©2022 Published in 6th International Symposium on Natural Hazards and Disaster Management
21-23 October 2022 (ISHAD2022 Bursa - Turkey)
https://doi.org/10.33793/acperpro.XXXX

# The ecological and health risks of heavy metal in vegetables irrigated with untreated wastewater under a semi-arid condition

[1*]Salar Rezapour and [2]Hawzhin M. Jalil
[1]Soil Science Department, Urmia University, Urmia, I. R. Iran
[2] Horticulture Department, University of Raparin, Qaladiza, Sulaymaniyah, I. Q. Iraq

## Abstract

The study evaluated the impact of wastewater irrigation (WW) on the severity of pollution and health risk potential of Zn, Cu, Cd, and Pb heavy metals in vegetables irrigated with WW versus those grown with fresh water (FW) across different vegetable kinds. This multifaceted assessment was conducted using the pollution index (PI), ecological risk (ER), bio-concentration factor (BCF), hazard quotient (HQ), overall hazard index (HI), carcinogenic risk (CR), and total carcinogenic risk (TCR). The findings revealed that WW irrigation increased metal concentrations across all soil samples and vegetable varieties in the order of Cd > Zn > Pb > Cu and Zn > Cu > Pb > Cd, respectively. When compared to the FW-irrigated soils, the values of PI and ER indicate a significant increase (an increase of 27%-3.3 times and 2.6-3.1 times, respectively) of the heavy metals in the WW-irrigated soils, falling their class from one to two (PI) and two (ER) grade. The order of heavy metals and BCF values exhibited the following pattern: Zn > Cu > Pb > Cd and Zn > Cu > Cd > Pb >, respectively, in all vegetables under WW irrigation and cabbage has the most potential to take up heavy metals compared with other crops. The mean HQ and HI were in the low category ($0.1 \leq$ HQ and HI $< 1$) and the CR and TCR were also within acceptable limits (1.00E-06 to 1.00E-04) by consuming veggies produced with WW, to children and adults implying that there are insignificant health risks to local inhabitants. However, the levels of HQ, HI, CR, and TCR were significantly higher in vegetables grown with WW than in those irrigated with FW, suggesting that wastewater irrigation has a major detrimental impact on human health.

**Key words:** Food pollution, Health risk assessment, Soil pollution, Fresh water, Vegetable

## 1. Introduction

Wastewater (WW) irrigation has gotten a lot of attention recently, particularly in water-scarce areas where producers have little choice but to use WW. WW irrigation, according to several studies, improves soil fertility and agricultural productivity by increasing nutrients in the form of N-P-K and trace metals (Cu, Mn, Fe, and Zn) [1]. Long-term continuous WW irrigation, on the other hand, may cause certain unintended modifications (negative impacts) in soil quality over time. Excessive dissolved salts, erosion, and mainly heavy metal contamination are all instances. Heavy metals are present in wastewater, and frequent irrigation with WW can cause to metal buildup in soil and crops. Consumption of crops and vegetables produced in heavy metal-polluted soil can leads to a variety of health issues, including blood pressure fluctuations, liver, lungs, and renal malfunction, and immune system insufficiency [2]. Given all the environmental cautions about the occurrence of heavy metals in WW-irrigated soils, the possibility of their uptake and accumulation by plants and the consequent possibility of entering the metals into human food chain, there is an evident need to assess the level and pollution of heavy metals in the soil–crop-human system under WW

*Corresponding author: Soil Science Department, Urmia University, P. O. Box 165, Urmia, 57134, I. R. Iran

irrigation in calcareous soils [3]. The following are the objectives of this research: (1) to analyze the accumulation and heavy metals content in soil and edible parts (lettuce, cabbage, green beans, and pepper) sampled from the wastewater and freshwater irrigated cropland, (2) to determine how WW irrigation affects the pollution index, ecological risk, and BCF of Zn, Cu, Cd, Ni, and Pb, (3) to determine the non-carcinogenic and carcinogenic risks, as well as human health concerns, associated with heavy metal buildup in vegetables irrigated with WW.

## 2. Materials and Method

### *2.1. Site description and soil-crop sampling*

The current research was conducted at Urmia (45º05' to 45º08' E and 37º32' to 37º38' N), the Western-Azarbaijan province, north-west Iran. The average rainfall and temperature in the region are 330 mm and 13 °C, respectively, resulting in a semi-arid Mediterranean climate with a cold winter and hot summer. Experimental sites for the study were selected in the vegetable fields (green bean, cabbage, pepper, and lettuce), growing mainly from April to September. The wastewater is generated from municipal, household, and local industry in Urmia city, used directly by local farmers through flood irrigation techniques without any refinement. Within a total of six farms, the composite soil samples were taken at random from the WW-irrigated site and the adjacent site (as control) irrigated by fresh water (FW), covering an area of 12 ha as six farms. The composite soil samples (36 pairs of samples) were mixtures of five sub-samples collected (depth of 0-50 cm) and were analyzed as described in the 2.3 section. Wastewater and freshwater samples, used for irrigation, were also collected from each experimental farm. At maturity, edible portions of green beans (Phaseolus Vulgaris), cabbage (Brassica Oleracea), peppers (Capsicum Annuum L.), lettuce (Lactuca Sativa L.), were collected from both the farms –induced by WW and FW. The edible parts were cleaned in double distilled water and dried at 70 ºC in an oven, crushed separately using a steel grinder, and passed through a1-mm sieve (100 mesh).

### *2.2. Laboratory analysis*

Using standard method, all soil samples were analyzed to determine their particle-size distribution, pH, electrical conductivity (EC), calcium carbonate equivalent (CCE), organic carbon (OC), and cation-exchange capacity (CEC) [4]. To extract the total proportion of Zn, Cu, Cd, and Pb, the soil samples were processed in strong nitric acid [5]. A 1.00 g of powdered samples of food crops were washed at 500 °C for 12 h, extracted with 2 M HCl, and filtered by a centrifuge to quantify select heavy metals. The concentration of Zn, Cu, Cd, and Pb in WW, FW, soil, and crop samples were analyzed using Shimadzu AA-6300. The certified standard solutions (1000 mg $l^{-1}$, Merck-Germany) of all five metals were applied as a stock solution for instrument standardization.

### *2.3. Pollution indicators and bioaccumulation factor*

**The soil pollution index (PI)** was calculated using Eq. (1) [6].

$$\boldsymbol{PI} = \frac{\boldsymbol{C_p}}{\boldsymbol{C_b}} \qquad (1)$$

Where $C_p$ and $C_b$ are the heavy metal values in the WW-irrigated soil and the control, respectively.

**The ecological risk index (ER)** was calculated as the sum of the risk factor of the heavy metals using Eqs 2 and 3 [7]:

$$ER = \sum_i^n Ei \qquad (2)$$

$$Ei = TiPI \qquad (3)$$

Where Ei denotes the monomial ecological risk factor and Ti is the toxicity response coefficient of heavy metals, considered as Zn =1, Cd = 30, and Cu and Pb = 5 to 5 for Cu and Pb [7]. The ER is the sum of all four risk factors for heavy metals which were grouped into four categories as: low potential ecological risk (ER ≤ 50), modest risk to the environment (50 < ER ≤ 100), considerable risk to the environment (100 < ER ≤ 200), and strong risk to the environmental (ER > 200) [7]

**The bio-concentration factor (BCF)** was assessed using Eq 4 to investigate the migration ability of heavy metals from soil to the edible portion of the study vegetables [3-8].

$$BCF = \frac{C_{crop}}{C_{soil}} \qquad (4)$$

Where C crop and C soil show the concentration of heavy metals in the vegetables (edible portion, on a dry weight) and soil, respectively.

**2.4.** ***Health risk assessment***

**Chronic daily intake (CDI)** of a metal was estimated as follows [9]:

$$CDI = \frac{(C_M \times C_F \times D_{FI})}{BW} \qquad (5)$$

Where CM is the HM concentration in vegetables (mg gk-1), CF is conversion factor of 0.085, DFI is the daily intake of vegetables with 0.232 and 0.345 $kg^{-1}$ $day^{-1}$ for children, and adults, respectively, and BW is the mean body weight (32.7 and 55.9 kg for children and adults).

The following Eqs were used to determine the **hazard quotient (HQ)** and **hazard index (HI),** showing the non-cancer risk during a lifetime for each metal and the overall potential non-carcinogenic impacts, respectively.

$$HQ = \frac{\mathrm{CDI}}{\mathrm{RfD}} \qquad (6)$$

$$HI = \sum_i^n HQ \qquad (7)$$

where RfD is the highest tolerable risk to humans from ingesting contact on a daily basis (mg $kg^{-1}$ $day^{-1}$). The RfD value is 0.3, 0.04, 0.001, and 0.04 mg kg-1 day-1 for Zn, Cu, Cd, and Pb, respectively. When HQ and HI are less than 1, the assumption is that potential non-carcinogenic effects are unlikely to occur; while if HQ and HI have to be >1, non-carcinogenic effects are possible.

Using Eq. (8), **cancer risk (CR)** was also assessed to estimate cancer risk over a lifetime because of exposure to a carcinogenic metal [9].

$$CR = CDI \times SF \qquad (8)$$

The SF is the slope factor. The case of 1 × 10-6<CR < 1 × 10-4 is an acceptable risk, and when CR>1 × 10-4, potential carcinogenic risk is possible.

## 3. **Results and Discussion**

### *3.1.Soil heavy metals*

Irrigation by WW resulted in a significant build-up in the values of Zn, Cu, Cd, and Pb compared with soil irrigated by FW. Depending on the farm site, an increase range of 29% to 3.6 times, on average, was observed in the concentration of all soil heavy metals under WW-irrigation compared with FW-irrigated soil. The greatest increase (2.2 -3.6 times) was found for Cd, followed by Zn (74.6-117.6%), Pb (43-71.4%), and Cu (29-54.3%), suggesting that all soils were substantially enriched by heavy metals produced from WW irrigation. In just 25% of the samples, however, the amount of Cd was over the acceptable limit [8] and the value of other metals were less than their threshold range in all soil samples. This means that some soils impacted by WW irrigation might not have been suitable for agricultural development due to phytotoxic issues caused by Cd for crops.

### **3.2.** *Indices of soil pollution*

The mean comparison of PI values showed increases of 65.5-108.7% for Zn, 30.84-57% for Cu, 3.04-3.3 time for Cd, and 27-101.8% for Pb, in the WW-irrigated soils compared with the FW-irrigated soils (Table 1). These increases have descended the PI class from low pollution (1<PI ≤ 2) in the FW-irrigated soils to moderate pollution (2<PI ≤ 3) in the WW-irrigated soils in 90% of the samples for Zn, 10.4% of the samples for Cu, and 47.9% of the samples for Pb. For PI-Cd, the majority of soil samples showed a scope of low or moderate pollution in the FW-irrigated, replaced by high pollution in 100% of the samples of WW-irrigated soils. These data imply that WW irrigation significantly increased both the PI values and the scope of the metals in the order of Cd> Zn> Pb> Cu and Cd was the most polluted metal in the study region.

**Table 3.** The level of PI of the investigated heavy metals in WW-irrigated and FW-irrigated soils.

| Data | | **Pollution index (PI)** | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | WW-irrigated soil | | | | FW-irrigated soil | | | |
| | | Zn | Cu | Cd | Pb | Zn | Cu | Cd | Pb |
| Min | | 1.82 | 1.4 | 4.76 | 1.14 | 1.1 | 1.07 | 1.18 | 1.11 |
| Max | | 3.13 | 2.0 | 13.39 | 3.43 | 1.5 | 1.21 | 3.08 | 1.7 |
| Mean | | 2.32 | 1.7 | 8.58 | 2.17 | 1.3 | 1.14 | 2.12 | 1.4 |
| SD | | 0.34 | 0.1 | 2.09 | 0.45 | 0.13 | 0.03 | 0.55 | 0.13 |
| %Class | LP | 100 | 100 | | 47.92 | 100 | 100 | 45.8 | 100 |
| of total | MP | | | | 43.75 | | | 41.67 | |
| samples | HP | | | 100 | 4.17 | | | 12.5 | |

PI; LP = low pollution (1< PI≤ 2), MP = Moderate pollution (2<PI ≤ 3), HP = High pollution (PI> 3).

The potential ecological risk (ER) analysis showed that WW irrigation resulted in a significant decrease in the ER values (min = 162.2, max = 423, mean = 287.8) compared with FW-irrigated soils (min = 48.7, max = 106.6, mean = 77.7). A considerable and high risk was found in the 45.8% and 54.2% of soil samples, respectively, under WW irrigation, while 25% and 75% of soil samples under FW irrigation showed a low and moderate risk, respectively. The results revealed that, on average, the ER degree falls in the high class of ecological risk by wastewater irrigation. Therefore, the data further confirmed that the quality of soils under WW irrigation was deteriorated by heavy metals. For both WW and FW-irrigated soils, Cd showed the highest contribution to ER, accounting in the range of 91– 94% and 80 – 83% for WW and FW-irrigated soils, respectively, followed by Pb, Cu, and Zn (Figure. 1).

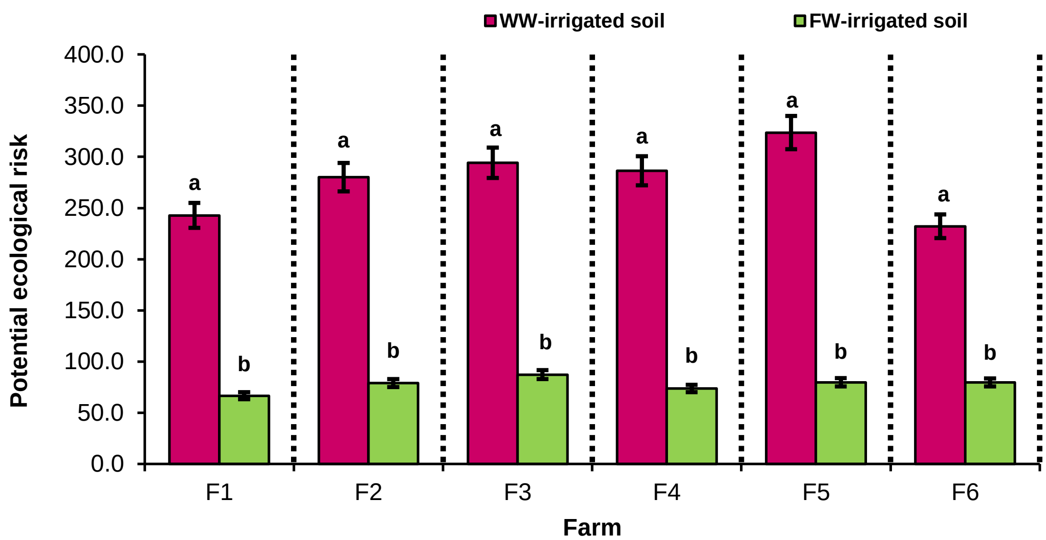



**Figure 1**. Comparison mean of ER values between the irrigated farms with WW and FW. In each farm, different letters show significant differences in ecological risk at $P < 0.05$ confidence level

### *3.3.Concentration of heavy metals in vegetables*

The following trend emerged in the order of heavy metal concentrations; Zn > Cu > Pb > Cd in all vegetables under WW and FW irrigation. This might be related to vegetables' increased capacity and demand for Zn accumulation compared to other metals, and it is dependent on the crop's physiology. Besides, the following is the sequence in which heavy metals accumulate in certain vegetables: Zn and Cu: cabbage> lettuce> green bean> pepper; Cd: cabbage> lettuce> pepper> green bean; and Pb: cabbage = lettuce> pepper> green bean. Such patterns were almost true for FW-irrigated soils. This result suggests that cabbage has a higher capacity to absorb heavy metals than other vegetables, which is consistent with earlier research findings [10].

The sequence of average BCF (bio-concentration factor) values decreased in the order of Zn > Cu > Cd > Pb > in both WW and FW- irrigated soils, indicating that transfer from the soil to leafy greens was easier for Zn than for other metals (Figure 2). Depending on vegetable type, an increase of 5% to 44%, on average, was found in the BCF of all vegetables under WW-irrigation compared with FW-irrigated soil. The greatest increase was observed for BCF-Cu (20-44%), followed by BCF-Pb (19-32%), Cd (7-21%), and Zn (5-16%) implying that the uptake and transport of heavy metals in most vegetables has been stimulated and encouraged by WW irrigation. For all heavy metals, however, the BCF values were<1 in all vegetables under WW and FW irrigation, suggesting a low potential of the vegetables to accrete these metals. Differences in soil characteristics (e.g., soil texture, pH, EC, OM, and CEC), crop species, metal bioavailability, interface between plant roots and surrounding soil, and metal translocation and transpiration rate in root-leaf vegetables may all influence metals transport from the soil to crop organs [8].

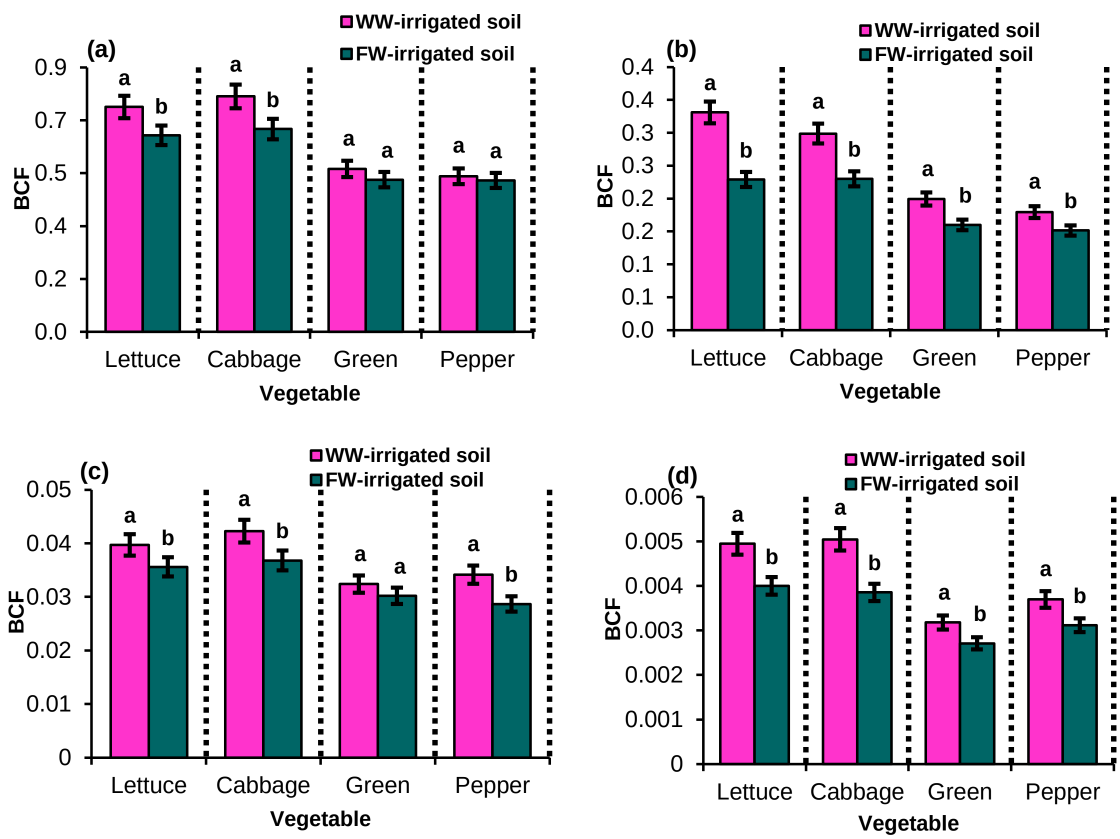



**Figure 2.** Means comparison of BCF values for Zn (a), Cu (b), Cd (c), and Pb (d)between WW-irrigated soils and adjacent FW-irrigated soil. For each vegetable, different letters show significant differences in ecological risk at P < 0.05 confidence level.

### *3.4.Health risk assessment*

#### *3.4.1. Non-carcinogenic risk*

The HQ and HI of heavy metals were employed to estimate non-carcinogenic risk in vegetables ingested by children and adults. Irrespective of the population group, HQ values in the vegetables irrigated with WW than vegetables irrigated with FW, ranging from 65.5 to 80.3% (HQ-Zn), 72.1 to 93.5% (HQ-Cu), 52.4 to 97.7% (HQ-Cd), and 74.1 to 94.1%. This shows that the possible human health hazards associated with the eating of WW-grown vegetables are a serious concern that should not be overlooked. Among vegetables irrigated with WW, the highest HI was found in cabbage (min = 0.3, max = 0.39, mean = 0.35) followed by lettuce (min = 0.29, max = 0.37, mean = 0.32), green beans (min = 0.2, max = 0.27, mean = 0.24), and pepper (min = 0.19, max = 0.25, mean = 0.23) in both children and adults. This information indicates that the risk of non-cancerous illnesses is minimal ($0.1 \leq HI < 1$) for the vegetable consumers in this region. The mean values of

HI were significantly elevated in the vegetables irrigated with WW than the vegetables irrigated with FW in both children and adults, ranging from 82.4-84,2% for lettuce, 88.2-94.7% for cabbage, 67-70% for green beans, and 66.7-69.2% for pepper (Figure 3). Among various metals, exposure to Cu (37-43%) was the largest contributor to the HI of the local population, followed by exposure to Cd, Zn and Pb, which accounted for 28–33%, 27-30%, and 0.9-1% of the total HI, respectively, for different vegetables (Figure 3). Copper, thus, would be the primary component leading to the possible health risk for those who consume WW-cultivated vegetables. Excessive copper consumption may have posed health risks to humans, including liver and kidney damage, anemia, immunotoxicity, and developmental toxicity [11].

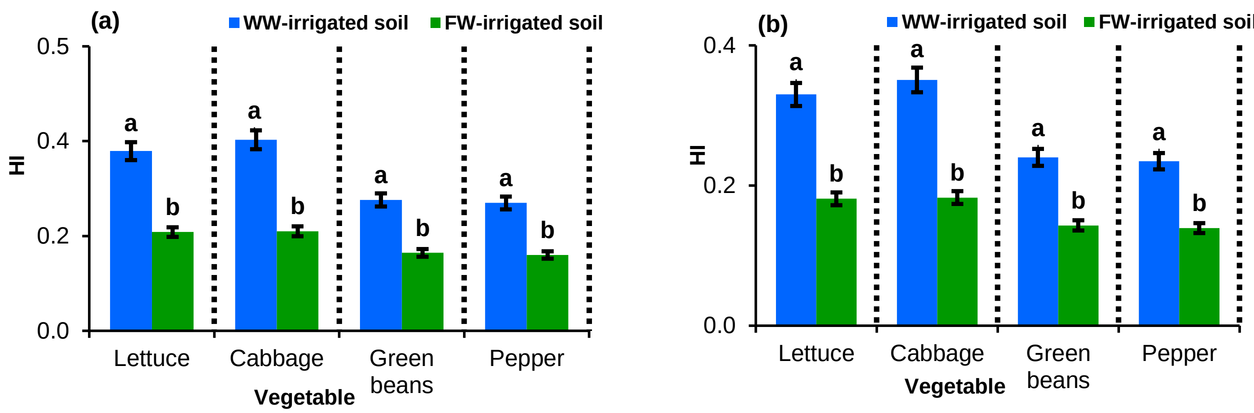
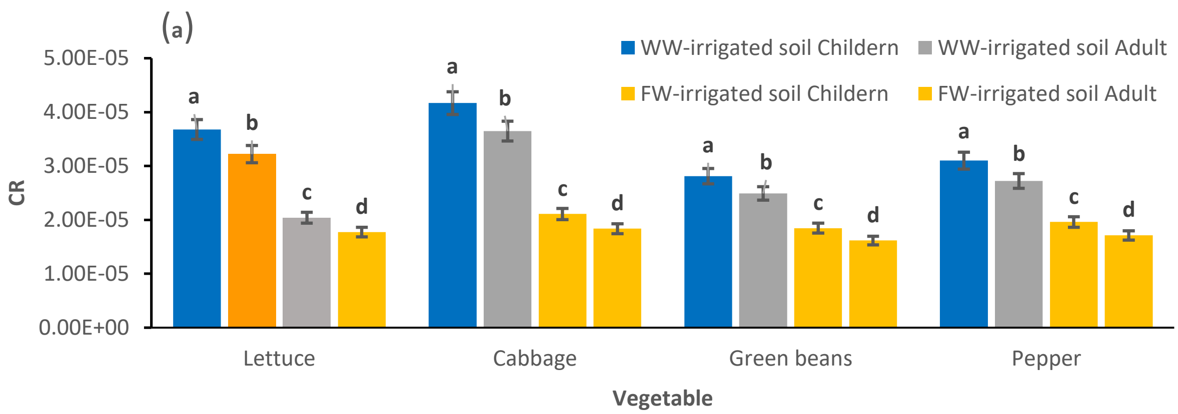



**Figure 3.** Comparison mean of Hi values between the irrigated farms with WW and FW for children (a) and adult (b). For each vegetable, different letters show significant differences at $P < 0.05$ confidence level.

### *3.4.2. Carcinogenic risk (CR)*

Cd and Pb were taken into account in this study while assessing cancer risk. The CR-Cd to children and adult population, via the eating of WW-grown vegetables was in the range of 2.98E-05 to 4.35E-05 and 2.59E-05 to 3.79E-05 for lettuce, 3.44E-05 to 4.81E-05 and 2.99E-05 to 4.19E-05 for cabbage, 1.83E-05 to 3.44E-05 and 1.59E-05 to 2.99E-05 for green beans, and 2.29E-05 to 3.667E-05 and 1.99E-05 to 3.19E-05 for pepper, respectively. These data suggest that CR is almost within the acceptable range (from 1.00E-06 to 1.00E-04) for vegetable-consuming children and adults in the region [9]. However, Cd had significantly higher CR than Pb for all the vegetables irrigated with WW (Figure 4) and its risk of cancer was 2.8: 100000 to 3.7: 100000 chance and 2.5: 100000 to 3.6: 100000 chance for children and adults, respectively. Children had substantially higher CRs of Cd and Pb than adults in all vegetables irrigated with WW and FW (Figure 4), demonstrating that children were more sensitive to cancer risks than adults. The total carcinogenic risk (TCR) for Cd and Pb was the highest for cabbage (min = 3.55E-05, max = 4.95E-05, mean = 4.29E-05) in children followed by lettuce (min = 3.55E-05, max = 4.95E-05, mean = 4.29E-05), pepper (min = 2.37E-05, max = 3.76E-05, mean = 3.19E-05), and green beans (min = 1.90E-05, max = 3.51E-05, mean = 2.89E-05). Such a sequence was observed for adults. Like CR-Cd and CR-Pb, the TCR values of children and adult population by the consumption of vegetables grown with WW were between 1.00E-06 to 1.00E-04, demonstrating that diverse demographic groups are acceptable with no substantial health risk associated with long-term intake of vegetables and processed foods. Given CR-Cd, CR-Pb, and TCR, the health risk of children and adults by consumption of vegetables irrigated with WW was significantly greater than those irrigated with FW (Figure 4), suggesting a significant negative influence of wastewater irrigation on human health.



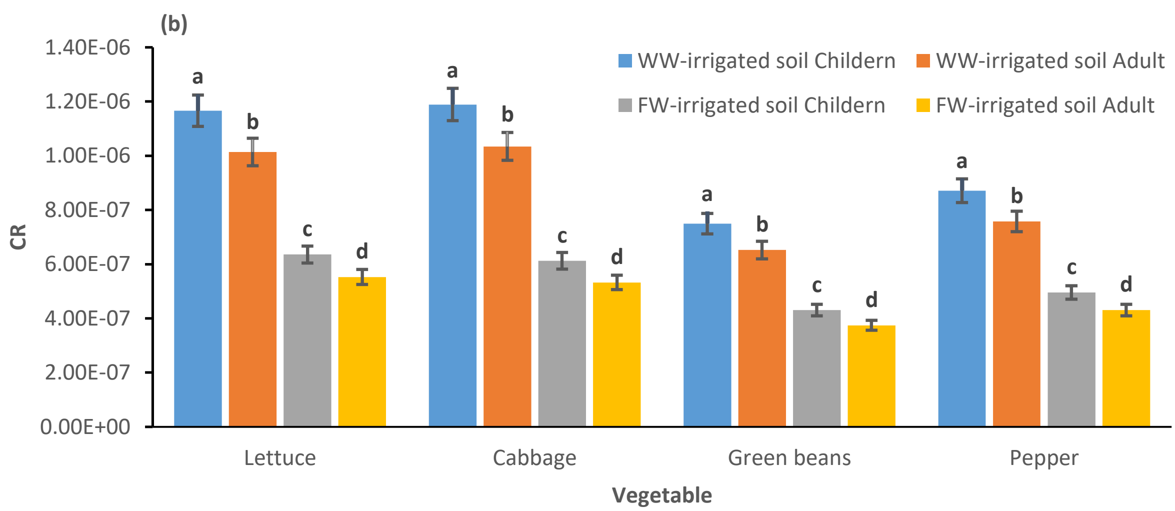
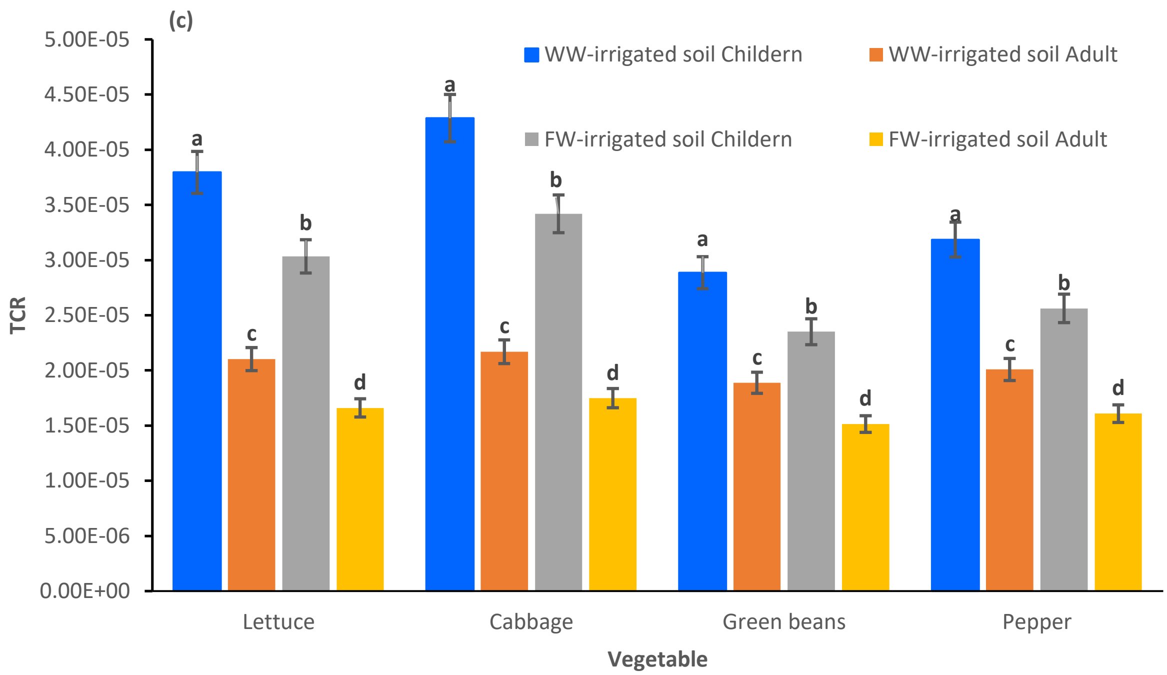




**Figure 4.** Means comparison of CR values for Cd (a) and Pb (b) and TCR values (c) between WW-irrigated soils and adjacent FW-irrigated soil in two different age groups. For each vegetable, different letters show significant differences at $P < 0.05$ confidence level.

**Figure 4 (continuous)**




## 4. Conclusion

Compared to the WW-irrigated soils, the metals concentration and PI value significantly increased in the order of Cd >Zn > Pb > Cu for all irrigated soils with WW. The value of ER was high class in the majority of WW-irrigated soils, which was mainly related to Cd, whereas it was moderate in the FW-irrigated soils, showing WW irrigation has fallen the ER by two grades. Among the vegetables irrigated with wastewater, the BCF value and heavy metal concentration were highest in cabbage, followed by lettuce, in comparison to other plants, showing cabbage has a greater capacity to absorb heavy metals. The results of the health risk assessment revealed that the HQ and HI values of vegetable consumers (adults and children) were <1 in the WW-irrigated soils, revealing that the local inhabitants are exposed to an insignificant non-carcinogenic risk. Although WW irrigation resulting in a substantial rise in the CR values caused by Cd and Pb and their combination (TCR) for both children and adults than those irrigated with FW, the carcinogenic risk of the metals was 2.5: 100000 to 3.6: 100000 chances, reflecting an acceptable range of CR and TCR (1.00E-06 <CR < 1.00E-04) for local inhabitants.

## References

[1] Tahtouh J, Mohtar R, Assi A, Schwab P, Jantrania A, Deng Y, Munster C. Impact of brackish groundwater and treated wastewater on soil chemical and mineralogical properties. Science of the total environment. 2019 Jan 10; 647:99-109.

[2] Yu MH, Tsunoda H. Environmental toxicology: biological and health effects of pollutants. crc press; 2004 Oct 28.

[3] Rezapour S, Atashpaz B, Moghaddam SS, Damalas CA. Heavy metal bioavailability and accumulation in winter wheat (Triticum aestivum L.) irrigated with treated wastewater in calcareous soils. Science of the Total Environment. 2019 Mar 15; 656:261-9.

[4] Klute A, Page AL. Methods of soil analysis. In: Klute, A. (Ed.), Chemical Methods. Part 3. American Society of Agronomy/Soil Science Society of America, Madison, WI, USA, 1996. pp. 417–475.

[5] Soon YR, Abboud, S. Cadmium, chromium, and nickel. In: Carter, M. R. (Ed) Soil Sampling and Methods of Soil Analysis. Lewis Publishers, Boca Raton FL, USA, 1993. pp. 101–108.

[6] Liu WH, Zhao JZ, Ouyang ZY, Söderlund L, Liu GH. Impacts of sewage irrigation on heavy metal distribution and contamination in Beijing, China. Environment international. 2005 Aug 1;31(6):805-12.

[7] Hakanson L. An ecological risk index for aquatic pollution control. A sedimentological approach. Water research. 1980 Jan 1;14(8):975-1001.

[8] Kabata-Pendias A. edition 4. Trace elements in soils and plants. Boca Raton, FL, USA: CRC Press/Taylor & Francis Group, 2010.

[9] EPA A. Risk assessment guidance for superfund. Volume I: human health evaluation manual (Part E, supplemental guidance for dermal risk assessment). EPA/540/R/99; 2004 Jul.

[10] Ji Y, Wu P, Zhang J, Zhang J, Zhou Y, Peng Y, Zhang S, Cai G, Gao G. Heavy metal accumulation, risk assessment and integrated biomarker responses of local vegetables: A case study along the Le'an river. Chemosphere. 2018 May 1; 199:361-71.

[11] Gupta N, Yadav KK, Kumar V, Kumar S, Chadd RP, Kumar A. Trace elements in soil-vegetables interface: translocation, bioaccumulation, toxicity and amelioration-a review. Science of the Total Environment. 2019 Feb 15; 651:2927-42.