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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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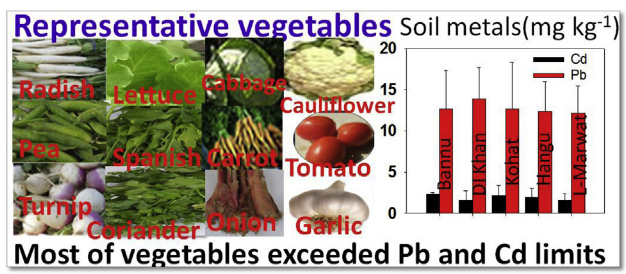
HIGHLIGHTS

- 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^{-}), 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 hispada, Momordi charantia, Aesculantus 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.

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 musculoskeletal, 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 rainwater 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 km^2, 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