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HEAVY METAL CONCENTRATIONS IN SOILS AND DATE PALMS IRRIGATED BY GROUNDWATER AND TREATED WASTEWATER

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Agricultural management of wastewater is a sustainable approach for arid- and semi-arid regions of the world. This research was conducted to evaluate the suitability of treated waste water for irrigating date palms and monitoring the partitioning of some heavy metals (e.g., Cu, Cr, Cd, Pb, Mn, Fe, Zn etc.) among soil, plant and fruits. Soil and plant samples including leaves and fruits were collected from different locations of the date palm plantations at the Sultan Qaboos University, Al-Hamra and izki which have been irrigated with treated water or ground water for seven years. Results showed that the concentrations of heavy metals in both groundwater and treated waste water were within the international standards. There were significant variations in heavy metal concentrations in soil at studied locations. In most of the cases, the concentrations of heavy metals were relatively higher in soils irrigated with treated waste water compared to the soils irrigated with groundwater. Generally, the concentrations of heavy metals in date palm leaves were not significantly different in plants irrigated with treated waste water or groundwater. However, there were significant differences in the concentrations of heavy metals in date fruits irrigated with different sources of water. The concentrations of some metals (Fe, Zn, and Ni) in date fruits were higher in waste water irrigated plants whereas other metals (Cu, Cd, Pb, and B) were higher in ground water treated plants. In all cases the concentration were within the permissible limits. Thus, the long-term effects of treated waste water did not indicate any adverse effects of irrigation using groundwater and waste water on fruit mineral composition, including heavy metals.
Keywords: Treated waste water, ground water, heavy metals, date palms, irrigation water management.

INTRODUCTION

Water is one of the important functional components of the terrestrial ecosystems. In water-deficit regions, like the Sultanate of Oman, the use of alternate source of water for irrigating crops is of paramount importance. The use of treated waste water in irrigated agriculture in arid- and semi-arid regions of the world is expected to free-up and prevent the contamination of good quality water resources for the use in urban and industrial sectors (Abdelrahman *et al.*, 2011). The use of treated waste water in agriculture could help to conserve freshwater and fertilizer application. In many parts of the world, treated waste water has long been used for irrigation (Levine and Sanot, 2004). However, waste waters reportedly contain salts, heavy metals, reactive nutrients, pathogens, and pollutants with unknown effects on the ecosystems (Mohammad and Mazahareh, 2003). Thus, indiscriminate use of treated waste water for irrigating crops could result in accumulation of total soluble salts including heavy metals in soil. This premise is supported by many reports in literature that heavy metals (including Fe, Zn, Cu, Mn, Pb and Co) contents in soil increased by treated waste water irrigation (Selem *et al.*, 2000). In some studies, an accumulation of heavy metals in the edible part of plants has also been reported (Abd-Elfattah *et al.*, 2002).

The Sultanate of Oman is considered to be one of the semi-arid countries of the world. Its agriculture is fully dependent on irrigation due to an average annual mean precipitation of 100 mm. While the surface water is very scarce, hence groundwater serves as the main water source for irrigation and domestic use. Consequently, an indiscriminate withdrawal of groundwater has led to a deficit level from lack of adequate recharge of freshwater from rainfall (MAF, 2007). Moreover, the rapid urbanization with high population growth and increasing need for agricultural production has led to a high demand of using treated waste water as alternative source of freshwater for irrigating crops. However, long-term use of treated waste water for irrigating field crops is expected to cause heavy metals accumulation in soil, and consequently in plant leaves and fruits. In Oman, very little information is available concerning the effect of treated waste water on the heavy metals concentrations and translocation among soil, plant and fruits. Therefore, this study was conducted to evaluate the long-term effect of treated waste water in irrigating date palms and monitoring their movements among soil, plant and fruits and edibility of date palm fruits.

MATRERIALS AND METHODS

For the treatment of large quantities of municipal waste water, the government of Oman have installed sewage treatment plants (STPs) where the waste water is subjected to tertiary treatment for removal of easily settle able solids, non-settle able and dissolved colloidal solids, organic and inorganic materials, nutrients and pathogens. Thus, this tertiary treatment of waste water is believed to produce effluents that have minimum concentrations of contaminants and is used for irrigation of different trees and crops. In order to study the long term effects of the treated waste water on accumulation of heavy metals in the soil and plants growing therein, the places were selected which have been irrigated for seven years with this tertiary treated waste water.

The study area included three different agricultural locations (location 1: Al-Hamra, 22° 56' 0" N / 57° 32' 0" E; location 2: Izki, 22° 56' 2" N / 57° 46' 30" E and location 3: Sultan Qaboos University, 23° 36' 47" N / 58° 35' 35" E) where sewage effluents originating from Government sewage treatment plants have been used to irrigate landscape and date palms trees for seven years. From the adjacent areas to the soils irrigated with treated waste water, samples were collected where ground water was used for irrigation purposes only. Ten replicates of composite soil samples were collected randomly from each location that have been irrigated either by groundwater (G) or tertiary treated wastewater (T). The distance between location 1 and 2 is around 70 Km, 2 and 3 is around 140Km and 1 and 3 is around 200 Km. Soil samples were air-dried, thoroughly mixed, ground and sieved through 2 mm sieve prior to chemical analysis. Plant leaves and fruits were oven-dried at 70^{o}C until a constant weight was obtained. All the samples were digested with concentrated hydrochloric acid to determine selected heavy metals using inductively coupled plasma (ICP) spectroscopy. Data were analyzed by 1-way analysis of variance and the means were compared by the Duncan's multiple range test at $p \leq 0.05$ using (SPSS, 1998).

RESULTS AND DISCUSSIONS

Concentrations of heavy metals in irrigation water from different sources: Information regarding the heavy metal concentrations in water used for irrigation in the studied locations and the standards for water reuse by Environmental Protection Agency (EPA), Food and Agriculture Organization (FAO) and Ministry of Agriculture and Fisheries (MAF), Oman, is presented in Table 1. It is evident from the data that heavy metal concentrations in all the sources of irrigation water were within the permissible limits set by EPA, FAO and MAF. Nonetheless, the concentration of some elements (for instance, Cu, Cd, Pb, Ni and B) were relatively higher in some treated waste water as compared with groundwater. Hence, it could be expected that long term usage of some treated waste waters may result in accumulation of some harmful elements in soil and plant. Similar results have been reported by Rattan *et al.* (2002) that the sewage effluents were found to contain higher content of Pb, Zn, Cu, Co, Cr, As, Cd, Fe, Mn and Ni compared to well water. Also Yadar *et al.* (2002) reported that the wastewaters were carrying substantial amounts of toxic metals and their concentrations varied from one city to another. Nevertheless, the higher concentrations of toxic metals not necessarily means toxicity to the plants. As metals availability and uptake by plants in agricultural environments is related to i) the activity of metal ions in the solution, which depends upon pH, pE, concentration and composition, ii) the concentration and ratios of accompanying elements that influence the uptake and transport of that metal ion by roots, and iii) numerous environmental factors (Pessarakli, 1999).

Effect of different sources of irrigation water on soil salinity and pH: The effect of different sources of irrigation water on soil pH and salinity is shown in Fig. 1. It is obvious from the Fig. 1, that there was no consistent trend in response of soil pH and salinity to irrigation with treated

Table 1. Concentrations of heavy metals (mg L^{-1}) in different sources of irrigation water and national and international standards for safe use of treated waste water

Water	Mn	Fe	Zn	Cu	Cr	Cd	Pb	Ni	B
Tap water	0.002	0.012	0.061	0.010	0.001	0.215	0.015	0.001	0.118
Groundwater	0.002	0.013	0.064	0.008	0.001	0.142	0.001	0.001	0.295
Treated wastewater (1)	0.002	0.016	0.064	0.024	0.001	0.431	0.066	0.001	0.508
Treated wastewater (2)	0.002	0.014	0.016	0.009	0.002	0.001	0.005	0.039	0.450
Treated wastewater (3)	0.002	0.017	0.036	0.018	0.003	0.001	0.009	0.031	0.600
EPA Standard*	0.200	5.000	5.000	0.500	0.100	0.010	0.100	0.100	0.750
FAO Standard	0.200	5.000	2.000	0.200	0.100	0.010	0.500	0.200	0.750
Omani Standard	0.500	5.000	5.000	1.000	0.050	0.010	0.200	0.100	0.750

*Summary of U.S. EPA guidelines for water reuse for irrigation (Adapted from U.S., 2004)

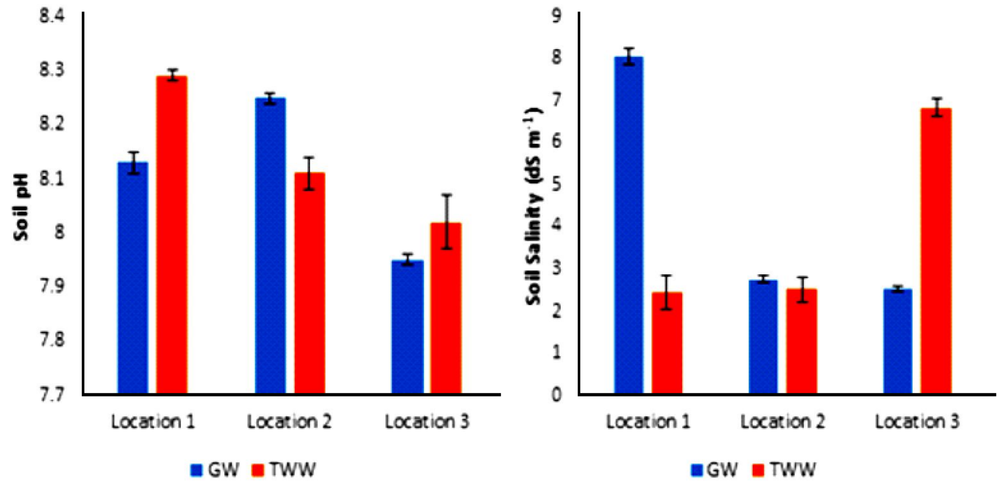

Figure 1. Effect of irrigation with treated wastewater and groundwater on soil pH and salinity.

waste water or ground water. The soil at all locations was slightly alkaline and have pH ranging from 7.95-8.29. On location 1, soil irrigated with treated waste water had higher pH as compared with the soil irrigated with ground water, conversely on location 2, soil irrigated with groundwater had relatively higher pH as compared with the soil irrigated with treated wastewater. On location 3, soil irrigated with treated waste water had slightly higher pH than the soil irrigated with the ground water. Generally, soils in Oman are calcareous in nature and therefore, mostly alkaline in nature (Al-Busaidi *et al.,* 2005). Likewise the soil pH, the response of soil salinity to irrigation with treated wastewater or ground water was not consistent, nonetheless, the variations in soil salinity were significant as compared with the soil pH. On location 1 the soil salinity was higher in soil irrigated with the ground water but on location 3 soil salinity was higher in wastewater irrigated soil. This could be due to the fact that not only the treated wastewater contains more salts, but the ground water is also getting saline due to over pumping in many parts of Oman (MAF, 2007).

Concentrations of heavy metals in soils irrigated with different sources of water: Data regarding the concentration of heavy metals in the soil irrigated with treated waste water and ground water are summarized in Table 2. It is obvious from the data, that the application of different sources of irrigation water has significant effects on the concentrations of many heavy metals in the soil. In most of the cases the soil receiving treated waste water has higher amounts of heavy metals as compared with the soils irrigated with ground water. However, in some cases, for instance, Cu and Cr, the concentrations were significantly higher in ground water irrigated soils as compared with the treated waste water irrigated soils on location 2, 3 and location 1, respectively, Similarly, on location 1, Pb concentrations were higher in soils irrigated with ground water as compared with the soils irrigated the treated waste water. The continuous application of treated waste water which contain relatively higher amounts of some toxic metals could lead to their accumulation in the soil. This premises has been supported by the work of many researchers for instance, Rattan *et al.* (2004) reported the accumulation of Zn, Pb, Ni, Mn, Fe, Cu, Cr, Co and As in the sewage irrigated soils as compared with the well water-irrigated ones. Similarly, Al-Omran *et al.* (2012) have reported 129.9%, 55.1%, 84.3%, 30.2%, 81.7%, 39.5%, 75.0%, 78.1% and 66.7% increases in the concentrations of Zn, Pb, Ni, Mn, Fe, Cu, Cr, Co and As, respectively, over a period of 13 years, in the sewage water irrigated soils as compared with the well water irrigated soils.

Table 2. Concentrations of heavy metals (mg L^{-1}) in soils irrigated with different sources of water

Soil	Mn	Fe	Zn	Cu	Cr	Cd	Pb	Ni	B
Location 1-T	0.200 c*	0.170 d	0.050 a	0.040 b	0.010 d	0.001 c	0.141d	0.001 b	0.211 d
Location 1-G	0.300 a	0.180 c	0.001 b	0.010 c	0.030 c	0.001 c	0.202c	0.001 b	0.320 c
Location 2-T	0.229a	0.457a	0.003b	0.012d	0.113a	0.067a	1.034a	0.006b	0.967a
Location 2-G	0.001 d	0.330 b	0.001 b	0.070 a	0.050 b	0.040 b	0.613b	0.001 b	0.524 b
Location 3-T	0.230 b	0.460 a	0.001 b	0.010 d	0.110 a	0.070 a	1.032a	0.010 a	0.971 a
Location 3-G	0.001d	0.328b	0.001b	0.066a	0.051b	0.038b	0.607b	0.001b	0.525b

*Means in the column with same letter indicate no difference at Duncan's Multiple Range Test at P< 0.05.

In our study, the concentrations of many toxic metals are less than reported in the previous studies, for example, Rattan *et al*. (2001, 2002, 2004) Al-Omran *et al*. 2012. This could be due to the reasons that Haya Water Company is subjecting the sewage wastewater to tertiary treatment which may result in reduction of the toxic metal loads which is evident from the concentrations of heavy metals reported in Table 1 and the coarse texture of soil which has less capacity to hold metals.

Heavy metal concentrations in date palm leaves: The effect of irrigation with treated waste water and groundwater on metals concentrations in date palm leaves is presented in Table 3. It is obvious from the data that in most of the cases there were no significant differences in the concentrations of heavy metals in date palm leaves e.g., Mn., Cu, Cr, Cd, Pb, B. The concentrations of Ni and Zn (excluding location 2) were little higher in groundwater treated soil as compared with the treated waste water irrigated soils. On the contrary, the Fe concentrations were slightly higher in treated waste water irrigated soils as compared with the ground water irrigated soils. The slight variations in heavy metal concentrations in date palm leaves could be attributed to some other factors, for example, the alkaline pH of the soil, which decrease the availability of most of the metals, sources of metals others than irrigation water, e.g metals present in the atmosphere etc. (Al-Rawahi *et al*. 2012). Some metals like, Fe, Zn, and Ni were higher in the date palm leaves but these were below the phytotoxic concentrations as described by Awashthi, 2000. Other researchers have reported accumulation of heavy metals in the leaves, for instance, Abd-Elfattah *et al*. (2002) reported significant differences between leaves grown in soils irrigated with treated wastewater as compared with the leaves grown in soils irrigated with Nile water. Similarly, Rattan *et al*. (2002) also reported that crops raised on the water irrigated soils accumulated metals in quantities excess enough to cause clinical problems both to animals and human beings consuming these metal rich plants. Also Elgala *et al*. (2003) found that Fe and Cu concentrations in clover tissues grown in Elgabal Elasfar soil exceeded the permissible limits. The differences found in our study as compared with previously reported studies could be ascribed to the soil pH which was alkaline in our case and other properties of soil like coarse texture of soil which has less retention of cations in the soil and these physico-chemical properties might have decreased the uptake of these metals by the date palms leaves.

Heavy metal concentrations in date palm fruits: Heavy metals concentrations in date palm fruits are summarized in Table 4. Fruits are the most important part of the plants for human consumption and concentrations of all elements should be within the range of safety standards. As shown in Table 4, there were significant variations in in concentrations of heavy metals in fruits irrigated with different sources of water. In case of Fe, Zn, Ni, the concentrations in date fruits were relatively higher in the treated waste water irrigated soil as compared with the groundwater irrigated soils. On the contrary, the concentrations of Cu, Cd, Pb and B were significantly higher in ground water irrigated plants as compared with the treated waste water irrigated plants. In almost all the treatments the metal concentrations in fruits were lesser than the standards proposed by different organizations, e.g., WHO/FAO (2007), European Union (2006) and Indian standards (Awashthi,

Table 3. Metals concentrations (mg L^{-1}) in date palms leaves irrigated with different sources of water

Plant leaves	Mn	Fe	Zn	Cu	Cr	Cd	Pb	Ni	B
Location 1-T	0.001b*	6.62a	2.26e	0.001a	0.001a	0.001a	0.001a	9.53c	0.001b
Location 1-G	0.001b	4.80d	6.50d	0.001a	0.001a	0.001a	0.001a	9.93b	0.001b
Location 2-T	0.001b	6.51b	45.31a	0.001a	0.001a	0.001a	0.001a	8.01d	0.001b
Location 2-G	0.001b	5.66c	12.92b	0.001a	0.00a	0.001a	0.001a	10.19a	0.001b
Location 3-T	0.04a	4.10e	7.52c	0.001a	0.001a	0.001a	0.001a	5.26e	2.60a
Location 3-G	0.001b	5.66c	12.92b	0.001a	0.001a	0.001a	0.001a	10.19a	0.001b

*Means in the column with same letter indicate no difference at Duncan's Multiple Range Test at P< 0.05.

Table 4. Metals concentrations (mg L^{-1}) in date palms fruits irrigated with different sources of water

Plant fruits	**Mn**	**Fe**	**Zn**	**Cu**	**Cr**	**Cd**	**Pb**	**Ni**	**B**
Location 1-T	0.001 c	5.93 b	7.92 a	0.001 d	0.001 a	0.001 d	0.001 d	4.52 b	0.001 d
Location 1-G	0.03 b	3.65 c	4.29 b	0.03 c	0.001 a	0.47 c	0.04 b	3.15 c	0.14 c
Location 2-T	0.001 c	7.29 a	1.02 d	0.001 d	0.001 a	0.001 d	0.001 d	8.82 a	0.001 d
Location 2-G	0.06 a	0.25 e	0.15 f	0.05 b	0.001 a	0.62 a	0.01 c	0.001 e	0.18 b
Location 3-T	0.06 a	3.11 d	3.58 c	0.05 b	0.001 a	0.51 b	0.001 d	1.97 d	0.18 b
Location 3-G	0.06 a	0.24 e	0.20 e	0.08 a	0.001 a	0.53 b	0.88 a	0.001 e	0.23 a

*Means in the column with same letter indicate no difference at Duncan's Multiple Range Test at P< 0.05

2000) as reported in Table 5. Many reasons could be responsible for these findings. For instance, the tertiary treatment of the wastewater which might have reduce the heavy metals load in water and soil physical and chemical properties like, soil texture and soil pH etc. might have decreased the availability of these metals to plant roots and ultimately the translocation to date fruits. The concentration of more metals in the ground water irrigated date fruits could be due to the mineralogical composition of soil and ground water and other sources of metals e.g., metals in the air etc. Other researchers have reported significant increase in the concentrations of metals in fruits irrigated with sewage water etc., for example, Mahdi *et al*. (1998) indicated that fruit crops have significant differences in the mineral composition of cultivated crops irrigated with treated water. These differences were largely attributed to the tree species rather than location or age of the plant and longer exposure to treated water did not show major effects on fruit minerals, including heavy metals. El Mardi *et al*. (1995) also reported that the concentration of various minerals in fig, lime, mango, pomegranates, and mulberry were influenced by the source of irrigation. Their results indicated that there was a significant correlation between leaf and fruit concentration of heavy metals ($y = 0.0069x + 1.6518$, $R^2 = 0.63$). Overall, the fruits had the highest macronutrients and heavy metals, whereas leaves contained higher concentration of micronutrients Fe, Mn, Al and B. This may be attributed to the ability of the crops to translocate certain elements within the plants. Similarly, Abd-Elfattah *et al*. (2002) found that the heavy metals contents in fruits produced by treated wastewater were significantly higher as compared with Nile water in two seasons.

Table 5. Guideline for safe limits of heavy metals in plants (mg kg^{-1})

Standards / Elements	Cd	Cu	Pb	Zn	As	Ni	Cr
WHO/FAO (2007)	0.2	40	5	60	-	-	-
European Union (EU 2006)	0.2	-	0.3	-	0.4	-	2.3
Indian Standard (Awashthi, 2000)	1.5	30	2.5	50	-	1.5	20

Conclusions: The treated waste water had relatively higher amounts of some metals but all were within the standards provided by different agencies. Irrigation of plants with treated waste water resulted in the accumulation of some metals in the soil. But the availability and uptake of the metals by the plants was not consistent with the concentrations of the metals in the soil and was found to be dependent upon other factors like soil physical and chemical properties and environmental conditions. In view of the shortage of water resources, wastewater treatment and reuse can play a major role in alleviating the water shortage problem, and hence, wastewater reuse should be considered within the framework of the overall water master plan. Nevertheless, to avoid any health or environmental problems, reuse of treated wastewater should be subjected to continuous monitoring and fruit qualities should be evaluated. Also, more studies are required to see the influence of irrigation water over longer period of time to exactly determine the effects of the treated waste water on concentrations of heavy metals in soil, plants and fruits.

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