and small-scale gold mining activities, located in the upstream areas of the tributaries that act as point sources, are the primary source of mercury in the sediment of the Ciujung watershed. The isotopes $\delta^{13}C$ and $\delta^{15}N$ successfully trace the Hg-contaminated sediment in the Ciujung watershed derived from the Cisimeut and Ciberang Rivers based on soil organic matter. The biological effect showed the Hg concentration in most of the sampling sites exceeded the effects-range medium and probable effect limit values. The risk quotient values of Hg indicated the Hg pollution had a possibility effect on the benthic organism. Several limitations have also been added in this study and needs for further investigation.

Keywords

artisanal small-scale gold mining; mercury; sediment; Ciujung

Funding

This study was funded by an international postgraduate scholarship for research on mercury from the Kumamoto Prefectural Government, Japan.

Received: 02.11.2023 ***Accepted: 24.09.2024*** ***Online: 11.02.2025*** ***Published: 25.04.2025***

Introduction

Metal contamination in the Ciujung watershed has recently received increasing attention due to urbanization and industrialization [1, 2]. The Ciujung watershed, the longest river in Banten Province, Indonesia, is used for sanitation and irrigation activities. It has a catchment area of 1,858 km^2 and flows from the south to the north with a total length of 179 km (based on Environmental and forestry agency of Banten Province in the Final Report of Environmental Protection and Management Plan. Banten, Indonesia, 2019).

In recent decades, the rapid development of industrialization has caused the Ciujung watershed to receive different pollutant sources discharged through two tributaries: Cisimeut and Ciberang River [3]. A published report by the Indonesian Center for Environmental Law in 2013 (Final Report: Strengthening the Right to Information for People and Environment. 2013, p. 1-57) estimated that the overall input

of industrial wastewater discharged into the Ciujung watershed reached 43,044.35 m^3/day from a significant number of industries disposing of, for instance, pulp and paper, fertilizer, coal combustion, electroplating, chemical industry as well as domestic waste. The untreated wastewater discharge flown directly into the river causes the degradation quality of the Ciujung watershed leading heavy metal concentrations in surface water and sediment to exceed the acceptable limit as prescribed in environmental standards or regulations. Therefore, the demurrage and instability among residents to local governments keep rising every year.

Most studies done in the Ciujung watershed have dwelt on heavy metals contamination, however, there is still no available information regarding Hg contamination in the Ciujung watershed sediment. One of the biggest artisanal small-scale gold mining (ASGM) hotspots in Indonesia is found in Cisitu village, Lebak District, Banten Province and these activities are unregistered and have informal operations located upstream of the Ciujung tributaries [4]. Blacksmith Institute in 2013 reported* that 1,000 t of mercury was still used in ASGM where 95 % of all Hg used illegally in the amalgamation process is released into the environment, and it was estimated to affect 300,000-500,000 people. This attracted many researchers to investigate the effect of ASGM activities in this area. Several studies have been conducted in Cisitu village, reporting that Hg concentration from the collected soil, sediment, human hair, blood also fish nearby ASGM activities exceeded the acceptable limit value [3, 5-7]. Worst of all, some studies reported that the fish collected in Cisitu pond were found to be high in Hg concentration. Other reports [8] reported that 70-90 % of the organic Hg that exists in fish and shellfish is methylmercury (CH_3Hg^+) which is the most toxic form of Hg [9]. It was revealed that ASGM activities were the point source of Hg contamination in these matrices.

Hg contamination from ASGM is likely to be highly heterogeneous [10]. Mercury can be deposited in specific hotspots where mining activities are concentrated, and its distribution can vary significantly over short distances due to differences in mining intensity, local hydrology, and sediment transport [11]. Understanding how Hg is transported within the river systems and the mainstream (e.g., through sediments, water column, biota) necessitates a denser network of sampling points to map out these pathways accurately. We can infer that Hg released into the river flows through settling, dissolving in the water, and getting deposited in sediments can pose long-term environmental and health risks in downstream areas. Therefore, monitoring of the Ciujung watershed is essential to be conducted.

Sediments can vary widely in their geochemical properties, such as organic content, particle size distribution, pH, and mineralogy, all of which affect Hg mobility and bioavailability [12]. In this study, sedimentary organic matter and isotope ($\delta^{13}C$ and $\delta^{15}N$) are used as an environmental pollutant tracer to identify the Hg source in downstream sediment. The Hg has a strong correlation with organic matter (OM) which affects the partitioning of mercury to suspended solids in the water column and the sequestration of mercury to sediments [12]. Therefore, the mercury source can be represented based on sedimentary organic matter. Stable isotopes ($\delta^{13}C$ and $\delta^{15}N$) in OM in lake sediments are frequently used to trace organic matter sources due to the isotopic composition of sedimentary organic matter being relatively unaffected by post-photosynthetic, or diagenetic processes [13]. By using the isotope mixing model, we can determine the source of Hg in a certain mixture.

We conducted a field survey in the wet season to trace and quantify the sedimentary OM composition and total mercury (THg) contamination due to ASGM in the Ciujung watershed in Indonesia. The investigation of this issue was conducted in the wet season because, during the wet season, heavy rainfall causes an increase in the water level of rivers (flood), soil run-off, and also flash out sediment, which can lead to increased sediment transport from upstream to downstream area of the Ciujung watershed. Moreover, biological effects and ecological risk assessment of Hg to the benthic organism in the sediment of the Ciujung watershed were evaluated based on sediment quality guideline values

* Mitigating Mercury Emission from Artisanal and Small Scale Gold Mining in Indonesia. Program Report. 2013, p. 39. URL: https://wedocs.unep.org/bitstream/handle/20.500.11822/31242/MercE.pdf?sequence=1 (accessed 02.11.2023).

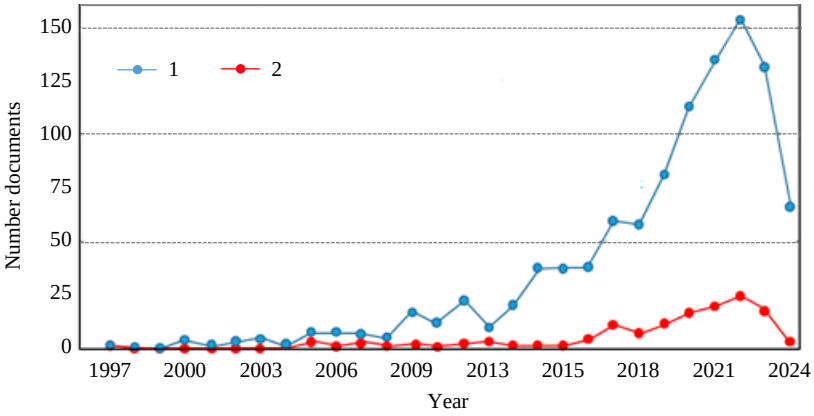

Fig.1. Bibliometric analysis for research trend using keyword "artisanal small-scale gold mining" (1) and "artisanal small-scale gold mining Indonesia" (2) [14]. Data was taken on 25 June 2024

(SQGV). This study was the first to examine the sediment associated with Hg pollution in the Ciujung watershed. This is the first time analytical reports in Indonesia since most papers focus primarily on only detecting heavy metals in river water and sediments, neglecting deep research on Hg. It is also confirmed by the bibliometric analysis based on Scopus database that only 136 documents until now for articles relating to ASGM in Indonesia (Fig.1), which is less number compared to general ASGM publication (reaching 1,024 documents). This makes the scientific publication relating ASGM in Indonesia needed to be reported.

This insists our novelties in this paper, including:

• evaluating Hg in two rivers in West Java Indonesia, namely Cisimeut and Ciberang Rivers and their tributaries, which are one of the important rivers in Indonesia;

• understanding the detection of Hg in sediment level that is possible from ASGM activities;

• evaluating sedimentary organic matter in the level of sediment, which were examined based on OM in soil organic matter (SOM);

• using stable isotopes of $\delta^{13}C$ and $\delta^{15}N$ to trace the source of Hg;

• evaluating Hg-contaminated sediment by the risk quotient;

• analyzing and understanding potential effects of Hg in rivers on benthic organisms (biological effects).

Methods

Study Area. The research was conducted in the Ciujung watershed, Banten Province, Indonesia. The sampling area was distributed in latitude 6°1'28.423"E to 6°34' 3.031"E and longitude 106°10' 9.354"S to 106° 9'53.67"S. We also showed the magnification from the map in Fig.2, *a*, which is

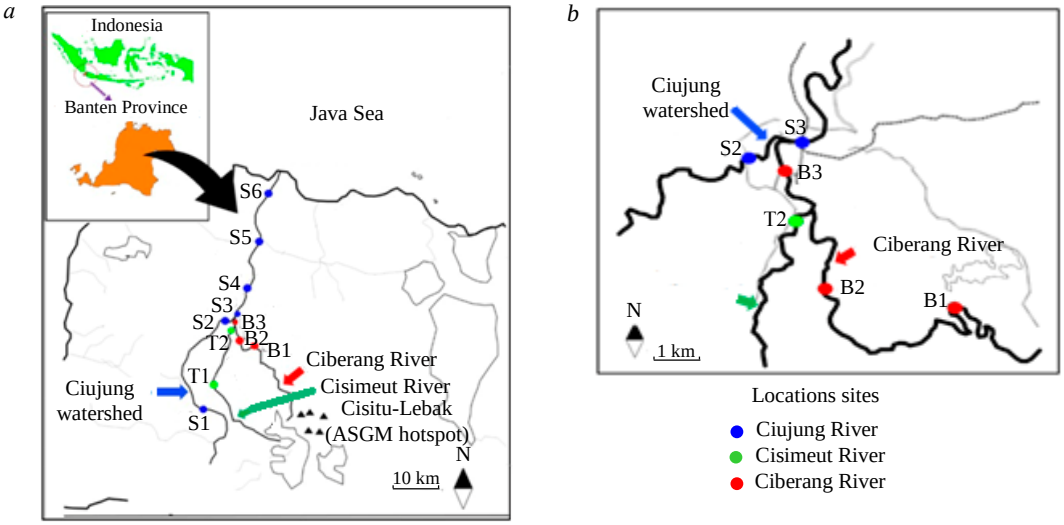

Fig.2. The location of surface sediment sampling (*a*) and detailed sediment sites (*b*) according to [15]

presented in Fig.2, *b*. In 2019, the Agency of Environment and Forestry, Banten Province, reported that the Ciujung watershed has two large tributaries: the Cisimeut River (458 km^2) and the Ciberang River (305 km^2). The water from these two rivers is used, specifically by the population for sanitation, daily activities, and agriculture. Thus, the research direction can have a wider impact on the importance of this study.

In the south of Rangkasbitung City, which is the capital city of Banten Province, these tributaries unite and flow to the North into the Java Sea. Interestingly, the upstream tributaries also come from the Karang and Halimun mountains [3] where the inhabitants of the upstream areas of the Ciujung watershed are mainly employed as farmers and gold miners, particularly in the Lebak District [3, 16]. Residents used the amalgamation processes in the forest, water river, and also their yard.

Sampling. In January 2021 (wet season) the sediment samples (0-10 cm) were collected from 11 sampling stations in the Ciujung watershed including two large tributaries: the Cisimeut and the Ciberang River (Fig.2, *a*). Six sites were distributed in the mainstream from the upstream (S1), middle stream (S2 and S3), and downstream (S4, S5, and S6). Two locations along the Cisimeut River: T1 and T2 and from three locations in the Ciberang River: B1, B2, and B3 (Fig.2, *b*). The sediment was collected one day after the big flood occurred in the Ciujung watershed which might lead to variations in the Hg content in the sediment [17].

The sediments were collected using a grab sampler and three times, were homogenized, and then stored in a cool box. They were then transported immediately to the laboratory before the analyses. Subsequently, the material was sieved through a 150 μm-sized sieve [18] and dried at 20 °C at room temperature. The dried samples were measured using a direct mercury analyzer.

Surface water was collected in each site using a point sampler horizontal based on Indonesian National Standard (SNI 6995:2021) method for water and wastewater, then surface waters were filtered with 0.45 μm of pore size and collected in polypropylene bottle, then acidified by using nitric acid until pH < 2. All bottles were transported to the laboratory in an ice-cooled container and stored in the refrigerator (4 °C) until analysis. THg concentration in the surface water sample was measured using Cold Vapour – Direct Mercury Analyzer.

Chemical and Sample Digestion. A high-purity Hg standard solution (1000 mg/l) was used for the calibration (Merck, Germany and Wako, Japan). The mercury concentration in the sediment was determined through the EPA method 7473, using thermal combustion with amalgamation atomic absorption spectrometry (Direct Mercury Analyzer 3,000 NIC, Japan). For determining the characteristics of the sediment, the total organic carbon (TOC) and total nitrogen (TN) were analyzed using an elemental analyzer (Thermo Fisher Scientific, ConFloIV).

The particulate organic phosphorous (POP) was determined by subtracting the sum of the inorganic phosphorus fractions from the total amount of phosphorus in the sediment after performing the ignition method [19]. Bulk surface sediment samples were collected for isotopic analysis (δ^{13}C and δ^{15}N). The sediment sample was acidified for 24 h in a bath containing 1N HCl solution to remove carbonates from the sediment matrix. The samples were washed with ultrapure water and oven-dried overnight at < 80 °C. The dry sediment sample was then ground to a fine powder using a mortar and pestle.

Isotopic analysis was performed using an elemental analyzer coupled online with an isotope ratio mass spectrometer using air nitrogen and Pee Dee Belemnite standards for N and C, respectively (Thermo Fisher Scientific: Flash 1112 Series, ConFloIV, DELTA Plus). The δ^{13}C and δ^{15}N values were compared with the Vienna Pee Dee Belemnite and air nitrogen standards, respectively [20]. The C:N, N:P, and C:P ratios were calculated by their respective molecular weights (‰),

$$\delta^{15}\mathrm{N} = \left\{\left(\frac{R_{\text{sample}}}{R_{\text{standard}}}\right) - 1\right\} \cdot 1000;$$

$$\delta^{13}\mathrm{C} = \left\{\left(\frac{R_{\text{sample}}}{R_{\text{standard}}}\right) - 1\right\} \cdot 1000,$$

where R corresponded to the ^{13}C/^{12}C or ^{15}N/^{14}N ratios.

Quality control and quality assurance. Certified reference material (CRM) marine sediment NMIJ 7302-a (Japan) was used for internal quality control to assess the validity of the data, and the number of replicates of CRM and samples were examined in duplo and triplicates. According to the analytical parameters, the accuracy of the method was 101.28 %, repeatability was 3.68 % of RSD, and the limit of detection (3SD of blank) was 20 ng/kg. This shows that the method was efficacious in determining the Hg content in the sediment.

Mixing model of isotope. All analyses were performed using freeware R version 4.0.2. The proportional contribution of each end-member (i.e., SOM of each tributary) to the surface sediment of each station was estimated using Bayesian mixing models according to the Markov chain Monte Carlo method [21]. The analyses were performed using the R package "simmr", which is the upgraded version of the "siar" and a prior distribution for this analysis is the Dirichlet, a generalization of the Beta distribution [22]. The trophic enrichment factor was set to zero concerning the non-trophic relationship among them. Four Markov chains with lengths of 10,000,000 were run, and the first 100,000 samples were discarded during burn-in. The thinning was adjusted to 100 depending on the convergence of the chains. The obtained Gelman – Rubin diagnostic statistics were close to 1 for all the estimated parameters, which satisfied the rules of thumb for the convergence assessment (less than 1.1) [23].

Biological Effects. According to Australian and New Zealand Environment and Conservation Council (ANZECC) guidelines, there are two recommended SQGV points, namely effective range low/median (ERL/ERM) trigger values representing the threshold value to trigger an adverse effect and threshold/probable effect level (TEL/PEL) value representing the high probability effect to evaluate the biological effect.

To evaluate the risk level of Hg in sediment, we used index RQ which is the most feasible method for risk assessment of pollutants in the environment [24, 25]:

$$\mathrm{RQ} = \frac{\mathrm{MEC}}{\mathrm{SQC}},$$

where MEC is the measured concentration for a single chemical measured at a sample, μg/kg; SQC is the sediment quality criterion, ng/g.

The sediment quality criterion of mercury

$$\mathrm{SQC} = \mathrm{K_p} \cdot \mathrm{WQC}, \quad (1)$$

where K_p is the partition coefficient of Hg between solid phase and interstitial water, l/kg.

The partition coefficient of Hg was calculated using equation

$$\mathrm{K_p} = \mathrm{S_s(Hg)}/\mathrm{S_w(Hg)},$$

where S_s (Hg) – Hg in sediment sample of each site, μg/kg; S_w (Hg) – Hg in water sample of each site, μg/kg.

In this study, the dissolved Hg concentrations ranged from n.d. (not detection) to 46 ng/l with an average of 14.2 ng/l. The partitioning coefficients (log K_p) of Hg between particulate matter and water in the Ciujung watershed were obtained at 0-5.21 with an average of 4.28. The average K_p derived in this study was calculated at 19,054 l/kg, and WQC is the water quality criterion (ng/l). The WQC was derived from tissue-based criterion (TBC) and bioaccumulation factor (BAF) using equation

$$\mathrm{WQC} = \frac{\mathrm{TBC}}{\mathrm{BAF}}. \quad (2)$$

The TBC is the tissue-based criterion derived by the species-sensitive distribution method. In this study, the TBC (HC5) value was 0.062 mg/kg based on data reported by [25]. BAF value was 3,023.00 l/kg based on data reported by previous reports [4]. Then using equation (2). The WQC of

Hg in the Ciujung watershed is 20.5 ng/l. Based on the average value of K_p (19,054 l/kg), the SQC of Hg was calculated to be 390.80 ng/g using equation (1). To evaluate the ecological risk of mercury: if RQ > 1, the risk posed by Hg is high; if 0.1 < RQ < 1.0, the ecological risk is moderate, and if RQ ≤ 0.1, the ecological risk is low [25].

Results and discussion

Hg distribution in the sampling sites. Table 1 lists the THg concentration in the mainstream of the Ciujung watershed ranging from 0.02 to 0.91 mg/kg of dry weight with average values of 0.59±0.32 mg/kg. Detailed analysis in Table 1 is shown in Fig.3. In the middle stream areas (S2 and S3), the THg concentration was relatively high, particularly in S3, it can be caused by confluence between tributaries (the Cisimeut (T1 and T2), the Ciberang River (B1-B3)) and mainstream (S1-S6) of the Ciujung watershed where the concentrations of Hg transported by these tributaries were extremely high ranging between (0.42-0.83 mg/kg). It showed that these high levels of Hg-contaminated sediments might have been caused by illegal small-scale gold mining activities that operated in the upstream areas of these tributaries as shown in Fig.2, *a*. To prove this, we examined the soil paddy field located within 500 m near the ASGM hotspot in Cisitu, Lebak District which acts as an Hg source in upstream of these tributaries. Based on research reported by other reports [6], the water used for the irrigation system is derived from Hg-contaminated water from the amalgamation process. The Hg concentration in the soil paddy field (n = 3) was obtained at 45.86±2.37 mg/kg of dry weight. The Hg concentration in soil paddy field was above than permissible level by the World Health Organization (Hg in soil for agriculture is 0.05 mg/kg) [26].

Table 1

Sediment characteristics and THg (dry weight) concentration in mainstream and its tributaries of the Ciujung River

Location sites	TOC*, μmol/g	TN*, μmol/g	POP*, μmol/g	C/N	N/POP	C/POP	$\delta^{13}C^*$, ‰	$\delta^{15}N^*$, ‰	THg**, mg/kg
Mainstream									
S1	174±30	20±2	1.0±0.9	8.8	18.2	138	–25.1	1.4	0.02±0.01
S2	800±33	81±3	3±1	9.9	27.3	232	–26.0	2.9	0.62±0.04
S3	1,174±110	110±2	6±2	10.7	18.5	170	–25.9	3.1	0.65±0.05
S4	958±370	94±37	6.0±0.8	10.2	15.3	134	–25.8	2.5	0.91±0.04
S5	707±190	74±8	5.0±7	9.7	13.8	114	–25.7	2.6	0.86±0.06
S6	357±150	38±12	4.0±0,1	9.3	9.8	78.8	–25.5	2.7	0.46±0.04
Min	174	20.0	1.0	8.8	9.8	78.8	–26.0	1.4	0.02
Max	1,174	110.0	6.0	10.7	27.3	232.0	–25.1	3.1	0.91
Mean	695.0	69.5	4.2	9.8	17.1	144.5	–25.7	2.6	0.59
SD	372.9	34.2	1.9	0.7	5.9	52.4	0.3	0.6	0.32
Cisimeut River									
T1	192±10	16±1	3±0.1	11.6	5.9	58	–26.2	2.0	0.62±0.04
T2	405±30	45±5	3±0.2	9.1	14.6	113	–25.3	3.2	0.52±0.04
Min	192	16	3	9.1	5.9	58	–26.2	2.0	0.52
Max	405	45	3	11.6	14.6	113	–25.3	3.2	0.62
Mean	298.5	30.5	3.0	10.4	10.2	85.5	–25.7	2.6	0.57
Ciberang River									
B1	320±30	33±4	4±0.1	9.8	7.8	65.4	–25.7	3.5	0.77±0.04
B2	397±63	44±3	4±0.8	9.1	12.0	94.3	–25.2	3.1	0.83±0.09
B3	281±21	28±1	3±0.8	10.0	10.4	89.3	–26.1	2.4	0.42±0.05
Min	281	28	3.0	9.1	7.8	65.4	–26.1	2.4	0.42
Max	397	44	4.0	10.0	12.0	94.3	–25.2	3.5	0.83
Mean	332.7	35.0	3.7	9.6	10.1	83.0	–25.7	2.0	0.67
SD	59.0	8.2	0.6	0.5	2.1	15.5	0.5	0.5	0.22

* Number of samples (n = 3); ** (n = 2).

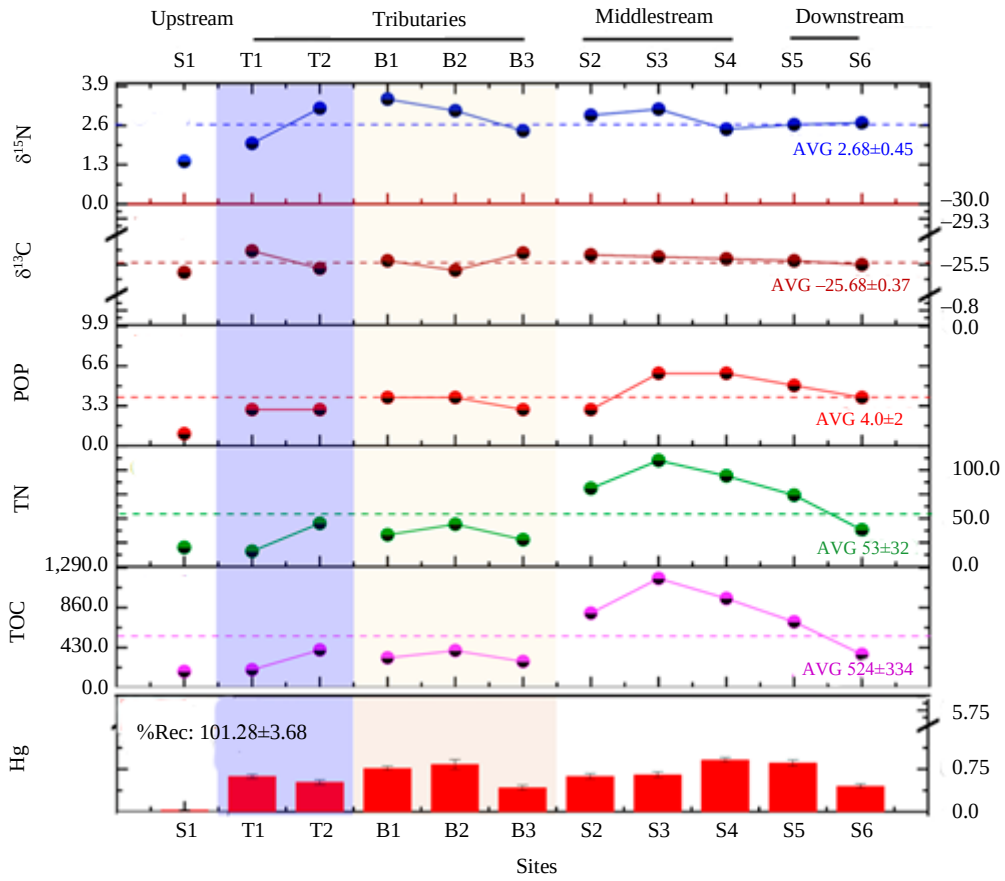

Fig.3. The distribution pattern of the $\delta^{13}C$ and $\delta^{15}N$ values (‰), organic matter, and THg concentration (μmol·g^{-1}) from each stream of rivers

When the Hg is discharged into the soil paddy field, it can be strongly bonded with the SOM. The soil will be transported into the river and accumulated in the sediment due to soil run-off during the wet season. Moreover, the highest Hg concentration occurred at S4 (0.91 mg/kg) followed by S5 (0.84 mg/kg) which is located downstream of the Ciujung watershed. The high Hg concentration at S4 and S5 might be due to these sites being the deepest sites in the Ciujung watershed with total depths of 6.72 and 4.22 m, respectively causing deposition of more suspended particles at these sites when the low flow rate occurs.

The accumulation of Hg in mainstream and tributaries was a serious problem where the THg concentrations in the mainstream and tributaries sediment of the Ciujung watershed were above the permissible limit set (0.15 mg/kg) by the Australian and New Zealand Environment and Conservation Council Interim Sediment Quality Guidelines (ANZECC ISQG) and Hongkong Interim Sediment Quality Guidelines (Hongkong ISQG) except in the S1 (upstream) [27]. Values of sediment quality indicators: the threshold values to trigger and adverse effect (ERL – 0.15; ERM – 0.7 mg/kg), evaluation the biological effect (TEL – 0.13; PEL – 0.7 mg/kg). The information of sediment quality from Indonesia government is still not available. Therefore, the sediment in the mainstream and tributaries had a high potential ecological risk due to Hg contamination, particularly for aquatic organisms.

Worldwide comparison of mean Hg. Among the Indonesia River, data from the Ciujung watershed are very limited. Results of other rivers from previous published are presented for comparison. Table 2 presents a comparison of the THg concentrations obtained in the study with the results of studies from other countries [28-31]. The THg concentrations in the Ciujung watershed were higher than those in the unpolluted areas, such as the Yangsuri lacustrine wetland, Korea; they were roughly comparable to the concentrations in Jiaozhou Bay, China, but were lower than those in other polluted areas, including Minamata Bay, Japan, and the Hunza River, Pakistan. The THg in the sediment of the Ciujung watershed and its tributaries indicated that they were moderately polluted by Hg.

Table 2

Comparison of THg in surface sediment in different basins/countries

Basin	Source of Hg	THg in surface sediment, mg/kg	Reference
Yangsuri Lacustrine wetland, Korea	Wetland	0.001-0.086	[28, 30]
Minamata Bay, Japan	Industrial activities	0.1-3.34	[29, 31]
Jiazhou Bay, China	Industrial activities	0.12-0.58	[30, 32]
Hunza River, Pakistan	ASGM	0.22-6.40	[31, 33]
Ciujung watershed, Indonesia	ASGM	0.02-0.91	This study

Characteristics of OM (TOC, TN, and POP) in sediment. TOC, TN, and POP concentrations are essential parameters for describing and determining the abundance of OM in sediments [32]. The distribution of TOC, TN, and POP in the Ciujung watershed sediments (S1-S6) was found to be similar. They were found to increase from upstream to the middle stream areas but decrease in the downstream areas (see Fig.3). The TOC, TN and POP concentrations in the mainstream were found in S3 (TOC – 1,174; TN – 110; POP – 6 μmol/g) followed by S4 (TOC – 958; TN – 94; POP – 6 μmol/g), and S5 (TOC – 707; TN – 74; POP – 5 μmol/g) and the mean concentration of TOC, TN and POP was 695, 69.5 and 4.17 μmol/g, respectively.

The TOC, TN, and POP were found to be highly concentrated in S3, it might be caused by high terrestrial input of OM, and, it might also impact the composition of OM in the sediment during heavy rain [33]. Meanwhile, the mean concentration of TOC, TN, and POP generated was 298.5, 30.50, and 3.00 μmol/g, respectively, in the Cisimeut River and 332.67, 35.00, and 3.67 μmol/g, respectively, in the Ciberang River. The varying concentrations of TOC, TN, and POP in these tributaries indicate the variation in the origin of these compound accumulations in the rivers. Other reports [34] reported similar research to our study that the concentrations of TOC, TN, and POP in the sediment of the Mianjiang River sub-watershed were significantly different.

Correlation between Hg, TOC, TN, and POP and their molar ratio. Pearson's correlation analysis was performed on the concentration data about the following: Hg, carbon, nitrogen, and phosphorus as well as for their molar ratios and isotopes, and the results have been presented in Table 3. The amount of TOC concentration was positively correlated with TN, POP, C/N, N/POP, C/POP, $\delta^{15}N$, and Hg content. A strong correlation existed between the amount of TOC and TN in the sediments of the Ciujung watershed and its tributaries ($r = 1.00$; $P < 0.01$), which suggests that a major part of TN was associated with TOC that can be considered as organic nitrogen [35]. Moreover, a strong correlation existed between the TOC and POP ($r = 0.78$; $P < 0.05$).

Table 3

Pearson's correlation matrix of Hg, $\delta^{15}N$, $\delta^{13}C$, TOC, TN, and POP and their molar ratio from the Ciujung watershed and its tributaries

Items	TOC	TN	POP	C/N	N/POP	C/POP	$\delta^{13}C$	$\delta^{15}N$
TOC								
TN	1.0**							
POP	0.78**	0.77**						
C/N	0.27	0.20	0.35					
N/POP	0.57	0.60	–0.04	–0.21				
C/POP	0.66*	0.68*	0.07	–0.06	0.99**			
$\delta^{13}C$	–0.28	–0.22	–0.29	–0.87	0.06	–0.08		
$\delta^{15}N$	0.37	0.40	0.51	–0.05	0.03	0.06	–0.03	
Hg	0.51	0.52	0.79**	0.33	–0.14	–0.06	–0.30	0.61

*$p < 0.05$; **$p < 0.01$.

The results suggested that the type of OM should be similar at all sites. Therefore, we can assume that the Hg accumulation in the sediment can be represented by OM. The findings of this study are consistent with the findings reported in the previous research conducted by other reports [36] that found that the Hg concentrations correlated significantly with the TOC contents in Bohay Bay, Northern China, and Tianjin's Haihe River caused by the thermally labile fraction of OM released during pyrolysis at 300 °C. It revealed that the strong correlations between the TOC, TN, POP, and Hg concentrations indicate the strong correlation between OM and Hg adsorption onto the sediment. The meaning of this result is Hg released from amalgamation processes is strongly bonded in the organic matter of sediment.

Identification of the type of OM. OM in the sediments in the mainstream and tributaries was analyzed to investigate the source of Hg in the mainstream of the Ciujung watershed (see Table 1). The molar ratio and stable isotope ratio of both organic C and N were used to determine the origin and transformation of OM affected by the anthropogenic impacts on local and regional ecosystems [32]. An analysis of the C/N ratio together with $\delta^{13}C$ and $\delta^{15}N$ can provide information on the OM sources and identify the type of OM). The narrow range (from −26.2 to −25.1 ‰) of δ13C in the sediment collected from the Ciujung watershed and its tributaries was attributed to the prevalence of higher hydrodynamic energy conditions in this watershed [36].

This study was conducted during the rainy season; therefore, the flow rate of the river was high, and the spatial distribution of sedimentary OM was expected to be strongly affected by river flow. Therefore, we assumed that during the wet season, the SOM in upstream of these tributaries can be transported in the mainstream of the Ciujung watershed. To identify the type of OM, we evaluated it according to $\delta^{13}C$ and $\delta^{15}N$ (Fig.4, *a*).

The results failed to distinguish between the types of OM based on some values derived from phytoplankton and OM in the soil. However, as predicted by the C/N ratio and $\delta^{13}C$ values, the mean $\delta^{13}C$ values indicated that the OM derived from the soil was abundant, which agrees with the soil organic properties of OM (Fig.4, *b*) [37]. This revealed that OM in the sediment of the Ciujung watershed and its tributaries was not generated by the phytoplankton and this result is consistent with that reported by other reports [38].

Mixing model of stable isotopes. A Bayesian mixing model was used to estimate the proportional contribution of each OM to the sediment at each site using the stable isotope and the three sources: S1 as uncontaminated upstream of the mainstream, T1 as a contaminated tributary (the Cisimeut River), and B1 + B2 as a contaminated tributary (the Ciberang River). In this case of B1 + B2 as points of sources, it can be caused because these areas have similar isotopic and geological characteristics. Therefore, we assume that these sites are treated as one group.

In this study, the proportion of the contribution of n + 1 different sources could be determined by different isotopic tracers. Based on the conservation of mass of the stable isotope. Based on the biplot of $\delta^{15}N$ and $\delta^{13}C$ (Fig.5), OM, which accumulates sediment downstream of the Ciujung watershed (mixtures S4, S5, and S6), had similar characteristics to the OM obtained from the mixtures T2 and B3; these sites were relatively close to the source (T1 and B1 + B2). In the case of S3 and B2 which are out of the triangle, we used the "siar" package based on a Monte Carlo simulation of mixing polygons to apply the point-in-polygon assumption to these models [22].

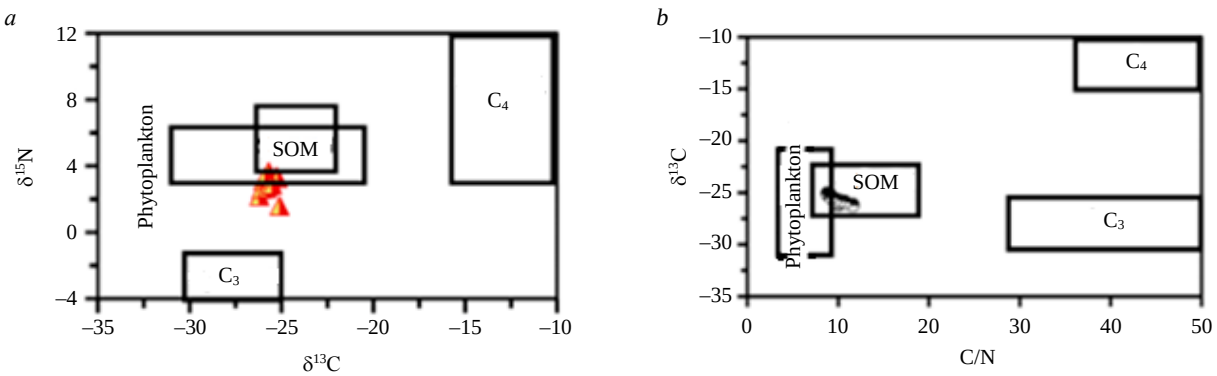

Fig.4. Plot of $\delta^{13}C$ vs $\delta^{15}N$ (*a*) and plot of C/N radio vs $\delta^{13}C$ (*b*)

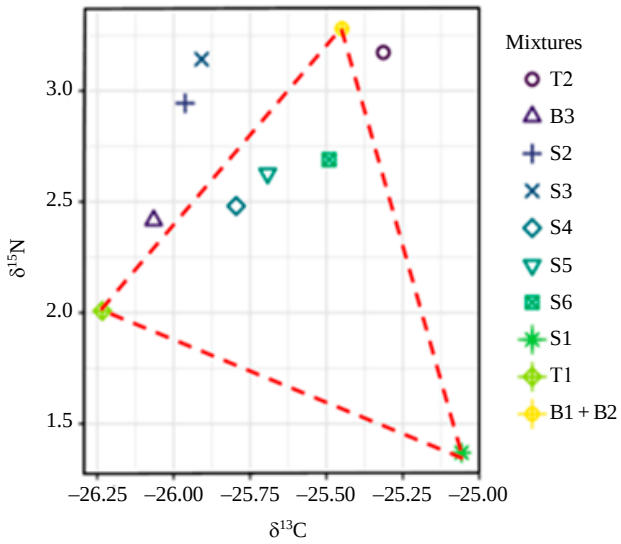

Fig.5. Mixing model of isotope using Bayesian mixing model

The mixing polygons (i.e., mixing model) were iterated using the distributions of the proposed OM sources, and the proportion of sources that have a solution was calculated. This proportion could be interpreted as the frequentist probability that the proposed mixing model could calculate source contributions to explain isotopic signatures. Since this analysis does not calculate a linear solution, we could obtain the contribution of the outside of the polygon.

The contribution of soil OM represented by (mean±SD) from the Cisimeut River (T1) was found in most of the downstream sediment in S3-S6, in which the highest was S4 (60.79±0.05 %), followed by S5 (49.32±0.07 %), S3 (39.00±0.0 %), and S6 (23.83±0.09 %). The OM from the Ciberang River contributed to the downstream sediment in S3-S6, with the highest contribution in S6 (47.51±0.01 %), S5 (36.29±0.01 %), and S4 (26.00±0.01 %). It was revealed that the soil OM from the Ciujung and Ciberang Rivers, which contained Hg, contributed to the accumulation of Hg in the sediment in the downstream area of the Ciujung watershed caused by a strong formation and complexation ionic bonds between mercury and SOM affected the sequestration of mercury to sediment [39]. Therefore, it can be assumed that the sediment organic matter in the downstream is predominantly derived from these tributaries.

Biological effect. Hg-contaminated sediment affects benthic organisms. Therefore, assessment of ecotoxicology of the Hg concentration in sediment was used effect range low (ERL)/effect range median (ERM) and threshold effect level (TEL)/threshold probable effect level (PEL). Due to the ERL and TEL values being less than expected to have adverse effects on sediment-dwelling organisms, we evaluated the biological effect using the ERM and PEL values indicating the chemical concentration above which adverse effects are likely to occur in the aquatic ecosystem [40]. These values provide a reliable tool to evaluate the quality of sediment and adverse biological effects on estuarine sediment [41].

As shown in Table 1, except for in S1, Hg concentrations in all sampling sites exceeded the TEL and ERL values. Based on PEL and ERM values, several sampling sites showed that extremely high concentrations of Hg exceeding the PEL and ERM values were found in downstream S4 and S5, and also in tributaries B1 and B2. Therefore, these sites can be categorized as potential toxicity to the benthic organisms. The impact of contaminated sediment on aquatic ecosystems is likely to be due to remobilization and re-suspension processes [40].

The data of Hg toxicity including abnormal and deformed aquatic organism-based tissue according to NOEC (no observed effects concentration) and LOEC (lowest observed effect concentration) values was reported by previous reports [25] for several benthic organisms, for instance, mollusks, crustaceans, aquatic insects, and fish. The deformed of growth in aquatic organism was found in mollusks (*Pyganodon grandis*, Hg = 0.686 mg/kg) and fish (*Perca flavescens*, Hg = 0.125 mg/kg; *Sandae vitreus*, Hg = 0.25 mg/kg); the abnormality of the cellular system was found in fish (*Liza aurata*, Hg = 0.1 mg/kg; *Bileophtalmus dussumieri*, Hg = 0.59 mg/kg); the biochemistry disorder was found in fish (*Ameiurus males*, Hg = 0.59 mg/kg; *Brycon amazonicus*, Hg = 0.63 mg/kg); crustaceans (*Dapnia magna*, Hg = 0.859 mg/kg); aquatic insects (*Hexagenia sp.*, Hg = 0.166 mg/kg), and the effect of mortality was found in molluscs (*Mytillus edulis*, Hg = 1.12 mg/kg); fish (*Poecilia reticulate*, Hg = 0.2 mg/kg; *Onchorhynchus keta*, Hg = 0.8 mg/kg). The previous study reported by other reports [3] showed the bioaccumulation factor of the selected fish namely *Barbonymus gonionotus* and *Channa striata* in the Ciujung watershed is found to be higher than 1,000 which includes dangerous categories if consumed and also consistent with the research reported by other reports [7] that the Hg concentrations found in Cisitu ponds ranging from 0.1-1.3 mg/kg which exceeding the maximum quantity for human consumption.

In this study, the ecological risk of Hg toxicity based on RQ at the 11 sampling sites show that the RQ values of 10 sediment sampling sites in the Ciujung watershed including mainstream and tributaries are more than 1, while the RQ of S1 is less than 0.1. RQ values of Hg at the 11 sampling sites of Ciujung watershed: mainstream – S1 – 0.05; S2 – 1.59; S3 – 1.66; S4 – 2.33; S5 – 2.20; S6 – 1.18; tributaries (Cisimeut River) – T1 – 1.59; T2 – 1.33; tributaries (Ciberang River) – B1 – 1.97; B2 – 2.12; B3 – 1.07.

A similar result was reported by a previous report [25] in Taihu Lake where almost the range of RQ values in the 31 sites was between 0.10-1.00, and two sites showed higher than 1 were S12 (RQ = 2.04) and S13 (RQ = 2.18). Based on that, the ecological risk of Taihu Lake is in the middle level.

The results of this study showed that the RQ values in the Ciujung Watershed were quite higher compared to the Taihu Lake, indicating that overall, the ecological risk level of Hg in Ciujung sediment was high. Regarding this, we can infer that Hg contamination in the Ciujung watershed has the potential effect on benthic organisms as a food chain in most of the sampling sites except S1. Future research is highly required to examine different species of fish and benthic organisms from those sampling sites in the Ciujung watershed.

Limitations of this study as follows:

• Indonesia's rivers are managed by local government authorities. To collect samples from these rivers, individuals or organizations must obtain specific permissions from local government.

• Local governments are responsible for the monitoring and supervision of rivers and have designated specific sampling points, individuals or organizations must adhere to the procedures established by local authorities.

• The challenging access to the rivers, which are surrounded by dense forests. Therefore, we have to follow the procedure established by local government. This process could limit the temporal and spatial representation of the collected data.

• Reliable identification of spatial patterns and correlations with other geochemical characteristics typically requires a larger dataset (30 samples at least) to ensure statistical significance and to reduce the impact of outliers.

• A small number of samples (11 samples) inadequately represents geochemical diversity and does not account for the influence of environmental factors such as vegetation cover, land use, and human activities, nor does it capture the temporal fluctuations in mercury levels due to seasonal changes such as varying water flow rates, sediment deposition, and erosion patterns; thus, it provides insufficient geographic coverage and risks missing significant variations in mercury contamination within the river system. Therefore, further investigation should be conducted to ensure sufficient geographic coverage for a comprehensive result.

• Besides ASGM, other potential sources of Hg contamination, such as industrial discharges, atmospheric deposition, and historical contamination, need to be considered. Understanding the relative contributions of these sources requires a more comprehensive sampling strategy.

Conclusion

The average concentration of total Hg on the surface sediment of the Ciujung watershed ranges from 0.02-0.91 mg/kg d.w., and based on the sediment quality guideline, the concentration of Hg on the surface sediment exceeded the normal limit value in all the sampling sites except S1. This study is important since it is the first to investigate Hg contamination in the mainstream of the Ciujung watershed, Banten Province as a result of illegal ASGM activities.

This study confirmed the presence of mercury contamination in the Ciujung watershed, where the average total concentration of Hg in the surface sediment of the Ciujung watershed ranges from 0.02-0.91 mg/kg d.w., and based on sediment quality guidelines, the concentration of Hg in surface sediment exceeds the normal limit value in all sampling locations except S1. The Bayesian mixing model using isotopes shows that the percentage of SOM distribution in the main stream of the Ciujung watershed consists of 23.8-60.8 % from the Cisimeut River and 26.0-47.51 % from the Ciberang River. The results utilizing C/N and $\delta^{13}C$ revealed that the OM sedimentary is composed of SOM

where THg concentration and C, N, and P was strongly correlated based on Pearson's correlation analysis. This revealed that these rivers have contributed to the Hg contamination of the mainstream of the Ciujung watershed due to illegal ASGM activities carried out in the upstream areas of the tributaries that act as point sources.

The biological effects evaluated by ERM and PEL showed that the concentration of Hg exceeded the normal limit value with a moderate index risk quotient level. It can be concluded that sediment in the Ciujung watershed confirms the high probability of its effect on residents with Hg. This study is significant because it addresses Hg pollution and provides suggestions for further environmental management of Hg and sediment remediation.

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The authors declare no conflict of interests.