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A STUDY ON THE CONTAMINATION OF KHABUR RIVER WITH HEAVY METALS DUE TO SPATIAL AND SEASONAL DISCHARGED WASTEWATER IN THE IRAQI KURDISTAN REGION

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ABSTRACT

The main objective of the present study is to spatially evaluate the water contamination of Khabur River, before it arrives in Zakho City, inside the city, and after it leaves the city of Zakho by heavy metals. Also, the seasonal effects of Zakho municipal wastewater discharged in the Khabur River were detected in this study. The results showed that some heavy metals, such as Ni, were not detected in all studied samples in this study due to their low concentration levels. It is indicated that heavy metals are statistically affected by spatial location and high amounts were detected after leaving, compared to before entering the city which indicates that municipal wastewater is the main source of metal pollution. However, all water tests met the WHO's authorized limits. The average detected concentrations of copper, iron, manganese, and lead ranges from 0.003 to 0.025 mg/L, 0.000 to 0.054 mg/L, and 0.057 to 0.112 mg/L, and 0.014 to 0.135 mg/L respectively, while the highest concentrations of copper and iron were recorded significantly in Bedare (0.025 mg/L, 0.054 mg/L) respectively. The highest concentration of Lead (Pb) was detected in Chamtre and Tawke 0.117 mg/L and 0.135 mg/L respectively and this increase may be due to the presence of oil in the discharged industrial wastewater, in this village. Regarding season's effects, higher mean concentrations for Mn 0.13423 mg/L, Fe, and 0.04208 mg/L were recorded in the autumn season. However, copper and lead had higher mean concertation 0.02389 mg/L, and 0.097 mg/L respectively, during the winter season, while the lowest mean concentration of copper was recorded in autumn, and lead in the summer season had a minimum concentration. There was a significant difference ($P < 0.01$) in the seasonal variation of Cu, Fe, Mn, and Pb ($P=0.001$).

KEYWORD: Heavy metal, Wastewater, River, WHO limits.

1. INTRODUCTION

Freshwater makes up about 3% of the planet's total water. The amount of freshwater that can be used by humans is very small (0.01%). Due to the population growth, urbanization, climate change, human activities, and untreated wastewater, even this tiny amount of fresh water is at extreme risk (Jamshaid et al.,2018). Any metal with a density greater than 5 $g.cm^{-3}$, not biodegradable, toxic at very low concentrations is considered a heavy metal, such as mercury (Hg), cadmium (Cd), Arsenic (As), chromium (Cr), Zinc (Zn), Nickel (Ni), and lead (Pb) (Rosenberg, 2015). In the natural world, high trace metal concentrations can be caused by geological events including, rock weathering of rocks, volcanic eruptions, and water-induced leaching into rivers, lakes, and seas (Sankhla et al.,2016). Artificial Sources, similar to how residential and agricultural waste gets into the environment, heavy metal traces also enter through car exhausts. Processing of ores of metal, and burning of fossil fuels like petrol, coal, and kerosene oil. Mining, Discharging farming waste. Discharging manufacturing waste. Heavy metals are hazardous because they bioaccumulate in living tissues. Bioaccumulation refers to the elevation of a chemical concentration in an organism's tissues over time compared to the chemical background in nature (Gerenfes & Teju, 2018). Hazardous chemical tends to accumulate in living tissues when they are absorbed and stored faster than their degradation or to be excreted from the organism body. Industrial and consumer waste can introduce heavy metals into water supplies. These harmful substances reach the bloodstream mainly by food and drink, and to a smaller amount through the breathing of contaminated air (Verma & Dwivedi, 2013). Toxic metals are released to the air mainly from industrial exhaust, rubbish biodegradation, and cars emissions. After the heavy metals build up in the soil with acid rains, it gradually reach freshwater bodies as surface runoff in rainy seasons (Jamshaid et al., 2018).

Some heavy metals are necessary for health, but only in small amounts such as (Cu, Fe, Zn, and Mn) as important trace metals, copper (Cu) is an essential micronutrient for better cell physiological functioning and acts a cofactor of photosynthesis enzymes, but it is toxic to the human body in larger amounts. However, the mining of copper may spread their dust to cause many illnesses in the human body. And chronic exposure may lead to kidney failure and even death (Bala et al., 2008). At high concentrations, Cu is hazardous to a wide range of aquatic organisms and induces growth, disturbing organisms, and metabolism, Cu exposure in the environment is mostly caused by mining, metallurgy, and industrial usage (Vitek et al., 2007).

Ni is a chemical element, most abundant in the iron/nickel core of the earth. It is used in the manufacture of many alloys and products such as ceramic paints, stainless steel, jewelry, kitchenware, batteries, textiles, and coins. Nickel is discharged into the ecosystem from energy generation stations, metal plants, and wastewater, it is also used in fertilizers and enters water via agricultural wastewater. (Rasheed et al.,2021) Furthermore, toxic non-essential metals such as Pb and Ni have a harmful effect on living things even at extremely low quantities. Because it has a toxic effects, very persistence to degradation, tends to bioaccumulate in living tissues, and biomagnification in the base of food chain, so high quantities of heavy metal in soil, water, and air are accompanies with high thread to natural ecosystem and human wellbeing (Adesiyan et al., 2018).

Pb is being so poisonous that it can harm the neurological system, kidneys, and reproductive system. Pb exposure results in encephalopathy symptoms and permanent brain damage (Chowdhury et al., 2014), there are several sources of lead in the environment, including industrial waste, residential paint, and vehicle exhausts. The Khabur River is mostly contaminated during the wet season by sewer pipes, industrial effluent, and municipal wastewater from Zakho city and villages around in addition to the agricultural runoff, and other sources, all of which may be untreated (Saleem et al., 2005). Along with the annual increase in the city population, the release amounts of pollutants into the river are also directly increased (AL-LAYLA & Al-Rizzo, 1989).The main objective of this study is to evaluate the spatial location before, inside, and when river leave the city as well as the seasonal effects of Zakho municipal wastewater discharged in Khabur river in the Iraqi Kurdistan Region.

2.MATRIALS AND METHODS

2.1 Study area

The study was carried out in different locations along Khabur River in Zakho City (Figure 1). The city of Zakho is located on the border of Iraq with Turkey and Syria in the northwest of Iraqi – the Kurdistan region. The city is located around 57 km southeast of Cizre and 52 Km northeast of Duhok City. Zakho city may have been established on a tiny island in the minor Khabur River, which now runs through the city (Raswol & Khorsheed, 2017). One of the five rivers that flow into the Tigris River, Khabur River contributes to the Tigris Flow at Zakho Station by roughly 2 billion cubic meters (BCM). The old perhaps Roman bridge of Delal crosses the Khabur River through Zakho City. After passing through the city, Khabur River serves as the natural boundary between Turkey and Iraq near Ibrahim Khalil. The study is situated at the point of intersection of latitude (37° 8' 53" N) and longitude (42° 42' 13").

2.2 Sample collection

Samples of water from different locations along the Khabur River were collected for this study following the standard procedure as described by APHA (1998). This study covered three seasons, including summer, autumn, and winter during 2021–2022. Three locations were selected for each season; and three water samples were collected from (Barzirke, Tawke, and Chamtre) before entering Zakho City; three samples passing through the city from (Pra Dalal, Sike, and Bedare); and three after leaving the city (Ibrahim Khalil, Shenava, and Qarawla) see (Figure1). Around two litters of water from the surface layer of each place were put in clean plastic bottles. Many physical, chemical and metabolic interactions would affect the quality of the water sample between collection and analysis. Thus, to limit this change, water samples were transferred to the laboratory by cooling box, then they were put in a refrigerator at around 4 °C to inhibit microbial activity before being analysed.

2 .3 Heavy Metal Determination

Heavy metals in water samples were analysed by Atomic absorption spectrometer (AAS) high performance type (Perkin- Elmer, PinAAcle 900T AAS), as described by (APHA,1998), according to standard analytical methods (APHA,1998). Samples of water have been filtered through the Whatman filter paper no.42, the concentration of Heavy metals such as (Pb), (Ni), (Cu), (Fe), and (Mn), were measured analytically by the atomic absorption spectrometer model. The results were expressed in mg/L.

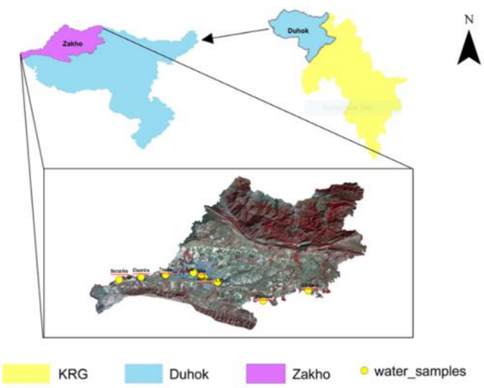

Figure 1. Zakho city map showing the location of sample collection.

2.4 Statistical analysis

The data of the experiments were submitted to SPSS Software (George & Mallery, 2019), and the procedures of Descriptive Statistics, MANOVA, and Correlation were performed. The used model for MANOVA via GLM was as follows:

$$Y1_{ijkl}.\ Y2_{ijkl}\ldots = \mu + S_i + R_j + A_k + (S*R)_{ij} + (S*A)_{ik} + (R*A)_{ik} + (S*R*A)_{ijk} + e_{ijkl}$$

Where:

$Y1_{ijkl}$= the first studied element (character), $Y2_{ijkl}$ = the second element…. etc.

μ= overall mean

S_i = the effect of Season.

R_j = the effect of Region

A_k = the effect of Area

$(S*R)_{ij}$+ $(S*A)_{ik}$+ $(R*A)_{ik}$ + $(S*R*A)_{ijk}$ = *Interactions between studied factors / treatments.*

e_{ijkl}= experimental error.

The difference between means of studied characteristics was compared using Duncan's multiple range test (Duncan, 1955).

3.RESULTS AND DISCUSSION

3.1 Spatial effects on Heavy metal in Khabur river

The mean value of heavy metals analysed in Khabur River such as Cu, Mn, Pb, Fe, Ni were affected by the location present in (Table 1 and Figure 2). Some heavy metals, such as Ni, were not detected in all sample locations in this study due to their low concentration levels. (Issa & Alshatteri, 2018) studied heavy metal pollutant in drinking water of Garmiyan, Kurdistan region, the results for Ni was not detected in all samples.

Table 1. Descriptive Statistics of heavy metals in Khabur river as affected by location.

Descriptive Statistics				Sig.
Element	**Location**	**Mean**	**SE**	***(p)***
Cu	Before	0.015 [ab]	0.003	** 0.001
	Inside	0.018 [a]	0.002	
	After	0.009[b]	0.003	
Fe	Before	0.012 [b]	0.004	NS 0.057
	Inside	0.032 [a]	0.009	
	After	0.012 [b]	0.006	
Mn	Before	0.069 [b]	0.008	** 0.001
	Inside	0.094 [a]	0.007	
	After	0.099 [a]	0.012	
Ni	Before	0.000 a	0.000	NS 0.203
	Inside	0.000 a	0.000	
	After	0.000 a	0.000	
Pb	Before	0.114 [a]	0.027	** 0.001
	Inside	0.036 [b]	0.011	
	After	0.045 [b]	0.014	

NS=Non Significant (p>0.05); *= significant at 0.05 level; **= significant at 0.01 level.
The means of having different superscript letters within each element/characteristic or element cell have differed significantly.

The mean value of Cu concentration was ranged from 0.009 mg/L to 0.018 the results of Cu inside Zakho City (Figure 2, a) higher than before and after the city. The high concentration of this location due to industrial effluents and Blue-green stains on plumbing fittings may indicate high copper levels in the water. Council, (2000) concluded that the content of Cu in water is affected mainly by the minerals type and their quantity, how long water lasts in the water pipes, temperature and acidity of the water. There was a significant difference at level ($p < 0.01$) in location variation for Cu ($p=0.001$). However, the pollution of Khabur River by Cu is negligible as compared to the WHO's permitted limit for copper in drinking water is 2 mg/L (Jamshaid et al., 2018). (Manne et al., 2022) studied the risk assessment of heavy metals in deteriorating water quality by copper, they observed that the concentration of copper was very low, similar to Khabur River.

I Fe is an essential micronutrients required for the human body at low amounts and have a vital role in the synthesis of haemoglobin (Jazza et al., 2022).The highest value of Fe concentration in Khabur was recorded inside ZC 0.032 mg/L. This might Iron may dissolve into the water as rain or when snow melts on the ground and water seeps through iron-enduring soil and bedrocks. Fe reach waters also by the steel corrosion in iron water pipelines or artesian well casing (Gibbs, 1979).while before and after Zakho City the concentration of Fe is similar. The mean concentration of Pb before Zakho Citywere higher than inside and after leaving the city see (Figure 2, b). The elevated Pb levels may be ascribed to pollution in the area, as well as a large number of oil wells and filtration stations. Large amounts of Pb caused to the burning of leaded fuel from an automobile(Rasheed et al., 2021). The concentration of Pb, the most toxic dangerous heavy metal for human being health is significantly greater than the most worldwide standards in three locations before, inside, and after leaving. So, the Pb should be removed by chemical and biological methods before it is used for drinking water as it was heavily toxic and carcinogenic. (Abdulla et al.,2020) Studied the heavy metal concentration in water and fish of Darbandikhan and Dokan Lakes in Sulaimaniyah governorate. The results of Pb, Cu, Cd and Zn concentrations showed 91.6 – 413.4, 6.2 – 56.9, 4.0 – 35.6, 42.6 – 388.8 µg/L at Darbandikhan lake, and 14.7 to 79.3, 15.9 to 43.9, 83.5 to 265.9,1.8 – 112.9 µg/L in Dokan lake respectively, the maximum allowed amount of lead and cadmium was present. All cells require manganese for phosphate transfer involving adenosine triphosphate and diphosphate. The mean concentration of Mn inside and after Zakho City nearly similar (Figure 2, c). Mn deposition in water is caused by leachate migration, neighbouring water bodies, and low-lying terrain (Oluyemi & Olabanji, 2011). Alam et al. studied heavy metal pollution in the area surrounding the waste open dump site of Mogla Bazar in Sylhet, Bangladesh. All metal concentrations were compared to the permitted limit stated by the WHO/FAO, EU, IS, USEPA. The concentrations of Fe, Mn, Cd, and Pb in water above the allowable limit. (Alam et al.,2020) significantly greater than the most worldwide standards in three location before, inside, and after leaching were 0.1,0.03, and 0.04 mg/L as the WHO allowable limit for Pb in drinking water is 0.01mg/L (Jamshaid et al., 2018). The direct use of Khabur water for drinking carries a lot of risks in increasing the incidence of cancer.

3.2 Heavy metal variation within same location in Khabur River

The concentration of heavy metals such as (Cu, Pb, Ni, Fe, Mn) affected by area are summarized in the (Table 2 and Figure 3). The average concentration of Cu , Fe, Mn, and Pb ranges from 0.003 to 0.025 mg/L, 0.000 to 0.054 mg/L, and 0.057 to 0.112 m/L, and 0.014 to 0.135 mg/L respectively. In this study, the highest concentrations of Cu, and Fe were recorded significantly in Bedare 0.025 mg/L, 0.054 mg/L respectively, and Cu concentration in other villages are not significantly different, especially Ibrahim Khalil, Shenava, and Tawke were minimum ranges were detected. It enters our water supply by mining, agricultural or industrial drainage from Pra-Dalal and Sike discharges into rivers, and as a result of population increase and untreated sewage for restaurants and cafes, the pollutants in the water increases (Vitek et al., 2007). The iron concentration in Shenava was not detected. While the highest concentration of manganese has been detected in Qarawla and Badare, the increase in manganese at this site may be due to human activities such as industrial discharges, mining, and landfill leaching. Fe and Mn are the most presenting heavy metals in groundwater; However, Mn content is usually much smaller than iron. Iron dissolved in water because it reduces from ferric to ferrous as well as manganese dissolve in water for the same reason of reduction and reduced form of metals are mostly soluble in water. Another important factor that effect the desolation of heavy metals is water pH , when pH draw down 7, the solubility of reduced Fe and Mn increased (Abdulla et al., 2020).The highest concentration of Pb was detect in chamtre and tawke 0.117 mg/L and 0.135 mg/L respectively, the simplest reason is that when lead containing plumbing pipes and fixtures corrode, the Pb can dissolve or flake into the water that runs from our faucets. Because lead cannot be seen, smelled, or tasted, it can be present in even clear water (Jazza et al., 2022).While the concentration of Ni in Khabur River not detect that mean there is no significant deferent p>0.05.

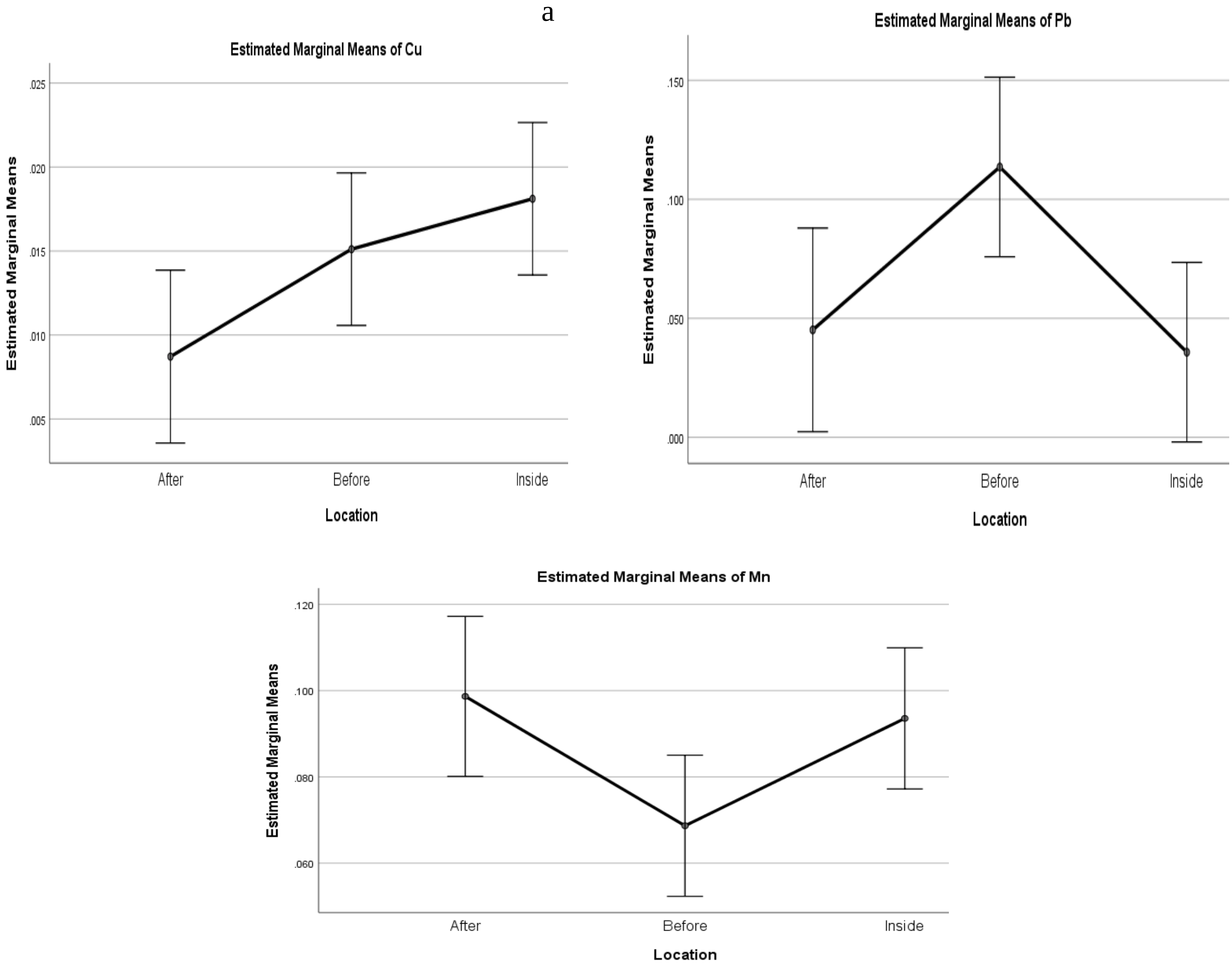

Figure 2. the main value of heavy metal affected by location :(a) Cu, (b) Pb, (c) Mn.

Manganese, and Pb ranges from 0.003 to 0.025 mg/L, 0.000 to 0.054 mg/L, and 0.057 to 0.112 m/L, and 0.014 to 0.135 mg/L respectively, in this study, the highest concentrations of Cu, and Fe were recorded significantly in Bedare 0.025 mg/L, 0.054 mg/L respectively, and Cu concentration in other villages are not significantly different, especially Ibrahim Khalil, Shenava, and Tawke were minimum ranges are detected and its save for drinking regarding Fe and Cu that not exceeds the level recommended by WHO. It enters our water supply by mining, agricultural or industrial drainage from Pra-Dalal and Sike discharges into rivers, and as a result of population increase and untreated sewage for restaurants and cafes increases pollutants in the water (Vitek et al., 2007). The iron concentration in Shenava was not detected. While the highest concentration of manganese has been detected in Qarawla and Badare, the increase in manganese at this site may be due to human activities such as industrial discharges, mining, and landfill leaching. Iron and manganese are often found together in groundwater.

Table 2. Descriptive statistics of heavy metal in Khabur river affected by area in same locations.

Descriptive Statistics				**Sig. (*p*)**
Element	**Area**	**Mean**	**SE**	
Cu	Barzire	0.023 [ab]	0.006	** 0.001
	Chamtre	0.012[ab]	0.003	
	Tawke	0.010[cd]	0.003	
	Pra-Dalal	0.013[ab]	0.001	
	Sike	0.016[ab]	0.004	
	Bedare	0.025 [a]	0.003	
	Ibrahim Khalil	0.003[d]	0.002	
	Shenava	0.009[c]	0.003	
	Qarawla	0.014[ab]	0.006	
Fe	Barzire	0.005 [b]	0.003	** 0.001
	Chamtre	0.032[ab]	0.008	
	Tawke	0.000[b]	0.000	
	Pra- Dalal	0.018[ab]	0.002	
	Sike	0.024[ab]	0.008	
	Bedare	0.054[a]	0.026	
	Ibrahum khalil	0.031[ab]	0.015	
	Shenava	0.000[b]	0.000	
	Qarawla	0.014[ab]	0.006	
Mn	Barzire	0.057 [b]	0.014	** 0.001
	Chamtre	0.072[ab]	0.015	
	Tawke	0.077 [ab]	0.013	
	Pra- Dalal	0.095 [ab]	0.008	
	Sike	0.080[ab]	0.014	
	Bedare	0.106[a]	0.010	
	Ibrahim Khalil	0.091[ab]	0.020	
	Shenava	0.093[ab]	0.021	
	Qarawla	0.112[a]	0.021	
Ni	Barzire	0.000 a	0.000	NS 0.206
	Chamtre	0.001 a	0.000	
	Tawke	0.000 a	0.000	
	Pra- Dalal	0.000 a	0.000	
	Sike	0.000 a	0.000	
	Bedare	0.001 a	0.000	
	Ibrahim Khalil	0.000 a	0.000	
	Shenava	0.000 a	0.000	
	Qarawla	0.001 a	0.001	
Pb	Barzire	0.089 [ab]	0.030	** 0.001
	Chamtre	0.117[ab]	0.059	
	Tawke	0.135[a]	0.053	
	Pra- Dalal	0.014[b]	0.007	
	Sike	0.048[ab]	0.024	
	Bedare	0.045[ab]	0.023	
	Ibrahim Khalil	0.057[ab]	0.0027	
	Shenava	0.064[ab]	0.030	
	Qarawla	0.014[b]	0.006	

NS=Non Significant ($p>0.05$); *= significant at 0.05 level; **= significant at 0.01 level.
The means having different superscript letters within each element/ characteristics or element cell are differed significantly.

3.3 Heavy metals in water as affected by the seasons:

The seasonal variation of heavy metals such as Cu, Mn, Pb, Fe, and Ni in Khabur River shown in (Table 3) higher mean concentration for Mn 0.13423 mg/L, Fe 0.04208 mg/L recorded in the autumn season. Might be attribute to the temperatures fluctuation that effect on the rates of photosynthesis in aquatic plants as well as the dissolvability of O_2 in water. As a result, when temperatures begin to decrease, plant growth activity reduces and plants begin to die, the dead debris consume oxygen when it breakdown by microorganisms and release nutrients , and Due to pollution, leaching from piping, or mineral contamination of the water supply (Arshad, Mehmood, Shah, & Abbasi, 2020). However, Cu and Pb had higher mean concertation 0.02389 mg/L, and 0.097mg/L respectively, during the winter season. While the lowest mean concentration of Cu was recorded in Autumn and Pb in the summer season had a minimum concentration.

A significant difference was detected ($p <0.01$) in the seasonal variation of Cu, Fe, Mn, and Pb ($P=0.001$), but no significant difference was detected in the seasonal variation of concentrations of Ni. Rasheed et al., studied the determination of some heavy metals in water in Sulaimanih City- Iraqi Kurdistan Region. All heavy metals in this study were analysed by Flame Absorption Spectrophotometer. The results of Pb in summer had maximum concentrations are aginst the result of the Khabur River. With the exception of nickel, heavy metals did not exceed the standard of Iraqi and the WHO for drinking water quality assessment (Rasheed et al., 2021). (Ali et al., 2016) studied a preliminary analysis of heavy metals in the water and sediment of Bangladesh's Karnaphuli River. The result of Pb was recorded in summer and winter at 38.33 and 49.04 mg/kg as a maximum .

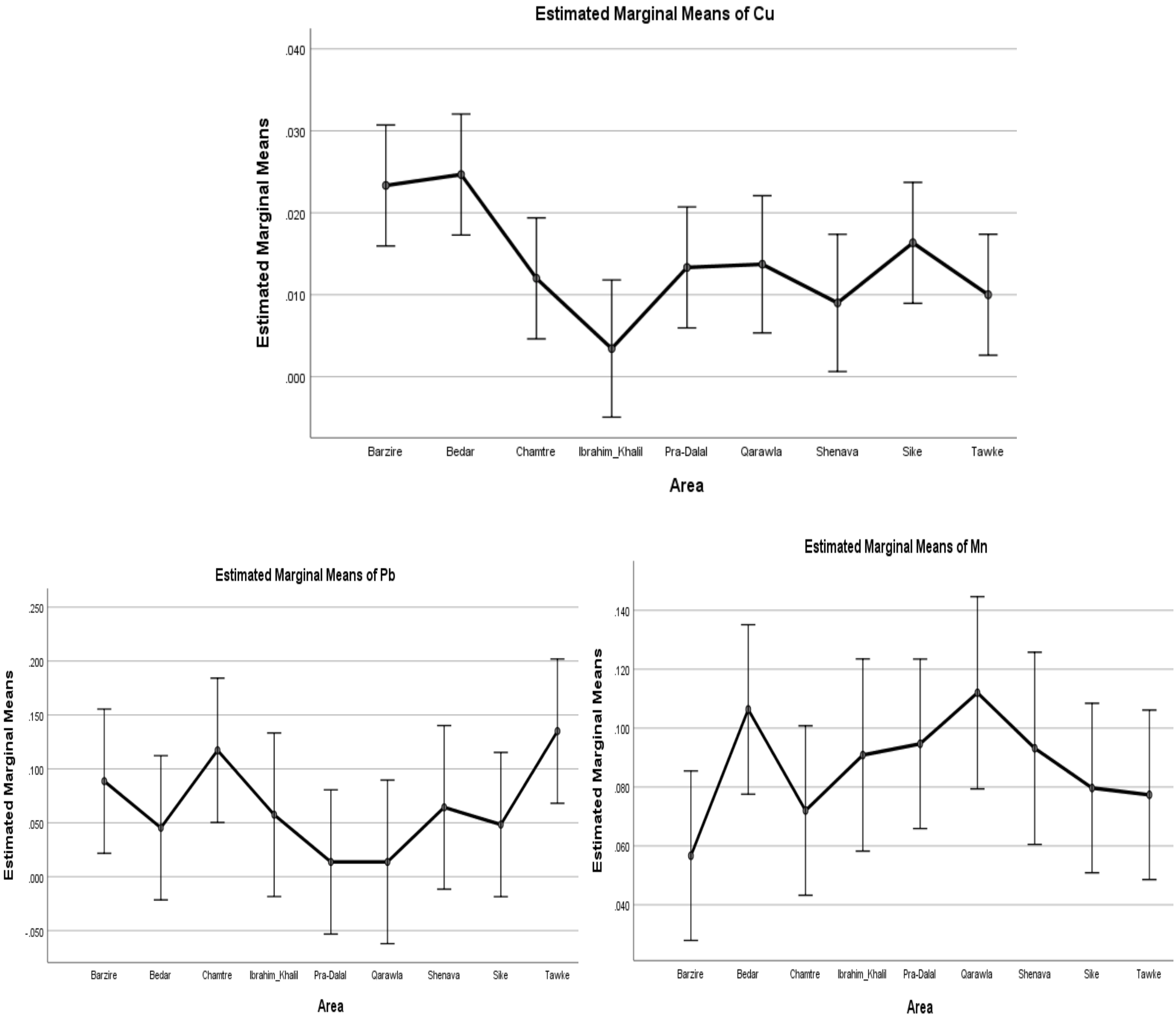

Figure 4.the mean concentrations of heavy metal affected by area: (a) Cu, (b) Pb, (c) Mn.

Table3. Descriptive Statistics of the characteristics of the studied elements as affected by the season.

Element	**Season**	**Mean**	**± SE**	**Sig. (*P*)**
Cu	Autumn	0.00462 [c]	0.00	** 0.001
	Summer	0.01386 [b]	0.00	
	Winter	0.02389 [a]	0.00	
Fe	Autumn	0.04208 [a]	0.01	** 0.001
	Summer	0.013 [b]	0.00	
	Winter	0.00314 [b]	0.00	
Mn	Autumn	0.13423 [a]	0.00	** 0.001
	Summer	0.07695 [b]	0.00	
	Winter	0.04807 [c]	0.01	
Ni	Autumn	0.00023 [a]	0.00	NS 0.26
	Summer	0.00048 [a]	0.00	
	Winter	0 [a]	0.00	
Pb	Autumn	0.087 [a]	0.01	** 0.001
	Summer	0 [b]	0.00	
	Winter	0.097 [a]	0.03	

NS=Non Significant (p>0.05); *= significant at 0.05 level; **= significant at 0.01 level.
The means of having different superscript letters within each element/characteristic or element cell have differed significantly.

4. CONCLUSION

Spatial variation of Khabur River was significantly influenced in their content of heavy metals when entering the centre of Zakho City comparing before arrival and when leaving the city, while the seasonal variation has less effects in changing their content of heavy metal. However, in summer season when the flow of the river is reduced, the detectable portion of heavy metals slightly increase. Nowadays, heavy metals are the most concerned pollutants in drinking water which associated with the human health, being the causer of cancer disease. However, none of studied heavy metal reach above permissible level set by WHO except Pb which need proper chemical and biological treatment before being used for drinking. Approximately all sewage water of Zakho City are discharged to the river without any proper treatments. Therefore, all precautions should be taken into consideration before discharging municipal wastewater to the rivers, and proper chemical and physical treatments of heavy metal removal from the water should be applied in future to prevent reaching this toxic metal to risky level.so it is recommended to collect the municipal wastewater of this city in one location to be treated after being discharged to the river in order not to contribute in further pollution of Mosul dam that support both Mosul and Duhok cities by drinking waters.

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