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ASSESSMENT OF HEALTH RISKS ASSOCIATED WITH CITRUS FRUITS EXPOSED TO HEAVY METAL TOXICITY IN DIVERSE IRRIGATION REGIMES

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Abstract

Heavy metals play a very important role in plant development, but exposure to these essential compounds at higher concentrations cause severe toxic effects in plants. Heavy metals are regarded as fundamental food supply pollutants due to their endurance, biomagnification, and non-biodegradability. Elevated trace metal levels in the diet imply a possible risk to human and environmental health. Despite that, these trace metals are an important part of our food. The present study was conducted at Tehsil Sargodha (Site FW-I and SW-II), Punjab, Pakistan to analyze Copper (Cu), Cobalt (Co), Nickel (Ni) and Zinc (Zn) concentration in water, soil and citrus fruits *(Citrus limetta* and *Citrus sinensis)*. The samples were analyzed with the help of atomic absorption spectrophotometer. The assessment of the bio-concentration factor (BCF), daily intake of metal (DIM), pollution load index (PLI), enrichment factor (EF), and health risk index (HRI) were carried out in the present findings. The EF of Co and PLI of Copper were remarked as more than 1 while other indices were found less than 1 for Cu, Co, Ni and Zn, indicated that fruits cultivated in water rich soil were not harmful, therefore heavy metal analysis was necessary to assess the extent of environmental contamination.

Key words: Sargodha; HRI; DIM; Metal contamination; PLI.

Introduction

The Rutaceae family includes the genus *Citrus*, represented by flowering shrubs and trees. Plants in the genus *Citrus* produce citrus fruits like lemons, limes, oranges, pomelos, and grapefruits. Citrus is native to Australia, East Asia, Southeast Asia, South Asia, and Mediterranean region (Zech-Matterne, *et al.*, 2018). In south Asia, citrus fruits account for about one-third of total fruit export value. The main developed citrus crops are mandarins, oranges, grapefruits, limes and, lemons which have been widely known for their nutritive worth (Satari & Karimi, 2018).

Heavy metals are considered essential contaminants in the food chains because of their persistence in the environment. Both humans and environment are potentially at risk due to elevated levels of these metals (Khan *et al.*, 2019a, Raja *et al.*, 2016). Assessment of risk is generally concerned with likelihood of any risk capable of connecting with exposure to pollutants. Human health risk assessment includes gathering, identifying, and integrating information about hazardous pollutant exposure and its negative health effects (Sobhanardakani, 2017a, 2017b).

The quantity of dangerous smoke and pollutants emitted into the atmosphere is strongly correlated with the number of heavy cars on the roads or highways. Some of these toxins are copper, chromium, cadmium, nickel, arsenic and lead which, although occasionally being helpful, pollute the environment (Chen *et al.*, 2021, Suvarapu & Baek, 2017). Cu is one of these contaminants, and while it is occasionally useful, it also pollutes the environment (Chen *et al.*, 2021).

In fact, the deficiency of copper, phosphorus, and manganese caused by zinc toxicity in plants is defined by the typical purple-red hue of foliage (Bhalakiya *et al.*, 2019). In addition, zinc limits the development of soil microorganisms by altering their shape and metabolic activity (Baran *et al.*, 2018). Because of the toxicity, abundance, non-biodegradability and accumulative nature of heavy metals, their contamination and accumulation pose a severe risk to human community worldwide. As a result of rapid industrialization, global trade and several other anthropogenic activities, the variety of environmental contaminants has substantially increased (Khan *et al.*, 2019a, Tóth *et al.*, 2016). Human activity in areas without household sewage or trash processing facilities may lead to discharge of metals into the environment. The fruits may become contaminated with trace and hazardous metals. As a result, human health is effected by this contamination (Ngo *et al.*, 2021).

Wastewater may be more reliable for irrigation than rainfall or groundwater in terms of availability and nutrient supply (Chaganti *et al.*, 2020, Martínez-Cortijo & Ruiz-Canales, 2018). Despite the availability of freshwater in some areas, farmers prefer wastewater to irrigate the fields because of high yield (Deh-Haghi *et al.*, 2020).

Many landowners in Pakistan employ eccentric water resources (treated and untreated) for forestry and farming due to the state's arid climate and lack of freshwater. City discharge, industrial effluent, wastewaters, and tainted canal water are important sources of irrigation water that several farmers have explored as an alternate supply of water. In practically every community across the country, this method is growing quickly. Wastewater has thus evolved into a practical method for lowering shortage of water (Hassan *et al.*, 2013, Riaz *et al.*, 2022, Ugulu *et al.*, 2019a).

The objective of this study was to analyze the bioconcentration of cobalt, nickel, zinc and copper metal in soil, fruits and water and to evaluate the risks posed to humans. The metal flow in the food chain can be assessed by examining the metal profile in the collected samples. This research also aimed to assess the mode of transfer of these trace metals in water–soil–fruits continuum with hazard risk assessments.

Material and Methods

Area of research: Sargodha is the third largest division in Punjab province of Pakistan. Sargodha district spreads over 5,864 square kilometers. Sargodha has a latitude of 32.082 and a longitude of 72.669 (Fig. 1). The average temperature of the area fluctuates between 42-105 ˚F. Sargodha has mostly fertile, flat plains, with only a few small hills along the Sargodha-Faisalabad Road (Khan *et al.*, 2019b, Khan *et al.*, 2019c).

Sample collection: The soil, water and plant samples were gathered from two different sites arbitrarily. Samples were collected from Chak 75 N.B. (named as Site-FW-I irrigated with fresh water) and from Risala No. 5 (named as Site-SW-II irrigated with sewage water) (Table 1).

Table 1. Botanical and common names of analyzed fruits.

Serial No.	English name	Botanical names
1.	Sweet lime	*Citrus limetta*
2.	Sweet orange	*Citrus sinensis*

Water and soil sampling: For collection of water samples, at each site, 500 mL plastic containers were used. The soil samples were gathered from selected sites and saved in polythene bag. The containers were sealed tightly, put in an ice box (5°C), and then brought to the lab to be tested for EC (electrical conductivity) and other physicochemical characteristics. After that, the water samples were filtered and refrigerated before the analysis of metals.

Fruit samples: The samples of edible portion of fruits (*C. limetta* and *C. sinensis*) were collected from the selected sites. Each sample was chosen randomly, sealed in a particular brown envelope, labelled, and transferred to the research lab for further evaluation. Each sample included three replicates.

Soil and fruit sample preparation: Soil and fruit (edible portion) samples were left to dry in the air before being placed in an oven for 3 days at 72˚C. Samples were air-dried until no moisture content remained and weighed using an electronic balance. The samples were crushed into a powder and kept in desiccators at room temperature. The samples were digested using the wet acid digestion method (Abbasi *et al.*, 2016, Sun *et al.*, 2017).

Wet acid digestion and analysis of samples: To ensure complete solubilization, three replicates of samples of soil, water, and fruits were digested in an acid mixture. Approximately, 1 ml water sample and 1.0 g of the powdered samples of soil and fruit were treated with 10 mL of concentrated HNO_3 (65%) and left unheated for overnight. Next day, the mixture was heated on electric burner for 70°C till evaporation took place (Abbasi *et al.*, 2015, 2016).

The mixture was cooled to room temperature and 5 ml of undiluted $HClO_4$ (70%) was added. The mixture was then reheated at 70°C until thick, white fumes began to appear, signifying that the digestion was complete. Whatman paper # 42 was then used to filter the samples. The digested samples were put in a flask of 50 mL and filled on-to the proper level with 0.1 N HNO_3 (Parveen *et al.*, 2020). The following metals were evaluated i.e., Cu, Co, Ni and Zn was explored with atomic absorption spectrophotometer (Añón & Calvelo, 1980).

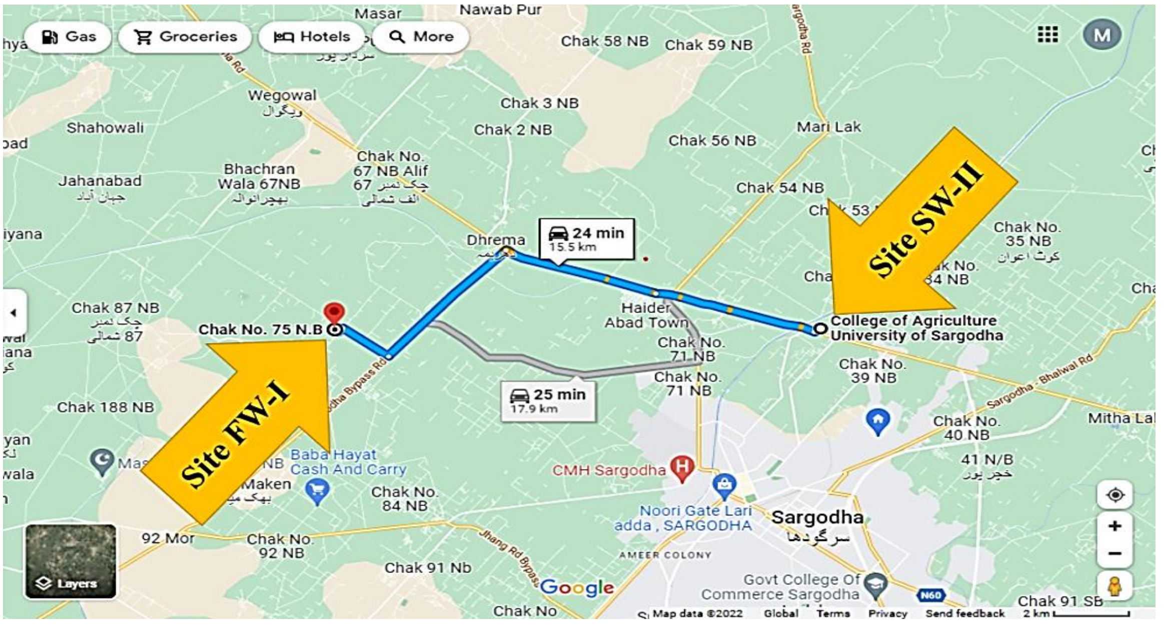

Fig. 1. Map of study area.

Evaluation of metal profile by statistical analysis: The data of soil, fruits and water samples was statistically analyzed. SPSS 23 software and Graphed Prism Pad was used to determine variance and correlations.

Pollution load index: The pollution load index is established on the rate of the metal in the soil, seeks to provide an estimate of the pollution status. The following formula (1) was used in this study according to Liu *et al*., (2005).

$$\text{PLI} = \frac{\text{Metal, content in tested soil}}{\text{Metal, reference value for soil}} \quad (1)$$

Reference values for cobalt, nickel, copper and zinc were 9.1, 9.06, 8.39 $mgkg^{-1}$ (Singh *et al.,* 2010) and 44.19 $mgkg^{1}$ (Hassan *et al.*, 2013) respectively.

Bio concentration factor: It was calculated using the following formula by Akhtar *et al*., (2022).

$$\text{BCF} = \frac{\text{MV (Fruits)}}{\text{MV soil}} \quad (2)$$

where,
M_v (Fruits) = Metal value. ($mgkg^{-1}$) in fruits; M_v (Soil) = Metal value. ($mgkg^{-1}$) in soil
Enrichment factor: EF is calculated by following formula (Ahmad *et al*., 2016).

$$\text{EF} = \frac{\text{MV in sampled fruit/MV in sampled soil}}{\text{MV}\left(\frac{\text{Fruit}}{\text{Soil}}\right)\text{ Standard value}} \quad (3)$$

Table 2. Standard concentrations of metals. All values are according to FAO/WHO (2010).

Heavy metals	Fruits
Cu	0.05-0.5
Co	2
Ni	2
Zn	60

Daily intake of metal (DIM): There is a diversity of pathways for heavy metals to accumulate in human body, including through skin contact, inhaling, and eating contaminated forages (Table 2). The regular consumption of metals was calculated using the following formula (Shahid *et al.*, 2015).

$$\text{DIM} = \frac{\text{C x F x D food intake}}{\text{W}} \quad (4)$$

C represents the concertation of metal in plants, F represents conversion factor, $D_{\text{food intake}}$ represents fruits intake per day, and W is Average body weight of humans (Table 3).

Health Risk Index (HRI): Health index was utilized to assess the potential metal exposure that might happen if humans consumed the sampled citrus fruit samples (*Citrus limetta* and *Citrus sinensis*). The daily metal intake (DIM) in fruit products separated by the oral reference dosage is used to determine HRI (Khan *et al.*, 2020b).

$$\text{HRI} = \frac{\text{Daily intake of metals (DIM)}}{\text{Oral reference dose (RfD)}} \quad (5)$$

RfD for Co, Ni, Zn and Cu was 0.043 (Campbell, 2004, Trumbo *et al.*, 2001), 0.02, 0.37 and 0.04 mg/kg (USEPA, 2011) respectively.

Results

Metal concentration (mgL^{-1}) in irrigation water: The mean concentration of Cu, Co, Ni and Zn in water varied from 0.062 to 0.37 mg/L, 0.08 to 0.14 mg/L, 1.54 to 2.24 mg/L and 0.56 to 0.761 mg/L respectively. The means of Cu, Co and Zn concentration was found higher in water at SW-II compared to FW-I, while highest level of Ni was found in water samples at FW-I (Tables 4, 5).

Metal concentrations in soil samples: The results from analysis of the variance of data revealed that the concentration of Cu was non-significant while Ni and Zn concentrations were highly significant $p<0.001$ and the concentrations for Cu, Co, Ni and Zn ranged from 6.01 to 10.88 $mgkg^{-1}$, 0.19-0.42 $mgkg^{-1}$, 0.29 to 5.041 $mgkg^{-1}$ and 0.86 to 2.39 $mgkg^{-1}$ respectively. Maximum mean concentrations of Cu, Co, Ni and Zn were found at site SW-II (Tables 6, 7).

Metals concentration in fruits, *Citrus limetta* and *Citrus sinensis*: The Cu, Co, Ni and Zn concentration ranged from 0.24 to 0.39 mg/kg, 0.10 to 0.36 $mgkg^{-1}$,0.062 to 0.37 $mgkg^{-1}$ and 0.23 to 1.52 $mgkg^{-1}$ respectively. The concentrations of Cu and Co were highly significant $p<0.001$ but Ni and Zn were non-significant ($p>0.05$) (Tables 8, 9).

Pollution load index for Cu, Co, Ni and Zn: The values of PLI fluctuated from 0.76 to 1.24 for Cu, 0.02 to 0.047 for Co, 0.032 to 0.56 and 0.019 to 0.054 for zinc (Table 10). Soil of *C. limetta* at SW-II confirmed elevated PLI value (1.24) for Cu compared to others. While, least value detected was 0.021 in the soil of *C. sinensis* at FW-I for Co.

Bio concentration factor for copper, cobalt, nickel and zinc: The BCF values for Cu, Co, Ni and Zn varied from, 0.025 to 0.062 $mgkg^{-1}$, 0.53 to 0.98 $mgkg^{-1}$ for Cu, 0.10 to 0.75 and 0.16 to 0.66 respectively (Table 11). BCF content of Zn (0.66 mg/kg) in C. limetta was higher at SW-II compared to other samples.

Enrichment Factor for **copper, cobalt, nickel and zinc:** The values for EF fluctuated among 0.42 to 1.047 for Cu, 2.43 to 4.46 for Co, 0.34 to 3.39 for Ni and 0.12 to 0.49 for Zn (Table 12). Cu metal showed the highest Enrichment Factor (1.047) at site SW-II in *C. sinensis*.

Daily intake of copper, cobalt, nickel and zinc: DIM of Cu, Co, Ni and Zn had variation among all locations. The values of daily intake in this study for Cu varied from 0.0041 to 0.014 $mgkg^{-1}day^{-1}$, 0.0090 to 0.015 $mgkg^{-1}day^{-1}$ for Co, 0.002 to 0.014 $mgkg^{-1}day^{-1}$ for Ni and for Zinc 0.0091 -0.041 $mgkg^{-1}day^{-1}$. Highest values of daily intake were detected for zinc (0.059 $mgkg^{-1}day^{-1}$) at SW-II in *C. limetta*, while lower values were noticed for Ni (0.002 $mgkg^{-1}day^{-1}$) at site FW-I in *C. limetta* (Table 13).

Table 3 Values for the daily food intake, conversion factor and average body weight.

Specimen	$D_{food\ intake}$	Conversion factor	Average body weight
Human	31.5 g/person/day*	0.085**	70 kg*

* WHO (2003), **Jan *et al*., (2010)

Table. 4 Analysis of variance for Cu, Co, Ni and Zinc in water.

Source	DF	Cu means	Co means	Ni means	Zn means
Sites	1	0.117^{ns}	0.001***	0.063^{ns}	0.061^{ns}
Error	4	0.003	0.000	0.157	0.008
Total	5				

Table 5. Mean and SE (standard error) for Cu, Co, Ni and Zn in water (mgL^{-1}).

Sites	Cu				Co				Ni				Zn			
	Mean	SE	Min	Max	Mean	SE	Min	Max	Mean	SE	Min	Max	Mean	SE	Min	Max
FW-I	0.06	0.00	0.05	0.07	0.07	0.002	0.07	0.09	1.54	0.06	1.41	1.61	0.55	0.034	0.45	0.67
SW-II	0.34	0.02	0.26	0.40	0.08	0.004	0.10	0.11	1.74	0.11	1.37	2.37	0.76	0.04	0.68	0.80
FW-I	0.09	0.018	0.05	0.17	0.09	0.003	0.08	0.09	2.12	0.094	2.01	2.24	0.56	0.024	0.45	0.67
SW-II	0.37	0.031	0.26	0.49	0.14	0.012	0.10	0.20	2.08	0.026	2.06	2.09	0.76	0.06	0.65	0.87

Table 6. Analysis of variance data for Cu, Co, Ni and Zn in soil (mgkg^{-1}).

Source	DF	Mean squares of Cu	Mean squares of Co	Mean squares of Ni	Mean squares of Zn
Sites	1	1.217^{ns}	0.082568*	37.1680***	4.10600***
Plants	1	22.325**	0.012224ns	2.8036**	0.38636**
Sites*Plants	1	8.098*	0.003482ns	4.5425**	0.21403^{ns}
Error	8	1.349	0.014954	0.2406	0.04816
Total	11				

*** Indicates highly significant data at level 0.001

Table 7. Cu, Co, Ni and Zn concentration (mean and standard error, SE) in soil mgkg^{-1}.

Sites	Cu				Co				Ni				Zn			
	Mean	SE	Min	Max	Mean	SE	Min	Max	Mean	SE	Min	Max	Mean	SE	Min	Max
FW-I	9.37	0.19	8.35	10.39	0.19	0.009	0.12	0.25	0.29	0.009	0.28	0.31	0.86	0.04	0.79	0.91
SW-II	10.38	0.27	9.40	11.40	0.36	0.031	0.26	0.47	5.04	0.08	4.92	5.16	2.30	0.05	2.20	2.38
FW-I	8.29	0.38	7.30	9.32	0.20	0.012	0.13	0.25	0.55	0.18	0.22	0.82	1.49	0.102	1.24	1.89
SW-II	6.01	0.68	4.37	7.39	0.42	0.04	0.35	0.47	2.84	0.32	1.86	3.70	2.39	0.14	2.20	2.66

Table 8. Analysis of variance data for Cu, Co, Ni and Zn in fruits (mgkg^{-1}).

Source of variation (S.O.V)	DF	Mean squares of Cu	Mean squares of Co	Mean squares of Ni	Mean squares of Zn
Sites	1	0.051798***	0.180811***	0.02708^{ns}	3.21782^{ns}
Plants	1	0.002380^{ns}	0.000000ns	0.05165^{ns}	0.19799*
Sites*Plants	1	0.000002^{ns}	0.000095ns	0.14734*	0.12706*
Error	8	0.001189	0.000672	0.02755	0.02042
Total	11	0.051798***			

*** Significant at when $p<0.001$ level; ns= Non-significant at $p>0.05$

Table 9. Cu, Co, Ni and Zn concentration in fruits.

Sites	Cu				Co				Ni				Zn			
	Mean	SE	Min	Max	Mean	SE	Min	Max	Mean	SE	Min	Max	Mean	SE	Min	Max
FW-I	0.24	0.025	0.17	0.28	0.10	0.00	0.09	0.12	0.062	0.005	0.05	0.07	0.28	0.03	0.21	0.33
SW-II	0.36	0.009	0.35	0.38	0.35	0.01	0.34	0.38	0.37	0.011	0.36	0.40	1.52	0.04	1.44	1.60
FW-I	0.26	0.014	0.24	0.28	0.11	0.00	0.10	0.12	0.41	0.01	0.13	0.77	0.23	0.07	0.11	0.36
SW-II	0.39	0.004	0.39	0.40	0.36	0.02	0.31	0.39	0.28	0.02	0.26	0.35	1.06	0.13	0.88	1.33

Table 10. Pollution load index for copper, cobalt, nickel and zinc.

Sites	Cu		Co		Ni		Zn	
	C. limetta	*C. sinensis*	*C. limetta*	*C. sinensis*	*C. limetta*	*C. sinensis*	*C. limetta*	*C. sinensis*
FW-I	1.12	0.99	0.022	0.021	0.032	0.061	0.019	0.034
SW-II	1.24	0.76	0.040	0.047	0.56	0.31	0.052	0.054

Table 11. Bio concentration factor for Cu, Co, Ni and Zn.

Sites	Cu		Co		Ni		Zn	
	C. limetta	*C. sinensis*	*C. limetta*	*C. sinensis*	*C. limetta*	*C. sinensis*	*C. limetta*	*C. sinensis*
FW-I	0.025	0.032	0.53	0.57	0.21	0.75	0.33	0.16
SW-II	0.035	0.062	0.98	0.82	0.075	0.10	0.66	0.45

Table 12 Enrichment factor for copper, cobalt, nickel and zinc.

Sites	Cu		Co		Ni		Zn	
	C. limetta	*C. sinensis*	*C. limetta*	*C. sinensis*	*C. limetta*	*C. sinensis*	*C. limetta*	*C. sinensis*
FW-I	0.42	0.53	2.43	2.61	0.97	3.39	0.25	0.12
SW-II	0.59	1.047	4.46	3.73	0.34	0.46	0.49	0.33

Table 13. Daily intake of Cu, Co, Ni and Zn (mgkg^{-1}day^{-1}).

Sites	Cu		Co		Ni		Zn	
	C. limetta	*C. sinensis*	*C. limetta*	*C. sinensis*	*C. limetta*	*C. sinensis*	*C. limetta*	*C. sinensis*
FW-I	0.0041	0.0043	0.0090	0.011	0.002	0.016	0.011	0.0091
SW-II	0.014	0.013	0.014	0.015	0.014	0.011	0.059	0.041

Table 14. Health risk index for copper, cobalt, nickel and zinc.

Sites	Cu		Co		Ni		Zn	
	C. limetta	*C. sinensis*	*C. limetta*	*C. sinensis*	*C. limetta*	*C. sinensis*	*C. limetta*	*C. sinensis*
FW-I	0.23	0.25	0.094	0.099	0.12	0.79	0.029	0.025
SW-II	0.35	0.38	0.32	0.31	0.73	0.55	0.16	0.11

Health risk index of copper, cobalt, nickel and zinc: The values of HRI for Cu, Co, Ni and Zinc varied from, 0.23 to 0.38 mgkg^{-1}day^{-1}, 0.094 to 0.34 mgkg^{-1}day^{-1}, 0.12 to 0.79 mgkg^{-1} day^{-1} and 0.025 to 0.16 mgkg^{-1}day^{-1}, respectively (Table 14). Site FW-I in C. sinensis showed maximum concentration of HRI for Ni, while least was examined at FW-I in C. limetta for Zn.

Discussion

Heavy metal contamination is a worldwide issue, especially in the food commodities, causing environmental challenges in the developing world. This study was particularly carried out to assess the level of contamination in citrus grown fields of district Sargodha and have an insight on risks posed to consumers of sweet orange and sweet lime. Sargodha region is well known for its citrus production and amid shortage of freshwater sources, farmers have now shifted to alternate sources of irrigation like wastewater (Khan *et al*., 2023).

Cu concentrations in water samples ranged from 0.062 to 0.37 mg/L. In comparison to the value of 0.10 mg/L stated by Chaoua *et al*., (2019), both irrigation waters had significantly higher Cu concentrations at SW-II. A study carried out on wastewater and groundwater, however, yielded higher mean levels of Cu (1.29, 1.03 mg/L) in comparison to the current study (Khan *et al*., 2017). Some samples had Cu concentrations above than the values determined by. Higher copper levels (1.842 mg L^{-1}) were also found as reported by Kumar & Chopra (2015).

In the current study, the mean concentration of cobalt in water ranged from 0.079 mg/L to 0.140 mg/L that was higher than the value of 0.0003mg/L (Khaskhoussy *et al.*, 2013). According to Chiroma *et al*., (2014) and USEPA (2011), the threshold limit of Co accumulation in water is 0.05 mg/L and the value of cobalt in irrigation water at both sites (FW-I and SW-II) exceeded this threshold value. In comparison to other studies, the current Co concentrations were below the values documented by Yaqub *et al*., (2021); Ugulu *et al*., (2021a) and Ugulu *et al*., (2022b).

All water samples had Ni concentrations that ranged between 1.54 and 2.24 mg/L. In waste water, Hassan *et al*., (2013) and Ugulu *et al*., (2020) recorded a lower Ni concentration of 0.05 mg/L compared to our results. According to Ahmad & Goni (2010) and Khan *et al*., (2020a) the concentration of Ni in recent study was higher than their findings. The present value of Ni in water was found to be higher than the findings of Pescod (1992) which was 0.2mg/L. Farmers utilize industrial and municipal waste excessively and untreated, which contaminates the water. This is a significant source of Ni contamination in water storage reservoirs.

Mean concentration of Zn (mgL^{-1}) in water ranged from 0.55 to 0.761. When the present research was compared with other studies, it was observed that the values of Zn in the present water samples were below the concentrations reported by Aurangzeb *et al*., (2014). In comparison to the results of the current investigation, Mousavi & Shahsavari (2014) recorded low values for Zn (0.010-0.021 mgL^{-1}) in ground water. Zn content in water was reported to be higher 7.2 mgL^{1} than the findings by Hassan *et al*., (2013). High Zn concentrations were recorded in Lahore canal water by Kashif *et al*., (2009).

For the soil samples, Cu concentration in this study varied from 6.01 to 10.38 mg/kg. Elevated Cu level (32.71 mg/kg) in soil treated with sewage water was reported by Alghobar & Suresha (2016). In contrast to this experiment, Wang *et al*., (2015) evaluated the low copper range (1.09-1.55 mg/kg). Compared to the level (1.09, 1.55 mg/kg) provided by Khan *et al*., (2017) our estimated value for Cu was greater. Ahmad *et al*., (2016) and Wajid *et al*., (2020) found that Cu concentrations ranged from 2.79-4.13 mg/kg which were below the concentrations of Cu in present study.

Co was found in soil at various concentrations. The mean value of Co in soil varied from 0.195 mg/kg to 0.428 mg/kg. Overall, Co accumulation in soil was within the WHO (2003) guidelines. Heavy metal contamination of soil raises the risk of metal uptake by plants and deposition in various edible parts (Ali & Al-Qahtani, 2012). USEPA (1997) reported that the permissible maximum limit of Co accumulation in soil was 750 mg/kg.

In both soils at FW-I and SW-II, the observed mean levels of Ni ranged from 0.19 to 0.42 mg/kg. Nickel buildup may be influenced by variables such soil pH, plant characteristics and kind. Nickel (Ni) accumulation is also influenced by anthropogenic soil contamination. According to Khan *et al*., (2017), Ni concentration in soil irrigated with control (1.36 mg/kg) and sewage water (1.61 mg/kg) varied a little, however, it was still higher than the results of the current study. In comparison to the current findings, Shah *et al*., (2009) found greater Ni level (36.0 mg/kg) in soil. Ni concentration in soil was lower than the value of 1.90-3.74 mg/kg reported by Ahmad *et al*., (2016). The concentration 13.29 to 13.99mg/kg of Ni in soil was observed by Leogrande *et al*., (2019) which was greater than the present study. The current values of Ni in soil were lower than the values recorded by Addis & Abebaw (2017).

Average concentration of zinc in soil samples collected from all sites fluctuated between 0.86 to 2.39 mg/kg. Eissa & Almaroai (2019) recorded the Zn level in the soil (600 mg/kg) to be much higher compared to current study. Zn levels in the soil of the current region were significantly lower than those of 3.205-6.910 mg/kg as reported by Orisakwe *et al*., (2017). The concentration of 1.09-1.55 mg/kg was provided by Khan *et al*., (2017) whereas the current reported range for Zn was lower. When compared to recent research values, Özyazici (2013) obtained the highest value for zinc, i.e., 83.3-58.8 mg/kg. Similarly, Yu *et al*., (2016) established that Zn had a higher value than attained in current work. Current Zn values in soil were also lower than calculated values of Ran *et al*., (2016).

The copper level in the analysis of fruits ranged from 0.24 to 0.39 mg/kg. The buildup of Cu is influenced by the types of plants and their needs. Small amounts of copper are necessary for plants to develop normally (5-20 mg/kg). However, a concentration of more than 20 mg/kg can make copper hazardous. In comparison to this study, Kumar (2016) reported the highest Cu content (5.5–18.7 mg/kg) in the green leafy vegetables. According to Chaoua *et al*., (2019) in the edible fraction, copper buildup was lower (6.57–6.86 mg/kg). The Cu content (0.52 mg/kg) suggested by Aurangzeb *et al*., (2014) was discovered to be lower than the present range.

For fruits, the mean concentration of cobalt varied from 0.106-0.357 mg/kg. The observed values were according to the permissible limits of FAO/WHO (2010). Co in fruits were below the global average and appeared to be less than the allowable limit (Al-Sayegh & Al-Yazichi, 2001). Plants require cobalt for functioning of certain enzymes which is why cobalt affects metabolism in plants. Co also interacts with other elements and forms complexes which may be phytotoxic depending on the type of complex.

Nickel was found in abundance in citrus fruits harvested from the FW-I irrigated site. The current results were thought to be above than those determined by Ihesinachi & Eresiya (2014). Ni was found in high concentrations (0.08 mg/kg) in oranges sold in Owerri, Nigeria (Orisakwe *et al.*, 2012). However, in another study, Ni mean level in orange was 0.129 mg/kg (Sobukola *et al*., 2010). According to our findings, the Ni concentration in citrus fruit was 0.415 mg/kg. These values were found to be above the allowable level of 0.14 mg/kg set by FAO/WHO (2010).

The zinc concentration in fruits was lower than the acceptable limit (100 mg/kg) established by Chiroma *et al*., (2014), which ultimately showed a deficiency of Zn in fruits in the current investigated area. In the study, Zn average content was varied from 0.23 to 1.52 mg/kg. Zinc is a vital nutrient required for plant growth and several metabolic processes. Zn is easily available to plants in solution forms and its accumulation depends on amount of zinc in nutrient solution and soil. Chaoua *et al*., (2019) reported maximum Zn values in plants (60.515, 86.35 mg/kg) than the current findings. Singla & Dhawan (2017) observed a zinc content ranging from 26.0 to 31.30 mg/kg, which was found to be significantly higher than the currently detected range. The concentrations of Zn from this study were found to be about ten times lower in all the samples compared to the permissible level of 99.40 mg/kg by (WHO, 2003).

The pollution load index values, which stayed within the range of 1, indicated that the soil in the current research region was not contaminated with copper and the fruits grown there were safe for human health. This study's PLI value was lower than the Cu reference values (8.39) as suggested by Singh *et al*., (2010). The value of PLI for Co in soil ranged from 0.047 to 0.0214 which was found to be lower compared to the value reported by Khan *et al*., (2015).

In these soil samples, PLI values ranged between 0.032 and 0.31. The PLI value for Ni found in this study was lower than earlier report (Proshad *et al*., 2017). Recent findings indicated that the soil in the studied site was safe for growing fruit. The pollutant load index value for Ni in the current analysis fell below the reference values of 9.06 and 2.89-3.67 as reported by Singh *et al*., (2010) and Ahmad *et al*., (2016) respectively when compared to the current study. It was determined that the Ni value of 1. 62 proposed by Ahmad *et al*., (2014) fell within the parameters noted in this study.

The soil of the researched region is not polluted, as shown by the current data, and the soil is safe for fruit cultivation. The Zn PLI level was also less than 1. PLI values for Zn were lower than the report of Singh *et al*., (2010) reference value (44.19). Ahmad *et al*., (2016) confirmed a higher PLI value for Zn (1.47) in contrast to the current study. It was found that the Zn PLI level (0.05) recommended by Ezemokwe *et al*., (2017), was below the current concentration.

It was found that the bio-concentration for Cu (4.38-5.05) provided by Ahmad *et al*., (2016) was greater than the present results. The highest concentrations observed in BCF for Co was 0.035. The highest concentration of nickel observed for BCF was 0.74. Compared to this experiment, Ahmad *et al*., (2016) found greater BCF values for nickel (2.948-4.149). According to Khan *et al*., (2017) the nickel BCF value was found to be greater than the current study.

The DIM for Cu (0.004 $mgkg^{-1}day^{-1}$) determined by Chaoua *et al*., (2019) was found to be consistent with the findings of this study. Ismail *et al*., (2015) observed that the suggested DIM for Cu (0.0002 $mgkg^{-1}day^{-1}$) was lower in comparison to the present study. The present Cu DIM was lower than the recommended highest acceptable level of 3 $mgkg^{-1}day^{-1}$.

The investigation showed that the Ni DIM ranged from 0.002 to 0.016 $mgkg^{-1}day^{-1}$. Khan *et al*., (2019d) investigated lower Ni DIM levels (0.006, 0.008, $mgkg^{-1}day^{-1}$). Likewise, Ismail *et al*., (2015) suggested a related DIM value of 0.002 $mgkg^{-1}day^{-1}$ for nickel as tested in this research. Ni consumption per day was lower in all samples than the expected daily permissible intake (1.4 $mgkg^{-1}day^{-1}$) (USEPA, 2011).

Research revealed that the Zn DIM ranged between 0.0091-0.059 $mgkg^{-1}day^{-1}$. There was no health risk for humans consuming polluted fruits in the study region because the DIM values for Zn were less than 1. Compared to the current study's DIM levels (0.0091 - 0.059 $mgkg^{-1}day^{-1}$), Nadeem *et al*., (2020) and Ugulu *et al*., (2022a), recorded a wider range of DIM values (0.039-0.769 $mgkg^{-1}day^{-1}$).

Cu health risk index values for both youth and adolescents in the study are below 1. The HRI<1 indicates, in accordance with USEPA (2011), that consumers don't face any immediate health risks (Liang *et al*., 2015, Tariq *et al*., 2021, Ugulu *et al*., 2019b). According to our findings, HRI occurred between 0.22 and 0.37. Therefore, it can be said that Cu in citrus fruit samples does not pose a risk to consumers' health. Similar results were also reported by Ugulu *et al*., (2021b, 2021c).

The mean concentration of HRI for Co ranged from 0.094 to 0.32. When the findings of HRI from this report's analysis were compared with those from Bibi *et al*., (2014) and Khan *et al*., (2019d) study, the HRI value was found to be slightly higher.

In the current study, HRI for Ni was less than 1, demonstrating that Ni had no harmful effects on the health of the humans who would consume these fruits grown on two different irrigation waters. The outcomes of the research showed that the HRI for Ni observed in this study ranged between 0.11-0.79. As a result, it is possible to conclude that Ni in citrus fruit samples posed no health risks to consumers. The current Ni HRI values were higher than that of the 0.39 described by Singh *et al*., (2010).

Ahmad *et al*., (2016) found a higher HRI for Zn concentration than what was found in the current study (0.0165-0.257). In contrast to the present investigation, Khan *et al*., (2017) discovered greater HRI Zn values (0.537–0.609). A higher HRI Zn value (0.040-0.021) was also recorded by Lawal *et al*., (2017).

Conclusion

The Cu, Co, Ni and Zn translocation from the soil and fruits to the human being food chain revealed that Cu, Co, Ni and Zn concentration in soil and fruit samples was within the acceptable USEPA and WHO limits. It was found that there were variations in concentration of these metals across the soil-fruit-human continuum at various locations. This study analyzed soil and citrus fruits contamination with Cu, Co, Ni and Zn as well as potential future health risks, was found a step towards pollution by these metals, however, it did not represent a concern to human health because the HRI and DIM values were less than 1.

Recommendations and Implications

Ecotoxicological assays are useful tools for analyzing Cu, Co, Ni, and Zn contamination levels chemically. The evaluation of the toxicity in soil and fruits should not simply utilize the pollution indices. Biological markers are another crucial tool for understanding the impact of pollutants on the environment. By examining dangerous metals, we can monitor how poisonous the environment is to people. It is important to be aware of the many poisons, metal pollutants, and significant plant stages during which HMs might infiltrate the environment and food chain, as well as the illnesses caused by them, in order to safeguard the public's health. In order to aid in the development of regulations, the government should regularly assess the quantity of HMs in biological and ecological aspects.

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