

## Research Article

# Source Identification and Spatial Distribution of Heavy Metal Concentrations in Shallot Fields in Brebes Regency, Central Java, Indonesia

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Shallots have been widely planted as the primary commodity crop in Brebes Regency, Central Java, Indonesia. Information on the distribution of heavy metals in the shallot fields of Brebes Regency, Central Java, Indonesia, is not yet available. Hence, the present study was conducted to identify the concentration and spatial distribution of several heavy metals (Pb, Cd, Co, Cr, and Ni) and their possible sources in the shallot fields through a field survey and a series of laboratory and statistical tests. The total concentration of heavy metals was analyzed from 184 sampling points of the shallot fields in Brebes Regency, Central Java, Indonesia, during the dry season from August to October 2019. The heavy metals concentration was as follows: Cr > Ni > Pb > Co > Cd. The values of total Pb, Cd, Co, Cr, and Ni concentrations in the soils were 7.84–18.94, 0.99–2.31, 3.02–9.98, 10.40–49.55, and 10.17–26.62 mg kg<sup>-1</sup>, respectively. All these concentration values of heavy metals are still lower than the critical values for agricultural soils and lower than the topsoil background values except Cd. Based on the concentration of heavy metals, the shallot fields in Brebes Regency, Central Java, are classified as uncontaminated soils. Multivariate and geostatistical analyses were employed to determine and describe the metals' origin. Pb, Cr, and Ni mainly originate from a natural source, while Cd and Co are from anthropogenic sources (agricultural practices and industry).

## 1. Introduction

Heavy metals are among the potentially harmful environmental pollutants in agricultural soils. Their presence in the environment is reported to come from natural and anthropogenic sources. Their natural sources are rocks, parent materials, vegetation, and volcanogenic particles, while the anthropogenic sources include transportation, mining, industrial activities, sewage, and wastewater irrigation, uncontrolled uses of chemical fertilizers and pesticides, and residual organic matters [1–3]. Atmospheric deposition is also a known source of pollution in urban areas and soils cultivated for agricultural purposes [4–6].

Heavy metals may accumulate in substantial amounts in agricultural soils following excessive agrochemical (pesticide and fertilizer) and ameliorant applications, atmospheric, and agriculture machinery usage. It is estimated that the contribution of anthropogenic sources is higher than natural sources [4, 7]. Here, cadmium (Cd), lead (Pb), and chromium (Cr) can be found in increasingly high levels due to the unchecked use of agricultural chemicals. When taken up by plant roots, they can damage the crops and reduce overall agricultural yields. Most importantly, heavy metal accumulation contaminates the environment that will, in turn, disturb the food chain and diminish groundwater quality [8, 9].

Brebes Regency is one of the most important shallot production centers in Java, if not Indonesia, that practices intensive agriculture, including uncontrolled pesticides and fertilizers. Generally, local farmers plant shallots three times a year and one-time planting for a rotation system involving other crops such as rice, chili, corn, and eggplant. However, the agricultural lands have not yet reached their optimal productivity because of some constraints such as low soil fertility, plant disease outbreaks, and pests. To overcome these problems, local farmers generally use chemical fertilizers and pesticides, but, in practice, it is found that their applications are excessive and uncontrolled, thus potentially polluting the environment. During its growth, shallot absorbs nutrients from the soil, which may include heavy metals that are later transferred to agricultural products. As contaminants, they can decrease crop yields and quality and food safety. On the other hand, shallot farming is expected to implement environmentally friendly practices to achieve sustainable agriculture in the future [10–13].

Many researchers have studied agricultural land in Brebes Regency: for example, organochlorine and organophosphate insecticide residues found in rice plants, soil, and water samples [14]; land suitability and availability evaluation for shallot farming development [15, 16]; degradation of soil quality due to pesticide application [11]; farmer's behavior in using pesticide and shallot cultivation [17, 18]; and Pb concentration in shallot bulbs [19]. No studies were reported on the spatial distribution of heavy metals in shallot fields in Brebes Regency, Central Java, Indonesia. Therefore, the present study is considered necessary.

A multivariate analysis is a statistical method for analyzing data that consist of many interdependent variables. Principal component analysis (PCA) has been widely used in geochemical applications to identify contamination sources and apportion natural and anthropogenic contributions. Cluster analysis (CA) is often coupled with PCA to check the results and provide a grouping of individual parameters and variables. A combination of PCA and GIS techniques in soil studies has been used to identify the spatial interpolation and variability-measured soil variables. Geostatistics is used to construct regional distribution maps, which are then compared to the geographical, geologic, and land-use regional using geographical information system (GIS) software. Based on GIS, the spatial distribution of metals can be used to identify their possible sources and contamination or risk hotspot [20–22].

This research aims to identify the spatial distributions and concentrations of several heavy metals (Pb, Cd, Co, Ni, and Cr) and investigate possible sources of these contaminants in shallot fields in Brebes Regency, Central Java Province, Indonesia.

## 2. Materials and Methods

**2.1. Study Sites.** The study was conducted in Brebes Regency on the northern coast of Central Java, Indonesia, stretching from 6°44' to 7°21' S and 108°41' to 109°11' E. A map showing the soil sampling points is shown in Figure 1. Brebes Regency has a tropical climate. Based on the 2019

data, Brebes receives a total rainfall of 3,729 mm, with the highest occurring in January and February, 670 mm and 686 mm, respectively. There are approximately 172 rainy days, most of which are recorded in March and December, counted as 29 days. Inceptisols (USDA soil taxonomy classification) are the predominant soil developing in the research location. Brebes covers an area of 1,769.62 km<sup>2</sup>, 38.1% of which are agricultural lands and the remaining 61.9% are for nonagricultural uses. The regency was selected as the research location based on its shallot productivity, and the persistent practice of excessive fertilizer and pesticide applications may cause heavy metal contamination and land degradation in the shallot fields.

**2.2. Soil Sampling.** The soil sampling was conducted during the dry season period from August to October 2019. In total, 184 topsoil samples (depth 0–20 cm) and five subsamples were collected from 50 to 100 acres of land using a soil auger. These soil samples were thoroughly mixed and bulked. The bulk samples were reduced to about 1–2 kg and labeled according to the soil observation procedure. GPS (global position system) was used to record the soil sampling coordinates. Upon soil collection, all samples were first air-dried in the shade, ground with a mortar and pestle, filtered with a 0.5 mm sieve, and then analyzed at an integrated laboratory at the Indonesian Agricultural Environment Research Institute (IAERI), Jakenan, Central Java, Indonesia.

**2.3. Chemical Analysis.** Soil pH was measured with a pH meter using a water extract and a soil-water ratio of 1 : 2.5 (weight/volume). The total heavy metal contents (Pb, Cr, Co, Cd, and Ni) in the soil were determined using the HNO<sub>3</sub>–HClO<sub>4</sub> mixture for digestion. The resultant liquid extract was measured with an atomic absorption spectrometer (AAS) and then compared with the standard series of each heavy metal. The heavy metal levels were measured at their respective wavelengths: Pb = 217.0 nm, Cd = 228.8 nm, Co = 240.7 nm, Ni = 232.0 nm, and Cr = 357.9 nm. They were calculated using their respective calibration curves. For each heavy metal, the absorbance of the extracted solution was subject to seven replicate measurements with an AAS at two different times to obtain the concentration values. The accuracy limit is % recovery = 75–125% [23]. Pb, Cd, Co, Cr, and Ni concentrations were calculated using the following formula:

$$\text{total heavy metal content (mg} \cdot \text{kg}^{-1}\text{)} = C \text{ curve} \times \frac{V}{W} \times f p x f k, \quad (1)$$

where  $C$  curve is the heavy metal concentration obtained from the AAS (mg kg<sup>-1</sup>),  $V$  is the final extractant volume (ml),  $W$  is the sample weight (g),  $fp$  is the dilution factor, and  $fk$  is the correction factor for sample moisture content. The limit of detection (LoD) for each metals was Pb = 0.0261 mg kg<sup>-1</sup>, Cd = 0.0196 mg kg<sup>-1</sup>, Cr = 0.0266 mg kg<sup>-1</sup>, Co = 0.0094 mg kg<sup>-1</sup>, and Ni = 0.0083 mg kg<sup>-1</sup>.

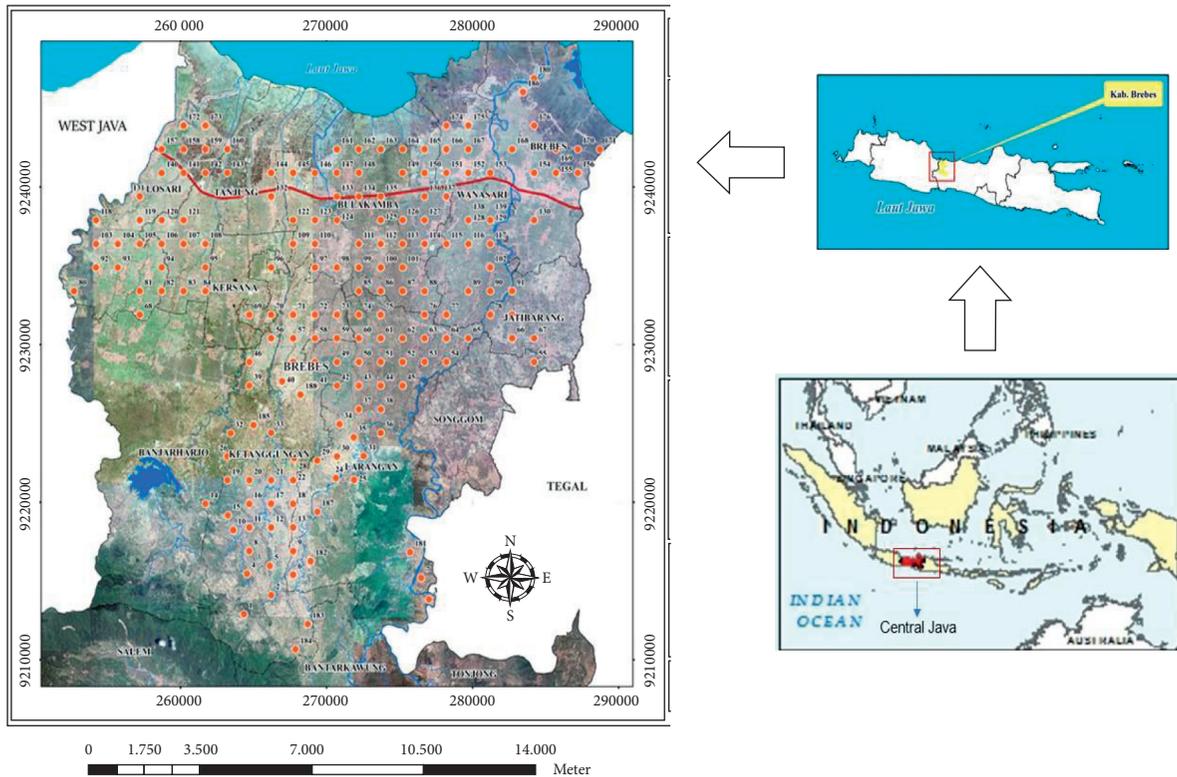


FIGURE 1: The study location and the soil sampling points in Brebes Regency, Central Java, Indonesia.

**2.4. Data Analysis.** The statistical analysis was performed on Minitab 16 program. The raw data of each heavy metal concentration were analyzed for their normal distribution using the Kolmogorov–Smirnov test. The research employed three multivariate analyses. Correlation analysis determined the relationship between heavy metals, principal component analysis (PCA) of similar metal groups identified their possible sources, and cluster analysis grouped the heavy metals observed based on their sources. Geostatistics is a set of statistical tools for inputting and processing temporal and spatial data by coordinates [24–26]. The spatial distributions (maps) of the five heavy metal levels in the shallot fields were generated with simple kriging interpolation on ArcGIS 10.4.

### 3. Results and Discussion

**3.1. Descriptive Statistics and Heavy Metal Concentrations in the Soil.** This section summarizes the statistical analysis results of the heavy metal concentration, soil pH, soil organic carbon (SOC), and cation exchange capacity (CEC) levels in the shallot fields in Brebes Regency, Central Java (Table 1), and the heavy metal concentrations by soil samples (Figure 2). The mean concentrations of the heavy metals in the soil ranged from  $1.78 \pm 0.27$  to  $21.52 \pm 7.42 \text{ mg kg}^{-1}$  in the ascending order of  $\text{Cr} > \text{Ni} > \text{Pb} > \text{Co} > \text{Cd}$ . Their background values are 59.5, 29.0, 27.0, 11.3, and  $0.41 \text{ mg kg}^{-1}$ ; thus, all mean concentrations are lower than their respective background values, except for Cd.

The soil pH ranged from 5.02 to 7.60, with 51.6% of the samples below 6.5, 47.3% between 6.5 and 7.5, and only 1.1%

above 7.5. The factors influencing soil pH are parent material, vegetation type, mineral composition, organic matter, climate, and human activities. The parent material's mineral composition is not a sole determinant of this property; human activities such as long-term chemical fertilization (mainly nitrogen) produce ammonium sulfate and lime that can change soil pH [28–30].

SOC contents in shallot fields are between 0.35 and 3.30%, generally 69% classified as very low (<1%), 30% low between 2 and 3%, and only 1% classified as moderate if SOC is between 2 and 3%. The CEC values ranged from 11.07 to  $62.15 \text{ cmol (+) kg}^{-1}$  with low to very high categories. Soil organic carbon improves the physical properties of the soil. It increases the cation exchange capacity (CEC) and the water-holding capacity. Besides, it contributes to the structural stability of clay soils by helping to bind particles into aggregates. Cation exchange capacity (CEC) is the maximum quantity of total cations that a soil can hold at a given pH value, available for exchange with the soil solution. CEC is used to measure fertility, nutrient retention capacity, and the capacity to protect groundwater from cation contamination. The relationship between CEC and SOC is generally affected by soil pH and local climate, largely the condition of soil physicochemical [31].

A low standard deviation means that the heavy metal concentrations are dispersed homogeneously in the research location. The skewness values of Pb, Cd, Co, and Ni were between  $-1$  and  $1$ , indicating normal distribution. Except for Cr, all heavy metals had kurtosis values lower than 1 and showed normal and platykurtic distribution. CV (coefficient

TABLE 1: Statistical analysis results of heavy metal concentrations in the soil of shallot fields in Brebes Regency, Central Java Province, Indonesia.

Variables	$n$	Mean	Min $\text{mg kg}^{-1}$	Max	SD	CV %	Skewness	Kurtosis	BGV $\text{mg kg}^{-1}$	K-S test
Pb	184	12.53	7.84	18.94	2.50	19.95	0.4	-0.45	27.0	0.058
Cd	184	1.78	0.99	2.31	0.27	15.00	-0.75	0.48	0.41	0.095
Co	184	6.33	3.02	9.98	2.17	34.34	0.06	-1.34	11.3	0.092
Ni	184	16.22	10.17	26.62	3.53	21.73	0.48	-0.35	29.0	0.073
Cr	184	21.52	10.40	49.55	7.42	34.50	1.33	1.86	59.5	0.117
pH	184	6.44	5.02	7.60	0.50	7.80	-0.27	-0.16	—	0.058
SOC <sup>e</sup>	184	0.91	0.35	3.30	0.37	40.35	2.70	11.66	—	0.169
CEC <sup>f</sup>	184	39.10	11.07	62.15	8.33	21.29	-0.15	0.39	—	0.061

SD, standard deviation; CV, coefficient of variation (%); BGV, background contents [27]; K-S, Kolmogorov–Smirnov test; SOC, soil organic carbon (%); CEC, cation exchange capacity ( $\text{cmol}(+) \text{kg}^{-1}$ ).

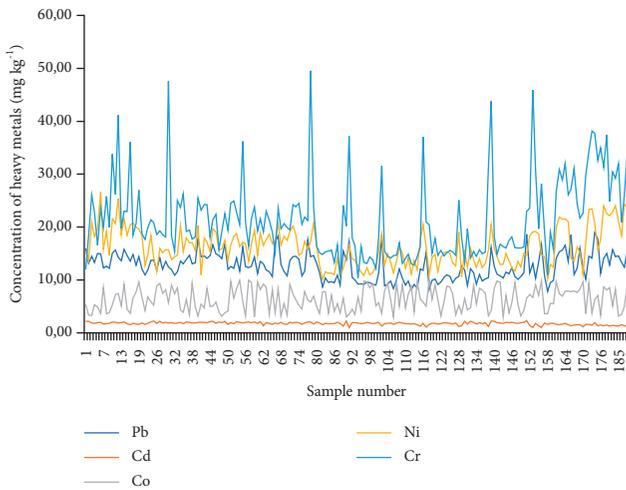


FIGURE 2: Heavy metal concentrations of the soil samples taken in the shallot fields in Brebes Regency, Central Java, Indonesia.

of variation) demonstrates disparate indicator measures. A low CV indicates that agricultural soils are contaminated with naturally occurring heavy metals, whereas a high CV is usually associated with heavy metals of anthropogenic sources [32–34]. The CVs of Pb, Cd, and Ni were 19.95%, 15.00%, and 21.73% (low variation,  $\text{CV} < 25\%$ ), meaning that, in the research location, these elements are sourced naturally (pedogenic processes and parent materials). Meanwhile, the CVs of Co and Cr were 34.34% and 34.50% (moderate variation,  $25\% < \text{CV} < 75\%$ ), indicating anthropogenic roles in Co and Cr entries into the soil.

**3.2. Correlation Coefficient Analysis Results of Heavy Metal Concentrations.** Correlation analysis defines the relation between several variables and helps determine the factors influencing the source of an element. A significant and positive correlation between heavy metals in the soil indicates a similar source of contamination. The Pearson correlation coefficients for each heavy metal pair is at a significance level of  $p < 1\%$ . Pb–Ni (0.622), Pb–Cr (0.577), and Ni–Cr (0.636) have a very significant and positive correlation ( $p < 0.01$ ); soil pH also strongly correlated with soil CEC (0.202) (Table 2); these metal pairs are assumed to come from the same source.

Meanwhile, Cd–Cr has a very significant and negative correlation ( $-0.292$ ): the higher the Cd content, the lower the Cr presence in the soil.

The research found that Ni and Cr were intercorrelated. It is believed to be associated with the type and amount of rock, allowing soil formation from rock layers. Besides, Ni and Cr can also come from anthropogenic sources, limestone, manure, and fertilizers, in which Ni and Cr result in the soil parent material with a different concentration in soils [35, 36].

**3.3. Principal Component Analysis (PCA) Results.** Principal component analysis was used to determine the origin of the soil contaminants. The PCA results and the loading plot of the five heavy metals are given in Table 3 and Figure 3. Table 3 provides that the eigenvalues of the two extracted components were higher than 1.0. The two components (PC1 and PC2) explained 67.10% of the total variance. According to the principal component matrix (Table 4), Pb, Ni, and Cr were included in PC1, which explained 45% of the total variance. Meanwhile, PC2 consisted of Cd (negative loading) and Co (positive loading) and explained 22.1% of the total variance.

PC1, consisting of Pb, Ni, and Cr, comes from the same natural source, such as lithogenic components controlled by parent materials. This finding is also confirmed in previous studies that report low Pb, Ni, and Cr contaminations from natural sources [20, 37, 38]. Based on the correlation coefficient and PCA results, Cd and Co in the second component (PC2) are not intercorrelated. Cd metal is mainly derived from anthropogenic sources, e.g., long-term applications of fertilizer, pesticide, and organic manure [39–41]. The correlation between heavy metals and factors in PCA shows the sources of each contaminant (anthropogenic, natural, or combined). However, it is rather complicated to distinguish the impacts of said elements on the environment [42]. Meanwhile, Co was mainly associated with lithogenic origin from parent materials and anthropogenic sources such as agricultural activities, industry, and household waste. The natural Co concentration in soil was up to  $40 \text{ mg kg}^{-1}$ . In Europe, the areas that are most exposed to the occurrence of high Co concentrations in the soil of anthropogenic sources are mainly industrial and transport areas [43]. The increase of chromium, cobalt, and nickel in

TABLE 2: Pearson correlation coefficients of heavy metal concentrations in the soil of shallot fields in Brebes Regency, Central Java, Indonesia.

	Pb	Cd	Co	Ni	Cr	pH	SOC	CEC
Pb	1							
Cd	0.025	1						
Co	-0.063	-0.071	1					
Ni	0.622**	-0.092	-0.010	1				
Cr	0.577**	-0.292**	-0.042	0.636**	1			
pH	0.004	-0.069	0.037	-0.050	0.039	1		
SOC	-0.017	0.122	0.122	-0.094	-0.118	-0.134	1	
CEC	0.015	-0.154*	0.108	0.039	0.141	0.202**	-0.043	1

\*Significant at  $p < 0.05$ . \*\*Significant at  $p < 0.01$ , (-) negative correlation.

TABLE 3: PCA results of heavy metal concentrations in the soil of shallot fields in Brebes Regency, Central Java, Indonesia.

Principal components	Eigenvalues	Proportions of the total variance %	Cumulative proportions
1	2.2485	45.00	45.00
2	1.1074	22.10	67.10
3	0.9404	18.80	85.90
4	0.3751	7.50	93.40
5	0.3286	6.60	100.00

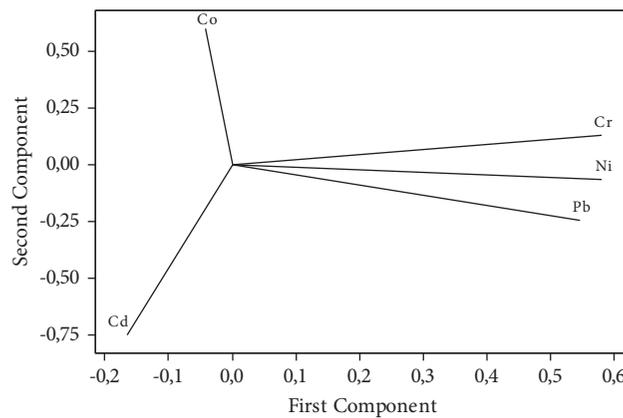


FIGURE 3: The loading plot of Pb, Cd, Co, Cr, and Ni from two components (PC1 and PC2).

TABLE 4: Principal component (PC) matrix correlating heavy metal concentrations in shallot fields in Brebes Regency, Central Java, Indonesia.

Heavy metals	PC1	PC2
Pb	0.546	-0.246
Cd	-0.0165	-0.749
Co	-0.042	0.598
Ni	0.580	-0.068
Cr	0.580	0.130

Vojvodina, Serbia, is explained by the distribution pattern and the presence of ultramafic and mafic parent rocks and anthropogenic sources from industry in several places [44].

3.4. Cluster Analysis (CA) Results. Hierarchical cluster analysis (HCA) aims to classify a variable and measure its

proximity to other variables in the form of a dendrogram: the lower the axis value, the more significant the relationship between said variables [45–47]. Based on the cluster analysis (CA) results (Figure 4), two different clusters were identified: cluster 1 consisted of Pb, Ni, and Cr, while cluster 2 included Cd and Co. The HCA results agreed with the PCA findings in that both analyses classified Pb, Ni, and Cr into cluster 1 while Cd and Co into cluster 2. The first cluster was related to natural factors, while the second was mainly associated with anthropogenic factors.

3.5. Spatial Distribution of Heavy Metals. The spatial distributions of Pb, Cd, Cr, Co, and Ni concentrations in shallot fields in Brebes Regency, Central Java Province, Indonesia, are presented in Figure 5. Pb, Ni, and Cr showed a similar distribution pattern in the shallot fields at lower concentrations, i.e., in the western and middle agricultural areas in

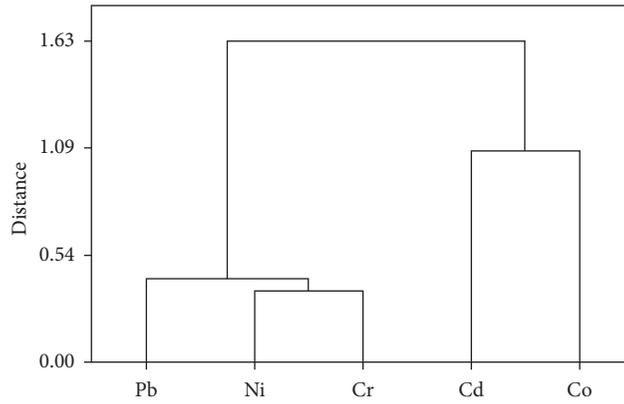


FIGURE 4: Dendrogram of the five heavy metals observed in the shallot fields (Ward's linkage).

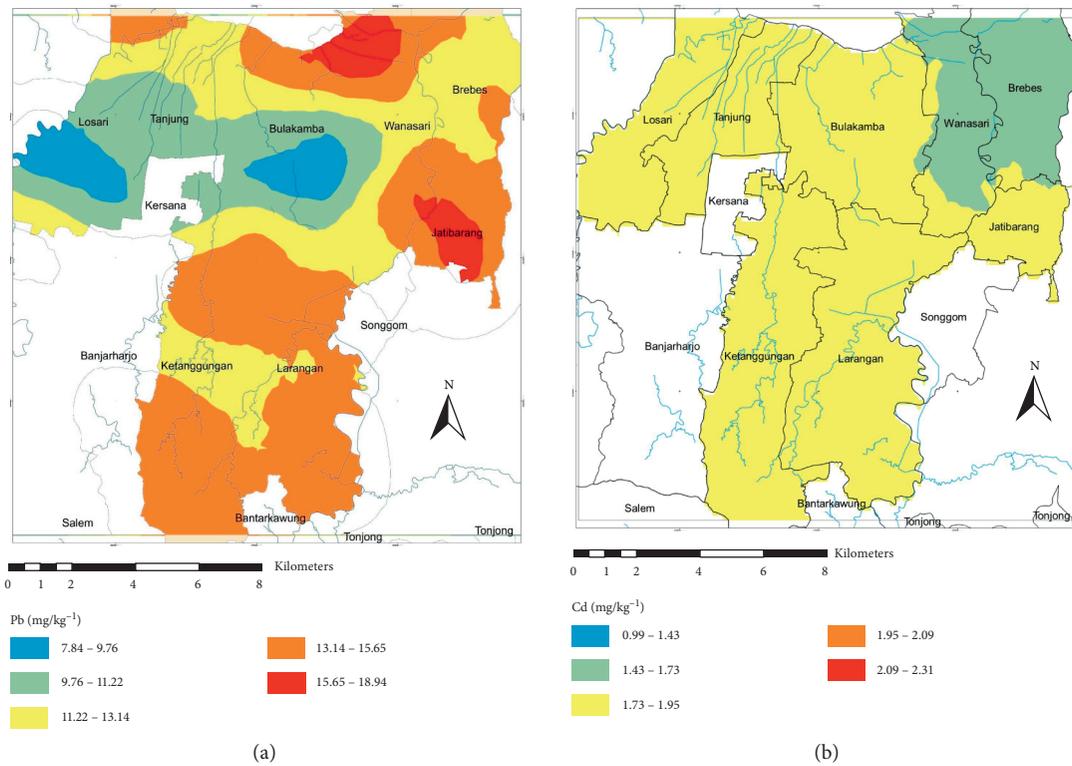


FIGURE 5: Continued.

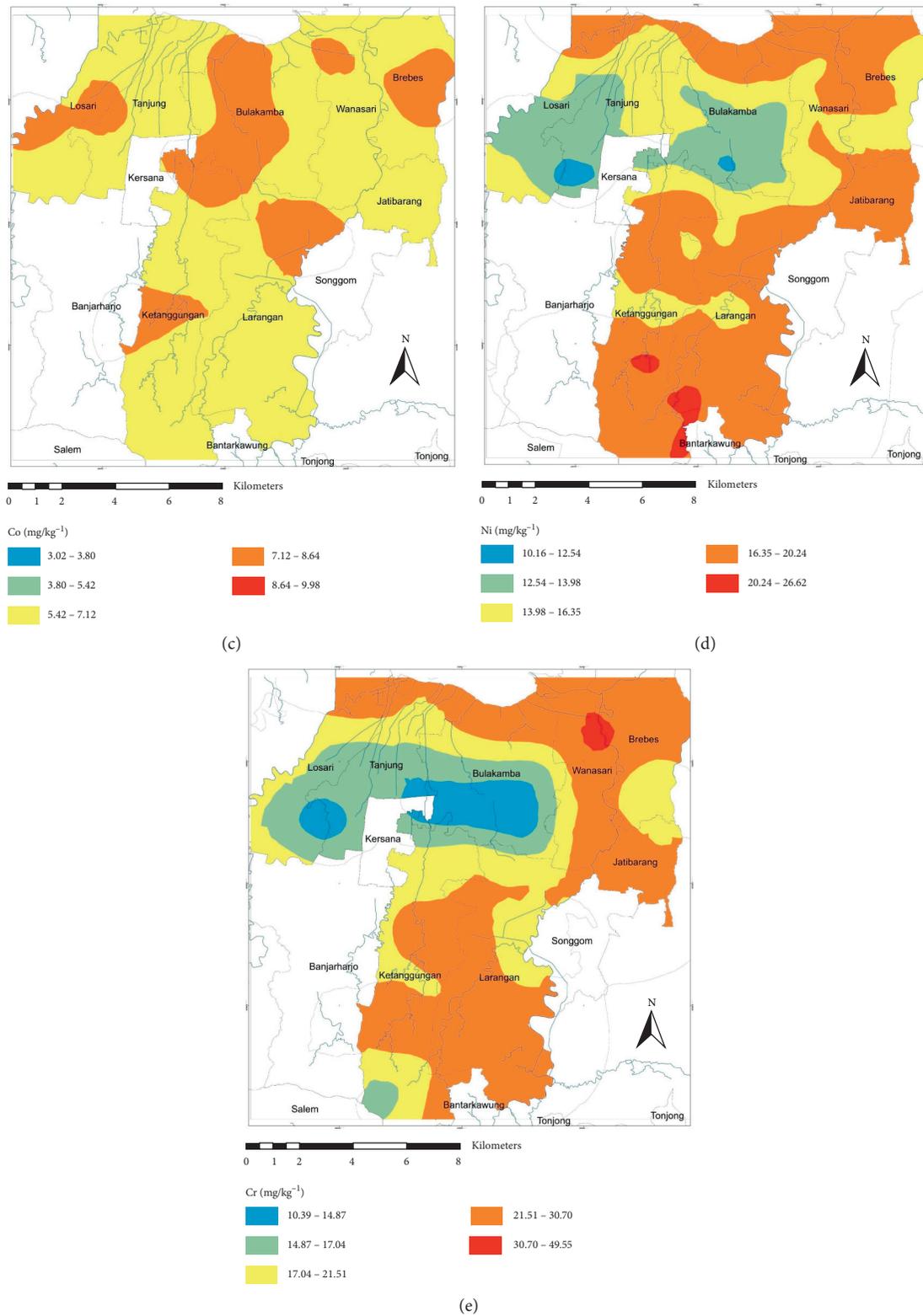


FIGURE 5: Spatial distribution maps of heavy metal concentrations in the soil of shallot fields in Brebes Regency, Central Java, Indonesia.

Losari and Bulakamba, respectively. High Pb contents were detected in the northern part of the Wanasari and the eastern part of Jatibarang. A high Ni concentration was found in the southern area of Brebes Regency, while a high Cr presence was in the northeast. Natural sources such as lithogenic factors and parent materials are believed to control the Pb, Ni, and Cr contents in the agricultural soils [32, 38, 48, 49].

Cd was mostly found in the range of 1.73–1.95 mg kg<sup>-1</sup>, spreading in almost the entire agricultural soils, except the northeastern part of the regency: Wanasari and Brebes. High Cd concentrations are caused by agricultural practices such as long-term applications of animal manure, pesticides, and phosphorus fertilizers. In other studies, the presence of Cd in agricultural soils is significantly correlated with the use of fertilizers (nitrogen, phosphorus, potassium, and fertilizer mixture) [36, 41]. Long-term applications of phosphate fertilizers will increase Cd concentrations in the soil without raising the risk of contamination. In contrast, combined fertilizers (70% phosphate fertilizer and 30% animal manure) create a higher risk of Cd contamination in a short time, with a maximum possibility of Cd causing damage to the soil at 55.21% [50].

The farmer's behavior in shallot cultivation in Brebes Regency, Central Java, was still excessive in using agrochemical. Furthermore, the usage was not in accordance with the recommended dosage in terms of the quality, type, and application method. The use of chemical pesticides and fertilizers that do not match the suggested doses impacts the environment. Besides, nowadays, farmers are also unwilling to use organic fertilizers and ameliorants in shallot cultivation, which are highly recommended to restore soil fertility. The farmer's behavior in the use of pesticides is the key to improving environmental safety. Several factors influenced the behavior. Among others is the level of awareness, attitude, perception of risk, knowledge, and self-efficacy. The majority of farmers applied pesticides without personal protective equipment, so that the pesticides are likely to be exposed to the respiratory system [51, 52].

The cobalt concentrations in shallot fields ranged from 3.02 to 9.98 mg kg<sup>-1</sup>. They were from industrial waste, agricultural activities, household waste, and mining activities. Co can also come from agrochemicals, various organic materials, sewage sludge, food waste, compost, and sewage sludge. The low concentration of Co soil originated from parent materials consists of sediment rock, limestone, claystone, sandstone conglomerate, sandy limestone, and a small amount of andesite [53, 54]. The development of the industrial area in Brebes Regency is currently increasing. As recorded in Banjarsari, Ketanggungan, and Bulakamba, the increase impacts the land conversion functions, from agriculture to industry, that impact the environment. There are several environmental problems in Brebes Regency, including water, air, and soil pollution. It is due to several factors, including many industries, vehicles emission, climate change, global warming, pesticides application, greenhouse gases, and others.

Co was spread with lower concentrations in the southern part, which is a higher land compared to the northern area of Brebes Regency, Central Java. The heavy metals distribution in soil depends on geogenic, anthropogenic, and climate. Several studies have shown that the distribution of heavy metals is influenced by many factors such as traffic levels, wind direction, and topography. Upland soils are known to be more leached than lowland soils due to higher rainfall and moisture conditions [55, 56]. Heavy metals may result from natural processes, such as redistribution and mobilization from rock weathering by surface and subsurface water, flood, and the processes of chemical and physical reactions occurring in the soil. Floods play a role in transporting heavy metals associated with particulates, especially in highly contaminated catchments. It was found that in the dry season, the concentration of heavy metals was higher in the sediment. In comparison, in the rainy season, the metal concentration was higher in the suspended sediment [57–60].

#### 4. Conclusions

The present study provides information on the heavy metal concentration, their possible sources, and distribution in shallot fields in Brebes Regency, Central Java Province, Indonesia. Pb, Cd, Cr, Co, and Ni metals in the soil were below the critical limit that could affect food safety. The combination of multivariate analysis and geostatistical indicates that Pb, Ni, and Cr mainly come from natural sources such as parent materials. Cd comes from anthropogenic sources such as the application of chemical fertilizers, manure, and pesticides. Meanwhile, Co comes from a mixture of natural and anthropogenic sources (agricultural activities, industry, and household waste). This study can be used to establish policies and plan remediation strategies to reduce heavy metal accumulations in agricultural land through environmentally friendly technologies by applying biochar, compost, and biopesticide.

#### Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

#### Conflicts of Interest

The authors declare that there are no conflicts of interest.

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