Health Risk Assessment of Heavy Metals Through the Consumption of Vegetables in the National Capital Region, India

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Health risk assessment of heavy metals through the consumption of vegetables in the national capital region, India

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Abstract

This study aimed to evaluate the heavy metal contamination in the vegetables growing in the national capital region of India and to assess the health risk in human resulting from consumption of these vegetables. A total of 99 vegetable samples were collected from the selected study area during the winter season and were analyzed for heavy metals contamination by atomic absorption spectrophotometer. The relative abundance of heavy metal in the vegetable samples was in the following order: $\text{Fe} > \text{Al} > \text{Mn} > \text{Zn} > \text{Cu} > \text{Cr} > \text{Ni} > \text{Pb} > \text{Cd} > \text{Co}$. The mean concentration of Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn, in the vegetables was $158.01 \text{ mg kg}^{-1} \text{ dw}^{-1}$, $0.23 \text{ mg kg}^{-1} \text{ dw}^{-1}$, $0.04 \text{ mg kg}^{-1} \text{ dw}^{-1}$, $3.70 \text{ mg kg}^{-1} \text{ dw}^{-1}$, $7.82 \text{ mg kg}^{-1} \text{ dw}^{-1}$, $297.87 \text{ mg kg}^{-1} \text{ dw}^{-1}$, $39.81 \text{ mg kg}^{-1} \text{ dw}^{-1}$, $1.78 \text{ mg kg}^{-1} \text{ dw}^{-1}$, $0.52 \text{ mg kg}^{-1} \text{ dw}^{-1}$, and $32.21 \text{ mg kg}^{-1} \text{ dw}^{-1}$ for respectively. The statistical analysis supported the formation of two primary clusters Al-Fe-Pb and Cu-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.

Keywords: 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). The human population is growing at a logarithmic rate in the national capital region of India due to the availability of livelihood sources. The increasing population is putting pressure on agriculture to fulfill the food supply demand. Farmers are forced to increase per hectare crop production with less available agricultural land, and hence they are using high doses of pesticides and fertilizers in their fields (Damalas and Eleftherohorinos 2011; Ahmadi et al. 2019). Wastewater irrigation is also getting popular 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. Besides these factors, several other factors are responsible for increasing heavy metals in vegetables such as high traffic movement, atmospheric deposition, or fugitive emissions from unknown sources and industrial emissions (Sharma et al. 2008; Delbari et al. 2012; Kwon et al. 2016; Giri & Singh, 2017). Agricultural products such as wheat, rice, fruits, and vegetables are an important part of the human diet. Vegetables are the sources of carbohydrates, proteins, vitamins, minerals, and trace metals that are essential for proper growth and development of the human body (Akoto et al. 2015). The vegetables growing on heavy metal contaminated soil have the potential to uptake these metals, and they may store metals in stem, root, fruits, and leaves (Khan et al. 2015a). The consumption of these contaminated agricultural products can cause health risks to humans. The adverse health effects of heavy metal exposure are well documented in the literature (WHO 2011;

Tchounwou et al. 2012). The main objective of the present study was to estimate the heavy metal contamination in different types of vegetables and to evaluate the human health risk to the general public of the national capital region via the consumption of these vegetables.

Material and methodology

Study area

Gurgaon, Faridabad, Panipat, and Sonipat are part of the national capital region (NCR) of India and known for fast industrial development in the past few decades. Further, the study area is significantly contributing towards agricultural products, as it has been covered under the green revolution belt and is having a suitable climate for agricultural activities. Most of the study area is irrigated by groundwater except for one site located in Sonipat city, which was irrigated with wastewater. Some industries are disposing of their waste illegally in the study area, which is polluting groundwater (Sarita and Rani 2016). These types of activities further increase the chances of heavy metal contamination in soil and water. So, there is a need to analyze the heavy metal contamination in this area to ensure the good health of the resident. There has been a limited number of published research data evaluating human health risk due to the ingestion of heavy metal contaminated vegetables in the national capital region of India. Keeping in view the facts, assessment of health risk on adults and children was computed due to the consumption of heavy metal contaminated vegetables. The different types of vegetables and the number of samples collected for the study are; beetroot (*Beta vulgaris*) (n=4), brinjal (*Solanum melongena* L.) (n=3), cabbage (*Brassica oleracea* var. *capitata*) (n=4), carrot (*Daucus carota* L.) (n=4), cauliflower (*Brassica oleracea* L. var. *botrytis*) (n=9), chenopodium (*Chenopodium album*) (n=3), coriander (*Coriandrum sativum* L.) (n=5), fenugreek (*Trigonella foenum-graecum*) (n=8), mustard (*Brassica nigra*) (n=10), onion (*Allium cepa*) (n=4), potato (*Solanum tuberosum*) (n=4), pumpkin (*Cucurbita pepo* var. *styriaca*) (n=4), radish (*Raphanus raphanistrum* subsp. *sativus* (L.)) (n=14), round gourd (*Praecitrullus fistulosus*) (n=4), spinach (*Spinacia oleracea*) (n=15), and turnip (*Brassica rapa* subsp. *Rapa*) (n=4).

Sampling

Samples of vegetables were collected from different locations (fig. 1) of NCR during October 2017 to February 2018 in zip-lock poly bags. Vegetable samples were collected from four corners and one center point of each field to prepare a composite sample weighing approximately 2 Kg. The vegetable samples were brought to the laboratory and 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°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. The sieved samples were kept at room temperature for further analysis (Sharma et al. 2008).

Digestion, analysis and quality control

1 gram dried sample of vegetable was digested by adding 15 ml of a tri-acid mixture (HNO₃, H₂SO₄, and HClO₄ in 5:1:1 ratio) at 80°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 by using an atomic absorption spectrophotometer (AAS) (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 (r²=0.999). Blank and standard were run after every 10 samples on the instrument. All the chemicals and standards used were of Merck Company.

Data analysis

The Statistical Program for Social Sciences (SPSS version 22.0) was used to find out the Pearson's correlation coefficient (r), principal component analysis (PCA), and hierarchical cluster analysis (CA) to understand the correlation between heavy metals.

Health risk assessment

Target hazard quotient (THQ)

Health risk can be estimated by calculating the target hazard quotient (THQ) caused by heavy metals exposure by ingestion. The equation used for estimating the THQ for non-carcinogenic risk are given below (Proshad et al. 2019).

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

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

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

$$HI = \sum THQ \quad (3)$$

Where, *Ef* – Exposure frequency (365 days/year for Non- carcinogenic risk and 350 days/year for carcinogenic risk), *Ed* – Exposure duration (70 Years), *Fir* – Ingestion rate of soil (345 g /person /day for adult and 232 g /person /day for children), *Cm* – Concentration of the contaminant in vegetable (mg/kg.dw), *Rfd* - Oral reference dose of heavy metals (mg/kg/day), *Bw* – Average body weight (55.9 kg for adult and 32.7 kg for children), *Ta* – Averaging time (365 days/year × number of exposure years (70 years)), Oral reference dose values (mg kg⁻¹ day⁻¹) for Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn were 1, 0.001, 0.043, 1.5, 0.040, 0.7, 0.014, 0.02, 0.0035, and 0.300, respectively (IRIS, 1995; US-EPA, 2006, 2008; Verma *et al.* 2015; Antoine *et al.* 2017).

Metal pollution index (MPI)

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).

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

Where, C_{fn} : Concentration of n^{th} metal in the sample

Result and discussion:

The relative abundance of heavy metal in the vegetable samples was in the following order: Fe > Al > Mn > Zn > Cu > Cr > Ni > Pb > Cd > Co. Metal concentrations were compared to the permissible limit of food and agricultural organization (FAO)/ World health organization (WHO) standards, as well as, the concentrations reported in earlier studies (Table 1). Mean concentration of metals in different types of vegetables collected from NCR, India is shown in fig. 2 (a-e).

Aluminium

A significant amount of aluminium was detected in all the vegetable samples with minimum and maximum value observed in pumpkin (13.85 mg/kg) and mustard (333.88 mg/kg). We could not find permissible limit for aluminium exposure through vegetable consumption in literature. Average concentration of aluminum in the studied vegetable samples was in the decreasing order of mustard > spinach > carrot leaves > coriander leaves > fenugreek > turnip > chenopodium > brinjal > beet leaves > radish > onion leaves > cauliflower > carrot > potato > beetroots > cabbage > onion bulb > round gourd > pumpkin. Very high concentration of Al was found in cabbage, carrot, pumpkin and turnip in the present study as compared to the concentration reported by Antoine et al. (2017). Aluminium is a toxic non-essential element having harmful effects on nervous and hematopoietic cells in human beings. It brings changes in neurons that are similar to Alzheimer's disease and are known to affect the enzymatic (hexokinase, phosphodiesterase, etc.) activities (ATSDR 2008; Jaishankar et al. 2014).

Cadmium

Cadmium was detected in all the vegetables except in onion leaves, round gourd, and potato. The minimum value of cadmium was found in cabbage and pumpkin (0.05 mg/kg) and the maximum value was in beet leaves (0.5 mg/kg). Amongst leafy vegetable mustard, beetroot, spinach, chenopodium, and carrot leaves exceeded the permissible limit of 0.2 mg/kg. All other vegetables have crossed the permissible limit of 0.05 mg/kg prescribed by FAO/WHO (Joint FAO/WHO food standards programme codex committee on contaminants 2017). The leafy vegetables have a high foliar surface area, high transpiration rate, and fast growth rate (Chabukdhara et al. 2016), and can absorb heavy metals from atmospheric deposition route (Sharma et al. 2008). The mean concentration of Cd in the vegetables was in decreasing order of beet leaves > spinach > mustard > carrot > chenopodium > carrot leaves > turnip > radish > coriander leaves > beetroot > onion bulb > fenugreek > cauliflower > brinjal > cabbage > pumpkin. The average concentration of Cd

reported in the present study was higher than the concentration reported in by other researcher in coriander (Dheri et al. 2007), cabbage and cauliflower (Bo et al. 2009; Guerra et al. 2012), cabbage (Roba et al. 2015; Eliku and Leta, 2017), carrot (Radwan and Salama 2006; Guerra et al. 2012), onion and beetroot (Guerra et al. 2012), onion (Roba et al. 2017), spinach (Dheri et al. 2007; Bo et al. 2009; Guerra et al. 2012), radish (Bo et al. 2009), pumpkin (Antoine *et al.* 2017), and fenugreek (Dheri et al. 2007). Chronic exposure to cadmium can affect the pancreas, kidneys, lungs, urinary bladder, breast, and prostate. Exposure to Cd during pregnancy induces premature birth and reduces birth weight in infants. Cd also induces diabetic complications and osteoporosis (Achparaki et al. 2012; Jaishankar et al. 2014).

Cobalt

Concentration of Cobalt was found above permissible limits (0.05-0.1 mg/kg) in mustard, fenugreek, beet leaves, carrot leaves, spinach, chenopodium, cauliflower, radish, brinjal, pumpkin, and round gourd (Nisa et al. 2020). The minimum value of Cobalt was found in brinjal (0.02 mg/kg) while the maximum (0.25mg/kg) was in beet leaves. Average Co concentration in the vegetables followed the decreasing order of beet leaves > chenopodium > cauliflower > radish > round guard > pumpkin > spinach > fenugreek > mustard > coriander leaves > brinjal. Chronic exposure to Cobalt causes allergy while a high dose of it can cause lung fibrosis and can destroy the male reproductive system. Cobalt also affects the heart, thyroid, liver, and kidney (ATSDR, 2004; Achparaki et al. 2012).

Chromium

All the studied leafy vegetables exceeded the permissible limit of chromium (2.3 mg/kg) set by FAO/WHO, except carrot leaves, chenopodium, and onion leaves. Among root vegetables, radish and turnip exceeded the permissible limits of chromium while in fruity vegetable only brinjal exceeded the permissible limit. In potato also the chromium concentration was higher than the permissible limit of WHO (Khan et al. 2015b). The value of chromium was the minimum in onion bulbs (1.37 mg/kg) and the highest was found in mustard (6.38 mg/kg). The mean concentration of Cr in the vegetables was in decreasing order of mustard > radish > fenugreek > cabbage > spinach > beet leaves > potato > coriander leaves > turnip > brinjal > pumpkin > chenopodium > cauliflower > carrot leaves > round guard > carrot > beetroot > onion leaves > onion bulb. A high concentration of Cr was found in the present study as compared to the other studies in cabbage and onion (Roba et al. 2015; Eliku and Leta, 2017), cabbage, (Bo et al. 2009; Guerra et al. 2012), cauliflower (Bo et al. 2009), carrot (Radwan & Salama, 2006; Guerra et al. 2012), onion and beetroot (Guerra et al. 2012), spinach and brinjal (Chary et al. 2008), radish and mustard (Verma *et al.* 2015), coriander (Dheri et al. 2007; Guerra et al. 2012), potatoes (Bo et al. 2009; Verma et al. 2015), spinach (Dheri et al. 2007; Chary et al. 2008; Bo et al. 2009; Guerra et al. 2012), radish (Bo et al. 2009; Verma et al. 2015), mustard (Verma et al. 2015), brinjal (Chary et al. 2008), and fenugreek (Dheri et al. 2007).

Copper

Copper concentration was found to be lowest in cabbage (3.02 mg/kg) and the highest in pumpkin (20.4 mg/kg). The mean concentration of Cu in vegetable samples was in the decreasing order of pumpkin > round guard > coriander leaves > beet leaves > beetroot > spinach > mustard > brinjal > fenugreek > chenopodium > carrot leaves > carrot > potato > onion bulb > onion leaves > cauliflower > radish > turnip > cabbage. All the samples were having copper concentration well below the permissible limit (40 mg/kg) of WHO/FAO (Manzoor et al. 2018). The mean concentration of Cu in different vegetable samples was higher in the present study than the concentration reported in cabbage (Bo et al. 2009), cabbage and onion (Roba et al. 2015; Eliku and Leta, 2017), carrot (Radwan and Salama 2006), cauliflower and radish (Bo et al. 2009; Verma et al. 2015), beetroot (Verma et al. 2015), brinjal (Chary et al. 2008), radish and mustard (Verma *et al.* 2015), potatoes (Bo et al. 2009; Roba et al. 2015; Verma et al. 2015), spinach (Dheri et al. 2007; Chary et al. 2008; Bo et al. 2009), mustard (Verma et al. 2015), and turnip (Rai and Tripathi 2008), reported by other researchers in India and other countries. Copper can cause mental disorders such as Alzheimer's. It also affects hepatic, renal, and central nervous systems (Achparaki et al. 2012; Hosseini Koupaie and Eskicioglu 2015).

Iron

High accumulation of Fe was found in nearly all vegetable samples whereas it exceeded the permissible limit (425 mg/kg) of FAO/WHO in mustard, fenugreek, spinach, and carrot leaves (Khan et al. 2015b). The minimum concentration of Fe was observed in pumpkin (32.5 mg/kg) and the maximum concentration was observed in mustard (730.72 mg/kg). Fe concentration in the studied vegetable samples was followed the order as mustard > carrot leaves > spinach > fenugreek > coriander leaves > chenopodium > turnip > beet leaves > brinjal > radish > potato > onion leaves > cauliflower > cabbage > carrot > beetroot > round guard > onion bulb > pumpkin. The average concentration of Fe reported in coriander, fenugreek and spinach in the present study is higher than the concentration reported in various parts of India (Arora et al. 2008; Rai & Tripathi, 2008; Punetha & Tewari, 2015). Higher uptake of iron increases the chances of the formation of gastrointestinal ulceration and the development of strictures in humans. Children are more susceptible to iron toxicity (Jaishankar et al. 2014). Fe and Al are present in abundant amounts in the earth's crust as well as used in the inorganic fertilizer and industries which may result in high accumulation of these metals in the vegetables.

Manganese

Manganese concentration was found to be highest in spinach (86.99 mg/kg) and the lowest concentration in potato (8.7 mg/kg). The mean concentration of Mn in vegetable samples was within the permissible limit of WHO/FAO (Khan et al. 2015b) and in the decreasing order of spinach > chenopodium > mustard > beet leaves > coriander leaves > fenugreek > pumpkin > carrot leaves > cabbage > brinjal > beetroot > round guard > cauliflower > turnip > radish > onion bulb > carrot > onion leaves > potato. Mn concentration in coriander (Arora et al. 2008; Punetha and Tewari 2015), spinach (Priya et al. 2014; Punetha and Tewari 2015), and fenugreek (Arora *et al.* 2008). Manganese affects the central nervous system in adults and causes behavioral changes; it reduces the learning ability in children (ATSDR 2012; Portier 2012).

Nickel

Nickel acts as a micronutrient; it catalyzes nitrogen metabolism and is a constituent of the urease enzyme molecule (Nagajyoti et al. 2010). The mean concentration of Nickel exceeded the permissible limit (1.5 mg/kg) in all the collected leafy vegetables except cabbage, carrot leaves, and onion leaves (Nisa et al. 2020). The mean concentrations were above the permissible limit in pumpkin, round gourd, potato, and onion bulb. The lowest mean concentration was found in beetroot (0.8 mg/kg) and the highest was in chenopodium (6.13 mg/kg). Concentration of Ni in the vegetables was in decreasing order of chenopodium > pumpkin > mustard > onion bulb > round guard > spinach > potato > fenugreek > beet leaves > coriander leaves > brinjal > carrot leaves > carrot > cauliflower > radish > cabbage > onion leaves > turnip > beetroot. The average concentration of Ni reported in mustard in the present study is two times higher than the concentration reported in wastewater irrigated mustard from Varanasi, India (Verma et al. 2015). Ni concentration in our study is higher than the concentration reported in cabbage (Guerra et al. 2012), cauliflower (Bo et al. 2009; Guerra et al. 2012), carrot, beetroot, and onion (Guerra et al. 2012), onion (Eliku and Leta, 2017), spinach (Chary et al. 2008; Bo et al. 2009; Guerra et al. 2012), radish (Bo et al. 2009), coriander (Dheri et al. 2007; Guerra et al. 2012), potato (Bo et al. 2009; Verma et al. 2015) and mustard (Verma et al. 2015). Nickel induces allergic contact dermatitis (ACD) and adult respiratory disease syndromes (ARSD). Ni may result in the development of suicidal thoughts and paralysis (Buxton et al. 2019).

Lead

Very high concentrations of lead were found in mustard, fenugreek, coriander leaves, and spinach, whereas in cauliflower, radish, brinjal, potato, and onion bulb it also exceeded the permissible limit of FAO/WHO standards. Permissible limit of Pb for leafy vegetables is 0.3 mg/kg and 0.1 mg/kg for tuberous and bulb vegetables (FAO/WHO, 2017). The minimum average concentration of lead was found in onion bulbs (0.07 mg/kg) while the maximum was in mustard (2.4 mg/kg). Pb concentration in the studied vegetable samples was followed the order as chenopodium > pumpkin > mustard > onion bulb > round guard > spinach > potato > fenugreek > beet leaves > coriander leaves > brinjal > carrot leaves > carrot > cauliflower > radish > cabbage > onion leaves > turnip > beetroot. Pb concentration in spinach, potatoes, cabbage, and cauliflower in present study was higher than the concentration reported by (Bo et al. 2009). Prenatal exposure to lead causes abnormality in offspring, reduces the Ca^{2+} ion absorption in the body and affects the soft tissues like the brain, heart, kidney, and central nervous system; and is also known as mutagen, teratogen, and carcinogens (Bhowmik et al. 2012; Jaishankar et al. 2014)

Zinc

Mean concentration of zinc was found minimum in onion leaves (15.55 mg/kg) and the maximum was in Pumpkin (70.35 mg/kg). Pumpkin exceeded 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 pumpkin > round guard > chenopodium > coriander leaves >

mustard > spinach > fenugreek > beet leaves > cauliflower > beetroot > brinjal > cabbage > turnip > onion bulb > radish > carrot leaves > carrot > potato > onion leaves. The average concentration of Zn reported in radish and mustard was 2 times higher than the concentration reported in Varanasi, India (Verma et al. 2015), and in potato it was 4 times higher than the concentration reported in India and China (Bo et al. 2009; Verma et al. 2015). Zn concentration in our study is higher than the concentration reported in cabbage and onion (Roba et al. 2015; Eliku and Leta, 2017), cabbage and cauliflower (Bo et al. 2009), carrot (Radwan and Salama 2006), beetroot (Verma et al. 2015), Spinach (Rattan et al. 2005; Chary et al. 2008; Bo et al. 2009; Guerra et al. 2012; Punetha & Tewari, 2015), radish (Bo et al. 2009; Verma et al. 2015), mustard (Verma et al. 2015), potato (Roba et al. 2015), and brinjal (Chary et al. 2008). Inhalation of zinc-containing smoke causes respiratory disorder while the ingestion of Zn elevates the risk of prostate cancer and causes sideroblastic anemia (Plum et al. 2010; Hosseini Koupaie & Eskicioglu, 2015).

Statistical analysis

Pearson correlation Matrix showed significantly positive correlation ($p < 0.05$) of Al, Fe, Mn with Pb, and Cu with Zn. Three principal components were obtained by principal component analysis using varimax with Kaiser Normalization. Percentages of variance explained by three principal components are 36.70%, 24.16%, and 17.03% with the cumulative percentage of 77.90 %. PC 1 has shown the maximum loading of Al, Fe, Cr, Pb, and Mn indicating their mixed source of origin i.e. natural as well as anthropogenic. Industries are potential sources of Al, Fe, Cr, Pb, and Mn while Al, Fe, and Mn are also present in the earth's crust relatively in abundant amounts. PC 2 is strongly correlated with Zn and Cu along with some amount of Ni. Zn and Cu can be added by the application of fertilizers in the soil while Ni may be present as a contaminant in fertilizer, irrigation water, and sewage sludge. Air can also be a potential source of metals deposition in the vegetables. PC 3 showed the maximum loading of Co and Cd. The dendrogram (fig. 3a) and PCA (fig. 4) suggested the formation of two primary clusters i.e. Al-Fe-Pb and Cu-Zn, indicating a mixed source of anthropogenic input for these metals which can be industrial and agricultural. Dendrogram using linkage between groups' method was used to classify the studied vegetables into several groups as shown in the fig. 3b. The vegetables were classified into different groups depending upon average heavy metal concentrations in them. Cluster analysis showed a strong significant correlation among all the vegetables except for turnip, pumpkin, and round gourd by forming primary clusters with a distance of 5 on the scale. Further, it can be seen that most of the leafy vegetables are grouped together like mustard, fenugreek, coriander leaves, carrot leaves, and fruit vegetables that are pumpkin and round guard were placed in the same group (fig. 3b).

Metal Pollution Index (MPI)

A metal pollution index is an effective tool for assessing metal pollution in vegetables. Among all collected different vegetable samples, the minimum value of MPI was found in brinjal while maximum in carrot leaves (fig. 5). MPI in the different collected vegetables from NCR, India followed the decreasing order as carrot leaves, mustard, spinach, chenopodium, beet leaves, turnip, potato, round guard, coriander leaves, carrot, beetroot, fenugreek, cauliflower, onion leaves, pumpkin, onion bulb, cabbage, radish and

brinjal with MPI values as 11.32, 10.32, 9.12, 8.55, 8.07, 7.8, 7.68, 7.04, 6.41, 6.06, 6.05, 5.9, 5.29, 4.77, 4.55, 3.6, 3.59, 3.43 and 3.30, respectively. Higher values of MPI for mustard, spinach, and chenopodium pointed towards high potential health risks due to the consumption of these leafy vegetables. MPI among vegetables followed the decreasing order as Fe > Al > Zn > Mn > Cu > Cr > Ni > Pb > Co > Cd. Thus, the highest metal pollution in the vegetables is due to iron and the lowest is due to cadmium.

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 (a-e). The estimation of THQ provides an indication of the potential risk probability in a population exposed to the toxicant. Among the studied metals THQ value of Al exceeded the threshold value of one for adults and children in mustard, fenugreek, coriander leaves, spinach, carrot leaves, radish, carrot, and turnip while THQ was greater than 1 for beetroot in children only. THQ was greater than 1 for Cd in adults and children both by the use of mustard, beet leaves, spinach, and carrot leaves, while it was greater than 1 for children only due to the consumption of coriander leaves and chenopodium. THQ was less than 1 for Cobalt in all the vegetable samples. THQ for Cr was greater than 1 for beetroot and carrot in adults and children. THQ value for Ni and Pb was >1 in Chenopodium and mustard for adults and children. THQ due to Zn was greater than 1 in radish, carrot, turnip, pumpkin, and round guard for adults and children while it was greater than 1 in children in case of coriander leaves, chenopodium, and beetroot. THQ was >1 for Cu, Fe, and Mn in Mustard, fenugreek, beet leaves, coriander leaves, spinach, carrot leaves, and chenopodium for adults and children. THQ was >1 for Cu in radish and onion bulb and for Fe in onion leaves, brinjal, and potato for children. Cu showed higher THQ values for round guard and potato, and Mn has THQ > 1 for brinjal and pumpkin. The total target hazard quotient expresses the non-carcinogen effect of multi-metals in a vegetable sample. 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. TTHQ value due to consumption of vegetables in the study area was in the following decreasing order mustard > spinach > coriander > carrot leaves > chenopodium > fenugreek > beet leaves > pumpkin > turnip > brinjal > round guard > carrot > beetroot > cauliflower > radish > onion bulb > cabbage > potato > onion leaves. The hazard index (HI) is the sum of individual THQ for vegetables. In the present study, HI was 140.85 (>1) for adults and 170.11 (>1) for children. Thus potential health risks from exposure to the vegetable are of great concern in the study area. The result of present study is consistent with the result of Hussain and Qureshi (2019).

Conclusion

The present study revealed that Fe, Co, Pb, Ni, Cd, and Zn have crossed the permissible limit set by FAO/WHO in most of the vegetable samples, indicating potential health hazards to humans due to consumption of these vegetables. Fe was the most prominent metal found in the vegetables followed by Al, Mn, and Zn while Co was the least reported metal in the present study. Multivariate analysis showed that Pb, Cr, Zn, Cu, Co, Cd, and Ni in the vegetable samples were mainly contributed by industrial activities. High

metal pollution indices values (MPI) for leafy vegetables are pointing towards their high tendency of metal accumulation. Mustard consumption may pose a health risk due to the high contents of Al, Cr, Fe, and Pb. The target hazard quotient for aluminium, copper, iron, manganese, and cadmium was greater than 1 for more than 50% of the collected vegetable sample. The total target hazard (TTHQ) value was greater than one in all the vegetables suggesting potential non-carcinogenic risk due to the presence of multi-metal in vegetable samples. TTHQ value for adults and children was 23.45 and 26.95 for the mustard which is 23 and 27 times higher than the threshold value. Thus, we can say that the consumption of mustard and other leafy vegetables are not safe in the study areas.

Declarations

Ethical approval and consent to participate

Not Applicable

Consent for Publication

Not Applicable

Availability of data and materials

All data generated or analyzed during this study are available from first author or corresponding author on request.

Competing interests

The authors declare they have no competing interests.

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Authors' contributions

JR provided and analysed data for the present study, SC helped in interpretation and writing of manuscript and TA provided consultation advice for project. All authors read and approved the final manuscript.

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Table1. Comparison of results with other reports of metal concentrations (mg kg⁻¹ dry weight⁻¹) in vegetables

Country	Al	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	Reference (s)
NCR, India	158.01±18.75	0.23±0.03	0.04±0.01	3.70±0.54	7.82±0.49	297.87±32.92	39.81±3.99	1.78±0.19	0.52±0.17	32.21±1.47	Present study
Coriander , Brazil	--	0.16	0.5	0.02	--	--	--	0.57	1.24	--	Guerra et al. 2012
Coriander leaves, India	--	--	--	125	--	250	10	--	--	7	Punetha and Tewari, 2015
Coriander, India	--	0.03	--	2.22	--	--	--	0.62	2.44	--	Dheri et al. 2007
Coriander, India	--	--	--	--	12.1	313	43.9	--	--	30.9	Arora et al. 2008
Cabbage, Brazil	--	0.12	0.51	0.39	--	--	--	0.54	1.66	--	Guerra et al. 2012
Cabbage, Romania (mg/kg.fw)	--	0.04	--	--	0.9	--	--	--	0.8	4.9	Roba et al. 2015
Cabbage, China (mg/kg.fw)	--	0.005	--	0.013	0.25	--	--	0.048	0.038	2.42	Bo et al. 2009
Cabbage, Ethiopia	--	0.41	--	1.33	2.84	--	--	1.09	0.54	14.4	Eliku and Leta, 2017
Cabbage, Jamaica (mg/kg. fw)	8.49	0.041	--	--	--	--	--	--	--	--	Antoine et al. 2017
Cauliflower, Brazil	--	0.08	0.15	ND	--	--	--	0.24	0.36	--	Guerra et al. 2012
Cauliflower, India	--	0.73	1.06	5.21	1.29	--	--	1.65	6.24	34.53	Verma et al. 2015
Cauliflower, China (mg/kg.fw)	--	0.014	--	0.018	0.58	--	--	0.068	0.032	5.45	Bo et al. 2009
Beetroot, India	--	0.53	1.09	3.05	5.23	--	--	1.35	5.14	25.10	Verma et al.

											2015
Beetroot, Brazil	--	0.09	0.33	0.21	--	--	--	0.26	0.58	--	Guerra et al. 2012
Carrot, Brazil	--	0.03	0.15	0.09	--	--	--	0.11	0.38	--	Guerra et al. 2012
Carrot, Egypt	--	0.01	--	--	1.5	--	--	--	0.18	8.03	Radwan and Salama, 2006
Carrot, Romania (mg/kg.fw)	--	0.59	--	--	7.1	--	--	--	5.4	21.7	Roba et al. 2015
Carrot, Jamaica (mg/kg. fw)	4.25	0.031	--	--	--	--	--	--	--	--	Antoine et al. 2017
Onion, Brazil	--	0.02	0.09	0.07		--	--	0.06	0.49	--	Guerra et al. 2012
Carrot, Romania (mg/kg.fw)	--	0.12	--	--	1.35	--	--	--	1.7	9.6	Roba et al. 2015
Onion, Ethiopia	--	0.22	--	1.25	2.01	--	--	0.53	0.34	9.17	Eliku and Leta, 2017
Spinach, India	--	--	--	2.9	0.09	--	--	0.07	0.03	2.99	Chary et al. 2008
Spinach, India	--	1.96	--	--	27.6	--	--	--	1.44	57.56	Sharma et al. 2009
Spinach, India	--	--	--	--	16.1	734	87.8	13.2	--	38.4	Rattan et al. 2005
Spinach, Brazil	--	0.13	0.46	0.27	--	--	--	0.48	1.05	--	Guerra et al. 2012
Spinach, India	--	--	--	322.5	--	392.5	75	--	--	39	Punetha and Tewari, 2015
Spinach, India	--	2.97	--	32.65	42.06	379	11.47	--	26.35	56.47	Priya et al. 2014

Spinach, India	--	0.08	--	2.53	--	--	--	3.11	5.43	--	Dheri et al. 2007
Spinach, China (mg/kg. fw)	--	0.018	--	0.89	0.7	--	--	0.07	0.03	2.99	Bo et al. 2009
Potato, India	--	0.45	0.26	1.85	3.05	--	--	1.28	4.73	3.88	Verma et al. 2015
Potato, China (mg/kg. fw)	--	0.015		0.03	1.03	--	--	0.054	0.067	3.77	Bo et al. 2009
Potato, Romania (mg/kg.fw)	--	0.03	--	--	2.9	--	--	--	1.2	8.2	Roba et al. 2015
Pumpkin, Jamaica (mg/kg. fw)	2.58	0.015	--	--	--	--	--	--	--	--	Antoine et al. 2017
Radish, India	--	0.62	1.20	3.14	2.92	--	--	1.08	4.36	9.8	Verma et al. 2015
Radish, China (mg/kg.fw)	--	0.012		0.03	0.34	--	--	0.07	0.07	2.48	Bo et al. 2009
Mustard, India	--	0.61	1.24	3.81	1.34	--	--	1.20	5.33	21.50	Verma et al. 2015
Fenugreek, India	--	0.03	--	1.30	--	--	--	2.45	4.7	--	Dheri et al. 2007
Fenugreek, India	--	--	--	--	18.2	302	38	--	--	27.1	Arora et al. 2008
Brinjal, India	--	--	--	1.1	0.7	--	--	3.1	3	4.5	Chary et al. 2008
Turnip, India	--	1.9	--	--	2.8	--	--	--	6.7	--	Rai and Tripathi, 2008
Turnip, Jamaica (mg/kg. fw)	36.69	0.286	--	--	--	--	--	--	--	--	Antoine et al. 2017

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

	Al	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Al	1									
Cd	0.512 [*]	1								
Co	0.050	0.325	1							
Cr	0.535 [*]	0.376	0.281	1						
Cu	0.092	0.079	0.297	-0.063	1					
Fe	0.963 ^{**}	0.438	0.052	0.594 ^{**}	0.128	1				
Mn	0.710 ^{**}	0.676 ^{**}	0.465 [*]	0.492 [*]	0.364	0.660 ^{**}	1			
Ni	0.108	0.070	0.451	-0.024	0.309	0.132	0.442	1		
Pb	0.698 ^{**}	0.336	0.047	0.695 ^{**}	0.144	0.767 ^{**}	0.509 [*]	0.113	1	
Zn	0.164	-0.001	0.525 [*]	0.093	0.832 ^{**}	0.181	0.482 [*]	0.535 [*]	0.223	1

*Correlation is significant at the 0.05 level (2-tailed), **Correlation is significant at the 0.01 level (2-tailed).

Table 3 PCA

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Fig. 1 Map of the study area

Fig. 2 (a) Aluminium and cadmium concentrations in vegetables collected during winter from NCR

Fig. 2 (b) Cobalt and chromium concentrations in vegetables collected during winter from NCR

Fig. 2 (c) Copper and iron concentrations in vegetables collected during winter from NCR

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55 **Fig. 2 (d)** Manganese and nickel concentrations in vegetables collected during winter from NCR

Fig. 2 (e) Lead and zinc concentrations in vegetables collected during winter from NCR

Fig. 3 (a) Dendrogram showing association between heavy metals

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Fig. 3 (b) Dendrogram showing association between vegetables depending on heavy metals concentrations

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Fig. 4 Principal components plot

Fig. 5 Metal pollution index for individual vegetables

Fig. 6 (a) THQ in adults and children due to heavy metals ingestion through vegetables

Fig. 6 (b) THQ in adults and children due to heavy metals ingestion through vegetables

Fig. 6 (c) THQ in adults and children due to heavy metals ingestion through vegetables

Fig. 6 (d) THQ in adults and children due to heavy metals ingestion through vegetables

Fig. 6 (e) THQ in adults and children due to heavy metals ingestion through vegetables