



Lead and cadmium contamination and exposure risk assessment via consumption of vegetables grown in agricultural soils of five-selected regions of Pakistan



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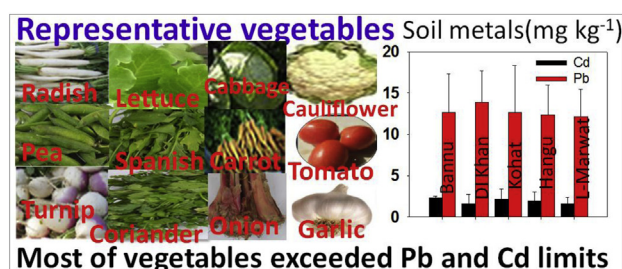
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HIGHLIGHTS

- Cd and Pb concentrations in some vegetables exceeded their respective limits.
- Health risk due to Cd and Pb exposure via ingestion of vegetable was determined.
- Pb concentrations in all soils were below the maximum allowable limit (350 mg kg^{-1}).
- Soil Cd exceeded the limits of European Union (1.5 mg kg^{-1}) and China (0.6 mg kg^{-1}).
- Pb and Cd in vegetables pose a potential health risk to the local residents.

GRAPHICAL ABSTRACT



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ABSTRACT

Rapid urbanization and industrialization result in serious contamination of soil with toxic metals such as lead (Pb) and cadmium (Cd), which can lead to deleterious health impacts in the exposed population. This study aimed to investigate Pb and Cd contamination in agricultural soils and vegetables in five different agricultural sites in Pakistan. The metal transfer from soil-to-plant, average daily intake of metals, and health risk index (HRI) were also characterized. The Pb concentrations for all soils were below the maximum allowable limits (MAL 350 mg kg^{-1}) set by State Environmental Protection Administration of China (SEPA), for soils in China, while Cd concentrations in the soils were exceeded the MAL (61.7–73.7% and 4.39–34.3%) set by SEPA (0.6 mg kg^{-1}), and European Union, (1.5 mg kg^{-1}) respectively. The mean Pb concentration in edible parts of vegetables ranged from 1.8 to 11 mg kg^{-1} . The Pb concentrations for leafy vegetables were higher than the fruiting and pulpy vegetables. The Pb concentrations exceeded the MAL (0.3 mg kg^{-1}) for leafy vegetables and the 0.1 mg kg^{-1} MAL for fruity and rooty/tuber vegetables set by FAO/WHO-CODEX. Likewise, all vegetables except *Pisum sativum* (0.12 mg kg^{-1}) contained Cd concentrations that exceeded the MAL set by SEPA. The HRI values for Pb and Cd were <1 for both adults and children for most of the vegetable species except *Luffa acutangula*, *Solanum lycopersicum*, *Benincasa hispida*, *Momordica charantia*, *Aesculantis malvaceae*, *Cucumis sativus*, *Praecitrullus*

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fistulosus, *Brassica oleracea*, and *Colocasia esculanta* for children. Based on these results, consumption of these Pb and Cd contaminated vegetables poses a potential health risk to the local consumers.

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1. Introduction

Food security and safety is of great concern throughout the world due to toxic heavy metal contamination and their associated health risks (Shaheen et al., 2016; Chen et al., 2016; Yousaf et al., 2016; Liu et al., 2008; Khan et al., 2014). The impact of Pb and Cd on human health has long been and continues to be of great concern, particularly for infants and children (Cao et al., 2016; Pan et al., 2016). Heavy metals enter the environment through natural sources (e.g. volcanic-emissions, erosion of soils, and weathering of parent rocks) and human practices (e.g., agricultural activities, manufacturing, mining etc) (ATSDR, 2012; Khan et al., 2014, 2015). For example, it is estimated that about 25,000 to 30,000 tons per year of Cd are released into the earth ecosystem through various sources; about half from the weathering of rocks and a further proportion from forest fires and volcanoes (ATSDR, 1999). Both Pb and Cd are listed as priority hazardous elements and are considered as two of the top 20 contaminants, ranked 2nd and 7th, respectively, by the US-EPA (ATSDR, 2012). The toxic heavy metals such as Pb and Cd can have severe impacts on biological processes ranging from microbial activities to primary production of plants (Khan et al., 2016a; Sinkkonen et al., 2010; Kauppi et al., 2012; Hansia et al., 2014).

Pb has a large spectrum of physical effects such as neurological and gastro-intestinal distress and oncogenic effects (Li et al., 2004a,b). Pb is a neurotoxin that can affect almost every organ or system in the human body, reducing cognitive development and intellectual performance in children and damage kidneys and the reproductive system (e.g., Qin and Chen, 2010). Most of the accumulated Pb is sequestered in the bone and teeth (e.g., Todd, 1996), causing brittle bones and weaknesses in the wrists and fingers. Pb that is stored in bones can re-enter the blood stream during periods of increased bone mineral recycling. Mobilized Pb can be redeposited in the soft tissues of the body and can cause musculo-skeletal, renal, ocular, immunological, and developmental effects (ATSDR, 1999). Pb can also cause chronic health illnesses such as abdominal-pain, nerve damage, lung and stomach cancer, irritability, and headache (e.g., Steenland and Boffetta, 2000; Jarup, 2003). As stated above, children are greatly vulnerable to Pb toxicity and their exposure to increased levels of Pb may cause severe health complications, such as behavioral disorders, memory weakening and reduced capability to understand, while long-term Pb exposure can lead to anemia (e.g., Jarup, 2003). Multiple routes of exposure to Pb exist, including unintentional soil ingestion, intake of Pb contaminated food-stuffs, and inhalation of soil-particles containing Pb. The quantity of Pb that can be transferred via inhalation is less than other pathways (Davies et al., 1990), and food ingestion is in some cases the most significant (Cao et al., 2016; Chen et al., 2016; Lanphear and Roghmann, 1997). Pb pollution of food-crops is critical to assess as vegetables are essential sources of human nutrition (Goswami et al., 2012).

Cd exposure may cause severe health effects including lung cancer, reproductive system impacts, gastro-intestinal, osteoporosis, prostate, endocrine disorder, cardio-vascular impacts (Martínez-Sánchez et al., 2011; Oliveira et al., 2014), bone fracture, hypertension, (Turkdogan et al., 2003; Khan et al., 2013a,b), anemia, injury of central nervous system, and liver disease (Prabu, 2009;

Asfaw et al., 2013). Cd intake due to the ingestion of environmentally contaminated food-crops was related also with a potential risk of post-menopausal breast cancer (Hiroaki et al., 2014). Furthermore, there are associations between Cd soil pollution and human health risks, for instance the contamination of soils by Japan's Jinzu River and its link with the 'itai-itai' disease (Robson et al., 2014). In the area adjoining to Hunan and Guangdong states (China), decades of metal production were revealed to have contaminated river sediments and agricultural soils (Robson et al., 2014) and residents are considered at risk of chronic health effects from consuming nearby grown food-crops (Zeng et al., 2009).

Pb and Cd are persistent in the environment, and are not removed by normal cropping practices nor easily leached by rain-water because of their strong affinities with the soil solid phase (Tandi et al., 2004; Rehman et al., 2013). Thus, soil constitutes a significant reservoir of Pb and Cd in many systems (e.g., Thuy et al., 2000). Vegetables take up and accumulate toxic metals in their edible and non-edible parts not only through the root system from the soil, but also through aerial deposition of contaminated dust from the air (Li et al., 2004a,b). The uptake of metals by plants depends on soil properties and various physiologic-factors of the plant (Khan et al., 2013a,b). These factors bring considerable uncertainties to estimating potential doses through oral intake compared to other exposure pathways such as soil ingestion and dust inhalation (Khan et al., 2016b).

The objective of this study was to determine the extent of Pb and Cd contamination in selected vegetables and associated soils of five growing areas of Khyber Pakhtunkhwa (KP) Province, Pakistan. Vegetables production is a significant profession in the selected parts of KP, Pakistan. Nevertheless, there has been to date no principal study of Pb and Cd in food crops and agricultural lands in this part of the KP. It is, therefore, critical to examine Pb and Cd accumulation in vegetable species and farmlands as well as their possible health risk. Thus, we conducted a detailed investigation of Pb and Cd concentrations in soils and vegetables grown in the study site, that may have been impacted by numerous sources, including agro-chemical fertilizers, pesticides, herbicides, sewage sludge, and waste-water irrigation.

2. Materials and methods

2.1. Study site description

The proposed study sites comprise five selected vegetable growing areas, Hangu, Kohat, Bannu, Lakki Marwat and Dera Ismail Khan (DI Khan), of KP Province, Pakistan, which lies between 31° 15' 00" to 33° 35' 00" north-latitude and 70° 11' 00" to 72° 01' 00" east-longitude with total population of 1,343,020. The total land under cultivation is approximately 5225.71 km², with planted with various vegetables and other food crops (Rehman et al., 2016; Waqas et al., 2014). Wheat, barley, maize, rice, sugarcane, cucumber, bitter melon, ridge gourd, onion, garlic, mint, lady finger, squash-melon, lettuce, spinach, pea, pumpkin, cabbage, cauliflower, potato, brinjal, turnips, pepper, carrot, radish, tomato, yam, perslane, Chinese onion, and coriander are the most important food-crops grown in the study area. The rivers Indus and Kurram, and their tributaries, as well as tube-wells are the chief sources of

irrigation. The growers sell their vegetables in the local markets and also supply to other regions of the country as a source of income generation (District Census Report (DCR), 1998a,1998b,1998c,1998d,1998e).

2.2. Sampling and pre-treatment

The sampling sites were located in the selected areas of KP, Pakistan as shown in Fig. 1. Soil samples were collected from boreholes 20 cm deep and 10 cm in diameter, drilled with a stainless-steel-auger. Each soil sample comprised 5 sub-samples collected at distances of about 10 m from each other in different directions using the quartile method, followed by bulking together to create a composite sample of 1 kg (Wu et al., 2010). The samples were placed into sealed plastic bags and taken to the laboratory. After air drying, the soil samples were mechanically ground and passed through a sieve of 2-mm mesh, and preserved in clean zip sealed plastic bags for further chemical analyses. Basic soil properties such as pH, electrical conductivity, total carbon, nitrogen and sulfur, dissolved organic carbon (DOC), soil texture, and bulk density are presented in our previous studies (Waqas et al., 2014; Rehman et al., 2016).

Vegetables grown in the study area (Table 1) were harvested from the same cultivated fields from where the soil samples were collected. The vegetable samples were packed into clean polyethylene bags and transported to the laboratory of Department of Environmental Sciences, University of Peshawar, Pakistan. The collected samples were washed with double deionized water to eliminate air borne pollutants, dirt, and dust particles, then the edible parts of the samples were dried in the oven at 65 °C for 72 h until constant weight. The dried vegetable samples were powdered using an electric grinder and stored in labeled paper bags for acid extraction and heavy metal analyses.

2.3. Extraction

The fine powdered soil samples (1 g) were placed in a typhlon beaker, adding 15 ml of aqua regia (HNO_3 : HCl at 1:3 ratio) following the method adopted by Khan et al. (2010). The soil samples were retained overnight and then next morning heated slowly on a hot plate in a fume hood, until near to dryness. After adding HClO_4 (5 ml), the samples were heated again gradually until near to dryness. Diluted acid solution was added and filtered through watt-man filter paper No. 42 into volumetric flasks (50 ml).

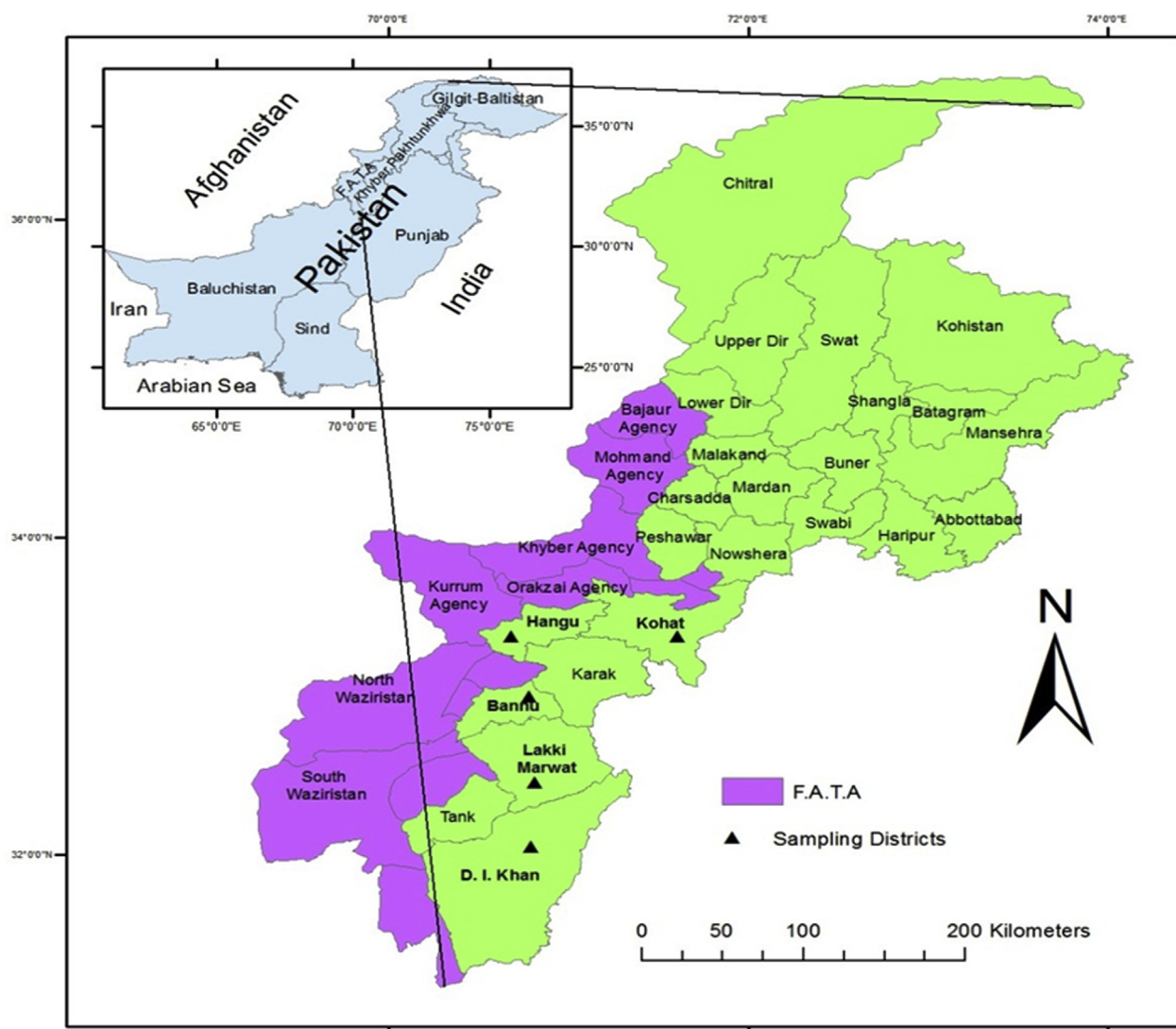


Fig. 1. Location map of the study area showing sampling sites in the selected region of KP, Pakistan (modified from Rehman et al., 2016).

Table 1
Description (botanical and vernacular names) of vegetables grown in the study area.

English name	Vernacular name	Botanical name	Abbreviations used
Bitter melon	Kareela	<i>Momordica charantia</i>	<i>M. charantia</i>
Ridge gourd	Tori	<i>Luffa acutangula</i>	<i>L. acutangula</i>
Tomato	Tamator	<i>Solanum lycopersicum</i>	<i>S. lycopersicum</i>
Lady finger	Bindhi	<i>Aesculantis malvaceae</i>	<i>A. malvaceae</i>
Cucumber	Keera	<i>Cucumis sativus</i>	<i>C. sativus</i>
Brinjal	Bangan	<i>Solanum melongena</i>	<i>S. melongena</i>
Squash-melon	Tinda	<i>Praecitrullus fistulosus</i>	<i>P. fistulosus</i>
Cauliflower	Gobhi	<i>Brassica oleracea</i>	<i>B. oleracea</i>
Pumpkin	Kadhu	<i>Benincasa hispida</i>	<i>B. hispida</i>
Pea	Matter	<i>Pisum sativum</i>	<i>P. sativum</i>
Mint	Podina	<i>Mentha arvensis</i>	<i>M. arvensis</i>
Lettuce	Salad	<i>Lactuca sativa</i>	<i>L. sativa</i>
Spinach	Paalak	<i>Beta vulgaris</i>	<i>B. vulgaris</i>
Cabbage	Band gobhi	<i>Brassica oleracea</i>	<i>B. oleracea</i>
Coriander	Dannya	<i>Coriandrum sativum</i>	<i>C. sativum</i>
Chinese onion	Gandana	<i>Allium chinense</i>	<i>A. chinense</i>
Perslane	Kolfeka sag	<i>Portulaca oleracea</i>	<i>P. oleracea</i>
Onion	Pyaz	<i>Allium cepa</i>	<i>A. cepa</i>
Garlic	Oga	<i>Allium sativum</i>	<i>A. sativum</i>
Radish	Molli	<i>Raphanus sativus</i>	<i>R. sativus</i>
Turnip	Shalgham	<i>Brassica rapa</i>	<i>B. rapa</i>
Carrot	Gajer	<i>Daucus carota</i>	<i>D. carota</i>
Yam	Kachalu	<i>Colocasia esculanta</i>	<i>C. esculanta</i>
Potato	Aaloo	<i>Solanum tuberosum</i>	<i>S. tuberosum</i>

The filtrates were diluted to 50 ml with double deionized water and stored for analysis.

Dried and powdered vegetable samples (2 g) were weighed in Pyrex beakers and 10 ml of high grade HNO_3 was added and placed overnight at room temperature (Khan et al., 2010). The samples were digested till near to dryness at 190 °C for 1.5 h on a hot plate. After cooling, 5 ml HClO_4 was added and the samples were heated slowly until the completion of digestion process. The samples were filtered, followed by the dilution with double deionized water to required volume (50 ml) in volumetric flasks rinsed with acidified water. The samples were sealed and stored at room temperature for further analyses.

2.4. Analytical procedures

The concentrations of Pb and Cd in soils and vegetable samples were analyzed using Atomic Absorption Spectrophotometry (AAS-700-Perkin-Elmer). Analytical-grade chemicals with a high-purity of 99.9% (Merck Darmstadt, Germany) were used for sample preparation and analysis. Pb and Cd standard solution was prepared by diluting its 1000 mg L^{-1} specified standard solutions (F. Kamica Busch, Switzerland). The blank reagents and standard reference soil (GBW-07406 (GSS-6) and plant (GBW-07602 (GSV-1) materials purchased from National Research Center for Certified Reference-Materials, China, were used to verify the accuracy and precision of digestion. The recovery of Cd ($101 \pm 4.5\%$) and Pb ($93 \pm 7.3\%$) was satisfactory from these reference materials. For data quality assurance, each digested sample was analyzed in a triplicate under standard conditions within the confidence level of 95%. All analyses were accomplished in the Centralized-Resource Laboratory (CRL), University of Peshawar, Pakistan.

2.5. Data analysis

The bioconcentration factor (BCF) of metals from soil to plant was computed as the ratio of metal concentration in edible parts of vegetables with the metal concentration in soils. The BCF was determined by the following equation (1) (Cui et al., 2005):

$$\text{BCF} = \frac{C(\text{vegetable})}{C(\text{soil})} \quad (1)$$

where $C_{\text{vegetable}}$ and C_{soil} represent the metal concentrations in the extracts of vegetables and soils on a dry weight basis, respectively.

The average daily intake of metals (DIMs) was calculated according to the following formula (2) as used by Khan et al. (2008, 2010) and Jan et al. (2010):

$$\text{DIM} = \frac{C_m \times C_f \times \text{IR}_{\text{veg}}}{\text{Bw}} \quad (2)$$

where C_m , C_f , IR_{veg} , and Bw represent the metal concentrations in vegetables (mg kg^{-1}), conversion factor (0.085) for conversion of fresh to dry weight vegetables (Jan et al., 2010), ingestion rate of vegetable, and average body weight, respectively. The average daily ingestion rate of food-crops for adults (both male and female) and child were considered to be 0.345 and 0.232 $\text{kg-person}^{-1}\text{-day}^{-1}$, respectively (Khan et al., 2008, 2010), while the average adult and child body weights were considered as 73 kg and 32.7 kg, respectively (Jan et al., 2010).

To estimate the chronic health risk, the health risk index (HRI) for Pb and Cd through contaminated food-crop consumption was determined using the following formula (3) (Khan et al., 2008; Jan et al., 2010):

$$\text{HRI} = \frac{\text{DIM}}{\text{RfD}} \quad (3)$$

Here, HRI, DIM, and RfD , represent the human health risk index, daily intake of metal, and reference dose of metal, respectively. According to USEPA (2005) database the oral toxicity reference dose value (RfD) for Pb and Cd is $3.50\text{E}-03$ and $5.0\text{E}-04$ $\text{mg kg}^{-1}\text{day}^{-1}$, respectively, (USEPA, 2005; Shah et al., 2012). The exposed population is considered to experience no significant risk when $\text{HRI} < 1$ (Khan et al., 2008; Muhammad et al., 2011).

The data were statistically analyzed using Microsoft Excel (2010) computer packages. The measurements were expressed in term of mean and standard deviation. The location map of the study area was prepared using Arc-Geographic Information System (Arc-GIS).

3. Results and discussion

3.1. Pb and Cd concentrations in soil samples

The concentrations of Pb in the soil samples collected from all study areas are summarized in Fig. 2, while the detail is given in Supporting Information (SI) (Table S1). The Pb concentrations varied from 12.2 to 13.9 mg kg^{-1} . The trend of mean Pb concentration in soils for the five locations were DI Khan > Kohat > Bannu > Hangu > Lakki Marwat. Pb concentrations for all samples are below the safe maximum allowable limits (MAL 350 mg kg^{-1}) established by State Environmental-Protection Administration, China (SEPA), for soils in China. These limits are used as a reference as no such values have been established in Pakistan. The Pb concentrations in soils of this study were lower than those reported in the previous studies undertaken by Jie et al. (2009) in contaminated vegetable soils from the Pb/Zn mining and smelting areas in Hunan Province of China, Khan et al. (2010), in Gilgit northern Pakistan; Lu et al. (2011), Douay et al. (2013), in northern France; Zheng et al. (2013), near the edge of the Lake Eyre Basin in the semi-arid region of continental Australia; Attanayake et al. (2014), in urban community garden located in the Washington Wheatley neighborhood in Kansas City, Missouri; and greater than the results of the previous studies at Ghazipur roadside, Bangladesh (Naser et al.,

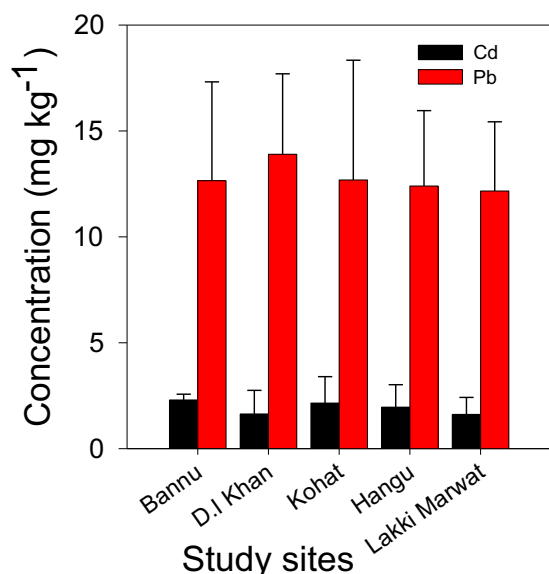


Fig. 2. Comparison of Pb and Cd concentrations (mg kg^{-1}) in the agricultural soils collected from the study area. The error bars indicate the standard deviations.

2012).

The results presented in Fig. 2 indicate that the mean concentration of Cd in soils collected from the five study areas varied from 1.6 to 2.3 mg kg^{-1} . Further detail is given in SI (Table S2). The lowest Cd concentration (0.15 mg kg^{-1}) was observed in the soils of Hangu sub-site, while the highest (3.43 mg kg^{-1}) was in soils of DI Khan area. The Cd concentrations were higher than its MAL (0.6 mg kg^{-1}) set by SEPA (1995) for soil in China, and higher than the MAL (1.5 mg kg^{-1}) set by the European Union (EU) (2000). The Cd concentrations in soils were higher than those reported by Khan et al. (2010) and lower than those investigated by Shah et al. (2010), Abanzun (2011) and Li et al. (2005) in China. The highest contents of Cd in the farm-field soils may be due to the extensive use of various agro-chemicals such as phosphate fertilizers, manures, and pesticides (Alam et al., 2003). Sabiha-Javied et al. (2009) reported that phosphate rock used for production of fertilizers contaminated with heavy metals such as Cd and Pb and acts as sources of heavy metal pollution of air, water, soil and food chain.

3.2. Pb and Cd concentrations in vegetables

The Pb concentrations in edible parts of various vegetable species collected from the study area are presented in Table 2 and Table S3. According to FAO/WHO (2001), the MAL of Pb is 0.3 mg kg^{-1} for fruits and vegetables, on a dry-weight-basis. Slight variations were observed in the concentrations of Pb in different vegetable species collected from the selected study area. The mean Pb concentration ranged between 1.8 and 11 mg kg^{-1} , all of which exceed the MAL of FAO/WHO (Fig. 3). The sequence of mean Pb concentration in the selected vegetable species was in the order of *P. oleracea* > *L. sativa* > *S. lycopersicum* > *B. hispida* > *M. arvensis* = *B. olemcea* > *L. acutangula* > *C. sativum* > *A. chinense* > *S. melongena* > *A. cepa* > *P. fistulosus* > *A. Malvaceae* > *C. sativus* > *B. vulgaris* > *S. tuberosum* > *B. rapa* > *B. oleracea* > *M. charantia* > *C. esculanta* > *D. carota* > *A. sativum* > *R. sativus* > *P. sativum*. The highest concentration of Pb was observed in samples of *P. oleracea* (leafy vegetable), while the lowest was observed in *A. sativum* (root vegetable). The results revealed that Pb concentration was higher than those reported by Mohajer et al. (2012), in Isfahan Province, Iran, and lower than the findings of Tewari and Pande (2013) in Tarai region of Kumaun

Table 2

DIM and HRI for Pb and Cd via consumption of different vegetables grown in the study area (Bold values have exceeded the safe limit i.e. one).

Species	Individual	DIM		HRI	
		Pb	Cd	Pb	Cd
<i>M. charantia</i>	Adult	7.33E-04	3.94E-04	2.09E-01	7.88E-01
	Children	1.10E-03	5.92E-04	3.14E-01	1.18E + 00
<i>L. acutangula</i>	Adult	2.43E-03	4.34E-04	6.93E-01	8.69E-01
	Children	3.64E-03	6.52E-04	1.04E + 00	1.30E + 00
<i>S. lycopersicum</i>	Adult	3.08E-03	4.83E-04	8.81E-01	9.66E-01
	Children	4.63E-03	7.25E-04	1.32E + 00	1.45E + 00
<i>A. Malvaceae</i>	Adult	1.69E-03	4.26E-04	4.82E-01	8.53E-01
	Children	2.53E-03	6.40E-04	7.24E-01	1.28E + 00
<i>C. sativus</i>	Adult	1.62E-03	4.79E-04	4.62E-01	9.57E-01
	Children	2.43E-03	7.19E-04	6.94E-01	1.44E + 00
<i>S. melongena</i>	Adult	1.87E-03	3.26E-04	5.33E-01	6.51E-01
	Children	2.80E-03	4.89E-04	8.01E-01	9.78E-01
<i>P. fistulosus</i>	Adult	1.85E-03	4.84E-04	5.28E-01	9.68E-01
	Children	2.77E-03	7.27E-04	7.93E-01	1.45E + 00
<i>B. oleracea</i>	Adult	1.25E-03	2.26E-04	3.57E-01	4.53E-01
	Children	1.87E-03	3.40E-04	5.35E-01	6.80E-01
<i>B. hispida</i>	Adult	3.17E-03	4.99E-04	9.05E-01	9.99E-01
	Children	4.76E-03	7.50E-04	1.36E + 00	1.50E + 00
<i>P. sativum</i>	Adult	5.49E-04	5.19E-05	1.57E-01	1.04E-01
	Children	8.24E-04	7.79E-05	2.35E-01	1.56E-01
<i>M. arvensis</i>	Adult	1.63E-03	1.55E-04	4.67E-01	3.09E-01
	Children	2.45E-03	2.32E-04	7.01E-01	4.64E-01
<i>L. sativa</i>	Adult	1.68E-03	1.99E-04	4.80E-01	3.98E-01
	Children	2.52E-03	2.99E-04	7.20E-01	5.97E-01
<i>B. vulgaris</i>	Adult	1.44E-03	2.82E-04	4.12E-01	5.63E-01
	Children	2.16E-03	4.23E-04	6.18E-01	8.46E-01
<i>B. olemcea</i>	Adult	1.39E-03	1.66E-04	3.96E-01	3.31E-01
	Children	2.08E-03	2.49E-04	5.94E-01	4.98E-01
<i>C. sativum</i>	Adult	1.37E-03	1.97E-04	3.92E-01	3.93E-01
	Children	2.06E-03	2.95E-04	5.89E-01	5.90E-01
<i>A. chinense</i>	Adult	2.20E-03	2.64E-04	6.28E-01	5.29E-01
	Children	3.30E-03	3.97E-04	9.43E-01	7.94E-01
<i>P. oleracea</i>	Adult	6.09E-04	4.12E-04	1.74E-01	8.24E-01
	Children	9.15E-04	6.18E-04	2.61E-01	1.24E + 00
<i>A. cepa</i>	Adult	2.01E-03	2.54E-04	5.73E-01	5.09E-01
	Children	3.01E-03	3.82E-04	8.60E-01	7.64E-01
<i>A. sativum</i>	Adult	1.04E-03	1.60E-04	2.96E-01	3.19E-01
	Children	1.55E-03	2.40E-04	4.44E-01	4.79E-01
<i>R. sativus</i>	Adult	8.95E-04	2.02E-04	2.56E-01	4.04E-01
	Children	1.34E-03	3.03E-04	3.84E-01	6.07E-01
<i>B. rapa</i>	Adult	9.63E-04	2.31E-04	2.75E-01	4.62E-01
	Children	1.45E-03	3.47E-04	4.13E-01	6.94E-01
<i>D. carota</i>	Adult	1.10E-03	1.27E-04	3.15E-01	2.54E-01
	Children	1.65E-03	1.91E-04	4.72E-01	3.82E-01
<i>C. esculanta</i>	Adult	1.27E-03	3.41E-04	3.62E-01	6.83E-01
	Children	1.90E-03	5.13E-04	5.43E-01	1.03E + 00
<i>S. tuberosum</i>	Adult	1.41E-03	2.98E-04	4.02E-01	5.96E-01
	Children	2.11E-03	4.47E-04	6.03E-01	8.95E-01

north-west Himalaya, India.

The results of this study revealed that the concentrations of Cd in vegetable samples collected from the five aforementioned sites of the study were ranged from 0.2 to 1.6 mg kg^{-1} . Cd concentrations for all vegetables, except *P. sativum* (0.12 mg kg^{-1}), exceed the MAL (0.1–0.2 mg kg^{-1}) set by SEPA (2005). Divergence was detected in Cd accumulation in various plants species, which suggest that different vegetables have different Cd accumulation rates (Yang et al., 2009).

3.3. Bioconcentration factor (BCF)

Fig. 4 summarizes the values of BCF for Pb from soil to the edible tissues of various selected vegetables collected from the study sites. Being the key components of human exposure, BCF is essential which reflects a high accumulating potential of the metal uptake by vegetables from the soils on which they are grown. Highest BCF

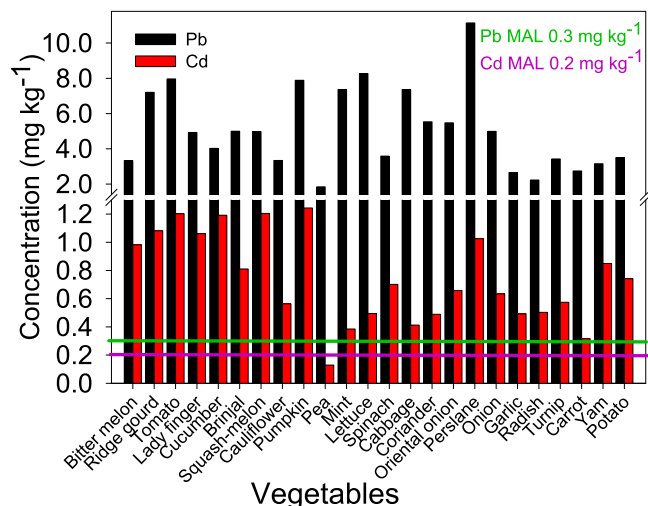


Fig. 3. Comparison of Pb and Cd concentrations (mg kg^{-1}) in vegetables collected from the study area. The horizontal lines indicate the MAL for Cd (purple color 0.2 mg kg^{-1}) and Pb (green color 0.3 mg kg^{-1}). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

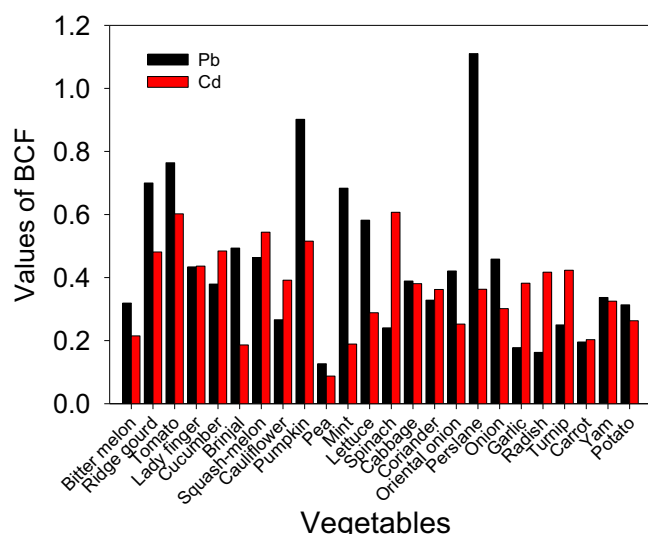


Fig. 4. Bioconcentration factor (BCF) of Pb and Cd for collected samples of vegetables grown in the study area.

(>1) for Pb was observed in *P. oleracea*, having maximum capability to bioaccumulate Pb content in the tissues of its edible parts as compared to all other vegetables investigated for this study, while the smallest BCF was determined for *P. sativum*. Among the groups, the trend of BCF was in decreasing order of leafy > fruity > rooty, which shows that leafy vegetables accumulated greater concentration of Pb as compared to other vegetables. The BCF for Cd was higher in leafy vegetables as compared to fruity and rooty food-crops, consistent with the Pb results.

BCF values for Pb are higher than those observed by Khan et al. (2008), in Beijing China; Garg et al. (2014), in a semi-urbanized area of Haryana state, India, and lower than the findings described by Khan et al. (2010), in Gilgit northern Pakistan. The reported Cd BCF values were lower than those observed by Khan et al. (2010), in Gilgit, northern Pakistan and by Khan et al. (2013a,b) in Swat District, northern Pakistan. This could be attributed to the variations in soil properties such as pH, organic matter, texture as well as

differences in sources of contamination (Khan et al., 2013a,b, 2014).

3.4. Daily intake of metals (DIM) and health risk index (HRI)

To assess the human health risk index of a toxin, it is mandatory to estimate the level of exposure by quantifying the routes of exposure of a contaminant to the target organisms. The exposure of toxic metals to humans occurs through several pathways including inhalation, food-chain, and dermal contact. The DIM and HRI of Pb and Cd were calculated for both adults and children to evaluate the potential human health risk in the study area due to ingestion of vegetables.

Table 2 summarizes the estimated values of DIM and HRI of Pb and Cd through the ingestion of selected food-crops in the entire study area. It is observed that the DIM values were relatively high through the consumption of various selected vegetables grown in the study area. However, all the values of DIM were observed less than one.

The highest intake of Pb was estimated for *P. oleracea*, while the lowest was for *P. sativum* for both adults and children. The calculated values of HRI for Pb and Cd through the ingestion of various vegetables grown in selected sites for both adults and children are also shown in Table 2. The results of this study indicate that the HRI values of Pb were <1 for most of the vegetable species. However, HRI values that exceeded the safe limit (>1) are calculated for *P. oleracea* for adults, and for *L. acutangula*, *S. lycopersicum*, *B. hispada*, *M. arvensis*, *L. sativa*, *B. olemcea*, *P. oleracea* for children. The HRI values for Pb of this study are consistent with the results reported by Singh et al. (2010).

The HRI of Cd was ranged from 0.1 to 1.3 for adults, while it was varied from 0.002 to 1.9 for children (Table 2). The results of Table 2 show that the Cd content was not within the acceptable limits (HRI >1) for several vegetables (US EPA, 1999), specifically *A. Malvaceae*, *C. esculanta*, *L. acutangula*, *M. charantia*, *P. fistulosus*, *P. oleracea*, *S. lycopersicum* for children and *B. hispada* and *P. fistulosus* for adults. HRI values exceeding 1 were reported for both Pb and Cd for a few species, namely *L. acutangula*, *S. lycopersicum*, and *P. oleracea* for children.

4. Conclusion

The concentrations of Pb in all soils were below the MAL (350 mg kg^{-1}) set by SEPA for soils. There were minimal differences in Pb concentration among all the soil samples of the study area. Conversely, Cd concentrations exceeded the MALs (0.6 mg kg^{-1}) and (1.5 mg kg^{-1}) set by SEPA (1995) for soils in China and EU (2000), respectively. Concentrations of Pb and Cd in the vegetables grown in the study area exceeded the MALs. The highest BCF was observed for leafy vegetables followed by fruity > rooty. The values of DIM for Pb and Cd in all the selected vegetables were below one. HRI values exceeded the safe limit (>1) were reported for several vegetables. Consumption of these vegetables poses a potential health risk to the local community.

Hence, it is suggested that the farmers of the study area should be informed about the proper use of fertilizers, pesticides, and herbicides and to test the soils of their agricultural fields prior to cultivation of vegetables. It is recommended that Government departments should formulate strategies for regular monitoring of toxic metal contamination of soils and food-crops to ensure food safety and to help to prevent health problems caused by ingestion of contaminated vegetables.

Conflict of interest

The authors declare that there are no conflicts of interest.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.chemosphere.2016.11.152>.

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