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ACCUMULATION OF HEAVY METALS IN CELERY PLANT APIUM GRAVEOLENS AND SOIL IRRIGATED WITH WASTEWATER WITHIN DUHOK CITY KURDISTAN OF IRAQ

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ABSTRACT

Wastewater contains poisonous metals which can be moved and accumulated in plants before entering the human body through the food chain. The aim of this research was to investigate the concentrations of toxic metals such as Nickel (Ni), lead (Pb), and copper (Cu) in wastewater, celery plant and farmlands soils of Duhok city Kurdistan of, Iraq. The heavy metals accumulation in the Celery plants, soil and water samples were analyzed by using an atomic absorption spectrophotometer. The results obtained showed that the mean concentrations of Pb, Ni, and Cu in the wastewater and Celery plants samples ranged from 0.45±0.08 to 5.95±0.2, 0.18±0.01 to 1.95±0.28, 0.02± 0.01 to1.13±0.23 mg/L respectively. The pattern of metal buildup in wastewater-irrigated soil is in the order: Cu > Pb > Ni, and the mean concentration of Pb, Ni and Cu in soil ranged between 21.68±0.42 to 118.57±0.07, 1.27±0.47 to 5.70±0.23, 1.28±0.24 to ± 15.12 0.53 mg/kg-1, respectively. While the mean concentration of Pb, Ni and Cu in celery leaves ranged from 1.04±0.22 to 5.22±0.60, 0.52± 0.22 to 4.31± 0.12, respectively. 1.34 ±0.22 to 19.47±0.82, 1.23±0.04 to 7.29± 2.53, 1.01±0.05 to 5.76±1.32 mg/ kg-1, respectively. According to this study, roots contain more heavy metals than leaves. According to the findings, a few of the sampling sites had Pb, Ni, and Cu values that were exceeded the permissible concentration. Celery plants cannot be planted in the Duhok Valley to prevent excessive heavy metal exposure to human health through vegetables, where the main irrigated water source is sewage from the local municipalities. Celery plant irrigation with wastewater has much greater levels of heavy metals than the controls.

KEYWORDS: Wastewater irrigation · Heavy metals · Celery plant · Contamination.

1. INTRODUCTION

Researchers from all around the world are indeed very concerned about the high levels of harmful substances found in soil, water, and vegetables, that are related to potential dangers to human health. Vegetable contamination by heavy metals frequently occurs, (Tariq, 2021). The term "heavy metals" often refers to metals that have a particular density of more than 5 g/cm3 and have a negative effect on the environment and living things (Jabeen et al., 2020). Heavy metals are deposited on plants via the air, polluted soil, and polluted irrigation water. Water. Urban vegetable cultivation frequently comes with a risk to human and animal health, especially in Iraqi nations, wherein rigorous attention is not made to food pollution. Farmers use wastewater that is high in dangerous heavy metals like cadmium (Cd), chromium (Cr), and copper (Cu)to irrigate their farms, nickel (Ni), Iron (Fe), lead (Pb), and zinc (Zn) manganese (Mn) are elements (Zn). (Abdel and Ibrahim,2018). The majority of the farmers in the study region were illiterate, and their only source of irrigation was unclean sewage. Water scarcity is a crucial issue for agricultural output in dry areas around the globe. Where water is not available, wastewater is frequently used for irrigation (Jun et al., 2018). *Apium graveolensL* is a member of the Apiaceae family of plants. The stems and leaves (petioles) are used raw or cooked in salads and soups. Celery has numerous health-promoting compounds, including dietary vitamins, fibre, minerals, and tryptophan amino acid (Helaly, 2014). The study analyzed the heavy metal level of selected Celery plants in Duhok administrative districts. Nonetheless, certain heavy metals receive special global attention. Due to its toxicity in the body when used large quantities. Because of their toxicity, biomagnification and bioaccumulation in the food chain, pollution with these metals pose a grave concern. (Dingkwoet et al., 2013). The concentration of heavy metals in wastewater is generally below the allowable limits, but long-term irrigated with wastewater effluents raises heavy metal concentrations in soil. The primary factor influencing the transfer of heavy metals from soil to plants is the nature of the sources. (Chaoua et al., 2018). The majority of farmers in the study area were illiterate, and their only source of irrigation was untreated wastewater. Water scarcity is a critical issue for agricultural production in arid regions around the globe. Where water is in short supply, wastewater is frequently used for irrigating. (Sajida et al., 2012). In order to characterize the extent and concentrations of heavy metals in wastewater, soils, and Apium graveolensL Celey plants, the present research was conducted.

2. MATERIALS AND METHODS

2.1 Study Area

The current study was conducted in the governorate of Duhok in Kurdistan. Region, Iraq (Fig 1). Forty-nine soil samples were taken from various locations. During the time (May 2021 to November 2021), monthly samples were collected. A total of 49 samples of wastewater, 49 samples of top-soil (5–15 cm depths) and 49 samples of Celery plants (Apium graveolens) were collected randomly between May and November 2021 from celery fields in Duhok, Iraq, in order to determine the total heavy metal content (Pb, Ni, and Cu) within those samples.

Chemical Analysis of Heavy Metals: The concentrations of Pb, Ni and Cu in the filtrate of digested water, soil, and the plant was estimated by using an Atomic Absorption Spectrophotometer chemical laboratory university of Zakho According to APHA 2005.

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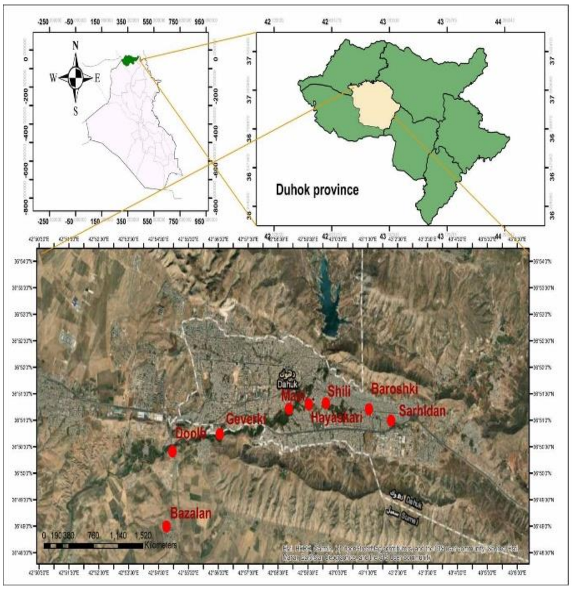

Figure 1: Illustrates the sampling location in Duhok valley.

2.2 Preparation of Samples

A. Total Dissolved solids (TDS) in mg/l: the amount of the total dissolved solids in water was estimated by TDS meter lonlab EC, TDS, level HANNA instrument WTW.

B.Biochemical Oxygen Demand (BOD5): Biochemical Oxygen Demand (BOD) in mg/l: the water sample was saved in an incubator for 5days under 20◦C after that determined dissolved oxygen by Winkler method (Azide modification) BOD5 = DOI – DOF as described by APHA, 2005 the results were reported by mg/l.

C.Phosphate: is measured according to APHA ,2005. used in ultraviolet spectrophotometer model 6800 UV/vis, Jenway, the results were expressed in mg /l.

D.Nitrates (NO3-) in mg/l: Nitrate ions were measured according to APHA ,2005 by using 2ml HCl (1N) added to the diluted sample (5ml of sample to 50ml deionized water), then measured by UV-spectrophotometer at wavelength 220 nm. Model 6800 UV/vis, Jenway.

.E. Water samples: In the laboratory, water samples were filtered with 0.45 m filters and preserved with 1 ml of 60% HNO3. The filtered samples were refrigerated but not frozen to minimize volatilization and biodegradation before analysis.

F. Vegetable samples: Vegetable samples were digested according to the procedure used by (Tandon, 1999). 0.5 g of finely grounded powder was wet digested in a 100-ml conical flask by adding 10 ml mixture of nitric acid and perchloric acid in a ratio of 9:4 on a hot plate in the digestion chamber (fume hood). Heating and digestion continued until the liquid became colorless. The liquid further heated to the volume 2-3 ml, then kept aside to lose the heat, then diluted by distilled deionized water in a 50 ml volumetric flask. Finally, the diluted sample was filtered by the filter paper and was stored in a polyethylene bottles for measuring heavy metals.

G. Soil samples: There were a total of 49 soil samples Three samples from top soil (0-30 cm) were collected from the farms irrigated by waste water. Air dried then crushed, sieved through 2-mm sieve and stored in polyethylene bags in order to be digested afterword. Sieved soil samples were grounded finely by a handle mill. 0.5 g of grounded soil was weighted and digested in a 50-ml conical flask by adding 10 ml of a mixture of sulfuric acid, nitric acid, and perchloric acid with a ratio of 3:1:1 respectively on a hot plate at 200˚ C in the digestion chamber (APHA, 1958). Heating continued until a color obtained, then diluted with distilled deionized water in a 50-ml volumetric flask and filtered by the filter paper. Filtered solution was stored in a polyethylene bottles and kept in the refrigerator until analysis.

2.3 Statistics Analytsis

Analysis of variance was used to conduct statistical analysis of the data; LSD 0.05 was used to determine significant differences across time periods and hospitals via SPSS version 19. All data were reported as mean, standard error.

Table 1: The latitudes and longitudes of sampling site of Duhok rivulet.

Samples	Location	Latitudes (North)	Longitudes (East)
Sample 1	Sarhldan	36.51'09"N	43.01'34"E
Sample 2	Baroshke	36.50'59"N	43.02'17"E
Sample 3	Shili	36.51'19"N	43.00'03"E
Sample 4	Hayaskari	36.51'17"N	42.59'32"E
Sample 5	Mazi	36.51'13"N	42.58'52"E
Sample 6	Geverke	36.50'44"N	42.56'34"E
Sample 7	Doolb	36.50'24"N	42.54'58"E
Sample 8	Bazalan	36.48'59"N	42.54'44"E

3. RESULTS AND DISCUSSION

3.1 Total dissolved solids (TDS) mg/l

Total dissolved solids (TDS) is the best individual value representing the salinity of the water. Figure (2) shows that the minimum value of 434.86 0.34 mg/l was recorded at Shili Site in October. While the maximum value of 1189.94 0.24 mg/l was recorded at Sarhldan Site in September. This increase is caused by domestic effluent and surface run-off from the cultivated fields, which might have increased the concentration of ions. All of the recorded values were within the minimum level of irrigating water standard for irrigation recommended by WHO and EPA 1000 mg/l EPA (2006). Except in Sarhldan Site.

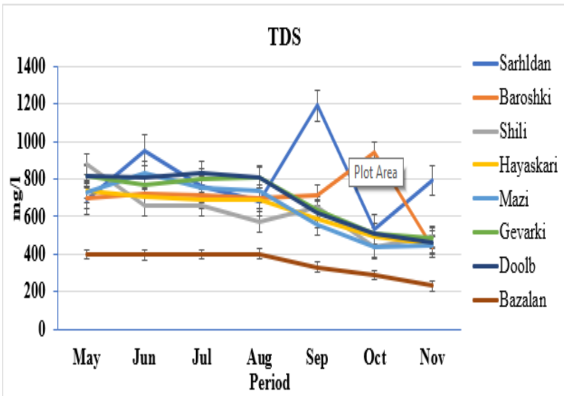

Figure 2: Monthly variation in TDS (mg/L) among wastewater for the study period.

3.2 Biochemical Oxygen Demand (BOD) mg/l

Figure (3) shows the highest concentration of BOD is recorded in Doolb Site was 147.43±0.13 mg/l in May, while the lowest value was recorded in Sarhldan Site was 44.04±0.55 mg/l in November, the increase result in Duhok valley may be due to domestic, industrial and agricultural waste discharge to Duhok Valley which contains organic matter different types of pollutants (Osibanjo et al., 2011).

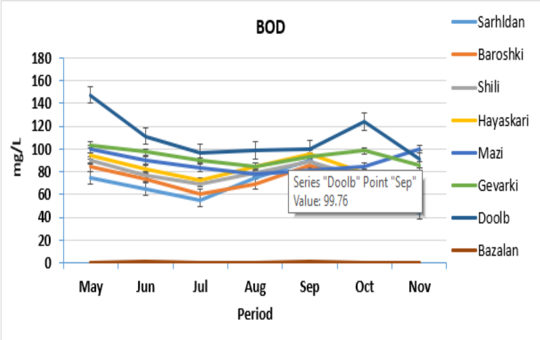

Figure 3: Monthly variation in BOD (mg/L) among wastewater for the study period.

3.3 Nitrate (NO3-) in mg/l

The seasonal variations of nitrate in the Duhok Valley are shown in Figure (4). Nitrate is essential for plant growth. During the study, nitrate in all seasons minimum range was recorded in the Sarhldan Site 0.21$\pm$ 0.48 mg/l in November, and maximum range recorded in the Doolb Site was 4.54 $\pm$0.04 mg/l in June. All the wastewater samples collected in the study area were within the permissible limit of nitrate which is 45 mg/l, (WHO, 2006). The sources of nitrate concentration in water might be anthropogenic or from the use of fertilizer on agricultural farms and septic system drainage (Olobaniyi etal.,2007).

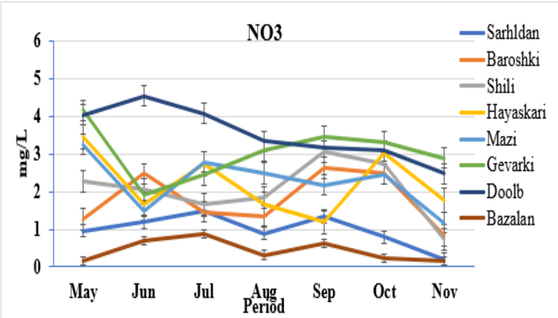

Figure 4: Monthly variation in NO3- (mg/L) among wastewater for the study period.

3.4 Phosphate in mg/l

(Orthophosphate) Phosphate comes from fertilizers, pesticides, industry and cleaning compounds. Natural sources include phosphate-containing rocks, and solid or liquid wastes Figure (5) show that a significant variation in phosphate value in Duhok Valley ranged from 4.01$\pm$ 0.41 to 0.14$\pm$ 0.09 mg/l. The highest concentration recorded in Doolb was 4.01$\pm$ 0.41 mg/l. This increase occurred as a result of domestic and agricultural wastes and may be due to solar radiation, which might have encouraged the biological degradation of the organic matter. While the lowest concentration recorded in Shili Site was 0.14 $\pm$ 0.09 mg/l in July.

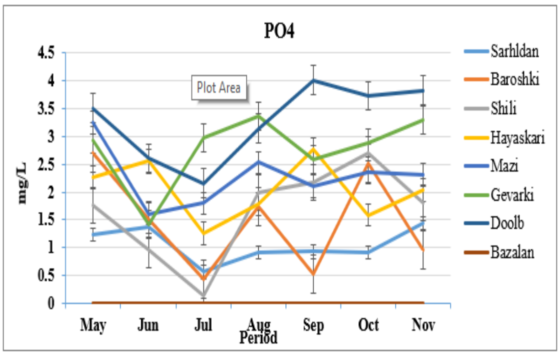

Figure 5: Monthly variation in PO4 (mg/L) among wastewater for the study period.

3.5 Lead (Pb) mg/L in wastewater

In this study, the mean concentrations of (Pb) in wastewater samples ranged from 0.45±0.08 to 5.95±0.2 mg/L Figure (6). The maximum allowable content of lead stated in wastewater is stated as 12 mg/l. The mean amounts of lead (Pb) in all the wastewater samples were lower than this value (WHO, 2014). The mean amounts of lead Pb in all the wastewater samples were much greater than this value (WHO, 2014). We see that the lead levels in crops watered with wastewater are substantially greater than in control (irrigated with freshwater). It indicates that the highest mean concentration of Pb was found at Gevarki, while the lowest mean concentration of Pb was found at Sarhldan. The high concentrations of heavy metals, including lead, are due to the subtraction of large quantities of the effluents move towards the Duhok Valley towards the south of the city, and that their concentrations between locations are due to the evaporation factor sedimentation, in addition to the discharge of industrial waste water, car repair workshops, and others. They showed a statistically significant difference ($p \leq 0.05$). The mean amounts of (Pb) measured in wastewater in the present study were less than those reported by Albarware., 2013; Zeliha et al., 2020.

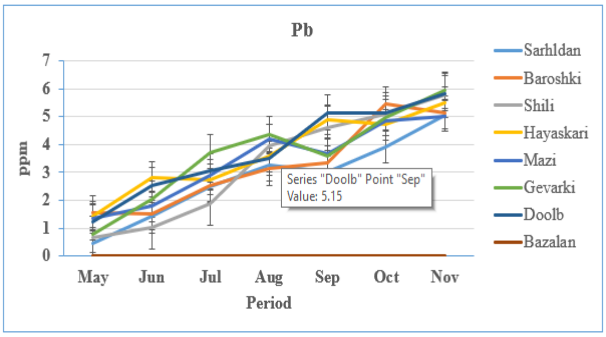

Figure 6: Mean concentration of Pb (mg/L) among wastewater for the study period

3.6 Nickel (Ni) in mg/Lin wastewater

In the present study, the mean concentration of Ni Figure (7) is found in the range of 0.18 $\pm$ 0.01 to 1.95 $\pm$ 0.28 mg /L, the minimum value of 0.18 0.01 mg/l is recorded at location Baroshki, and the maximum value of 1.95 0.28 mg/l is recorded at Gevarki Site. The Ni concentrations showed significant variation at $p \leq 0.05$. The result arrived at in the present study are significantly higher than those reported by Fariha et al., 2020) and Gupta et al., 2010). The mean concentrations of Ni in irrigated water were above the limit set by the WHO (2004). This demonstrates the Continuous application of wastewater for agricultural land which could raise the soil's concentration of heavy metals. We observe that Ni content of vegetables watered with wastewater is substantially greater than that of the control (irrigated with freshwater).

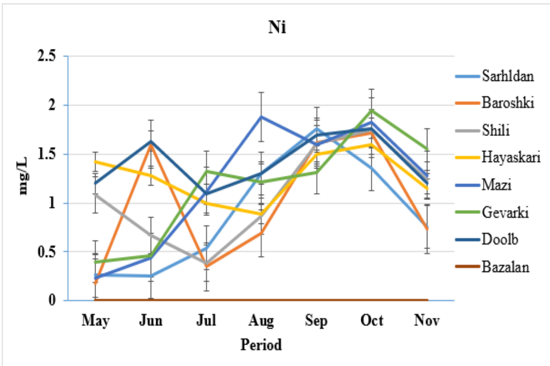

Figure 7: Mean concentration of Ni in wastewater – irrigated soil.

3.7 Copper (Cu) mg/L in wastewater

The mean concentrations of Cu in the investigated wastewater are given in Figure (8). The observed concentration range was determined to be 0.02±0.01to1.13±0.23 mg/L. It demonstrates that the highest mean concentration of Cu was found at Mazi Site, whereas the lowest concentration of Cu was found at Shili Site. All of these concentrations were significantly greater than the control (irrigated with freshwater) concentrations, and all of these concentrations were higher than the permissible levels recommended by WHO.,2014. Cu concentrations in wastewater measured in the present study were lower than those reported Albarware (2013), in Duhok and Leblebici & Kar (2018) in Nevsehir. In general, the sequence of heavy metal values was Pb > Ni > Cu.

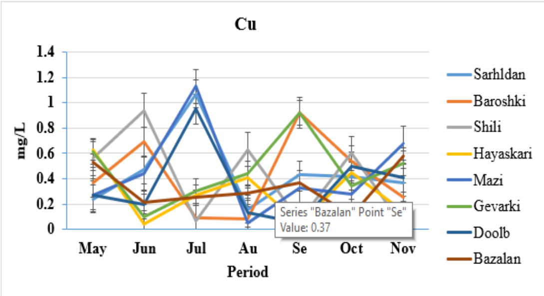

Figure 8: Mean concentration of Cu in wastewater – irrigated soil.

3.8 Lead (Pb) mg/kg-1 in soil

In the present study, the mean concentration is measured to vary within the range of 21.68 ±0.42 to 118.57± 0.07 mg/kg-1 Figure (9). Doolb Site recorded a high concentration of lead, whereas Sarhldan Site recorded a low concentration. Typically, field soils irrigated with wastewater have higher lead concentrations. According to the results, some soil samples fall within the acceptable limit, while others exceed them, which was significantly higher than the control (irrigated with freshwater). The permissible limit recommended by WHO (2005) is 85 mg/kg. They have a significant variation at p≤ 0.05. In this work they are significantly lower than those reported by Dingkwoet et al (2013) and higher than those reported by. Albarware (2013).

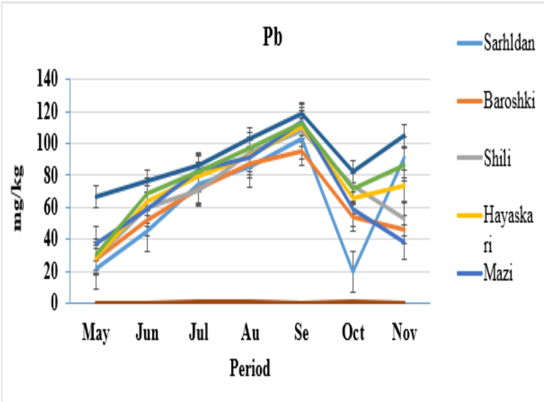

Figure 9: Mean concentration of Pb mg/kg (dry weight) in soil samples.

3.9 Nickel (Ni) mg/kg in soil samples

In this study, the average Ni concentration in the field soils of selected sites ranged from 1.27±0.47 to 7.44±0.73 mg/kg-1 Figure (10). The lowest Ni concentration was observed at Mazi, and the highest concentration was recorded at Shili. The high concentration may be attributable to Mazi industrial discharge into the waterway. The WHO recommended limit or Ni is 35 mg/kg-1 (WHO, 1996). The results indicate that the soil samples are within acceptable parameters. They own a statistically significant difference at the level of significance (P ≤ 0.05), similar to the results reported by Rolli (2014) Maukeeb et al (2022). We see that the Ni contents in soil irrigated by wastewater are significantly higher than those in control irrigated with freshwater.

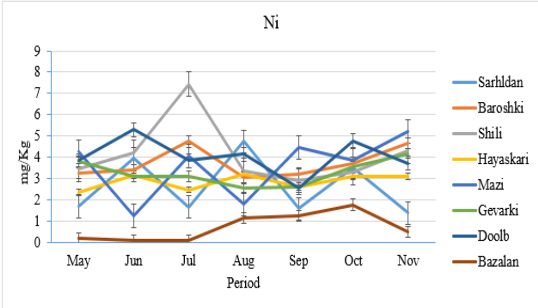

Figure 10: Mean concentration of Ni mg/kg (dry weight) in soil samples.

3.10 Copper (Cu) mg/kg soil samples

The mean concentration of Cu in field soils ranged from 1.28 ±0.24 to 15.12 ± 0.53 mg /kg-1 Figure (11). Maximum Copper was observed at Gevarki Site, and minimum concentration was observed at Baroshki Site. Copper concentration in such sites was primarily the result of industrial and commercial victory effluents to the waterway (Ratul., 2018). The results indicate that the soil samples lie within the permissible limits. The permissible limit recommended by WHO (2005) is 36 mg/kg. They have a significant variation at p≤ 0.05), similar results reported by Noor et al (2019). The sequence of the heavy metal values was generally Pb > Cu > Ni.

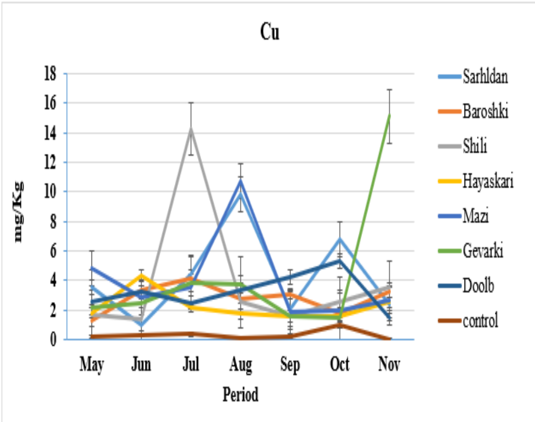

Figure 11: Mean concentration of Cu mg/kg (dry weight) in soil samples.

3.11 Lead (Pb) (mg/ kg-1) in celery leaves

Lead is the most dangerous non-biodegradable heavy metal to the population. Pb is not an essential metal for plants, but it is taken up by plants due to its presence in soil from anthropogenic sources such as fertilizers and automobile exhaust (Lamhamdi et al., 2011). The concentration of Pb in the celery leaves ranged from 1.04±0.22 to 5.22±0.60 mg/ kg−1 (Figure 12). The highest range of Pb concentration in celery leaves was found in Gevarki Site, which was irrigated with wastewater, while the lowest mean concentration of Pb was found in Mazi, which was substantially higher than the remainder of the control (0.05±0.01 mg kg-1). The mean concentrations of Pb measured in Celery leaves in the present study were lower than those reported by Kooti & Daraei (2017), but all of these concentrations were higher than the levels recommended by the Environmental Protection Agency (WHO., 2014). All of these concentrations were higher than the permissible levels recommended by WHO (2014). The permissible limit of Pb in vegetables recommended by (WHO (2009) is 0.05 mg/kg. The lowest Pb content recorded in the control (Celery plant) was 0.04± 0.05 mg/kg. The accumulation of minerals depends on some environmental factors such as salinity, pH and solidity and temperature. The solubility of heavy metals in aqueous media depends on the pH, Dissolved oxygen, hardness, and an increase in the pH of aqueous (Wang, 2015).

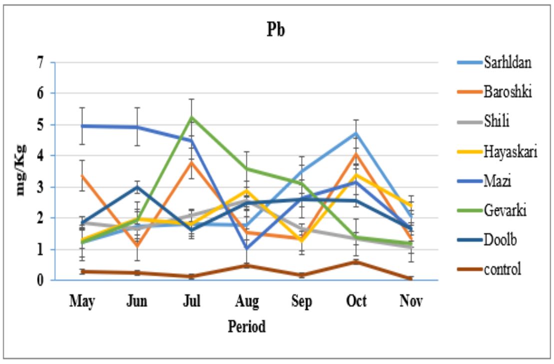

Figure 12: Mean concentration of Pb mg/kg (dry weight) in leaves grown in wastewater.

3.12 Nickel (Ni) (mg/kg) in celery leaves

According to WHO (2014), the Nickel tolerance limit in vegetables is 1.6 mg/kg. The levels of Ni in the investigated celery leaves of this study ranged from 0.57±0.03 to 4.31 ± 0.12 mg/kg -1 Figure (13). The lowest concentration of Ni was seen at Gevarki Site, while the highest level was seen at Shili Site. The accumulation of Ni is greater in the case of wastewater irrigated celery plants than in the control. The mean concentrations of Ni measured in celery leaves in the present studt were lower than those reported by Albarware (2013). All of these concentrations were lower than the permissible levels recommended by WHO (1996). The sequence of the heavy metal values was generally Pb > Cu > Ni.

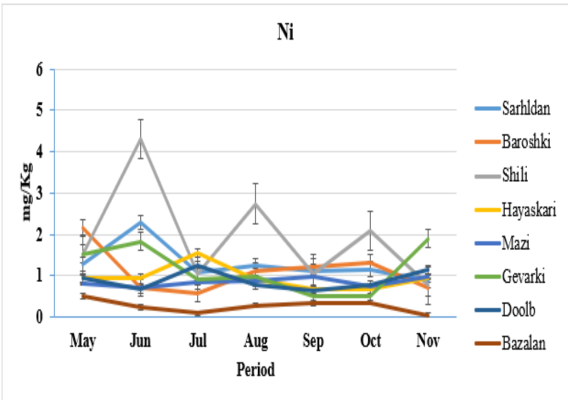

Figure 13: Mean concentration of Ni mg/kg (dry weight) in leaves grown in wastewater.

3.13 Copper (Cu) (mg/ kg-1) in celery leaves

The mean concentrations of Cu in the examined celery leaves of this study ranged from 0.34± 0.10 to1.71 ± 0.49 mg/kg-1 Figure (14). The highest range of (Cu) concentration in celery leaves was found at Doolb Site that was irrigated by wastewater, while the lowest concentration was found at Sarhldan Site. Table (8) shows that the concentration of Cu is higher in celery leaves irrigated with wastewater as compared with control irrigated with fresh water. The maximum admissible value of copper should be 3 mg/kg in vegetables set by WHO (1996). The mean concentration of Cu in celery leaves was below the WHO standards. The mean concentrations of Cu measured in Celery plant leaves in the present research were lower than reported by Satyanand et a (2013).

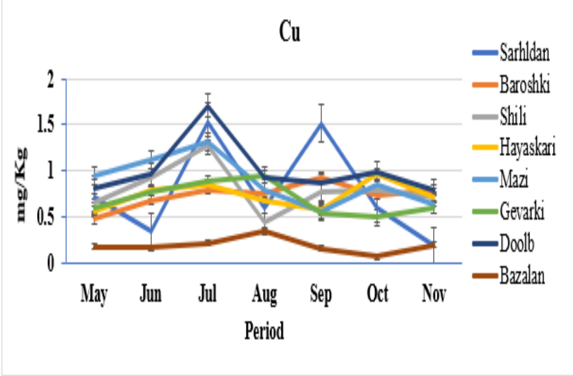

Figure 14: Mean concentration of Cu mg/kg (dry weight) in leaves grown in wastewater.

3.14 lead (Pb) (mg/ kg-1) in celery roots

The average of Pb concentration in the of celery roots ranged from 1.34±0.22 to19.47±0.82 mg/ kg-1. Figure (15). The greatest range of Pb concentration in celery roots was reported in Mazi, which was irrigated with wastewater, while Baroshki Site had the lowest concentration. The accumulation of heavy metals is greater in vegetable irrigation with wastewater than in the control group. The mean concentration of heavy metals was the highest in celery root samples compared to leaves. They were significantly higher in this study than those reported by Musher and Najlaa (2019). The results indicate that Lead absorbed by plants from the soil is first accumulated in their roots.

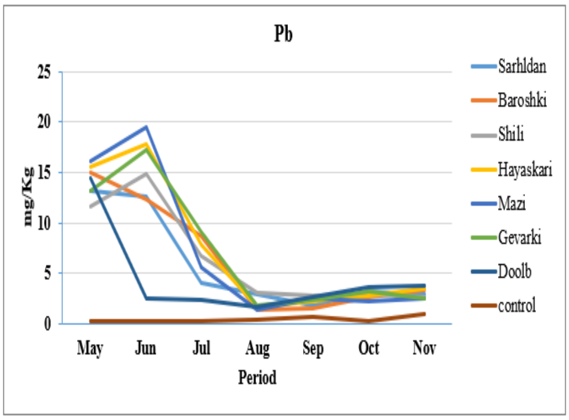

Figure 15: Mean concentration of Pb mg/kg (dry weight) in roots grown in wastewater.

3.15 Nickel (Ni) in mg/kg in celery roots

The mean concentration of Ni in celery roots was found in the range of 1.23± 0.04 to 7.29 ±2.53 mg/kg-1 Figure (16). The level of Ni in all the samples was below the maximum permissible level (67.90 mg/kg) set by WHO (1996). The highest mean level was found at Baroshki Site, whereas the lowest value was found at Shili Site. The considerable concentration of Ni in the aerial parts of celery plants indicated their strong bioaccumulation. The general behaviour for the accumulation of Ni in various parts of the celery plants was found to be root> stems. The result of the present study are significantly lower than those reported by Albarware (2004). The sequence of the heavy metal values was generally Pb > Cu > Ni.

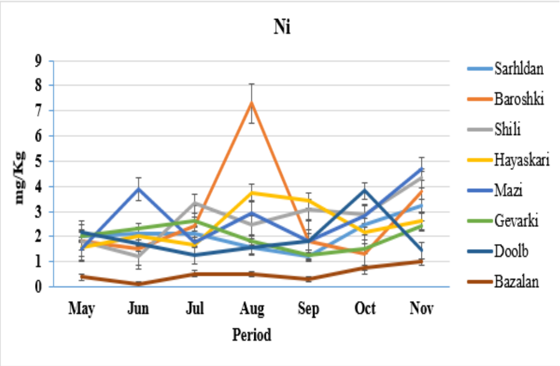

Figure 16: Mean concentration of Ni mg/kg (dry weight) in roots grown in wastewater.

3.16 Copper (Cu) in mg/kg in celery roots

The mean concentration of Cu in the celery roots samples was found in the range of 1.01±0.05 to 5.76 ±1.32 mg/kg-1 Figure (17). Among the sites, the mean concentration of Cu in root parts was found to be the highest at Mazi Site, and the lowest concentration was found at Shili Site. The concentration of Cu in all samples was observed to be lower than the permissible limit in crops (73.30 mg/kg). The results indicated that the pattern of Cu accumulation in the celery parts was root> stem. The results also showed that Cu taken up by plants from the soil is accumulated first of all in roots and then transported slightly to leaves. These results were significantly lower than those reported by Ewa (2013).

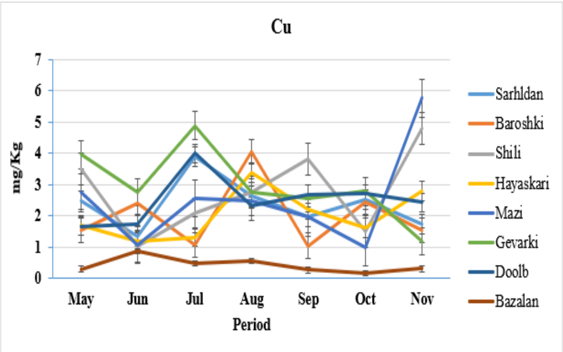

Figure 17: Mean concentration of Cu mg/kg (dry weight) in roots grown in wastewater.

Element	Drinking water(mg/l)	Wastewater (mg/l)	Vegetable (mg/kg)	Soil (mg/kg)
Pb	0.05	5.0	0.05	2
Ni	0.1	0.2	0.10	10
Cu	1.0	0.2	0.5-0.05	10

Table 2: The permissible limits of some heavy metal ions in water, vegetable and soil recommended by WHO.

4. CONCLUSION

The celery plant is consumed by the population of Duhok city, thus exposing the population to a dangerous concentration of toxic metals. The results demonstrate that there is a risk associated with the consumption of celery plants grown in Duhok Valley. The heavy metal concentrations in the celery plant were below the acceptable levels. In order to prevent excessive accumulation of heavy metals in the body, residents of this region are cautioned against consuming large quantities of celery, according to this study. However, the results of the present study indicate that heavy metals in celery plants have not yet affected human health. The heavy metals examined were present in all the samples studied, with most of them falling within the WHO permissible limits. To prevent their excessive accumulation in the food chain, regular monitoring of these toxic heavy metals from effluents and wastewater in vegetables and other food materials is essential. We concluded that using untreated wastewater for irrigation will severely contaminate soil and celery plants.

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