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## Trace metals in vegetables and fruits cultivated around the surroundings of Tummalapalle uranium mining site, Andhra Pradesh, India

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## ABSTRACT

Vegetables (Tomato – *Solanum lycopersicum*, green chilli – *Capsicum annum* and bitter gourd – *Momordica charantia*) and fruits (Banana – *Musa acuminata colla*, papaya – *Carica papaya* and mosambi – *Citrus limetta*) from the cultivated areas around the Tummalapalle uranium mining site were analyzed for trace metals (Al, Cr, Mn, Fe, Ni, Cu, Zn, Pb, Be, V, Co, Cd and U) using inductively coupled plasma-mass spectrometer (ICP-MS). As per the estimated data, the concentrations of trace metals in vegetables and fruits are found in the range of 47.5–7.8 mg/kg for Al, 9.7–1.0 mg/kg for Cr, 3.8–1.0 mg/kg for Mn, 75.5–13.9 mg/kg for Fe, 1.4–0.2 mg/kg for Ni, 2.3–0.8 mg/kg for Cu, 9.2–3.1 mg/kg for Zn, 0.2–1.4 mg/kg for Pb, 19.2–1.9 µg/kg for Be, 96.1–15.8 µg/kg for V, 48.2–12.9 µg/kg for Co, 46.5–2.3 µg/kg for Cd and 16.4–2.7 µg/kg for U. The trace metals observed are compared to the literature reported values. Trace elemental data were subjected to statistical analysis to examine the interrelationship between the investigated trace elements and possible source identification of the trace metal contamination in vegetable and fruits. Daily intake of trace metals through ingestion of vegetables and fruits are also calculated.

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## 1. Introduction

Demonstration and determination of trace metals in food are an important task for nutritionists, environmentalists and scientists. Vegetables and Fruits are some of the most common foods of human diet in all around the human kind. These are rich sources of vitamins, minerals, fibres and also take on a dependable anti-oxidative effects. Other than water and soil, foods may also be contaminated with trace metals due to increased usage of chemicals,

sprays, preservatives, industrialization, mining activities, fertilizers, etc. Trace metal contamination of the food is one of the most significant aspects of food quality [1–3]. Trace metals can be classified as potentially toxic (arsenic, cadmium, lead, mercury, etc.), probably essential (nickel, vanadium and cobalt) and essential (iron, manganese, copper, zinc, selenium, etc.). Toxic metals can be very harmful even at low concentration when ingested over a long time period. The essential metals may also create toxic effects when metal intake is too elevated [4–10].

The levels of trace elements in vegetables and fruits have been widely reported in the literature (Table 3). However, the data on the trace metals in vegetables and fruit samples at the surroundings of mining industry in India are very

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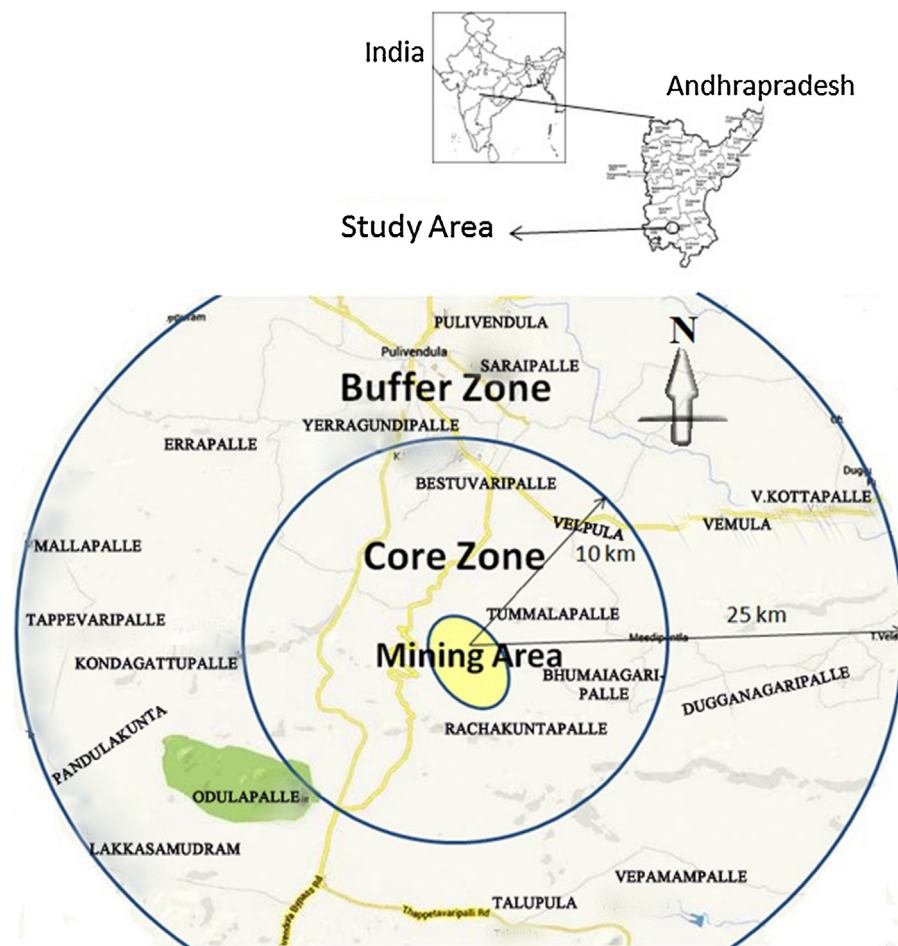


Fig. 1. Map showing the study zones.

limited. In the present study, the cultivated vegetables and fruits in and around the Tummalapalle uranium mining site were collected and analyzed for metal contents of Al, Cr, Mn, Fe, Ni, Cu, Zn, Pb, Be, V, Co, Cd and U using inductively coupled plasma-mass spectrometer (ICP-MS).

## 2. Experimental

### 2.1. Study area

The study area, Tummalapalle uranium mining is located between latitudes  $14^{\circ}18'36''$  N and  $14^{\circ}20'20''$  N and longitudes  $78^{\circ}15'16''$  E and  $78^{\circ}18'03.3''$  E according to survey of India Toposheet Nos. 57 J/3 and 57 J/7.

### 2.2. Sample collection

The study area is divided into two parts of the sample collection based on the radial distance from the mining site. (1) Core zone – which is having 10 km radial distance around the mining site; (2) buffer zone – which is spreading over the radial distance from 10 to 25 km around the mining site. The map showing the study area is described in Fig. 1.

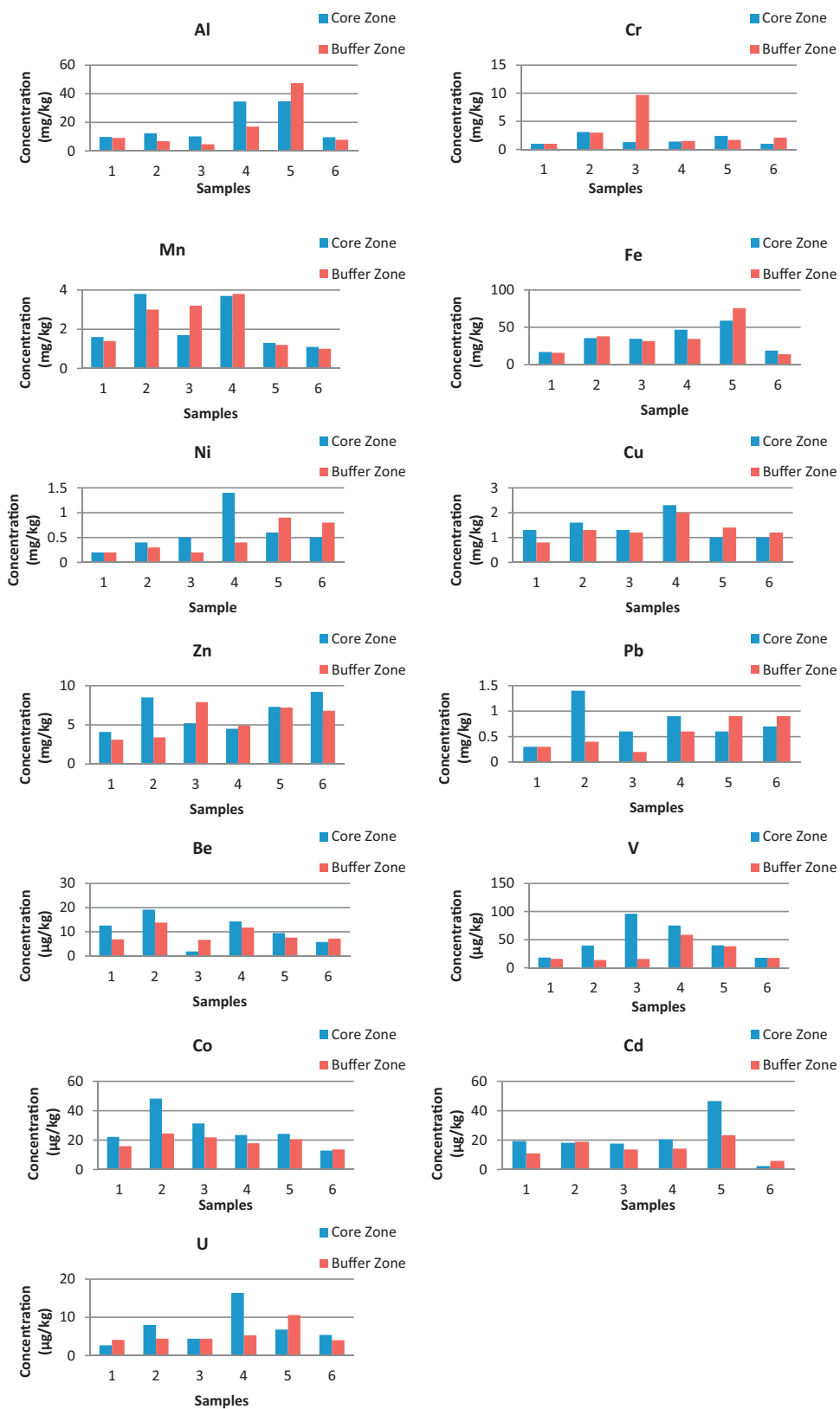
A total of 72 samples containing commonly cultivated vegetables of *Solanum lycopersicum* (tomato), *Capsicum annum* (green chilli), *Momordica charantia* (bitter gourd) and the fruits of *Musa acuminata colla* (banana), *Carica papaya* (papaya) and *Citrus limetta* (mosambi) were collected in the study area around the uranium mining site during 2011–2012.

### 2.3. Reagents

All reagents used for the analysis were of analytical grade and Milli-Q water is used for all dilutions. Supra pure grade reagent (Merck, Germany) of nitric acid is utilized for the digestion of samples. All the glass wares were cleaned by soaking it in dilute nitric acid and then rinsed with distilled water before use.

### 2.4. Sample processing and analysis

The samples were collected in polyethylene bags and preserved in the refrigerator prior to the processing and analysis. The samples were cleaned with Milli-Q water and the known quantity of edible portion of the sample (vegetable or fruit) was weighed using 0.01 mg sensitive



**Fig. 2.** Trace metal concentration in vegetable and fruits. (1) *Solanum lycopersicum* (tomato); (2) *Capsicum annuum* (green chilli); (3) *Momordica charantia* (bitter gourd); (4) *Musa acuminata colla* (banana); (5) *Carica papaya* (papaya); (6) *Citrus limetta* (mosambi)

**Table 1**  
Operation conditions for the analysis of trace metals by ICP-MS.

Parameter	Value
RF power (W)	1500
RF matching (V)	2.1
Sample depth (mm)	6.8
Carrier gas flow rate (L/min)	0.7
Makeup gas flow rate (L/min)	0.57
Nebulizer pump rate (rps)	0.1
Data acquisition	Peak hopping
Replicates	6

weighing balance and transferred to 100 ml glass beaker. To the weighed sample 15 ml of supra pure HNO<sub>3</sub> and 3 ml of HClO<sub>4</sub> were added and covered with the watch glass. After an hour, the beaker was placed on a hot plate, heated up to 140 °C until the sample completely digested and the total volume was reduced to approximately 2 ml. The digested sample was cooled, diluted with 2 N supra pure HNO<sub>3</sub> and made up to 50 ml in a standard flask [11–14]. Reagent blanks were made in each batch of the samples processed. The treated samples have been examined for trace metal contents using inductively coupled plasma-mass spectrometer (ICP-MS), make Agilent Technologies (Model No. 7700). The instrumental parameter for ICP-MS operation is presented in Table 1.

### 2.5. Quality assurance

The quality assurance of the procedure for the estimation of trace metals has been assessed by calibrating the instrument using multi element calibration standard 2A (Lot No. 28–68 JB) obtained from Agilent Technologies. The validity of the method was further ascertained by spike recovery and replicate analysis.

## 3. Results and discussion

The averaged trace metal concentrations in three varieties of vegetables and fruits from core zone and buffer zone in and around the uranium mining site are tabulated in Table 2 and the literature reported values from various regions of the world from corresponding samples are listed in Table 3. All metal concentrations are reported in wet weight basis. The concentrations of examined trace elements in vegetables and fruits were found to be in the range of 1.2–98.5 mg/kg for aluminium (Al); 0.3–13.7 mg/kg for chromium (Cr); 0.3–8.1 mg/kg for manganese (Mn); 6.5–126.3 mg/kg for iron (Fe); 0.1–4.1 mg/kg for nickel (Ni); 0.3–3.2 mg/kg for copper (Cu); 0.1–19.7 mg/kg for zinc (Zn); 0.1–3.4 mg/kg for lead (Pb); <0.01–44.1 µg/kg for beryllium (Be); 6.9–98.7 µg/kg for vanadium (V); 6.7–78.9 µg/kg for cobalt (Co); 0.7–83.9 µg/kg for cadmium (Cd); and 1.7–27.6 µg/kg for uranium (U). It is seen that, Fe has the highest concentration followed by Al, Zn and Cr. The U has the lowest concentration followed by Be and Co. The graphical representation of average trace metals content in the samples is shown in Fig. 2. The daily intake of trace metals through an average ingestion of 100 g of these vegetables and fruits are calculated and tabulated in Table 4.

The standards available for an acceptable dosage limit of daily intake are also given in this table.

The highest average concentration of aluminium (47.5 ± 25.3 mg/kg) was observed in *C. papaya* (papaya) at buffer zone and the lowest mean concentration (6.8 ± 4.5 mg/kg) in *C. annuum* (green chilli) at buffer zone. 1.7 mg/day intake of Al was found through the intake of these vegetables and fruits in the study area and is well within the maximum permissible dose limit of aluminium for an adult, 60 mg/day [15,4].

The minimum and maximum chromium contents of the samples were found to be 1.0 mg/kg in *S. lycopersicum* (tomato) at both core zone and buffer zone and 9.7 mg/kg in *M. charantia* (bitter melon) at buffer zone, respectively. In the literature chromium content has been reported in the range of 0.07–6.0 mg/kg in vegetable and fruits [16,20,29]. The average recommended daily intake of chromium is 50–200 µg [15,17,29] and the intake of Cr in the study area was found to be 0.2 mg/day (Table 5).

The average concentration of the manganese (Mn) observed in vegetables and fruits are about 2.2 mg/kg which contains minimum (1.0 mg/kg) is seen in *C. limetta* and maximum (3.8 mg/kg) in *C. annuum* (green chilli) and *M. acuminata* (banana). The intake of Mn (0.2 mg/day) in the investigated samples is well below the tolerable daily upper limit of 4.1 mg/day [4,15]. The literature values of observed concentrations are ranging from 0.04 to 27.85 mg/kg for both vegetables and fruits. The obtained values in this study are comparable to the literature reported values (Table 3).

The obtained Fe concentration in vegetable and fruit samples is compared to the literature reported values (Table 3). Minimum and maximum levels of Fe were found in *C. limetta* (13.9 mg/kg) and *C. papaya* (75.5 mg/kg), respectively. The acceptable daily limit of Fe is 15 mg/day [4,15]; and the average intake in the study area is 3.5 mg/day.

Obtained concentration of nickel in the samples is showing good comparison with the literature (Table 3). The mean concentration of Ni in the samples is 0.53 mg/kg. The intake of Ni at the study area is found to be 0.1 mg/day. This is equal to the acceptable dosage limit of Ni (0.1 mg/day) and is alarming.

The median concentration of Cu in the samples is approximately 1.37 mg/kg and is comparable to the reported literature values, which are in the range of 0.1–14.1 mg/kg (Table 3). The daily intake of Cu in adults from food generally varies from about 1 to 2.5 mg, corresponding to 15–45 µg/kg body weight in adults [15]. The daily intake of Cu in the study area is about 0.1 mg/day.

The highest and lowest zinc concentrations were observed in *C. limetta* (9.2 mg/kg) and *S. lycopersicum* (3.1 mg/kg), respectively. The reported literature values range from 0.06 to 10.6 mg/kg (Table 3). The average intake of 0.6 mg/day of zinc was observed in the field area. The recommended daily intake of zinc is 15 mg for men and 12 mg for adult female [4,15].

The median value of Pb obtained in the samples is 0.64 mg/kg and is showing good comparison with the reported literature values, in the range of 0.06–2.78 mg/kg (Table 3).

**Table 2**  
Average trace metal content (mean  $\pm$  SD) in cultivated vegetable and fruits (N=72).

Type	Sample	Zone	Al (mg/kg)	Cr (mg/kg)	Mn (mg/kg)	Fe (mg/kg)	Ni (mg/kg)	Cu (mg/kg)	Zn (mg/kg)	Pb (mg/kg)	Be ( $\mu$ g/kg)	V ( $\mu$ g/kg)	Co ( $\mu$ g/kg)	Cd ( $\mu$ g/kg)	U ( $\mu$ g/kg)
<i>Vegetables (wet weight basis)</i>															
1	<i>Solanum lycopersicum</i> (tomato)	Core zone	9.8 $\pm$ 4.2	1.0 $\pm$ 0.4	1.6 $\pm$ 0.5	16.9 $\pm$ 3.4	0.2 $\pm$ 0.1	1.3 $\pm$ 0.3	4.1 $\pm$ 0.8	0.3 $\pm$ 0.1	12.6 $\pm$ 3.3	18.3 $\pm$ 7.9	22.2 $\pm$ 10.8	19.3 $\pm$ 14.0	2.7 $\pm$ 0.4
		Buffer zone	9.1 $\pm$ 2.9	1.0 $\pm$ 0.6	1.4 $\pm$ 0.4	15.7 $\pm$ 3.2	0.2 $\pm$ 0.1	0.8 $\pm$ 0.4	3.1 $\pm$ 0.9	0.3 $\pm$ 0.1	6.9 $\pm$ 3.6	15.8 $\pm$ 4.5	15.9 $\pm$ 6.3	11.0 $\pm$ 9.9	4.1 $\pm$ 3.3
2	<i>Capsicum annuum</i> (green chilli)	Core zone	12.3 $\pm$ 9.8	3.1 $\pm$ 2.2	3.8 $\pm$ 2.2	35.5 $\pm$ 13.9	0.4 $\pm$ 0.2	1.6 $\pm$ 0.6	8.5 $\pm$ 6.5	1.4 $\pm$ 1.2	19.2 $\pm$ 18.0	39.6 $\pm$ 22.2	48.2 $\pm$ 21.8	18.2 $\pm$ 6.0	8.0 $\pm$ 3.4
		Buffer zone	6.8 $\pm$ 4.5	3.0 $\pm$ 1.9	3.0 $\pm$ 1.0	37.8 $\pm$ 10.2	0.3 $\pm$ 0.1	1.3 $\pm$ 0.7	3.4 $\pm$ 0.7	0.4 $\pm$ 0.2	13.8 $\pm$ 3.5	13.9 $\pm$ 4.0	24.5 $\pm$ 12.1	18.9 $\pm$ 11.4	4.4 $\pm$ 2.0
3	<i>Momordica charantia</i> (bitter Gourd)	Core zone	10.2 $\pm$ 3.4	1.3 $\pm$ 0.3	1.7 $\pm$ 0.5	34.5 $\pm$ 17.2	0.5 $\pm$ 0.3	1.3 $\pm$ 0.5	5.2 $\pm$ 0.8	0.6 $\pm$ 0.2	1.9 $\pm$ 2.2	96.1 $\pm$ 15.5	31.3 $\pm$ 9.6	17.6 $\pm$ 7.9	4.4 $\pm$ 0.2
		Buffer zone	4.7 $\pm$ 4.0	9.7 $\pm$ 2.7	3.2 $\pm$ 2.3	31.5 $\pm$ 19.2	0.2 $\pm$ 0.1	1.2 $\pm$ 0.9	7.9 $\pm$ 6.6	0.2 $\pm$ 0.1	6.7 $\pm$ 0.4	15.6 $\pm$ 10.8	21.9 $\pm$ 7.6	13.6 $\pm$ 5.6	4.4 $\pm$ 0.3
<i>Fruits (wet weight basis)</i>															
4	<i>Musa acuminata colla</i> (banana)	Core zone	34.5 $\pm$ 29.4	1.4 $\pm$ 0.8	3.7 $\pm$ 2.7	46.7 $\pm$ 26.2	1.4 $\pm$ 1.1	2.3 $\pm$ 0.6	4.5 $\pm$ 2.9	0.8 $\pm$ 0.6	14.3 $\pm$ 13.2	75.2 $\pm$ 21.2	23.5 $\pm$ 4.3	20.5 $\pm$ 15.1	16.4 $\pm$ 9.5
		Buffer zone	17.0 $\pm$ 11.8	1.5 $\pm$ 0.5	3.8 $\pm$ 1.7	34.4 $\pm$ 18.5	0.4 $\pm$ 0.2	2.0 $\pm$ 0.6	4.9 $\pm$ 3.7	0.6 $\pm$ 0.4	11.8 $\pm$ 6.3	58.6 $\pm$ 17.4	17.9 $\pm$ 9.7	14.2 $\pm$ 10.1	5.3 $\pm$ 2.1
5	<i>Carica papaya</i> (papaya)	Core zone	34.7 $\pm$ 27.4	2.4 $\pm$ 1.9	1.3 $\pm$ 0.8	58.8 $\pm$ 50.6	0.6 $\pm$ 0.4	1.0 $\pm$ 0.7	7.3 $\pm$ 5.1	0.6 $\pm$ 0.4	9.5 $\pm$ 5.4	40.0 $\pm$ 28.3	24.3 $\pm$ 13.4	46.5 $\pm$ 25.4	6.8 $\pm$ 2.8
		Buffer zone	47.5 $\pm$ 25.3	1.7 $\pm$ 0.8	1.2 $\pm$ 0.4	75.5 $\pm$ 24.5	0.9 $\pm$ 0.3	1.4 $\pm$ 0.3	7.2 $\pm$ 3.4	0.9 $\pm$ 0.5	7.6 $\pm$ 3.8	38.3 $\pm$ 17.1	20.6 $\pm$ 13.9	23.3 $\pm$ 11.9	10.6 $\pm$ 4.1
6	<i>Citrus limetta</i> (mosambi)	Core zone	9.6 $\pm$ 2.5	1.0 $\pm$ 0.5	1.1 $\pm$ 0.1	18.7 $\pm$ 5.8	0.5 $\pm$ 0.3	1.0 $\pm$ 0.3	9.2 $\pm$ 4.2	0.7 $\pm$ 0.4	5.8 $\pm$ 1.2	17.8 $\pm$ 8.2	12.9 $\pm$ 4.9	2.3 $\pm$ 1.0	5.4 $\pm$ 1.6
		Buffer zone	7.8 $\pm$ 1.5	2.1 $\pm$ 0.9	1.0 $\pm$ 0.2	13.9 $\pm$ 4.1	0.8 $\pm$ 0.3	1.2 $\pm$ 0.3	6.8 $\pm$ 2.7	0.9 $\pm$ 0.3	7.2 $\pm$ 5.6	17.6 $\pm$ 7.1	13.6 $\pm$ 6.3	5.9 $\pm$ 1.8	4.0 $\pm$ 0.9
Range			1.2–98.5	0.3–13.7	0.3–8.1	6.5–126.3	0.1–4.1	0.3–3.2	0.1–19.7	0.1–3.4	<0.01–44.1	6.9–98.7	6.7–78.9	0.7–83.9	1.7–27.6

**Table 3**  
Trace metal content in various parts of the world (literature values).

Sample	Country	Al	Cr	Mn	Fe	Ni	Cu	Zn	Pb	Be	V	Co	Cd	U
<i>Solanum</i>	India (mg/g) [22]	-	-	-	11.56	1.26	0.56	3.28	0.05	-	-	0.02	0.01	-
<i>Lycopersicum</i> (tomato)	Spain (mg/kg) [19] Nigeria (mg/kg) [24] Saudi Arabia (mg/kg) [21] Egypt (mg/kg) [25]	-	-	0.63 17.88 27.84	2.19 26.59 364.60	7.75	0.31 14.07 7.46 1.83	0.86 - 22.91 9.81	2.36 2.78 0.26	-	-	-	0.12 2.45 0.10	-
<i>Capsicum Annum</i> (Green Chilli)	India (mg/g) [22] Nigeria (mg/kg) [26] Egypt (mg/kg) [25]	-	0.06	-	9.54 0.69	2.03	0.84 0.13 5.20	2.77 0.18 13.20	0.07 0.18 0.61	-	-	0.01	0.01 0.19 0.06	-
<i>Momordica charantia</i> (Bitter Gourd)	India (mg/g) [22] Pakistan (mg/kg) [27]	-	-	1.40	11.32 116.00	0.22 1.30	0.27 1.10	2.74 4.00	0.14	-	-	0.01 1.14	0.01	-
<i>Musa acuminata</i> colla (banana)	Egypt (mg/kg) [25] Pakistan (mg/kg) [27] USA (mg/kg) [28] Nigeria (mg/kg) [26]	-	-	1.16 4.80	163.00 3.15	0.60	2.51 1.10 0.96	5.59 1.10 1.80 0.06	0.05	-	-	1.06	0.02	-
<i>Carica papaya</i> (papaya)	Pakistan (mg/kg) [20] Nigeria (mg/kg) [26] Malaysia (mg/kg) [29]	-	0.13	-	6.58	0.70	0.77 0.40 3.48	26.20 0.06 8.13	0.64 0.11 1.38	-	-	0.02	0.34 0.01 0.55	-
<i>Citrus limetta</i> (mosambi)	Egypt (mg/kg) [25] USA (mg/kg) [28] Pakistan (mg/kg) [20]	-	-	0.56	0.99 7.26	-	1.89 0.35 1.41	2.63 0.30 10.60	0.25	-	-	-	0.08	-
			0.13	-					0.27				1.61	

**Table 4**

Daily intake of trace metals through the ingestion of vegetables and fruits in comparison along with the permissible exposure limit.

Trace metal	Intake through ingestion of average 100 g of vegetables and fruits per day	Permissible exposure limit of daily dietary intake [15,4]
Al	1.7 mg/day	60 mg/day
Cr	0.2 mg/day	35 µg/day
Mn	0.2 mg/day	4.1 mg/day
Fe	3.5 mg/day	15 mg/day
Ni	0.1 mg/day	0.1 mg/day
Cu	0.1 mg/day	2.5 mg/day
Zn	0.6 mg/day	15 mg/day
Pb	64 µg/day	-
Be	1.0 µg/day	-
V	3.7 µg/day	50 µg/day
Co	2.3 µg/day	55 µg/day
Cd	1.8 µg/day	60 µg/day
U	0.6 µg/day	1.5 µg/day

Minimum and maximum levels of beryllium were found in *M. charantia* (1.9 µg/kg) and *C. annum* (19.2 µg/kg), respectively.

The highest and lowest levels of V were found in *C. annum* (15.8 µg/kg) and *M. charantia* (96.1 µg/kg), respectively. The dietary intake of vanadium reported is 5–50 µg/day [4,15] and is comparable to the observed value of V (3.7 µg/day) in the study area.

The highest and lowest contents of Co was founded in *Capsicum Annum* (48.2 µg/kg) and *citrus limetta* (12.9 µg/kg), respectively. This is comparable to the reported literature values (Table 3). Human dietary intake of cobalt varies between 5–55 µg/day [4,15]. An average intake of 2.3 µg/day of Co is estimated for the study area.

The highest and lowest content of Cd was found in *C. papaya* (46.5 µg/kg) and *C. limetta* (2.3 µg/kg), respectively. The reported literature values to Cd are in the range of 0.01–2.45 mg/kg (Table 3). The estimated average intake of Cd at study area is 1.8 µg/day and shows a good comparison with the literature reported intake value of 10–60 µg/day for a 70 kg person [4,15].

The average concentration of uranium in the present study for vegetable and fruit samples is in the range of 1.7–27.6 µg/kg. The median intake of uranium is 0.6 µg/day and shows a good comparison with literature reported values, in the range of 0.9–1.5 µg/day [15].

The trend of concentration points of various measured trace metals in vegetables is observed in the following sequence: Fe > Al > Zn > Mn > Cr > Cu > Pb > Ni > Co > V > Cd > Be > U (Table 2). In general, it is a common belief that most of the vegetables are enriched with Fe [16] and the same is also seen in our observation (15.7–37.8 µg/kg). The elements Fe, Mn, Al, Cr, Cu, Pb, Be, Co, Cd and U were found to be highest in *C. annum* (green chilli), among the three vegetables analyzed in this study. According to Indian Ministry of Agriculture, Report of the post harvest profile of chilli [23], the nutritional mineral and trace metal levels in *C. annum* are Cu – 1.5 mg/100 g; Mn – 1.38 mg/100 g; Zn – 1.78 mg/100 g and Cr – 0.04 mg/100 g. The elements Ni, Zn and V were found to be highest in *M. charantia* (bitter gourd) and lowest in *S. lycopersicum* (tomato) amongst the vegetables analyzed in this study. Nirmal Kumar et al. [22]



**Table 5**  
Correlation between metal concentrations of the vegetables and fruit samples (N=72).

	Al	Cr	Mn	Fe	Ni	Cu	Zn	Pb	Be	V	Co	Cd	U
Al	1.00												
Cr	0.05	1.00											
Mn	-0.13	0.20	1.00										
Fe	<b>0.71</b>	0.18	0.03	1.00									
Ni	<b>0.70</b>	-0.02	-0.12	<b>0.42</b>	1.00								
Cu	<b>0.37</b>	0.06	<b>0.59</b>	0.30	<b>0.50</b>	1.00							
Zn	0.23	0.30	0.30	0.16	0.07	0.21	1.00						
Pb	0.26	-0.01	0.08	0.15	0.17	0.21	0.25	1.00					
Be	0.09	0.37	<b>0.50</b>	-0.02	0.19	<b>0.45</b>	0.35	-0.05	1.00				
V	0.29	-0.06	0.33	0.34	0.32	<b>0.43</b>	0.09	0.15	0.15	1.00			
Co	0.17	0.19	<b>0.41</b>	0.27	0.12	0.31	<b>0.47</b>	0.31	<b>0.40</b>	0.27	1.00		
Cd	<b>0.41</b>	0.28	0.09	<b>0.47</b>	0.02	0.07	0.21	0.15	-0.11	0.25	0.26	1.00	
U	0.30	0.18	<b>0.41</b>	0.37	0.10	0.30	0.27	0.18	0.14	<b>0.43</b>	0.15	<b>0.40</b>	1.00

Bold values shows the significant values.

reported that the trace metals Fe – 9.54, 11.56, 11.32 mg/g; Ni – 2.03, 1.26, 0.22 mg/g; Cu – 0.84, 0.56, 0.27 mg/g; Zn – 2.77, 3.28, 2.74 mg/g; Pb – 0.07, 0.06, 0.14 mg/g; Co – 0.01, 0.02, 0.01 mg/g and Cd – 0.01, 0.01, 0.01 mg/g in *C. annum*, *M. charantia*, *S. lycopersicum*, respectively. The levels of toxic metals like Pb and Cd in vegetables from India and that from various other parts of the world are given in Table 3. Comparison with the compactness of the toxic metals in vegetables in the present study reveals that the concentration levels are safe and comparable to those reported by others.

The succession of concentration levels of the measured trace metals in fruits analyzed is as follows: Fe > Al > Zn > Mn > Cu > Cr > Pb > Ni > V > Cd > Co > U > Be (Table 2). This is most alike to the style observed in vegetables. The measured trace elements are higher in *M. acuminata colla* and *C. papaya* and low in *C. limetta*. The reported levels of trace metals in fruits from diverse parts of the world are given in Table 3. The reported literature values from various parts of the world are comparable to the values observed in this study. The concentration of toxic metals like Pb and Cd are comparable with the reported literature (Table 3), which are in the range of 0.05–1.38 mg/kg for Pb and 0.02–0.55 mg/kg for Cd.

### 3.1. Correlation analysis

A linear regression correlation analysis was performed to investigate the correlation between the trace elements and to determine the likely sources of contamination of the vegetables and fruit samples analyzed. The values of the correlation coefficients between metal concentrations are given in Table 5. Al is showing good correlation with Fe, Ni, Cu and Cd, with a corresponding regression coefficient (*r*) values of 0.71, 0.70, 0.37 and 0.41, respectively. Fe is showing good correlation with Ni and Cd with corresponding *r* values of 0.42 and 0.47, respectively. This indicates that, the contamination of these metals (Al, Fe, Ni, Cu and Cd) in these samples is likely from the earth's crust as Al and Fe are most abundant crustal elements.

Further, Mn is showing good correlation with Cu, Be, Co and U; Ni with Cu; Cu with Be and V; Zn with Co; Be with Co; V with U and Cd with U. This indicates that, the contamination of these metals is occurring through various

**Table 6**  
Crustal mineral composition of Tummalapalle uranium mine and surroundings [18].

Mineral	% Weight
Carbonates	83.2
Quartz + feldspar	11.3
Collophane	4.3
Pyrite	0.47
Chalcopyrite	0.05
Galena	Traces
Magnetite	0.15
Ilmenite + leucoxene	0.25
Iron hydroxide (goethite)	0.27
Pitchblende in intimate association with pyrite	0.1
Total	100.0

sources like crustal contamination, use of polluted water for cultivation, use of pesticides, fertilizers, atmospheric particulate deposition, anthropogenic activities, industrialization, mining activities, etc. Table 6 gives the better understanding of heavy metal content of rock and soils inside and surroundings of the Tummalapalle uranium mining site [18].

## 4. Conclusion

The present study has generated data on trace metals in vegetables and fruits, which are cultivated in and around the surrounding villages of the uranium mining site. The levels observed are comparable with the literature reported values from various regions of the globe. The concentration trend of various measured trace metals in vegetables and fruits is in the order of 'Fe > Al > Zn > Mn > Cr > Cu > Pb > Ni > Co > V > Cd > Be > U'. Further, it has been observed that *C. annum* (green chilli) is showing high levels of trace elemental content, among the three varieties of vegetables, similarly *M. acuminata colla* (banana) is showing high levels of trace metal content among the three types of fruits analyzed. A linear regression correlation analysis of trace metal concentrations in vegetables and fruits indicates that, the contamination of Al, Fe, Ni, Cu and Cd is likely from the earth's crust, whereas, Mn, Be, Co, Zn, V and U is from multisource origin. Concentration of trace elements in the studied vegetables and fruits is acceptable for human consumption and the percentage of trace metal

contribution through ingestion of these vegetables and fruits are in the range of 2–10% of the daily permissible exposure limit of intake.

### Conflict of interest

None.

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### References

- [1] K. Bakkali, N. Romos Martos, B. Souhail, E. Ballesteros, Characterization of trace metals in vegetables by graphite furnace atomic absorption spectrometry after closed vessel microwave digestion, *Food Chem.* 116 (2009) 590–594.
- [2] M.N. Matos-Reyes, M.L. Cervera, R.C. Campos, M. de la Guardia, Total content of As, Sb, Se, Te and Bi in Spanish vegetables, cereals and pulses and estimation of the contribution of these foods to the Mediterranean daily intake of trace elements, *Food Chem.* 122 (2010) 188–194.
- [3] R.K. Sharma, M. Agarwal, F.M. Marshall, Heavy metals in vegetables collected from production and market sites of a tropical urban area of India, *Food Chem. Toxicol.* 47 (2009) 583–591.
- [4] O.D. Uluozlu, M. Tuzen, D. Mendil, M. Soylak, Assessment of trace element contents of chicken products from turkey, *J. Hazard. Mater.* 163 (2009) 982–987.
- [5] J. Falandysz, A. Frankowska, A. Mazur, Mercury and its bio concentration factors in King Bolete (*Boletus edulis*), *Bull. Fr. J. Environ. Sci. Health* 42A (2007) 2089–2095.
- [6] M. Tuzen, Determination of heavy metals in fish samples of the middle Black sea (Turkey) by graphite furnace atomic absorption spectrometry, *Food Chem.* 80 (2003) 119–123.
- [7] K. Ganesh Chandra, P. Pandey, N. Mahendra Pratap Singh, V. Mishra, Uptake and accumulation of potentially toxic metals (Zn, Cu and Pb) in soils and plants of Durgapur industrial belt, *J. Environ. Biol.* 32 (2011) 831–838.
- [8] I.U. Adams, I.U. Happiness, Quantitative specification of potentially toxic metals in expired canned tomatoes found in village markets, *Nat. Sci.* 8 (2010) 54–58.
- [9] V. Singh, A.N. Garg, Availability of essential trace elements in Indian cereals, vegetables and spices using INAA and the contribution of spices to daily dietary intake, *Food Chem.* 94 (2006) 81–89.
- [10] S.F. Sulaiman, N.A.M. Yusoff, I.M. Eldeen, E.M. Seow, A. Abu Bakar Sajak, K. Leong Ooi, Correlation between total phenolic and mineral contents with antioxidant activity of eight Malaysian bananas, *J. Food Compo. Anal.* 24 (2011) 1–10.
- [11] A.M. Basha, N. Yasovardhan, S.V. Satyanarayana, G.V.S. Reddy, A.V. Kumar, Assessment of heavy metal content of hen eggs in the surroundings of uranium mining area India, *Ann. Food Sci. Tech.* 14 (2013) 344–349.
- [12] B. Suseela, S. Bhalke, A. Vinod Kumar, R.M. Tripathi, V.N. Sastry, Daily intake of trace metals through coffee consumption in India, *Food Addit. Contam.* 18 (2001) 115–120.
- [13] R.M. Tripathi, S. Mahapatra, R. Raghunath, A. Vinod Kumar, S. Sadasi-van, Daily intake of aluminium by adult population of Mumbai, India, *Sci. Total Environ.* 299 (2002) 73–77.
- [14] R.M. Tripathi, R. Raghunath, A. Vinod Kumar, T.M. Krishnamoorthy, Intake of chromium by the adult population of Mumbai city, *Environ. Monit. Assess.* 53 (1998) 379–389.
- [15] G.F. Nordberg, B.A. Fowler Monica Bordberg, L. Friberg, *Handbook on the Toxicology of Metals*, third ed., European Environmental Agency, Copenhagen, Denmark, 2005.
- [16] A. Demirbas, Oil, micronutrient and heavy metal contents of tomatoes, *Food Chem.* 118 (2010) 504–507.
- [17] A. Shokrollahi, M. Ghaedi, M.S. Niband, H.R. Rajabi, Selective and sensitive spectrophotometric method for determination of sub micro molar amounts of aluminium ion, *J. Hazard. Mater.* 151 (2008) 642–648.
- [18] A.K. Suri, Innovative process flow sheet for the recovery of uranium from Tummalalpal ore, *BARC News Lett.* 317 (2010) 6–12.
- [19] M. Hernandez Suarez, E.M. Rodriguez, C. Diaz Romero, Mineral and trace element concentrations in cultivars of tomatoes, *Food Chem.* 104 (2004) 489–499.
- [20] M. Jaffar, K. Masud, Selected toxic metal levels in seasonal fruits of Pakistan, *J. Nutri. Food Sci.* 33 (2003) 9–15.
- [21] A.H.H. Mohammed, K.M. Al-Qahtani, Assessment of some heavy metals in vegetables, cereals and fruits in Saudi Arabian markets, *Egypt. J. Aqua. Res.* 38 (2012) 31–37.
- [22] J.I. Nirmal Kumar, H. Soni, N. Rita Kumar, Characterization of heavy metals in vegetables using inductive coupled plasma analyzer (ICPA), *J. Appl. Sci. Environ. Manage.* 11 (2007) 75–79.
- [23] Report of post harvest profile of Chilli, Government of India, Ministry of Agriculture (Department of Agriculture & Cooperation), 2009.
- [24] U.A. Birnin-Yauri, Y. Yahaya, B.U. Bagudo, S.S. Noma, Seasonal variation in nutrient content of some selected vegetables from Wamakki, Sokoto State, Nigeria, *J. Sci. Environ. Manage.* 2 (2011) 117–125.
- [25] M.A. Radwan, A.K. Salama, Market basket survey for some heavy metals in Egyptian fruits and vegetables, *Food Chem. Toxicol.* 44 (2006) 1273–1278.
- [26] O.P. Sobukola, O.M. Adhnriran, A.A. Odedairo, O.E. Kajihusa, Heavy metal levels of some fruits and leafy vegetables from selected markets in Lagos, Nigeria, *African. J. Food Sci.* 4 (2010) 389–393.
- [27] F. Ismail, M.R. Anum, A.N. Mamon, T.G. Kazi, Trace metal content of vegetables and fruits of Hyderabad retail market, Pakistan, *J. Nutri.* 10 (2011) 365–372.
- [28] N.J. Miller-Ihli, Atomic absorption and atomic emission spectrometry for the determination of the trace element content of selected fruits consumed in the United States, *J. Food Compo. Anal.* 9 (1996) 301–311.
- [29] L.H. Ang, L.T. Ng, Trace element concentration in Mango (*Mangifera indica* L.) deedless Guava (*Psidium guajava* L.) and Papaya (*Carica papaya* L.) grown on agricultural and ex-mining lands of bidor, perak, Pertanika *J. Trop. Agri Sci.* 23 (2000) 15–22.