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A Comparative Study of Heavy Metal Toxicity in the Vegetables Using ICP-MS and AAS

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ABSTRACT

Our study was conducted between January and September 2023, and aimed to evaluate the presence of toxic heavy metals in vegetables cultivated using effluent and sewage water in Jodhpur city. Eight vegetable samples were obtained from four distinct locations, and the concentrations of seven harmful heavy metals (Cu, Zn, Pb, Cd, Cr, Ni, and Fe) were examined. The initial analysis employed ICP-MS, followed by determining the concentration of heavy metals in the vegetables through atomic absorption spectrometry (AAS). The Heavy Metal Pollution Index (HPI) was computed to assess pollution levels at each site. Comparative analysis of vegetable samples using both ICP-MS and AAS indicated elevated levels of lead, cadmium, and chromium pollution in vegetables from all four sites. Calculations further confirmed the contamination of the sites with toxic heavy metals. The order of metal concentration based on allowable limits was Fe>Cd>Cr>Pb>Ni>Zn>Cu. The overall order of observed heavy metal concentrations site-wise was site 4 > site 3 > site 2 > site 1. Many samples were obtained from local vendors near residential areas and community markets frequented by the public. This highlights a crucial issue of heavy metal toxicity exposure to the general population, emphasizing urgent government action, including essential restrictions and measures, to address this pressing problem.

Keywords: Contamination, Toxic metal ions, Wastewater, Vegetables, Irrigation, Concentration, HPI.

INTRODUCTION

Vegetables play a vital role in human nutrition, supplying essential micronutrients such as copper, zinc, calcium, iron, magnesium, iodine, sodium, potassium, antioxidants, vitamins, and various bioactive compounds. Given that both cooked and raw foods are commonly consumed, it is imperative to consider the presence of harmful heavy metals in vegetables, as they can pose risks to human health.[1-2] Numerous global reports extensively detail the contamination of heavy metals in the food chain, with plants having the capacity to absorb these metals from the soil, resulting in elevated concentrations in their edible parts.[3-4] The primary catalyst for heavy metal accumulation in plants is soil pollution caused by human activities like industrialization, urbanization, and natural geological factors.

While certain heavy metals like Cu, Zn, and Ni are beneficial in trace amounts as micronutrients, others such as lead (Pb), mercury (Hg), cadmium (Cd), and arsenic (As) are highly toxic even at low levels, particularly for vulnerable groups like pregnant women and young children[5]. Cadmium is a notable concern due to its mobility in soil and plants, posing health risks to various organs and increasing the likelihood of certain cancers, including pulmonary adenocarcinoma and prostatic lesions, in addition to compromising the immune system. Elevated blood levels of lead can lead to hypertension, impairments in the skeletal, immune, and endocrine systems, and diminished cognitive abilities in children, affecting renal and cardiac functions in adults.[6-7] Vegetables and fruits serve as the primary sources of heavy metal exposure in humans, constituting 90% of metal intake, while the remaining 10% comes from skin contact and inhalation of contaminated dust[8-12]. Therefore, quantifying the concentration of heavy metals in commonly consumed vegetables and fruits is essential for assessing potential health risks[13-16]. There have been reports of heavy metal toxicity in the Jodhpur city[17-19]. However, continuous assessment of toxic metal ions in vegetables is very important for people's health. Despite the National Green Tribunal (NGT) ban on direct disposal of industrial effluents and their subsequent use for irrigation, many farmers are using polluted water for the irrigation of vegetables and settling them in the local market small shops, and man carts[20-21]. Given the ongoing use of toxic effluents in the city, and increasing Cancer cases, regular monitoring of heavy metal infiltration in the edibles is crucial including evaluating the health risks linked to heavy metal-infused commonly consumed vegetables.

Methodology

Sample Collection Sites

Our study areas were divided into four sites, namely Salawas, Madhuban Housing Board mandi, Basni phase 1 and 2, Bhadwasia Nallah area, and Sangaria region of Jodhpur City, to ascertain the impact of heavy metal contamination in vegetables irrigated with polluted and untreated wastewater from the sewage and industrial effluents. Eight vegetables, namely, Carrot, radish, tomato, coriander, spinach, radish leaves, mustard leaves, and wild spinach were gathered from various locations and were stored in marked sampling packets.

Preparation of Samples

The samples collected and their preparation are represented in Fig. 1, All the collected samples were rinsed with water and dried in the oven Finally, they were ground into a homogeneous fine powder and digested in acids. 1 g of a single sample (fine powder of dried vegetables) was weighed and digested with 15 mL of triacid mixture (70% high purity HNO_3, 65%$HClO_4$ and 70% H_2SO_4;5:1:1) in a conical flask. They were thoroughly stirred for 30 min on a hot plate at 250°C till a translucent solution was obtained. The digested materials were filtered, and diluted with distilled water and were used for the analysis. For each sample, we prepared blank solutions. Then, these samples were analyzed by ICP-MS[22], and later the concentration of all heavy metal ions was measured using AAS.

Fig. 1. Samples collected and preparation

RESULTS AND DISCUSSION

Heavy metals are important in eco-chemistry and eco-toxicology due to their low-level toxicity and tendency to accumulate in human organs. FAO/WHO has published vegetable metal content limits, and these limitations are an essential part of a healthy diet Table 6. Heavy metals stay around for a long time and are not chemically degradable. Physical methods are needed for their removal, or they should be changed into nontoxic compounds.

In our research, eight species of vegetables were collected from four different sites. We collected four samples of each vegetable from 4 different sites in different areas in the remote areas of the Blue City and later analyzed them for seven heavy metals. The study was divided into three parts.

(1) The samples were analyzed by ICP-Ms and polluted sites were marked.
(2) The samples were assessed by AAS for heavy metal concentration.
(3) The heavy metal pollution index was calculated to ascertain site-wise pollution.

Analysis by Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

ICP-MS analysis of 32 samples collected from different sites in Jodhpur city was done. The below graph 1-11 shows the ICP-MS analysis.

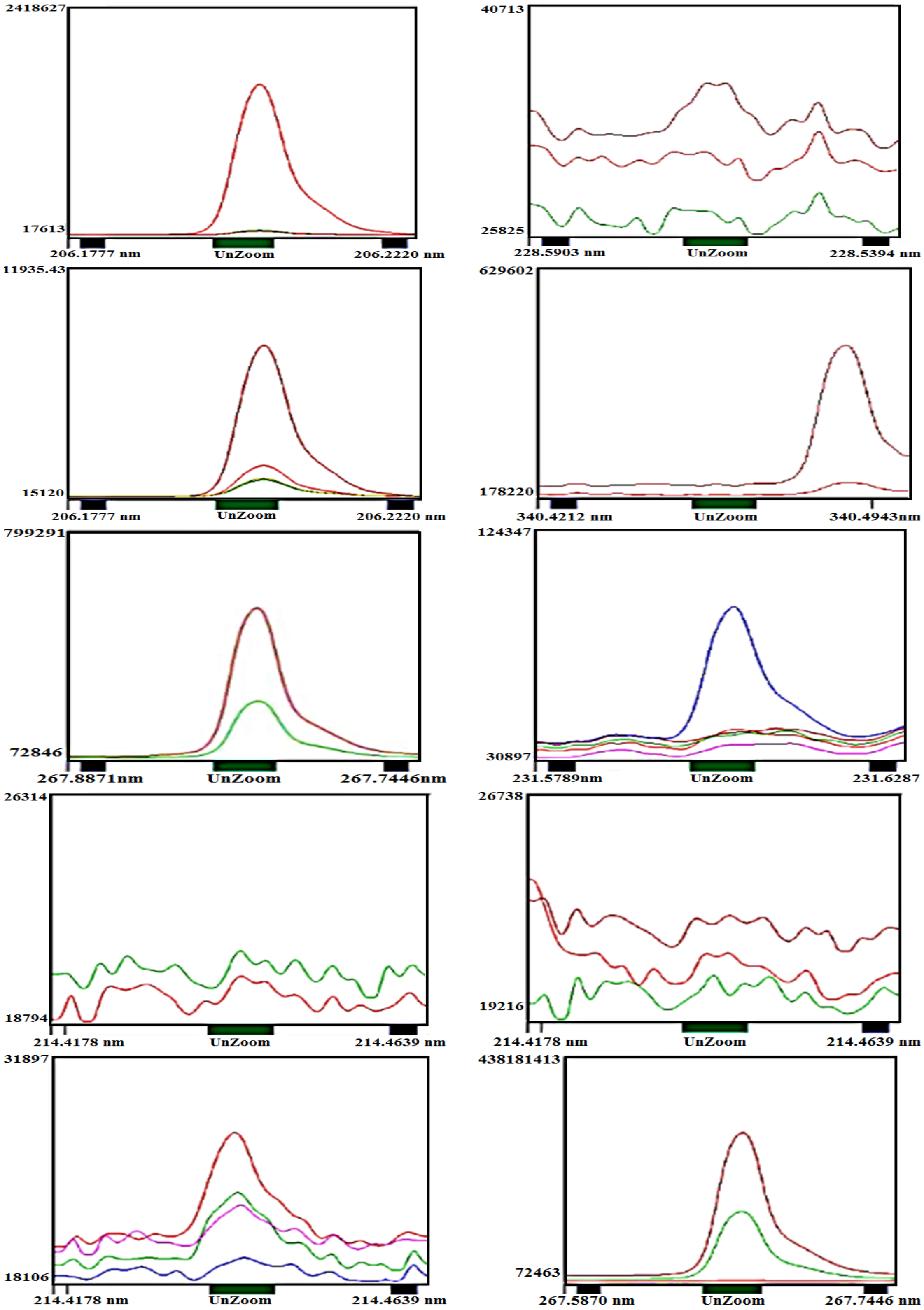

Fig. 2. ICP-MS Analysis of vegetable samples at four different sites

Site 1-Bhadvasia area samples were collected from the sabzi mandi shops and farms. The intensities observed are represented in Fig. 2 and Table 1. The results indicated that the vegetables were least polluted in Site 1 areas.; maximum intensity variations were observed in tomato and radish for Zn metal ions. Site 1 looked the least polluted.

Site 2 Sangaria-Here mainly Jojari river water is used for irrigation. The intensities observed are shown in Fig. 2 and Table 1. Most of the vegetable samples were not polluted and did not show much variation in the intensity. However, coriander, spinach, and radish leaves showed high-intensity variation for Pb, as shown in Table 1, and tomato and radish showed high-intensity variation for Zn metal ion, as represented in. So, site 2 was moderately polluted.

Site-3-Salawas- Here mainly industrial wastewater and Jojari river water is used for irrigation The intensities observed are shown in Fig. 2 and Table 1. The intensity of Pb, Cr, and Ni showed a remarkable increase, especially in wild spinach and tomato, for Pb metal ions as shown in wild spinach, coriander and wild spinach for Cr metal ions as shown in wild spinach, coriander for Ni metal ion as indicated in Fig 2. Site 3 was very much polluted.

Site-4-Madhuban Housing Board and Basni Phase 1 and 2- This site has mainly local vegetable shops, mandi and farms where vegetables are sold, which are mainly grown with toxic polluted water. Site four showed maximum variation in the intensity for Cd, Cr, and Ni metal ions. Spinach, radish leaves, coriander, tomato, carrot, radish in mustard leaves, and wild spinach for Cd metal ion. Radish leaves for Cr metal ion and tomato radish for Ni metal ion as indicated in Fig. 2 and Table 1. The most intensity variation was observed in site 4. Hence, site 4 was more populated than other sites.

Table 1: Observed metal intensity for different vegetable samples at all four sites by ICP-MS

Metal vegetable	Cd	Pb	Cr	Ni	Zn
			λ_{max}		
	214.441	220.353	267.716	231.604	206.200
			Blank Intensity		
	24	534	34382	1338	3533
			Observed Intensity		
Tomato	685*	1042#	-	2349*	530450ϕ 1089231Ψ
Coriander	1537*	971ϕ	131140#	4930#	-
Wild Spinach	138*	1079#	348676#	40054#	-
Radish	461*	-	192969234*	1986*	62592ϕ 34452Ψ
Carrot	677*	-	-	-	-
Spinach	4631*	610ϕ	-	-	-
Mustard leaves	357*	-	1452348*	2600#	-
Radish leaves	3388*	523ϕ	92170349*	-	-

Site 4-Intensity*, Site 3-Intensity#, Site 2-Intensityϕ, Site 1-IntensityΨ

The overall order observed for heavy metal concentration by ICP-Ms was-site 4> site 3> site 2> site 1.

Based on the above data, we advanced our study and did an AAS analysis for all 32 samples to get an exact amount of heavy metal concentration in ppm.

Analysis by AAS and HPI:[23-24]

Atomic Absorption Spectroscopy was then used to analyse all the samples again. The results of the concentration measurements are given in parts per million (ppm). The results are summarized in Tables 2, 3, 4 and 5.

Table 2: Metal concentration and HPI values for eight different vegetables at site 1

S. No	Vegetable Name	Cd	Cu	Pb	Ni	Cr	Zn	Fe	HPI
1	Carrot	0.033	0.1536	0.1876	0.955	0.0026	1.4315	5.354	0.2169
2	Radish	0.147	0.0471	1.331	0.495	0.0041	2.583	3.462	0.2962
3	Tomato	0.010	1.058	1.314	0.122	0.021	55.452	7.436	0.5538
4	Coriander	0.003	0.477	1.175	0.513	0.0043	1.753	19.59	0.2875
5	Spinach	0.020	0.5036	1.078	0.317	0.0049	1.5161	13.50	0.3285
6	Radish leaves	0.002	0.3694	1.2459	0.587	0.0040	2.290	11.46	0.2609
7	Mustard Leaves	0.006	0.384	0.9340	0.109	0.0067	1.845	6.779	0.2227
8	Wild spinach	0.034	0.406	1.4163	0.7108	0.0023	2.930	7.985	0.3653

Table 3: Metal concentration and HPI values for 8 different vegetables at site 2

S. No	Vegetable Name	Cd	Cu	Pb	Ni	Cr	Zn	Fe	HPI
1	Carrot	0.073	0.199	0.5077	0.411	0.0040	2.011	4.306	0.2774
2	Radish	0.028	0.023	0.8325	0	0.01264	4.169	2.7743	0
3	Tomato	0.024	0.344	1.791	0.661	0.006	26.99	4.451	0.4997
4	Coriander	0.034	0.599	1.559	0.533	0.0055	1.717	14.031	0.4258
5	Spinach	0.024	0.472	1.210	0	0.0064	2.625	11.363	0
6	Radish leaves	0.028	0.344	0.840	0.341	0.0037	1.382	11.67	0.2959
7	Mustard leaves	0.036	0.335	1.5159	0.252	0.00527	2.883	12.777	0.3743
8	Wild spinach	0.023	0.473	0.9951	1.146	0.0043	3.184	11.756	0.4189

Table 4: Metal concentration and HPI values for 8 different vegetables at site 3

S. No	Vegetable Name	Cd	Cu	Pb	Ni	Cr	Zn	Fe	HPI
1	Carrot	0.150	0.210	1.363	0.2995	0.00511	1.628	5.5671	0.3536
2	Radish	0.0415	0.039	0.8214	0.2041	0.0199	5.0239	3.6696	0.2752
3	Tomato	0.023	0.942	1.7346	0.486	0.032	52.859	5.637	0.7829
4	Coriander	0.0222	0.6974	0.9195	2.291	0.0244	2.668	27.80	0.6729
5	Spinach	0.051	0.460	0.5851	0.3677	0.0086	3.137	11.709	0.4058
6	Radish leaves	0.027	0.3595	0.966	0.9275	0.0134	1.976	19.184	0.4681
7	Mustard leaves	0.0309	0.4906	1.28	1.20	0.0043	1.6611	19.083	0.4469
8	Wild spinach	0.029	0.6511	1.67	18.622	0.065	2.976	15.028	1.0777

Table 5: Metal concentration and HPI values for 8 different vegetables at site 4

S. No	Vegetable Name	Cd	Cu	Pb	Ni	Cr	Zn	Fe	HPI
1	Carrot	0.55	1.318	0.345	0.050	0.038	2.757	8.690	0.5344
2	Radish	0.234	0.174	0.732	0.893	28.5342	7.230	15.265	1.8589
3	Tomato	0.220	1.648	0.589	1.363	0.0625	0.798	2.590	0.6317
4	Coriander	0.031	1.296	0.330	0.081	0.042	1.078	4.166	0.3040
5	Spinach	0.052	1.040	0.294	0.063	0.022	1.010	8.081	0.2990
6	Radish leaves	0.238	0.510	0.560	0.587	1.928	0.962	12.655	0.9909
7	Mustard leaves	0.127	0.821	0.702	0.411	0.4089	3.029	23.245	0.9801
8	Wild spinach	0.121	0.814	0.758	0.639	0.0198	2.645	12.965	0.6187

Table 6: Variations in the HPI of heavy metal ions observed in the vegetables

Metals	Permissible limits in ppm for vegetables	The mean concentration of metal ions for all sites (ppm)	Increased Percentage (%)	Diseases caused by excess metal concentration
Cd	0.003	0.0659	2096.67	It can cause cancer.
Cu	1	0.4972	Within limits	Vomiting, jaundice, depression, fatigue
Pb	0.3	0.9797	226.57	Muscular, skeletal and neurological disorders
Ni	0.5	0.8505	70.1	Headache, gastrointestinal and respiratory disorders
Cr	0.05	0.6571	1214.2	Cardiac, neurological, and gastrointestinal disorders
Zn	5.15	5.0439	Within limits	Vomiting, nausea, diarrhoea
Fe	0.3	9.5518	3083.93	Diabetes, hemochromatosis, stomach problems and nausea
HPI	0.2376	0.4208		

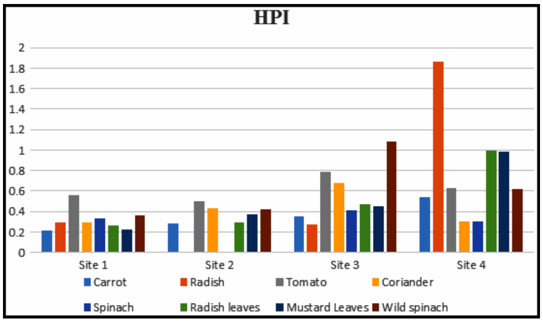

Fig. 3. HPI of all the vegetable samples at four different sites

Analysis of vegetable samples by AAS

Cadmium

Cadmium levels in vegetables averaged 0.0659 parts per billion. The mean concentration of all four sites per vegetable was -Radish leaves> Carrot> Radish> Tomato> Wild Spinach> Mustard leaves> Spinach> Coriander. The site-wise cadmium concentration was - Site 4> site 3> site 2 > site 1

Copper

The copper concentrations in the vegetables were within safe limits and averaged 0.4972 ppm. The mean concentration of copper concentration at all the four sites per vegetable was-Tomato > Coriander > Spinach> Wild spinach> Mustard leaves> Carrot> Radish leaves> Radish The site-wise copper concentration was -Site 4> site 3> site 1> site 2

Lead

This research revealed that among eight types of vegetables sampled across four sites, tomato had the highest lead content (1.791 ppm) at site 2. The average amount of lead found in vegetables was 0.9797 parts per billion. The mean concentration of all four sites per vegetable was -Tomato> Coriander > Wild spinach > Mustard leaves> Radish > Radish leaves> Spinach > Carrot. The site-wise Lead concentration was -Site 3 > site 2 > site 1 > site 4.

Nickel

The highest nickel content was discovered in wild spinach (18.622 ppm) at site three among the eight types of vegetables tested at four sites. The average nickel content of vegetables suitable for human consumption was 0.8505 ppm. The mean concentration of all four sites per vegetable was Wild spinach > Coriander > Tomato > Radish leaves > Mustard leaves > Carrot > Radish > Spinach. The site-wise Nickel concentration was -Site 3> site 4 > site1 > site 2.

Chromium Radish had the highest chromium levels (28.5342 ppm) at site 4 out of the eight vegetable groups tested. Wild spinach at the first site had the lowest concentration (0.0023 ppm). The average chromium content of vegetables suitable for human consumption was 0.6571 ppm. The mean concentration of all four sites per vegetable was -Radish > Radish leaves > Wild spinach > Mustard leaves > Tomato > Coriander > Carrot > Spinach. The site-wise chromium concentration was -Site 4 > site 3 > site 1 > site 2.

Zinc

The highest quantity of zinc was discovered in tomatoes (55.45 ppm) at site 1 out of the eight vegetable groups tested. However, at site 4, the concentration was lowest in tomatoes (0.798% ppm). Zinc levels in consumable veggies averaged 5.0439 ppm. The mean concentration of all four sites per vegetable was- Tomato > Mustard leaves > Radish > Wild spinach > Spinach > Carrot > Coriander > Radish leaves. The site-wise Zinc concentration was- Site 3 > site 1 > site 2 > site 4.

Iron

Coriander had the highest zinc levels (27.80 ppm) at site 3 out of the eight vegetable groups tested across the four locations. Site 4's tomato sample had the lowest value (2,590 ppm). Zinc levels in vegetables that may be consumed averaged 9.5518 ppm. The mean concentration of all four sites per vegetable was- Radish > Coriander > Mustard leaves > Radish leaves > Wild spinach > Spinach > Carrot >Tomato. The site-wise Iron concentration was- Site 3 > site 4 > site 1 > site 2.

Heavy Metal Pollution Index (HPI).[23-24]

HPI of all sites vegetable-wise is represented in Fig. 3. It is based on the heavy metal ion concentrations obtained by AAS and it evaluates the total metal content in each variety of vegetables grown at different sites; using the formula initially proposed by Usero *et al.*, The HPI was calculated as the geometric mean of the metal concentrations in the edible sections of the plants at each site 24.

HPI = (Cf1× Cf2×Cf3×..........Cfn)^1/n

Cf is the Concentration of ‘nth' heavy metal in vegetable samples. So, in our study, n=7, we analysed 8 vegetable samples for seven metals. Results are given in Tables 2, 3, 4, 5 and Fig. 3 for all four sites. The HPI indicated the following order for site-wise heavy metal toxicity- Site 4 > Site 3 > Site 2 > Site 1, which was also confirmed by the high frequencies of heavy metal ions observed in the same areas.

CONCLUSION

The study was based on HPI, AAS and ICP-MS studies. The range of HPI in carrot, radish, tomato, coriander, spinach, radish leaves, mustard and wild spinach was between 0.2169-0.5344, 0-1.8589, 0.4997-0.7829, 0.2875-0.6729, 0-0.4058, 0.2609-0.9909, 0.2227-0.9801 and 0.3653-1.0777 respectively. Site 4 samples were the most toxic, and the highest levels of heavy metals were found in radish among all the sites. The overall order of observed heavy metal concentration site-wise was- site 4 > site 3 > site 2 > site 1. This was also confirmed by the AAS and ICP-MS analysis. In our study, Zn and Cu were found within limits, whereas Fe, Cd, Cr, Pb, and Ni were found 3-10 times more in concentration than the prescribed limits.

There has been an alarming increase in cancer, bronchial issues, and neurological disorders in Jodhpur City in the past few years[25-28] Site four where vegetables were most toxic is a slum area, where mostly lower-income group people reside, they buy these toxic cheap vegetables from local vendors near residential areas, small shops, and community markets where vegetables are brought from the areas irrigated with toxic effluent waters could well be due to severe and pressing issues of heavy metal toxicity exposure among the general population. Enforcing a strict ban on irrigation with effluent water should be the utmost priority. We also recommend continuous monitoring of these heavy metals in vegetables sold in the local market to keep a check on the toxicological changes. People should be aware of the health-related issues from heavy metals consumption in vegetables. Implementing these measures can save people from life-threatening diseases caused by toxic vegetables.

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Conflict of interest

The authors declare no conflict of interest in the present work.

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