Eco. Env. & Cons. 28 (3) : 2022; pp. (1516-1525)
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ISSN 0971–765X

DOI No.: http://doi.org/10.53550/EEC.2022.v28i03.059

Evaluation of Heavy Metals in Vegetables and their Associated Health Risk through the consumption of Vegetables in the National Capital Region, India

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(Received 16 October, 2021; Accepted 15 December, 2021)

ABSTRACT

Evaluation of the heavy metals concentration in the vegetables growing in the national capital region of India was done in the present study. A total of 96 samples of vegetables were collected from the selected study area during the summer season and were analyzed for heavy metals contamination by atomic absorption spectrophotometer. The relative abundance of heavy metal (mg/kg) in the vegetable samples was in the following order: Fe > Al >Mn> Zn > Cu > Cr > Ni >Pb> Cd > Co with the average concentrations of 260.26, 149.70, 32.59, 29.10, 17.18, 3.97, 1.45, 1.38, 0.27, and 0.10 respectively. The statistical analysis supported the formation of two primary clusters i.e. Al-Fe-Cr-Ni and Pb-Zn, indicating their common source of origin. Most of the vegetable samples exceeded the permissible limit of heavy metals prescribed by the Food and agricultural organization/ World health organization (FAO/WHO) standards. The total target hazard quotient was greater than 1 for all types of vegetables, indicating appreciable health risk due to the consumption of these vegetables in the study area.

***Key words**: Metals, Vegetables, Total hazard quotient, Principal component analysis, Dendrogram, Metal pollution index.*

Introduction

Lack of urban planning has resulted in the establishment of industries either on or near agricultural lands in the national capital region of India. The industries are responsible for the emission of various types of pollutants (Tiwari *et al.,* 2015). These pollutants get dispersed into the environment and can harm plants, animals, and humans. Amongst pollutants, heavy metals are of major concern as they are toxic and non-biodegradable. Some heavy metals also show bio-magnification through the food chain at different trophic levels (David *et al.*, 2012). Wastewater irrigation is also one of the source of contamination in agricultural fields due to the unavailability of freshwater resources for agricultural uses (Nayek *et al.,* 2010). These factors are increasing heavy metals concentration in agricultural soils and ultimately to vegetables. The consumption of contaminated vegetables can cause health risks to humans (Tchounwou *et al.*, 2012). The study area is irrigated by groundwater and drains carrying wastewater of the city (Sharma, 2020). The contamination of groundwater by various small scale industries in the region has also been observed by Sarita and Rani, 2016, which may be a source of heavy metal accumulation in vegetables. The main objective of the present study was to estimate the heavy metal contamination in different types of vegetables grown in the National Capital region, Delhi and to evaluate the human health risk via the consumption of these vegetables.

Experimental

Study area

The national capital region, Delhi (Gurgaon, Faridabad, Panipat, and Sonipat) is known for their fast development in the past few decades. Further, the study area is significantly contributing towards agricultural products. A total of 94 vegetable samples were collected for the study from different fields from four corners and one centre point of each field to prepare a composite sample (weighing approximately 2 Kg) during April 2017 to August 2017 (Figure 1). The description of the different types of vegetable collected from the study area is given in Table 1.

Preparation of sample

The vegetable samples were rinsed with clean tap water as per normal household practice to remove the dirt from the surface. The washed vegetables were chopped into small pieces and oven-dried at 80 ^{0}C till constant weight was achieved. The dried vegetables were powdered in a stainless steel blender and sieved through a 0.1 mm mesh sieve (Sharma *et al.,* 2008).

Digestion, analysis and quality control

1 g dried sample of vegetable was digested by adding 15 ml of a tri-acid mixture (HNO_3, H_2SO_4, and $HClO_4$ in 5:1:1 ratio) at 80 ^{o}C until a transparent solution was obtained. After cooling, the digested sample was finally maintained to 50 ml with double distilled water and was filtered using Whatman No. 42 filter paper (Sharma et al., 2008). Concentrations of Al, Cd, Cu, Co, Cr, Fe, Mn, Ni, Pb, and Zn in the filtrate of digested vegetable samples were estimated using an atomic absorption spectrophotometer (Model Lab India 8000A). The instrument was calibrated with a blank and reference standard of heavy metals solution of different concentrations ranging between 0.02–10 ppm at standard conditions (r^2=0.999). Blank and standard were run after every 10 samples on the instrument.

Data analysis

The Statistical Program for Social Sciences was used to find out the Pearson's correlation coefficient (r), principal component analysis (PCA), and hierarchi-

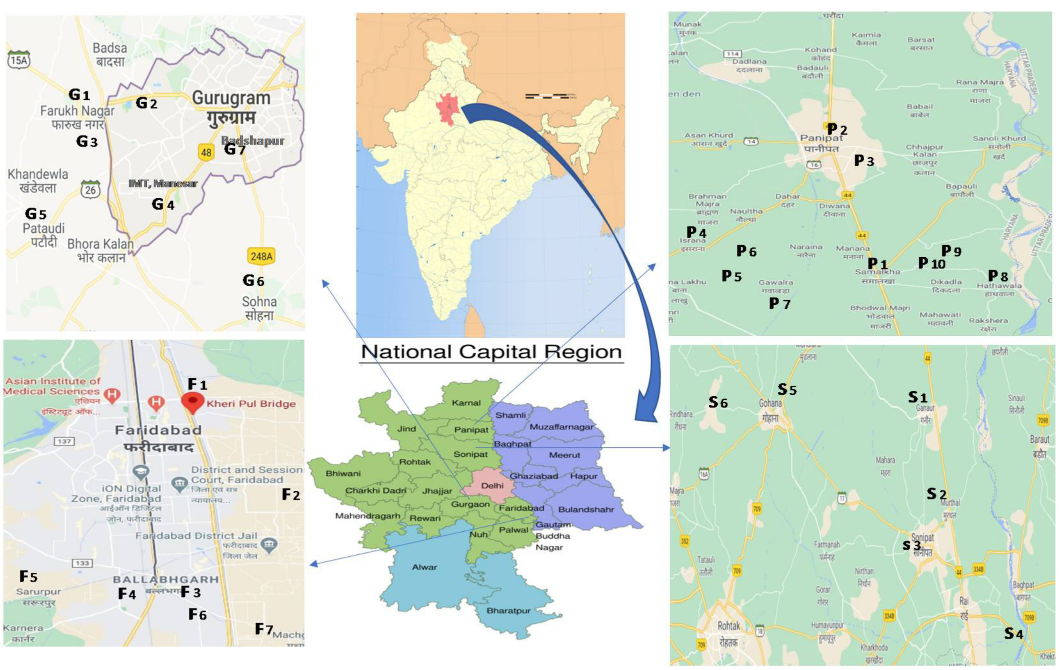

Fig. 1. Map of the study area

Table 1. Description of the vegetable samples collected from the NCR, India

Vegetable (s)	Scientific name of vegetables	No. of samples samples collected (n)	Classification
Amaranth	*Amaranthus hybridus* L.	(n=3)	Leaves
Beans	*Phaseolus vulgaris*	(n=3)	Legume
Brinjal	*Solanum melongena* L.	(n=5)	Fruit
Bitter gourd	*Momordica charantia*	(n=4)	Fruit
Bottle gourd	*Lagenaria siceraria*	(n=11)	Fruit
Cabbage	*Brassica oleracea var. capitata*	(n=4)	Leaves
Cauliflower	*Brassica oleracea* L. *var. botrytis*	(n=3)	Flower
Cucumber	*Cucumis sativus* L.	(n=3)	Fruit
Chilli	*Capsicum annuum*	(n=6)	Fruit
lady's finger	*Abelmoschus esculentus*	(n=12)	Fruit
Indian round gourd	*Praecitrullus fistulosus*	(n=6)	Fruit
Torai	*Luffa cylindrica*	(n=14)	Fruit
Tomato	*Solanum lycopersicum*	(n=3)	Fruit
Onion	*Allium cepa*	(n=4)	Bulb
Mint	*Mentha arvensis* L.	(n=3)	Leaves
spinach	*Spinacia oleracea*	(n=10)	Leaves

cal cluster analysis (CA) to understand the correlation between heavy metals.

Health risk assessment

Health risk can be estimated by calculating the target hazard quotient (THQ) as a result of heavy metals ingestion by humans. The equations (1), (2) and (3) were used for health risk to the consumers due to ingestion of heavy metals contaminated vegetables (Proshad *et al.*, 2019).

$$THQ = \frac{(Ef \times Ed \times Fir \times Cm)}{(\mathrm{Rfd} \times \mathrm{Bw} \times \mathrm{Ta})} \times 10^{-3} \qquad (1)$$

The total target hazard quotient (THQ) for all studied metals in a vegetable can be calculated as:

$$Total\ THQ\ (individual\ vegetable) = THQ_{M1} + THQ_{M2} ...... + THQ_{MN} \qquad .. (2)$$

$$HI = \Sigma TTHQ \qquad .. (3)$$

Where, ^{Ef} (Exposure frequency) 365 days/year, ^{Ed} (Exposure duration) 70 years, ^{Fir} (Ingestion rate of vegetable in g /person /day) 345 for adult and 232 for children, ^{Cm} (Concentration of the heavy metal in vegetable (mg/kg. dw), ^{Rfd} (Oral reference dose of heavy metals (mg/kg. day) Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn were 1.0, 0.001, 0.043, 1.5, 0.040, 0.7, 0.014, 0.02, 0.0035, and 0.300, respectively. (US department of health, 2008; US-EPA, 2006; Verma *et al.* 2015; Antoine *et al.* 2017), Bw (Average body weight) 55.9 kg for adult and 32.7 kg for children, and Ta (Averaging time) 365 days/year × number of exposure years (70 years).

Metal pollution index (MPI)

$$MPI = (C_{f1} \times C_{f2} \times .... \times C_{fn})^{1/n} \qquad ..(4)$$

The Metal pollution index was used to assess the total metal content for a vegetable by calculating the geometric mean concentrations of all the studied metals in the vegetable as per equation (4) (Usero *et al.*, 1997).

$$MPI = (C_{f1} + C_{f2} \times ......\times C_{fn})^{1/n} \qquad .. (4)$$

Where, is Concentration of nth metal in the sample.

Results and Discussion

Heavy metals concentration for the present study varied among different vegetables (Fig. 2). The average concentration (mg/kg dry weight) of Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn were 149.7, 0.27, 0.10, 3.97, 17.18, 260.26, 32.59, 1.45, 1.38, and 29.10. The relative abundance of heavy metal in the vegetable samples was in the order of Fe > Al > Mn> Zn > Cu > Cr > Ni > Pb > Cd > Co in leafy vegetables, Fe > Al > Zn > Cu >Mn > Pb > Cr > Ni > Cd > Co in fruity vegetables, Fe > Al > Zn > Mn> Cu > Cr > Pb > Ni > Cd > Co in bulb, Fe > Al > Zn > Mn > Cu > Ni > Cr > Pb > Cd > Co in legume and, Fe > Zn > Al >Mn> Cu > Cr > Ni > Pb > Cd > Co in flower vegetables. The metal concentrations of vegetables were compared with the permissible limit prescribed by food and agricultural organization (FAO)/World health organization (WHO) standards and it was found that the average concentrations of Cd, Co and Pb have exceeded the permissible limits in almost all the vegetable samples. The detailed description of heavy metals contamination in vegetables was as given below.

Aluminium

A significant amount of aluminium was detected in all the vegetable samples varying from 1462.50 mg kg^{-1} in mint to 35.40 mg kg^{-1} in beans. The average aluminium concentration in various vegetable was found as mint > onion bulb >amaranthus> bitter gourd > onion bulb > cucumber > lady finger > tomato > ridge gourd > bottle gourd > round gourd > brinjal > cauliflower > cabbage > chilli > beans. A high concentration of Al was observed in leafy vegetables like mint (1462.50 mg kg^{-1}), spinach (499.82 mg kg^{-1}) and amaranths (157.24 mg kg^{-1}). Very high concentration of Al was found in cabbage in the present study as compared to the concentration reported in the same vegetable in Jamaica by Antoine *et al.* (2017). The potential sources of Al may be coal based power plant, aluminium alloy, die casting industries, and cosmetic industries (ATSDR, 2008).

Cadmium

All the vegetables exceeded the permissible limit of Cd prescribed by FAO/WHO except cabbage (FAO/WHO, 2017). The maximum value of cadmium was found in green chilli (0.91 mg kg^{-1}) and the minimum value was in cabbage (0.1 mg kg^{-1}). The mean concentration of Cd in the vegetables was in order of chilli > brinjal> spinach > lady finger > mint > tomato > amaranth > ridge gourd > cucumber > bitter gourd > bottle gourd > beans > round gourd > cauliflower > onion > cabbage. Average concentration of Cd was 2.2 to 18 times higher than the permissible limits in the vegetables. The average concentration of Cd reported in the present study was higher than the concentration reported in by other researcher in brinjal and amaranth (Haque *et al.*, 2021), cabbage and spinach (Rehman *et al.*, 2019), cucumber (Jalali and Mojahed, 2020), mint (Naghipour *et al.*, 2018), tomato (Jalali and Mojahed, 2020), and ridge gourd (Latif *et al.*, 2018). High concentration of Cd in vegetables may be attributed to atmospheric deposition, application of Cd- containing fertilizers and combustion of fossil fuels (Sharma *et al.*, 2008; Roberts, 2014). Cadmium may result in cancer and several non-carcinogenic effects (gastrointestinal irritation, vomiting excessive salivation etc.) in human body.

Cobalt

Concentration of Cobalt was found above permissible limits (0.05 mg kg^{-1}) in all the vegetables except for onion bulb and Cabbage. The minimum value of Cobalt was found in onion (0.025 mg kg^{-1}) while the maximum (0.25 mg kg^{-1}) was in chilli. Average concentration of Co was 4 and 5 times higher than the permissible limits in mint and chilli. Average Co concentration in the vegetables followed the order of chilli > mint > ridge gourd > amaranth > bottle gourd > cauliflower > round gourd > spinach > lady finger > beans > bitter gourd > brinjal > Cucumber > tomato > onion. A high concentration of Co was found in the present study in mint (Naghipour *et al.*, 2018) and bitter gourd (Kosanovic *et al.*, 2009). Sources of cobalt in the study area may be printing industries, glass and ceramics, chemical and fertilizer, and steel industries (MSME, 2015).

Chromium

Spinach, amaranth, cabbage, mint, cucumber and tomato exceeded the permissible limit of chromium (2.3 mg kg^{-1}) set by FAO/WHO. The value of chromium was the minimum in round gourd (0.77 mg kg^{-1}) and the maximum was found in mint (49.5 mg kg^{-1}). In mint the concentration of Cr was 21.5 times higher than the permissible limits. The mean concentration of Cr in the vegetables was in order of mint > Cabbage > tomato > Amaranth > Spinach > cucumber > onion > cauliflower > ridge gourd > bottle gourd > bitter gourd > brinjal > lady finger > chilli > beans > round gourd. A high concentration of Cr was found in the present study as compared to the other studies in cabbage, onion and spinach (Cheshmazar *et al.*, 2018), and tomato (Jalali and Mojahed, 2020; Zafarzadeh *et al.*, 2018). Ingestion of high concentration of chromium may cause gastrointestinal burns, liver and kidney damage, pulmonary and cardiac arrest, and death. Sources of chromium in the study area may be wastewater irrigation, metal processing industries, and steel industries (MSME, 2015).

Copper

Copper concentration was found to be lowest in onion (4.43 mg kg^{-1}) and the highest in round gourd

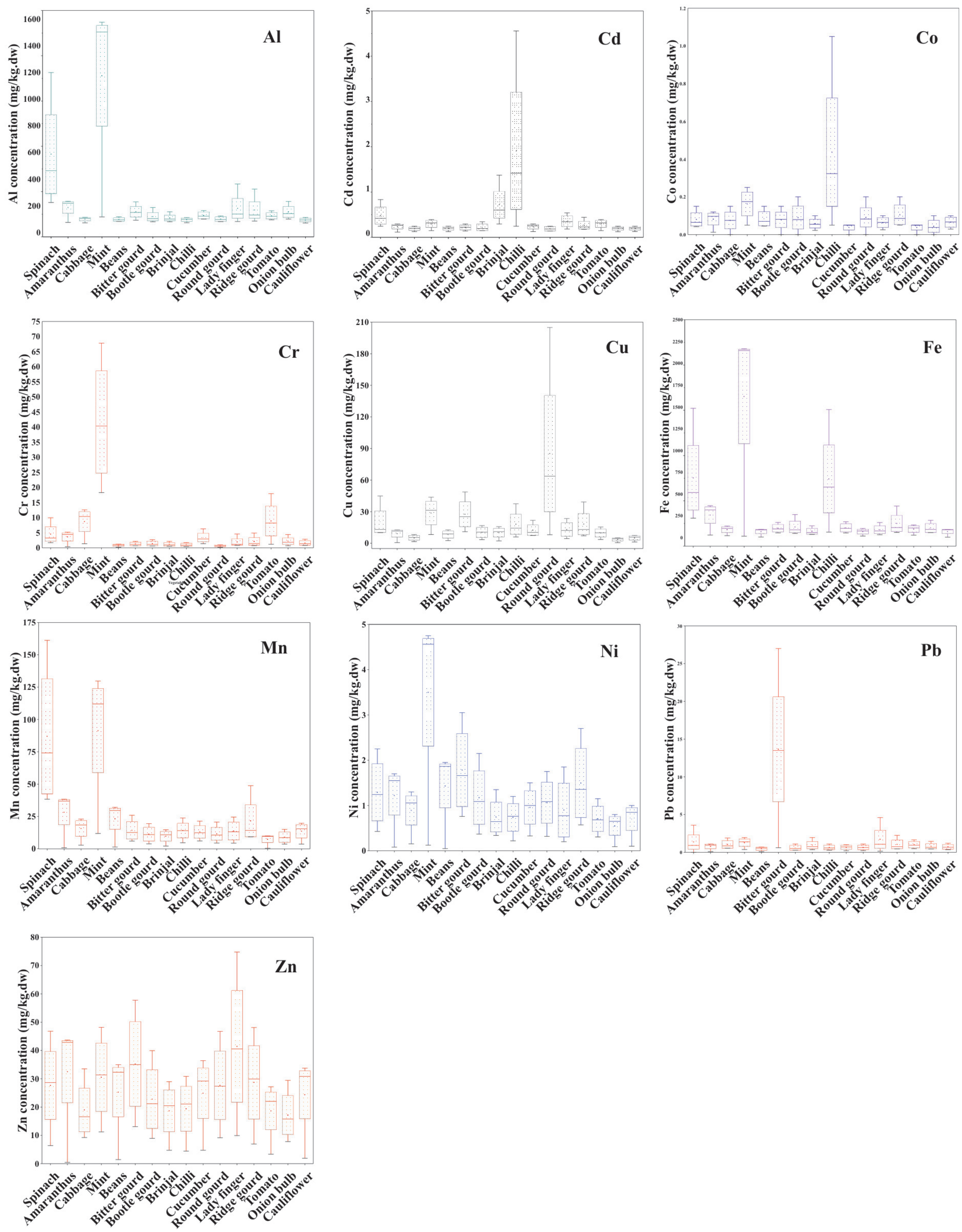

Fig. 2. Heavy metals distribution in vegetables collected during summer from NCRStatistical analysis

(81.87 mg kg^{-1}). The mean concentration of Cu in vegetable samples was in the order of round gourd > mint > bitter gourd > chilli > ridge gourd > Spinach > lady finger > bottle gourd > Amaranth > cucumber > brinjal > tomato > beans > cauliflower > Cabbage > onion. All the samples were having copper concentration well below the permissible limit (40 mg kg^{-1}) of WHO/FAO (Manzoor *et al.*, 2018) except round gourd.

Iron

High accumulation of Fe was found in nearly all vegetable samples whereas it exceeded the permissible limit (425 mg kg^{-1}) of FAO/WHO in spinach, mint and chilli (Latif *et al.*, 2018). The minimum concentration of Fe was observed in brinjal (81.85 mg kg^{-1}) and the maximum concentration was observed in mint (2153.25 mg kg^{-1}). Average concentration of Fe was 5, 1.5 and 1.19 times higher than the permissible limits in mint, spinach, and chilli, respectively. In all the vegetable studied Fe concentration has followed the order mint > Spinach > chilli > amaranth > ridge gourd > cucumber > cabbage > tomato > bitter gourd > onion > bottle gourd > lady finger > cauliflower > beans > round gourd > brinjal. The concentration of Fe reported in the present study was higher than the concentration reported in various parts of India and world in cabbage, cauliflower and onion (Rehman *et al.*, 2019), spinach and tomato (Cheshmazar *et al.*, 2018), ridge gourd (Latif *et al.*, 2018), and bitter gourd (Yap *et al.*, 2019). Steel fabrication and machine manufacturing industries in the vicinity of sampling sites may be a source of Fe in the present study (MSME, 2011).

Manganese

Manganese concentration was found to be highest in mint (117.88 mg kg^{-1}) and the lowest concentration in tomato (9.62 mg kg^{-1}). The mean concentration of Mn in vegetable samples was within the permissible limit of WHO/ FAO (Khan *et al.*, 2015) and followed the order: mint > spinach > amaranth > beans > cabbage > ridge gourd > cauliflower > lady finger > bitter gourd > chilli > cucumber > bottle gourd >brinjal> round gourd > onion > tomato. A high Mn concentration was also reported in cabbage (Rehman *et al.*, 2019) and spinach (Cheshmazar *et al.*, 2018).

Nickel

The mean concentration of Nickel exceeded the permissible limit (1.5 mg kg^{-1}) in amaranth, spinach, mint, beans, bitter gourd and ridge gourd (Nisa *et al.*, 2020). The lowest mean concentration was found in onion (0.7 mg kg^{-1}) and the highest was in mint (4.63 mg kg^{-1}). Ni concentration was found to be 3 times the permissible limit in mint. Concentration of Ni in the vegetables was in the order: mint > bitter gourd > beans > ridge gourd > amaranth > spinach > bottle gourd > round gourd > cucumber > lady finger > cabbage > cauliflower > chilli >brinjal> tomato > onion. A high average concentration of Ni was found in the present study as compared to other studies on cauliflower and cabbage (Rehman *et al.*, 2019) and cucumber (Jalali and Mojahed, 2020).

Lead

Concentration of Pb have exceeded the permissible limit 0.3 mg kg^{-1} for tuberous and 0.1 mg kg^{-1} for bulb vegetables (FAO/WHO, 2017). The minimum average concentration of lead was found in beans (0.65 mg kg^{-1}) while the maximum was in bitter gourd (12.8 mg kg^{-1}). Average concentration of Pb was 2.7 times to 12 times higher than the permissible limits in the different vegetables analyzed in the present study. Pb concentration in the studied vegetable samples followed the order: bitter gourd > mint > tomato > amaranth > cabbage > brinjal > ridge gourd > lady finger > cauliflower > cucumber > Spinach > onion > round gourd > chilli > bottle gourd > beans. A very high Pb concentration was found in the present study in cabbage (Rehman *et al.*, 2019). Metallurigical, automobile, paint industries and road dust are the potential source of Pb in the study area (MSME, 2015).

Zinc

Mean concentration of zinc was found minimum in cabbage (16.6 mg kg^{-1}) and the maximum was in lady finger (47.55 mg kg^{-1}). All the vegetables were within the permissible limits of Zn (60 mg kg^{-1}) prescribed by FAO/WHO standards (Sharma *et al.*, 2008). Zn concentration in the studied vegetable samples followed the order as lady finger > amaranth > bitter gourd > mint > ridge gourd > beans > round gourd > spinach > cauliflower > cucumber > bottle gourd > brinjal > chilli > tomato > onion > cabbage. Zn concentration in our study is higher than the concentration reported in amaranth, bottle gourd and brinjal (Haque *et al.*, 2021), cauliflower (Rehman *et al.*, 2019), cucumber and tomato (Jalali and Mojahed, 2020).

Table 2. Inter-metal Pearson's correlation for vegetables collected from NCR, India

Heavy metals	Al	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Al	1									
Cd	.019	1								
Co	.450	***.559****	1							
Cr	***.934*****	-.059	.381	1						
Cu	.376	-.020	.369	.315	1					
Fe	***.975*****	.162	***.607****	***.931*****	.372	1				
Mn	***.896*****	.060	.364	***.735*****	.254	***.854*****	1			
Ni	***.886*****	-.150	.473	***.847*****	.498*	***.861*****	***.758*****	1		
Pb	-.008	-.136	-.046	-.034	.292	-.042	-.070	.232	1	
Zn	.218	-.162	.238	.081	.383	.170	.246	.440	.358	1

**Correlation is significant at the 0.01 level (2-tailed).

Pearson correlation Matrix showed significantly positive correlation ($p<0.01$) of Al, Cr, Fe, Mn with Ni (Table 2). PCA was performed using varimax with Kaiser Normalization method (Table 3). PC 1 has shown the maximum loading of Al, Fe, Cr, Ni, and Mn indicating their mixed source of origin i.e. natural as well as anthropogenic. Industries are potential sources of Al, Fe, Cr, Ni, and Mn while Al, Fe, and Mn are also present in the earth's crust. PC 2 is strongly correlated with Pb and Zn. Zn can be added by the application of fertilizers in the soil while aerosols may be a potential source of Pb deposition in the vegetables. PC 3 showed the maximum loading of Co and Cd. The dendrogram (Figure 3a) and PCA (Figure 4) suggested the formation of two primary clusters i.e. Al-Fe and Cu-Co. Cluster analysis showed a strong significant correlation among the vegetables by forming primary clusters with a distance of 5 on the scale like bottle gourd, ridge gourd, brinjal, cauliflower, cucumber, tomato and amaranth. Further, it has been observed that spinach, mint and onion bulb is grouped together (Fig. 3b).

Table 3. PCA analysis

	Factor 1	Factor 2	Factor 3
Al	***.967***	-.087	-.193
Cd	.072	-.622	***.697***
Co	.599	-.299	***.639***
Cr	***.898***	-.113	-.295
Cu	.510	.417	.375
Fe	***.971***	-.201	-.057
Mn	***.862***	-.125	-.196
Ni	***.937***	.234	-.085
Pb	.078	***.728***	.276
Zn	.352	***.639***	.314
Eigen Value	5.061	1.720	1.383
% Variance	50.606	17.203	13.826
Cumulative %	50.606	67.809	81.636

Metal Pollution Index (MPI)

MPI in the different collected vegetables was as mint

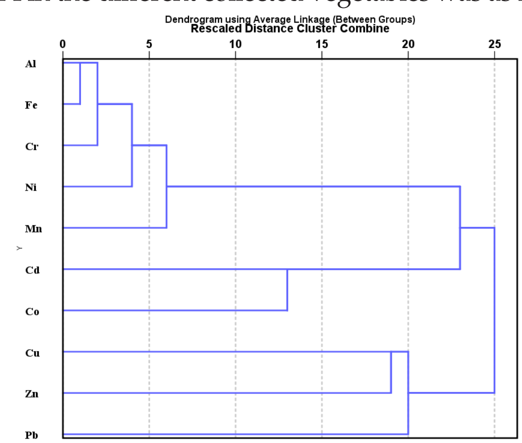

Fig. 3(a). Association between heavy metals

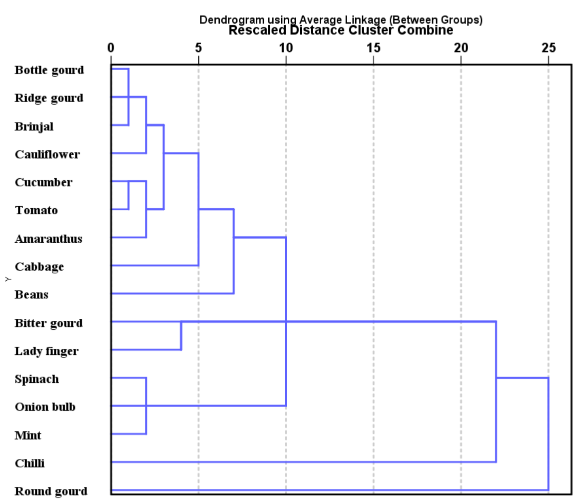

Fig. 3(b). Association between vegetables

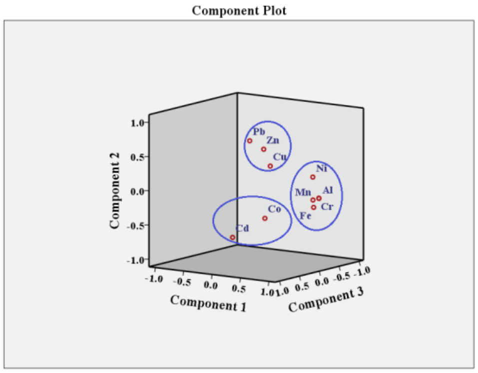

Fig. 4. Principal components plot

> spinach > amaranth > bitter guard > cabbage > chilli > ridge guard > lady finger > tomato > cucumber > round guard > brinjal > bottle guard > beans > cauliflower > onion. Higher values of MPI for spinach, amaranth and cabbage pointed towards high potential health risks due to the consumption of these leafy vegetables (Fig. 5). The MPI reported in the current study is much higher than the MPI reported by Kulshrestha, 2021.

Health risk assessment

The target hazard quotient (THQ) for non-carcinogenic risk of the studied metal from consuming vegetables for adults and children are shown in the fig. 6. THQ was greater than 1 for Cd, Cu, Fe and Pb in most of the vegetables for both adults and children. THQ was greater than 1 for Mn in spinach, mint, cabbage, amaranth, and beans in children. The highest TTHQ value was observed for mustard in adults (23.45) and children (26.96). The high value indicating the potential non-carcinogen may occur due to the consumption of mustard in the population. HI was 214.8 (>1) for adults and 246.92 (>1) for children. Total hazard quotient was also studied for group of vegetables like leafy, fruity, flowery and bulb. THQ was high for leafy vegetables due to contamination of Al, Cd, Cu, Fe, Mn, Pd, and Zn. THQ was > 1 for Fe and Pb in Bulb, for Cu and Pb in flowery, and for Cd, Cu, Fe and Pb in fruity vegetables. Thus potential health risks from exposure to the vegetable are of great concern in the study area.

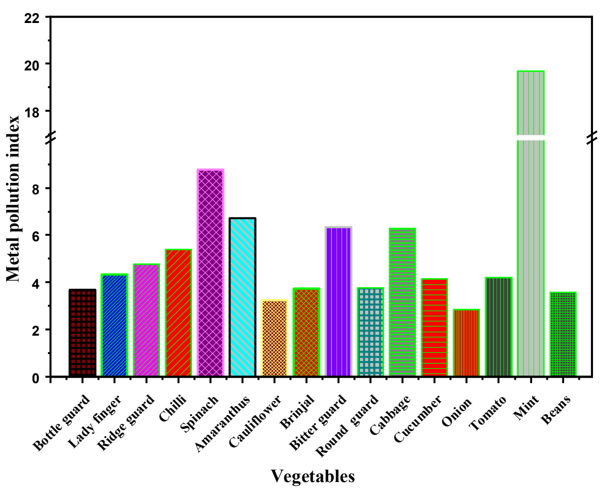

Fig. 5. Metal pollution index for individual vegetables

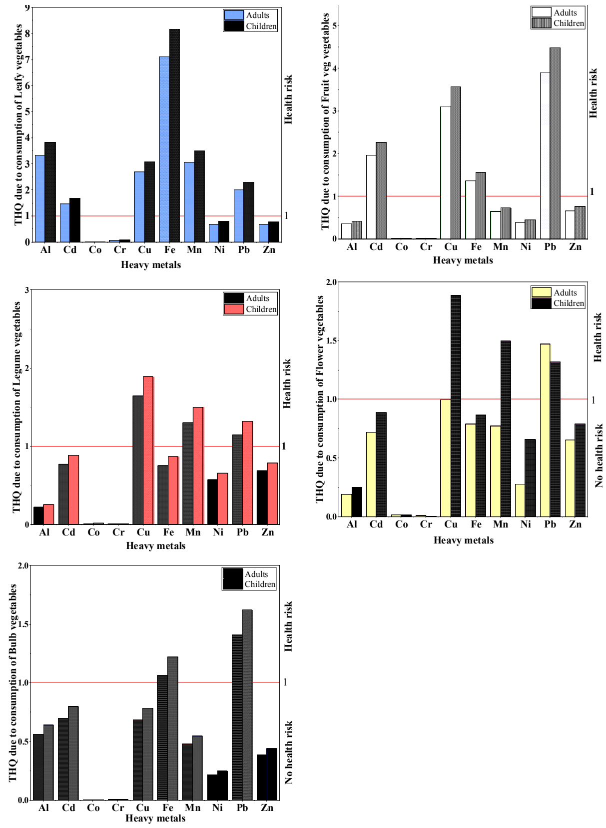

Fig. 6. THQ in adults and children due to ingestion of heavy metals contaminated vegetables

Conclusion

The present study revealed that Cd, Co, Cr and Pb have crossed the permissible limit set by FAO/WHO in most of the vegetable samples while concentrations of Fe, Co and Ni were exceeded their respective permissible limits in few vegetables. Fe was the most prominent metal found in the vegetables while Co was the least reported metal in the present study. Multivariate analysis showed that potential sources of Al, Fe, Cr, Cd, Ni, Pb, and Mn in the vegetable samples is mainly due to industrial activities. The metal pollution index was followed the following order as Leafy > Fruity > bulb > legume > flower. High MPI values for leafy vegetables are pointing towards their high tendency of metal accumulation. Mint consumption may pose a health risk due to the high contents of Al, Cd, Cu, Fe, Mn, and Pb. TTHQ value for adults and children was 45.42 and 52.21 for the mint which is 45 and 52 times higher than the threshold value suggesting potential non-carcinogenic risk due to the presence of multi-metal in vegetable samples.

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