

# IMPACTS OF IRRIGATION WITH WATER CONTAINING HEAVY METALS ON SOIL AND GROUNDWATER – A SIMULATION STUDY

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**Abstract.** This research work intended to study the impacts of irrigation water containing various levels of copper, lead, and zinc on adsorption capacity of soil packed in 4'' plastic columns and obtained from two locations around the city of Nablus: Salem (A) and Deir Sharaf (B). Results of simulation experiments showed an increase in the copper, lead, and zinc concentrations in soil and in leachate with increasing the amount of metal in irrigation water. Copper, lead, and zinc concentrations increased also with soil depth and duration of application. The results also indicate that the self purification of both soils was highly affected by physical factors, i.e. the intermittent application of irrigation water to the soils in the columns caused soil wetting and drying cycles which resulted in the formation of cracks in shrunk soils specially in the top half of the columns. Crack formation is common in such clay soils due to the climatic conditions (Mediterranean type: dry summers and wet winters) and type of clay minerals in the soil. Thus, short circuiting of water through cracks results in moving contaminants fast and deep in the soil profile.

**Keywords:** groundwater, heavy metals, irrigation water, leachate, mediterranean climate, soil pollution

## 1. Introduction

Generally, wastewater is a liquid waste that is removed from residential, institutional and commercial establishments. Contaminants of domestic wastewater are categorized as: Disease causing microorganisms, essential plant nutrient elements, dissolved minerals and toxic chemicals and biodegradable organic matter (Manahan, 1990). Discharging raw wastewater to the environment causes pollution problems, therefore, the treatment of wastewater is essential to enhance overall water availability and conserve water resources (Aziz *et al.*, 1996; Moatgomery, 1988).

Urban wastewater collection practices in Palestine are such that many small industries are located within municipal boundaries and drain their wastewater into the municipal systems. Due to scarcity of fresh water, farmers use raw wastewater in irrigation (Haddad, 1990, 1993). The long term use of land application as a disposal method of raw wastewater and/or sludge may result in limiting soils agricultural ability to produce (Martin Edward, 1991).



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Several studies were conducted on the toxicity of urban wastewater in Palestine and on its impacts on groundwater, plants and soils (Haddad, 1994; Radi *et al.*, 1988; Environmental Protection, 2000; Haddad, 2000). The reported heavy metal concentrations of wastewater in Palestine range from 0 to 2075 mg L<sup>-1</sup> for zinc, 0 to 10 mg L<sup>-1</sup> for copper, and from 0 to 15 µg mL<sup>-1</sup> for lead (Environmental Protection, 2000; Haddad, 2000; Haddad *et al.*, 1999). Zinc has the highest level due to the fact that galvanized steel tanks mounted on the roofs of buildings and houses are used in Palestine for water supply storage.

In conventional wastewater treatment, considerable portions of heavy metals remain in the treated effluent if special advanced treatment is not conducted. Thus, long term effects of irrigation with wastewater might include pollution of ground water and soil with heavy metals such as: Pb, Cu and Zn ions (Lebourg *et al.*, 1998). Other impacts of treated wastewater in agriculture include the health impacts of possible contamination of crops by pathogenic bacteria and heavy metals (Farid *et al.*, 1993).

There is a rapidly growing awareness of the threat to water resources caused by highway drainage and sewage effluents. Some of the most significant contaminants are heavy metals such as copper, zinc and lead (Farid *et al.*, 1993; Selim and Iskandar, 1992; Laxen and Harrison, 1977; Chatzoudis and Rigas, 1998; Mendoza *et al.*, 1996).

Though copper is not a cumulative systemic poison, large dose (>100 mg) of copper are harmful to humans and might cause central nervous system disorder, failure of pigmentation of hair and adverse effects on Fe-metabolism that results in liver damage. Excess copper may also be deposited in the eyes, brain, skin, pancreas and myocardium (McAnally *et al.*, 1997). Lead is a cumulative poison to humans. Its major effects are impairment of hemoglobin and porphyrins synthesis. Zinc cause muscular weaknesses and pain, irritability and nausea (AWWA, 1990).

In Palestine, the availability of renewable water resources to maintain various human needs is poor and scarcity is accelerating with time. Therefore, alternative water resources development options such as brackish water desalination and the reuse of treated wastewater is gaining much importance at present. The use of these options is expected to be obligatory with time.

The present work aims to conduct a column study to simulate Pb, Cu and Zn ions adsorption on soil and in leachate from two locations near the city of Nablus in order to recommend if these soils are suitable for wastewater application, based on simulation results.

## 2. Methodology

All chemicals were Analytical Grade reagents, deionized water was used for preparation and dilution of metal solutions. All bottles and other containers (except

columns) were treated with 1 M HNO<sub>3</sub> solution before being washed with deionized water and dried.

### 2.1. EXPERIMENTAL SETUP

The experimental setup consisted of 20 PVC columns, 4" in diameter and 2 m long. Soil samples from the top 100 cm layer soil were collected from the two locations near Nablus city, Salim (A) and Deir Sharaf (B), before winter 1998. Small stones (if any) were removed by hand from soil samples and 19 kg of soil was mixed and then placed in each column in layers of 10 cm.

To allow drainage flow freely without eroding soil from columns, a thin layer of gravel and sand was placed in the lower end of the column, with a plastic mesh screen at the bottom of the column. A plastic container was placed under each column to collect drainage water.

For each soil, three treatments were carried out. These treatments represent simulation of irrigation for 2, 10 and 20 yr periods in triplicates. Two other columns were used as blanks. Rainwater was simulated for the blanks by applying 250 mL of rainwater to each column as needed.

### 2.2. WATER AND HEAVY METALS APPLICATIONS

To each column, a solution containing known combinations of Pb<sup>2+</sup>, Cu<sup>2+</sup> and Zn<sup>2+</sup> was added. The concentration of each metal was estimated based on following (Table I):

1. Average rainfall, evapotranspiration, crop irrigation requirements and leaching in Nablus area.
2. Volume of irrigation water = 1025 mm (m<sup>3</sup>/dunum), assuming fruit trees will be planted in these areas.
3. Volume of leaching water = 403 mm (m<sup>3</sup>/dunum). Considering the column volume, the volume of irrigation water for simulation was 8.05 L per column per year.

### 2.3. WATER APPLICATION

Two, 10 and 20 yr were selected for simulation to study short, medium and long terms' effects of simulation. Details are found in Table I.

### 2.4. HEAVY METAL APPLICATION

Metal solutions were prepared from their nitrate salts and stored in polyethylene bottles.

The amount of heavy metals applied in irrigation was based on the maximum allowable limit by FAO (10, 5 and 10 mg L<sup>-1</sup> for Zn<sup>2+</sup>, Cu<sup>2+</sup> and Pb<sup>2+</sup>, respectively) (FAO, 1980). Details are shown in Table I.

TABLE I  
Water and heavy metal application in both soils<sup>a</sup>

Description	Years			
	1	2	10	20
Depth of irrigation water (mm)	1025	2050	10250	20500
Depth of leachate water (mm)	403	806	4030	8060
Volume of irrigation water (L/column)	8.05	16.1	80.5	161
Volume of leachate water (L/column)	3.17	6.34	31.7	63.4
Weight of zinc added to each column (mg)	80.5	161	805	1609
Weight of copper added to each column (mg)	40.25	80	402	805
Weight of lead added to each column (mg)	80.5	161	805	1609
Duration (from 28-12-1998 to		23-4-1999	8-5-1999	19-5-1999

<sup>a</sup> Humidity was 49–69% and pan evaporation was 2.4–7.8 mm day<sup>-1</sup>.

## 2.5. LEACHATE

Water leaching from the columns drained into the plastic containers under the columns. To illuminate evaporation of drainage water, the containers were covered by plastic sheets. Depth of drainage water in the collection containers was monitored and when it reach about 10–15 cm (this depth equals the height of the small layer of gravel and sand in the bottom of each column), water was collected and transferred into storage containers. Storage containers were polyethylene bottles of 4 L in volume. To each storage bottle, 10 mL of 1 M HNO<sub>3</sub> has been added.

## 2.6. METALS IN SOIL

Metal concentrations in each soil were measured before treatment. After simulation was finished, soil was evacuated from the PVC column, which was cut into three pieces or 4 cross sections at 10, 67, 133 and 200 cm heights. Each soil patch was dried and the metal ions were extracted from a given weight using NH<sub>4</sub>Ac/EDTA solutions. The extracts were then analyzed for Pb, Zn and Cu ions. Extractable ions were determined as this research is concerned with the amounts adsorbed on soil surfaces which could be a fraction of total amounts of metals in the soil. Adsorbed metals influence plant and environment as they interact with soil solution and plant roots.

## 2.7. SOIL ANALYSIS

Soils were analyzed for chloride, carbonate, sodium, potassium, magnesium, calcium, copper, zinc, lead, phosphorus, total dissolved salts (TDS), organic matter

content and pH according to standard procedure (Laboratory Manual, 1992; Reeve, 1994). Concentrations of chlorides, sodium, potassium, magnesium and copper were determined in soil extracts. Concentrations of zinc, lead and copper were determined from extracts by  $\text{NH}_4\text{Ac}/\text{EDTA}$ .

Moisture content, particle size distribution, bulk density and specific gravity of soils were also measured following standard methods (Das Braja Soil Mechanics, 1941).

## 2.8. INSTRUMENTATION

Analysis of Cu, Zn, and Pb were carried out by atomic absorption spectrophotometry using Atomic Absorption Spectrophotometer VIDEO 11, which was calibrated, using supplied standards, prior to each use. Electrical conductivity was measured using Conductivity Meter 4010 instrument. pH was recorded using a Corning pH Meter Model 12.

## 3. Results and Discussion

### 3.1. CHEMICAL ANALYSIS

Results of soil chemical analysis are shown in Table II. The concentration of total dissolved solids (TDS) and electrical conductivity readings were low for both soils, which indicate that neither soil is saline (FAO, 1980; McNeal *et al.*, 1982). It also indicate that the average precipitation in the area of  $600 \text{ mm yr}^{-1}$  is sufficient with time to wash salts from soil especially that both soils showed low sodicity and good permeability, in spite of soil clay nature.

The conductivity readings are supported by the low concentration found for calcium, magnesium, potassium, sodium, copper, zinc, lead, phosphorous chlorides and nitrates. For all cations and anions tested, the found concentration was below the acceptable limits of agricultural soil.

The relatively high percentage composition of calcium carbonates in soil (13–20%) is attributed to the fact that parent materials of these soils were originated from rocks rich in calcium carbonates such as limestone and dolomite. The soil pH (7–8) and the high buffer capacity make soils suitable for most plants, as the nutrient availability of most macronutrient is high at this pH, though some micronutrient such as iron and manganese demand more acidic soil.

The low sodicity resulting from low sodium carbonate content indicates that both soils were not alkaline ( $\text{pH} < 8.3$ ) (Schwab *et al.*, 1993). The high calcium carbonate content and the low exchangeable sodium on the surfaces of these soils result in the formation of highly stable aggregates with suitable permeability and hence good drainage ability (Sposito, 1989).

TABLE II  
Chemical and physical analysis of soil

Type of analysis	Salim	Deir Sharaf
Electric conductivity of soil extract (mmho cm <sup>-1</sup> )	1.2	1.3
Total dissolved solids for soil extracts (mg g <sup>-1</sup> )	3.84	4.16
Soil extract pH	7.29	7.11
Chlorides in soil extract (μg g <sup>-1</sup> )	87.5	175
Calcium and magnesium in soil extract (meq L <sup>-1</sup> ) <sup>a</sup>	11.5	10
% Organic matter content	2	12
Phosphorous in soil extract (μg g <sup>-1</sup> )	460	240
Potassium in soil extract (μg g <sup>-1</sup> )	140	180
Sodium in soil extract (μg g <sup>-1</sup> )	1140	880
% CaCO <sub>3</sub>	13.75	20
Copper extracted by NH <sub>4</sub> Ac/EDTA (μg g <sup>-1</sup> )	4.84	2.52
Zinc extracted by NH <sub>4</sub> Ac/EDTA (μg g <sup>-1</sup> )	2.86	1.24
Lead extracted by NH <sub>4</sub> Ac/EDTA (μg g <sup>-1</sup> )	2.66	0.94
% Moisture content	9.4	8.5
Specific gravity (gm cm <sup>-3</sup> )	2.6	2.7
Bulk density (gm cm <sup>-3</sup> )	1.7	1.8
% Silt	43.2	41.6
% Clay	37.6	35.6
% Sand	19.2	22.8
Soil texture	Clay loam	Clay loam

<sup>a</sup> 5.0 g dry soil was extracted with 100 mL NH<sub>4</sub>Ac (1 N).

### 3.2. PHYSICAL ANALYSIS

Due to the formation of aggregates, sieve analysis was not suitable to determine soil texture. Therefore, hydrometer analysis was utilized and results are summarized in Table II.

Soil from both locations was found to contain high clay percentage and classified as clay loam soil based on textural triangle (Beaton *et al.*, 1975; Fitzpatrick, 1986). Both sites from which soil was collected are located within alluvial plains of wadis. The low erosion in these plains and the high annual precipitation allow the formation of clay. However, medium weathering rates are characteristics of the environmental conditions of the area in these plains. These conditions result in forming montmorillonite clay minerals in the area. This could be easily observed in the Plains of the West Bank and the response of soils there to the weather conditions. The most common response of such clay minerals is the formation of cracks in summer (dry weather) and the expansion of soils in winter (wet weather). Depth of these cracks exceeds 1 m in these soils and might reach several meters in some

deep soil profiles due to the long dry summer which might exceed 6 months. The soil bulk density is high due to shrinkage and formation of aggregates during the long months of the dry hot summer season. However, the specific gravity of soil particles is typical for such soils with calcium carbonates parent materials.

### 3.3. SOIL ANALYSIS AFTER SIMULATION

Application of heavy metals was carried out to simulate their impact on soil and leachate.

The concentrations of metals were analyzed before and after the simulation experiment and the results are presented in Table III.

Because the industrial zone is located in the eastern side of Nablus City, Soil A was more polluted with the three heavy metals than in Soil B.

After simulation experiments, soil samples were taken from columns at different depths (10, 67, 133 and 200 cm), thereafter analyzed for copper, zinc and lead content (Table III).

For all metals employed in the three terms of treatments, the metal concentration increased with depth. This could be attributed to one or more of the following factors:

1. The experimental setup allows better ion exchange between applied solution and soil particles in the lower part of the column.
2. The applied metal concentrations could be low enough to be washed by the running water of irrigation.
3. The possibility of short circulating on the walls of the PVC column and through the soil cracks due to wetting and drying conditions and thus preventing ion exchange between soil and applied solution in the upper part of the column.

Heavy metals residue was calculated for each element in each column (Table IV). In all cases, the residue increased with increasing the concentration of metal applied and simulation period. This indicates that heavy metals application in irrigation water is accumulative. However, Soil A retained more heavy metals than Soil B.

Although the present results show an increase in heavy metals concentrations with depth, the actual increase in the field might be different as a result of different evapotranspiration rates from different soil layers depending on plant physiology and distribution of plant roots.

### 3.4. LEACHATE ANALYSIS

Figure 1 shows the changes in electrical conductivity of soil with time for the long term treatment for both soils. The electrical conductivity of soil was enhanced with duration of treatment whereas salinity decreased.

TABLE III

Concentrations of copper, zinc and lead in soil at different depths at the end of simulation period

Column	Residue concentration in soil batches ( $\mu\text{g g}^{-1}$ )			Total (mg)	
	Lower	Middle	Upper		
<b>Cu</b>					
2 yr	Salim	11.11 $\pm$ 1.07	3.63 $\pm$ 0.46	3.45 $\pm$ 0.12	115.14 $\pm$ 9.77
	D. Sharaf	10.00 $\pm$ 0.07	2.03 $\pm$ 0.16	2.25 $\pm$ 0.08	90.48 $\pm$ 1.53
10 yr	Salim	37.61 $\pm$ 0.47	4.45 $\pm$ 0.58	3.57 $\pm$ 0.59	288.97 $\pm$ 9.84
	D. Sharaf	36.49 $\pm$ 0.77	3.03 $\pm$ 0.74	2.67 $\pm$ 0.84	267.27 $\pm$ 13.6
20 yr	Salim	81.61 $\pm$ 0.81	5.03 $\pm$ 1.13	4.50 $\pm$ 0.82	577.18 $\pm$ 11.70
	D. Sharaf	78.67 $\pm$ 0.19	3.38 $\pm$ 1.02	2.83 $\pm$ 0.55	53057 $\pm$ 9.38
<b>Zn</b>					
2 yr	Salim	19.89 $\pm$ 0.36	1.67 $\pm$ 0.22	1.80 $\pm$ 0.16	147.9 $\pm$ 2.91
	D. Sharaf	17.41 $\pm$ 0.99	0.99 $\pm$ 0.41	0.84 $\pm$ 0.34	121.9 $\pm$ 5.07
10 yr	Salim	93.94 $\pm$ 1.35	2.04 $\pm$ 0.15	1.87 $\pm$ 0.27	619.7 $\pm$ 9.91
	D. Sharaf	91.29 $\pm$ 1.05	1.30 $\pm$ 0.05	1.09 $\pm$ 0.15	593.3 $\pm$ 5.93
20 yr	Salim	181.33 $\pm$ 1.85	4.15 $\pm$ 0.06	2.29 $\pm$ 0.22	1189.2 $\pm$ 13.43
	D. Sharaf	170.13 $\pm$ 7.99	1.85 $\pm$ 0.49	1.43 $\pm$ 0.17	1089.2 $\pm$ 21.25
<b>Pb</b>					
2 yr	Salim	18.61 $\pm$ 0.99	2.51 $\pm$ 0.08	2.29 $\pm$ 0.05	148.2 $\pm$ 5.8
	D. Sharaf	17.75 $\pm$ 0.62	1.75 $\pm$ 0.66	1.37 $\pm$ 0.17	132.2 $\pm$ 8.1
10 yr	Salim	90.69 $\pm$ 0.72	3.53 $\pm$ 0.08	3.04 $\pm$ 0.17	615.9 $\pm$ 4.79
	D. Sharaf	88.97 $\pm$ 0.71	2.03 $\pm$ 0.24	1.59 $\pm$ 0.30	586.4 $\pm$ 1.40
20 yr	Salim	184.60 $\pm$ 0.53	5.57 $\pm$ 0.81	3.36 $\pm$ 0.26	1225.7 $\pm$ 6.89
	D. Sharaf	176.64 $\pm$ 11.29	2.75 $\pm$ 0.32	2.09 $\pm$ 0.30	1149.4 $\pm$ 68.52

The amount of heavy elements in leachate (Table IV) was also dependent on the simulation period as expected. However, no significant difference in the amount of heavy elements was found in leachate from the two soils. This could result from similar physical and chemical characteristics of the two soils. Therefore their self-purification capacities are also similar. The threat to groundwater, if happens, will rather depend on the hydrogeological characteristics of the two areas.

Mass balance of metal requires that the added amount in irrigation water plus that present initially in soil, should equal to the metal residue in soil plus that in

TABLE IV  
Mass balance of copper, zinc and lead

		Actual	Mass balance calculations			
			Initial (mg)	Applied (mg)	Leachate (mg)	Residue (mg)
<b>Cu</b>						
2 yr	Salim	115.14	91.96	80.00	2.21	169.75
	D. Sharaf	90.48	47.88	80.00	3.25	124.64
10 yr	Salim	288.97	91.96	402.00	38.45	455.51
	D. Sharaf	267.27	47.88	402.00	39.43	410.45
20 yr	Salim	577.18	91.96	805.00	45.12	845.23
	D. Sharaf	537.57	47.88	805.00	46.61	806.27
<b>Zn</b>						
2 yr	Salim	147.9	54.37	161	1.01	214.36
	D. Sharaf	121.9	23.55	161	1.41	183.14
10 yr	Salim	619.7	54.37	805	17.07	842.30
	D. Sharaf	593.3	23.55	805	18.12	810.43
20 yr	Salim	1189.2	54.37	1609	66.33	1597.0
	D. Sharaf	1098.2	23.55	1609	82.32	1550.2
<b>Pb</b>						
2 yr	Salim	148.2	50.578	161	0.61	210.96
	D. Sharaf	132.2	17.805	161	1.01	177.80
10 yr	Salim	615.9	50.578	805	19.08	836.49
	D. Sharaf	586.4	17.805	805	19.67	805.88
20 yr	Salim	1225.7	50.578	1609	33.18	1623.1
	D. Sharaf	1149.4	17.805	1609	34.26	1592.5

leachate (Table IV). The difference between the amount of metal found by mass balance calculations and that found experimentally in soil, especially for two years treatment could be explained by one or more of the followings:

1. Precipitation of metal ions as insoluble salts.
2. Adsorption of heavy metals on the columns' surfaces.
3. The variation in temperature during the experimental period.

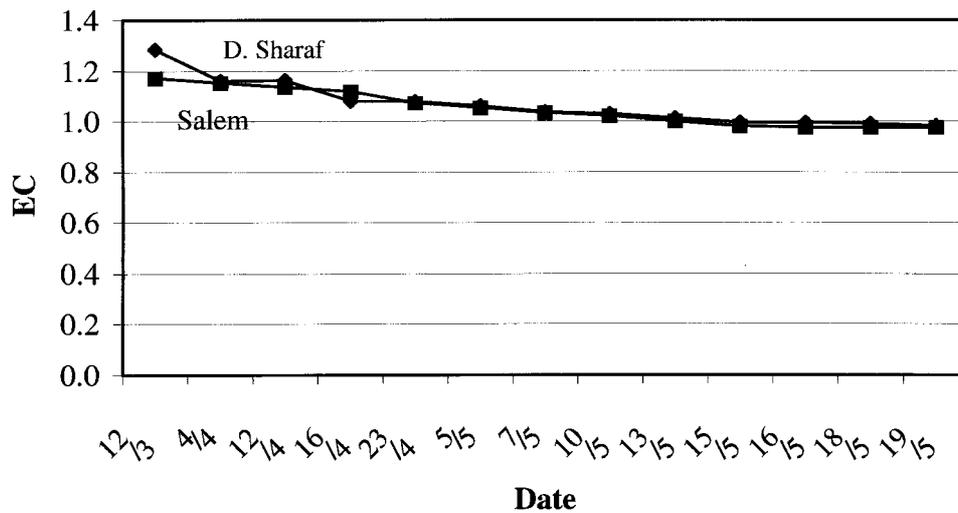


Figure 1. Electric conductivity ( $\text{mmho cm}^{-1}$ ) of the leachate versus date of experiment for the 20 yr term treatment.

#### 4. Conclusions

The two soils were found to have similar chemical and physical properties and thus showed similar response to simulation experiments.

The nature of metal, soil properties and the metal loading level affected redistribution of metals in soil. The three metals used showed cumulative effect on soil and in leachate, and thus the possibility of groundwater contamination does exist. The concentrations of these elements and other heavy metals should be reduced as much as possible and industrial waste should be either separated or treated before dumped in domestic wastewater.

The clay nature of soils and their high content of montmorillonites were responsible for the formation of large and deep cracks due to wetting and drying weather conditions. As a result, cation exchange between soil and running solution was not allowed which reduces self-purification capacity of soil. Continuous monitoring of wastewater, soil and groundwater qualities are essential for any sustainable reuse of wastewater in Palestine.

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