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Impact of Sewage Water Irrigation on Soil and Radish (*Raphanus sativus* L.) with Respect to Heavy Metals in Tarlai, Islamabad

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ABSTRACT

With increasing population and urbanization, the sewage water generated by domestic, industrial and commercial sources has increased. In the Urban areas, sewage water is used as a source of irrigation water as it contains organic and inorganic elements essential for plant growth. However, sewage water may also contain heavy metals which contaminate the soil and toxic for plants which is a potential health risk to consumers. For this purpose, a study was conducted at Tarlai farms, Islamabad, Pakistan in 2016 to observe the impact of sewage water on heavy metals (Pb, Cd, As) content in soil and its accumulation in radish. The sewage water, soil and radish samples were collected at 7, 14, 20, 40 and 60 days after sowing (DAS) from farmer's field where use of sewage water for irrigation purpose is a regular practice for many years. The samples were analyzed using Atomic Absorption Spectrometry (AAS) and Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES). The physicochemical analysis of experimental soil at 0-15 cm depth indicate that it is sandy clay loam in texture, alkaline in reaction, non-saline with 0.08% total nitrogen and 1.8% organic matter. The results show that the concentration of heavy metals in sewage water and soil are in safe limits. However, in radish, the concentrations of heavy metals (Pb and Cd) are above than the permissible levels set by the World Health Organization (WHO). The highest concentrations of Pb (1.0 mg kg⁻¹) and Cd (0.65 mg kg⁻¹) in radish are found at 7 DAS while maximum level of As $(0.42 \text{ mg kg}^{-1})$ is recorded at 40 DAS. The levels of heavy metals in radish plant is found in the order of Pb > Cd > As.

1. Introduction

The production of sewage water has been increased due to increased industrialization, population and commercial activities. In the urban areas, where sewage water flows like a canal and due to ease of availability, farmers use it commonly for irrigation purposes, especially for raising vegetables [1]. It is recommended to use sewage water after proper treatment but people apply it directly [2]. The two main aspects regarding sewage water application in fields are enhancing soil fertility, as it carries organic matter and inorganic nutrients which are essential for plant growth and the other one is soil and plant contamination with hazardous chemicals, pathogens and undesired metals especially heavy metals which are under observation in this article [3, 4].

Irrigation with sewage water carrying heavy metals, first accumulates in soil and then via roots transports through xylum towards plant edible parts. In this way, these are being added to our food chain. Heavy metals are persistent in sewage water, soil and also in human body due to their nonbiodegradable nature [5, 6]. There are two sources of heavy metals contamination in sewage water that are natural and anthropogenic. Natural sources includes soil erosion, urban runoff and volcanos while the human sources includes metal finishing, minings, electroplating, textile industry, lead battery industry and metallurgical industries that are constantly emitting various trace metals (heavy metals) and through effluents reach in the urban sewage water stream [5, 7, 8]. Heavy metals toxicity adversely affect the human body. Among heavy metals, lead (Pb), cadmium (Cd), mercury (Hg) and arsenic (As) are considered most dangerous for health [9]. The toxicity of Pb in human body causes anaemia, hypertension, renal impairment, immunotoxicity, affects children's brain, reduced IQ and attention, increased antisocial behavior while its high exposure causes coma, convulsions and even death [10]. Cadmium toxicity affects the brain, IQ, blood pressure, classified carcinogen and may result in kidney failure [9, 11].Pollution of As in our body may cause cancer, skin lesion, pigment change and in severe cases vomiting, pain and diarrhea may also be observed [12].

The present work has been carried out inorder to investigate the status of heavy metals (Pb, Cd and As) in radish (*Raphanus sativus* L.) and soil irrigated with sewage water at Tarlai, farms, Islamabad.

2. Materials and Methods

The study was conducted during September – December 2016 at the farmer's field at Tarlai farms, Islamabad.

2.1 Site Characterization

The experimental site is situated at P&V Scheme1,Tarlai farms, Islamabad, Pakistan (Fig. 1). These farms have a long history of irrigation with sewage water canal named "Gumrah Kas". The experimental site is located at 33^0 38 North and 73^0 08 East with an altitude of 610 m above the sea level, with cool winter and warm to hot summer. Average annual temperature is 21.2 °C with 80 mm annual rainfall.

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Fig. 1: Location of the experimental site (P&V Scheme-1) at Tarlai farms, Islamabad.

2.2 Sample Collection

Composite samples of sewage water, soil and radish were collected from farmer's field at 7, 14, 20, 40 and 60 days after sowing (DAS) of radish. The sewage water pumping system used for irrigation at Tarlai farms, Islamabad is shown in Fig. 2. All three types of samples were collected randomly from the same site. The soil samples were collected at 0-15 cm depth using soil auger.

Moreover, soil samples were also collected from two sites adjacent to experimental area, where source of irrigation water was other than sewage water (tube well and submersible pump). Collected samples were properly labeled and directly transported to laboratory at Central Analytical Facility Division, PINSTECH, Islamabad for preperation and immediately performed coliform test in sewage water samples.



Fig. 2: Pumping of sewage water used for irrigation at Tarlai farms.

2.3 Sample Preparation

Soil samples were air dried and ground with pestle and mortar to fine powder and then sieved through $250\mu m$ sieve. For moisture determination, soil samples were dried in hot air oven at 105 °C for 48 hours.

Radish samples were washed first with tap water and then with distilled water to remove soil particles. The samples were air dried at room temperature. Pestle and mortar was used to convert the samples into powder form and stored for digestion. The radish samples collected in the last three stages (20, 40, 60 DAS) were divided into two parts (root and leaves) and after preparation were analyzed seperatley for heavy metals concentration.

2.4 Sample Digestion

Soil samples were digested with hydrofluoric acid (HF) and perchloric acid (HClO₄) method [13]. Radish samples were digested with di-acid mixture (nitric acid (HNO₃) and HCLO₄) [14].

The sewage water samples were analyzed without treatment/digestion (only filtered); the technique used are Atomic Absorption spectrometry (AAS) and Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES). For the analyses of Pb and Cd, Graphite Furnace Atomic Absorption spectrophotometer (GFAAS) was used. In this, sample is electrically heated with controlled fashion in four different steps, i.e., drying, ashing, atomization and cleaning. In the ashing, temperature is maintained at 300-1200 °C to convert organic matter to H₂O and CO₂; this is to confirmed that all organic matter is combusted and inert gas (Ar) is passed to remove the volatile materials. Arsenic was measured through hydride generation AAS in which only volatile arsenic hydrides (AsH₃) are routed to burner slot and then it is atomized and measured.

For ICP-OES technique, in plasma only ions and electrons can reside at high temperature (10000 K) so organic materials get combusted completely at such a high temperature.

2.5 Physicochemical Analysis

Before sowing radish, composite soil samples were collected at 0-15cm depth from farmer's field and analyzed for textural class, pH, conductivity, moisture content, total organic carbon/organic matter, total nitrogen, extractable potassium and extractable phosphorus using various instruments at Nuclear Institute of Food and Agriculture (NIFA), Peshawar.

The sewage water, digested soil and radish samples were analyzed using Atomic Absorption Spectrophotometer (AAS) for heavy metals and Inductively Coupled Plasma Optical Emission Spectrometry for macro nutrients determination. In Table 1, the list of instruments is given which are used for characterization in the present study. For Pb and Cd analysis, Graphite Furnace AAS (Hitachi Z 8000) was used, whereas As and Hg analysis were performed on HG-AAS (GBC 932). However, ICP-OES (Thermo, iCAP-6500) was used for Ca, Mg, Na and K analysis. Other parameters (Flouride, Chloride, Sulfide, Alaklinity) were measured by using Photometer and Spectrodirect RS-232. Table 1: List of instruments with their models used for analyses for various parameters.

Parameters	Instruments/model	
Heavy Metals		
Pb	CEAAS Litechi 7 9000	
Cd	GFAAS, Hitachi Z-8000	
As	UCAAS CDC 022-b	
Hg	HGAAS, GBC-932plus	
Macronutrients		
К		
Ca	ICP-OES, Thermo, iCAP-6500	
Na	ICF-OES, Inemio, ICAF-0500	
Mg		
Physicochemical parameters		
pН	pH meter, Oakton pH 2100 series	
Conductivity	Conductivity motor Incl.ch	
TDS	Conductivity meter, InoLab	
TSS		
Ammonia	Photometer, MultidirectLovibond	
COD		
Sulfide		
Fluoride	Photometer Spectrodizet PS 222	
Chloride	Photometer, Spectrodirect RS-232	
Alkalinity		

2.6 Standard Reference Material (SRM)

Three Standard Reference Materials, CL IAEA-359 (cabbage leaves), PL NIST-1547 (peach leaves) and IAEA Soil-7 were used in the experiments for heavy metals analyses. The SRMs were digested simultaneously with soil and plant samples and thereafter analyzed by Atomic Absorption Spectrophotometer.

2.7 Environmental Parameters

2.7.1 Plant concentration factor

Heavy metals transfer from soil to the plant through roots and the transportation is calculated by Plant Concentration Factor (PCF) and is given as [15]:

$$PCF = \frac{Concentration of heavy metal in plant}{Concentration of heavy metal in soil}$$

2.7.2 Pollution load index

The irrigation with sewage water may increase the concentration of heavy metals in the soil as compared to reference soil (the soil having no contamination of heavy metals). Therefore, to calculate how much soil is polluted is measured by Pollution Load Index (PLI) parameter [15]:

$$PLI = \frac{C_{soil}}{C_{reference}}$$

where

 C_{soil} = Concentration of heavy metals in sample soil $C_{reference}$ = Concentration of heavy metals in reference soil The data obtained was calculated for PLI in terms of Pb, Cd and As.

3. Results and Discussions

The samples were analyzed and results obtained are given below:

3.1 Physicochemical Analysis of Experimental Soil

Before starting the experiment, composite soil samples were collected from the experimental site at 0-15 cm depth and were analyzed for various parameters. The results reveal that soil has 56% sand, 22% silt and 22% clay (Table 2). The texture of soil is sandy clay loam. This type of soil has a great impact on root growth and ability to absorb nutrients and water for optimum plant growth. Roots of the plant easily penetrate and grow well in sandy loam soil [16]. The results show that soil is alkaline in reaction with pH 7.46, non-saline (EC 0.7 ds m⁻¹) with 0.08 % total nitrogen, 10 mg kg⁻¹ extractable phosphorus and 400 mg kg⁻¹ extractable potassium (Table 2). The soil analysis further revealed that soil has 4.35% calcium, 0.5% sodium and 0.6% magnesium.

Table 2: Physicochemical parameters of experimental soil.

Properties	Units	Values
Sand	%	56
Silt	%	22
Clay	%	22
Textural Class		Sandy Clay Loam
pH (1:2.5)		7.46
EC _e (1: 2.5)	dS m ⁻¹	0.7
Moisture	%	17
Total organic carbon	%	1.0
Organic Matter	%	1.80
Total Nitrogen	%	0.08
Extractable Phosphorus	mg kg ⁻¹ soil	10
Extractable Potassium	mg kg ⁻¹ soil	400
Calcium	%	4.35
Magnesium	%	0.60
Sodium	%	0.50

The analysis disseminate that soil organic matter content is 1.8% with total organic carbon of 1.0%. Soil organic matter is composed of various components including plant residues, microorganisms and stable organic matter. Soil organic carbon is an important quality indicator as it affects physical, biological and chemical properties of soil [17]. The presence of organic matter content is important as it improves soil structure, provides essential nutrients or modifying soil properties that can improve plant growth [18]. Moreover it plays a significant role in micronutrients availability and their uptake by plants. It increases cation exchange capacity due to high negative charge of organic matter. It keeps nutrient retention and availability to plants [17]. The experimental soil show high organic matter, the reason might be due to use of sewage water for irrigation for last many years for growing vegetables. As sewage water also contains organic matter [19] that resulted into build up of soil organic matter.

3.2 Physico-chemical Properties of Sewage Water

The physicochemical analysis of sewage water are presented in Table 3. These result show that sewage water has pH 6.3-6.6 and electrical conductivity in the range of (765-914 μ S cm⁻¹). Total dissolved solids (TDS) is an important parameter which indicate the presence of inorganic salts and small amount of organic matter. Our results show that TDS value is in the range of 420-500 mg L⁻¹ which is below than the safe limit. These results further reveal that sulfide, fluoride, chloride and sulphate in sewage water are in permissible limits as described by WHO [20].

It is also observed that sewage water contain total coliforms (89 CFU/100 mL) and ammonia in the range of $(0.03-0.46 \text{ mg } \text{L}^{-1})$. Another important parameter, i.e., chemical oxygen demand (COD) is useful which directly measures the amount of organic compounds in water.

Table 3: Physico-chemical properties of sewage water with safe limits.

Parameters	Units	Value	*Safe Limits
pH		6.3 - 6.6	6.5-8.4
Conductivity	µs/cm	765 - 914	250 - 3000
TDS	mg/L	420 - 500	<2000
TSS	mg/L	4 -17	\leq 50
Ammonia	mg/L	0.03- 0.46	5
Alkalinity	mg/L	335 - 419	<610
Sulfide	mg/L	0 - 4.1	
Fluoride	mg/L	0.5 - 0.95	1.0
Chloride	mg/L	4.7 - 24.1	<350
Sulphate	mg/L	30.59	500
TotalColiforms	CFU/100mL	89	150
COD	mg/L	40	250

*WHO [20].

The analysis showed that COD (40 mg L^{-1}) is well below than the safe limit of WHO. Alkalinity, sulfide, fluoride, chloride and sulphate measured valus were also within the safe limits (Table 3).

The results regarding the elemental profile of sewage water collected at the start of the experiment show that most of the heavy metals (Pb, Cd, Hg, Cu, Se and Zn) are not detected in the sewage water (Table 4). The sewage water has Cr (5.6 ng mL⁻¹) and As (2.17 ng mL⁻¹) but these values are well below than the safe limits as mentioned by WHO [20].

The analysis of sewage water performed using ICP-OES technique revealed that sewage water has some contents of macronutrients including, Na, Ca, K and Mg (Table 4). On the

Table 4: Elemental profile of sewage water used for irrigation.				
Elements	Units	Concentrations	lod ^a	Safe Limits*
Pb	ng/ml	ND	8.76	5000
Cd	ng/ml	ND	0.35	10
Cr	ng/ml	5.6	2.59	100
Cu	ng/ml	ND	14.66	200
Hg	ng/ml	ND	0.52	2
As	ng/ml	2.17	0.69	100
Se	ng/ml	ND	0.816	20
Zn	µg/ml	ND	0.01	2
Na	µg/ml	35.6 - 86.0	0.198	230
Ca	µg/ml	34.4 - 84.0	0.002	230
Κ	µg/ml	6.3 - 17.0	0.005	<2.0
Mg	µg/ml	23.2 - 34.0	0.165	<61

basis of these results, sewage water may be termed suitable

for irrigation but the excessive use of this sewage water for

irrigation purpose may accumulate As in the soil.

*WHO [20], ND =Not detected, ^alimit of detection

3.3 Lead, Cadmium and Arsenic Content in Sewage Water at Different Time Intervals

The sewage water used as source of irrigation for radish was analyzed for heavy metals (Pb, As, Cd) contents at 7, 14, 20, 40 and 60 DAS; these results are presented in Table 5. The results show that Pb and Cd are not detected in sewage water at different time intervals during the experiment. Arsenic contents are found in the range of 2.16 - 5.12 ng mL⁻¹ but these values are well below than the permissible limits of WHO.

Table 5: Pb, Cd & As levels in sewage water at different time intervals.

Days after Sowing	Pb	AS	Cd
Mean +S.D (ng mL ⁻¹)			
7 DAS	ND	2.16 <u>+</u> 0.08	ND
14 DAS	ND	3.81 <u>+</u> 0.07	ND
20 DAS	ND	2.23 <u>+</u> 0.12	ND
40 DAS	ND	5.12 <u>+</u> 0.07	ND
60 DAS	ND	2.43 <u>+</u> 0.07	ND

ND = Not detected

3.4 Lead, Cadmium and Arsenic Content in Soil

The results regarding heavy metals (Pb, Cd and As) content in soil at 0-15 cm depth at different time intervals during the experiment are presented in Fig. 3. The results show that Pb content in soil are in the range of 1.76-3.34 mg kg⁻¹. A gradual increase in Pb contents are observed in soil during the experiment with minimum $(1.76 \text{ mg kg}^{-1})$ level is recorded at 7 DAS and maximum level (3.34 mg kg⁻¹) at 60 DAS. However, the Pb content in soil are below the safe limit (84 mg kg⁻¹) as described by WHO [20].

The results regarding As content in soil show that no significant change in As contents are observed throughout the

study. The range of As in soil is 2.70-3.01 mg kg⁻¹. Moreover, Pb and As contents are almost similar in soil.

The Cd concentration in soil is found in the range of 0.11-0.31 mg kg⁻¹. There is no any significant change observed in Cd levels during the study. The results further disseminate that Cd concentration in soil is almost similar at 20, 40 and 60 DAS. The Cd and As level in soil are also found under permissible limit as mentioned by WHO [20]. Similar results are reported by Naser et al. [21] when they observed the behavior of heavy metals at different time intervals in vegetables.



Fig. 3: Concentration of Pb, Cd and As in experimental soil with safe limit at different time intervals.

3.5 Heavy Metals Content in Radish Plant

The behavior of the radish plant towards the individual heavy metals (Pb, Cd and As) are different at various time intervals. The mean values of As Pb and Cd concentration at different time intervals are presented in Fig. 4.

The results show that As, Pb and Cd in the radish plant in different growth stages are in the range of ND (not detected) to 0.42 mg kg⁻¹, 0.09 to 1.0 mg kg⁻¹ and 0.09 to 0.65 mg kg⁻¹, respectively. The highest concentration of Pb (1.0 mg kg⁻¹) and Cd (0.65 mg kg⁻¹) in radish are found at 7 DAS while maximum level of As (0.42 mg kg⁻¹) is found at 40 DAS. The level of heavy metals in the radish are found in the order of

Pb > Cd > As. It can be seen from Fig. 4(a) that As content in radish plant is under the threshold limit (0.43 mg kg⁻¹) set by WHO [22]. At initial plant growth stage, As contents are not detected in radish plant. However at 40 DAS, As level is increased to maximum (0.42 mg kg⁻¹). After that a decrease in As content (0.14 mg kg⁻¹) is observed at harvesting stage.

Among all the heavy metals, Lead (Pb) level in soil is higher so its uptake is relatively higher than the rest of the heavy metals. Lead level in radish plant is decreasing successively at 14 and 20 DAS then jumps high at 40 DAS (Fig. 4(b). At the harvesting stage, its concentration again decrease and is just below the threshold limit. The decreased concentration at the harvesting stage may be attributed due to the dilution effect as at that stage the weight and size of the radish sharply increases. Naser et al [21] also reported more or less similar trend of heavy metals in vegetables.

In case of Cd, the concentration remained above the threshold limit (0.1 mg kg^{-1}) in all growth stages except at last stage (Fig. 4 (c)), it is dropped to just below safe limit. In this study, Pb, Cd and As content are decreased at maturity level, this may be due to dilution effect with plant growth [23].

These findings are in line with Parveen et al. [24] who studied the impact of municipal waste water on turnip, where the concentration of heavy metals in the waste water, used for irrigation, was within the safe limits. However, in plant parts, heavy metals showed significantly higher values due to continuous use of waste water; similar findings are also reported by Parashar and Prasad [25].

3.6 Distribution of Heavy Metals in Radish (Root & Leaves)

The distribution of heavy metals in root and leaves depends upon the genotype of the plant. Fig. 5(a) shows the As concentration in root and leaves. It can be seen that with growth stage advancement, the As is not only absorbed by the root but also transported to the leaves of the radish as well. There is substantial increase in the uptake of As with the growth stages. The concentration of As in both root and leaves remained below the threshold value recommended by WHO [22] in all three growth stages.



Fig. 4: Concentration of (a) As, (b) Pb and (c) Cd in radish plant with permissible limits at different time intervals.





Concentration of (a) As, (b) Pb and (c) Cd in root and leaves of radish at different time intervals. Fig. 5:

The permissible value of Pb by the WHO is 0.25 mg kg⁻¹ on the basis of dry weight. Moreover, the Pb uptake by the root is greater at earlier stage than the 4th and 5th stage Fig. 5(b). However, transportation of Pb to radish leaves was greater at later stages. It is clear from Fig. 5(a & b) that uptake and distribution of heavy metals is not linear with growth stage.

In contrast to As, Cd uptake is higher at initial growth stage (Fig. 5(c)) and then successively decrease with the maturity of the plant. This sharp decrease is due to dilution effect as size and weight of the plant increases rapidly during last stages. It may also be clear that Cd uptake at 4th and harvesting stages are relatively less as compared to Pb and As. It is observed that radish plant behavior towards different heavy metals uptake is different.

From Fig. 5(b) it can be seen that Pb level in leaves has crossed the safe limit at 40 and 60 days after sowing. So the consumption of leaves may cause health risks at harvesting stage.

If we compare the results of heavy metals (Pb, Cd and As) shown in Figs. 3, 4 and 5 we can infer that Pb concentration is higher in both soil and plant followed by As and then Cd. Hence, it may be concluded that higher concentration of heavy metal in soil leads to more uptake of that heavy metal in the plant. Paeveen et al [17] described more or less similar distribution of heavy metals in their study.

3.7 SRM Analyses for Heavy Metals

Standard reference material (SRM) is treated similar as other samples to check the accuracy and precision of the experimental facility (digestion and instrument) by matching the experimental data with certified values. In this study, SRMs of International Atomic Energy Agency (IAEA) and National Institute of Standards and Technology (NIST) are used to determine the recoveries of the As and Cd. The detail of specific SRMs that are used in the experiments and their comparison between measured and certified values are presented in Table 6. It can be seen that As is recovered 100% in both cabbage leave (CL IAEA-359) and peach leave (PL NIST-1547) whereas in soil (IAEA Soil-7) its recovery is 88.5%. Cadmium (Cd) percentage recovery in SRM (IAEA Soil-7) is 96% which show a great precision and accuracy of

Element Reference Value Value

Standard

adopted during the experiment.

reference materials.

	Material	(mg kg ⁻¹)	(mg kg ⁻¹)	lecovery
	CL IAEA-359	0.10 ± 0.02	0.10	100.0
As	PL NIST- 1547	0.06±0.02	0.06	100.0
	IAEA Soil-7	11.86±0.1	13.40	88.50
Cd	IAEA Soil-7	1.40±0.03	1.50	96.00

the processes. Overall, the results disseminate a very good

precision and accuracy of the methods (digestion a analysis)

Table 6: Comparison of measured values with certified values of standard

Measured

Certified

Percentage

3.8. Environmental Parameters

The plant concentration factor (PCF) and pollution load index (PLI) of Pb, Cd and As for radish are shown in the Table 7. The uptake of the metals are functions of different factors as described earlier. The PCF values describe the result of competition of metals in the same biological system [3]. It is good estimation about the transfer of heavy metals from soil to the plant. The results show that PCF values for all the heavy metals are less than one. The comparison of PCF values among heavy metals indicate that PCF value of Cd is higher than Pb and As, which reflects it's relatively high uptake (i.e., with respect to concentration in soil and plant). Moreover, PLI reflects a comparison between background and polluted soil.

Table 7: Plant concentration factor and pollution load index values for lead, cadmium and arsenic.

Heavy Metals	PCF	PLI	
Pb	0.003	2.33	
As	0.053	4.28	
Cd	0.818	1.57	

The pollution may assessed as [26]:

$1 < PLI \ge 3$	medium level of pollution
PLI>3	high level of pollution

Our results indicate that PLI values for Pb, Cd and As are higher than 1. Cadmium and lead show medium level of pollution, whereas Arsenic reflects high level of pollution in experimental soil for radish.

4. Conclusions

On the basis of the present results, the following conclusions can be drawn:

The concentration of heavy metals (Pb, Cd and As) in irrigated sewage water and soil are found in safe limits. However, Pb and Cd levels in radish are above safe limits, which decrease with the plant maturity. Cadmium being the most toxic heavy metal remained above safe limit in radish till 40 days after sowing, after that its concentration decreases sharply due to dilution effect. It is concluded that harvesting at early stage may cause more heavy metals contamination and greater health risk associated with the consumers for long term use.

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