HEALTH RISK OF ZINC POLLUTANT IN AGRICULTURAL SOIL IN SOME LEAVES OF SELECTED LEAFY VEGETABLES IN KIRKUK, NORTH IRAQ

¹Hassan A. A. Al-Jumaily And ¹Eman W.W. Al-Berzanje*

¹Department of Applied Geology, College of Science, University of Kirkuk, Iraq

*E-mail: Emanalberzanje@gmail.com

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ABSTRACT

Evaluation of the environmental geochemistry needs studying element content in different environmental profiles, for that reason the zinc content has been studied in the soil-plant to fill gaps in Kirkuk City. Ten soil samples (L1–L10) are collected from agricultural fields (0–20 cm depth), with twelve leafy vegetable samples for four types, and each type has three samples: celery (N1, N2, N3), basil (N4, N5, N6), cress (N7, N8, N9), and arugula (N10, N11, N12) from the fields (L2, L7, L8) which the soil sample were chosen. Samples were analyzed using ICP-MS technique. The results show the average concentration value of Zn in soil samples is 70.91 mg/kg the highest concentration is recorded in sample L7 (169.2 mg/kg), whereas in the plant samples the highest average of Zn was found in cress 67.73> basil 52.53> arugula 38.9> celery 29.3 in mg/kg, respectively. Transfer factor value showed that the highest value in Cress 0.729> TF Basil 0.612> TF Arugula 0.511> TF Celery 0.395, respectively. Igeo result values show that all soil sampling sites are unpolluted with Zn, except L7 is unpolluted to moderate with Igeo value (0.8). The contamination factor values in L7, L8, L9, L10 samples (2.603, 1.284, 1.086 and 1.129) refers to a moderate contamination level with Zn. Target hazard quotient values for children >1 in samples N4, N5, N7, N8, N9, N10, and target hazard quotient values for adult >1 in samples N5, N8, N9, that indicate they are exposed to different health effects. The increase in the concentration of the Zn in soils is due to irrigated field crops with wastewater, and through it, the plant absorbs it by root depending on the type of plant, the location of the sample, and the soil properties.

Keywords: Zinc; Transfer factor; Leafy vegetable; Pollution level; Health risk assessment

INTRODUCTION

Zinc (Zn) is a chemical element discovered by German scientist Andreas Marggraf in 1774 (Heiserman, 1992), the geochemical principles for it is located in period four and group twelve

in the periodic table, with atomic number 30, atomic mass 65.38, atomic radius 153 pm, and density 7.13 gm/cm^3 (Reimann and Caritat, 1998 and Kabata- Pendias, 2011). Contamination with heavy elements in worldwide has disrupted the environment and causes health impact and risks to humans (Rai et al., 2019). Nu et al. (2020) mentioned that soil salinization is a serious problem that restricts agricultural productivity worldwide due to climate change. A heavy element including zinc is accumulating in the top layer of the soils (Kooner et al., 2014). Soil is an important basin for many nutrients and pollutants in it, which plays an important role in environmental and social safety and stability (Wu et al., 2018). In an uncontaminated environment with zinc source is from weathering source rocks, deposition from the atmosphere, agricultural fertilizers, and accumulates over time (Imseng et al., 2019). Continuous increase in the levels of heavy elements in the soil results from practices agricultural and anthropogenic sources (Baltas et al., 2019). The most important anthropogenic that release the Zn to the environment are the wastewater, fertilizers, pesticide (Mičijević et al., 2019), Where absorption the Zn from soil and water by root tissue and transferred it to other parts of leafy vegetables (Gupta et al., 2019), that transferred to plant in form of Zn^{+2} or organic acids (Hefferon, 2019), and available for absorption by ionic exchange (Gupta et al., 2016), it occurs as a natural constituent in plant tissues (WHO/FAO, 2017), and it is a major micronutrient and necessary for the natural growth of agricultural crops (Akhtar et al., 2019), it participates in several major physiological functions as formation of chlorophyll, structure of membrane, immunity against drought, disease and photosynthesis (Noulas et al., 2018). The high concentrations of Zn may cause toxic for plants and microorganisms that live in the soil and this in turn has an impact on human health (Alloway, 2013). Al-Hamadani et al., 2016 mentioned that the organic fraction in Kirkuk soil contained the next significantly portion of metals, with the following decreasing order: Cd, V, Ag, Cu and Cr whereas Fe-Mn oxide fraction was contained high levels of Zn and Co. Cd and Pb occupied the important portion of carbonate fraction. With the exception of residual fractions, most metal fractions decrease with soil profiles, but there are some metal such as Ag, Zn and V increase. The main pathway of human exposure to Zn is via eating vegetables (Akande and Ajayi, 2017). In wise of human health, Zn is one of the most important trace elements for organism (WHO/FAO, 2017), it has three biological roles: organizational, structural, and catalytic (Akhtar et al., 2019 and Chasapis et al., 2012). Zinc is essential for neurological, immune system function, DNA and protein synthesis, cell growth and division, male and female reproduction (Alloway, 2013), and has an important role in taste acuity, blood clotting, wound healing, sperm production, fetal growth, bone hardening, and thyroid gland functions (Chasapis et al., 2012), it also stimulates growth, height, weight, smell, appetite,

vision, skin, and hair (Das and Green, 2016). Zinc is available in all tissues, cells and fluids in human body and estimated around 2 gm (Das and Green, 2016 and FAO/WHO, 2001) in lung it is 10 mg/kg and in liver equals to 57 mg/kg (Kabata-Pendias and Mukherjee, 2007). The Zn average daily intake has been estimated for adults by food chain to be maximally 20 mg/d, if the taken dose more than 4-8 gm, may cause poisoning with main symptoms are vomiting, nausea, diarrhea, and fever (, 2017). There is an urgent need to assess the soil pollution with heavy element and determine their sources to meet the risks that they cause on soil quality, food safety and human health (Ren et al., 2019). The aims of the current study are determination the concentration of zinc in agricultural soil and leafy vegetable samples (Celery, Cress, Basil, Arugula) in agricultural field crops, determine transfer factor (TF) of leafy vegetables of zinc from soil to plant, pollution assessment of Zn in soil samples, and Assessing the health risks of zinc for children and adults via their consumption of leafy vegetables in the Kirkuk city.

MATERIALS AND METHODES

Location of Study Area

The study area Kirkuk city is located 250 Km north of the capital city Baghdad, between latitudes ($35^{\circ} 20' 00''$ N- $35^{\circ} 40' 00''$ N) and longitude ($44^{\circ} 15' 00''$ E- $44^{\circ} 30' 00''$ E) (Fig. 1). The sides of Al Khassa River are used for growth crops there, the sampling site position coordinates recorded by GPS (Global Position System) (Table 1).

Fig. 1. Location of study area and distribution of sample points (Arc GIS)

Sample Preparation

Ten agricultural bulk soil samples are selected from study area crop fields, and collected from 0-20 cm depth across selected sites throughout the study area in September 2019 by using auger, they are kept in closed bags with written sample symbol on them. Totally twelve leafy vegetables samples are collected from three different fields with three sample for each four kinds of plant Celery, Basil, Cress and Arugula, and coding each sample from vegetables. These plant samples were selected from soil sampling sites (L2, L7, L8) (Table 1) (Fig. 1). The vegetables were chosen for their abundant in the study area and consumed by local population, that irrigated from Al Khassa river water, which is a seasonal river that sewage water flows into, the soil and vegetable samples were collected from those fields that distributed on both sides of Khassa River. The preparation of all collected bulk soil samples (mixture of irregularly sized fraction granules) include drying it in room temperature then grinding and sifting the samples through a sieve with 2 mm mesh size, then stored in polyethylene bags until further laboratory analysis. While, vegetable samples were washed by distilled water, dried at room temperature and then saved in polythene bags. Thus, all the samples for both of soil and plant are prepared according to the laboratory requirements and being ready for chemical analysis to determine zinc concentration using an Inductively Coupled Plasma-Mass Spectrometer (ICP-MS) technique at Acme Laboratory in Canada.

Table 1. Coordinate's and symbols for collected soil and leafy vegetable samples

Samples			Location	
Number	Type	Location	Lat.	Long.
L1	Soil	Near Lilan Bridge	N35° 23' 24"	E44° 21' 44"
L2	Soil	Opposite to 1st of March district	N35° 23' 47"	E44° 22' 02"
N1, N4, N7, N10	Plant			
L3	Soil	Behind galaxy hall	N35° 24' 07"	E44° 22' 21"
L4	Soil	Opposite to Aden district	N35° 24' 20"	E44° 22' 43"
L5	Soil	Opposite to life fitness hall	N35° 24' 42"	E44° 22' 53"
L6	Soil	Opposite to Almualmeen district	N35° 25' 05"	E44° 23' 03"
L7	Soil	Opposite to Almansour mosque	N35° 27' 30"	E44° 23' 26"
N2, N5, N8, N11	Plant			
L8	Soil	Behind AL rehab games city	N35° 28' 25"	E44° 23' 34"
N3, N6, N9, N12	Plant			
L9	Soil	Behind Imam qasim footbridge	N35° 28' 38"	E44° 23' 40"
L10	Soil	Sarchnar	N35° 29' 14"	E44° 24' 35"

Transfer Factor of Zinc

The most important paths that human exposure to health risk from diet are transfer of heavy metals from soil to plant tissue (Jolly et al., 2013), and this transfer of the element from soil to plant is measured by transfer factor (TF), where it is an indicator for assessing the movement

of heavy metal, and its value varies depending on the environment and type of plant (Cervantes-Trejo et al., 2018). TF is used to assess the potential ability of the plant to transfer the elements from soil to the plant edible tissues (Proshad et al., 2019). Transfer factor of Zn from agricultural soil-leafy vegetables samples were calculated as follow equation (Harrison and Chirgawi, 1989 and Cui et al., 2004):

$$TF = \frac{C_{plant} \text{ (dry wt)}}{C_{soil} \text{ (dry wt)}} \quad (1)$$

Where, C_{plant} and C_{soil} are representing the zinc concentration in the edible part of plant tissue (mg/kg)/ zinc concentration in soil (mg/kg) dry weight basis respectively, from where the plant was grown (Harrison and Chirgawi, 1989). The ratio of $TF > 1$, indicate that plants are enriched in the element and accumulate the element in edible parts (Rehman et al., 2017), $TF \sim 1$ indicate uniformity in the absorption process and transport the elements to the plant, and the ratio of $TF < 1$ indicate that plants excluding the elements from absorption (Olowoyo et al., 2010).

Pollution Assement of Zinc in Soil Samples

The geo-accumulation index (I_{geo}) is used to evaluate pollution of heavy elements in soil and sediments (Adimalla and Wang, 2018), where calculated by follow equation that introduced by (Müller, 1986):

$$I_{geo} = \log_2 \left[\frac{C_n}{1.5 \cdot GB} \right] \quad (2)$$

Where, C_n is the measured concentration of individual heavy element (Zn) in soil (mg/kg); 1.5 is the multiplication factor reduces the effect of possible variation of lithogenic effects in the background value (Müller, 1986); GB represent geochemical background of elements in earth crust, which is 65 mg/kg for zinc according to Wedephol (1995). According to Müller (1986), the I_{geo} index for each metal consists of seven grades as: unpolluted ($I_{geo} \leq 0$); unpolluted to moderately polluted ($0 < I_{geo} < 1$); moderately polluted ($1 < I_{geo} < 2$); moderately to strongly polluted ($2 < I_{geo} < 3$); strongly polluted ($3 < I_{geo} < 4$); strongly to extremely polluted ($4 < I_{geo} < 5$) and extremely polluted ($5 < I_{geo}$).

Contamination factor (C_f) is individual index utilized to evaluate soil contamination by Håkanson (1980) according to follows equation:

$$C_f = \frac{C_n}{C_{BG}} \quad (3)$$

Where, C_n is the measured content of zinc in soil (mg/kg); C_{BG} represents pre-industrial element concentration, and in general the elements concentration in the earth's crust is compensated as element concentration before the influence of industrial processes (Loska et al., 2004), which is 65 mg/kg for zinc (Wedepohl, 1995). Where C_f value is classified into five contamination classes according to Håkanson (1980): low contamination ($C_f < 1$); moderate contamination factor ($1 \leq C_f < 3$); considerable contamination factor ($3 \leq C_f < 6$) and very high contamination factor ($6 \leq C_f$).

Health Risk Assessment of Zinc in Leafy Vegetables

The health risks of heavy metal via consumption of leafy vegetables by local inhabitants were assessed by target hazard quotient (THQ) (Wang et al., 2005). The THQ was determined based on methods by Chien et al. (2002), that described as follows equation:

$$THQ = \frac{C_i * E_f * E_d * VI}{W * T_a * Rfd} \quad (4)$$

Where, C_i is the zinc concentration in leafy vegetables (mg/kg); E_f is the exposure frequency (350 days/yr); E_d is the exposure duration (70 yr); VI is the vegetable ingestion rate for children and adult (0.232 kg/person/day)(0.345 kg/person/day) respectively (Wang et al., 2005); W is the average body weight (32.7 kg) for children and (55.9 kg) for adults as used in previous researches (Wang et al., 2005); T_a is the noncarcinogens average time ($E_d * 365$ day/yr); Rfd is the oral reference dose, ($Zn = 0.3$ mg/kg/day) (Chien et al., 2002). If value is less than one ($THQ < 1$) that means the population haven't exposed to health effects (Wang et al., 2005 and Chien et al., 2002), while, if THQ value is equal to or greater than one ($THQ \geq 1$) that means there are potential health risks from exposure and must be taken deterrent to her (Islam et al., 2014 and Adedokun et al., 2016).

RESULTS AND DISCUSSION

Zinc Content in Soils and Vegetables

The concentration of zinc in the study area agricultural soils (mg/kg) is presented in Table 2. Total Zn content ranges from 48.1-169.2 mg/kg. Mean of Zn concentration in soils equals to 70.91 mg/kg higher than the total Zn mean concentration for worldwide soils (55 mg/kg), (70 mg/kg), and (64 mg/kg) as Alloway (2008), Kabata-Pendias (2011), and Kabata-Pendias and Mukherjee (2007) respectively, the source of Zn in the studied samples is anthropogenic.

Table 2. Zinc average and concentration in agricultural soil of the study area

Sample No.	Zn (mg/kg)
L1	48.1
L2	52.8
L3	54.2
L4	58.6
L5	50.6
L6	48.1
L7	169.2
L8	83.5
L9	70.6
L10	73.4
Range	48.1-169.2
Average	70.9

Maximum content of zinc is observed in site L7 then L8, L9 and L10 samples (169.2, 83.5, 70.6, 73.4) in mg/kg respectively, this increases are due to using wastewater for irrigation the field crops, this interpretation corresponds to the study of (Awadh and Al-Hamdani, 2019; Khan et al., 2014 and Khan et al., 2008), and use of agricultural fertilizers or vehicle traffic (Ozyazici et al., 2017), or enrichment by intensive agricultural activities for long time (Liu et al., 2019), but notice Zn concentration in L1, L2, L3, L4, L5, and L6 samples are lower than the content from the above mentioned samples (48.1, 52.8, 54.2, 58.6, 50.6, and 48.1) in mg/kg respectively, that indicate these sampling sites are not under the exhibition of any anthropogenic or industrial source that increase Zn content in the soil.

The zinc concentrations in four kinds of vegetable samples for each location are listed in Table 3. Zinc total concentration in leafy vegetable samples ranges from 18.9-95.3 mg/kg, with mean concentration of 47.11 mg/kg. In detail, Zn concentration ranged from 18.9-35.5 mg/kg in celery, 41.3-66.1 mg/kg in basil, 47.9-95.3 mg/kg in cress, and 33.1-49.4 mg/kg in arugula samples. According to the results, the highest average of Zn was in Cress 67.73> Basil 52.53> Arugula 38.9 > Celery 29.3 mg/kg (Table 3) and (Fig. 2), this increasing in Zn concentration is attributed to irrigation fields with wastewater (Ismail and Rashid, 2017), where the concentration of Zn in same or different type of vegetables are varied from one sampling location to another is dependent on the Zn concentration in sampling soil location (Mohamed et al., 2003), and ability of vegetable to absorb the zinc through their roots and transfer them to different parts of plant from soil (Kananke et al., 2015).

Fig. 2. Average zinc concentration of leafy vegetables in the study area

Table 3. Zinc concentration and transfer factor (TF) values of leafy vegetable in study area samples

Sample No.	Plant type	Zn concentration in soil (mg/kg)	Symbol of Sample	Zn concentration in plant (mg/kg)	Symbol of Sample	TF
1	Celery 1	52.8	L2	35.5	N1	0.672
2	Celery 2	169.2	L7	18.9	N2	0.111
3	Celery 3	83.5	L8	33.5	N3	0.401
Av.				29.3		0.395
4	Basil 1	52.8	L2	50.2	N4	0.950
5	Basil 2	169.2	L7	66.1	N5	0.390
6	Basil 3	83.5	L8	41.3	N6	0.494
Av.				52.53		0.612
7	Cress 1	52.8	L2	47.9	N7	0.907
8	Cress 2	169.2	L7	95.3	N8	0.563
9	Cress 3	83.5	L8	60	N9	0.718
Av.				67.73		0.729
10	Arugula 1	52.8	L2	49.4	N10	0.935
11	Arugula 2	169.2	L7	34.2	N11	0.202
12	Arugula 3	83.5	L8	33.1	N12	0.396
Av.				38.9		0.511
Range				18.9-95.3		0.111-0.950
Total Av.				47.11		0.562

Transfer Factor of Zinc in Leafy Vegetables

Calculated transfer factor (*TF*) values for Zinc in various leafy vegetables of the study area samples are varied between plant kinds (Table 3). The *TF* total value of Zn ranged from (0.111-0.950), with total average (0.562) for all kinds of plant. *TF* values ranged from 0.111-0.672 in celery, 0.390-0.950 in basil, 0.563-0.907 in cress, and 0.202-0.935 in arugula. Where record the highest average in transfer ability of zinc in cress and lower value is in celery (*TF* Cress 0.729>

TF Basil $0.612 > TF$ Arugula $0.511 > TF$ Celery 0.395) respectively, the difference depends on the plant type and their environment (Swasti Prabasiwi et al., 2020), and sampling site to another (Kananke et al., 2015).

The vegetable samples N4, N7, N10 that were growth on soil sample L2 (field opposite to 1st of March), Zn concentration reached 52.8 mg/kg , from that observed is very near to concentration value in vegetables as basil 1 (N4= 50.2 mg/kg); cress 1 (N7= 47.9 mg/kg); arugula 1 (N10= 49.4 mg/kg), and the transfer factor values in the vegetable samples are around one ($TF \sim 1$), this indicates to uniformity in transfer of the zinc from soil to leafy vegetables in this site samples, and that convergence in concentration is depending on the properties of the soil in the sampling area and its environmental conditions (Rehman et al., 2017).

Pollution Assessment of Zinc in Soils

Pollution assessment indices of zinc in agricultural soil samples are shown in Table 4. All sample site is unpolluted with zinc, whose I_{geo} values ranged from -1.02 - -0.22 , which is within the class ($I_{geo} \leq 0$), except in sample L7 the I_{geo} value for it was 0.8 , that is in class ($0 < I_{geo} < 1$), indicate unpolluted to moderately polluted site with Zn due to occurrence of wastewater.

Table 4. Geo-accumulation index (I_{geo}) and contamination factor (C_f) values in studied soil samples

Sample No.	I_{geo}	C_f
L1	-1.02	0.74
L2	-0.88	0.812307692
L3	-0.84	0.833846154
L4	-0.73	0.901538462
L5	-0.94	0.778461538
L6	-1.02	0.74
L7	0.8	2.603076923
L8	-0.22	1.284615385
L9	-0.46	1.086153846
L10	-0.41	1.129230769

While contamination factor with Zn results show the soil samples (L1, L2, L3, L4, L5, and L6) have C_f value (0.74 , 0.81 , 0.83 , 0.9 , 0.77 , and 0.74) respectively, indicate low contamination level by Zn in this site ($C_f < 1$), and the sites (L7, L8, L9, L10) have C_f value (2.603 , 1.284 , 1.086 and 1.129) in class ($1 \leq C_f < 3$) with moderate contamination level by Zn, where the highest C_f value is recorded in L7 sample, this increase is due to irrigation by waste water.

Health Risk Assessment of Zinc in Leafy Vegetables

The THQ results for children and adults from zinc content in study area leafy vegetable samples for each type (Table 5). Where the THQ for zinc in celery for children was higher than the adults (THQ celery child > THQ celery adult), but generally the THQ values are less than one (THQ celery <1), that indicate the local population is not exposed to health effect from Zn. Where THQ value for Zn in basil for children in sample N4 and N5 is 1.138 and 1.498 respectively, while for adults in sample N5 is 1.303. THQ basil >1 indicate the children and adults in this samples are exposed to health effect from Zinc compared with another sample.

Table 5. THQ of zinc for children and adults in leafy vegetable

Sample No.	Plant type	THQ child	THQ adult
N1	Celery 1	0.805	0.7
N2	Celery 2	0.428	0.372
N3	Celery 3	0.759	0.66
N4	Basil 1	1.138	0.99
N5	Basil 2	1.498	1.303
N6	Basil 3	0.936	0.814
N7	Cress 1	1.086	0.944
N8	Cress 2	2.161	1.879
N9	Cress 3	1.36	1.183
N10	Arugula 1	1.12	0.974
N11	Arugula 2	0.775	0.674
N12	Arugula 3	0.75	0.652

As well as THQ value for children and adults in cress samples (N7, N8 and N9) are greater than one (THQ child cress >1), that local population are exposed to health effect from zinc in that sites, except THQ for N7 sample adults is less than one. Regarding arugula sample N10 the THQ value for children is 1.120, that exposed to the zinc create health effect, while THQ for another samples is smaller than one, that indicate Zn did not exposed any health effects for local population.

CONCLUSIONS AND RECOMMENDATIONS

This study examines the zinc concentration in the agricultural soils in which the leafy vegetables grow and these vegetables are consumed by local population in the Kirkuk city, by assessing pollution indexes on Zn content in the soil, and health risks of Zn that may cause in four kinds of leafy vegetables to consumer. The reason for increasing of zinc concentration in soil samples L7, L8, L9, and L10 (169.2, 83.5, 70.6, 73.4 mg/kg) are mainly due to the drainage of waste water into Al Khassa River and field crops irrigated with this water then runoff to the soil, uses

agricultural fertilizers, enrichment by intensive agricultural activities, and vehicle traffic as well. Pollution assessment result *Igeo* value is proved that was moderately polluted with Zn in sample L7 (0.8), and uncontaminated to moderately contaminated factor C_f value for zinc in L7, L8, L9, and L10 samples (2.603, 1.284, 1.086, 1.129) respectively. While the Zn concentration in vegetable samples records the highest average in Cress 67.73> Basil 52.53> Arugula 38.9 > Celery 29.3 in mg/kg respectively, then this results corresponds to the transfer factor results (TF Cress 0.729> TF Basil 0.612> TF Arugula 0.511> TF Celery 0.395) in terms of the sequence of vegetable kinds, which is the reason for its increased content in celery compared with other type is the concentration of zinc in the soil, ability to transfer it, absorption it from the soils by roots, type of vegetables, and soil properties. Through an assessment the health risks of Zn on local population for both age groups via consumption leafy vegetable studied samples, results show the THQ values for children (THQ child >1) in samples N4, N5, N7, N8, N9, N10 except N1, N2, N3, N6 and for adult THQ values (THQ adult >1) in samples N5, N8, N9 except N1, N2, N3, N4, N6, N7, N10 they are exposed to different health effects. Sum up and suggest the recommendations from the results obtained, it shall not be irrigated vegetable fields with wastewater that source from Khassa River, and reduce the use of fertilizers, that in order to increase the zinc concentration which leads it to absorption by studied vegetables and effect on the human health of local population.

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