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Heavy metal accumulation in soils and grains, and health risks associated with use of treated municipal wastewater in subsurface drip irrigation

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Abstract Constant use of treated wastewater (TWW) for irrigation over prolonged periods may cause buildup of heavy metals up to toxic levels for plants and animals, and entails environmental hazards in different aspects. However, application of TWW on agricultural land might be an effective and sustainable strategy in arid and semi-arid countries where fresh water resources are under great pressure, as long as potential harmful effects on the environment including soil, plants, and fresh water resources, and health risks to humans are minimized. The aim of this study was to assess the effect of deep emitters on limiting potential heavy metal accumulation in soils and grains, and health risk under drip irrigation with treated municipal wastewater. A field experiment was conducted according to a split block design with two treatments (fresh and wastewater) and three sub-treatments (0, 15, and 30 cm depth of emitters) in four replicates on a sandy loam Calcic Argigypsids, in Esfahan, Iran. The annual rainfall is about 123 mm, mean annual ET_o is 1457 mm, and the elevation is 1590 m above sea level. A two-crop rotation of wheat (*Triticum* spp.) and corn (*Zea mays*) was established on each plot with wheat growing from February to June and corn from July to September. Soil samples were collected before planting and after harvesting for each crop in each year. Edible grain samples of corn and wheat were collected at harvest. Elemental concentrations (Cu, Zn, Cd, Pb, Cr, Ni) in soil and grains were determined using an atomic absorption spectrophotometer. Results showed that the concentrations of heavy metals in the wastewater-irrigated soils were not significantly different ($P>0.05$) compared with the freshwater-irrigated soils. No significant difference ($P>0.05$) in heavy metal content in soil between different depths of emitters was found. A pollution load index (PLI) showed that there was no substantial buildup of heavy metals in the wastewater-irrigated soils compared to the freshwater-irrigated soils. Cu, Pb, and Zn concentrations in wheat and corn grains were within the permissible US Environmental Protection Agency (EPA) limits, but concentrations of Cd (in wheat and corn) and Cr (in corn) were above the safe limits of the EPA. In addition, concentrations of Ni in wheat and corn seeds were several folds higher than the EPA standards. A health risk index (HRI) which is usually adopted to assess the health risk to hazard materials in foods showed values higher than 1 for Cd, particularly for wheat grain (HRI >2.5). Results also showed that intake of Cu through consumption of edible wheat grains posed a relatively high potential health risk to children (HRI >1.4), whereas children might also be exposed to health risk from Cd and Cr from corn grains (HRI >1.4). Based on aforementioned results, it can be concluded that the emitter depth in drip irrigation does not play a significant role in the accumulation of heavy metals from TWW in our sandy loam soil. Although their accumulation in the soil was limited and similar to using freshwater, uptake of Cd and Cr by wheat and corn was relatively large and hence resulted in health risk. The results suggest that

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more attention should be directed towards cultivation of other crops with drip irrigation system for a safe and more productive use of wastewater for irrigation. Alternatively, methods that filter the wastewater before it enters the soil environment might be an option that needs further investigation.

Keywords Municipal wastewater · Subsurface drip irrigation system · Health risk index · Plant concentration factor · Pollution load index · Esfahan

Introduction

Water scarcity is a main issue in most countries in the Middle East and North Africa. According to the International Water Management Institute (IWMI), by 2025, 1.8 billion people will live in countries or regions with serious water shortage (IWMI 2000). Using poor quality water could be an option for reducing pressure on water resources (Oweis and Hachum 2009). Nonconventional water such as wastewater is an indispensable part of irrigation water resources particularly in arid regions, which are facing drought during a considerable part of the crop growing season. In Esfahan region (Iran), municipal (urban) treated wastewater (TWW) is widely used for irrigating agricultural lands for fruit and vegetable production. Besides providing water and nutrients to meet crop requirements, this usage could solve disposal problems of wastewater, in that it alleviates environmental pollution in city suburbs. Research has shown that properly using TWW for irrigation of agricultural crops is the best solution for the abovementioned problem (Pescod 1992).

However, application of TWW and sludge, separated from urban wastewater, on agricultural land entails environmental hazards in different aspects. Wastewater contains metals that are toxic when given thresholds are exceeded. Constant use of TWW for irrigation over prolonged periods may cause buildup of heavy metals up to toxic levels for plant and animal health (Jagtap et al. 2010; Ehsan et al. 2011.; Kelepertzis 2014). Crops irrigated with such waters take up substantial high amounts of heavy metals (Arora et al. 2008). Wei and Yang (2010) reported that the concentrations of Cr, Cu, Pb, Zn, Ni, Cd, Hg, and As in agricultural soils were higher than their background values in most of the 12 studied cities in China. A study in Lechang, Guangdong, showed that irrigating rice fields with untreated mining wastewater caused rice grains to become heavily contaminated by Cd (Yang et al. 2006; Williams et al. 2009). Lu et al. (2015) made a comprehensive map of soil and water pollution threats to food safety and human health in some villages in eastern China where the morbidity rate of cancer was significantly higher than the average level. Usage of TWW in irrigation thus necessitates special management to take advantage of nutrients while ensuring that harmful effects on the environment including soil, plants, and fresh water resources, and health risks to humans are minimized.

Therefore, new irrigation management strategies and technologies must be developed and integrated with farming systems in order to effectively utilize the precious existing water resources for achieving high water use efficiency and increasing productivity (Naeem and Rai 2005). Pescod (1992) presented benefits and disadvantages of different wastewater irrigation systems and concluded that drip irrigation is able to solve most problems of using wastewater in agriculture. Reuse criteria can be relaxed somewhat when using drip irrigation (DI) and primarily subsurface drip irrigation (SDI) because the soil acts as a complementary biofilter hence reducing soil contamination (Oron et al. 1999). Heidarpour et al. (2007) demonstrated that concentrations of chemical constituents in soil layers were influenced by water movement patterns, chemical concentrations in irrigation water, and plant uptake. Drip irrigation applies water precisely and distributes it uniformly in comparison with furrow and sprinkler irrigation resulting in potential reduction of subsurface drainage, controlled soil salinity, improved water and nutrient management, and potentially improving yields and crop quality (Ayars et al. 1999; Hanson and May 2004). Other researchers showed that this irrigation system could reduce water losses to evaporation, runoff, and percolation; decrease weed growth and pollution of soil, water, and plants, improve control of irrigation delivery systems; and reduce physical contact between farmers and wastewater (Gushiken 1993; Oron et al. 1999).

As contaminants enter into the human body through the food chain, accumulation of heavy metals in soil and plants has become an important issue for human health. Heavy metals in contaminated water and soil could increase the potential risk of cancers (He et al. 2014). Several researchers reported that contaminated water and food could even increase the morbidity and mortality of cancers, especially digestive cancers (Sun et al. 2014; Zhang et al. 2014; Zhao et al. 2014). Factors such as efficiency of uptake of given heavy metals which is plant species dependent, and soil characteristics need to be considered to evaluate this phenomenon (Rattan et al. 2005). Mn, Cu, and Zn are non-protein substances of a large number of enzymes, and show toxic effects on human body when their concentration exceeds given thresholds. Cu surplus had been associated with liver damage, and Zn may produce adverse nutrient interactions with Cu. Other metals like Pb and Cd are toxic even at low concentrations. Cereals, which are the main source of food in most countries, are also an important source of these metals. An increased metal uptake by food crops, vegetables, and fruits grown on soils contaminated with these metals has been widely observed (Iyengar and Nair 2000). When critical levels of heavy metal intake are exceeded over time, various harmful effects can be discerned, which are responsible for some refractory diseases, disorder in immunological defenses, fetal defects, disabilities, and different kind of cancers (Iyengar and Nair 2000; Turkdogan et al. 2003).

Regarding these effects, international organizations such as the Organization for Economic Co-operation and Development (OECD), the European Union (EU), and the US Environmental Protection Agency (EPA) recommend guidelines, and suggested procedures and regulations of risk assessment (Gushiken 1993; Adams and Chapman 2003). Besides that, the Food and Agriculture Organization (FAO), the World Health Organization (WHO), the EPA, and other regulatory bodies of various countries strictly regulate the allowable concentrations or maximum permitted concentrations of toxic heavy metals in foodstuffs (FAO/WHO 1984; USEPA 2000; US-EPA 2012).

In order to assess health risks, it is necessary to identify the potential of a heavy metal source for introducing risk agents into the environment, to estimate the amount of risk agents exposed to the human–environment boundaries, and to quantify the health consequence of the exposure (Ma and Chen 2007; Zhi-Fan et al. 2010). The objective of this study was therefore (i) to investigate the effect of emitter depth in (subsurface) drip irrigation with treated wastewater on the extent of soil contamination with heavy metals, (ii) to assess the heavy metal uptake by wheat and corn, and thus their accumulation in (consumable) grains, and (iii) to evaluate the potential health risks associated with human consumption of food crops grown under the above conditions.

Material and methods

Field site characteristics

The study area was located on the border of the city of Esfahan, Iran (32° 37′ N 51° 43′ E and 1590 m above sea level). Esfahan is situated on the semi-arid plateau of central Iran, with dry and hot summers and mild winters. Mean annual rainfall is about 123 mm, mean annual ET_0 is 1547 mm, and mean monthly temperature ranges from 3.4 to 28.9 °C. During the study period, from February 2010 to October 2011, the temperature varied from −6 to 41 °C and relative humidity from 3 to 100 %. The soil was a sandy loam Calcic Argigypsids (NRCS 2014) with 60 % sand, 25 % silt and 15 % clay, 12 g kg^{-1} organic carbon, and an overall ECe of 2.5 dS m^{-1} in the top 50 cm at the beginning of the experiment (Table 1). The water table at the experimental site was at more than 2 m below the soil surface.

Experimental setup

Twenty-four 3 m×3 m plots were laid out according to a split block design with two treatments and three subtreatments in four replicates. A two-crop rotation of wheat (*Triticum aestivum* L.) and corn (*Zea mays*) was established on each plot with wheat growing from February to June and corn from July to September. This rotation was repeated in the second year. A recommended amount of 150 kg of urea fertilizer (based on soil analysis) was evenly distributed manually over the land before plowing. A plant distance of 3 and 30 cm was used for wheat and corn, respectively. Row distance was 30 and 60 cm, respectively. The fields were irrigated with an irrigation system installed at the border of the farm. The system included one electromotor for transferring wastewater from the channel passing besides the farm, one sand filter to segregate large suspended solids from TWW, and one disk filter for small solids. All plots were under drip irrigation with inline emitters (Iran Dripper Co., Esfahan, Iran). A supply rate of 4 L h^{-1} was used based on an irrigation schedule following the ET-HS model (Najafi and Tabatabaei 2007).

The treatments were irrigation water quality with dripper depth as sub-treatment. Emitters were

Table 1 Initial physicochemical properties of the soil at the experimental site in the top 50 cm

pH	ECe (dS/m)	OC (g/kg)	Sand (%)	Silt (%)	Clay (%)	Cu (mg/kg)	Zn (mg/kg)	Cd (mg/kg)	Pb (mg/kg)	Cr (mg/kg)	Ni (mg/kg)
8.1	2.5	12	60	25	15	27.85	81.9	2.3	7.45	13.8	18.65

positioned at the soil surface (DI_0), at 15 cm depth (SDI_{15}), and at 30 cm depth (SDI_{30}). Irrigation water was freshwater (FW) and treated wastewater (TWW). FW was taken from an urban water supply network, whereas TWW came from the Esfahan wastewater plant.

Sampling and analysis

To evaluate the irrigation water quality, 20 mL was collected at every irrigation time. pH and electrical conductivity EC of the samples were immediately determined after which they were frozen (−5 °C). At the end of each farming season, all water samples were defrosted and mixed, and the elemental concentrations were determined using an atomic absorption spectrophotometer (PerkinElmer, USA) (Table 2).

Soil samples were collected in June after harvesting wheat and in September after harvesting corn in each year (2010 and 2011). In each plot, three separate samples were taken at depth of 0–50 cm with an auger (20 cm diameter) and mixed to have a composite sample. They were transported to the laboratory in air-tight bags, air-dried and sieved through a mesh (<2 mm), and then sealed in envelopes until analysis. Samples were used to measure pH, ECe, and heavy metal concentration according to standard procedures (Carter and Gregorich 2007). The measurement of soil pH H_20 was done by means of a glass electrode and a calomel electrode as reference (pH meter, Model Corning M220, USA). Before the measurements, the pH meter was calibrated using standard solutions of pH 4 and pH 7. EC was measured in 1:5 soil water extract by means of a conductivity meter (Istek, Model 915PDC, Korea) (Sonmez et al. 2008). The determination of soil organic carbon was based on the Walkley-Black (Walkley and Black 1934) chromic acid wet oxidation method. To determine total concentrations of Cd, Cr, Cu, Ni, Pb, and Zn in the soil, immediately after each soil sampling time, 0.100 g of air-dried soil was digested and the elemental concentrations were determined using an atomic absorption spectrophotometer (PerkinElmer, USA). All determinations were repeated three times to minimize the risk of error.

Edible grain samples of corn and wheat were collected at harvest from two to three randomly selected plants within each plot. The grains were properly washed with deionized water to remove all dust and surface pollution, dried at 65 °C for 24 h and then reduced to a fine powder in a porcelain mortar. Precisely weighed quantities of 0.2000 g were digested with a mixture of nitric acid and hydrogen peroxide, and analyzed as above. All analyses were made at the Khorasgan University reference laboratory (Isfahan, Iran), which follows quality management procedures according to international standards.

Data analysis

As a measure for the degree of soil pollution by each metal, we used the widely used pollution load index (PLI) (Liu et al. 2005):

$$\mathrm{PLI} = C_{\mathrm{WW}}/C_{\mathrm{FW}} \tag{1}$$

where C_{WW} and C_{FW} represent the heavy metal concentrations in the TWW- and FW-irrigated soils, respectively.

A plant concentration factor (PCF) was calculated as follows (Cui et al. 2004):

$$\mathrm{PCF} = C_{\mathrm{plant}}/C_{\mathrm{soil}} \tag{2}$$

where C_{plant} and C_{soil} represent the heavy metal concentration in extracts of plants and soils on a dry weight basis, respectively.

The daily intake of metals (DIM) was estimated as:

$$\mathrm{DIM} = \left(C_{\mathrm{metal}} \times C_{\mathrm{factor}} \times D_{\mathrm{food\ \ intake}}\right)/B_{\mathrm{average\ \ weight}} \tag{3}$$

where C_{metal} represents heavy metal concentrations in plants (mg kg^{-1}), C_{factor} is a conversion factor, $D_{\mathrm{food\ intake}}$ is daily intake of vegetables, and $B_{\mathrm{average\ weight}}$ is average body weight. A conversion factor of 0.085 was used to convert fresh green vegetable weight to dry weight as described by Rattan et al. (2005). The average

Table 2 Chemical properties of the treated wastewater and the freshwater

Water Source	pH	EC (ds/m)	Cu (mg/kg)	Zn (mg/kg)	Cd (mg/kg)	Pb (mg/kg)	Cr (mg/kg)	Ni (mg/kg)
Freshwater	7	1.2	0.07	0.06	0.010	0.01	0.010	0.010
Wastewater	7.5	1.4	0.13	0.37	0.012	0.02	0.013	0.022

Values shown are means (n=45)

daily intakes for adults and children was considered to be 0.345 and 0.232 kg per person per day, respectively, while the average adult and child body weights were considered to be 55.9 and 32.7 kg, respectively, as used in previous studies (Ge 1992; Wang et al. 2005; Khan et al. 2008; Zhi-Fan et al. 2010).

The health risk index (HRI) for the local population through the consumption of contaminated grains was calculated using DIM and reference oral dose (RfD) for each metal:

$$\mathrm{HRI} = \mathrm{DIM}/\mathrm{RfD} \tag{4}$$

An HRI <1 means that the exposed population is assumed to be safe (US-EPA 2012). The data were statistically analyzed using the package STATISTICA 10 (StatSoft 2011). Statistical significance was detected using the independent samples t test and analysis of variance at α=0.05. Values shown are averages over two farming seasons.

Results and discussion

Physicochemical properties and heavy metal accumulation in soil

Table 3 shows the basic physicochemical soil properties and heavy metal concentrations under the different treatments and crops. pH was not significantly (p>0.05) affected by the quality of irrigation water, depth of emitter or type of crop. ECe under TWW irrigation was significantly higher (p<0.01) than under irrigation with FW, with the first showing values approaching 4 dS m^{-1}, above which soils are expected to experience salinity problems according to USDA criteria (Corwin and Lesch 2013). Mohammad and Mazahreh (2003) stated that the increase in EC for soil irrigated with TWW compared with soil irrigated with freshwater originates from the high level of totally dissolved solids in TWW. Similarly, our study showed that reuse of TWW leads to a significant (p<0.05) increase of Na and K in soil. This observation was confirmed by Khai et al. (2008) and Mojiri and Abdul Aziz (2011). They reported that higher concentration of cations as Na and K in TWW led to an increase in EC and exchangeable Na and K in soils irrigated with TWW. Depth of emitters did not result in significant differences (p>0.05). In addition, differences between wheat and corn, though showing a different irrigation schedule with corn receiving water at higher frequency, were not significant (p>0.05).

With values below 4 g kg^{-1}, soil organic carbon (SOC) content was low. This is to be expected because of rapid mineralization of SOC under semiarid conditions (Giongo et al. 2011) and intensive use of these soils. No significant differences in SOC (p>0.05) were found when comparing water quality or emitter depth. Adding wheat straw during crop maturity of wheat slightly increased SOC compared to corn (p<0.05). Long-term planting of crops with residue remaining on the field after harvesting could gradually increase the amount of SOC in soils that have low amounts of it (Jin et al. 2008).

Regarding heavy metal concentration, no significant differences (p>0.05) were observed between emitter depth. The high frequency of irrigation needed to address the excessive crop water requirement in our arid study area leads to similar wetting patterns in soil, which could explain the minor effect of emitter depth.

Although the used TWW showed concentrations that were for several heavy metals much higher as compared to FW (Table 2), differences in heavy metal concentrations in the soil were not significant across the water qualities. Recent studies (Asgari et al. 2007a, b) showed higher production of total dry mater in TWW-irrigated plots compared with FW-irrigated plots. Consequently, higher amounts of nutrient were taken up by plants and overall low concentrations of heavy metals were observed in sampled soils in TWW-irrigated plots. Crop type did play a role for Cd (p<0.01), Pb (p<0.01), and Ni (p<0.05), but not for Cu, Zn, and Cr (p>0.05). The findings showed that each crop has the ability to take up specific heavy metals from soil. Several studies

Table 3 Basic properties and total metal concentration in the top 50-cm soil after drip irrigation DI of wheat and corn with wastewater (WW) and freshwater (FW).

Property and element in soil	Wheat						Corn						EPA soil, MCL
	WW-irrigated			FW-irrigated			WW-irrigated			FW-irrigated			
	DI_0	DI_{15}	DI_{30}	DI_0	DI_{15}	DI_{30}	DI_0	DI_{15}	DI_{30}	DI_0	DI_{15}	DI_{30}	
pH	8.2	8.2	8.1	8.3	8.2	8.3	8.2	8.2	8.2	8.3	8.2	8.2	–
EC (ds/m)	3.7	4.0	4.3	2.1	2.5	2.2	3.7	3.1	4.2	2.0	2.7	2.3	–
OC (g/100 g)	3.9	3.8	3.5	3.8	3.7	3.7	2.6	2.6	2.8	2.5	2.6	2.9	–
Cu (mg/kg)	57.7	60.0	60.4	64.5	54.0	57.1	63.6	57.3	51.9	61.5	64.3	64.3	170
Zn (mg/kg)	161.1	190.1	160.7	156.8	153.7	154.6	197.3	230.2	238.9	146.1	227.6	197.7	350
Cd (mg/kg)	5.5	5.4	4.9	5.3	5.5	5.0	4.1	3.8	4.0	3.9	3.8	4.0	3
Pb (mg/kg)	20.2	19.8	20.8	18.9	17.5	20.2	46.4	47.3	46.9	41.6	37.5	44.8	250
Cr (mg/kg)	29.1	26.8	27.2	29.3	33.9	31.9	28.4	23.8	35.3	45.3	27.0	44.7	100
Ni (mg/kg)	25.6	23.5	24.6	25.0	25.4	24.2	30.0	29.8	34.8	28.9	29.0	30.3	100

Emitters were located at the surface (DI_0), 15 cm depth (DI_{15}), and 30 cm depth (DI_{30}). MCL is a maximum concentration level (US-EPA 2012)

confirmed different phytoextraction ability of heavy metal fractions by different plant species (Van Ginneken et al. 2007; Wang et al. 2009; Bieby Voijant et al. 2011).

Overall, mean values of Cu, an essential element for living organisms, were 58.9 and 60.5 mg kg^{-1} under wheat and corn, respectively. These concentrations were below the Environmental Quality Standards set by the US Environmental Protection Agency for soil samples (US-EPA 2012). Zn and Ni are also essential elements, and their mean values were 162.8 and 27.4 mg $kg_{soil}{}^{-1}$ for wheat and 206.3 and 30.5 mg $kg_{soil}{}^{-1}$ for corn, respectively, i.e., below the EPA standards. The mean values of Cd were 5.3 and 3.9 mg $kg_{soil}{}^{-1}$ for wheat and corn, respectively. In spite of its low values, Cd concentrations at much lower levels create toxicity compared to the other elements analyzed in this study, and its concentration was higher than the EPA standard in all treatments. Finally, Pb and Cr are not essential elements for plants and are toxic. Their mean values were 29.7 and 19.6 mg $kg_{soil}{}^{-1}$ for wheat and 34.1 and 44.1 mg $kg_{soil}{}^{-1}$ for corn, respectively, and lower than the EPA standards.

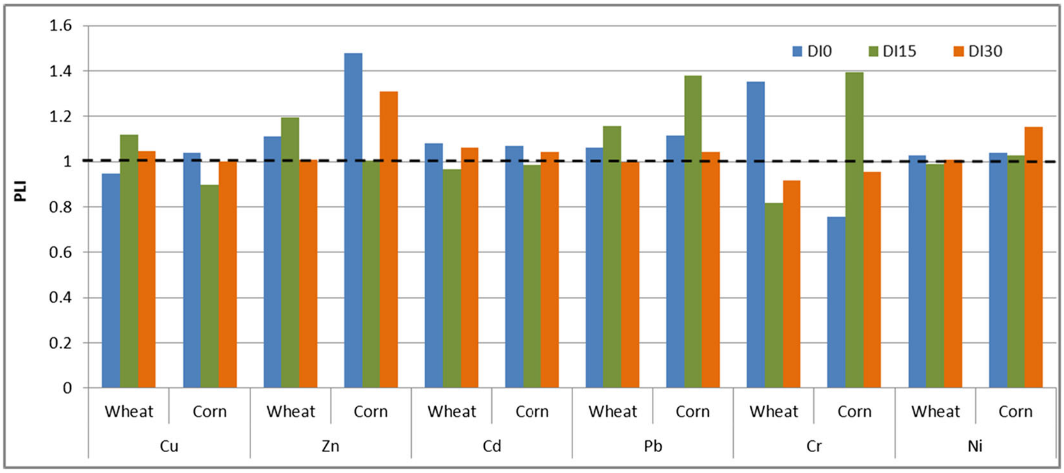

Fig. 1 Pollution load index (PLI) in wastewater-irrigated and freshwater-irrigated soils of wheat and corn. Emitters were located at the surface (*DI0*), 15 cm depth (*DI15*), and 30 cm depth (*DI30*). (*N*=96)

Table 4 Heavy metals concentration in wheat and corn seeds grown under wastewater WW and fresh water FW drip irrigation

Element in seeds	Wheat						Corn						EPA crop,MCL
	WW-irrigated			FW-irrigated			WW-irrigated			FW-irrigated			
	DI_0	DI_{15}	DI_{30}	DI_0	DI_{15}	DI_{30}	DI_0	DI_{15}	DI_{30}	DI_0	DI_{15}	DI_{30}	
Cu (mg/kg)	11.7	12.8	13.8	12.5	12.4	13.5	3.7	3.9	3.1	4.5	3.5	3.3	20
Zn (mg/kg)	32.7	34.8	39.2	28.0	29.6	27.7	43.2	42.7	36.2	42.0	37.0	41.2	50
Cd (mg/kg)	1.1	1.1	0.9	1.2	1.0	1.2	0.2	0.3	0.3	0.4	0.3	0.1	0.1
Pb (mg/kg)	0.1	0.1	0.1	0.1	0.1	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.2
Cr (mg/kg)	0.1	0.0	0.0	0.0	0.0	0.0	1.0	0.4	0.6	1.4	1.9	0.6	1
Ni (mg/kg)	2.3	3.0	3.2	2.9	3.3	3.4	1.8	2.2	1.6	1.5	1.7	2.0	0.04

Emitters were located at the surface (DI_0), 15 cm depth (DI_{15}) and 30 cm depth (DI_{30}). MCL is a maximum concentration level (US-EPA 2012). Values shown are means (n=96)

The PLI indices in Fig. 1 show that there was no substantial buildup of heavy metals in the TWW-irrigated soils compared to FW-irrigated soils. No significant differences (p>0.05) were found between crop and emitter depth. Continuous irrigation with TWW containing heavy metals is expected to increase their total concentration in soil. However, this study showed PLI values near 1 for most treatments and elements. Furthermore, no significant differences between soils irrigated with TWW and FW were found. This could be partly explained by the subsurface drip irrigation system with higher water use efficiency, limited wetted soil volume, and concentration of roots within the dripper's bulb (Coelho and Or 1999; Wang et al. 2006). As a result, less irrigation water was needed to achieve similar yields as compared with other irrigation systems (Najafi and Tabatabaei 2007; Asgari and Najafi 2008). Because of lower water usage, fewer amounts of heavy metals were added to the soil through irrigation with TWW. To the best of our knowledge, specific studies on the effect of emitter depth on accumulation of heavy metal released from TWW under subsurface drip irrigation in grain crops have not been reported so far. Kabata-Pendias and Pendias (2001) show that both roots and leaves absorbed a significant amount of Cd. Hinesly et al. (1984) reported that soil pH had great influence on Cd transport in corn (*Z. mays* L.). In studies related to water and solute transport, the Hydrus model was used

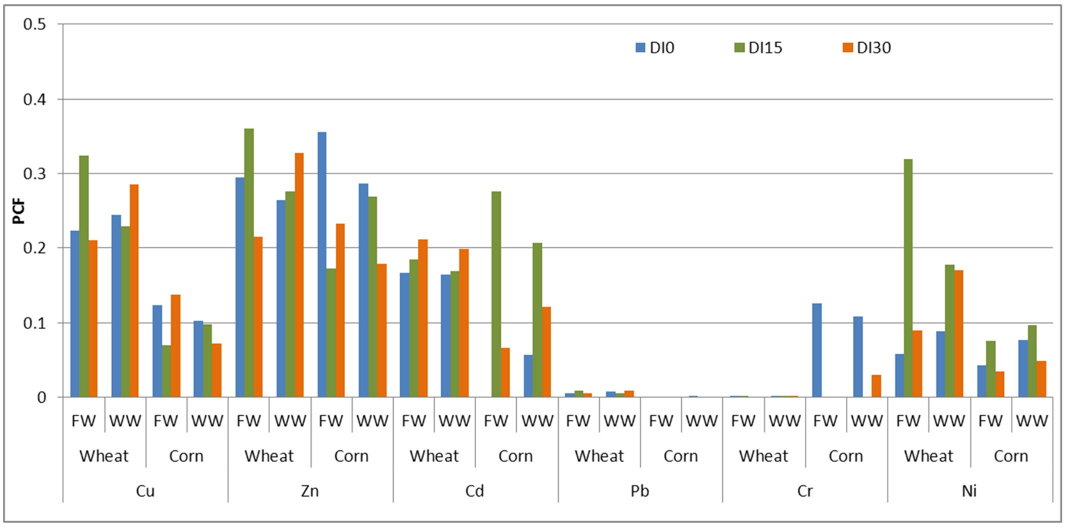

Fig. 2 The plant concentration factor (PCF) in edible seeds of wheat and corn grown in wastewater (*WW*)-irrigated and freshwater (*FW*)-irrigated soils. Emitters were located at the surface (*DI0*), 15 cm depth (*DI15*), and 30 cm depth (*DI30*). (*N*=96)

to simulate soil water and solute transport under subsurface drip irrigation (Kandelous and Šimůnek 2010; Kandelous et al. 2011; Shan and Wang 2012). Cote et al. (2003) indicated that in highly permeable coarse textured soils, water moved quickly downwards from the dripper. Skaggs et al. (2004) demonstrated that for drip tape irrigation the soil water distribution predicted with Hydrus-2D agreed well with experimental observations. Globally, recent studies demonstrated that the number of drippers, discharge rate, and the irrigation frequency of a drip irrigation system could adopt wetted soil volume as close as possible to the crop-rooting pattern (Wang et al. 2006; Kandelous et al. 2011).

Heavy metals in seeds

Concentrations of heavy metals in the edible seeds of wheat and corn are shown in Table 4. No significant differences were found between water source and emitter depth ($p>0.05$). Because of the high frequency of irrigation as well as roots growing primarily in the wetted zone near the emitters, emitter depth did not affect the level of absorbed heavy metals for a given crop. Our results (not presented here) show that because of the high amount of nutrients, especially nitrogen and phosphorus in TWW, the wheat and corn seed dry matter per unit area under TWW drip irrigation was higher than under FW use, which is consistent with other studies (Asgari and Najafi 2008; Zema et al. 2012). These higher seed dry matter with TWW might compensate for the higher uptake hence resulting in non-significant differences. Crop type on the other hand did significantly affect concentrations of Cu, Cd, Pb, and Ni in wheat seeds being significantly higher than in those of corn ($p<0.01$), whereas Zn and Cr concentrations were larger in corn ($p<0.01$). This could be ascribed to the plant-specific ability to absorb trace elements from soil (Van Ginneken et al. 2007; Wang et al. 2009; Bieby Voijant et al. 2011).

The maximum permissible limit of Pb, Cd, Cu, Zn, Cr, and Ni are respectively 0.2, 0.1, 20, 50, 1, and 0.04 mg kg^{-1} dry seed based on a dry weight (US-EPA 2012). Mean values of Cu, Zn, and Cr elements were below the EPA limits and appear to be safe. However, concentrations of Cr, Cd, and Ni did exceed standard limits. Both wheat and corn were heavily contaminated with Ni and Cd, and corn was contaminated with Cr. The mean values of Cd in wheat and corn were 10.5 and 2.7 times the EPA standard limits, respectively.

Table 5 DIM for children and adults of heavy metals associated with the crops grown in wastewater and freshwater drip irrigated soils

Emitter depth		Cu		Zn		Cd		Pb		Cr		Ni	
		Wheat	Corn	Wheat	Corn	Wheat	Corn	Wheat	Corn	Wheat	Corn	Wheat	Corn
DI_0	Adults	3.3E−02	1.1E−02	9.4E−02	1.2E−01	3.0E−03	4.7E−04	2.4E−04	5.7E−05	1.5E−04	2.8E−03	6.5E−03	5.0E−03
	Children	5.7E−02	1.8E−02	1.6E−01	2.1E−01	5.2E−03	8.0E−04	4.1E−04	9.7E−05	2.6E−04	4.8E−03	1.1E−02	8.6E−03
DI_{15}	Adults	3.7E−02	1.1E−02	1.0E−01	1.2E−01	3.0E−03	8.4E−04	1.7E−04	0.00	6.0E−05	1.2E−03	8.7E−03	6.3E−03
	Children	6.3E−02	1.9E−02	1.7E−01	2.1E−01	5.2E−03	1.4E−03	2.8E−04	0.00	1.0E−04	2.1E−03	1.5E−02	1.1E−02
DI_{30}	Adults	3.9E−02	8.7E−03	1.1E−01	1.0E−01	2.7E−03	9.8E−04	2.5E−04	0.00	7.7E−05	1.6E−03	9.1E−03	4.6E−03
	Children	3.9E−02	1.5E−02	1.9E−01	1.8E−01	4.6E−03	1.7E−03	4.3E−04	0.00	1.3E−04	2.8E−03	1.5E−02	7.9E−03

Emitters were located at the surface (DI_0), 15 cm depth (DI_{15}), and 30 cm depth (DI_{30})

For Ni, concentrations were even 75 and 45 times higher than the EPA limits for wheat and corn, respectively. These concentrations are of serious concern owing to the potential public health impacts.

Several researches also showed that irrigating rice fields using TWW increased the accumulation of heavy metals in plants as compared to FW irrigation resources (Chung et al. 2011; Jung et al. 2014). Li et al. (2012) confirmed that heavy metals (Cd, Cr, Cu, Zn, and Ni) from soil are transferred to edible seeds such as of rice and to root of vegetables. Some studies in contaminated mining areas showed that Zn and Cu concentration exceed the maximum allowable limit in parsley and carrot roots (Harmanescu et al. 2011), and roots of other vegetables compared with those grown in clean reference areas (Lacatusu and Lacatusu 2008). Cd is a mobile element, easily absorbed by roots and transported to shoots where it is uniformly distributed in plant (Sekara et al. 2005). Several studies observed high concentrations of Cu in seeds (Fytianos et al. 2001; Sridhara Chary et al. 2008; Olawoyin et al. 2012; Khan et al. 2013b; Liu et al. 2013). In some studies related to soil and crop pollution, average level of heavy metals, such as Cd and Pb were more than 0.2 mg kg^{-1} in vegetables (Arora et al. 2008; Zhuang et al. 2009; Singh et al. 2010; Huang et al. 2014). Pandey et al. (2012) studied dietary intake of heavy metals via vegetables. The results indicated substantial accumulation of heavy metals in vegetables, which was found to be maximum in leaves (spinach) followed by fruits (tomato) and roots (radish). They determined concentrations of Cd, Ni, and Pb in vegetables exceeding the safe limits of prevention of food adulteration and the values recorded were 5.36 mg g^{-1} for Cd, 13.77 mg g^{-1} for Cr, 27.46 mg g^{-1} for Cu, 5.94 mg g^{-1} for Ni, and 19.77 mg g^{-1} for Pb.

The plant concentration factor (PCF) values were not significantly different (p>0.05) among the two water qualities and three depths of emitters. Higher wheat and corn yields with TWW drip irrigation resulted in similar PCF values as compared to FW irrigation. Overall, there was a great variability in PCF values for each heavy metal per treatments and sub-treatments. The mean values of PCF for heavy metals including Cu, Zn, Cd, Pb, Cr, and Ni were respectively 0.07 to 0.32, 0.17 to 0.36, 0.0 to 0.28, 0.0 to 0.01, 0.0 to 0.13, and 0.03 to 0.32 (Fig. 2). On the other hand, PCF values of Cu and Pb were significantly different (p<0.01) between crop type. For Pb and Cu, the values of PCF in corn seeds were lower than those in wheat, while the PCF values of other elements in corn seeds were comparable to the levels found in wheat seeds. Some studies (Jan et al. 2010) showed that Cd and Cr had higher value of PCF than other elements, which was not confirmed in our study.

Human health risks resulting from daily intake of heavy metals through food consumption

There are various exposure pathways of pollutants to humans, such as the food chain, dermal contact, and

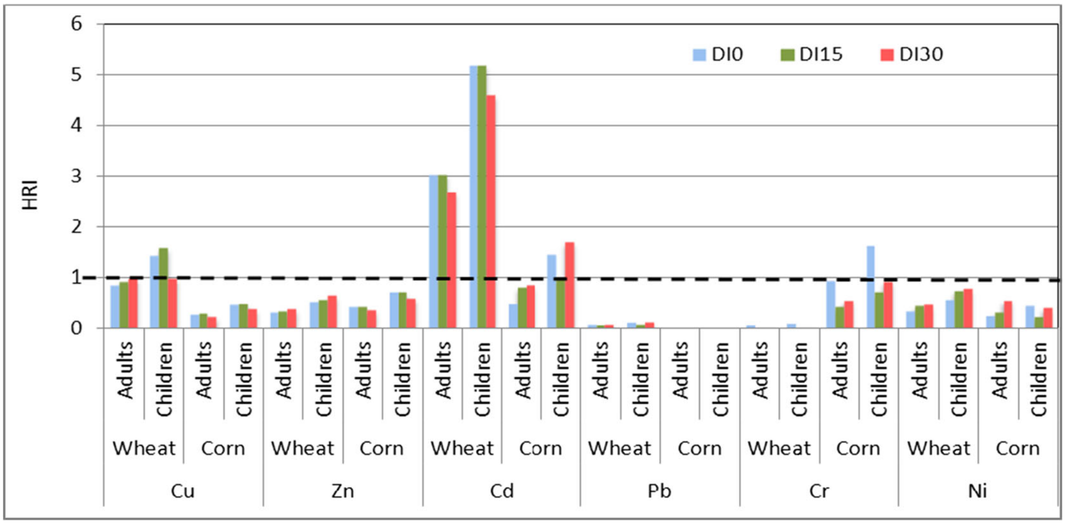

Fig. 3 The health risk index (HRI) for children and adults of heavy metals associated with wheat and corn grown in wastewater drip-irrigated soils. Emitters were located at the surface (*DI0*), 15 cm depth (*DI15*), and 30 cm depth (*DI30*). (*N*=96)

inhalation. Compared to oral intake, all other pathways are negligible. The daily intake of heavy metals was assessed according to average consumption estimations (Table 5). DIM was significantly higher when consuming food crops grown in TWW-irrigated soils. The highest intake of Cd, Cu, Ni, and Pb were observed for wheat consumption, and of Zn and Cr for corn consumption for both adults and children. For adults, the contribution of seeds to dietary intake of Cu, Zn, Cd, Pb, Cr, and Ni per individual per day was respectively 36.5, 101.7, 2.9, 0.2, 0.09, and 8.1 μg for wheat, and 10.2, 116.49, 0.7, 0.01, 1.9, and 5.3 μg for corn. For children, the related contribution per day was 53.1, 174, 4.97, 0.3, 0.17, and 0.13.8 μg for wheat, and 17.5, 199, 1.3, 0.03, 3.2, and 9 μg for corn. However, these values are still below the tolerable limits recommended by the FAO/WHO in the study area. Khan et al. (2013b) studied the effects of irrigation with TW for some crops including wheat and corn. They estimated values of DIM for both crops in the same ranges as those observed in the current study. Several studies indicate a wide variation in daily intake of trace elements via grain crop and vegetables, which often depends on variation in crop ability, as well as element content, growth period and seasonal influences (Khan et al. 2013a, b; Liu et al. 2013; Wang et al. 2013; Wongsasuluk et al. 2014; Huang et al. 2014).

The HRI of metals through the consumption of edible seeds for both adults and children is given in Fig. 3. HRI is usually adopted to assess the health risk to hazard materials in foods (Chen et al. 2011; Osman et al. 2011). HRI more than 1 is considered not safe for human health (USEPA 2000; US-EPA 2012). Results show that Cu intake through the consumption of edible wheat seeds posed a relatively high potential health risk to children, and it is also somewhat dangerous for adults, irrespective of emitter depth. For corn, HRI always remained <0.5. Values of Zn, Pb, Cr, and Ni also remained below 1 (except for Cr under corn with surface emitters). Cd, however, showed a high potential health risk, particularly for wheat, with HRI values far above 1. Similar findings have been observed in Nanning, China (Cui et al. 2004), Boolaroo, Australia (Kachenko and Singh 2006), Swat District, Pakistan (Khan et al. 2013b), and Zhejiang, China (Huang et al. 2014).

Jan et al. (2010) reported that some vegetables irrigated with TWW have high HRI of Mn, which was also observed by Singh et al. (2010) in rice and wheat grains, whereas Chen et al. (2010) reported safe HRI (<1) in edible seeds of plants grown in sewage-irrigated soils. Wang et al. (2013) studied thallium (Tl) contamination in soils and reported that HRI values were generally higher than 1, indicating that health risks associated with Tl exposure are significant and assumed to be dangerous to the health of local villagers. Pandey et al. (2012) found that HRI for Cd and Pb exceeded EPA's safe limits.

Conclusion

Several studies reported that irrigation of agricultural lands with TWW has caused a substantial buildup of heavy metals in soils compared to background values and control soils. In contrast to this, we found that irrigation of wheat and corn with TWW did not significantly elevate the concentration of heavy metals compared to the FW-irrigated soils. This could be attributed to less usage of water and higher uptake of nutrients due to higher dry matter production under drip irrigation.

The concentration of heavy metals in wheat grains were significantly higher than those of corn except for Zn, illustrating that wheat had a higher ability to take up heavy metals from soil as compared with corn. Therefore, wheat production in contaminated soil seems to lead to higher health risk for humans as indicated by the HRI values of heavy metals. It is worth to mention that crops grown in arid or semi-arid region naturally suffer from Zn and Cu deficiency. Therefore, HRI expedience of Zn and Cu in crops may not be taken as seriously as other nonessential trace elements.

Based on our results, drip irrigation system could be a potential solution for reducing the metal contamination of edibles crops irrigated with TWW and cultivated in contaminated soils. The depth of the emitters though did not play a significant role. The results suggest that more attention should be directed toward cultivation of other crops with drip irrigation system for a safe and more productive use of TWW for irrigation.

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