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The effect of long-term irrigation using wastewater on heavy metal contents of soils under vegetables in Harare, Zimbabwe

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Abstract

The magnitude of contamination, regulatory compliance and annual loadings of soils with copper (Cu), zinc (Zn), cadmium (Cd), nickel (Ni), chromium (Cr) and lead (Pb) were determined at three sites in Harare where wastewater was used to irrigate vegetable gardens for at least 10 years. Heavy metal total concentrations (mg kg⁻¹) in sandy and sandy–clay soils of pH 5.1–8.1 from all sites ranged from 7.0 to 145 for Cu, 14 to 228 for Zn, 0.5 to 3.4 for Cd, <0.01 to 21 for Ni, 33 to 225 for Cr and 4 to 59 for Pb in the 0–20 cm soil depths. The concentrations had increased significantly in the gardens compared with control soils and subsoil. Annual heavy metal loading rates showed that within 5–60 years, all studied heavy metals would have exceeded their permitted limits in soils, depending on site. It was concluded that the use of wastewater in urban horticulture enriched soils with heavy metals to concentrations that may pose potential environmental and health risks in the long-term.

Keywords: Heavy metals; Soil; Contamination; Wastewater; Irrigation

1. Introduction

Horticultural production has contributed to rapid economic growth in Zimbabwe, and demand for horticultural products has continued to increase with increasing population (Jackson, 1997). This has resulted in more horticultural enterprises, one of which is vegetable production at municipal farms and along riverbanks in Harare using wastewater for irrigation. Wastewater use occurs either indirectly, when partially and untreated effluent is discharged into rivers that supply water for agriculture, or directly, at municipal farms when partially treated sewage effluent is conveyed into some gardens.

Past experience had shown that these developmental projects, created with the aim of producing socio-economic benefits, have also produced adverse environmental impacts (FAO, 2000) such as land degradation. Earlier studies (Zaranyika et al., 1993; Mangwayana, 1995; Oloya and Tagwira, 1996;...
Nyamangara and Mzezewa, 1999) showed that the concentrations of heavy metals in wastewater that is used for irrigation at Pension, Crowborough and Mukuvisi vegetable production sites in Harare were several-fold (3 to <13-fold) higher than the recommended limits in wastewater in Zimbabwe. The studies also implicated land disposal of wastewater as the chief source of Cu, Zn, Cd and Pb enrichment of pasturelands. However, it has not been clear whether vegetable production sites irrigated with wastewater have also been enriched with heavy metals in the same magnitude or not. Although no cases of heavy metal poisoning due to the ingestion of vegetables irrigated with wastewater have been reported in humans in Harare, heavy metals remain important cumulative poisons (Kitagishi and Yamane, 1981).

Soils, as filters of toxic chemicals, may adsorb and retain heavy metals from wastewater. But when the capacity of soils to retain toxic metals is reduced due to continuous loading of pollutants or changes in pH, soils can release heavy metals into groundwater or soil solution available for plant uptake. The amount of heavy metals mobilized in a soil environment is a function of pH, clay content, organic matter content, cation exchange capacity and other soil properties making each soil unique in terms of pollution management (Kimberly and William, 1999). With the exception of Mo, Se and As, heavy metal mobility decreases with increasing soil pH due to precipitation of hydroxides, carbonates or formation of insoluble organic complexes (Smith, 1996). Heavy metals are capable of forming insoluble complex compounds with soil organic matter and according to Sauve et al. (2000) solid-solution partitioning of Cd, Cu, Ni, Pb and Zn is dependent on soil solution pH, total metal content and soil organic matter.

Heavy metals contribute to environmental pollution because of their unique properties, mainly that they are non-biodegradable, non-thermo-degradable and generally do not leach from the topsoil. Unlike petroleum hydrocarbons and litter that visibly build up on soils, heavy metals can accumulate unnoticed to toxic concentrations (Bohn et al., 1985) that affect plant and animal life. The duration of contamination by heavy metals may be for hundreds or thousands of years, even after their addition to soils had been stopped. The time taken for Cd, Cu and Pb to reach half their concentrations (half lives) in soil were found to be 15–1100, 310–1500 and 740–5900 years, respectively, depending on soil type and physicochemical parameters (Alloway and Ayres, 1993).

Metals added in small concentrations find specific adsorption sites in soil where they are retained very strongly, either on inorganic or organic colloids (Sauve et al., 2000). Following addition to soil, organic loading of wastewater undergoes decomposition to CO₂, low molecular weight soluble organic acids, residual organic matter and inorganic constituents (Boyd et al., 1980). Decomposition can also release heavy metals into soil solution. But, because of their low solubility and limited uptake by plants, heavy metals tend to accumulate in surface soil and become part of the soil matrix (McGrath et al., 1994). With repeated wastewater applications, heavy metals can accumulate in soil to toxic concentrations for plant growth (Chang et al., 1992).

Not all heavy metals in soil are results of human activity. Trace metals in soil originally arose from the net effects of geological and soil-forming processes of the elements (Kabata-Pendias and Adriano, 1995) and the concentration in soil is governed by the parent material, climate, topography and human activities, factors which are responsible for soil formation. Sandy soils from granite rocks normally contain lower concentrations of heavy metals than clay soils derived from mafic rocks (Ross, 1994a). According to Alloway and Ayres (1993) heavy metals may enter the soil from agricultural related sources such as pesticides, fertilizers, composts and manure, and sewage sludge.

There are currently no locally derived permissible limits of heavy metals in soils amended with wastewater in Zimbabwe. However, there are limits for heavy metal concentrations in wastewater applied on agricultural land, which are presented in the ‘Zimbabwean Waste Discharge and Disposal (Water Pollution) Regulations’ of 1998. These limits were derived primarily from international sources of information like the United States Environmental Protection Agency (USEPA), World Health Organization (WHO) and the European Union (EU), with special considerations to local water resources and utilization in Zimbabwe (Mtetwa, 1996). Under the Zimbabwean Public Health Protection Act (1972),
the use of wastewaters or sludge is prohibited for irrigation of vegetables, and crops to be eaten raw and berry fruits in order to protect consumers from exposure to pathogens and toxic chemicals found in some wastewaters. However, the history of urban cultivation in Harare shows that institutional responses to wastewater use have not been very prohibitive (Mbiba, 1994).

Within each country in the EU, the concentrations of heavy metal contaminants are controlled under an EC Directive (86/278) on the protection of the environment and in particular, of the soil, when wastewater and sewage sludge are used in agriculture. The UK limits for heavy metals in agricultural soils (MAFF, 1993) are based on the EC Directive, which insists that soils should be monitored before and after wastewater or sludge is applied. They are more adaptable to Zimbabwean environment than the stricter Dutch policy, which states that all soils should not be contaminated and heavy metal concentrations in effluent should be very low (Mtewa, 1996). Furthermore, UK maximum permissible limits were set in relation to soil pH, accounting for the varying availability of heavy metals for crop uptake with soil pH. In this study, the limits of heavy metals in soils and their annual loading rate limits refer to the UK permissible limits.

The objectives of the paper were to determine the total concentrations of Cu, Zn, Cd, Ni, Pb and Cr, and estimate their annual loading rates in soils at the Mukunsvi, Pension and Crowborough vegetable production sites where wastewater has been applied for at least 10 years. This would provide knowledge that guides future research into the protection of the environment and people from exposure to heavy metals with potential to cause health problems. Although total concentrations of heavy metals in soil poorly indicate their availability for plant uptake (Kimberly and William, 1999), existing permissible limits of heavy metals in soils are based on total concentrations. Thus, the information would be useful from a policy point of view. In addition, the total concentration also indicates the potential risks from other contamination pathways such as soil ingestion by children (and some adults, commonly pregnant women), inhalation of dust from the sites, soil adsorption on edible leaves and other potential risks associated with handling the soil.

2. Materials and methods

2.1. Site description and management practices

Three vegetable production sites, Pension, Crowborough and Mukunsvi, were selected from Harare (Fig. 1) where many pollution problems as well as high commercial activities related to horticulture are found. Harare has cold–dry winters and hot–wet summers (subtropical). Average annual rainfall is about 850 mm and average annual temperature is 18–20 °C. Crowborough Farm extends from 17°49’S to 17°52’S and lies between 30°52’E and 30°58’E, while Pension Farm extends from 17°52’S to 17°55’S and lies between 30°52’E and 30°55’E. The vegetable gardens at the Crowborough and Pension Farms are located near Crowborough and Firle Sewage Treatment Works, respectively, and they receive treated effluent (sometimes mixed with anaerobically digested sludge) for irrigation. The vegetable gardens at the Mukunsvi site extends from 30°58’E to 31°03’E, and lies between 17°54’S and 17°57’S, and are irrigated with the Mukunsvi river water contaminated by domestic and industrial effluent from Harare.

Vegetable production by farm workers at Crowborough and Pension sites started in 1975, but some new gardens at the Pension site started operating in 1996. Over 50 households from Pension and over 35 households from Crowborough cultivate the gardens, each with an approximate area ranging from 400 to 3500 m². Vegetable production along the Mukunsvi river started in the late 1980s and is practiced by households from the surrounding high-density areas of Glen Norah and Highfields, on areas ranging from a few square meters to over a hectare. The crops commonly grown at Crowborough, Pension and Mukunsvi sites include maize (Zea mays), leafy vegetables (mostly Brassica spp.), tomatoes (Lycopersicon esculentum) and onions (Allium cepa). Leafy vegetables are produced all year round, and the types commonly and extensively grown are Brassica juncea, B. napus (covo and rape) and B. oleracea (viscose). The crops are produced for home consumption and for sale to vendors from residential areas of Glenview, Glen Norah and Highfields.

Vegetables grown at the Pension and Crowborough sites are flood and furrow irrigated, respectively, during the dry season (February–October). The
Gardens at these sites are found in clusters on gently undulating terrains of about 2–5% slope. Many of the gardens at the Mukuvisi site are irrigated with buckets (20 L capacity), while a few are irrigated with hosepipes connected to petrol pumps drawing the water from the river. The gardens are scattered along the riverbanks on gently undulating to undulating terrains of up to about 8% slope. The river flows through industrial and residential areas of Harare receiving waste discharges at various points along its length. Increases in industrial and residential structures have increased pressure on the sewer systems leading to incessant sewage pipe blockages and bursts in some areas and the subsequent discharge of unknown toxic chemicals and sewage water into the river (Zaranyika, 1996).

The volumes of river water applied at Mukuvisi site, estimated from the number of 20 L buckets applied per unit area and frequency of irrigation (two to three times per week), ranged from 6 to 29 ML ha\(^{-1}\) year\(^{-1}\) (average, 16 ML ha\(^{-1}\) year\(^{-1}\)). The volumes of wastewater applied at the Pension site, estimated from garden area, depth of application and frequency of irrigation (twice a week) ranged from 24 to 36 ML ha\(^{-1}\) year\(^{-1}\) (average, 30 ML ha\(^{-1}\) year\(^{-1}\)). Quantity of wastewater applied at Crowborough site was not captured, but Hranova (2002) indicated that on average the active farmland receives >20 ML ha\(^{-1}\) year\(^{-1}\). The average concentrations of heavy metals in the Mukuvisi river (Table 2) covering the dry season (May–July) were obtained from a monitoring study by Zaranyika et al. (1993), while the average concentrations heavy metals in treated sewage effluent (Table 2) were obtained from a 4-year study on final effluent quality at Firle Sewage Treatment Works (Madyiwa et al., 2002). The volumes of wastewater applied and the concentrations of heavy metals in the wastewater were used to estimate the annual heavy metal loading rates.
2.2. Soil sampling

Soil samples were collected from wastewater-irrigated gardens as well as from the non-irrigated (control) sites at Pension, Crowborough and Mukuvisi sites (Fig. 2). A composite sample, made up of five sub-samples collected using a Dutch auger along zigzag paths (Zigzag sampling) to achieve randomness from each garden, was obtained from each garden. The samples were collected at 0–20 and 30–50 cm depths at the Pension and Crowborough sites, and at 0–10 and 20–30 cm depths at the Mukuvisi sites. The 21–29 cm depths at the Pension and Crowborough sites and the 11–19 cm depths at the Mukuvisi sites were discarded as boundaries between the topsoil and subsoil. Shallower depths were sampled at the Mukuvisi sites because the effective soil depths were only up to about 35 cm from the surface. A total of 210 samples were collected, 96 from the Pension site, 76 from the Crowborough site and 38 from the Mukuvisi site. These samples were collected from each of the wastewater-irrigated gardens and from the non-irrigated (control) sites numbered in Fig. 2. Paddocks that received heavy application of a mixture of effluent and sludge for more than two decades surrounded the gardens at Crowborough site and therefore, no suitable control site was found in the vicinity.

Selected properties of the studied soils are shown in Table 1. The underlying geology at Pension Farm and area surrounding the Mukuvisi river is coarse-grained granite, while the geology at Crowborough Farm is dolerite and meta-arenite (Baldock, 1991). The textures of the studied soils at Pension, Crowborough and Mukuvisi sites range from the sands to the sandy loams (Arenosols). The concentrations of heavy metals in the topsoil and in the subsoil were used to derive heavy metal enrichment indices, given as concentration of a metal in topsoil divided by concentration of that metal in the corresponding subsoil (Ross, 1994b).

2.3. Soil analysis

Soil pH from all production sites was measured with a pH meter using the water method (McNeal, 1982). Soil texture was determined using the hydrometer method (Gee and Bauder, 1986). Cations were determined using atomic (emission for Na and K) absorption spectrophotometry after extraction using ammonium acetate. Cation exchange capacity was determined by saturating the soil 1 M ammonium acetate buffered at 5.2. Organic C was determined by the modified Walkley Black method (Houba et al., 1989) with additional heat applied under reflux. Total N was determined using the Kjeldahl procedure (Bremner and Mulvaney, 1982).

Soil samples were digested for heavy metal analysis using the aqua regia digest method (Baker and Amacher, 1982). One gram soil for each sample, in duplicates, was transferred into a 100 mL digestion flask to which 10 mL of aqua regia (a mixture of concentrated HCl and concentrated HNO₃ in the HCl:HNO₃ ratio of 3:1) was added before covering the digestion flask with a watch glass and allowing the mixture to react overnight (for at least 12 h). The next day, the mixture was heated progressively and boiled under reflux for 2 h after which the digestion flask was cooled. The cooling column was rinsed with 15 mL of distilled water recovering rinse water in the digestion flask. The mixture was separated using a centrifuge at 1500 rpm for 5 min after which a supernatant solution was collected into a 50 mL volumetric flask before diluting to the mark with hot 2 M HNO₃. The soil extract was analysed for Cu, Zn, Cr, Cd, Ni and Pb using an atomic absorption spectrophotometer (model: Philips AA-10). Standard solutions were prepared in the concentration ranges of 0–5 mg L⁻¹ for Cu, Zn, Ni, Cr and Pb, and 0–1 mg L⁻¹ for Cd. A blank determination was also carried out. Data were subjected to analysis of variance using Genstat statistical package to compare the topsoil and subsoil.

3. Results

3.1. Annual loading rates

The estimated annual heavy metal loading rates at Pension and Mukuvisi sites (Table 2) showed that all selected heavy metals, except Cd, had annual loading rates below the maximum permissible limits at the Mukuvisi site. On the contrary, the annual loading rates of all selected heavy metals, except Zn, were above their maximum permissible limits at the
Fig. 2. Soil sampling sites: the wastewater-irrigated gardens were numbered 1–46 for Pension, 1–38 for Crowborough and 1–17 for Mukuvisi, while C1 and C2 were approximate sites where control soils were collected. The Mukuvisi river is disjoined by a slash to separate the two sites that were approximately 5 km apart along the river. No controls were suitable for Crowborough site.
Pension site. Cadmium loading rate had the greatest magnitude of deviation from the maximum permissible limit (0.15 kg ha\(^{-1}\) year\(^{-1}\)), being over 1000-fold higher. No data were found on the monitoring of Zn levels in wastewater at the Pension site, and other heavy metals at the Crowborough site.

3.2. Soil pH

The results of the soil pH are given in Fig. 3. Topsoil pH at Pension site ranged from 5.1 to 6.3 while the mean pH in control soils was 5.3. The topsoil pH was not significantly different (\(P > 0.05\)) from subsoil pH (5.0–6.6). Topsoil pH at Crowborough site ranged from 5.4 to 7.0 and was significantly (\(P < 0.05\)) lower than subsoil pH (6.1–7.0). No suitable control sites were found at Crowborough. The soil pH at Mukuvisi sites ranged from 5.1 to 8.2, while mean pH in control soils was 5.5. Topsoil pH was not significantly different (\(P > 0.05\)) from subsoil pH (5.3–8.2) at Mukuvisi sites. Significant differences (\(P < 0.05\)) in soil pH were found among the sites and the order was: Mukuvisi > Crowborough > Pension for both topsoil and subsoil.

3.3. Copper

Topsoil Cu at the Pension site ranged from 22 to 94 mg kg\(^{-1}\) while control soils had less than 10 mg Cu kg\(^{-1}\) (Fig. 4). About 9% of gardens from Pension had total Cu above the maximum permitted limits (50 mg kg\(^{-1}\) at pH < 5.5; 100 mg kg\(^{-1}\) at 5.5 ≤ pH < 6.0) and topsoil Cu was significantly (\(P < 0.05\)) higher than subsoil Cu (3–40 mg kg\(^{-1}\)). The Cu concentrations in the topsoil at Crowborough site ranged from 21 to 145 mg kg\(^{-1}\) and about 16% of the gardens had total Cu above their maximum limits.

<table>
<thead>
<tr>
<th>Metal</th>
<th>Site</th>
<th>Limits</th>
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<tbody>
<tr>
<td></td>
<td>Pension</td>
<td>Wastewater irrigated</td>
</tr>
<tr>
<td>Sewage effluent</td>
<td>ALR River water</td>
<td>ALR Wastewater</td>
</tr>
<tr>
<td>Cu</td>
<td>0.66</td>
<td>20  0.15</td>
</tr>
<tr>
<td>Zn</td>
<td>–</td>
<td>–   0.42</td>
</tr>
<tr>
<td>Cd</td>
<td>6.6</td>
<td>195 0.41</td>
</tr>
<tr>
<td>Ni</td>
<td>3.37</td>
<td>99   0.14</td>
</tr>
<tr>
<td>Cr</td>
<td>6.62</td>
<td>195 0.14</td>
</tr>
<tr>
<td>Pb</td>
<td>2.72</td>
<td>80   0.31</td>
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(11–34 mg kg\(^{-1}\)). Significant differences \((P < 0.05)\) in Cu concentrations were found among sites and the concentrations were in the order: Crowborough > Pension > Mukuvisi for the topsoil, and Mukuvisi ≈ Crowborough > Pension for the subsoil.

### 3.4. Zinc

Topsoil Zn at the Pension site ranged from 60 to 228 mg kg\(^{-1}\) while control soils had less than 14 mg Zn kg\(^{-1}\) (Fig. 5). About 4\% of gardens from Pension had Zn concentrations above the permitted limit (200 mg kg\(^{-1}\) at pH < 5.5) and topsoil Zn was significantly \((P < 0.05)\) higher than subsoil Zn (14–72 mg kg\(^{-1}\)). Zinc concentrations in the topsoil at the Crowborough site ranged from 42 to 131 mg kg\(^{-1}\), and no garden had total Zn concentration above the maximum permitted limit. Topsoil Zn concentrations were significantly \((P < 0.05)\) higher than in the subsoil (9–58 mg kg\(^{-1}\)). Total Zn concentrations in the topsoil at the Mukuvisi sites ranged from 14 to 58 mg kg\(^{-1}\) while control...
soils had not more than 11 mg Zn kg\(^{-1}\), but no garden was above the permitted limits for Zn. Subsoil Zn ranged from 16 to 54 mg kg\(^{-1}\) and was not significantly different \((P > 0.05)\) from the topsoil Zn. Significant differences \((P < 0.05)\) in topsoil Zn concentrations were found among the sites and the order was: Pension > Crowborough > Mukuvisi. However, no significant differences \((P > 0.05)\) in subsoil Zn concentrations were found among the sites.

3.5. Cadmium

Cadmium concentrations in the topsoil at the Pension site (Fig. 6) ranged from 2.0 to 3.4 mg kg\(^{-1}\) and the average concentration in control soils was 2.0 mg Cd kg\(^{-1}\). About 13% of the gardens from Pension had their concentration above the limit of 3 mg Cd kg\(^{-1}\). Topsoil Cd was significantly higher \((P < 0.05)\) than subsoil Cd (1.8–2.9 mg kg\(^{-1}\)). The concentrations of Cd in the topsoil at the Crowborough
site ranged from 1.0 to 2.3 mg kg\(^{-1}\) and no garden was above permitted limit (3 mg kg\(^{-1}\)). Topsoil Cd concentrations were significantly higher \((P < 0.05)\) than subsoil Cd (0.9–1.5 mg kg\(^{-1}\)) at Crowborough site. Total Cd at Mukusi ranged from 0.5 to 2.5 mg kg\(^{-1}\) while control soils had not more than 0.5 mg Cd kg\(^{-1}\). No garden had total Cd above the permissible limit at the Mukusi site and the topsoil Cd concentrations were not significantly different \((P > 0.05)\) from subsoil Cd (0.5–1.5 mg kg\(^{-1}\)). Significant differences \((P < 0.05)\) in Cd concentrations were found among the sites and the order was: Crowborough > Pension > Mukusi for both topsoil and subsoil.

3.6. Nickel

No garden at all the study sites had total Ni above the permitted limits (50 mg kg\(^{-1}\) at pH < 5.5) (Fig. 7). Nickel concentrations in the topsoil of the wastewater-irrigated gardens at the Pension site ranged from 9 to 19 mg kg\(^{-1}\) while control soil had less than 3.2 mg Ni kg\(^{-1}\). Topsoil Ni was significantly higher \((P < 0.05)\) than subsoil Ni (4–17 mg kg\(^{-1}\)). Concentrations of Ni at the Crowborough site ranged from 9 to 21 mg kg\(^{-1}\) and subsoil Ni (7–16 mg kg\(^{-1}\)) was significantly less \((P < 0.05)\) than the topsoil Ni. Total Ni concentrations in the topsoil at the Mukusi site ranged from 0 to 14 mg kg\(^{-1}\) and not more than 1 mg kg\(^{-1}\) in the non-irrigated (control) soil. There were no significant differences \((P > 0.05)\) in Ni concentrations between the topsoil and subsoil (0–8.4 mg kg\(^{-1}\)) at the Mukusi site. Significant differences in Ni concentrations were found among the sites and the order was: Crowborough > Pension > Mukusi for both topsoil and subsoil.

3.7. Chromium

No Cr concentration was above the maximum permitted limit of 400 mg kg\(^{-1}\) (Fig. 8). Total Cr concentrations in the wastewater-irrigated soils from the gardens at the Pension site ranged from 33 to 225 mg kg\(^{-1}\) while the non-irrigated soil had 3.0 mg Cr kg\(^{-1}\). Topsoil Cr was significantly higher \((P < 0.05)\) than subsoil Cr (8–34 mg kg\(^{-1}\)). Chromium concentrations in the wastewater-irrigated gardens at the Crowborough site ranged from 48 to 100 mg kg\(^{-1}\) and the topsoil Cr was significantly \((P < 0.05)\) higher than subsoil Cr (21–47 mg kg\(^{-1}\)). Chromium concentrations in the gardens at the Mukusi site ranged from 33 to 76 mg kg\(^{-1}\) and the non-irrigated (control) soils had 54 mg Cr kg\(^{-1}\). There were no significant differences \((P > 0.05)\) in Cr concentrations between the topsoil and subsoil (49–79 mg kg\(^{-1}\)) at the Mukusi site. Significant differences were found in Cr concentrations...
among the sites and the order was: Pension > Mukuvisi ≈ Crowborough for the topsoil and Mukuvisi > Crowborough > Pension for the subsoil.

3.8. Lead

Topsoil Pb concentrations in the wastewater-irrigated soils from all the study sites (Fig. 9) were below the permitted limit (300 mg kg\(^{-1}\)). Lead concentrations in the topsoil of wastewater irrigated gardens ranged from 22 to 41 mg kg\(^{-1}\) at the Pension site, while non-irrigated (control) sites had 18 mg Pb kg\(^{-1}\). Topsoil Pb concentration at the Pension site was significantly higher (\(P < 0.05\)) than subsoil Pb (17–38 mg kg\(^{-1}\)). Topsoil Pb concentrations at the Crowborough site ranged from 17 to 59 mg kg\(^{-1}\) and was significantly higher than the subsoil Pb (4.1–38 mg kg\(^{-1}\)). The concentrations of Pb in the topsoil at the Mukuvisi site ranged from 4 to 38 mg kg\(^{-1}\), while
the mean Pb concentration in the non-irrigated soil was 9.2 mg kg\(^{-1}\). The topsoil Pb concentration was not significantly \((P > 0.05)\) different from the subsoil Pb (3–28 mg kg\(^{-1}\)) at the Mukvisi site. Significant differences in Pb concentrations were found among the sites and the order was: Crowborough > Pension > Mukvisi for the topsoil and Crowborough ≈ Pension > Mukvisi for the subsoil.

3.9. Relative topsoil metal enrichment

The topsoil heavy metal enrichment indices for all sites were grouped into percentages of gardens with a given enrichment index ranging from <2 to >20 (Fig. 10). Control soils (corresponding bars below) are also shown. The relative enrichment indices for Cr, Zn and Cu in Pension and Crowborough gardens indicated moderate to extremely strong enrichment for these heavy metals. Relative enrichment indices for Ni, Cd and Pb indicated slight to moderate enrichment. The topsoil enrichment indices for control soils indicated that control soils were not enriched by all studied heavy metals. The relative topsoil enrichment indices for Mukvisi garden soils indicated slight to strong enrichment for all selected heavy metals but no garden were either very strongly or extremely enriched. In the control soils there was no evidence of significant enrichment with for studied heavy metals except Pb in which 50% of control soils that showed some moderate enrichment.

4. Discussion

The application of wastewater at the Pension and Mukvisi sites increased soil pH by 0.5–3 units comparing the wastewater-irrigated sites to the non-irrigated soils. Past research (Zaranyika et al., 1993; Oloya and Tagwira, 1996) has indicated that the wastewaters applied for irrigation at the Pension, Crowborough and Mukvisi sites have in most cases neutral to alkaline pH (6.5–8.0) in addition to the high concentrations of basic cations such as Ca, Mg and K. Oloya and Tagwira (1996) also found out that the pH of wastewater-irrigated soils in Harare ranged from 6.2 to 8.0, while the highest pH in the virgin soil was 6.4.

The solid-solution partitioning of Cd, Cu, Ni, Pb and Zn is dependent on soil solution pH and total metal content (Brallier et al., 1996; Sauve et al., 2000) among other soil factors. Heavy metals are generally less mobile at pH > 7 than at pH < 5.5. Thus, the risk of heavy metal uptake by plants would be highest at the wastewater-irrigated gardens from the Pension site where soil pH was lowest. Heavy metal absorption by plants generally increases with decreasing pH, due to dissolution of metal–carbonate complexes, releasing free metal ions into solution (Connell and Miller, 1984).

The concentrations of the studied heavy metals in the wastewater-irrigated soils were significantly above
the concentration found in the non-irrigated (control) soil indicating that the application of wastewater had enriched the soils with heavy metals. Exceptions were found at the Mukuvisi site where the Cr concentrations were significantly higher in the non-irrigated soil than in the wastewater-irrigated soils, although overall the concentrations were generally low at the site. Higher concentrations of Cr (47–60 mg kg\(^{-1}\)) in control soils relative to the wastewater-irrigated gardens (33–76 mg Cr kg\(^{-1}\)) at Mukuvisi site could be attributed to some unknown historical background of the site since granitic parent material would normally contribute little amounts of Cr in soil. The concentration of Cr in rocks varies from an average of 5 mg kg\(^{-1}\) (range of 2–60 mg kg\(^{-1}\)) in granitic rocks to an average of 1800 mg kg\(^{-1}\) (range of 1100–3400 mg kg\(^{-1}\)) in ultrabasic and serpentine rocks (NAS, 1974; Ross, 1994a). Thus, chemical weathering could contribute trace amounts of Cr in soils at the Mukuvisi site since they are derived from granite.

The distribution of selected heavy metals in gardens at each site was mainly affected by the ages and location of the gardens (notably in Pension gardens). Older gardens (>10 years old), located upslope (closer to the Compound; Fig. 2a) had the highest level of contamination because of a longer enrichment time than downslope gardens (last quarter of the garden cluster in the direction of the slope) where younger gardens (5–10 years old) were located. Similar studies by Nakshabandi et al. (1997) on heavy metal accumulation in soils irrigated with treated effluent indicated that enrichment of soils with heavy metals increased with time, although they found normal concentrations of heavy metals in plant tissues. The increase in concentrations of heavy metals with time implies that heavy metals persist in soil, and in the long-term, productivity of soils from all sites that use wastewater in Harare may go down significantly, because of heavy metal toxicity, despite the fact that soils are nutrient-rich.

The differences in the level of contamination of soils at the study sites were mainly attributed to the period when wastewater was used. Pension and Crowborough sites with over two decades of irrigation with wastewater had higher concentrations of heavy metals than the Mukuvisi site with a period ranging from 5 to 15 years. Some heavy metals, notably Cu, Zn and Cd, at the Pension and Crowborough sites had already started exceeding the maximum permissible concentrations above which substantial amounts of heavy metals may be taken up by plants or may cause associated environmental problems.

The level of topsoil contamination with Cu, Pb and Ni was in the order: Crowborough > Pension > Mukuvisi. The wastewater applied at the Crowborough site is treated effluent (sometimes mixed with digested sludge) from the Crowborough Sewage Treatment Works (CSTW). Similarly the Pension site receives the sewage effluent and sludge mixture from the Firle Sewage Treatment Works (FSTW). Both CSTW and FSTW receive domestic and industrial effluent mixtures. Higher Cu, Pb and Ni concentrations at the Crowborough site than at the Pension site indicated that the industries that supply the CSTW produce higher Cu, Pb and Ni loaded effluent than those that supplied FSTW. The wastewater flow received at CSTW comes from about 690 companies located on the western industrial side of Harare that discharge their effluent directly into the sewage system (Steneva, 1996). High concentrations of Cu, Pb and Ni in effluent may come from electroplating and battery industries located west of Harare.

Zinc, Cd and Cr concentrations at the study sites were generally in the order: Pension > Crowborough > Mukuvisi. The gardens at the Pension sites are flood irrigated while those at the Crowborough site are furrow irrigated. It is possible that the Pension site receives more irrigation of wastewater than the Crowborough site since less, if any, run-off would be expected from the flood irrigated gardens than the furrow irrigated gardens. This is because the gardens at Crowborough, unlike at Pension, do not have ridges at the boundaries to control run-off and the wastewater flows from the gardens into the nearby Marimba river.

The annual heavy metal loading rates in soils at Mukuvisi and Pension sites indicated that the biggest problem was of high Cd loading rates. The volumes of wastewater that could be applied without exceeding the permissible annual loading rate limit (0.15 kg Cd ha\(^{-1}\) year\(^{-1}\)) is 0.4 ML ha\(^{-1}\) year\(^{-1}\) at Mukuvisi, which is only about 2.5% of the estimated mean volume of wastewater that is being applied (16 ML ha\(^{-1}\) year\(^{-1}\)). For the Pension site, only 0.02 ML ha\(^{-1}\) year\(^{-1}\) may be used without exceeding...
the same limit, which is less than 1% of the estimated average volume of wastewater that is being applied (30 ML ha\(^{-1}\) year\(^{-1}\)) at Pension site.

5. Conclusions

Given the prevailing conditions of generally low pH, light textured soils and significantly high concentrations of Cu, Zn, Cd, Cr, Pb and Ni compared with control sites, it was concluded that soil contamination by wastewater use present long-term environmental and health risks. Some heavy metals, notably Cu, Zn and Cd, have begun to exceed their maximum permitted limits, especially at Pension and Crowborough gardens, and had high annual loading rates. Contamination at Mukuvisi gardens was generally lower but potential long-term problems from Cd were noted. Distribution and concentrations of studied heavy metals and pH of soils indicated higher contamination in older gardens, gardens irrigated with treated effluent mixed with sludge and gardens irrigated with larger volumes of wastewater.

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