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Health Risks Assessment Diagnosis of Toxic Chemicals (Heavy Metals) via Food Crops Consumption Irrigated with Wastewater

(Diagnosis Penilaian Risiko Kesihatan Bahan Kimia Toksik (Logam Berat) melalui Penggunaan Tanaman Makanan yang Diairkan dengan Air Buangan)

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ABSTRACT

*The present study investigated the concentration of metals in commonly grown vegetables (*Luffa acutangula *L.,* Zea mays *L.,* Solanum melongena *L.) irrigated with waste water in District Bannu, Khyber Pakhtunkhwa, Pakistan. The pH (5.80) and electrical conductivity (13 dS/m) of waste water indicated the acidic nature that is not suitable for irrigation purposes. Soil and vegetables samples were analyzed for metals concentration through flame atomic absorption spectrometry (Varian FAAS-240). The findings showed that waste water irrigated soil was highly contaminated with Cd (4.62 mg/kg) which was above permissible limits set by European Union Standard (EU 2006, 2002). The concentrations of heavy metals such as Cr and Cd in vegetables were higher than the permissible limits set by World Health Organization/Food and Agriculture Organization U.S.A guidelines 2001. The health hazard quotient (HQ) of waste water irrigated vegetables was observed higher for Ni (0.699-0.1029 mg/kg), (0.0456-0.1040 mg/kg), (0.731-0.0994 mg/kg) in* Luffa acutangula, Solanum melongena *and* Zea mays, *respectively. The study concluded that the consumption of commonly grown vegetables in waste water zone of the study area may pose potential health threats in local population.*

Keywords: Commonly grown vegetables; health risks; heavy metals; waste water

ABSTRAK

*Penyelidikan ini mengkaji kepekatan logam dalam sayur-sayuran yang biasa ditanam (*Luffa acutangula *L.,* Zea mays *L.,* Solanum melongena *L.) diairkan dengan air buangan di Daerah Bannu, Khyber Pakhtunkhwa, Pakistan. PH (5.80) dan pengaliran elektrik (13 dS/m) air buangan menunjukkan sifat asid yang tidak sesuai untuk tujuan pengairan. Sampel tanah dan sayur-sayuran telah dianalisis untuk kepekatan logam melalui spektroskopi serapan atom api (Varian FAAS-240). Keputusan kajian menunjukkan bahawa tanah yang diairkan dengan air buangan adalah sangat tercemar dengan Cd (4.62 mg/kg) melebihi had yang dibenarkan yang ditetapkan oleh piawai Kesatuan Eropah (EU 2006, 2002). Kepekatan logam berat seperti Cr dan Cd dalam sayur-sayuran adalah lebih tinggi daripada had yang dibenarkan yang ditetapkan oleh Pertubuhan Kesihatan Dunia/Organisasi Makanan dan Pertanian Amerika Syarikat garisan panduan 2001. Darjah bahaya kesihatan (HQ) daripada sayur-sayuran yang diairkan dengan air buangan diperhatikan lebih tinggi nilai Ni (0.699-0.1029 mg/kg), (0.0456-0.1040 mg/kg), (0.731-0.0994 mg /kg) masing-masing dalam* Luffa acutangula, Solanum melongena *dan* Zea mays. *Kajian ini menyimpulkan bahawa pengambilan sayur-sayuran yang biasa ditanam di zon air buangan dari kawasan kajian berpotensi menimbulkan ancaman kesihatan kepada penduduk tempatan.*

Kata kunci: Air buangan; logam berat; sayur-sayuran yang biasa ditanam; risiko kesihatan

INTRODUCTION

The industrial revolution and suburbanization of 20th century has accelerated the waste water discharge which not only contains toxic chemicals but is also a rich source of organic matter and nutrients. Such waste waters are being used for irrigation purposes (Liu et al. 2011). A total of 20 million ha of land are irrigated with waste water throughout the world and the food crops contaminated with these waste waters are consumed by 10% of the world's population (Hamilton et al. 2007; WHO 2006). In poor countries the waste water is extensively used for food crops cultivation without any prior treatment, which is not only harmful for the crops production but it is also an illegal agricultural practice (Huibers et al. 2004). The waste water practices for irrigation have adverse effects on food crops and pose serious health risks by the consumption of such contaminated food crops (Hua et al. 2013). The edible parts of the food crops accumulate the toxic metals which have adverse effects on their consumers (Arora et al. 2008). The food crops grown on contaminated soil with waste water practice have risky effects on human health by their consumption (Gosh et al. 2012; Khan et al. 2008; Ullah et al. 2011).

Heavy metals such as cadmium, lead, chromium, nickel, arsenic and mercury are highly toxic due to their insistent and bio accumulation nature. The continuing exposure to such toxic metals in the food crops has harmful effects on human health and causes bone, heart, brain and kidney diseases from their consumption. Furthermore, cadmium and mercury have cancer causing effects (Hussain et al. 2001; Jarup 2003). Accumulation of a high concentration of toxic metals causes health risks in plants, animals and humans and there is no medical method through which these metals can be removed from their body (Bhuiyan et al. 2011). Therefore, it is necessary to assess and quantify the health consequences of the exposure to such a contaminated environment (Khan et al. 2015a; Ma et al. 2006).

The present investigation aims to quantify the concentration of various metals in the soil and commonly grown vegetables irrigated with waste water, to calculate daily intake rate of metals (DIM) by intake of particular vegetables via the local human population and to evaluate the human health hazards related to heavy metals through the consumption of vegetables in District Bannu Khyber Pakhtunkhawa, Pakistan.

MATERIALS AND METHODS

INTRODUCTION TO STUDY AREA

The study areas for research work were selected in District Bannu. The District Bannu lies at 32.9889°N, 70.6056°E Latitude. The waste water irrigated zone was located at Hinjal, Meta Khel and the entire Mandan area. These areas are irrigated from the stream known as Mandan Vial. The Kachkot stream which comes from the Baran reservoir (Baran Dam) gives a stream at 15500 R.D (Running Distance) known as Mandan feeder which irrigate Fatmakhel and Isaki Villages of District Bannu. The Kachkot stream at 11100 R.D gives off three Branches of stream known as Vial Sadat, Vial Landi and Vial Mandan. The entire length of the Vial Mandan is three miles which irrigates Hinjal, Meta Khel and Mandan villages of the District Bannu. All the drainage of associated areas through which the Vial Mandan stream flows runs in to the Mandan vial stream. Along with the drainage system of the related areas, the city drainage and many small industries in the city release their effluents directly into the Vial Mandan stream without proper treatment.

COLLECTION OF WATER SAMPLES AND THEIR ANALYSES

One litter of waste water samples were taken from different localities of the study area and were analyzed for pH and electric conductivity (EC). The pH of water samples was analyzed by immerging pH electrode into the water sample. The EC of sample water was determined by electrical conductivity meter (model Mc-1).

COLLECTION OF SOIL SAMPLES

The soil samples (20 cm depth) were collected randomly from various localities of the waste water irrigated zone. The 500 g soil sample was placed in polythene bag, dried in an oven and ground to a fine powder which was passed through a 2-mm mesh sieve.

SOIL SAMPLES DIGESTION AND THEIR ELEMENTAL ANALYSES

The 1 g of soil sample was digested in aqua regia (HCl: HNO_3) with 3:1 ratio. The solution was heated to 200°C until the cessation of brown fumes from the solution. The solution was filtered through Whatman filter paper no 42. The solution was diluted with deionized distilled water to raise the volume of the solution up to 50 mL (USEPA method: 3005A). The prepared samples were analyzed by flame atomic absorption spectrometry (Varian FAAS-240).

COLLECTION OF VEGETABLE SAMPLES

Solanum melongena L. (brinjal), *Luffa acutangula* L. (gourd) and *Zea mays* L. (maize) were collected in late summer of September, 2015 from waste water irrigated zone of study area. The whole plant including their roots were collected and wrapped in blotting paper. The fruits of the collected vegetables were dried in an oven at 75°C for 3 days and grind into a fine powder by using an electric grinder.

VEGETABLES SAMPLE DIGESTION

The 0.5 g fine powder of the vegetables samples was digested in the tri-acid mixture of HNO_3, H_2SO_4 and $HClO_4$ with 5:1:1 ratio, respectively. The solution was heated to 80°C until clear. The solution was filtered through Whatman filter paper no 42 and diluted with deionized water to raise the volume of the solution up to 50 mL (USEPA method: 3005A).

METALS ANALYSIS

The prepared samples were analyzed for concentration of metals through flame atomic absorption spectrometry (Varian FAAS-240).

DETERMINATION OF TRANSLOCATION FACTOR

The metals are transferred from the contaminated soil to the edible parts of the vegetables. The transformations of metals from the contaminated soil to the edible parts of the vegetables are shown by the accumulation factor (AF) formula (Chary et al. 2008).

$$\text{AF} = \text{metals concentration in vegetables/metals concentration in soil} \quad (1)$$

HEALTH RISK ASSESSMENT

The health risk assessment of selected waste water irrigated vegetables of the study area was calculated through the daily intake rate (DIR) and the hazard quotient (HQ). For that the average body weight and average daily intake of metals were calculated on the basis of a questionnaire (n=40 for each vegetable) of waste water irrigated vegetables of the study area (Table 1).

TABLE 1. Respondents interviewed for health risk assessment of waste water irrigated vegetables consumption in study area

Respondents	Age group (years)		Total interviewed persons
	<45	>45	
Male	28	45	73
Female	20	27	47
Total interviews			120

DAILY INTAKE RATE (DIR)

The daily intake rate of metals through consumption of selected vegetables viz., *Luffa acutangula* L., *Solanum melongena* L. and *Zea mays* L. was calculated by the equation of Orisakwe et al. (2012);

$$\text{DIR} = \frac{\text{C metals} \times \text{D average food intake}}{\text{B average weight}}, \quad (2)$$

where C represents the concentration of metals; D is the average daily intake of vegetables; and B is the average body weight of consumers.

HAZARD QUOTIENT (HQ)

Hazard quotient or chronic threat assessment in the study area was calculated through the equation of Khan et al. (2008).

$$\text{Hazard quotient (HQ)} = \frac{\text{DIR}}{\text{RfD}}, \quad (3)$$

where DIR represents the daily intake rate and RfD represents the reference dose of metals. The oral reference dose for Fe, Zn, Ni, Cr and Cd are 3.0, 3.0, 2.0, 1.5 and 1.0 mg/kg-day, respectively (Khan et al. 2008).

STATISTICAL ANALYSIS

The data of the research work was statistically calculated in mean, range and standard deviation. The mean, range and standard deviation values of metals in soil and vegetables samples were compared by using independent sample t-test. Statistics version 8.1 was used for basic descriptive statistical analysis of the data. The identification, location and for development of the sites of study area map the Arc-GIS software version 9.3.1 was used.

Quality control (QC) analysis Analytical grade (Merck, Germany) reagents were used in all the experimental procedures of the research study. All the glass ware and plastic ware used in the experimental work of the research work were thoroughly rinsed with 10% HNO_3 solution followed by washing with de-ionized water. To ensure the quality data of samples of research work, each sample was analyzed in triplicate.

For the concentration of respective metals analysis through atomic absorption three standards; one blank and two of 2.5 and 5.0 µg/L were used after every five samples. The standard solution of all the metals was prepared by dilution of certified standard (NIST-SRM, 1570a for plants and SRM 2709 for soil) of corresponding metals ions with de-ionized water.

RESULTS AND DISCUSSION

The pH of waste water was 5.80. The electrical conductivity (EC) of waste water was 13.00 dSm^{-1}. The pH and electrical conductivity (E.C) of waste water irrigated soil was 6.78 and 16 dSm^{-1}, respectively (Table 2).

The results of metal concentrations in waste water irrigated soil are shown in Table 3. The mean concentration of Ca, Mg and Na were 1326.7, 358.78 and 57.143 mg/kg, respectively. The mean concentrations of heavy metals like Zn, Ni, Fe, Cr and Cd were 56.433, 8.770, 46.390, 3.24 and 4.62 mg/kg, respectively. The heavy metals concentration in waste water irrigated soil of study area was observed in order of Zn > Fe > Ni > Cd > Cr. The waste water irrigated zone (soil) was highly contaminated with Cd as its concentration was higher than the permissible limit set by European Union Standards (EU 2006, 2002). The concentration of other screened heavy metals like Ni, Cr and Fe fell within the range of European Union Standards. The higher concentration of Cd in waste water irrigated soil might be due to small scale industries such as dyeing, electroplating, fabrics printing, batteries and paints, which discharge their effluents directly into the surface water used for irrigation purposes (Khan et al. 2015b). The Cd has easy availability and solubility in soil and hence accumulates in the edible parts of the plants (Luo et al. 2011). Similar findings were reported by Gupta et al. (2008) in the waste water irrigated soil samples collected from Titagarh, India. The present investigation was carried out to compare the level of metals in selected vegetables viz., *Solanum melongena* (Brinjal), *Luffa acutangula* (Gourd) and *Zea mays* (Maize) irrigated with municipal waste water. Table 4 shows that the highest (118.22 mg/kg) metal concentration level of Na was observed in *Luffa acutangula* while the lowest concentration (47.221 mg/kg) level of Na was noted in *Solanum melongena*. The concentration of K was higher (245.74 mg/kg) in *Solanum melongena* and lower (88.644 mg/kg) in *Zea mays*. The Mg showed the highest (392.61 mg/kg) value in *Zea mays* and lowest (223.49 mg/kg) value in *Solanum melongena*. The highest (1286.6 mg/kg) concentration of Ca was observed in *Solanum melongena* and lowest (1034.5 mg/kg) in *Luffa acutangula*.

TABLE 2. Physico-chemical properties of waste water irrigated soil of study area

Parameters	Waste water	Waste water irrigated soil
pH	5.80	6.78
EC (dSm^{-1})	13	16

TABLE 3. Metals concentration in waste water irrigated soil of study area

Metals	Mean ± SD	Range	EU (2002)	Awashthi et al. (2000)
Na	57.143±2.3756	54.410-58.710	NA	NA
Ca	1326.7±88.202	1260.8-1426.9	NA	NA
Mg	358.78±81.800	283.54-445.85	NA	NA
Fe	46.390±9.5252	36.490-55.490	NA	NA
Zn	56.433±48.371	0.6500-86.750	300	300-600
Ni	8.770±8.770	6.670-10.470	75	75-150
Cr	3.24±0.4339	2.560-3.670	150	NA
Cd	4.62±0.7561	3.240-5.220	3	3-6

Eu = European Union standard (2002), Awashthi et al. (2000), SD = Standard deviation, NA = Not allotted. The values are in mg/kg

In the heavy metals Zn showed highest (58.453 mg/kg) in *Luffa acutangula* and lowest (23.752 mg/kg) in *Solanum melongena*. The Fe indicated the highest (69.012 mg/kg) in *Luffa acutangula* while lowest (57.834 mg/kg) in *Solanum melongena*. The Ni showed highest (7.391 mg/kg) in *Zea mays* and lowest (6.065 mg/kg) in *Luffa acutangula*. The Cr has highest (4.418 mg/kg) concentration in *Zea mays* and lowest (2.420 mg/kg) in *Luffa acutangula*. The Cd showed highest (1.771 mg/kg) concentration in *Solanum melongena* and lowest (0.751 mg/kg) in *Luffa acutangula*. In the selected waste water irrigated vegetables Cr has highest (4.418 mg/kg) concentration in *Zea mays* and lowest (2.420 mg/kg) in *Luffa acutangula*. The Cd showed highest (1.771 mg/kg) concentration in *Solanum melongena* and lowest (0.751 mg/kg) in *Luffa acutangula*. The concentration of Cr and Cd in all the tested vegetables was higher than permissible limits set by WHO/FAO guidelines 2001.

The higher concentration of these heavy metals in waste water irrigated vegetables may result from the irrigation sources such as garages, sewage wastes and solid waste dumping sites that discharge their effluents in to the irrigation water sources from where they are deposited in the soil and finally accumulated in the edible parts of vegetables (Khan et al. 2015a, 2015b, 2008). Similar results for higher Cd concentration in selected vegetables were

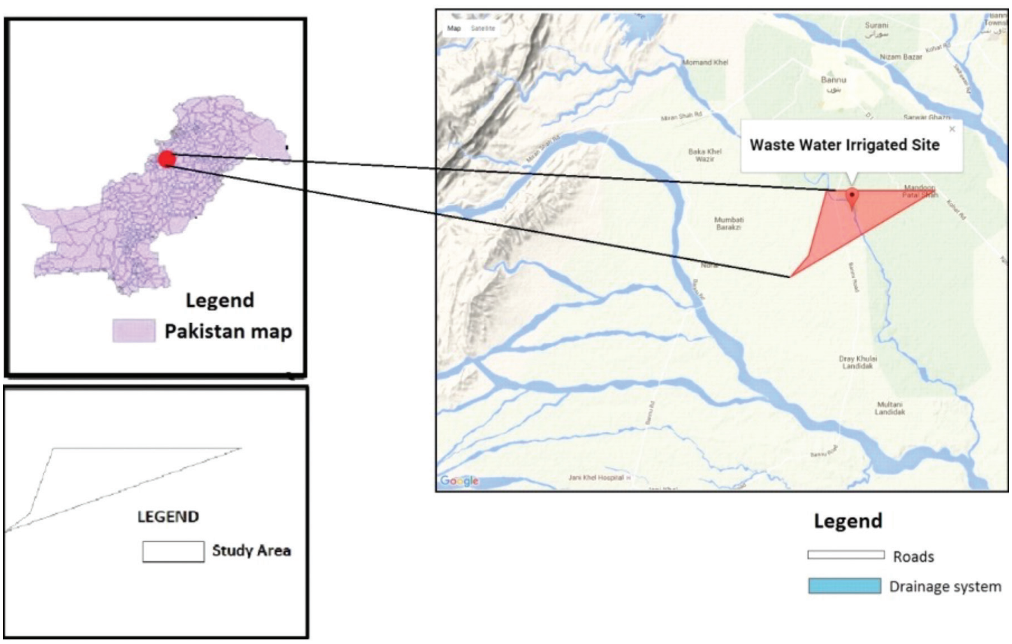

FIGURE 1. Map of study area

TABLE 4. Metals concentration in waste water irrigated vegetables of study area

Plants	Statistics	Na	K	Mg	Ca	Fe	Zn	Ni	Cr	Cd
L. acutangula	Mean±SD	118.22± 104.26	118.28± 148.35	247.74± 117.33	1034.5± 623.93	69.012± 32.012	58.453± 50.346	6.065±1.995	2.420± 0.597	0.751±.0.140
	Range	25.920-314.36	43.040-491.05	110.02-445.85	230.90-1956.7	36.490-142.39	0.6500-139.14	3.897-8.887	1.8780- 3.208	0.588-0.887
S. melongena	Mean±SD	47.221± 21.560	245.74± 158.34	223.42± 127.51	1286.6± 344.04	57.834± 30.531	23.752± 37.364	6.267±1.104	4.054± 0.648	1.771± 0.802
	Range	15.920-85.660	58.350-515.24	110.37-459.89	706.40-1726.1	14.590-125.89	0.6600-116.80	5.097-7.497	2.968-4.618	0.993-2.797
Z. mays	Mean±SD	53.487± 13.093	88.644± 20.319	392.61± 228.22	1204.5± 241.88	62.957± 33.113	36.606± 12.784	7.391±0.809	4.418± 0.617	1.335± 0.305
	Range	29.850-66.510	55.940-110.90	101.90-745.85	787.40-1556.7	28.790-129.69	18.860-54.250	6.307-8.577	3.408-5.038	1.017-1.798
WHO/FAO Guidelines 2001						425	100	67	2.3	0.2

The values are in mg/kg

reported by Torabitni and Mahjouii (2002) with waste water irrigation in South Tehran. Gardiner et al. (1995) reported that Cd is a highly mobile metal that easily accumulates in the edible parts of vegetables. Higher concentration of Cd is marked as a causative agent of tetragenosis, gastrointestinal cancer and mutagenesis (Jarup 2003). Dietary intakes of Cr and Ni cause headache, skin rashes, heart problems, breast cancer and respiratory problems (Khan et al. 2010).

The results of the accumulation factor (AF) or transfer factor are given in Figure 2. The *Luffa acutangula* has the highest transfer factor values for Fe (21.34) followed by Mg (9.90), Ca (7.46), Zn (5.71), Cr (0.746), Ni (0.691) and Cd (0.164) and Na (0.074). While *Solanum melongena* has the highest transfer factor values for Fe (36.65) followed by Mg (10.511), Ca (8.430), Cr (1.210), Ni (0.714), Cd (0.383), Zn (0.289), Na (0.077). The *Zea mays* has the highest transfer factor values for Fe (15.65) which was followed by Mg (8.164), Ca (7.28), Zn (4.302), Cr (1.29), Ni (0.84), Cd (0.289) and Na (0.084).

For calculation of daily intake rate (DIR) of metals and hazard quotient (HQ), 40 questionnaires for each vegetable were conducted in the study area. On the basis of the questionnaires, the average body weight of consumers for *Luffa acutangula, Solanum melongena* and *Zea mays* were 68.975, 69.55 and 72.25 kg, respectively. Similarly, the average daily intake rate of metals for *Luffa acutangula, Solanum melongena* and *Zea mays* were 1.624, 1.599 and 1.5 mg/kg/day, respectively.

Table 5 shows that the DIR of Ca in *Luffa acutangula* was high with (6.3359-53.692 mg/kg/day) range and low with (0.7112-8.6260 mg/kg/day) range for Na. Among the heavy metals the DIR was higher with (0.1399-0.2057 mg/kg/day) range for Ni and lower with (0.0178-3.8180 mg/kg/day) range for Zn. Similarly the HQ of *Luffa acutangula* was higher with (0.0699-0.1029 mg/kg) range for Ni and lower with (0.0256-0.0767 mg/kg) range for Cd.

Table 6 shows that the DIR in *Solanum melongena* was higher with (16.530-40.391 mg/kg/day) range for Ca and lower with (0.3725-2.0044 mg/kg/day) range for Na. In the heavy metals the Fe has highest DIR with (0.3414-2.9458 mg/kg/day) range and lowest with (0.0144-0.0288 mg/kg-day) for Cd. Moreover the HQ was high with (0.1225-0.2218 mg/kg) range for Zn and lowest with (0.0144-0.0288 mg/kg) range for Cd.

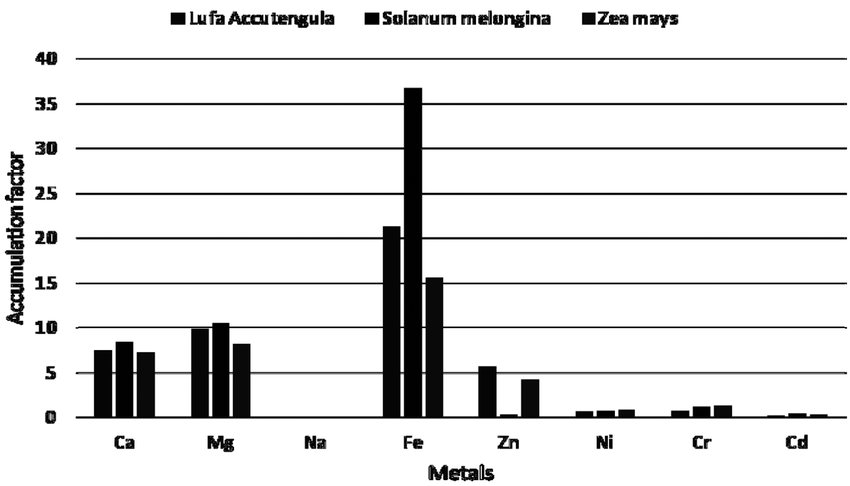

FIGURE 2. Metals accumulation factor (AF) or transfer factor in waste water irrigated vegetables

TABLE 5. Daily intake rate and Health Hazard Quotient of *Luffa acutangula* with waste water practice

Metals	Daily intake rate mg/kg-day		Hazard quotient
	Mean±SD	Range	Mean±SD
Na	3.2440±2.8608	0.7112-8.6260	NA
Mg	6.7980±3.2196	3.0189-12.234	NA
Ca	28.388±17.121	6.3359-53.692	NA
K	3.2457±4.0708	1.1810-13.474	NA
Fe	1.8937±0.8784	1.0013-3.9072	0.6224±0.2775
Zn	1.6040±1.3815	0.0178-3.8180	0.5088±0.4418
Ni	0.1720±0.0303	0.1399-0.2057	0.0860±0.0152
Cr	0.1112±0.0178	0.0814-0.1267	0.0742±0.0119
Cd	0.0486±0.0220	0.0256-0.0767	0.0486±0.0220

NA = Not allotted

TABLE 6. Daily intake rate and Health Hazard Quotient of *Solanum melongena* with waste water practice

Metals	Daily intake rate mg/kg-day		Hazard quotient
	Mean±SD	Range	Mean±SD
Na	1.1241±0.4754	0.3725-2.0044	NA
Mg	5.2279±2.9838	2.5826-10.761	NA
Ca	30.105±8.0504	16.530-40.391	NA
K	5.7504±3.7051	1.3654-12.057	NA
Fe	1.3533±0.7144	0.3414-2.9458	0.4511±0.2381
Zn	0.5558±0.8743	0.0154-2.7331	0.1853±0.2914
Ni	0.1422±0.0468	0.0912-0.2080	0.0711±0.0234
Cr	0.0566±0.1040	0.0439-0.0751	0.03399±0.0064
Cd	0.0204±0.0609	0.0144-0.0288	0.0216±0.0059

NA = Not allotted

TABLE 7. Daily intake rate and Health Hazard Quotient of *Zea mays* with waste water practice

Metals	Daily intake rate mg/kg-day		Hazard quotient
	Mean±SD	Range	Mean±SD
Na	1.2402±0.3036	0.6922-1.5422	NA
Mg	9.1036±5.2919	2.3629-17.294	NA
Ca	27.930±5.6087	18.258-36.096	NA
K	2.0554±0.4712	1.2971-2.5715	NA
Fe	1.4598±0.7678	0.6676-3.0072	0.4795±0.2727
Zn	0.8488-0.2964	0.4373-1.2579	0.2837±0.1056
Ni	0.1714 ±0.0188	0.1462-0.1989	0.0857±0.0093
Cr	0.1024±0.0143	0.0790-0.1168	0.0683±0.0095
Cd	0.0310±0.0220	0.0236-0.0417	0.0310±0.0070

NA = Not allotted

Table 7 shows that DIR in *Zea mays* was high (18.258-36.096 mg/kg/day) for Ca and low (0.6922-1.5422 mg/kg/day) for Na. In the heavy metals the highest DIR (0.6676-3.0072 mg/kg/day) was recorded for Fe and lowest (0.0236-0.0417 mg/kg/day) for Cd. Similarly the HQ of Fe was high with (0.2225-1.0024 mg/kg) range and low for Cd with (0.0236-0.0417 mg/kg) range. Similar results for the daily intake rate of heavy metals in various plant species have been reported by earlier works (Khan et al. 2015a, 2015b).

CONCLUSION

The soil irrigated with waste water exhibited accumulation of toxic heavy metals. Similarly, the vegetables irrigated with waste water were contaminated with various types of heavy metals. The concentrations of heavy metals such as Cr and Cd in vegetables were higher than the permissible limits. For heavy metals the daily intake rate was high for Fe and low for Cd. Hazard quotient was high for Fe and low for Cd. It is inferred from the findings of the present investigation that the consumption of vegetables irrigated with waste water in the study area pose potential health risks and may result in serious health issues. Therefore, proper treatment of waste water and public awareness is necessary to reduce the risks associated with the consumption of such vegetables irrigated with waste water.

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