Assessment of Heavy Metals Pollution For Cultivated Soils, Irrigated by Several Irrigation Waters Varying in Their Qualities in Al-Hassa Oasis, Kingdom of Saudi Arabia

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ABSTRACT:

Twenty four composite surface soil samples(0-30cm depth) representing cultivated soils irrigated for long-term (more than fifteen years) with different irrigation water qualities:(i-ground water (GW), ii-ground water + agricultural drainage water (DW), iii-ground water + tertiary treated wastewater (TTWW) and iv- ground water,(GW) + agricultural drainage water,(DW) + tertiary treated wastewater,(TTWW), were analyzed for their contents, from Cu, Mn, Fe, Zn, Co, Cd, Pb, As and Ni. The results showed that, these contents can be arranged in the following descending order: Fe > Cu > Mn > Zn > Ni > Pb > Co > As > Cd. Generally, the different irrigation water qualities can be arranged according to their effects on total heavy metal contents in the soils in the following order: (GW+DW+TTWW) > (GW+TTWW) > (GW+DW) > (GW). Based on the geo-accumulation index, I_{geo} values for Mn, Fe, Co, and Cd, the soil irrigated with groundwater is uncontaminated with these elements. On the other hand I_{geo} values for Cu, Zn, Pb, and Ni are > 0 and <1, indicating that the soil is uncontaminated to moderately contaminated with these elements. In general, I_{geo} values for the soil irrigated with (GW+ DW+TTWW) showed patterns of heavy metals contamination similar to those of the soils irrigated with (GW+TTWW) and those irrigated with (GW+DW) but with different levels. Based on the Enrichment factor (EF) the studied soils are significantly contaminated with Cu, Ni, and Zn due to irrigation with, ground water, Cu, Ni, Pb, and Zn in the soil irrigated with (GW+DW), Cu, Pb, Zn, Ni, and As in both soils irrigated with (GW+TTWW) and (GW+ DW+TTWW). The results reveal that the *EF* mean values of heavy metals in the studied soils irrigated with different irrigation water qualities, can be arranged in the following descending order: (GW + DW+TTWW)> (GW+TTWW) > (GW+DW) > (GW).

Key words: Irrigation water quality. Geo-accumulation index. Enrichment factor. Heavy metals pollution and Pollution index.

INTRODUCTION:

Al-Hassa Oasis is located in the south of the Eastern area of Saudi Arabia about 65 km from the Arabian Gulf with an area of about 120 km^2 with a population of more than one million person. It is bounded by the Ad- Dahna deserts. Al-Hassa Oasis is an important agricultural area in east of Saudi Arabia. Agriculture is the most significant sector in the Oasis and recently large agricultural enterprises were established in the Oasis with the support provided by Saudi Arabian government. There are about of 16000 ha cultivated area in Al-Hassa Oasis. Around three million date palms produce wide ranges of varieties of high quality dates, among the other crops rice, citrus and other fruits are prominent. The deficiency of water resources is the most significant problem in the Oasis. Although all these lands were planned to be irrigated by spring water, ground water resource is insufficient today. Therefore, unconventional water resources such as drainage water and treated waste water were used in irrigation practices. With all these water sources, the available amount of irrigation water is still insufficient under the prevailing irrigation practices and conditions.

The reuse of treated wastewater is a good option for increasing water supplies for agricultural use. One of its benefits is the plant use of the water nutrients and therefore a reduction in the pollution load that wastewater contributes to the surface water supply (Zekri and Koo., 1994). There is considerable interest and concern in the long-term effects of treated wastewater on crops intended for human consumption. Presence of heavy metals in soils above the permissible limits poses threats to public health. Naveedullah *et al.* (2013) determined concentrations of seven metals in cultivated soils from Yuhang county, Zhejiang, China. Multipartite statistical approaches were used to study the variation of metals in soils during summer and winter seasons. Contamination of soils was evaluated on the basis of enrichment factor (EF), geo-accumulation index (*I*geo), contamination factor (*Cf*), and degree of contamination (*C*deg). They found that the heavy metal concentrations were higher in winter as compared to summer season. Cr and Cd revealed random distribution with diverse correlations in both seasons. Principal component analysis and cluster analysis showed significant anthropogenic intrusions of Zn, Cd, Pb, Cr, and Cu in the soils. Enrichment factor revealed significant enrichment (EF > 5) of Zn, Cd, and Pb, whereas geo-accumulation index and contamination factor exhibited moderate to high contamination for Zn, Cr, Cd, and Pb. In light of the studied parameters, permissible limit to very high degree of contamination (*C*deg> 16) was observed in both seasons.

Pollution of the natural environment by heavy metals is a universal problem because these metals are indestructible and most of them have toxic effects on living organisms, when permissible concentration levels are exceeded. Heavy metals frequently reported in literature with regards to potential hazards and occurrences in contaminated soils are Cd, Cr, Pb, Zn, Fe and Cu (Akoto *et al.*, 2008). Soil samples represent an excellent media to monitor heavy metal pollution because anthropogenic heavy metals are usually deposited in top soils (Govil et al., 2001). Heavy metal contaminated soil affects the ecosystem when heavy metals migrate into groundwater or are taken up by flora and fauna, this results in great risk to ecosystems due to bioaccumulation (Bhagure and Mirgane, 2010).

Vegetables cultivated in soils polluted with toxic and heavy metals take up such metals and accumulate them in their edible and non-edible parts in quantities high enough to cause clinical problems both to animals and human beings consuming these metal-rich plants as there is no good mechanism for their elimination from the human body (Bhuiyan *et al.*, 2011). Heavy metals and trace elements are also a matter of concern due to their non-biodegradable nature and long biological half-lives. (Singh *et al.*; 2012). The anthropogenic sources of heavy metals in agricultural soils include mining, smelting, waste disposal, urban effluent, vehicle exhausts, sewage sludge, pesticides, fertilizers application. (Luo *et al.* 2012). Due to spatial variability in lithology and mineralogy, world reference has been known to be erratic when used to determine enrichment factors (Abrahim and Parker, 2008).

The geo-accumulation index (I_{geo}) has been used since the late 1960 and has been widely employed in European trace studies. Originally, it is used for bottom sediments (Muller, 1969), and has been successfully applied to the measurement of soil contamination (Loska *et al.,* 2003). The I_{geo} enables the assessment of contamination by comparing current and pre-industrial concentrations, although it is not always easy to reach pre-industrial sediment layers. Enrichment factor (EF) has been employed for the assessment of contamination in various environmental media by several researchers (Lue *et al.*, 2009). Enrichment Factor (EF) of an element in a sample is based on the standardization of a measured element against a reference element. A reference element is often the one characterized by low occurrence variability. It is used to differentiate heavy metal sources. To assess the extent of contamination of heavy metals in soil and also provide a measure of the degree of overall contamination along a particular soil, pollution index has been applied (Hakanson, 1980).

The pollution index reflects the metal enrichment in the soil. The geochemical background values in continental crust averages of the trace metals under consideration reported by Taylor and Mclennan (1985) were used as back ground values for the metal.

The objective of the present study is to: (1) assess heavy metal contamination of agricultural soil irrigated with different irrigation water qualities in Al-Hassa Oasis, Saudi Arabia using three parameters which are namely; the geo accumulation index (I_{geo}), Enrichment Facto (EF) and Pollution Index (PI).

MATERIALS AND METHODS: Soil Samples Collection and Analysis Twenty four composite surface soil samples (0-30cm depth) were collected from farms representing cultivated soils irrigated with different irrigation water qualities for long-term (more than fifteen years). The collected soil samples were air-dried, gently crushed, sieved through a 2 mm sieve and stored in plastic bags for chemical and physical analyses. Soil paste pH value and EC values of soil paste extracts were determined according to Sparks *et al.* (1996). Particle size distribution was carried out using the hydrometer method according to Gee and Bauder,(1996). Organic matter was determined according to the method described by Nelson and Sommers,(1982). The concentrations of soluble cations and anions (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, CO_3^{-2}, HCO_3^{-}, Cl^{-} and SO_4^{-2}) were determined according to the method described by Loeppert and Suarez, (1996). Soil samples were digested for total metal analysis using a concentrated acid mixture of H_2SO_4, HF and $HClO_4$ according to Hossner (1996). The filtrated digests were analyzed for the total contents of Cu, Mn, Fe, Zn, Co Cd, Pb, As and Ni using Shimadzu Atomic Absorption Spectrophotometer (AAS 6300).

Moreover, the contamination assessment of the study soils was calculated. The assessment of soil or sediment enrichment with metal ions was carried out by the index of geo- accumulation I_{geo} and enrichment EF factor (Lue *et al.*,2009); beside of the Pollution Index (PI).

Geo-accumulation Index (I_{geo}):

In this study, the I_{geo} for cultivated soil irrigated with different irrigation water qualities was calculated using the following equation:

$$I_{geo} = \log_2 (C_n/1.5\ B_n) \qquad (1)$$

Where, C_n is the measured concentration of the element in the tested sediment (soil) and B_n is the geochemical background value of the element in fossil argillaceous sediment. The authors in this study used the world background values reported by Taylor and Mclennan (1985) due to unavailability of local background ones. The constant 1.5 is introduced to minimize the effect of possible variation in the background values which may be attributed to lithological variations in the sediment. Lue *et al.* (2009) gave the following interpretation for the geo-accumulation index (I_{geo}) levels in soil (Teng *et al.*, 2002 and Ji *et al.* 2008):-

I_{geo}	Class I_{geo}	Contamination Level
0	Igeo ≤ 0	Uncontaminated
1	0 < Igeo < 1	Uncontaminated/moderately contaminated
2	1 < Igeo < 2	Moderately contaminated
3	2 < Igeo < 3	Moderately/strongly contaminated
4	3 < Igeo < 4	Strongly contaminated
5	4 < Igeo < 5	Strongly/extremely contaminated
6	5 < Igeo	Extremely contaminated

Enrichment factor (EF): The enrichment factor, due to its universal formula, is relatively simple and easy tool for assessing enrichment degree and comparing the contamination of different environments (Reimann and De-Caritat, 2000). It is determined by the relation:

$$EF = [\ C_x/C_{ref}\ ]_{sample}\ /\ [\ B_x\ /B_{ref}\ ]_{Background} \qquad (2)$$

where;

C_x = content of the examined element in the examined environment.
C_{ref} = content of the examined element in the reference environment.
B_x = content of the reference element in the examined environment and
B_{ref} = content of the reference element in the reference environment.

An element is regarded as a reference element if it is of low occurrence variability. It is also possible to apply an element of geochemical nature whose substantial amounts occur in the environment but has no characteristic effects i.e. synergism or antagonism towards an examined element. Five contamination categories are recognized on the basis of the enrichment factor:

EF category	EF value	Contamination Level
1	EF < 2	Deficiency to minimal enrichment
2	EF = 2 - 5	Moderate enrichment
3	EF = 5 - 20	Significant enrichment
4	EF = 20 - 40	Very high enrichment
5	EF > 40	Extremely high enrichment

Pollution index (PI):

The pollution index (PI) parameter is expressed as:

$$PI = C_{metal}\ /\ C_{background} \qquad (3)$$

Where, PI is the pollution index, C metal is the concentration of pollutant in soil, C background is the background value for the metal. The pollution index (PI) was classified into four groups (Mmolawa *et al.*, 2011 and Al Omran *et al.* 2011), as follow:

PI ≤ 1 refers to low contamination;

$1 \leq$ PI < 3 means moderate contamination;

$3 \geq$ PI < 6 indicates considerable contamination and

PI > 6 indicates very high contamination.

Quality control and data analyses:

Before analysis, the devices were rinsed with acidified water (10% HNO3) and weighted to dissolve metals. Also, all the equipment's and containers were soaked in 10% NHO3 for 24 h then washed and cleared using de-ionized water before use. Moreover, quality control was assured by performing duplicate analysis on all samples and by using reagent blanks and standards. Also values of the studying metals below the detection limits of the Atomic Absorption Spectrophotometer (AAS) Model AA-6300 Shimadzu Corporation, Japan, were refused according to Mapanda et al. (2005). Finally, descriptive statistics (maximum, minimum, average and LSD, etc....) were calculated. Statistical analysis was performed using analysis of variance technique (ANOVA) by means of the computer program and statistical analysis systems (SAS, 2001).

RESULTS AND DISCUSSION:

The estimated physical and chemical properties of the collected soil samples are statistically summarized in Table (1). The texture class of soil generally, ranged from sandy loam, to loamy sand. In the surface soil samples (0 – 30 cm depth) irrigated with the different irrigation water qualities i.e. (GW), (GW+DW), (GW+TTWW) and (GW+DW+TTWW); the average percentages of sand were 81.18, 80.15, 78.35 and 78.56, respectively. The respective average percentages of silt were 7.74, 8.62, 9.10 and 9.52. The corresponding clay percentages reached 11.08, 11.23, 12.55 and 11.91, respectively. The EC values were 2.81, 5.04, 3.15 and 4.21 $dS.m^{-1}$ for the study soil irrigated with (GW), (GW+DW), (GW+TTWW) and (GW+DW+TTWW) respectively. The corresponding pH values were 7.63, 7.67, 7.61 and 7.70, respectively; while the organic matter contents ($g\ kg^{-1}$) were 6.5, 6.7, 8.8 and 6.4, respectively. The Ca^{++} and Na^{+} ions were the most dominant cations, meanwhile the Cl^{-} and $SO4^{--}$ ions were the most dominant anions. Also, the exchangeable sodium percentage values reached 11.80, 10.27, 6.21 and 5.70 in soils irrigated with (GW+DW), (GW+ DW + TTWW), (GW+TTWW) and (GW), respectively. These results are in harmony with those obtained by Abdel- Nasser et al. (2000) who reported that increasing salinity of irrigation water led to an increase in the exchangeable sodium percentage (ESP %) on soil complex.

Table (1): The physical and chemical properties of the studied irrigated soil with different irrigation water qualities.

Parameter	Irrigation Water Quality				
	GW	GW+DW	GW+TTWW	GW+DW+TTWW	
	Average	Average	Average	Average	LSD at 5%
Sand %	81.18 a	80.15 b	78.35 d	78.56 c	0.002
Silt %	7.74 d	8.62 c	9.10 b	9.52 a	0.002
Clay %	11.08 d	11.23 c	12.55 a	11.92 b	0.002
Soil Texture	Sandy Loam	Loamy Sand	Sandy Loam	Sandy Loam	--
pH	7.63 c	7.67 b	7.61 d	7.70 a	0.002
O.M. g kg^{-1}	5.4 d	5.7 b	7.3 a	6.4 c	0.002
EC (dS/m^{-1})	2.74 b	3.16 ab	2.57 b	3.52 a	0.582
Ca^{++} (m mole.L^{-1})	9.02 c	11.14 b	8.70 d	11.76 a	0.004
Mg^{++} (m mole.L-1)	1.34 c	3.22 a	1.60 b	1.72 c	0.004
Na^{+} (m mole.L-1)	16.46 c	22.24 a	15.12 d	21.02 b	0.004
K^{+} (m mole.L-1)	0.54 d	0.74 a	0.58 c	0.70 a	0.004
HCO_3^- (m mole.L-1)	7.72 d	12.52 a	8.64 c	10.40 b	0.004
Cl^- (m mole.L-1)	10.42 d	23.18 b	13.54 c	23.46 a	0.004
SO_4^{--} (m mole.L-1)	9.24 a	1.70 c	3.74 b	1.46 d	0.004
ESP	5.70 d	11.80 a	6.21 c	10.27 b	0.089

(GW) ground water, (DW) agricultural drainage water, (TTWW) tertiary treated wastewater.
The means in each row followed by the same letter(s) did not differ at < 0.05 according to Duncan's multiple-range value of each property is the average of 6 soil samples collected over two successive seasons (2010,2011).

Data presented in Table (2) show the average chemical composition of the different water qualities used for irrigation. Apparently, the values of EC were (2.81, 5.04 , 3.15 , and 4.21 dS m^{-1} for (GW), (GW+ DW), (GW + TTWW), and (GW+DW+TTWW) water samples, respectively, whereas the corresponding values of TDS were 1798.4, 3225.6, 2016.0 and 2694.4 mg/L, respectively. The data illustrate that the highest value of EC was recorded for (GW+DW) followed by (GW+DW+TTWW) and (GW + TTWW) while the lowest value of EC was recorded for (GW). The values of pH were 7.63, 7.80, 7.55 and 7.77, for (GW), (GW+ DW), (GW + TTWW), and (GW+DW+TTWW) water samples, respectively. With respect to heavy metal contents of the different irrigation water qualities, data show that (GW+DW+TTWW) followed by (GW + TTWW) contained higher concentrations of Cu, Mn, Fe, Zn, B, Ni, Pb, Cd, As and Co compared to (GW+ DW) or (GW) irrigation water. The concentrations of these metals in all irrigation water qualities were within the permissible limits for irrigation purposes. In this respect, Pescod (1992) showed that the threshold values of heavy metals in irrigation water leading to crop damage are 2.0mg L^{-1}

for Cu, 0.2mg L^{-1} for Mn, 5.0mg$L^{-1}$for Fe, 2.0mg L^{-1} for Zn,0.2 mg L^{-1} for Ni, 5.0 mg$L^{-1}$for Pb and 0.01 mgL^{-1} for Cd.

Heavy metal total contents (mgkg^{-1}) in the cultivated soils:

Total amount of heavy metals in cultivated soil under study i.e. iron [Fe], manganese [Mn], zinc [Zn], copper [Cu], lead [Pb], cadmium [Cd], arsenic [As], cobalt [Co], and nickel [Ni] and their corresponding background values are listed in Table 3.

The results showed that (GW+ DW+TTWW), (GW+TTWW) and (GW+DW) increased total heavy metal contents of Fe, Mn, Cu , Zn, Co, Cd, Pb, As and Ni in the cultivated soils irrigated with these water qualities as compared to the cultivated soil irrigated with ground water. Results also showed that, the total soil contents of these metals could be arranged in the following descending order: Fe > Cu > Mn > Zn > Pb > Co > As > Cd> Ni. The concentration of iron [Fe] ranged from 1820.0 to 2525.67, 2089.0 to3711.50, 2367.17 to 4701.33 and from 2724.67 to 5038.67 mg/kg at the depth of (0-30) cm for soils irrigated with (GW), (GW+ DW), (GW+TTWW) and (GW+ DW +TTWW), respectively. The concentration range of Cu was 26.61 to 57.33 with an average of 41.97 mg/kg for soil irrigated with (GW), the mean concentration of Cu is higher than the average value of common range in agricultural soil. The concentration of Mn ranged from 31.93 to 46.74 mg/kg with an average of 39.34 mg/kg for soil irrigated with (GW) while the corresponding range for soil irrigated with (GW+ DW +TTWW) was 77.31 to 106.18 with an average 91.46 mg/kg soil. The mean concentrations of Zn were 27.77, 37.26, 50.48 and 58.47 for soils irrigated with (GW), (GW+ DW), (GW+TTWW) and (GW+ DW +TTWW), respectively. The mean concentrations of Cd were 0.06, 0.12, 0.16 and 0.21 mg/kg for soils irrigated with (GW), (GW+ DW), (GW+TTWW) and (GW+ DW +TTWW), respectively.

Generally, the data showed that the effects of different irrigation water qualities on total heavy metals content in soil are in the following order: (GW+ DW+TTWW) > (GW+TTWW) > (GW+DW) > (GW). These results are also in agreement with those obtained by Hussein (1991) who found that agricultural drainage water significantly increased Fe, Mn, Cu and Zn in sandy clay loam soil, sandy soil and calcareous soil. These results are in harmony with those obtained by Shahin and Hussein (2005) who reported that the (GW+ DW +TTWW) resulted in the highest effect on Cd content of soil followed by (GW+TTWW), (GW+ DW) and then (GW).

Table (2): The chemical composition contents of the different irrigation water qualities used for irrigation of Al-Hassa soil.

Characteristic	Irrigation Water Quality				LSD at 5%
	GW	GW+DW	GW+TTWW	GW+DW+TTWW	
EC (dS/m)	2.81 d	5.04 a	3.15 c	4.21 b	0.002
TDS (mg/L)	1798.4 d	3225.6 a	2016.0 c	2694.4 b	3.700
pH	7.63 c	7.80 a	7.55 d	7.77 b	0.002
Soluble Cations, m molc L^{-1}					
Ca^{2+}	7.94 d	13.26 a	9.40 c	10.44 b	0.004
Mg^{2+}	4.36 d	7.58 a	4.90 c	6.90 b	0.004
Na^{+}	14.9 d	28.42 a	16.26 c	23.92 b	0.004
K^{+}	0.90 c	1.14 a	0.94 b	0.84 d	0.004
Soluble Anions, m molc L^{-1}					
CO_3^{2-}	-	-	-	-	-
HCO_3^-	4.46 c	8.84 a	3.62 d	5.70 b	0.004
Cl^-	10.00 d	17.34 c	20.32 b	22.34 a	0.120
SO_4^{2-}	13.64 c	24.22 a	7.56 d	14.06 b	0.004
NO_3^-, , mg L^{-1}	5.23 d	10.21 c	11.34 b	13.53 a	0.240
Micronutrients, mg L^{-1}					
Cu	0.012 b	0.016 ab	0.019 c	0.026 a	0.060
Mn	0.017 d	0.022 b	0.027 c	0.032a	0.002
Fe	0.072 d	0.085 c	0.095 b	0.099 a	0.002
Zn	0.045 d	0.076 c	0.085 b	0.090 a	0.110
B	0. 35 b	0. 48 a	0. 26 b	0. 57 a	0.110
Heavy metals, ug L^{-1}					
Ni	0.005 d	0.008 b	0.013c	0.015 a	0.002
Pb	0.009 d	0.019 b	0.014 c	0.017 a	0.002
Cd	0.002 a	0.006 c	0.015 d	0.019 b	0.002
As	0.003 b	0.008 d	0.009a	0.011 c	0.002
Co	0.004 a	0.009 c	0.012 d	0.016 b	0.002

(GW) ground water, (DW) agricultural drainage water, (TTWW) tertiary treated wastewater.
The value of each property is the average of 24 water samples for each irrigation water quality during two successive seasons (2010,2011).

The Geo-Accumulation Index (I_{geo}) for the studied soils:

The I_{geo} values for the nine heavy elements in the cultivated soils irrigated with different irrigation water qualities are listed in Table (4). Applying the classification system devised by Lue et al., (2009). Ji et al., (2008) and Tenget al. (2002); the elements identified in the irrigated soils may be divided into three categories. The I_{geo} values, of the soils irrigated with groundwater , for Mn, Fe, Co, Cd and As fell into (class 0). This indicates that the cultivated soil irrigated with groundwater is uncontaminated with these elements. On the other hand I_{geo} values for Cu, Zn, Pb and Ni are > 0 and < 1 (Table 4). This indicates that the soil irrigated with groundwater is uncontaminated to moderately contaminated with these elements may be du to the moving engine parts, fungicide, insecticides and phosphate fertilizers anthropogenic activities, (Sutherland *et al.*, 2000 and Ji *et al.*, 2008).

The I_{geo} values for the soil irrigated with groundwater mixed with agricultural drainage water, indicate a contamination with the same elements that contaminated the soil irrigated with groundwater but with different levels. I_{geo} values are > 0 < 1, for Cu, Zn, Pb, and Ni. This indicates that the soil irrigated with (GW+ DW) are classified according to the level of contamination (classes) into the category of uncontaminated to moderately contaminated and this probably due to anthropogenic activities.

The I_{geo} values for the soil irrigated with (GW+TTWW), showed that the I_{geo} for Mn, Fe, Co, and Cd are classified as class 0. This indicates that the soil irrigated with (GW+ TTWW) is uncontaminated with these elements. On the other hand the I_{geo} values for Zn, As, and Ni are > 0 < 1, This indicates that the soil irrigated with (GW+TTWW) was uncontaminated to moderately contaminated with these elements (class 1). The I_{geo} values for Cu and Pb are more than1 and less than 2, (> 1 and < 2) This indicates that the soil irrigated with groundwater mixed with tertiary treated wastewater are classified according to the level of contamination by these elements as moderately contaminated (class2), probably due to anthropogenic activities.

The I_{geo} values (Table 4), of the soil irrigated with (GW+ DW + TTWW) for Mn, Fe, and Co, are negative (class 0); i.e. uncontaminated soils, while the I_{geo} values for Cd and As are more than 0 and less than 1 indicating that these soils can be classified as uncontaminated to moderately contaminated with Cd and As elements. On the other hand the I_{geo} values for the elements Cu, Zn, Pb, and Ni are 1.67, 1.10, 1.49, and 1.32, respectively. This indicates that the soils irrigated with (GW+DW+TTWW) are classified as moderately contaminated with Cu, Zn, Pb, and Ni (class2). The most likely source of these elements may be the agricultural materials added to the soil through irrigation water polluted with (DW) and/or (TTWW) water. (Lue *et al.*, 2009).

Table (3): **Total content of heavy metals (mgkg^{-1} soil) in the soils (farms) irrigated with different irrigation water qualities (at 0-30 cm depth) over two seasons compared to common ranges in soil of Al- Hassa oasis.**

Metal (mgkg^{-1} soil)	GW			GW + DW			GW+TTWW			GW+DW+TTWW			Common range in soil* (mg/kg^{-1}soil)		
	Max.	Min	Ave.	Max.	Min	Ave.	Max.	Min	Ave.	Max.	Min	Ave.	Max.	Min	Ave.
Cu	57.33	26.61	41.97	81.97	32.39	57.19	98.80	49.86	74.30	113.56	74.97	93.74	100.0	2.00	30.0
Mn	46.74	31.93	39.34	63.09	55.13	59.11	81.50	58.05	74.56	106.18	77.31	91.46	4180	182	1476
Fe	2525.67	1820.00	2172.50	3711.50	2089.00	2900.50	4701.33	2367.17	3534.33	5038.67	2724.67	3881.50	55000.0	7000.0	38000.0
Zn	40.16	15.45	27.77	54.04	20.46	37.26	68.39	32.58	50.48	79.27	48.31	58.47	300.00	10.00	50.00
Co	4.14	2.46	3.30	6.28	3.91	5.09	7.92	5.27	6.35	9.85	6.40	8.13	40.00	1.00	8.00
Cd	0.08	0.04	0.06	0.16	0.08	0.12	0.21	0.10	0.16	0.26	0.14	0.21	0.7	0.01	0.06
Pb	4.14	2.77	3.62	6.76	4.81	5.79	9.84	6.11	7.97	11.43	7.50	9.47	200.00	2.00	10.00
As	2.08	1.53	1.81	3.88	2.14	3.02	4.24	2.53	3.39	5.22	3.08	3.88	50.00	1.00	5.00
Ni	10.14	4.32	7.23	12.73	5.56	9.14	15.65	8.25	11.95	19.80	11.44	15.62	500.00	5.00	40.00

Gw= (ground water); GW+DW= (ground water + agricultural drainage water); GW+TTWW= (ground water + tertiary treated wastewater); GW+DW+TTWW= (ground water + agricultural drainage water + tertiary treated wastewater).

*Common range of element concentrations in soils reported by Lindsay (1979), Kabata and Pendias (1992), Marschner (1995), Adriano (2001), and Al-Omran *et al.*(2011). Cobalt rangeis after Bowen (1996) {c.f. Cataldo *et.al.* (1999)}.

Table (4): Average values of Geo-accumulation indexes (I_{geo}), Enrichment Factor (EF) and Pollution index (PI) for soils (layer at 0 - 30 cm depth) irrigated with different irrigation water qualities in Al- Hassa Oasis.

Metal	Average value of Geo-accumulation Index (I_{geo})					Average value of Enrichment Factor (EF)					Average value of Pollution index (PI)				
	Back Ground* (mg.kg^{-1} soil)	Irrigation water qualities				Back Ground* (mg.kg^{-1} soil)	Irrigation water qualities				Back Ground* (mg.kg^{-1} soil)	Irrigation water qualities			
		GW	GW+DW	GW+TTWW	GW+DW+TTWW		GW	GW+DW	GW+TTWW	GW+DW+TTWW		GW	GW+DW	GW+TTWW	GW+DW+TTWW
Cu	19.66	0.51	0.96	1.33	1.67	19.66	5.42	7.38	9.59	12.10	19.66	2.92	2.91	3.78	4.77
Mn	688	-4.71	-4.13	-3.79	-3.50	688	1.29	1.63	1.63	2.00	688	0.07	0.09	0.11	0.13
Fe	43193	-4.90	-4.48	-4.20	-4.06	43193	0.76	1.01	1.23	1.35	43193	0.06	0.07	0.08	0.09
Zn	18.23	0.02	0.45	0.88	1.10	18.23	3.87	5.19	7.03	7.03	18.23	2.20	2.04	2.77	3.21
Co	15.90	-2.85	-2.23	-1.91	-1.55	15.90	0.62	0.95	1.19	1.52	15.90	0.26	0.32	0.40	0.51
Cd	0.12	-1.58	-0.58	-0.17	0.22	0.12	1.17	2.54	3.38	4.44	0.12	0.67	1.00	1.33	1.75
Pb	2.24	0.11	0.79	1.25	1.49	2.24	4.10	6.56	9.03	10.73	2.24	1.85	2.58	3.56	4.23
As	2.12	-0.81	-0.07	0.09	0.29	2.12	2.17	3.62	4.06	4.65	2.12	0.98	1.42	1.60	1.83
Ni	4.18	0.21	0.54	0.93	1.32	4.18	4.39	5.55	7.26	9.49	4.18	2.43	2.19	2.86	3.74

Gw= (ground water); GW+DW= (ground water + agricultural drainage water); GW+TTWW= (ground water + tertiary treated wastewater); GW+DW+TTWW= (ground water + agricultural drainage water + tertiary treated wastewater).

(*) The background values were obtained according Al-Omran *et al.*(2011).

In general, I_{geo} values for the soil irrigated with (GW+DW+TTWW) showed patterns of heavy metals contamination similar to those of the soils irrigated with (GW+TTWW).

On average, levels of Mn and Fe found in this study were below concentrations which are deemed pollutants, therefore, Mn and Fe may be chosen as reference elements for research on agricultural cultivated soils. It should be also noticed that according to the I_{geo} mean values of metals in the soil irrigated with different irrigation water qualities could be arranged in the following descending order: (GW +DW+TTWW) > (GW +TTWW) > (GW +DW) > (GW).

The Enrichment Factor (EF) for studied soils:

For a better estimation of anthropogenic inputs, EF was calculated for each metal by dividing its ratio to a normalized element by the same ratio found in a baseline. The use of EF for identification of anomalous metal concentration requires geochemical normalization of the heavy metal data to a conservative element such as Al or Fe (Ghrefat and Yusuf, 2006). Several authors have successfully used Fe or suggested the use of Fe to normalize metal contamination (Bhuiyan *et al.* 2011). The current study had also employed Fe as a conservative tracer to differentiate natural from anthropogenic source of metal contamination in the cultivated soils irrigated with different irrigation water qualities. In order to estimate quantitatively the anthropogenic trace metals in the cultivated soils; their background concentrations must be known. Previous researchers often used lactogenic background value as an average concentration in shale (Ghrefat and Yusuf, 2006; Bhuiyan *et al.*, 2011) or an average value of measured concentration before industrialization (Hakanson, 1980) to assess trace metal concentration in sediment. In this study the background value was taken from average of cultivated soils (Turekian and Wedephol, 2011; Al- Omran *et al.*,2011).

The average levels of the sampling representing the cultivated soils irrigated with different irrigation water qualities for EF are displayed in Table (4). The EF values for soil irrigated with groundwater reveal that EF values for studied metals could be arranged in the following descending order: Cu > Ni > Pb > Zn > As > Mn > Cd > Fe > Co. The highest average value for Cu, 5.42 indicating significant enrichment, (EF = 5-20) while the EF values for Ni, Pb, Zn, and As are 4.39, 4.10, 3.87, and 2.17, respectively moderate significant enrichment (EF = 2-5) while the EF values for Mn, Fe, Co and Cd are 1.29, 0.76, 0.62 and 1.17, respectively indicating moderate enrichment. The EF values for Cd, Mn, Fe and Co, are 1.69, 1.38, 0.88 and 0.77 respectively indicating deficiency to minimal enrichment (EF < 2).

The calculated results of EF values for heavy metals in the soils irrigated with (GW +DW) are shown in Table (4). The results show that Cu, Pb, Ni, and Zn have significant enrichment (EF value =5-20) with highest values reaching: 7.38, 6.56, 5.55 and 5.19, respectively indicating severe enrichment; while the As and Cd have EF values of 3.62 and 2.54, respectively therefore this soil is moderately enriched with As and Cd (EF value =2-5). Meanwhile, the EF values for Mn, Fe and Co reaching: 0.95, 1.01and 1.63, respectively therefore this soil is deficiency to minimal enriched with Mn, Fe and Co (EF value < 2). This reveals that the cultivated soils irrigated with (GW + DW) are depleted in these minerals (deficient category).

In general, EF values for the soils irrigated with (GW +TTWW) or for the soils irrigated with (GW +DW+TTWW) are similar to those of the cultivated soils irrigated with (GW +DW), where the EF values for Cu, Pb, Zn, and Ni are 9.59, 9.03, 7.03, and 7.26, respectively for the cultivated soils irrigated with (GW + TTWW).

The EF values for these metals in the soils irrigated with (GW+DW+TTWW) are 12.10, 10.73, 7.03, and 9.49, respectively. The EF values for these elements which are greater than 5, (i.e. EF value = 5 to 20) indicate significant enrichment. This suggests that the sources of contamination with these elements are anthropogenic due to previous agricultural activities such as fungicides, algaecides, pesticides, wood preservatives, antifouling paint and nutritional supplements in animal feed (Edwards,1976). Heavy metal accumulations in plant and soil from natural and artificial sources represent important environmental pollution problems. Food safety issues and potential adverse health risks make this one of the most serious environmental concerns (Cui *et al.*, 2004). Fe and Co are the two deficient to minimal enrichment metals and therefore contamination may be traced to a natural source. The differences in the EF values may be due to the difference in the magnitude of input for each metal in the soil and/or differences in the removal rate of each metal from the soil (Akoto, *et al.*, 2008).

It should be also noticed that the *EF* mean values of metals in the studied cultivated soils irrigated with different irrigation water qualities, when compared to the EF severe enrichment level adopted in many studies (Ghrefat and Yusuf, 2006; Abrahim and Parker, 2008; Akoto *et* *al*.,2008; Olubunmi et al., 2010) which is (5 to 20), can be arranged in following descending order: (GW +DW+TTWW) with 66% of metals falling within the EF severe enrichment level ; > (GW+TTWW) with 55% of metals falling within EF severe enrichment level; > (GW +DW) with 44% of metals falling within EF severe enrichment level > (GW) with 33% of metals falling within EF severe enrichment level.

Pollution index (PI) for the studied soils:

Based on the results of the calculated pollution index shown in Table (4), it is observed that the lowest PI value was shown for the soil irrigated with (GW), while the highest PI values are shown for the soil irrigated with (GW +DW+TTWW) . Based on PI values for the studied soils, PI value for the different heavy metals fall into three categories. The first category with PI value ≤ 1 indicating low contamination or unpolluted cultivated soils with the metals: Mn, Fe, Co, Cd, and As in the cultivated soil irrigated with (GW), Mn, Fe, and Co in the soils irrigated with (GW+DW), (GW+TTWW) and (GW+DW+TTWW).

The second category, with PI value from1 to 3 indicating moderate contamination by the heavy metals: Cu, Zn, Pb, and Ni in cultivated soil irrigated with (GW), Zn, Cd, and As in cultivated soil irrigated with (GW +DW), Cd, and As in both the soils irrigated with (GW+ TTWW) and (GW+DW+TTWW). More detailed study and monitoring are required to monitor the source of pollution.

The third category, (3 ≥ PI < 6) with PI value = 3 to 6 indicating considerable soil heavy metal contamination which require intervention to ameliorate the pollution. The soils falling in this category also require regular monitoring and the investigation of the major source of pollution. The current results indicate that the third category is not included in both the soils irrigated with ground water, and the soil irrigated with (GW +DW), while the highest PI values are shown for the soil irrigated with (GW+TTWW) for Cu and Pb, the PI values falling within the third category, reaching: 3.78 and 3.56 respectively, also the soils irrigated with (GW +DW+TTWW) show Cu, Zn, Pb and Ni severe pollution with PI values falling within the third category, reaching: 4.47, 3.21,4.23 and 3.74, respectively.

CONCLUSION:

The present study represents a useful tool for the evaluation heavy metal hazards of cultivated soil, in relation to different irrigation water qualities and how it may affect the soil heavy metal contents. The diagnostic parameters, including geo-accumulation index, enrichment factor, pollution index and correlation analysis, provide important tools for better understanding of the pollutants among the cultivated soil sampling in relation to the environmental matrices employed for the study. The relatively different concentrations of the studied heavy metals clearly indicate that the main source of pollution may come from the agricultural activities. The use of geo-accumulation index, enrichment factor, and pollution index has provided essential information for the assessment of pollution level in the cultivated soils. Enrichment Factor (EF) has shown a significant enrichment with elements such as Cu, Zn, Pb, and Ni. The possible source of pollution was expected to be originated from land base agricultural activities and the different irrigation water qualities used for soils irrigation.

Due to the unavailability of studies defining the regional background values of the heavy metal contamination of soils as a result of agricultural activities in Al- Hassa area, this study used values from other areas with similar conditions. However, taking into consideration the plans for the expansion in the use of treated wastewater for irrigation in the future in Al Hassa area, determining these background values becomes very important. It is highly recommended that the relevant government agencies and research centers should be conduct studies in that direction, with the aim of protecting the soils from heavy metal pollution.

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الملخص العربي

تقييم التلوث بالعناصر الثقيلة فى أراضى مروية بنوعيات مختلفة من مياه الري بواحة الأحساء بالمملكة العربية السعودية.

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تم جمع ٢٤ عينة تربة سطحية مركبة من عمق (صفر -٣٠ سم) تمثل الأراضى المروية بنوعيات مختلفة من مياه الري لفترة تزيد عن ١٥ سنة وهي (المياه الجوفية)،(المياه الجوفية المخلوطة بمياه الصرف الزراعي)، (المياه الجوفية المخلوطة بمياه الصرف الصرف الصحي المعالجة ثلاثياً)، (المياه الجوفية المخلوطة بمياه الصرف الزراعي ومياه الصرف الصحي المعالجة ثلاثياً)، وقد تم تقييم المحتوى الكلي للمعادن الثقيلة بكل تربة تروي من كل مصدر من مصادر مياه الري المختلفة لتقييم معدل تلوث كل تربة بالمعادن الثقيلة تحت الدراسة: (النحاس، المنجنيز، الحديد، الزنك، الكوبالت، الكادميوم، الرصاص، الخارصين، النيكل).

أظهرت النتائج أن المحتوى الكلى من هذه العناصر الثقيلة في الأراضى المروية بالنوعيات المختلفة من مياه الري يمكن ترتيبها في ترتيب تنازلي كالآتي : الحديد> النحاس> المنجنيز> الزنك> النيكل> الرصاص> الكوبالت> الخارصين> الكادميوم. وكان تأثير نوعيات مياه الري على المحتوي الكلى للعناصر الثقيلة بالأراضى المروية بكل مصدر من مصادر مياه الري المختلفة بالترتيب التالي: (المياه الجوفية المخلوطة بمياه الصرف الزراعي ومياه الصرف الصحي المعالجة ثلاثياً) أكبر من (المياه الجوفية المخلوطة بمياه الصرف الصحي المعالجة ثلاثياً) أكبر من (المياه الجوفية المخلوطة بمياه الصرف الزراعي) أكبر من (المياه الجوفية) على أساس مقاييس التراكم والتي استخدمت في هذه الدراسة لتحديد معدل

Geo–accumulation Index (I_{geo}), Enrichment Factor (EF), Pollution Index (PI). التراكم من كل عنصر بالتربة التى تروي بنوعية من نوعيات مياه الري المختلفة.

أظهرت النتائج إلى أن قيم معدل التراكم لعناصر المنجنيز، الحديد، الكوبالت، الكادميوم جاءت (الدرجة صفر) في التربة المروية بالمياه الجوفية. وهذايشير إلى أن التربة التى تروي بالمياه الجوفية لفترات طويلة غير ملوثة بهذه العناصر. بينما قيم عناصر النحاس، والزنك، والرصاص، والنيكل كانت أكبر من الصفر وأقل من الواحد وهذايعني أن التربة تقع ضمن المجموعة غير الملوثة إلى معتدلة التلوث بهذه العناصر. في حين بلغت قيمة معدل التراكم لعنصر الخارصين في التربة المروية بالمياه الجوفية في الفئة أكبر من ٢ وأقل من ٣ من معتدلة التلوث إلى شديدة التلوث بعنصر الخارصين في التربة المروية بالمياه الجوفية.

بشكل عام أظهرت قيم معدل التراكم طبقاً للمقاييس المختلفة المستخدمة في تقييم معدل التلوث بكل تربة تروي بنوعية محددة من نوعيات مياه الري لفترات طويلة أن الأراضي المورية (بالمياه الجوفية المخلوطة بمياه الصرف الزراعي ومياه الصرف الصحي المعالجة ثلاثياً) ذات معدل تراكم وتلوث بالعناصر الثقيلة تحت الدراسة مقارنة بالأراضي المروية (بالمياه الجوفية المخلوطة بمياه الصرف الصحي المعالجة ثلاثياً) أو الأراضي المروية (بالمياه الجوفية المخلوطة بمياه الصرف الزراعي) وتأتي الأراضي المروية بالمياه الجوفية فقط أقلها في معدلات التراكم والتلوث بالعناصر الثقيلة طبقاً لمقاييس التراكم والتلوث المستخدمة في هذه الدراسة.