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Comparative analysis of heavy metal concentration in secondary treated wastewater irrigated soils cultivated by different crops

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ABSTRACT: The use of treated urban wastewater for irrigation is a relatively recent innovation in Botswana and knowledge is still limited on its impact on soil heavy metal levels. The aim of this study is to analyze and compare heavy metal concentration in secondary wastewater irrigated soils being cultivated to different crops: olive, maize, spinach and tomato in the Glen Valley near Gaborone City, Botswana. The studied crop plots have been cultivated continuously under treated wastewater irrigation for at least 3 years. Most crop farms have sandy loam, loamy sand soils. Based on food and agriculture organization, heavy metal threshold values for crop production have been studied. Results showed that the wastewater irrigated soils in the Glen Valley have higher cadmium, nickel and copper than desirable levels, while the levels of mercury, lead and zinc are lower than the maximum threshold values recommended for crop production. The control sites show that the soils are naturally high in some of these heavy metal content. Comparing between the crops, mercury and cadmium levels are highest in soils under maize and decline linearly from maize to spinach to olive to tomato and control site. By contrast, concentrations of the other metals are at their lowest in maize and then increase from maize to spinach to olive to tomato and to control site.

Keywords: Heavy metals; Maize; Olive; Spinach; Tomato; Wastewater

INTRODUCTION

In Botswana, the use of treated urban wastewater for irrigation is a relatively recent innovation. Although a number of studies has been carried out on various aspects of the system (Li *et al.*, 2001; Cornish and Kielen, 2004; Emongor and Ramolemana, 2004; Samarghandi *et al.*, 2007), knowledge is still limited on its impact on soils and the associated heavy metal accumulation in soils. Most studies on the effects of treated waste effluents on vegetable crops had been done with sewage sludge (e.g. Ngole, 2005) rather than on wastewater. Therefore, there is need for more (Kirkham, 2006). Thus, there is need for more studies focused on the benefits and limitations of the use of wastewater for irrigation (Kirkham, 2006).

Thus far, according to Emongor and Ramolemana (2004), the use of secondary treated wastewater, rather than raw effluent water, appears to have had few adverse physical, chemical, or biological effects on vegetables and fruits. This agrees with the findings of researchers in some other parts of the world (Heidarpour *et al.*,

2007; Wang *et al*, 2007a; b). Thus, these findings differ from the experience in some other parts of Africa, where the application of large volumes of partially treated or untreated wastewater has adversely affected both surface water bodies and the urban and peri-urban farmers using these water bodies as sources of irrigation (Keraita and Drechsel, 2004). Also, in their studies in the Lake Victoria Basin, Nabulo *et al.* (2008) found that heavy metals accumulated in wetlands used for wastewater disposal, the highest levels being recorded in wetlands receiving wastewater from multiple industrial sources.

In spite of the favorable results from existing studies in Botswana, there is still need for further investigation because of evidence from other parts of the world showed that there is accumulation of heavy metals such as zinc, nickel and chromium in the food chain (Oloya and Tagwira, 1996a; b) when sludge is used as a fertilizer for growing crops (Nouri, 1980). Further, the use of wastewater and sludge in agricultural lands was found to enrich soils with heavy metals to concentrations that may pose potential environmental and health risks

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in the long-term (Sánchez-Martín et al., 2007; Tabari et al., 2008). Nwuche and Ugoji (2008) found heavy metal concentrations adversely affected the biological health of the soil manifested in lower rates of nitrogen mineralization, lower soil microbial biomass carbon and reduced rate of respiration by soil microbial population. It is true that heavy metals commonly enter soils through addition of sludge (e.g from treated waste water), composts, or fertilizers (Kirkham, 2006; Kaonga et al., 2010). But the risk is still there that some residual heavy metals may get into the soil and crops through the treated wastewater itself. Inadequately, treated wastewater can pose a major risk to the physicochemical characteristics of the watershed and environments where it is disposed as found by Igbinosa and Okoh (2009) in Eastern Cape Province, South Africa. In assessing Gaborone's secondary treated sewage water crop irrigation suitability for selected physical variables, anions and metal content, Khotlhao et al., (2006) attested to the generally high quality of the secondary treated water by the low levels of all investigated metals and physical variables. However, there were significant (P = 0.05) variations in the levels of the metals, and in pH and EC, between certain time periods and at different sampling points along the Notwane River. But the issue of wastewater irrigation exceed the physical, chemical and biological quality of the wastewater and its variability. Equally important are the different types of crops being cultivated and the soils themselves. For example, it has been noted that the suitability of soils for receiving wastewaters without deterioration varies widely, depending on their infiltration capacity, permeability, cation exchange capacities and phosphorus adsorption capacity, water holding capacity, texture, structure and type of clay mineral (Ivan and Earl, 1972; Donahue et al., 1977). Sandy soils usually allow the greatest rates of water percolation but the least adsorption and sieving action. Alkaline soils will remove most heavy metals by precipitation. Rusan et al. (2007) concluded that continuous irrigation with wastewater could lead to accumulation of salts, plant nutrients and heavy metals (with the exception of Pb and Cd) in soils beyond crop tolerance levels. Also according to Nabulo et al. (2008), uptake of trace metals from soil differs from plant to plant and from site to site. Thus, there is need for sustained studies in order to fully analyze and understand the long term environmental impacts of the use of treated sewage water for crop irrigation.

This study, carried out in 2009 in the Glen Valley, near Gaborone City, capital of Botswana, focuses primarily on the effects of secondary treated wastewater irrigation on heavy metal accumulation in soils under different types of crops. Thus, the study aims to assess and compare heavy metal concentration in soils where different types of arable crops are being cultivated under secondary treated wastewater irrigation. The main hypothesis being tested is that the impact of treated wastewater on heavy metals varies with the crop type.

The study area

This study was carried out in the Glen Valley, about 10 km northeast of Gaborone City beside the Notwane River where about 234 ha of cropland are being cultivated with secondary treated wastewater. The farms lie between the Botswana Defence Force camp and the Gaborone sewage ponds between Latitudes 24°35'23.56"S and 24°37'01.14"S and between Longitudes 25°58'43.29"E and 25°5816.74"E. There are 47 different farms, varying in size from 1 to 10 habeing managed by private farmers raising a wide variety of arable crops. In addition, a government agency, the national master plan for arable agriculture and dairy development (NAMPAAD) is running a 13 ha farm for demonstration purposes to develop and introduce new technologies (mainly olive, alfalfa/Lucerne) to the local farmers. The crops cultivated under waste water irrigation in the Glen valley include tomatoes, spinach, okra, maize, cabbage, olive, Lucerne, butternuts, and green pepper. The variety of operators and crop types provides a good opportunity for assessing the impact of different management systems on soil quality in the Glen Valley. The soils are predominantly sandy loam to sand occurring in an alluvial-cum-colluvial landscape, with patches of vertisolic clayey materials alternating with areas of more sandy and, even, gravelly deposits. The soils, mapped on a scale 1:20 000, are classified as luvisols, lixisols, cambisols, calcisols, regosols and arenosols (FAO, 1988) but they are all texturally very similar irrespective of taxonomic classification.

Gaborone secondary treated wastewater

Detailed analysis of the physico-chemical properties of the Gaborone secondary treated wastewater indicate that the treated wastewater is of high quality and suitable for unrestricted irrigation of horticultural crops based on the EC, SAR, Cl, NaCl, faecal coliforms, available plant nutrients and the low concentration of heavy metals (Pb, Cr, Cu, Co, Cd, As and Se (Emongor *et al.*, 2005). The treated wastewater has the following physico-chemical properties: EC, 0.51 dS/m, pH 9.08, total dissolved solids, 358 mg/L, total alkalinity, 234 mg/L as CaCO₃, Cl 70 mg/L, NaCl 115 mg/L, sodium adsorption ratio (SAR), 2.26, NO₃ 5.56 mg/L, NH₄ 0.3 mg/L, NO₂ 0.0184 mg/L, total Fe 0.623 mg/L and faecal coliforms, 5/100 mL.

MATERIALS AND METHODS

Farm selection and soil sampling

The study was conducted in farms that had been cultivated under irrigation continuously for at least 3 years to give a sufficient long period for the impact of irrigation to begin to show. Four types of crops most widely grown in the Glen Valley for sale in the urban market were selected for study, namely, olive, maize, spinach and tomato. Five sampling points were systematically selected on each farm with sampling points selected along crop planted rows at specified intervals of 20 cm on each farm plot. In addition, a control site was selected for soil sampling (5 replicates) in the neighborhood of the Glen Valley farms. Given the shallow rooting of the crops, soil sampling was limited to the top 30 cm of the soil and the soils were sampled at 0-15 cm and 15-30 cm depths. Altogether, 10 soil samples were collected on each farm and control site giving a total of 30 samples per crop type and control site.

Soil analysis and heavy metal analysis

The soil samples from the crop farms and the control site were analyzed for basic soil characteristics, including particle size distribution (sand %, silt %, clay %), pH, electrical conductivity, soil organic matter, exchangeable bases (Ca++, Mg++, K⁺, Na⁺), exchange acidity, trace elements, Mn, Fe, Al and cation-exchange capacity (CEC), using laboratory standard methods (Breitbart, 1988; van Reeuwijk, 1993). However, cost considerations lead to the restriction of the heavy metal analyses to a smaller number of soil samples. Fifty two soil samples were analyzed for lead (Pb), mercury (Hg), cadmium (Cd), nickel (Ni), zinc (Zn) and copper (Cu). Forty eight samples came from two soil depths at two sampling points per farm for each of the four crop types, while the remaining 4 samples came from the two control sites (one sampling point per control site). Fine sized particles (particle size < 0.053 mm) samples were prepared to measure heavy metals using AAS (atomic absorption spectrometer), ICP-MS (inductively coupled plasm-mass spectrometer) and GFAAS (graphite furnace atomic absorption spectrometry).

Data analysis

The values for each parameter were averaged for each soil depth (0-15 cm; 15-30 cm) for all the five sampling points on each crop farm. These average values are displayed on a series of multiple line graphs for purposes

Table 1: Basic soil characteristics for surface soils under different crops irrigated with treated secondary wastewater

Cron form	Class	C:14	Cand		EC	00	CEC	Ca	Ma	V	Na
Crop farm	Clay	SIII	Sand	рп-	EC	UC	CEC	Ca	Mg	<u>к</u>	INa
	(%)	(%)	(%)	$CaCl_2$	(µs/cm)	(%)	(cmol/kg)		Exchange	able bases	
									(meq/10	0 g soil)	
Maize Crop											
Farm 1	11.5	9.7	78.8	5.94	90.5	0.69	12.27	23.22	4.70	7.07	16.00
Farm 2	9.4	7.8	82.8	5.92	106.5	0.98	7.33	18.24	3.03	4.46	9.22
Farm 3	31.8	14.5	53.7	6.96	144.9	1.22	4.44	44.88	10.21	6.61	7.61
Spinach Crop	2										
Farm 1	9.1	10.6	80.4	6.09	373.6	2.57	14.60	35.46	5.71	6.26	14.11
Farm 2	7.7	15.2	77.1	6.22	287.9	2.67	12.25	47.52	8.03	8.78	19.07
Farm 3	31.8	15.9	52.3	6.24	288.4	2.57	31.90	39.42	5.93	8.84	7.55
Olive Crop											
Farm 1	28.2	14.7	57.1	6.23	158.7	1.72	23.76	21.42	3.31	11.83	11.25
Farm 2	34.4	15.2	50.4	6.45	203.0	1.76	9.73	26.10	3.79	7.61	16.14
Farm 3	31.8	18.2	50.0	7.17	257.8	2.06	19.33	62.28	8.81	10.76	5.90
Tomato Crop)										
Farm 1	20.7	14.3	65.0	5.97	215.3	2.67	10.41	25.02	3.88	7.34	6.64
Farm 2	14.8	15.1	70.1	6.79	198.9	2.57	10.86	35.10	3.65	9.18	4.41
Farm 3	35.5	16.4	48.1	6.96	293.2	2.68	4.57	62.40	7.64	8.54	6.19
Control	31.1	12.3	56.6	5.55	105.1	1.47	9.21	36.6	5.47	7.19	2.95

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of direct comparison among and between crop types. The data are tabulated in Table 1. Also correlation analysis has been carried to show the relationship between some of the heavy metals and trace elements.

RESULTS AND DISCUSSION

Characterization of soils

Surface soil texture for most crop farms is characterized as loamy sand to sandy loam but there are also sandy clay loams particularly on the olive farms and on one maize and one spinach farm plot where the clay fraction is on average over 30 %. The soil is sandy clay on one of the tomato farms with a clay content of over 35 %. The soils of the control site also are sandy clay loams. Overall, the crop types are similar in the range of soil textural classes displayed and provide a basis for comparison of soil quality parameters between them. There is little variation in soil pH between the soils under irrigation (Table 1). But, the pH values indicate the soils to be generally slightly acidic to neutral in reaction. Almost everywhere, soil pH is higher on all the irrigated crop fields than in the control sites, the highest values being recorded on the spinach and tomato plots. Soil pH values are lowest on the olive plots and, except in one case, maize plots. Altogether, treated wastewater irrigation has had a positive impact on soil pH. Soil pH is important because it influences the availability and plant uptake of micronutrients including heavy metals (e.g. Kirkham, 2006).

Heavy metals

Table 2 gives the FAO threshold values for soil trace elements for crop production. The heavy metal and trace element concentrations in the treated wastewater irrigated soils in the Glen Valley (Figs. 1-4) may be compared with these threshold values. The first observation to make from a comparison of the soils of the irrigated plots and control sites is that it appears that the Glen Valley soils are naturally high in some of these heavy metal trace elements and that crop cultivation under wastewater irrigation has actually lowered the trace element content of the soils. This is true for the example of Cu, Zn, Ni and Pb. It is only in respect of Hg and Cd that the control site soils have lower concentration levels than the soils under cultivation. The somewhat high heavy metal trace element content of the soils may be due to their colluvial-cum-alluvial origin and the imperfectly drained ground conditions experienced during periods of heavy rainfall.

The comparatively low soil trace element levels on the cultivated plots relative to the control site soils, with the exception of Cd and Hg, might suggest that, perhaps, plant uptake of some of these trace elements under irrigation has been high to the extent that their levels in the soils have been significantly lowered. On the other hand, the higher levels of Cd and Hg in the cultivated soils relative to the levels in the control sites might suggest that crop uptake of these toxic elements is presently not high although Kirkham (2006) has noted

Element	Recommended maximum concentration (mg/L)	Remarks
Al	5.0	Can cause non-productivity in acid soils (pH \leq 5.5), but more alkaline soils at pH \geq 7.0 will precipitate the ion and eliminate any toxicity.
Cd	0.01	Toxic to beans, beets and turnips at concentrations as low as 0.1 mg/L in nutrient solutions. Conservative limits recommended due to its potential for accumulation in plants and soils to concentrations that may be harmful to humans.
Cu	0.20	Toxic to a number of plants at 0.1 to 1.0 mg/L in nutrient solutions.
Fe	5.0	Not toxic to plants in aerated soils, but can contribute to soil acidification and loss of availability of essential phosphorus and molybdenum. Overhead sprinkling may result in unsightly deposits on plants, equipment and buildings.
Mn	0.20	Toxic to a number of crops at a few-tenths to a few mg/L, but usually only in acid soils.
Ni	0.20	Toxic to a number of plants at 0.5 mg/L to 1.0 mg/L; reduced toxicity at neutral or alkaline pH.
Pb	5.0	Can inhibit plant cell growth at very high concentrations.
Zn	2.0	Toxic to many plants at widely varying concentrations; reduced toxicity at $pH > 6.0$ and in fine textured or organic soils.
Hg		

Table 2: Recommended maximum levels of trace elements for crop production (FAO, 1985)

Source: Wastewater quality guidelines for agricultural use series title: FAO irrigation and drainage papers - 47 1992 T0551/E



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Fig. 4: Mn, Fe and Al concentration in treated waste irrigated soils under different crops

that elevated levels of Cd in plants are being reported in recent studies. But, the high Cd and Hg levels might also suggest that secondary wastewater is causing a buildup of soil cadmium and mercury levels on the cultivated plots. In particular, Cd has a relatively higher adsorption capacity to clays than other heavy metals (Sanchez *et al.*, 1999) and higher mobility index (Sánchez-Martín *et al.*, 2007).

The second major observation that might be made is that, based on FAO (1985) recommended maximum levels of trace elements for crop production, the wastewater irrigated soils in the Glen Valley have higher than the recommended levels of Cd (e"0.01), Ni (e"0.20) and Cu (e"0.20) while the levels of Hg, Pb and Zn are lower than the maximum threshold values recommended for crop production. According to Kirkham (2006), Cd is a heavy metal that is of great concern in the environment, because of its toxicity to animals and humans. Moreover, with long-term use of sewage sludge, heavy metals can accumulate to phytotoxic levels and result in reduced plants growth and/or enhanced metal concentrations in plants, which consumed by animals then enter the food chain (Behbahaninia and Mirbagheri, 2008). The Cd, Ni and Cu levels recorded in the Glen Valley soils are close to or higher than the toxic levels for crops; these levels can become problematic at low pH values. According to Barancikova *et al.* (2004), soil pH and organic matter are the most critical factors in controlling Cd availability and plant uptake. Low pH favors accumulation of Cd in the soil (Kirkham, 2006).

Comparing between the crops, Cd and Hg levels are highest in soils under maize and decline linearly from maize to spinach to olive to tomato and control

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Cd (mg/L)		Hg (mg/L)		Pb (mg/L)		Ni (mg/L)		Zn (mg/L)		Cu (mg/L)	
L1	L2										
1.00	0.95*	0.92*	0.94*	-0.59*	0.79*	-0.25	-0.41	-0.22	-0.12	-0.59*	-0.64*
	1.00	0.90*	0.93*	-0.56*	0.79*	-0.33	-0.28	-0.22	0.06	-0.61*	-0.58*
		1.00	0.84*	-0.66*	0.76*	-0.34	-0.38	-0.04	-0.15	-0.63*	-0.48
			1.00	-0.47	0.81*	-0.41	-0.49	-0.26	-0.02	-0.65*	-0.79*
				1.00	0.66*	0.32	0.40	0.22	0.28	0.52	0.34
					1.00	0.58*	0.42	0.29	0.14	0.62*	0.58*
						1.00	0.60*	0.22	-0.03	0.67*	0.49
							1.00	0.42	0.43	0.64*	0.81*
								1.00	0.21	0.58*	0.59*
									1.00	0.09	0.23
										1.00	0.72*
											1.00

Table 3a: Correlation matrix of soil heavy metal concentration levels

*Figs. in bold show r values significant at p < 0.05, L1, L2 = 0-15 cm, 15-30 cm soil depths, respectively.

Table 3b: Correlation matrix of soil trace elements Al, Fe, Mn

Mn (r	ng/L)	Fe (1	mg/L)	Al	(mg/L)
0-1 5 cm	15-30 cm	0-1 5 cm	15-30 cm	0-1 5 cm	15-30 cm
1.00	0.83*	0.13	-0.18	0.10	-0.02
	1.00	0.09	0.03	-0.05	0.16
		1.00	0.14	0.27	-0.16
			1.00	-0.38	0.45
				1.00	0.15
					1.00

*Significant at p<0.05

site (Figs 1-3). The pattern for the other heavy metals is broadly in the reverse order, with the lowest values being recorded in maize and then rising through spinach to olive, tomato to the control site soils. Therefore, broadly speaking there is an apparent inverse relationship between the concentration of Cd and Hg on the one hand and of all the other heavy metals on the other.

Table 3a shows the correlation matrix between the heavy metal concentration levels. The highest positive correlation is between Hg and Cd while the two elements in turn show strong negative correlations with top layer Pb and both top and subsoil layer Cu. But, in the subsoil layer Pb is positively correlated with both Cd and Hg. Cu is correlated with most other elements at least in one soil layer with the exception of Zn. Zn is the only element that is not significantly correlated with any other element at any of the two soil depths. Ni also only shows significant positive correlation with Pb in the subsoil layer. With the exception of Zn, all elements show strong positive correlation levels between the top and subsoil layers.

Micronutrients

In general, wastewater irrigation appears to have raised the Al saturation levels of the soils under cultivation while lowering the Fe and Mn levels (Fig. 4). At low soil pH, the growth of certain plants which are highly sensitive to Al toxicity is greatly affected. Mn toxicity in addition may adversely cause leaf distortion, vellowing and necrosis in some plants. Fortunately, thus far, acidity levels in the soils under study are low to moderate. Al saturation levels exceed the threshold maximum of 20 mg/kg only on four of the farms (2 tomato, 1 olive and 1 spinach). Al levels are highest in the topsoils under olive and spinach (about 10-32 cmols/kg) while they are also high in the subsoils under tomato, olive and spinach. Al levels under maize are about the same as in the control site soils (5-10 cmol/kg).

Fe and Mn saturation levels vary widely between and among crop types but in general the highest levels are found under olive, tomato and spinach in that order. Similarly, Lone *et al.* (2003) reported higher accumulation of heavy metals and micronutrients in spinach than in other crops (e.g okra). Fe and Mn

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saturation levels are everywhere well below the recommended maximum levels for crop production as indicated in Table 2 above. Table 3b indicates very low correlation between the trace elements at both soil depths with the singular exception of Mn. Also, there is almost no relationship between these trace elements and the heavy metals. The only significant correlations are between Mn layer 1 and Hg layer 2 (-0.56) and between Fe layer 2 and Cu Layer 1 (+0.56).

CONCLUSION

Rusan et al. (2007) rightly noted that there is inconsistency in research findings on the impact of wastewater irrigation on soil micronutrients and heavy metals. However, results in this study tally with the findings of some previous studies, including: 1) the build-up of heavy metals such as Cd, Hg, Ni and Pb and micronutrients such as Cu, Fe, Mn and Zn in the upper layers of the soil or sediment cores (Mancino and Pepper, 1992; Rusan et al, 2007; Harikumar et al, 2009); 2) increases in soil Al, Fe and Mn with wastewater irrigation (Mohammad and Mazahreh, 2003); 3) variable and inconsistent responses of soil micronutrients under different crop types; and 4) the significance of soil textural differences in influencing the accumulation of heavy metals and micronutrients in soils under wastewater irrigation (Ivan and Earl, 1972; Donahue et al., 1977).

The Glen Valley soils under treated wastewater irrigation crop farming have higher heavy metal quality than the non-irrigated control site soils. For example, the soils under irrigation have lower concentrations of Cu, Zn, Ni and Pb than the control site soils. It is only in respect of Hg and Cd that the control site soils have lower concentration levels than the soils under cultivation. However, the concentration levels for almost all the metals analyzed are higher or close to the maximum threshold values for crop production recommended by FAO.

Altogether, the picture that has emerged from the pattern of heavy metal concentration in the soils is that the risk of heavy metal uptake by wastewater irrigated crops is high even if it is not already taking place. Unfortunately, this study did not analyze the trace element contents of the crops being grown under treated wastewater irrigation in the Glen Valley. It would be worthwhile doing a comprehensive analysis of both soil and crop trace element levels to assess the risk posed to both human and animal health. At present, favorable soil pH levels, which range from slightly alkaline to slightly acidic, are probably serving as buffer against any heavy metal or trace element toxicity on these treated wastewater irrigated plots. But, the potential is there for the buildup of heavy metals and other trace element saturation levels unless there is careful management of soil acidity and the wastewater irrigation. This is the more so as the soils in the Glen Valley appear to have comparatively high levels of many of the trace elements.

In terms of crops, maize cultivation appears to hold the greatest risk for Cd and Hg pollution of the soil under treated wastewater irrigation. In both elements, the comparative levels in soils under the various crops are in the order maize > spinach > olive > tomato > control soil. But the good side of this is that it may well be that maize uptake of these elements is low compared to other crops. The downside is that any other crops planted on the same plots after maize may suffer the toxic effects of these trace elements. This is important because it was discovered that most Glen Valley farmers plant different crops on their plots at different periods of the year. Thus, a plot planted to tomatoes might be sown to spinach or maize later on. This crop rotation on the irrigated plots might be a basis for managing the treated wastewater irrigation system in the area to minimize the risk of trace element toxicity to crops and humans. Kirkham (2006) has noted that the best solution to trace element toxicity e.g. cadmium, is to remove the sources of the trace elements in the environment. This is impossible at the moment but something can be done about the quality of the treated wastewater being used for irrigation to ensure that much of the heavy metals and other trace elements are removed before its use for irrigation.

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