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A Comparative Study on the Bioavailability and Soil-to-Plant Transfer Factors of Potentially Toxic Element Contamination in Agricultural Soils and Their Impacts: A Case Study of Dense Farmland in the Western Region of Saudi Arabia

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Abstract: Soil and aquatic pollution by heavy metal (Pb, Cr, Cu, Fe, Zn, and Ni) ions has become one of the prime problems worldwide. Thus, the purpose of the current study is to conduct hydrogeological research and quantify the main trace metals in the edible vegetables, soil, irrigation water, pesticides, and fertilizers in the farmland near Jeddah city, Saudi Arabia. Samples of soil, water, and plants such as coriander (*Coriandrum sativum*), dill (*Anethum graveolens*), parsley (*Petroselinum crispum*), and arugula (*Eruca sativa*) were collected, acid-digested, and analyzed using an inductively coupled plasma–optical emission spectrometer (ICP–OES). The levels of the elements in soil were determined in the order of Fe > Zn > Cu > Cr > Ni > Pb, whereas the sequence in plants was Fe > Cr > Zn > Pb> Ni > Cu, and in water, the order was Pb > Fe > Cu > Zn> Ni = Cr. In soil, the levels of Fe, Cr, and Pb were higher than the recommended values set by the World Health Organization (WHO) and the Food Administration Organization (FAO). In soil, Pb and Zn uptake increased with an increase in the availability of both elements, whereas in plants, Zn and Pb uptake occurs primarily through the plant roots, and some specific proteins facilitate metal transport and movement across the membrane. In soil, the root cell walls first bind to metal ions, which are taken up across the plasma membrane. The levels of the investigated elements in water and vegetables samples were below the permissible limits set by the FAO and within the allowable limits in the available pesticides and fertilizers. The transfer factor (TF) of metal absorption from soil to plant ($TF_{soil\text{-}plant}$) and from irrigated water to plant ($TF_{water\text{-}plant}$) in the study area was determined, followed by correlation and statistical treatment according to the date. The TF values were used to assess the metal levels in collected plant, soil, and water samples. The computed values of TF implied that plant leaves and soil were safe from the risk of heavy metals. Water irrigation causes heavy metal accumulation in soil and vegetables, with varying concentrations. The results of this study revealed no abnormal metal accumulation due to irrigation and no health risks to consumers.

Keywords: agricultural soil; pollution assessment; toxic elements; edible vegetables; soil-to-plant transfer factors

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1. Introduction

Safe plant-based foodstuff has played an important role in human life [1,2]. Edible plants, e.g., raw vegetables, are consumed prior to processing and represent the first link in the food chain [3]. Macro- and micrometals can be transferred directly from raw vegetables into the body, causing toxic effects if they exceed the allowable limits set by the World Health Organization (WHO) and the Food Administration Organization (FAO) [4]. Trace elements including heavy metals such as Pb, Cd, etc., and trace micronutrients such as Cu, Zn, Fe, Mn, Mo, and B (required for plant growth) are highly variable and influenced by both physiological and environmental factors. They are essential to the normal growth and health of plants, animals, and human beings at certain allowable levels [5,6]. Elements such as Cu, Zn, Pb, and Cd have been reported to cause contamination of soil, water, and plants [1,2].

In the modern era, trace elements, e.g., Ni, Zn, Cd, Cu, and Pb, have been reported as soil contaminants via agricultural fertilizers and pesticides, municipal waste, traffic, mining, and industrial emissions [7]. In both urban and agricultural areas, soil is a medium and a drain for the release of metals into the atmosphere. Some of these trace elements are permanent due to their immobile existence, whereas other metals are more mobile [8,9]. In soil, the heavy metal concentration plays an important role in controlling uptake of metal bioavailability in plants, as well as root uptake and the eventual transition to the food chain, causing a prospective hazard to human health [10]. A subset of plants known as hyperaccumulators can accumulate high levels of elements, e.g., Cu, As, and Cd, through multiple pathways [11]. The impact on the transfer of trace elements between plants and soil depends on the redox state and chemical form of the heavy elements, soil clay matter, iron and manganese oxide concentration, and climatic circumstances, as reported in [12]. The availability and mobility of elements are also considerably impacted by several factors, e.g., plant species, transpiration rate, soil pH, cation exchange capacity, organic matter, microorganisms, and other coexisting elements [13,14]. The occurrence of elements in surface and groundwater may be attributed to the dissolution of minerals in soil and aquifer materials [15] and/or to human activities and unsuitable disposal of industrial waste [16,17]. Consumption of contaminated food leads to uptake of toxic elements, which can disrupt mental development and disturb the function of organs such as the kidneys, lungs, and liver [15,18].

Saudi Arabia has experienced rapid agricultural development, resulting in an increase in organic and inorganic pollutants in agricultural soils. In North America and Europe, studies have shown that herbivores contain high concentrations of minerals due to their consumption of vegetation with a high concentration of minerals [19]. The toxic effects of trace elements have been studied in humans, animals, and plants [3,20]. The average concentrations of Zn, Cu, Fe, and Pb in well water in the Al-Bahah region in Saudi Arabia were found to be higher than the mean mineral concentration in irrigation water [21]. Edaphic factors and the distribution of diverse vegetation groups in Wadi Fatimah, Saudi Arabia, were found to be significantly correlated [22]. In central and western Saudi Arabia, significant increases in the levels of toxic elements were reported, which were attributed to agricultural activities, including the use of chemical and organic fertilizers and pesticides [23,24]. In the local market of Jeddah city, KSA, herbal plants were found to contain high levels of hazardous trace toxic elements, exceeding the maximum allowable levels set by the World Health Organization (WHO) [25,26] and indicating a need for precise monitoring of the levels of these toxic elements in agricultural areas that are vulnerable to the heavy use of agrochemicals. Total elemental concentrations in plants, soil, water, fertilizers, and pesticides can be precisely assessed by inductively coupled plasma–optical emission spectrometry (ICP–OES), as this technique provides good sensitivity, does not require large samples for measurements, and is an easy and rapid approach for conducting multiple automated core elemental analyses [6,26–29]. Hence, the assessment of Pb, Cr, Cu, Fe, Zn, and Ni uptake by plants is important to draw attention to the monitoring of the levels of these elements in agricultural areas that are vulnerable to the heavy use of agrochemicals.

The accumulation of potentially harmful elements (PHEs) in lettuce (*Lactuca sativa* L.) and coriander (*Coriandrum sativum* L.) irrigated with wastewater, as well as health risk assessment, was reported in a probabilistic meta-analysis of selected Ethiopian spices [30,31]. Trace determination of heavy metals in edible vegetables, soil-to-plant transfer factors, and toxic metal content in parsley (*Petroselinum crispum*), as well as associated health risks in vegetables, were reported for samples obtained from local farms in the Baz Kia Gorab region of western Iran [32–36].

Thus, the aims of this study were to (i) assess the levels of contamination with Pb, Cr, Cu, Fe, Zn, and Ni elements in cultivated soil, irrigation water, fertilizers, pesticides, and different tissues of selected plants coriander (*Coriandrum sativum*), dill (*Anethum graveolens*), parsley (*Petroselinum crispum*), and arugula (*Eruca sativa*), (ii) calculate the TF to assess the concentrations of metals in plants and soil; and, finally, (iii) explore the relationships and combined impacts of these parameters in plants, soil, water, fertilizers, and pesticides in order to properly address environmental risk in the study area through statistical treatment. Overall, this study provides decision-makers with an appropriate solution the level of heavy metals in plants representing the most important food crops in the world. The proposed solution can be helpful in reducing the risk of food chain contamination. The findings of this study can also help the government sector to formulate more stringent management procedures for the discharge of the elements and industrial activities.

2. Materials and Methods

2.1. Study Area and Sampling

The agriculture study area is located in a valley with a low rainfall regime approximately 100 km east of Jeddah city (Huda AL sham) in the Hijaz Mountains. The study area extends for about 70 km across the territory of the western coastal plains (Tihamah) and is surrounded by mountains with heights in the range of 0.0 to 500 m above the mean sea level [29]. The basin covers an area of approximately 4.860 km^2 at a longitude of 39°40′5′′ E and a latitude of 21°33′0′′ N, as demonstrated in Electronic Supplementary Information Figure S1.

Irrigated and wastewater samples were collected from the farm under study in pre-cleaned low-density polyethylene (LDPE) bottles with nitric acid (1% *v*/*v*). The water samples were immediately stored at 4 °C until analysis. The water pH, electrical conductivity (EC), and heavy metal concentrations were measured by ICP–OES as reported in [37].

Fertilizer and pesticide samples were taken from commercial products: urea (carbonic diamide 46% N) and NPK (complesal fluid, 8 + 8 + 6 + TE from Germany) fertilizers, as well as runner (methoxyfenozide 24%) and tiller (glyphosate IPA 48%) pesticides, used on the selected farm. Then, the samples were transported to the laboratory of King Abdulaziz University (KAU), Jeddah, Saudi Arabia, for processing and metal analysis.

Cultivated soil samples were collected from planting fields at a depth of 0.0 to 15 cm with the help of a garden shovel pre-cleaned with concentrated HNO_3. The soil samples were collected in plastic containers that had all been cleaned with detergent and tap water. The soil samples were air-dried, crushed, and passed through a sieve with a particle size of 0.40 mm. Then, soil samples were homogenized, transferred to clean polyethylene bags, and stored at room temperature for laboratory analysis. The pH, electrical conductivity (EC), and organic matter (OM) were measured according to soil analysis methods [4].

Coriander (*Coriandrum sativum*), parsley (*Petroselinum crispum*), arugula (*Eruca vesicaria*), and dill (*Anethum graveolens*) are the most widely grown vegetables in the study area. An approximate mass of 500 ± 0.1 g of each plant was collected from the farm at harvest (n = 40). In the lab, each individual vegetable sample was separated into roots, shoots, and leaves. The leaves of the subsamples were washed well with deionized water, dried, and ground to a fine powder using a stainless grinder. The samples were placed in labeled polyethylene bags and stored in a desiccator.

2.2. Recommended Wet Acid Digestion Procedures

2.2.1. Wet Digestion of Vegetable Samples

A total of n = 40 plant samples were irrigated with well water and collected at harvest. An accurate mass (0.500 ± 0.001 g) of each part of plant sample was digested in a 50 mL beaker containing 8 mL HNO_3 (69%, CDH). The beaker was covered with a watch glass and left overnight at room temperature, followed by the addition of 2 mL of H_2O_2 (30%, Sigma-Aldrich, Gillingham, England), and heated on a hot plate to 90–120 °C until the lightbrown-colored fume disappeared. The digest solutions were filtered through Whatman No. 50 Ashley filter paper and diluted to 50 mL using HNO_3 (0.1 M). The USEPA SW-846 (method 3050) vegetable digestion methodology was followed [38]. Then, the metal concentration was evaluated by inductively coupled plasma–optical emission spectrometry (ICP–OES).

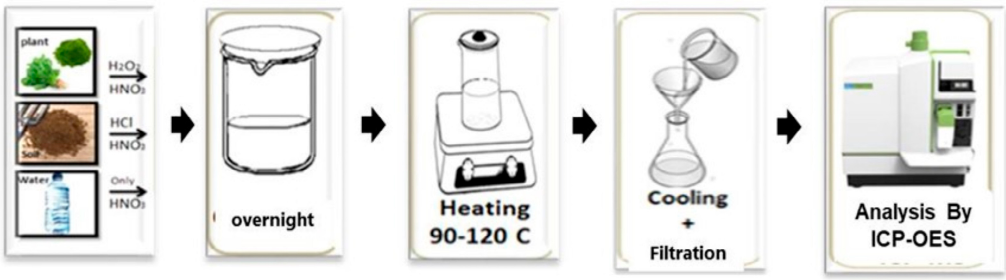
2.2.2. Wet Digestion of Soil, Fertilizer, and Pesticide Samples and Total Organic Carbon (TOC) Content

The soil samples were collected from the farm fields (n = 8), and fertilizer samples (n = 2) (0.500 ± 0.001 g) were placed into beakers (50 mL) containing 10 mL of aqua regia (65% HNO_3 and 37% HCl in a 3:1 ratio CDH). Pesticides (n = 2) were digested by adding 10 mL of 1:4 H_2SO_4 (95% m/v)/HNO_3 (69% m/v) mixtures. In the soil samples, the TOC content was determined as follows. Soil samples were first dried in an oven to remove the excess moisture and obtain the accurate mass (5 ± 0.01 g) of each soil sample. The samples were then placed into a crucible, and the weight of the soil crucible was recorded. The soil samples were heated in a muffle furnace (VULCAN, A-550) up to 300 °C for 5 h then cooled in a desiccator, and the TOC was calculated by employing Equation (1) [39]:

$$\text{TOC}\ \% = \frac{pre\text{-}ignition\ wight\ (\text{g}) - post\text{-}ignition\ wight\ (\text{g})}{pre - ignition\ wight\ (\text{g})} \times 100. \quad (1)$$

2.2.3. Digestion of Water Samples

Wet digestion of the water samples (n = 3) was performed as follows. An accurate volume (100 mL) of water sample was digested by adding 5 mL of concentrated HNO_3 (69%, CDH), followed by heating of the solution mixture to 90–120 °C until the solution became transparent. The solutions were then filtered and diluted to 50 mL with deionized water and analyzed as reported in [40]. The overall recommended procedures for preparation and ICP–OES analysis are demonstrated in Scheme 1.



Scheme 1. Preparation of the studied samples and their ICP–OES elemental analysis.

2.3. Recommended Sample Preparation and ICP–OES Measurements

The Pb, Ni, Zn, Fe, Cr, and Cu elements in the sample filtrates were measured using an inductively coupled plasma–optical emission spectroscope (ICP–OES, Optima 8300–PerkinElmer) according to the optimal parameters of each element. The parameters of ICP–OES were a wet plasma aerosol type of axial view, nebulizer startup instant condition, a flow rate (Ar) of 15 L/min, auxiliary flow (Ar) of 0.2 L/min, nebulizer flow (Ar) of 0.8 L/min, sample uptake rate of 1.5 mL/min, and sample flush time of 5 s. The wavelengths used for the observed elements were 217 nm for Pb, 231.6 nm for Ni, 206.2 for Zn, 259.94 nm for Fe, 283.56 nm for Cr, and 324.75 nm for Cu. Based on the IUPAC [41], the values of LOD and LOQ were calculated by employing the following Equations (2) and (3):

$$\text{LOD} = 3\sigma/b \quad (2)$$

$$\text{LOQ} = 10\sigma/b \quad (3)$$

where σ is the standard deviation of five replicate determination values of the blank under the optimized experimental conditions and b is the sensitivity factor, i.e., the slope of the linear calibration plot of the analyte. The percentage of relative standard deviation (%RSD) lies within the range 2.70 and 32.76%.

Digestion was performed on replicates on a routine basis to determine precision. A series of more diluted solutions was prepared from the stock of the standard solution (1000 mg/L, Appli Chem Panreac-ITW Companies) containing Pb, Cr, Cu, Fe, Zn, and Ni to afford working standard solutions of 1.0, 5.0, 10.0, 20.0, 50.0, 100.0, 200.0, and 300.0 mg/L for each metal, the absorbances of which were determined using ICP–OES to construct the calibration curve. The repeatability, expressed as relative standard deviation (RSD), ranged from 1.77% to 3.57% for replicate analyses of the calibration standard for all detected metals. To determine each metal's detection limit, data from replicate measurements of low-concentration samples were used. The detection limit was then derived from the metals' standard deviations. Metal standards were employed to calibrate the instrument for each element being analyzed. The maximum limit of Pb, Cr, Cu, Fe, Zn and Ni) in the samples was adopted as reported by the WHO [4].

2.4. Data Analysis

2.4.1. Estimation of the Soil and Water Transfer Factor to the Plant (TF)

The transfer factor is defined as the ratio of the metal concentration in plant tissue above the ground divided by the total metal concentration in the soil ($TF_{soil\text{-}plant}$) or water $TF_{water\text{-}plant}$ samples. The values of $TF_{soil\text{-}plant}$ and $TF_{water\text{-}plant}$ can be estimated for each element using Equations (4) and (5) [42]:

$$TF_{soil-plant} = C_p \text{ (mg kg–1 dry wt)}/C_s \text{ (mg kg–1 dry wt)}; \quad (4)$$

$$TF_{water-plant} = C_p \text{ (mg kg–1 dry wt)}/C_w\text{(mg kg–1 dry wt)}, \quad (5)$$

where C_p, C_w, and C_s are the elemental concentrations in plant, water, and soil samples, respectively.

2.4.2. Statistical Analysis

The mean, minimum, maximum, and standard deviation (±SD) of the analytical results were calculated using Microsoft Excel. The transfer factor (TF) of trace metals from soil to plants was computed using Equation (2). Data collected for all variables were first subjected to statistical analysis. Data on heavy metal concentrations in soil, water, fertilizers, and plants are presented as the means (±SD) for each sampling. Three-way analysis of variance (ANOVA) was performed on levels of trace element variations in the various water samples (well water, irrigated water, and wastewater) (block 1), fertilizers (urea and NPK) (block 2), and pesticides (tiller and runner) (block 3). The elemental uptake in the leaves of the four plants (coriander, parsley, arugula, and dill) was also critically determined as demonstrated in Figure S2. Three-way ANOVA interactions between the factors were considered significant at $p < 0.05$.

3. Results and Discussion

3.1. Physicochemical Characteristics of Water and Soil Samples

A preliminary study of the uptake of the tested trace elements in the various tissues of the four plants revealed dependence on water, soil pH, and conductivity [43]. Therefore, the physical characteristics (pH and conductivity) of the studied water and soil samples were a primary concern. In well water, irrigated water, and wastewater samples, the pH ranged between 6.53 and 7.73, which is within the permissible limit set by the FAO (pH 6–8.5) [44]. The conductivity of the well water and irrigated water samples was around 2860 µS/cm, which is within the recommended range reported by the FAO/WHO [4] (3100 µS/cm), whereas in the wastewater samples, high conductivity (4980 µS/cm) was noted, with higher concentrations of the labile (free) ions of the elements and fewer complexed metal ions and impurities.

The pH of soil samples Is a criti"al p'rameter of metal content. In normal agricultural soils with low pH (pH 5.0 to 7.0), the solubility of trace elements is generally high, as reported in [45]. Thus, low pH values measured in the soils in the current study account for the elemental transfer from soil to plants. The electrical conductivity (EC) of the soil is an important characteristic that can be used to determine nutrient availability and the presence of soluble salt in the soil. The EC values ranged from 410 to 1480 µS/cm; thus, the level of soluble salt increased, making it more difficult for plants to extract water from soil and resulting in water stress in plants [46]. Total organic carbon (TOC) is an important property of agricultural soil. The soil organic carbon content ranged from 6.30 to 10.60% in the agricultural soil samples, with organic matter affecting both the chemical and physical characters of the soil [46].

3.2. Levels of Trace Elements in the Studied Samples

The analyzed metals (Pb, Cr, Cu, Fe, Zn, and Ni) in the studied water samples were determined and compared according to the guidelines prescribed by the FAO/WHO [4] for water value. The average levels of metals in water samples in the study area are shown in Figure 1. The recorded values of Ni, Zn, and Cr metals were below the methodological detection limit in all water samples. The Pb concentration in well water was 1.171 mg/L, and the levels of Pb concentration in irrigation and wastewater samples were 0.01 mg/L, which is lower than the recommended levels set by international standard guidelines for irrigation water of the FAO (5.00 mg/L) [4]. The Fe levels in water samples ranged from 0.21 to 0.30 mg/L, which is two times greater than the recommended value (0.1 mg/L) set by FAO standards. The farm location may also contribute to the Fe level naturally occurring in the atmosphere and in soil that enters water bodies through natural processes and as a result of human activities. In Al-Kharj region, Saudi Arabia, and the Nangodi region of northern Ghana, the level of Fe in well water was found to be high compared to that determined in the present study area [47–49]. The concentration of Cu varied between 0.003 and 0.21 mg/L. The concentration of Cu was below the permissible limit according to the international standard guideline for irrigation water of FAO (0.02 mg/L) [4]. Furthermore, Cu content was found to be slightly higher in well water (0.2 mg/L) compared to the other water samples, in agreement with the data reported by Brar et al. [48].

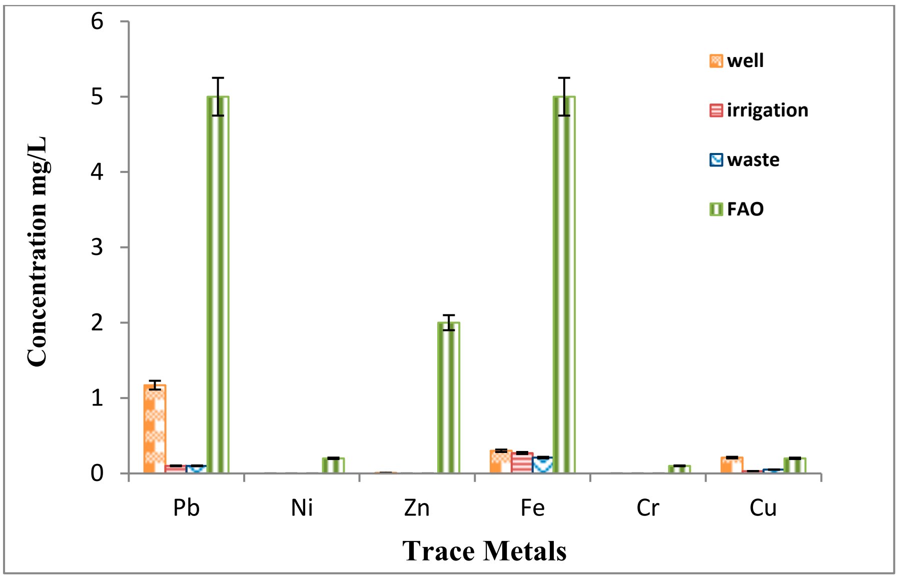


Figure 1. Distribution pattern of trace elements (Pb, Ni, Zn, Fe, Cr, and Cu) in the tested water samples.

In the modern era, soil pollution by trace toxic elements most likely arises from the constant use of element-enriched chemicals, fertilizers, and pesticide, as well as organic alterations, e.g., sewage sludge, wastewater, and other industrial activities [50]. Thus, the average levels (n = 3) of the toxic elements were critically studied in tiller and runner pesticides, the results of which are shown in Figure 2. The average concentrations of Pb, Ni, Cu, and Zn in tiller and runner were found to be in the ranges of 0.03–0.04, 0.0006–0.01, 0.004–0.02, and 0.20–0.28 mg/kg, respectively. In tiller, the average Fe level was not detected (ND), whereas in runner pesticides, the average Fe level was determined to be 0.77 mg/kg. In tiller, the Cr level was 0.03 mg/kg, whereas in runner, it was not detected. Thus, the average levels of the measured elements were lower than the levels in other pesticides (0.75–14.25 mg/kg) [51].

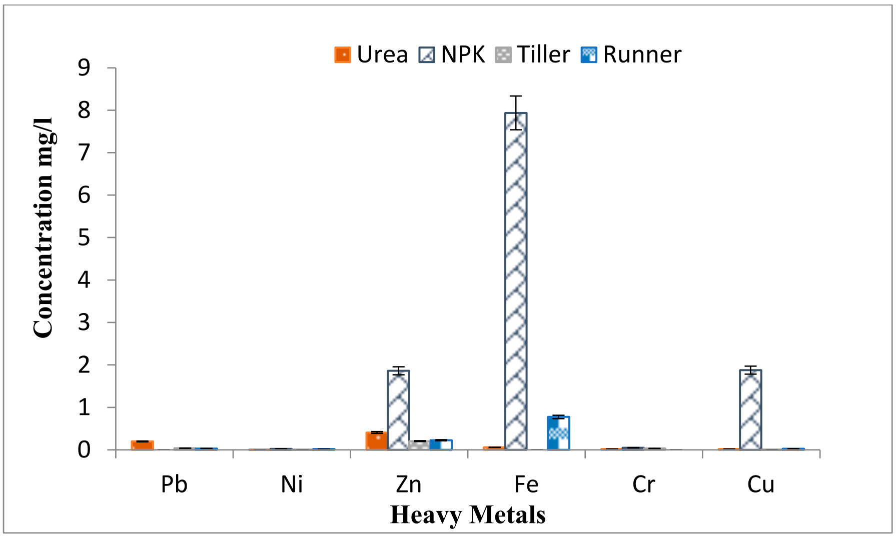


Figure 2. Average concentrations (mg/kg) of trace elements (Pb, Ni, Zn, Fe, Cr, and Cu) in fertilizers and pesticides.

The levels of Zn, Fe, Cu, and Ni in NPK fertilizer were 1.86, 7.39, 1.88, and 0.02 mg/kg, respectively. These values were found to be higher than those measured in urea for Zn (0.41), Fe (0.06), Cu (0.02), and Ni (0.003 mg/kg), whereas the average Ni level in phosphatic fertilizers in Saudi Arabia was in the range of 52.80 to 85.8 mg/kg [52]. The average levels of Pb and Cr contents were below the limit of detection in NPK and urea fertilizers. In urea, the average level of Fe in composite fertilizers was 347.3 mg/kg, revealing that the concentration of trace elements varied considerably depending on the measured element and fertilizer type. In superphosphate fertilizer samples, the average levels of the tested trace elements were substantially higher than those in urea fertilizer. Thus, it can be concluded that the average levels of trace elements in phosphorus fertilizers are above the levels found in other fertilizers. Superphosphate fertilizers resulting from phosphate ores contain a wide range of impurities, including trace elements [23,53]. However, according to Canadian standards, the average levels of trace elements in available urea and superphosphate fertilizers were within the allowable limits [54].

The average levels of trace elements in the agricultural soil in the study area are illustrated in Figure 3. The levels of the measured elements were below the maximum permissible limits set by the WHO/FAO [4]. In soils in which the arugula and dill samples were grown, the average levels of Pb were 33.10 and 37.18 mg/Kg—lower than the permissible limits set by the FAO/WHO [4]. In soils in which the coriander and parsley samples were grown, Pb was not detected, revealing that the Pb originated from other sources, such as leaching during the rainy season, and simplified by soil microbial activities [23]. Natural soil contains Pb, which may be discharged from natural sources, such as the decomposition of plants and animals. The impact of Pb toxicity depends upon its solubility, which is influenced by soil pH and other sources of Pb, including herbicides and insecticides [23]. In the agricultural soils in the current study, the Pb level was lower than the average Pb (1.20 mg/kg) in Al-Taif district, Saudi Arabia [55].

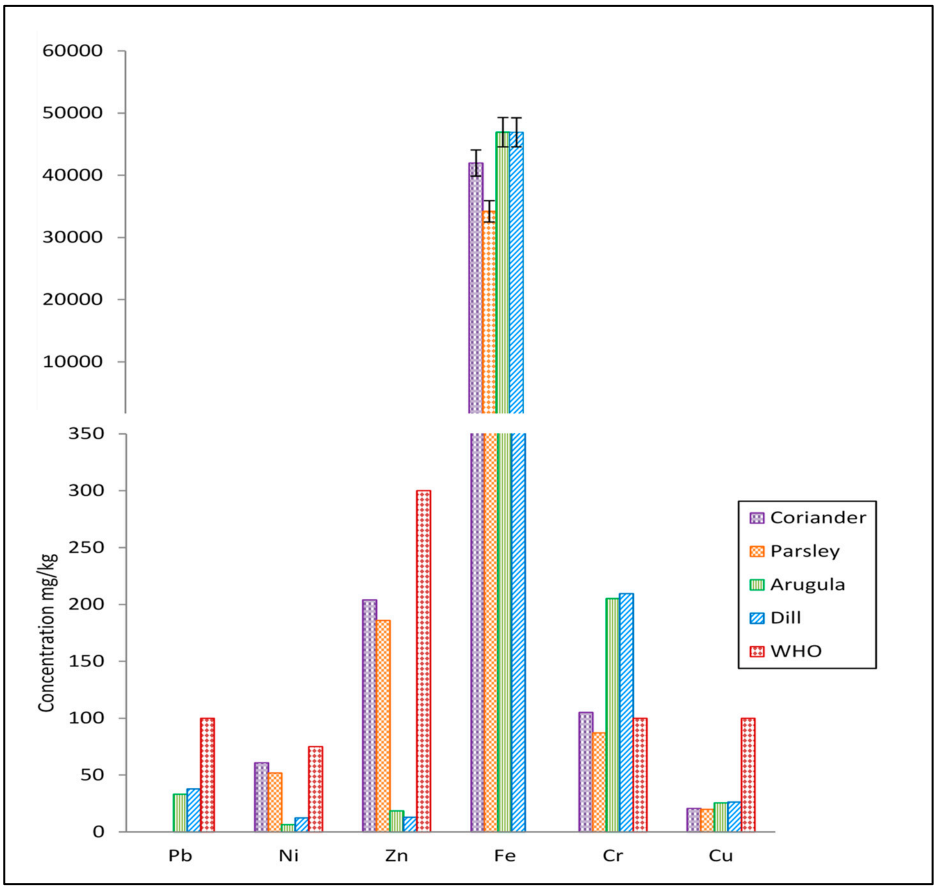


Figure 3. Average concentrations (mg/kg) of trace elements in cultivated soil.

The average Ni level in all locations varied from 6.40 to 60.80 mg/kg, which is less than the maximum permissible standard according to WHO/FAO (75 mg/kg). The average level of Ni in the study area was found to be lower than the Ni level (117.2 to 117.6 mg/kg) in India [48]. Ni levels can be ascribed to the dust and aerosols emitted during plant transportation and the use of organic fertilizers [17,56]. The average Zn level was highest in coriander soil (204.0 mg/kg), followed by parsley (185.90 mg/kg), arugula (18.60 mg/kg), and dill (12.90 mg/kg) soils (Figure 3). These results differ from those reported by Al-Hammad and El-Salam [47] for Zn in soils (38.45 to 174.52 mg/kg). This trend can most likely be attributed to soil conditions and other environmental factors, e.g., high soil acidity may also release the bound Zn pool, resulting in an increase in the level of labile Zn ions in the soil solution [7,23].

The average iron contents in the soil samples are illustrated in Figure 3. The average Fe content was very high (34,196.60 to 46,913.30 mg/kg). In soil samples in which arugula and dill were grown, Fe levels were found to be high, suggesting that the physicochemical conditions of the soil, e.g., redox potential, humidity, morphological conditions, flows, and iron fixation, are affected by different soil components [57]. These data agree with the results reported by Harmanescu et al. (55,489.20 mg/kg and 44,582.45 mg/kg) [17] and Ramteke et al. (18,328 mg/kg to 37,980 mg/kg) [7]. The average Cr level in soil samples is also shown in Figure 3. The average levels of Cr were in the range of 87.20 to 209.50 mg/kg in arugula and dill, which are above the maximum permissible limits set by the WHO/FAO for the Cr ion concentration in soil, with the exception of the soil in which parsley was grown. Additionally, the average Cr levels of soils in the present study were higher than the average level (60.43 mg/kg) in agricultural soil in Al-Kharj region, Saudi Arabia [47]. The Cr content in soil can be attributed to agricultural activities, including the use of chemical and organic fertilizers and pesticides and the high solubility of chromium (VI) species. Cu levels in all soil samples were found to be in the range of 19.90 to 26.30 mg/kg, as shown in Figure 3, which is within the national permissible levels set by the WHO. These values are lower than the Cu levels reported in in Al-Kharj region, Saudi Arabia (68.78 mg/kg), and in India (153 mg/kg) [7,47]. Thus, in the soil in the study area, the potentially toxic trace elements are mainly initiated from anthropogenic activity involving the use of agrochemicals, e.g., fertilizers, pesticides, and organic manure, in the condensed agricultural soils.

The average levels of toxic trace elements in crops are controlled by the contents of trace elements; the pathways of uptake of these elements by plant tissues, such as roots, leaves, and edible parts [58,59]; pH [60]; and the structure of heavy metals [61]. Trace element toxicity directly affects the physiology, structure, and growth of plants, as reported by Jorgensen et al. [62]. Table 1 demonstrates the levels of trace elements in leaves, shoots, and roots of the four vegetables analyzed in the study area. Vegetables can absorb elements from soil, which can be deposited on parts of vegetables exposed to the air in contaminated environments [63]. The average levels of the trace elements in different organs of leafy green plants (coriander, parsley, arugula, and dill) were successfully compared with the permissible levels set by the FAO/WHO [4], as summarized in Table 1. The Pb, Ni, Zn, Fe, Cr, and Cu levels in the leafy green plants were determined in the following order: roots > leaves > shoot. In parsley plants, the levels of Zn, Fe, and Cu were determined in the following order: leaves > roots > shoots. The average Pb levels were determined in the following order: shoots > roots > leaves. The mean experimental results indicate that Cr was not detected in parsley plants. In arugula plants, the Pb, Ni, Zn, and Fe concentrations were determined in the following order: leaves > roots > shoots.

Table 1. Average levels (mg/kg) of trace elements in selected leafy green plants.

Plant Sample	Part of Plant	Pb	Ni	Zn	Fe	Cr	Cu
Coriander	Leaves	3.05 ± 1.36	ND *	29.9 ± 3.2	157.80 ± 74.9	1.20 ± 2	9.20 ± 2.1
	Shoots	4.70 ± 0.70	0.23 ± 1.4	14.2 ± 3.7	57.90 ± 17.9	ND	7.40 ± 0.5
	Roots	5.70 ± 0.42	9.70 ± 10.1	38.6 ± 8.6	212.10 ± 45.1	4.40 ± 7.8	11.30 ± 0.7
Parsley	Leaves	5.50 ± 5.23	0.20 ± 0.2	70.9 ± 14.4	101.50 ± 13.2	ND	14.80 ± 1.4
	Shoots	12.50 ± 8.34	0.1 ± 0.1	48 ± 8.5	64.20 ± 10.05	ND	10.50 ± 1.1
	Roots	8.90 ± 4.38	0.03 ± 0.05	58.5 ± 11.8	57.80 ± 17.4	ND	13.40 ± 2
Arugula	Leaves	7.06 ± 1.97	4.40 ± 0.9	37.2 ± 11.3	157.40 ± 30.3	ND	ND
	Shoots	2.75 ± 1.62	2.60 ± 1.0	26.7 ± 5.6	60.06 ± 37.4	0.40 ± 0.7	ND
	Roots	6.85 ± 2.05	2.20 ± 0.2	45.5 ± 4.7	60.10 ± 11.2	ND	ND
Dill	Leaves	7.10 ± 0.14	12.03 ± 14.2	36.8 ± 6.4	158.30 ± 8.6	ND	0.70 ± 1.2
	Shoots	6.35 ± 0.07	3.80 ± 2.2	13.1 ± 0.9	13.90 ± 17.2	3.40 ± 6.1	1.30 ± 2.2
	Roots	4.75 ± 11.68	3.06 ± 0.2	26.9 ± 3.1	327.40 ± 24.5	ND	ND
WHO/FAO		0.3	67.9	99.4	425	0.5	73

* ND = not detected; average concentrations ± standard deviation.

In leaf tissues, Pb levels were higher for all leafy green plants, and the overall Pb uptake was determined in the following sequence: dill > arugula > coriander. Parsley recorded higher levels of Pb in the shoots than in the leaves. The Pb concentration ranged between 2.75 mg/kg in the shoots of arugula to 12.5 mg/kg in the shoots of parsley. It is well known that the leaf tissues of plants are capable of Pb uptake, mainly through the plant roots via passive absorption, and some specific proteins facilitate metal transport through movement across the membrane [60]. The leaf tissue root cell walls first bind metal ions from the soil; then, the metal ions are taken up across the plasma membrane. The difference in the binding sites and the protein structures in the various tissues and organs most likely account for the observed trend. The Pb levels in all the leafy green plants from the farm exceeded the maximum permissible limit set by the FAO/WHO [4] because of the age-dependent accumulation, defense, or tolerance mechanisms of the plants to avoid toxic levels in organs [64]. Muamar et al. [65] reported high Pb levels in leaves of parsley (16.67 mg/kg) in the Skhirat region of Morocco. The Pb level (0.08 mg/kg) reported in roots in the current study is lower than that reported in maize roots (2020 mg/Kg) [23]. Similar levels of Pb (0.54–6.98 mg/kg) in leafy green plant samples grown in industrial and urban cities in Tabouk, Riyadh, Damamm, and Jazan, Saudi Arabia were reported [66]. In leafy vegetables, the average Pb levels in arugula and spinach were found to range from 2.14 to 4.67 and from 1.26 to 4.14 mg/kg, respectively [66].

The average level of Ni in leafy green plant samples is presented in Table 1 (0.03 to 12.03 mg/kg). The highest Ni level was recorded in dill leaves (12.03 mg/kg), followed by coriander roots (9.70 mg/kg), with the lowest level (0.03 mg/kg) in roots of the parsley sample. These values were lower than the recommended limit set by the FAO/WHO [4]. The average level of nickel recorded in the current study was lower than the mean Ni concentrations (28.29 mg/kg) reported in India [67]. In leafy green plants, the Zn level was in the range of 13.1 to 70.9 mg/kg (Table 1), whereas the Zn level in shoot tissue was considerably less than that in leaf tissue and roots. In the leafy green plants, the Zn content was determined in the following order: leaves > roots > shoots. Therefore, we conclude that Zn was highly accumulated in harvestable parts of the plant [8,60]. Zn transport in plants is a complicated physiological process primarily controlled by metal chelators and Zn transporters [68,69]. The leafy green plants exhibited low Zn levels compared to the permissible limit set by the FAO/WHO [4], in agreement with the data reported by Abrham and Gholap [62]. The Zn level (25.86–126.30 mg/kg) in the Aseer region was higher than that in the study area in this work [16].

The average Fe levels in the four leafy green plant species are summarized in Table 1. In all plants, the levels of Fe ranged between 13.90 to 327.40 mg/kg, and the differences between root, shoot, and leaf Fe levels in the tested plants were highest among all elements measured in the present study. Fe levels in the leaves of dill (158.30 mg/kg), coriander (157.80 mg/kg), and arugula (157.40 mg/kg) were higher than those found in the leaves of parsley (101.50 mg/kg). Fe concentrations were higher in root tissue than those in shoot tissue, except in the shoot tissue of parsley. Fe levels did not surpass the maximum permissible limit set by the FAO/WHO in any of the leafy green plants [4]. The Fe level recorded in this study is similar to the significant mean values of Fe (17.01–22.94 mg/L) reported by Malede et al. [54] and Seyyed et al. [28] in landfill leachate, affecting the municipal solid waste composition.

Cr uptake is essential and must lie within a certain range of concentrations ($\leq$ 200 μg/day) for carbohydrate and lipid metabolism in human beings and animals [70]. In arugula shoots, a low level of Cr (0.40 mg/kg) was noticed, whereas the maximum level was recorded in coriander roots (4.40 mg/kg). The Cr levels in coriander leaves and the shoots of dill were 1.20 mg/kg and 3.40 mg/kg, respectively (Table 1), exceeding the allowable Cr limit reported by the FAO/WHO [4]. In parsley plants, Cr was not detected because a variety of contemporary methods are currently being used to remove heavy elements from soil by phytoremediation, phytodetoxification, soil rinsing, and leaching [19]. The Cu content in coriander varied between 7.40 and 11.30 mg/kg, with variation of 10.50 to 14.80 mg/kg in parsley, 0.70 to 1.30 mg/kg in dill, and no Cu detected in arugula (Table 1). In leafy green plant samples, the average Cu content was below the recommended permissible limit (73 mg/kg) set by the FAO/WHO. In this study, the average Cu level was lower than the reported value (32.45 mg/kg) in Gamo, Ethiopia [63]. According to the WHO and FAO, the values of Cu and Cr contents in vegetables should not exceed 30 mg/kg.

3.3. Transfer Factor (TF)

The transfer route and deposition of heavy elements from soil and irrigated water to the edible part of leafy green plants represent the main means of entrance of heavy elements to food chain [20]. Thus, in the current study, considerable attention has been oriented towards heavy element transferability from soil to leafy green plants and from irrigated water to leafy green plants. The transfer factors from soil to plants ($TF_{soil\text{-}plant}$) and from irrigated water to plants ($TF_{water\text{-}plant}$) were calculated using Equations (4) and (5) [20]. The results are summarized in Tables 2 and 3. As shown in Table 2, the $TF_{soil\text{-}plant}$ values, regardless of plant type, were 1.09. 0.27, 0.26, 0.08. 0.005, and 0.002 for Zn, Cu, Ni, Pb, Cr, and Fe, respectively. The mean $TF_{soil\text{-}plant}$ of heavy metals in soil–edible plant transfer decreased in the following order: Zn > Cu > Ni > Pb > Cr > Fe. In different leafy green plants, the $TF_{soil\text{-}plant}$ of the studied elements varied significantly, with Ni and Zn exhibiting maxima. Arugula and dill revealed higher TF from soil to plants than did the other leafy green plants. In leafy vegetables, the average level of trace element uptake was high compared to that in other vegetables, as leafy vegetables have a high transpiration rate to sustain their growth and the moisture content of the plant [71]. Absorption and accumulation also vary from species to species in the same category of leafy vegetable, e.g., spinach has higher levels of heavy elements (Fe, Zn, Mn, Cu, Pb, Cr, and Co) than lettuce [72]. The total TF decreased in the following order: Barkin Ladi (1.0) mg/kg > Jos South and Jos East (0.7) mg/kg > Bassa and Mangu (0.6) mg/kg [73]. Metal ions play a considerable role in plant growth, moving through various parts of the plant to distribute nutrients for various biological functions (photosynthesis, energy generation, metabolic reactions, and mitochondrial function). Thus, metal chelators such as phytochelatin, metallothionein, nicotianamine, and histidine play a vital role in the metal translocation mechanism, which helps in reducing the toxicity level of heavy elements in plants [74]. In terms of soil–crop transfer, Mingtao et al. reported the synergistic and antagonistic impacts of trace elements following Zn, Pb, and Cr adsorption from soil to crops [75]. Another study revealed that the TF values of Pb^{2+}, Fe^{2+}, and Cu^{2+} were higher than those of Ni^{2+} and Zn^{2+}, leading to the conclusion that Ni^{2+} and Zn^{2+} ions have a restrictive effect on internal transport in crops [69]. The amount of accumulation of Zn, Cu, and Fe in edible vegetables is contingent on the concentration of their water-soluble metal forms in the soil [19]. The $TF_{water–plant}$ summarized in Table 3 indicate that Fe has an effect on plants through transfer from water, which is an essential element found in most living things.

Table 2. Transfer factor (TF) of selected trace elements from soil to plants.

Plant Sample	Part of Plant	Pb	Ni	Zn	Fe	Cr	Cu
Coriander	Leaves	ND *	ND	0.15	0.004	0.01	0.44
	Shoots	ND	0.003	0.06	0.001	ND	0.35
	Roots	ND	0.15	0.18	0.005	0.04	0.54
Parsley	Leaves	ND	0.004	0.39	0.003	ND	0.74
	Shoots	ND	0.001	0.25	0.001	ND	0.52
	Roots	ND	0.0005	0.31	0.001	ND	0.67
Arugula	Leaves	0.22	0.69	2.00	0.003	ND	ND
	Shoots	0.08	0.40	1.43	0.001	0.001	ND
	Roots	0.20	0.34	2.44	0.001	ND	ND
Dill	Leaves	0.18	0.99	2.85	0.003	ND	0.03
	Shoots	0.16	0.31	1.01	0.0002	0.01	0.04
	Roots	0.12	0.25	2.08	0.006	ND	ND

* ND = not detected.

Table 3. Transfer factor ($TF_{water\text{-}plant}$) of selected trace elements from irrigation water to plants.

Plant Sample	Part of Plant	Pb	Ni	Zn	Fe	Cr	Cu
Coriander	Leaves	30.5	ND *	ND	584.44	ND	306.66
	Shoots	47	ND	ND	214.44	ND	246.66
	Root	57	ND	ND	785.55	ND	376.66
Parsley	Leaves	55	ND	ND	375.92	ND	493.33
	Shoots	30.5	ND	ND	237.77	ND	350
	Roots	89	ND	ND	214.07	ND	446.66
Arugula	Leaves	70.6	ND	ND	582.96	ND	ND
	Shoots	27.5	ND	ND	222.44	ND	ND
	Roots	68.5	ND	ND	222.59	ND	ND
Dill	Leaves	71	ND	ND	586.29	ND	23.33
	Shoots	63.5	ND	ND	51.48	ND	43.33
	Roots	47.5	ND	ND	1212.59	ND	ND

* ND = not detected.

3.4. Statistical Analysis

The results of our analysis of the correlation between the level of the trace elements based on the combined data of soil and plants ($TF_{soil\text{-}plant}$) and some other factors affecting metal uptake (three blocks (water, fertilizer, and pesticide samples), plants, and plant tissues) are summarized in Table S1, with $p < 0.05$ indicating a significant correlation between the element content and the suitability of leafy green plants for human consumption. The Zn, Fe, and Cu levels in the four vegetables in various plant tissues are displayed in Figure 4. Block 1 was found to have a more significant effect on the plant tissues of dill and arugula than blocks 2 and 3. The uptake of Zn was significantly higher in leaves than shoots in block 1 (water samples), as shown in Figure 5. Zn bioaccumulation varies in green leafy plants due to differences in the physiology, morphology, and anatomy of each plant [11]. To determine the probable relationship between Fe content and $TF_{soil\text{-}plant}$, correlations with three blocks were calculated. The results reported in Table S1 and Figure 4 reveal a positive correlation between block 1 and coriander plants. The correlation varies widely between the three blocks and the plant tissues. In the plant tissues, the binding sites are totally different from those in soil containing large amounts of humic, fluvic acids, and other phenolic compounds. The highest Fe content was found roots, whereas the lowest Fe content was found in shoots, as shown in Figure 5; however, in some cases, trace elements such as Co, Cr, and Fe were retained in the roots, and only a minor portion reached the shoots [76,77]. In soil and plants, the correlation depends on available forms of elemental ions and the particular conditions of plants. In soil, Fe absorption efficiency depends on the ability of roots to reduce Fe^{3+} to Fe^{2+} to secrete of mugineic acid, which changes the soil conditions and causes reduction [25].

The highest Cu levels of different vegetables are presented in Figure 4. The strong binding of Cu with the available organic matter and other soil colloids and the high mobility of Cu from soil to plants can most likely account for the observed trend. However, the interactive effects of the three blocks used in this study contributed effectively to Cu contents, leading to elevated Cu levels in parsley relative to arugula plants, as shown in Figure 5. The level of Pb was significantly higher in parsley than arugula plants Figure 5. On the other hand, a positive and significant correlation between heavy metal contents was observed, although Ni and Cr were positively but non-significantly correlated (Table S1). Our analysis of the correlation between the level of trace elements based on the combined data of TF and the three factors affecting metal uptake (block, part of plant, and plant) revealed non-significant correlations between element content and blocks 2 and 3, i.e., fertilizer and pesticide samples. The reported data further support the notion of multiple sources from agricultural activities accounting for significant differences.

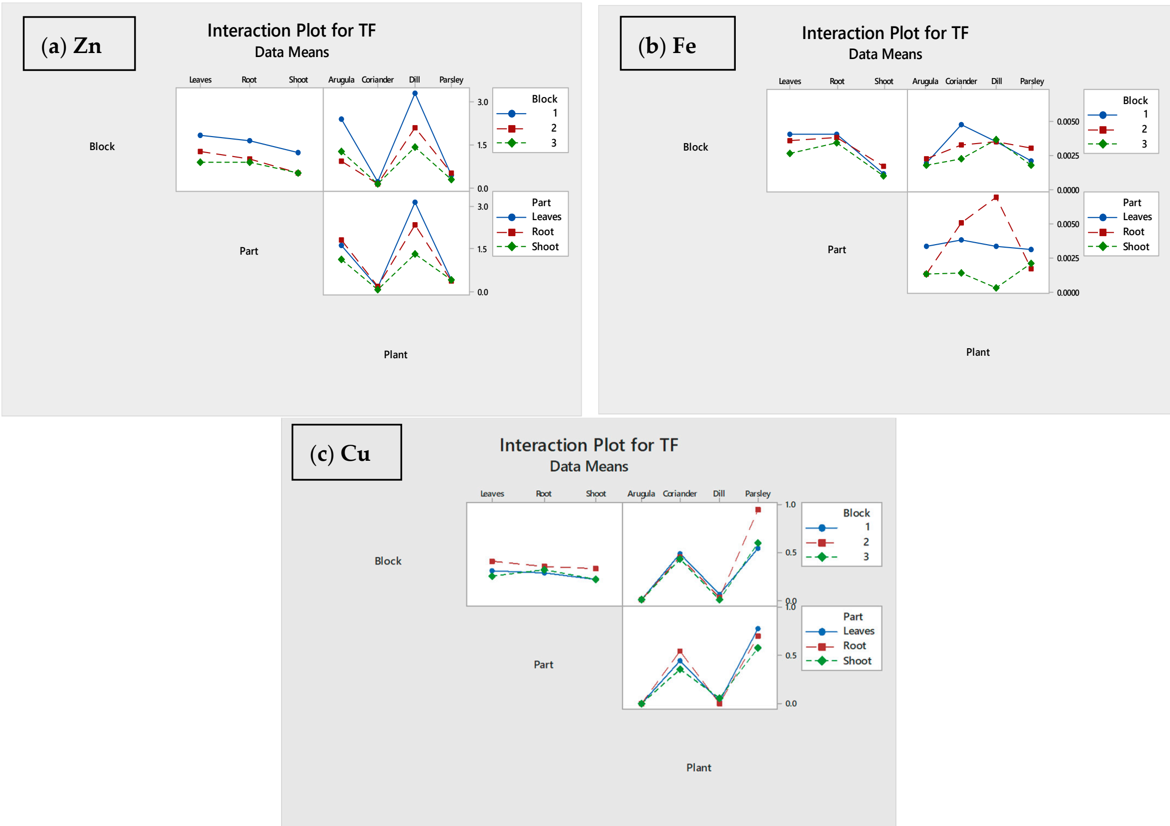


Figure 4. Interaction of trace element uptake (Zn (**a**), Fe (**b**), and Cu (**c**)) in various plant tissues.

Figure 5. Correlation significance of trace element levels for TF of the plants and various plant tissues.

4. Conclusions and Future Perspectives

In conclusion, the identification and quantification of sources of trace elements in soil and crops, as well as contamination status, are of considerable environmental and scientific importance. The outcomes of this study establish that the water and edible plants in the study area are suitable for public consumption, although regular checking for heavy metals in the study area is recommended. The correlation factors that influence the bioaccumulation of elements in green leafy plants were determined. The concentrations of trace elements in soil, water, pesticides, and fertilizers were found to be within the allowed limits set by the FAO/WHO. The soil and water resources in the study area were not contaminated, whereas certain vegetable species were found to contain trace elements in levels that exceeded the allowed limits. The most significant toxic elements additions constituting soil impurities were Zn, Fe, and Cu. $TF_{soil\text{-}plant}$ values were determined in the following order: Zn > Cu > Ni > Pb > Cr > Fe. The mean TF values, irrespective of plant type, were 1.09. 0.27, 0.26, 0.08. 0.005, and 0.002 for Zn, Cu, Ni, Pb, Cr, and Fe, respectively. Hence, Zn was the most bioavailable to plants because it can be transferred from soil to plants more easily than Cu, Ni, Pb, Cr, and Fe. The results reported in this study can provide benefits with respect to improving soil fertility, reducing waste, improving soil health, identifying slow-release sources of nutrients for plant growth, and reducing environmental impacts. Thus, systematic monitoring of the concentrations of potentially toxic trace metals in soil and green vegetables by public environmental agencies is necessary in order to verify operations and to protect the environment from hazardous pollutants. Finally, the study results indicate the need for a more current management program with respect to discharge into agricultural soil. In future work, we will use principal component analysis (PCA) for each of the matrices to be tested.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/pr11092515/s1, Figure S1. A map describing the study area and sampling sites, Figure S2. An agricultural experiment design for statistical treatment of data, Table S1. Mean element (Pb, Ni, Zn, Fe, Cu and Cr) concentrations based on the combined data of soil and plant.

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