Tropical Soil Labile Fractions of Copper in Experimental Plots ±Ten Years after Treatment with Copper-Containing-Waste

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Abstract

Copper is reported to be retained in soils for a quite long time particularly in soil treated with some amendments. This research was intended to evaluate the soil labile fractions of Cu ±10 years after treatments with Cu-containing industrial waste, lime, and cassava-leaf compost. Soil samples were taken from topsoils and subsoils of ±10 years old experimental plots set in 1998 and factorially treated with a metal-spoon industrial waste at 0, 15, and 60 ton ha^{-1}, lime at 0 and 5 ton ha^{-1}, and cassava-leaf compost at 0 and 5 ton ha^{-1}. The measured Cu labile fractions were compared to those in soils sampled ±1.5 years and ±3 years after treatments. The results showed that the soil Cu labile fractions in waste treated soil were higher than those in the control treatments even though their concentration decreased with the years of sampling. Lime showed a decreasing effect on soil labile Cu fractions, but the effect decreased with the years of sampling. Effect of cassava-leaf compost was in general not observed ±10 years after treatment.

Keywords: Cassava-Leaf Compost, Copper, Heavy Metals, Industrial Waste, Lime

Introduction

Heavy metals are by definition the elements with relatively high atomic density of > 6 g cm^{-3} or specific gravity of > 5 g cm^{2}. This category of elements is one of the research major focuses in soil science because heavy metals are very important in the environment. Some heavy metals are needed by plants and animals at relatively small amounts. For example, Cu and Zn are considered microelements for plant growth; therefore, they are needed by plants, even though only at relatively small amounts. On the contrary, some of the elements such as Cd and Pb are not needed by the living things. Both categories of these elements show similarity, they are toxic to the living things at concentrations relatively higher than their critical levels. For examples, the total concentrations of

Cu in soils must not exceed 60 mg kg^{-1}; that of Zn must be below 70 mg kg^{-1}; while Cd and Pb must be < 9 and 100 mg kg^{-1}, respectively (Ross 1994a). These values are toxic boundaries for plants.

However, the concentrations of heavy metals in soils are subjected to natural and anthropogenic sources (Juracek and Ziegler 2006; Biasioli et al. 2007; Benke et al. 2008; Berenguer et al. 2008; Lin et al. 2008; Cakmak et al. 2010; Alloway 2012; Tu et al. 2012; Kargar et al. 2013; Adams et al. 2014). Berenguer et al. (2008) reported that long-term use of liquid swine manure significantly elevated the concentrations of Cu and Zn in soils. Long-term application of cattle manure significantly increased the soil total and EDTA-extractable Cu and Zn (Benke et al. 2008). The concentrations of Cu, Ni, Hg, Pb, Cd, and As in sediment samples at electronic waste recycling plant at Guiyu, Guangdong, China significantly increased above the reference background concentrations (Guo et al. 2009). Therefore, heavy metals in the soil environment must be one of our concern.

Heavy metals in soil environment are distributed in several foms with decreasing bioavailability: free ions, complex ions, exchangeable forms, precipitates, and minerals (Salam 2017). Among these, free ions and to some extent complex ions and exchangeable forms, are the most mobile and potential to affect the living things because these forms are directy related to plant root absorption and heavy metal toxicities (Salam 2001; Daoust et al. 2006; Salam 2017). Therefore, the behaviors of these labile fractions of heavy metals in soils must be understood. This may include their behaviours related to the dynamics of some soil key properties such as soil cation exchange capacity (CEC), pH, and reaction time.

Soil adsorptive capacity – part of it is expressed as CEC – is by no means the most important soil property for heavy metal immobilization. This property may be improved by addition of ameliorants. For examples, some researchers empoyed organic matters/biosolids (Salam et al. 2001 2005 2017; Tokunaga et al. 2003; Brown et al. 2004; Stehouwer et al. 2006; Schroder et al. 2008; Brown et al. 2009; Kukier et al. 2010; Mamindy-Pajany et al. 2014; Tang et al. 2014; Pukalchik et al. 2017) or P fertilizers such as TSP and rock phosphates (Brown et al. 2004; Kilgour et al. 2008;

Moseley et al. 2008). Organic matters are believed to give functional groups such as phenolic and carboxyls in significant amounts and to increase soil CEC (Ross 1994b; Parfitt et al. 1995; Rodella et al. 1995; Salam 2001). Lin et al. (2008) reported that Cu tended to be preferentially retained by Fe-oxides and organic matters. Some other ameliorants were also developed to immobilize heavy metals by precipitation, for examples by employing Na_2HPO_4, hidroxyapatite, or rock phosphates (Ma et al. 1990 1994 1995; Rabinowitz 1993; Ruby et al. 1994).

As repeatedly reported by several researchers (Salam 1999 2001; Adams et al. 2004; Bang and Hesterberg 2004; Quaghebeur et al. 2005; He et al. 2006; Brown et al. 2009; Bolan et al. 2014; Malinowska 2017), the soil adsorptive capacity or CEC is positively related to the changes in soil pH. In general, soil CEC increases with the increase in pH of soils with variable charges. The increase is due to dehydogenation of soil particle surfaces in the presence of increased concentration of OH^-. The increase in soil CEC increases the adsorption of heavy metals. For example, it was reported that amendment of soil contaminated with industrial waste with 5 ton ha^{-1} $CaCO_3$ significantly increased the soil pH dan decreased the labile heavy metals extracted by chemical extractors (Salam 2000 2001 2017). This behavior then increases the residence time of heavy metals in soils and decreases their availabilities to plants and mobilities in soil environment. Daoust et al. (2006) lately reported that Cu partitioning and its toxicity were significantly affected by pH greater than by organic matter and clay content.

This research was intended to evaluate the soil labile fraction of Cu ±10 years after soil treatments with Cu-containing industrial waste, lime, and cassava-leaf compost.

Materials and Methods

Soil samples were taken from topsoils and subsoils in the experimental plots set in July 1998, located in Sidosari, Natar, South Lampung. Treatments were previously set in a randomized block design, consisted of 3 factors: industrial waste of metal spoon, lime as dolomite, and cassava-leaf compost, with 3 replications. Industrial waste was metal-spoon industry of PT Star Metal Wares

Jakarta, containing high Cu and Zn. Some chemical properties of the industrial waste were pH 7.30, Pb 2.44 mg kg^{-1}, Cd 0.12 mg kg^{-1}, Cu 754 mg kg^{-1}, and Zn 44.6 mg kg^{-1} (Salam et al. 2005).

Industrial waste was given and thoroughly plowed to 30 cm soil depth at rates: 0, 15, and 60 ton ha^{-1}. Lime and cassava-leaf compost were both given and thoroughly mixed to the same soil depth one week later at 0 and 5 ton ha^{-1}. Lime was ($CaMg(CO_3)_2$). Casava-leaf compost was prepared as reported previously by Salam (2001). Rates of industrial waste, lime, and cassava-leaf compost were selected through a series if preliminary experiments reported previously (Salam 2000). Each plot measured 4.5 m long and 4 m wide, 50 cm apart between plots and 100 cm apart between blocks. Complete experimental treatments are listed in Table 1. The experimental plots were since 1998 planted with corn, dryland paddy, and left bare in between.

Soil sampling was conducted in 15 February 2009 (±10 years after treatment). Composite topsoil (0-15 cm) and subsoil (15-30 cm) samples were taken diagonally from 5 points in each plot. Soil samples were air-dried, ground to pass a 2-mm-sieve, and mixed thoroughly before analysis. Analysis included soil Cu labile fraction with DTPA method (Baker and Amacher 1987) and soil pH with a pH electrode. This data was compared to those of ±1.5 years after treatment (Amirulloh 2000) and ±3 years after treatment (Prihatin 2002).

Insert Table 1

Results and Discussion

Copper was chemically retained in soil body for more than 10 years since soil treatment with Cu-containing industrial waste in July 1998. Analysis of variance (Anova) shows that the industrial waste significantly enhanced the soil labile factions of Cu (Table 2). Test of Least Significant Difference (LSD-Test) also shows the significance of this effect, particularly by the addition of the industrial waste of 60 ton ha^{-1} (Table 3). The concentration of Cu at the addition of 60 ton ha^{-1} was 3.93 times compared to that at Control at 0 – 15 cm, while that at 15- 30 cm was 2.55 times. The addition of industrial waste at 15 ton ha^{-1} also shows tendencies to increase the concentrations of Cu; 1.82 times and 1.49 times at 0 – 15 and 15 – 30 cm, respectively. The increases were clearly attributed to the fact that the industrial waste contained relatively high amount of Cu (Salam et al. 2005).

Insert Table 2

Insert Table 3

However, the concentrations of soil labile Cu in general decreased significantly with year of sampling, particularly at the addition level of 60 ton ha^{-1}. The concentrations of labile Cu at ±1.5 years were higher than those at ±3 years and those at ±3 years were higher than those at ±10 years (Figs. 1, 2, and 3). The relative concentrations of labile Cu in topsoils treated with 60 ton waste ha^{-1} and 5 ton lime ha^{-1} decreased from 0.67 at ±1.5 years to 0.54 at ±3 years and to 0.39 at ±10 years. The relative concentration of Cu is expressed as Cu/Cu_0-1.5 i.e. the ratio of the concentration of Cu to that of the Control (No Compost and No Lime) at ±1.5 years. The relative concentrations of Cu at waste level of 60 ton ha^{-1} are listed in Tabel 4.

Insert Table 4

The decreasing trend in the concentration of Cu for the last ±10 years was due to several possibilities: (1) enhanced retainment by soil adsorptive surfaces or by precipitation, (2) absorbed by plant roots, and (3) leached by percolating water to subsoil or ground water, as described in Fig. 4.

Based on the observation, the first possibility was unlikely. First, at ±10 years after treatment, the effect of lime had decreased significantly and, based on Anova, the effect on decreasing Cu labile fractions was not significant (Table 1). Second, the interaction of Industrial Waste of 15 ton ha^{-1} with lime tended to decrease the concentration of soil labile Cu. However, the effect was not significant; and, on the contrary, the interaction with Industrial Waste of 60 ton ha^{-1} in fact increased the concentration of soil labile Cu. The interaction of Industrial Waste and Cassava-Leaf Compost also significantly decreased the soil labile Cu. However, this material was easily decomposed. Therefore, the logical possibility causing the decrease in the concentrations of labile Cu with time was Cu absorption by plant roots or Cu translocation into subsoil or ground water (Fig. 4). The complexation of Cu with soil humic substances may have mobilized Cu ions so that it was more easily transported and/or absorbed by plant roots. In fact, Cu is more easily complexed by organic substances.

Insert Fig. 1

Insert Fig. 2

Insert Fig. 3

Insert Fig. 4

Summary and Conclusion

Copper of industrial-waste-origin was retained in soil for more than 10 years. The concentration of labile Cu decreased with year of soil sampling. However, part of the Cu from industrial waste was translocated, probably absorbed by plant roots and/or moved into subsoil and/or ground water through soil body.

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Table 1. The existing treatment units in the experimental plots at Sidosari, Natar, Lampung.

Waste/W	Lime /L	Compost/C	
		C_0	C_1
W_0	L_0	$W_0L_0C_0$	$W_0L_0C_1$
	L1	$W_0L_1C_0$	$W_0L_1C_1$
W_1	L_0	$W_1L_0C_0$	$W_1L_0C_1$
	L_1	$W_1L_1C_0$	$W_1L_1C_1$
W_2	L_0	$W_2L_0C_0$	$W_2L_0C_1$
	L_1	$W_2L_1C_0$	$W_2L_1C_1$

Notes:
W = Waste (W_0 0, W_1 15, and W_2 60 ton ha^{-1}), L = Lime (L_0 0 and L_1 5 ton ha^{-1}); and C = Compost (C_0 0 and C_1 5 ton ha^{-1}).

Table 2. Analysis of variance of the changes in labile Cu concentration in a tropical soil treated with Cu-containing industrial waste, lime, and cassava-leaf compost after a period of time since treatment (Transf √x) (Ginanjar 2009).

Treatment	Time (years)					
	± 1.5 [1)]		± 3 [2)]		± 10	
	0-15 cm	15-30 cm	0-15 cm	15-30 cm	0-15 cm	15-30 cm
W	**	**	**	**	**	**
C	ns	ns	ns	**	ns	ns
L	**	ns	ns	ns	ns	ns
WxC	ns	ns	ns	**	**	**
WxL	ns	ns	**	ns	**	**
CxL	ns	ns	ns	**	ns	ns
WxCxL	ns	ns	**	**	ns	**

Notes: W = Waste; C = Cassava-Leaf Compost; L = Lime; * = Significant at 5%; ** = Significant at 1%; ns = Not Significant at 5% and 1%; [1)] Amirulloh (2000); [2)] Prihatin (2002).

Table 3. The effect of waste on the concentrations of Cu labile fraction in a tropical soil ±10 years after treatment with industrial waste (Trans √x) (Ginanjar 2009).

Waste Levels ($t\ ha^{-1}$)	Soil Depth (cm)	
	0-15	15-30
	-------------------------- $\sqrt{mg/kg}$ --------------------------	
0	2,68 a	1,78 a
15	3,62 a	2,17 a
60	5,31 b	2,84 b
LSD 5%	1,01	0,64

Notes: Figures in one column with the same characters are not significantly different at 5%.

Table 4. The relative concentrations of Cu (Cu/Cu_0-1.5) at waste level of 60 ton ha^{-1}.

No.	Treatment Unit	± 1.5 years	± 3 years	± 10 years
1	No C, No L	1.00	0.77	0.19
2	No C, With L	0.67	0.54	0.39
3	With C, No L	0.80	0.60	0.21
4	With C, With L	0.55	0.64	0.17

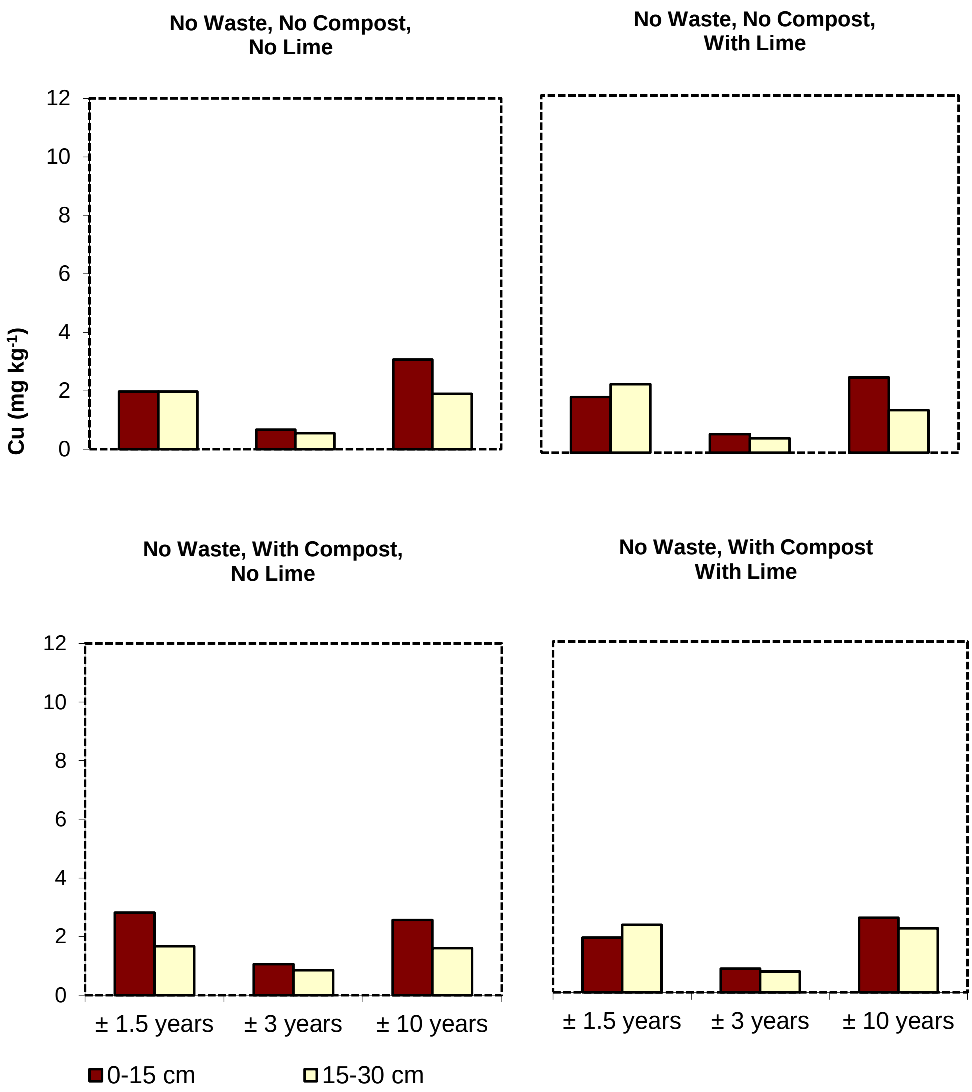

Fig 1. The effect of industrial waste, lime, and cassava-leaf compost on the concentration of a tropical soil labile fraction of Cu (Trans√x) at waste level 0 ton ha^{-1} (Compost 5 ton ha^{-1} and Lime 5 ton ha^{-1}). [1)] Amirullah (2000); [2)]Prihatin (2002) (Ginanjar 2009).

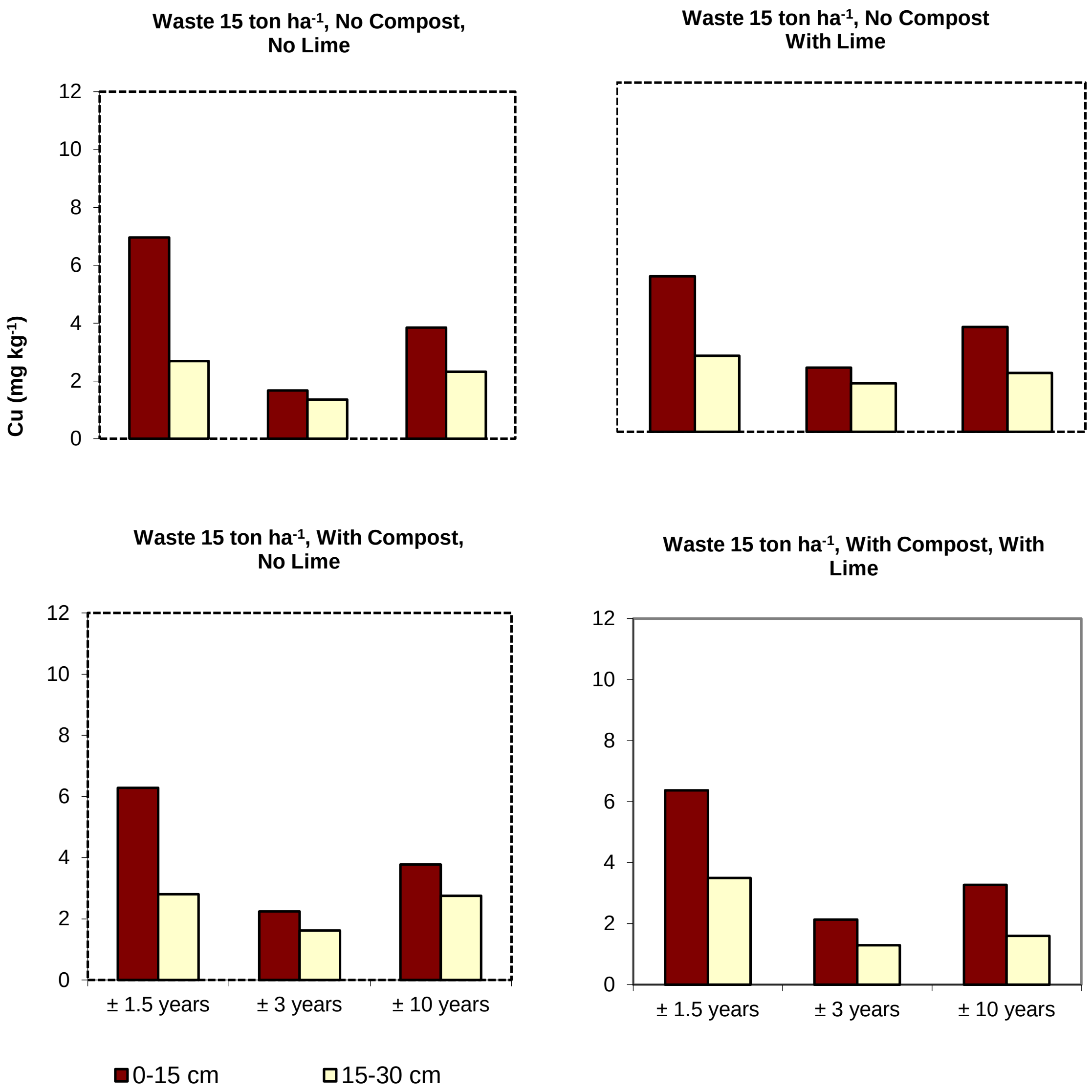

Fig 2. The effect of industrial waste, lime, and cassava-leaf compost on the concentration of labile fraction of Cu (Transf√x) at waste level of 15 ton ha^{-1} (Compost 5 ton ha^{-1} and Lime 5 ton ha^{-1}). [1] Amirullah (2000); [2]Prihatin (2002) (Ginanjar 2009).

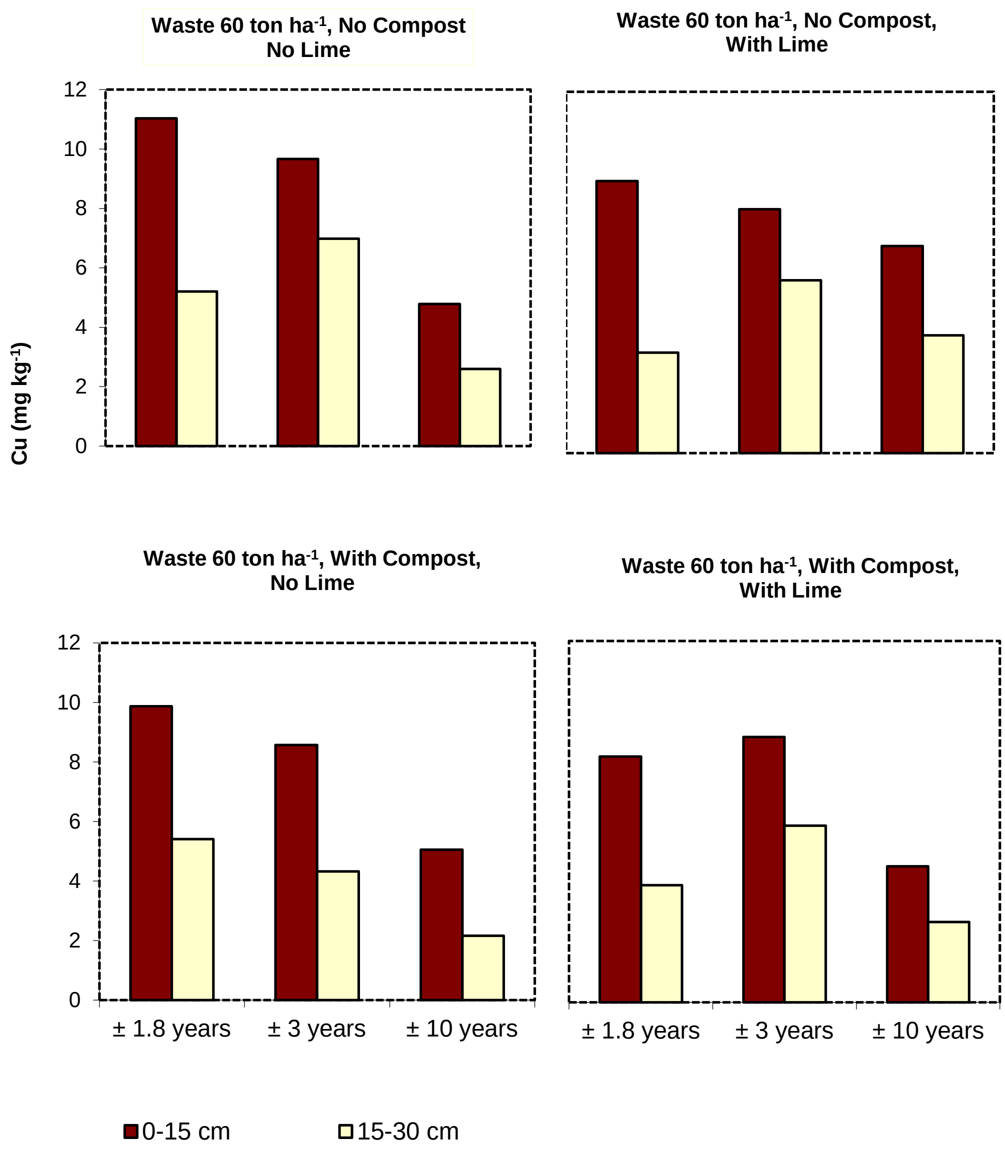

Fig 3. The effect of industrial waste, lime, and cassava-leaf compost on the concentration of a tropical soil labile fraction of Cu (Transf√x) at waste level of 60 ton ha^{-1} (Compost 5 ton ha^{-1} and Lime 5 ton ha^{-1}). [1)] Amirullah (2000); [2)]Prihatin (2002) (Ginanjar 2009).

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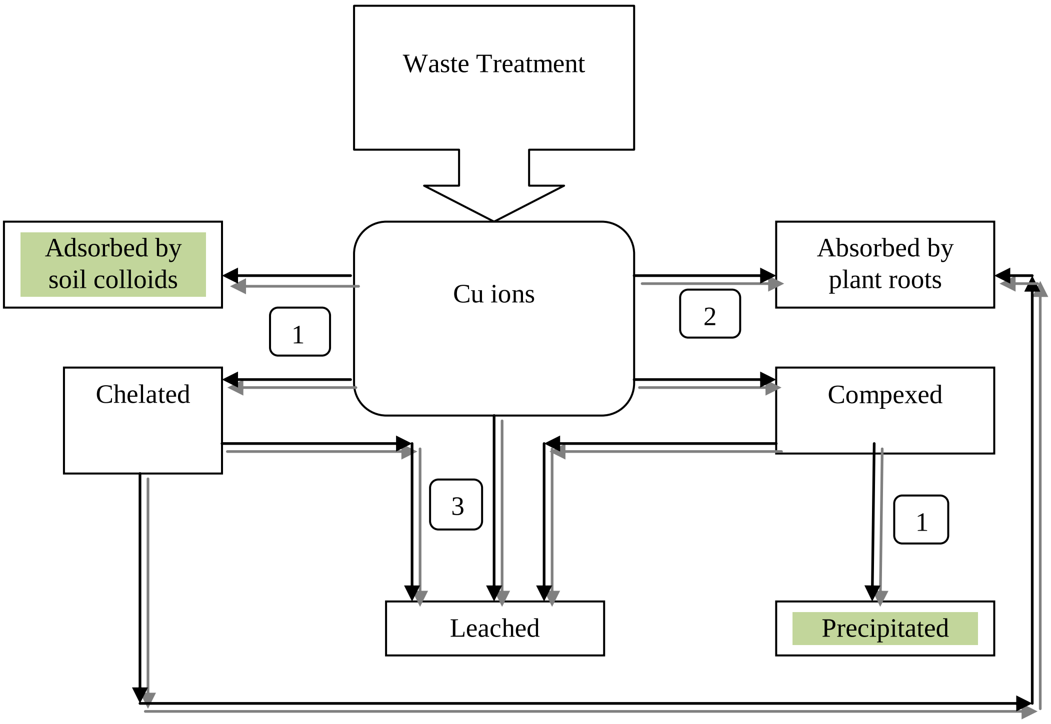

Fig. 4. The possibility of the waste-origin Cu removal from soil labile fractions ±10 years after waste treatment (1 – Adsorption or precipitation, 2 – Absorption by plant roots, 3 – leaching).