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ORIGINAL PAPER

HEALTH RISK ASSESSMENT OF HEAVY METALS IN IRRIGATION WATER, SOIL AND VEGETABLES FROM DIFFERENT FARMS IN RIYADH DISTRICT, SAUDI ARABIA*

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Abstract

The safety and security of different foodstuffs are considered important and have a high priority in global sustainable development. The contents of Fe, Mn, Cu, Zn, Ni, Pb and Cd were estimated in edible parts of 60 vegetable samples, irrigation water and agricultural soil collected from 5 farms in suburban areas around Riyadh district, Saudi Arabia. Also, soil pollution indices, pollution load index and contamination factor (PLI, CF), bioaccumulation factor (BAF), non-carcinogenic and carcinogenic risk indices (HQ, THQ, and ILCR) were estimated. The content of heavy metals in irrigation water and soil was within the recommended permissible levels, except for Cd in soil, which exceeds permissible levels. The trend of metal concentrations in both agricultural soil and vegetables was found in the order: Fe > Mn > Zn > Cu > Ni > Pb > Cd. Cluster analysis of the metal content in vegetables showed a tendency towards the accumulation of heavy metals in the order of leaves > roots > fruits > flowers > tuber. The BAFs values found were < 1.0, HQs values for Fe, Mn, Cu, Zn and Ni were found to be < 1, while the estimated HQs for Pb and Cd were > 1, posing greater risk to the health of adults and children. THQ values were higher for children compared to adults. ILCR values for Cd showed a serious issue, as it exceeds the threshold risk limit ($>10^{-4}$) in all parts of the investigated vegetables in all sampling sites.

Keywords: heavy metal, vegetables, dietary intake, health risk, HQ, THQ, ILCR.

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INTRODUCTION

The consumption of vegetables grown in urban and suburban areas has increased extremely as a result of the progressive growth of the population's food demand (Olsson et al. 2016). Moreover, several studies have concluded that vegetables cultivated in urban and suburban regions accumulate higher levels of different chemical pollutants compared to those cultivated in rural regions (Christou et al. 2017). These studies state that rapid industrialization, heavy traffic and irrigation with treated wastewater play an important role in foodstuff contamination (Christou et al. 2017, Margenat et al. 2019). The consumption of polluted vegetables grown in suburban and urban regions has raised concern due to the considerable health implications they pose on human populations (Augustsson et al. 2018).

Heavy metal pollution has spread widely all over the world, causing environmental perturbation and serious human health hazards. The rapid urbanization, changes in land use and heavy industrialization in developing countries are main causes of this problem. Consequently, different types of environmental contaminants have increased leading to potential health risks and various negative impacts on foodstuffs' safety. Therefore, heavy metal pollution requires great attention, especially in association with public health (Cherfi et al. 2016, Rai 2018). Subsequently, the evaluation of heavy metals in agricultural crops has acquired great importance due to their share in the average human diet either consumed quantitively or as a nutritional value (Khillare et al. 2012).

The uptake of trace metals by vegetables can occur through roots, via the absorption process from soils containing metals, or by deposition on leafy surfaces (Al-Jassir et al. 2005). Many factors control the uptake of heavy metals through roots, such as the solubility of trace metals in soil, pH of the soil, the growth stage of the plant (Sharma et al. 2007). Moreover, during the transportation, marketing and storage process, crops may be more susceptible to contamination due to heavy metal deposition on the surface of crops from vehicular and industrial emissions (Al-Jassir et al. 2005). Thus, the soil/vegetable system provides a perfect example of abiotic-biotic interactions. Soil, a fundamental element of crop cultivation, can be widely contaminated with trace metals from point sources and non-point sources (Rai 2018).

The heavy metals that tend to accumulate in vegetables, such as copper, iron and zinc, are classified as micronutrients if found in low quantities. Nevertheless, they pose obvious health risks on humans as a result of prolonged intake and their accumulation (Sharma, Prasad 2009, Marschner 2012). On the other hand, several metalloids and metals, such as Hg, Pb, As, Pb and Cd, are known to be non-essential for various metabolic processes in the human body, and these elements are considered unhealthy in several respects (Khalid et al. 2017). Food security and safety and human health concerns encourage many researchers to investigate foodstuffs intensively in many countries, e.g. Bangladesh (Shaheen et al. 2016, Sultana et al. 2017), South Korea (Kwon et al. 2017), Pakistan (Khan et al. 2013, 2015*a*,*b*) and Hong Kong (Hu et al. 2013). The adverse effects of the accumulation of heavy metals in vegetables have drawn attention of many researchers in Saudi Arabia as well (Al-Farraj 2003, Adham et al. 2011, Ali, Al-Qahatani 2012, Adam et al. 2014, Assubaie 2015, Alsunni, Badar 2015, Alshaikh et al. 2018, Nassar et al. 2018, Faridi, Sulphey 2019).

The evaluation of dietary exposure to different chemical pollutants requires data on food consumption and contaminant occurrence, in addition to which comparisons are made with the corresponding health guidance values for each chemical of concern (WHO 2009). Either probabilistic or deterministic approaches can be used for a risk assessment. A single value used in the deterministic approaches, such as a mean or a percentage to illustrate the variables, has a main disadvantage, which is the lack of deep insight because such information only offers the knowledge of a possible exposure range and a proportion of the population at risk. In contrast, probabilistic approaches are used more broadly (Quijano et al. 2017) since they consider the variability of food consumption and the body weight of the consumer in addition to the contaminant occurrence variability. Thereby, they cover variability and uncertainty (Mondal, Polya 2008).

Traditional risk assessment indices include pollution load index (PLI), contamination factor (CF), bioaccumulation factor (BAF), non-carcinogenic and carcinogenic risk indices (HQ, THQ, and ILCR), postulating that the exposure effect of individual chemical pollutants was additive, and they have not had any effects either antagonistic or synergistic (Prosser, Sibley 2015). Therefore, using these indices may assist in obtaining a proper health risk assessment with relatively rapid and accurate results, which would be useful for decision making authorities.

This manuscript aimed to investigate the toxic metals Fe, Mn, Cu, Ni, Pb and Cd in irrigation water, soil and different vegetables grown in suburban areas around Riyadh district, KSA. This was performed by (a) measuring toxic heavy metal content in water and soil (b) measuring toxic heavy metals in different types of vegetables (c) estimating some soil pollution indices and (d) assessing human health risk indices of selected vegetables.

MATERIALS AND METHODS

Sampling sites description

Irrigation water, soil and the selected vegetable samples were collected from five farms located in Riyadh district namely: Al-Ammaria, Al-Kharj, Al-Uyaynah, Al Muzahimiyah and Al-Sahbaa cities (Table 1). The 5 farms

Table 1

Description of selected studied farms located around Riyadh District

Farm	City	Long & Lat.	Description
1	Al-Ammaria	24.8068, 46.4278	about 50 km north-west of Riyadh City
2	Al-Kharj	24.1690, 47.2941	about 100 km south-east of Riyadh City
3	Al-Uyaynah	24.7916, 46.3599	about 60 km north-west of Riyadh City
4	Al-Sahbaa	24.1965, 47.4004	about 110 km south-east of Riyadh City
5	Al Muzahimiyah	24.4231, 46.2024	about 60 km south-west of Riyadh City

irrigated their crops with underground water through deep wells found in the terminals of each farm. All farms are categorized by farming different vegetables, which are mainly consumed in Riyadh.

Sampling collection

The collected vegetables were chosen due to their high consumption in Riyadh district, 60 vegetables, 5 soil and 5 water samples were collected from the 5 farms. Vegetable samples were selected according to their edible parts, for example, root (radish and carrot), the tuber (potatoes and ginger), the leafy parts (lettuce, spinach, dill, leeks, basil plant, mint, arugula, green onion and coriander), the fruits (cucurbita, cucumber, lemon, pepper, and eggplant) and the flowers (broccoli and cauliflower) – Figure 1. The samples were collected during the harvesting season in May 2019 and each selected farm was divided to 4 sections. Soon afterwards, vegetable samples were collected from each quarter and then four specimens for each vegetable species were mixed forming a composite sample for each vegetable to ensure normal distribution. The samples were stored in a freezer following

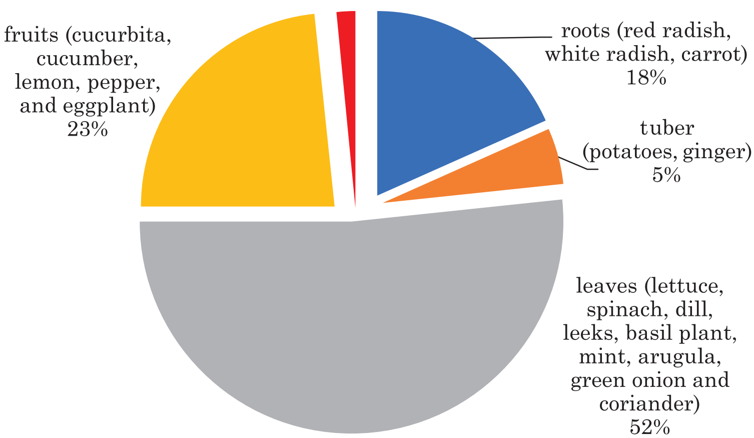

Fig. 1. Type, names, percentage of collected vegetables from Riyadh district

the commencement of the analysis. The same steps were followed for the collection of water and soil samples. After collecting water samples, they were kept in clean polyethylene bottles, acidified with 5 ml of concentrated HNO_3 and stored in a refrigerator. Whereas, samples of agricultural soil were taken from 10 cm beneath the subsurface, eliminating the non-soil parts, such as gravels, stones, wooden parts and any organic particles. The soil and vegetable samples were dried in an oven at 105°C overnight, after cooling they were ground using an electric mill, passed through 2 mm sieve and stored in clean labelled polyethylene bags (Lei et al. 2008)

Digestion of samples

For water samples, 500 ml of each water sample was digested by adding 20 ml of concentrated HNO_3 at 100°C to minimize the volume to a large extent; after that the samples were centrifuged at 10 000 rpm for 10 minutes, the final sample volumes were replenished to 100 ml using deionized water (APHA 2005). For soil analyses, 1 g of each well fine ground dried sample was digested using a mixture of HNO_3 (63%), $HClO_4$ (70%), H_2SO_4 (98%), 5:1:1 ratio at 300°C until the solution became transparent (US EPA 2006). Vegetable samples were digested by 1 g of the dried fine powder of each vegetable tissue, then 10 ml of HNO_3/HCl mixture was added at 95°C for 3 h (EPA 1994). All digested samples reached the final volume of 100 mL using deionized distilled water and centrifugation to eliminate any suspended particles. The samples were analyzed with Perkin Elmer inductively coupled Plasma model Optima 3000XL ICP-OES.

Soil-plant transfer contamination and risk assessment indices

Metal interactions with soil-plant and health risks indices could be used to illustrate the translocation of heavy metals between soil and plant systems also to determine the degree of risks associated with the dietary intake of vegetables (Yang et al. 2018).

Bioaccumulation factor (BAF) is important to describe the hazards of transferring heavy metals and other pollutants from soil to plant (Chang et al. 2014, Yang et al. 2018). It can be calculated using the following equation:

$$\text{BAF}=\frac{\text{concentration of metal in plant}}{\text{concentration of metal in soil}}.$$

Contamination factor (CF):

$$CF=\frac{C_t}{C_{Ref}}$$ (Antoniadis et al. 2019)

where: C_t – content of total metal in soil (mg kg^{-1}), C_{Ref} – reference concentration of the metal in pre-industrial soils. Reference values were Mn=488, Cu=38.9, Zn=70, Ni=29, Pb=27 and Cd=1 mg kg^{-1} (Kabata-Pendias 2011). With respect to CF, CF values are classified, based on the degree of contamination, as follows; low if CFT<1; moderate if 1≤CFT<3; considerable if 3≤CFT<6 and very high if CFT≥6 (Islam et al. 2015, Rinklebe et al. 2019).

Pollution load index (PLI) is a unitless index (integrated pollution index) used to estimate the level of pollution at a given site for given metals (Shaheen et al. 2019):

$$PLI = \sqrt[n]{Cf_1 \times Cf_2 \times Cf_3 \ldots \times Cf_n},$$

where: Cf_1, Cf_2, Cf_n – contamination factors for given metals 1, 2, …, n; if the value of PLI>1, that indicates significant pollution.

Human health risk assessment

Hazard quotient (HQ) for trace elements

The transfer of various kinds of pollutants from vegetables to humans is of a matter of the concern for scientists around the world. Therefore, many studies have been established using indices such as hazard index (HI), hazard quotient (HQ) and estimated daily intake (EDI) to assess the health risk (Zhuang et al. 2009). The hazard quotient (HQ), which reflects the non-carcinogenic risk level of the contaminants (Dziubanek et al. 2017), was calculated as follows:

$$HQ = \frac{EDI}{Rf_D},$$

where: Rf_D – oral reference dose for specific heavy metals (μg kg^{-1} bw/day). for Fe – 700, Mn – 140, Cu – 40, Zn – 300, Ni – 20, Pb – 3.5 and Cd – 1 (WHO 2018), which have not caused any harmful health effect, and EDI is the estimated daily intake (μg kg^{-1} bw/day), which was calculated as follows:

$$EDI = \frac{E_p \times E_D \times F_{1R} \times E_f \times C_m}{W_{AB} \times T_A},$$

where: E_F – exposure frequency (365 days/year), E_D – exposure duration, which is equivalent to the average lifetime, F_{IR} – fresh vegetable ingestion rate (the value of FIR for Saudi citizens were used according to Balkhair, Ashraf (2016)), C_f – conversion factor (0.085) for wet weight to dry weight, C_m – trace metal content in foodstuffs (mg kg^{-1} Dwt.), W_{AB} – average body weight (70 kg for adults and 24 kg for child), and T_A – average exposure time for non-carcinogens (equal to $E_F \times E_D$) (US EPA 1989).

If the value of HQ > 1, a consumer potential risk is indicated (IRIS 2018). Finally, THQ "the total hazard quotient" was calculated by the summation of all HQs for all the analyzed metals.

Incremental Lifetime Carcinogenic Risk (ILCR) implies the probability of developing cancer in consumers over a lifetime due to the exposure to a potential carcinogen. Acceptable risk levels for carcinogens range from 10^{-4} to 10^{-6} (the risk of developing cancer over a human lifetime is 1:10^5). The risk of cancer can be calculated using the equation:

$$CR = CSF \times EDI$$

where: CSF – carcinogenic slope factor as listed by (US EPA 2010). EDI – estimated daily intake of heavy metals.

Statistical analysis of the results obtained was carried out using Statistica 5 software for windows (StatSoft 1995) for cluster analysis and correlation coefficient.

RESULTS AND DISCUSSION

Heavy metals in irrigation water

The current results concerning the content of heavy metals in irrigation water of the selected farms in Riyadh district are given in Table 2. The content of selected heavy metals showed normal values within the recommended permissible levels by the FAO (1985) and the national guidelines provided in the Saudi Arabian environmental standards (MWE 2004). However, the irrigation water sample from farm 1 showed elevated values of heavy metals compared to the other farms. The irrigation water sample from farm 4 had the lowest heavy metal content. The high values from farm 1 sample may be due to some anthropogenic impacts, such as the heavy traffic in this region, in addition to industrial and domestic activities (Ali et al. 2012). The mean values of Pb and Cd (2.82 and 2.02 μg l^{-1}) in our study were higher than reported for the irrigation water of leafy plants cultivated in the Al-Kharj area, Saudi Arabia (2 and 1 μg l^{-1} for Pb and Cd, respectively), while the mean values of Fe, Mn, Zn and Cu (88.4, 15.7, 11.4 and 6.1 μg l^{-1}) are lower than the corresponding ones (436, 44, 157 and 38 μg l^{-1}, respectively) in the same region (Al-Hammad, El-Salam 2016). Moreover, Assubaie (2015) reports higher mean values of Fe, Mn, Zn, Cu, Pb and Cd (280, 120, 14, 14,

Table 2

Concentrations of heavy metals (μg l^{-1}) in irrigation water used in the selected farms compared with different guidelines

Farm	pH	Fe	Mn	Cu	Zn	Ni	Pb	Cd
1	7.25	215.6	26.50	12.34	22.15	8.69	4.46	2.20
2	7.47	25.60	7.50	2.20	5.60	2.17	1.36	nd
3	7.25	110.60	25.60	8.72	15.17	3.77	2.37	2.36
4	7.95	11.60	3.40	1.60	3.60	1.60	nd	nd
5	7.28	78.60	15.6	5.60	10.60	4.50	3.12	1.50
PL (MWE 2004)	6.0 - 8.4	5000	200	400	4000	200	100	20
PL (FAO 1985)	6.0 - 9.0	5000	200	400	4000	200	500	10

PL – Permissible limits, nd – non detected

Table 3

Concentrations of some heavy metals (µg g^{-1}) in the agricultural soil of the selected farms compared with the maximum permissible limits

Farm	Fe	Mn	Cu	Zn	Ni	Pb	Cd
1	465.8	39.7	24.69	43.65	21.6	10.6	5.2
2	257.6	48.6	14.69	26.5	13.5	9.65	3.6
3	336.5	51.9	20.6	36.5	6.48	7.89	8.9
4	415.6	62.6	25.62	45.6	23.05	15.63	9.5
5	412.6	55.4	22.23	38.17	26.4	18.6	9.07
PL (FAO/WHO 2001)	*	400	100	300	75	100	3

* no guideline

7 and 10 µg l^{-1}, respectively) in groundwater used in irrigation in Alahsa Oasis.

Heavy metals in agricultural soil

The concentration of heavy metals varied widely in the selected sites (Table 3). The results showed a decreasing trend of metal concentrations in agricultural soil, found in the order Fe > Mn > Zn > Cu > Ni > Pb >Cd. Iron reached the highest mean value (377.6 µg g^{-1}), with the maximum content found on farm 1 (465.8 µg g^{-1}), while its lowest value (257.6 µg g^{-1}) was recorded on farm 2. The mean value of Mn was 51.6 µg g^{-1} (ranging from 39.7 to 62.6 µg g^{-1}). Cu ranged between 14.69 and 24.69 µg g^{-1}, Zn – 26.5 to 45.6 µg g^{-1}, Ni – 6.48 to 26.4 µg g^{-1}, and Pb varied from 7.89 to 26.4 µg g^{-1}, which was all below the maximum permissible values defined by the FAO/ /WHO (2001). On the other hand, the mean value of Cd equal 7.25 µg g^{-1} (ranging 3.6 to 9.5 µg g^{-1}) was higher than the permissible threshold value (FAO/WHO 2001). Our current data are similar to those obtained by Mahmood and Malik (2014), who reported that the mean values of Mn, Zn, Cu, Ni and Pb in the agricultural soil in Lahore, Pakistan, were 39.0, 50.45, 28.8, 28.3 and 15.5 µg g^{-1}, respectively.

There is a significant positive correlation between the concentration of Fe in soil and the presence of Fe, Cu, Zn and Ni in irrigation water (r=0.55, 0.49, 0.48 and 0.66 at P<0.05), while a significant negative correlation was observed between Mn in soil and all metals in irrigation water at P<0.05 (Table 4).

Contamination indices of soil (CF and PLI)

The calculated values of CF and PLI for the selected farms were tabulated in Table 5. CF values for Mn, Cu, Zn, Ni and Pb are lower than one, indicating low agricultural soil contamination with those metals. However, the recorded values of Cd (3.6 and 5.2) indicate "moderate contamination"

Table 4

The Pearson's correlation coefficients between heavy metal content in irrigation water and in agricultural soil on the selected farms

Specification		Heavy metals in agricultural soil						
		Fe	Mn	Cu	Zn	Ni	Pb	Cd
Heavy metals in irrigation water	Fe	**0.55**	**-0.80**	0.35	0.34	0.01	-0.32	-0.21
	Mn	0.31	**-0.67**	0.17	0.15	-0.30	-0.44	-0.03
	Cu	**0.49**	**-0.74**	0.32	0.31	-0.12	-0.37	-0.11
	Zn	**0.48**	**-0.78**	0.29	0.28	-0.10	-0.38	-0.17
	Ni	**0.62**	**-0.79**	0.37	0.35	0.22	-0.14	-0.26
	Pb	0.44	**-0.78**	0.15	0.11	0.12	-0.13	-0.25
	Cd	0.40	**-0.51**	0.28	0.25	-0.21	-0.28	0.17

Bold values show the significant values.

Table 5

The contamination factor (CF) and pollution load index (PLI) of the agricultural soil of the selected farms

Farm	Contamination Factor (CF)						PLI
	Mn	Cu	Zn	Ni	Pb	Cd	
1	0.081	0.635	0.624	0.745	0.393	5.200	0.650
2	0.100	0.378	0.379	0.466	0.357	3.600	0.506
3	0.106	0.530	0.521	0.223	0.292	8.900	0.559
4	0.128	0.659	0.651	0.795	0.579	9.500	0.816
5	0.114	0.571	0.545	0.910	0.689	9.070	0.795
C_{ref} (mg kg^{-1})	488	38.9	70	29	27	1	

on farm 2 and 1, respectively, while a high contamination degree with cadmium was recorded for all the other farms with CF values > 6. Pollution load index (PLI), which demonstrates that all CF index values were lower than unity (< 1), indicates a significantly low heavy metal enrichment in the soil of the studied farms. The results were much lower than those obtained by Gabersek and Gosar (2018), who demonstrated that the soil of the industrial areas of Volos and Maribor was contaminated with multiple elements as a consequence of the intensive industrial activities in these regions.

Occurrence of heavy metals in vegetables

The mean content of the heavy metals studied in different edible parts of vegetables and fruit from the selected farms around Riyadh district are tabulated in Table 6, and results of a comparison between different values of heavy metals reported in the literature from various local and global areas of the world are found in Table 7. The concentrations of various metals are calculated on the dry weight basis.

Table 6

Range, mean concentration values and standard deviation of some heavy metals (μg g^{-1} dwt) in vegetables grown in the selected farms (n=60, P<0.05)

Farm	Plant		Fe	Mn	Cu	Zn	Ni	Pb	Cd
1	root (red & white radish)	range	82.9 : 89.19	7.9 : 10.6	4.31 : 4.6	7.56: 8.70	2.39 : 4.15	1.01: 1.62	0.69 : 0.81
		mean±SD	86.1 ±4.4	9.25± 1.9	4.5± 0.21	8.1±0.81	3.3±1.25	1.3±0.43	0.75± 0.085
	leaves (dill, leeks, spinach, purslane)	range	118.6 : 251.6	19.8: 25.4	7.07 : 12.89	12.36 : 23.72	3.65: 5.69	1.29: 2.15	0.63 : 1.04
		mean±SD	191.8± 35.8	22.9± 2.05	10.1±1.45	18.3±4.1	4.7±0.52	1.9±0.32	0.9± 0.16
2	root (carrot, red and white radish)	range	75.60: 115.6	8.9 : 13.6	4.15 : 6.49	8.07 : 12.65	1.18 : 2.98	0.74 : 1.66	0.42 : 0.89
		mean±SD	89.9± 10.33	11.1±2.33	5.5±1.19	10.6±2.3	2.4±0.97	1.0±0.47	0.55 ±0.24
	tuber (potato, ginger, onion)	range	58.60 : 91.5	6.60 : 11.30	3.18 : 4.67	7.16 : 10.65	1.08 : 2.65	0.68 : 0.98	0.31 : 0.56
		mean±SD	66.9±14.5	8.4±2.33	3.7±0.64	8.1±1.41	1.5±0.65	0.81±0.13	0.40±0.11
	leaves (dill, leeks, spinach, purslane, coriander, mint, Jew's mallow, rugula, lettuce)	range	48.60 : 189.40	6.20 : 36.90	3.49 : 10.69	8.78 : 20.65	1.78 : 5.17	0.69 : 2.25	0.42 : 1.09
		mean±SD	113.5±39.19	19.4±8.44	6.3±2.00	13.3±4.35	3.1±0.91	1.41±0.55	0.76±0.25
	fruit (eggplant, cucumber, lemon, peppers, cucurbit, okra)	range	55.60 : 169.6	6.50 : 18.60	3.17 : 9.69	6.32 : 17.69	1.09 : 4.56	0.57 : 1.87	0.33 : 0.91
		mean±SD	96.1± 23.5	11.6± 4.96	5.5± 2.31	10.1± 4.01	2.7± 1.15	0.95±0.49	0.53±0.21
	flowers (broccoli and cauliflower)	range	35.6 : 48.9	5.80 : 6.5	3.16 : 5.95	3.65 : 6.48	2.47 : 3.2	0.87 : 1.01	0.42 : 0.49
		mean±SD	42.6±9.40	6.2±0.49	4.1± 1.96	4.2±2.00	2.8±0.55	0.91±0.11	0.44±0.05
3	root (carrot, red radish)	range	143.60 : 168.90	31.30 : 35.60	9.15 : 10.60	14.56 : 16.78	4.57 : 5.64	2.78 : 3.54	1.35 : 1.72
		mean±SD	156.2±17.88	33.5±3.04	9.9±1.02	15.7±1.56	5.1±0.75	3.2±0.54	1.5± 0.26
	leaves (arugula, coriander, mint, green onion)	range	129.60 : 215.60	27.50 : 33.30	7.48 : 12.74	13.93 : 24.36	3.78 : 6.78	1.92 : 3.65	1.01 : 1.90
		mean±SD	165.3± 39.17	31.0±2.37	9.8± 2.62	18.3±4.83	4.6±1.24	2.5±0.67	1.27±.36
4	root (carrot and red radish)	range	198.30 : 234.30	41.30 : 46.40	12.25 : 13.69	21.60 : 23.01	7.88 : 9.18	4.96 : 5.78	2.41 : 2.75
		mean±SD	216.5± 25.45	43.90±3.60	13.00±1.02	22.30±1.01	8.50±0.91	5.40±0.57	2.58±0.24
	leaves (mint, parsley, coriander, basil plant)	range	218.60 : 389.60	37.70 : 61.30	12.69 : 21.60	22.41 : 36.58	7.88 : 10.65	4.35 : 6.74	1.19 : 3.50
		mean±SD	278.5±58.66	48.60± 11.6	16.10± 4.01	27.80±6.44	9.40±1.24	5.50±1.28	2.51±1.07
	fruit (cucurbit, cucumber)	range	215.60 : 225.36	43.80 : 45.60	12.36 : 15.47	26.68 : 22.16	6.98 : 8.01	3.06 : 4.39	1.26 : 1.98
		mean±SD	220.5±7.07	44.70±1.27	13.90±2.20	24.40±3.19	7.50±0.72	4.20±0.94	1.62± 0.51
5	root (carrot and red radish)	range	188.30 : 214.30	33.30 : 43.40	11.25 : 13.69	21.60 : 25.01	5.88 : 9.82	3.01 : 4.78	1.49 : 2.75
		mean±SD	205.2± 27.45	39.36±3.30	12.58±1.24	23.14±0.91	7.88±1.01	4.02±0.57	2.08±0.22
	leaves (mint, parsley, basil plant)	range	187.60 : 345.60	27.90 : 38.10	10.64 : 18.17	18.87 : 32.69	5.41 : 8.70	4.48 : 5.15	2.18 : 2.50
		mean±SD	215.6±53.87	34.15±5.42	15.6±3.79	28.7±7.09	7.15±1.67	4.98±0.34	2.31±0.16
Permissible limits (FAO/WHO 2001)			*	400	100	300	75	100	3

The content of heavy metals in the selected vegetables varied between 48.6 to 369.6 μg g^{-1} for iron, 5.8 to 61.3 μg g^{-1} for manganese, 3.16 - 21.6 μg g^{-1} for copper, 6.32 to 36.58 μg g^{-1} for zinc, 1.08 to 10.65 μg g^{-1} for nickel, 0.57 - 674 μg g^{-1} for lead and 0.31 to 3.5 μg g^{-1} for cadmium. Evidently, iron reached the highest values among the studied metals, with a decreasing concentration order of iron > manganese > zinc > copper > nickel > lead > cadmium (Table 6). Moreover, the content of heavy metals was greater in vege-

Table 7

Comparison between heavy metal content (mg kg^{-1} d wt.) in some vegetables from different regions with the current results

Plant	Current data	KSA	Pakistan	India	China	Spain
Radish	Fe 89 Cu 4.6 Zn 8.70 Pb 1.62 Cd 0.81	Fe 115 Cu 9.6 Zn 18.7 Pb 5.12 Cd 3.21	Cr > 0.18 Pb 0.91–3.96	Cu 5.96* Zn 22.5 Pb ND Cd ND Ni ND	Cu 0.34 Zn 2.48 Pb 0.07 Cd 0.012 Ni 0.07	-
Carrot	Fe 143 Cu 6.49 Zn 12.65 Pb 2.15 Cd 1.04	Fe 77.9 Cu 6.15 Zn 15.6 Pb 1.42 Cd 1.02	Cr > 0.18 Pb 0.91–3.96	-	Cu 0.23 Zn 1.59 Pb 0.23 Cd 0.023	-
Potato	Fe 75.0 Cu 4.15 Zn 8.69 Pb 1.59 Cd 0.98	Fe 69.6 Cu 6.08 Zn 14.15 Pb 6.19 Cd 0.99	Cu 0.1 Ni 0.83	Cu 0.94** Zn 4.28 Ni 0.49	Zn 3.77 Cu 1.03 Pb 0.067 Cd 0.015 Ni 0.054	
Onion	Fe 58.5 Cu 3.18 Zn 8.54 Pb 0.68 Cd 0.33	Fe 50.5 Cu 3.29 Zn 20.8 Pb 3.52 Cd 0.93	Cu 0.09 Ni 0.83	Cu 1.09** Zn 4.72 Ni 0.49	Ni 0.13 Pb 0.49 Cd 0.2	
Spinach	Fe 187.5 Cu 9.6 Zn 18.24 Pb 1.94 Cd 1.03	Fe 156.7 Cu 11.38 Zn 35.54 Pb 2.88 Cd 4.02	Pb 0.91–3.96 Cr > 0.18	Zn 10** Cu 0.09 Pb 3.1 Ni 3.2	Zn 0.16 : 0.53 Cu 0.16 : 0.85 Pb 0.02 : 0.013	
Lettuce	Fe 92.5 Cu 4.74 Zn 9.05 Pb 2.69 Cd 0.69	-	-	Cu 0.95** Zn 42.0 Pb 3.7 Cd 1.04	Zn 0.16 : 0.53 Cu 0.16 : 0.85 Pb 0.02 : 0.013	Cu 0.49 : 0.74* Zn 1.2 : 2.5 Pb 0.05 : 0.16 Cd 0.01 : 0.02
Cucumber	Fe 215.6 Cu 12.36 Zn 22.16 Pb 4.39 Cd 1.6	Fe 112.6 Cu 3.21 Zn 29.78 Pb 3.67 Cd 1.13	-	Cu 18.2*** Pb 1.3 Cd 1.4	-	Cu 0.046 ** Pb 0.005 Cd 0.006
Eggplant	Fe 148.8 Cu 7.63 Zn 12.86 Pb 1.52 Cd 0.78	-	Cu 2.93 Zn 50.7 Pb 4.57 Cd 0.69	Zn 7.25*** Cu 1.7 Pb 0.13	Zn 1.7 Cu 0.77 Pb 0.4	
Cauliflower	Fe 35.6 Cu 3.17 Zn 3.65 Pb 2.47 Cd 0.42	-		Cu 12.5*** Pb 6.1 Cd 1.4	Zn 5.45 Cu 0.6 Pb 0.03 Cd 0.014	Zn 4.60* Cu 0.61 Pb 0.004 Cd 0.004
Reference		Ali, Al-Qahtani (2012)	Khan et al. (2013) Parveen et al. (2003)	* Arora et al. (2008) ** Roychowdhury et al. (2003) *** Sonawane (2015)	Song et al. (2009) Zhou et al. (2016)	* Margenat et al. (2019) ** Marín et al. (2018)

tables grown on farm 5 followed by farm 4, while the lowest values were recorded on farm 1. On the other hand, both farm 2 and 3 have moderate heavy metal distribution in their crops. A cluster analysis was conducted according to the distribution of the heavy metals among the sampled farms (Figure 2). The cluster analysis exhibited that the distribution of heavy

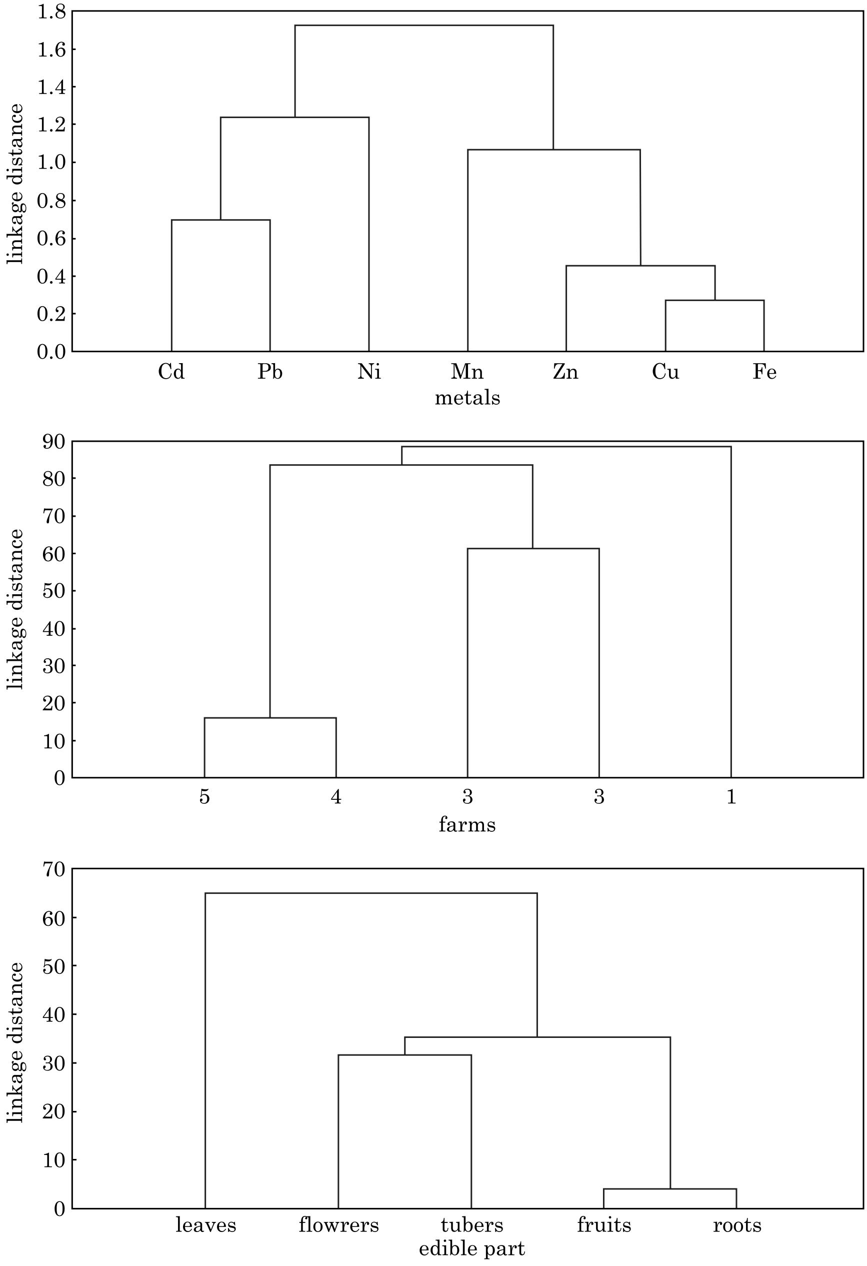

Fig. 2. Cluster analysis of heavy metal distribution, sampling sites and types of edible parts in vegetables from selected farms

metals was the highest on Farm 5. Then, the sampling sites were divided to three classes: farm 5 and farm 4 (Class I); farm 2 and farm 3 (Class II) and farm 1 (Class III), where Class III > Class II > Class I. These results are in agreement with a previous study performed in Al-Kharj, Saudi Arabia, and around Riyadh city, in which metals achieved their higher concentrations in urban and suburban regions than in rural areas (Ali, Al-Qhatani 2012).

Parsley leaves from farm 5 had the highest concentrations of Fe, Mn, Cu, and Zn (389.6, 61.3, 21.6 and 36.58 μg g^{-1}, respectively), and basil from the same farm had the highest content of Ni, Pb and Cd (10.56, 6.74 and 3.5 μg g^{-1}, respectively), the reasons being a high content of heavy metals in the irrigation water having and proximity of the farm to a highway. Broccoli flowers grown on farm 2 had the lowest concentrations of Fe, Mn, Co and Zn (35.6, 5.8, 3.16, 3.65 μg g^{-1}, respectively), whereas Ni, Pb and Cd fell the lowest in onion from the same farm (1.08, 0.68 and 0.31 μg g^{-1}, respectively). The results agree with those from other studies, either locally in the KSA or globally as mentioned in Table 7, which showed a relatively similar range of heavy metal concentration in vegetables grown in suburban and urban areas.

It is clear that tuberous plants (ginger, onion and potato) in this study tend to accumulate heavy metals less than other edible parts of vegetables (66.9 μg g^{-1} for Fe; 8.37 μg g^{-1} for Mn; 3.66 μg g^{-1} for Cu; 8.13 μg g^{-1} for Zn; 1.51 μg g^{-1} for Ni; 0.81 μg g^{-1} for Pb and 0.4 μg g^{-1} for Cd, on average), while the leafy vegetables presented the strongest tendency towards accumulating these elements among the edible plant parts studied (199.9 μg g^{-1} for Fe; 30.7 μg g^{-1} for Mn; 11.3 μg g^{-1} for Cu; 20.5 μg g^{-1} for Zn; 5.7 μg g^{-1} for Ni; 3.2 μg g^{-1} for Pb and 1.6 μg g^{-1} for Cd, on average). The general trend of heavy metals to accumulate in different edible parts of vegetables followed the decreasing order of leaves > roots > fruits > flowers > tuber. These findings were supported by a cluster analysis, which demonstrated three distinct classes based on heavy metal accumulation behaviour in different plant parts (Figure 2): class I which has the weakest accumulation tendency comprises tubers and flowers, class II shows a moderate accumulation tendency and comprises roots and fruits, and class III, which has the highest accumulation tendency (leafy parts).

Soil-plant metal transfer and health risk assessment indices

Some soil-plant indices such as metal transfer from soil to plants and health risks indices could be used to illustrate soil-plant metal translocation (uptake factors) and to determine the presence of dietary risks from vegetable or other crops to human health (Yang et al. 2018).

Bioaccumulation factor (BAF)

Bio-concentration or Bioaccumulation factors are defined as the ratio of a metal concentration in the edible parts of the vegetable to the concentration of the metal in soil. Furthermore, the BAF expressed well vegetables' ability to accumulate metals in different parts. A clear difference was recorded in the BAFs for the heavy metals in the different edible parts of vegetables (Figure 3).

The BAF values were less than 1.0 for all sampled vegetables and in all sampling sites, although their values are closer to unity in leaves of vegetables grown on farm 5 (Figure 3). Moreover, BAF values demonstrated a sim-

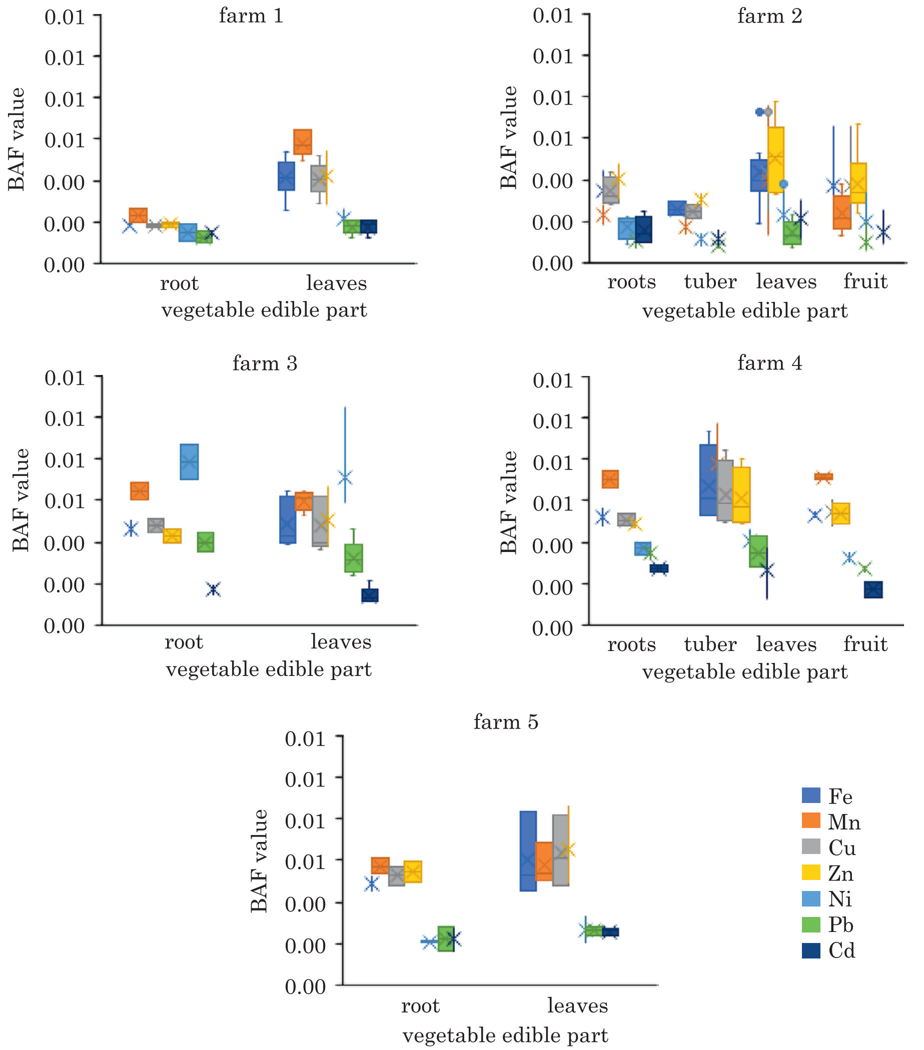

Fig. 3. Bioaccumulation factor of heavy metals in different edible parts of types of vegetables

ilar trend of heavy metals' distribution in vegetables, since it reached higher values on farms 5 and 4 than on the other farms. The BAF values indicated a considerable ability of leafy vegetables to accumulate metals more than other edible parts in addition to the tendency of Ni, Mn, and Fe to be accumulated more than Cu, Zn, Pb and Cd. The results are in agreement with several studies, which report that the highest BAF values are found in leafy vegetables, whereas horticultural crops and fruits score the minimal values (Liu et al. 2012, Chang et al. 2014, Zhou et al. 2016).

Health risk assessment indices (HQ and THQ)

Assessment of the health risk, either carcinogenic or non-carcinogenic, through the three ordinary exposure pathways: inhalation, ingestion and dermal contact, is a very important tool to identify the risk indices for decision-makers (Hu et al. 2016). The Estimated Daily Intake (EDI) of the investigated heavy metals indicate that Cd metal has the lowest daily intake in onions (0.20 and 0.27 μg day^{-1} for adults and children, respectively), whereas parsley maintained the highest EDI of iron (249.3 - 344.8 μg day^{-1} for adults and children, respectively). The EDI of Pb in our study showed alarming values in the range of 0.50 - 5.96 and 0.36 - 4.31 μg kg^{-1}.bw/day for children and adults, respectively. These values exceed the three values cited in the Benchmark Dose Lower Confidence Limits (BMDLs), which determine the side-effects of exposure to lead in humans: $BMDL_{10}$ 0.63 μg kg^{-1}.bw/day causes development of chronic kidney disease, $BMDL_{01}$ 1.5 μg kg^{-1}.bw/day has effects on systolic blood pressure and $BMDL_{01}$ 0.5 μg kg^{-1}.bw/day may causes neurotoxicity for fetuses and infants (WHO 1986).

The calculated values of the health risk assessment quotient (HQs) for the studied heavy metals using oral reference doses (R_{fD}) for adults and children are listed in table 8. It was determined that HQ values of Fe, Mn, Cu, Zn and Ni were < 1 for all sampled vegetables in all sampling sites for adults and children, respectively. Whereas, HQ values for Pb and Cd in several cases were > 1, especially on farm 3, 4 and 5 (Table 8). Therefore, they pose the greatest risk to children's and adults' health. These results are in agreement with those obtained in several other studies, which show that HQ values exceed unity, e.g. Cui et al. (2004), Singh et al. (2010), in experiments carried out in China and India.

The values of THQ were found in the ranges of 0.73 - 6.06 and 0.53 - 4.38 for children and adults, respectively (Table 8). Leaves of mint showed the highest THQ values (6.06 and 4.38 for children and adults, respectively), while the lowest values were recorded in onion (0.53 and 0.73 for children and adults, respectively). It was clear that the THQ values of the heavy metals studied were higher in children than in adults, indicating a real potential risk facing children. Therefore, future studies must be carried out to evaluate the risk associated with the consumption of vegetables.

Table 8

HQ, THQ and ILCR of heavy metals in vegetable samples for adults (70 kg) and children (24 kg)

Farm	Plant	HQ and THQ										ILCR		
		Ad. Chl.	Fe	Mn	Cu	Zn	Ni	Pb	Cd	THQ		Ni	Pb	Cd
1	root (red and white radish)	ad.	0.08	0.04	0.07	0.02	0.10	0.24	0.48	**1.03**		1.05E-03	3.79E-05	3.02E-03
		chl.	0.11	0.06	0.10	0.02	0.14	0.33	0.66	**1.43**				
	leaves (dill, leeks, spinach, purslane)	ad.	0.18	0.11	0.16	0.04	0.15	0.34	0.59	**1.56**		1.51E-03	5.40E-05	3.71E-03
		chl.	0.24	0.15	0.22	0.05	0.21	0.47	0.81	**2.16**				
2	root (carrot, red and white radish)	ad.	0.08	0.05	0.08	0.02	0.08	0.19	0.36	**0.86**		7.57E-04	2.96E-05	2.25E-03
		chl.	0.11	0.07	0.11	0.03	0.10	0.26	0.49	**1.18**				
	tuber (potato, ginger, onion)	ad.	0.06	0.04	0.06	0.02	0.05	0.15	0.26	**0.63**		4.84E-04	2.34E-05	1.61E-03
		chl.	0.08	0.05	0.08	0.02	0.07	0.21	0.35	**0.87**				
	leaves (dill, leeks, spinach, purslane, coriander, mint, Jew's mallow, arugula, lettuce)	ad.	0.10	0.09	0.10	0.03	0.10	0.26	0.49	**1.16**		1.00E-03	4.05E-05	3.06E-03
		chl.	0.14	0.12	0.14	0.04	0.14	0.36	0.67	**1.61**				
	fruit (eggplant, cucumber, lemon, peppers, cucurbit, okra)	ad.	0.09	0.06	0.09	0.02	0.09	0.18	0.34	**0.86**		8.76E-04	2.76E-05	2.12E-03
		chl.	0.13	0.08	0.13	0.03	0.12	0.24	0.46	**1.19**				
	flowers (broccoli and cauliflower)	ad.	0.04	0.03	0.05	0.01	0.08	0.16	0.27	**0.64**		7.90E-04	2.51E-05	1.69E-03
		chl.	0.06	0.04	0.07	0.02	0.11	0.22	0.37	**0.89**				
3	root (carrot, red radish)	ad.	0.14	0.15	0.16	0.03	0.16	0.58	0.98	**2.21**		1.63E-03	9.10E-05	6.19E-03
		chl.	0.20	0.21	0.22	0.05	0.23	0.80	1.36	**3.06**				
	leaves (arugula, coriander, mint, green onion)	ad.	0.15	0.14	0.16	0.04	0.15	0.46	0.81	**1.91**		1.48E-03	7.30E-05	5.13E-03
		chl.	0.21	0.20	0.22	0.05	0.20	0.64	1.13	**2.65**				
4	root (carrot and red radish)	ad.	0.20	0.20	0.21	0.05	0.27	0.98	1.65	**3.56**		2.73E-03	1.55E-04	1.04E-02
		chl.	0.27	0.28	0.29	0.07	0.38	1.36	2.28	**4.92**				
	leaves (mint, parsley, coriander, basil plant)	ad.	0.25	0.22	0.26	0.06	0.30	1.00	1.61	**3.71**		3.01E-03	1.57E-04	1.02E-02
		chl.	0.35	0.31	0.36	0.08	0.42	1.38	2.23	**5.13**				
	fruit (cucurbit, cucumber)	ad.	0.20	0.20	0.22	0.05	0.24	0.77	1.04	**2.73**		2.40E-03	1.22E-04	6.53E-03
		chl.	0.28	0.28	0.31	0.07	0.33	1.07	1.43	**3.77**				
5	root (carrot and red radish)	ad.	0.20	0.15	0.20	0.05	0.18	0.55	0.95	**2.29**		1.81E-03	8.67E-05	6.01E-03
		chl.	0.27	0.21	0.28	0.07	0.25	0.76	1.32	**3.16**				
	leaves (mint, parsley, basil plant)	ad.	0.23	0.15	0.23	0.05	0.22	0.89	1.51	**3.27**		2.21E-03	1.39E-04	9.49E-03
		Chl.	0.32	0.20	0.31	0.07	0.31	1.22	2.08	**4.52**				
R_{fD} µg/g			**700**	**140**	**40**	**300**	**20**	**3.5**	**1**		**CFS**	**0.5**	**0.042**	**6.3**

Ad. – adults, Chl. – child

Incremental lifetime cancer risk (ILCR)

The carcinogenic slope factors for Fe, Mn, Cu and Zn are unavailable, so the index of carcinogenic hazard (CR) was only calculated for Ni, Pb and Cd (Table 8). The values of CR were found in the range of 4.84×10^{-04} to 3.01×10^{-03} for Ni, 2.34×10^{-05} to 1.57×10^{-04} for Pb and 1.61×10^{-03} to 1.04×10^{-02} for Cd.

US-EPA (2010) reported the safe limit for ILCR ranging between $>10^{-4}$ (threshold risk limit) to $<10^{-6}$ about 1: 1 000 000 lifetime exposure. The results of ILCR for Cd showed a serious issue, as it exceeds the threshold risk limit ($>10^{-4}$) in all parts of the investigated vegetables in all sampling sites. Also, the ILCR values for Ni were alarming, especially on farm 4 and 5. The general trend of the risk factor of cancer development in different parts of the vegetables studied was found in the order: leaves > roots > fruits > flower > tuber. Similar results were obtained by SULTANA et al. (2017) in their study on the health risk assessment for carcinogenic exposure to heavy metals in vegetables and fruits from Bangladesh. They found that the risk trend for developing cancer due to the consumption of the vegetables was in the order of leaves > roots > fruits.

CONCLUSION

The recent status of the distribution of heavy metals (Fe, Mn, Cu, Zn, Ni, Pb and Cd) in irrigation water, agricultural soil and grown vegetables in some farmlands was analyzed in suburban areas around Riyadh district, Saudi Arabia. In addition to the estimation of some pollution indices, bioaccumulation factor (BAF), non-carcinogenic and carcinogenic risk indices were calculated. Mean values of the heavy metals in irrigation water and soil showed normal distribution values within the recommended permissible levels, except for Cd in soil, which exceed the permissible threshold. The trend of metal concentrations in both agricultural soil and selected vegetables was found in the order Fe > Mn > Zn > Cu > Ni > Pb > Cd. The concentrations of the heavy metals in different parts of vegetables were found within the permissible limits, except for Cd, which reached values close to the upper permissible limits. Heavy metals tend to accumulate in the order of leaves > roots > fruits > flowers > tuber. The BAF values were less than 1.0 for all sampled vegetables in all sampling sites, The BAF values indicate that leafy vegetables have the greatest ability to accumulate metals more than other edible parts. On the other hand, HQ values for Fe, Mn, Cu, Zn and Ni were estimated to be < 1, while Pb and Cd values were >1, posing the greatest risk to children's and adults' health. THQ values were higher for children than for adults indicating a real potential risk facing children. ILCR values for Cd implied a serious issue, as they exceed the threshold risk limit ($>10^{-4}$) in all edible parts of the investigated vegetables in all sampling sites. Therefore, further studies must be conducted to evaluate the risk associated with the consumption of vegetables

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