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Heavy Metal Contamination of Soil and Olive Trees (*Olea europaea* L.) in Suburban Areas of Tehran, Iran

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Abstract: The concentrations of Zn, Pb, Cr and Ni were measured in the soil, fruits and leaves of *Olea europaea* L. irrigated with municipal effluent. In this study, site irrigated with municipal effluent and site irrigated with well water for 7 years were sampled for soil and plant chemical analysis to evaluate its long term effect. Samples of irrigation water, soil and leaves and fruits of olive were analyzed for Zn, Pb, Cr and Ni. Heavy metals in municipal effluent were higher than the standard (WHO). Irrigation with municipal effluent increased Zn, Pb, Cr and Ni in soil compared to well water. The mean heavy metal concentrations in topsoil were higher than the standard (FAO) for all heavy metals except Cr. Plant Zn, Pb, Cr and Ni increased with 7 years of wastewater irrigation. The study concludes that the use of municipal effluent for irrigation has increased the contaminated products of Zn, Pb, Cr and Ni in soil and plant, but the contaminated products of Zn, Cr and Ni in fruits were below the permissible limits of the standard except Pb, after 7 years from this practice. Based on these results, it can be concluded that proper management of municipal effluent irrigation and periodic monitoring of soil and plant quality parameters are required to ensure successful, safe, long-term municipal effluent irrigation.

Key words: Municipal effluent, irrigation, heavy metals, accumulation, contamination, *Olea europaea* L.

INTRODUCTION

Water is a vital resource but a severely limited one in most countries for afforestations especially in the dry zones. Therefore, there is an urgent need to conserve and protect fresh water and to use the water of lower quality for irrigation (Al-Rashed and Sherif, 2000). In suburban areas, the use of industrial or municipal wastewater is common practice in many parts of the world (Singh *et al*., 2004). Woody species may utilize wastewater and uptake heavy metals through extensive root systems. It is known that serious systemic health problems can develop as a result of excessive accumulation of dietary heavy metals such as Cd, Cr and Pb in the human body (Oliver, 1997). On the other hand, wastewater may contain undesirable chemical constituents and pathogens that pose negative environmental and health impacts (Papadopoulos, 1995). Consequently, mismanagement of wastewater irrigation would create environmental and health problems to the ecosystem and human beings (Mohammad and Ayadi, 2004). When wastewater will be used continuously as the sole source of irrigation water for field crops in arid regions, excessive amounts of nutrients and toxic chemical substances could simultaneously be applied to the soil-plant system. This would cause unfavorable effects on productivity and quality parameters of the crops and the soil (Vazquezmontiel *et al*., 1996). Heavy metals are extremely persistent in the environment; they are nonbiodegradable and

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nonthermodegradable and thus readily accumulate to toxic levels. Wastewater irrigation is known to contribute significantly to the heavy metal content of soils (Mapanda *et al*., 2005). One important dietary uptake pathway could be through crops irrigated with contaminated wastewater. Soils irrigated by wastewater accumulate heavy metals such as Zn Pb, Cr and Ni in surface soil. When the capacity of the soil to retain heavy metals is reduced due to repeated use of wastewater, soil can release heavy metals into ground water or soil solution available for plant uptake. The objective of this study was to evaluate the impact of long-term land application of municipal effluent on soil and possible accumulation of heavy metals in the soil-plant system.

MATERIALS AND METHODS

Study Area and Species of Tree

The field study took place in the urban fringe area of Tehran city (Rey town), 5 km South of Tehran (35.37 N latitude and 51.31 E longitude, elevation of 1005 a.s.l.). The site of the experiments lies in a semi-arid continental climatic zone characterized by relatively mide-cold winters, mean annual rainfall is 232 mm (for the period 1993-2003, in Mehrabad Station) with maximum falls during winter. The highest precipitation falls in March and the lowest in August. Maximum and minimum mean monthly temperatures are 31.01°C in August and 5.49°C in January and average annual temperature is 13.3°C. The warmest month occurs in August and the coldest in January and vital growing season (dry season) of seven months (Mid April-Mid November). The soil is clay (according to US soil taxonomy) with 45% clay, 30% sand and 25% silt. The most common tree grown at the study site is olive (*Olea europaea* L.).

Sampling and Analysis

Industrial effluents, in addition to untreated domestic sewage, flow through open drains in some areas and are directly used for irrigation of trees. In this study, two even-aged (7 years) *Olea europaea* L. afforestations have been selected. The first stand (20000 m^2) was being irrigated by municipal effluent and the second (10000 m^2) by well water. The irrigations were carried on 8 day durations for 8 months/year (during April-November). Water, soil and plant samples were taken from all the sites. Water samples (Municipal effluent and well water) were collected from the sites monthly from May to November 2005. This collection provided 21 samples of municipal effluent in each month (7 days and each day 3 samples, at three-hour intervals 7 am, 13 pm and 19 pm). Three replicate polyethylene bottles (acid-washed) of capacity 100 mL were immersed one by one at an interval of 15 sec in an open drain that was being used for irrigation purposes and immediately after filling, 1 mL of concentrated HNO_3 was added to the water to avoid microbial utilization of heavy metals. The bottles were brought back to the laboratory and digestion was completed within a week. Each sample was analyzed for pH, Electrical Conductivity (EC) and concentration of heavy metals (Zn, Pb, Cr and Ni). The irrigation water samples (50 mL) were digested with 10 mL of concentrated HNO_3 at 80°C until the solution became transparent (APHA, 1985). The solution was filtered through Whatman No. 42 filter paper and the solution was diluted to 50 mL with distilled water.

For the field measurements, four plots of 20×20 m were randomly identified in either of both areas. Soils were collected in July 2005. In each plot, soil samples were taken from the root zone of each tree at three depths (0-15, 15-30 and 30-60 cm), by using a spiral auger of 2.5 cm diameter. Soils were randomly sampled and bulked together to form a composite sample per tree (Hbibi Kaseb, 1992). After transportation to the laboratory, Soil samples were air dried, crushed and passed through 2-mm-mesh sieve and stored at ambient temperature before analysis of soil properties and concentrations of heavy metals. The soil pH was measured with H_2O (1:2.5 ratio, dry wt/v), Electrical Conductivity (EC) and soil Organic Matter (OM) were determined using standard procedures (Jackson, 1973). For soil samples, 0.5 g of dried samples were digested with 15 mL of HNO_3, H_2SO_4 and $HClO_4$ in 5:1:1 ratio at 80°C until a transparent solution was obtained (Allen *et al*., 1986). The solution was filtered through Whatman No. 42 filter paper and the solution was diluted to 50 mL with distilled water.

The concentrations of heavy metal (Zn, Pb, Cr and Ni) in municipal effluent and well water, soil and plant samples were determined with an atomic absorption spectrophotometer (PU9400X) fitted with a specific lamp of a particular metal using appropriate drift blanks (Jackson, 1973). Quality control measures were taken to assess contamination and reliability of data. Blank and drift standards were run after five determinations to calibrate the instrument. The coefficient of variation of replicate analysis precision of analysis. Variations were found to be less than 10%. Precision and accuracy of analysis were also ensured through repeated analysis of samples against National Institute of Standards and Technology standard reference material (SRM 1570) for all the heavy metals. The results were found to be within 72% of the certified value.

In each plot, five olive individuals and Leaves and fruits were sampled from harvested as well as the standing trees (non-destructive) immediately at the time of harvest, washed with tap water, rinsed with distilled water and dried at 80°C for 24 h. Dry mass of the leaf and fruit was recorded after oven drying the samples for 72 h at 80°C. Dried plant samples were grounded and retained for mineral analysis. Data were statistically analyzed using an SPSS package (Lindaman, 1992).

Initially, normal distribution of the data was performed using the Shapiro-Wik's test. The difference between the concentrations of nutrient elements in the municipal effluent and well water, soils, leaves and fruits in the treatment and control sit were assessed by the independent-sample t-test. All statistical analyses were carried out with the program SPSS 12.5 for Windows.

RESULTS AND DISCUSSION

Municipal Effluent and Well Water

The pH of the municipal effluent was 7.60±0.01 (mean±SE), Electrical conductivity was 1.91±0.02 dS m^{-1} (Table 1). The concentration of Zn, Pb, Cr and Ni in municipal effluent was more than well water. The comparison of mean heavy metal concentrations in municipal effluent from the data of other countries suggests that the values of the present study were manifold lower than the levels observed at Harare, Zimbabwe (Mapanda *et al*., 2005). The elevated levels of Zn, Pb, Cr and Ni in municipal effluent at Rey town may be due to effluents discharged from various heavy-metal-based industries such as the fabric printing (Zn, Ni and Cr), battery (Pb) and paint industries (Zn and Pb). The higher Cr content at Lohta site is due to effluent discharged from more than 20 industries where chromium and its compounds are used as color and pigment, plating and alloys for metal surface treatment and as catalysts, situated in the catchment area of drains discharging wastewater at Rey town. It is evident that treatment of municipal effluent is not helpful in elimination of heavy metals at Rey town.

Effect of Irrigation with Municipal Effluent on Soil Properties

Table 2 summarizes the physicochemical characteristics of all samples, including both municipal effluent-irrigated and well water-irrigated soils. The soils tested are classified as clay. Independent-

Table 1: Main characteristics of municipal effluent and well water (mean±SE)

			Zn	Pb	Cr	Ni
Variations	pH	EC	------------------------------	------------ ($mg\ kg^{-1}$)	------------	------------------------------
Municipal effluent	7.63±0.01[a]	1.91±0.02[a]	3.30±0.06[a]	0.10±0.02[a]	0.10±0.005[a]	0.080±0.007[a]
Well water	7.32±0.50[b]	0.59±0.008[b]	0.73±0.01[b]	0.03±0.02[b]	0.04±0.002[b]	0.028±0.005[b]
WHO standard*	6.50-8.50	3.00	3.00	0.01	0.05	0.50

*Source: Hach (2002)

Table 2: Effect of 7 years application of municipal effluent on soil physico-chemical properties under the plantation of *Olea europaea* L.

	Soil depth (cm)					
	0-15		15-30		30-60	
Quality parameters	T_1	T_2	T_1	T_2	T_1	T_2
Tissue	Clay	Clay	Clay	Clay	Clay	Clay
pH	7.95 ± 0.016^a	7.68 ± 0.032^b	7.93 ± 0.075^a	7.67 ± 0.017^b	7.95 ± 0.014^a	7.71 ± 0.017^b
EC (dS m^{-1})	0.89 ± 0.05^a	0.69 ± 0.07^b	0.85 ± 0.068^a	0.64 ± 0.008^b	0.81 ± 0.085^a	0.63 ± 0.008^b
OM (%)	0.83 ± 0.13^a	0.37 ± 0.045^b	0.41 ± 0.038^a	0.24 ± 0.045^b	0.29 ± 0.06^a	0.11 ± 0.03^b

T_1: Irrigation of seedlings with municipal effluent; T_2: Irrigation of seedlings with well water. Unsimilar superscripts within column indicates significant (p<0.01) difference. Values are mean of three replications with±SE

Table 3: Accumulation of heavy metal in soil irrigated with municipal effluent and well water (mean±SE)

	Zn	Pb	Cr	Ni
Soil depth (cm)	(mg kg^{-1})			
0-15				
Municipal effluent	187.30 ± 10.72^a	78.40 ± 2.01^a	82.83 ± 2.51^a	46.00 ± 1.52^a
Well water	94.66 ± 2.90^b	50.00 ± 2.04^b	34.66 ± 0.88^b	27.37 ± 1.54^b
15-30				
Municipal effluent	129.61 ± 3.17^a	53.04 ± 1.03^a	61.33 ± 0.88^a	36.41 ± 0.57^a
Well water	49.16 ± 5.08^b	23.02 ± 1.04^b	27.24 ± 1.15^b	20.24 ± 0.57^b
30-60				
Municipal effluent	116.30 ± 9.13^a	42.12 ± 0.90^a	43.33 ± 2.84^a	28.29 ± 0.57^a
Well water	37.66 ± 4.50^b	18.72 ± 1.91^b	20.11 ± 0.57^b	13.33 ± 0.88^b
FAO standard*	150.00	50.00	100.00	30.00

*Source: FAO (2000)

samples t-test indicated that a seven-year application of municipal effluent increased (0-60 cm soil layer) in pH, EC and OM% when compared to soil irrigated with well water (p<0.01). The range of EC indicated no acute problems with soil salinity. By comparison, Boulding (1994) classified EC of soils (in dS m^{-1}) as: non-saline <2; moderately saline 2-8; very saline 8-16; extremely saline >16. The increase in the soil pH and EC may have been due to alkaline nature of and salt concentration in the effluent (Mitra and Gupta, 1999). Increase in OM content might be due to municipal effluent application (Kumar *et al*., 1998). Baddesha *et al*. (1985) also suggested an increase in OM may be due to municipal effluent application. These results agreed with the findings of previous studies (Mapanda *et al*., 2005; Rattan *et al*., 2005).

The results showed that continuous application of municipal effluent led to elevated levels of heavy metals in the soil (Table 3). The concentrations of all the heavy metals showed spatial and temporal variations, which may be ascribed to the variations in heavy metal sources and the quantity of heavy metals discharged through the sewage and effluents in irrigation water. This trend suggests that continuous application of municipal effluent influenced the soil physico-chemical properties (Willett *et al*., 1984). Similar results were also found in the previous studies (Liu *et al*., 2005). The mean concentrations of Zn, Pb, Cr and Ni in treated municipal effluent during the present study were similar to those in the earlier report of Singh *et al*. (2004). The range of concentrations of Zn, Pb and Ni in topsoil observed during the present study was higher than the FAO standard. The comparison of the data from the present study with earlier findings of Singh *et al*. (2004), suggested that the range of concentrations of Cr in soil was higher; however, Zn, Pb and Ni were lower than the previously reported range. This variation may be ascribed to the differences in sites, frequency of soil collection and crops under cultivation between the two studies. Among the soil layers, pH, EC, OM, Zn, Pb, Cr and Ni decreased (p<0.01) with soil depth. Bansal *et al*. (1992) studied nutrient and heavy metal accumulation in sewage water irrigated soil and suggested a decrease concentration heavy metals with soil depth. This trend correlated with the source of irrigation and development of roots in below layers may and continuous uptake of heavy metals by plants during their growth and development and higher

Table 4: Heavy metal concentrations in leaves and fruits of *Olea europaea* L.

	Zn	Pb	Cr	Ni
Variations	------------------------------------ (mg kg^{-1})			---------------------------------------
Leaves				
Municipal effluent	26.80±1.30^{a}	26.50±1.29^{a}	3.80±0.10^{a}	4.83±0.16^{a}
Well water	12.76±0.91^{b}	12.33±0.28^{b}	0.76±0.33^{b}	1.25±0.16^{b}
Permissible limits of SEPA standard	50.00	0.50	1.00	2.00
Fruits				
Municipal effluent	14.00±1.00^{a}	2.00±0.38^{a}	0.43±0.066^{a}	1.76±0.16^{a}
Well water	11.16±0.58^{b}	1.14±0.38^{b}	0.26±0.033^{b}	0.99±0.16^{b}
Permissible limits of SEPA standard	50.00	0.50	1.00	2.00

concentrations of heavy metals in the topsoil layer owing to higher evaporation (Sharma *et al*., 2007). The trend in topsoil might be due to clay nature of the soil and accumulation Zn, Pb, Cr and Ni in the topsoil layer probably was due to surface application of municipal effluent and their retention in soil micelles.

Concentrations of Heavy Metals in Leaves and Fruits of *Olea europaea* L.

The mean concentrations of all the heavy metals in *Olea europaea* L. (leaves and fruits) at sites irrigated with municipal effluent were considerably higher than the trees irrigated with well water (Table 4). The comparison of mean values of heavy metals recorded in the plant material during the present study with SEPA standard (2005) showed that Pb, Cr and Ni concentrations were higher in leaves of trees irrigated with municipal effluent, however, Zn was not higher than SEPA standard (2005). Mean concentrations of Zn, Cr and Ni were below the SEPA standard (2005) in fruits of trees irrigated with municipal effluent. Results from this study agreed with the data reported by Rattan *et al*. (2005). Results from present and previous studies (Liu *et al*., 2005; Muchuweti *et al*., 2006; Sharma *et al*., 2007) demonstrate that the plants grown on wastewater-irrigated soils contaminated with heavy metals and pose a major health concern. Absorption and accumulation of heavy metals in plant tissue depend upon many factors, which include temperature, moisture, organic matter, pH and nutrient availability. The site irrigated with municipal effluent showed higher organic matter content than the site irrigated with well water. The site irrigated with municipal effluent also showed similar contamination levels for heavy metals, which may be at least partially explained by the high organic matter. Organic complexing molecules of Low Molecular Weight (LMW) serve as carriers of micronutrients. LMW has been shown to increase heavy metals uptake (Chen and Aviad, 1990), whereas the presence of organic matter has been reported to increase the uptake of Zn in the wheat plant (Rupa *et al*., 2003). The pH of the soil was close to 7.5-8 in all depths. The alkaline range of soil is known to restrict the mobilization of heavy metals, thus reducing their uptake. However, the high nutrient input from irrigation water at these sites could result in relatively high uptake of heavy metals as a result. The field data support this argument in that a higher uptake was recorded at sites that were irrigated with municipal effluent as compared to the area receiving well water. Long-term application municipal effluent and presence of N in municipal effluent is known to be associated with an increase in plant uptake and bioavailability of heavy metals (Nambiar and Ghosh, 1984). In the present study it seems that many soil factors such as pH, organic matter, nitrogen bioavailability, soil moisture and temperature have interacted to impact on uptake.

CONCLUSIONS

The study concludes that irrigation by municipal effluent has increased the heavy metal concentrations in soil and plants of receiving area. Zn, Pb and Cr concentration in municipal effluent was found above the permissible limit set by WHO. The pollution load index values indicated that the wastewater-irrigated soils were moderately enriched with Zn, Pb, Cr and Ni in topsoil. Heavy metal concentrations in plants show significant spatial and temporal variations. Furthermore, it was concluded the wastewater-irrigated grown plants were contaminated with those heavy metals and exceeded the permissible limits for vegetables set by SEPA standard (2005). This study clearly showed that consumption of fruits of *Olea europaea* L. by the urban and suburban population might pose health hazards due to Pb contamination. An important issue is that the contamination levels were frequently higher than permissible limits in the plant tissue, at the same sites as water and soil samples that comply with established safe standards. This has important implications for policy in that programmes aimed at monitoring and controlling heavy metal concentrations in irrigation water sources will not necessarily result in acceptable levels in plants. Policies and programs need to be adapted so that local edaphic conditions and appropriate local measures developed for ameliorating heavy metal uptake by crops for a given set of local conditions. These measures need to be regularly reviewed to take in to account factors such as the accumulation of heavy metals in the soil over time.

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