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Distribution Analysis And Environmental Impact Of Heavy Metal Contamination In Rice Fields: A Case Of Sleman Regency, Yogyakarta, Indonesia

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Abstract

Examining the heavy metals' contamination in soil is important yet haven't much done in Sleman, Yogyakarta, Indonesia. The research aims to determine the distribution and pollution levels of Pb, Cu, As, Cr, Ni in Sleman's paddy fields. The composite soil samples were collected from 23 irrigated lowland fields of 15 subdistricts in Sleman using the grid sampling method. The analysis of heavy metal contamination including contamination factor (CF), pollution load index(CLI), ecological risk assessment

(Er),potential ecologica risk index (RI),enrichment factor(EF), and geoaccumulation index (Igeo). The results detected the soil contamination of Cu, Pb, As, Cr and Ni from all sampling points. There was high-level contamination factor of Cu (CF =7.69), medium-level contamination factor of Pb (CF = 2.23) and As (CF = 1.67), and low-level contamination factor of Cr and Ni (CF < 1). Cr and Ni were derived from natural processes, but Cu, Pb, and As contaminants were anthropogenically produced. Cu, Pb, As, Cr, and Ni metals have a low potential ecological risk (Er<40). The enrichment factor (EF) for heavy metals in the paddy fields of Sleman are low category (Cr, Ni), medium category (Cu, Pb), and high category for As. The excessive soil pollutant in the soil gives a serious caution on chemicals use. Agricultural practices need to be eco-friendly and sustainable. Future studies can assess to what extend the transfer of heavy metal from soil to plants or rice.

Keywords: contamination, ecological risk assessment, heavy metals, pollution load index, potential ecological risk index.

Introduction

Obtaining maximum yield, high productivity, and added value are the goals of agricultural research and development. As the population increased, so did the demand for food, with rice as the staple food. Land exploitation with intensive cultivation to overcome limited land due to land conversion to non-agriculture can damage the environment. Problems arising from intensive agricultural land use include excessive use of agrochemicals by farmers, thereby contaminating the surrounding environment and agricultural products (Dewi et al., 2023).

On the other hand, climate change may lead to a proliferation of plant-interfering organisms that destabilize crop production. Sutrisno and Kuntyastuti (2015) reported that efforts to realize the availability of sufficient and safe food for the community through intensification of crop cultivation could cause land damage (soil physics, chemistry, and biology) and impact heavy metals pollution from the active ingredients of chemical fertilizer. Heavy metals’ concentration in the water, soil, and plants might rise above permissible thresholds when chemical fertilizers and pesticides are applied excessively to maintain and improve crop yields.

Inorganic and organic fertilizers, pesticides, mining waste, industrial exhaust, and solid or liquid municipal or industrial waste that reaches paddy fields are some of the materials that can contaminate the soil and cause heavy metal accumulation (Wuana and Okieimen, 2011). The sources of heavy metals in soil are from natural and anthropogenic sources; natural sources originate from soil parent material. The heavy metal concentrations in the undeveloped soil depend on soil parent material, geochemistry, and soil processes responsible for soil formation (Hardy and Cornu, 2006). The use of pesticides, insecticides, and fertilizers containing heavy metals is a secondary source of heavy metal pollution in agriculture, according to anthropological studies. The primary sources of heavy metal pollution include metal castings, industries based on metal, and metal leaching from a variety of sources, including landfills, waste dumps, excretions, livestock and chicken manure, water runoff, cars, and road construction. (Briffa et al., 2020).

From a safety perspective, the use of agrochemicals can reduce the quality of agricultural products because the potential of these materials contains pesticide residues and heavy metals. Agricultural land contaminated with heavy metals can affect plant growth, inhibit the processes of photosynthesis, and decrease yields. Since these factors are connected to food security, heavy metal contamination, pollution, and land marginalization brought about by the loss of the original soil's organic matter and related nutrients have sparked a lot of interest in developing environmentally friendly and sustainable agriculture. (Yuniarti et al., 2022). Food products containing heavy metals consumed by humans over a long period will accumulate in the body tissues and eventually adversely affect human health, such as cancer (carcinogenic), birth deformities (teratogenic), nerve damage (neurotoxin), reproductive endocrine, and lowers the immune systems (Kiran and Sharma, 2021). A positive correlation was found between heavy metal content in blood and urine with cases of cardiovascular disease and death from cancer (Duan et al., 2020).

The organic matter content status map of rice fields in Sleman Regency (Mulyadi et al., 2017) shows that 98% contain very low to low organic matter. Moreover, the Sleman Regency is a high source of rice production, and the rice produced is supposed to be of good quality and acceptable to the industrial segment and the wider community. Ensuring food quality and safety by preventing food from being chemically contaminated by possible heavy metals or other contaminants so that the food consumed does not endanger human health by the government regulations of the Republic of Indonesia number 86, the year 2019 for food safety (Sukarjo et al., 2021).

Investigations into heavy metal contaminants in paddy fields need to be carried out, including assessing their concentrations in agricultural soil to determine the potential risk of paddy fields been contaminated with heavy metals (Hang et al., 2009). Potential ecological risk is the possibility of a risk to human health caused by environmental factors. Assessment of the potential ecological risk of heavy metals is beneficial for finding out information about the risk value of this contaminant in the environment so that it can be reduced or overcome. The aimed of this study were to determine heavy metals distribution and contamination level of Pb, Cu, As, Cr, and Ni in paddy fields in Sleman Regency based on the parameters of contamination factor (CF), contamination load index (CLI), ecological risk assessment (Er), potential ecological risk index (RI), enrichment factor (EF), and accumulation index (Igeo).

Materials and Methods

Location and sampling of paddy soil

The study was carried out in the rice fields of Sleman Regency and used desk work study method, field survey, and soil sample analysis was undertaken in the aboratory. The activity stage includes the pre-survey (quick assessment) and the primary survey. In the preliminary survey activities, we coordinated with relevant agencies, conducted interviews with farmers and on-site positioning of the surveying and mapping areas to prepare and collect information for the smooth implementation of the primary survey. Primary survey activities include taking soil samples and collecting main data on plant cultivation, especially rice fertilization techniques commonly used by farmers in paddy fields. Survey activities were held in paddy fields based on soil maps of 23 rice field center points in Sleman Regency. Soil samples were taken using a grid system with observation points distributed across agricultural land irrigated with paddy fields. The composite soil sample is a mixture of five individual sub-samples. Individual soil samples were taken from the tillage (root) layer (0–20 cm).

From the results of the field survey, 23 soil sampling points were selected spread across 15 sub-districts from 17 sub-districts in Sleman Regency, namely Ngaglik, Turi, Mlati, Seyegan, Minggir, Moyudan, Godean, Moyudan, Gamping, Berbah, Prambanan, Kalasan, Ngemplak, Cangkringan and Pakem. Only two districts, Depok and Tempel, did not collect soil samples (Table 1). The coordinate points were plotted on the administrative map and overlapped with the semi-detailed map of Sleman Regency (IAARD, 2016). The combined results represent a series of soil map units that make it possible to identify soil physical properties, soil units, topography, and parent materials.

Analysis of heavy metal content and distribution map of heavy metals

The soil samples from the field were air dried, crushed, and sieved through a 100-mesh sieve. The analysis of heavy metals concentration Cu, Pb, As, Cr, and Ni following the Omnian standard method (X-Ray Fluorescence Analysis = XRF). Data on the concentration of each heavy metal were analyzed using descriptive statistics, which included minimum, maximum, and average values, coefficient variation, skewness tests, and kurtosis tests, using Excel software. The distribution of each heavy metal content on map is categorized by score (low, medium, high). The mean and standard deviation are used as a reference to determine the heavy metal classification score on the map. The map was created by overlaying the administrative map with a semi-detailed map of the Sleman Regency (IAARD, 2016).

Table 1. Soil sampling locations representing soil map units in the rice fields of Sleman Regency.

No.	Location (village)	Sample code (composite)	Altitude (meters above sea level)	Coordinate point
1.	Donoharjo, Ngaglik	A	293.7	-7°41'59"; 110°23'15"
2.	Donokerto, Turi	B	356.8	-7°40'22"; 110°22'22"
3.	Sumberadi, Mlati	C	208.9	-7°43'32"; 110°20'3"
4.	Margokaton, Seyegan	D	195.3	-7°43'4"; 110°17'59"
5.	Sendangsari, Minggir	E	143.1	-7°43'7"; 110°14'57"
6.	Sumberagung, Moyudan	F	120.8	-7°45'50"; 110°15’23"
7.	Sidoluhur, Godean	G	117.0	-7°46'23"; 110°16'22"
8.	Sumbersari, Moyudan	H	108.5	-7°46'58"; 110°16'19"
9.	Sidomulyo Godean	I	116.1	-7°46'40"; 110°17'24"
10.	Ambarketawang, Gamping	J	124.1	-7°47'33"; 110°19'4"
11.	Banyuraden, Gamping	K	120.4	-7°47'37"; 110°19'48"

12.	Kalitirto, Berbah	L	129.0	-7°47'14"; 110°27'16"
13.	Jogotirto, Berbah	M	119.7	-7°48'44"; 110°27'56"
14.	Madurejo, Prambanan	N	135.4	-7°47'28"; 110°28'20"
15.	Sumberharjo, Prambanan	O	110.0	-7°48'11"; 110°29'33"
16.	Tamanmartani, Kalasan	P	178.3	-7°45'01"; 110°28'51"
17.	Selomartani, Kalasan	Q	233.2	-7°43'31"; 110°28'24"
18.	Bimomartani, Ngemplak	R	246.2	-7°42'09"; 110°28'00"
19.	Argomulyo, Cangkringan	S	439.9	-7°39'53"; 110°27'08"
20.	Pakembinangun, Pakem	T	456.3	-7°39'39"; 110°25'38"
21.	Argomulyo, Cangkringan 2	U	439.9	-7°39'53"; 110°27'08"
22.	Sendangsari, Minggir	V	156.2	-7°43'36"; 110°15'5"
23.	Sendangrejo, Minggir	W	157.0	7°43'37"; 110°15'32"

Soil contamination analysis

Contamination Factor (CF)

Given that it may give an overview of the degree of heavy metal contamination in soil, contamination factor (CF) is a useful tool for periodically evaluating soil contamination. The ratio of the soil metal concentration to the background metal concentration, or the metal concentration in the soil that is naturally present in the earth's crust, is the value of the contamination factor. Equation 1's formula was used to calculate the contamination factor's value (Hakanson, 1980).

$$CF = \frac{C\ (heavy\ metal)}{C\ (background)} \quad (1)$$

The concentration of metals in soil samples is denoted by C (heavy metal), whereas the concentration of metals naturally found in the earth's crust is denoted by C (background). Alfaro et al. (2015) state that background heavy metal concentrations in soil have been tested in numerous countries, with China being the only Asian nation to do so. Four categories are identified based on the CF value: I) low polluted soil (value < 1), ii) moderately contaminated soil (1 ≤ CF ≤ 3), iii) high contaminated soil (3 ≤ CF < 6), and highly contaminated soil (value > 6). (Gupta et al., 2021).

Pollution load index (PLI)

The amount of heavy metal in the area can be calculated using pollution load index (PLI). The method was initially used by Tomlinson et al. (1980). The PLI value < 1 presumes no soil metal contamination, while a PLI value > 1 indicates that soil is contaminated. The PLI value can be determined using the formula in equation 2.

$$PLI = (CF1 \times CF2 \times CF3 \times CF4 \times CF5)^{1/n} \quad (2)$$

CF1 = Contamination Factor of Cu
CF2 = Contamination Factor of Pb
CF3 = Contamination Factor of As
CF4 = Contamination Factor of Cr
CF5= Contamination Factor of Ni
n = contamination factors' number

Ecological risk assessment (Er) and potential ecological risk index (RI)

The ecological risk value (Er) was employed to evaluate the ecological risk associated with heavy metals in soil samples. The potential ecological risk index (RI) value is utilized to quantify the combined ecological risk posed by various heavy metals found in the soil samples. Er values were established to gauge the detrimental ecological consequences stemming from human activities aimed at safeguarding the environment. The risk index provides insight into the environmental and ecological impact of heavy metals (Saleh et al., 2018) and is computed using the equation outlined in Equation 3 (Hakanson, 1980)

$$Er = Tr \times CF \quad (3)$$

The value of the toxic response factor (Tr) is different for each heavy metal, for Cu = Pb = Ni = 5; Cr = 2; and As = 10 (Hakanson, 1980) and CF indicated the contamination factor. Er in soil samples was assessed using the following 4 categories: (a) low potential ecological risk at Er < 40, (b) moderate risk at 40< Er, < 80, (c) moderately high risk at 80 < Er < 320, and (d) Er > 320 the risk is very high.

RI values were obtained from the sum of all Er values of heavy metals observed in soil samples according to the formula in Equation 4 (Hakanson, 1980). The value of the toxic response factor (Tr) is different for each heavy metal, for Cu = Pb = Ni = 5; Cr = 2; and As = 10) and CF indicated the contamination factor.

$$RI = \sum Er \quad (4)$$

The index value of potential ecological risk can be grouped into 4 categories, namely "(a) RI < 150 indicates low risk, (b) 150 RI < 300 indicates moderate risk, (c) 300 RI < 600 indicates large risk, and (d) RI > 600 indicates a very significant risk" (Hakanson, 1980).

Enrichment Factor (EF)

The Enrichment Factor (EF) is employed to assess the extent of heavy metal contamination originating from human activities, allowing differentiation between anthropogenic and natural sources, as outlined by Swarnalatha et al. (2015). Iron (Fe) serves as a reference element for geochemical standardization, following the approach suggested by Syakti et al. (2015). The EF proves to be a valuable tool in gauging the degree of anthropogenic heavy metal pollution (Sakan et al., 2009). The EF is calculated using the following relation (Syakti et al., 2015).

$$EF = \frac{Mc_{sample} / Fe_{sample}}{Mc_{background} / Fe_{background}} \quad (5)$$

Mc sample is the concentration of a heavy metal taken from a sample of paddy soil, Fe sample is the concentration of Fe in a sample of paddy soil, Mc background is the natural concentration of a reference metal according to (Wedepohl, 1995), and Fe background is the natural concentration of Fe metal reference. Interpretation of EF values follow the suggestion of Sakan et al.,2009, where "EF <1 indicates no enrichment, 1–3 is minor enrichment, 3–5 is moderate enrichment, 5–10 is moderately severe enrichment, 10–25 is severe enrichment, 25–50 is very severe enrichment, and >50 is extremely severe enrichment."

Geoacculmulation index (Igeo)

The extent of heavy metal accumulation, particularly those resulting from human activities on the soil surface, is discerned through the accumulation index value. This value is determined using the formula presented in equation 6 (Syakti et al., 2015):

$$Igeo = log_2\left(\frac{M_c}{1.5 \times B_c}\right) \quad (6)$$

C (heavy metal), representing the measured metal concentration in the soil, stands in contrast to C (background), which signifies the naturally occurring concentration of the heavy metal in the Earth's crust before any human activities introduced heavy metals into soil pollution. The factor 1.5 is an adjustment factor accounting for variations in the background matrix resulting from lithospheric influences (Syakti et al., 2015. The geoaccumulation index value can be categorized into seven categories, namely "(i) uncontaminated = 0, (ii) not polluted to moderately polluted = 0-1, (iii) moderately contaminated = 1–2, (iv) moderately to heavily contaminated = 2– 3, (v) heavily contaminated = 3–4, (vi) heavily contaminated to extreme= 4-5, (vii) extremely contaminated > 5" (Mandal et al., 2022).

Results

Location overview

Geographically, the area of Sleman Regency lies at 110°15'13" to 110°33'00" East Longitude and 7°34'51" to 7°47'03 South Latitude. Sleman Regency has an area of 57,482 ha, or 574,82 km2, and covers around 18% of the Yogyakarta Special Region Province area (3,185.80 km2). Sleman Regency has 17 sub-districts, 86 villages, and 1,212 Padukuhan/sub-villages. This area lies at an altitude of 79–1000 m above sea level, which is classified as follows: 1) <100, 2) 100–499, and 3) 500–999 m above sea level. The area of land in Group 1 is 58.16 ha (10.12% of the total area), including Godean and Sleman District; Group 2 is 473.57 ha (82.39% of the total area), covering Gamping, Moyudan, Minggir, Depok, Prambanan, Kalasan, Mlati, Seyegan, Berbah, Ngaglik, Ngemplak, Tempel, Pakem, and Cangkringan District; Group 3 is 43.09 ha (7.50% of the total area), namely Turi District. In 2018, the total land area used for rice cultivation was around 24,517.36 ha (42.65%), with the remaining land area being yards, fields, forests, barren land, and others.

Physical and chemical properties of the soil coordinates of the survey location

The 23 paddy field coordinate points selected as the sampling points were determined using a survey of a semi-detailed map (IAARD, 2016), where each point represents a series of soil map units. Therefore, the physical property, soil unit, landform, relief, and parent material of the soil samples presented in Figure 1 can be known (Sulaeman et al., 2009).

The soil characteristics from this study are as follows: The texture varies between sand, clay, and dust fractions; organic C content ranges from low to high (1.03–4.48%); and soil cation exchange capacity (CEC) values range from low to high (10.70–42.29 me/100 g). Soil cation exchange capacity is an important parameter because it prepares an indication of the dominant type of clay mineral present in the soil and retains nutrients against leaching. The CEC is affected by the nature and amount of mineral and organic colloids in soil. The CEC of clay and organic matter-rich soils is higher than that of sandy soils (Jia et al., 2022).

Figure 1. Overlay of the survey location coordinates with the Semi-Detailed Soil Map of Sleman Regency, Special Region of Yogyakarta (Source: primary data).

Concentrations of heavy metals (Cu, Pb, As, Cr, and Ni) in paddy fields in Sleman Regency

The heavy metals Cu, Pb, and As were found at all soil sampling points (23 locations); in contrast to the heavy metals Ni and Cr, the two metals were not detected in a few sampling points in paddy fields in Sleman Regency. The heavy metal concentrations were Cu>Pb>As>Cr>Ni, and their concentrations exceeded normal limits, according to the Ministry of State for Population and Environment of Indonesia University, Canada (1992), Pickering (1980), and the National Standardization Agency (2004). Cu, Pb, As, Cr, and Ni, respectively, are 153.89 ± 19.99 mg kg^{-1}; 53.45 ± 11.25 mg kg^{-}

^{1}; 9.75 ± 10.01 mg kg^{-1}; and 3.78 ± 4.88 mg kg^{-1} (Table 2).

The coefficient of variation (CV) value determines the distribution and spatial variations of heavy metals. The CVs of heavy metals Cu, Pb, As, Cr, and Ni in paddy fields in Sleman Regency were 12.99%, 21.05%, 35.56%, 102.61%, and 128.92%, respectively. The heavy metals Cu, Pb, and As had < 50% values and were classified as moderate, which indicated their distribution in the soil was relatively uniform. The CV values of Cr and Ni metals were >50% and belonged to broad categories. Figure 2 (box plot) shows the heavy metal distribution of Cu, Pb, As, Cr, and Ni in paddy fields in 15 sub-districts in Sleman Regency.

In the study area, the slope coefficients for heavy metals Pb, As, Cr, and Ni show positive skewness, while those for Cu metals show negative skewness. Positive skewness values indicate no differences in fluctuating heavy metal concentration values. Thus, it is possible that anthropogenic activities carried out have little effect on the heavy metals content in paddy fields (Handayani et al., 2022) in Sleman Regency and vice versa for negative skewness values.

Table 2. Descriptive statistics of heavy metal concentrations in rice fields in Sleman Regency

Description	**Heavy metals**				
	Cu	**Pb**	**As**	**Cr**	**Ni**
Count	23	23	23	23	23
Minimum	118.90	33.80	4.20	0.00	0.00
Maximum	184.00	78.53	26.40	29.50	14.50
Average	153.89[a,b,c]	53.45[b,c]	15.39[b,c]	9.75[a]	3.78 [c]
Standard deviation	19.99	11.25	5.63	10.01	4.88
Coefficients variation (%)	12.99	21.05	36.56	102.61	128.92
Varian	399.72	126.60	31.65	100.13	23.78
Skewness	-0.32	0.24	0,36	0.50	0.97
Kurtosis	-0.86	-0.13	0.37	-0.91	-0.48
VMR*	2.60	2.37	2.06	10.27	6.29
	60 -125[a]	100[a]	0.1-4[b]		20[a]
	2 -100[b]	2-200[b]	0.07 [c]	2.5[a]	10-1000[b]
Critical limit (ppm)**	0.04 [c]	0.07 [c]			0.07 [c]

*) VMR = Variant of Mean Ratio, if the VMR value >1 data is grouped but if < 1 data is spread out,

**) Critical limits of heavy metals in soil according to [a] Ministry of State for Population and Environment of Indonesia and

Dahousie University, Canada (1992); [b] Pickering, 1980; [c] National Standardization Agency. 2004.

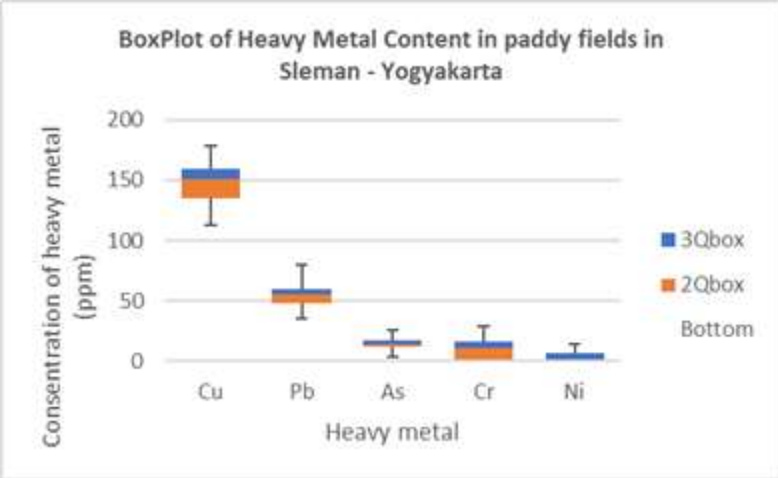

Figure 2. Boxplot of heavy metal concentration in paddy fields in Sleman Regency

Distribution map of heavy metals Cu, Pb, As, Cr, and Ni

The distribution map of heavy metals in paddy field in 15 sub-districts in Sleman Regency (Figure 3) was created by overlaying a semi-detailed map (IAARD, 2016) with the administrative map of Sleman Regency, each sub-district representing one soil map unit. It was composed based on data of mean and standard deviation (Table 2) as references and then classified into high, medium, and low score ranges using Excel software.

Contamination factor (CF) and pollution load index (PLI) of heavy metals

Assessments of soil contamination can be carried out utilizing methods such as the contamination factor, pollution load index, and risk index as outlined by Hakanson in 1980. It is anticipated that the contamination factor value for heavy metals in agricultural soil will fall within the low range, with a contamination factor value below 1. The average value of the contamination factor is Cu>Pb>As>Cr>Ni, and the pollution factor values are 7.69, 2.23, 1.67, 0.18, and 0.16 (Table 3). The maximum contamination factor values for Cu, Pb, and As metals were > 1, but for Cr and Ni metals, they were still included in the low contamination category with contamination factor values < 1. Cu metal at all soil sampling points has a contamination factor percentage value of 100% moderate and Pb metal 100% in the extreme category. For As metal, 8.70% of the soil sampling sites included in the low contamination category and 91.3% included in the fair contamination category. The CF values of Cr and Ni metals at all soil sampling locations showed the contamination low category (Table 4).

The pollution load index values show the pollution values of all heavy metals observed in paddy fields in Sleman Regency (Table 3). The PLI values for five heavy metals (Cu, Pb, As, Cr, and Cu) range from 0.00–1.63, with an average of PLI of 0.41 (<1), which indicates that paddy fields were not expose to five heavy metals (Cu, Pb, As, Cr, and Ni). However, the PLI value for three heavy metals (Cu, Pb, As) was > 1, which indicated that the paddy field contaminated with heavy metals (Cu, Pb, and As).

Table 3. Descriptive statistics of contamination factor (CF) and pollution load index (PLI) of heavy metals in paddy fields in Sleman Regency

	Contamination factor (CF)					**Pollution load index (PLI)**	
	Cu	**Pb**	**As**	**Cr**	**Ni**	5 metals (Cu, Pb, As, Cr, Ni)	3 metals (Cu, Pb, As)
Mean	7.69	2.23	1.67	0.18	0.16	0.41	1.93
Median	7.84	2.24	1.55	0.21	0.00	0.00	1.94
Standard deviation	1.00	0.47	0.61	0.19	0,21	0.59	0.20
Min	5.95	1.41	0.46	0.00	0.00	0.00	1.52
Max	9.20	3.27	2.87	0.55	0.63	1.63	2.24
Sum	176.98	51.22	38.47	4.15	3.78	9.38	44.29
Count	23.00	23.00	23.00	23.00	23.00	23.00	23.00

Table 4. Percentage of contamination factor (CF) value categories

Heavy metals	Low	Moderate	Considerable	Extreme
Cu	0.00	0.00	0.00	100.00
Pb	0.00	100.00	0.00	0.00
As	8.70	91.30	0.00	0.00
Cr	100.00	0.00	0.00	0.00
Ni	100.00	0.00	0.00	0.00

Ecological risk assessment (Er) and potential ecological risk index (RI) of heavy metals

The average ecological risk assessment (Er) value from highest to lowest is Cu>As>Pb>Ni>Cr, with an average Er value of 38.47; 16.72; 11.14; 0.82; 0.36 (Table 5). The Er values for all Cu, Pb, As, Cr, and Ni metals are <40, indicating that the concentrations of these metals have a low potential ecological risk. The RI values represent potential ecological risks posed by all metals observed. The RI value is between 50.82 and 87.55, and the average RI value is 67.52 (Table 5), so the RI value is <150, which belongs to the category of low ecological risk.

Enrichment factor (EF) and geoaccumulation index (Igeo)

The enrichment factor (EF) for heavy metals in the paddy fields of Sleman Regency includes low/deficient category for Cr and Ni, medium category for Cu and Pb, and high category for As (Table 6). The average I-geo value in this series is Cu (2.33) > Pb (0.54) > As (0.05) > Cr (-2.33) > Ni (- 2.85). Base on I-geo value, the paddy fields are moderate to heavy contamination of Cu (2-3), not contamination to moderately contamination of Pb and As (0-1); and not contamination of Cr and Ni heavy metal (Table 6) (Mandal et al., 2022).

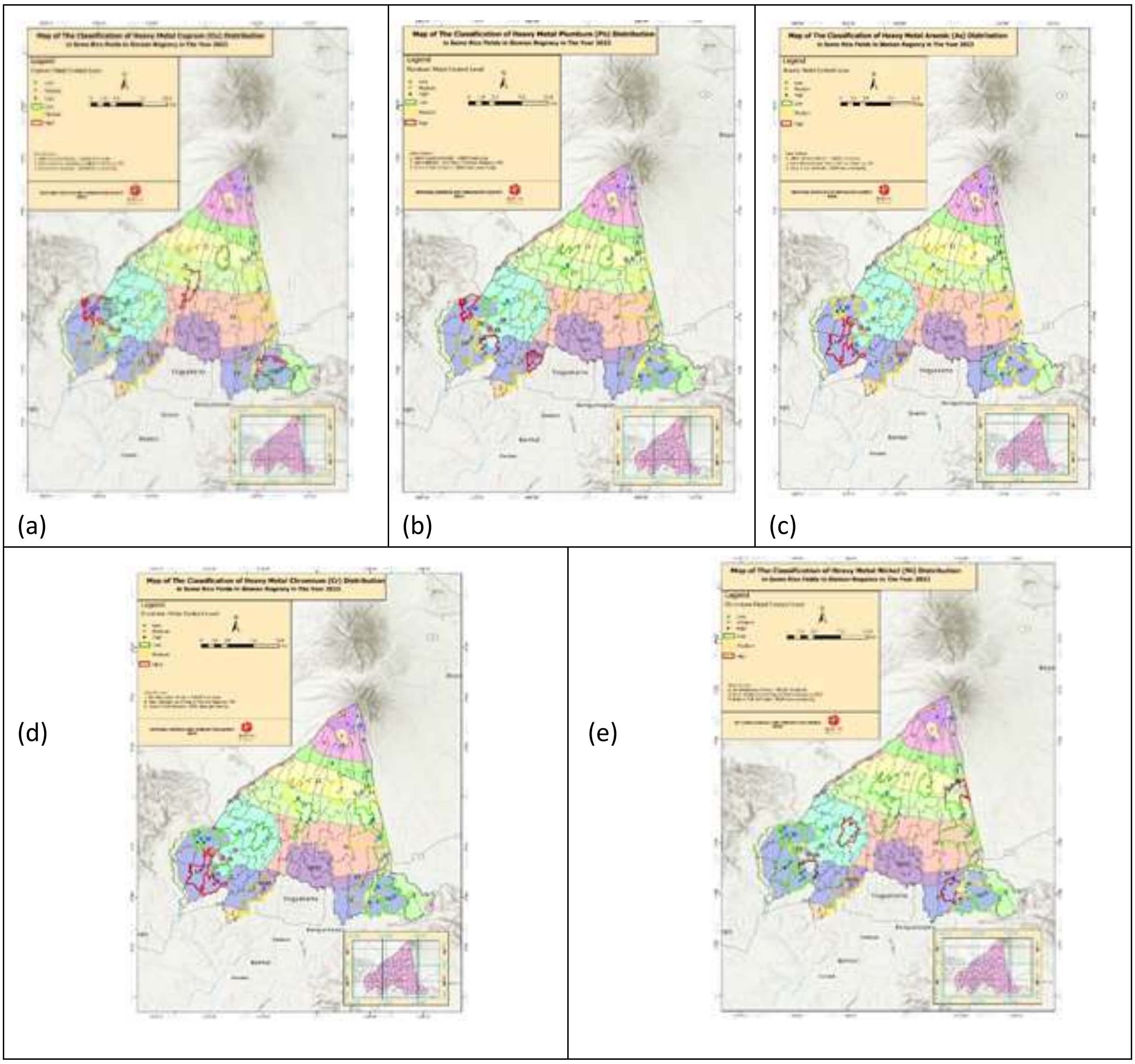

Figure 3. Distribution map of heavy metals Pb (a), Cu (b), As (c), Cr (d), and Ni (e) in rice fields in Sleman Regency (Source: primary data).

Table 5. Descriptive statistics of ecological risk assessment (Er) and potential ecological risk index (RI) of heavy metals in paddy fields in Sleman Regency

Statistical	**Ecological risk assessment (Er)**					**Potential ecological risk index (RI)**
	Cu	**Pb**	**As**	**Cr**	**Ni**	
Mean	38.47	11.14	16.72	0.36	0.82	67.52
Standard error	1.04	0.49	1.28	0.08	0.22	2.03
Median	39.18	11.21	15.54	0.42	0.02	67.62
Standard deviation	5.00	2.34	6.11	0.37	1.06	9.73
Sample variance	24.98	5.50	37.39	0.14	1.12	94.62
Kurtosis	-0.86	-0.13	0.37	-0.91	-0.48	-0.61
Skewness	-0.32	0.24	0.36	0.50	0.97	0.22

Min	29.73	7.04	4.57	0.00	0.00	50.82
Max	46.00	16.36	28.70	1.09	3.15	87.55
Sum	884.88	256.11	384.67	8.31	18.91	1552.88
Count	23.00	23.00	23.00	23.00	23.00	23.00
Confidence level	2.16	1.01	2.64	0.16	0.46	4.21

Table 6. Enrichment factor (EF) and geoaccumulation index (Igeo) of heavy metals in paddy fields in Sleman Regency

Heavy metals	Enrichment factor (EF)	Geoaccumulation index (Igeo)
Cu	3.06 ± 0.70	2.35 ± 0.19
Pb	1.78 ± 0.46	0.54 ± 0.31
As	4.33 ±1.23	0.05 ± 0.62
Cr	0.03 ± 0.03	- 2.33 ± 0.56
Ni	0.03 ± 0.04	-2.85 ± 1.95

Discussion

Location distribution map, concentration of heavy metals (Cu, Pb, As, Cr, Ni)

Soil samples were taken from rice fields in Sleman Regency, with coordinate points representing soil map units points 1, 4, 11, 14, 15, 16, and 17. The lowland landforms are flow paths, volcanic plains, volcanic slopes, and tectonic hills; relief is flat (0–3%), slightly flat (1–3%), undulating (3–8%), undulating (8–15%), and a little hilly (15–25%); parent materials are andesite, clay sediment, and sandy clay stone. The heavy metal content (Cu, Pb, As, Cr, and Ni) varies from low to high. Based on this result, it appears that paddy fields with high Cu, Pb, As, Cr, and Ni metal contents have the same parent material, that is, andesite. Sources of heavy metals in the soil come from natural and anthropogenic sources (Sukarjo et al., 2019), as well as climate (Dewi et al., 2023). Soil contains heavy metals inherited from the parent soil material. From an anthropogenic perspective, in addition to irrigation water, the sources of heavy metals in farmland (Affum et al., 2020) can also come from industrial activities and agricultural cultivation carried out by farmers by applying chemical fertilizers and pesticides during the planting period (Handayani et al., 2022). Numerous studies have demonstrated that various factors influence the distribution of heavy metals (Dong et al., 2021).

The study examined the distribution of heavy metals (enrichment or aggregation of copper, lead, arsenic, chromium, and nickel) in paddy soil in the Sleman Regency using VMR values (Table 3). Concentrations of copper and lead, chromium, and nickel metals are declared hazardous or critical metals in the soil if each of these metals crosses the threshold, according to the Ministry of State for Population and Environment of Indonesia and Dahousie University, Canada (1992); Pickering (1980); and the National Standardization Agency (2004). Cu > 125 mg/kg; Pb > 0.07 mg/kg (National Standardization Agency, 2004); As > 4 mg/kg (Ministry of State for Population and Environment of Indonesia and Dahousie University, Canada 1992); Cr > 2.5 mg/kg; and Ni > 0.07 mg/kg (National Standardization Agency 2004).

Each country has quality standards for determining the maximum limit of heavy metals in soil. Australia limits heavy metals content in soil for Pb, Cr, As, and Ni at 100, 50, 20, and 70 mg/kg, respectively. Russia sets the maximum limit for heavy metals in soil at 55 mg/kg for Pb, 3.8 mg/kg for Cr, 4.5 mg/kg for As, and 2.6 mg/kg for Ni. In Bulgaria, the maximum values for the concentrations of heavy metals Pb, Cr, As, and Ni in soil were 40, 90, 15, and 6 mg/kg, respectively (Chen et al., 2018). In Indonesia, it is even stricter than in other countries, namely 0.04 mg/kg for Cu, 0.07 mg/kg for Pb, 0.07 mg/kg for As, 2.5 mg/kg for Cr, and 0.07 mg/kg for Ni (National Standardization Agency, 2004).

The concentration of heavy metals at each location is not always fixed but depends on land management, the source of metal pollution, and the rock (the main ingredient) as the source of the metal itself. Metal contamination in agricultural soil depends on several factors, including the amount of metal in the rock from which it originates, the amount of minerals added to the soil as fertilizer, the amount of metal deposits from the atmosphere that enter it, and the amount transported by plants during the harvest process (Darmono, 2001).

Environmental pollution in Sleman Regency often occurs due to agricultural activities that do not implement conservation principles, such as chemical fertilizers and pesticides used, and pollution from motor vehicles. The concentration of Cu, Pb, As, Cr, and Ni in paddy fields from 15 sub-districts in Sleman Regency shown in Figure 2 (box plot). The high concentrations of Cu, Pb, As, Cr, and Ni that exceed the specified limits (based on descriptive statistics in Table 3) may come from natural sources such as the weathering of source rocks, volcanic ash from Mount Merapi, and intensive agricultural cultivation by using chemical fertilizers and pesticides. Fertilizers and some pesticides are sources of heavy metals such as cadmium (Cd), cobalt (Co), nickel (Ni), lead (Pb), and chromium (Cr). Commercial pesticides with active ingredients such as organophosphate, avermectin, pyrrole, pyrethroid, carbamate, dithiocarbamate, triazole, imidazole, and glycine groups contain Pb and Cd with concentrations of 2.70–22.31 mg/kg and 0.04–0.50 mg/kg, respectively. Moreover, five types of commercial inorganic fertilizers also contain Pb and Cd with concentrations of 10.53–28.09 mg/kg and 0.07–0.52 mg/kg, respectively.

Contamination factor (CF) and pollution load index (PLI), Ecological risk assessment (Er) and potential ecological risk index (RI) of heavy metals

The contamination factor value for Cu metal has reached the polluted category at all sampling point locations because this metal is naturally found in the earth's crust. Its concentration is relatively small (20) compared to the other metals's concentrations except As (9.2). In addition, because the Cu metal content is higher (153.89 ppm) than the other metals, the average value of the metal pollution factor Pb at all sampling location points and As at several location points is included in the low category. The CF values for Cu are high, and those for Pb and As are moderate due to the presence of residues from the use of inorganic fertilizers, herbicides, fungicides, and chemical pesticides. Senesil et al. (1999) reported that pesticides contain “As 0.8–60 mg/kg, Cu 4–56 mg/kg, Hg 0.6–42 mg/kg, Mn 1–17 mg/kg, Pb 11–60 mg/kg, and Zn 1–30 mg/kg”. These are some of the sources of contamination for components.

The pollution load index (PLI) aims to determine the pollution status of a collection of metals in a locality. The PLI value determines whether the water is polluted or not by heavy metals [16]. The PLI value of the three heavy metals (Cu, Pb, and As) is > 1, which indicates these three metals exposed paddy fields in Sleman Regency. It is in line with the high contamination factor (CF) value. Cu, Pb, As, Cr, and Ni metals are included in the category of low ecological risk potential for all locations because they have an Er value < 40. Based on the lower Er value, potential ecological risk index (RI) value, namely <150 (average 67.52), belongs to the low-risk category. The RI value shows the potential risk posed by all the metals observed.

Enrichment factor (EF) and geo-accumulation index (Igeo)

The enrichment factor (EF) value for Cr and Ni metals is <1, which indicates that there is no enrichment of these metal elements in the sediment, and anthropogenic factors do not play a significant role in the enrichment of these elements. The EF values for Cu, Pb, and As heavy metals are 3.06, 1.78, and 4.33, respectively, which indicates there is enrichment of these elements by anthropogenic activities. In general, if the EF value is <1, the heavy metal comes from natural processes, but if the EF value is >1, the pollution comes from human or anthropogenic activities. The EF value is the concentration comparison of a metal in the sediment with a reference metal, for example, Fe in the sediment (Jahan and Strezov, 2018). Fe is a reference metal for several reasons, including: (1) Fe is associated with the surface layer of sediment, (2) the geochemical properties of Fe are relatively identical with many metals, and (3) the natural concentration of Fe in sediment tends to be uniform (Syakti et al., 2015).

Igeo determination is a process that assesses the presence of heavy metal pollution in coastal sediments by analyzing variations in the earth's lithosphere layers (Syakti et al., 2015). The average Igeo value for rice fields in Sleman Regency is – 2.85 – 2.33, which indicates that the rice fields are very polluted by heavy metals Cu, Pb, As are originating from anthropogenic sources but not contamination of Cr and Ni. It is in line with the EF value, which means that there is enrichment due to anthropogenic activities that cause the accumulation of Cu, Pb, and As metals (ER > 1), while the enrichment of the heavy metals Cr and Ni comes from natural processes. Chemical pesticides and inorganic fertilizers overuse is one example of anthropogenic activity (Sukarjo et al., 2019).

Conclusions

The results detected the soil contamination of Cu, Pb, As, Cr and Ni from all sampling points in Sleman Regency representing soil map units number 1, 4,11, 14, 15, 16 and 17. The average heavy metal concentration is Cu>Pb>As>Cr>Ni. There was high-level contamination factor of Cu (CF =7.69), medium-level contamination factor of Pb (CF = 2.23) and As (CF = 1.67), and low-level contamination factor of Cr and Ni (CF < 1). Paddy fields were not at once exposed to the five soil metals (Cu, Pb, As, Cr, and Ni with a PLI value, <1); but showed pollution for Cu and As metals together (PLI value > 1). Cr and Ni were derived from natural processes, but Cu, Pb, and As contaminants were anthropogenically produced. Cu, Pb, As, Cr, and Ni metals have a low potential ecological risk (Er<40). The enrichment factor (EF) for heavy metals in the paddy fields of Sleman are low category (Cr, Ni), medium category (Cu, Pb), and high category for As. The excessive soil pollutant in the soil gives a serious caution on chemicals use. Agricultural practices need to be eco-friendly and sustainable. Future studies can assess to what extend the transfer of heavy metal from soil to plants or rice.

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Conflict of Interest

The authors declare that there are no conflicts of interest.

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