

The absence of environmental law to protection of irrigation water with from accumulation of heavy metals in soil and crops in the region.

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ABSTRACT

Heavy metals exposure through consumption of contaminated crops is becoming a serious concern for human health in the absence of environmental law. Present study is based on the environmental and health risks associated with the use of treated wastewater produced from Al-Rustamia old plant for irrigation various crops cultivated in the soils around the station. The physicochemical parameters pH and electrical conductivity, were determined for the samples. Irrigated water, soil and vegetable samples for Zn, Pb, Ni and Cd concentrations and transfer factor from soils to plants (TF) were analyzed, while, their crop's and multi targeted risk, daily intake of metals (DIM) and health risk index (HRI) were calculated. Results showed, the mean values of Zn, Pb, Ni and Cd were lower in soil and crop than Food and Agriculture Organization guidelines for food additives and contaminants. The irrigated soil was contaminated with Zn 56.5, Pb 15.1, Ni 9.30 and Cd 0.850 mgkg⁻¹. The trend of heavy-metal concentrations in all samples was Zn > Pb > Ni > Cd. daily intake of metals in crops species for Zn (0.011 – 0.019 mgkg⁻¹), Pb (2.0×10⁻⁵ – 5.9×10⁻⁵ mgkg⁻¹), Ni (2.4×10⁻⁴ – 5.2×10⁻⁴ mgkg⁻¹) and Cd (1.3×10⁻⁵ – 3.3×10⁻⁵ mgkg⁻¹) are unexceeded the permissible limits. The Health Risk Index (HRI) varied from 0.037 – 0.063 for Zn, 5. ×10⁻³ – 1.4 ×10⁻² for Pb, 1.2 ×10⁻² – 2.6 ×10⁻² for Ni and 1.3 ×10⁻² – 3.3 ×10⁻² for Cd. The HRI was >1 for the elements indicating a unpotential health risk. Crop irrigation by wastewater irrigation is a prominent alternative option for water scarce countries, however prior testing and treatment of such wastewater streams must be employed to minimize the adverse impacts on human health and environment.

Keywords: Wastewater, Heavy metals, Crops, Health risk index, environmental law.

Article Received: 22 Nov 2020, Revised and Accepted: 15 Dec 2020 and published in January 2021

INTRODUCTION

Environmental pollution by the heavy metals and associated food safety is a major global concern now a day. These metals can pose a serious health implication to all living things in general and humans in particular if accumulated in elevated concentration above body requirements. Iraq is a country with limited renewable fresh water resources [1]. In Iraq, most of the arable land falls within the arid and semi-arid regions [2], which necessitates reducing the escalating deficit between the available water resources, the growing needs of the agricultural sector from irrigation water, and up to achieving the desired water balance and reaching the stage of water security, which is the solid basis for sustainable agricultural development. Based on that, and the cumulative growing deficit in the water resources available for agricultural purposes, it was necessary to search for alternative water sources that support the water budget and aim to set an important strategy to provide surface water and improve its quality through the reuse of (Wastewater) after its treatment in the field of agricultural irrigation and the introduction of this type of unconventional water as an additional renewable water resource within the national water budget in the irrigation of some crops is restricted, as the area of agricultural lands in which this quality of water is used is 37,11 hectares in the various governorates which constitute 6.2% of the total irrigated area. So, wastewater becomes

an attractive source to irrigate crops most probably in urban areas because of easy accessibility, cheap and reduced fresh water availability [3]. In developing countries, municipal or industrial wastewater are commonly used for the purpose of irrigation [4]. Irrigation with wastewater for long term might lead towards the heavy metals accumulation is increased in farming soils and plants, heavy metals are reserved under the category of environmental pollutant because of their poisonous effects to animals, plants and human and residents near such facilities is likely to occur. Heavy metals above allowable limits will often lead to disadvantageous effects in humans, other organisms and the environment at large . Allowable safe limits of heavy metals in food samples are associated with low health risks in humans [5] . Crop and vegetables comprise a significant part of the human diet since they restrain proteins, carbohydrates, minerals, vitamins and trace elements. Various studies showed heavy metals as main vegetables contaminants, crops and vegetables that grown in soil irrigated with sewage water contaminated with heavy metals poses a potential health risk to human beings [6,7,8,9] .

Accumulation of heavy metals in crop plants is of great concern due to the probability of food contamination through the soil root interface. Though the heavy metal viz. Cd, Pb and Ni are not essential for plant growth, they are readily taken up and accumulated by plants in toxic levels [10]. Ingestion of vegetables irrigated with waste water and grown in soils contaminated with heavy metals

poses a possible risk to human health and wildlife [11,12]. Heavy metal concentration in the soil solution plays a critical role in controlling metal bioavailability to plants. Most of the studies show that the use of waste water contaminated with heavy metals for irrigation over long period of time increases the heavy metal contents of soils above safe limits [13]. Ultimately, increasing the heavy metal content in soil also increases the uptake of heavy metals by plants depending upon the soil type, plant growth stages and plant species [14]. Due to the noxious effects of these heavy metals, and in order to guarantee food safety, it is imperative to periodically monitor sewage effluents for heavy metal contamination levels to determine the quality of waste water being discharged to the environment.

In view of the ever increasing evidence and significance of heavy metal contamination, the current work reported daily metal intake by human body on average basis, health risk index related with heavy metal ingestion and heavy metal concentration in water, soil and selected vegetables irrigated with the wastewater of AlRistmia region in Baghdad, as the Drain receives large volumes of effluent from households and industries, in order to reach binding environmental laws and legislations for the optimal use of wastewater in accordance with national environmental determinants and to limit the expansion of environmental and food pollution.

1. Materials and Methods

The current study was carried during spring 2016 in Baghdad city, Iraq. This region has a wastewater treatment plant (WWTP) Al-Rustumiya Sanitation with a capacity $175000 \text{ m}^3\text{day}^{-1}$ of 3 million population equivalents (About a third of the population)[15]. The WWTP is located on a lot of 40.7 ha besides the Diyala river bed to the south of Baghdad city. The wastewater is discharged from the WWTP into Diyala river and thus directed through the irrigation channels into the field where farmers fetch to irrigate their crops. The Discharge point and irrigation area are located at latitude 33.274446N, 33.262611N and longitude 44.53724E, 44.524999E and at the elevation of approximately 454 and 379 meters above sea level, respectively (Fig. 1).

Water sampling and treatment

The water samples were collected arbitrarily from the similar sites from where vegetables and soil samples were collected along the canal from the discharge of raw sewage, from WWTP of Al-Rustumiya Sanitation to the irrigation area. twenty samples were taken in polyethylene bottles (250ml) which were sterilized with HNO_3 (1%), the HNO_3 acid was added for the purpose to avoid any microbial activity and preservation of the water samples [16]. Then samples were filtered through Whatman 42 filter paper and stored in storage flasks at 4°C until analyses [16]. Then, pH, electrical conductivity (EC) and heavy metals concentrations were measured according to standard methods and standard procedures were maintained to avoid false reading and contamination [17].

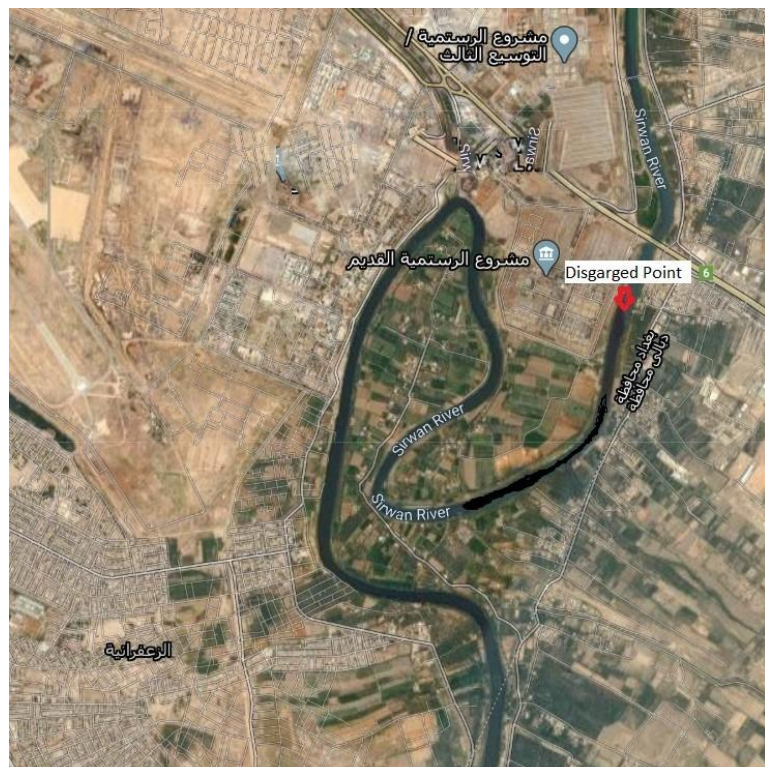


Fig. 1. A map showing sampling points around WWSP in AlRistmia (Google map, 15th July 2019).

Soil sampling and characterization

Eight representative soil samples from irrigation area were collected randomly from each plant sampling plot, the samples were mixed thoroughly to obtain an individual composite sample in a clean polyethylene bag. The soil sub-samples were obtained from the depth of 0–20 cm using a stainless steel hand soil auger. In the laboratory, all samples were air dried in ambient conditions and passed through a 2-mm mesh sieve. Non-soil particles e.g. stones, wooden pieces, rocks, gravels, organic debris were removed. pH, electrical conductivity (EC) were measured according to soil analysis methods [18].

Plant sampling and identification

The common vegetables and crops that was used as a botanical guide grown in the study area are Wheat (*Triticum turgidum*), Millet (*panicum miliaceum*) and Tomato (*Solanum lycopersicum*) irrigated with wastewater. 500 g of each plant was collected with three replications. In the laboratory, each individual vegetable was separated into root, leaf and seed subsamples and properly washed with deionized water to remove all visible soil particles.

Heavy metals analysis

All samples were dried in an oven at 75°C for 72 h and then ground to powder. 0.5 g of each soil and vegetable samples were digested in the presence of aqua regia (mixture of nitric acid HNO_3 concentrated with hydrochloric acid HCl (1:3)) overnight at room temperature. Then, the mixture was boiled for 2 h

according to the standard method [18]. After cooling, the digested samples were filtered using Membrane (0.45 mm), the filtrate was diluted to 50 ml with deionized water and kept at room temperature for further analysis

Heavy metals determination

Determination of the four heavy metals, Fe, Zn, Cd and Pb in the filtrate of samples was carried out and achieved by using Shimadzo atomic absorption spectrophotometer (AAS). The standard solutions were prepared for heavy metal determination by diluting a stock solution containing 100 ppm of single element AAS grade standard with distilled water. The standard solutions for the heavy metals were used to construct the calibration curves with the help of AAS. The Guideline for maximum limit of heavy metals (Zn, Ni, Pb and Cd) in irrigation water, soil and vegetable was adopted from the reference by WHO/FAO.

Data analysis

Transfer factor (TF)

The bioaccumulation of metals in plants from soils can be predicted using a transfer factor (TF) is the ratio of metal concentration in plant to the concentration of metals in soil (mg k⁻¹) [12].

$$TF = C_{\text{Edible part}} / C_{\text{soil}}$$

where : C_{Edible part} and C_{soil} represent the heavy metal concentration in edible part of vegetables and soils, respectively.

Daily Intake of Heavy Metals (DIM)

The daily intake of heavy metals DIM depends on both the metal concentration in crops and the amount of consumption of the respective food crop, was calculated using the following equation:

$$DIM = C_m \times C_f \times I_d / B_w$$

Where: C_m, C_f, I_d and B_w represent the heavy metal concentrations in plants (mgkg⁻¹), conversion factor (fresh vegetable weight into dry vegetable weight), daily intake of vegetables and average body weight, respectively.

The conversion factor 0.085 was used to convert fresh green vegetable weight to dry weight, as described by [19]. The average daily vegetable intake for adults and children was considered to be 0.345 and 0.232 kg/person/day, respectively, while the average adult and child body weights were considered to be 70.0 and 35.7 kg, respectively [19]. Upper tolerable daily intake limit (safe limits) for both adults and children through the consumption of vegetables were presented in Table 1.

Health Risk Index (HRI)

The health risk index is calculated by dividing daily intake of metals (DIM) by reference oral dose (RFD). This index represents the harmful to people in which consume vegetables contaminated with heavy metals. If the value of HRI is less than 1, people will safe to eat those kinds of vegetables [20].

$$HRI = DIM / RFD$$

Oral reference doses (mg kg⁻¹ day⁻¹) for each heavy metal including Zn (0.3), Ni (0.02), Pb (0.004) and Cd (0.001) [21].

Table 1 Upper tolerable daily intake (mg person⁻¹ day⁻¹)

limit for both adults and children.

Heavy Metals	Integrated Risk Information System [22]
Zn	40E-00
Ni	1.0E-00
Pb	2.40E-01
Cd	6.40E-02

Statistical analysis

Statistical analyses were carried out using the Statistical Analysis Systems Computer Package (SAS Institute, 2004). A completely randomized block design (CRBD) was used in these experiments. The measurements were expressed in term of mean and standard error. The comparison of heavy metals concentration in wastewater from discharge point and irrigation area were done by a paired t-test. Treatment means were compared by the least significant difference between heavy metal concentration in vegetables grown in irrigation area test at p < 0.05.

Results and discussion

The results of the physicochemical parameters of the wastewater discharged by the WWTP of AlRistmua used in the irrigation of crops are presented in the Table 2. The average pH values for wastewater samples from discharge point and Irrigation area were 7.83 and 7.70 respectively. These values are within the permissible limit of [23,24] set for irrigation purposes . The mean electrical conductivity for wastewater at the two sampling points was 6.32 and 5.90 dSm⁻¹ respectively. The comparison of the values of the electrical conductivity in the wastewater analyzed with the water quality standards for irrigation can be deduced that these wastewater samples are unacceptable for irrigation of crops [24]. The mean heavy metal concentrations (mg l⁻¹) in wastewater between the sites were 0.168 – 0.150 for Zn, 0.137 – 0.100 for Ni, 0.227 – 0.220 for Pb and 0.009 – 0.009 for Cd. The concentrations of heavy metals in wastewater channel were highest relatively for the Pb, followed by Zn, Ni, and Cd. Concentrations of Zn, Ni and Cd in the wastewater were below the permissible limits of heavy metals allowed in the irrigation water, but Pb exceeded the limit set by Iraqi national limits (Table 1). Heavy metals that are present in wastewater used for irrigation tend to accumulate in the soils where there is a potential that they could become bioavailable for crops.

Table 2. Average values (±Std.Error) of physico-chemical parameters and concentrations (mg l⁻¹) of heavy metals in wastewater from discharge point and irrigation area.

Parameter	Discharge point	Irrigation area	P value	WHO/FAO standards for irrigation [23]	Iraqi national limits [24]
E.C (dSm ⁻¹)	6.32±0.184	5.90±1.04	0.711	0.703 – 3.12	3.91
pH	7.83±0.7	7.70±1.1	0.9	6.5 – 8.0	4.0 –

	65	5	08		8.6
Zn	0.168±0.036	0.150±0.047	0.647	2.0	2.0
Pb	0.227±0.018	0.220±0.024	0.828	5.0	0.100
Ni	0.137±0.063	0.100±0.051	0.194	0.200	0.200
Cd	0.009±0.002	0.009±0.003	0.136	0.01	0.01

Results presents in table 3. shown the characteristics and total concentration of physico-chemical parameters in soils sampled at irrigation area. The average pH value of the soil samples was 7.80. The pH is one of the factors influencing the bioavailability and the transport of heavy metals in the soil. Heavy metal mobility decreases with increasing soil pH due to the precipitation of hydroxides, carbonates or the formation of insoluble organic complexes, the soil pH has a major influence on the mobility and bioavailability of heavy metals and its changes depend on pH of the wastewater used for irrigation [25]. The mean of electrical conductivity in soil sample was 4.07 dSm⁻¹, classifying it as moderately saline according to [26]. Increase in conductivity has been shown to enhance the solubility of heavy metals, thus causing greater availability of metals to the plant from the soil [26]. The mean concentrations of Zn, Pb, Ni and Cd in studied soil were 56.5, 15.1, 9.30 and 0.850 mgkg⁻¹. The mean highest concentration recorded in the soil was for Zn followed by Pb, Ni and the minimum concentration was observed for Cd. The concentrations of Zn, Pb, Ni and Cd was found less than those reported were observed within their respective permissible limits set by [22]. Continuous removal of metals by food crops (vegetables and cereals) grown at the wastewater irrigated soil and heavy metals leaching into the deeper layers of soil may be a reason of low concentration of heavy metals than the permissible limits [27].

Table 3. Heavy metals concentration of soils collected from the study area irrigated with wastewater

Physico-chemical parameters		mgkg ⁻¹			
E.C	pH	Zn	Pb	Ni	Cd
4.07	7.80	56.5	15.1	9.30	0.850
Recommended levels in the soil [22]		200	300	50	3.00

Table 4. summarizes the concentration of heavy metals in the various of plant grown in this region. The results indicated that all metals in the food crops exceeded acceptable limits. The trend of heavy-metal concentrations in plants was Zn > Ni > Pb > Cd. [28] found highest concentration of Fe and Zn and lowest of Pb in various imported and locally produced vegetable crops, including root crops (Turnips and Carrots), stem crops (Potatoes and Onion), leaves (Lettuce), and fruits (Tomatoes) collected from Baghdad, Iraq markets. According to plant species, the lowest Zn concentration (30.3 mgkg⁻¹) was

observed in Millet, while the highest (33.6 mgkg⁻¹) in Wheat. The lowest and highest Pb concentrations were observed in Millet (0.048 mg/kg) seeds and Tomato fruit (0.107 mgkg⁻¹). For Ni, the lowest concentration was detected in Millet (0.579 mgkg⁻¹) from seeds, while the highest (0.948 mgkg⁻¹) was detected in Tomato. The highest Cd concentration (0.060 mgkg⁻¹) was observed in Tomato and the lowest (0.032 mgkg⁻¹) in Wheat. The results of the statistical analysis (table 4) and using the least significant difference index (p<0.05) showed that there are no significant differences between the different plants through the overall average heavy metal concentration. Also there are no significant differences between the various heavy metals, with the exception of zinc, it has significant differences from the rest of the studied elements through the overall mean plants. Metal concentrations has shown variations in their availability to the plant, this may be attributed to differences in the uptake mechanism of individual ions and competition among the heavy metals for absorption by different plants. The interaction between different heavy metals occurs at root surface and also within the plant, which ultimately affects the uptake and translocation of heavy metals. The interaction between different heavy metals occurs at root surface and also within the plant, Uptake and kinetic for nutrient ions and chemically related nonnutrient analogs suggest that metabolic processes associated with root absorption of nutrients regulate both the affinity and rate of absorption of specific nonnutrient ions [29]. The results from the present study and earlier reports [8,14,17] demonstrated that plants grown on irrigated soils by wastewater are contaminated with heavy metals and pose a health concern.

Table 4. Multivariate analysis of variance, mean comparisons using LSD test, P value 5% and standard

Error (Sd.Err) for heavy metals concentration in edible plant part (mgkg⁻¹).

Plants	Zn	Pb	Ni	Cd	Row s average
Tomato	26.5±0.866	0.107±0.004	0.948±0.005	0.060±0.003	6.90a
Wheat	33.6±1.73	0.062±0.013	0.831±0.017	0.032±0.010	8.63a
Millet	30.3±3.06	0.048±0.016	0.579±0.035	0.042±0.007	7.74a
Columns average	30.1a	0.072b	0.786b	0.045b	
Guide line for safe limits [23]	50	0.3	5.0	0.2	

Results of the current study showed the values of bioconcentration factor from soil to crop samples (figure

1). Zinc had the highest BCF value among all elements. The maximum BCF of Zn was found in Wheat soil 0.595 mgkg⁻¹ followed by Millet and Tomato soil 0.536 and 0.469 mgkg⁻¹ respectively. The trend of these BCF values decreased in the order of Zinc > Nickel > Cadmium > lead. Among all the crop samples Wheat exhibit the highest BCF value followed by Millet and then Tomato. These results indicated that there is significant difference between the BCF mean value among the four metals, and there is no a significant difference between the BCF mean value among the three plants. Toxic elements that have more value of bioconcentration factor, have more chances and easier way of accumulating in plants/crops and translocate to edible parts than toxic elements with low bioconcentration factor and can cause more health risks [30]. Parallel BCF results were detected from earlier researches in different areas of Iraq. These results were protuberant in terms of long-term wastewater irrigation in which the same properties in the soil would not be the part of food chain [31]

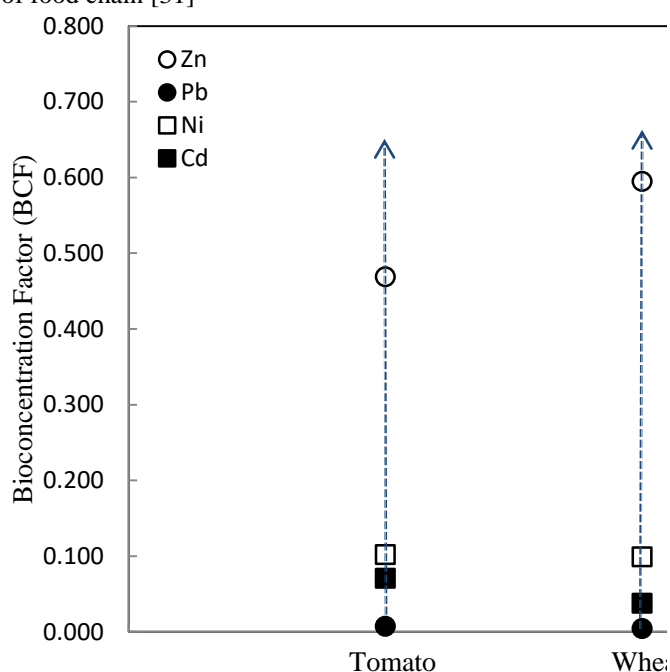


Figure 2. Metals Bioconcentration factor for crop grown in wastewater.

To assess the health risk index due to heavy metals intake from crops consumption, it is essential to estimate the level of exposure by quantifying the routes of exposure of a pollutant to the target organisms. There are various possible exposure pathways of pollutants to humans, but the food chain is one of the key pathways of heavy metal exposure to humans. Therefore, to assess the potential human health risks in this area, both the DIM and HRI for adults and children were calculated and the results are presented in Tables 5. The DIM results in were compared with the recommended daily intake of metals and the upper tolerable daily intake level (UTDL) established by the Institute of Medicine for people between the ages of 19 to 70 years [32]. It is very clear from the table that daily intake of metals in crops species for Zn (0.011 – 0.019 mgkg⁻¹), Pb (2.0×10⁻⁵ – 5.9×10⁻⁵

mgkg⁻¹), Ni (2.4×10⁻⁴ – 5.2×10⁻⁴ mgkg⁻¹) and Cd (1.3×10⁻⁵ – 3.3×10⁻⁵ mgkg⁻¹) are significantly lower than the recommended daily intake of metals and fall within the upper tolerable daily intake level (UTDL).

As well as, the results of this study indicated (table 5), the heavy metals, have HRI<1 for all the crops species cultivated in study area, HRI < 1 means that the exposed population is safe of metals health risk while HRI > 1 means a possible future risk for human health via the intake of food crops. [12] had also observed in his study, accumulation of seven different heavy metals (Cd, Pb, Zn, Cr, Cu, Ni and Fe) was determined in four different green vegetables were studied, namely, tomato, onion, pepper and beans irrigated with different water resources (tube well water, river water, and wastewater), and it was determined that DIM values were free of risk due to consumption of vegetables grown under wastewater irrigated areas of Nevsehir Province. The study of [15] focuses on the environmental and health risks associated with the use of treated wastewater produced from Al-Rustamia plant for irrigation vegetable. The average $LSD_{(0.05)Plant} = 0.024$ $LSD_{(0.05)Metal} = 0.028$ $lex (CPI)$ which indicates ns and the average organic F es slight pollution in Winter season, while for the other seasons indicated as being to be contaminated.

Table 5. Daily intake of metal rate (mg person⁻¹day⁻¹) and health risk index (HRI) for individual heavy metals caused by the consumption of different agricultural crops grown in the study area.

Crops	Individuals	Health risk index	Zn	Pb	Ni	Cd
Tomato	Adults	DIM	0.011	4.5E-05	4.0E-04	2.5E-05
			0.015	5.9E-05	5.2E-04	3.3E-05
	Children	HRI	0.037	1.1E-02	2.0E-02	2.5E-02
			0.050	1.4E-02	2.6E-02	3.3E-02
Wheat	Adults	DIM	0.014	2.6E-05	3.5E-04	1.3E-05
			0.019	3.4E-05	4.6E-04	1.8E-05
	Children	HRI	0.047	6.5E-03	1.7E-02	1.3E-02
			0.063	8.5E-03	2.3E-02	1.8E-02
Millet	Adults	DIM	0.013	2.0E-05	2.4E-04	1.8E-05
			0.017	3.4E-05	3.2E-04	2.3E-05
	Children	HRI	0.043	5.0E-03	1.2E-02	1.8E-02
			0.057	8.5E-03	1.6E-02	2.3E-02

*RDI M	mg person ⁻¹ day ⁻¹	8	0.00 0	0.50 0	0.00 0
*UTD L	mg person ⁻¹ day ⁻¹	40	0.24 0	1.00	0.06 4

* Recommended daily intake (RDIM) and upper tolerable daily intake (UTDL) levels of heavy metals in foodstuffs [32].

Conclusion

In this study, three different crops cultivated were studied, namely, tomato, wheat and millet irrigated with treated wastewater discharged from the Rustumiya plant, in Baghdad Province. Accumulation of four different heavy metals (Zn, Pb, Ni and Cd) was determined in wastewater, soils and different of crops. As a result, continuous irrigation of agricultural lands by wastewater for several years increased heavy metals in soils and plants it can be stated that there is no danger in using wastewater in the near term for irrigation of vegetables, in terms of heavy metal contamination. The results of the study clearly indicate that the wastewater-irrigated soils were moderately enriched with Zn, Pb, Ni and Cd, while the crops contaminations exhibited variations relative to FAO/WHO permissible limit. Therefore, the dietary intake of these vegetables results in long term low level body accumulation of heavy metals and the detrimental impact becomes apparent only after several years of exposure. The Health Risk Index (HRI < 1) indicated also that Zn, Pb, Ni and Cd uncontainment in most of the vegetables and not had potential for human health risk due to consumption of plants grown in this area having long term uses of waste water for irrigation. Therefore, this region needs some effective measures to maintain sustainability .

Author Contributions

Conceptualization, ; methodology, ; formal analysis, ; investigation, ; resources, *Author name* ; writing—original draft preparation, ; writing—review and editing, **Fayath salamat**; visualization, ; project administration, . All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding

Conflicts of Interest: The authors declare no conflict of interest.

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