Highlights

- In Pakistan, Punjab province is the backbone of agricultural food crops
- Cd concentrations (0.13–8.75 mg kg^{-1}) exceeded the limit in agricultural soils
- CF values showed high degree of contamination to considerable contamination for Cd
- I_{geo} and PERI were moderately however, EF were extremely severe polluted for Cd
- HQ and HI values for adult and child were ≤1, while RI was $\geq 1\times10^{-4}$ via ingestion

Farmlands Degradation with Intensive Agricultural Practices and Human Health Risk Assessment: A Case Study of Province Punjab, Pakistan

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Abstract

Farmlands contamination with heavy metals (HMs) can be considered as a global issue especially in developing countries. The current study investigates the various pollution indices, potential ecological risk index (PERI) and human health risk caused by HMs in some selected regions of Punjab Province, Pakistan. Farmlands soil samples were collected, acid extracted and analyzed via ICP-MS (Agilent 7500c) for selected HMs. All the HMs were found within permissible limits set by worldwide regulatory authorities except Cd which exceeded its limit in 68% agricultural soils of the study area. The CF values for Cd showed high (CF≥6) degree of contamination to considerable contamination (3≤CF≤6). The CD values indicated low (CD≤6) contamination to moderate (6<CD<12) level of contamination. The I_{geo} indices for all HMs were unpolluted except for Cd (moderately polluted $1\leq I_{geo}\leq 2$¿. For Cd the EF values were extremely severe (EF≥50) in the farmland soils of two regions Lahore and Faisalabad, while moderately severe (5≤ EF≤10) in Multan region. The PERI

values were moderate (150≤RI≤300) in the same two regions and low (RI≤150) in other locality. The HQ and HI values for adults and children were observed ≤1 in order of ingestion>dermal>inhalation. Furthermore, the RI values were observed higher than 1.0×10^{-4} for Cd (Lahore and Faisalabad regions) and Cr (Multan and Faisalabad regions) in children working in farmlands and likely exposed to high cancer risk. So, minimization of pollutants must be the top priority of the state to reduce contaminants inputs and immobilization in soil through environmental protection laws and regulations.

Keywords: Heavy metals; Farmlands soil contamination; Ecological risk assessment; Health risk assessment; Carcinogenic risk; Non-carcinogenic risk;

1. Introduction

In the recent decades soil pollution has drawn tremendous attention worldwide (Niazi et al., 2017). Soil not only play a vital role as a resource, that supports the development and survival of humanity but it also acts as a sink for HMs (Antoniadis et al., 2017). Metals having densities greater than 5 g cm^{-3} are generally classified as HMs because of their environmental behaviors and chemical properties (Oves et al., 2012; Khan et al., 2014). HMs can be released from both natural (erosion of parent rocks, weathering, volcanic activities and atmospheric deposition, etc.) and anthropogenic activities (road traffic, sewage, irrigation, mining industries, waste disposal, pesticides and fertilizers use etc.) are the main sources of crops and soil contamination with HMs (Nawab et al., 2019; Nawab et al., 2018a, 2018b; Karbassi et al., 2016; Sekomo et al., 2011; Shah et al., 2010). The release of HMs into the soil from anthropogenic activities is greater than the release of HMs from natural processes (Teng et al., 2014; Desaules, 2012; Li et al., 2012).

HMs can adversely affect water bodies, atmosphere and food chain, as well as they also threaten the well-being and health of humans and ecosystems through materials and energy cycling (Sun et al., 2010; Li and Feng, 2010). Toxic HMs such as chromium (Cr), nickel (Ni), cadmium (Cd), zinc (Zn), lead (Pb), copper (Cu) and manganese (Mn) can create environmental problems due to their persistence and non-biodegradable nature (Khan et al., 2018; Nawab et al., 2018a; Nawab et al., 2016; Nawab et al., 2015; Khan et al., 2010; Radwan and Salama, 2006). Accumulation of HMs in fatty tissues affect the functions of immune system, nervous system, endocrine system, normal cellular metabolism, urogenital system, etc., has confirmed by many investigations (Li et al., 2013; Wang, 2013; Bocca et al., 2004; Waisberg et al., 2003).

In recent years the agricultural soil pollution by HMs has drawn growing public and academic concern globally due to their adverse impacts on human's health and food safety (Liu et al., 2017; Zhang et al., 2016a; Wang et al., 2015; Xue et al., 2014; Sun et al., 2013). Some of the toxic HMs are very persistent in the environment and they are non-biodegradable and thus their accumulation leads to critical levels (Nawab et al., 2018c; Khan et al., 2009). There are no known homeostasis mechanisms for HMs because of their non-degradable nature. Thus, any high concentration of HMs will threaten biological life (Tong and Lam, 2000) and will also cause adverse effect on human's health, ranging from acute to chronic illness (Pei et al., 2015; Wang et al., 2015; Kampa and Castanas, 2008; Shi et al., 2008).

The concentration of HMs in agricultural soil is increasing due to the over use of different chemicals for the better crop yields. The pollutants from agricultural practices include organic, inorganic substances and metals (Bhatti et al., 2018). Due to the harmful health impacts of HMs, several studies have been conducted to determine the concentration of HMs in contaminated food plants. HMs could adversely affect human's health mainly due to their interference in biochemistry of normal body through different metabolic processes (Okunola et al., 2011). Usually, farmer prefer to use wastewater for irrigation purposes to get beneficial nutrients and they ignore the impacts and presence of toxic HMs (Chen et al., 2005; Singh et al., 2004). Although lower concentration of these toxic metals are naturally found in soils, but anthropogenic activities have tremendously increased their concentration (Karbassi et al., 2015). Here, we evaluated the province Punjab current status of soil contamination through various pollution indices, human health status and also proposed strategies to deal with this worsening problem. The focus of this analysis is on HMs in farmlands soils, as these can present a serious threat to human health through the food chain. Special importance is placed on Cd which is the most toxic metal threatening food safety and agricultural sustainability. Thus, it is very important to evaluate and assess the soil pollution by HMs and take immediate and necessary remediation strategies. Therefore, the aim of the study was to investigate the HMs concentrations in farmlands soil of the study area and to estimate ecological and health risk of the selected HMs in both children and adults.

2. Material and methods

2.1 Description of the study area

Punjab is Pakistan's second largest province by area, after Baluchistan, and is the most populous province, with an estimated population of 110,012,442 according to the census report 2017. The province lies between 31.1704° N, north latitudes and 72.7097° E east longitudes. Three regions of province Punjab, Pakistan were selected for HMs analysis including Lahore, Multan and Faisalabad as shown in Fig 1. The regions were selected on the basis of dense population, high number of industries and large agriculture areas. Samples were collected randomly from different location in three regions from agricultural land near to populated areas.

2.2 Soil sampling

The study was carried out around the urban areas in three districts, the farmlands near to industries were selected for sampling. Total (150) composite samples of soil were collected from farmlands in three districts. From each district 50 composite samples were collected with the weight of 1–2 kg at a depth of 0-30 cm with stainless steel auger. Non soil particles such as organic debris, plastics, wooden pieces and stone were removed from soil sample. The samples were collected randomly from different agricultural land at different locations. The collected samples were then stored, labeled in plastic Zip bags and transported to the laboratory for further analysis.

2.3 Sample preparations

In the laboratory, at room temperature, the soil samples were first air dried for 72 hours and then oven dried for 24 hours at 40˚C to obtain constant weight. The dried soil samples were then ground using wooden hammer and sieved through 0.15mm sieve. The samples were mixed to obtain composite sample. The samples were kept in well labelled plastic Zip bags for further analysis. The soil samples were weighed using a digital balance to obtain 1.00 g soil.

2.4Soil extraction

For HMs extraction the samples were acid digested according to the standard procedures adopted by Nawab et al. (2015). For acid digestion 1 g of soil samples were taken in digestion tube and digested with two strong acids HNO_3 and $HCLO_4$ with 3:1 ratio. 10 ml of HNO_3 were added first to the samples and kept overnight at 40˚C. After night 5 ml of HCl were added to the samples and were kept on hot plate on 80˚C for 30 min, then gradually temperature raised up to 180˚C until the solution become transparent. For digestion processes fume hood was used to avoid chemical release. The samples were then filtered through Whatman No. 42. After filtration the final volume of 100 ml was made with double de-ionized water in a clean volumetric flask.

2.5 Heavy metal analysis

The concentrations of HMs such as Pb, Cd, Cr, Zn, Ni and Cu were determined at the Institute of Urban Environment, Chinese Academy of Sciences Xiamen, China using Inductive Coupled Plasma Mass Spectrometry (ICP-MS) (Agilent Technologies, 7500 CX, USA).

2.6 Risk assessment

The different types of pollution indices risk assessment such as contamination factor (CF), geo accumulation index(I¿¿geo)¿, contamination degree (CD), enrichment factor (EF), ecological risk index (ERI), potential ecological risk index (PERI) are summarized in supporting information (S1). Further the risk assessment is a multiple procedure of data collection, evaluation, toxicity evaluation, risk characterization and exposure assessment (USDOE 2011; USEPA 1989). Human's expose to HMs through inhalation, ingestion and dermal contact. In the present study the exposure to selected metals were calculated using the methodology adopted by U.S environmental protection agency (USEPA 1989, 2001a, 2002, 2011). The values for factor used in calculation are given in Table S2.

2.7 Ingestion of HMs via soil

Average daily intake (ADI_{ing}¿ via ingestion (µg/kg/day) for adults and children were calculated in the three-district using the following equation (1).

$$DI_{ing} = \frac{C_S \times IR_{soil} \times ED \times EF \times CF}{BW \times AT} (1)$$

Where C_S, is the concentration of measured HMs in soil sample, IR_{soil} is the ingestion rate mg/day, ED is the exposure duration, EF is the exposure frequency days/year, CF is the conversion factor in mg kg^{-1}, BW is the body weight in (kg) and AT is the average time for individual exposed over a period of time in (days).

2.8 Inhalation of HMs via soil

Average daily intake ADI_{inh} via inhalation (µg/kg/day) values for adults and children were calculated using the following equation (2).

$$ADI_{inh} = \frac{C_S \times IR_{air} \times ED \times EF}{PEF \times BW \times AT} (2)$$

Where C_S, is the concentration of HMs in sample, IR_{air} is the inhalation rate of soil m^3/day and PEF is the particle emission factor in m^3/kg. The rest of factors are already discussed in the previous section.

2.9 Dermal contact with soil

Average daily intake ADI_{derm} via dermal contact (µg/kg/day) for adults and children were calculated using the following equation (3).

$$_{derm} = \frac{C_S \times SA \times AF \times |\times| ED \times EF \times CF}{BW \times AT} (3)$$

Where C_S, is the concentration of heavy metal in the sample, SA is the skin area exposed in cm3, AF is the adherence factor of soil in mg/cm^2 and ABS is the absorption factor through skin. ED, EF, CF, BW and AT are discussed in the previous equation (1) and (2). Furthermore the carcinogenic and non-carcinogenic risk assessment is summarized in supporting information (S1).

2.10 Quality control

The standard reference material of soil (GBW07406-GSS-6) and blank reagents were included in each batch to validate the accuracy of the extraction procedures. All the samples were digested and studied in triplicate. The recovery rates ranged from 95.3±5.2 to 103.1±7.4%.

2.11 Data analysis

Statistical analyses of obtained data in this work were performed by using Special package for Social Sciences (SPSS) version 21, Statistical Software packages of Statistix Version 10 as complementary Software. Means values and standard deviation were calculated using MS EXCEL, 2016.

3. Results and discussion

3.1 HMs in agricultural soil

The spatial distribution of HMs in the study area presented in Figure 2, 3 & 4. The release of HMs and its adverse consequences results in soil deterioration and load mainly in high natural-values regions (Bą̧belewska, 2010; Vrbek and Buzjak, 2004). The quality of soil is impacted with progressive HMs contamination. Additionally, HMs enriched contaminated soil did not decline with the passage of time and reduce the potential of soil resources (Dudzik et al., 2010; Vrbek and Buzjak, 2004). In this study the content of six HMs including Pb, Cd, Cr, Zn, Ni and Cu in the farmland soil of three most important agricultural regions (Lahore, Multan and Faisalabad) province Punjab with the range and mean values are presented in Table 4. The mean concentration of HMs in Lahore region were in order of Pb>Zn>Cr>Cd>Cu>Ni, in Multan region were Cr>Pb>Zn>Cu>Ni>Cd while, in Faisalabad region were in order of Cr>Zn>Pb>Cd>Cu>Ni as shown in Table 4. All the mean concentration value were compared with different regularities authorities globally for HMs in agricultural soil including State Environmental Protection Administration (SEPA, 2015) Environmental Protection Authority Australia (EPPA AUS, 2016) Bulgarian Soil Pollution Standards (BSPS, 2007) Regulatory Standards of Heavy Metal Pollutants in Soil Taiwan (RSHP, 2011) Canadian Ministry of the Environment (CCME, 2009) European Commission on Environment (ECE EU, 2002) Department of Environmental Affairs South Africa (DEA SA, 2010) and United States Environmental Protection Agency (USEPA, 2002). The detailed information are presented in Table 4.

Pb concentration in Lahore, Multan and Faisalabad regions ranged from 10.02–66.27, 0.77–10.79 and 3.47–19.26 mg kg^{-1} while the mean values were 25.04, 6.44 and 8.98 mg kg^{-1} respectively. Pb concentration was observed higher in Lahore region followed by Faisalabad and Multan regions. The maximum concentration of Pb (66.27 mg kg^{-1}) was observed in Lahore regions. Although the Pb concentration in Lahore, Multan and Faisalabad regions were below SEPA (2015), EPPA AUS (2016), CCME (2009), ECE EU (2002), and USEPA (2002) and higher than BSPS (2007) and DEA SA (2010) permissible limits. Moreover, the Pb concentrations in this study was observed higher when compared with Aiman et al. (2015) in Lahore, Iqbal and Shah (2011), Mehmood et al. (2018) and lower than Tahir et al. (2016) and Karim et al. (2014) in Pakistan. The Pb results were also found higher when compared with other countries Cai, et al. (2012) and Xioa et al. (2018) China, and similar to Martín et al. (2013) Spain and Kelepertis (2014) Greece.

Cd concentrations in the same three regions ranged from 0.18–7.54, 0.57–0.94 and 0.13–8.75 mg kg^{-1} while the mean concentrations were 4.26, 0.74 and 4.65 mg kg^{-1} respectively as shown in Table 1. The concentrations of Cd were higher in Faisalabad region followed by Lahore and Multan. The maximum Cd concentration was found in Faisalabad region (8.75 mg kg^{-1}). Cadmium is a highly toxic and carcinogenic metal used in metal plating, nickel-cadmium batteries, plastic stabilizers and pesticides. Cadmium is also present as a pollutant in phosphate fertilizers. Moreover, Faisalabad and Lahore regions are the hubs of industrial and agriculture activities so, these high concentration of Cd in agricultural soil may be due to the different industrial and intensive agricultural practices in the regions. The concentration of Cd in Faisalabad and Lahore regions were higher than SEPA (2015), EPPA AUS (2016), BSPS (2007), RSHP (2011), CCME (2009), ECE EU (2002 and USEPA (2002) permissible limits. The concentration of Cd in Multan region was only higher than SEPA (2015), BSPS (2007) and USEPA (2002) permissible limits. High level of Cd in the study are may be due to vehicle emissions, industrial pollution and intensive agricultural practices (Rehman et al., 2018). Previously high Cd concentration was also reported by Murtaza et al. (2008), Huq et al. (2003) and Butt et al. (2005) in

Pakistan. The Cd values were also observed higher when compared with other parts of the world such as Cai et al. (2012) and Xioa et al. (2018) China, and similar to Martín et al. (2013) Spain and Kelepertis (2014) Greece.

In the same three regions the Cr concentrations ranged from 2.48–9.54, 6.48–63.8 and 3.21–90.6 mg kg^{-1} and the mean values were 6.39, 32.4 and 21.9 mg kg^{-1} respectively as shown in Table 1. Cr concentration was observed higher in Multan region followed by Faisalabad and Lahore region. The maximum concentration of Cr (90.6 mg kg^{-1}) was found in Multan. Although the Cr concentration in (Lahore, Multan and Faisalabad) regions were below than SEPA (2015), EPPA AUS (2016), CCME (2009) and ECE EU (2002) and higher than USEPA (2002 and DEA SA (2010) permissible limits. Similar studies conducted in other regions of Pakistan in terms of Cr content in soil were compared with this study Aiman et al. (2015), Iqbal and Shah (2011), Mehmood et al. (2018) and Hamid et al. (2017) and the results were found lower. Similarly, our results were also observed lower when compared with Doabi et al. (2018).

Zn concentrations ranged from 0.16–107, 0.38–18.3 and 0.22–83.6 mg kg^{-1} while the mean values were 14.8, 4.48 and 20.1 mg kg^{-1} respectively in the same three regions. The concentration of Zn was higher in Faisalabad region followed by Lahore and Multan regions. The maximum Zn concentration was observed in Lahore region (107 mg kg^{-1}). Although the Zn concentration in Lahore, Multan and Faisalabad regions were within the given permissible limits of different countries stated in the previous paragraph. Although the Zn concentration as compared to Tahir et al. (2016) in Pakistan and Xioa et al. (2018) in China were found lower.

Ni concentration in Lahore, Multan and Faisalabad regions ranged from 0.44–0.82, 0.46–2.20 and 0.46–0.76 mg kg^{-1} while the mean concentrations were 0.58, 0.84 and 0.64 mg kg^{-1} as shown in Table 1. Ni concentration was observed higher in Multan region followed by Faisalabad and Lahore region. The maximum concentration of Ni (2.20 mg kg^{-1}) was observed in Multan region. The concentration Ni was found lower than other five HMs in all the three regions. Even though the Ni concentration in Lahore, Multan and Faisalabad regions were found below when compared with other countries permissible limits stated in previous paragraphs. The Ni concentration in this study as compared to Tahir et al. (2016) were found very low and almost similar to Mehmood et al. (2018). The concentration of Ni was also observed lower when compared with Doabi et al. (2018).

Cu concentration in the same three regions ranged from 1.11–10.10, 0.15–2.71 and 0.97–4.27 mg kg^{-1} while the mean concentrations were 3.32, 1.80 and 1.90 mg kg^{-1} respectively as presented in Table 1. The highest Cu concentration was observed in Lahore region followed by Faisalabad and Multan region. Among the three regions the maximum Cu concentration (10.1 mg kg^{-1}) was observed in Lahore region. Moreover, the Cu concentration in the same three regions were within the permissible limits of different countries as presented in the previous paragraphs. The Cu values followed a similar pattern as reported by other studies in Pakistan such as Aiman et al. (2015), Iqbal and Shah (2011), Mehmood et al. (2018) and Hamid et al. (2017).

In the study area all the selected HMs were within permissible limits except Cd which showed elevated levels than the listed countries permissible limits. Moreover, province Punjab is the hub of different fertilizers and pesticides companies because of its agricultural production. This elevated levels of Cd in the agricultural soils may be possibly due to the extensive agricultural practices including the usage of numerous agro-chemicals such as pesticides, phosphate fertilizers and manures (Alam et al., 2003). Another study conducted by Sabiha-Javied et al. (2009) also stated that phosphate rock used for fertilizers production are polluted with HMs such as Pb and Cd and acts as HMs contamination source for surrounding ecosystem. The management strategies to minimize intensive agricultural practices in developing countries like Pakistan is provided in supporting information (S1).

3.2 Contamination factor

The contamination factor (CF) values for all the HMs showed low degree of contamination (CF<1) except Cd as presented in Table 2. The CF values for all HMs in Lahore region were in order of Cd>Pb>Zn>Cr>Cu>Ni; for Multan region the order was Cd>Cr>Pb>Zn>Cu>Ni and Faisalabad region the order was Cd>Cr>Pb>Zn>Ni>Cu respectively. The CF values for Cd in Lahore and Faisalabad regions showed high (CF>6) degree of contamination and were considerable contaminated (3<CF<6) in Multan region. The CF values in Lahore region for Pb, Cd, Cr, Zn, Ni and Cu were 0.31, 8.52, 0.03, 0.05, 0.011 and 0.033 in Multan region 0.08, 1.48, 0.16, 0.01, 0.016 and 0.018 and in Faisalabad region 0.11, 9.9, 0.1, 0.08, 0.012 and 0.019 respectively as shown in Table 2. The degree of contamination showed that Lahore and Faisalabad regions are moderately contaminated (6<CD<12) while Multan regions showed low degree of contamination (CD<6). The contamination degree (CD) values in Lahore, Multan and Faisalabad regions were 8.95, 1.75 and 10.21 respectively as shown in Table 2. A similar study carried out in Pakistan by Iqbal and Shah (2011) also reported moderate level of contamination for Pb and Cd.

3.3 Geo accumulation index

The geo accumulation index values were assessed base on the descriptive classes proposed by (Müller, 1969) as shown in Table 1. The geo accumulation indices were calculate based on the local geochemical background values. The geo accumulation indices ranged from unpolluted ($I_{geo} \leq 0$¿ to moderate polluted ($1< I_{geo} \leq 2$¿ in Lahore, Multan and Faisalabad regions as shown in Table 2. The geo accumulation indices in Lahore region for Pb, Cd, Cr, Zn, Ni and Cu were 0.0628, 1.7098, 0.0064, 0.0118, 0.0023, and 0.0066, for Multan region 0.0161, 0.2970, 0.0324, 0.0035, 0.0033 and 0.0036 and for Faisalabad region were 0.0225, 1.8663, 0.0219, 0.0161, 0.0025, and 0.0038 respectively. Although the geo accumulation indices for all HMs were unpolluted except for Cd in Lahore and Faisalabad were moderately polluted ($1<I_{geo} \leq 2$¿ as shown in Table 2. The previously studies conducted by Aiman et al. (2015) and Karim et al. (2014) also showed unpolluted to moderately polluted level of contamination by HMs in Pakistan. In China Xioa et al. (2018)

also reported that Cd was the main contributor in agricultural soil contamination. Previously in Iran Doabi et al. (2018) showed I_{geo} higher only for Ni in contrast to other HMs.

3.4 Enrichment factor

Enrichment factor (EF) is a technique used for normalization of metals fractions associated with soil and sediments. The calculated EF values for each metal in Lahore, Multan and Faisalabad regions are presented in Table 2. The EF values ranged from low enrichment (EF < 1) to extremely sever (EF > 50) for some HMs in the study areas. The EF values in Lahore region were in order of Cd>Pb>Zn>Cu>Cr>Ni, in Multan region Cd>Cr>Cu>Pb>Ni>Zn, and in Faisalabad region Cd>Pb>Cr>Zn>Cu>Ni respectively. The EF values for Pb, Cd, Cr, Zn, Ni and Cu in Lahore region were 1.82, 50.11, 0.17, 0.17, 0.29, 0.06 and 0.19, in Multan region were 0.47, 8.7, 0.94, 0.05, 0.09 and 0.1, and in Faisalabad region were 0.64, 58.23, 0.58, 0.47, 0.07, 0.11 respectively. The EF values for Cd were extremely severe (EF>50) in Lahore and Faisalabad regions and moderately severe (5<EF<10) in Multan region. Although the overall EF values for other selected HMs low level of enrichment as shown in Table 2. In comparison with Iqbal and Shah (2011) the EF values were moderately contaminated for Pb and Cd as compared to other HMs. Similar study by Karim et al. (2014) showed EF values higher for Pb in contrast to other HMs.

3.5 Potential and ecological risk index

Ecological risk factor (ER) and potential ecological risk index (PERI) are the most useful techniques responsible for risk identification for HMs in soil. Ecological risk factor (ER) can illustrate the risk of individual metal and potential ecological risk index (PERI) can illustrate the risk caused by the overall metals contamination. The investigate soil metals ER and PERI values in Lahore, Multan and Faisalabad regions are presented in Table 2. The order of ER mean values in Lahore region were Cd>Pb>Cu>Cr>Zn>Ni, in Multan region were Cd>Pb>Cr>Cu>Ni>Zn and in Faisalabad region were

Cd>Pb>Cr>Cu>Zn>Ni respectively. The ER values for Pb, Cd, Cr, Zn, Ni and Cu in Lahore region were 1.55, 255.6, 0.06, 0.05, 0.055 and 0.165, in Multan region were 0.4, 44.4, 0.32, 0.01, 0.08 and 0.095 and in Faisalabad region were 0.55, 297, 0.2, 0.08, 0.06, and 0.095 respectively. According to the Hakanson (1980) classification the ER values in the study area ranged from low (E_r^i<40) to high (160 ≤ E_r^i < 320) ecological risk. Although the ER value for all the selected HMs were low except for Cd which showed high ER values. The ER values for Cd in Lahore and Faisalabad regions showed high (160 ≤ E_r^i < 320) and moderate (40≤ E_r^i <80) in Multan region. The PERI values in Lahore, Multan and Faisalabad regions were 257, 45.3 and 298 respectively. The PERI values were moderate (150≤RI<300) in Lahore and Faisalabad regions and low (RI<150) in Multan region as shown in Table 2. The previous study carried out by Aiman et al. (2015) ranged from low to moderate PERI values except Cd which exceeded to high PERI. Furthermore, the study conducted by Xioa et al. (2018) in China also reported that Cd was the main contributor in high ecological risk in their study.

3.6 Health risk assessment

Health risk assessment is considered as a useful technique to characterize the potential adverse effects on human health (USEPA, 2011). Risk assessment is used in different tools of science and statistical measurement and also identify the routes of exposure to HMs and calculate the potential health risk. Health risk assessment is classified in two categories carcinogenic risk and non-carcinogenic risk. Health risk assessment average daily intake (ADI) values were calculated using through different routes of exposure including ingestion, inhalation and dermal. The ADI values via ingestion, inhalation and dermal were calculated for both children's and adults in Lahore, Multan and Faisalabad regions. Non carcinogenic assessment were calculated via HQ and HI values in both children's and adults. Carcinogenic risk assessment values were measured using cancer risk (RI), total cancer risk (TRI) and combined cancer risk (CRI) values.

3.7 ADI via ingestion, inhalation and dermal

The average daily intake (ADI) of HMs via ingestion, inhalation and dermal contact by adults and children in Lahore, Multan and Faisalabad regions are summarized in Table 3. The ADI in adults and children in Lahore were in order of ingestion>dermal>inhalation for all the selected HMs. The highest ADI value for Pb were observed in Lahore region (3.18×10^{-04}) via ingestion in children and lowest (8.24×10^{-10}) via inhalation in Multan region for adults. For Cd highest (5.91×10^{-05}) value was observed for Faisalabad region via ingestion and lowest (9.47×10^{-11}) for Multan region via inhalation; in case of Cr highest (4.11×10^{-04}) was observed for children via ingestion in Multan regions and lowest (8.18×10^{-10}) for adults via inhalation in Lahore region; for Zn highest (2.55×10^{-04}) was found for children in Faisalabad region via ingestion and lowest (5.73×10^{-10}) for adults via inhalation in Multan region; similarly for Ni highest (1.07×10^{-05}) was observed for children in Multan region via ingestion and lowest (7.42×10^{-11}) for adults in Lahore region via inhalation and highest Cu (4.22×10^{-05}) was observed for Lahore region via ingestion and lowest (2.30×10^{-10}) for adults in Multan region via inhalation respectively as shown in Table 3. This study revealed that adults and children are mainly exposed through ingestion and inhalation to these selected HMs. Furthermore, this research work also highlighted that children are more exposed to HMs than adults via ingestion which are agreements with previous studies carried out by Zeng et al. (2015a) and Chen et al. (2018).

3.8 Non carcinogenic risk

The non-carcinogenic risks for adults from selected HMs through exposure routes (inhalation, ingestion and dermal) are summarized in Table 4. The HQ values of Pb, Cd, Cr, Zn, Ni and Cu for adults via ingestion in Lahore region were 9.73×10^{-03}, 5.79×10-0, 2.90×10^{-03}, 6.70×10^{-05}, 3.94×10^{-05} and 1.13×10^{-04}, through inhalation the values were 9.11×10^{-07}, 2.27×10^{-04}, 2.86×10^{-05}, 6.31×10^{-07}, 3.60×10^{-09} and 1.06×10^{-08} and through dermal contact the values were 2.65×10^{-03}**,** 2.37×10^{-02}**,** 5.92×10^{-03}**,** 1.37×10-0, 5.97×10^{-06} and 1.54×10^{-05} respectively as shown in Table 4. Similarly the HQ values via ingestion in Multan region for adults were 2.50×10^{-03}, $1.01\times10^{-}$

^{03}, 1.47×10^{-02}, 3.81×10^{-06}, 5.71×10^{-05} and 6.12×10^{-05}, through inhalation the values were 2.34×10^{-07}, 3.95×10^{-05}, 1.45×10^{-04}, 1.91×10^{-07}, 5.22×10^{-09} and 5.73×10^{-09} through dermal the values were 6.82×10^{-04}, 4.11×10^{-03}, 3.00×10^{-02}, 4.15×10^{-06}, 8.65×10^{-06} and 8.34×10^{-06} respectively. Moreover in Faisalabad region the HQ values via ingestion for adults were 3.49×10^{-03}, 6.32×10^{-03}, 9.92×10^{-03}, 9.10×10^{-05}, 4.35×10^{-05} and 6.46×10^{-05}, through inhalation the values were 3.27×10^{-07}, 2.48×10^{-04}, 9.80×10^{-05}, 8.56×10^{-07}, 3.98×10^{-09} and 6.05×10^{-09} and through dermal the values were 9.51×10^{-04}, 2.59×10^{-02}, 2.03×10^{-02}, 1.86×10-0, 6.59×10^{-06} and 8.80×10^{-06} respectively as shown in Table 4. The HI values for adults in Lahore region via ingestion, inhalation and dermal were 1.86×10^{-02}, 2.57×10^{-04} and 3.23×10^{-02}, in Multan region the values were 1.83×10^{-02}, 1.85×10^{-04} and 3.48×10^{-02} and in Faisalabad region the values were 1.99×10^{-02}, 3.47×10^{-04} and 4.71×10^{-02} respectively as presented in Table 4. In the study area the calculated values of HQ for adults were observed less than 1 in all the exposure pathways. This meant that adults were at low risk of non-carcinogenic effects. The HQ values in adults were in order of ingestion>dermal>inhalation in Lahore, Multan and Faisalabad regions. The HI values for adults in Lahore region were in order of dermal>ingestion>inhalation also vice versa for Multan and Faisalabad regions. Furthermore, the results of HQ and HI were observed lower when compared with other studies conducted by Doyi et al. (2018) and Lian et al. (2018) and were similar with the results of Wei et al. (2015) and Khan et al. (2014).

Similarly, the non-carcinogenic risks for children from selected HMs through all exposure routes are presented in Table 5. The HQ values of Pb, Cd, Cr, Zn, Ni and Cu for children through ingestion in Lahore region were 9.09×10^{-02}, 5.41×10^{-02}, 2.71×10^{-02}, 6.26×10^{-04}, 3.68×10^{-04} and 1.05×10^{-03}, through inhalation the values were 2.55×10^{-06}, 6.35×10^{-04}, 8.00×10^{-05}, 1.76×10^{-06}, 1.01×10^{-08} and 2.96×10^{-08} and through dermal contact the values were 1.26×10^{-03}, 1.12×10^{-02}, 2.81×10^{-03}, 6.51×10^{-06}, 2.84×10^{-06} and 7.30×10^{-06} respectively as shown in Table 5. The HQ values through ingestion in Multan region were 2.34×10^{-02}, 9.40×10^{-03}, 1.37×10^{-01}, 1.90×10^{-04}, 5.33×10^{-04} and 5.72×10^{-04}, through inhalation the values were 6.55×10^{-07}, 1.10×10^{-04}, 4.05×10^{-04}, 5.35×10^{-07},

1.46×10^{-08} and 1.60×10^{-08} and through dermal contact the values were 3.24×10^{-04}**,** 1.95×10^{-03}**,** 1.42×10^{-02}**,** 1.97×10^{-06}**,** 4.11×10-0 and 3.96×10^{-06} respectively in Table 5. The HQ values thorough ingestion in Faisalabad region were 3.26×10^{-02}, 5.91×10^{-02}, 9.27×10^{-02}, 8.50×10^{-04}, 4.06×10^{-04} and 6.03×10^{-04}, through inhalation the values were 9.13×10^{-07}, 6.94×10^{-04}, 2.74×10^{-04}, 2.40×10^{-06}, 1.11×10^{-08} and 1.69×10^{-08} and through dermal contact the values were 4.52×10^{-04}**,** 1.23×10^{-02}**,** 9.63×10^{-03}**,** 8.83×10^{-06}**,** 3.13×10^{-06} and 4.18×10^{-06} respectively. The HI values for children in Lahore region through ingestion, inhalation and dermal were 1.74×10^{-01}, 7.20×10^{-04} and 1.53×10^{-02}, in Multan region the values were 1.71×10^{-01}, 5.17×10^{-04} and 1.65×10^{-02} and in Faisalabad region the values were 1.86×10^{-01}, 9.71×10^{-04} and 2.24×10^{-02} respectively as shown in Table 5**.** The HQ values in children were in order of ingestion>dermal>inhalation in Lahore, Multan and Faisalabad regions. The HI for children were in order of ingestion >dermal>inhalation in the study area. The HI values for children were observed higher as compared to adults in Lahore, Multan and Faisalabad regions. However, the HI values less than 1 represent that the exposed population is considered to be safe (Khan et al., 2014). In contrast with other studies carried out by Hu et al. (2017), Lian et al. (2018) and Rehman et al. (2018) the HQ and HI values were lower for children as compared to adults.

3.9 Carcinogenic risk

The results of carcinogenic risk (RI) values in agricultural soil of Lahore, Multan and Faisalabad for adults and children are summarized in Table 6. Due to low carcinogenic risk for Cu, Zn and Ni the RI values were only estimated for Pb, Cd and Cr for both adults and children. The RI values of Pb for adults in Lahore via ingestion, inhalation and dermal were 2.89×10^{-07}, 2.72×10^{-11} and 1.18×10^{-08}, for Cd the values were 2.09×10^{-05}, 1.96×10^{-09} and 8.53×10^{-07} and for Cr the values were 4.35×10^{-06}, 4.09×10^{-10} and 1.78×10^{-07} respectively. The RI values of Pb in Multan were 7.44×10^{-08}, 7.01×10^{-12} and 3.04×10^{-09}, for Cd the values were 3.62×10^{-06}, 3.41×10^{-10} and 1.48×10^{-07} and for Cr the values were 2.20×10^{-05}, 2.07×10^{-09} and 9.00×10^{-07} respectively. Furthermore the RI values of Pb in Faisalabad were 9.77×10^{-12}, 9.77×10^{-12} and 4.24×10^{-09} , for Cd the values were 2.28×10^{-05}, 2.14×10-0 and 9.31×10^{-07} and for Cr the values were 1.49×10^{-05}, 1.40×10^{-09} and 6.09×10^{-07} respectively. The total risk (TRI) values in Lahore via ingestion, inhalation and dermal were 2.55×10^{-05}, 2.40×10^{-09} and 1.04×10^{-06} in Multan were 2.57×10^{-05}, 2.42×10^{-09} and 1.05×10^{-06} and in Faisalabad were 3.77×10^{-05}, 3.55×10^{-09} and 1.54×10^{-06} respectively as shown in Table 6. The combined risk (CRI) values in Lahore for Pb, Cd and Cr were 3.41×10^{-05}, 6.65×10^{-06} and 8.87×10^{-06} in Multan were 8.76×10^{-06}, 1.15×10^{-06} and 4.49×10^{-05} and in Faisalabad were 1.22×10^{-05}, 7.26×10^{-06} and 3.04×10^{-05} respectively.

Similarly the RI values of Pb for children in Lahore via ingestion, inhalation and dermal were 2.70×10^{-06}, 7.62×10^{-11} and 5.62×10^{-09}, for Cd the values were 1.95×10^{-04}, 5.49×10^{-09} and 8.53×10^{-07} for Cr the values were 4.06×10^{-05}, 1.14×10^{-09} and 8.43×10^{-08} respectively. Further the RI values of Pb in Multan were 6.95×10^{-07}, 1.96×10^{-11} and 1.45×10^{-09}, for Cd the values were 3.38×10^{-05}, 9.54×10^{-10} and 7.03×10^{-08} and for Cr the values were 2.05×10^{-04}, 5.79×10^{-09} and 4.27×10^{-07} respectively. The RI values of Pb in Faisalabad were 9.69×10^{-07}, 2.73×10^{-11} and 2.02×10^{-09}, for Cd the values were 2.13×10^{-04}, 5.99×10^{-09} and 4.42×10^{-07} and for Cr the values were 1.39×10^{-04}, 3.92×10^{-09} and 2.89×10^{-07} respectively as shown in Table 6. The TRI values in Lahore via ingestion, inhalation and dermal were 2.38×10^{-04}, 6.71×10^{-09} and 9.43×10^{-07} in Multan were 2.40×10^{-04}, 6.77×10^{-09} and 4.99×10^{-07} and in Faisalabad were 3.53×10^{-04}, 9.94×10^{-09} and 7.33×10^{-07} respectively. The CRI values in Lahore for Pb, Cd and Cr were 3.18×10^{-04}, 5.45×10^{-05} and 8.12×10^{-05}, in Multan were 8.18×10^{-05}, 9.47×10^{-06} and 4.11×10^{-04} and in Faisalabad were 1.14×10^{-04}, 5.95×10^{-05} and 2.78×10^{-04} respectively as presented in (Table 6). Although according to Wu et al. (2015), if the RI values exceeded than 1.0×10^{-04} then it might cause carcinogenic risk to human health and if the RI values ranged from 1.0×10^{-04} to 1.0×10^{-06} indicates tolerable or acceptable risk to human health. The RI values higher than 1.0×10^{-04} are likely to develop high risk of cancer in humans (Qing et al., 2015). The RI values of Cd exceeded for both children and adults in Lahore, Multan and Faisalabad via ingestion. High exposure of Cd may cause, skeletal damage, kidney damage, fractures and other severe disease (Jarup, 2003). In comparison with other studies the RI values in this study were similar to Rehman et al. (2018) and lower than Hu et al. (2017) and Lian et al. (2018).

Conclusion

Farmlands degradation due to intensive agricultural practices is a serious problem in developing country like Pakistan. The HMs including Pb, Cd, Cr, Zn, Ni and Cu concentrations were investigated in Lahore, Multan and Faisalabad regions. All the HMs were within the permissible limits except Cd in Lahore and Faisalabad regions while the Cd concentration in Multan region were only higher than USEPA (2002). The high concentration of Cd might be due to industrial activities and intensive agricultural practices. In the study area the pollution indices such as CF, CD, I_{geo}, EF and PERI values for Cd showed some level of contamination in Lahore and Faisalabad region as compared to Multan region. The HQ values in adults and children were in order of ingestion>dermal>inhalation Lahore, Multan and Faisalabad regions. Although the calculated values of HQ for adults and children were < 1 in all the exposure pathways. The HI values in Lahore region were in order of dermal>ingestion>inhalation also similar in Multan and Faisalabad regions while, other metals were within safe limits. Due to low carcinogenic risk for Cu, Zn and Ni the RI values were only estimated for Pb, Cd and Cr for both adults and children. The RI values of Cd exceeded for both children and adults in Lahore, Multan and Faisalabad regions via ingestion. The RI values higher than 1.0×10^{-04} might develop cancer risk in humans. The finding of this study suggests that farmlands soil contaminated with HMs need more attention and remediation strategies including (pollution source identification, phyto-availability of HMs, fertilizer management, variation in cropping system and phytoremediation) should be practiced in the selected regions to suppress metals accumulation in food crops and to achieve greater effectiveness for public health safety in the near future.

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Conflict of interest

The authors declared no conflict of interest

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