Heavy Metal Contents of Some Soils of Tomato Growing Fields in Hai District, Tanzania, as Influenced by Duration of use of Metal-Containing Fungicides

[1]Munisi, N. and Semu, E.*

[1]Department of Soil Science, Sokoine University of Agriculture, P.O. Box 3008, Chuo Kikuu, Morogoro, Tanzania

Abstract

Hai district is one of the major vegetable producing areas of Tanzania. Copper fungicides have been used for a long time in the district, but no studies have been conducted to evaluate accumulation of Cu in the soils in that district. This study was, therefore, initiated to survey and to document the status of fungicide use in some tomato fields. Generally, the total or DTPA-extractable Cu contents were significantly ($P<0.05$) higher in fungicide-treated soils than in control soils, the levels reaching 7305 mg total Cu/kg soil where fungicides have been used for 15 to 30 years (long-term use). The total and extractable Cu contents of the soils increased with increasing duration of Cu fungicide use. This resulted in lower microbial populations with increasing duration of fungicide use, the populations decreasing from $log_{10} = 6.4$ in control soils to $log_{10} = 5.1$ in long-term use soils. Zinc and manganese did not show clear trends. There was no significant relationship between total or DTPA-extractable Cu, Zn or Mn and soil pH. Total Cu was significantly and positively correlated with organic carbon, while total Mn was significantly but negatively correlated with organic carbon. It is concluded that long-term use of Cu fungicides in Hai district has resulted in a build-up of Cu residues in the soils, and this may impair microbial processes in those soils.

Key words: Heavy metals, pollution, heavy metal-containing fungicides, tomato, microbial populations.

Introduction

Vegetable production in Tanzania, if fully developed, can contribute to increased food security. In addition to food security, the vegetable industry provides income and employment to farmers and other workers (Nyange, 1994). However, diseases have threatened the continued cultivation of vegetables in major producing areas of the country, especially during rainy periods. To control diseases and sustain production, fungicide use has been indispensable. Copper (Cu) compounds have been the most widely used fungicides in vegetable production. But use of these compounds may lead to build-up or accumulation of their residues in the environment.

In Tanzania, accumulation and fate of the heavy metal containing fungicides has not been adequately studied. Some of the limited research done includes assessment of Cu accumulation in a few soils of coffee growing farms in Moshi (Mkindi, 1990), Lushoto (Baruti, 1997) and in some soils of tobacco and vegetable growing areas of southern Tanzania (Semu and Singh, 1996; Mwalilino, 1997).

In Iringa district, Semu and Singh (1996) found that soils under tomato production which had received Cu fungicides contained significantly ($P<0.05$) higher levels of total Cu than in the virgin soils. In Lushoto, Baruti (1997) reported an average of 52 mg total Cu/kg soil in vegetable fields which had received Cu

*Corresponding author

fungicides for more than 20 years. Mwalilino (1977) reported that total Cu was significantly ($P<0.05$) higher in fungicide-treated vegetable growing soils than in virgin ones in Mbeya district.

Hai district is one of the major vegetable producing areas of Tanzania. Despite use of Cu fungicides for a long time in that district, no studies have been conducted to evaluate accumulation of Cu in soils in the district. Such studies would provide data on the basis of which recommendations on the course of future use of Cu fungicides in the district could be made. This study was, therefore, initiated to survey and to document the status of fungicide use in some tomato fields. Accumulation of Cu and other heavy metals in soils as a result of fungicide use was assessed. Because Cu in high amounts is toxic to biological systems, effects of the Cu fungicide residues on soil microorganisms, which are the agents of organic matter decomposition and nutrient cycling, were assessed in those soils.

Materials and Methods

This study was undertaken in Hai District, which is situated in the Northeastern part of Tanzania, in Kilimanjaro Region. The District lies on the Southwestern slopes of Mt. Kilimanjaro, between 2°50' and 3°29' South and 30°30' and 37°18' East. It has a bimodal rainfall pattern, of which the long rains are from March to June, with the short rains from October/November to December. Rainfall varies from 900mm in the lowland areas to 1800mm in the highlands. The dominant soils are volcanic and are deep, dark reddish, dark yellowish or brown clay loams on lower slopes. The upper slopes have dark brown silt loams over reddish brown silty clay loams.

A survey was undertaken, using a questionnaire, to document the main vegetable crops grown and different durations of time fungicides have been used in various farms. The durations were grouped into short-term use of fungicides (1-5 years), intermediate term (6-14 years), and long-term (15-30 years). Locations, which previously had not received intentional applications, of fungicides were identified and taken as being controls. These were chosen away from the cultivated fields, which receive fungicide treatments, so as to avoid unintentional fallout of fungicide residues to them.

Soil samples were collected, in four replicates, at a depth of 0-10cm. Two sets of samples were collected. In one set, replicates were taken within each field. In another set, entire fields served as replicates. The sampled fields were drawn from the different categories of duration of tomato cultivation and, hence, of fungicide use as already described.

The soils were air-dried, ground and passed through the 10 mesh (2mm) sieve. The soils' pH was determined in water (1:2.5 soil:water ratio) (MacLean, 1982) using the glass electrode. Organic carbon (O.C.) was determined by the wet digestion method of Walkley and Black (Nelson and Sommers, 1982). These properties are presented in Table 1. From those data, pH of the soils was generally around neutral, while O.C. values were low to medium.

Table 1: Some properties of the soils

Duration of fungicide use, years.	Soil sampling protocal			
	Replicates within a field		Fields as replicates	
	pH(water)	%O.C.	pH(water)	%O.C.
0(=control)	6.8±0.1	3.1±0.1	6.7±0.2	4.2±1.0
1-5(=short term)	7.0±0.1	3.1±0.3	7.1±0.2	4.2±0.2
6-14)=medium term)	6.7±0.4	2.5±0.1	7.2±0.3	4.5±0.5
15-30 (=long term)	6.9±0.1	2.9±0.1	7.0±0.3	5.3±0.4

Total heavy metal contents of the soils were determined using an atomic absorption spectrophotometer (AAS), following aqua regia digestion of the samples (Jeng and Bergseth, 1992). Copper, Zn and Mn in the soils were extracted using 0.005M DTPA (Lindsay and Norvell, 1978) and determined using AAS.

Viable microbial populations in the samples were determined in nutrient agar using the plate count method (Wollum, 1982). The petri dishes were incubated at 25-28°C for 2 weeks and colonies counted. The data were expressed as microbial populations per gramme of soil and transformed into $\log_{10}$ values prior to statistical analysis.

Results and Discussion

Range of Vegetable crops grown in Hai District

The different types of vegetables grown in the surveyed sites include tomatoes (*Lycopersicum esculentum*), onions (*Alium cepa*), egg plant (*Solanum melongena*), sweet pepper (*Capsicum annuum*) carrots (*Daucus carota*), chinese cabbage (*Brassica chinensis*), and African spinach (*Amaranthus* sp). Tomatoes and sweet pepper account for approximately 80% of all the vegetables produced in this district, and receive large quantities of Cu fungicides for controlling diseases.

In Hai, these vegetables are sometimes intercropped with maize or sunflower and in some other cases they are raised as sole crop in small plots of less than 0.2 hectare. The majority of growers rotate vegetables with other field crops such as maize, beans, or sunflower.

Pests and their control strategies in Hai district

The vegetable growers in the district face high levels of pest infestation during the vegetable production cycle. Pest problems, especially diseases, have been exacerbated by the fact that irrigation, widely practised in the area, enables off-season production from which the outbreak of secondary inoculum is very high, resulting in high infection rates. The main fungal diseases are late blight (*Phytophthora infestans*) and *Fusarium* wilt of tomatoes, blight (*Phytophthora palmivora*) of peppers, and white rot (*Sclerotium cepivorum*) of onions. Use of fungicides, especially during rainy seasons, becomes a prerequisite for crop protection and assurance of high yields.

Farmers in the vegetable growing areas have for long been using Cu fungicides such as those bearing the trade names Blue copper, Red copper, Cobox, Copper Nordox, Copper Sandoz, Recop, Delan, and Kocide. Other formulations in use contain Mn, and include trade names such as Brestan, Dithane, Ridomil, and Octave. These fungicides are used especially on tomatoes and, to a lesser extent, on sweet pepper and onions, to control fungal diseases. Other formulations which contain Mn (e.g. Maneb), Zn (e.g. Zineb), and Mn + Zn (Mancozeb) may also have been used as they have been gazetted in this country in the past. The rates of fungicide use are usually high during rainy seasons. According to some growers interviewed, the amounts of fungicide used per plot of 0.2 hectare in a single spray may range from 0.5-3.0 kg of the product, depending on crop growth stage. When the crop is young, with a less developed canopy, the application rate is lower than when the crop is well developed with a full canopy. High rates are necessary because the high humidity during rainy seasons favours and accelerates growth of pathogens, leading to increased levels of infestation. Also, following spraying, the pesticides are often washed away by the rains, thus reducing their effectiveness. This necessitates shorter intervals of spraying, eventually increasing the number of sprays and quantity of fungicide used in the season.

Again, the rates of application depend on availability of the fungicides. When a certain chemical is easily available, growers tend to overdose, thinking that the larger the dose the better the control. When pesticides are scarce, growers tend to underdose in order to meet the prescribed number of sprayings. In Hai, where pesticides are readily available, the tendency usually has been to overdose.

After many years of fungicide use on these vegetable plots, substantial amounts of heavy metals such as Cu, Mn and Zn are likely to have accumulated in these Hai soils. This is because metal removal from the top soil via crop uptake, and/or leaching to the sub-soil, are usually relatively little (Lexmond, 1980).

Distribution of heavy metals in soils of tomato growing areas in relation to duration of fungicide use

Copper levels in the soils

The total and DTPA-extractable Cu in the soils are presented in Table 2. In the case where replicates were taken within a field, the total and extractable Cu levels increased with increasing duration of Cu fungicide use. Generally, the total or DTPA-extractable Cu contents were significantly ($P<0.05$) higher in fungicide-treated soils than in control soils. No significant ($P<0.05$) difference was observed in total or DTPA-extractable Cu between short and medium term soils.

Table 2: Copper levels in the soils, mg/kg

Duration of fungicide use, years	Fields as replicates			
	Replicates within a field		Fields as replicates	
	Total Cu	DTPA-Cu	Total Cu	DTPA-Cu
0(=C)	1013.4±62.6c	52.1±6.1c	816±275.6c	33.3±3.6b
1-5(=ST)	3130.9±258.5b	376.3±75.9b	1315±273.2bc	214.2±63.4b
6-4(=MT)	3569.5±367.2b	471.9±64.4b	2435.2±4421ab	268.6±85.6ab
15-30(=LT)	7305.4±284.2a	1365.5±68.6a	3100±99.0a	459.2±146.4a

C=Control; ST=Short term; MT=Medium term; LT= Long term. Means within a column followed by the same letter are not significantly different at P<0.05 according to the Duncan's New Multiple Range Test

Similarly, in the case where entire fields were used as replicates, there were significantly (P<0.05) higher total and extractable Cu contents in the soils of long term use of fungicides than in the control ones. There was no significant (P<0.05) difference in Cu contents between the short term and medium term soils.

The lack of significant difference between short and medium term soils may be due to differential intensity of fungicide use within the individual farms, such that some medium term soils may have received lower dosages while some short term soils could have received higher doses of the fungicides.

The trend of these data vis-a-vis duration of fungicide use was generally similar in the two sampling protocols. The difference between the protocols was in the fact that higher Cu levels were found in the situation where replication was done within each field than where fields were used as replicates. The possible reasons for this difference may be related to differential intensity of sampling between the two protocols. The lower levels of Cu revealed from using fields as replicates would mask the true levels of pollution due to its low intensity of sampling, thereby leading to erroneous conclusions. Thus, any sampling programme should integrate replication within fields to reflect the true situation.

From this study it was observed that the mean Cu concentrations in the soils increased with increase in duration of fungicides use. The easy availability of Cu-based fungicides from the neighbouring villages of Mbosho and Kibohehe, where there is intensive use of these fungicides in coffee production, has facilitated their widespread use in tomato production also. Apart from using Cu fungicides such as Kocide and Blue copper to control fungal diseases, most growers also use Red copper for enhancing ripening of tomato fruits, especially during the cold season (Ndosi, 1998, personal communication). In addition, most growers use doses higher than those recommended by manufacturers. With time, all these factors increased the rate of build-up of Cu fungicide residues in these soils, with greater accumulation associated with longer durations of use of the fungicides.

According to Baker (1991) total Cu levels in most uncontaminated soils averaged 30mg/kg, while the normal range of Cu for most soils based on extraction by DTPA was 0.1-2.5mg/kg (Sims and Johnson, 1991). Mwalilino (1997) found that total and DTPA-extractable Cu levels in control soils of Mbeya were 18.5 and 1.44mg/kg, respectively. In the present studies it was found that the total Cu levels in control soils were 1013.4 and 816.8mg/kg, while DTPA-extractable Cu levels were 52.1 and 33.3mg/kg, in the case where replications were taken within a field and where fields were taken as replicates, respectively. These high levels imply that Cu-based fungicides may have been used in these soils in the past. Perhaps some of these soils had been cropped to coffee or vegetables previously.

The significantly higher amounts of total Cu in the soils receiving copper fungicides in this study agree with other findings. Semu and Singh (1996), working with soils of tomato growing areas of Iringa district, reported significantly higher amounts of Cu in soils with long term use of fungicides compared to those from virgin sites. Similarly Baruti (1997) and Mwalilino (1997) revealed that soils of vegetable growing areas of Lushoto and Mbeya districts treated with fungicides for long periods had significantly (P<0.05) higher Cu levels as compared to soils of virgin sites.

Zinc levels in the soils

There were no significant (P<0.05) differences in the total Zn levels between soils of fungicide treated and non-treated sites where replicates were taken within fields (Table 3). However, DTPA-extractable Zn was significantly higher in the long-term fungicide-treated soils compared to the rest. When fields were used as replicates, there were significant ($P<0.05$) differences in the total as well as in the DTPA-extractable Zn levels between pesticide-treated and non-treated soils.

The fact that the control soils, in the case where fields were used as replicates, had higher levels of Zn compared to treated sites may imply that little or no Zn-based fungicides have been used in this area. If they ever were used, it is probable that small quantities were used, and hence their contribution to the Zn content of the soils was insignificant.

Manganese levels in the soils

The total and DTPA-extractable Mn in the soils are presented in Table 4. When replicates were taken within fields there were significant ($P<0.05$) differences in total and DTPA-extractable Mn levels between control and treated soils. No differences were observed when fields served as replicates.

Table 3. Zinc levels in the soils, mg/kg

Duration of fungicide use (year)	Soil sampling protocol			
	Replicates within a field		Fields as replicates	
	Total Zn	DTPA-Zn	Total Zn	DTPA-Zn
0(=C)	6209.8±364.9a	29.7±2.7b	8128.2±325.1a	13.3±1.6b
1-5(=ST)	8128.1±81.8a	19.4±3.5b	6502.5±2.4b	27.2±3.5b
6-14(=MT)	11379.4±369.7a	14.1±1.8b	4226.6±520.0c	27.9±6.2b
15-30(=LT)	3576.4±325.1a	63.7±11.2a	5202.0±1.2c	70.2±18.3a

C=Control; ST=Short term; MT=Medium term; LT=Long term.
Means within a column followed by the same letter are not significantly different at $P<0.05$ according to the Duncan's New Multiple Range Test.

In both protocols of soil sampling there were inconsistent trends of total and extractable Mn levels in the soil with relation to duration of fungicide use. Also, levels of Mn in control soils were higher than those in treated soils. This implies that levels of naturally occurring Mn are high and that if there was any use of Mn-based fungicides in this area, it must not have been extensive.

Table 4: Manganese levels in the soils, mg/kg

Duration of fungicide use	Soil sampling protocol			
	Replicates within a field		Fields as replicates	
Year	Total Mn	DTPA-Mn	Total Mn	DTPA-Mn
0(=C)	99813.4± 144.2a	780.4±66.1a	77379.8±611.3a	194.8±8.83.9a
1-5(=ST)	73250.6±134.1ab	629.2±40.4ab	64374±1.2a	90.2±15.4a
6-14(=MT)	59497.9±432.6b	432.6±39.0c	74453±720.0a	176.7±41.3a
15-30(=LT	72828.0±341.7ab	589,9±24.9b	70552.2±540.6a	130.1±15.9a

C=Control; ST=Short term; MT=Medium term; LT=Long ter.
Means within a column followed by the same letter are not significantly different at $P<0.05$ according to the Duncan's New Multiple Range Test

Influence of heavy metals on microbial populations in fungicide-treated soils

One consequence of presence of heavy metals in the environment is toxicity to living organisms, including soil microorganisms. The distribution of total (viable) microbial populations in the soils as influenced by the heavy metals is presented in Table 5. The populations were significantly ($P<0.05$) higher in control sites than in all the fungicide-treated sites. As the duration of fungicide use increased the populations of microorganisms in those soils showed a consistently decreasing trend, although the differences in populations between the treated sites themselves were not significant ($P<0.05$).

This consistent trend of decrease of microbial populations in soils with increasing duration of fungicide use implies that the levels of heavy metals in soils exerted deleterious effects on soil microorganisms and that this reduced their numbers. This may be due to the fact that heavy metals interfere with metabolic activities of soil microorganisms. For instance Hiroki (1991) found that simple bacteria as well as actinomycetes were strongly affected when soils contained 310-751mg Cu/kg. Chang and Broadbent (1981) and Ohya et al. (1985) reported a decrease in microbial respiration in soils with high amounts of Cu and Zn. Such effects may have operated in the present soils which had high levels of these metals, leading to a reduction in microbial populations.

Table 5: Log10 of microbial population in the soils in relation to the duration of pesticide use

Duration of fungicide use, years	Soil sampling protocol
	Replicates within a field
	Log^{10} of microbial populations
0(=C)	6.4 ± 0.2a
1-5 (=ST)	6.0 ± 0.3b
6-14(=MT)	5.4 ± 0.1b
15-30(=LT)	5.1 ± 0.1b

C=Control; ST= Short term; MT=Medium term; LT=Long term. Means within a column followed by the same letter are not significantly different at P<0.05 according to the Duncan's New Multiple Range Test

Relationships between heavy metals and some soil properties

Regression equations of total and DTPA-extractable heavy metal contents on pH and % organic carbon are presented in Tables 6 and 7. In both sampling procedures no significant relationship was found to exist in these soils between total or DTPA-extractable Cu, Zn or Mn and soil pH. But it was observed that where replicates were taken within a field, total Mn was significantly (P<0.05) and positively correlated with the soils' organic carbon content. In both sampling procedures, DTPA-extractable Mn was correlated with organic carbon content. Where different fields served as replicates, total Cu was significantly and positively correlated with organic carbon, while total Mn was significantly but negatively correlated with organic carbon.

This lack of correlation between heavy metals and soil pH may be due to the narrow range of pH (6.7 - 7.2) in the present soils. However, the results obtained in this study agree with those of Piper and Beckwith (1949) who found that copper was not affected much even over a wider range of soil pH, between 4.5 and 7.5. Similar results have also been reported by Archer (1971) and Brennan et al. (1980).

The positive correlations between total heavy metals with soil organic carbon contents indicate the contribution of soil organic matter in the retention of heavy metals in these soils. This relationship will prolong the residence time as well as toxicity of these metals in this environment. The positive and significant correlations presently observed agree with the findings of Sangwan and Singh (1993) and Vadivelu and Bandyopadhyay (1995).

Table 6: Relationships between total heavy metals and some soil properties.

Regression equation	R^2	Significance
A: Replicates taken within a fied		
Total Cu = 11135.8 + 2167.9 pH	0.035	NS
Total Zn = 76388.7 - 8020.8 pH	0.002	NS
Total Mn= 42315.6 + 18053.2 pH	0.044	NS
Total Cu = 7044.5 - 1142.0 O.C	0.019	NS
Total Zn = 31102.2 + 11189.7 O.C	0.016	NS
Total Mn = 39149.8 + 41948.3 O.C	0.463	**
B: Fields taken as replicates		
Total Cu = -12707.3 + 2076.2pH	0.223	NS
Total Zn = 2228.9 - 2310.6 pH	0.167	NS
Total Mn= 74508.6 - 400.2 pH	0.001	NS
Total Cu = 140.37 + 397.3 O.C	0.045	*
Total Zn = 11285.6 - 1178.6 O.C	0.24	NS
Total Mn = 120656.9 - 10949.9 O.C	0.28	*

Table 7: Relationships between DTPA - extractable heavy metals and some soil properties

Regression equation	R^2	Significance
A: Replicates taken within a fied		
Total Cu = 11135.8 + 2167.9 pH	0.035	NS
Total Zn = 76388.7 - 8020.8 pH	0.002	NS
Total Mn= 42315.6 + 18053.2 pH	0.044	NS
Total Cu = 7044.5 -142.0 O.C	0.019	NS
Total Zn = 31102.2 + 11189.7 O.C	0.016	NS
Total Mn = 39149.8 + 4148.3 O.C	0.463	**
B: Fields taken as replicates		
Total Cu = -12707.3 + 2076.2pH	0.223	NS
Total Zn = 2228.9 - 2310. pH	0.167	NS
Total Mn= 74508.6 - 400.2 pH	0.001	NS
Total Cu = 140.37 + 397.3 O.C	0.045	*
Total Zn = 11285.6 - 1178.6 O.C	0.24	NS
Total Mn = 120656.9 - 10949.9 O.C	0.28	*

Relationships between DTPA-extractable metals and total heavy metals

Regression equations of DTPA-extractable heavy metals on total heavy metals are presented in Table 8. In both sampling protocols, extractable Cu and Mn were significantly (P=0.05) and positively correlated with total Cu and Mn. This positive correlation indicates that the more total heavy metals there are in soil, as can be introduced via intensified use of fungicides, the greater the extent of extractability and, probably, bioavailability to growing plants and soil-inhabiting microorganisms. Such mobilized metals may be dispersed and eventually may contaminate water resources. Long-term consumption of contaminated crops and water may then affect human health.

Table 8: Relationships between DTPA-extractable heavy metals and total heavy metals

Regression equation	R^2	Significance
A: Replicates taken within a field		
DTPA Cu = -237.9 + 0.2 T. Cu	0.98	**
DTPA Zn = 33.5 - 0.000084 T. Zn	0.023	NS
DTPA Mn= 98.6 + 0.006 T. Mn	0.52	**
B: Fields taken as replicates		
DTPA Cu = - 123.6 + 0.5 T. Cu	0.72	**
DTPA Zn= 56.2 + 0.00072 T.Zn	0.40	NS
DTPA Mn= 241.8 + 0.09 T. Mn	0.02	*

Relationships between microbial populations and soil heavy metal contents

Regression equations of microbial populations (expressed as $\log_{10}$) on total and DTPA-extractable heavy metals are presented in Table 9. In both sampling protocols microbial populations were significantly and negatively correlated with total Cu and DTPA-extractable Cu. The populations were also significantly but positively correlated with total Mn and DTPA-Mn.

The negative correlations observed in this study indicate that Cu exerted deleterious effects on some metabolic functions of microorganisms in these soils. An example is the inhibition of soil microbial respiration (Bernhard et al., 1986). Similar results have been reported by some other researchers, for example Fritze et al. (1989) and Philip (1998), who found that microbial populaions were reduced in soils polluted with heavy metals.

Table 9: Relationships between microbial populations and heavy metal contents in soil

Regression equation	R^2	Significance
A: Replicates taken within a field		
$\log_{10}$ C.F.U = 6.5 - 0.0002 Total Cu	0.57	**
$\log_{10}$ C.F.U = 5.6+0.00006 Total Zn	0.15	NS
$\log_{10}$ C.F.U =3.48+0.00003Total Mn	0.59	***
$\log_{10}$ C.F.U =6.23 - 00009 DTPA Cu	0.54	***
$\log_{10}$ C.F.U = 6.12 - 0.012 DTPA Zn	0.19	NS
$\log_{10}$ C.F.U = 4.38 +0.0022 DTPA Mn	0.28	*

Note: C.F.U= Colony Forming Units of microbial population

In conclusion, the present study has shown that long-term use of fungicides in Hai district has led to a build-up of Cu residues in those soils. Soil organic matter contributed to the retention of Cu in the soils. The Cu fungicide residues reduced microbial populations, and this may affect organic matter decomposition and plant nutrient cycling in these soils.

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