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## EFFECT OF TREATED WASTEWATER IRRIGATION ON VEGETABLES

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*Treated waste water is normally used for irrigation purposes in countries suffering from water shortages to narrow the gap between supply and demand. The concept behind this is to save water consumed for agricultural activities, which consumes most of the water, for municipal and industrial uses. The Alsukhna area in Jordan is used to grow vegetables which are irrigated by treated wastewater. Surface and groundwater samples from the Zarqa region were analyzed for their major cations, anions and heavy metals. The impact of the treated waste water on the chemical components of vegetables was studied using Zn, Mn, Fe, Pb and Ni in sweet and hot pepper, tomato, cauliflower, cabbage, squash, cucumber and eggplant which were compared with similar vegetables irrigated by natural unpolluted water from the Mafraq region. The four metals, namely Zn, Fe, Pb, and Ni, had concentrations higher than in the reference vegetables by 3423%, 155%, 397%, 2949% and 289%, 187%, 211%, 214% for tomato and cauliflower, respectively. Sweet pepper was mainly influenced by an increased content of Fe, which was almost 180% higher than that in sweet pepper from the Mafraq region. Hot pepper had highly elevated concentrations of Ni (6980%) and Zn (419%), while squash demonstrated high Zn (207%) and Pb (666%). When all the heavy metals are considered, the most affected vegetable is the hot pepper with an average percent of heavy metals accumulation of 1559% while the least effected is cabbage at 116%.*

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## INTRODUCTION

Most of the water consumption in many countries is allocated to agricultural practices. Countries suffering from water shortages are forced to use non-conventional resources, mainly water harvesting and treated wastewater. The Middle East is one of the areas experiencing shortfalls of freshwater (Roger and Lydon, 1994; Biswas, 1994; Al-Ansari, 1998). Jordan, with an area of 89 900 km<sup>2</sup> is a typical example of such a country, one of the poorest in its water resources. The estimated population in 2010 was 6.8 million with a rate of growth of 3.2% (Al-Ansari and Salameh, 2006) to 3.6% (Alkhaddar et al., 2003). The water shortage experienced by the country is well documented (Salameh and Udluft, 2001; MWI, 1998; Alkhaddar et al., 2003; Alkhaddar et al., 2005; Al-Ansari and Salameh, 2006). Jordan exceeds the limits of its available renewable freshwater supply (Biswas, 1994; Murakawi and Musiaka, 1994; Gleick, 1998). According to the Ministry of Water (2001) the deficit will reach 408 Mm<sup>3</sup>/year in 2020. To overcome the gap between supply and demand, non-conventional water resources are used including water harvesting and treated wastewater.

Agricultural practices consume 66% of the available water (Hiniker, 1999). Currently major efforts are being made to transfer as much as possible from water allocated for agriculture to other sectors of demand. A transfer of 10% of the water allocated for agriculture would provide 40% for the needs of the domestic water supply (Sadik and Barghouti, 1994). Accordingly, the practice of utilizing treated wastewater for crop irrigation is well established in Jordan and 17 wastewater treatment plants are in operation now with a design capacity of about 134 000 m<sup>3</sup>/day and the inflow rate of 82.6 Mm<sup>3</sup>/year in 2001 (Al-Ansari and Salameh, 2006). The World Bank (1997 and 2001) expects that this volume will reach 232 Mm<sup>3</sup>/year in 2020. However, problems concerning poor water quality and quantity exist which can lead to crop damage and pollution (World Bank, 1997).

Irrigation using treated wastewater has been a common practice worldwide. In India, treatment of crops with wastewater caused increased uptake of heavy metals, namely Fe, Mn, Pb, Cd, Cr, Cu, and Ni, in cauliflower, mustard, radish, celery, spinach etc. (Gupta et al., 2008; Gupta et al., 2010). Sharma et al. (2006) showed that variation of vegetables' adsorption and accumulation led to different uptake rates for various crops during different seasons. Thus, e.g. palak plants accumulated mainly Pb and Ni during summer while Cd accumulated in the winter season. Mint and spinach favored build-up of mainly Fe and Mn, whereas carrot showed the highest uptake rates of Cu and Zn (Arora et al., 2008). Irrigation using sewage sludge in Zimbabwe led to concentrations of heavy metals in crops of up to 20 times higher than the European levels (Muchuweti et al., 2006). Accumulation of heavy metals in olives and olive leaves as a result of irrigation with municipal wastewater showed greater uptake of Fe, Zn, Mn, and Cu than Ni, Pb, and Cd (Batarseh et al., 2011). Additionally, olive leaves and fruits had different uptake rates of these metals. Rimawi et al. (2009), comparing agricultural plots irrigated with freshwater and effluents from the phosphate mining industry, concluded that apart from a diminished crop yield of 50%, there was no risk of contamination in plants and soils. Similarly, some studies indicated a relative absence of health risk even though a large amount of heavy metals associated with contaminated vegetables was ingested by humans (Muchuweti et al., 2006; Khan et al., 2008; Avci, 2012). However, most of the investigations were performed on a short-time scale, and therefore limitations of such time-limited studies must be kept in mind. A positive practice of using wastewater from a stabilization pond for irrigation in Jordan showed that under the conditions described in Al-Nakshabandi et al. (1997) eggplant production increased due to the nutritive value of the effluent while heavy metal concentrations in the vegetable crops remained below the permissible limits.

The use of contaminated water for irrigation will only increase in the future, being controlled by the rising water demand especially in areas of water deficit. Effects of irrigation using polluted effluents still remain underestimated. For this research, an area which is irrigated by treated wastewater was chosen. The water used for irrigation was analyzed for its major cations and anions and selected heavy metals. To study the impact of this water on agricultural produce, 8 vegetables were analyzed for heavy metal concentrations and compared with similar vegetables irrigated by natural unpolluted water.

## STUDY AREA

The River Zarqa is the second largest river in Jordan (Salameh and Al-Ansari, 2000). The drainage area of the river reaches 4025 km<sup>2</sup> (Figure 1). The headwaters of the river rise in the Ain Ghazal spring on the eastern side of the Gilead Mountain northeast of Amman. The river course is 105 km long before joining the River Jordan. Two tributaries (Wadi Dhuleil and Sael Zarqa) join together at Sukhna forming the Zarqa River (Figure 1). Wadi Dhuleil drains the eastern part of the catchment and the Sael Zarqa (also known as the Amman-Zarqa River) drains the western part of the catchment. The average annual precipitation on the catchment reaches 237 mm. The mean annual flow of the river is 63.3 million cubic meters. The Zarqa catchment is heavily populated and contains about 65% of the population of Jordan and about 80% of the industry of the country (Salameh and Al-Ansari, 2000). The studied area is bounded by the Khirb alsamara waste water plant in the north, Hashimeya, Sukhna and Bera plants from the southwest and west, and the Dhulleil valley from the east respectively. The data obtained from the Jordan Meteorological Office (2009) indicates that it is characterized by a mean annual temperature of 17.7 °C. The maximum and minimum recorded temperatures have been 44.8 °C and -8.6 °C respectively. The mean annual rainfall is 146 mm and the average number of rainy days is 25. The average humidity is 53.3% and the average daily evaporation rate is 7.35 mm.

Ground water aquifers in the area are mainly of Na'ur, Hummar and Wadi Assir Formations (Marssi, 1965). They are composed of limestone, dolomite, marl, marly limestone. It should be mentioned however, that the surface runoff within the area percolates through the basaltic rocks exposed at the surface of the catchment area of the river Zarka (Marssi, 1965).

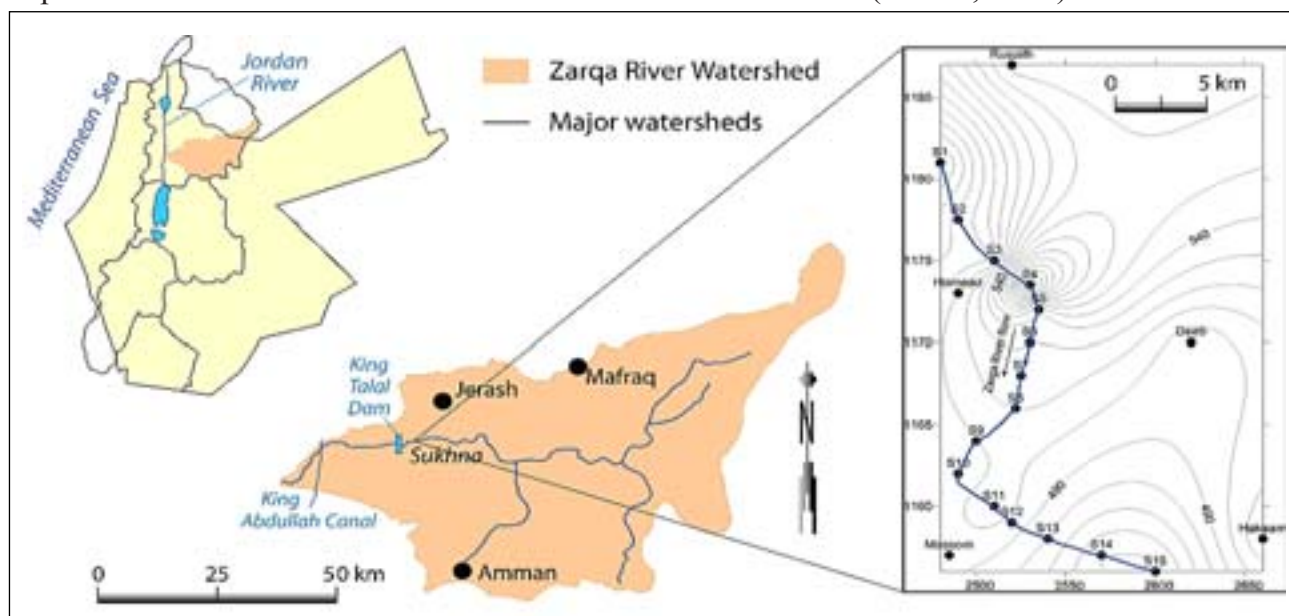


Figure 1. Catchment area of the Zarqa River (left, center) and location of the surface water (S1-S15) and groundwater samples at the Sukhna area on the elevation map (right).

Domestic and industrial waste water are usually treated to varying degrees and discharged to the surface water system. In some cases, however, raw sewage is discharged to dry river beds of nearby valleys. In addition, solid waste disposal sites are located within the catchment. Their leachates are known to reach the surface and groundwater resources causing pollution. The domestic and industrial waste water contribution to the flow of the river is estimated to be 50% (Salameh and Al-Ansari, 2000). The Zarqa River discharges its water into the reservoir of the King Talal dam. The dam which was constructed in 1977 with a total storage capacity of 56 million cubic meters was raised to store 89 million cubic meters in 1988.

Salameh and Al-Ansari (2000) showed that after the operation of the Khirbet Alsamara waste water treatment plant pollution parameters ( $BOD_5$ , COD, phosphorus and nitrates) increased drastically in Wadi Dhullel, in the reservoir of the King Talal Dam and in some areas of the Jordan valley. This treatment plant is very much overloaded and samples taken 12 km downstream from the plant in the Zarqa River were polluted. In addition, seepages of wastewater within the stabilization ponds of the plant to the groundwater aquifers are another negative factor on the groundwater quality of the area. Over-pumping of ground water in the Amman-Zarqa basin increases both salinity and pollution levels (Figure 2). Pollution is caused by seepage of treated wastewater, agricultural fertilizers, pesticides, leaching of seepage from landfill, and swamps of oil and factory waste.

## METHODOLOGY

Five groundwater samples from the wells distributed within the area and 15 surface water samples from the main river were collected in summer (Tables 1, 2, and 3; Figure 3). The temperature and electrical conductivity (EC), total dissolved solids (TDS) of the samples were immediately taken in the field using field portable meters. GPS was used to fix the exact sampling locations. One liter plastic bottles, previously rinsed carefully in the laboratory, and used for the collection of the samples. All samples were analyzed in the university lab. Titration using 0.02N

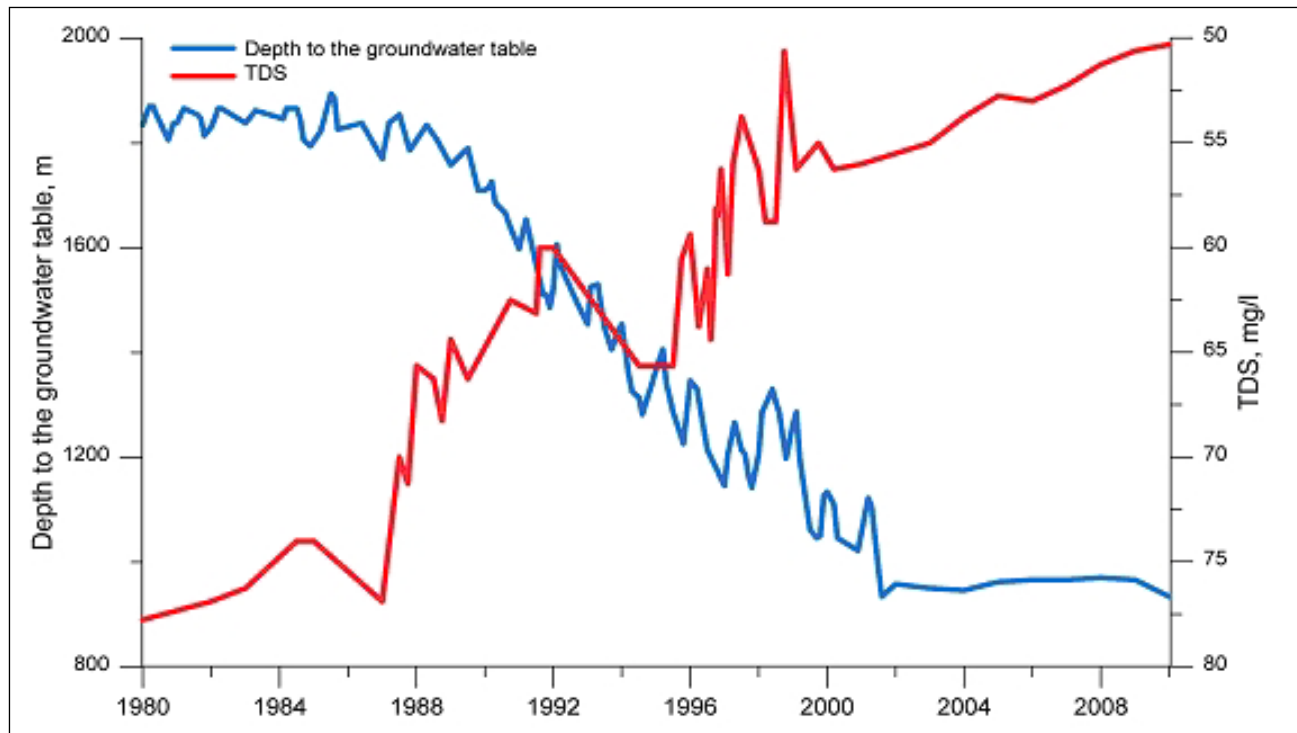


Figure 2. Variation of depth to the groundwater table (m) and TDS (mg/l) in Amman-Zarqa basin.



Table 1. EC ( $\mu\text{S}/\text{cm}$ ), TDS (sum of the major cations and anions;  $\text{mg}/\text{l}$ ), pH, and concentration of major ions ( $\text{mg}/\text{l}$ ) in surface and groundwater samples at Sukhna area.

Sample	EC	TDS	pH	K	Mg	Na	Ca	Cl	NO <sub>3</sub>	HCO <sub>3</sub>	SO <sub>4</sub>
S1	2510	2405	8,14	22.0	151	405	240	440	45.0	793	354
S2	2420	2051	8,09	30.0	164	294	77	347	49.0	1007	132
S3	1470	1408	8,00	10.0	156	450	95.4	294	36.0	366	36,9
S4	2450	2110	8,04	19.0	180	299	99	421	65.0	763	329
S5	1990	2207	7,91	19.0	158	299	205	431	50.0	671	423
S6	2390	1956	8,31	21.0	71	289	200	431	66.0	641	302
S7	2410	2155	8,28	19.0	78	299	210	455	75.0	641	453
S8	2290	1754	8,80	20.0	98	-	95	425	70.2	641	471
S9	2390	2256	8,11	26.0	89	249	250	421	60.3	1083	138
S10	2350	2544	8,15	27.0	133	269	50	421	66.8	1144	500
S11	2420	2155	8,10	26.0	82	259	100	401	73.9	1037	250
S12	2530	2259	8,20	26.0	180	470	55	372	72.8	1007	149
S13	2390	2308	8,18	26.0	69	259	40	406	76.5	1007	501
S14	2300	2776	8,22	27.0	151	259	240	426	78.0	1083	590
S15	2430	2972	8,16	27.0	160	402	260	445	80.0	1098	580
Deeb well	1910	1583	7,01	7.6	98	188	110	333	40.0	656	190
Resaifa well	1370	1108	7,08	8.2	73	190	81	98	68.0	458	200
Hakeem well	1810	1368	7,77	9.0	150	220	73	333	75.0	381	201
Ma'asoom well	1980	2191	7,42	11.0	150	399	210	415	77.4 8	503	502
Hamead well	2460	2254	6,93	21.0	153	406	261	421	79.0	442	550

EDTA was applied to analyze Ca and Mg, using 0.01M  $\text{AgNO}_3 - \text{Cl}$ , using 0.02M  $\text{H}_2\text{SO}_4 - \text{CO}_3$  and  $\text{HCO}_3$ . Na and K were analyzed using a flame photometer, and  $\text{SO}_4$ ,  $\text{NO}_3$ ,  $\text{PO}_4$  and  $\text{NH}_4$  – using a spectrophotometer at wave lengths of 492, 206, 700, and 309 nm, respectively. The heavy metals (Zn, Mn, Fe, Pb, and Ni) were analyzed using an atomic absorption spectrophotometry.

To find out the effect of irrigating vegetables with waste water, 24 samples of 8 different vegetables (Sweet and hot pepper, tomato, cauliflower, cabbage, squash, cucumber and eggplant) were collected from the farms along the river. These were irrigated from the water of the Zarqa River and the wells in the area. Similar vegetables were collected from the Ma'asoom area which was irrigated from unpolluted wells.

All samples were analyzed using the standard procedures recommended by American Public Health Association (APHA) (Clesceri et al., 1999; Horwitz, 2005). Prior to analyses, the vegetables were sliced into small pieces. One hundred gram of each type of the vegetable was taken in a 50 ml beaker. Three ml of perchloric acid ( $\text{HClO}_4$ ) was added to each sample which was then put on a hotplate. When the samples were dried, 10 ml of  $\text{HNO}_3$  was added. The sample was digested and a white vapor generated. The samples were allowed to cool, after which 10 ml of  $\text{HCl}$  and water (1:1 ratio) was added. Then the samples were transferred to flasks. Distilled water was added till the total volume was 50 ml. Each sample was duplicated and the procedure was repeated twice; their average values were used for in this paper.

## RESULTS AND DISCUSSION

### Water samples

All natural water samples showed relatively high concentrations of total dissolved salts (TDS)

Table 2. Concentration of heavy metals in surface and groundwater samples.

Sample	Zn (mg/L)	Mn (mg/L)	Fe (mg/L)	Pb (mg/L)	Ni (mg/L)
S1	5.2	0.0172	0.1588	0.099	0.045
S2	3.4	0.02	0.087	0.084	0.049
S3	60.2	0.09	0.12	0.05	0.05
S4	5.02	0.1	0.055	0.062	0.0524
S5	8.12	0.132	0.35	0.081	0.06
S6	15.1	0.14	0.0556	0.089	0.069
S7	15.98	0.16	1.56	0.09	1.61
S8	16.45	0.19	1.17	0.1	1.68
S9	16.25	0.2	1.57	0.18	1.7
S10	18.52	0.623	1.25	0.2	1.89
S11	17.4	0.56	1.48	0.25	1.9
S12	19.1	0.99	1.99	0.29	1.99
S13	18.56	1.006	2.99	0.34	2
S14	20.05	1.05	2.7	0.4	2.01
S15	20.1	2.03	2.01	0.64	2.09
Deeb well	4.98	0.1	0.5	0.045	0.01
Resaiifa well	13.8	0.19	0.99	0.05	0.09
Hakeem well	14.08	0.21	1.3	0.051	0.1
Ma'asoom well	15.75	0.2208	1.45	0.053	0.19
Hamead well	15.98	0.38	1.5	0.056	0.2

where the average reached 1936 mg/l (max 7400 mg/l and min 1160 mg/l) in surface water and 1200 mg/l in groundwater. The average pH value for the surface water was 8.2 and 7.2 for the groundwater. The average electrical conductivities (EC) of the surface water and groundwater samples was 2316 and 1906  $\mu\text{S}/\text{cm}$ , respectively (Table 1). There were local increases in TDS, pH and EC as result of water discharge from nearby factories, the Khirbet Alsamara wastewater treatment plant and the returned irrigation water from the farms on both sides of the river. As far as the groundwater is concerned, infiltration of irrigation water and seepages from the river are the main reasons for quality deterioration. In addition it was noted that some of the farmers pumped water directly from the river using wells installed in the river bed.

Surface and groundwater samples showed Na as the prevailing cation followed by Ca, Mg and K (Figure 4; Table 1). The most dominant anion in the surface and ground water samples was  $\text{HCO}_3$

Table 3. Concentration of heavy metals in vegetables at Mafraq and Zarqa (H:highest %,L:lowest %).

Vegetable type	Source	Zn	Mn	Fe	Pb	Ni
Sweet Prpper	Mafraq	0.0124	0.0457	0.1332	0.0587	0.0049
	Zarqa	0.0161	0.0494	0.2378	0.0703	0.0054
Tomato	Mafraq	0.0013	0.0434	0.2484	0.0453	0.0045
	Zarqa	0.0445(H)	0.0614	0.3842	0.1798	0.1327
Hot pepper	Mafraq	0.0153	0.0513	0.2324	0.061	0.0060
	Zarqa	0.0641	0.0779(H)	0.3141	0.0666	0.4188(H)
Cauliflower	Mafraq	0.0098	0.047	0.1241	0.0569	0.3181
	Zarqa	0.0283	0.0507	0.2326(H)	0.1201(L)	0.6807
Cabbage	Mafraq	0.0306	0.0646	0.1583	0.0459	0.3038
	Zarqa	0.0341(L)	0.0769	0.1957	0.0555	0.3518
Squash	Mafraq	0.0225	0.0474	0.2351	0.0599	0.2803
	Zarqa	0.0465	0.0547	0.2534(L)	0.3987(H)	0.3124
Eggplant	Mafraq	0.0097	0.0481	0.1855	0.0507	0.1994
	Zarqa	0.0303	0.0498(L)	0.2454	0.0668	0.2307(L)
Cucumber	Mafraq	0.0097	0.0373	0.114	0.0463	0.0044
	Zarqa	0.0156	0.0491	0.1379	0.0552	0.0046

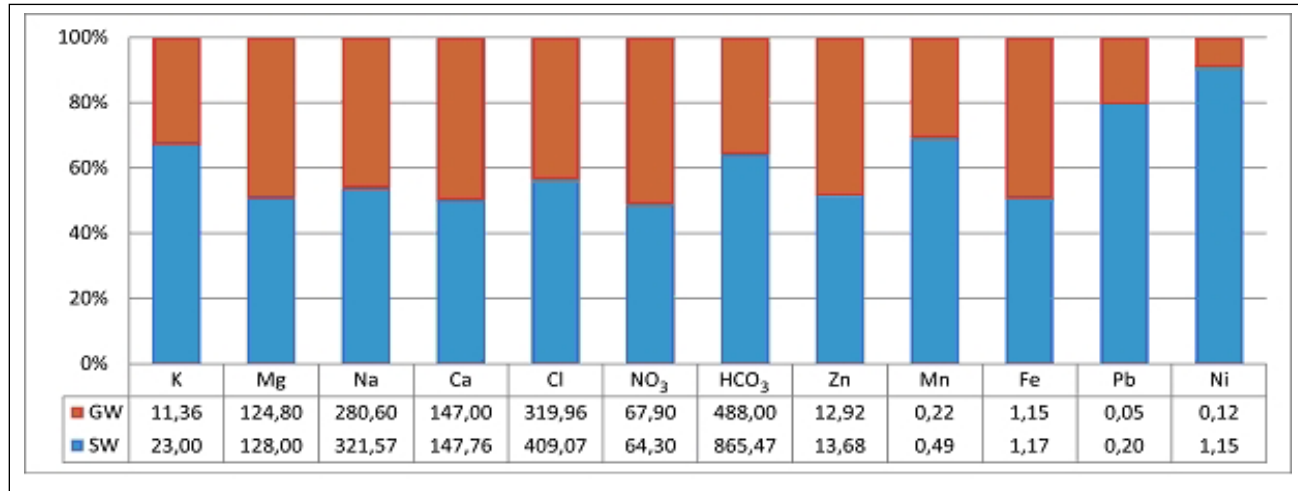


Figure 3. Average concentrations (mg/l) and relative proportion (%) of major and trace ions in surface water (SW) and groundwater (GW) samples.

followed by Cl and SO<sub>4</sub>. The origin of the cations and anions in the water samples is partly due to weathering of the sedimentary and igneous rocks in the catchment area. However, it is also severely affected by the wastewater in the area. The treated wastewater discharged from different factories distributed within the area, in particular those producing detergents, is rich in calcium carbonate. The use of nitrogen fertilizers and pesticides used in agricultural practices contributes also to increased concentrations of different cations and anions in the water.

Biochemical Oxygen Demand (BOD) and Chemical Oxygen Demand (COD) showed average values of 58.7 and 71.5 mg/l, respectively, in surface water samples. It is suggested that these values may be partly due to the effects of water discharged from the Khirb Esamra wastewater treatment plant where the annual average BOD and COD readings at the outlet are considerably

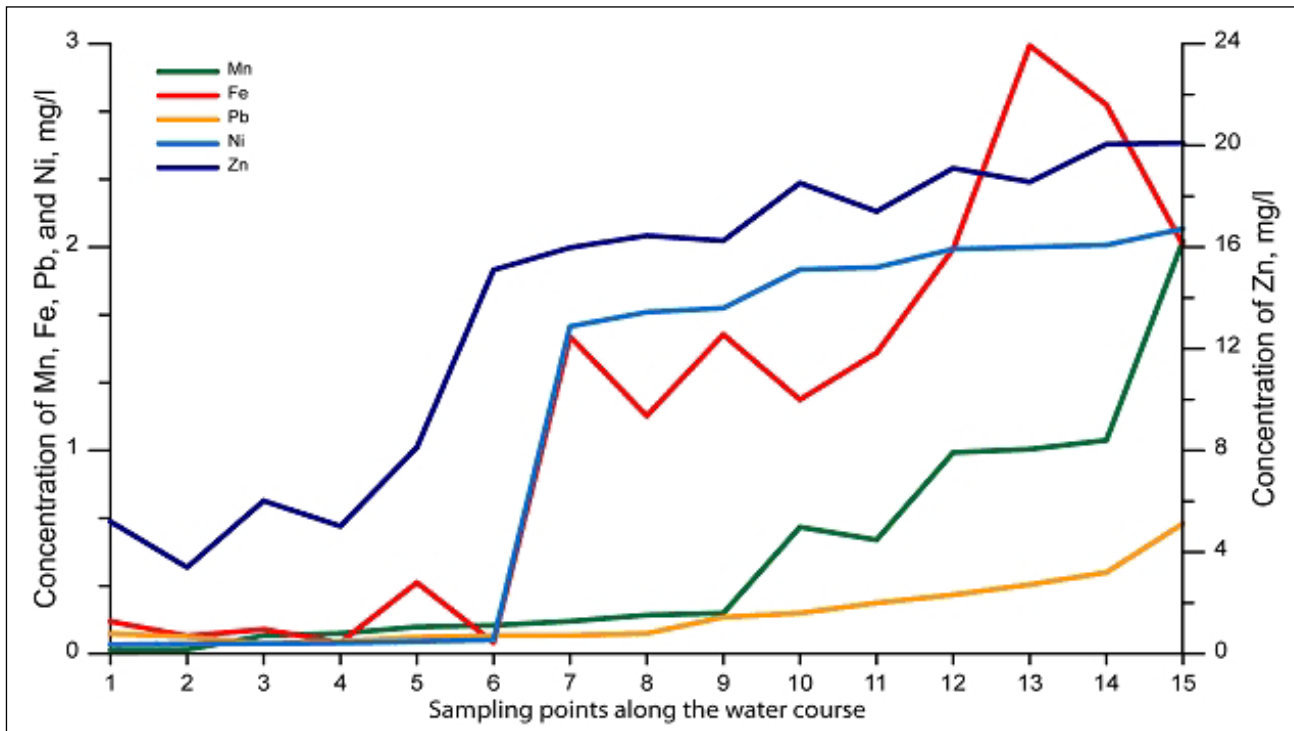


Figure 4. Evolution of concentration of heavy metals (mg/l) along the flowpath from sampling location S1 downstream.

higher, 144 and 356 mg/l, respectively (Salameh and Al-Ansari, 2000). In addition, food and paper producing factories contribute to the increase of BOD and COD in the surface water.

Heavy metals in all water samples showed very much higher concentrations (Table 2) and exceeded the threshold values (3.0, 0.4, 0.01, and 0.07 mg/l for Zn, Mn, Pb, and Ni, respectively) than in the drinking water guidelines given by WHO (2004 and 2011).

### **Surface water-groundwater interaction**

Irrigation strategies in the catchment are practiced by farmers in a non-systematic manner and provoke irregular mixing of surface water and groundwater leading to uncontrolled changes of the quality of water used for irrigation. When groundwater is extracted beneath the river bed its quality may deteriorate due to possible clogging of the river bed, precipitation or dissolution of redox sensitive elements, transformation of nitrogen and phosphorous etc. Decline of the quality of water used for irrigation may pose a risk to soils, to vegetables and the humans that ingest them (Al-Nakshabandi et al., 1997; Muchuweti et al., 2006; Khan et al., 2008).

In the Sukhna area, mixing of surface water and groundwater can be noticed from the similarity of their quality parameters, primarily TDS and EC. Additionally, a trend of increasing heavy metals content in the surface water can be observed from the sampling location S1 downstream. While Mn and Pb demonstrated a slow and steady rise, Ni, Zn, and Fe showed an abrupt concentration jump, indicating changes in redox conditions along the water flow (Figure 4). More conservative measures (e.g. EC, Cl, Ca, Na, HCO<sub>3</sub>) showed that there are two locations along the investigated water course that show altered chemical conditions. The first one is around sampling points S3-S5 and the second – S10-S13. At these locations, the river water composition presumably mixes with inflowing groundwater which suggests water extraction which has implications on the quality of water used for irrigation.

Interaction zones along the water flowpath are also indicated by the evolution of pH which ranges between 7.9 and 8.2 in places where the groundwater contribution is high and increases to 8.8 with minimum or no groundwater inflow. The rise of pH is also associated with an increase of Fe and Ni concentrations around S6 and S7. Further, increase of Fe at S12 is not related to pH which remains stable, however the increase may depend on the contributions of more Fe-rich groundwater found in the southern part of the area, namely in the Hakeeb and Masoon wells.

Due to limited hydrogeological knowledge of the area, identification of the exact groundwater flowpath is impossible. Therefore, we can merely define river stretches which have different water compositions. If uncontrolled pumping of the surface water exists in the area, it is assumed that in the vicinity of the sampling points S3-S5 and S10-S13 mixing of the river water and groundwater is controlled by groundwater/surface water extraction and provokes deleterious impacts on the water quality.

### **Vegetable samples**

The analyses of heavy metal contents (Zn, Mn, Fe, Pb and Ni) in eight types (three from each type) of common vegetables (sweet and hot pepper, tomato, cauliflower, cabbage, squash, cucumber, and eggplant) irrigated with treated wastewater (Zarqa region) showed higher concentrations than similar vegetables irrigated with clean water (Mafraq region).

Comparison between vegetables treated with clean and with treated wastewater showed that tomato and cauliflower are affected by four out of five metals analyzed in this study and therefore



considered the most polluted. Pure concentrations of heavy metals in vegetables from the two areas are shown in Figure 5. Further, the relation between vegetables from the Zarqa and Mafrqa regions are discussed. The four metals, namely Zn, Fe, Pb, and Ni, had concentrations higher than in the reference vegetables by 3423%, 155%, 397%, 2949% and 289%, 187%, 211%, 214% for tomato and cauliflower, respectively (Figure 6). Sweet pepper was mainly influenced by an increased content of Fe, which was almost 180% higher than that in sweet pepper from the Mafrqa region. Hot pepper had highly elevated concentrations of Ni (6980%) and Zn (419%), while squash demonstrated high Zn (207%) and Pb (666%). When all the heavy metals are considered, then the most effected vegetable is the hot pepper with average percent of heavy metals accumulation of 1559% while the least effected is cabbage – 116%.

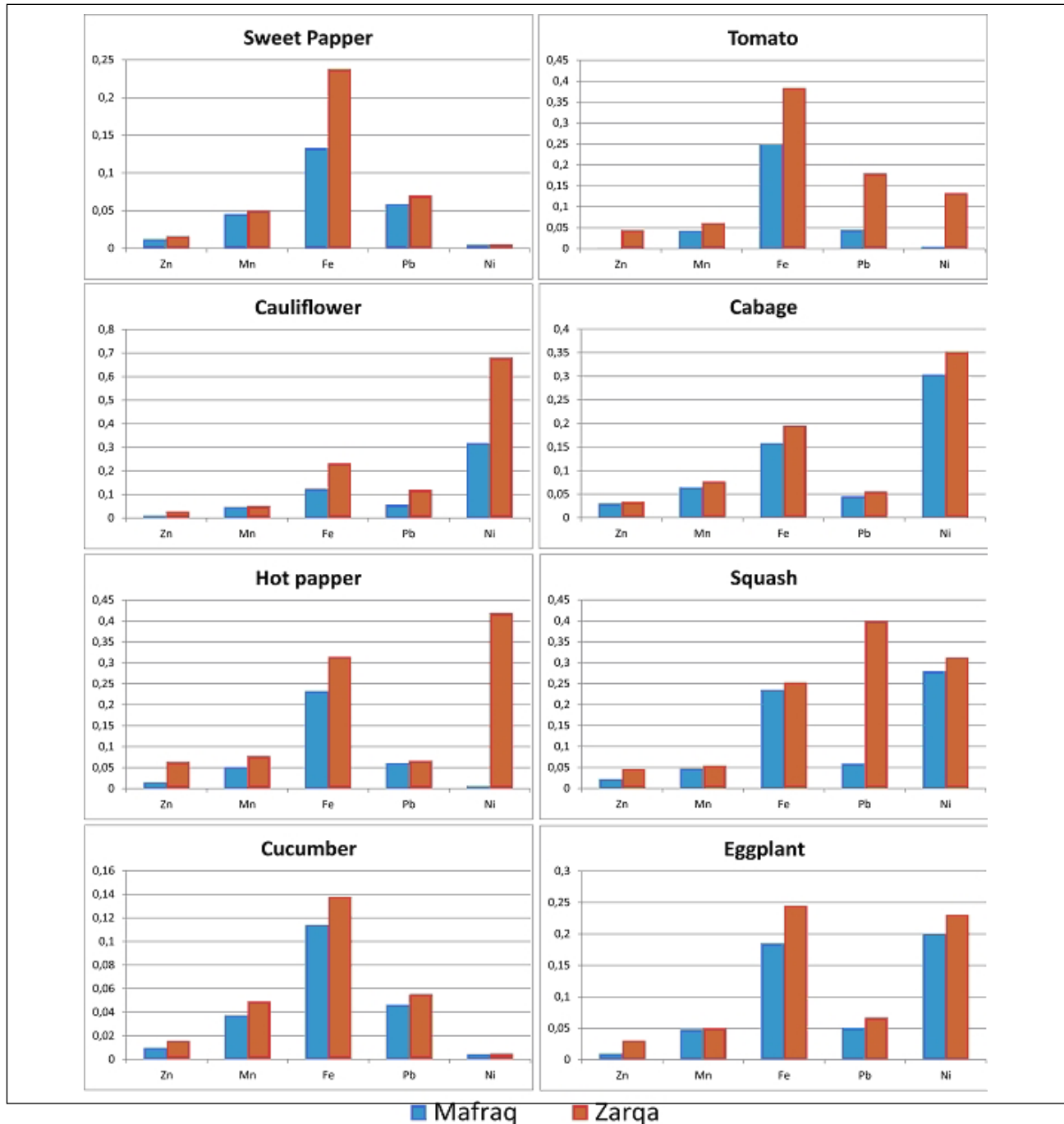


Figure 5. Concentration of Zn, Mn, Fe, Pb, and Ni in vegetables (mg/l).

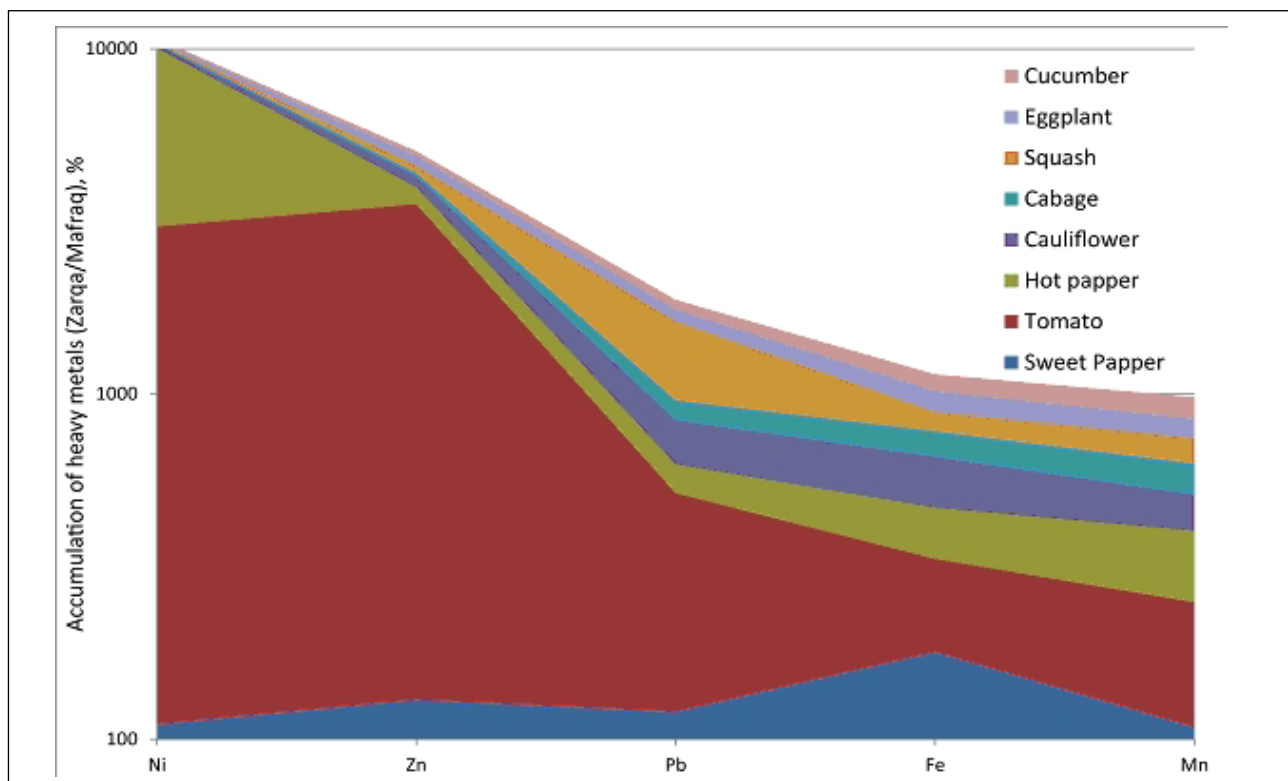


Figure 6. Relative accumulation of heavy metals (%) by vegetables irrigated with wastewater from the Zarqa area and with freshwater from the Mafraq area.

These figures indicate the effect of treated wastewater on the accumulation of heavy metals in the vegetables and suggest that certain vegetable crops are more vulnerable to heavy metal pollution than the others. Thus, Zn is mainly accumulated by cauliflower, tomato, hot pepper, and eggplant (in descending order); Fe – by sweet pepper, tomato, cauliflower; Pb – by tomato and squash; Ni – by tomato and hot pepper; Mn showed no any pronounced effect on any of the investigated crops. Cabbage and cucumber do not seem to be influenced by the treated wastewater applied for irrigation in the Zarqa region. Overall, Ni and Zn are the elements that vegetables uptake the most (Figure 5 and 6).

## CONCLUSIONS

Pumping of water for irrigation purposes in the area of Sukhna provokes unnatural interaction between the Zarqa River and the surrounding groundwater. Pollution and water withdrawal creates hazardous water quality issues. The river water, sampled at 15 locations along the flow path, showed elevated concentrations of heavy metals. It was noticed that the concentrations evolve along the river flow which can be related to interactions between the river and the underlying aquifer(s). Inflowing groundwater causes an immediate decrease of the heavy metal's concentrations, and this is followed by an increase downstream. At all locations, the river water and the groundwater showed concentrations of heavy metals far exceeding limits prescribed by WHO. Water from the area is intensively used for agriculture and thus poses a health risk for adults and children ingesting vegetables irrigated with the treated wastewater. Most of the investigated vegetables showed elevated concentrations of heavy metals. The most affected were tomato, hot and sweet pepper, cauliflower, squash, and eggplant. Ni and Zn were the metals accumulated the most, while Mn – the least. Tomato and cauliflower favored uptake of mainly Zn, Pb, Ni, and Fe; hot pepper accumulated chiefly Zn and Ni, while squash – Zn and Pb. Very little effect of irrigation by treated wastewaters

was observed in cabbage and cucumber. The results suggest that irrigation practice require governmental attention and control of water quality used for agriculture. The effects of irrigation by treated wastewaters have been used for only a limited period, and no adverse health impacts have been reported. However, these may become evident in the long term.

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