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Assessment of Chromium in orchard soil and fruit irrigated with treated chromite mine effluent and potential dietary health implications

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Abstract: *Contamination of agricultural soil by trace elements poses threats to food safety, soil quality and human health. The concentration of chromium (Cr) for soil and tissues of two plant species sampled from chromite mine effluent-irrigated and rain-fed soils was determined using ICP-OES. The concentration of Cr in treated effluent (mean: 26.10±2.48mg/l) was higher than the irrigation criteria for irrigation water quality. The contamination factor (0.304) and the index of geoaccumulation (-1.595) showed no pollution of irrigated soil. The relatively elevated soil Cr concentration in effluent-irrigated soil was within acceptable soil quality values. The observed Cr concentrations trend for both Mangifera indica and Musa paradisiaca was soil>roots>leaves>fruits. Chromium transference factors suggested poor accumulation and very poor translocation to the above-ground parts. Estimated daily metal intake and health risk index indicated that dietary exposure to Cr by consuming these fruits at the study intake rates was unlikely to constitute a health hazard.*

Keywords: *bioaccumulation, Chromium, contamination index, dietary exposure, health risk, mine effluent*

I. Introduction

Trace elements are found in low concentrations in soil [1, 2] but they may be elevated by natural processes and anthropogenic activities [3]. Chromite mining and ferrochromate production release Cr into the environment [4]. The contamination of soil and water in chromite mining areas is a widespread and serious problem [5]. Mine effluents may contain trace elements in varying proportions depending upon mining processes and the extent of wastewater treatment. Wastewater reuse for irrigation may have adverse effects on soil quality and food safety due to trace element toxicity [6]. Agricultural plants represent an important pathway for the movement of potentially toxic trace elements from soil to humans [7, 8]. Ecological and global public health concerns associated with soil contamination by trace elements have been raised [9]. Trace element enrichment in soil through irrigation water, subsequent uptake and accumulation by plant tissues has been reported [10-12]. Therefore, care must be taken when considering industrial wastewater reuse for crop irrigation or disposal.

Chromium is a redox active element that exists in the environment in stable forms mainly as Cr(III) or Cr(VI) [13, 14]. The concentration of Cr in various environmental matrices is influenced by its chemical speciation through redox transformations mainly between Cr(III) and Cr(VI) species [15]. Chromium (III) is an essential nutrient which exerts biochemical and physiological functions in humans but Cr(VI) is carcinogenic and affects the respiratory and digestive tracts [13, 16]. Chromium (VI) has been reported to accumulate mainly in plant roots where it is reduced to Cr(III) in the vacuole and poorly translocate to above-ground plant parts [1]. However, concentrations of Cr, a non-essential element to plants, have been recorded in above-ground tissues of particular plant species [17, 10-12]. Since plants can take up Cr to above-ground parts, the consumption of Cr-contaminated edible plant tissue may expose consumers to health risk. The potential dietary exposure of consumers to toxic trace elements should therefore be evaluated.

There seems to be limited reports on the potential uptake, accumulation and translocation of Cr by *Musa paradisiaca* (*M. paradisiaca*) and *Mangifera indica (M. indica)* growing on soil irrigated with chromite mine effluent. Available studies seem to have been focused on the concentration of Cr in fruits of these plants bought from markets [18–20]. In the present study we determined the concentration of Cr in irrigation effluent, irrigated soil and in tissues of *M. paradisiaca* and *M. indica*. We then evaluated the potential health risk posed by the consumption of fruits from these plants using the Daily Metal Intake (DMI) and the Health Risk Index (HRI) for Cr. No attempt was made to assess the speciation of Cr in soil or plants.

II. Materials and Methods

2.1 Description of the study area

Chromite is mined in ultramafic-mafic igneous rocks with deposits at Selukwe and along Great Dyke in Zimbabwe [21]. The study area (18055´S; 29049´E at 1197m asl) has a semi humid tropical climate which is characterised by low rainfall (500-750mm/year) which is spread from November to March. The average annual temperature is 19°C. Chromite mining and ferrochrome production release process water which is treated and used to irrigate *M. indica* and *M. paradisiaca* twice weekly in an orchard as a disposal option. The study was a randomised block design where irrigation was the blocking factor with plant species and soil depth as treatments. Soil depth had two levels (0-10 and 10-30cm) and plant species had three levels (root, leaf and fruit). The concentration of Cr was the independent variable. The rain-fed soils were considered a reference for the study.

2.2 Sampling and sample analysis

Three grab effluent samples (100ml) were collected at the point of discharge in the orchard per day (8a.m, 12 Noon and 4p.m) to give a time-integrated composite effluent sample. This was repeated for two random days per week for five consecutive weeks. Samples were filtered (0.45mm), preserved in HNO_3 (2ml) and concentrated to 10-fold by evaporation on a water bath. The concentrate was digested in *aqua regia* and diluted to 50 for analysis of total Cr. Five plants of each species were randomly selected from the effluent-irrigated orchard soil and two plants for each species from rain-fed soils for the sampling of soil and plant tissue in February 2015. Three grab soil samples (about 200g) around each plant within a circle (0.5m radius) were sampled at two depths (0-10 and 10-30cm) to give composite samples for each soil depth (0.6kg). Soil preparation was done following the procedures by [8]. Air-dried samples were used for the analysis of selected physicochemical soil properties (Table 1). Filtered and acid-digested (HNO_3/$HClO_4$/HF) split soil sample (0.5g) solutions were diluted to 100 before being analysed for Cr (Table 1). Field sampling of plant tissues was done as described by [22] and sample preparation by [23]. A new ultra-sharp carbon steel surgical blade (Swann-Morton, India) was used to get clean sharp cuts of leaves. A clean garden hoe was used to sample roots. Fresh fruits were harvested. A split sample (about 05g) of each oven-dried and finely ground plant tissue was acid digested (*aqua regia*), filtered and diluted to 100 and analysed for Cr (Table 1).

2.3 Data analysis

The index of geo-accumulation (I-geo) was determined following procedures by [24]. The concentration factor (CF) for Cr in soil was estimated following procedures by [25]. This was determined as a quotient of Cr concentrations of the irrigated soil and the geochemical average shale (90mg/kg) suggested by [26]. Chromium transference factors (concentration in plant tissue: soil; plant tissue: plant tissue) were determined using procedures as described by [8]. The average daily fruit intake of 80g/cap/day [27] from 10 sub Saharan African countries was used to determine the Daily Metal Intake. An African average mass of about 60kg [28] was used in the present study. Additionally, the oral reference doses (RfD) of 1.5 for Cr(111) and 0.003mg/kg.bw/d for Cr(V1) [29] were used to evaluate the Health Risk Index (HRI) as described by [30]. The mean concentration of Cr in irrigation mine effluent, soil and fruit were compared with international maximum values.

2.4 Statistical analysis

Normality and equality of variance-tested data were subjected to One-Way Analysis of Variance to determine any significant differences between means in the concentration of Cr. An independent samples t test was run to determine if there was any significant differences between (i) soil characteristics of effluent-irrigated and rain-fed soils, (ii) concentrations of Cr in similar plant tissues of different species and (iii) the sample mean concentration of Cr and the maximum allowable limits for irrigation, soil and fruit quality. A Pearson correlation test was performed to determine whether there was any association between the concentrations of Cr in (i) irrigation mine effluent and soil and (ii) soil and roots. All statistical analyses were performed at 95% level of confidence ($p<0.05$) unless specified. MS Excel and IBM SPSS version 21 were used throughout the analyses.

2.5 Quality control procedures

All apparatus used were thoroughly washed before use, and then rinsed thrice using deionised water. Glassware used throughout the study was acid-resistant pyrex glass (Electrical and Engineering Supplies, SA).

Table 1: Analytical procedures for the characterisation of soil samples for selected Physicochemical parameters and the determination of Cr in plant tissues

Parameter	Procedure and equipment used
Soil texture	Gravity: Hygrometer
Soil pH (H_2O)	Electrochemical: (Ecosan pH meter)
EC	Electrochemical: Conductivity meter (HJM Electronics C7)
Soil OM	Walkley–Black digestion
Soil water	Gravimetric analysis on drying at 105±5°C:
Cr (total)	Spectrometric: ICP-OES (ARCOS FHS16)

All equipment and analytical grade reagents were purchased from Electrical and Engineering, Supplies, SA)

Reagents used were analytical grade. Soiled, diseased and stressed plant tissue was not sampled [23]. To exclude contamination, plant samples were washed thoroughly by hand using deionised water. Samples were replicated five times. Reagents blanks were run. The analytical procedure was validated by using prepared standards.

III. Results And Discussion

3.1 Quality control for the analytical procedure

The procedural drift in the measurement of Cr concentration by analysis of replicate blanks after a batch of every ten samples was negligible. The limit of detection (LOD) was 0.003mg/L for soil (0.60mg/kg) and plant tissue (0.15mg/kg).

3.2 Physicochemical characteristics of soil

Table 2 Selected physicochemical properties of mine effluent-irrigated soil and rain-fed soil. Values are expressed as mean±SE of replicate measurements.

Parameter	Effluent-irrigated soil	Rain-fed soil
pH (H_2O)	7.51 ± 0.1^a	7.70 ± 0.94^a
Particle distribution (%)		
clay	20.7 ± 1.2^a	28.5 ± 0.8^a
silt	43.9 ± 0.6^a	27.3 ± 4.8^b
fine sand	35.4 ± 0.9^a	44.21 ± 0.3^a
Moisture content (%)	25.51 ± 2.59^a	7.8 ± 0.44^b
Organic Matter (%)	5.60 ± 0.20^a	2.03 ± 0.32^b
EC (µS/cm)	315.25 ± 10.25^a	89.63 ± 2.15^b

Superscripts a, b... same superscripts in the same row denote no significant differences while different superscripts denote significant differences at 95% confidence level (Independent samples *t* test).

Table 2 shows the variation of physicochemical properties of mine effluent-irrigated and rain-fed soils. There were no significant differences in the soil properties (pH, clay content) between the soils ($p>0.05$). These soil characteristics influence the availability of Cr in soil [7, 17]. Effluent-irrigated soil tends to have significantly higher EC, OM and moisture values than the rain-fed soil ($p<0.05$). These variations could be attributed to irrigation with mine effluent. Moisture from irrigation effluent may facilitate microbial decomposition of fallen foliage thus increase OM content (Table 2). Mine effluent may contain process wastes including ions derived from minerals in the ores that could have elevated soil EC since Cr is not found pure in nature [31]. The distribution and bioavailability of trace elements in soil depend on soil characteristics, plant species, soil management and the interrelationships between these factors [2, 12].

Figure 1 shows the concentrations of Cr in effluent-irrigated and rain-fed soils at two soil depths. The concentration of Cr in effluent irrigated soil (27.38±0.42) was significantly higher than that of rain-fed soil (5.39±0.10mg/kg) ($F_{1,58}=12.92$; $p=0.001$). The top soil layer (0-10cm) had significantly higher Cr concentration than the lower layer (10-30cm) in each treatment ($p<0.05$). The presence of Cr in the 10-30cm depth could be a result of irrigation which influences the amount, distribution and movement of trace elements in the soil [7]. Under neutral to basic pH, as in the present study (pH =7.50±0.10), the CrO_4^{2-} species predominates [14] as Cr(VI) and would therefore preferentially spread across the soil media at the two soil depths as observed. The presence of Cr in the rain-fed soil could be the natural content in soil. Naturally occurring soil generally contains low concentrations of Cr ranging from 10–50mg/kg [1] and from 1-1500mg/kg [2]. The concentrations of Cr in soil in the two treatments were within the natural total Cr background concentrations. When compared with other studies, the concentration of soil Cr in the present study (5-30mg/kg) was lower than values reported by [32] in an irrigated area in China (58.3-62.5mg/kg) but higher than values for agricultural soils (0.44-0.51mg/kg) at five sites reported by [33]. However, our values compare very well with findings reported by [5] in soil near a chromite area (11.17mg/kg) and by [12] for wastewater irrigated soil (23.17–39.54; mean:

29.86mg/kg). Variation in the concentration of Cr in soil could be a function of physicochemical properties of soils and the sources of Cr.

The concentration of Cr in mine effluent ranged from 18.5-33.7mg/l (mean: 26.10±2.48mg/l). This value is higher than 0.03-0.08mg/l reported by [6] but lower than 0.65mg/l recorded by [5] in irrigation wastewater near a chromite mine. The concentration of Cr in the irrigation effluent from this study is about 260 times more than the international threshold level of Cr (0.1mg/l) in irrigation water [34]. The observed differences in the concentration of Cr in mine effluent could be due to variations in the sources and the wastewater treatment processes.

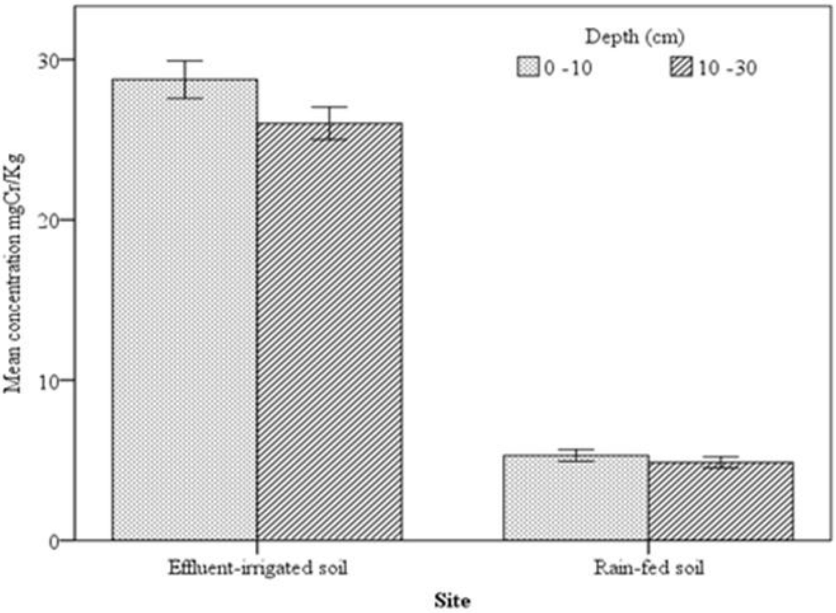

Figure 1 The concentration of Cr in mine effluent-irrigated and rain-fed soils. Values are mean±SE of five replicate samples measured in triplicate expressed in mg/kgCr dry soil.

The increase in heavy metal concentration in soil due to wastewater application has been reported [32, 6, 12]. In the present study, the concentration of Cr in soil remained significantly lower than the maximum allowable limit of 1200mg/kg [35] ($p<0.05$). The I-geo showed no pollution (-1.595) and the CF revealed no contamination (0.304) of the effluent-irrigated soil. Our study findings suggest no need for any drastic measures to remediate soil quality.

3.3 The concentration of Cr in plant tissues

Figure 2 shows the mean concentrations of Cr in plant tissue of *M. indica* and *M. paradisiaca* grown in mine effluent-irrigated soil as dry weight. No Cr was recorded for plant tissues from the rain-fed site. [13] reported that the concentration of Cr in plants growing in ‘normal’ soil is usually <1mg/kg, with typical values of 0.02-0.2mg/kg. The moisture content of the peeled edible fruits was 82.6% for *M. indica* and 75.2% for *M. paradisiaca. Musa parasidiaca* had significantly higher concentrations of Cr in roots (5.90±0.18), leaves (1.90±0.01) and fruits (0.55±0.00mg/kg) than *Mangifera indica*; roots (4.31±0.28), leaves (0.83±0.02) and fruits (0.35±0.01mg/kg) ($p<0.05$).

Chromium concentrations followed the trend: root>leaf>fruit for both plant species. This trend seems common for various trace elements in many plant species [5]. The concentrations of Cr in soil and plant roots were poorly correlated in *M. indica* ($r^2=0.327$; $p>0.05$) and *M. parasidiaca* ($r^2=0.126$; $p>0.05$). When compared with other studies, the concentration of Cr in fruit of *M. indica* in the present study (0.35mg/kg) was lower than values (1.92-2.43mg/kg) reported by [20] and those (0.56mg/kg) by [19] for fruits sampled from market survey studies. The concentration of Cr in fruit of *M. parasidiaca* in the present study (0.55mg/kg) was lower than the value (1.72mg/kg) reported by [19]. In a market survey, [18] recorded 0.001-0.03mg/100g (0.01-0.3mg/kg) for *M. indica* and 0.003-0.004mg/100g (0.03-0.04mg/kg) for *M. parasidiaca* fruits. Comparison with values from other studies may be complicated under situations where values are based on wet weight (WW) yet no moisture content was given. Differences in the concentrations of Cr in plant tissues could be due to the differences in plant physiology: while *M. indica* is a tree, *M. parasidiaca* is a grass. The concentration of trace elements in plants has been reported to vary with plant species [9]. Plant roots retained most of the Cr in each case with very little translocation to the above-ground tissues.

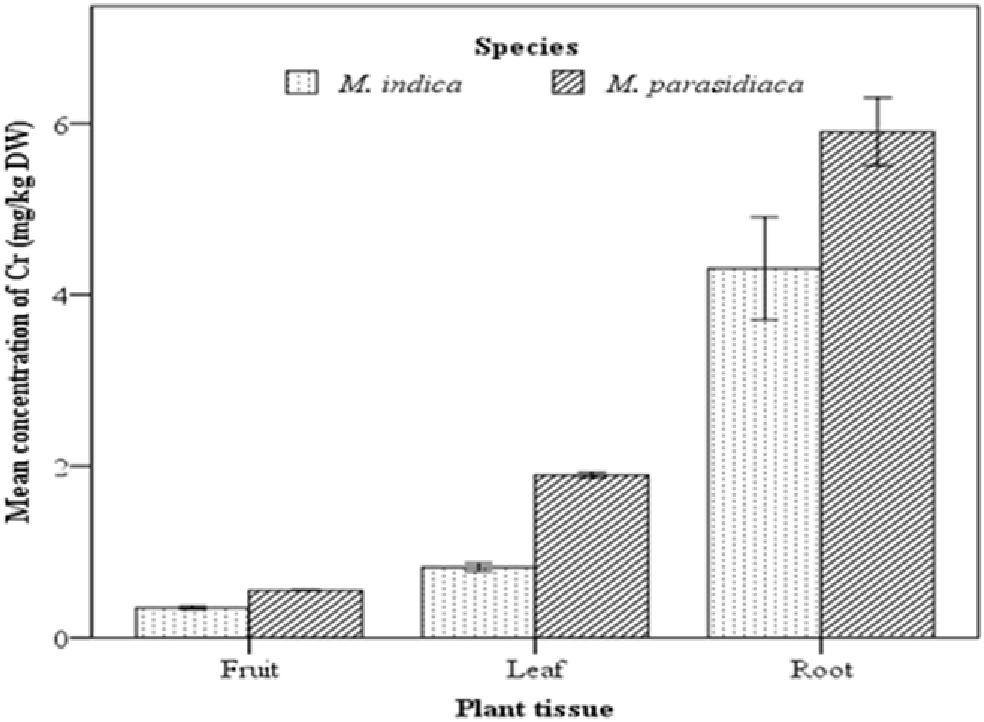

Figure 2 The concentration of Cr in plant tissues of *M. indica and M. paradisiaca* grown on mine effluent-irrigated soil. Concentrations are of replicates measured in triplicate expressed in mg/kgCr of dry weight

Table 3 The concentration of Cr in mine effluent, soil and fruits of *M. indica* and *M. parasidiaca*

Sample	Mean concentration of Cr	Maximum allowable limit
Irrigation water (mg/l) *n=5*	26.10±2.48	0.1[34]
Irrigated soil (mg/kg) *n=60*	27.38±0.42	1 200[35]
Fruit: *M. indica* (mg/kg) *(n=15)*	0.35±0.01	2.3[36]
M. parasidiaca (mg/kg) *(n=15)*	0.55±0.00	2.3[36]

3.4 Transference of Cr in tissues of *M. indica* and *M. paradisiaca*

Table 4 shows the transference (soil-plant tissue, tissue–tissue) factors for Cr. Transference values were less than unity for both plant species. This indicates that Cr slowly accumulates from the soil into the roots, slowly translocate to leaves and very slowly or not at all further to the fruit. However, the transference values in this study for both plant species are higher than typical values (0.0045, i.e. 0.45%) reported by [14].

Table 4 Transference factors for Cr from soil to plant tissues of *M. indica* and *M. paradisiaca* growing in mine effluent- irrigated soil. Values are used in dry weight (DW) basis.

Factor	Plant species	
	M. indica	*M. paradisiaca*
Metal Transference Factor	0.027	0.100
Bio-concentration Factor (Root/Soil)	0.160	0.220
Translocation Factor: AG/soil	0.040	0.090
: Leaf/root	0.200	0.33
: Fruit/root	0.100	0.086

3.5 Assessment of potential health risk due dietary exposure to fruits contaminated with Cr

The daily fruit intake (80g/cap/day) used in the present study is much higher than values given by [27] for Malawi (30.42g/cap/day) and Mozambique (23.29g/cap/day), Zimbabwe's neighbours. The estimated daily Cr intake values for *M. indica* (3.97E-05mg/kg) and M. *paradisiaca* (6.23E-05mg/kg) were much lower than [29] oral reference doses (RfD) of 1.5mg/kg for Cr(111) and 0.003mg/kg for Cr(V1). The HRI values for *M. Parasidiaca* fruits were 4.15E-05 for and 0.0208mg/kg for Cr(V1). *For M indica* the HRI values were 3.97E-05 for Cr(111) and 0.0132mg/kg for Cr(V1). These values are less than unity suggesting that there is no concern for potential adverse health effects [30].

IV. Conclusion

Results suggest that irrigating plants with effluent from a chromite mine elevated the concentration of Cr in soil to levels within the maximum allowable limits. *Mangifera indica* accumulates and translocate Cr more than *Musa parasidiaca*. However, both plant species poorly accumulate and very poorly translocate Cr from the soil to the above-ground parts. The consumption of fruits from the two plant species posed no health risk based on DMI and HRI assessments. The present study can be useful to farmers who irrigate fruit plants with treated or untreated wastewater. Furthermore, it may assist consumers in making informed decisions on the quality of fruits they eat. Mine effluents should be treated to the degree required to meet surface discharge safety or reuse quality standards. Continuous monitoring of soil and fruits irrigated with industrial effluents may be routinely done to ensure the safety of the consumers and quality of agricultural soil in the long term.

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