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Research Article

Heavy Metals Accumulation In Conventional Rice Production

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Abstract

Rice grains produced in the conventional system are a prime source of heavy metal exposure in the human body leading to various health problems. The study aims to asses heavy metals concentration in soil and rice grain produced with the conventional system in Banyumas, Indonesia. Furthermore, it examines the pollution index and bioaccumulation factor. There were 37 samples collected based on soil type, elevation, and irrigation system. The heavy metals level in soil ranged from 2.7 to 39.92 ppm of Pb, 0.11 to 3.01 ppm of Cd, and 4.79 to 61.32 ppm of Cr. Pb and Cr accumulation levels were below the maximum permissible concentration (MPC). A different result in Cd exceeded the MPC in 21 samples. Rice grain shows a high accumulation in Pb (6.85 ppm) and Cr (5.73 ppm) that exceeds the maximum standard. Cd was exceeding in 9 samples with an average of 0.54 ppm. The Pollution Index (PI) was medium in Cd and low in Pb and Cr. The Bioaccumulation Factor (BAF) of heavy metals in rice from the higher is Cd>Pb>Cr with averages of 0.64, 0.53, and 0.30.

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Introduction

Rice is the most grown crop in Indonesia as a staple food for the people. The government has consistently pushed national productivity through intensification to meet the needs of the growing population. Banyumas is a region that contributes about 380 tons per year to national rice production (Banyumas, 2017).

The increasing use of chemical fertilizer without sufficient input of organic matter could increase heavy metal accumulation in the soil, such as Lead (Pb) and Cadmium (Cd), then transferred to plants (Merismon et al., 2017). Excessive pesticide application in conventional cultivation leads to heavy metal pollution in soil (Sutrisno and Kuntyastuti, 2015), including Chromium (Cr)(Manurung et al., 2018).

The accumulation of heavy metals such as Pb, Cd, and Cr in soil and food may harm the environment and human health. Heavy metals in soil may affect human health by consuming plant products grown in contaminated soil (Alexander et al., 2006; Zhuang et al., 2009). Consuming food grown in high Pb and Cd concentrations of soil is considered to risk health (Zhuang et al., 2009). Therefore the contamination of heavy metals has been a serious concern to agricultural land management to mitigate environmental disasters and health problems from several diseases. (Alexander et al., 2006; Zhuang et al., 2009; Portier, 2012).

Exceeding the tolerable level, heavy metals pollution in agricultural soil of some regions in Indonesia requires serious consideration (Marselius and Laoli, 2021; Handayani et al., 2022). Several studies demonstrated the danger of heavy metals accumulation due to chemical fertilizer and pesticides application on rice crops (Amelia and Rachmadiarti, 2015), onion (Kusumaningrum et al., 2012), potatoes (Kusdianti et al., 2014), and agricultural soil in general (Suastawan et al., 2016; Komarawidjaja, 2017). According to (Sutrisno and Kuntyastuti, 2015), heavy metal pollutants originate from industrial waste disposal, agrochemical application, and household waste disposal to waterways.

Accumulation of Pb, Cd, or Cr at a certain level causes growth inhibition, yield decline, and premature death of the crop. In contaminated soil, the plant intakes heavy metals and accumulates them in root, leaf, fruit, and grain (Fang and Zhu, 2014). Pollution of Pb, Cd, and Cr in agricultural soil of some regions in Indonesia has surpassed the tolerable limit (Susanti et al., 1992; Sutrisno and Kuntyastuti, 2015). Therefore, it needs serious attention from the farmers, government, and industrial society.

According to the current state, it is essential to evaluate the distribution of heavy metals in the conventional rice production system. The assessment output is a baseline to determine remediation strategies and agricultural development policies. The study aims to assess the accumulation of Pb, Cd, and Cr in soil and rice grain, in addition to the pollution index and bioaccumulation factor in the rice field in Banyumas regency, Indonesia.

Materials and Methods

The study was conducted from July to December 2020. The coverage area of the study is in the Banyumas regency of Indonesia. The study was conducted in several stages, including spatial analysis, survey, and heavy metals analysis.

Spatial analysis was carried out to determine the number and location of samples to be collected. Various maps, including a geographical map, satellite image, irrigation map, and soil map of Banyumas, were overlayed by applying software of geographical information system (GIS) to obtain a homogenous land unit. Sampling sites of Soil and rice were determined based on the number and area of fixed homogeneous land units (Ahadiyat et al., 2021).

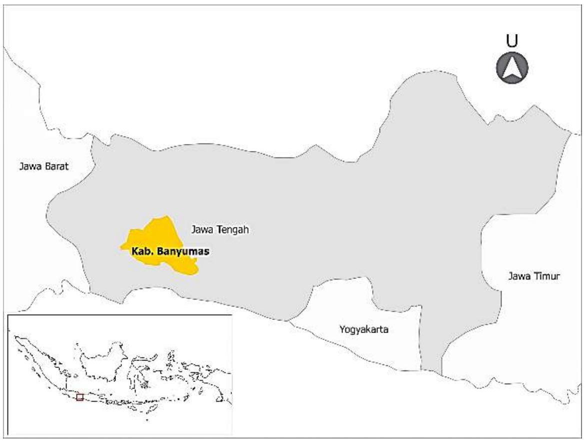

Figure 1. The geographical location of the study area

Soil and rice grain samples from each site were analyzed to assess heavy metals level. The soil was prepared by air dried, crushed, and sieved in 2 mm size before the concentration of Pb, Cd, and Cr were measured. 100 rice grains were prepared for heavy metals level measurement.

Approximately 0,5 grams of the mashed soil and rice sample was put into a porcelain cup, then heated at 300 °C in the kiln for half an hour. The temperature of the furnace was increased to 500 °C so that the sample was scorched (white). Then the cup and its contents were cooled in a closed state and then were added 1 mL of 25% HCl until dissolved and then heated using a hot plate until the HCl evaporated. The sample in the cup that had been evaporated was dissolved with 10 mL of 1 N HCl and filtered. The filtrate was diluted ten times, and the solution was read for Pb, Cd, and Cr metals using an atomic absorption spectrophotometer (SSA) (Bauer et al., 1978; Sulaeman et al., 2005).

To assess the Pb, Cd, and Cr contamination, a pollution index (PI) was calculated using the formula:

$$PI = Cn/ Bn$$

where Cn (mg/kg) is the measured concentration of each heavy metal, and Bn is the background value for each metal (Siti Norbaya et al., 2014).

The PI of each metal was classified as low contamination ($IP<1$), moderate contamination ($1\leq IP\leq 3$), high contamination ($3<IP\leq 6$), and very high contamination ($IP<6$) (Gupta et al., 2021). The background values provide a guide to ascertain the presence of anthropogenic effects that could lead to hazardous or beneficial consequences. In environmental geochemistry and health, the background is considered as the natural concentration in environmental material devoid of human influences (Kazapoe and Arhin, 2021). The definition of background (geogenic) values, as opposed to baseline values, is very important in environmental legislation, which fixes, at different levels in various countries, the intervention limits for both organic and inorganic substances in soils (Cicchella et al., 2005). The background value in this research was adopted from the official background value of the Netherlands (Brus et al., 2009).

The heavy metal concentration in soil was standardized to Maximum Permissible Concentrations and Negligible Concentrations for metals released by the National Institute of Public Health and The Environment, Netherlands (Crommentujin et al., 1997), for the official Indonesian standard for maximum concentration of heavy metals in soil was not available yet (Erfandi and Juarsah, 2013).

The heavy metal concentration in rice grain was compared to standards provided by FAO/WHO (General Standard for Contaminants and Toxins in Food and Feed CXS 193-1995, 2019), The National Standardization Body of Indonesia (BSN, 2009), and The National Standard of China (National Food Safety Standard Maximum Levels of Contaminants in Foods, 2017).

The translocation capability of heavy metals from soil to plant was calculated using bioaccumulation factor (BAF) as in the following equation:

$$BAF = [CR]/[CS]$$

where CR (mg kg−1) and CS (mg kg−1) represent the concentration(s) of heavy metals in rice grains and soil on a dry-weight basis (Khan et al., 2013; Kong et al., 2018).

Results and Discussion

Sample distribution

Sampling sites were determined using homogenous land units (HLU) generated in the spatial analysis stage based on irrigation type, soil type, and elevation.

Irrigation type:
SI: irrigated soil
ST: rainfed soil
Soil type:
1: Hidromorf alluvial
2: Yellowish-grey alluvial
3: Asociation of grey alluvial and grey brown alluvial
4: Asociation of brown andisol and brown regosol
5: Asociation of andisol
6: Asociation of brown latosol
7: Complex of yellowish-red latosol and yellow-red podzolic
8: Complex yellow-red podzolic
9: Brown latosol
Elevation:
a: 25-250 masl
b: 250-500 masl
c: 500-750 masl
d: 750-1000 masl

There were 37 sampling sites representing 25 types of HLU which resulted from maps overlay (Figures 2 and 3).

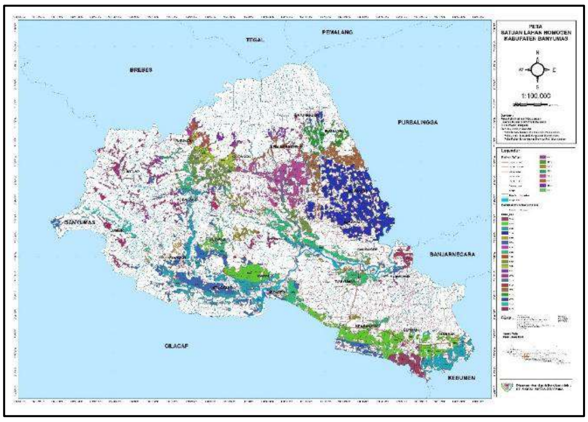

Figure 2. Map of Homogenous Land Unit in Banyumas

Figure 3. Distribution of sampling sites

Table 1. Types of homogenous land units resulted from spatial analysis

No.	Homogonous Land Unit	Area (ha)	Sampling site
1.	SI1a	1144.9	1
2.	SI2a	4502.24	3
3.	SI3a	4469.52	3
4.	SI5a	5898.93	4
5.	SI6a	230.75	1
6.	SI7a	3662.21	2
7.	SI7b	38.33	1
8.	SI8a	3785.83	3
9.	SI8b	70.66	1
10.	SI9a	1836.5	1
11.	SI9b	696.52	1
12.	ST2a	117.81	1
13.	ST3a	102.26	1
14.	ST4c	27.7	1
15.	ST5a	1743.25	1
16.	ST5b	917.10	1
17.	ST5c	168.18	1
18.	ST6a	453.41	1
19.	ST7a	2276.59	2
20.	ST7b	317.72	1
21.	ST8a	925.54	1
22.	ST8b	151.31	1
23.	ST9a	2524.48	2
24.	ST9b	1017.61	1
25.	ST9c	434.99	1
	TOTAL	***37514.34***	***37***

Accumulation of heavy metals in soil

Accumulation of Pb and Cr in soil ranged from 2.7 – 39.92 ppm and 4.79 – 61.32 ppm. This level was lower than the Maximum Permissible Concentrations and Negligible Concentrations for metals (MPC), according to the National Institute of Public Health and The Environment Netherlands (Crommentujin et al., 1997). A different result was found in Cd that exceeded the maximum standard in 21 sites (Table 2.).

Cd has contaminated most of the ricefield area in Banyumas. There were 21 from 37 sites containing Cd more than the allowable limit. Those sites, according to the homogenous land unit from spatial analysis, represent 22,199.27 ha or equal to 59% of the total ricefield in Banyumas (Figure 7.). The soil excess in Cd may produce contaminated rice which is harmful to consume

Table 2. Heavy metal accumulation in soil and rice grain

Statistics	Accumulation in soil (ppm)			Accumulation in rice grain (ppm)		
	Pb	Cd	Cr	Pb	Cd	Cr
Sample	37	37	37	37	37	37
Min	2.70	0.11	4.79	0.22	0.10	2.54
Max	39.92	3.01	61.32	32.13	2.16	10.36
Median	26.44	1.80	27.65	3.46	0.24	5.49
Mean	22.64	1.62	27.80	6.85	0.54	5.73
stdev	11.43	0.88	14.27	9.95	0.67	1.93
Maximum Permissible Concentration	140 [1]	1.6 [1]	100 [1]	0.2 [2,4] 0.3 [3]	0.4 [2,3] 0.2 [4]	1.0 [4]

[1] National Institute of Public Health and The Environment Netherlands 1997
[2] SNI 7387:2009
[3] FAO/WHO Codex Alimentarius International Food Standards
[4] National Standard of the People's Republic of China

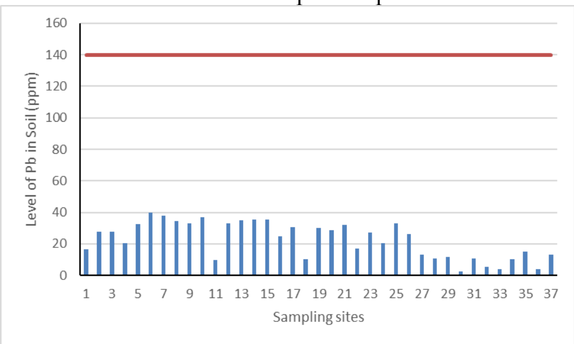

Figure 4. Distribution of Pb levels in sampling sites

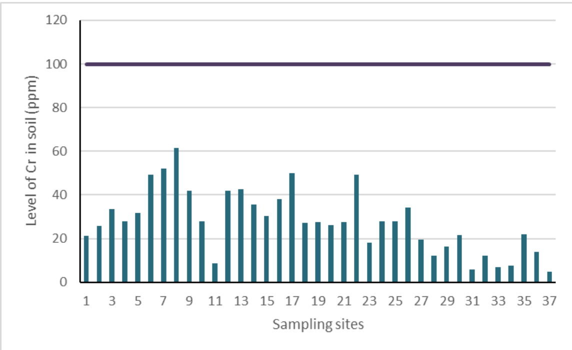

Figure 5. Distribution of Cr levels in sampling sites

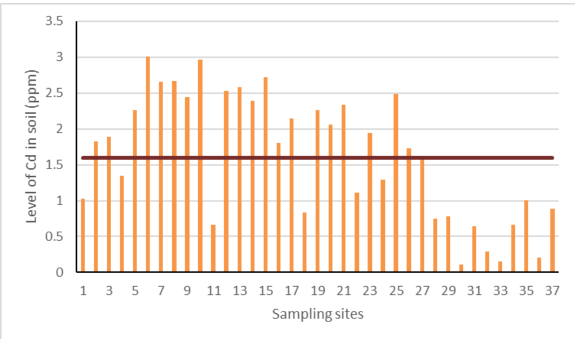

Figure 6. Distribution of Cd levels in sampling sites

The high level of Cd in the soil after harvesting was caused by the intense use of chemical fertilizers and pesticides in rice cultivation. Cd in soil was released by inorganic fertilizers, mainly N and P, which were categorized as the source of contaminants for containing high amounts of heavy metals. For example, SP-36 contains 11 ppm of Cd and 67 ppm of Pb. Pb and Cd in phosphate fertilizer originated from natural rock phosphate as the raw material of the fertilizer. Natural rock phosphate generally consists of large amounts of phosphate, carbonate, and heavy metals (Merismon et al., 2017).

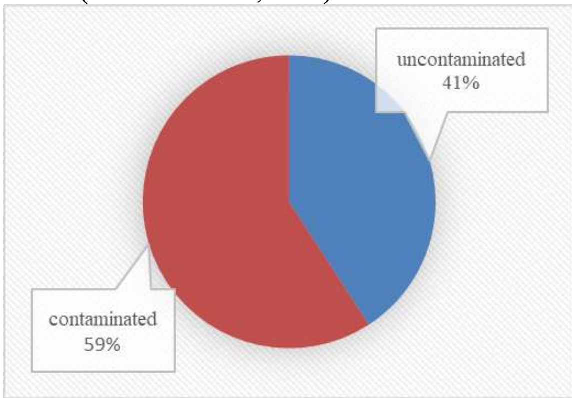

Figure 7. Cd contaminated soil in Banyumas

Banyumas farmers commonly apply chemical fertilizer twice in a growing season at 7-14 and 21-28 days after planting. In pest control, farmers regularly spray pesticides at least twice for a season or more, depending on the pest population. According to the current level of heavy metals in soil due to those chemical inputs, it is crucial to bring a conservative effort to minimize the accumulation in soil. Some organic-based fertilizers and pesticides could be applied as alternative inputs in rice cultivation after conducting soil remediation from heavy metals in Banyumas.

Heavy Metals Accumulation in Rice Grain

Rice grain samples were collected from each site of soil sampling. It was found that Pb, Cd, and Cr accumulation in rice grain ranged from 0.22 – 32.13 ppm, 0.1 – 2.16 ppm, and 2.54 – 10.36 ppm. The level of Pb and Cr in rice grain from each site exceeds the maximum tolerable level according to Indonesian and China National Standards (0,2 ppm). On the other hand, Cd accumulation in rice grain exceeds the highest standard only in 9 of 37 sampling sites (0.4 ppm).

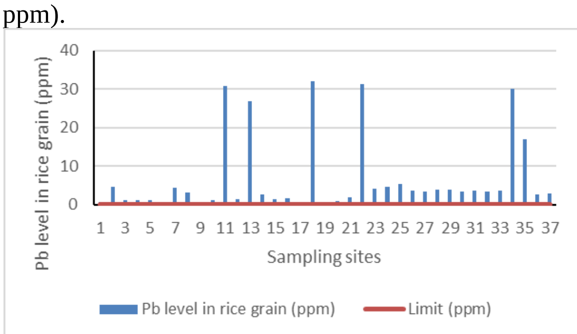

Figure 8. Pb level in rice grain

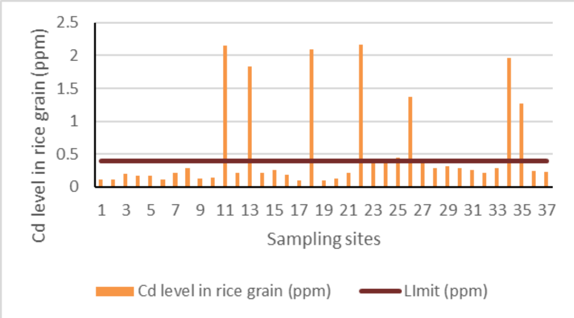

Figure 9. Cd level in rice grain

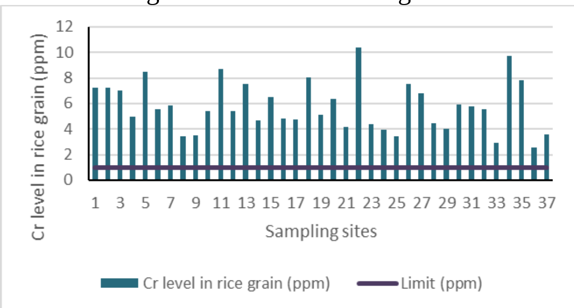

Figure 10. Cr level in rice grain

There was no correlation between heavy metal accumulation in soil and rice grain. The Pb concentration in soil was relatively low, in contrast to the high Pb concentration in rice grain which was beyond the safety limit of food. Pb is a carcinogenic element for humans therefore, its exposure at a chronic level could cause headaches, convulsions, palsy, and nerve damage, especially in children (Guo et al., 2020). The level of Pb accumulation in rice was determined by the cultivation method. Agrochemical applications, including fertilizers and pesticides, contribute to the high accumulation of heavy metals in rice.

The high level of measured Pb in rice was also caused by direct sampling of a rice grain from the field at harvesting without the milling process. Pb level in rice grain could be decreased due to processing to produce white rice. This result is in line with the study of (Norton et al., 2014) and (Tatahmentan et al., 2020), which found a higher level of Pb in rice grain collected from the field than that of white rice from the market.

Cadmium was found to exceed the food safety standard (0.4 ppm) in 9 sites from villages of Kebasen (2.15 ppm), Ajibarang Kulon (1.83 ppm), Beji (2.1 ppm), Kranggan (2.16 ppm), Pasiraman Kidul (0.43 ppm), Kalisari (0.44 ppm), Kasegeran (1.37 ppm), Kaliputih (1.96 ppm) and Semedo (1.27 ppm). The high level of Cd found in rice was suspected to be associated with Cd in contaminated soil of sampling sites which represent 59% of the total area in the Banyumas ricefield.

Intensification of rice cultivation through superior variety and agrochemical input, which was implemented widely since the industrial revolution, has led to national food sufficiency and heavy metals contamination in crop products. The average concentration of Cd in rice grain was 0.5 ppm. This level of Cd shows that Cd accumulation has increased significantly from 40 years ago when (Suzuki et al., 1980) reported that the lowest Cd concentration in Asian rice (0.04 ppm) was cultivated in Java, with 0.02 ppm of the world average (Watanabe et al., 1989). The study by (Shi et al., 2020) also reported that the Cd concentration of Indonesian rice was 0.005 – 0.597 ppm with 0.019 ppm of the global median value.

Cadmium is one of the most mobile and toxic elements in the soil. Cd could be easily transferred into plant tissue and accumulated in harvested part. When Cd enters the human body, it will cause some harmful effects on the lung, cardiovascular, and musculoskeletal systems. The same as Pb concentration, Cd in whole grain (brown rice) is higher than that in white (polished rice) because the outer layer of rice grain accumulates more heavy metals (Sun et al., 2008). Therefore rice milling would lower the Cd concentration in rice (Meharg et al., 2013).

Indonesian national standard for maximum Cr level in rice is not available yet, therefore, this study adopted other countries' standards. Each rice grain from all sample sites contains Cr more than the safety limit of food, according to the National Standard of China (1.0 ppm). Consuming rice grown in soil containing high levels of Cd, Cr, or Zn could seriously risk human health because 24 to 22% of total heavy metals in plant biomass are accumulated in grains (Wang et al., 2003).

Pollution Index (PI)

Most of the measured heavy metals in this study were lower than the background value adopted from the Netherlands' official background value of heavy metals in soil. Pb levels in soil that ranged from 2.7 to 39.92 ppm were lower than the background value (50 ppm). Only one site of soil samples contained Cr more than the background value (55 ppm), in contrast to Cd, which mostly exceeded the background value (Brus et al., 2009).

Table 3. Pollution Index and Bioaccumulation Factor

	Pollution index			Bioaccumulation factor		
Statistics	Pb	Cd	Cr	Pb	Cd	Cr
Samples	37	37	37	37	37	37
Min	0.05	0.18	0.09	0.01	0.04	0.06
Max	0.80	5.02	1.11	3.11	3.26	1.30
Median	0.53	3.00	0.50	0.16	0.18	0.21
Mean	0.45	2.71	0.51	0.53	0.64	0.30
stdev	0.23	1.46	0.26	0.86	0.92	0.28

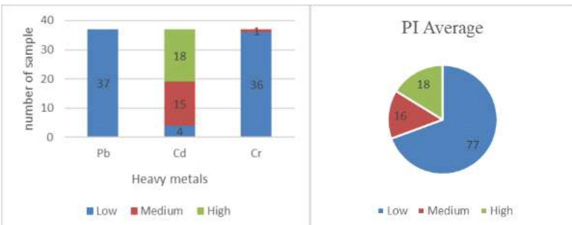

Figure 11. Distribution of pollution index

The evaluation of the pollution index (PI) showed that each site has a low PI for Pb (IP< 1). PI's of Cd were low in 4 sampling sites, medium (1≤ IP≤ 3) in 15 sampling sites, and high (3<IP≤6) in 18 samples. In addition, PI's of Cr were low in 36 sampling sites and medium in 1 sampling site. This result indicates that land management carried out by Banyumas farmers in rice cultivation affects the increased heavy metals accumulation in soil. The anthropogenic contamination of heavy metal in the Banyumas ricefield from lower to higher was Pb< Cr < Cd.

Bioaccumulation factor (BAF)

Bioaccumulation factors (BAF) of rice crops were 0.01 – 3.11 for Pb, 0.04 – 3.26 for Cd, and 0.06 – 1.30 for Cr. Sequentially, heavy metals with higher bioaccumulation factor average to the lower were Cd>Pb>Cr. The bioaccumulation factor of heavy metals in the plant depends on plant and metal species. This fact is in line with what was reported by (Kabata-Pendias, 2010) that cadmium is the most preferred plant to absorb with intensive accumulation, followed by Pb with medium accumulation, and the last is Cr with low accumulation.

Furthermore, there are several sites in Banyumas where the accumulation factor is >1 (6 sites in Pb, 8 sites in Cd, and 2 sites in Cr), and 2 sites show BAF >1 for the three of Pb, Cd, and Cr. According to (Usman et al., 2019), a plant with BAF >1 indicates that the plant is a bio-accumulator and has the potential to be a phytoremediation agent, and BAF<1 shows that the plant is an excluder.

Several sites with BAF>1 might be caused by foliar application of chemical fertilizer or pesticides during cultivation. Metals from aerial sources may increase contamination in a plant through foliar uptake. Heavy metals from foliar uptake could be translocated to other parts of the plant (Kabata-Pendias, 2010) including rice grain. Plant ability to uptake metals from the atmosphere varies among species. Furthermore, (Kabata-Pendias, 2010) reported that some plants were able to uptake large amounts of metals through leaves, including cereal crops which are sensitive to air pollution for Pb and Ni accumulation in leaves

Conclusion

The concentration of Pb and Cr in the soil of the Banyumas ricefield were below the maximum permissible concentration (MPC) with an average of 22.64 dan 27.80 ppm. In contrast to Pb and Cr, the concentration of Cd had exceeded MPC in 21 sites which represent 59% of ricefield in Banyumas regency. The accumulation of Pb and Cr in rice grain exceeded the safety limit with an average of 6.85 dan 5.73 ppm. Furthermore, Cd in rice grain exceeded the limit in 9 of 37 sample sites. Pb and Cr showed a low pollution index (PI), while Cd was moderate. Bioaccumulation factor (BAF) of heavy metals in rice crops sequentially from the highest to the lowest was Cd>Pb>Cr with averages of 0.64, 0.53, and 0.30. These conditions of heavy metals in the rice production system should become a baseline to arrange strategies or policies in protecting people from heavy metal exposure from rice. Minimizing the use of chemical fertilizer and more organic material input could become supporting action to regenerate the contaminated soil, in addition to remediation practices such as bioremediation or phytoremediation.

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to YUGI

YUGI R. AHADIYAT:

Your manuscript entitled "Heavy Metals Accumulation In Conventional Rice Production" has been reviewed by the Journal of Degraded and Mining Lands Management reviewers. Based on the reviewer comments (attached), the authors are suggested to resubmit after Major Revision and Rewriting. A substantial amount of work is necessary to raise the present manuscript to the standard of a research journal article.

All the best

Prof Eko Handayanto PhD
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REVIEW-1

Research Article

In its current form, this manuscript cannot be accepted for publication as a research article due to the following reasons:

(1) the content of the manuscript does not fit well within the scope of this journal (the management of degraded and mining lands); the authors have to make adjustments in order to fit within the scope of this journal
(2) this manuscript only presents the results of problem identification (distribution of heavy metals in rice plants) to be used as a basis for a research
(3) The Introduction section does not describe the current problems to be solved (in the study area), but it only presents the theories and research results of others. Thus, the rationale behind this study of this manuscript is unclear
(4) The materials and Methods section only presents sampling and sample analysis methods, not research methods
(5) Results are mixed with methods; For example, the sample distribution section is presented in the Results and Discussion section; this should be placed in the Materials and Methods section
(6) Maps are blurred, and legends are letters too small
(7) The authors did not seriously prepare this manuscript for publication in a journal; Tables and Figures are simply presented in JPEG format so they cannot be edited
(8) Many references are too old, more than 20 years

Recommendation: resubmission after **Major Revision, Rewriting**, and **Adjustments** to fit within the scope of this journal

Heavy Metals Accumulation In Conventional Rice Production

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Abstract

Rice grains produced in the conventional system are a prime source of heavy metal exposure in the human body leading to various health problems. The study aims to asses heavy metals concentration in soil and rice grain produced with the conventional system in Banyumas, Indonesia. Furthermore, it examines the pollution index and bioaccumulation factor. There were 37 samples collected based on soil type, elevation, and irrigation system. The heavy metals level in soil ranged from 2.7 to 39.92 ppm of Pb, 0.11 to 3.01 ppm of Cd, and 4.79 to 61.32 ppm of Cr. Pb and Cr accumulation levels were below the maximum permissible concentration (MPC). A different result in Cd exceeded the MPC in 21 samples. Rice grain shows a high accumulation in Pb (6.85 ppm) and Cr (5.73 ppm) that exceeds the maximum standard. Cd was exceeding in 9 samples with an average of 0.54 ppm. The Pollution Index (PI) was medium in Cd and low in Pb and Cr. The Bioaccumulation Factor (BAF) of heavy metals in rice from the higher is Cd>Pb>Cr with averages of 0.64, 0.53, and 0.30.

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Introduction → The rationale behind this study is unclear; the existing problems to be solved have to be described instead of just presenting theories and research results of others

Rice is the most grown crop in Indonesia as a staple food for the people. The government has consistently pushed national productivity through intensification to meet the needs of the growing population. Banyumas is a region that contributes about 380 tons per year to national rice production (Banyumas, 2017).

The increasing use of chemical fertilizer without sufficient input of organic matter could increase heavy metal accumulation in the soil, such as Lead (Pb) and Cadmium (Cd), then transferred to plants (Merismon et al., 2017). Excessive pesticide application in conventional cultivation leads to heavy metal pollution in soil (Sutrisno and Kuntyastuti, 2015), including Chromium (Cr)(Manurung et al., 2018).

The accumulation of heavy metals such as Pb, Cd, and Cr in soil and food may harm the environment and human health. Heavy metals in soil may affect human health by consuming plant products grown in contaminated soil (Alexander et al., 2006; Zhuang et al., 2009). Consuming food grown in high Pb and Cd concentrations of soil is considered to risk health (Zhuang et al., 2009). Therefore the contamination of heavy metals has been a serious concern to agricultural land management to mitigate environmental disasters and health problems from several diseases. (Alexander et al., 2006; Zhuang et al., 2009; Portier, 2012).

Exceeding the tolerable level, heavy metals pollution in agricultural soil of some regions in Indonesia requires serious consideration (Marselius and Laoli, 2021; Handayani et al., 2022). Several studies demonstrated the danger of heavy metals accumulation due to chemical fertilizer and pesticides application on rice crops (Amelia and Rachmadiarti, 2015), onion (Kusumaningrum et al., 2012), potatoes (Kusdianti et al., 2014), and agricultural soil in general (Suastawan et al., 2016; Komarawidjaja, 2017). According to (Sutrisno and Kuntyastuti, 2015), heavy metal pollutants originate from industrial waste disposal, agrochemical application, and household waste disposal to waterways.

Accumulation of Pb, Cd, or Cr at a certain level causes growth inhibition, yield decline, and premature death of the crop. In contaminated soil, the plant intakes heavy metals and accumulates them in root, leaf, fruit, and grain (Fang and Zhu, 2014). Pollution of Pb, Cd, and Cr in agricultural soil of some regions in Indonesia has surpassed the tolerable limit (Susanti et al., 1992; Sutrisno and Kuntyastuti, 2015). Therefore, it needs serious attention from the farmers, government, and industrial society.

According to the current state, it is essential to evaluate the distribution of heavy metals in the conventional rice production system. The assessment output is a baseline to determine remediation strategies and agricultural development policies. The study aims to assess the accumulation of Pb, Cd, and Cr in soil and rice grain, in addition to the pollution index and bioaccumulation factor in the rice field in Banyumas regency, Indonesia.

Materials and Methods → The followings seem to be sampling and sample analysis methods, not a research method

The study was conducted from July to December 2020. The coverage area of the study is in the Banyumas regency of Indonesia. The study was conducted in several stages, including spatial analysis, survey, and heavy metals analysis.

Spatial analysis was carried out to determine the number and location of samples to be collected. Various maps, including a geographical map, satellite image, irrigation map, and soil map of Banyumas, were overlayed by applying software of geographical information system (GIS) to obtain a homogenous land unit. Sampling sites of Soil and rice were determined based on the number and area of fixed homogeneous land units (Ahadiyat et al., 2021).

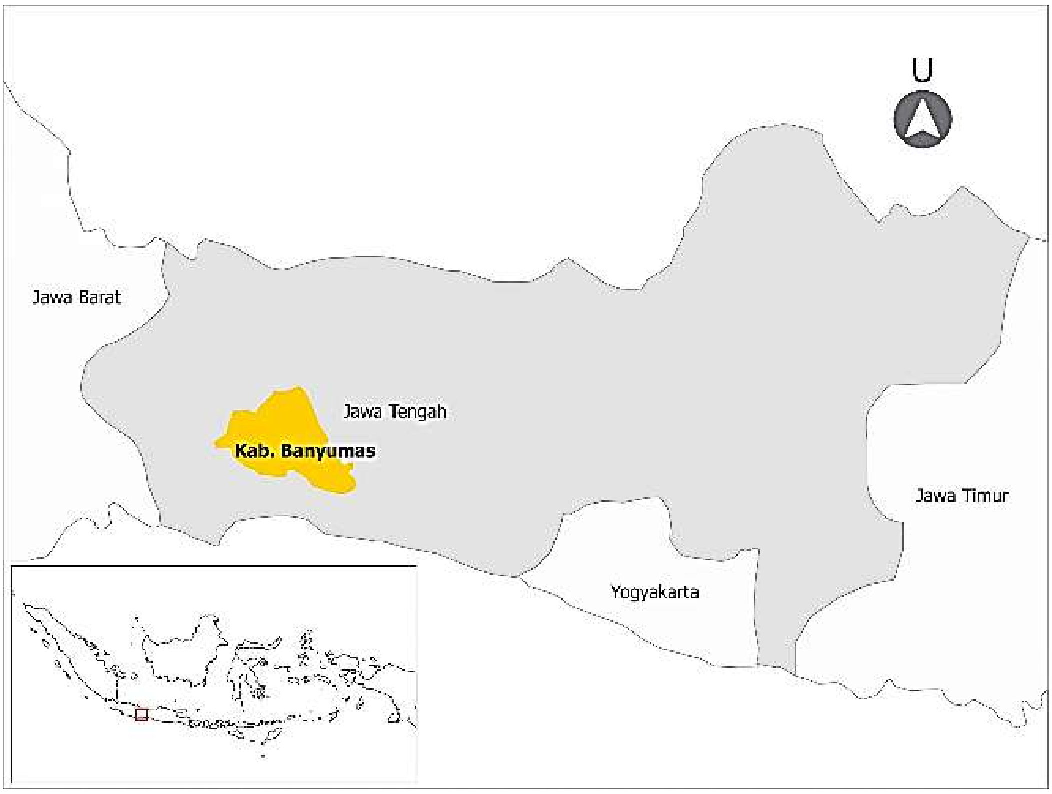

Figure 1. The geographical location of the study area → Legends ahould be in English and written in Times New Roman font (as for the whole texting of this MS)

Soil and rice grain samples from each site were analyzed to assess heavy metals level. The soil was prepared by air dried, crushed, and sieved in 2 mm size before the concentration of Pb, Cd, and Cr were measured. 100 rice grains were prepared for heavy metals level measurement.

Approximately 0,5 grams of the mashed soil and rice sample was put into a porcelain cup, then heated at 300 °C in the kiln for half an hour. The temperature of the furnace was increased to 500 °C so that the sample was scorched (white). Then the cup and its contents were cooled in a closed state and then were added 1 mL of 25% HCl until dissolved and then heated using a hot plate until the HCl evaporated. The sample in the cup that had been evaporated was dissolved with 10 mL of 1 N HCl and filtered. The filtrate was diluted ten times, and the solution was read for Pb, Cd, and Cr metals using an atomic absorption spectrophotometer (SSA) (Bauer et al., 1978; Sulaeman et al., 2005).

To assess the Pb, Cd, and Cr contamination, a pollution index (PI) was calculated using the formula:

$$PI = Cn/ Bn$$

where Cn (mg/kg) is the measured concentration of each heavy metal, and Bn is the background value for each metal (Siti Norbaya et al., 2014).

The PI of each metal was classified as low contamination (IP<1), moderate contamination (1≤ IP≤ 3), high contamination (3<IP≤6), and very high contamination (IP<6) (Gupta et al., 2021). The background values provide a guide to ascertain the presence of anthropogenic effects that could lead to hazardous or beneficial consequences. In environmental geochemistry and health, the background is considered as the natural concentration in environmental material devoid of human influences (Kazapoe and Arhin, 2021). The definition of background (geogenic) values, as opposed to baseline values, is very important in environmental legislation, which fixes, at different levels in various countries, the intervention limits for both organic and inorganic substances in soils (Cicchella et al., 2005). The background value in this research was adopted from the official background value of the Netherlands (Brus et al., 2009).

The heavy metal concentration in soil was standardized to Maximum Permissible Concentrations and Negligible Concentrations for metals released by the National Institute of Public Health and The Environment, Netherlands (Crommentujin et al., 1997), for the official Indonesian standard for maximum concentration of heavy metals in soil was not available yet (Erfandi and Juarsah, 2013).

The heavy metal concentration in rice grain was compared to standards provided by FAO/WHO (General Standard for Contaminants and Toxins in Food and Feed CXS 193-1995, 2019), The National Standardization Body of Indonesia (BSN, 2009), and The National Standard of China (National Food Safety Standard Maximum Levels of Contaminants in Foods, 2017).

The translocation capability of heavy metals from soil to plant was calculated using bioaccumulation factor (BAF) as in the following equation:

BAF = [CR]/[CS]

where CR (mg kg−1) and CS (mg kg−1) represent the concentration(s) of heavy metals in rice grains and soil on a dry-weight basis (Khan et al., 2013; Kong et al., 2018).

Results and Discussion

Sample distribution

→ for a research article, this part should be in the Method section, not in the Result section

Sampling sites were determined using homogenous land units (HLU) generated in the spatial analysis stage based on irrigation type, soil type, and elevation.

Irrigation type:
SI: irrigated soil
ST: rainfed soil
Soil type:
1: Hidromorf alluvial
2: Yellowish-grey alluvial
3: Asociation of grey alluvial and grey brown alluvial
4: Asociation of brown andisol and brown regosol
5: Asociation of andisol
6: Asociation of brown latosol
7: Complex of yellowish-red latosol and yellow-red podzolic
8: Complex yellow-red podzolic
9: Brown latosol
Elevation:
a: 25-250 masl
b: 250-500 masl
c: 500-750 masl
d: 750-1000 masl

There were 37 sampling sites representing 25 types of HLU which resulted from maps overlay (Figures 2 and 3).

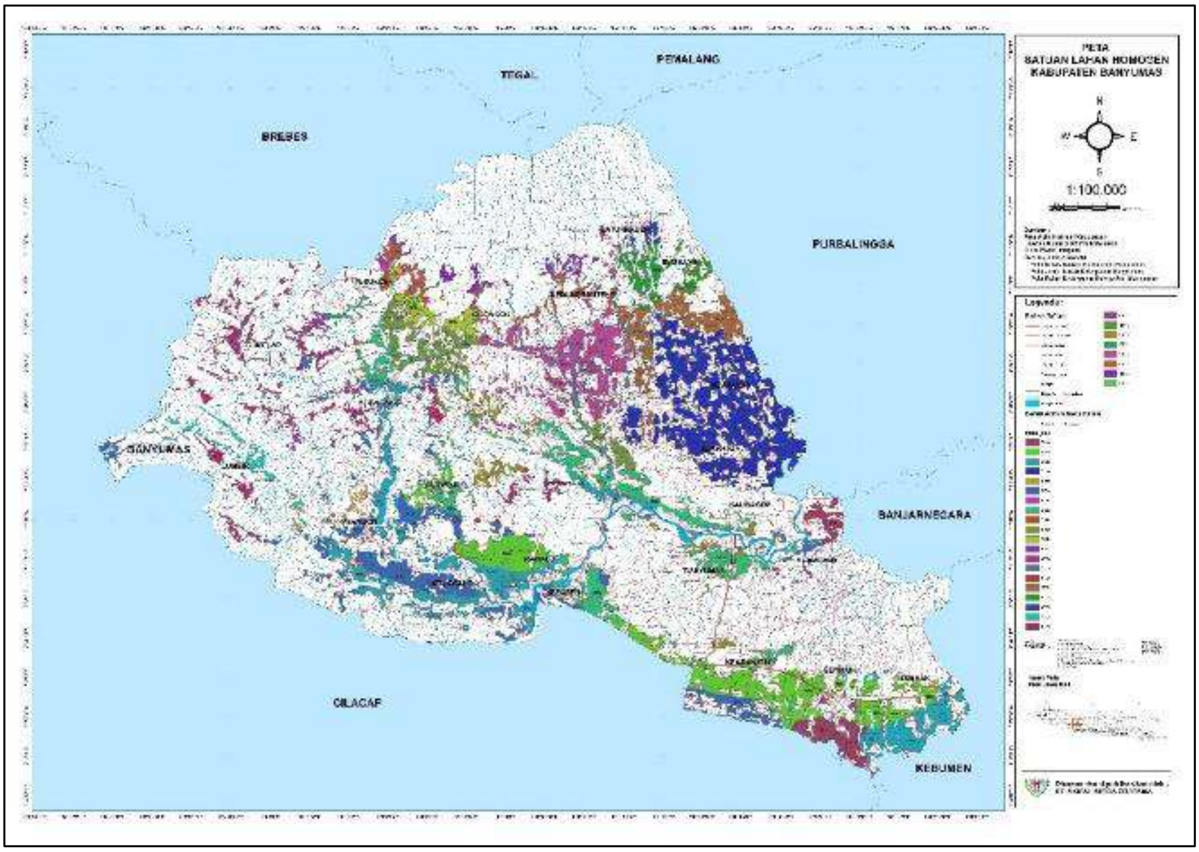

Figure 2. Map of Homogenous Land Unit in Banyumas → the map is blurry; legends are too small

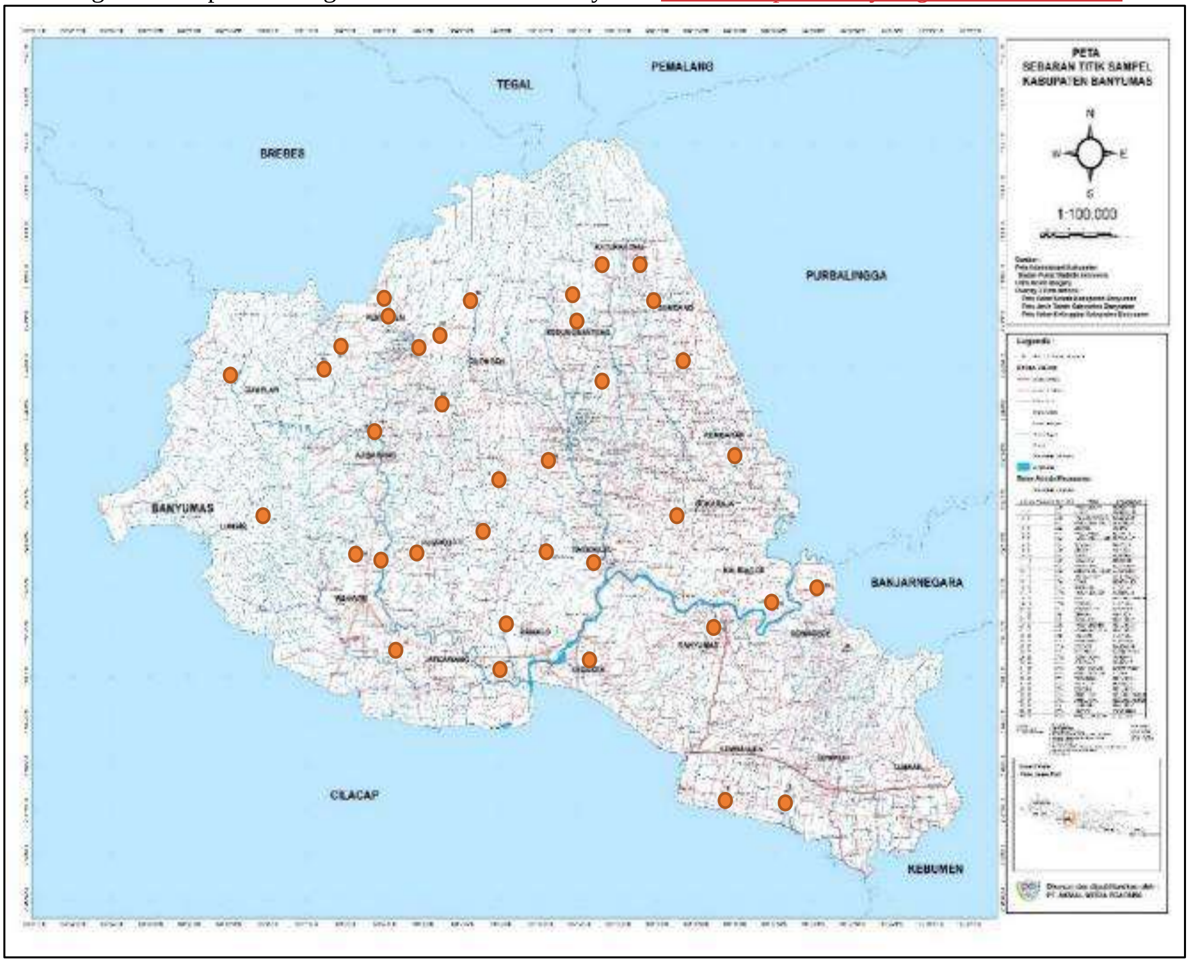

Figure 3. Distribution of sampling sites→ the map is blurry; legends are too small

Table 1. Types of homogenous land units resulted from spatial analysis→ should be presented in an editable table format (word or excel); not a JPEG format

No.	Homogonous Land Unit	Area (ha)	Sampling site
1.	SI1a	1144.9	1
2.	SI2a	4502.24	3
3.	SI3a	4469.52	3
4.	SI5a	5898.93	4
5.	SI6a	230.75	1
6.	SI7a	3662.21	2
7.	SI7b	38.33	1
8.	SI8a	3785.83	3
9.	SI8b	70.66	1
10.	SI9a	1836.5	1
11.	SI9b	696.52	1
12.	ST2a	117.81	1
13.	ST3a	102.26	1
14.	ST4c	27.7	1
15.	ST5a	1743.25	1
16.	ST5b	917.10	1
17.	ST5c	168.18	1
18.	ST6a	453.41	1
19.	ST7a	2276.59	2
20.	ST7b	317.72	1
21.	ST8a	925.54	1
22.	ST8b	151.31	1
23.	ST9a	2524.48	2
24.	ST9b	1017.61	1
25.	ST9c	434.99	1
	TOTAL	***37514.34***	***37***

Accumulation of heavy metals in soil

Accumulation of Pb and Cr in soil ranged from 2.7 – 39.92 ppm and 4.79 – 61.32 ppm. This level was lower than the Maximum Permissible Concentrations and Negligible Concentrations for metals (MPC), according to the National Institute of Public Health and The Environment Netherlands (Crommentujin et al., 1997). A different result was found in Cd that exceeded the maximum standard in 21 sites (Table 2.).

Cd has contaminated most of the ricefield area in Banyumas. There were 21 from 37 sites containing Cd more than the allowable limit. Those sites, according to the homogenous land unit from spatial analysis, represent 22,199.27 ha or equal to 59% of the total ricefield in Banyumas (Figure 7.). The soil excess in Cd may produce contaminated rice which is harmful to consume

Table 2. Heavy metal accumulation in soil and rice grain→ should be presented in an editable table format (word or excel); not a JPEG format

Statistics	Accumulation in soil (ppm)			Accumulation in rice grain (ppm)		
	Pb	Cd	Cr	Pb	Cd	Cr
Sample	37	37	37	37	37	37
Min	2.70	0.11	4.79	0.22	0.10	2.54
Max	39.92	3.01	61.32	32.13	2.16	10.36
Median	26.44	1.80	27.65	3.46	0.24	5.49
Mean	22.64	1.62	27.80	6.85	0.54	5.73
stdev	11.43	0.88	14.27	9.95	0.67	1.93
Maximum Permissible Concentration	140 (1	1.6 (1	100 (1	0.2 (2,4) 0.3 (3	0.4 (2,3) 0.2 (4	1.0 (4

(1 National Institute of Public Health and The Environment Netherlands 1997
(2 SNI 7387:2009
(3 FAO/WHO Codex Alimentarius International Food Standards
(4 National Standard of the People's Republic of China

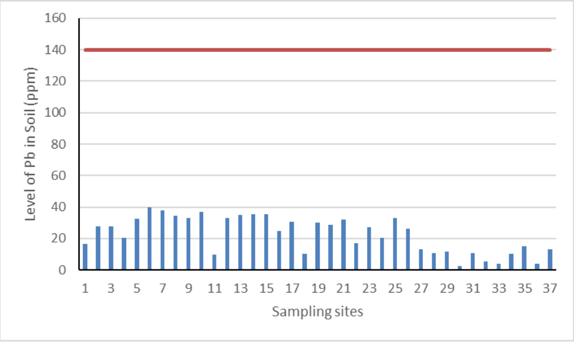

Figure 4. Distribution of Pb levels in sampling sites → Figures should be presented in editable figure format generated from Excel, not in JPEG format

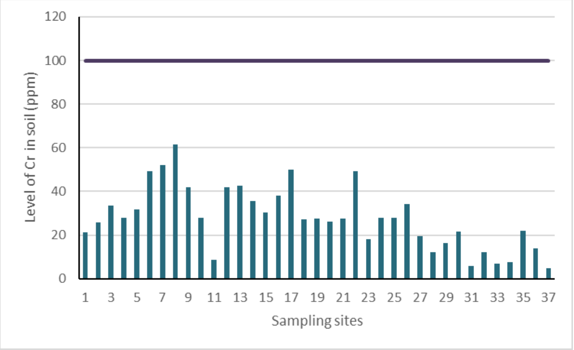

Figure 5. Distribution of Cr levels in sampling sites

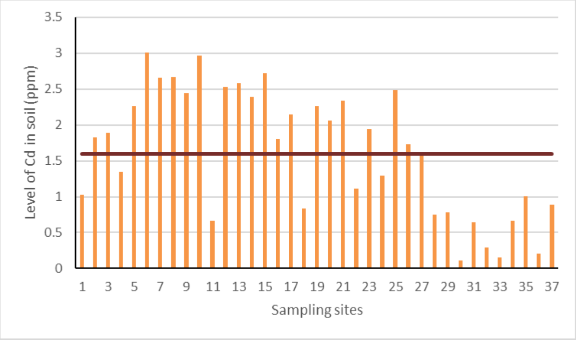

Figure 6. Distribution of Cd levels in sampling sites

The high level of Cd in the soil after harvesting was caused by the intense use of chemical fertilizers and pesticides in rice cultivation. Cd in soil was released by inorganic fertilizers, mainly N and P, which were categorized as the source of contaminants for containing high amounts of heavy metals. For example, SP-36 contains 11 ppm of Cd and 67 ppm of Pb. Pb and Cd in phosphate fertilizer originated from natural rock phosphate as the raw material of the fertilizer. Natural rock phosphate generally consists of large amounts of phosphate, carbonate, and heavy metals (Merismon et al., 2017).

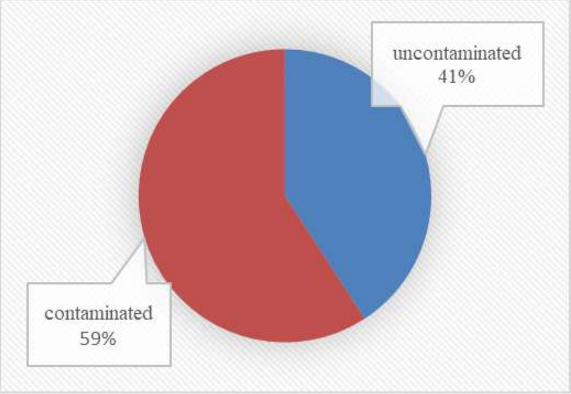

Figure 7. Cd contaminated soil in Banyumas

Banyumas farmers commonly apply chemical fertilizer twice in a growing season at 7-14 and 21-28 days after planting. In pest control, farmers regularly spray pesticides at least twice for a season or more, depending on the pest population. According to the current level of heavy metals in soil due to those chemical inputs, it is crucial to bring a conservative effort to minimize the accumulation in soil. Some organic-based fertilizers and pesticides could be applied as alternative inputs in rice cultivation after conducting soil remediation from heavy metals in Banyumas.

Heavy Metals Accumulation in Rice Grain

Rice grain samples were collected from each site of soil sampling. It was found that Pb, Cd, and Cr accumulation in rice grain ranged from 0.22 – 32.13 ppm, 0.1 – 2.16 ppm, and 2.54 – 10.36 ppm. The level of Pb and Cr in rice grain from each site exceeds the maximum tolerable level according to Indonesian and China National Standards (0,2 ppm). On the other hand, Cd accumulation in rice grain exceeds the highest standard only in 9 of 37 sampling sites (0.4 ppm).

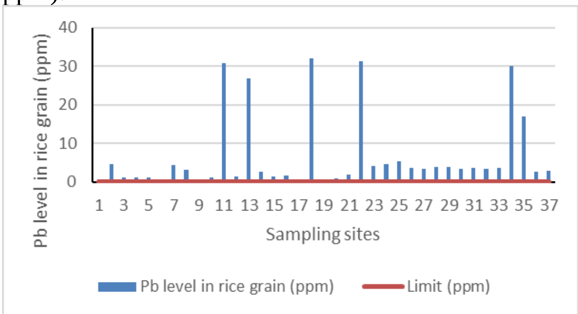

Figure 8. Pb level in rice grain

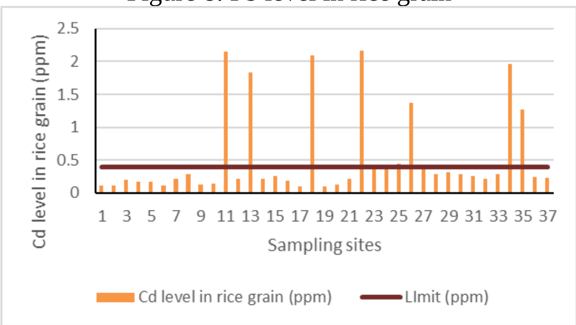

Figure 9. Cd level in rice grain

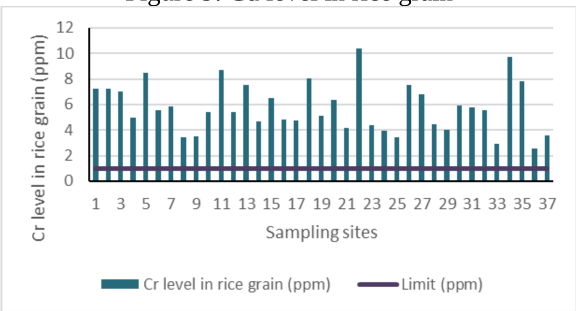

Figure 10. Cr level in rice grain

There was no correlation between heavy metal accumulation in soil and rice grain. The Pb concentration in soil was relatively low, in contrast to the high Pb concentration in rice grain which was beyond the safety limit of food. Pb is a carcinogenic element for humans therefore, its exposure at a chronic level could cause headaches, convulsions, palsy, and nerve damage, especially in children (Guo et al., 2020). The level of Pb accumulation in rice was determined by the cultivation method. Agrochemical applications, including fertilizers and pesticides, contribute to the high accumulation of heavy metals in rice.

The high level of measured Pb in rice was also caused by direct sampling of a rice grain from the field at harvesting without the milling process. Pb level in rice grain could be decreased due to processing to produce white rice. This result is in line with the study of (Norton et al., 2014) and (Tatahmentan et al., 2020), which found a higher level of Pb in rice grain collected from the field than that of white rice from the market.

Cadmium was found to exceed the food safety standard (0.4 ppm) in 9 sites from villages of Kebasen (2.15 ppm), Ajibarang Kulon (1.83 ppm), Beji (2.1 ppm), Kranggan (2.16 ppm), Pasiraman Kidul (0.43 ppm), Kalisari (0.44 ppm), Kasegeran (1.37 ppm), Kaliputih (1.96 ppm) and Semedo (1.27 ppm). The high level of Cd found in rice was suspected to be associated with Cd in contaminated soil of sampling sites which represent 59% of the total area in the Banyumas ricefield.

Intensification of rice cultivation through superior variety and agrochemical input, which was implemented widely since the industrial revolution, has led to national food sufficiency and heavy metals contamination in crop products. The average concentration of Cd in rice grain was 0.5 ppm. This level of Cd shows that Cd accumulation has increased significantly from 40 years ago when (Suzuki et al., 1980) reported that the lowest Cd concentration in Asian rice (0.04 ppm) was cultivated in Java, with 0.02 ppm of the world average (Watanabe et al., 1989). The study by (Shi et al., 2020) also reported that the Cd concentration of Indonesian rice was 0.005 – 0.597 ppm with 0.019 ppm of the global median value.

Cadmium is one of the most mobile and toxic elements in the soil. Cd could be easily transferred into plant tissue and accumulated in harvested part. When Cd enters the human body, it will cause some harmful effects on the lung, cardiovascular, and musculoskeletal systems. The same as Pb concentration, Cd in whole grain (brown rice) is higher than that in white (polished rice) because the outer layer of rice grain accumulates more heavy metals (Sun et al., 2008). Therefore rice milling would lower the Cd concentration in rice (Meharg et al., 2013).

Indonesian national standard for maximum Cr level in rice is not available yet, therefore, this study adopted other countries' standards. Each rice grain from all sample sites contains Cr more than the safety limit of food, according to the National Standard of China (1.0 ppm). Consuming rice grown in soil containing high levels of Cd, Cr, or Zn could seriously risk human health because 24 to 22% of total heavy metals in plant biomass are accumulated in grains (Wang et al., 2003).

Pollution Index (PI)

Most of the measured heavy metals in this study were lower than the background value adopted from the Netherlands' official background value of heavy metals in soil. Pb levels in soil that ranged from 2.7 to 39.92 ppm were lower than the background value (50 ppm). Only one site of soil samples contained Cr more than the background value (55 ppm), in contrast to Cd, which mostly exceeded the background value (Brus et al., 2009).

Table 3. Pollution Index and Bioaccumulation Factor→ should be presented in an editable table format (word or excel); not a JPEG format

Statistics	Pollution index			Bioaccumulation factor		
	Pb	Cd	Cr	Pb	Cd	Cr
Samples	37	37	37	37	37	37
Min	0.05	0.18	0.09	0.01	0.04	0.06
Max	0.80	5.02	1.11	3.11	3.26	1.30
Median	0.53	3.00	0.50	0.16	0.18	0.21
Mean	0.45	2.71	0.51	0.53	0.64	0.30
stdev	0.23	1.46	0.26	0.86	0.92	0.28

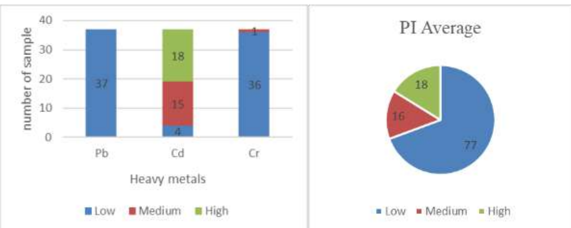

Figure 11. Distribution of pollution index

The evaluation of the pollution index (PI) showed that each site has a low PI for Pb (IP< 1). PI's of Cd were low in 4 sampling sites, medium (1≤ IP≤ 3) in 15 sampling sites, and high (3<IP≤6) in 18 samples. In addition, PI's of Cr were low in 36 sampling sites and medium in 1 sampling site. This result indicates that land management carried out by Banyumas farmers in rice cultivation affects the increased heavy metals accumulation in soil. The anthropogenic contamination of heavy metal in the Banyumas ricefield from lower to higher was Pb< Cr < Cd.

Bioaccumulation factor (BAF)

Bioaccumulation factors (BAF) of rice crops were 0.01 – 3.11 for Pb, 0.04 – 3.26 for Cd, and 0.06 – 1.30 for Cr. Sequentially, heavy metals with higher bioaccumulation factor average to the lower were Cd>Pb>Cr. The bioaccumulation factor of heavy metals in the plant depends on plant and metal species. This fact is in line with what was reported by (Kabata-Pendias, 2010) that cadmium is the most preferred plant to absorb with intensive accumulation, followed by Pb with medium accumulation, and the last is Cr with low accumulation.

Furthermore, there are several sites in Banyumas where the accumulation factor is >1 (6 sites in Pb, 8 sites in Cd, and 2 sites in Cr), and 2 sites show BAF >1 for the three of Pb, Cd, and Cr. According to (Usman et al., 2019), a plant with BAF >1 indicates that the plant is a bio-accumulator and has the potential to be a phytoremediation agent, and BAF<1 shows that the plant is an excluder.

Several sites with BAF>1 might be caused by foliar application of chemical fertilizer or pesticides during cultivation. Metals from aerial sources may increase contamination in a plant through foliar uptake. Heavy metals from foliar uptake could be translocated to other parts of the plant (Kabata-Pendias, 2010) including rice grain. Plant ability to uptake metals from the atmosphere varies among species. Furthermore, (Kabata-Pendias, 2010) reported that some plants were able to uptake large amounts of metals through leaves, including cereal crops which are sensitive to air pollution for Pb and Ni accumulation in leaves

Conclusion → a conclusion should articulate the state of the art of this study. A conclusion is not a summary

The concentration of Pb and Cr in the soil of the Banyumas ricefield were below the maximum permissible concentration (MPC) with an average of 22.64 dan 27.80 ppm. In contrast to Pb and Cr, the concentration of Cd had exceeded MPC in 21 sites which represent 59% of ricefield in Banyumas regency. The accumulation of Pb and Cr in rice grain exceeded the safety limit with an average of 6.85 dan 5.73 ppm. Furthermore, Cd in rice grain exceeded the limit in 9 of 37 sample sites. Pb and Cr showed a low pollution index (PI), while Cd was moderate. Bioaccumulation factor (BAF) of heavy metals in rice crops sequentially from the highest to the lowest was Cd>Pb>Cr with averages of 0.64, 0.53, and 0.30. These conditions of heavy metals in the rice production system should become a baseline to arrange strategies or policies in protecting people from heavy metal exposure from rice. Minimizing the use of chemical fertilizer and more organic material input could become supporting action to regenerate the contaminated soil, in addition to remediation practices such as bioremediation or phytoremediation.

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Mapping heavy metals accumulation in conventional rice farming system at Banyumas Regency of Central Java, Indonesia

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Abstract

Rice grains produced in the conventional system are a prime source of heavy metal exposure in the human body leading to various health problems. The objective of this study was to asses heavy metals concentration in soil and rice grain under conventional rice farming system in Banyumas, Indonesia, the pollution index and bioaccumulation factor. There were thirty seven samples collected based on soil type, elevation, and irrigation system. The heavy metals level in soil ranged from 2.7 to 39.92 ppm of Pb, 0.11 to 3.01 ppm of Cd, and 4.79 to 61.32 ppm of Cr. Pb and Cr accumulation levels were below the maximum permissible concentration (MPC). A different result in Cd exceeded the MPC in 56.75% sampling sites. Rice grain showed a high accumulation in Pb (6.85 ppm) and Cr (5.73 ppm) that exceeds the maximum standard. Cd was exceeding in 24.32% sampling sites with an average of 0.54 ppm. The Pollution Index (PI) was medium in Cd and low in Pb and Cr. The Bioaccumulation Factor (BAF) of heavy metals in rice was Cd>Pb>Cr with averages of 0.64, 0.53, and 0.30, respectively. High correlation resulted on Pb and Cd in soils and rice grain. Remediation is must done to reduce the heavy metals accumulation in soils and rice grain for environmental and health safety to prevent the further contamination.

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Introduction

Rice is the most grown crop in Indonesia as a staple food for the people. The government has consistently pushed national productivity through intensification to meet the needs of the growing population. In Indonesia, most farmers practices conventional farming systems while total organic agricultural land in Indonesia was only 0.6 percent (208,042 hectares) in 2017 (Willer and Lernoud, 2019). The worldwide application of pesticides has estimated at approximately 6 billion pounds (Atwood and Paisley-Jones, 2017). This number keeps increasing, especially in developing countries. In Indonesia, detailed information on pesticide uses in agricultural activities, according to rice production, is still limited (Mariyono et al., 2018) . Consumption of NPK fertilizer in Indonesia is about 10-11 million tons (BPS Indonesia, 2021), and in Central Jawa about 400 tons (BPS Jawa Tengah, 2021). Banyumas is applied NPK fertilizer of about 7 tons with 371,827.52 tons of rice production in 2020 (BPS Banyumas, 2021). Even though the application of synthetics pesticide and fertilizers improves yield production but impact of its application on the environment such as in agricultural lands is unexploited.

The high application of chemical fertilizer could increase heavy metal accumulation in the soil, such as Lead (Pb) and Cadmium (Cd) (Budianta et al., 2016) then transferred to plants (Merismon, et al., 2017). Also, excessive pesticide application in conventional cultivation leads to heavy metal pollution in soil (Sutrisno and Kuntyastuti, 2015), including Chromium (Cr) (Manurung et al., 2018). The contamination of heavy metals has been a serious concern to agricultural land management to mitigate environmental disasters and health problems from several diseases (Faroon et al., 2012).The accumulation of heavy metals such as Pb, Cd, and Cr in soil and food may harm the environment and human health. Heavy metals in soil may affect human health by consuming plant products grown in contaminated soil (Gupta et al., 2021). Consuming food grown in high Cd concentrations of soil is considered to risk health (Ekere et al., 2020).

Exceeding the tolerable level, heavy metals pollution in agricultural soil of some regions in Indonesia requires serious consideration (Laoli et al., 2021; Handayani et al., 2022). Several studies demonstrated the danger of heavy metals accumulation due to chemical fertilizer and pesticides application on onion (Kusumaningrum et al., 2012), rice (Amelia et al., 2015), potatoes (Manurung et al., 2018), and agricultural soil in general Suastawan et al., 2015; Komarawidjadja, 2017). According to (Sutrisno and Kuntyastuti, 2015), heavy metal pollutants originate from industrial waste disposal, agrochemical application, and household waste disposal to waterways.

Accumulation of Pb, Cd or Cr at a certain level causes growth inhibition, yield decline, and premature death of the crop. In contaminated soil, the plant intakes heavy metals and accumulates them in root, leaf, fruit, and grain (Fang and Zhu, 2014). Pollution of Pb, Cd, and Cr in agricultural soil of some regions in Indonesia has surpassed the tolerable limit (Sutrisno and Kuntyastuti, 2015). Therefore, it needs serious attention from the farmers, government, and industrial society. According to the current state, it is essential to evaluate the distribution of heavy metals in the conventional rice production system. The assessment output is a baseline to determine remediation strategies and agricultural development policies. The objective of this study was to map the accumulation of Pb, Cd, and Cr in soil and rice grain, the pollution index, and bioaccumulation factor in the rice field in Banyumas Regency of Central Jawa, Indonesia.

Materials and Methods

The study area

The coverage area of the study was in the Banyumas regency of Indonesia. The geographic positions of the study sites are spread over 108º39'17”-109º27'15” E, and 7º15'5”-7º3710”' S, with a total area of 1.335,30 km². Banyumas Regency consists of 27 districts, with an average rainfall of 4,354 mm per year. Land uses in Banyumas Regency consist of 31,896 ha (24.03 %) of paddy field, 68,088 ha (51.39 %) of agricultural land, and 32,774 (24,69%) of non-farmland, dominated by intensive rice farming management (BPS Kabupaten Banyumas, 2021). According to the USDA Soil Taxonomy Classification System, the soils in Banyumas Regency can be grouped into Entisols, Inceptisols, Litosol, Regosol, Oxisol, and Ultisol.

Table 1. Soil sampling location

No. sampling site	Location (village)	No. sampling site	Location (village)
1	Karangsoka	20	Somakaton
2	Pliken	21	Cikakak
3	Kalisogra Wetan	22	Kranggan
4	Karangkedawung	23	Karangkemiri
5	Lebeng	24	Pasiraman Kidul
6	Pesawahan	25	Kalisari
7	Bangsa	26	Kasegeran
8	Menganti	27	Srowot
9	Cikakak	28	Ketenger
10	Notog	29	Kebanggan
11	Kebasen	30	Kotayasa
12	Pekunden	31	Karangsalam
13	Ajibarang Kulon	32	Karanggayam
14	Tinggarjaya	33	Petahunan
15	Sirau	34	Kaliputih
16	Gumelar	35	Semedo
17	Karangendep	36	Windujaya
18	Beji	37	Windujaya
19	Pejogol		

Spatial analysis and soil sampling

Spatial analysis was carried out to determine the number and location of samples to be collected. Various maps, including a geographical map, satellite image, irrigation map, and soil map of Banyumas, were overlayed by applying software of geographical information system (GIS) of QGIS Version 3.22 to obtain a homogenous land unit. Sampling sites of Soil and rice were determined based on the number and area of fixed homogeneous land units. Descriptive method was applied in this study based on survey by explorative approach. The site sample was determined through purposive sampling based on the similarity of the soil mapping unit resulted from overlay soil types, elevation and irrigation types (Fig. 1 and 2).

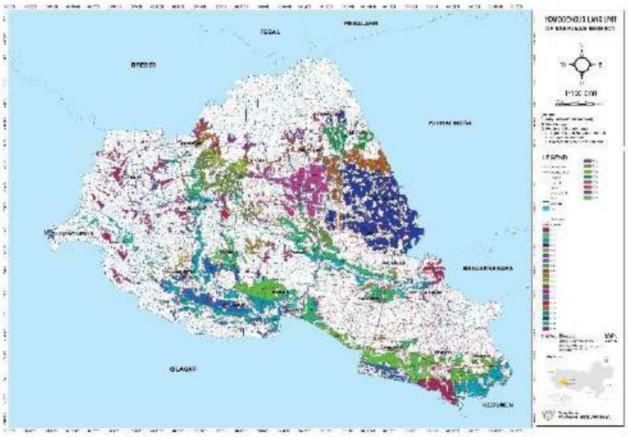

Figure 1. Map of overlay soil types, elevation and irrigation types in Banyumas

Figure 2. Distribution of sampling sites

Assessment of heavy metals contamination

Soil and rice grain samples from each site were analyzed to assess heavy metals level. The soil was prepared by air dried, crushed, and sieved in 2 mm size before the concentration of Pb, Cd, and Cr were measured. 100 rice grains were prepared for heavy metals level measurement.

Approximately 0,5 grams of the mashed soil and rice sample was put into a porcelain cup, then heated at 300 °C in the kiln for half an hour. The temperature of the furnace was increased to 500 °C so that the sample was scorched (white). Then the cup and its contents were cooled in a closed state and then were added 1 mL of 25% HCl until dissolved and then heated using a hot plate until the HCl evaporated. The sample in the cup that had been evaporated was dissolved with 10 mL of 1 N HCl and filtered. The filtrate was diluted ten times, and the solution was read for Pb, Cd, and Cr metals using an atomic absorption spectrophotometer (SSA) (Bauer et al., 1978).

To assess the Pb, Cd, and Cr contamination, a pollution index (PI) was calculated using the formula: PI = Cn/ Bn, where Cn (mg/kg) is the measured concentration of each heavy metal, and Bn is the background value for each metal (Amin et al., 2019). The PI of each metal was classified as low contamination ($IP<1$), moderate contamination ($1\leq IP\leq 3$), high contamination ($3<IP\leq 6$), and very high contamination ($IP<6$) (Gupta et al., 2021). The background values provide a guide to ascertain the presence of anthropogenic effects that could lead to hazardous or beneficial consequences. In environmental geochemistry and health, the background is considered as the natural concentration in environmental material devoid of human influences (Kazapoe and Arhin, 2021).

The official Indonesian standard for maximum concentration of heavy metals in soil was not available yet. The heavy metal concentration in soil was standardized to Maximum Permissible Concentrations and Negligible Concentrations for metals (Vodyanitskii, 2016). The heavy metal concentration in rice grain was compared to standards provided by FAO/WHO (IFS, 2019), requirements for heavy metal contamination in food of Indonesia (BPOM, 2022), and The National Standard of China (Woolsey and Bugang, 2010). The translocation capability of heavy metals from soil to plant was calculated using bioaccumulation factor (BAF) as equation BAF = [CR]/[CS], where CR (mg kg^{-1}) and CS (mg kg^{-1}) represent the concentration(s) of heavy metals in rice grains and soil on a dry-weight basis (Khan et al., 2013; Kong et al., 2018).

Statistical analysis

Kolmogorov Smirnov test was used to analyzed the normality of data. One Way Analysis of Variance (ANOVA) by IBM SPSS Statistics version 26 was applied for statistical analysis of the heavy metals content in study site area. Heavy metals content in soil and rice grain were performed by Pearson correlation analysis.

Results and Discussion

Accumulation of heavy metals in soil

Accumulation of Pb and Cr in soil ranged from 2.7 – 39.92 ppm and 4.79 – 61.32 ppm. This level was lower than the Maximum Permissible Concentrations and Negligible Concentrations for metals (MPC), according to the standard for content of heavy metals in soils (Vodyanitskii, 2016). A different result was found in Cd that exceeded the maximum standard in 21 sites. Cd has contaminated most of the ricefield area in Banyumas. There were 21 from 37 sites containing Cd more than the allowable limit. Those sites, according to the homogenous land unit from spatial analysis, represent 22,199.27 ha or equal to 59% of the total ricefield in Banyumas. In general, accumulation Pb and Cr in soils was lower than Cd (Table 1).

Table 1. Heavy metal accumulation in soil and rice grain

	Accumulation in soil (ppm)			Accumulation in rice grain (ppm)		
Statistics	Pb	Cd	Cr	Pb	Cd	Cr
Sample	37	37	37	37	37	37
Min	2.70	0.11	4.79	0.22	0.10	2.54
Max	39.92	3.01	61.32	32.13	2.16	10.36
Median	26.44	1.80	27.65	3.46	0.24	5.49
Mean	22.64	1.62	27.80	6.85	0.54	5.73
stdev	11.43	0.88	14.27	9.95	0.67	1.93
Maximum Permissible Concentration	140 [1]	1.6 [1]	100 [1]	0.2 [2,4] 0.3 [3]	0.4 [2,3] 0.2 [4]	1.0 [4]

(1) Standard for content of heavy metals in soils of some states (Vodyanitskii, 2016)
(2) SNI (2009) and BPOM (2022)
(3) FAO/WHO Codex Alimentarius International Food Standards (IFS, 2019)
(4) National Standard of the People's Republic of China (Woolsey and Bugang, 2010)

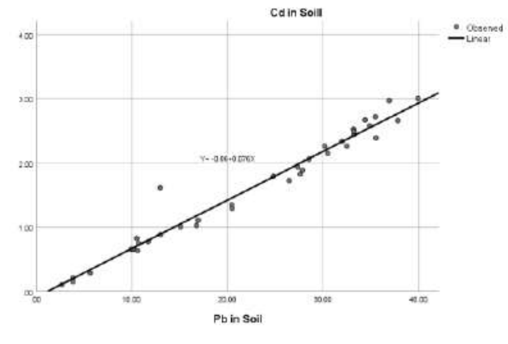

Figure 3. Correlation accumulation Pb and Cd in soil

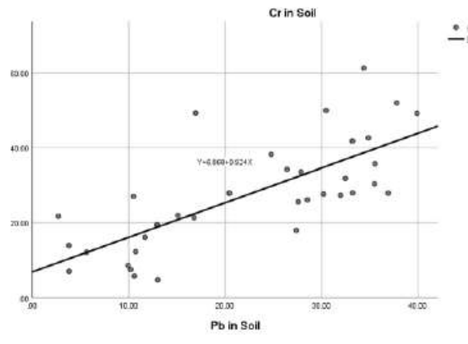

Figure 4. Correlation accumulation Pb and Cr in soil

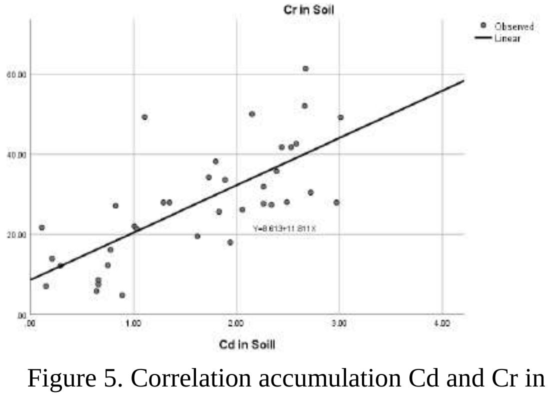

Figure 5. Correlation accumulation Cd and Cr in soil

The high level of Cd in the soil was caused by the intense use of chemical fertilizers and pesticides in rice cultivation. Cd in soil was released by inorganic fertilizers, mainly N and P, which were categorized as the source of contaminants for containing high amounts of heavy metals. For example, SP-36 contains 11 ppm of Cd and 67 ppm of Pb. Pb and Cd in phosphate fertilizer originated from natural rock phosphate as the raw material of the fertilizer. Natural rock phosphate generally consists of large amounts of phosphate, carbonate, and heavy metals (Merismon et al., 2017). Therefore, there has high correlation between accumulation of Pb and Cd in soils (Figure 3). But, there has no correlation between Pb and Cr, and Cd and Cr according to its accumulation in soil (Figure 4 and 5). This indicated that the presence of Pb and Cd in the soil was in the same proportions from ther source of synthetic of fertilizers and pesticides. Some studies indicated that accumulation of Pb and Cd had high correlation (Merismon et al., 2017; Guo et al., 2020; Tatahmentan et al., 2020).

Banyumas farmers commonly apply chemical fertilizer twice in a growing season at 7-14 and 21-28 days after planting. In pest control, farmers regularly spray pesticides at least twice for a season or more, depending on the pest population. According to the current level of heavy metals in soil due to those chemical inputs, it is crucial to bring a conservative effort to minimize the accumulation in soil. Some organic-based fertilizers and pesticides could be applied as alternative inputs in rice cultivation after conducting soil remediation from heavy metals in Banyumas.

Heavy Metals Accumulation in Rice Grain

Rice grain samples were collected from each site of soil sampling. It was found that Pb, Cd, and Cr accumulation in rice grain ranged from 0.22 – 32.13 ppm, 0.1 – 2.16 ppm, and 2.54 – 10.36 ppm, respectively. The level of Pb and Cr in rice grain from each site exceeds the maximum tolerable level according to accumulation of heavy metal standards (0.2 – 0.4 ppm). On the other hand, Cd accumulation in rice grain exceeds the highest standard only in 9 of 37 sampling sites (> 0.4 ppm) (Table 1).

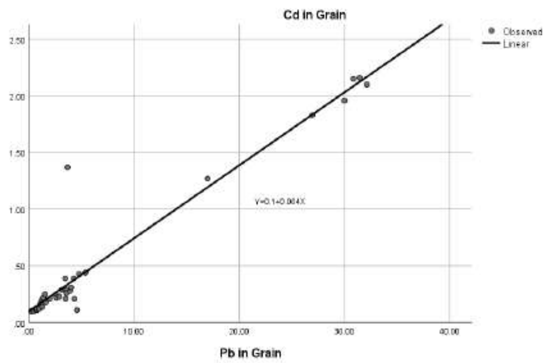

Figure 6. Correlation accumulation Pb and Cd in rice grain.

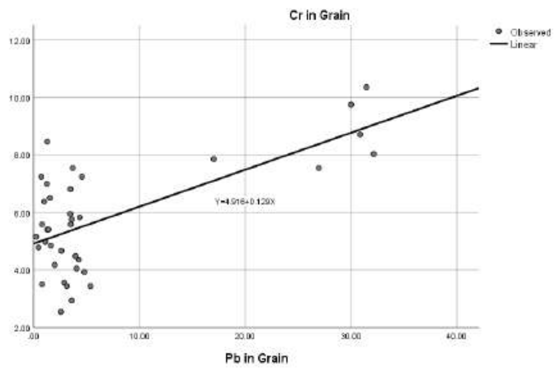

Figure 7. Correlation accumulation Pb and Cr in rice grain.

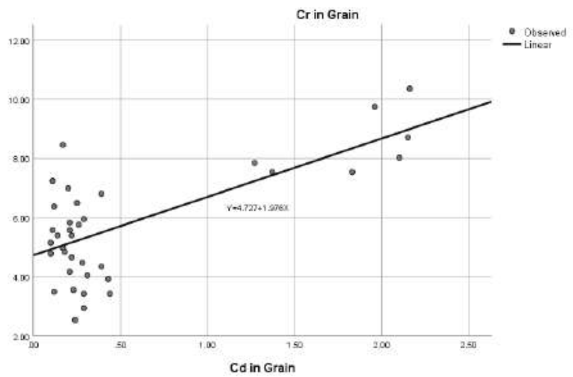

Figure 8. Correlation accumulation Cd and Cr in rice grain

There had correlation between heavy metal accumulation of Pb and Cd in rice grain (Figure 6). The Pb concentration in soil was relatively low, in contrast to the high Pb concentration in rice grain which was beyond the safety limit of food. Pb is a carcinogenic element for humans therefore, its exposure at a chronic level could cause headaches, convulsions, palsy, and nerve damage, especially in children (Guo et al., 2020). The level of Pb accumulation in rice was determined by the cultivation method. Agrochemical applications, including fertilizers and pesticides, contribute to the high accumulation of heavy metals in rice.

The high level of measured Pb in rice was also caused by direct sampling of a rice grain from the field at harvesting without the milling process. Pb level in rice grain could be decreased due to processing to produce white rice. This result is in line with the study of (Norton et al., 2014) and (Tatahmentan et al., 2020), which found a higher level of Pb in rice grain collected from the field than that of white rice from the market.

Cadmium was found to exceed the food safety standard (0.4 ppm) in 9 sites from villages of Kebasen (2.15 ppm), Ajibarang Kulon (1.83 ppm), Beji (2.1 ppm), Kranggan (2.16 ppm), Pasiraman Kidul (0.43 ppm), Kalisari (0.44 ppm), Kasegeran (1.37 ppm), Kaliputih (1.96 ppm) and Semedo (1.27 ppm). The high level of Cd found in rice was suspected to be associated with Cd in contaminated soil of sampling sites which represent 59% of the total area in the Banyumas ricefield.

Intensification of rice cultivation through superior variety and agrochemical input, which was implemented widely since the industrial revolution, has led to national food sufficiency and heavy metals contamination in crop products. The average concentration of Cd in rice grain was 0.5 ppm. Shi et al. (2020) reported that the Cd concentration of Indonesian rice was 0.005 – 0.597 ppm with 0.019 ppm of the global median value. Cadmium is one of the most mobile and toxic elements in the soil. Cd could be easily transferred into plant tissue and accumulated in harvested part. When Cd enters the human body, it will cause some harmful effects on the lung, cardiovascular, and musculoskeletal systems. Therefore rice milling would lower the Cd concentration in rice (Meharg et al., 2013).

Indonesian national standard for maximum Cr level in rice is not available yet, therefore, this study adopted other countries' standards. Each rice grain from all sample sites contains Cr more than the safety limit of food, according to the National Standard of China (1.0 ppm). Consuming rice grown in soil containing high levels of Cd, Cr, or Zn could seriously risk human health because 24 to 22% of total heavy metals in plant biomass are accumulated in grains (Kong et al., 2018). Therefore, The soil excess in Cd may produce contaminated rice which is harmful to consume.

Pollution Index (PI)

Most of the measured heavy metals in this study were lower than the preceeding value adopted from the standard for content of heavy metals in soil. Pb levels in soil that ranged from 2.7 to 39.92 ppm were lower than the preceeding value (50 ppm). Only one site of soil samples contained Cr more than the preceeding value (55 ppm), in contrast to Cd, which mostly exceeded the preceeding value.

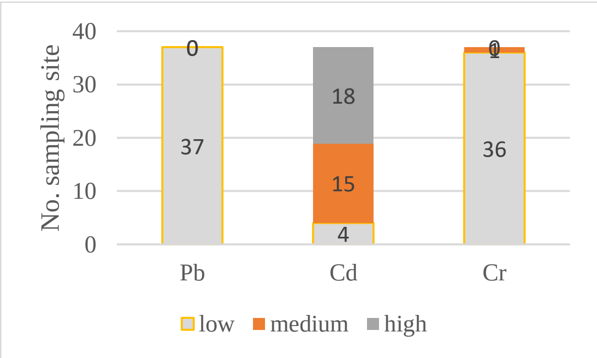

Figure 9. Distribution sampling site of pollution index

Based on the study was shown in Figure 9 that the pollution index (PI) in each site had a low PI for Pb (IP< 1). PI's of Cd were low in 4 sampling sites, medium (1≤ IP≤ 3) in 15 sampling sites, and high (3<IP≤6) in 18 samples. In addition, PI's of Cr were low in 36 sampling sites and medium in 1 sampling site. This result indicates that land management carried out by Banyumas farmers in rice cultivation affects the increased heavy metals accumulation in soil. The anthropogenic contamination of heavy metal in the Banyumas ricefield from lower to higher was Pb< Cr < Cd.

Bioaccumulation factor (BAF)

Bioaccumulation factors (BAF) of rice crops were 0.01 – 3.11 for Pb, 0.04 – 3.26 for Cd, and 0.06 – 1.30 for Cr. Sequentially, heavy metals with higher bioaccumulation factor average to the lower were Cd>Pb>Cr. The bioaccumulation factor of heavy metals in the plant depends on plant and metal species. This fact is in line with what was reported by (Kabata-Pendias, 2011) that cadmium is the most preferred plant to absorb with intensive accumulation, followed by Pb with medium accumulation, and the last is Cr with low accumulation.

Furthermore, there are several sites in Banyumas where the accumulation factor is >1 (6 sites in Pb, 8 sites in Cd, and 2 sites in Cr), and 2 sites show BAF >1 for the three of Pb, Cd, and Cr. According to (Usman et al., 2019), a plant with BAF >1 indicates that the plant is a bio-accumulator and has the potential to be a phytoremediation agent, and BAF<1 shows that the plant is an excluder.

Several sites with BAF>1 might be caused by foliar application of chemical fertilizer or pesticides during cultivation. Metals from aerial sources may increase contamination in a plant through foliar uptake. Heavy metals from foliar uptake could be translocated to other parts of the plant (Kabata-Pendias, 2011) including rice grain. Plant ability to uptake metals from the atmosphere varies among species. Furthermore, (Kabata-Pendias, 2011) reported that some plants were able to uptake large amounts of metals through leaves, including cereal crops which are sensitive to air pollution for Pb and Ni accumulation in leaves

Conclusion

The concentration of Pb and Cr in the soil of the Banyumas ricefield were below the maximum permissible concentration (MPC) but the concentration of Cd had exceeded MPC in 21 sites which represent 59% of ricefield in Banyumas regency. The accumulation of Pb, Cd and Cr in rice grain exceeded the safety limit in mostly sample sites. Pb and Cr showed a low pollution index (PI), while Cd was moderate. Bioaccumulation factor (BAF) of heavy metals in rice crops sequentially from the highest to the lowest was Cd>Pb>Cr Correlation of Pb and Cd was high according to accumulation in soil and rice grains.

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to me

We inform you that the revised manuscript entitled "Mapping heavy metals accumulation in conventional rice farming system at Banyumas Regency of Central Java, Indonesia" will be ACCEPTED for publication in the Journal of Degraded and Mining Lands Management. Prior to publication, the draft of the attached article needs to be slightly corrected,

(1) Figures 1 and 2, (blurred and legend too small, unreadable),

(2) Figures 3, 4, 5, 6, 7, and 8, (the letters / legends are too small, and must be replaced with the Times New Roman font, the same used throughout the text).

We inform you that the article mentioned above will be published in the JDMLM vol 10 no 4 (1 July 2023), because the JDMLM vol 4 no 3 (1 April 2023) edition has reached a maximum of 24 articles on 31 January 2023. Apart from the 24 ready-to-published articles, as of February 4, 2023 there are 47 manuscripts that are currently "under review" and "waiting list", while JDMLM is only able to publish 24 articles per issue (January, April, July, October).

If you are willing to have your article published in JDMLM vol 10 no 4 (July 1, 2023), then we will immediately send you LoA, Galley Proof, and APC payment invoice (after the Figures in the attached draft are corrected). However, if you do not want your article to be published in JDMLM vol 10 no 4 (1 July 2023), then you are welcome to "withdraw" your article.

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REVIEW 2

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Research Article

Mapping heavy metals accumulation in conventional rice farming system at Banyumas Regency of Central Java, Indonesia

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Abstract

Rice grains produced in the conventional system are a prime source of heavy metal exposure in the human body leading to various health problems. The objective of this study was to assess heavy metals concentration in soil and rice grain under the conventional rice farming system in Banyumas, Indonesia, the pollution index, and the bioaccumulation factor. There were thirty seven samples collected based on soil type, elevation, and irrigation system. The heavy metals level in soil ranged from 2.7 to 39.92 ppm of Pb, 0.11 to 3.01 ppm of Cd, and 4.79 to 61.32 ppm of Cr. Pb and Cr accumulation levels were below the maximum permissible concentration (MPC). A different result in Cd exceeded the MPC in 56.75% of sampling sites. Rice grain showed a high accumulation in Pb (6.85 ppm) and Cr (5.73 ppm) that exceeds the maximum standard. Cd exceeded 24.32% of sampling sites with an average of 0.54 ppm. The Pollution Index (PI) was medium in Cd and low in Pb and Cr. The Bioaccumulation Factor (BAF) of heavy metals in rice was Cd>Pb>Cr with averages of 0.64, 0.53, and 0.30, respectively. A high correlation resulted in Pb and Cd in soils and rice grains. Remediation is must done to reduce the heavy metals accumulation in soils and rice grains for environmental and health safety to prevent further contamination.

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Introduction

Rice is the most grown crop in Indonesia as a staple food for the people. The government has consistently pushed national productivity through intensification to meet the needs of the growing population. In Indonesia, most farmers practice conventional farming systems, while total organic agricultural land in Indonesia was only 0.6 percent (208,042 hectares) in 2017 (Willer and Lernoud, 2019). The worldwide application of pesticides has been estimated at approximately 6 billion pounds (Atwood and Paisley-Jones, 2017). This number keeps increasing, especially in developing countries. In Indonesia, detailed information on pesticide uses in agricultural activities, according to rice production, is still limited (Mariyono et al., 2018). Consumption of NPK fertilizer in Indonesia is about 10-11 million tons (BPS Indonesia, 2021), and in Central Jawa, about 400 tons (BPS Jawa Tengah, 2021). Banyumas farmers applied NPK fertilizer of about 7 tons with 371,827.52 tons of rice production in 2020 (BPS Banyumas, 2021). Even applying of synthetics pesticide and fertilizers improves yield production, the impact of its application on the environment, such as in agricultural lands, is unexploited.

The high application of chemical fertilizer could increase heavy metal accumulation in the soil, such as Lead (Pb) and Cadmium (Cd) (Budianta et al., 2016),

then transferred to plants (Merismon et al., 2017). Also, excessive pesticide application in conventional cultivation leads to heavy metal pollution in soil (Sutrisno and Kuntyastuti, 2015), including Chromium (Cr) (Manurung et al., 2018). The contamination of heavy metals has been a serious concern to agricultural land management to mitigate environmental disasters and health problems from several diseases (Faroon et al., 2012). The accumulation of heavy metals such as Pb, Cd, and Cr in soil and food may harm the environment and human health. Heavy metals in soil may affect human health by consuming plant products grown in contaminated soil (Gupta et al., 2021). Consuming food grown in high Cd concentrations of soil is considered to risk health (Ekere et al., 2020).

Exceeding the tolerable level, heavy metals pollution in agricultural soil of some regions in Indonesia requires serious consideration (Laoli et al., 2021; Handayani et al., 2022). Several studies demonstrated the danger of heavy metals accumulation due to chemical fertilizer and pesticides application on onions (Kusumaningrum et al., 2012), rice (Amelia et al., 2015), potatoes (Manurung et al., 2018), and agricultural soil in general Suastawan et al., 2015; Komarawidjadja, 2017). According to (Sutrisno and Kuntyastuti, 2015), heavy metal pollutants originate from industrial waste disposal, agrochemical application, and household waste disposal to waterways.

Accumulation of Pb, Cd, or Cr at a certain level causes growth inhibition, yield decline, and premature death of the crop. In contaminated soil, the plant intakes heavy metals and accumulates them in root, leaf, fruit, and grain (Fang and Zhu, 2014). Pollution of Pb, Cd, and Cr in agricultural soil of some regions in Indonesia has surpassed the tolerable limit (Sutrisno and Kuntyastuti, 2015). Therefore, it needs serious attention from the farmers, government, and industrial society. According to the current state, it is essential to evaluate the distribution of heavy metals in the conventional rice production system. The assessment output is a baseline to determine remediation strategies and agricultural development policies.

The objective of this study was to map the accumulation of Pb, Cd, and Cr in soil and rice grain, the pollution index, and the bioaccumulation factor in the rice field in the Banyumas Regency of Central Jawa, Indonesia.

Materials and Methods

The study area

The coverage area of the study was in the Banyumas regency of Indonesia. The geographic positions of the study sites are spread over 108º39'17”-109º27'15” E, and 7º15'5”-7º37'10” S, with a total area of 1.335,30 km². Banyumas Regency consists of 27 districts, with an average rainfall of 4,354 mm per year. Land uses in Banyumas Regency consist of 31,896 ha (24.03%) of paddy field, 68,088 ha (51.39%) of agricultural land, and 32,774 (24,69%) of non-farmland, dominated by intensive rice farming management (BPS Kabupaten Banyumas, 2021). According to the USDA Soil Taxonomy Classification System, the soils in Banyumas Regency can be grouped into Entisols, Inceptisols, Litosol, Regosol, Oxisol, and Ultisol. Soil sampling locations are presented in Table 1.

Table 1. Soil sampling location.

No. sampling site	Location (village)	No. sampling site	Location (village)
1	Karangsoka	20	Somakaton
2	Pliken	21	Cikakak
3	Kalisogra Wetan	22	Kranggan
4	Karangkedawung	23	Karangkemiri
5	Lebeng	24	Pasiraman Kidul
6	Pesawahan	25	Kalisari
7	Bangsa	26	Kasegeran
8	Menganti	27	Srowot
9	Cikakak	28	Ketenger
10	Notog	29	Kebanggan
11	Kebasen	30	Kotayasa
12	Pekunden	31	Karangsalam
13	Ajibarang Kulon	32	Karanggayam
14	Tinggarjaya	33	Petahunan
15	Sirau	34	Kaliputih
16	Gumelar	35	Semedo
17	Karangendep	36	Windujaya
18	Beji	37	Windujaya
19	Pejogol		

Spatial analysis and soil sampling

Spatial analysis was carried out to determine the number and location of samples to be collected. Various maps, including a geographical map, satellite image, irrigation map, and soil map of Banyumas, were overlayed by applying software of geographical information system (GIS) of QGIS Version 3.22 to obtain a homogenous land unit. Sampling sites of soil and rice were determined based on the number and area of fixed homogeneous land units. The descriptive method was applied in this study based on a survey by explorative approach. The site sample was determined through purposive sampling based on the similarity of the soil mapping unit resulting from overlay soil types, elevation, and irrigation types (Figures 1 and 2).

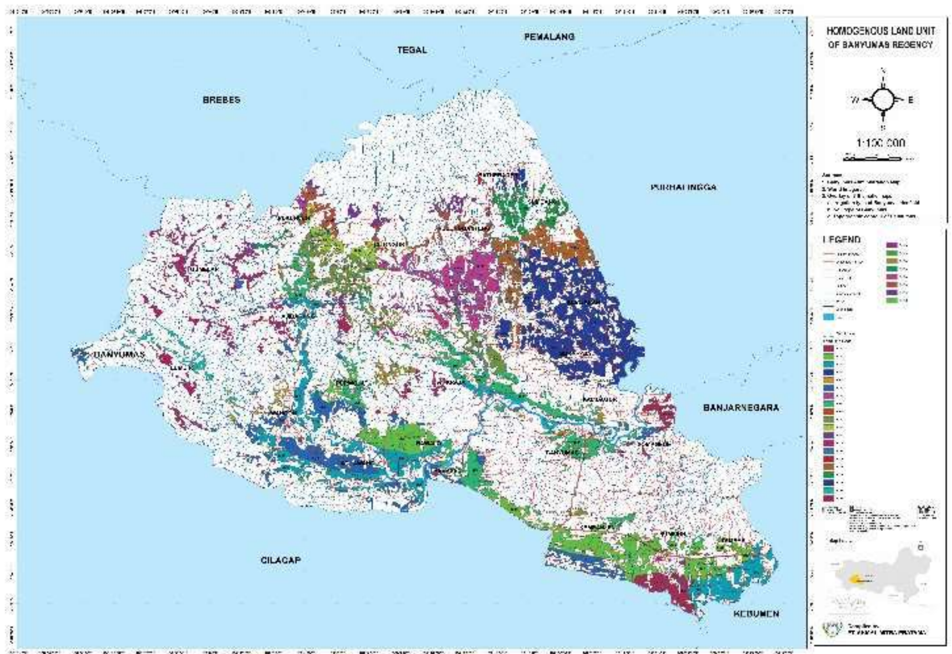

Figure 1. Map of overlay soil types, elevation, and irrigation types in Banyumas.

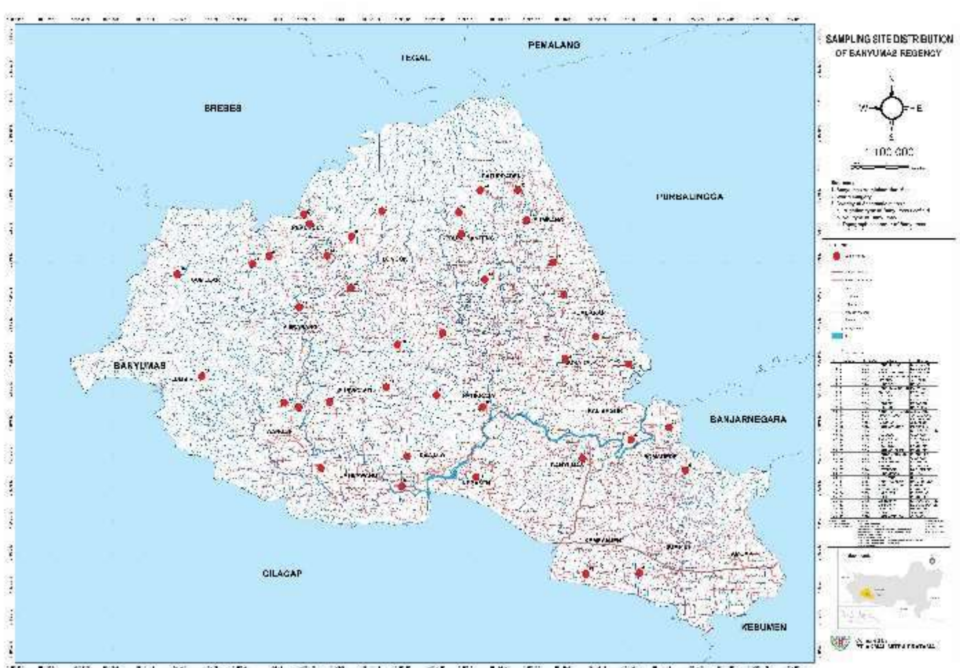

Figure 2. Distribution of sampling sites.

Assessment of heavy metals contamination

Soil and rice grain samples from each site were analyzed to assess heavy metals level. The soil was prepared by air dried, crushed, and sieved in 2 mm size before the concentration of Pb, Cd, and Cr were measured. One hundred rice grains were prepared for heavy metals level measurement.

Approximately 0,5 g of the mashed soil and rice sample was put into a porcelain cup and then heated at 300 °C in the kiln for half an hour. The temperature of the furnace was increased to 500 °C so that the sample was scorched (white). Then the cup and its contents were cooled in a closed state, and 1 mL of 25% HCl was added until dissolved and then heated using a hot plate until the HCl evaporated. The sample in the cup that had been evaporated was dissolved with 10 mL of 1 N HCl and filtered. The filtrate was diluted ten times, and the solution was read for Pb, Cd, and Cr metals using an atomic absorption spectrophotometer (SSA) (Bauer et al., 1978).

To assess the Pb, Cd, and Cr contamination, a pollution index (PI) was calculated using the formula: PI = Cn/Bn, where Cn (mg kg^{-1}) is the measured concentration of each heavy metal, and Bn is the background value for each metal (Amin et al., 2019). The PI of each metal was classified as low contamination (IP<1), moderate contamination (1≤ IP≤ 3), high contamination (3<IP≤6), and very high contamination (IP<6) (Gupta et al., 2021). The background values provide a guide to ascertain the presence of anthropogenic effects that could lead to hazardous or beneficial consequences. In environmental geochemistry and health, the background is considered as the natural concentration in environmental material devoid of human influences (Kazapoe and Arhin, 2021).

The official Indonesian standard for the maximum concentration of heavy metals in soil was not available yet. The heavy metal concentration in soil was standardized to Maximum Permissible Concentrations and Negligible Concentrations for metals (Vodyanitskii, 2016). The heavy metal concentration in rice grain was compared to standards provided by FAO/WHO (IFS, 2019), requirements for heavy metal contamination in food of Indonesia (BPOM, 2022), and The National Standard of China (Woolsey and Bugang, 2010). The translocation capability of heavy metals from soil to plant was calculated using bioaccumulation factor (BAF) as equation BAF = [CR]/[CS], where CR (mg kg^{-1}) and CS (mg kg^{-1}) represent the concentration(s) of heavy metals in rice grains and soil on a dry-weight basis (Khan et al., 2013; Kong et al., 2018).

Statistical analysis

Kolmogorov Smirnov test was used to analyze the normality of data. One-way analysis of variance (ANOVA) by IBM SPSS Statistics version 26 was applied for statistical analysis of the heavy metals content in the study site area. Heavy metal content in soil and rice grain was performed by Pearson correlation analysis.

Results and Discussion

Accumulation of heavy metals in soil

Accumulation of Pb and Cr in soil ranged from 2.7-39.92 ppm and 4.79-61.32 ppm. This level was lower than the Maximum Permissible Concentrations and Negligible Concentrations for metals (MPC), according to the standard for the content of heavy metals in soils (Vodyanitskii, 2016). A different result was found in Cd that exceeded the maximum standard in 21 sites. Cd has contaminated most of the ricefield area in Banyumas. There were 21 from 37 sites containing Cd more than the allowable limit. Those sites, according to the homogenous land unit from spatial analysis, represent 22,199.27 ha or equal to 59% of the total ricefield in Banyumas. In general, the accumulation of Pb and Cr in soils was lower than Cd (Table 2).

Table 2. Heavy metal accumulation in soil and rice grain.

Statistics	Accumulation in soil (ppm)			Accumulation in rice grain (ppm)		
	Pb	Cd	Cr	Pb	Cd	Cr
Sample	37	37	37	37	37	37
Min	2.70	0.11	4.79	0.22	0.10	2.54
Max	39.92	3.01	61.32	32.13	2.16	10.36
Median	26.44	1.80	27.65	3.46	0.24	5.49
Mean	22.64	1.62	27.80	6.85	0.54	5.73
stdev	11.43	0.88	14.27	9.95	0.67	1.93
Maximum Permissible Concentration	140 (1)	1.6 (1)	100 (1)	0.2 (2,4) 0.3 (3)	0.4 (2,3) 0.2 (4)	1.0 (4)

(1) Standard for the content of heavy metals in soils of some states (Vodyanitskii, 2016).
(2) SNI (2009) and BPOM (2022).
(3) FAO/WHO Codex Alimentarius International Food Standards (IFS, 2019).
(4) National Standard of the People’s Republic of China (Woolsey and Bugang, 2010).

The high level of Cd in the soil was caused by the intense use of chemical fertilizers and pesticides in rice cultivation. Cd in soil was released by inorganic fertilizers, mainly N and P, which were categorized as the source of contaminants for containing high amounts of heavy metals. For example, SP-36 contains 11 ppm of Cd and 67 ppm of Pb. Pb and Cd in phosphate fertilizer originated from natural rock phosphate as the raw material of the fertilizer.

Natural rock phosphate generally consists of large amounts of phosphate, carbonate, and heavy metals (Merismon et al., 2017). Therefore, there has a high correlation between the accumulation of Pb and Cd in soils (Figure 3). But, there has no correlation between Pb and Cr, and Cd and Cr according to its accumulation in soil (Figures 4 and 5). This indicated that the presence of Pb and Cd in the soil was in the same proportions as the source of synthetic fertilizers and pesticides. Some studies indicated that the accumulation of Pb and Cd had a high correlation (Merismon et al., 2017; Guo et al., 2020; Tatahmentan et al., 2020).

Banyumas farmers commonly apply chemical fertilizer twice in a growing season at 7-14 and 21-28 days after planting. In pest control, farmers regularly spray pesticides at least twice for a season or more, depending on the pest population. According to the current level of heavy metals in soil due to those chemical inputs, it is crucial to bring a conservative effort to minimize the accumulation in soil. Some organic-based fertilizers and pesticides could be applied as alternative inputs in rice cultivation after conducting soil remediation from heavy metals in Banyumas.

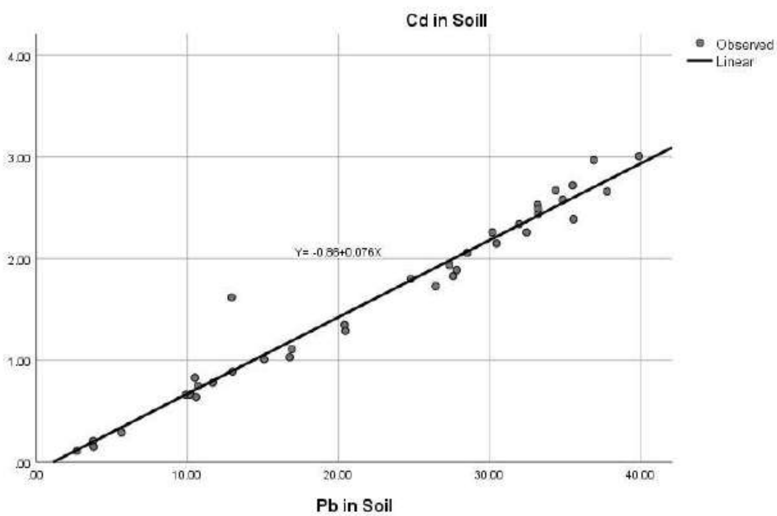

Figure 3. Correlation accumulation of Pb and Cd in soil.

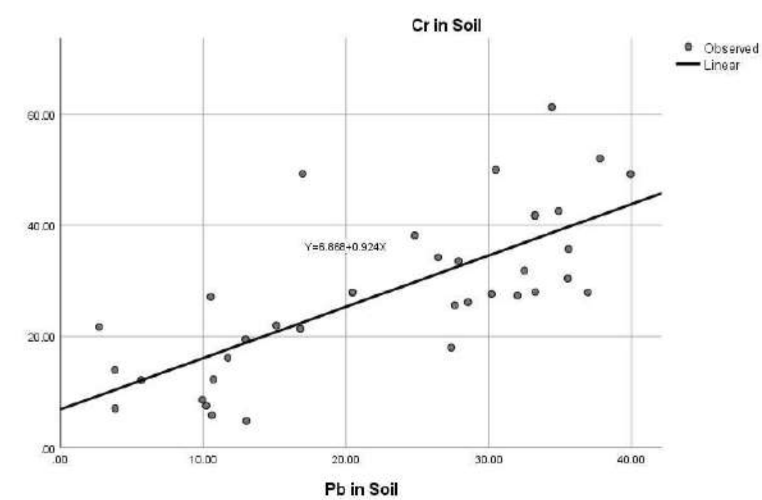

Figure 4. Correlation accumulation of Pb and Cr in soil.

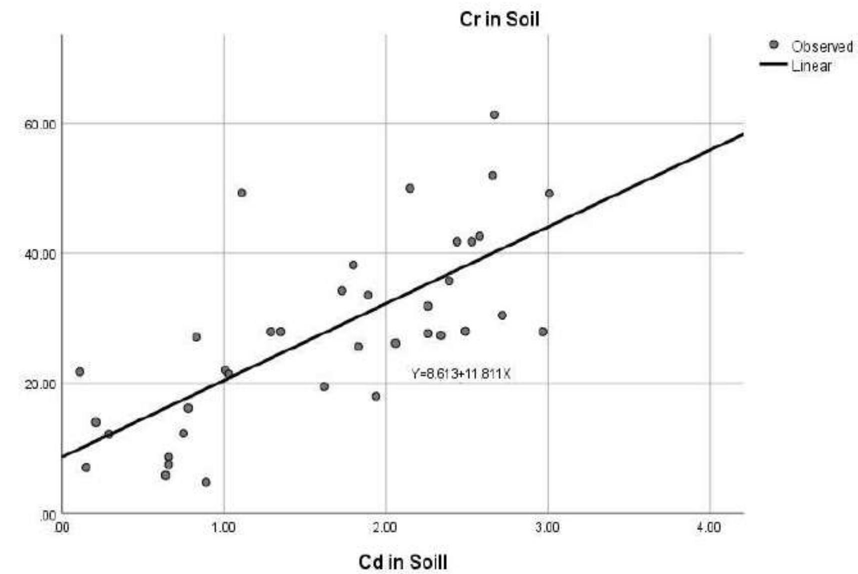

Figure 5. Correlation accumulation of Cd and Cr in soil.

Heavy metals accumulation in rice grain

Rice grain samples were collected from each site of soil sampling. It was found that Pb, Cd, and Cr accumulation in rice grain ranged from 0.22-32.13 ppm, 0.1-2.16 ppm, and 2.54-10.36 ppm, respectively. The level of Pb and Cr in rice grain from each site exceeds the maximum tolerable level according to accumulation of heavy metal standards (0.2-0.4 ppm). On the other hand, Cd accumulation in rice grain exceeds the highest standard only in 9 of 37 sampling sites (>0.4 ppm) (Table 2). There had correlations between heavy metal accumulation of Pb and Cd in rice grains (Figure 6), Pb and Cr in rice grains (Figure 7), and Cd and Cr in rice grains (Fifure 8). The Pb concentration in soil was relatively low, in contrast to the high Pb concentration in rice grain, which was beyond the safety limit of food. Pb is a carcinogenic element for humans. Therefore, its exposure at a chronic level could cause headaches, convulsions, palsy, and nerve damage, especially in children (Guo et al., 2020). The level of Pb accumulation in rice was determined by the cultivation method. Agrochemical applications, including fertilizers and pesticides, contribute to the high accumulation of heavy metals in rice.

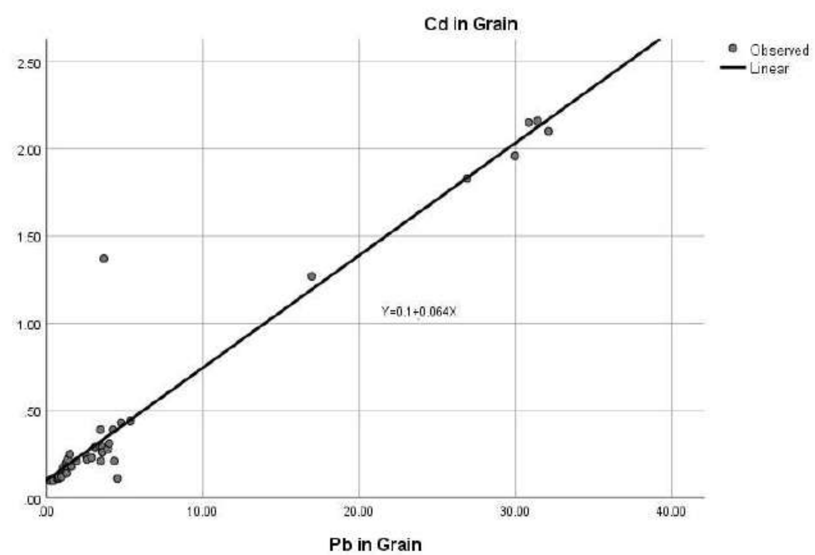

Figure 6. Correlation accumulation of Pb and Cd in rice grain.

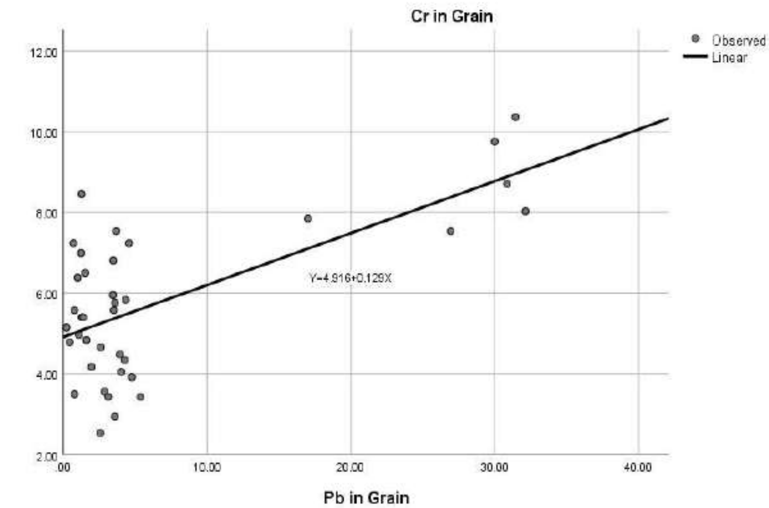

Figure 7. Correlation accumulation of Pb and Cr in rice grain.

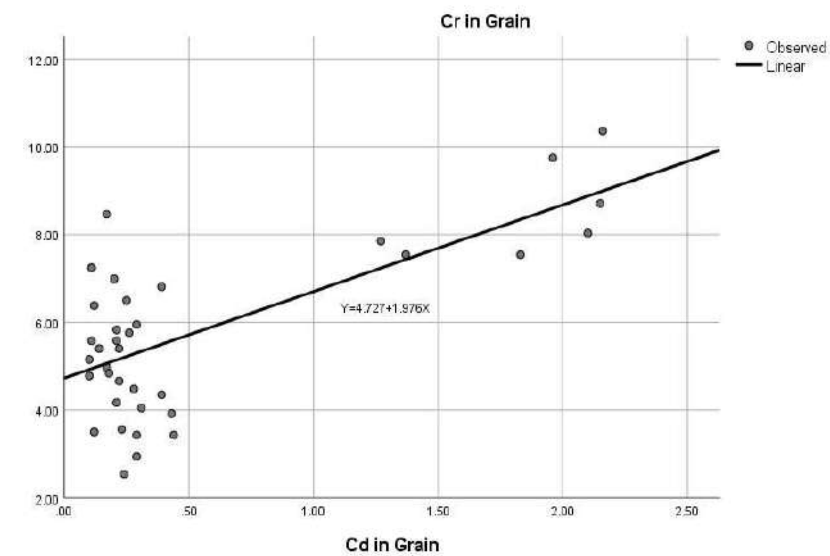

Figure 8. Correlation accumulation of Cd and Cr in rice grain.

The high level of measured Pb in rice was also caused by direct sampling of a rice grain from the field at harvesting without the milling process. Pb level in rice grain could be decreased due to processing to produce white rice. This result is in line with the study of (Norton et al., 2014) and (Tatahmentan et al., 2020), which found a higher level of Pb in rice grain collected from the field than that of white rice from the market.

Cadmium was found to exceed the food safety standard (0.4 ppm) in 9 sites from villages of Kebasen (2.15 ppm), Ajibarang Kulon (1.83 ppm), Beji (2.1 ppm), Kranggan (2.16 ppm), Pasiraman Kidul (0.43 ppm), Kalisari (0.44 ppm), Kasegeran (1.37 ppm), Kaliputih (1.96 ppm) and Semedo (1.27 ppm). The high level of Cd found in rice was suspected to be associated with Cd in contaminated soil of sampling sites which represent 59% of the total area in the Banyumas ricefield.

Intensification of rice cultivation through superior variety and agrochemical input, which was implemented widely since the industrial revolution, has led to national food sufficiency and heavy metals contamination in crop products. The average concentration of Cd in rice grain was 0.5 ppm. Shi et al. (2020) reported that the Cd concentration of Indonesian rice was 0.005-0.597 ppm with 0.019 ppm of the global median value. Cadmium is one of the most mobile and toxic elements in the soil. Cd could be easily transferred into plant tissue and accumulated in harvested part. When Cd enters the human body, it will cause some harmful effects on the lung, cardiovascular, and musculoskeletal systems.

Therefore rice milling would lower the Cd concentration in rice (Meharg et al., 2013).

Indonesian national standard for maximum Cr level in rice is not available yet, therefore, this study adopted other countries' standards. Each rice grain from all sample sites contains Cr more than the safety limit of food, according to the National Standard of China (1.0 ppm). Consuming rice grown in soil containing high levels of Cd, Cr, or Zn could seriously risk human health because 24 to 22% of total heavy metals in plant biomass are accumulated in grains (Kong et al., 2018). Therefore, The soil excess in Cd may produce contaminated rice which is harmful to consume.

Pollution Index (PI)

Most of the measured heavy metals in this study were lower than the preceding value adopted from the standard for the content of heavy metals in soil. Pb levels in soil that ranged from 2.7 to 39.92 ppm were lower than the preceding value (50 ppm). Only one site of soil samples contained Cr more than the preceding value (55 ppm), in contrast to Cd, which mostly exceeded the preceding value.

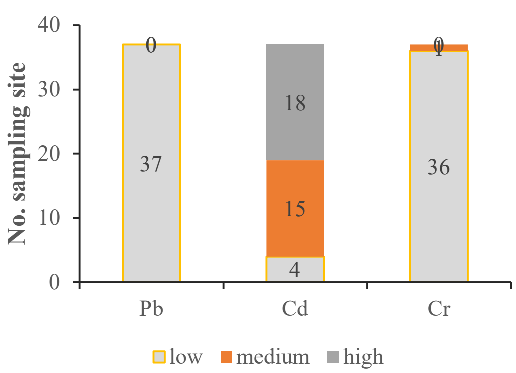

Figure 9. Distribution of sampling site of pollution index.

Based on the study shown in Figure 9 that the pollution index (PI) in each site had a low PI for Pb (IP< 1). PI's of Cd were low in 4 sampling sites, medium (1≤ IP≤ 3) in 15 sampling sites, and high (3<IP≤6) in 18 samples. In addition, PI's of Cr were low in 36 sampling sites and medium in 1 sampling site. This result indicates that land management carried out by Banyumas farmers in rice cultivation affects the increased heavy metals accumulation in soil. The anthropogenic contamination of heavy metal in the Banyumas ricefield from lower to higher was Pb< Cr < Cd.

Bioaccumulation factor (BAF)

Bioaccumulation factors (BAF) of rice crops were 0.01-3.11 for Pb, 0.04-3.26 for Cd, and 0.06-1.30 for Cr. Sequentially, heavy metals with higher bioaccumulation factor average to the lower were Cd>Pb>Cr. The bioaccumulation factor of heavy metals in the plant depends on plant and metal species. This fact is in line with what was reported by (Kabata-Pendias, 2011) that cadmium is the most preferred plant to absorb with intensive accumulation, followed by Pb with medium accumulation, and the last is Cr with low accumulation.

Furthermore, there are several sites in Banyumas where the accumulation factor is >1 (6 sites in Pb, 8 sites in Cd, and 2 sites in Cr), and 2 sites show BAF >1 for the three of Pb, Cd, and Cr. According to (Usman et al., 2019), a plant with BAF>1 indicates that the plant is a bio-accumulator and has the potential to be a phytoremediation agent, and BAF<1 shows that the plant is an excluder.

Several sites with BAF>1 might be caused by foliar application of chemical fertilizer or pesticides during cultivation. Metals from aerial sources may increase contamination in a plant through foliar uptake. Heavy metals from foliar uptake could be translocated to other parts of the plant (Kabata-Pendias, 2011), including rice grain. Plant ability to uptake metals from the atmosphere varies among species. Furthermore, (Kabata-Pendias, 2011) reported that some plants were able to uptake large amounts of metals through leaves, including cereal crops which are sensitive to air pollution for Pb and Ni accumulation in leaves

Conclusion

The concentration of Pb and Cr in the soil of the Banyumas ricefield was below the maximum permissible concentration (MPC) but the concentration of Cd had exceeded MPC in 21 sites which represent 59% of the ricefield in the Banyumas regency. The accumulation of Pb, Cd, and Cr in rice grain exceeded the safety limit in most sample sites. Pb and Cr showed a low pollution index (PI), while Cd was moderate. The bioaccumulation factor (BAF) of heavy metals in rice crops sequentially from the highest to the lowest was Cd>Pb>Cr. The correlation of accumulated Pb and Cd was high according to soil and rice grains.

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Research Article

Mapping heavy metals accumulation in conventional rice farming system at Banyumas Regency of Central Java, Indonesia

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Abstract

Rice grains produced in the conventional system are a prime source of heavy metal exposure in the human body leading to various health problems. The objective of this study was to assess heavy metals concentration in soil and rice grain under the conventional rice farming system in Banyumas, Indonesia, the pollution index, and the bioaccumulation factor. There were thirty seven samples collected based on soil type, elevation, and irrigation system. The heavy metals level in soil ranged from 2.7 to 39.92 ppm of Pb, 0.11 to 3.01 ppm of Cd, and 4.79 to 61.32 ppm of Cr. Pb and Cr accumulation levels were below the maximum permissible concentration (MPC). A different result in Cd exceeded the MPC in 56.75% of sampling sites. Rice grain showed a high accumulation in Pb (6.85 ppm) and Cr (5.73 ppm) that exceeds the maximum standard. Cd exceeded 24.32% of sampling sites with an average of 0.54 ppm. The Pollution Index (PI) was medium in Cd and low in Pb and Cr. The Bioaccumulation Factor (BAF) of heavy metals in rice was Cd>Pb>Cr with averages of 0.64, 0.53, and 0.30, respectively. A high correlation resulted in Pb and Cd in soils and rice grains. Remediation must be done to reduce the heavy metals accumulation in soils and rice grains for environmental and health safety to prevent further contamination.

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Introduction

Rice is the most grown crop in Indonesia as a staple food for the people. The government has consistently pushed national productivity through intensification to meet the needs of the growing population. InIndonesia, most farmers practice conventional farming systems, while total organic agricultural land in Indonesia was only 0.6 percent (208,042 hectares) in 2017 (Willer and Lernoud, 2019). The worldwide application of pesticides has been estimated at approximately 6 billion pounds (Atwood and Paisley-Jones, 2017). This number keeps increasing, especially in developing countries. In Indonesia, detailed information on pesticide uses in agricultural activities, according to rice production, is still limited (Mariyono et al., 2018). Consumption of NPK fertilizer in Indonesia is about 10-11 million tons (BPS Indonesia, 2021), and in Central Jawa, about 400 tons (BPS Jawa Tengah, 2021). Banyumas farmers applied NPK fertilizer of about 7 tons with 371,827.52 tons of rice production in 2020 (BPS Banyumas, 2021). Even applying of synthetics pesticide and fertilizers improves yield production, the impact of its application on the environment, such as in agricultural lands, is unexploited.

The high application of chemical fertilizer could increase heavy metal accumulation in the soil, such as Lead (Pb) and Cadmium (Cd) (Budianta et al., 2016),

then transferred to plants (Merismon et al., 2017). Also, excessive pesticide application in conventional cultivation leads to heavy metal pollution in soil (Sutrisno and Kuntyastuti, 2015), including Chromium (Cr) (Manurung et al., 2018). The contamination of heavy metals has been a serious concern to agricultural land management to mitigate environmental disasters and health problems from several diseases (Faroon et al., 2012). The accumulation of heavy metals such as Pb, Cd, and Cr in soil and food may harm the environment and human health. Heavy metals in soil may affect human health by consuming plant products grown in contaminated soil (Gupta et al., 2021). Consuming food grown in high Cd concentrations of soil is considered to risk health (Ekere et al., 2020).

Exceeding the tolerable level, heavy metals pollution in agricultural soil of some regions in Indonesia requires serious consideration (Laoli et al., 2021; Handayani et al., 2022). Several studies demonstrated the danger of heavy metals accumulation due to chemical fertilizer and pesticides application on onions (Kusumaningrum et al., 2012), rice (Amelia et al., 2015), potatoes (Manurung et al., 2018), and agricultural soil in general Suastawan et al., 2015; Komarawidjadja, 2017). According to (Sutrisno and Kuntyastuti, 2015), heavy metal pollutants originate from industrial waste disposal, agrochemical application, and household waste disposal to waterways.

Accumulation of Pb, Cd, or Cr at a certain level causes growth inhibition, yield decline, and premature death of the crop. In contaminated soil, the plant intakes heavy metals and accumulates them in root, leaf, fruit, and grain (Fang and Zhu, 2014). Pollution of Pb, Cd, and Cr in agricultural soil of some regions in Indonesia has surpassed the tolerable limit (Sutrisno and Kuntyastuti, 2015). Therefore, it needs serious attention from the farmers, government, and industrial society. According to the current state, it is essential to evaluate the distribution of heavy metals in the conventional rice production system. The assessment output is a baseline to determine remediation strategies and agricultural development policies.

The objective of this study was to map the accumulation of Pb, Cd, and Cr in soil and rice grain, the pollution index, and the bioaccumulation factor in the rice field in the Banyumas Regency of Central Jawa, Indonesia.

Materials and Methods

The study area

The coverage area of the study was in the Banyumas regency of Indonesia. The geographic positions of the study sites are spread over 108º39'17”-109º27'15” E, and 7º15'5”-7º37'10” S, with a total area of 1.335,30 km². Banyumas Regency consists of 27 districts, with an average rainfall of 4,354 mm per year. Land uses in Banyumas Regency consist of 31,896 ha (24.03%) of paddy field, 68,088 ha (51.39%) of agricultural land, and 32,774 (24,69%) of non-farmland, dominated by intensive rice farming management (BPS Kabupaten Banyumas, 2021). According to the USDA Soil Taxonomy Classification System, the soils in Banyumas Regency can be grouped into Entisols, Inceptisols, Litosol, Regosol, Oxisol, and Ultisol. Soil sampling locations are presented in Table 1

Table 1. Soil sampling location.

No. sampling site	Location (village)	No. sampling site	Location (village)
1	Karangsoka	20	Somakaton
2	Pliken	21	Cikakak
3	Kalisogra Wetan	22	Kranggan
4	Karangkedawung	23	Karangkemiri
5	Lebeng	24	Pasiraman Kidul
6	Pesawahan	25	Kalisari
7	Bangsa	26	Kasegeran
8	Menganti	27	Srowot
9	Cikakak	28	Ketenger
10	Notog	29	Kebanggan
11	Kebasen	30	Kotayasa
12	Pekunden	31	Karangsalam
13	Ajibarang Kulon	32	Karanggayam
14	Tinggarjaya	33	Petahunan
15	Sirau	34	Kaliputih
16	Gumelar	35	Semedo
17	Karangendep	36	Windujaya
18	Beji	37	Windujaya
19	Pejogol		

Spatial analysis and soil sampling

Spatial analysis was carried out to determine the number and location of samples to be collected. Various maps, including a geographical map, satellite image, irrigation map, and soil map of Banyumas, were overlayed by applying software of geographical information system (GIS) of QGIS Version 3.22 to obtain a homogenous land unit. Sampling sites of soil and rice were determined based on the number and area of fixed homogeneous land units. The descriptive method was applied in this study based on a survey by explorative approach. The site sample was determined through purposive sampling based on the similarity of the soil mapping unit resulting from overlay soil types, elevation, and irrigation types (Figures 1 and 2).

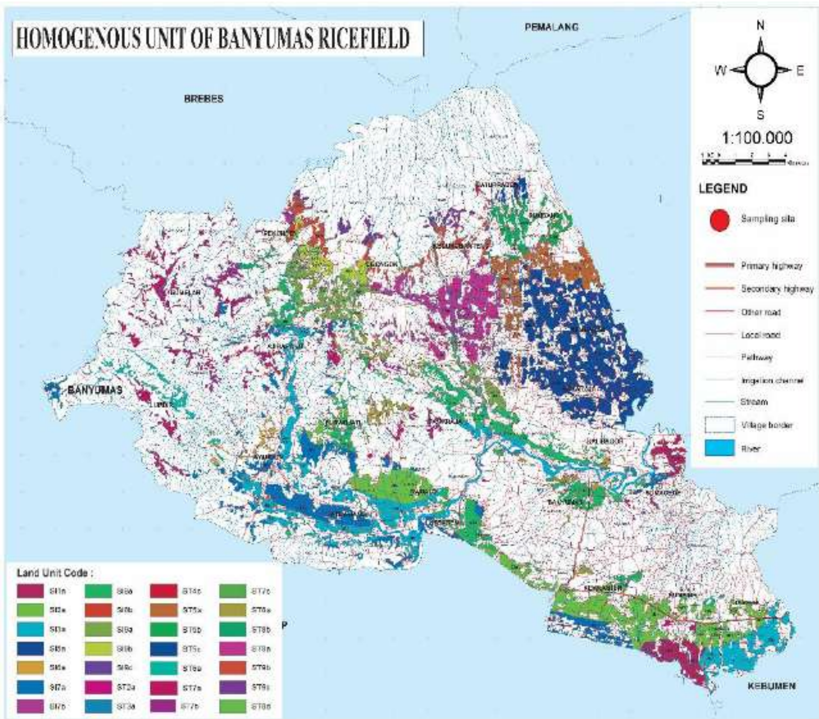

Figure 1. Map of overlay soil types, elevation, and irrigation types in Banyumas.

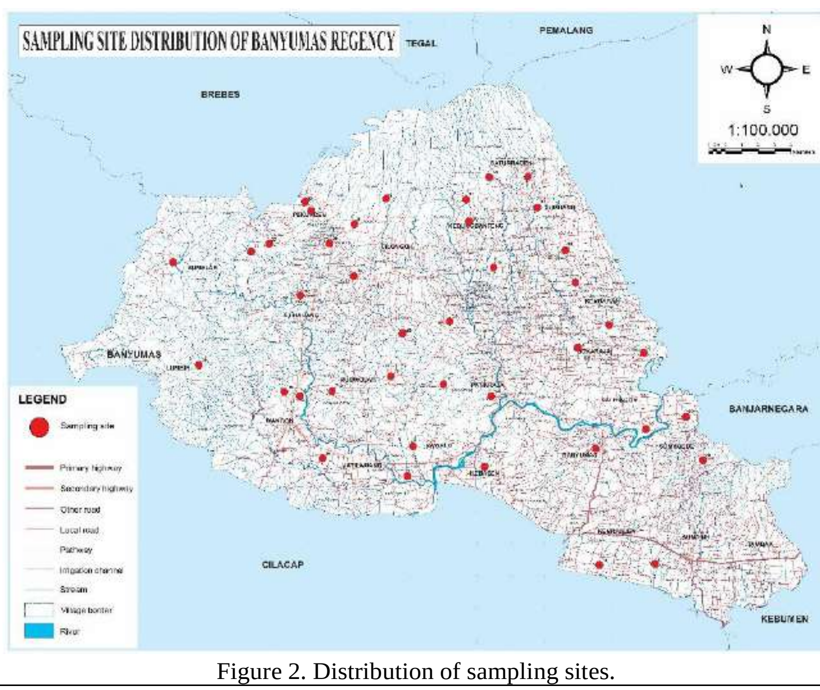

Figure 2. Distribution of sampling sites.

Assessment of heavy metals contamination

Soil and rice grain samples from each site were analyzed to assess heavy metals level. The soil was prepared by air dried, crushed, and sieved in 2 mm size before the concentration of Pb, Cd, and Cr were measured. One hundred rice grains were prepared for heavy metals level measurement.

Approximately 0,5 g of the mashed soil and rice sample was put into a porcelain cup and then heated at 300 °C in the kiln for half an hour. The temperature of the furnace was increased to 500 °C so that the sample was scorched (white). Then the cup and its contents were cooled in a closed state, and 1 mL of 25% HCl was added until dissolved and then heated using a hot plate until the HCl evaporated. The sample in the cup that had been evaporated was dissolved with 10 mL of 1 N HCl and filtered. The filtrate was diluted ten times, and the solution was read for Pb, Cd, and Cr metals using an atomic absorption spectrophotometer (SSA) (Bauer et al., 1978).

To assess the Pb, Cd, and Cr contamination, a pollution index (PI) was calculated using the formula: PI = Cn/Bn, where Cn (mg kg^{-1}) is the measured concentration of each heavy metal, and Bn is the background value for each metal (Amin et al., 2019). The PI of each metal was classified as low contamination (IP<1), moderate contamination (1≤ IP≤ 3), high contamination (3<IP≤6), and very high contamination (IP<6) (Gupta et al., 2021). The background values provide a guide to ascertain the presence of anthropogenic effects that could lead to hazardous or beneficial consequences. In environmental geochemistry and health, the background is considered as the natural concentration in environmental material devoid of human influences (Kazapoe and Arhin, 2021).

The official Indonesian standard for the maximum concentration of heavy metals in soil was not available yet. The heavy metal concentration in soil was standardized to Maximum Permissible Concentrations and Negligible Concentrations for metals (Vodyanitskii, 2016). The heavy metal concentration in rice grain was compared to standards provided by FAO/WHO (IFS, 2019), requirements for heavy metal contamination in food of Indonesia (BPOM, 2022), and The National Standard of China (Woolsey and Bugang, 2010). The translocation capability of heavy metals from soil to plant was calculated using bioaccumulation factor (BAF) as equation BAF = [CR]/[CS], where CR (mg kg^{-1}) and CS (mg kg^{-1}) represent the concentration(s) of heavy metals in rice grains and soil on a dry-weight basis (Khan et al., 2013; Kong et al., 2018).

Statistical analysis

Kolmogorov Smirnov test was used to analyze the normality of data. One-way analysis of variance (ANOVA) by IBM SPSS Statistics version 26 was applied for statistical analysis of the heavy metals content in the study site area. Heavy metal content in soil and rice grain was performed by Pearson correlation analysis.

Results and Discussion

Accumulation of heavy metals in soil

Accumulation of Pb and Cr in soil ranged from 2.7-39.92 ppm and 4.79-61.32 ppm. This level was lower than the Maximum Permissible Concentrations and Negligible Concentrations for metals (MPC), according to the standard for the content of heavy metals in soils (Vodyanitskii, 2016). A different result was found in Cd that exceeded the maximum standard in 21 sites. Cd has contaminated most of the ricefield area in Banyumas. There were 21 from 37 sites containing Cd more than the allowable limit. Those sites, according to the homogenous land unit from spatial analysis, represent 22,199.27 ha or equal to 59% of the total ricefield in Banyumas. In general, the accumulation of Pb and Cr in soils was lower than Cd (Table 2

Table 2. Heavy metal accumulation in soil and rice grain.

Statistics	Accumulation in soil (ppm)			Accumulation in rice grain (ppm)		
	Pb	Cd	Cr	Pb	Cd	Cr
Sample	37	37	37	37	37	37
Min	2.70	0.11	4.79	0.22	0.10	2.54
Max	39.92	3.01	61.32	32.13	2.16	10.36
Median	26.44	1.80	27.65	3.46	0.24	5.49
Mean	22.64	1.62	27.80	6.85	0.54	5.73
stdev	11.43	0.88	14.27	9.95	0.67	1.93
Maximum Permissible Concentration	140 (1	1.6 (1	100 (1	0.2 (2,4) 0.3 (3)	0.4 (2,3) 0.2 (4)	1.0 (4)

(1) Standard for the content of heavy metals in soils of some states (Vodyanitskii, 2016).
(2) SNI (2009) and BPOM (2022).
(3) FAO/WHO Codex Alimentarius International Food Standards (IFS, 2019).
(4) National Standard of the People's Republic of China (Woolsey and Bugang, 2010).

The high level of Cd in the soil was caused by the intense use of chemical fertilizers and pesticides in rice cultivation. Cd in soil was released by inorganic fertilizers, mainly N and P, which were categorized as the source of contaminants for containing high amounts of heavy metals. For example, SP-36 contains 11 ppm of Cd and 67 ppm of Pb. Pb and Cd in phosphate fertilizer originated from natural rock phosphate as the raw material of the fertilizer.

Natural rock phosphate generally consists of large amounts of phosphate, carbonate, and heavy metals (Merismon et al., 2017). Therefore, there has a high correlation between the accumulation of Pb and Cd in soils (Figure 3). But, there has no correlation between Pb and Cr, and Cd and Cr according to its accumulation in soil (Figures 4 and 5). This indicated that the presence of Pb and Cd in the soil was in the same proportions as the source of synthetic fertilizers and pesticides. Some studies indicated that the accumulation of Pb and Cd had a high correlation (Merismon et al., 2017; Guo et al., 2020; Tatahmentan et al., 2020).

Banyumas farmers commonly apply chemical fertilizer twice in a growing season at 7-14 and 21-28 days after planting. In pest control, farmers regularly spray pesticides at least twice for a season or more, depending on the pest population. According to the current level of heavy metals in soil due to those chemical inputs, it is crucial to bring a conservative effort to minimize the accumulation in soil. Some organic-based fertilizers and pesticides could be applied as alternative inputs in rice cultivation after conducting soil remediation from heavy metals in Banyumas.

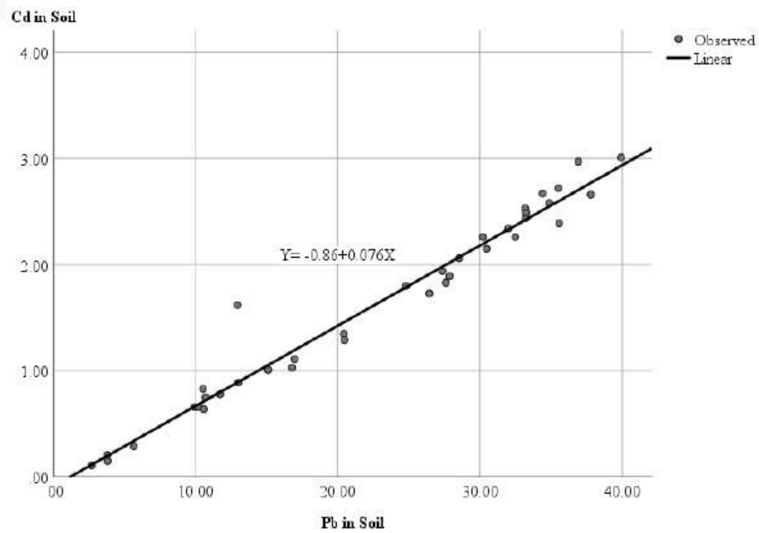

Figure 3. Correlation accumulation of Pb and Cd in soil.

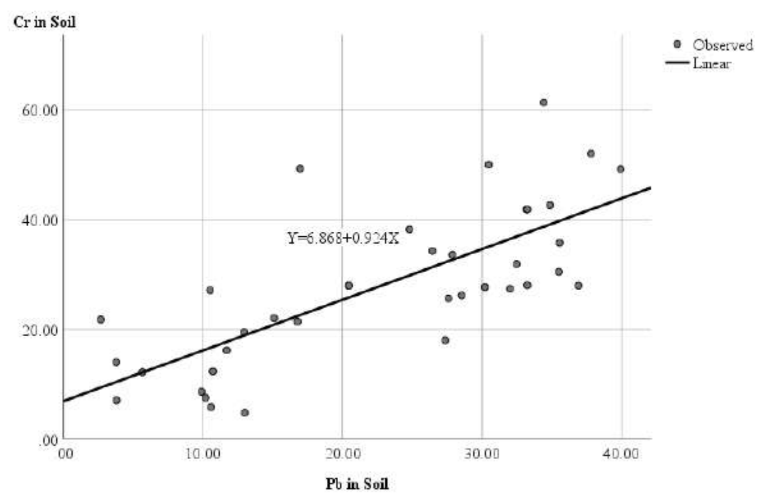

Figure 4. Correlation accumulation of Pb and Cr in soil.

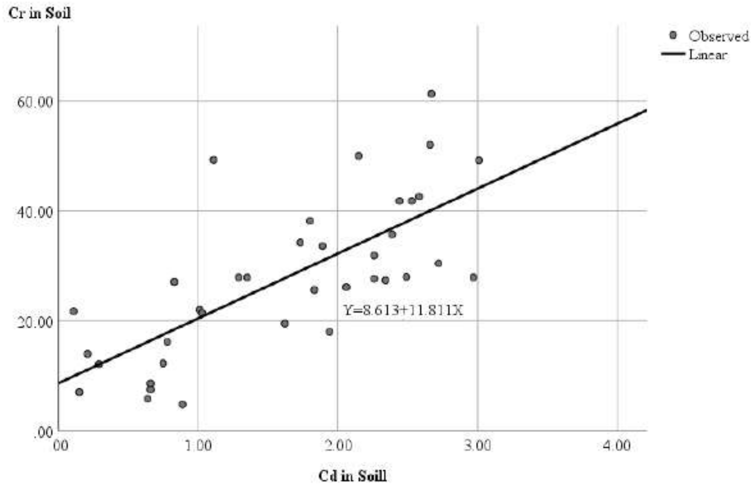

Figure 5. Correlation accumulation of Cd and Cr in soil.

Heavy metals accumulation in rice grain

Rice grain samples were collected from each site of soil sampling. It was found that Pb, Cd, and Cr accumulation in rice grain ranged from 0.22-32.13 ppm, 0.1-2.16 ppm, and 2.54-10.36 ppm, respectively. The level of Pb and Cr in rice grain from each site exceeds the maximum tolerable level according to accumulation of heavy metal standards (0.2-0.4 ppm). On the other hand, Cd accumulation in rice grain exceeds the highest standard only in 9 of 37 sampling sites (>0.4 ppm) (Table 2). There had correlations between heavy metal accumulation of Pb and Cd in rice grains (Figure 6), Pb and Cr in rice grains (Figure 7), and Cd and Cr in rice grains (Fifure 8). The Pb concentration in soil was relatively low, in contrast to the high Pb concentration in rice grain, which was beyond the safety limit of food. Pb is a carcinogenic element for humans. Therefore, its exposure at a chronic level could cause headaches, convulsions, palsy, and nerve damage, especially in children (Guo et al., 2020). The level of Pb accumulation in rice was determined by the cultivation method. Agrochemical applications, including fertilizers and pesticides, contribute to the high accumulation of heavy metals in rice.

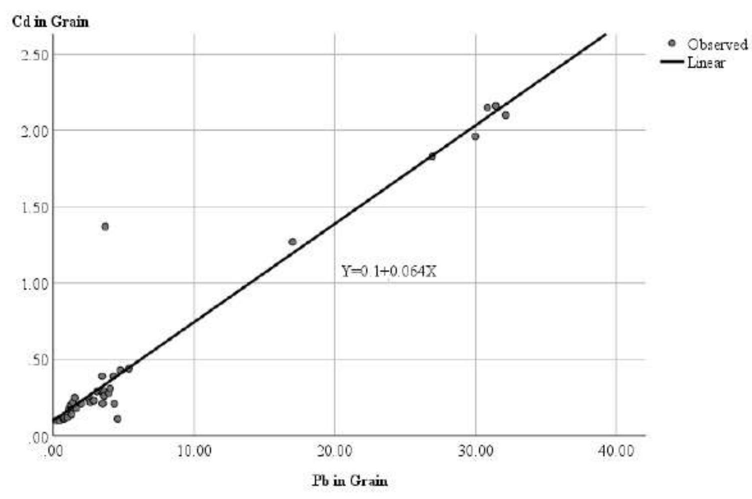

Figure 6. Correlation accumulation of Pb and Cd in rice grain.

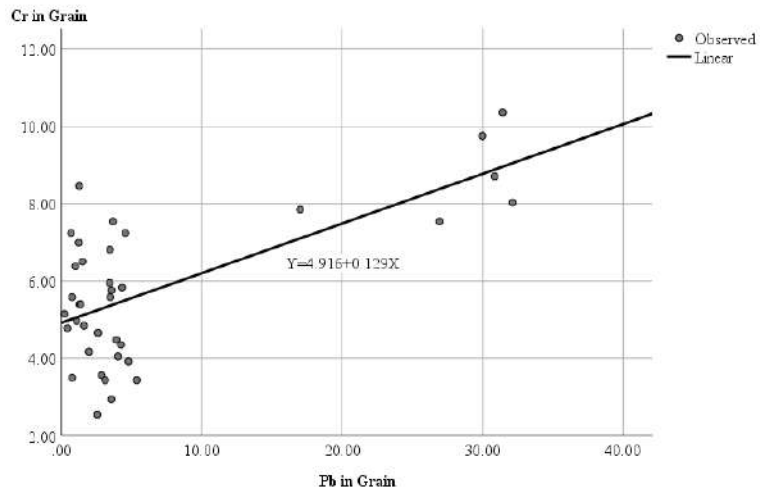

Figure 7. Correlation accumulation of Pb and Cr in rice grain.

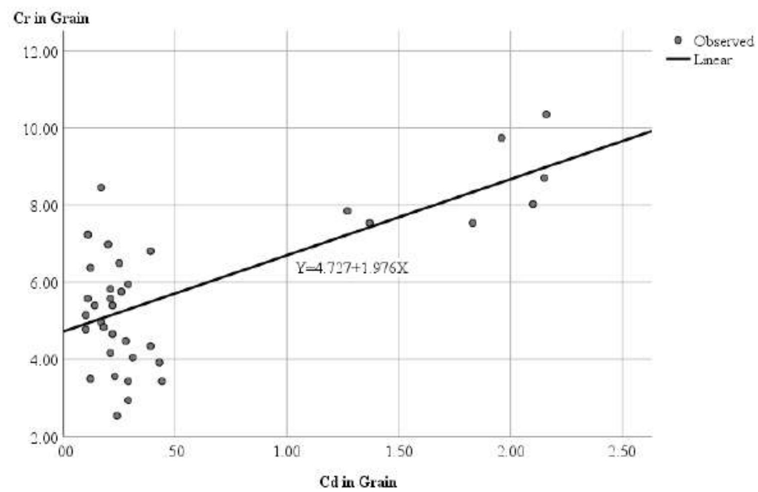

Figure 8. Correlation accumulation of Cd and Cr in rice grain.

The high level of measured Pb in rice was also caused by direct sampling of a rice grain from the field at harvesting without the milling process. Pb level in rice grain could be decreased due to processing to produce white rice. This result is in line with the study of (Norton et al., 2014) and (Tatahmentan et al., 2020), which found a higher level of Pb in rice grain collected from the field than that of white rice from the market.

Cadmium was found to exceed the food safety standard (0.4 ppm) in 9 sites from villages of Kebasen (2.15 ppm), Ajibarang Kulon (1.83 ppm), Beji (2.1 ppm), Kranggan (2.16 ppm), Pasiraman Kidul (0.43 ppm), Kalisari (0.44 ppm), Kasegeran (1.37 ppm), Kaliputih (1.96 ppm) and Semedo (1.27 ppm). The high level of Cd found in rice was suspected to be associated with Cd in contaminated soil of sampling sites which represent 59% of the total area in the Banyumas ricefield.

Intensification of rice cultivation through superior variety and agrochemical input, which was implemented widely since the industrial revolution, has led to national food sufficiency and heavy metals contamination in crop products. The average concentration of Cd in rice grain was 0.5 ppm. Shi et al. (2020) reported that the Cd concentration of Indonesian rice was 0.005-0.597 ppm with 0.019 ppm of the global median value. Cadmium is one of the most mobile and toxic elements in the soil. Cd could be easily transferred into plant tissue and accumulated in harvested part. When Cd enters the human body, it will cause some harmful effects on the lung, cardiovascular, and musculoskeletal systems.

Therefore rice milling would lower the Cd concentration in rice (Meharg et al., 2013).

Indonesian national standard for maximum Cr level in rice is not available yet, therefore, this study adopted other countries' standards. Each rice grain from all sample sites contains Cr more than the safety limit of food, according to the National Standard of China (1.0 ppm). Consuming rice grown in soil containing high levels of Cd, Cr, or Zn could seriously risk human health because 24 to 22% of total heavy metals in plant biomass are accumulated in grains (Kong et al., 2018). Therefore, The soil excess in Cd may produce contaminated rice which is harmful to consume.

Pollution Index (PI)

Most of the measured heavy metals in this study were lower than the preceding value adopted from the standard for the content of heavy metals in soil. Pb levels in soil that ranged from 2.7 to 39.92 ppm were lower than the preceding value (50 ppm). Only one site of soil samples contained Cr more than the preceding value (55 ppm), in contrast to Cd, which mostly exceeded the preceding value.

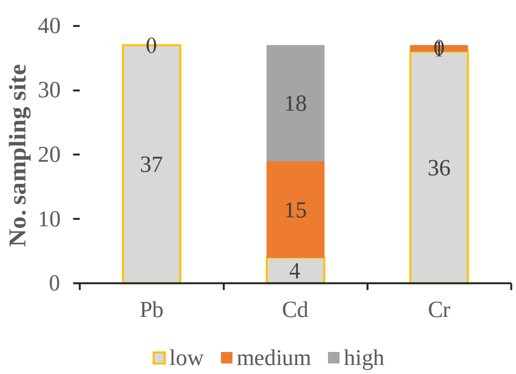

Figure 9. Distribution of sampling site of pollution index.

Based on the study shown in Figure 9 that the pollution index (PI) in each site had a low PI for Pb (IP< 1). PI's of Cd were low in 4 sampling sites, medium (1≤ IP≤ 3) in 15 sampling sites, and high (3<IP≤6) in 18 samples. In addition, PI's of Cr were low in 36 sampling sites and medium in 1 sampling site. This result indicates that land management carried out by Banyumas farmers in rice cultivation affects the increased heavy metals accumulation in soil. The anthropogenic contamination of heavy metal in the Banyumas ricefield from lower to higher was Pb< Cr < Cd.

Bioaccumulation factor (BAF)

Bioaccumulation factors (BAF) of rice crops were 0.01-3.11 for Pb, 0.04-3.26 for Cd, and 0.06-1.30 for Cr. Sequentially, heavy metals with higher bioaccumulation factor average to the lower were Cd>Pb>Cr. The bioaccumulation factor of heavy metals in the plant depends on plant and metal species. This fact is in line with what was reported by (Kabata-Pendias, 2011) that cadmium is the most preferred plant to absorb with intensive accumulation, followed by Pb with medium accumulation, and the last is Cr with low accumulation.

Furthermore, there are several sites in Banyumas where the accumulation factor is >1 (6 sites in Pb, 8 sites in Cd, and 2 sites in Cr), and 2 sites show BAF >1 for the three of Pb, Cd, and Cr. According to (Usman et al., 2019), a plant with BAF>1 indicates that the plant is a bio-accumulator and has the potential to be a phytoremediation agent, and BAF<1 shows that the plant is an excluder.

Several sites with BAF>1 might be caused by foliar application of chemical fertilizer or pesticides during cultivation. Metals from aerial sources may increase contamination in a plant through foliar uptake. Heavy metals from foliar uptake could be translocated to other parts of the plant (Kabata-Pendias, 2011), including rice grain. Plant ability to uptake metals from the atmosphere varies among species. Furthermore, (Kabata-Pendias, 2011) reported that some plants were able to uptake large amounts of metals through leaves, including cereal crops which are sensitive to air pollution for Pb and Ni accumulation in leaves

Conclusion

The concentration of Pb and Cr in the soil of the Banyumas ricefield was below the maximum permissible concentration (MPC) but the concentration of Cd had exceeded MPC in 21 sites which represent 59% of the ricefield in the Banyumas regency. The accumulation of Pb, Cd, and Cr in rice grain exceeded the safety limit in most sample sites. Pb and Cr showed a low pollution index (PI), while Cd was moderate. The bioaccumulation factor (BAF) of heavy metals in rice crops sequentially from the highest to the lowest was Cd>Pb>Cr. The correlation of accumulated Pb and Cd was high according to soil and rice grains.

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Research Article

Mapping heavy metals accumulation in conventional rice farming system at Banyumas Regency of Central Java, Indonesia

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Abstract

Rice grains produced in the conventional system are a prime source of heavy metal exposure in the human body leading to various health problems. The objective of this study was to assess heavy metals concentration in soil and rice grain under the conventional rice farming system in Banyumas, Indonesia, the pollution index, and the bioaccumulation factor. There were thirty seven samples collected based on soil type, elevation, and irrigation system. The heavy metals level in soil ranged from 2.7 to 39.92 ppm of Pb, 0.11 to 3.01 ppm of Cd, and 4.79 to 61.32 ppm of Cr. Pb and Cr accumulation levels were below the maximum permissible concentration (MPC). A different result in Cd exceeded the MPC in 56.75% of sampling sites. Rice grain showed a high accumulation in Pb (6.85 ppm) and Cr (5.73 ppm) that exceeds the maximum standard. Cd exceeded 24.32% of sampling sites with an average of 0.54 ppm. The Pollution Index (PI) was medium in Cd and low in Pb and Cr. The Bioaccumulation Factor (BAF) of heavy metals in rice was Cd>Pb>Cr with averages of 0.64, 0.53, and 0.30, respectively. A high correlation resulted in Pb and Cd in soils and rice grains. Remediation is must done to reduce the heavy metals accumulation in soils and rice grains for environmental and health safety to prevent further contamination.

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Introduction

Rice is the most grown crop in Indonesia as a staple food for the people. The government has consistently pushed national productivity through intensification to meet the needs of the growing population. In Indonesia, most farmers practice conventional farming systems, while total organic agricultural land in Indonesia was only 0.6 percent (208,042 hectares) in 2017 (Willer and Lernoud, 2019). The worldwide application of pesticides has been estimated at approximately 6 billion pounds (Atwood and Paisley-Jones, 2017). This number keeps increasing, especially in developing countries. In Indonesia, detailed information on pesticide uses in agricultural activities, according to rice production, is still limited (Mariyono et al., 2018). Consumption of NPK fertilizer in Indonesia is about 10-11 million tons (BPS Indonesia, 2021), and in Central Jawa, about 400 tons (BPS Jawa Tengah, 2021). Banyumas farmers applied NPK fertilizer of about 7 tons with 371,827.52 tons of rice production in 2020 (BPS Banyumas, 2021). Even applying of synthetics pesticide and fertilizers improves yield production, the impact of its application on the environment, such as in agricultural lands, is unexploited.

The high application of chemical fertilizer could increase heavy metal accumulation in the soil, such as Lead (Pb) and Cadmium (Cd) (Budianta et al., 2016),

then transferred to plants (Merismon et al., 2017). Also, excessive pesticide application in conventional cultivation leads to heavy metal pollution in soil (Sutrisno and Kuntyastuti, 2015), including Chromium (Cr) (Manurung et al., 2018). The contamination of heavy metals has been a serious concern to agricultural land management to mitigate environmental disasters and health problems from several diseases (Faroon et al., 2012). The accumulation of heavy metals such as Pb, Cd, and Cr in soil and food may harm the environment and human health. Heavy metals in soil may affect human health by consuming plant products grown in contaminated soil (Gupta et al., 2021). Consuming food grown in high Cd concentrations of soil is considered to risk health (Ekere et al., 2020).

Exceeding the tolerable level, heavy metals pollution in agricultural soil of some regions in Indonesia requires serious consideration (Laoli et al., 2021; Handayani et al., 2022). Several studies demonstrated the danger of heavy metals accumulation due to chemical fertilizer and pesticides application on onions (Kusumaningrum et al., 2012), rice (Amelia et al., 2015), potatoes (Manurung et al., 2018), and agricultural soil in general Suastawan et al., 2015; Komarawidjadja, 2017). According to (Sutrisno and Kuntyastuti, 2015), heavy metal pollutants originate from industrial waste disposal, agrochemical application, and household waste disposal to waterways.

Accumulation of Pb, Cd, or Cr at a certain level causes growth inhibition, yield decline, and premature death of the crop. In contaminated soil, the plant intakes heavy metals and accumulates them in root, leaf, fruit, and grain (Fang and Zhu, 2014). Pollution of Pb, Cd, and Cr in agricultural soil of some regions in Indonesia has surpassed the tolerable limit (Sutrisno and Kuntyastuti, 2015). Therefore, it needs serious attention from the farmers, government, and industrial society. According to the current state, it is essential to evaluate the distribution of heavy metals in the conventional rice production system. The assessment output is a baseline to determine remediation strategies and agricultural development policies.

The objective of this study was to map the accumulation of Pb, Cd, and Cr in soil and rice grain, the pollution index, and the bioaccumulation factor in the rice field in the Banyumas Regency of Central Jawa, Indonesia.

Materials and Methods

The study area

The coverage area of the study was in the Banyumas regency of Indonesia. The geographic positions of the study sites are spread over 108º39'17”-109º27'15” E, and 7º15'5”-7º37'10” S, with a total area of 1.335,30 km². Banyumas Regency consists of 27 districts, with an average rainfall of 4,354 mm per year. Land uses in Banyumas Regency consist of 31,896 ha (24.03%) of paddy field, 68,088 ha (51.39%) of agricultural land, and 32,774 (24,69%) of non-farmland, dominated by intensive rice farming management (BPS Kabupaten Banyumas, 2021). According to the USDA Soil Taxonomy Classification System, the soils in Banyumas Regency can be grouped into Entisols, Inceptisols, Litosol, Regosol, Oxisol, and Ultisol. Soil sampling locations are presented in Table 1.

Table 1. Soil sampling location.

No. sampling site	Location (village)	No. sampling site	Location (village)
1	Karangsoka	20	Somakaton
2	Pliken	21	Cikakak
3	Kalisogra Wetan	22	Kranggan
4	Karangkedawung	23	Karangkemiri
5	Lebeng	24	Pasiraman Kidul
6	Pesawahan	25	Kalisari
7	Bangsa	26	Kasegeran
8	Menganti	27	Srowot
9	Cikakak	28	Ketenger
10	Notog	29	Kebanggan
11	Kebasen	30	Kotayasa
12	Pekunden	31	Karangsalam
13	Ajibarang Kulon	32	Karanggayam
14	Tinggarjaya	33	Petahunan
15	Sirau	34	Kaliputih
16	Gumelar	35	Semedo
17	Karangendep	36	Windujaya
18	Beji	37	Windujaya
19	Pejogol		

Spatial analysis and soil sampling

Spatial analysis was carried out to determine the number and location of samples to be collected. Various maps, including a geographical map, satellite image, irrigation map, and soil map of Banyumas, were overlayed by applying software of geographical information system (GIS) of QGIS Version 3.22 to obtain a homogenous land unit. Sampling sites of soil and rice were determined based on the number and area of fixed homogeneous land units. The descriptive method was applied in this study based on a survey by explorative approach. The site sample was determined through purposive sampling based on the similarity of the soil mapping unit resulting from overlay soil types, elevation, and irrigation types (Figures 1 and 2).

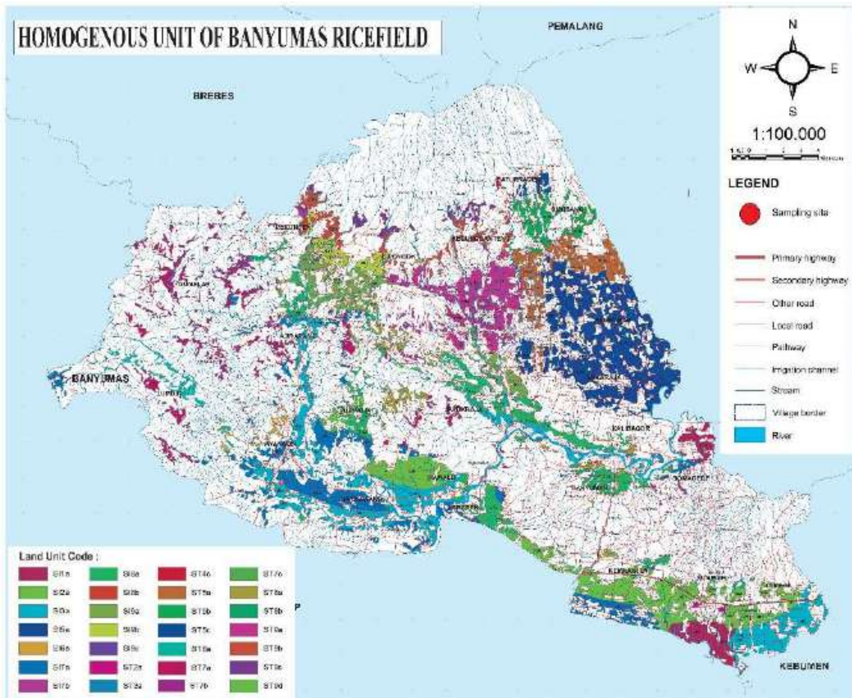

Figure 1. Map of overlay soil types, elevation, and irrigation types in Banyumas.

Figure 2. Distribution of sampling sites.

Assessment of heavy metals contamination

Soil and rice grain samples from each site were analyzed to assess heavy metals level. The soil was prepared by air dried, crushed, and sieved in 2 mm size before the concentration of Pb, Cd, and Cr were measured. One hundred rice grains were prepared for heavy metals level measurement.

Approximately 0,5 g of the mashed soil and rice sample was put into a porcelain cup and then heated at 300 °C in the kiln for half an hour. The temperature of the furnace was increased to 500 °C so that the sample was scorched (white). Then the cup and its contents were cooled in a closed state, and 1 mL of 25% HCl was added until dissolved and then heated using a hot plate until the HCl evaporated. The sample in the cup that had been evaporated was dissolved with 10 mL of 1 N HCl and filtered. The filtrate was diluted ten times, and the solution was read for Pb, Cd, and Cr metals using an atomic absorption spectrophotometer (SSA) (Bauer et al., 1978).

To assess the Pb, Cd, and Cr contamination, a pollution index (PI) was calculated using the formula: PI = Cn/Bn, where Cn (mg kg^{-1}) is the measured concentration of each heavy metal, and Bn is the background value for each metal (Amin et al., 2019). The PI of each metal was classified as low contamination (IP<1), moderate contamination (1≤ IP≤ 3), high contamination (3<IP≤6), and very high contamination (IP<6) (Gupta et al., 2021). The background values provide a guide to ascertain the presence of anthropogenic effects that could lead to hazardous or beneficial consequences. In environmental geochemistry and health, the background is considered as the natural concentration in environmental material devoid of human influences (Kazapoe and Arhin, 2021).

The official Indonesian standard for the maximum concentration of heavy metals in soil was not available yet. The heavy metal concentration in soil was standardized to Maximum Permissible Concentrations and Negligible Concentrations for metals (Vodyanitskii, 2016). The heavy metal concentration in rice grain was compared to standards provided by FAO/WHO (IFS, 2019), requirements for heavy metal contamination in food of Indonesia (BPOM, 2022), and The National Standard of China (Woolsey and Bugang, 2010). The translocation capability of heavy metals from soil to plant was calculated using bioaccumulation factor (BAF) as equation BAF = [CR]/[CS], where CR (mg kg^{-1}) and CS (mg kg^{-1}) represent the concentration(s) of heavy metals in rice grains and soil on a dry-weight basis (Khan et al., 2013; Kong et al., 2018).

Statistical analysis

Kolmogorov Smirnov test was used to analyze the normality of data. One-way analysis of variance (ANOVA) by IBM SPSS Statistics version 26 was applied for statistical analysis of the heavy metals content in the study site area. Heavy metal content in soil and rice grain was performed by Pearson correlation analysis.

Results and Discussion

Accumulation of heavy metals in soil

Accumulation of Pb and Cr in soil ranged from 2.7-39.92 ppm and 4.79-61.32 ppm. This level was lower than the Maximum Permissible Concentrations and Negligible Concentrations for metals (MPC), according to the standard for the content of heavy metals in soils (Vodyanitskii, 2016). A different result was found in Cd that exceeded the maximum standard in 21 sites. Cd has contaminated most of the ricefield area in Banyumas. There were 21 from 37 sites containing Cd more than the allowable limit. Those sites, according to the homogenous land unit from spatial analysis, represent 22,199.27 ha or equal to 59% of the total ricefield in Banyumas. In general, the accumulation of Pb and Cr in soils was lower than Cd (Table 2).

Table 2. Heavy metal accumulation in soil and rice grain.

Statistics	Accumulation in soil (ppm)			Accumulation in rice grain (ppm)		
	Pb	Cd	Cr	Pb	Cd	Cr
Sample	37	37	37	37	37	37
Min	2.70	0.11	4.79	0.22	0.10	2.54
Max	39.92	3.01	61.32	32.13	2.16	10.36
Median	26.44	1.80	27.65	3.46	0.24	5.49
Mean	22.64	1.62	27.80	6.85	0.54	5.73
stdev	11.43	0.88	14.27	9.95	0.67	1.93
Maximum Permissible Concentration	140 (1	1.6 (1	100 (1	0.2 (2,4) 0.3 (3)	0.4 (2,3) 0.2 (4)	1.0 (4)

(1) Standard for the content of heavy metals in soils of some states (Vodyanitskii, 2016).
(2) SNI (2009) and BPOM (2022).
(3) FAO/WHO Codex Alimentarius International Food Standards (IFS, 2019).
(4) National Standard of the People's Republic of China (Woolsey and Bugang, 2010).

The high level of Cd in the soil was caused by the intense use of chemical fertilizers and pesticides in rice cultivation. Cd in soil was released by inorganic fertilizers, mainly N and P, which were categorized as the source of contaminants for containing high amounts of heavy metals. For example, SP-36 contains 11 ppm of Cd and 67 ppm of Pb. Pb and Cd in phosphate fertilizer originated from natural rock phosphate as the raw material of the fertilizer.

Natural rock phosphate generally consists of large amounts of phosphate, carbonate, and heavy metals (Merismon et al., 2017). Therefore, there has a high correlation between the accumulation of Pb and Cd in soils (Figure 3). But, there has no correlation between Pb and Cr, and Cd and Cr according to its accumulation in soil (Figures 4 and 5). This indicated that the presence of Pb and Cd in the soil was in the same proportions as the source of synthetic fertilizers and pesticides. Some studies indicated that the accumulation of Pb and Cd had a high correlation (Merismon et al., 2017; Guo et al., 2020; Tatahmentan et al., 2020).

Banyumas farmers commonly apply chemical fertilizer twice in a growing season at 7-14 and 21-28 days after planting. In pest control, farmers regularly spray pesticides at least twice for a season or more, depending on the pest population. According to the current level of heavy metals in soil due to those chemical inputs, it is crucial to bring a conservative effort to minimize the accumulation in soil. Some organic-based fertilizers and pesticides could be applied as alternative inputs in rice cultivation after conducting soil remediation from heavy metals in Banyumas.

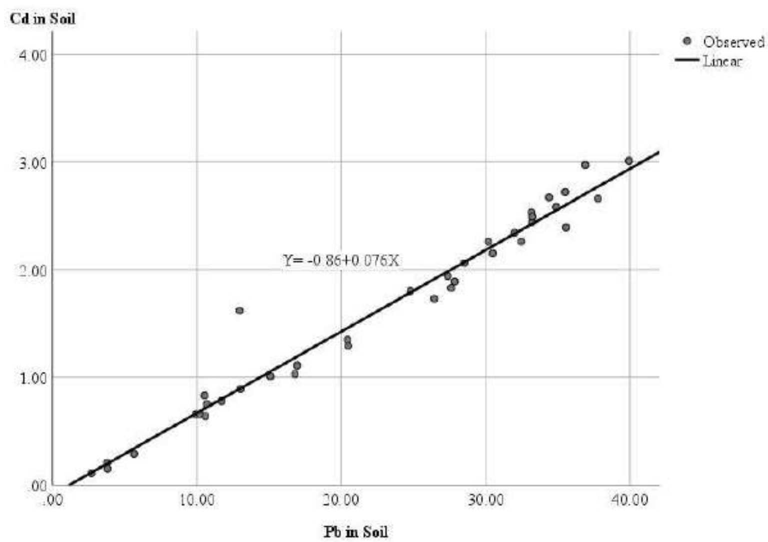

Figure 3. Correlation accumulation of Pb and Cd in soil.

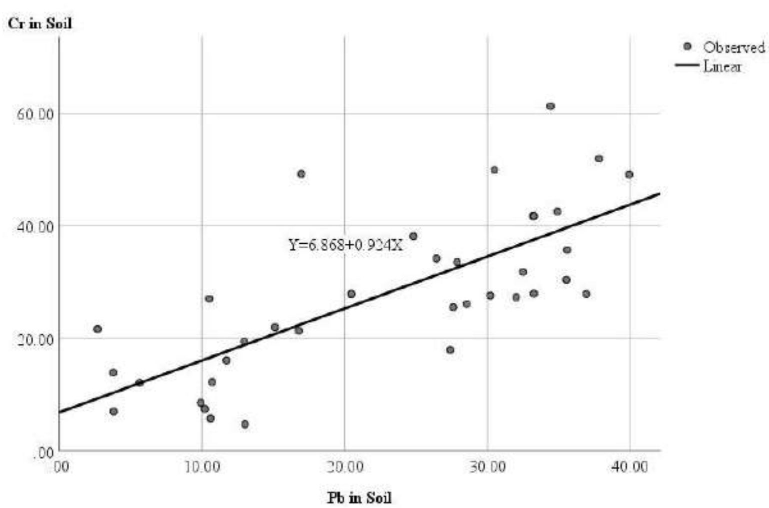

Figure 4. Correlation accumulation of Pb and Cr in soil.

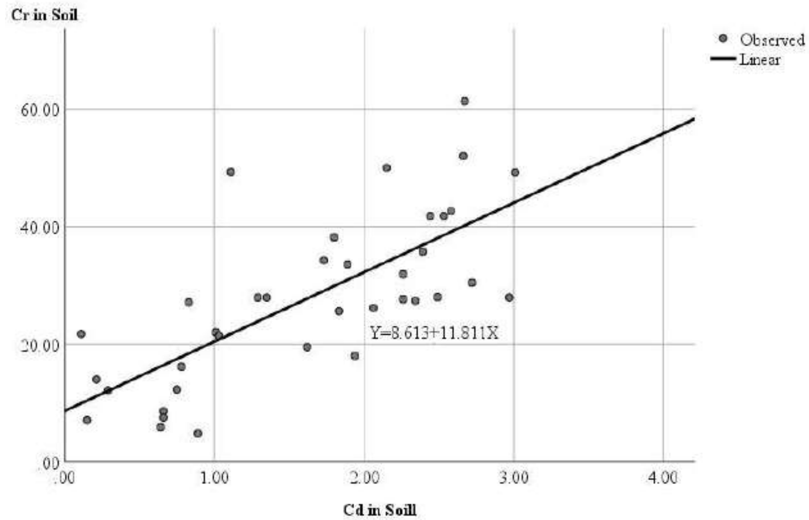

Figure 5. Correlation accumulation of Cd and Cr in soil.

Heavy metals accumulation in rice grain

Rice grain samples were collected from each site of soil sampling. It was found that Pb, Cd, and Cr accumulation in rice grain ranged from 0.22-32.13 ppm, 0.1-2.16 ppm, and 2.54-10.36 ppm, respectively. The level of Pb and Cr in rice grain from each site exceeds the maximum tolerable level according to accumulation of heavy metal standards (0.2-0.4 ppm). On the other hand, Cd accumulation in rice grain exceeds the highest standard only in 9 of 37 sampling sites (>0.4 ppm) (Table 2). There had correlations between heavy metal accumulation of Pb and Cd in rice grains (Figure 6), Pb and Cr in rice grains (Figure 7), and Cd and Cr in rice grains (Fifure 8). The Pb concentration in soil was relatively low, in contrast to the high Pb concentration in rice grain, which was beyond the safety limit of food. Pb is a carcinogenic element for humans. Therefore, its exposure at a chronic level could cause headaches, convulsions, palsy, and nerve damage, especially in children (Guo et al., 2020). The level of Pb accumulation in rice was determined by the cultivation method. Agrochemical applications, including fertilizers and pesticides, contribute to the high accumulation of heavy metals in rice. The high level of measured Pb in rice was also caused by direct sampling of a rice grain from the field at harvesting without the milling process. Pb level in rice grain could be decreased due to processing to produce white rice. This result is in line with the study of (Norton et al., 2014) and (Tatahmentan et al., 2020), which found a higher level of Pb in rice grain collected from the field than that of white rice from the market.

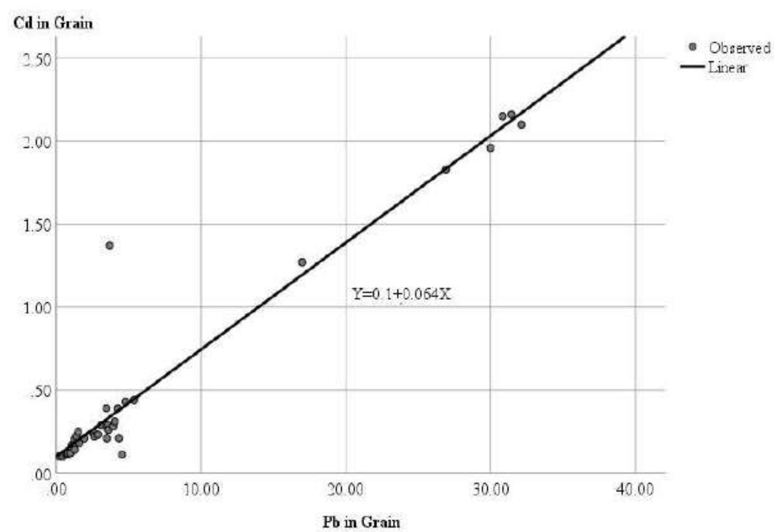

Figure 6. Correlation accumulation of Pb and Cd in rice grain.

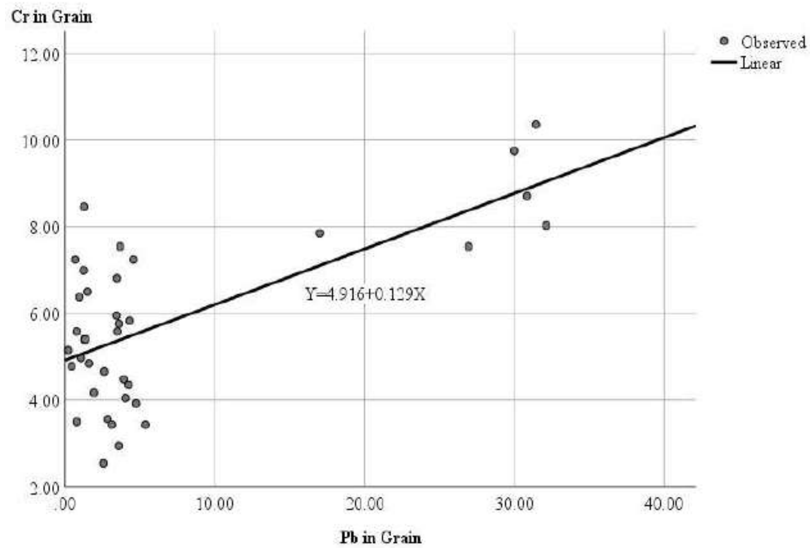

Figure 7. Correlation accumulation of Pb and Cr in rice grain.

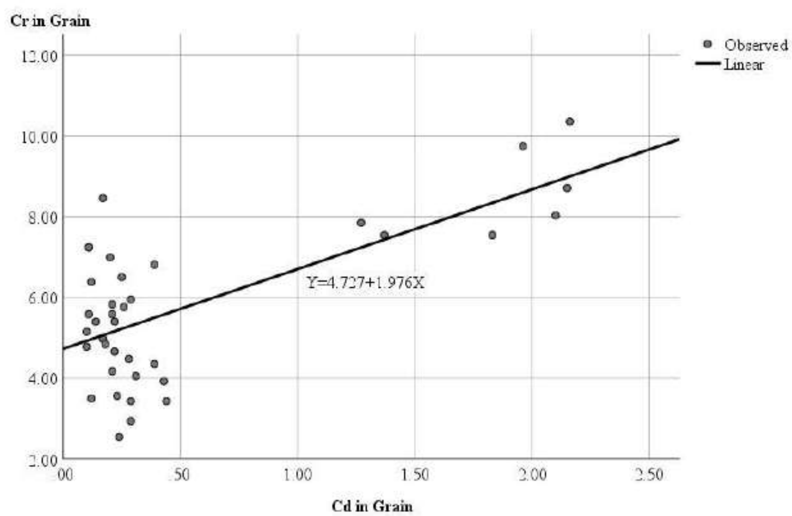

Figure 8. Correlation accumulation of Cd and Cr in rice grain.

Cadmium was found to exceed the food safety standard (0.4 ppm) in 9 sites from villages of Kebasen (2.15 ppm), Ajibarang Kulon (1.83 ppm), Beji (2.1 ppm), Kranggan (2.16 ppm), Pasiraman Kidul (0.43 ppm), Kalisari (0.44 ppm), Kasegeran (1.37 ppm), Kaliputih (1.96 ppm) and Semedo (1.27 ppm). The high level of Cd found in rice was suspected to be associated with Cd in contaminated soil of sampling sites which represent 59% of the total area in the Banyumas ricefield.

Intensification of rice cultivation through superior variety and agrochemical input, which was implemented widely since the industrial revolution, has led to national food sufficiency and heavy metals contamination in crop products. The average concentration of Cd in rice grain was 0.5 ppm. Shi et al. (2020) reported that the Cd concentration of Indonesian rice was 0.005-0.597 ppm with 0.019 ppm of the global median value. Cadmium is one of the most mobile and toxic elements in the soil. Cd could be easily transferred into plant tissue and accumulated in harvested part. When Cd enters the human body, it will cause some harmful effects on the lung, cardiovascular, and musculoskeletal systems. Therefore rice milling would lower the Cd concentration in rice (Meharg et al., 2013).

Indonesian national standard for maximum Cr level in rice is not available yet, therefore, this study adopted other countries' standards. Each rice grain from all sample sites contains Cr more than the safety limit of food, according to the National Standard of China (1.0 ppm). Consuming rice grown in soil containing high levels of Cd, Cr, or Zn could seriously risk human health because 24 to 22% of total heavy metals in plant biomass are accumulated in grains (Kong et al., 2018). Therefore, The soil excess in Cd may produce contaminated rice which is harmful to consume.

Pollution Index (PI)

Most of the measured heavy metals in this study were lower than the preceding value adopted from the standard for the content of heavy metals in soil. Pb levels in soil that ranged from 2.7 to 39.92 ppm were lower than the preceding value (50 ppm). Only one site of soil samples contained Cr more than the preceding value (55 ppm), in contrast to Cd, which mostly exceeded the preceding value.

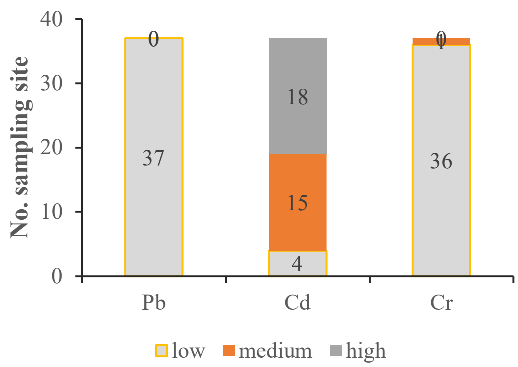

Figure 9. Distribution of sampling site of pollution index.

Based on the study shown in Figure 9 that the pollution index (PI) in each site had a low PI for Pb (IP< 1). PI's of Cd were low in 4 sampling sites, medium (1≤ IP≤ 3) in 15 sampling sites, and high (3<IP≤6) in 18 samples. In addition, PI's of Cr were low in 36 sampling sites and medium in 1 sampling site. This result indicates that land management carried out by Banyumas farmers in rice cultivation affects the increased heavy metals accumulation in soil. The anthropogenic contamination of heavy metal in the Banyumas ricefield from lower to higher was Pb< Cr < Cd.

Bioaccumulation factor (BAF)

Bioaccumulation factors (BAF) of rice crops were 0.01-3.11 for Pb, 0.04-3.26 for Cd, and 0.06-1.30 for Cr. Sequentially, heavy metals with higher bioaccumulation factor average to the lower were Cd>Pb>Cr. The bioaccumulation factor of heavy metals in the plant depends on plant and metal species. This fact is in line with what was reported by (Kabata-Pendias, 2011) that cadmium is the most preferred plant to absorb with intensive accumulation, followed by Pb with medium accumulation, and the last is Cr with low accumulation. Furthermore, there are several sites in Banyumas where the accumulation factor is >1 (6 sites in Pb, 8 sites in Cd, and 2 sites in Cr), and 2 sites show BAF >1 for the three of Pb, Cd, and Cr. According to (Usman et al., 2019), a plant with BAF>1 indicates that the plant is a bio-accumulator and has the potential to be a phytoremediation agent, and BAF<1 shows that the plant is an excluder.

Several sites with BAF>1 might be caused by foliar application of chemical fertilizer or pesticides during cultivation. Metals from aerial sources may increase contamination in a plant through foliar uptake. Heavy metals from foliar uptake could be translocated to other parts of the plant (Kabata-Pendias, 2011), including rice grain. Plant ability to uptake metals from the atmosphere varies among species. Furthermore, (Kabata-Pendias, 2011) reported that some plants were able to uptake large amounts of metals through leaves, including cereal crops which are sensitive to air pollution for Pb and Ni accumulation in leaves

Conclusion

The concentration of Pb and Cr in the soil of the Banyumas ricefield was below the maximum permissible concentration (MPC) but the concentration of Cd had exceeded MPC in 21 sites which represent 59% of the ricefield in the Banyumas regency. The accumulation of Pb, Cd, and Cr in rice grain exceeded the safety limit in most sample sites. Pb and Cr showed a low pollution index (PI), while Cd was moderate. The bioaccumulation factor (BAF) of heavy metals in rice crops sequentially from the highest to the lowest was Cd>Pb>Cr. The correlation of accumulated Pb and Cd was high according to soil and rice grains.

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