

## Research Article

# The Residual Copper and Zinc in Tropical Soil over 21 Years after Amendment with Heavy Metal Containing Waste, Lime, and Compost

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This research aimed to evaluate the residual Cu and Zn in tropical soil over 21 years after amendment with industrial waste, lime, and compost. Soil samples were collected from a well-maintained experimental field amended one time with industrial waste high in Cu and Zn set in 1998 at about 1.5–20 years after amendment. Treatments were arranged in a randomized block design with a metal-ware industrial waste at rates 0, 15, and 60 Mg·ha<sup>-1</sup>, lime at 0 and 5 Mg·ha<sup>-1</sup>, and compost at 0 and 5 Mg·ha<sup>-1</sup>. Soil samples were also taken vertically in the exact plot centers at depths of 0–75 cm of plots not amended with lime and compost and also from topsoils (0–15 cm) at 90 points with distances 50 cm between each other in each of the respective plots 21 years after amendment. Significant increases in Cu and Zn caused by waste were observed over 21 years. The increase in soil pH by lime was observed over 10 years but its effect in decreasing Cu and Zn was observed only at 1.5 years, particularly in topsoils. However, the effect of lime in lowering Cu and Zn was observed over 20 years when waste and/or compost were also given. Similarly, the effect of compost was absent over 20 years but significant when waste and/or lime were also given. The relative concentration of Cu and Zn in the 60 Mg waste ha<sup>-1</sup> plots decreased over 20 years to 17–53% for Cu and to 12–33% for Zn; their concentrations were, in general, lower with lime and/or compost addition. The relative concentrations of Cu and Zn significantly decreased over 21 years in plots with 15 or 60 Mg·ha<sup>-1</sup> and increased in the control plots. Heavy metals moved in soil significantly through soil tillage and erosion and partially by leaching.

## 1. Introduction

High concentrations of heavy metals in soils must be properly managed [1–11]. Several chemical mechanisms have been intensively investigated to immobilize heavy metals in soils involving adsorption and precipitation [12–19]. These mechanisms have been reported to lower the heavy metal labile fractions in the soils [20–22]. Previous researches showed that the increase in soil pH by liming and in soil colloid negative charges by organic matter amendment significantly reduced the DTPA-extracted Cu, Cd, and

Zn in some tropical soils including Ultisols, Oxisols, and Alfisols. The negative effects of industrial waste-borne Cu and Zn on the growth of spinach (*Amaranthus spinosus* L.) and corn (*Zea mays* L.) were attenuated by lime and/or cassava-leaf compost amendment [23]. Some other soil workers also reported the reducing effects of lime and/or organic matters on soil heavy metal concentrations [22, 24–27].

However, heavy metals in soils are temporarily retained. Some soil workers [28, 29] showed the movement of some heavy metals in soils treated with heavy metal containing

industrial waste years before the soil sampling. A particular amount of heavy metals may stay in the soil system due to their immobilization through chemical reactions like precipitation at high soil pH and adsorption at low soil pH, particularly in soils with high cation exchange capacity (CEC) [12, 15, 22, 23]. Parts of these heavy metals may with time release into the soil solution through desorption and dissolution processes [15, 30, 31]. Absorption of heavy metals by plant roots may speed up the release of the retained heavy metals on soil colloids [32–34]. Some physical processes like mass flow, diffusion, and leaching of heavy metals, as well as water runoff, erosion, and plowing/tillage, may also accelerate the release and movement of the temporary adsorbed heavy metals [29, 35–40]. The study in [41] reported that heavy metals moved vertically to deeper layers by soil tillage. The study in [42] also demonstrated that the waste-borne Cu and Zn moved to subsoil layers, probably by water leaching. To some extent, plowing and tillage may cause the most significant movement of heavy metals in the soil system. The study in [41] reported that soil tillage moved heavy metals to deeper layers of soils. Previously, the study in [35] reported that plowing may have moved heavy metals including Cd, Cu, and Zn in sludge amended soil out of the experimental plot boundaries. The destruction of soil matrix through mineral weathering and organic matter decomposition may also decrease the soil's ability to immobilize heavy metals [30, 43, 44]. All these processes may with time release the immobilized heavy metals into soil solution and cause dangers on the living things [2, 7, 45–47].

Many research reports addressing the concentrations of heavy metals in soils years after heavy metal amendment are available in the literature [28, 30, 35, 37, 48, 49]. The study in [48] reported the increase in soil heavy metals upon yearly treatment of municipal waste compost at rates 0–80 Mg·ha<sup>-1</sup>. The study in [35] reported the redistribution of sludge-borne heavy metals in the experimental plots years after treatment. The data on the spatial distribution of heavy metals in tropical soils, particularly those in soils treated with high amounts of heavy metals, are not available. The objective of this research was to study the residual spatial and vertical distribution of Cu and Zn in the experimental plots treated with a Cu and Zn containing waste over 21 years after amendment. Tropical soils are unique and different from those of temperate soils due to their high contents of secondary minerals with low CEC affected by high water precipitation that may cause high water leaching and heavy metal movement.

## 2. Materials and Methods

**2.1. Soil Samples, Experimental Field, and Experimental Design.** Soil samples were collected from a well-maintained experimental field amended one time with an industrial waste high in Cu and Zn set in 1998 located in Sidosari, Natar, South Lampung, Indonesia, reported previously (Figure 1) [23, 50]. The experimental field measured 30.5 m × 26.5 m and was a part of an active upland agricultural land with a slope of 2.5–3.0%

(Figure 2). The soil in the experimental field was an Ultisol characterized by textural properties 41.2% sand, 26.0% silt, and 32.8% clay (Textural Class of Sandy Clay Loam); pH of 5.11, organic C content of 1.28 g·kg<sup>-1</sup>, and Cu and Zn of 1.28 and 1.60 mg·kg<sup>-1</sup>, respectively [23], listed in Table 1. The average rainfall in South Lampung was fairly high. The total rainfall in 2015 was 1.628 mm with 143 rainy days [51].

Treatments in the experimental field were arranged in a randomized block design and consisted of 3 factors: industrial waste, lime, and cassava-leaf compost, replicated 3 times. Industrial waste was a metal-ware industrial waste taken from CV Star Metal Wares Jakarta characterized by high Cu and Zn (Table 1). Some properties of the industrial waste were Cu 754 mg·kg<sup>-1</sup>, Zn 44.6 mg·kg<sup>-1</sup>, Pb 2.44 mg·kg<sup>-1</sup>, Cd 0.12 mg·kg<sup>-1</sup>, and pH 7.30 [42]. The waste was air-dried and ground to pass a 1 mm sieve and given one time at 3 levels: 0, 15, and 60 Mg·ha<sup>-1</sup>. Lime was CaCO<sub>3</sub> (calcite), given one time at 0 and 5 Mg·ha<sup>-1</sup>. Organic compost was made of cassava (*Manihot utilissima*) leaf prepared as reported previously [23], also given one time at 0 and 5 Mg·ha<sup>-1</sup>. Rates of industrial waste, lime, and cassava-leaf compost were selected through several preliminary laboratory and glasshouse experiments conducted in 1995–1998 reported previously [23]. The rates of industrial waste were determined to set the heavy metal level into 3 categories: control, low heavy metal, and high heavy metal, particularly for Cu, which was found the highest in the waste (Table 1). The soil extracted Cu of more than 100 mg·kg<sup>-1</sup> was considered high [48]. Lime at 5 Mg CaCO<sub>3</sub>·ha<sup>-1</sup> was to increase the soil pH 1 unit higher than the initial value. The field arrangement of the experimental plots in the field is depicted in Figure 2

Each plot measured 450 cm long and 400 cm wide, with distances 50 cm between plots and 100 cm between blocks. The total number of the experimental plots was 36 separated into 3 blocks. Borders were imaginary, permanently marked only in several points, to ease plowing, planting, and plant maintenance, and therefore, it was possible for the soil masses to move between plots and blocks during plowing and harrowing. The industrial waste was broadcasted on the surface of the soil and mixed thoroughly by plowing (0–15 cm). Lime and cassava-leaf compost were both given into 0–15 cm depth by plowing one week after waste treatment. The experimental plots were planted with various crops since 1998, first planted with corn, and then dryland paddy, cassava, peanut, and sometimes left bare in between. Plowing and harrowing before planting were conducted twice, each in the direction N-S and E-W vice versa.

To evaluate the residual Cu and Zn in soils, three evaluations were conducted. The first evaluation was to study the residual Cu and Zn in topsoil (0–15 cm) and subsoil (15–30 cm) in all experimental plots over a time span of 20 years. The second evaluation was to study the vertical spatial distribution of Cu and Zn over time in the experimental plots treated only with industrial waste at 0, 15, and 60 Mg·ha<sup>-1</sup>. The third evaluation was to study the horizontal spatial distribution of Cu and Zn in the experimental plots treated only with industrial waste at 0, 15, and 60 Mg·ha<sup>-1</sup>.



FIGURE 1: Geographical location of the experimental plots.

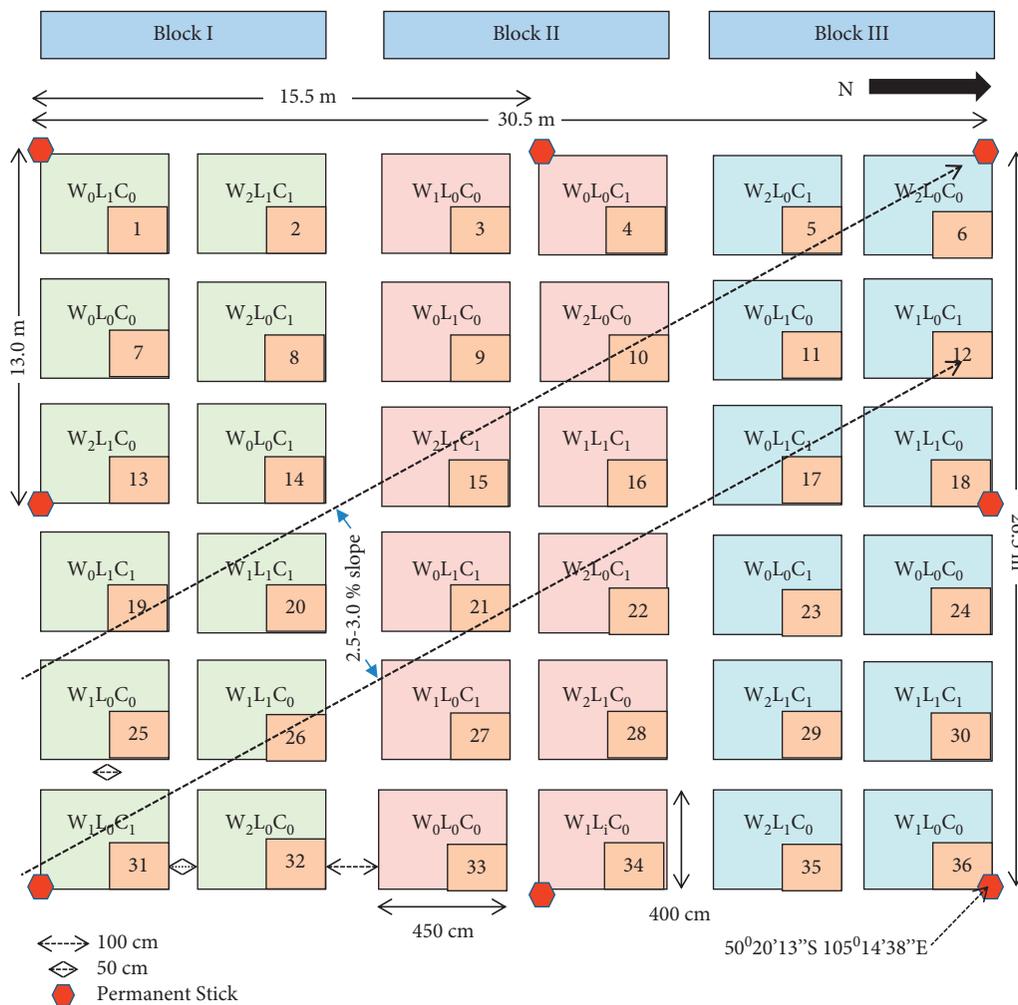


FIGURE 2: The arrangement of the experimental plots set in 1998 ( $W$  = industrial waste,  $W_0 = 0$ ,  $W_1 = 15$ ,  $W_2 = 60 \text{ Mg}\cdot\text{ha}^{-1}$ ;  $L$  = lime,  $L_0 = 0$ ,  $L_1 = 5 \text{ Mg calcite}\cdot\text{ha}^{-1}$ ;  $C$  = cassava-leaf compost,  $C_0 = 0$ ,  $C_1 = 5 \text{ Mg}\cdot\text{ha}^{-1}$ ).

2.2. Evaluation of the Residual Cu and Zn over 20 Years. To reach the purpose of this research, soil samples were collected and analyzed for Cu and Zn from all experimental units (plot nos. 1–36 in Figure 2) at particular times over the last 20 years (1998–2018). The latest soil sampling was

conducted on July 20, 2018 (almost 20 years after the amendment). Composite topsoil (0–15 cm) and subsoil (15–30 cm) samples were taken diagonally from 5 points in each plot. For the purpose of this research, this data was compared to those of  $\pm 1.5$  years,  $\pm 3$  years [42], and  $\pm 10$  years

TABLE 1: Selected initial properties of materials used in the experiment.

Properties	Methods	Materials		
		Soil	Compost	Industrial waste
Sand (%)	Hydrometer	41.2		
Silt (%)	Hydrometer	26.0		
Clay (%)	Hydrometer	32.8		
pHH <sub>2</sub> O (1 : 2)	Electrode	5.11		7.3
Organic C (g kg <sup>-1</sup> )	Walkley and black	1.28	275	
Total N (g kg <sup>-1</sup> )	Kjeldahl		40.1	
C/N			6.85	
Cu (mg kg <sup>-1</sup> )	DTPA	1.60		754
Zn (mg kg <sup>-1</sup> )	DTPA	0.89		44.6
Pb (mg kg <sup>-1</sup> )	DTPA	0.08		2.44
Cd (mg kg <sup>-1</sup> )	DTPA	nd		0.12

nd stands for below detection limit; after [50].

after amendment [50]. The method of soil sampling and analysis for this research was the same as that conducted for  $\pm 10$  years after amendment.

**2.3. Evaluation of the Vertical Spatial Distribution of Cu and Zn in the Experimental Plots.** Depth-wise soil samples were also collected from the exact centers of the experimental plots to investigate the vertical distribution of Cu and Zn (Table 2). The soil samples were taken depth-wisely using a Belgian auger at 0–15, 15–30, 30–45, 45–60, and 60–75 cm (Figure 3), conducted in January 2019 ( $\pm 21$  years after amendment) from 9 plots comprising 3 plots with no addition of industrial waste with no lime and no organic compost (control plot (CP)) including plot nos. 7, 24, and 33, 3 plots with 15 Mg waste ha<sup>-1</sup>, no lime, and no organic compost treatment (low metal plot (LMP)) including plot nos. 3, 25, and 36, and 3 plots with 60 Mg waste ha<sup>-1</sup>, no lime, and no organic compost treatment (high metal plot (HMP)) including plot nos. 6, 10, and 32 in Figure 2. The total experimental units sampled were 9 plots comprising 3 levels of industrial waste treatment levels with 3 replications.

**2.4. Evaluation of the Horizontal Spatial Distribution of Cu and Zn in the Experimental Plots.** Soil samples were also purposively collected in January 2019 ( $\pm 21$  years after amendment) from the A<sub>p</sub> horizons (0–15 cm) of the experimental plots to evaluate the horizontal spatial distribution of Cu and Zn. The soil samples were collected only from 3 plots representing CP (plot no. 7), LMP (plot no. 36), and HMP (plot no. 10 in Figure 2). The soil samples were collected using an auger from 90 sites in each plot located 50 cm apart between sites (Figure 4).

**2.5. Soil and Data Analysis.** Each soil sample was air-dried, ground to pass a 2 mm sieve, thoroughly mixed, and then analyzed for the soil pH, organic C, and the concentrations of Cu and Zn. The soil pH (1 : 2) was determined with a pH electrode, organic C with the method of Walkley and Black, and heavy metal analysis with the DTPA method [52]. The extracting solution was prepared following the procedure in the method. Exactly 1.96 g diethylene triamine pentaacetic

acid powder and 14.92 g triethanolamine were dissolved in about 900 ml distilled water in a 1000 ml beaker glass. After added with 1.47 g CaCl<sub>2</sub>·2H<sub>2</sub>O and the pH adjusted to 7.30, the solution was quantitatively moved into a 1000 ml volumetric flask and the volume was adjusted to 1000 ml with deionized water, after which the solution was thoroughly mixed.

The heavy metal analysis was conducted by extracting 10 g oven-dry equivalent (105 °C 24 hours) of air-dry soil sample with 20 ml of the DTPA extracting solution in an end-to-end shaker for 2 hours. The filtrate of the extract (using Whatman Paper No. 42) was determined for Cu and Zn using the iCE 3000 flame atomic absorption spectrophotometer (flame AAS) at  $\lambda = 324.7$  nm for Cu and  $\lambda = 213.9$  nm for Zn.

The data for evaluation 1 was evaluated using variance analysis (ANOVA) at 1 and 5% significant levels. The changes in the concentration of Cu and Zn over time were presented as the relative concentrations ( $R_M$ ), calculated by

$$R_M = 100 \times \frac{M}{M_0 - 1.5}, \quad (1)$$

where  $R_M$  was the relative concentration of heavy metal  $M$  (expressed in %),  $M$  is the concentration of heavy metal  $M$ , and  $M_0 - 1.5$  was the concentration of heavy metal  $M$  in the control plot (no compost and no lime) observed at 1.5 years which equaled to 122 mg·kg<sup>-1</sup> for Cu and 116.6 mg kg<sup>-1</sup> for Zn at the waste level of 60 Mg·ha<sup>-1</sup>, which were considered the highest. This data was needed to evaluate the trend of heavy metal immobilization over 20 years as affected by lime and/or compost.

The changes in the concentration of Cu and Zn over soil depth were also presented as  $R_M$ , calculated by

$$R_M = 100 \times \frac{M}{M_0}, \quad (2)$$

where  $R_M$  was the relative concentration of heavy metal  $M$  (expressed in %),  $M$  is the concentration of heavy metal  $M$  at a particular soil depth, and  $M_0$  was the highest concentration of  $M$  in the 60 Mg waste ha<sup>-1</sup> plot.

TABLE 2: The soil samples used in the evaluation of the Cu and Zn vertical and spatial distribution.

Soil plot	Industrial waste (W)	Lime (L) Mg·ha <sup>-1</sup>	Organic compost (C)
Control, CP	0	0	0
Low metal, LMP	15	0	0
High metal, HMP	60	0	0

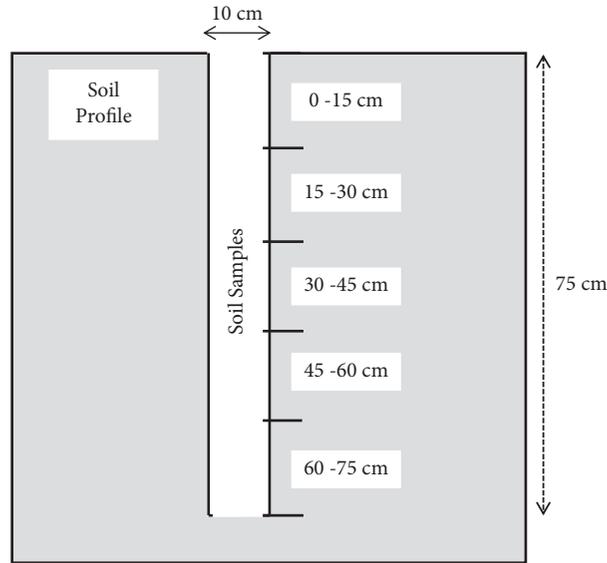


FIGURE 3: Depth-wise soil sampling with 15 cm intervals.

### 3. Results

**3.1. Changes in the Residual Cu and Zn over 20 Years after Waste Amendment.** The soil concentrations of Cu and Zn were in general enhanced by the addition of the industrial waste, as were also reported previously [42, 50]. Analysis of variance (ANOVA) shows the significant increases in the soil concentrations of Cu and Zn by waste amendment over 20 years after amendment in 1998 with the exception for Zn at 20 years (Tables 3 and 4).

Even though organic compost was reported to increase the soil adsorption capacity toward heavy metals, particularly right after treatment [23], it did not affect the retainment of Cu and Zn after a long time (Tables 3 and 4). However, when combined with waste or lime treatment, organic compost significantly decreased the soil concentrations of Cu and Zn (Tables 3 and 4) at 3 and 10 years after treatment. However, lime treatment increased the soil pH until 10 years while waste only until 3 years after amendment particularly in topsoils (Table 5). The significant effect of organic compost on soil concentrations of Cu and Zn was also observed at 3, 10, and 20 years when compost was combined with waste and lime (Tables 3 and 4).

The relative concentrations of Cu and Zn ( $R_{Cu}$  and  $R_{Zn}$ ) as affected by lime and/or cassava-leaf compost and time of sampling in plots with the highest waste addition of 60 Mg·ha<sup>-1</sup> (HMP) are shown in Figures 5 and 6, respectively. These data clearly show that in general the  $R_{Cu}$  and  $R_{Zn}$  decreased with time of sampling over 20 years. These

values indicate that the concentrations of Cu and Zn decreased with time. The expected effect of compost and/or lime, where compost and/or lime lowered the concentrations of Cu and Zn, was observed. In general, with some exception on the particular time of sampling, the  $R_{Cu}$  (Figure 5) and the  $R_{Zn}$  (Figure 6) follow the order of (control or no compost + no lime) > (C or with compost + no lime) > (L or no compost + with lime) > (C+L or with compost + with lime). This trend is more obvious until 3 years for Cu and until 10 years after the amendment for Zn.  $R_{Cu}$  and  $R_{Zn}$  decreased over 20 years to 17–53% for Cu and to 12–33% for Zn; their concentrations were in general lower with lime and/or compost.

**3.2. Vertical Spatial Distribution of the Residual Cu and Zn 21 Years after Waste Amendment.** The vertical spatial distribution of the residual Cu and Zn is depicted in Figures 7 and 8, respectively. The concentrations of Cu and Zn followed the order of high metal plot (HMP) > low metal plot (LMP) > control plot (CP). The concentrations of Cu and Zn were the highest in the 0–15 cm layers of LMP and HMP plots where waste was amended one time in 1998 and then in the 15–30 cm layers. Some significant amounts of Cu and Zn were observed in 30–60 cm layers, particularly in HMP, and insignificant amounts in CP and LMP.

The concentrations of Zn were lower at 20 years than those at 3 years (Figure 8). The concentrations of Cu were also lower at 20 years than those at 3 years in HMP but

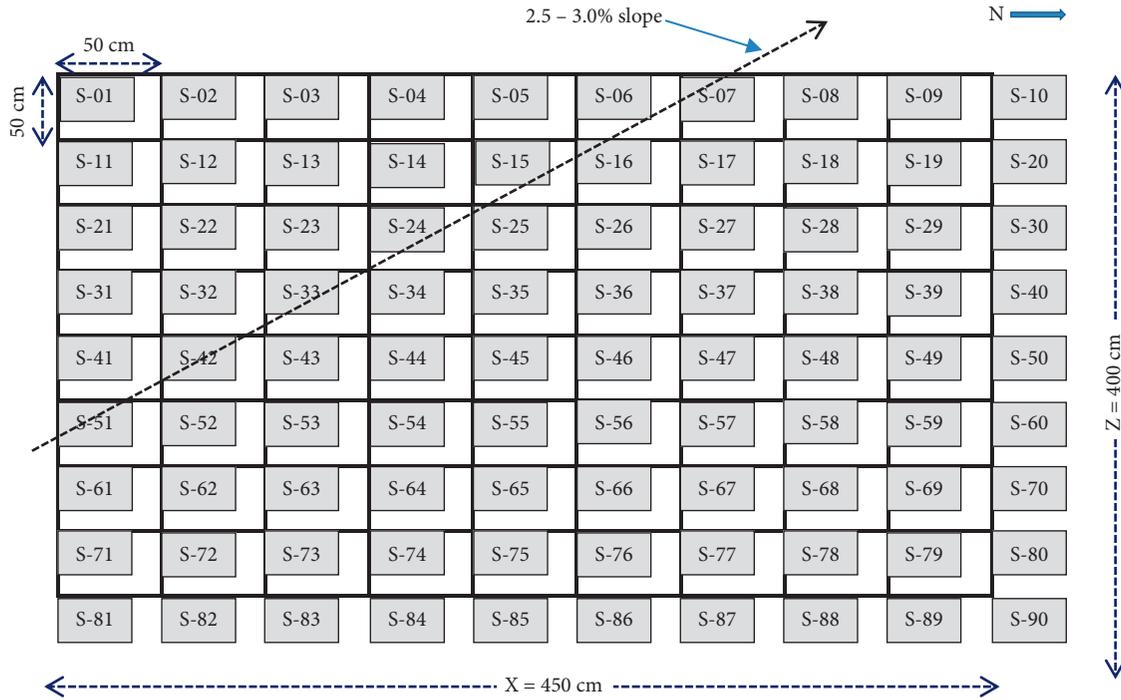


FIGURE 4: The sampling sites of soil in each experimental plot (the experimental plot measured 450 cm (X) × 400 cm (Z); the number of soil samples = 90 per plot, i.e., S-01 through S-90).

TABLE 3: The effect of the 20-year-old amendment of industrial waste, lime, and cassava-leaf compost on the soil Cu concentration.

Treatment	Time (years)							
	±1.5 <sup>1)</sup>		±3 <sup>1)</sup>		±10 <sup>1)</sup>		±20 <sup>2)</sup>	
	0-15 cm	15-30 cm	0-15 cm	15-30 cm	0-15 cm	15-30 cm	0-15 cm	15-30 cm
W	**	**	**	**	**	**	*	ns
C	ns	ns	ns	**	ns	ns	ns	ns
L	**	ns	ns	ns	ns	ns	ns	ns
WxC	ns	ns	ns	**	**	**	**	ns
WxL	ns	ns	**	ns	**	**	**	ns
CxL	ns	ns	ns	**	ns	ns	*	ns
WxCxL	ns	ns	**	**	ns	**	**	ns

Note: W = waste, C = cassava-leaf compost, and L = lime; \* = significant at 5%, \*\* = significant at 1%, and ns = not significant at 5% and 1% ANOVA; <sup>1)</sup>after [50] and <sup>2)</sup>after Susilowati and Salam (unpublished data 2018).

TABLE 4: The effect of the 20-year-old amendment of industrial waste, lime, and cassava-leaf compost on the soil Zn concentration.

Treatment	Time (years)							
	±1.5 <sup>1)</sup>		±3 <sup>2)</sup>		±10 <sup>3)</sup>		±20 <sup>4)</sup>	
	0-15 cm	15-30 cm	0-15 cm	15-30 cm	0-15 cm	15-30 cm	0-15 cm	15-30 cm
W	**	**	**	**	**	**	ns	ns
C	ns	ns	ns	ns	ns	ns	ns	ns
L	**	*	ns	ns	ns	ns	ns	ns
WxC	ns	ns	*	ns	**	**	*	ns
WxL	ns	**	*	ns	**	**	**	*
CxL	ns	ns	**	ns	*	*	*	*
WxCxL	ns	ns	**	ns	*	**	*	*

Note: W = waste, C = cassava-leaf compost, and L = lime; \* = significant at 5%, \*\* = significant at 1%, and ns = not significant at 5% and 1% ANOVA; <sup>1)</sup>after Amirulloh and Salam (unpublished data 2000), <sup>2)</sup>after [42], <sup>3)</sup>after [50], and <sup>4)</sup>after Susilowati and Salam (unpublished data 2018).

TABLE 5: The effect of the 20-year-old amendment of industrial waste, lime, and cassava-leaf compost on the soil pH.

Treatment	Time (years)							
	$\pm 1.5^1$		$\pm 3^2$		$\pm 10^3$		$\pm 20^4$	
	0–15 cm	15–30 cm	0–15 cm	15–30 cm	0–15 cm	15–30 cm	0–15 cm	15–30 cm
W	*	ns	**	ns	ns	ns	ns	ns
C	ns	ns	ns	ns	*	ns	ns	ns
L	**	**	**	**	**	**	ns	ns
WxC	ns	ns	ns	*	ns	ns	ns	ns
WxL	**	ns	ns	ns	**	**	ns	ns
CxL	ns	ns	**	ns	**	**	ns	ns
WxCxL	ns	ns	ns	ns	ns	*	ns	ns

Note: W = waste, C = cassava-leaf compost, and L = lime; \* = significant at 5%, \*\* = significant at 1%, and ns = not significant at 5% and 1% ANOVA; <sup>1</sup>after Amirulloh and Salam (unpublished data 2000), <sup>2</sup>after [42], <sup>3</sup>after [50], and <sup>4</sup>after Susilowati and Salam (unpublished data 2018).

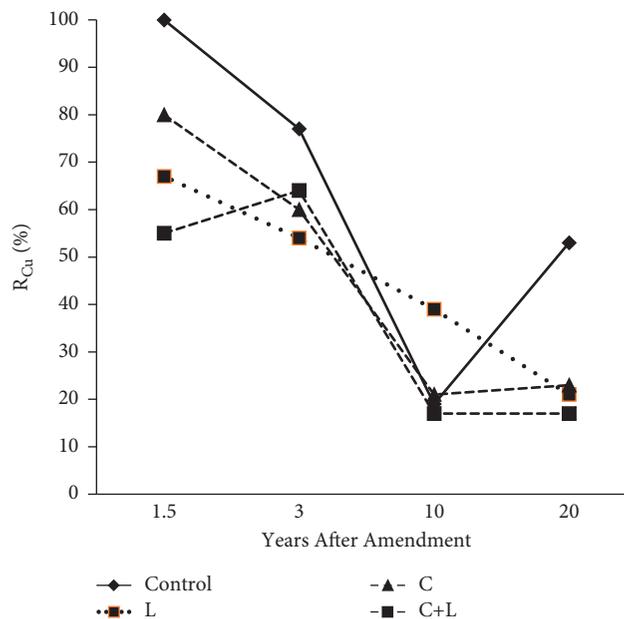


FIGURE 5: The relative concentrations of Cu ( $R_{Cu} = 100 \times Cu/Cu_{0-1.5}$ ) in topsoil amended with 60 Mg waste  $ha^{-1}$  (HMP) (after Amirulloh and Salam (unpublished data, 2000) for 1.5 years, after [42] for 3 years, after [50] for 10 years, and after Susilowati and Salam (unpublished data 2018) for 20 years; the concentration of Cu in the topsoil of control at 1.5 years ( $Cu_{0-1.5}$ ) was  $122 \text{ mg}\cdot\text{kg}^{-1}$  (L: lime at  $5 \text{ Mg}\cdot\text{ha}^{-1}$ ; C: cassava-leaf compost at  $5 \text{ Mg}\cdot\text{ha}^{-1}$ ).

higher in 20 years than those at 3 years in CP and LMP (Figure 7). Since the industrial waste was given only one time, this phenomenon indicates the decrease in soil heavy metal concentration with time.

**3.3. Horizontal Spatial Distribution of the Residual Cu and Zn 21 Years after Waste Amendment.** The horizontal spatial distributions of Cu and Zn in the 0–15 cm layer of the soil in the experimental plots are depicted in Figures 9 and 10, respectively. The Cu and Zn in the topsoils were not evenly distributed. The concentration of Cu was on average below  $10 \text{ mg}\cdot\text{kg}^{-1}$  in the CP and LMP ranging from  $<10$  to about  $55 \text{ mg}\cdot\text{kg}^{-1}$  in HMP (Figure 9). However, the Cu in topsoils treated with  $60 \text{ Mg}$  waste  $ha^{-1}$  (HMP) was observed significantly high.

Similar phenomena were also observed in the concentration of Zn in the 0–15 cm of the soil. The concentrations of Zn in the plots were also not evenly distributed in topsoils. The average concentration of Zn was  $<5 \text{ mg}\cdot\text{kg}^{-1}$  in CP, ranging from about 0 to  $12 \text{ mg}\cdot\text{kg}^{-1}$  in LMP and ranging from about 5 to  $60 \text{ mg}\cdot\text{kg}^{-1}$  in HMP (Figure 10). The concentrations of Zn in topsoils treated with  $60 \text{ Mg}$  waste  $ha^{-1}$  (HMP) were also observed significantly high. The concentrations of Cu and Zn were also observed to be higher in the northwest parts of the plots, particularly in plots treated with  $60 \text{ Mg}$  waste  $ha^{-1}$  (Figures 9 and 10).

The concentrations of Cu and Zn in waste amended plots (LMP and HMP) significantly decreased over the period of 1998–2019. On the other hand, those in the control (CP) increased. The changes in the concentrations of Cu and Zn over the last 21 years are listed in Table 6.

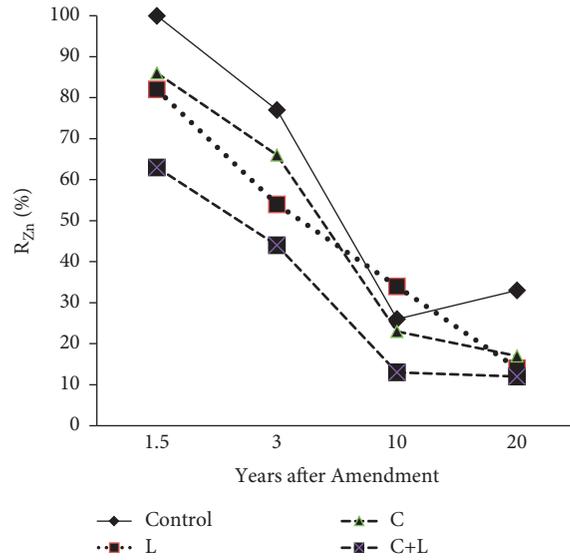


FIGURE 6: The relative concentrations of Zn ( $R_{Zn} = 100 \times Zn/Zn_{0-1.5}$ ) in topsoil amended with 60 Mg waste  $ha^{-1}$  (HMP) (after Amirulloh and Salam (unpublished data, 2000) for 1.5 years, after [42] for 3 years, after [50] for 10 years, and after Susilowati and Salam (unpublished data, 2018) for 20 years; the concentration of Zn in the topsoil of control at 1.5 years was 116.6  $mg \cdot kg^{-1}$  ( $Zn_{0-1.5}$ ) (L: lime at 5  $Mg \cdot ha^{-1}$ ; C: cassava-leaf compost at 5  $Mg \cdot ha^{-1}$ ).

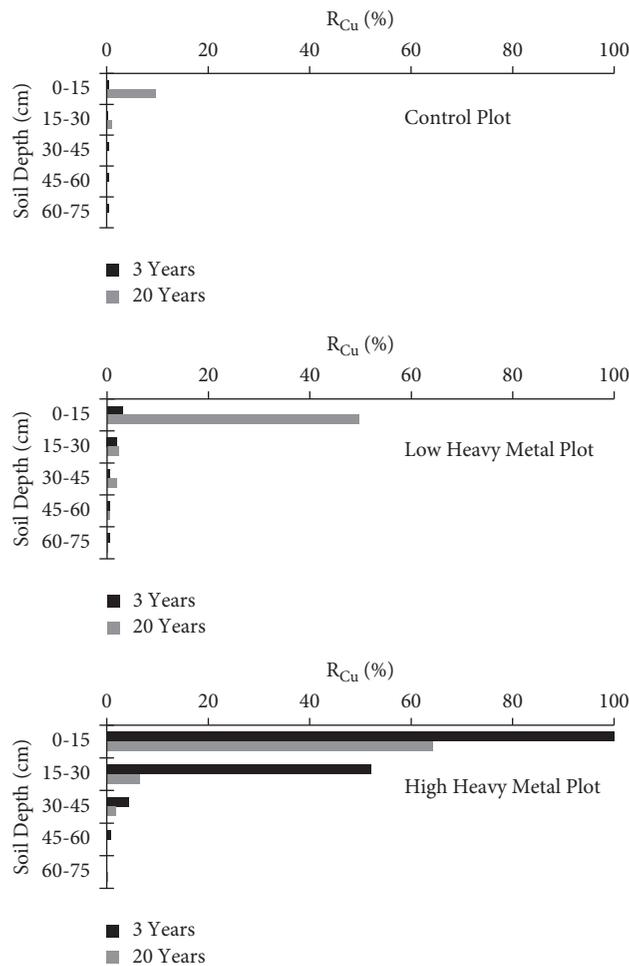


FIGURE 7: The vertical spatial distribution of soil Cu 3 years<sup>1)</sup> and 20 years<sup>2)</sup> after amendment with Cu-Zn containing industrial waste ( $R_{Cu}$  is the ratio of the actual concentration to the highest concentration of Cu in the 60 Mg waste  $ha^{-1}$  plot (93.3  $mg \cdot kg^{-1}$ ); waste at CP = 0, LMP = 15, and HMP = 60  $Mg \cdot ha^{-1}$ ; <sup>1)</sup>after [42]; <sup>2)</sup>after Fernando and Salam (unpublished data 2018)).

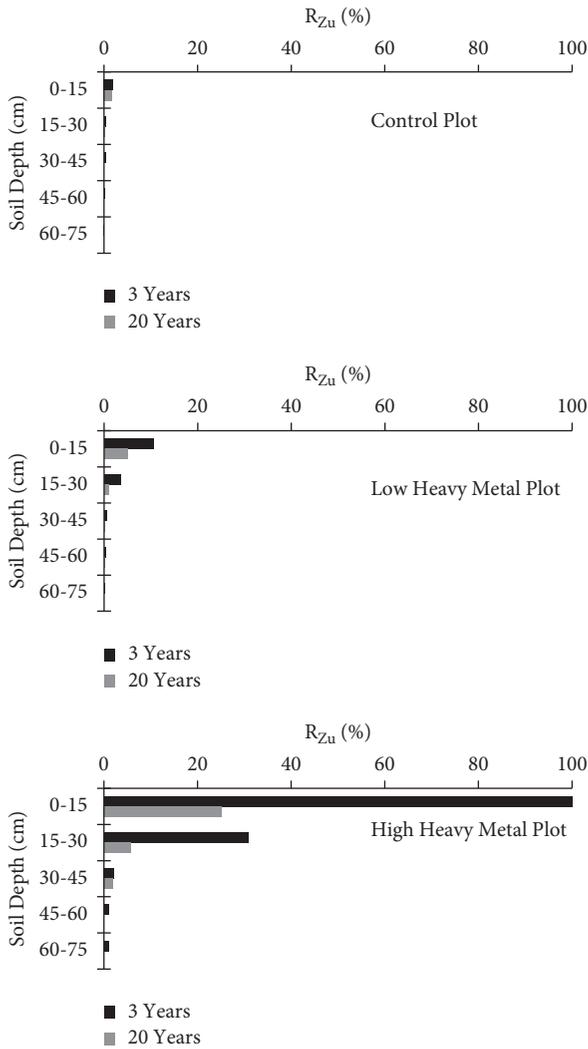


FIGURE 8: The vertical spatial distribution of soil Zn 3 years<sup>1)</sup> and 20 years<sup>2)</sup> after amendment with industrial waste ( $R_{Zn}$  is the ratio of the actual concentration to the highest concentration of Zn in the 60 Mg waste ha<sup>-1</sup> plot (90.0 mg·kg<sup>-1</sup>); waste at CP=0, LMP = 15, and HMP = 60 Mg waste ha<sup>-1</sup>; <sup>1)</sup>after [42]; <sup>2)</sup>after Fernando and Salam (unpublished data 2018).

#### 4. Discussion

The increases in the soil concentrations of Cu and Zn by waste amendment observed over 20 years as shown in Figure 5 through Figure 10 clearly indicate that parts the added Cu and Zn amended in 1998 stayed in topsoil and/or subsoil for a quite long time. The DTPA-extracted Cu and Zn in the last observations in the 60 Mg waste ha<sup>-1</sup> plots (HMP) 20 years after the amendment were 17–53% for Cu and 12–33% for Zn (Figures 5 and 6). Most of the amended Cu and Zn disappeared or were unextracted by DTPA 20 years after the amendment.

The high precipitation rates in the tropics in fact did not leach out all the Cu and Zn from soils. Parts of these metals were retained by soils, most probably through adsorption reactions by the soil colloid variable charges. The presence of precipitation reactions may have also possibly occurred

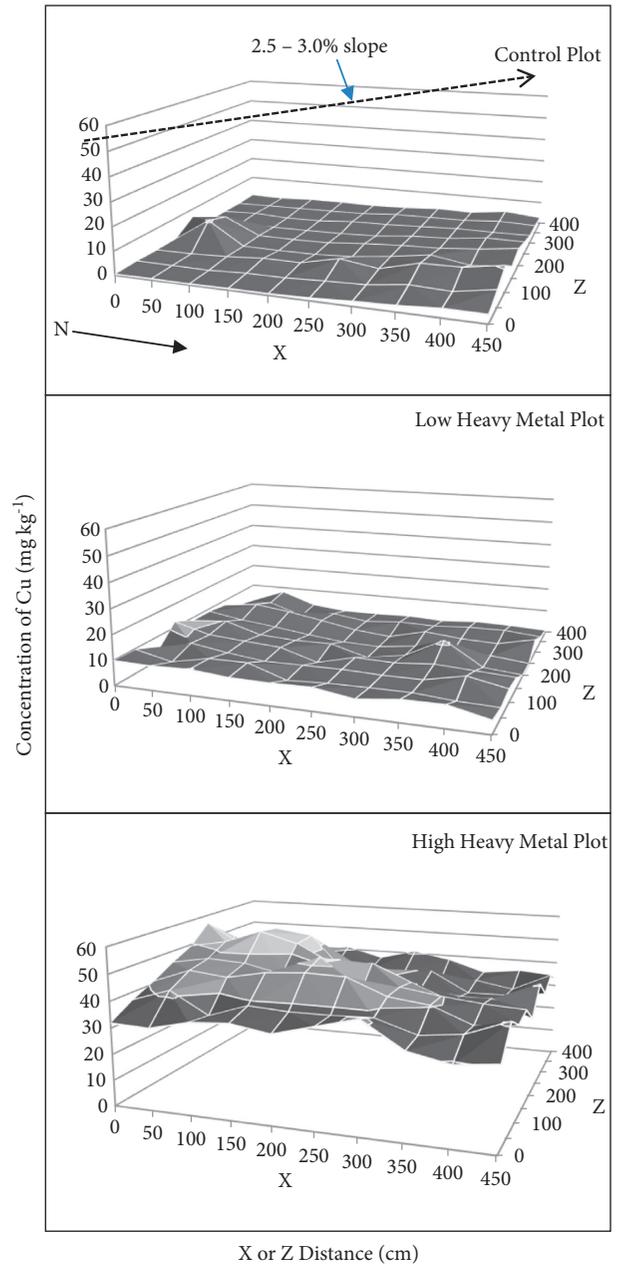


FIGURE 9: The horizontal spatial distribution of Cu in the waste-treated plots 21 years after amendment (control plot = 0, low metal plot = 15, and high metal plot = 60 Mg waste ha<sup>-1</sup>; plots measured 450 cm (X) x 400 cm (Z)).

because the addition of waste and/or lime and/or compost also significantly increased the soil pH, particularly until about 10 years since their amendment in 1998 (Table 5). This observation shows that the tropical soil was effective in retaining part of heavy metals for a quite long time. The effectiveness of this soil in retaining soil Cu and Zn increased in the presence of organic compost and/or lime (Figures 5 and 6). In general, organic compost and/or lime increased the soil adsorption capacity and thereby effectively lowered the soil concentrations of Cu and Zn. As reported, organic compost may have provided some organic functional groups

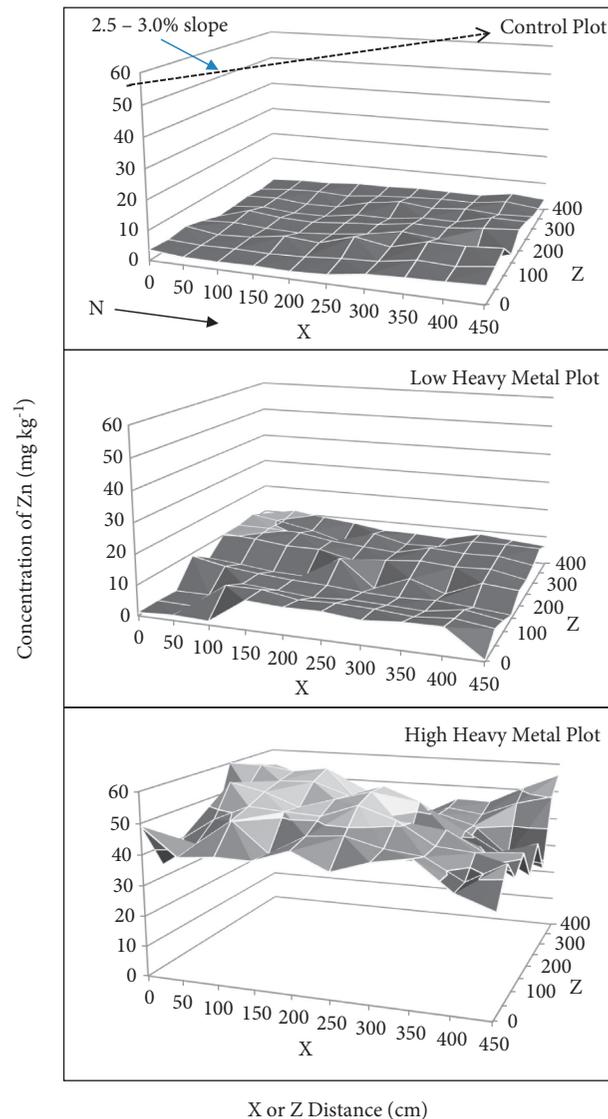


FIGURE 10: The horizontal spatial distribution of Zn in the waste-treated plots 21 years after amendment (control plot=0, low metal plot=15, and high metal plot=60 Mg waste ha<sup>-1</sup>; plots measured 450 cm (X) x 400 cm (Z)).

which may, when combined with the increase in soil pH, have increased the soil negative charges able to increase the Cu and Zn immobilization by soils [20–24].

This inability of organic compost alone to lower the concentrations of Cu and Zn like right after amendment in 1998 [23] (Tables 3 and 4) was obviously caused by the probable quick decays of organic matter over time, particularly in the tropics where the relatively high moisture content may accelerate the enzymatic organic matter decomposition [53]. The decomposition process of organic matter may lower the effectiveness of organic matter in decreasing the soil concentrations of Cu and Zn. Based on their researches, [43] reported that organic matter incorporation was indeed useful to retain part of heavy metals in mine soils, but they suggested that the decomposition of organic matter by soil microorganisms may cause the release of heavy metals to more readily available forms in the future. It was also reported that the oxidation of soils with dilute

H<sub>2</sub>O<sub>2</sub> significantly decreased the adsorbed Cu and slightly decreased the adsorbed Zn onto organic substances [44]. Organic matter decomposition may then in general increase the Cu and Zn mobility and availability in soils. More durable organic matter like biochar may probably give a more significant effect over a longer time of process [54].

However, when combined with waste or lime, organic compost significantly decreased the soil concentrations of Cu and Zn (Tables 3 and 4, Figures 5 and 6) for some time of observation. The increase in soil pH due to waste or lime treatment (Table 5) may have enhanced the adsorption sites on the remaining organic matter functional groups. This process may have eventually decreased the soil concentrations of Cu and Zn. The significant effect of organic compost was also observed after 3, 10, and 20 years when compost was combined with waste and lime (Tables 3 and 4, Figures 5 and 6). The presence of OH<sup>-</sup> ions at higher soil pH by lime may have detached bonded H on the organic matter functional

TABLE 6: The changes in the concentrations of Cu and Zn in soil over 21 years from 1998 to 2019.

Metal	Waste treatment in 1998 <sup>1)</sup> mg·ha <sup>-1</sup>	Concentration in		Changes over 21 years	
		1998 <sup>2)</sup>	2019 <sup>3)</sup>		%
Cu	0	0.240	2.43	+2.19	+9.12
	15	89.0	4.90	-84.1	-94.0
	60	94.2	39.6	-54.6	-54.6
Zn	0	0.98	3.51	+2.53	+258
	15	61.2	6.10	-55.1	-90.0
	60	53.5	45.2	-8.35	-15.0

<sup>1)</sup>Waste was amended one time; <sup>2)</sup>after Suryanti and Salam (unpublished data 1998); <sup>3)</sup>average of 3 blocks.

groups causing the increase in the adsorption sites that were important for heavy metal adsorption [20–24].

At an earlier time, i.e., at  $\pm 1.5$  and  $\pm 3$  years, the role of lime and/or cassava-leaf compost was clearly observed (Figures 5 and 6). The soil concentrations of Cu (Figure 5) and Zn (Figure 6) were depressed by compost and/or lime treatment. The organic matter enhanced the soil capacity to retain heavy metal cations as negatively charged functional groups and the raised soil pH by lime enlarged the quantity of the negatively charged organic matters and soil clays [12, 55] that were able to retain more Cu and Zn cations. However, the role of these factors faded with the time of sampling more than 3 years. After 3 years, the presence of compost solubilized Cu and Zn. However, the interactions of lime and compost in the presence of waste were in general observed to lower the soil concentrations of Cu and Zn (Figures 5 and 6). The chemical reactions which include adsorption and precipitation of Cu and Zn might have also been involved in decreasing the soil concentration of Cu and Zn over 20 years. Since the initial soil pH was low (pH = 5.11), the adsorption reaction probably dominated [12, 23]. Even though the soil CEC was relatively low, to some extent, the Cu and Zn adsorption may have caused a decrease in the concentrations of Cu and Zn in soil [12].

As clearly shown in Figure 5,  $R_{Cu}$  in the 60 Mg waste ha<sup>-1</sup> plot (HMP) decreases with the time of sampling. For example,  $R_{Cu}$  at  $\pm 1.5$ ,  $\pm 3$ ,  $\pm 10$ , and 20 years is 100, 77, 19, and 53%, respectively (Figure 5). A similar pattern was observed for Zn (Figure 6).  $R_{Zn}$  in general also decreases with the time of sampling. These values indicate that the soil concentrations of Cu and Zn decreased with time. Some of the Cu and Zn were removed from their labile pools and were shifted to other forms over time depending on compost and/or lime treatment. Several possibilities may explain the phenomenon of Cu and Zn decreases over time in Figures 5 and 6. The heavy metals released into the soil solution may have encountered various fates as suggested by [31, 55] shown in Figure 11, which include absorption by plants [32, 34] or leached through the soil bodies [29, 40] or transported away by mass flow, diffusion, water runoff and erosion, and plowing/tillage [35, 37, 39, 41, 56, 57].

One of the important physical processes that may have changed the concentrations of Cu and Zn was the heavy metal movement through mass flow and diffusion. Mass flow follows the direction of water movement which

includes heavy metal leaching by percolating water. The high rain intensity during the rainy season in South Lampung might have fastened the movement of Cu and Zn from topsoils to subsoils through mass flow and leaching. As previously mentioned, the total rainfall in the area was 1.628 mm in 2015 with 143 total rainy days [51]. The higher concentrations of Cu and Zn in subsoils (soil depths >0–15 cm) in plots amended with industrial waste (LMP and HMP) than those in CP (Figures 7 and 8) suggest that Cu and Zn may have one way or another moved from topsoils to subsoils. The current data (Figures 7 and 8) also show that the concentrations of Cu and Zn were the highest in the 0–15 cm layers of LMP and HMP where waste was amended in 1998 even though in HMP significant amounts of Cu and Zn were also observed in 30–60 cm layers. The movement is, however, minuscule, particularly to soil depth >30 cm. Figures 7 and 8 show that in general the concentrations of Cu and Zn at 20 years were lower than those at 3 years, with the exception of Cu at waste level  $\leq 15$  Mg·ha<sup>-1</sup>. The paper [42] also previously showed that after  $\pm 3$  years, Cu and Zn were leached in smaller amounts, and, overall, the Cu and Zn were not significantly moved to subsoils. Most of the heavy metals may have been adsorbed onto the higher energy sites and probably also absorbed by plant roots.

The physical processes like plowing and harrowing were probably the most important in moving the masses of Cu and Zn. These physical activities may have shifted the heavy metals in soils out of the plot boundaries and to other adjacent and nearby plots. The decreases in Cu and Zn concentrations in LMP and HMP and the increases in their concentrations in CP over 20 years after waste amendment (Table 6) support this suggestion. The higher concentrations of Cu at 20 than those at 3 years in 0–15 cm layers of CP and LMP (Figure 7) may also support these suggestions. Parts of Cu and Zn from the adjacent LMP and HMP may have been transported to CP. Plowing may have also exchanged the masses of topsoils and subsoils and surely decreased the new topsoil concentrations of Cu and Zn. These phenomena were in accordance with those reported by [41]. The significant decreases in the topsoil Cu and Zn concentrations (Table 6) were probably also caused by this physical process. The paper [42] also shows that the concentrations of Cu and Zn in the underlying soil layers in plots treated with the industrial waste were significantly higher than the same layers in the control plots. The underlying layers of heavy metals

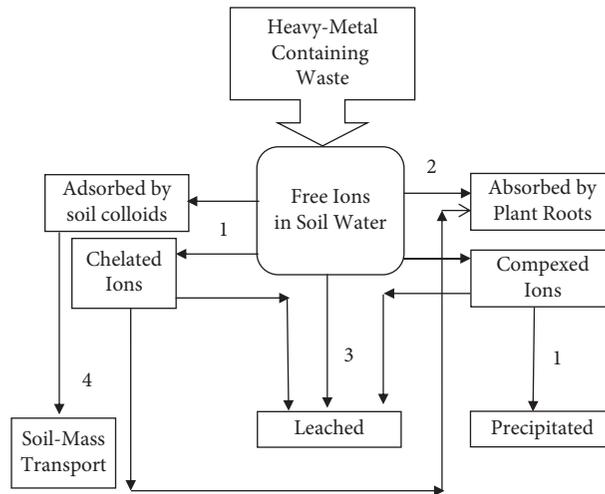


FIGURE 11: The possibilities of the waste-origin Cu and Zn removals from soil labile fractions after waste amendment (1: adsorption or precipitation, 2: absorption by plant roots, 3: leaching, 4: soil mass transport) [50, 55].

were probably contributed by topsoil Cu and Zn in which the heavy metals were initially given in topsoils (0–15 cm) in 1998. Therefore, the Cu and Zn may have moved by soil tillage (particularly that in 0–30 cm) and also by water leaching. However, most Cu and Zn residues were observed to remain in topsoils.

The horizontal movement of Cu and Zn like this may have also occurred by water runoff and soil erosion suggested by [36] because the experimental land in Sidosari tilted about 2.5–3.0% to the northwest direction (Figure 2). Figures 9 and 10 show that the concentrations of Cu and Zn were generally higher in the northwest corners of the plots than those in other parts of the plots, particularly in plots with 60 Mg waste ha<sup>-1</sup>. The plowing and harrowing may ease the water runoff and soil mass erosion. Therefore, soil tillage may cause not only vertical but also horizontal movement of soil mass containing heavy metals.

In addition to the physical and chemical processes, the extraction of Cu and Zn by plants grown and harvested from the experimental plots might have also caused significant decreases in the soil concentrations of Cu and Zn [13]. The experimental soils in Sidosari had been planted with various plant crops including cassava (*Manihot utilissima*), upland rice (*Oryza sativa*), spinach, and corn (*Zea mays*) since the industrial waste was amended in 1998. As cited by [13, 29], crop removal and leaching may have been the main causes of heavy metal losses from the soil system. The results of all these factors may have eventually decreased the concentrations of Cu and Zn in topsoils. However, this suggestion needs further research.

Our current glasshouse investigation using the soil samples from CP, LMP, and HMP showed that water spinach (*Ipomoea aquatica*) was able to decrease the soil Zn by as much as 38.5% in CP, 21.6% in LMP, and 16.0% in HMP. Other investigations also revealed that the uptake of Cu increased with corn plant biomass with a high correlation coefficient ( $R^2 = 0.95^*$ ). The decrease in soil Cu

and Zn was assumed to be caused by plant absorption and immobilization by soils because plants were watered capillary. Therefore, no heavy metal leaching occurred.

The above experimental data clearly showed that significant parts of amended Cu and Zn were immobilized by soil colloids for a quite a long time. Some other parts were translocated through several chemical, physical, and biological mechanisms like soil chemical immobilization, physical movement such as soil erosion, plowing, and leaching, and crop removals.

## 5. Conclusion

Significant increases in the soil concentrations of Cu and Zn caused by waste were observed over 20 years after treatment. The increase in pH by lime was observed over 10 years but its effect on the soil concentrations of Cu and Zn was observed only at 1.5 years, particularly in topsoils. However, the effect of lime was observed over 20 years when waste and/or compost were also given. Similarly, the effect of compost was absent over 20 years but was significant when waste and/or lime were also given. The relative concentration of Cu and Zn in the 60 Mg waste ha<sup>-1</sup> plots decreased over 20 years to 17–53% for Cu and to 12–33% for Zn; their concentrations were, in general, lower with lime and/or compost. Copper and Zn were not evenly distributed over each plot. The relative concentrations of Cu and Zn significantly decreased over 21 years in plots with 15 or 60 Mg·ha<sup>-1</sup> and increased in the control plots. The concentrations of Cu and Zn were the highest in the 0–15 layers of low metal plots and high metal plots where waste was amended in 1998 and then in 15–30 layers. Some significant amounts of Cu and Zn were observed in 30–60 cm layers, particularly in high metal plots, indicating Cu and Zn vertical movement. Heavy metals moved in soil mostly through soil tillage and erosion and partially through leaching.

## Data Availability

The raw and analyzed data used to support 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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