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Heavy Metal Accumulation in Fruits and Vegetables and Human Health Risk Assessment: Findings From Maharashtra, India

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ABSTRACT

BACKGROUND: Vegetables are consumed enormously by humans all over the world. Consumption of contaminated fruits and vegetables is the most likely route of heavy metal exposure. Hence, it is important to quantify heavy metal concentration in frequently consumed fruits and vegetables.

MATERIALS AND METHODS: The main aim of our study is to investigate heavy metal (Pb, Cd, As, and Hg) contamination in 24 different kinds of vegetables and fruits grown in the industrialized city of Solapur, Maharashtra, India. Potential health risks due to the consumption of fruits and vegetables were assessed. Heavy metal concentration and quality of native soil were also determined. Vegetable and fruit samples were analyzed using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) Agilent 7500.

RESULTS: The mean concentrations of selected heavy metals in fruits and vegetables analyzed were: Lead (0.17 ± 0.38 mg/kg) > Mercury (0.06 ± 0.09 mg/kg) > Cadmium (0.02 ± 0.007 mg/kg) > Arsenic (0.002 ± 0.003 mg/kg). Among them, garlic showed the highest heavy metal accumulation followed by potato.

CONCLUSION: Overall, vegetables showed higher metal accumulations than fruits. Some vegetables showed alarming levels of human health risk indices such as the Metal Pollution Index (MPI), Health Risk Index (HRI) and Hazard Index (HI), suggesting that reducing the intake amount of these vegetables may lower the adverse health effects.

KEYWORDS: Human health risk assessment, heavy metals, vegetables, fruits, soil

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Introduction

Vegetables and fruits are the major components of the human diet, as it is the source of essential micronutrients like copper (Cu), zinc (Zn), calcium (Ca), iron (Fe), magnesium (Mg), iodine (I), sodium (Na), potassium (K), antioxidants, vitamins, and various metabolites.^{1,2} They are consumed in both cooked and raw forms, thus vegetables containing toxic metals can cause detrimental effects on human health. Consumption of heavy metals through the food chain has been thoroughly documented worldwide.^{3–6} Plants can absorb heavy metals from soil and build up in higher concentrations in them.^{7–9} Heavy metal pollution in the soil is the main contributing factor to the bioaccumulation of metals in plants.¹⁰ Meanwhile, as the economy develops and industrialization, urbanization, and agricultural modernization accelerate, the risk of polluted farmland increases.¹¹ Anthropogenic activities determine heavy metal contamination in the soil surface, but geological background also has a role to play.⁵ The basic

primal material of soil is rock, which is broken down by weathering to generate loose debris known as soil parent material, the physical and chemical properties of which vary. Previous research has discovered that the soil parent material is a significant natural source of heavy metals, determining the soil's initial heavy metal composition.^{12,13}

Heavy metals such as Cu, Zn, and Ni, when present in trace amounts, are useful as micronutrients for the growth of human beings.¹ However, some toxic non-essential heavy metals, such as lead (Pb), mercury (Hg), cadmium (Cd), and arsenic (As), are detrimental to human health even at trace levels, particularly in pregnant women and young children, who are more susceptible to heavy metal toxicity.^{1,14,15} Cd has the greatest potential for transmission through the food chain since it is more mobile in soil and plant than any other heavy metal.¹⁶ Increased intake of Cd causes damage to the liver, lung, kidney and bones. Cd increases the risk of developing pulmonary adenocarcinoma and proliferative prostatic lesions and hampers



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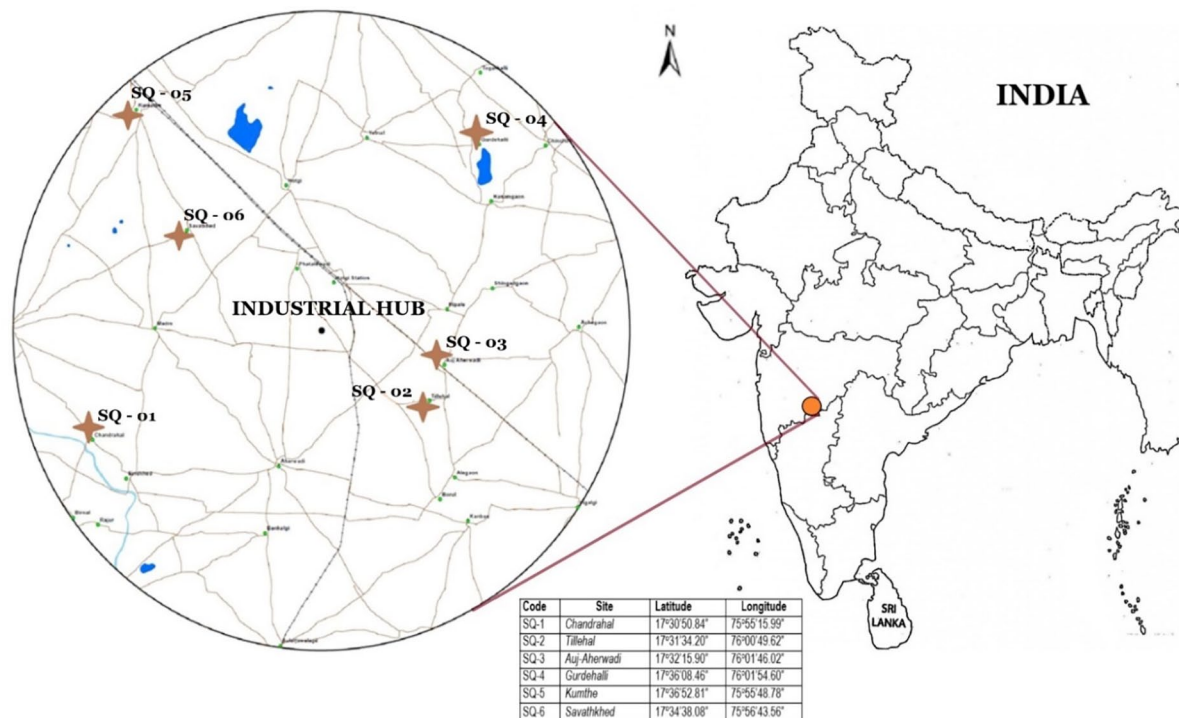


Figure 1. Study area and sampling locations.

the immune system.^{17,18} Excessive levels of blood Pb can induce hypertension, impair the skeletal, immune and endocrine system, and lower intellectual capacity in children. It also disrupts renal and heart functions in adults.^{2,19,20} Toxicity of As depends on the nature of the compound and its valency. Trivalent and inorganic compounds of As are more hazardous than organic, pentavalent compounds. As is a carcinogen that has a variety of acute and long-term detrimental impacts on human health.²¹ Hg increases reactive oxygen species (ROS) generation, which causes widespread oxidative damage. Covalent bonds are formed between cysteine residues of protein and Hg (II), which forms a chemical carcinogen and initiates carcinogenesis.^{3,22,23} Also, heavy metal contamination in soil affects the productivity and fertility of the farmlands.²⁴

Central Pollution Control Board (CPCB) reported that the states of Gujarat, Maharashtra, and Andhra Pradesh contribute to 80% of hazardous waste including heavy metal pollution in India.²⁵ Solapur which is located in the Indian state of Maharashtra is home to many textile industries.²⁶ Studies have shown the role of textile industries in heavy metal pollution.^{27,28} Vegetables and fruits are the most common source of heavy metal exposure in humans, accounting for 90% of metal intake, with the remaining 10% coming via skin contact and inhalation of contaminated dust.²⁹⁻³² Therefore, quantification of heavy metal concentration in frequently consumed vegetables and fruits is necessary to assess the potential risk to human health.³³ Previous studies have mainly focused on a few kinds of vegetables.³⁴⁻³⁶ To the best of our knowledge, no studies have reported heavy metal accumulation in fruits and vegetables of Solapur region. Also, heavy metal contamination and public

health problems are more pronounced in developing nations compared to developed ones.³⁷⁻⁴⁰ Hence, human health risk assessment of heavy metals through commonly consumed vegetables and fruits is necessary. The main objectives of the study were, (1) to investigate the soil quality and selected heavy metal contamination in the study area, (2) to investigate heavy metal accumulation (Pb, Cd, As and Hg) in locally grown 24 different kinds of vegetables and fruits, and (3) to evaluate the potential health risk caused by these vegetables and fruits.

Materials and Methods

Study area and sample collection

The study was conducted in the industrialized city of Solapur district, which is located in the Indian state of Maharashtra. Handlooms and power-looms are the main industries in the area.^{26,41} The city experiences a tropical monsoon-type climate.⁴²

Soil sampling was carried out at 6 locations around a 10 km radius of the industrial hub for 2 seasons viz. winter (January) and summer (April) in the year 2017 to understand soil quality (Figure 1). After removing the surface vegetation cover, visible roots, plant litter, soil fauna, stones, plastic materials and artificial matter, soil samples were collected using stainless steel auger soil sampler at a depth of 50, 150, and 300 cm and mixed thoroughly and analyzed as a single unit sample. The land area near the root zones and foundations of civil constructions were avoided. Samples were analyzed for physical parameters [texture, bulk density (%) and porosity (%)] and heavy metals content (As, Cd, Cr⁶⁺, Cu, Hg, Mn, Ni, Pb, and Zn) (mg/kg).

Table 1. Names of vegetables and Fruits analyzed.

NAME OF VEGETABLES	SCIENTIFIC NAME
Cabbage	<i>Brassica oleracea</i>
Carrot	<i>Daucus carota</i>
Fenugreek (Methi-leafs)	<i>Trigonella foenum-graecum</i>
Garlic	<i>Allium sativum</i>
Ginger	<i>Zingiber officinale</i>
Potato	<i>Solanum tuberosum</i>
Radish	<i>Raphanus sativus</i>
Onions	<i>Allium cepa</i>
Sorghum	<i>Sorghum arundinaceum</i>
Sugarcane	<i>Saccharum officinarum</i>
NAME OF FRUITS	SCIENTIFIC NAME
Apple	<i>Malus domestica</i>
Brinjal	<i>Solanum melongena</i>
Broad Beans	<i>Vicia faba</i>
Chick Pea	<i>Cicer arietinum</i>
Cucumber	<i>Cucumis sativus</i>
Cream Beans	<i>Mucuna pruriens</i>
Green Peas	<i>Pisum sativum</i>
Mung beans	<i>Vigna radiata</i>
Okra	<i>Abelmoschus esculentus</i>
Orange	<i>Citrus sinensis</i>
Papaya	<i>Carica papaya</i>
Red gram	<i>Cajanus cajan</i>
Tamarind	<i>Tamarindus Indica</i>
Tomatoes	<i>Solanum lycopersicum</i>

An adequate amount (1 kg) of soil samples was collected from each site. The samples were packed in a waterproof zip lock pouch bag and transported to the laboratory for testing.

Frequently consumed vegetables and fruits (Table 1) by the human population grown in the agricultural land within the study area were collected from farms located in Chandrahah (SQ-1) (Figure 1) for investigating the concentration of As, Cd, Hg, and Pb.

Sample preparation and analysis

Vegetables and fruit samples were washed with distilled water and then with deionized water to remove various airborne and soil contaminants. Then the samples were air-dried, crushed, and stored at room temperature in the laboratory. For analysis, 1 to 2 g of samples were digested with the repeated infusion of

conc. HNO₃, conc. HCl, and 30% H₂O₂ along with heating as indicated in EPA method 3050B⁴³ and FSSAI guidelines.⁴⁴ After cooling, the solution was diluted by adding deionized water to 100 ml and then centrifuged. The obtained solutions were analyzed for heavy metals (Pb, Cd, As, Hg) using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) Agilent 7500. Laboratory equipment and utensils were washed with soap and water, then rinsed with 1.5% v/v HNO₃ and distilled water after every use to prevent any contamination.

Human health risk assessment

Metal Pollution Index (MPI). The overall heavy metal concentration (Pb, Cd, As, and Hg) contained in vegetable and fruit samples were estimated by Metal Pollution Index (MPI). This index was calculated using the equation⁴⁵:

$$MPI \text{ (mg / kg)} = (Cf_1 \times Cf_2 \times \dots \times Cf_n)^{1/n}$$

Where Cf_n = concentration of metal in nth sample.

Daily intake of metal (DIM) and Health Risk Index (HRI). To calculate the human exposure dose, the daily intake of metal was calculated by using the equation^{6,46}:

$$\text{Daily intake of metal (DIM)} = (C_{\text{metal}} \times D_{\text{food intake}}) / B_{\text{average weight}}$$

Where, C_{metal} = Concentration of heavy metals in vegetables (mg/kg), D_{food intake} = Daily intake of vegetables (average consumption = 0.280 kg/person/day),⁴⁷ B_{average weight} = Average body weight "56 kg/person."⁴⁸

The ratio of estimated test vegetables and fruits exposure to oral reference dose was used to estimate the health risk index.⁴⁹ The health risk index is also known as Hazard Quotient (HQ). It indicates potential hazards to human health. The index was calculated using the equation:

$$HRI = DIM / RfD$$

The oral reference dose (RfD) of As, Cd, Hg, and Pb are 0.0003,⁵⁰ 0.001,⁵⁰ 0.01, and 0.004 mg/kg/day,⁵¹ respectively.

Hazard Index (HI). To evaluate the potential risk to human health through more than one heavy metal, the hazard index (HI) has been developed.⁵² The hazard index was calculated using the equation:

$$HI = \sum HQ = HQ_{Hg} + HQ_{Cd} + HQ_{As} + HQ_{Pb}$$

It is assumed that magnitude of the adverse effect will be proportional to the sum of multiple metal exposure. It also assumes similar working mechanisms that linearly affect the target organ.⁵³ When the value of HI > 1, there is a greater possibility of non-carcinogenic health effects, and the

probability increases with a rising value of HI.^{3,54} The hazard index of the toxic elements (As, Cd, Hg, and Pb) has been calculated in the present study.

Results and Discussion

Soil parameters

The results of all the specified parameters at each location were then compiled for both seasons and are tabulated in Table 2.

The soil samples were observed to be neutral to slightly basic in nature with pH ranging from 7.64 to 8.0 for both seasons. The soil samples were mostly sandy clay and sandy clay loam in texture. Sand percentage varied between 55.0% and 67.5%, silt content was between 1.5% and 5.7% and clay content was between 29% and 42.5%. The bulk density varied between 1.36% and 1.50% and porosity was between 42% and 47% for both seasons.

Table 2. Soil characteristics and heavy metal concentration.

PARAMETER	UNIT	SQ-1		SQ-2		SQ-3	
		WINTER	SUMMER	WINTER	SUMMER	WINTER	SUMMER
<i>pH</i>	–	7.8	7.64	7.8	7.69	7.8	7.69
<i>Bulk Density</i>	%	1.36	1.42	1.4	1.41	1.4	1.41
<i>Porosity</i>	%	44	42	45	44	45	44
<i>Texture</i>	–	S. Clay	S. Clay	S. Cl. Loam	S. Cl. Loam	S. Cl. Loam	S. Cl. Loam
<i>Sand</i>	%	57.5	57	61.8	62.5	61.8	62.5
<i>Clay</i>	%	40	39.5	32.5	32	32.5	32
<i>Silt</i>	%	2.5	3.5	5.7	5.5	5.7	5.5
<i>Hg</i>	mg/kg	<0.01	<0.01	Nil	<0.01	Nil	<0.01
<i>As</i>	mg/kg	<0.5	<0.5	Nil	<0.5	Nil	<0.5
<i>Cd</i>	mg/kg	<0.01	<0.01	Nil	<0.01	Nil	<0.01
<i>Pb</i>	mg/kg	12	9.6	5	4.1	5	4.1
<i>Ni</i>	mg/kg	<0.05	<0.05	Nil	<0.05	Nil	<0.05
<i>Cu</i>	mg/kg	11	10.4	4	4.7	4	4.7
<i>Zn</i>	mg/kg	24	20.6	21	20.8	21	20.8
<i>Mn</i>	mg/kg	5	1.3	Nil	1.8	Nil	1.8
<i>Cr</i>	mg/kg	<0.1	<0.1	Nil	<0.1	Nil	<0.1
PARAMETER	UNIT	SQ-4		SQ-5		SQ-6	
		WINTER	SUMMER	WINTER	SUMMER	WINTER	SUMMER
<i>pH</i>	–	7.8	7.69	7.72	7.84	7.76	7.79
<i>Bulk Density</i>	%	1.4	1.41	1.43	1.46	1.48	1.47
<i>Porosity</i>	%	45	44	47	46	46	44
<i>Texture</i>	–	S. Cl. Loam	S. Cl. Loam	S. Clay	S. Clay	S. Cl. Loam	S. Cl. Loam
<i>Sand</i>	%	61.8	62.5	57.5	58	67	67.5
<i>Clay</i>	%	32.5	32	39.5	38.5	29.9	29
<i>Silt</i>	%	5.7	5.5	3	3.5	3.1	3.5
<i>Hg</i>	mg/kg	Nil	<0.01	<0.01	<0.01	<0.01	<0.01
<i>As</i>	mg/kg	Nil	<0.5	<0.5	<0.5	<0.5	<0.5

(Continued)

Table 2. (Continued)

PARAMETER	UNIT	SQ-4		SQ-5		SQ-6	
		WINTER	SUMMER	WINTER	SUMMER	WINTER	SUMMER
Cd	mg/kg	Nil	<0.01	<0.01	<0.01	<0.01	<0.01
Pb	mg/kg	5	4.1	14	11.3	16	14.5
Ni	mg/kg	Nil	<0.05	<0.05	<0.05	<0.05	<0.05
Cu	mg/kg	4	4.7	9	8.3	14	12.6
Zn	mg/kg	21	20.8	25	23.7	28	26.2
Mn	mg/kg	Nil	1.8	Nil	1.1	2	1.8
Cr	mg/kg	Nil	<0.1	<0.1	<0.1	<0.1	<0.1

Table 3. Standard levels of selected heavy metals (mg/kg) in soil, fruits, and vegetables.

	SOIL		FRUITS AND VEGETABLES	
	INDIAN STANDARD ⁵⁵	EU STANDARD ⁵⁶	INDIAN STANDARD ⁵⁵	WHO/FAO STANDARD ⁷⁵
Pb	250-500	60	2.5	5.0
Cd	3-6	1	1.5	0.2
Hg	—	0.5	—	0.03
As	—	5	—	0.5
Zn	300-600	200		
Ni	750-150	50		
Cu	135-270	100		
Mn	—	—		
Cr	—	100		

The values for the Pb for all locations vary between 4.1 and 16 mg/kg, Cu ranges from 2.0 to 14 mg/kg, Zn from 6.0 to 28 mg/kg, and Mn ranges between 1.1 and 2.0 during both seasons (Table 2). Heavy metal concentrations were found to be below the permissible levels set by Indian⁵⁵ and European standards⁵⁶ (Table 3). The mean concentration of heavy metals in soil shows the trend: Zn > Cu > Pb > Mn.

Soil pH plays a critical role in determining solubility, mobility, and final bioavailability of metals.⁵⁷⁻⁵⁹ Numerous studies have found a negative correlation between soil pH and heavy metal mobility and availability to plants. For example, in the case of Cd, Pb, and Zn, decreasing soil pH resulted in substantial increases in heavy metal desorption from soil components and dissolution in soil solution.^{60,61} Reduced soil pH increases heavy metal mobility and bioavailability,⁶²⁻⁶⁴ promoting heavy metal absorption by plants and posing a health risk.⁶⁵ Also, the particle size of soil has been shown to have a significant impact on heavy metal concentrations in soils.^{66,67} Due to greater surface areas, the finer particles show higher concentrations of

heavy metals.^{68,69} In addition, it was reported that in sandy clay soil, soil pH appeared to be the most important predictor of Cr, Pb, and Zn solubility and mobility.⁷⁰ In comparison to other sampling sites with sandy clay loam soil, Chandrahah (SQ-1) and Kumthe (SQ-5) with sandy clay soil show higher Pb content (Table 2). Additionally, soil pH varied very slightly across all locations, and no discernible variation in the quantities of selected heavy metals were observed.

Heavy metals in fruits and vegetables

Heavy metal contamination was found in nearly all samples examined. Some items contain a higher proportion of one metal compared to the others. This can be explained by differential absorptions of heavy metal from soil and subsequent translocation within the plant.^{6,71,72} Table 4 represents the concentration of heavy metals in fruit and vegetable samples. Among them, sugarcane has the highest Pb accumulation (1.741 mg/kg) while garlic has the highest Cd accumulation (0.37 mg/kg). Sugarcane has preferential absorption for Pb;

Table 4. Concentration of heavy metals, MPI, DIM, HRI and HI of different fruits and vegetables.

NAME OF THE VEGETABLES AND FRUITS	LEAD (MG/KG)	CADMIUM (MG/KG)	ARSENIC (MG/KG)	MERCURY (MG/KG)	GEO MEAN OF ALL METAL (MPI)	DIM LEAD (MG/KG)	DIM CADMIUM (MG/KG)	DIM ARSENIC (MG/KG)	DIM MERCURY (MG/KG)	HRI CD	HRI PB	HRI AS	HRI HG	HI
Carrot	0.087	0.008	0.013	0.001	0.009753	0.000435	0.00004	0.000065	0.000005	0.04	0.10875	0.2166667	0.0005	0.3659167
Radish	0.1	0.008	0.004	0.052	0.0201971	0.0005	0.00004	0.00002	0.00026	0.04	0.125	0.0666667	0.026	0.2576667
Ginger	0.11	0.001	0.005	0.14	0.016658	0.00055	0.000005	0.000025	0.0007	0.005	0.1375	0.0833333	0.07	0.2958333
Potato	0.542	0.023	0.007	0.013	0.0326356	0.00271	0.000115	0.000035	0.000065	0.115	0.6775	0.1166667	0.0065	0.9156667
Cabbage	0.003	0.006	0.002	0.001	0.0024495	0.000015	0.00003	0.00001	0.000005	0.03	0.00375	0.0333333	0.0005	0.0675833
Garlic	0.774	0.37	0.007	0.123	0.1253102	0.00387	0.00185	0.000035	0.000615	1.85	0.9675	0.1166667	0.0615	2.9956667
Fenugreek	0.001	0.003	0.001	0.235	0.0051528	0.000005	0.000015	0.000005	0.001175	0.015	0.00125	0.0166667	0.1175	0.1504167
Sugarcane	1.741	0.001	0.001	0.035	0.0157115	0.008705	0.000005	0.000005	0.000175	0.005	2.17625	0.0166667	0.0175	2.2154167
Sorghum	0.034	0.001	0.001	0.356	0.010489	0.00017	0.000005	0.000005	0.00178	0.005	0.0425	0.0166667	0.178	0.2421667
Onions	0.046	0.009	0.001	0.001	0.0045108	0.00023	0.000045	0.000005	0.000005	0.045	0.0575	0.0166667	0.0005	0.1196667
Orange	0.002	0.001	0.005	0.007	0.0028925	0.00001	0.000005	0.000025	0.000035	0.005	0.0025	0.0833333	0.0035	0.0943333
Papaya	0.019	0.001	0.001	0.016	0.0041756	0.000095	0.000005	0.000005	0.00008	0.005	0.02375	0.0166667	0.008	0.0534167
Cream Beans	0.001	0.002	0.001	0.05	0.0031623	0.000005	0.00001	0.000005	0.00025	0.01	0.00125	0.0166667	0.025	0.0529167
Apple	0.068	0.002	0.003	0.001	0.0044943	0.00034	0.00001	0.000015	0.000005	0.01	0.085	0.05	0.0005	0.1455
Brinjal	0.053	0.004	0.001	0.111	0.0123855	0.000265	0.00002	0.000005	0.000555	0.02	0.06625	0.0166667	0.0555	0.1584167
Cucumber	0.005	0.0005	0.001	0.046	0.0032747	0.000025	0.0000025	0.000005	0.00023	0.0025	0.00625	0.0166667	0.023	0.0484167
Tomatoes	0.063	0.008	0.001	0.001	0.0047381	0.000315	0.00004	0.000005	0.000005	0.04	0.07875	0.0166667	0.0005	0.1359167
Okra	0.149	0.005	0.001	0.17	0.0188648	0.000745	0.000025	0.000005	0.00085	0.025	0.18625	0.0166667	0.085	0.3129167
Mung beans	0.007	0.004	0.004	0.012	0.0060548	0.000035	0.00002	0.00002	0.00006	0.02	0.00875	0.0666667	0.006	0.1014167
Tamarind	0.244	0.001	0.001	0.147	0.0137618	0.00122	0.000005	0.000005	0.000735	0.005	0.305	0.0166667	0.0735	0.4001667
Green Peas	0.022	0.001	0.004	0.001	0.0030628	0.00011	0.000005	0.00002	0.000005	0.005	0.0275	0.0666667	0.0005	0.0996667
Broad Beans	0.003	0.021	0.002	0.015	0.0065935	0.000015	0.000105	0.00001	0.000075	0.105	0.00375	0.0333333	0.0075	0.1495833
Chick Pea	0.001	0.034	0.001	0.001	0.0024147	0.000005	0.00017	0.000005	0.000005	0.17	0.00125	0.0166667	0.0005	0.1884167
Red gram	0.086	0.007	0.003	0.001	0.006519	0.00043	0.000035	0.000015	0.000005	0.035	0.1075	0.05	0.0005	0.193

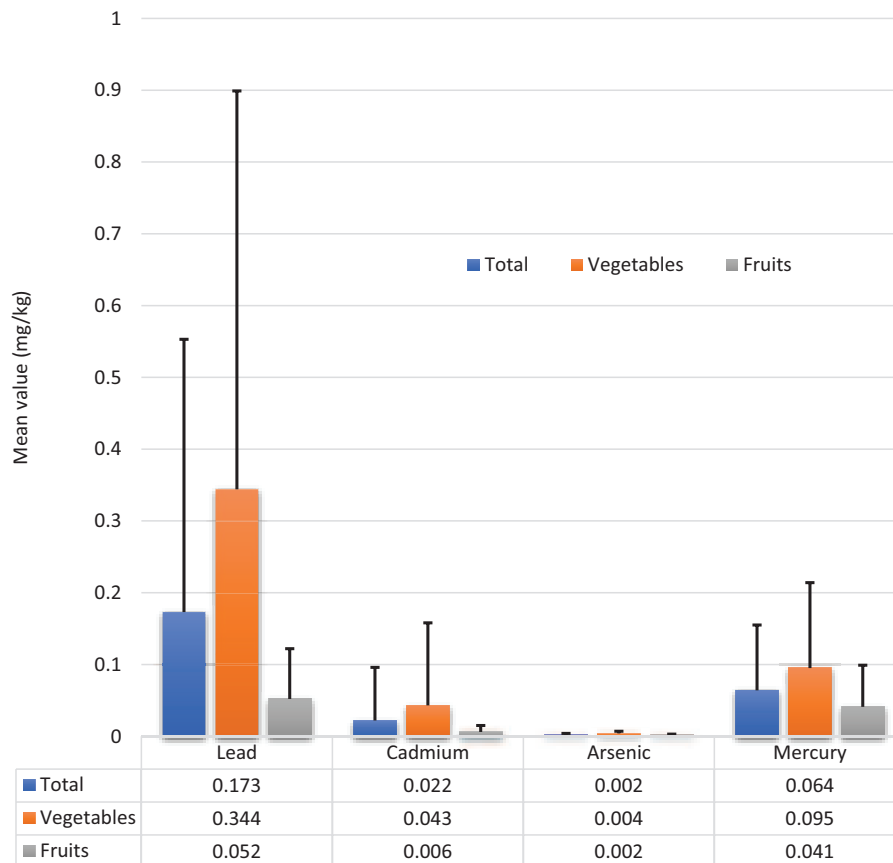


Figure 2. Level of heavy metals in fruits and vegetables.

hence it is used to reduce the bioavailability of Pb in soil.⁷³ Garlic has a higher mean concentration of Cd and Pb than other fruits and vegetables, suggesting it has the potential to hyper-accumulate Cd and Pb. Similar observations about garlic were reported earlier and this principle is used for phytoremediation of soil.⁷⁴

In the majority of fruits and vegetables, heavy metal concentrations were found to be within permissible levels set by Indian⁵⁵ and WHO/FAO standards.⁷⁵ The level of Cd in garlic (0.37 mg/kg) exceeds the WHO/FAO standard (0.2 mg/kg) but was within the Indian threshold (1.5 mg/kg). While in garlic (0.123 mg/kg), okra (0.17 mg/kg), fenugreek (0.235 mg/kg), sugarcane (0.035 mg/kg), tamarind (0.147 mg/kg), and sorghum (0.356 mg/kg) Hg concentration exceeds WHO/FAO standard (0.03 mg/kg).

It was found that selected heavy metal concentration in fruits and vegetables was not proportionate to the native soil. For example, Cd levels in soil samples were found to be below detectable levels, but Cd levels in garlic were found to be considerable. This might be related to garlic's tendency to hyper-accumulate Cd from the soil,⁷⁴ as well as other probable sources of Cd exposure, such as wastewater irrigation.⁶

Fruits showed lower metal accumulation compared to vegetables. Figure 2 represents the mean concentration of metals in fruits and vegetables. Similar observations were reported widely suggesting vegetables have a higher heavy metal accumulation

capacity than fruits.^{36,76-78} There are several reasons given to explain this phenomenon. Firstly, leafy vegetables have higher growth and transpiration rates than fruits. It facilitates increased heavy metal absorption through roots and subsequent translocation from roots to leaves. Secondly, in fruit plants, large quantities of heavy metals absorbed are stored in other organs, especially leaves than the edible part. Thirdly, leaves have a higher surface area, which makes vegetables more vulnerable to physical contamination by rainwater and dust from soil.^{76,79}

Human health risk assessment

MPI varied from 0.002414736 (Chickpea) to 0.12531021 mg/kg (Garlic). Figure 3 represents the MPI of different fruit and vegetable samples examined. A higher value of MPI signifies a greater concentration of heavy metal pollution. If vegetables and fruits with higher MPI values are consumed in greater quantities, it can lead to higher heavy metal accumulation in the body. In our study, among different vegetables, garlic showed the highest value of MPI followed by potato (Table 4). As compared to vegetables, fruits showed a lower metal pollution index which is explained by the lower metal accumulative capacity of fruits compared to vegetables.^{76,79}

HRI of Pb, As, Cd, and Hg were determined for both vegetable and fruit samples (Table 4). In our study, HRI of more than 1 was noted in garlic (HRI Cd=1.85) and sugarcane

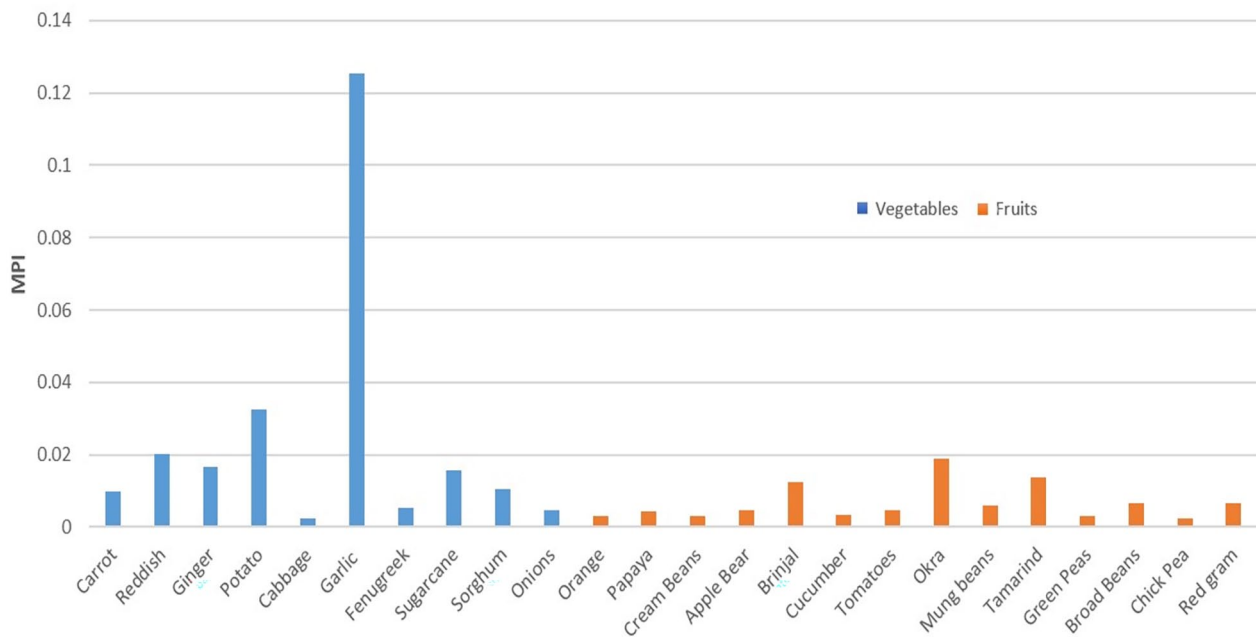


Figure 3. Metal Pollution Index (MPI) in different vegetables and fruits analyzed.

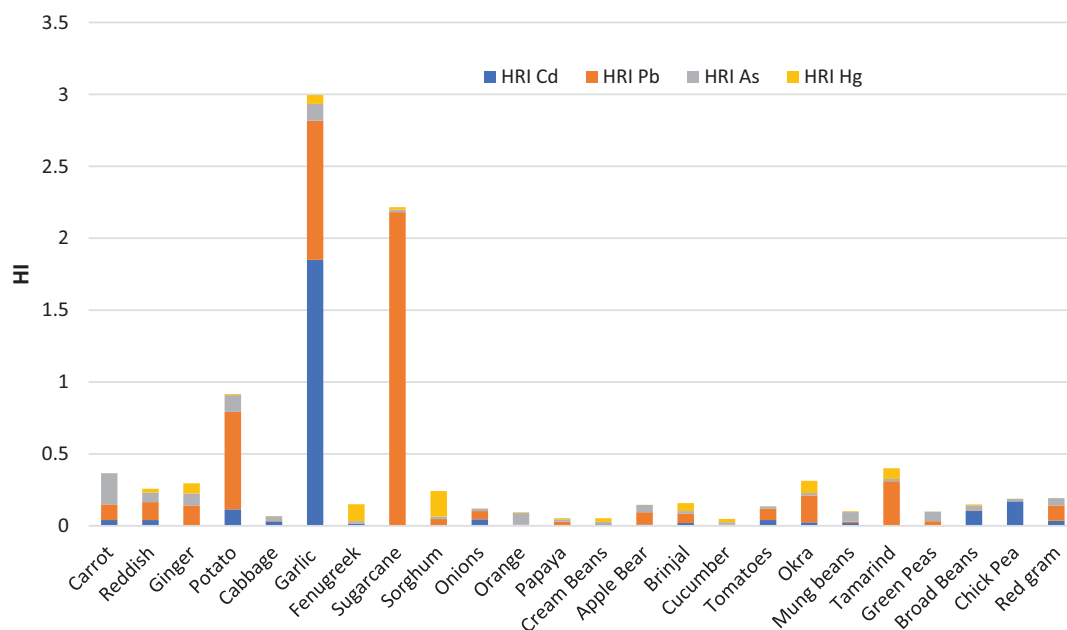


Figure 4. Hazard Index of toxic elements (Hg, As, Cd, Pb) for different vegetables.

(HRI Pb = 2.176). HI varied from 0.048416667 (Cucumber) to 2.99566667 (Garlic). Figure 4 represents the HI of various vegetables and fruits examined. For reference, an index of more than 1 is considered not safe for human health (USEPA, 2002). The hazard index gives an overall estimation of adverse health effects. A higher hazard index means more magnitude of health problems.^{3,54} HI of garlic is mainly determined by Cd followed by Pb whereas, in the case of sugarcane and potato, HI is determined by Pb (Figure 4). The result showed HI of Garlic > Sugar cane > Potato. Hence these vegetables cause more human health risks compared to others.

The study only included the human health risk assessment of heavy metals (Pb, Cd, As, and Hg) through contaminated

fruit and vegetable consumption. There are other ways of heavy metal exposure such as inhalation, dermal contact and ingestion of contaminated soil (for children). All these factors may increase the overall heavy metal intake but were not considered in this study.

Conclusion

The result of this study indicated that the concentration of heavy metals in soil was below the permissible levels. Heavy metal (As, Cd, Hg, and Pb) accumulation of fruits and vegetables was not proportional to the native soil. Cd was below detectable limits in soil samples, whereas garlic contains Cd accumulation. Hence, the soil may not be the only source of

heavy metals, other possible factors of heavy metal exposure to fruits and vegetables such as wastewater irrigation should be examined.

From the overall study, it can be revealed that heavy metal accumulation in the majority of vegetables and fruits of Solapur district is within permissible levels. In some vegetables, the MPI, HRI, and HI values for some elements are at alarming levels, and the adverse effect can be reduced by consuming those vegetables in lesser amounts. Moreover, it is suggested that regular monitoring should be enforced in this area as metal accumulation can be toxic to the consumers when they are present in excess or cause different diseases when present in quantities not suitable for the human body.

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Author Contributions

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Ethical Approval

Instructional Ethical Committee Maulana Azad Medical college.

Informed Consent

Informed consent taken from the patient.

Registry and the Registration No. of the Study/Trial


N/A

Animal Studies

NO

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REFERENCES

- Liang G, Gong W, Li B, Zuo J, Pan L, Liu X. Analysis of heavy metals in foodstuffs and an assessment of the health risks to the general public via consumption in Beijing, China. *Int J Environ Res Public Health*. 2019;16:909.
- Huang Z, Pan XD, Wu PG, Han JL, Chen Q. Heavy metals in vegetables and the health risk to population in Zhejiang, China. *Food Control*. 2014;36:248-252.
- Ahmad W, Alharthy RD, Zubair M, Ahmed M, Hameed A, Rafique S. Toxic and heavy metals contamination assessment in soil and water to evaluate human health risk. *Sci Rep*. 2021;11:17006.
- Muchuweti M, Birkett JW, Chinyanga E, Zvauya R, Scrimshaw MD, Lester JN. Heavy metal content of vegetables irrigated with mixtures of wastewater and sewage sludge in Zimbabwe: Implications for human health. *Agric Ecosyst Environ*. 2006;112:41-48.
- Gao Z, Dong H, Wang S, et al. Geochemical characteristics and ecological risk assessment of heavy metals in surface soil of Gaomi City. *Int J Environ Res Public Health*. 2021;18:8329.
- Singh A, Sharma RK, Agrawal M, Marshall FM. Health risk assessment of heavy metals via dietary intake of foodstuffs from the wastewater irrigated site of a dry tropical area of India. *Food Chem Toxicol*. 2010;48:611-619.
- Tang M, Lu G, Fan B, Xiang W, Bao Z. Bioaccumulation and risk assessment of heavy metals in soil-crop systems in Liujiang karst area, southwestern China. *Environ Sci Pollut Res Int*. 2021;28:9657-9669.
- Ding Z, Li Y, Sun Q, Zhang H. Trace elements in soils and selected agricultural plants in the Tongling mining area of China. *Int J Environ Res Public Health*. 2018;15:202.
- Mahar A, Wang P, Ali A, et al. Challenges and opportunities in the phytoremediation of heavy metals contaminated soils: a review. *Ecotoxicol Environ Saf*. 2016;126:111-121.
- Khan A, Khan S, Khan MA, Qamar Z, Waqas M. The uptake and bioaccumulation of heavy metals by food plants, their effects on plants nutrients, and associated health risk: a review. *Environ Sci Pollut Res*. 2015;22:13772-13799.
- Chen WX, Li Q, Wang Z, Sun ZJ. [Spatial distribution characteristics and pollution evaluation of heavy metals in arable land soil of China]. *Huan Jing Ke Xue*. 2020;41:2822-2833.
- Facchinelli A, Sacchi E, Mallen L. Multivariate statistical and GIS-based approach to identify heavy metal sources in soils. *Environ Pollut*. 2001;114:313-324.
- Franco-Uría A, López-Mateo C, Roca E, Fernández-Marcos ML. Source identification of heavy metals in pastureland by multivariate analysis in NW Spain. *J Hazard Mater*. 2009;165:1008-1015.
- Kim Y, Ha EH, Park H, et al. Prenatal lead and cadmium co-exposure and infant neurodevelopment at 6 months of age: the Mothers and Children's Environmental Health (MOCEH) study. *Neurotoxicol*. 2013;35:15-22.
- Reuben A, Caspi A, Belsky DW, et al. Association of childhood blood lead levels with cognitive function and socioeconomic status at age 38 years and with IQ change and socioeconomic mobility between childhood and adulthood. *JAMA*. 2017;317:1244-1251.
- Li Y, McCrory DF, Powell JM, Saam H, Jackson-Smith D. A survey of selected heavy metal concentrations in Wisconsin dairy feeds. *J Dairy Sci*. 2005;88:2911-2922.
- Klaassen CD, Liu J, Diwan BA. Metallothionein protection of cadmium toxicity. *Toxicol Appl Pharmacol*. 2009;238:215-220.
- Lin YS, Caffrey JL, Lin JW, et al. Increased risk of cancer mortality associated with cadmium exposures in older Americans with low zinc intake. *J Toxicol Environ Health A*. 2013;76:1-15.
- Navas-Acien A, Guallar E, Silbergeld EK, Rothenberg SJ. Lead exposure and cardiovascular disease--a systematic review. *Environ Health Perspect*. 2007;115:472-482.
- Nieboer E, Tsuji LJ, Martin ID, Liberda EN. Human biomonitoring issues related to lead exposure. *Environ Sci Process Impacts*. 2013;15:1824-1829.
- Medina-Pizzali M, Robles P, Mendoza M, Torres C. [Arsenic intake: Impact in Human Nutrition and health]. *Rev Peru Med Exp Salud Publica*. 2018;35:93-102.
- Valko M, Rhodes CJ, Moncol J, Izakovic M, Mazur M. Free radicals, metals and antioxidants in oxidative stress-induced cancer. *Chem Biol Interact*. 2006;160:1-40.
- Tchounwou PB, Yedjou CG, Patlolla AK, Sutton DJ. Heavy metal toxicity and the environment. *Experientia Suppl*. 2012;101:133-164.
- Sadeghi H, Fazlzadeh M, Zarei A, Mahvi AH, Nazmara S. Spatial distribution and contamination of heavy metals in surface water, groundwater and topsoil surrounding Moghan's tannery site in Ardabil, Iran. *Int J Environ Anal Chem*. 2022;102:1049-1059.
- Marg BZ. *Hazardous Metals and Minerals Pollution in India: Sources, Toxicity and Management. A Position Paper*. Indian National Science Academy; 2011.
- The Gazetteers Department. SOLAPUR; 2006. Accessed May 26, 2022. https://cultural.maharashtra.gov.in/english/gazetteer/Solapur/ind_large%20small.html
- Deepali KK, Gangwar K. Metals concentration in textile and tannery effluents, associated soils and ground water. *NY Sci J*. 2010;3:82-89.
- Kashem MA, Singh BR. Heavy metal contamination of soil and vegetation in the vicinity of industries in Bangladesh. *Water Air Soil Pollut*. 1999;115:347-361.

29. Martorell I, Perelló G, Martí-Cid R, Llobet JM, Castell V, Domingo JL. Human exposure to arsenic, cadmium, mercury, and lead from foods in Catalonia, Spain: Temporal trend. *Biol Trace Elem Res.* 2011;142:309-322.
30. Kim H, Song B, Kim H, Park J. Distribution of trace metals at two abandoned mine sites in Korea and arsenic-associated health risk for the residents. *Toxicol Environ Health Sci.* 2009;1:83-90.
31. Khan S, Reid BJ, Li G, Zhu YG. Application of biochar to soil reduces cancer risk via rice consumption: A case study in Miaoqian village, Longyan, China. *Environ Int.* 2014;68:154-161.
32. Ferré-Huguet N, Martí-Cid R, Schuhmacher M, Domingo JL. Risk assessment of metals from consuming vegetables, fruits and rice grown on soils irrigated with waters of the Ebro River in Catalonia, Spain. *Biol Trace Elem Res.* 2008;123:66-79.
33. González-Muñoz MJ, Peña A, Meseguer I. Monitoring heavy metal contents in food and hair in a sample of young Spanish subjects. *Food Chem Toxicol.* 2008;46:3048-3052.
34. Chen TB, Song B, Zheng YM, et al. A survey of arsenic concentrations in vegetables and soils in Beijing and the potential risks to human health. *Acta Geogr Sin.* 2006;61:297-310.
35. Wang B, Ma Z, Feng X, Wang J. Concentrations and health risk evaluation of heavy metals in vegetables in Beijing. *J Food Saf Qual.* 2015;6:2736-2745.
36. Nolos RC, Agarín CJM, Domino MYR, et al. Health risks due to metal concentrations in soil and vegetables from the Six municipalities of the Island Province in the Philippines. *Int J Environ Res Public Health.* 2022;19:1587.
37. Chen T, Liu X, Li X, et al. Heavy metal sources identification and sampling uncertainty analysis in a field-scale vegetable soil of Hangzhou, China. *Environ Pollut.* 2009;157:1003-1010.
38. Cai L, Xu Z, Bao P, et al. Multivariate and geostatistical analyses of the spatial distribution and source of arsenic and heavy metals in the agricultural soils in Shunde, Southeast China. *J Geobchem Explor.* 2015;148:189-195.
39. Wang L, Tao W, Smardon RC, Xu X, Lu X. Speciation, sources, and risk assessment of heavy metals in suburban vegetable garden soil in Xianyang City, Northwest China. *Earth Sci Front.* 2018;12:397-407.
40. Bayissa LD, Gebeyehu HR. Vegetables contamination by heavy metals and associated health risk to the population in Koka area of central Ethiopia. *PLOS ONE.* 2021;16:e0254236.
41. Solapur. Industries|District Solapur, Govt. of Maharashtra, India; 2006. Accessed May 27, 2022. <https://solapur.gov.in/en/industries/>
42. The Gazetteers Department. SOLAPUR; 2006. Accessed May 26, 2022. https://cultural.maharashtra.gov.in/english/gazetteer/Solapur/agri_rainfall.html
43. USEPA. *EPA Method 3050B: Acid Digestion of Sediments, Sludges.* USEPA; 1996.
44. FSSAI. *Manual of Methods of Analysis of Foods- Metals.* FSSAI; 2016. Accessed April 12, 2022. [https://fssai.gov.in/upload/uploadfiles/files/Manual_Metals_25_05_2016\(1\).pdf](https://fssai.gov.in/upload/uploadfiles/files/Manual_Metals_25_05_2016(1).pdf)
45. Usero J, González-Regalado E, Gracia I. Trace metals in the bivalve molluscs *Ruditapes decussatus* and *Ruditapes philippinarum* from the Atlantic coast of Southern Spain. *Environ Int.* 1997;23:291-298.
46. Rabiul Islam GM, Habib MR, Waid JL, et al. Heavy metal contamination of freshwater prawn (*Macrobrachium rosenbergii*) and prawn feed in Bangladesh: a market-based study to highlight probable health risks. *Chemosphere.* 2017;170:282-289.
47. Guilbert J. The World Health Report 2002-reducing risks, promoting healthy life. *Educ Health Change Learn Pract.* 2003;16:230-230.
48. Gupta SK, Chabukdhara M, Singh J, Bux F. Evaluation and potential health hazard of selected metals in water, sediments, and fish from the Gomti River. *Hum Ecol Risk Assess.* 2015;21:227-240.
49. Cui YJ, Zhu YG, Zhai RH, et al. Transfer of metals from soil to vegetables in an area near a smelter in Nanning, China. *Environ Int.* 2004;30:785-791.
50. Antoine JMR, Fung LAH, Grant CN. Assessment of the potential health risks associated with the aluminium, arsenic, cadmium and lead content in selected fruits and vegetables grown in Jamaica. *Toxicol Rep.* 2017;4:181-187.
51. Wang X, Sato T, Xing B, Tao S. Health risks of heavy metals to the general public in Tianjin, China via consumption of vegetables and fish. *Sci Total Environ.* 2005;350:28-37.
52. United States Environmental Protection Agency (USEPA). *Risk Assessment Guidance for Superfund (RAGS), Volume I: Human Evaluation Manual (HHEM)-Part A, Baseline Risk Assessment.* Office of Emergency and Remedial Response, US Environmental Protection Agency; 1989.
53. Cao LL, Yang J, Wang P, et al. Potentially toxic metal and metalloid fractionation contamination in sediments of Daya Bay, South China Sea. *Kemija Ind.* 2015;64:255-262.
54. Qasemi M, Shams M, Sajjadi SA, et al. Cadmium in groundwater consumed in the rural areas of Gonabad and Bajestan, Iran: Occurrence and Health Risk Assessment. *Biol Trace Elem Res.* 2019;192:106-115.
55. Awashthi SK. *Prevention of Food Adulteration Act No 37 of 1954. Central and State Rules as Amended for 1999.* Ashoka Law House; 2000.
56. Union E. *Heavy Metals in Wastes.* European Commission on Environment; 2002.
57. Mühlbachová G, Šimon T, Pechová M. The availability of Cd, Pb and Zn and their relationships with soil pH and microbial biomass in soils amended by natural clinoptilolite. *Plant Soil Environ.* 2011;51:26-33.
58. Zhao K, Liu X, Xu J, Selim HM. Heavy metal contaminations in a soil-rice system: Identification of spatial dependence in relation to soil properties of paddy fields. *J Hazard Mater.* 2010;181:778-787.
59. Zeng F, Ali S, Zhang H, et al. The influence of pH and organic matter content in paddy soil on heavy metal availability and their uptake by rice plants. *Environ Pollut.* 2011;159:84-91.
60. Sukreeyapongse O, Holm PE, Strobel BW, Panichsakpatana S, Magid J, Hansen HC. pH-Dependent release of cadmium, copper, and lead from natural and sludge-amended soils. *J Environ Qual.* 2002;31:1901-1909.
61. Bang J, Hesterberg D. Dissolution of trace element contaminants from two coastal plain soils as affected by pH. *J Environ Qual.* 2004;33:891-901.
62. Badawy SH, Helal MI, Chaudri AM, Lawlor K, McGrath SP. Soil solid-phase controls lead activity in Soil Solution. *J Environ Qual.* 2002;31:162-167.
63. Wang AS, Angle JS, Chaney RL, Delorme TA, Reeves RD. Soil pH effects on uptake of Cd and Zn by *Thlaspi caerulescens*. *Plant Soil.* 2006;281:325-337.
64. Du Laing G, Vanthuyne DR, Vandecasteele B, Tack FM, Verloo MG. Influence of hydrological regime on pore water metal concentrations in a contaminated sediment-derived soil. *Environ Pollut.* 2007;147:615-625.
65. Brallier S, Harrison RB, Henry CL, Dongsen X. Liming effects on availability of Cd, Cu, Ni and Zn in a soil amended with sewage sludge 16 years previously. *Water Air Soil Pollut.* 1996;86:195-206.
66. Tiller KG. The geochemistry of basaltic materials and Australia associated soils of south-eastern South Australia. *J Soil Sci.* 1958;9:225-241.
67. Qian J, Shan XQ, Wang ZJ, Tu Q. Distribution and plant availability of heavy metals in different particle-size fractions of soil. *Sci Total Environ.* 1996;187:131-141.
68. Haque MA, Subramanian V, Gibbs RJ. Copper, lead, and zinc pollution of soil environment. *CRC Crit Rev Environ Control.* 1982;12:13-68.
69. Förstner U, Salomons W. Trace metal analysis on polluted sediments. *Environ Technol Lett.* 2008;1:494-505.
70. Speir TW, Van Schaik AP, Percival HJ, Close ME, Pang L. Heavy metals in soil, plants and groundwater following high-rate sewage sludge application to land. *Water Air Soil Pollut.* 2003;150:319-358.
71. Harmanescu M, Alda LM, Bordean DM, Gogoasa I, Gergen I. Heavy Metals Health risk assessment for population via consumption of vegetables grown in old mining area; a case study: Banat County, Romania. *Chem Cent J.* 2011;5:64.
72. Voutsas D, Grimanis A, Samara C. Trace elements in vegetables grown in an industrial area in relation to soil and air particulate matter. *Environ Pollut.* 1996;94:325-335.
73. Puga AP, Abreu CA, Melo LCA, Paz-Ferreiro J, Beesley L. Cadmium, lead, and zinc mobility and plant uptake in a mine soil amended with sugarcane straw biochar. *Environ Sci Pollut Res Int.* 2015;22:17606-17614.
74. Hussain J, Wei X, Xue-Gang L, et al. Garlic (*Allium sativum*) based interplanting alters the heavy metals absorption and bacterial diversity in neighboring plants. *Sci Rep.* 2021;11:5833.
75. WHO/FAO. Joint FAO/WHO Food Standards Programme Codex Alimentarius Commission Twenty-Seventh Session Report of the Thirty-Sixth Session of the Codex Committee on Food Hygiene. Food & Agriculture Org.; 2007.
76. Luo C, Liu C, Wang Y, et al. Heavy metal contamination in soils and vegetables near an e-waste processing site, south China. *J Hazard Mater.* 2011;186:481-490.
77. Pipoyan D, Beglaryan M, Sireyan L, Merendino N. Exposure assessment of potentially toxic trace elements via consumption of fruits and vegetables grown under the impact of Alaverdi's mining complex. *Hum Ecol Risk Assess.* 2019;25:819-834.
78. Manea DN, Ienciu AA, Ștef R, Șmuleac IL, Gergen II, Nica DV. Health risk assessment of dietary heavy metals intake from fruits and vegetables grown in selected old mining areas-a case study: the Banat area of Southern Carpathians. *Int J Environ Res Public Health.* 2020;17:E5172.
79. Roba C, Roșu C, Pișteea I, Ozunu A, Băciu C. Heavy metal content in vegetables and fruits cultivated in Baia Mare mining area (Romania) and health risk assessment. *Environ Sci Pollut Res.* 2016;23:6062-6073.