ASSESSMENT OF ENVIRONMENTAL POLLUTION BY RESIDUES OF COPPER FUNGICIDES USED ON COFFEE IN ARUSHA REGION, TANZANIA

BY

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ABSTRACT

This study was undertaken to examine environmental pollution by residues of Cu fungicides in coffee fields of Arusha and Arumeru districts. Soils were analysed from coffee fields sprayed with fungicides for duration of 15 - 20 years to over 50 years under both small and large-scale cultivation. Young bean plants and their associated soils were collected from randomly selected coffee fields based on the duration of fungicide use. Copper was determined to examine the relationship between plant Cu concentrations with Cu content in soils. The study also assessed the contribution of the topography (hence erosion) in Cu transportation and possibility of polluting water resources. Water and sediments were collected from Lake Duluti to examine the possibility of the lake being polluted with Cu originating from coffee farms. Total Cu from the soil and sediments was determined by aqua regia digestion and extractable Cu using 0.005 M DTPA. Plant samples were digested using aqua regia. Copper contents were determined by AAS. The results indicated that both total and DTPA extractable Cu increased with increased frequencies, amounts and duration of Cu fungicide use. Copper was significantly ($P=0.05$) higher in fungicide treated sites and lower levels in absolute control (virgin forest) soils. Young bean plants sampled from fungicide-treated coffee fields had high levels of Cu while lower Cu levels were found in samples collected from control soils. Plant Cu concentrations and plant Cu uptake were significantly ($P=0.05$) correlated with the Cu contents in soils. Slope contributed to transportation of Cu through erosion, with higher levels of Cu accumulating downslope. Water and sediments from one location of Lake Duluti were found to have levels of Cu comparable to natural background levels. It is concluded that higher Cu levels in soils resulted in increased Cu uptake by bean plants. Long-term consumption of such crops with higher Cu levels may pose health risks.

DECLARATION

I, SADA BAKARI MZIMBA, do hereby declare to the senate of the Sokoine University of Agriculture, Morogoro, that this dissertation is my original work and that it had never been submitted for a degree award in any other University.

Signature ....

Date ...16\02\2001...

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DEDICATION

This work is dedicated to my parents Mwadawa Shabani and the late Bakari Selemani, uncle Omari Fyeku, brothers Mursali, Selemani, Bilal, sister Halima, Zainabu, and the late Halima Haji.

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AAS	- Atomic Absorption Spectrophotometry
C.B.D.	- Coffee berry disease
Cu	- Copper
DASP	- Department of Animal Science and Production
DSS	- Department of Soil Science
DTPA	- Diethylene-triamine pentaacetic acid
N	- Sample size
NORAD	- Norwegian Agency for International Development
M	- Molarity
sd	- Standard deviation
SUA	- Sokoine University of Agriculture
TEA	- Triethanolamine
USDA	- United States Department of Agriculture

CHAPTER ONE

1.0 INTRODUCTION

Copper (Cu) fungicides are routinely used in coffee fields in Tanzania. This is because coffee plants are often times infested by many pests, which result in reduction of yields of this important cash-earning crop. The major pests are the fungi causing the coffee leaf rust disease (*Hemileia vastatrix*) and coffee berry disease (C.B.D.) (*Colletotrichum coffeanum*). Coffee leaf rust is a major disease of coffee requiring continual use of fungicides for its control. Coffee berry disease is capable of causing up to 60% loss of yields if not controlled (Okioga, 1978). Bujulu (1978) estimated that about 50% of coffee crop can be lost due to coffee leaf rust. Control of these diseases is difficult, but satisfactory results are obtained through the use of Cu fungicides. Use of Cu fungicides, therefore, becomes a prerequisite for crop protection and assurance of high yields.

The use of Cu fungicides for the control of fungal diseases of coffee plants started in 1956/57 and has been going on since then (Bujulu, 1978; Okioga, 1978). During this period, the most effective fungicides were found to be formulations containing 50% copper and phenyl mercuric acetate sprayed up to 8 times per season. The phenyl mercuric acetate proved phytotoxic and was later abandoned (Okioga, 1978). Studies by Bujulu (1978) and on-going research at Lyamungo Coffee Research Centre (1990) still recommend use of Cu compounds in Tanzania for the control of coffee diseases.

These Cu compounds, such as cuprous oxide, copper oxychloride and cupric hydroxide are likely to continue playing an important role in the control of fungal diseases of coffee until economic and more effective fungicides are found to replace them (Agrios, 1988; Mkindi, 1990). As a result, farmers in the coffee growing areas in Arusha region have found themselves committed to near monthly sprays of Cu fungicides and they are forced to apply as frequent as they can to maximize production and eliminate diseases in their farms. Therefore, soils in these areas are likely to accumulate copper residues due to frequent use of Cu fungicides. Reuther and Smith (1953) reported large accumulations of copper in soils of citrus groves in Florida as a result of Cu fungicide applications.

At higher concentrations in the environment, toxic heavy metals, including Cu, may enter food chains and result in health hazards. Toxicity problems can occur in crops grown on polluted soils and in livestock grazing herbage growing on polluted soils. Copper is known to result in a marked depression in feed intake and growth, especially in birds, and anaemia in mammals including man (Underwood, 1971). When such fungicides are used in the same location over long periods of time, as would happen every year in attempts to control the coffee diseases, residues accumulate in those soils.

Most of the information available at present regarding heavy metal pollution of soils pertains to the industrialized world. Although more localized, pollution due to use of pesticides may be as intense in such locations of the developing world (Semu and

Singh, 1996). Because use of these fungicides which contribute to pollution is on the increase in these countries, Tanzania included, the situation deserves careful monitoring. Much work has been done in other countries on soil contamination due to pesticides application (Chang and Broadbent, 1982; Doelman, 1985; Fritze *et al.*, 1989; Hiroki, 1991). In Tanzania, this type of pollution has not been adequately studied. Some studies have been carried out to determine the accumulation of copper in soils in some coffee growing areas of Moshi district (Moshi *et al.*, 1982; Mkindi, 1990), in some soils of vegetables and tobacco growing areas in Iringa district (Semu and Singh, 1996), for coffee and vegetable growing soils in Lushoto district (Baruti, 1997) and in those of vegetable growing areas of Mbeya district (Mwalilino, 1997).

However, there is only little information on the extent of heavy metal pollution due to agricultural activities in Arusha region, a major coffee growing area in which Cu fungicides are used. Arusha and Arumeru districts are among those areas, where frequent and large amounts of these fungicides are used. A study was conducted in 1998 (Loland, 1998) which indicated some accumulation of Cu in some soils. But it may be expected that high levels of residues may exist since coffee production and fungicide use in those areas have been undertaken for a very long time, since the 1920s in some locations. Contamination of soils may imply the risk of water resources contamination. Leaching of Cu may not be a serious problem, even in highly contaminated sites, because Cu is specifically adsorbed on organic matter and clay surfaces. But upon introduction to soil, the Cu may be translocated to other locations through soil erosion. This, too, has not been studied much in Tanzania.

Thus, there is a need to carry out studies on the accumulation and dispersion of residues due to longterm use of copper fungicides on coffee, in terms of pollution of soils, waters and crop plants grown in or near the fields treated with these pesticides.

In view of the above, the research reported herein was undertaken with the overall objective of assessing environmental pollution by Cu fungicides residues as a result of use of Cu fungicides on coffee in Arusha and Arumeru districts.

The specific objectives of the study were:

1. To determine the levels of copper residues in soils and plants of large and small coffee farms which are still operational.

2. To determine the levels of copper residues in soils of abandoned farms.

3. To assess the effect of topography on the distribution of Cu in those areas.

4. To determine the influence of soil properties on Cu retention.

5. To determine the possibility of pollution of water sources in the vicinity of coffee farms.

CHAPTER TWO

2.0 LITERATURE REVIEW

2.1 Use of copper fungicides

Copper fungicides are well known for their effective control of many fungal diseases such as leaf rust, leaf spots, fruit diseases, anthracnose and downy mildew (Agrios, 1988). Bacterial blight of coffee (*Pseudomonas syringae*) can be controlled by the same copper fungicides applied at the same rate as recommended for coffee berry disease (*Colletotricum coffeanum*), but a higher frequency of treatment is required (Kairu, 1983). The use of copper fungicides for plant disease control was reported as early as 1917. Floyd (1917) as cited by Camp (1945) used copper sulphate as soil application and Bordeaux mixture as a foliar spray to control dieback of citrus. Bordeaux mixture, made by mixing copper sulphate, quick lime and water has long been used to protect vines in Europe against fungal attack, and it, or a similar mixture, is still often widely applied to potatoes to combat blight, and to fruit trees to control apple and pear scab (Mellanby, 1971). The major group of fungicides whose use is widely adopted amongst coffee producers is blue and red copper. These fungicides contain Cu as a structural constituent of the active ingredient and the fungicidal component is the Cu^{2+} ion.

2.2 Copper accumulation in soils due to fungicide use

A continual high input of copper fungicides into agricultural soils may result in adverse effects due to copper accumulation. Due to poor mobility of heavy metal in plants, an appreciable quantity remains on the plant body rather than being translocated to, and be removed through, the harvested economic parts. The plant residues are incorporated in the soil during weeding and subsequent land preparation, and in this way these metals accumulate in soils. Calculations by Moolenaar *et al.* (1998) showed that the Cu_{sol} after 50 years of fungicide use would be 4 times higher while the Cu^{2+} adsorbed on colloids would be 750 times higher than in control soils.

Preliminary investigations by Venkataramaiah and Singh (1974) were conducted to assess the extent of copper accumulation in coffee soils as a result of continual spraying with copper fungicides over a period of 30 - 40 years. An average of 217 kg of extractable copper had accumulated per hectare in the top 15 cm of the soil when arabica coffee was sprayed with these fungicides as compared to 24 kg/ha in the unsprayed robusta or control forest soils. Moshi *et al.* (1982) estimated an increase in soil copper to be 8 ppm per year in the top 15 cm of copper soils in Moshi district.

Mkindi (1990) reported values of 5.6 ppm to 186.5 ppm Cu in the top 5 cm soils of farms which had never been sprayed and those sprayed for more than 30 years, respectively. Alloway (1990) estimated that world-wide more than 7 x 10^7 kg of Cu as Bordeaux mixture were sprayed each year in vine yard, banana, citrus and other crops. Semu and Singh (1996) observed significantly (P=0.05) higher copper contents in some soils of Iringa district, Tanzania, that had received fungicides as compared to virgin sites. Alloway (1990) reported that the Cu accumulated in those soils was most often found to be toxic to the new plantings of the same or different crop.

Baruti (1997) reported that Cu contents in coffee soils of Lushoto district were consistently higher, in both extractable and total Cu, compared to controls. Total Cu ranged between 109 - 229 mg/kg and 47.50 - 55.5 mg/kg of soil for treated and control soils, respectively, within the 5 cm depth. Extractable Cu levels were found to be 27 - 73 mg/kg and 5 - 9 mg/kg of soil for treated and control soils, respectively, within the 5 cm depth. Mwalilino (1997) reported that virgin soils of Mbeya district had very low total Cu compared to the mean total Cu for soils as reported by Baker (1991). Kerin (1983) reported 25 kg/ha of Cu deposits in vine yard soils when copper-based fungicides were used.

The Cu contents of some Cu fungicides have been summarized by Thomson (1988) as shown below.

Name	Formula	Cu content %
Bordeaux mixture	$CuSO_4.H_2$+$Ca(OH)_2$	12.75
Blue copper	$CuSO_4.5H_2O$	25.2
Copper lime mixture	$CuSO_4.H_2O$	35
Kocide	$Cu(OH)_2$	65.13
Red copper	Cu_2O	89
Britox	$3Cu(OH)_2.CuCl_2$	50
Basic copper carbonate	$Cu(OH)_2.CuCO_3$	55
Copper 5 dust	$Cu_s.O_4.3Cu(OH)_2.H_2O$	53

This shows that continual use of Cu fungicides will contribute to Cu accumulation in agricultural soils.

2.3 Retention of Cu added to soils

Pollution by heavy metals is of great concern because whatever their source the metals tend to be immobilized in the top layer of soils, except under extremely acid conditions (McGrath, 1993). Copper may be adsorbed by soil in amounts in excess of their conventional exchange capacities, assuming adsorption as the Cu^{2+} ion (Menzel and Jackson, 1950; De Mumbrum and Jackson, 1956; Bingham *et al.*, 1964). This type of phenomenon as applied to copper and other heavy metal cations has been termed as specific adsorption and is exhibited by all major types of soil colloids, that is clay minerals (Bingham *et al.*, 1964; Tiller, 1964) organic matter (De Mumbrum and Jackson, 1956) and free oxides (McKenzie, 1967). However, some of this Cu will be partitioned into the solution phase in the soil.

Copper in soil solution exists as free ions (Cu^{2+}) and complex species as $Cu(OH)^{+}$ and $CuSO_4$. The total concentration of copper is the sum of its free ions and complex species. It is generally accepted that solution copper concentrations are controlled by specific adsorption/desorption reactions of copper and soil colloidal materials (McLaren *et al.*, 1990). Adsorption of 'free' heavy metal ions on soil components {soil particles, (in)organic ligands} depends on factors such as pH, clay content, organic matter and competition with other chemical species. The free metal ion fraction in the soil solution is often important with respect to mobility (Plette, 1996; Temminghoff *et al.*, 1997).

2.4 Effects of soil pH on adsorption

The concentrations of soluble copper and its free ionic activities in soil solution have been reported to be affected by soil pH (Harsh and Doner, 1984; Kuo and Baker, 1980). Generally, concentrations of dissolved Cu and its free ionic activities in soil solution decrease with the increase in soil pH. The decrease in free Cu^{2+} activities with soil pH is due to increase in the adsorptive capacity of the soil as pH is increased. This is in agreement with previous reports, which proposed that as soil pH increased the adsorptive capacity of soil solids increased due to the deprotonation of surface-bound H^+ (Alloway, 1990; Kishk and Hassan, 1973). Cavallaro and McBride (1980) found that an increase in pH adds to soil power to bind Cu thus reducing concentration of Cu^{2+} ions in solution, which is generally assumed to be the biologically active form of Cu. In this way, pH is important soil factor, which can have direct influence on availability, deficiency or toxicity of Cu in soil.

Low soil pH has been reported to reduce the capability of soils to sorb Cu (Kuo and Baker, 1980). It has also been observed that the relationship between copper and soil pH can be variable. Baruti (1997) reported that total copper showed significant negative correlation with pH. Similar results with respect to total copper and soil pH have been reported by Semu and Singh (1996), working with vegetable and tobacco growing soils of Iringa. Mayona (1977), however, reported a non significant, negative correlation (R^2 = 0.21) between extractable Cu and soil pH for soils of Tukuyu, Tanzania where the soils were generally acidic. Chattopadhyay *et al.* (1996) showed that the available Cu was significantly and negatively correlated with pH (R^2 = 0.23). Brennan *et al.* (1980) concluded that pH did not affect the availability of Cu in soil.

2.5 Effect of organic matter

All soils contain organic matter, although the amounts and types may vary considerably. Inhibition of litter decomposition tends to increase the storage capacity of heavy metals by soil organic matter, because soil organic matter has high (specific) binding capacity for heavy metals. Organic matter appears to have differing roles in controlling trace metal uptake by plants, depending upon whether it is the soluble (fulvic acid) or the insoluble (humic acid) fraction. The stability of metal complexes with fulvic acid and humic acids increased with increasing pH from 3 to 7.

Metals complexed by fulvic acids were more available to plant roots and soil biota than those complexed by humic acids (Kabata-Pendias and Pendias, 1984). Insoluble organic matter inhibits uptake of metal cations such as Cu^{2+} by strongly binding the metals, thereby preventing them from diffusing to roots. Conversely, soluble organics raise the carrying capacity of soil solution for Cu^{2+} and other metal cations at any particular pH by forming soluble metal-organic complexes (McBride, 1994). The plant is able to extract trace metals from these complexes once they diffuse to the root (Nor and Cheng, 1986). Several workers have reported that soils with high organic matter adsorbed Cu strongly thereby reducing its availability. For example, humus in which fulvic acid predominates fixed a noticeable amount of Cu (Stepanova, 1974). Stepanova (1974) further reported that Cu was the best flocculator of humic substances and in acid soils large amounts of Cu could be fixed by organic matter. The study by Hodgson *et al.* (1965) on the effect of organic matter on the relative mobility of Cd, Zn, Ni, Cu and Co in soils showed that Cu was less mobile and more strongly complexed with humic acids as compared to other metals.

McBride (1978) reported that complexation by organic matter was an effective mechanism of Cu retention in soils. He further reported that Cu^{2+} ions were directly bonded to two or more specific organic functional groups (mainly carboxylic, carbonyl and phenolic) so that the ion was immobilized in a rigid inner sphere complex. Stevenson and Fitch (1981) reported that the concentration of Cu in soils could be regulated through complexation by organic matter. When excess Cu was present, complexation reduced the concentration of Cu^{2+} ions to non toxic levels. Senesi *et al.* (1985) reported that fulvic acid have a high binding capacity for Cu added to aqueous solutions.

The above observations indicate that availability of Cu largely decreases with increasing organic matter content. This is because large amounts of Cu are retained as highly stable organo-copper complexes which are not immediately available to plants. Retention was highest in peat and muck soils, commonly resulting in deficiencies of Cu (Singh *et al.*, 1986). The varying amounts and types of organic matter in different soils may explain the differing roles of organic matter in metal retention in soil.

2.6 Environmental impacts of heavy metals

2.6.1 Uptake/accumulation of metals in the edible plant parts

According to Alloway (1990) some locations around the world have anomalous concentrations of heavy metals in soils, though not so high as to cause acute toxicity problems. However, these metals may accumulate in plants. Rice has been reported to accumulate Cu in grain (Kitagishi and Obata, 1981). Long term consumption of such produce may result in adverse health effects. Chelation of metals in soil may also affect amount of heavy metals that accumulate in edible plant parts. Copper toxicity may also be induced by the forms of other nutrients supplied during growth stage. Ozawa and Tazuki (1990), working with the form of nitrogen supplied and pH level of nutrient solution on Cu toxicities in crops, reported a relatively high accumulation of Cu in roots than in leaves of beans, spinach, lettuce and radish grown at pH 4 than at pH 6.

2.6.2 Toxicity to growing plants

The most critical soil parameter which indicates whether that soil has exceeded the threshold of metal phytotoxicity for a particular crop is not the heavy metal content, but the activity of the free metal cation in soil solution. Free metal cations are generally more toxic than the same metals in soluble complexed forms, although soluble metal -fulvic acid complexes are probably plant available (Nor and Cheng, 1986). For most crop species, the critical toxicity level of copper in the leaves is considered to be above 20 to 30 μg/g dry weight (Robson and Reuter, 1981). There are, however, marked differences in copper tolerance among plant species, for example, bean is much more tolerant than maize, these differences are directly related to the copper content of the shoots (Bachthaler and Stritesky, 1973).

Early studies of Cu toxicity have been associated with Cu accumulations in surface horizons of acid soils (Delas, 1963). Reuther and Smith (1953) reported toxicity symptoms in those citrus fields, which had extremely high soil Cu levels in the range of 320 - 382 kg/ha. Characteristic chlorotic disorders in citrus were observed, while these disorders were not observed in uncultivated fields of citrus, which had only 1.8 - 6.6 kg Cu/ha. Other characteristic disorders which were found in these soils were dead fibrous roots and abnormally dark coloured stubby roots. Copper toxicity to citrus seedlings growing in sandy soils with pH less than 5 and total Cu concentrations higher than 150 mg/kg have been reported by Reuther and Smith (1953). Using a culture solution, Wallace *et al.* (1981) reported that when the Cu concentration in trifoliate leaves of bush beans (*Phaseolus vulgaris*) was 89 mg/kg the yield reduction was 42% compared to control. Aduayi (1972) found characteristic symptoms such as corrugation, yellowing and necrosis of leaves following dieback of leaves in coffee seedlings when grown in solution containing 750 mg/kg Cu. Petterson (1971) reported only slight toxic symptoms in maize when soils contained 245 mg/kg Cu when extracted by 0.05 M EDTA. Lindsay and Norvel (1978) reported yield decreases of about one third when Cu extractable by 0.1 N HCl or DTPA was greater than 40 mg/kg. Tu and Qing (1990) working with purple soils in China with lettuce and cayenne pepper as test crops, reported that the same level of toxicity could be obtained in different soils with 0.5 μg/ml soil solution for lettuce and 2.5 μg Cu/ml for cayenne pepper. Tu (1993) reported a rice yield reduction of 15%, when Cu was added at a rate of 60, 100 and 130 mg Cu/kg soil in acid, neutral and calcareous soils, respectively. Heavy metals can also have toxic effects on roots and mycorrhiza (Colpaert and Van Assche, 1992) and nutrient uptake can be seriously reduced. The vegetation cover may ultimately be completely destroyed, increasing the risk of soil erosion (Hutchinson and Whitby, 1977; Freedman and Hutchingson, 1980; Lukina *et al.*, 1993).

2.6.3 Water contamination

Copper fungicides are applied as foliar sprays. Much of the Cu is eventually returned to the soil as spray droppings from leaves, through drift during application, washing off from leaves by rains, dumping/washing of containers or during decomposition of dead sprayed leaves. Clean, quality water can be preserved through understanding of the products and processes that endanger this valuable resources. Fungicides represent only a portion of the contaminants introduced into our water systems that affect water quality. The standards determining water quality depends on its intended use. Drinking water standards are higher than irrigation water or water used in industry. Copper is specifically adsorbed in soils, making it one of the trace metals which move the least. Water contamination resulting from the use of Cu may, therefore, be due to soil erosion or direct application on the water surfaces. Groundwater contamination by other metals has been reported. Kalta *et al.* (1995), for example, reported that selenium concentrations ranging from 3 – 330 µg/L were detected in groundwaters in the USA. Gaciri and Mailu (1989) reported that some streams in Kenya were particularly enriched with Cu, Zn and Pb, which was attributed to flushing from coffee estates, where large amounts of fertilizers and pesticides were used.

If the capacity of soils to bind heavy metals is overwhelmed due to accumulation of large quantities of heavy metals like Cu, this increases the likelihood of the heavy metals gradually leaching into the groundwater.

2.6.4 Heavy metal contamination and human health

Excessive concentrations of heavy metals in soil may affect human health through consumption of crops, and it is well known that uptake by plants is one of the most important mechanisms by which heavy metals enter man's food chain. Il'in (1991), working with vegetable - garden soils and crops in three Siberian cities, reported that among other ways in which heavy metals entered human body, about 75 - 80 % of heavy metal burden in humans was ingested through consumption of garden vegetables. Il'in (1991) observed high levels of Zn, Pb and Cd, beyond safe limits, in tissues of potatoes, cabbage and carrots grown in soils near non-ferrous metallurgical plants. Therefore, toxicity of humans through vegetable consumption can be minimized by growing the vegetables on uncontaminated soils and/or selectively growing crops which have mechanisms that restrict heavy metal entry into the edible parts.

Industrial exposure to Cu has been reported to result in the Wilson's disease, which is characterized by excessive concentration of Cu in the tissues, arising from metabolic defects involving absorbed Cu but not through ingestion (Underwood, 1971). Literature on the hazardous effects of Cu is rather scant. However, examples of other heavy metals are available. Underwood (1971) reported that Cadmium (Cd) is accumulative poison. Some of the prominent effects associated with Cd toxicity included pulmonary and testicular lesion, renal dysfunction, poor bone mineralization, anaemia, liver and kidney damage, retarded growth, disturbed carbohydrate metabolism and inhibition of drug metabolising enzymes (Piotrowski and Coelman, 1980). The most serious case of health hazards due to chronic Cd was observed in Japan, after world war II through ingestion of rice grown in the fields irrigated by polluted industrial waste water (UNEP, 1980; Hay, 1984). The disease, then called *itai itai kyo*, occurred most commonly among post menopausal women above 45 years of age.

2.6.5 Effects of heavy metals on soil microbiological processes

Heavy metals accumulated in soils may interfere with essential microbial functions in soils, such as organic matter decomposition, nitrogen fixation and nitrification. Tiller and Merry (1981) reported that Cu retarded the rate of ammonification in vineyard soils in France. Nitrification was found to be reduced by excessive levels of Cu or Zn in soils (Liang and Tabatabai, 1978).

CHAPTER THREE

3.0 MATERIALS AND METHODS

3.1 Description of the study area

Arusha region is situated in the Northern Highlands of Tanzania, in the north-eastern part of the country. It lies between 2^0 and 6^0 South and 35^0 and 38^0 East (Figure 2). Arusha region is comprised of nine districts. Arumeru and Arusha districts are among of nine districts of Arusha region. Arumeru district is divided into two zones. The first one is the undulating lands surrounding Mt. Meru, with annual precipitation of 1000 - 1400 mm. The other is the plains, with isolated hills/depressions, receiving less than 800 mm annually. The district covers an area of about 2 900 square km and the population is about 322 000 people. Arusha district, a largely urban area with a population of about 135 000 people, is located inside rural Arumeru. This means that the agricultural production in Arumeru should, therefore, be regarded as supplying both districts.

3.1.1 Soils

Soils of Arumeru district are of volcanic origin, well drained moderately deep clay loams and clays of low natural fertility with good moisture holding capacities and generally classified as andosols. Due to their volcanic origin the soils are very fertile but easily erodible. The most fertile land with high rainfall is on the moderate and steep slopes of Mt. Meru and associated hills. The plains, with low rainfall, are used mainly as rangelands and they support dense livestock populations.

3.1.2 Water sources

There are seven major perennial rivers in the Arumeru district, some of which also traverse through Arusha district. They include Nduruma, Ngaramtoni, Maji ya Chai, and Tengeru rivers. Others are Makumira, Themi and Kikuletwa. All these rivers originate from the natural forests high up on the Mount Meru slopes. These rivers contribute to economic development of the district as they are utilised intensively for small scale irrigation.

3.1.3 Farming systems

The predominant farming system in Arusha is that of small holdings. There are state owned and private large scale farms. The majority of small scale farmers practise mixed farming systems. Family labour is the main production force although some farmers, having relatively large farms, are bound to hire labour. Coffee is the main cash crop in the district. This crop receives Cu fungicides as the main strategy of pest management.

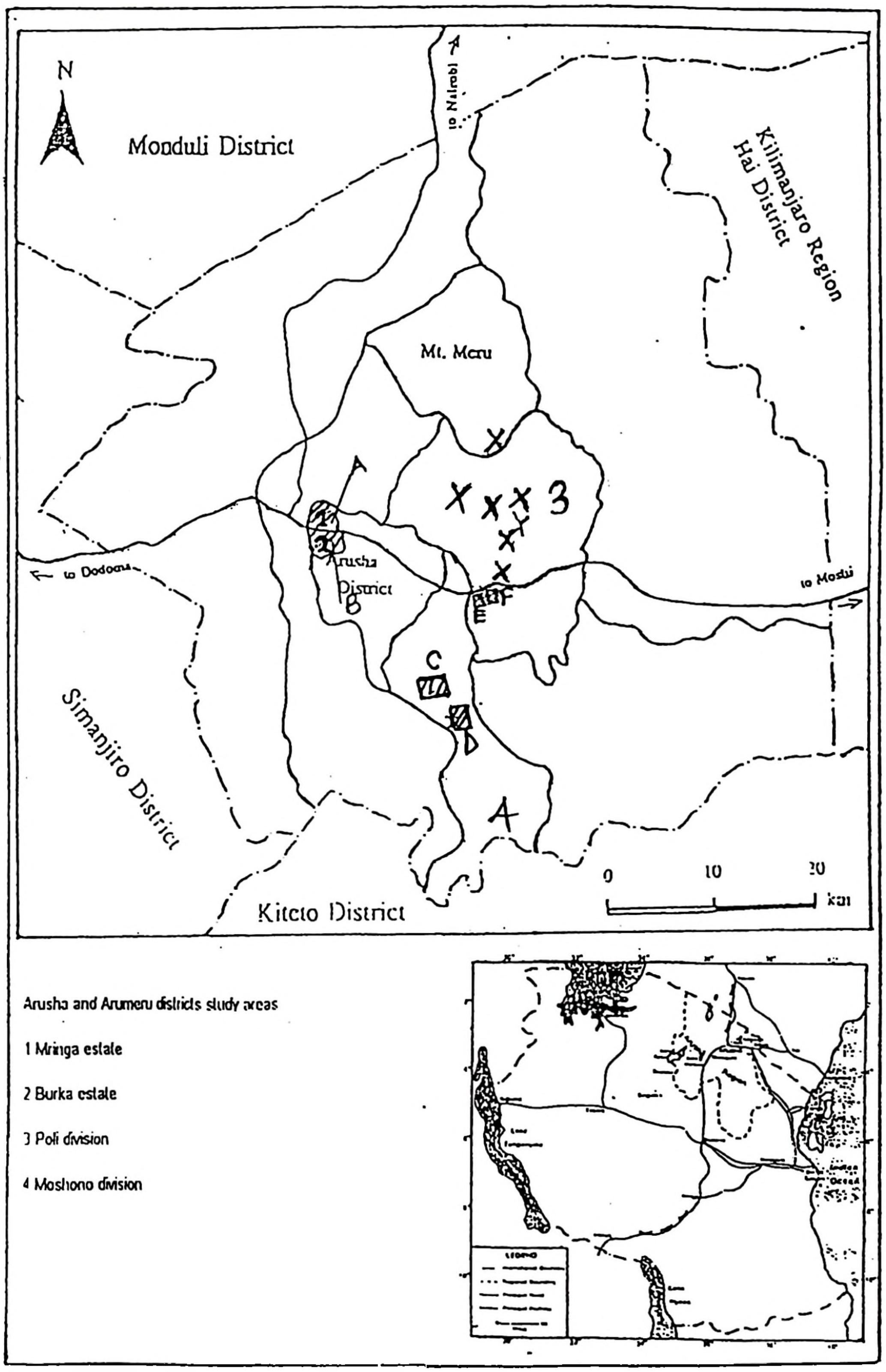

Figure 1. Maps showing the location of Arusha region, and Arusha and Arumeru districts where soil/water sampling was undertaken.

3.2 Site Selection

Prior to sampling, a survey was conducted in coffee growing areas of Arusha and Arumeru districts. The history of the fields i.e. age of the coffee, types, rates and the length of time Cu fungicides have been used were obtained through discussions with farmers and agricultural extension officers. Locations of sampling were randomly selected in some large scale farms, including Mringa (A) and Burka (B) estates found in Arusha district, Lá Fleur De Afrique (Malalua) (C) and Ragper (D) estates in Moshono division (Arumeru district), Ogaden (E) and Apostles of Jesus (F) estates in Poli division (Arumeru district). Those from small scale farms included Nguruma, Mraiwenyi Kimundo, Damu, Maksoru, Akheri, and Patandi villages (each village presented with a star) in Poli division (Arumeru district) and control samples were taken from the base of Mt. Meru as shown in Figure 1. Based on the information obtained, six categories of coffee fields were established as listed below:

(i) Fields which had not been under fungicide use (control fields),

(ii) Farms which had received Cu fungicides for 15 - 20 years and are still existing, under smallholder cultivation,

(iii) Fields which had been under Cu fungicides use for 50 years (or more) and are still existing, under smallholder farmers,

(iv) Fields which had been under Cu fungicides use for 50 years (or more) and are still existing, under large scale farmers,

(v) Fields which had been under Cu fungicides use for 20 - 30 years (or more) under large scale farmers, and are now abandoned for about 10 years, and

(vi) Fields which had been under Cu fungicide use for 25 - 30 years under small holder farmers, and are now abandoned for about 8 - 10 years.

3.3 Establishing the trends of Cu fungicide use

The trends of Cu fungicide use in selected fields was obtained by interviewing the farmers. The interviews were conducted in farmers fields prior to soil sampling. Coffee farmers in each of the categories established in section 3.2 were picked at random with the assistance of the local agricultural extension officers. About 14 coffee farmers whose farms are still operating, four farmers whose farms were used as fungicide controls sites were in the vicinity of coffee farms cultivated maize, fodder crops or grass for animals and three whose farms were abandoned were interviewed. The absolute control sites were virgin forest soil and a farm nearby cultivated beans without fungicide use on the slopes of the mountain. Information from the farmers was collected through interviews using a structured questionnaire as detailed in Appendix 1. The questionnaire was designed to capture information on the length of time the farm had existed, and on trends and types of Cu fungicides used.

3.4 Soil Sampling

Soil samples were obtained by opening minpits of about 10 cm deep by using hand hoe, while a field knife was used to dermacate and sample the top 5 cm layer. During this sampling four subsamples were collected and mixed to give composite samples. Four composite samples were collected from each field. The samples were placed in plastic bags and brought to the laboratory at the Sokoine University of Agriculture.

Due to the fact that some coffee farms are located in sloping areas, sampling in sloping fields was undertaken to evaluate the influence of slope on Cu movement (through erosion). Four samples or replicates were collected from upslope, midslope or downslope, to make a total of twelve samples.

All the soil samples were air-dried, ground and passed through the 2 mm sieve prior to determination of their physicochemical properties.

3.5 Soil analysis

3.5.1 Soil pH

Soil pH was determined in water and in 1M KCl solution. Ten gram soil samples were weighed into 100 ml plastic bottles and 25 ml of water or KCl were added. The mixture was shaken for 0.5 h on a reciprocating shaker and pH determined electrometrically using a pH meter.

3.5.2 Organic carbon

The soil's percent organic carbon was determined by the wet digestion method of Walkley and Black method as described by Page *et al.* (1982). One gram soil sample was weighed into a 500 ml erlnemeyer flasks. To the sample 10 ml of 0.1667 M potassium dichromate solution were added and the flask shaken for about 5 seconds. Then 20 ml of conc. H_2SO_4 were added. The flask was swirled for one minute and left on a wooden pad for 0.5 h. Then 200 ml of distilled H_2O, 10 ml of conc. H_3PO_4 and one ml of diphenyalamine indicator were added. The mixture was titrated with 0.5 M ferrous ammonium sulphate to the end point (brilliant green). A blank (only the reagents but no soil) was also treated in the same way for purposes of standardizing the ferrous ammonium sulphate solution. The volume of ferrous sulphate was used for calculating the per cent organic carbon content.

3.5.3 Particle size analysis

This was done by using Bouyoucos method as described by Gee and Bauder (1986), using sodium hexametaphosphate as the dispensing agent. A 50 g air dried soil sample was weighed into 250 ml plastic bottle. Fifty ml of 4% sodium hexametaphosphate were added, followed by 200 ml of distilled water. The mixture was shaken at 150 revolutions per minute (rpm) overnight in the horizontal position on a reciprocating shaker. It was then transferred to a one litre sedimentation cylinder. Distilled water was added to the mixture to bring it to the mark. The mixture was stirred for one minute and left to stand. Hydrometer readings were taken after 40 seconds, 4 minutes and 3 hours and the relative proportions of the soil particles (i.e. sand, silt and clay) were calculated. The texture was determined using the USDA textural class triangle.

Table 1 summarizes selected physical and chemical properties of the soils. The soil's clay contents were between 11.2 and 76%; silt separates ranged from 11.7 to 58.4% and sand separates ranged from 4.8 to 61.2%. The textural classes of the soils varied between sandy loam and clay loam, which should ensure good physical conditions. The pH of the soils ranged from 5 to 7.74 which are rated as being very strongly acidic to mildly alkaline by (Landon, 1991). The organic carbon of these soils ranged from 0.28 to 13.07%, which rated as being very low to very high (Landon, 1991). The higher organic matter in the top soil may imply that most of the Cu input from the fungicides applied may be retained in the top soil.

Table 1: Some physical and chemical properties of the soils sampled

Parameter	n	Minimum	Maximum	Mean ± sd
pH water	116	5.0	7.74	6.57 ± 0.60
pH KCl	92	3.68	6.68	5.26 ± 0.61
% OC	116	0.28	13.07	5.32 ± 2.38
% sand	116	4.8	61.2	34.4 ± 13.0
% silt	116	17.2	58.4	35.3 ± 6.9
% clay	116	11.2	76.0	30.3 ± 16.1
Textural class	116	sandy clay loam	clay loam	

n = sample size   sd = standard deviation

3.5.4 Total copper determination by the aqua regia digestion method

From the 2 mm sieved samples, 0.5 g of soil (1.0 g for control soils) was weighed and transferred into a 150 ml conical flasks. To the sample in the flask 6 ml concentrated nitric acid and 14 ml concentrated hydrochloric acid (this mixture is called aqua regia) were added (Jeng and Bergseth, 1992). The samples were swirled to mix and left to stand overnight. The conical flasks were placed on a hot plate and boiled at 90 - 100 ^{0}C under reflux conditions for two hours. Refluxing was stopped and boiling continued until the solutions almost dried, but without caking. The samples were cooled and the residues dissolved in 10 ml conc. HNO_3. The samples were transferred quantitatively into 100 ml volumetric flasks and made to the volume using distilled water, thus making a matrix of 10% HNO_3. They were filtered through Whatman no. 1 filter paper and transferred to washed 100 ml plastic bottles. These plastic bottles had been prewashed by soaking in 6 N HCl and rinsing thoroughly with distilled H_2O. Total Cu content in the digests was determined using flame Atomic Absorption Spectrophotometry (AAS) at 324.7 nm, with appropriate standards prepared in a similar matrix (i.e. 10% HNO_3).

3.5.5 Extraction of Cu from soils using DTPA solution

The aim of this experiment was to determine the amounts of extractable Cu released into the soil solution, since this may correlate with forms solubilized under natural soil conditions and thus translocated/leached through water movement or which may be taken up by growing plants and consequently circulate in food chains. A DTPA extracting solution was prepared which contained 0.005 M DTPA, 0.01 $CaCl_2$, 0.1 M Triethanolamine (TEA), adjusted to pH 7.3 with 1:1 N HCl (Lindsay and Norvell, 1978). Twenty grams of the 2 mm sieved soil samples were weighed into 250 ml plastic bottles, and 40 ml of the 0.005 M DTPA solution were added and the bottles screw-capped tightly. The mixture was then shaken for two hours on a reciprocating shaker at 175 rpm. The suspension was filtered through Whatman No. 1 filter paper and the Cu in the extract determined by AAS, with appropriate standards.

3.6 Plant sampling, processing and analysis

Five-week old bean (*Phaseolus vulgaris,* L.) plants were sampled by cutting at the ground level. Twenty such plant samples were separately taken from coffee farms of different categories (i.e. coffee farms aged 25 years, between 30 - 40, over 50 years) and from a vegetable field as a control sample. At the points of plant sampling, soil samples (which were associated with the plants) were also collected.

The plant samples were washed using distilled water to remove any adhering soil and other contaminants and placed in clean paper bags. They were dried to constant weight at 65 °C. They were chopped to small pieces using a pair of scissors and then finely ground using a mortar and pestle. About 0.5 g or less (depending on plant material available) were weighed and placed in clean porcelain crucibles and ashed in a muffle furnace at 550 °C for 8 hours. To the ash 1 ml of conc. HNO_3 and 2 ml conc. HCl were added and the mixture boiled to dryness on a hot plate. The residue was then dissolved in 5 ml conc. HNO_3 and made to the volume of 50 ml using distilled water (making a 10% HNO_3 matrix). The suspension was filtered using Whatman no. 42 filter paper and Cu determined using AAS.

The soil samples associated with the plants were processed as in section 3.4 and analysed routinely (sections 3.5.1 - 3.5.3) and for Cu (as in sections 3.5.4 and 3.5.5).

3.7 Water and sediment sampling and analysis

Runoff water and eroded sediments were collected from sloping land of coffee field. Also water samples (approximately 300 ml) were taken from four different points along the northern shore of lake Duluti, which is nearby or adjacent to abandoned coffee fields further north. Sediments were taken at the same points where water was sampled.

The water samples were filtered and Cu contents determined by AAS. The sediments were air-dried, ground and sieved as for soils in section 3.4. They were extracted using DTPA and digested using aqua regia as in the case of soil samples, and Cu determined by AAS.

3.8 Statistical analysis of data

The total Cu contents of soils, sediments, plant materials, as well as DTPA-extractable Cu contents were subjected to analysis of variance using the Winks computer software. This was done using the completely randomized design to evaluate the effects of duration of fungicide use and type of farming (either small holder farmer or large scale farmer, abandoned farm or farm still under coffee production) on Cu contents of soils. The Tukey multiple comparison test was used to rank the means.

The DTPA-extractable soil Cu and total Cu were regressed on soil properties (pH in water, percentage organic carbon and percentage clay) using the simple linear regression model. Copper concentrations of the *Phaseolus vulgaris* plant material as well as Cu uptake were similarly regressed on the total Cu as well as on DTPA-extractable Cu contents of the soils from which the plants were sampled.

CHAPTER FOUR

4.0 RESULTS AND DISCUSSION

4.1 Trends of Cu fungicide use on coffee in Arusha and Arumeru districts

Occurrence of fungal diseases in coffee has necessitated the use of fungicides. The main fungal disease of coffee reported by farmers during interviews was C.B.D. Farmers reported that frequent occurrence of C.B.D. infestation resulted in low yield and poor quality of their coffee. Infestation was highest during the rainy seasons and the spread of the disease was very fast during these seasons. They observed that if fungicides were not used during these periods, death of coffee seedlings, failure of plants to give good quality berries, or total loss of produce (coffee berries) usually resulted in.

Control of fungal diseases is based on use of Cu fungicides. The most common fungicides used in Arusha and Arumeru districts are those of Cu, namely Blue Cu ($CuSO_4.5H_2O$, 25.2 %Cu) and Red Cu (Cu_2O, 89 %Cu). According to the farmers, Blue Cu was used to control fungal diseases, and Red Cu was used in controlling diseases and to enhance the ripening of the coffee berries.

Some 30 years back, farmers were given incentives such as credit for fertilizers and fungicides. The money used in the purchase of these chemicals was directly deducted from the sale of coffee and farmers did not feel the impact of fungicide prices. During that period, many farmers managed to adhere to recommended rates of fungicide use. However, some other farmers believed that the recommended rates were not sufficient to effectively control diseases. Because the availability of fungicides was not a constraint, this resulted in misuse of fungicides by adopting higher rates and more frequent spray regimes within the growing period than were recommended.

Following the removal of credit as the co-operatives disintegrated, and as prices of fungicides increased, most of the farmers were unable to purchase the fungicides. Therefore, they applied the fungicides at lower rates and with less frequent regimes than those recommended. Todate, the high cost of production of coffee is claimed by most farmers as being caused mainly by persistent increases in the prices of Cu fungicides. The interviews revealed that coffee production would not be possible during the rainy seasons without the use of Cu fungicides. Still, some farmers operated within the fungicide levels and regimes as recommended, with very few farmers even exceeding the recommendations. These differential practices in the use of these fungicides may result in differential accumulation of Cu in soils of different fields. This is further discussed in sections 4.2 and 4.3.

During discussions with the farmers, it was observed that some of them did not have exact recollection in terms of the number of years they had been using fungicides in their farms and when exactly the farms were established. This was particularly the case with the farmers, who had inherited the farms from their forebears. Therefore, some of the ranges of years reported herein could be somewhat under-or overestimated. However, the general consensus is that long-term use of Cu fungicide would lead to enrichment of soils with Cu, over and above background values.

4.2 Accumulation of Cu in soils of small scale coffee farms

4.2.1 Total copper

The total Cu contents in soils of small scale coffee farms are presented in Table 2. Total Cu in soils receiving Cu fungicides was significantly ($P=0.05$) higher in comparison with the absolute control and fungicide control soils. The levels of Cu in the absolute control (virgin) soils were consistently lower than in sites where fungicide control soils were sampled. Relative to control soils, the total Cu contents of the soils generally increased with length of duration of fungicide use.

The significantly higher amounts of total Cu in the soils receiving Cu fungicides may be attributed to the use and dosage or intensity of application within the individual farms. Use of Cu fungicides presents an external input of Cu, which raises Cu levels in those soils to exceed background levels. In terms of levels or doses of use, it was reported by farmers in interviews (section 4.1) that some farmers believed that the recommended rates were not sufficient to effectively control diseases. This has resulted in misuse of fungicides in terms of using higher rates and frequencies of spraying within the growing period than were recommended. This contributed to a faster build up of Cu levels in the soils. The maximum total Cu contents in farms of over 50 years in small scale farmers, of 2670 mg/kg relative to a maximum of 96 mg/kg in absolute control soils, is testament to the contribution of continual use of Cu fungicides on Cu build up in those soils. Similarly, Dickison and Nicholas (1984) found the surface litter of a 68 years old coffee plantation receiving continued applications of Cu based fungicides to contain about 2000 mg/kg total Cu (extracted with 71% HNO_3), while surface (0 - 10 cm) soil beneath the litter contained 236 mg/kg. Total Cu of up to 3150 mg/kg in soils receiving Cu fungicides were reported by McBride and Bouldin (1984). The higher total Cu contents in the soils of the present studies imply greater possibility of that Cu being a source of contamination of crop plants.

The differences in Cu contents between the fungicide control soils and absolute control or virgin soils may be attributed to the influence of wind drift from the nearby sprayed fields since the collection of soil samples from fungicide control farms was done in areas, which were in the vicinity of sprayed coffee fields. On the other hand, absolute control soils were obtained from a natural forests high up on the slopes of Mt. Meru away from the coffee growing zone and from influences of wind drift. The fact that the lower levels were observed in the absolute control sites suggests that the controls are a reflection of natural levels in these soils. The Cu concentrations of the absolute controls soils, at 39 - 96 mg/kg soil, are within the range reported by Thornton and Abrahams (1984), who pointed out that total Cu contents in uncontaminated soils lied between 2 - 100 mg/kg.

Table 2: Total copper contents (mg/kg) in soils of small scale coffee farms

Soil category	n	Copper contents Minimum	Maximum	Mean ± sd
		---------- mg/kg soil ----------		
Absolute control (virgin forest soil)	8	39	96	57.0c ± 16.9
Fungicide control	16	75	430	262.8bc ± 108.4
Farms operated for 20-30 years, then abandoned	4	320	700	515.0abc ± 162.8
Farms operated for 15 - 20 years	16	50	1730	715.6ab ± 524.8
Farms operated for over 50 years	20	480	2670	1210.0a ± 656.1

Means within the same column followed by the same letter(s) were not significantly (P=0.05) different according to Tukey's multiple comparisons test.

n = sample size sd = standard deviation

Relative to control soils, the total Cu contents of the soils generally increased with length of duration of fungicide use. This implies that fungicide used over time had a residual effect and its accumulation depend on the duration, intensity and amount of fungicide use. The Cu concentration of farm operated for 15 - 20 years soils had lower 50 - 1730 mg/kg soils in comparison to soils in farm operated for over 50 years as indicated in Table 2.

4.2.2 DTPA-extractable Cu

The levels of DTPA extractable Cu in soils of small scale coffee farms are presented in Table 3. There were variations in the amounts of DTPA extractable Cu among sites, which received fungicides and in controls soils.

Farms operated for over 50 years had significantly higher ($P=0.05$) Cu contents than farms of the other durations of fungicide use. The DTPA Cu concentrations in the absolute control and fungicide control sites were consistently lower than in soils receiving Cu fungicides.

With the exception of the soils of abandoned fields, the trends of DTPA extractable Cu in the different categories of soils were similar to those shown by total Cu (section 4.2.1). The factors deemed responsible for the trends as discussed in section 4.2.1, that is use and dosages of Cu fungicides, may also apply in this case. The DTPA extractable Cu and total Cu followed same trends as they have a significant positive correlation as discussed in section 4.5.

Table 3: DTPA extractable Cu (mg/kg) in soils under small scale coffee farms

Soil category	n	Copper contents		
		Minimum	Maximum	Mean ± sd
		----------- mg/kg soil -------------		
Absolute control (virgin forest soil)	8	0.6	8.4	3.9c ± 3.40
Fungicide control	16	1.4	13.2	6.9c ± 5.0
Farms operated 20-30 years, then abandoned	4	156.0	222.0	195.3ab ± 28.0
Farms operated 15-20 years	16	21.6	304.0	103.7bc ± 95.4
Farms operated for over 50 years	20	59.0	556.0	209.7a ± 157.8

Means within the same column followed by the same letter(s) were not significantly (P=0.05) different according to Tukey's multiple comparisons test.

4.2.3 Differences in Cu contents in soils of different small scale farms

4.2.3.1 Differences in soils farms operated between 15 - 20 years

The total and DTPA extractable Cu contents of soils in these farms are presented in Table 4. The two indices of Cu showed significant (P=0.05) differences between farms.

Table 4: Copper contents (mg/kg) in soils of farms of 15 - 20 years of operation and fungicide use

		Total Cu		DTPA extractable Cu	
Farm number	n	Range	Mean ± sd	Range	Mean ± sd
		--------------------	mg/kg	--------------------	
Farm 10	4	1080 - 1730	1380.0a ± 270.7	61.0 - 229.0	143.0ab ± 70.4
Farm 11	4	610 - 1120	947.5a ± 234.2	55.2 - 304.0	198.6a ± 118.8
Farm 12	4	50 - 450	240.0b ± 209.4	21.6 - 48.0	33.3b ± 12.2
Farm 13	4	180 - 390	295.0b ± 86.6	25.2 - 58.8	39.9b ± 15.3

Means within the same column followed by the same letter(s) were not significantly (P=0.05) different according to Tukey's multiple comparisons test.

The differences in Cu levels between the farms with four sample size each may be attributed mainly to farm management in terms of fungicide use, according to information obtained through interviewing farmers. The farmers reported wide variability in rates and intensities (or regimes) of application of Cu fungicides. Some farmers reported using lower, some higher, rates and intensities than those recommended. This, then, resulted in differences in Cu accumulation in the soils of the different farms.

4.2.3.2 Differences in soils of farms operated for over 50 years

Total Cu and DTPA extractable Cu contents of soils in farms operated for over 50 years by small-scale holders are summarized in Table 5. Both total Cu and DTPA extractable Cu in soils of farms 14, 15, 19 and 20 showed no significant (P=0.05) differences as indicated in Table 5. For farm 21, both parameters were significantly (P=0.05) higher than those in the rest of the farms.

The lack of significant (P=0.05) differences in Cu in soils of farms 14 to 20 may be due to the fact that low rates and fewer regimes of fungicide use were adopted. Such farmers would, reportedly, below recommended rate due to unaffordable high price and therefore apply 1 - 1.5 kg of Cu per acre at a frequency of 2 - 3 times per season. This would contribute to slow down the rate of Cu accumulation in the soils.

Soils of farm 21 had Cu concentrations, which were significantly (P=0.05) higher in comparison with those of the other four farms. This is attributed to the fact that the owner of this farm reported to have adopted higher rates and intensity of fungicide use than the rest of the farmers surrounding him (Ndelilio, personal communication, 1999). His farm was observed to have healthy and evergreen coffee plants, and production was reportedly high. However, it was learnt that this farmer has other sources of income such as a carpentry workshop, dairy cattle, and educated sons working in different places. All these could also have contributed to enable this farmer to afford a higher level of use of the Cu fungicides. However, this was not explicitly conceded. This farmer reported a 5 - 6 time fungicide application regime in one season, each application consuming about 2 kg of Cu fungicide per acre. This might have been contributed by rain washing off with fungicide shortly after a previous spraying. Therefore, the need to spray again, as much as is feasible, to attain effective control of diseases was felt. This was a higher level of use of these fungicides as compared to the farmer's neighbors. Accordingly, more Cu was accumulated in the soils of his farm as shown in Table 5.

Table 5: Copper contents (mg/kg) in soils of farms of over 50 years of operation and fungicide use

		Total Cu		DTPA extractable Cu	
Farm number	n	Range	Mean ± sd	Range	Mean ± sd
		- - - - - - - - - - - - - - - - - -	mg/kg	- - - - - - - - - - - - - - - - - -	
Farm 14	4	480 - 1070	762.5b ± 242.5	71 - 181	148.30b ± 51.8
Farm 15	4	650 - 1450	1185.0b ± 361.9	91 - 249	190.8b ± 72.9
Farm 19	4	650 - 1330	912.5b ± 306.4	59 - 181	95.8b ± 57.8
Farm 20	4	760 - 1110	895.0b ± 151.6	72 - 184	146.1b ± 51.4
Farm 21	4	1440 - 2670	2295.0a ± 576.0	223 - 556	467.7a ± 163.2

Means within the same column followed by the same letter(s) were not significantly (P=0.05) different according to Tukey's multiple comparisons test.

4.3 Accumulation of Cu in soils of large scale coffee farms

4.3.1 Total copper

The total Cu contents in soils of large-scale farms are presented in Table 6. The levels of total Cu in the absolute control or (virgin) soils were significantly (P=0.05) lower than fungicide control soils and in farms where fungicide are used. Copper contents of fungicide control soils were not significantly (P=0.05) different from those of farms operated for 25 - 30 years and then abandoned. Copper contents in soils collected from soils of farms operated for over 50 years were significantly (P=0.05) higher than those from the soils of the other categories of farms.

The absolute control or (virgin) soils ranked the lowest and, again, this is a reflection of background Cu concentrations in these soils. Tornton and Abrahams (1984) reported the total Cu concentrations of uncontaminated soils to lie between 2 - 100 mg/kg. However, fungicide control soils in the present studies were out of that range. The fact that the higher Cu concentrations were recorded in fungicide control soils than absolute control or virgin soils could be attributed to wind drifts of fungicide droplets in nearby fields during spraying. Also, soil erosion and run off might have contributed to increased Cu contents of fungicide control soils by depositing Cu from nearby fields.

The significantly (P=0.05) higher contents of Cu observed in soils of farms operated for over 50 years compared with the rest of the soils in other soil categories may be attributed to the long duration of farm operation and fungicide use.

Table 6: Total copper contents (mg/kg) in soils of large scale coffee farms

		Total Cu		
Soil category	n	Minimum	Maximum	Mean ± sd
		------------	-mg/kg ------	--------
Absolute control (virgin forest soil)	8	39	96	57.0c ± 16.9
Fungicide control	16	75	430	262.8b ± 108.4
Farms operated for 25-30 years, then abandoned	8	90	760	407.5b ± 216.1
Farms operated for over 50 years	16	630	1400	967.5a ± 237.3

Means within the same column followed by the same letter were not significantly (P=0.05) different according to Tukey's multiple comparisons test.

4.3.2 DTPA-extractable Cu

The levels of DTPA-extractable Cu in soils of large scale farms are presented in Table 7. Generally, there were no variations in the amounts of DTPA-extractable Cu in soils of absolute control sites, fungicide controls and farm operated for 25 - 30 years before being abandoned. But the concentration of extractable Cu in fungicide control is very low in comparison with high total Cu as presented in Table 6 in the same category. This might imply that the applied Cu became tightly held or adsorbed by the soil's colloidal complex that it would not be easily extracted, hence the low level of extraction by DTPA. The Cu contents in soils of farms operated for over 50 years were significantly (P=0.05) higher than those in the rest of the soils.

The significantly higher amounts of DTPA extractable Cu in soils of farms operated for over 50 years may be a consequences of the high total Cu contents in these soils. The dependence or relationship of DTPA extractable Cu on total Cu is discussed in section 4.5.

Table 7: DTPA-extractable Cu (mg/kg) in soils of large scale coffee farms

Soil category	N	Extractable Cu minimum	Extractable Cu maximum	Mean ± sd
		------------ mg/kg	-------------	
Absolute control (virgin forest soil)	8	0.6	8.4	3.9b ± 3.4
Fungicide control	16	1.4	13.2	6.9b ± 5.0
Farms operated for 25 - 30 then abandoned	8	27.6	112.8	57.6b ± 27.7
Farms operated for over 50 years	16	86.0	385.6	162.2a ± 82.1

Means within the same column followed by the same letter are not significantly (P=0.05) different according to Tukey's multiple comparisons test.

4.3.3 Differences in Cu contents in soils of different large scale farms

4.3.3.1 Differences in soil Cu of farms operated between 25 - 30 years and then abandoned

The total and DTPA extractable Cu contents of soils in these farms are presented in Table 8. Both parameters showed no significant (P=0.05) differences between farms.

The lack of significant (P=0.05) differences in soil Cu levels between these farms may be attributed to similar durations of farm operation and fungicide use across these farms. The DTPA extractable Cu normally is correlated with the total Cu contents and, therefore, factors affecting total Cu may have direct influence on DTPA Cu. Other factors attributed to this can be soil characteristic such as organic carbon and clay contents responsible for retention or storage of Cu by the time farms were abandoned.

Although the farms have stayed abandoned for a long period now, the Cu residues still exist in soils of these farms. The major factor contributing to this may be the high amounts of organic matter, which may have contributed to Cu retention in soils. Therefore, soils of these abandoned fields will continue to be potential sources for further dispersion of Cu to the rest of the environment, including water supplies, edible plant/crop parts and other organisms. This could, eventually, pose risks to human health (Underwood, 1971).

Table 8: Total and DTPA Cu in (mg/kg) soils of large scale farms abandoned after 25 - 30 years of coffee production and fungicide use.

		Total Cu		DTPA Cu	
Farm number	n	Range	mean ±sd	Range	mean±sd
		mg/kg			
Farm 8	4	360 - 620	470.0a ± 122.8	27.6 - 76.8	48.9a ± 20.5
Farm 9	4	90 - 760	345.0a ± 289.0	32.4 - 112.8	66.3a ± 34.1

Means within the same column followed by the same letter were not significantly (P=0.05) different according to analysis of variance.

4.3.3 2 Differences in soils Cu content of farms operated for over 50 years

The total Cu and DTPA extractable Cu contents in soils of farms operated for over 50 years by large scale farmers are presented in Table 9. Both total Cu and DTPA extractable Cu showed significant (P=0.05) differences between farms.

The major factor contributing to the differences in soil Cu may be the management practices in terms of fungicide use. It was observed during interviews with farmers that large farmers also differed in the levels, intensities and method of fungicide spraying. For example, farm 23 was observed to have high frequencies of fungicide use, using sprinklers. In farm 24 the fungicides were also applied using sprinklers but the frequencies and intensities were not as high as in farm 23. Therefore, Cu accumulation was higher in soils of farm 23 than in those of farm 24. The DTPA extractable Cu was found to follow the same trends of total Cu.

Table 9:Copper contents in soils of farms of over 50 years of operation and fungicide use

		Total Cu		DTPA extractable Cu	
Farm number	n	Range	Mean ± sd	Range	Mean ± sd
		--------------------	-mg/kg-	------------------	
Farm 22	4	1010 - 1340	1127.5a ± 147.5	92 - 177	119.8b ± 38.9
Farm 23	4	810 – 1400	1072.5a ± 276.6	162 - 386	246.2a ± 103.9
Farm 24	4	630 – 740	685.0b ± 53.2	86 - 92	88.3b ± 2.6
Farm 25	4	820 – 1200	985.0ab ± 157.8	163 - 225	194.5ab ± 30.1

Means within the same column followed by the same letter(s) were not significantly (P=0.05) different according to Tukey's multiple comparisons test.

4.4 Comparisons of Cu contents in soils between small and large scale farms

4.4.1 Comparison of Cu contents between soils of small and large scale farms, which were abandoned

The Cu contents of soils in abandoned large and small farms are presented in Table 10. Total Cu contents did not differ significantly. However, the concentrations of Cu were found to be significantly different (P=0.05) only in DTPA extractable Cu, with small scale farms having the higher value. Differences in intensity of Cu application between these categories of farmers during the operational years may account for the differences in Cu contents in those soils.

Table 10: Comparison of Cu contents (mg/kg) in soils of small and large scale abandoned fields

		Total Cu		DTPA extractable Cu	
Farm category	n	Range	Mean ± sd	Range	Mean ± sd
		mg/kg			
Small scale farms	4	320 - 700	515.0a ± 162.8	156.0 - 222.0	195.3a ± 28.0
Large scale farms	8	90 - 760	407.5a ± 216.1	27.6 - 112.8	57.6b ± 27.7

Means within the same column followed by the same letter were not significantly (P=0.05) different according to the analysis of variance.

4.4.2 Comparisons of Cu contents between soils of small and large scale farms of over 50 years of operation and fungicide use

The soils' total and DTPA extractable Cu in large and small-scale farms of over 50 years of fungicide use are presented in Table 11. The two indices of Cu showed no significant (P=0.05) differences between farms as indicated in Table 11.

The lack of significant differences in Cu between soils of large and small scale farms of over 50 years could be due to adoption of essentially similar fungicide use practices, although the levels were slightly higher in small scale farm soils. Also, the large variability observed in the organic carbon and clay contents of these soils might have contributed to mask any differences in Cu contents between these categories of soils.

Table 11: Comparison of farmers over 50 years of farm operation under large scale and small holder

		Total Cu		DTPA extractable Cu	
Farm category	n	Range	Mean ± sd	Range	Mean ± sd
		mg/kg			
Small scale farms	20	480 - 2670	1210.0a ± 656.1	59 - 556	209.7a ± 153.8
Large scale farms	16	630 - 1400	967.5a ± 237.3	86 - 386	162.2a ± 82.1

Means within the same column followed by the same letter were not significantly (P=0.05) different according to the analysis of variance.

4.5 Relationships between Cu contents and some soil physical and chemical properties

The regressions of total Cu or DTPA Cu on some soil properties were made from raw data presented in Appendix 2.0. The significant relationships were those of DTPA extractable Cu on total Cu with the coefficient of determination (R^2=0.79) as graph shows in Appendix 2.1, clay content (Appendix 2.2) but coefficient of determination was low. Those of total Cu on clay content and DTPA extractable Cu on % organic carbon were not significant with very low coefficient of determination value, hence they were not shown as graphs.

The significant relationship between DTPA extractable Cu and total Cu implies that the level of DTPA extractable Cu, as reflection of available Cu in soil solution, depends on the total Cu present in soils. The DTPA extractable Cu is a simulation of the Cu deemed available to be taken up by plants. Therefore, a significant correlation implies that an increase of total Cu in soils will result in increased levels of DTPA extractable Cu and, hence, plant available Cu. This is discussed in section 4.6.

The significant negative relationships between total or DTPA extractable Cu and clay content implies a significant role of clay in the retention of the Cu added to these soils through fungicide use. This means that DTPA extractable Cu was high at low clay content implying that clay content bound little Cu in the binding sites and the rest was in the soil solution which was easily extractable. These observations were contrary to those of Baruti (1997), who found no significant relationship between total Cu, DTPA extractable Cu and per cent clay in coffee-growing soils of Lushoto district, which also received Cu fungicides.

The significant relationship observed between DTPA extractable Cu and soil organic carbon content shows that organic matter has a high binding capacity for Cu and hence increases storage capacity of Cu in soils. This relationship may imply that soil organic matter is another major soil characteristic controlling total as well as DTPA-extractable Cu. Similarly, Semu and Singh (1996) reported significant correlation between soil organic carbon and extractable Cu.

4.6 Copper contents in young bean plants in relation to Cu concentrations in soils

The relationship between Cu contents in young bean plants with total and DTPA extractable Cu are summarized in Table 12. The DTPA extractable Cu showed a significant correlation (R^2=0.96) with total Cu in soils where plant samples were collected. Plant Cu concentration showed a significant relationship with DTPA extractable Cu (Appendix 3.1). Plant Cu uptake showed significant relationships both with DTPA extractable and total Cu (Appendix 3.2 and 3.3).

The significant relationship between DTPA extractable Cu with total Cu implies that increase of DTPA extractable Cu is determined by increase of total Cu in soil. This means that higher levels DTPA extractable Cu will make Cu to be available for plant absorption and hence high Cu uptake.

Lack of significant correlation between plant Cu concentrations with total Cu could be attributed to the fact that total metals in the soil are not good indices of availability of elements to plants.

Table 12: Relationship between Cu contents in young bean plants in relation to Cu concentrations in soils.

Regression equation	R^2	Probability
$Plant\_Cu_{conc.} = -13.43 + 1.06Ext.Cu_{soil}$	$R^2 = 0.244$	P=0.014*
$Plant\_Cu_{conc.} = -12.71 + 0.44TotalCu_{soil}$	$R^2 = 0.147$	P=0.065NS
Cu_uptake = -14.91 + 0.58Ext.Cu	$R^2 = 0.447$	P<0.001***
Cu_uptake = -20.92 + 0.27TotalCu	$R^2 = 0.332$	P=0.003**

NS = not significant

In the present study plant Cu concentrations and plant Cu uptake observed for beans were found to differ from one site to another as a result of variability in Cu contents in soils. These different contents of soil Cu were due to different duration of fungicide use (Appendix 3.0). Young bean plant samples from control sites contained

Cu concentrations, which were low (3.46 to 26.55 mg/kg) and were within the acceptable values, below toxicity values, reported by Geraldson *et al.* (1973) and Jarvis (1981). Geraldson *et al.* (1973) reported the normal range of Cu in mature trifoliate leaves of bean to be 15 - 30 mg/kg. Jarvis (1981) reported that 30 mg/kg Cu in plant tissue was probably the limit for most plants, above which toxicity symptoms may be observed.

Copper concentrations in plants from soils of farms aged 20 - 25 years of fungicide use ranged from 20.90 - 46.55 mg/kg and Cu uptake ranged from 6.55 - 10.80 μg per plant, which were slightly higher than the critical values reported by Jarvis (1981). For the plant samples taken from soils of farm operated between 25 and 40 years, Cu concentrations ranged from 45.71 - 1180.68 mg/kg and Cu uptake ranged from 12.8 - 400.3 μg per plant, which were much higher than the plant toxicity limit. However, these plants did not show Cu toxicity symptoms. The lack of plant toxicity under such conditions may be explained in two ways. Firstly, the plants were too young to show toxicity symptoms. Secondly, plants may be tolerant to high Cu levels in the tissues as has been observed by Reilly (1969) for flowers of some plants in Rhodesia (Zimbabwe).

However, Cu concentrations in bean collected from soil of farms of over 50 years of operation ranged from 19.26 - 376.05 mg/kg, with only three of the values above the toxicity limit. The Cu uptake in the same samples ranged from 5.75 - 314.00 μg per weight of plant as presented in Appendix 3.0.

Since the bean grain is widely consumed by humans in the Arusha area, with bean vegetative residues fed to animals, accumulation of high levels of Cu by plants is an important aspect in determining the amounts of metals circulating in the food chain in this area. Should high amounts of Cu be retained in the grain and vegetative residues, and consumption of such materials sustained over long periods of time, adverse health effects might result in.

4.7 Dispersion of Cu in the environment

4.7.1 Soil erosion and Cu movement in sloping lands

The Cu contents of soils in a sloping field are presented in Table 13. Generally, total and DTPA-extractable Cu in soil samples collected from the sloping farm were statistically (P=0.05) different in these three different slopes. The Cu contents in soils of the upper side of the slope were lower (P=0.05) while the Cu contents in the downslope side were highest.

The increasing trend of Cu contents downslope may be attributed mainly to heavy rains usually received in this area, which may have eroded and transported the top soil layers from the upper slopes and deposited the soils, with the Cu they contain, at the downslope part.

The higher Cu levels in the soils downslope confirm the contribution of soil erosion in the dispersion of Cu, its deposition or accumulation at a lower elevation, and the potential for polluting water sources/streams around or near these farms once the sediments are released directly into these sources. This conclusion is arrived at because of the fact that sampling was done in a single small field, with distances of about 5m between the upslope, mid slope or downslope points. Hence, there was not much variation in soil type and properties up and down the slope. This is further discussed in section 4.7.2. Wilcke and Kaupenjohann (1994; 1997) also raised the possibility of polluting water sources from polluted soils.

Table 13: Total Cu and DTPA-extractable Cu contents of soils in a sloping field, mg/kg

Position on slope	Total copper	DTPA-extractable Cu
	-----------------mg/kg soil	---------------
Upslope	957.5c ± 147.7	62.0c ± 6.2
Midslope	1425.0b ± 126.1	215.0b ± 26.5
Downslope	1820.0a ± 146.5	360.4a ± 47.4

Means within the same column followed by the different letter were significantly (P=0.05) different according to Tukey's multiple comparisons test.

4.7.2 Copper contents in some water resources

4.7.2.1 Copper contents in water and eroded sediments in a sloping farm

The Cu contents in runoff water collected from another sloping farm were determined. The total Cu contents in sediments and that in water were not statistically analysed but the amount detected are good indicators that the eroded soils and runoff water do normally move with the Cu they adsorbed. The amount of total Cu in sediments ranged from 297.6 - 996 mg Cu/kg soil in five selected points in the farm. These are much higher than the background levels of 96 mg Cu/kg soil as obtained from control soils. The Cu contained in such eroded sediments would contaminate nearby streams if the sediments are eroded into such streams. Gaciri and Mailu (1989) reported that some streams in Kenya were particularly enriched with Cu, Zn and Pb, which was attributed to flushing from coffee estates where large amounts of fertilizers and pesticides were used. The Cu content in runoff water ranged from 0.058 - 0.337 mg Cu/L collected in the same points as the sediments.

4.7.2.2 Copper contents in sediments and water sampled from Lake Duluti

The Cu contents in water and in sediments from lake Duluti are presented in Table 14. These were only occasional samples and were not subjected to statistical analysis. The pH ranged from 6.95 - 8.11. Total Cu contents in the sediments ranged from 21.1 - 46 mg Cu/kg soil, DTPA-extractable Cu in sediments ranged from 2.95 - 3.45 mg

Cu/kg soil and Cu content in water 0.04 - 0.06 mg Cu/L.

In comparison with the higher Cu contents in sediments collected from sloping land (section 4.7.2.1) the Cu contents in sediments from Lake Duluti were consistently lower. The amounts observed at Lake Duluti may reflect the natural levels of Cu as was observed in control soils (section 4.2.1). This seemingly uncontaminated state may be due to the fact that Lake Duluti is situated at the lower end of a gentle slope. This gentle slope, and the grass vegetation on an abandoned field adjacent to the lake may have reduced the movement of any eroded sediments carrying Cu, thus preventing the sediments and water from being polluted. The amount of Cu detected in water was similarly low in comparison with those observed in runoff water in section 4.7.2.1. This was only from essentially one location. It should not be interpreted as representing the rest of Lake Duluti. More sampling is necessary to confirm this trend, or otherwise.

Table 14: Copper contents in sediments and water from Lake Duluti

Sampling points *	pH water	Total Cu in sediments	Extractable Cu in sediments	Copper content in water
		(mg/kg)	(mg/kg)	(mg/L)
1	6.95	46.0	3.0	0.04
2	7.10	32.4	3.2	0.04
3	7.13	22.4	3.0	0.06
4	8.11	21.1	3.5	0.06

* Distances between sampling points were about 15 m.

4.8 General discussion

The study indicated that long-term use of Cu fungicides increases the amounts of Cu in the soil. The varying practices of farm management in terms of amounts of fungicide used, frequencies, and durations of farm operation and fungicide use resulted in variations of Cu contents in soils and plants of both small scale and large scale farms.

Generally, smallholder farmers seemed not to be aware of the adverse effects of using larger dosages of the Cu fungicides. The variations observed in fungicide use practices were attributed to cost and availability of these chemicals. High cost of the chemicals have forced farmers to use lower rates. Some chemicals or fungicides meant for coffee were diverted to vegetables, as they use same chemicals. This may have created scarcity, forcing some farmers to use lower rates and doses to meet prescribed number of sprayings. Washing off of fungicide by rain shortly after application leads to higher numbers of sprayings in order to ensure effective disease control. This may be commendable from the viewpoint of disease control, but it accelerates Cu accumulation in soils. Education on the effects of fungicides in the environment at large should be given to farmers, so that they adhere to recommended practices. Expired fungicides may lead to change of dosages or regimes as they may be of lower effectiveness in controlling the disease. Use of these may lead to repetition in fungicide sprays in the hope of meeting the strength of the unexpired chemical. Government should ensure that expired chemicals do not find their way to the market.

Therefore, these differential practices lead to different rates of Cu accumulations in soils of different farmers. The agricultural extension service should address itself to these matters so that recommended practices are followed so as to reduce the rate of accumulation of residues of these fungicides in the environment.

CHAPTER FIVE

5.0 CONCLUSION AND RECOMMENDATIONS

5.1 SUMMARY AND CONCLUSIONS

The results of the present studies have shown the presence of higher levels of Cu in coffee farms in Arusha and Arumeru districts and there were defined trends with increasing length of time of Cu fungicide use. The increase in Cu contents indicated significant (P=0.05) differences with respect to duration of fungicide use, as well as frequencies and amounts applied. The highest levels of Cu recorded were 2670 mg Cu/kg soil for total Cu and 556 mg Cu/kg soil for DTPA extractable in small-scale farms. The respective levels were 1440 mg/kg and 385.6 mg/kg in large-scale farms. This indicates that fungicide use practices and Cu contents in soils of these categories of farms were not uniform.

The Cu concentrations in plants were high in soils, which received Cu fungicides. The Cu accumulation in soils presently may cause toxicity to other crops such as bean plants grown in the fields receiving fungicide as compared to the lower levels in control soils. The highest-level recorded in fungicide treated soils was 376 and 1180.7 mg Cu/kg of plant material and 6.0 mg/kg in control soils. Therefore, long-term consumption of such crops grown in these soils may result in accumulation of high Cu in tissues of consumers and pose health risks.

5.2 RECOMMENDATIONS

In view of the findings from this study, the following can be recommended:

Farmers be educated, through the agricultural extension service, on the dangers of these chemicals that they are poisons, and that rampant use of them has negative environmental consequences.

Farmers be informed on the levels of Cu accumulated in these soils and that annual crops such as beans should not be intercropped in coffee farms with high soil Cu levels to avoid passing on high amounts of Cu to humans.

Expanded studies be conducted to evaluate Cu content of different crops and water resources in the vicinity of coffee farms as follow-up to the soil pollution already observed.

Further coffee breeding research be conducted on long-term basis to come up with cultivars resistant to fungal diseases to avoid further use of fungicides, thereby halting subsequent Cu accumulation and pollution.

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7.0 APPENDICES

Appendix 1.0: Checklist questions (questionnaire) used in farmer interviews

Trends of fungicide use in Arusha and Arumeru districts

Date . . . . Subvillage . . . . Village . . . .

Division . . . . District . . . .

A. Background information

Farmers name (Optional) . . . . . . Gender . . . . Marital status . . . .

Age of farmer . . . . . . Years in farming . . . .

B. Coffee and vegetable growing

Do you grow both coffee and vegetable? Yes/No

If yes which one . . . .

C. Information on the use of Cu fungicides

1. For how long the farm has been under coffee production?. . . . . .(years).
2. For how long the abandoned fields are not under coffee production and what other crops are grown in those farms?. . . . .(years).
3. What are the effects of fungal diseases farmers facing in their farms in terms of yields and/or crop quality.
4. Do farmers use copper fungicides in their production .Yes/No.

5. If yes what type of copper fungicides have they been using?.

(1). . . . .(2). . . . .(3) . . . .(4) . . . .

6. What crops do they intercrop in coffee farms?.

7. For how long have farmers been using these fungicides?.

8. At what rates do farmers use these fungicides and how many times per season/year?.

9. Do farmers know any adverse effects of using copper fungicides? Yes/No. If yes mention any. . . . .

10. Has production increased due to use of copper fungicides? Yes/No.

11. Are there any water resources/streams around or near the farms. Yes/No.

12. If yes, for what purposes is the water mainly for. . . . .

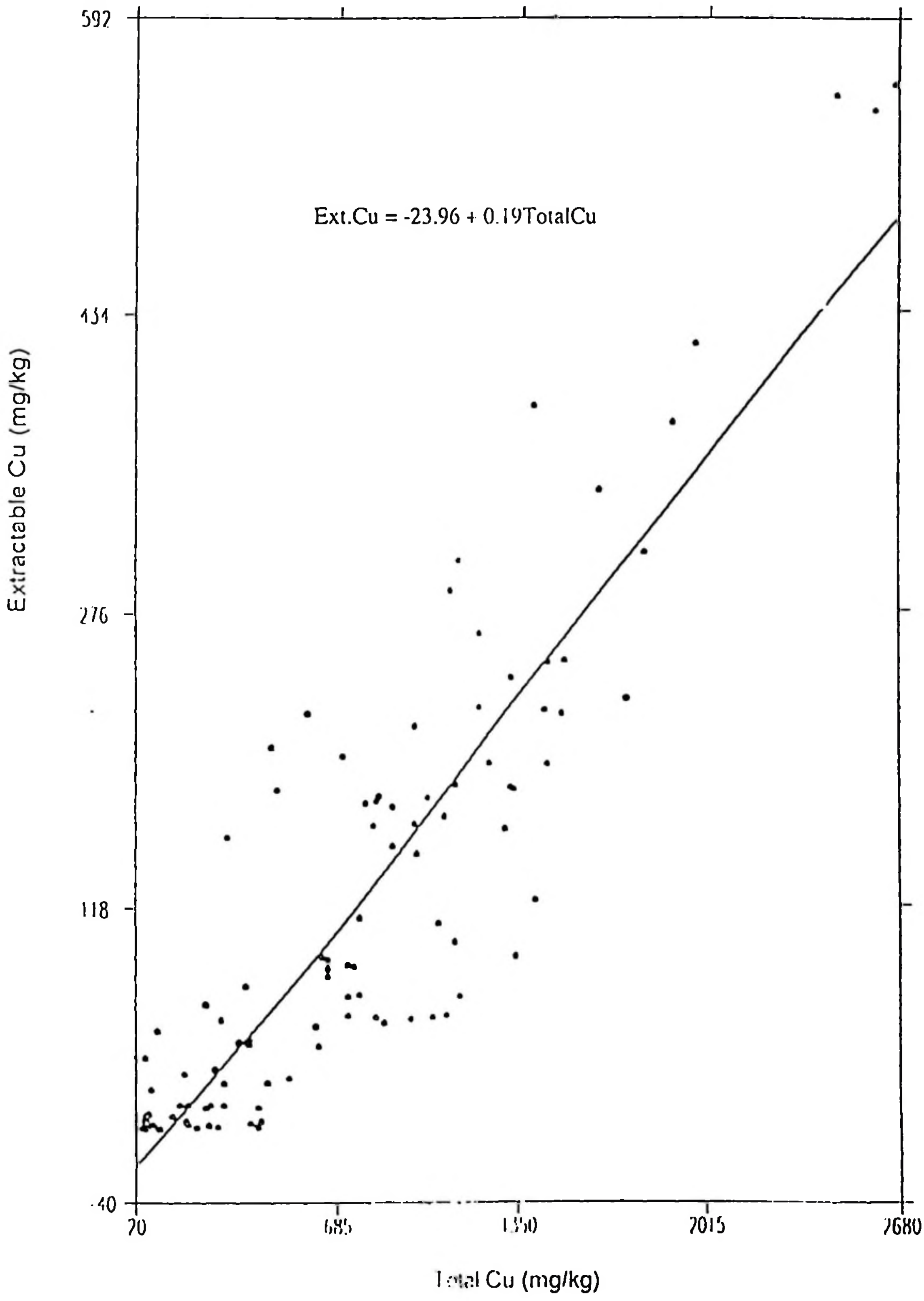
Appendix 2.0: Raw data for total and DTPA extractable Cu (mg/kg) with some physical and chemical properties of the soil

Farm type	Farm number	Total Cu	DTPA Cu	pH water	%OC	% Particle size distribution			Textural class
						%Sand	%Silt	%Clay	
Absolute control	Farm 1	96	0.7	5.05	13.07	51.2	33.2	15.6	SL
		57	1.5	6.0	10.34	53.2	29.2	17.6	SL
		39	0.8	5.91	12.71	59.2	27.2	13.6	SL
		50	0.6	6.19	11.52	61.2	23.2	15.6	SL
	Farm 2	54	6.7	6.45	7.72	55.2	31.2	13.6	SL
		52	7.7	6.49	7.56	53.2	35.2	11.6	SL
		60	8.4	6.25	7.48	53.2	35.2	11.6	SL
		49	5.1	6.12	7.56	53.2	35.2	11.6	SL
Fungicide control	Farm 3	190	3.1	6.92	2.89	41.6	42.4	16	L
		260	2.3	6.64	3.33	46.8	39.2	14	L
		220	1.4	6.92	3.05	42.8	45.2	12	L
		290	1.8	7.03	3.17	44.8	43.2	12	L
	Farm 4	250	12	7.39	3.88	25.2	41.6	33.2	CL
		430	4.8	7.32	4.20	22.8	37.2	40	CL
		420	12	7.64	4.20	27.2	37.6	35.2	CL
		395	3.6	7.13	4.60	26.8	37.2	36	CL
	Farm 5	420	1.6	7.03	4.78	27.2	35.6	37.2	CL
		75	2.8	6.85	3.05	27.2	29.6	43.2	C
		185	4.8	6.61	2.34	21.6	29.2	49.2	C
		140	7.2	6078	3.33	26.8	27.2	46	C
	Farm 6	190	13.2	6.9	1.50	6.8	23.2	70	C
		165	13.2	6.96	1.82	12.8	17.2	70	C
		265	13.2	6.68	1.50	8.8	19.2	72	C
		310	13.2	6.83	1.50	8.8	19.2	72	C
Small farm abandoned	Farm 7	700	199.2	7.19	11.44	42.8	29.2	28	C
		320	156	6.33	9.27	36.8	31.2	32	CL
		460	204	7.02	7.05	30.8	33.2	36	CL
		580	222	6.99	7.33	38.8	30	31.2	CL
Large farm abandoned	Farm 8	380	76.8	6.37	3.64	20.8	34	45.2	C
		360	46.8	6.42	3.25	12.8	35.2	52	C
		620	44.4	6.42	3.64	12.8	35.2	52	C
		520	27.6	6.74	2.45	10.8	35.2	54	C
	Farm 9	760	112.8	6.56	5.11	4.8	27.2	68	C
		90	52.8	5.83	3.68	6.8	21.2	72	C
		250	67.2	6.04	3.88	6.8	23.2	70	C
		280	32.4	5.36	2.81	4.8	19.2	76	C
Small farm 15 - 20 years	Farm 10	1080	61	6.84	3.96	48.8	35.2	16	L
		1300	160	6.85	4.43	42.8	41.2	16	L
		1410	122	6.52	4.63	44.8	37.2	18	L
		1730	229	6.26	6.53	44.8	37.2	18	L
	Farm 11	1120	304	6.43	5.39	46.8	37.2	16	L
		970	147	6.72	4.99	56.8	27.2	16	SL
		1090	288	6.42	6.22	44.8	37.2	18	L
		610	55.2	6.36	3.17	45.6	28.4	26	L
	Farm 12	390	48	6.57	3.72	41.6	31.2	27.2	CL
		450	25.2	7.04	4.87	22.8	35.2	42	C
		70	21.6	6.88	4.01	20.8	37.2	42	C
		50	38.4	6.47	3.60	20.8	35.2	44	C
	Farm 13	300	58.8	6.44	3.05	16.8	39.2	44	C
		180	30	7.18	3.25	20.8	37.2	42	C
		310	25.2	6.89	3.05	20.8	35.2	44	C
		390	45.6	6.76	2.73	20.8	31.2	48	C
Over50 years small farms	Farm 14	480	181	7.28	5.64	45.6	36.4	18	L
		780	174	6.54	4.66	41.6	36.4	22	L

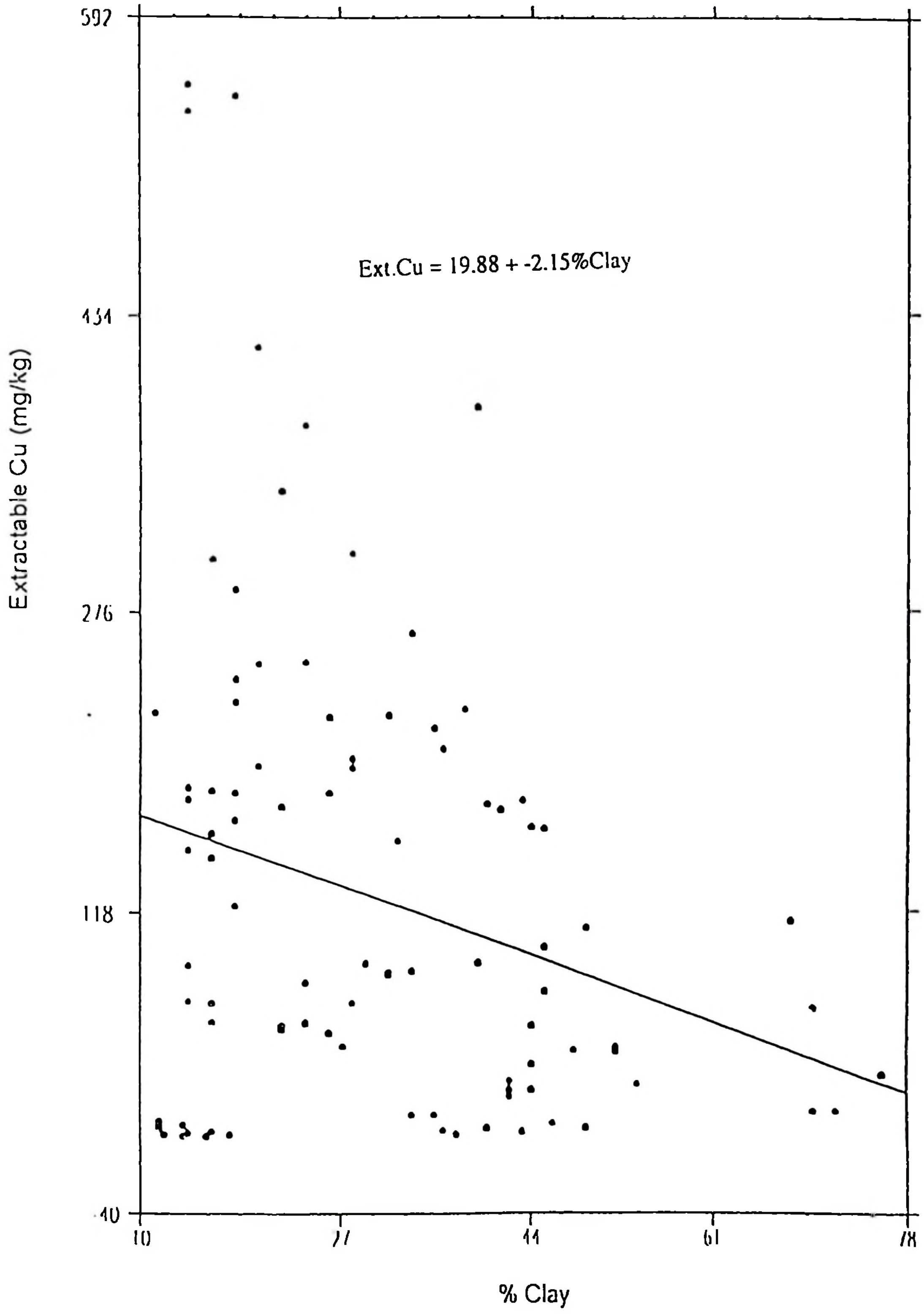
Appendix 2 continue

		1070	167	6.53	5.25	45.6	36.4	18	L
		720	71	7.74	5.02	45.6	38.4	16	L
	Farm 15	1320	241	6.55	7.18	35.6	46.4	18	L
		1450	249	6.91	9.47	41.6	38.4	20	L
		1320	182	6.84	6.11	43.6	40.4	16	L
		650	91	7.09	0.28	49.6	36.4	14	L
	Upslope	820	60	6.27	1.86	35.6	40.4	24	L
		850	57	6.03	3.92	37.6	40.4	22	L
		1030	60	5.87	4.12	39.6	36.4	24	L
		1130	71	5.56	5.80	36.8	325.2	28	CL
	Mid slope	1240	195	5.78	7.73	43.6	36.4	20	L
		1510	250	5.6	8.40	41.6	34.4	24	L
		1500	221	5.55	7.65	41.6	32.4	26	L
		1450	194	5.46	6.63	33.6	38.4	28	CL
	Down slope	1970	417.6	7.02	7.30	21.6	58.4	20	sL
		1890	376	6.15	6.70	37.6	38.4	24	L
		1630	340.8	5.82	7.81	50.8	27.2	22	SCL
		1790	307.2	5.37	9.62	38.8	33.2	28	CL
	Farm 19	650	82	6.7	2.38	25.6	30.4	24	sL
		720	61	7.06	6.74	27.6	48.4	24	sL
		1330	181	6.56	7.38	28.8	45.2	26	L
		950	59	7.01	6.19	32.8	45.2	22	L
	Farm 20	830	177.6	6.69	4.91	42.8	43.2	14	L
		760	72	6.93	4.32	42.8	43.2	14	L
		880	151.2	6.73	4.75	40.8	45.2	14	L
		1110	183.6	6.71	6.14	44	42	14	L
	Farm 21	2600	542	6.41	2.45	30	56	14	sL
		2470	550	5.98	5.90	44	38	18	L
		2670	556	6.5	6.53	47.2	38.8	14	L
		1440	223.2	6.99	4.87	49.2	39.6	11.2	L
Over 50 years large farms	Farm 22	1010	177	6.37	5.14	19.2	37.6	43.2	C
		1050	110	6.27	4.66	19.2	31.6	49.2	C
		1110	100	7.06	4.55	21.2	33.6	45.2	C
		1340	92	6.82	5.17	23.2	37.6	39.2	CL
	Farm 23	810	162	6.83	3.41	25.2	29.6	45.2	C
		1400	385.6	6.1	6.9	25.2	35.6	39.2	CL
		880	172	5.0	7.99	25.2	33.6	41.2	C
		1200	265	5.61	7.92	29.2	37.6	33.2	CL
	Farm 24	720	88	5.3	4.55	31.2	35.6	33.2	CL
		630	92	5.92	4.94	35.2	35.6	29.2	CL
		650	86	5.66	4.43	39.2	29.6	31.2	CL
		740	87	5.23	3.84	39.2	29.6	31.2	CL
	Farm 25	820	175	7.12	2.79	31.6	28.4	40	CL
		960	215	6.92	4.82	37.2	27.6	35.2	CL
		960	163	6.26	3.73	25.6	30.4	44	C
		1200	225	5.51	3.73	29.6	32.4	38	CL

C = Clay
CL = ClayLoam
L = Loam
SCL = SandyClayLoam
SL = SandyLoam
sL = SiltLoam



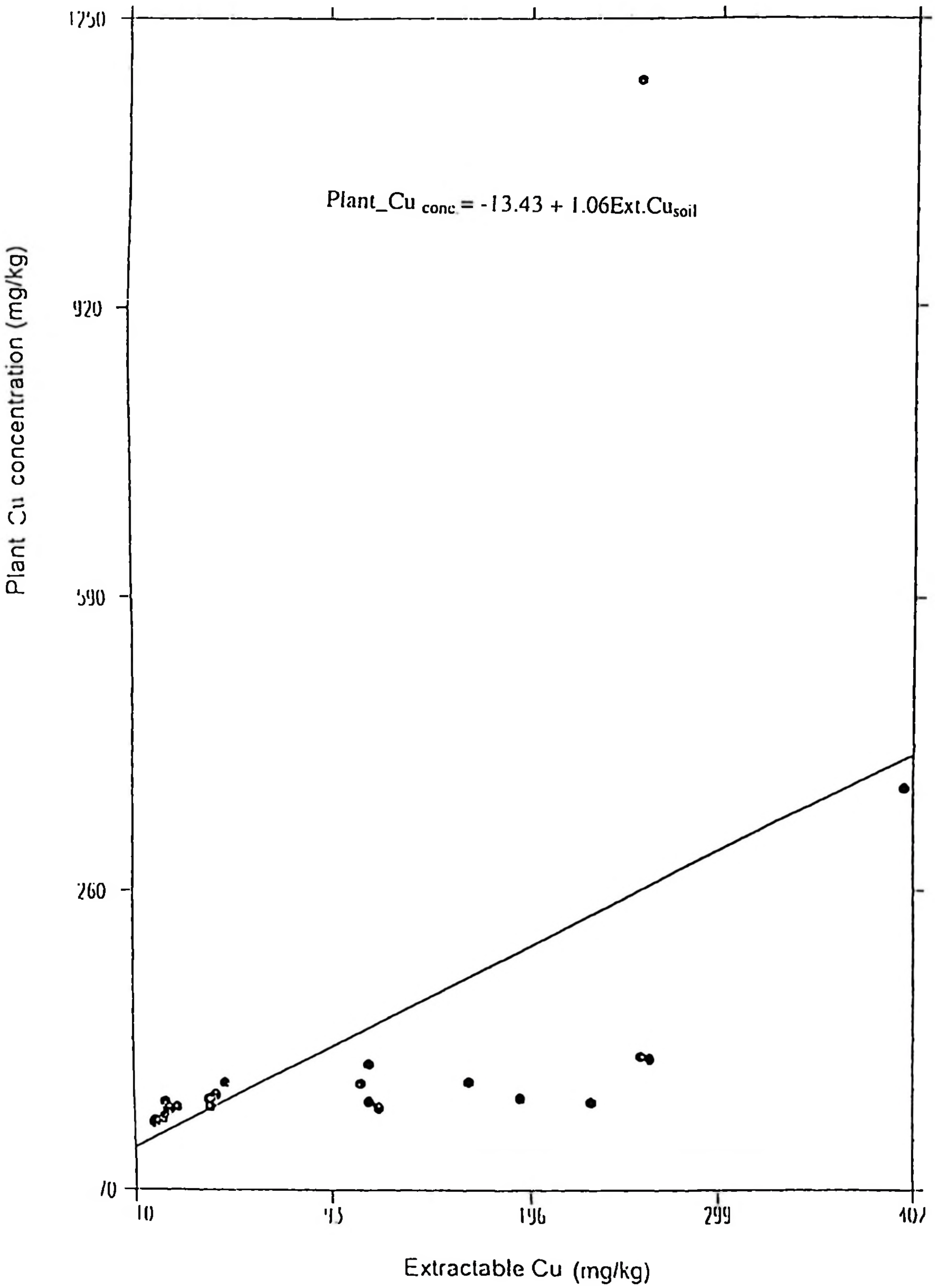
Appendix 2.1: Relationships between extractable Cu and total Cu (mg/kg).



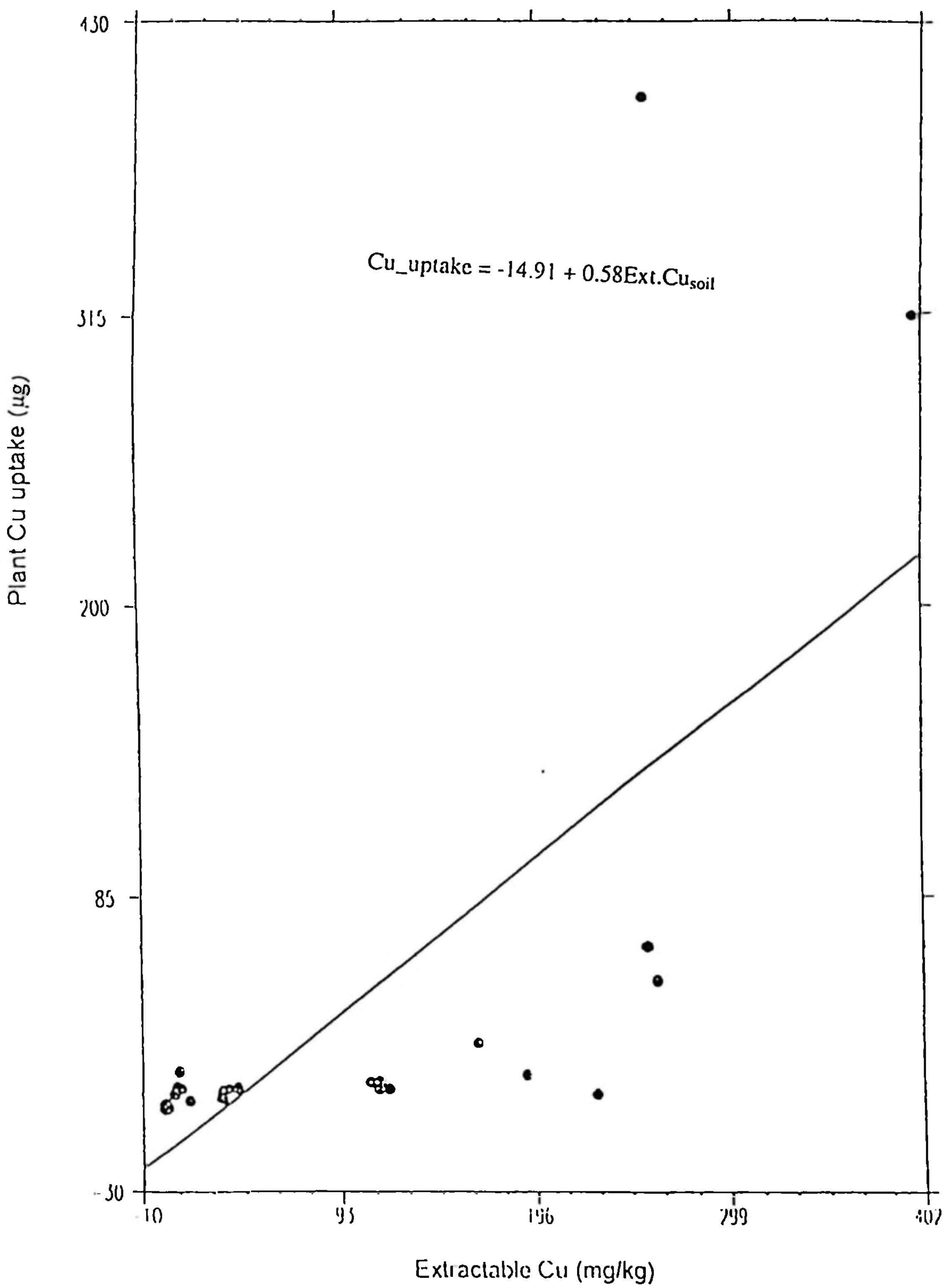
Appendix 2.2: Relationships between extractable Cu (mg/kg) and % clay.

Appendix 3.0: Total Cu contents, DTPA-extractable Cu, plant weight, plant Cu contents and Cu uptake.

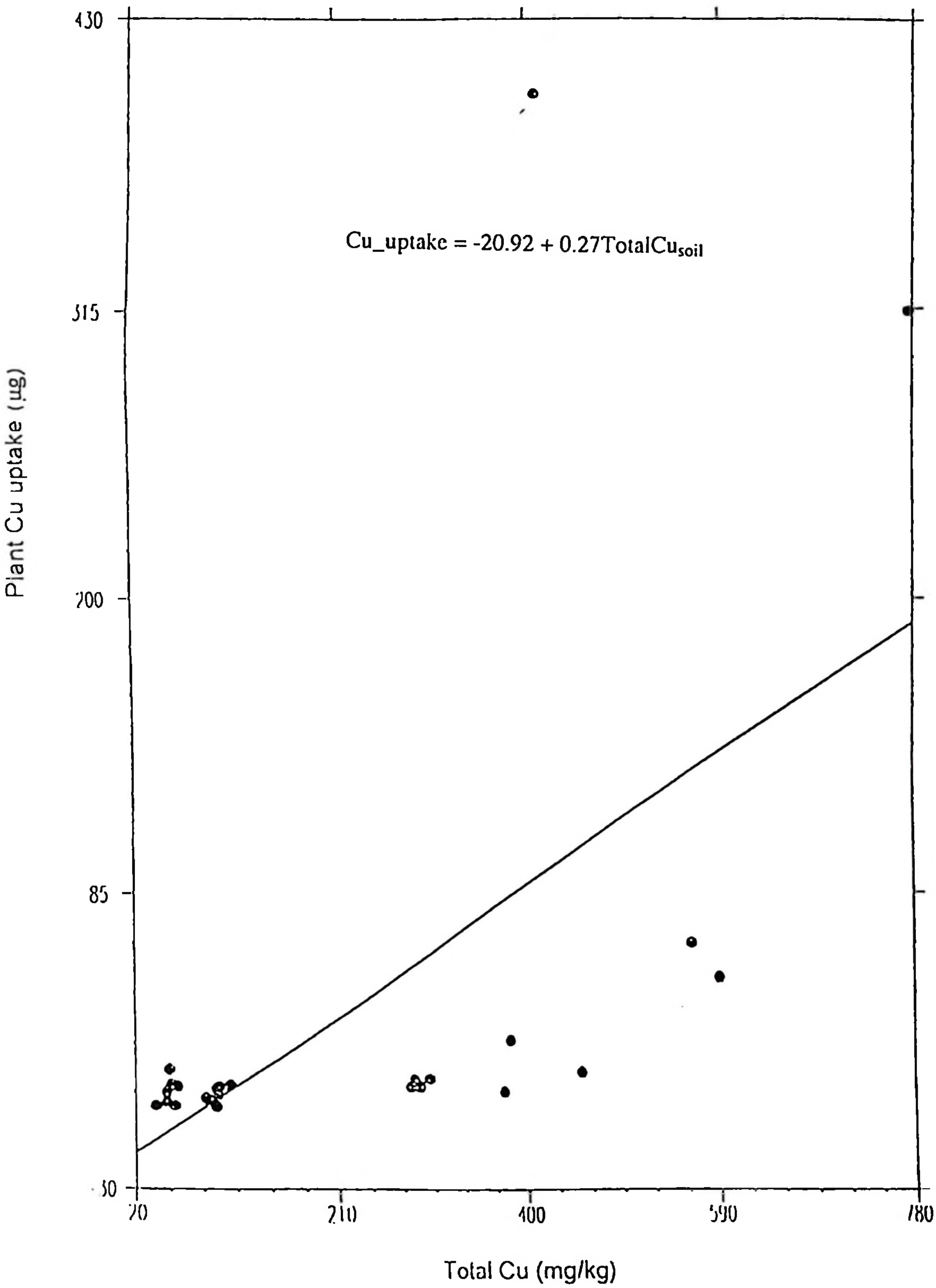
Sample number		Total Cu in soils mg/kg	DTPA Cu mg/kg	Plant weight (g)	Cu_{conc} in plants µg/g	Cu uptake µg/plant
Control site	1	96.1	0.692	0.693	3.463	2.40
	2	56.9	1.532	0.49	6.020	2.95
	3	39	0.756	0.528	5.492	2.90
	4	49.5	0.584	0.815	5.399	4.40
Vegetable site	5	53.6	6.722	0.42	26.548	11.15
	6	52.2	7.706	0.811	21.147	17.15
	7	59.6	8.404	0.636	16.588	10.55
	8	49.4	5.136	0.976	8.48	8.28
Between 20-25 years	9	96.8	29.8	0.457	20.897	9.55
	10	109.6	37.44	0.232	46.552	10.80
	11	99	32.6	0.306	33.824	10.35
	12	97.2	29.12	0.223	29.372	6.55
Between 25-40 years	13	301.4	112.64	0.2	66.75	13.35
	14	411.4	254.58	0.339	1180.678	400.25
	15	286	108.52	0.28	45.714	12.80
	16	385.2	164.64	0.593	47.808	28.35
Over 50 years	17	776.8	397.76	0.835	376.048	314.00
	18	591.8	261.54	0.712	73.244	52.15
	19	564.4	256.68	0.865	75.665	65.45
	20	454	190.88	0.529	30.057	15.90
	21	291.6	112.6	0.389	26.093	10.15
	22	377.8	228.9	0.323	25.232	8.15
	23	282.6	117.88	0.527	19.26	10.15
	24	87	12.88	0.281	20.462	5.75



Appendix 3.1: Relationships between plant Cu concentration and extractable Cu (mg/kg).



Appendix 3.2: Relationships between plant Cu uptake (µg) and extractable Cu (mg/kg).



Appendix 3.3: Relationships between plant Cu uptake (µg) and total Cu (mg/kg).