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Influence of wastewater irrigation on soil chemical properties and buildup of heavy metals in soil

Amina Kalsom*[1], Abdul Kareem[2], Ana Aslam[1], Abid Niaz[1], Nisa Mukhtar[1], Aamer Sattar[1], Raheela Naz[1], Asifa Naz[1], Farah Rasheed[1], Arfan Rasheed[1], Sadia Sultana[1], Muhammad Aftab[1] , Ehsan ul Haq[3]

1 Soil Chemistry Section, Ayub Agricultural Research Institute, Faisalabad, Pakistan
2 Institute of Soil and Environmental Sciences, University of Agriculture, Faisalabad, Pakistan
3 Institute of Soil Chemistry and Environmental Sciences, Kala Shah Kaku, Pakistan

* Corresponding author e-mail: amina.kalsom.aari@punjab.gov.pk

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Abstract
The contamination of soil and plants by heavy metals is one of the crucial issues to be faced in the world including Pakistan and requires attention because heavy metals above their normal ranges are highly threatened to both plants and animals. To monitor this situation, a survey study was conducted to diagnose the concentration of toxic metals and other chemical properties of wastewater irrigated soils. For this purpose, soil samples were collected from various districts of Punjab, Pakistan and analyzed for heavy metals (cadmium, lead, chromium, nickel) and chemical properties such as electric conductivity, pH, organic matter and macronutrients (Nitrogen, Phosphorus, Potassium and Sodium contents). Result showed that pH of wastewater irrigated surface soil was alkaline in nature. Results also revealed that use of wastewater irrigation improved the O.M content (upto 2%) in soils. Macronutrient dynamics also increased specially build up in total N up to 0.12%, Olsen P (45 mg kg^{-1}), Extractable K and Na (608, 1525 mg kg^{-1} respectively) was also observed in soils of wastewater irrigated areas. Furthermore, results also indicated that the concentration of heavy metal, cadmium was ranged from 0.004-3.44 mg kg^{-1}, lead 0.05-279.3 mg kg^{-1}, chromium 0.004-683 mg kg^{-1} and nickel 0.01-221.7 mg kg^{-1} in wastewater irrigated areas of Punjab. It was observed that, 28, 19, 15 and 5% samples were found above the permissible limits (35, 100, 0.8 and 85 mg kg^{-1}) of Ni, Cr, Cd and Pb, respectively. Based on the results, the order of heavy metal contamination in the wastewater irrigated soil of Punjab occurred as Ni>Cr>Cd>Pb. This study concluded that untreated wastewater affect chemical properties of soil as well as degrades soil quality and at the same time it's continues use ultimately responsible for buildup of heavy metals in soils.

Keywords
Soil contamination, Heavy metals, Chemical characteristics, Effluents

Introduction

In the developing countries, demand for a huge quantity of water for domestic, municipal and industrial purpose is increasing due to rapidly growing population. With the increasing demand of water, the usage of waste water for irrigation has been enhancing particularly in the arid and semi-arid regions of the world. In most regions of Pakistan sewage and industrial wastewater is used to irrigate the crops, which is a rich source of heavy metals (Shakoor *et al.*, 2016). All the factories are discharging large amount of unprocessed effluents into the drains. Since amount of good quality of water for agriculture is reducing and farmers are using this polluted water for crops production, especially vegetables (Farid *et al.*, 2015). Farmers think that wastewater is a reliable source of nutrients, substitute of good quality water and good source of irrigation round the clock. Wastewater

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of different factories may differ in composition based on the source of production (Al Zabir *at al.*, 2016). It may contain essential nutrients and some harmful substances.

In agriculture sector the waste water irrigation affects soil characteristics and crop production. The use of raw city effluent for irrigating agricultural soil causes a number of potential advantageous changes like an increase in organic carbon, available nitrogen, phosphorus and potassium concentration (Kordlaghari, 2013). The macro and micronutrients present in wastewater can enhance soil fertility, but eutrophication occur when soil irrigated by industrial water over a long period of time.

On the other hand, untreated waste water discharge contains large amount of health hazard elements such as heavy metals, organic contaminants, pathogens and salts (Farid *et al.*, 2015). These substances have deleterious impact on the productivity of soils, plants, animals and the whole environment if exceed permissible limits (Mapanda *et al.*, 2005; Farid *et al.*, 2015). Even in low concentration, heavy metals persist for longer period in soil from where they enter into food chain via plant uptake (Farid *et al.*, 2015). Heavy metals are hazardous for different crops due to their non-biodegradability, bio-availability and toxicity effect (Mahler, 2003). The presence of heavy metals can influence the soil ecology, agricultural crop productions, product value and quality of ground water that will ultimately damage health of living organism through entries into food chain (Nazir *et al.*, 2015). Heavy metals caused various disabilities in humans including cancer, injury of kidneys, nervous system, skin, and bones, cardiovascular diseases, cognitive impairment and chronic anemia (Jarup, 2003). Many analyzers reported that concentration of heavy metals increased in to the soil due to continuous irrigation with wastewater (El-Nennah *et al.*, 1982). Ultimately, increasing the concentration of heavy metal in soil also enhance the uptake of heavy metals via plants based on the type of soil, plant species and growth stages (Farooq *et al.*, 2008; Khan *et al.*, 2011). Another possible risk to soil is the salt accumulation caused by addition of wastewater. Continued irrigation with waste water in semiarid and arid climate condition, could cause secondary soil salinity to levels that will inhibit the crop growth (Murtaza *et al.* 2010) and degrade the soil profile (Duan *et al.* 2009).

Keeping in sight the above aspects, a survey study was conducted to discuss the chemical characteristics and to quantify the buildup of heavy metals (Cd, Pb, Cr & Ni) of wastewater irrigated soils of various districts of Punjab.

Materials and Methods

Collection and preparation of samples

A survey study was planned and carried during 2017-2019 in Soil Chemistry Section, Ayub Agricultural Research Institute, Faisalabad, Pakistan. For samples collection, different districts of Punjab including Gujranwala, Faisalabad, Lahore, Sargodha, Bahawalpur, Rawalpindi, Sahiwal and Multan were selected which are being irrigated with wastewater from 8-10 years. Ten waste water and ninety soil samples were collected from different sites of each district.

Soil samples were dried, ground, sieved and stored for analysis. These samples were analyzed for soil properties i.e., pH_s, electric conductivity (Richards, 1954), and organic matter (Nelson and Sommers, 1982). Fertility status of soil was determined by analyzing total nitrogen (Bremner, 1996), Olsen P (Olsen, 1954), extractable K, Na (Richards, 1954). For the determination of heavy metals (Cd, Pb, Cr & Ni), waste water and soil samples were digested with triacid mixture (H_2SO_4: $HClO_4$: HNO_3) (HF: HCl: $HClO_4$) respectively (Estefan *et al.*, 2013) and analyzed on Atomic Absorption Spectrophotometer (Model: AA-7000). The data were subjected to simple means with standard deviation for comparison.

Results and Discussion

Results present in Table 1 showed the concentration of heavy metals (Cd, Pb, Cr, Ni) in the samples of waste water used for irrigation in various districts of Punjab. It was found that the heavy metals (Cd, Pb, Cr & Ni) contents in wastewater were beyond the maximum permissible limits set by WHO (2008). Results showed that the heavy metals content in the waste water is beyond the normal value but it is highly threatened for soil quality and human consumption (Nazir *et al.*, 2015).

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Districts	Cadmium	Lead	Chromium	Nickel
	(mg L^{-1})			
	Range	Range	Range	Range
Gujranwala	0.000-0.036	0.000-0.025	0.000-0.429	0.008-0.024
Faisalabad	0.000-0.024	0.000-0.228	0.000-0.020	0.000-0.209
Lahore	0.002-0.028	0.000-0.372	0.071-2.969	0.125-0.028
Sargodha	0.004-0.015	0.000-0.049	0.000-2.759	0.000-0.122
Bahawalpur	0.000-0.021	0.041-0.221	0.000-0.427	0.000-0.210
Rawalpindi	0.000-0.010	0.000-0.165	0.000-0.571	0.034-0.513
Sahiwal	0.006-0.312	0.025-0.174	0.000-3.703	0.000-0.665
Multan	0.000-0.016	0.028-0.330	0.000-0.577	0.000-0.282

Table 1 *Heavy Metal (Cd, Pb, Cr, Ni) contents in waste water*

Chemical properties of wastewater irrigated soil

Results for pH_s, EC_e (electrical conductivity) and organic matter content of collected soil samples readings are shown in Table 2. Analysis showed alkaline pH_s range from 6.4 to 8.3 in surface soil. It represents the number of samples lies in low, medium and high pH range. Mean values are within the acceptable value of WHO and FAO stand for irrigation purposes (WHO, 2008). The standard limit of pH for irrigation water ranges from 6.0 to 9.0 (Singh *et al.*, 2009; Chaoua *et al.*, 2018). Soil pH directly influences the growth of plants by affecting the nutrients availability in the soil (FAO, 2003). Low pH values is possibly due to the existence of acidic constituents in wastewater which transform to acidic compounds which lead to limiting pH value (Muamar *et al.*, 2014). High pH values support the precipitation and sorption of vast range of metals (Mehmood *et al.*, 2019; Ma *et al.* 2015).

Districts	pH_s		Electrical conductivity EC_e (dS m^{-1})		Organic Matter OM (%)	
	Range	Av.± S.D	Range	Av. ± S.D	Range	Av.± S.D
Gujranwala	6.40-7.50	7.05 ± 0.24	0.91-7.02	2.75 ± 1.30	0.69-2.86	1.59 ± 0.58
Faisalabad	7.01-8.00	7.51 ± 0.26	2.22-14.9	8.21 ± 4.01	0.66-2.14	1.18 ± 0.33
Lahore	6.79-8.10	7.25 ± 0.30	1.01-10.7	4.16 ± 2.10	0.38-2.79	1.29 ± 0.38
Sargodha	7.00-8.10	7.38 ± 0.25	1.86-12.4	5.51 ± 2.35	0.52-2.04	1.07 ± 0.32
Bahawalpur	6.70-7.50	7.22 ± 0.18	2.22-14.2	7.00 ± 3.23	0.41-2.28	1.02 ± 0.31
Rawalpindi	6.90-7.80	7.38 ± 0.23	0.54-3.64	1.55 ± 0.70	0.17-2.21	1.15 ± 0.39
Sahiwal	6.90-8.30	7.43 ± 0.28	1.80-12.9	5.81 ± 3.04	0.45-2.59	1.15 ± 0.50
Multan	7.10-7.90	7.44 ± 0.18	2.00-12.6	5.51 ± 2.47	0.41-1.21	0.93 ± 0.16

Av. = average S.D = standard deviation

Table 2. *Chemical properties (pH_s, EC_e and OM) of waste water irrigated surface soils*

The same table represents the increase in EC_e of the soil. EC_e ranged from a minimum of 0.54 dS m^{-1} at Rawalpindi to a maximum of 14.9 dS m^{-1} at Faisalabad in wastewater irrigated surface soil. Averages of all district samples have higher EC (> 2.5 dS m^{-1}) except Rawalpindi that is injurious to most crops. EC_e is related to the quantity of dissolved minerals in water and higher value of EC_e showed the presence of contaminants alike sodium, chloride or sulphate (Nazir *et al.*, 2015) but above permissible limit EC_e describe the existence of salinity which is the most vital indicator respecting to fields irrigated with wastewater (FAO, 2003). Enhance in conductivity had shown to increase the solubility of heavy metals, hence causing more availability of metals to the plant from the soil (Singh *et al.*, 2009; Chaoua *et al.*, 2018).

Similarly soil organic matter content increase (up to 2.86%) in surface soil of Punjab over a continue irrigation with wastewater. Maximum concentration of organic matter (1.59%) was also observed at Gujranwala

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and minimum concentration (0.93%) was at Multan in 0-15 cm soil depth. The organic matter is considered as a vital ingredient of soil fertility due to its vital role in physical, chemical and biological activities to supply the nutrients to the plants and also support soil to keep the moisture (Muamar *et al.*, 2014). Wastewater irrigation enhanced soil organic carbon content, which have resulted in increase in organic complexing molecules of low molecular weight, which serve as transporter of heavy metals and enhanced their uptake (Chen and Aviad, 1990; Chaoua *et al.,* 2018).

Total nitrogen, Olsen P & Extractable K, Na

Data regarding fertility status of soil i.e. total nitrogen (N), Olsen phosphorus (P), ext. potassium (K) and sodium (Na) presented in Table 3. Results indicate that total nitrogen ranged from 0.009 to 0.12% in different districts of Punjab. Maximum N contents were observed in district Gujranwala. On average basis, soils of all districts had less than 0.1% of total N. Although, FAO suggestions reported that soil has less than 0.1% of total N is poor soil (Muamar *et al.*, 2014). However, P content increased in surface soil with waste water irrigation. On average basis, maximum concentration (36 mg kg^{-1}) was observed at Sargodha and minimum (28 mg kg^{-1}) was at Faisalabad. It is known that N and P are considered as the important macro nutrients that has direct effect on crop growth and productivity.

Districts	Total N (%)		Olsen P (mg kg^{-1})		Ext. K (mg kg^{-1})		Ext. Na (mg kg^{-1})	
	Range	Av.± S.D	Range	Av.± S.D	Range	Av.± S.D	Range	Av.± S.D
Gujranwala	0.034-0.12	0.080±0.03	8-42	32±8.58	126-538	344±86.85	114-384	211±63.16
Faisalabad	0.033-0.10	0.059±0.02	4-40	28±9.79	154-608	389±98.65	153-1381	596±256.3
Lahore	0.019-0.10	0.051±0.02	7.5-39	29±9.70	79-416	245±94.90	86-1461	265±51.85
Sargodha	0.026-0.10	0.054±0.02	21-45	36±6.49	185-533	341±83.19	140-451	261±76.67
Bahawalpur	0.021-0.11	0.051±0.02	21-44	35±6.26	263-489	382±62.73	137-1525	497±248.1
Rawalpindi	0.009-0.11	0.057±0.02	19-38	29±5.16	206-438	354±60.87	92-192	157±22.32
Sahiwal	0.022-0.09	0.057±0.03	16-39	32±5.92	244-493	376±74.56	210-442	292±53.27
Multan	0.021-0.06	0.047±0.01	14-41	32±6.47	147-410	301±74.32	140-451	261±76.40

Table 3. NPK contents of waste water irrigated surface soil

Extractable K ranged from 79 mg kg^{-1} at Lahore to 608 mg kg^{-1} at Faisalabad in the wastewater irrigated surface soil. Data (Table 3) also indicated that there was no deficiency of K in surface soil. It is said that K usually required for agricultural crop production would be supplied by the wastewater (Pescod, 1992; Muamar *et al.*, 2014).

Results also revealed that the mean maximum concentration of Na in soil irrigated with wastewater was 596 mg kg^{-1} in Gunjranwala district. Na is one of most concerned among the specific noxious ions. It is described that Na directly influences the availability of water to crop and causes physicochemical changes in to the soil, especially to soil structure. It has the potential to disseminate soil hence leading to reducing permeability, decreased shear strength and enhanced compressibility (Pescod, 1992; Muamar *et al.*, 2014).

Heavy metal concentration in soil

Cadmium (Cd) concentration in soil. The results showed that the Cd level in soil ranged from 0.004-3.44 mg kg^{-1} in 0-15 cm (Table 4). The highest concentration of Cd (3.44 mg kg^{-1}) was observed at Gujranwala while lowest Cd concentration (0.004 mg kg^{-1}) was at Rawalpindi in 0-15 cm soil depth. Overall, the concentration of Cd in the soil samples collected from the Gujranwala district was recorded above the WHO recommended permissible limit >0.8 mg kg^{-1}. Cd is highly fatal even at low concentration. It causes learning abnormalities and hyperactivity in children (Shah *et al.*, 2013; Hunt, 2003).

Lead (Pb) concentration in soil. Analysis revealed that Pb content in soil samples was found between the ranges of 0.05 to 279.28 mg kg^{-1} (Table 4). The highest concentration of Pb (279.3 mg kg^{-1}) was observed at Gujranwala while lowest concentration of Pb (0.05 mg kg^{-1}) was observed at Sahiwal. On average basis, concentration of Pb in all the collected soil samples was recorded below the acceptable limit set by WHO. Pb enters in to the human body by uptake of food (65%), water (20%) and air (15%) (Nazir *et al.*, 2015). Pb at

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high concentration cause several disorders in human body including anemia, headache, damage of brain and central nervous system (Rehman *et al.*, 2013; Shah *et al.*, 2013).

Districts	Cadmium (mg kg^{-1})		Lead (mg kg^{-1})		Chromium (mg kg^{-1})		Nickel (mg kg^{-1})	
	Range	Av.± S.D	Range	Av.± S.D	Range	Av.± S.D	Range	Av.± S.D
Gujranwala	0.05-3.44	1.17±0.9	0.82-279.3	51.82±62.9	0.04-683	95.4±177	0.18-221.7	47.8±57.9
Faisalabad	0.01-0.77	0.12±03	0.67-169.7	21.05±33.3	0.01-184	31.6±57.9	0.03-91.21	25.1±57.9
Lahore	0.01-0.79	0.13±0.2	0.65-38.14	8.38±8.3	0.004-194.8	26.9±48.5	0.01-69.45	18.5±24.7
Sargodha	0.02-0.96	0.21-0.3	1.08-66.66	14.53±1.1	0.008-216.9	45.1±63.0	0.03-100.1	16.9±29.4
Bahawalpur	0.013-1.35	0.24±0.4	1.44±46.56	12.24±11.2	0.01-241.1	47.3±60.4	0.06-61.88	14.6±16.3
Rawalpindi	0.004-0.83	0.16±0.2	0.63-755.5	20.59±96.4	0.005-198.7	24.1±45.4	0.06-70.1	9.67±20.2
Sahiwal	0.006-1.14	0.08±0.26	0.05-48.04	13.33±11.9	0.004-230.9	41.9±57.6	0.11-95.16	22.4±26.1
Multan	0.009-1.66	0.45±0.6	0.59-61.91	12.48±18.2	0.008-188.7	42.3±63.8	0.02-52.05	12.4±17.4

Table 4. Concentration of Heavy Metals (Cd, Pb, Cr & Ni) in waste water irrigated soils

Chromium (Cr) concentration in soil. Analytical data (Table 4) showed that the concentration of Cr in wastewater irrigated soil samples ranged between 0.004 to 683 mg kg^{-1}. The highest concentration of Cr (683 mg kg^{-1}) was observed at Gujranwala. At high level it is toxic and health hazard (Chishti *et al.*, 2011; Shah *et al.*, 2013). WHO recommended limit of Cr in soil is 100 mg kg^{-1} (Table 5).

Nickel (Ni) concentration in soil. The experimental data showed that in the wastewater irrigated soil samples, Ni concentration occurred in a range of 0.01-221.7 mg kg^{-1} in 0-15 cm depth. The highest concentration of Ni (221.7 mg kg^{-1}) was observed at Gujranwala while lowest concentration of Ni (0.01 mg kg^{-1}) was found at Lahore. On average basis, the concentration of Ni in the soil samples collected from the Gujranwala district was recorded above the WHO acceptable limit 35 mg kg^{-1} (Table 5). At high concentration, it becomes toxic and causes severe disorders like loss of body weight and vision, heart and liver failure and skin infuriation (McGrath, 1990; Shah *et al.*, 2013).

Serial No.	Element	*Target Values of soil (mg/kg)	**Intervention Values of soil (mg/kg)
1	Cd	0.8	12
2	Cr	100	360
3	Pb	85	530
4	Ni	35	210

*Target values are specified to designate desirable maximum levels of elements in unpolluted soils.
**Intervention when remedial action is mandatory (Denneman and Robberse, 1990; Ministry of Housing, Netherland, 1994). Source: WHO (1996)

Table 5. *Permissible limits of heavy metals in soil and plants*

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Summary of accumulation of heavy metals in soil of Punjab

Based on the results obtained, it was observed (Fig. 1) that the order of heavy metal concentration (> permissible limit) in the wastewater irrigated soil samples of Punjab is Ni>Cr>Cd>Pb.

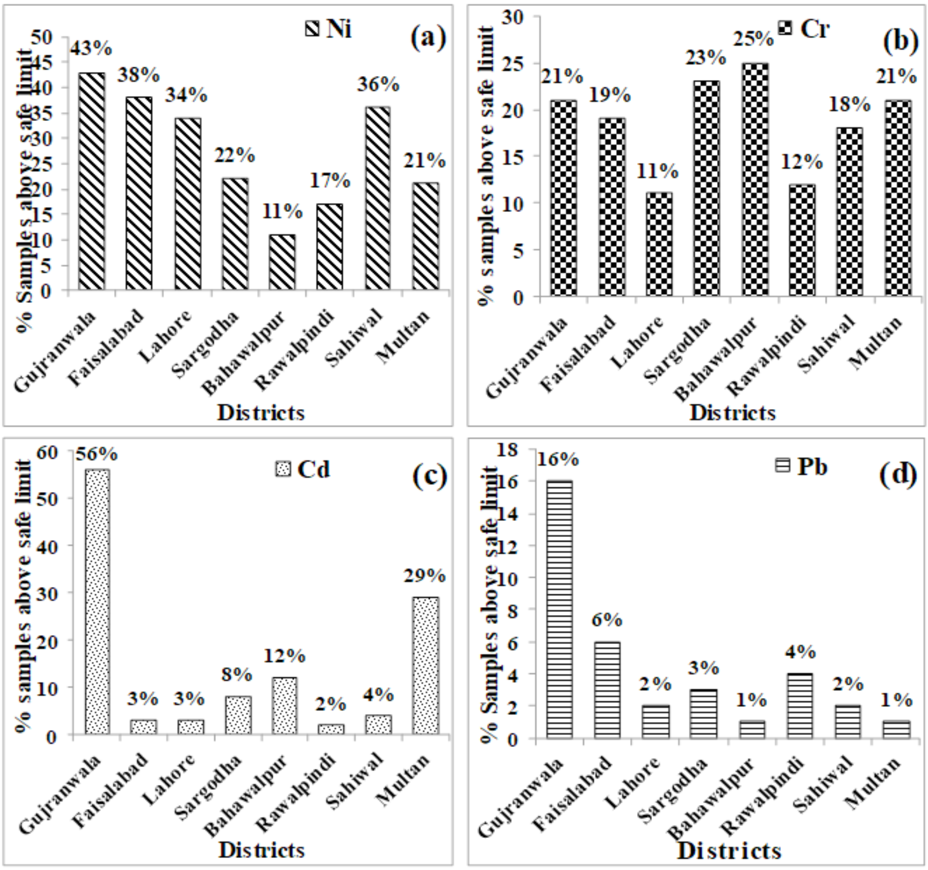

Figure 1 *Percentage of soil samples found contaminated with Ni* **(a)**, *Cr* **(b)**, *Cd* **(c)** *and Pb* **(d)**

In Gujranwala, Faisalabad, Lahore, Sargodha, Bahawalpur, Rawalpindi, Sahiwal and Multan district, 43, 38, 34, 22, 11, 17, 36, 21% wastewater irrigated soil samples were contaminated with Ni, 21, 19, 11, 23, 25, 12, 18, 21% with Cr, 56, 3, 3, 8, 12, 2, 4, 29% with Cd and 16, 6, 2, 3, 1, 4, 2, 1% with Pb, respectively (Fig. 1). The maximum heavy metal concentration was observed in Gujranwala district. Wastewater irrigation enhanced the heavy metals concentration in the soils where possibly they could become bioavailable to crops (Chaoua *et al.,* 2018).

Conclusions

The purpose of this survey study was to determine the concentration of toxic heavy metals and also to discuss the chemical properties of wastewater irrigated soil of Punjab. The results and discussion indicated that pH of wastewater irrigated soil was within range of WHO recommended acceptable limits while the values of EC were observed to be significantly higher than WHO limits. Organic matter contents and macronutrients recorded higher in wastewater irrigated areas. Similarly the results also indicated that the wastewater irrigation increased the heavy metal contents in soil. However, the concentration of heavy metals in wastewater irrigated soil of Punjab, presented levels in the following sequence: Ni>Cr>Cd>Pb. Considering these results, it is concluded that irrigation of agricultural soils with untreated wastewater for numerous years increased salts level and heavy metals concentration in soil which led to potential health hazards for humans through uptake of food.

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