Assessment of health and ecological risks of heavy metal contamination: a case study of agricultural soils in Thall, Dir-Kohistan

Irshad Ullah · Allah Ditta ⬤ · Muhammad Imtiaz ·
Sajid Mehmood · Muhammad Rizwan · Muhammad
Shahid Rizwan · Amin Ullah Jan · Iftikhar Ahmad ⬤

Abstract Heavy metal (HM) contamination in agricultural soils has been a significant health concern worldwide due to their persistent and non-biodegradable nature and biomagnification to higher trophic levels. The present study was conducted to assess Cd and Pb concentrations in soil samples collected from potato-growing areas in Thall, Dir-Kohistan, and to determine their associated health and ecological risks. The contamination factor calculated for Cd (0 to 1.74) and Pb (0 to 0.91) showed their moderate to low contamination in the study area. Geo-accumulation indices of Cd and Pb were less than 1, indicating moderate soil pollution of these HM in the study area. However, the ecological risk factor (Eri) of Cd was greater than 40 for only three soil

I. Ullah · A. Ditta (✉)
Department of Environmental Sciences, Shaheed Benazir Bhutto University, Sheringal, Dir (Upper), Khyber Pakhtunkhwa 18000, Pakistan
e-mail: ad_abs@yahoo.com

e-mail: allah.ditta@sbbu.edu.pk

A. Ditta
School of Biological Sciences, The University of Western Australia, Perth, WA 6009, Australia

M. Imtiaz
Soil and Environmental Biotechnology Division, National Institute for Biotechnology and Genetic Engineering, Faisalabad, Pakistan

S. Mehmood
Guangdong Provincial Key Laboratory for Radionuclides Pollution Control and Resources, School of Environmental Science and Engineering, Guangzhou University, Guangzhou 510006, People's Republic of China

S. Mehmood
School of Civil Engineering, Guangzhou University, Guangzhou 510006, People's Republic of China

S. Mehmood · M. Rizwan
College of Resources and Environment, Huazhong Agricultural University, Wuhan 430070 Hubei, People's Republic of China

M. S. Rizwan
Cholistan Institute of Desert Studies, The Islamia University of Bahawalpur, Bahawalpur 63100, Pakistan

A. U. Jan
Department of Biotechnology, Shaheed Benazir Bhutto University Sheringal, Dir (U) Khyber Pakhtunkhwa 18000, Pakistan

I. Ahmad
Department of Environmental Sciences, COMSATS University Islamabad, Vehari-Campus, Vehari 61100, Pakistan

I. Ahmad
Key Laboratory of Urban Agriculture by Ministry of Agriculture of China, School of Agriculture and Biology, Shanghai Jiao Tong University, Shanghai, People's Republic of China

samples, indicating the moderate potential of ecological risks of respective soils. The principal component analysis (PCA) and Pearson correlation suggested that the contamination in different soils was lithogenic followed by anthropogenic activities. The hazard quotient (HQ) in children and adults was found in the following order: dermal > ingestion > inhalation. Moreover, the values of HQ through various exposure routes were higher in children compared to adults, which showed that adults were at a lower level of risk associated with HM contamination. The results of the present study can serve as baseline data for government agencies related to environmental protection, which could devise policies to minimize Cd and Pb contamination in the agricultural soils.

Abbreviations
HRA   Health risk assessment
Cd   Cadmium
Pb   Lead
OOC   Oxidizable organic carbon
OM   Organic matter
TOC   Total organic carbon
EC   Electrical conductivity
CF   Contamination factors
PLI   Pollution load index
Igeo   Geo-accumulation index
Eri   Ecological risk factors
RI   Potential ecological risk index
ADD   Average daily dose
HQ   Hazard quotients
THI   Total hazard index

Introduction

Heavy metals have become the major factors responsible for soil contamination, especially in agricultural soils around the world (Cao et al. 2018; Mehmood et al. 2019), where these heavy metals are accumulating through various agronomic practices such as the application of chemical phosphatic fertilizers, manures, pesticides, fungicides, and wastewater (Khan et al. 2013; Ahmad et al. 2018; Cao et al. 2018; Elbehiry et al. 2019; Guo et al. 2019; Parente et al. 2019; Ahmad et al. 2020;

Githaiga et al. 2020; Leblebici et al. 2020). Heavy metals present in the soil when transformed into either ionic or organometallic form and that enter into the food chain via the consumption of plant biomass produced in that contaminated soils may cause severe deleterious effects on human health (Liang et al. 2015). Keeping in view the above concerns, international organizations such as the World Health Organization (WHO), United States Environmental Protection Agency (USEPA), and National Environmental Quality Standards of Pakistan (NEQS-Pak), have set the maximum allowable limits of heavy metals in agricultural soils to protect environmental quality and human health (US-EPA 2010).

With industrialization and urbanization over the past few decades, heavy metal pollution has become a severe threat in many countries including Pakistan (Muhammad et al. 2019). There are many reports in several parts of the country about the health issues arisen from heavy metal contamination (Karim and Qureshi 2013; Khan et al. 2013; Gul et al. 2015; Mehmood et al. 2019; Muhammad et al. 2019). The studies have also found moderate to severe contamination of heavy metals such as nickel (Ni), copper (Cu), zinc (Zn), lead (Pb), chromium (Cr), arsenic (As), and cadmium (Cd) in soil, water, and food items grown either on heavy metal-contaminated soils or soils irrigated with contaminated water (Wang et al. 2012; Cao et al. 2018; Ahmad et al. 2020).

Assessment of ecological risks associated with these contaminations and their subsequent effect on human health are the main pillars in highlighting intolerable risks to humans and the environment. Also, the driving factors for these ecological risks need to be identified. Various researchers around the world have reported three main exposure routes, i.e., ingestion, dermal, and inhalation of heavy metals from contaminated soils (Khan et al. 2013; Muhammad et al. 2019; Dzul-Caamal et al. 2020). Moreover, the associated ecological risks mainly depend on the nature and concentration of heavy metals under study (Cao et al. 2018; Mehmood et al. 2019; Askari et al. 2020).

Heavy metals react with sulfhydryl enzyme and inhibit cellular production in biological systems through the displacement of essential metals on active sites (Khan et al. 2013; Jan et al. 2015; Mehmood et al. 2019). Physiological and neurological disorders have been reported due to mild Pb toxicity in humans, while severe toxicity may cause the problem in the liver and kidney leading to cancer (Odum 2000; Manahan 2001).

Induction of nausea, headache, bone fractures, and kidney failure have been reported due to Cd toxicity (Fowler 2009; Fu et al. 2011).

Many researchers around the world have found anthropogenic activities as the main responsible factors behind heavy metal pollution (Khan et al. 2013; Cao et al. 2018; Mehmood et al. 2019). Recently, agricultural soils contaminated with heavy metals have received more attention due to more emphasis on soil quality and sustainability. For example, the Chinese government launched 8-year soil pollution survey program and found large-scale soil pollution in the study areas (Teng et al. 2010). Also, Liang et al. (2015) found that the cultivated soils in Zhangjiakou City of Hebei Province, China, have heavily been contaminated with Cd, Hg, and Cu, which require periodic monitoring.

Earlier, assessment of groundwater quality regarding heavy metal contamination has been investigated in many research studies conducted in different parts of Pakistan like Muzaffargarh, Lahore and Kasur in Punjab, Jamshoro and Manchar Lake in Sindh, Peshawar, Kohistan region, Mohmand Agency, and Mardan in Khyber Pakhtunkhwa (Nickson et al. 2005; Gul et al. 2015; Muhammad et al. 2019). From these studies, it has been clear that groundwater quality in different parts of Pakistan is poor due to the presence of heavy metals. Groundwater is usually being used as a sole source of irrigation in most areas of Pakistan including Khyber Pakhtunkhwa, so it can build up heavy metals in agricultural soils.

Anthropogenic activities like the use of chemical phosphatic fertilizers, pesticides, fungicides, manures, and wastewater irrigation are frequently observed in developing countries like Pakistan, which can cause accumulation of heavy metals especially Cd and Pb in the agricultural soils and pose serious health threats to the local community. Besides, there is no regulation on the permissible limits for agricultural soils contaminated with heavy metals. These heavy metals (Cd, Pb) remain persistent in the soils for longer periods because these are non-biodegradable and not easily removed from the soil through normal cropping practices and have strong affinities with soil particles. Therefore, soil constitutes a significant reservoir of Cd and Pb. Recent findings showed that Cd contamination in soil varied from 0.02–184 mg kg^{-1}; however, it depends upon soil type and area (Waseem et al. 2014; Latif et al. 2020). A better understanding of agricultural soils contaminated with heavy metal and subsequent management practices would ultimately lead us to minimize heavy metal contamination and towards better soil quality. Based on this hypothesis, the present study was conducted to assess the heavy metal (Cd and Pb) concentrations in the agricultural soils of the Thall area, located in Dir-Kohistan. The objectives of the present study were (1) to determine the concentrations of Cd and Pb in soils under potato cultivation areas of Thall, Dir-Kohistan, and (2) to determine their associated environmental and human health risks.

Materials and methods

Study area

The present study was conducted in the surrounding areas of Thall (35° 28′ 42″ North, 72° 14′ 33″ East), located in Dir-Kohistan, District Dir Upper, Khyber Pakhtunkhwa, Pakistan. Union council Thall is situated in Tehsil Kalkot, District Dir Upper, and is about 50 km away from Sheringal Khass. It consists of four village councils and thirty-eight sub-villages. In the North of Thall, one of the most beautiful valleys of Pakistan, i.e., Kumrat valley is located; in South, a beautiful town, i.e., Patrak is located; in North-West, another one of the most beautiful valleys of Pakistan, i.e., Gwaldi valley is located; and in South-East, Badghoi valley is located. The whole union council is lush green with some plain area till the end of the Kumrat boundary. The total distance Thall Union Council from Dir Khass is about 75 km by road. The valley is mostly mountainous and a few patches of plain land are available where agriculture is practiced. The height of mountains, present in the study area, ranges from 2000 to 2200 m mean sea level. The peaks are mostly covered with snow throughout the year. Most of the downhill basin is covered with the round shape stones brought with the flood from the upper areas of the Panjkora river. Most of the rocks are metamorphic and igneous in nature. The thick forest in the valley covers the mountains. The pictorial view of the study area is presented in Fig. 1.

The climate of the study area remains pleasant throughout the year. In summer, the temperature is mild with an average temperature ranging from 25 to 30 °C. The summer season is quite short and June is the hottest month of the year. In winter (November, December, January, and February), the temperature is very low due to frequent and heavy snowfall up to 2–4 ft on the
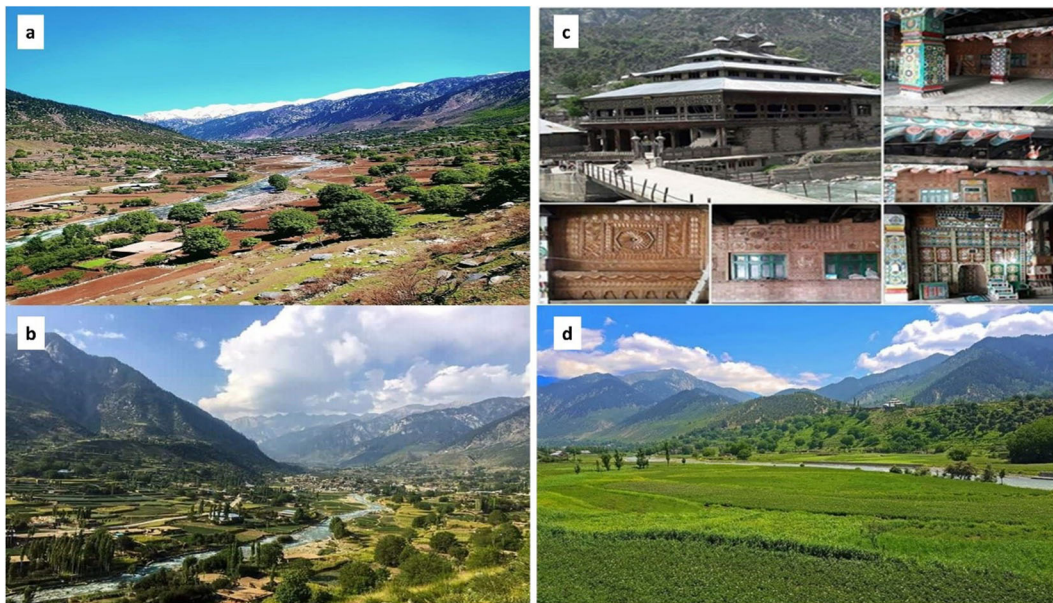
Fig. 1 Pictorial view and geographical location of (**a**) the whole study area before potato cultivation, (**b**) after potato cultivation (35° 28′ 42″ North, 72° 14′ 33″ East), (**c**) Thall mosque in Thall Khas (35° 28′ 42″ North, 72° 14′ 33″ East), and (**d**) potato cultivation area in Haji Shai Payeen Thall (35° 27′ 46.9″ North, 72° 13′ 58.9″ East)

mountains with an average temperature ranging from − 4 to 10 °C. The valleys remain under snowfall for about 4 months. The spring season remains for 2 months, i.e., March and April. Annual rainfall in the study area ranges from 100 to 255 mm.

The soil in the study area is very fertile but due to long severe winter, only one crop, i.e., maize or potato, can be cultivated. Various types of fruits such as apples, walnuts, many wild berries, and almonds are naturally present and offer a delicious taste. The soil is loamy and slightly gravel in texture.

Soil sampling

The present study involved the collection of soil samples from potato-harvested fields in the study area during August–September, 2017. During the sampling period, the maximum and minimum temperature was 30.8 and 14.1 °C, respectively, while the mean monthly rainfall was 75 mm (Pakistan Meteorological Department 2017). Soil samples ($n = 52$) were collected with the help of an auger from potato-growing agricultural fields located in the surrounding villages of Thall, Dir-Kohistan. The sample distribution among different villages was Bijligar Thall ($n = 4$), Kalan Bala Thall ($n = 11$), Kalan Payeen Thall ($n = 7$), Thall Khass ($n = 4$), Darwazo Thall ($n = 4$), Haji Shai Payeen Thall ($n = 8$),

and Haji Shai Bala Thall ($n = 14$). Non-uniform sampling was done due to uneven, sloppy areas, and only the fields cultivated with potato were selected for soil sampling. In some villages, the area under potato cultivation was small with few samples collected while a greater number of samples were collected from large areas with potato cultivation.

Composite homogeneous soil samples in triplicate were taken randomly from each site from a depth of 0–15 cm (topsoil) which was selected due to the predominant effect of agricultural practices such as chemical fertilizer, irrigation water, and pesticides in this layer. All the samples were put in airtight polythene bags, properly labeled, taken back to the laboratory, and stored at 4 °C under dark conditions until the analysis was done. The location of sampling sites was identified by using a global positioning system (GPS, Garmin Etrex-30x), and each point was drawn in a map using ArcGIS (version 10.5). The map regarding the sampling points in the study area is given in Fig. 2.

Heavy metals analysis

Each collected soil sample in triplicate from the study area was air-dried, ground, and passed through a 2-mm sieve. An acid mixture of $HNO_3:HClO_4:HF$ in the ratio 3:1:3 was used for digestion of 1 g of each sample taken
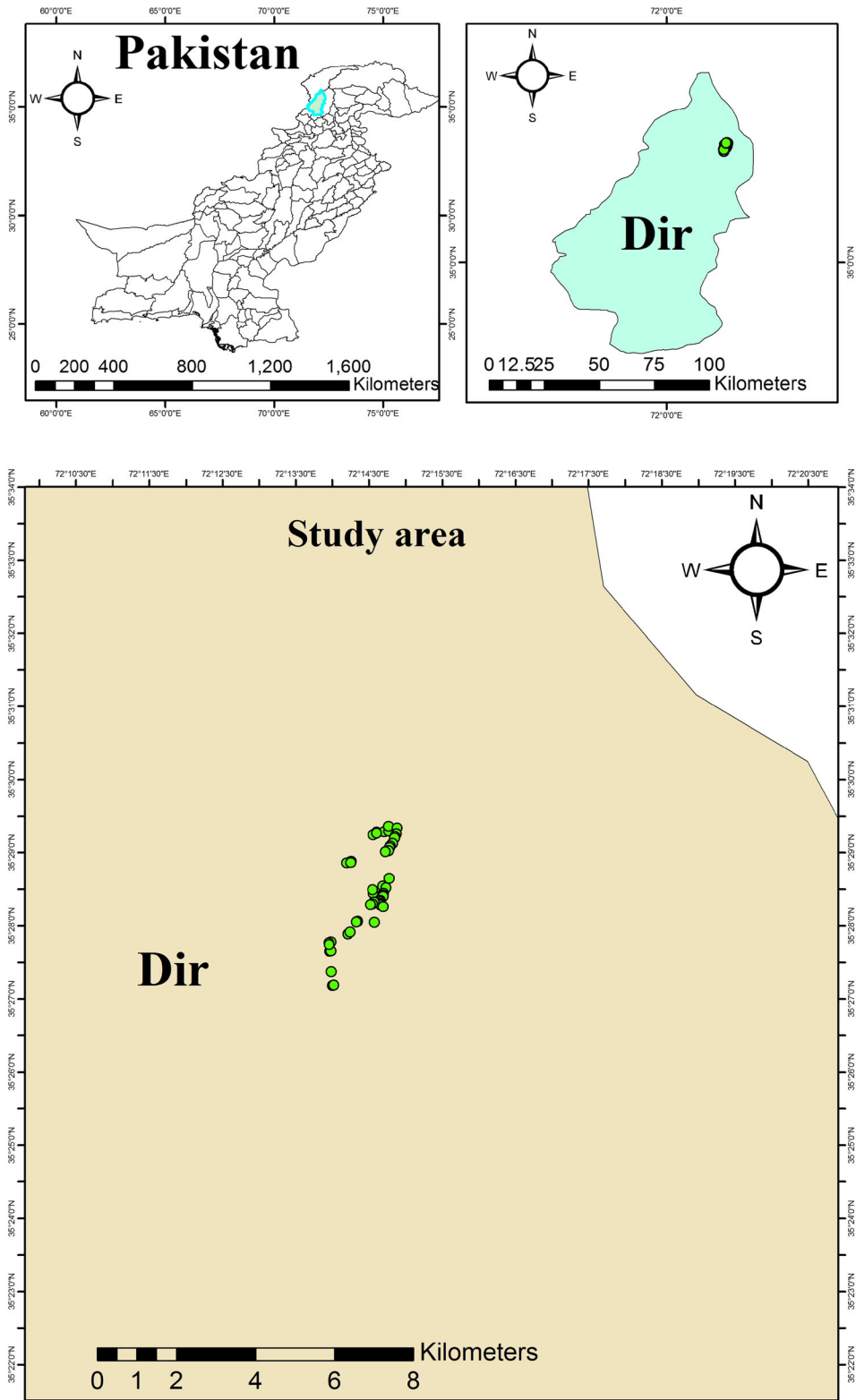
Fig. 2 Map of study area showing sampling points

in a 100-mL conical flask by placing it on a hot plate and heating until a clear point reached (Nwajei and Gagophein 2000). The clear digest after acid digestion was filtered through Whatman 40 filter paper and diluted to 50 mL using deionized water. Atomic absorption spectrophotometer (VARIAN 240FS, Sweden) with Cd and Pb cathode lamps was used for the determination of Cd and Pb in the soil samples. A blank with all the chemicals without sample was also run to remove any heavy metal concentration in the chemicals.

Physicochemical characteristics analysis

Immediately after the transportation of samples into the laboratory, the soil suspension (1:2.5 w/v dilutions) was prepared to determine pH and electrical conductivity (EC) using pH (inoLab® pH 7110) and conductivity (inoLab® Cond 7110) meters, respectively. Potassium dichromate ($K_2Cr_2O_7$) titration method was employed to estimate oxidizable organic carbon (OOC) (Walkley 1947) while total organic carbon (TOC) and organic matter (OM) contents in collected soil samples were calculated from the values of OOC.

Pollution indices

In agricultural soils, various pollution indices such as contamination factor, pollution load index, geo-accumulation index, and enrichment factor are very helpful parameters during the processing and analysis of data related to environmental pollution (Caeiro et al. 2005). According to Lacatusu (2000), the interpretation of standards of different pollution indices varies from country to country and is mainly dependent on the values of chosen factors. The details regarding each index are given below:

Contamination factor

It is used to monitor the extent of heavy metal contamination in soil and is the ratio between the concentration of a particular heavy metal to its background value (Hakanson 1980). The background values (B_n) used during the calculation of contamination factor (CF) for Cd and Pb were 0.3 and 20 mg kg^{-1}, respectively (Martin and Meybeck 1979; Hakanson 1980).

$$CF = \frac{C \text{ (Heavy metal)}}{B_n}$$

where C is the concentration of heavy metal (mg kg^{-1}) and B_n is the background value of respective metal (mg kg^{-1}).

Pollution load index

For the quantification of a pollutant such as heavy metals in agricultural soils, pollution load index (PLI) is used and calculated according to the equation given by Tomlinson et al. (1980).

$$PLI = (CF_1 \times CF_2 \times CF_3 \cdots \times CF_n)^{1/n}$$

where n is the number of heavy metals analyzed from the collected soil samples and CF is the contamination factor. The PLI values give an estimate of the metal contamination status (Tomlinson et al. 1980).

Geo-accumulation index

This index is mainly concerned with the contamination in soils and sediment under study and helps in the determination of heavy metal contamination in soil with respect to natural background level as a reference (Muller 1969).

$$Igeo = \log_2 \left[\frac{C_n}{1.5 \times B_n} \right]$$

where Cn represents the measured concentration of the metal (n) in the sample, and Bn is used to explain the geochemical background concentration of the metal. To remove the influence of lithological variations in the soil, factor 1.5 is applied (Stoffers et al. 1986). Base on the values of Igeo, seven classes have been proposed by Muller (1969).

Ecological risk factor

The potential ecological risk of a given single contaminant, ecological risk factor (Eri), was calculated by the following equation suggested by Hakanson (1980):

$$Er_i = T_r^i . C_f^i$$

where T_r^i = toxic response factor of a given substance (lead = 5, cadmium = 30); C_f^i is the contamination factor. According to Hakanson (1980), five categories of ecological risks have been proposed based on the values of Eri.

Potential ecological risk index

This index determines the degree of contamination and is the sum of the ecological risk factors (Eri) as clear from the equation given below:

$$RI = \sum_{i=1}^{m} Eri$$

where Eri is the ecological risk factor and m is the count of the heavy metal species. According to Hakanson (1980), four categories have been proposed based on the values of risk index (RI).

Human health risk assessment

Average daily dose

In agricultural soils, heavy metal exposure to humans can occur through ingestion, inhalation, and dermal pathways. Average daily dose (ADD) (mg kg^{-1} day^{-1}) for heavy metals (Cd and Pb) through each exposure pathway to the soil in children (06 years) and adults (25 years) separately was calculated using the following equations proposed by the United States Environmental Protection Agency (USEPA 1989, 1996):

Average Daily Dose Ingestion (ADD_{ing})

$$= \frac{C \times EF \times ED \times R_{ing} \times 10^{-6}}{BW \times AT}$$

Average Daily Dose Inhalation (ADD_{inh})

$$= \frac{C \times EF \times ED \times R_{inh} \times 10^{-6}}{PEF \times BW \times AT}$$

Average Daily Dose Inhalation (ADD_{derm})

$$= \frac{C \times SL \times SA \times ABS \times EF \times ED \times 10^{-6}}{BW \times AT}$$

where ADD_{ing}, ADD_{inh}, and ADD_{derm} are the average daily dose for ingestion, inhalation, and dermal exposure routes, respectively, C = concentration of heavy metals (mg kg^{-1}), EF = exposure frequency (days year^{-1}), ED = exposure duration (year), R_{ing} = ingestion rate (mg day^{-1}), BW = body weight (kg), AT = average time (days), R_{inh} = inhalation rate (m^3 day^{-1}),

PEF = particular emission factor (m^3 kg^{-1}), SL = skin adherence factor (mg cm^{-2}), SA = exposed skin area (cm^2), and ABS = dermal absorption factor. A detailed explanation of each parameter is presented in Table 1.

Hazard quotient

According to USEPA (2010), HQ was estimated according to the following equation:

$$HQ = \frac{ADD}{RfD}$$

where ADD = average daily dose (mg kg^{-1} day^{-1}) for the different exposure routes and RfD = reference dose (mg kg^{-1} day^{-1}).

Total hazard index

Total hazard index (THI) is the overall potential risks posed by a mixture of heavy metals determined in the soil samples and expressed as the sum of the HQ for each heavy metal (USEPA 1989):

$$THI = \sum HQ$$

Accordingly, THI value ≤ 1.0 shows that significant additive or toxic interactions would not occur and there is no need for further evaluation. However, THI > 1.0 poses a serious concern regarding potential health issues (USEPA 1989).

Statistical analysis

The data obtained from this study were analyzed for descriptive statistics, principal component analysis (PCA), and Pearson correlation using SPSS version 25. ArcGIS version 10.5 was used to make a study area map showing the distribution of collected soil samples. PCA measures the relationship among different metals and physicochemical characteristics of soil samples. PCA was also utilized to identify sources, apportioning them to natural vs. anthropogenic sources. The Pearson correlation coefficient measures the strength of relationships between pairs of different metals and physicochemical characteristics of soil samples. Correlations and *P* values (two-tailed tests) were calculated utilizing Pearson's correlation coefficients using SPSS version 25.

Table 1 Symbols and values of different parameters used to estimate the health risks associated with heavy metal contamination in agricultural soils located in Thall, Dir-Kohistan

Symbol	Parameters	Units	Values	References
C	Concentration of heavy metals	mg kg^{-1}		
BW	Body weight	kg	70a, 15b	USEPA 1989
RfD	Chronic reference dose	mg kg^{-1} day^{-1}	Oral RfD: 0.001 (Cd), 0.0035 (Pb) Dermal RfD: 1.0E-05 (Cd), 5.25E-05 (Pb) Inhalation RfD: 1.00E-05 (Cd), 3.5E-03 (Pb)	Ferreira-Baptista and De Miguel 2005; Askarova and Mussagaliyeva 2014
ABS	Dermal absorption factor	Unitless	0.14 (Cd), 0.006 (Pb)	HC, 2004; Chen et al. 2015; Kelepertzis 2014
SA	Exposed skin area	cm^2	3300a, 2800b	USEPA 2001
ED	Exposure duration	year	25a, 6b	USEPA 2001
EF	Exposure frequency	days year^{-1}	250	Ferreira-Baptista and De Miguel 2005
Ring	Ingestion rate	mg d^{-1}	100a, 200b	
Rinh	Inhalation rate	m^3 day^{-1}	20a, 10b	Van den Berg 1995
PEF	Particular emission factor	m^3 kg^{-1}	1.32E+09	USEPA 2001
SL	Skin adherence factor	mg cm^{-2}	0.2a, 0.2b	USEPA 2001
Bn	Background concentration of metal	mg kg^{-1}	Pb = 20 and Cd = 0.3	
AT	Average time	days	ED*365	Ihedioha et al. 2017
Tr	Toxic response factor	Unitless	Cd = 30 and Pb = 5	Ihedioha et al. 2017

[a] Adults

[b] Children

Results and discussion

The present study was conducted to assess heavy metal contamination and their associated health risks in different soil samples collected from the surrounding areas of Thall-Kohistan, Khyber Pakhtunkhwa, Pakistan. The results obtained have been discussed in the following sections:

Heavy metal concentration and physicochemical properties of soil

The data regarding concentrations of Cd and Pb and physicochemical properties of soil, viz. pH, EC, and OM, oxidizable organic carbon (OOC), and total organic carbon (TOC) are presented in Table 2. The metal

Table 2 Heavy metal concentration, organic matter, pH, and electrical conductivity of the soils in the study area

	Pb (mg kg^{-1})	Cd (mg kg^{-1})	OOC (%)	TOC (%)	OM (%)	pH	EC (dS m^{-1})
Mean (n = 52)	3.850	0.182	3.029	4.040	6.966	4.870	0.430
Std. error of mean	0.80	0.02	0.14	0.18	0.31	0.15	0.05
Median	0.520	0.186	3.100	4.140	7.140	4.990	0.325
Std. deviation	5.710	0.150	0.973	1.298	2.238	1.048	0.388
Variance	32.603	0.023	0.946	1.686	5.007	1.098	0.150
Minimum	0.00	0.00	0.21	0.28	0.48	2.12	0.06
Maximum	18.00	0.52	4.92	6.57	11.32	6.77	1.92
Sum	196.00	9.29	154.49	206.05	355.28	248.36	21.91

Pb, lead; *Cd*, cadmium; *OOC*, oxidizable organic carbon; *TOC*, total organic carbon; *OM*, organic matter; *pH*, acidity/alkalinity; *EC*, electrical conductivity

Table 3 Contamination factors, pollution load index, geo-accumulation index (Igeo), and potential ecological risk index (RI) of different heavy metal-contaminated soil samples

Sr. No.	Village name	Contamination factors		Pollution load index	Geo-accumulation index (Igeo)		Potential ecological risk index (RI)
		Cd	Pb	PLI	Cd	Pb	
1	Bijlighar Thall	1.50 ± 0.006*	0.22 ± 0.001	0.58 ± 0.001	0.00 ± 0.006	-2.75 ± 0.006	46.22
2		1.74 ± 0.002	0.23 ± 0.004	0.63 ± 0.006	0.22 ± 0.002	-2.70 ± 0.025	53.46
3		1.61 ± 0.004	0.26 ± 0.001	0.65 ± 0.002	0.11 ± 0.004	-2.52 ± 0.007	49.71
4		1.14 ± 0.001	0.23 ± 0.001	0.52 ± 0.001	-0.39 ± 0.001	-2.69 ± 0.007	35.46
5	Kalan Bala Thall	1.10 ± 0.009	0.29 ± 0.004	0.57 ± 0.006	-0.45 ± 0.012	-2.35 ± 0.021	34.47
6		1.03 ± 0.026	0.29 ± 0.003	0.55 ± 0.007	-0.54 ± 0.038	-2.38 ± 0.016	32.34
7		1.08 ± 0.014	0.34 ± 0.003	0.61 ± 0.002	-0.48 ± 0.019	-2.13 ± 0.011	34.02
8		1.08 ± 0.012	0.31 ± 0.003	0.58 ± 0.001	-0.48 ± 0.015	-2.27 ± 0.016	33.86
9		0.56 ± 0.006	0.21 ± 0.001	0.34 ± 0.003	-1.43 ± 0.016	-2.85 ± 0.010	17.74
10		0.57 ± 0.003	0.24 ± 0.001	0.37 ± 0.002	-1.39 ± 0.007	-2.62 ± 0.007	18.42
11		0.62 ± 0.003	0.22 ± 0.000	0.37 ± 0.001	-1.27 ± 0.007	-2.77 ± 0.001	19.70
12		0.61 ± 0.003	0.26 ± 0.003	0.40 ± 0.003	-1.30 ± 0.006	-2.50 ± 0.017	19.62
13		0.72 ± 0.003	0.59 ± 0.003	0.65 ± 0.002	-1.07 ± 0.006	-1.35 ± 0.008	24.44
14		0.74 ± 0.003	0.78 ± 0.002	0.76 ± 0.002	-1.01 ± 0.006	-0.94 ± 0.004	26.21
15		0.77 ± 0.006	0.73 ± 0.003	0.75 ± 0.002	-0.96 ± 0.011	-1.04 ± 0.006	26.86
16	Kalan Payeen Thall	0.71 ± 0.001	0.78 ± 0.006	0.74 ± 0.003	-1.09 ± 0.002	-0.94 ± 0.011	25.10
17		0.55 ± 0.003	0.00	0.00	-1.46 ± 0.008	0.00	16.40
18		0.58 ± 0.003	0.00	0.00	-1.37 ± 0.007	0.00	17.40
19		0.51 ± 0.003	0.00	0.00	-1.57 ± 0.007	0.00	15.20
20		0.55 ± 0.006	0.00	0.00	-1.46 ± 0.015	0.00	16.40
21		0.60 ± 0.003	0.76 ± 0.002	0.68 ± 0.002	-1.33 ± 0.007	-0.97 ± 0.004	21.72
22		1.05 ± 0.006	0.88 ± 0.003	0.96 ± 0.001	-0.51 ± 0.008	-0.76 ± 0.005	36.02
23	Thall Khas	1.06 ± 0.009	0.91 ± 0.004	0.98 ± 0.006	-0.50 ± 0.012	-0.73 ± 0.006	36.44
24		0.96 ± 0.002	0.87 ± 0.002	0.92 ± 0.002	-0.64 ± 0.003	-0.78 ± 0.004	33.26
25		0.98 ± 0.003	0.00	0.00	-0.61 ± 0.005	0.00	29.40
26		0.82 ± 0.002	0.01 ± 0.000	0.07 ± 0.000	-0.87 ± 0.003	-7.85 ± 0.007	24.63
27	Darwazo Thall	0.85 ± 0.002	0.00	0.05 ± 0.001	$-0.82 \pm .003$	-9.23 ± 0.061	25.51
28		0.72 ± 0.001	0.03 ± 0.000	0.14 ± 0.003	-1.06 ± 0.002	-5.85 ± 0.064	21.73
29		0.81 ± 0.003	0.00	0.00	-0.89 ± 0.005	0.00	24.30
30		1.03 ± 0.019	0.00	0.00	-0.54 ± 0.026	0.00	31.00
31	Haji Shai Payeen Thall	1.10 ± 0.005	0.01 ± 0.001	0.11 ± 0.003	-0.45 ± 0.006	-7.09 ± 0.076	32.96
32		1.06 ± 0.002	0.00	0.00	-0.50 ± 0.002	0.00	31.80
33		1.08 ± 0.001	0.00	PLI	-0.47 ± 0.002	0.00	32.50
34		1.06 ± 0.002	0.01 ± 0.001	0.11 ± 0.002	-0.50 ± 0.002	-7.09 ± 0.042	31.96
35		0.00	0.03 ± 0.000	0.00	0.00	-5.85 ± 0.070	0.13
36		0.00	0.00	0.00	0.00	0.00	0.00
37		0.00	0.00	0.00	0.00	-8.38 ± 0.052	0.02
38		0.00	0.04 ± 0.001	0.00	0.00	-5.42 ± 0.080	0.18
39	Haji Shai Bala Thall	0.00	0.00	0.00	0.00	0.00	0.00
40		0.00	0.00	0.00	0.00	0.00	0.00
41		0.00	0.00	0.00	0.00	0.00	0.00

Table 3 (continued)

| Sr. No.	Village name	Contamination factors		Pollution load index	Geo-accumulation index (Igeo)		Potential ecological risk index (RI)
		Cd	Pb	PLI	Cd	Pb	
42		0.00	0.00	0.00	0.00	0.00	0.00
43		0.00	0.00	0.00	0.00	0.00	0.00
44		0.00	0.00	0.00	0.00	0.00	0.00
45		0.00	0.00	0.00	0.00	0.00	0.00
46		0.00	0.00	0.00	0.00	0.00	0.00
47		0.00	0.00	0.00	0.00	0.00	0.00
48		0.00	0.00	0.00	0.00	0.00	0.00
49		0.00	0.17 ± 0.002	0.00	0.00	-3.16 ± 0.019	0.84
50		0.00	0.17 ± 0.001	0.00	0.00	-3.11 ± 0.009	0.87
51		0.00	0.14 ± 0.003	0.00	0.00	-3.42 ± 0.036	0.70
52		0.00	0.10 ± 0.001	0.00	0.00	-3.95 ± 0.008	0.49

[*] The values following mean values represent standard error where $n = 3$

concentrations were found in the order of Pb > Cd. The data showed that the mean concentration of Pb, Cd, OOC, TOC, and OM were 3.85 mg kg^{-1}, 0.18 mg kg^{-1}, 3.03%, 4.04%, and 6.97%, respectively. The soil samples tested were acidic and non-saline revealed from the mean values of pH (4.87) and EC (0.43 dS m^{-1}). The minimum and maximum concentration of Pb, Cd, OOC, TOC, and OM ranged from 0 to 18 mg kg^{-1}, 0–0.52 mg kg^{-1}, 0.21–4.92%, 0.28–6.57%, and 0.48–11.32%, respectively. The minimum and maximum values of pH and EC ranged from 2.12–6.77 and 0.06–1.92 dS m^{-1}, respectively.

The concentration of Cd in three soil samples from Bijlighar Thall was higher than the permissible limits set by the WHO (2004), i.e., 0.4 mg kg^{-1} for Cd and Canadian Council of Ministers of the Environment

(CCME 1991, i.e., 0.5 mg kg^{-1} for Cd) guidelines, while concentrations of Cd and Pb in rest of soil samples were under permissible limits set by the WHO and CCME, i.e., 18 mg kg^{-1} for Pb. The lower pH might be due to a high amount of organic matter in the soils as clear from the data regarding different organic matter-related parameters in the present study. Earlier, it has been found that the solubility of heavy metals was drastically increased as the soil pH was decreased to 3.3 (Chuan et al. 1996). Higher concentrations of Cd above the permissible limits might be due to the weathering of Zn sulfide and calcium silicate minerals in rocks of the studied area (Shah et al. 2004). These soil samples were collected from the potato-cultivated area, which usually receives a greater amount of phosphate fertilizers, and might be a reason for the higher concentration of Cd in different

Fig. 3 Heavy metal (Cd and Pb) mean contamination factor (CF), pollution load index (PLI), and geo-accumulation index (Igeo) values in different soil samples of the study area
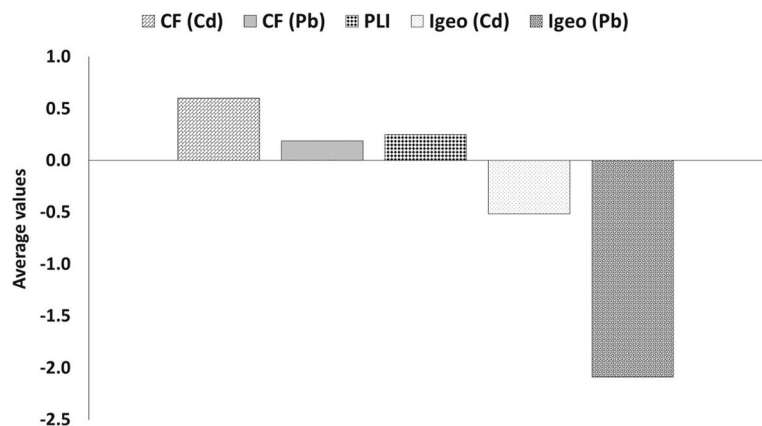
Fig. 4 Ecological risk factors (Eri) of different heavy metal-contaminated soil samples
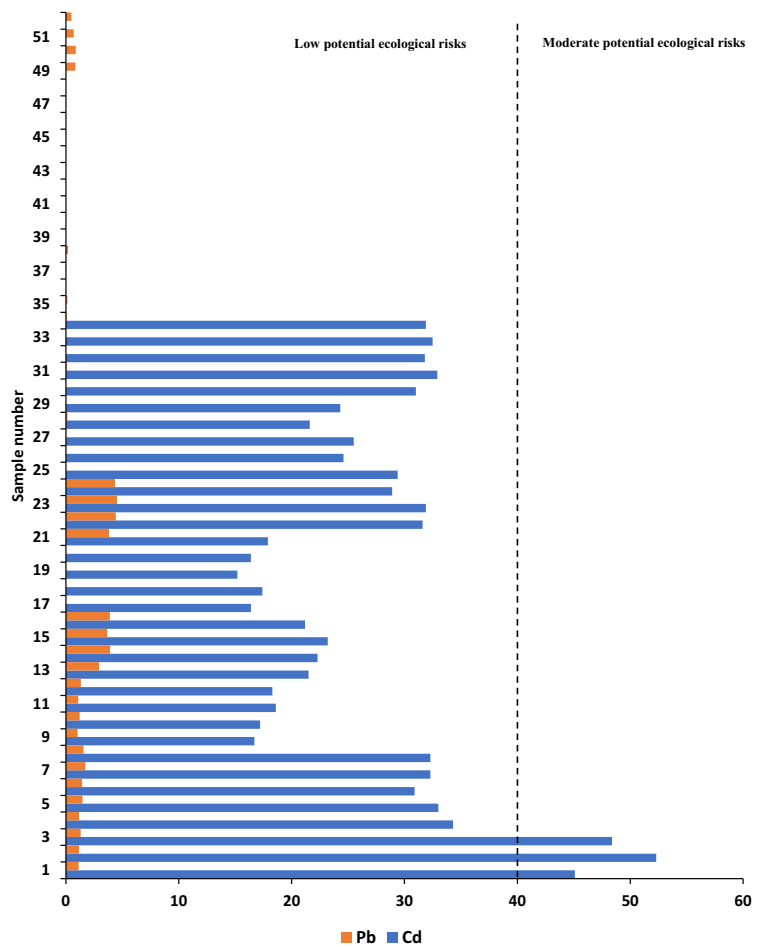
soil samples of the study area (Adimalla et al. 2019; Hou et al. 2019; Parente et al. 2019; Dzul-Caamal et al. 2020). Moreover, the low pH and high organic matter contents observed in the present study might have resulted in increased leaching and contamination of heavy metals to the surrounding areas (Muhammad et al. 2019).

Contamination factors, pollution load index, and geo-accumulation index

The data regarding contamination factors, pollution load, and geo-accumulation indices of the soil samples ($n = 52$) collected from the study area are presented in Table 3. The contamination factor calculated was in the order of Cd > Pb. The contamination factor (CF) calculated for Cd ranged from 0 to 1.74 which showed moderate contamination of Cd, whereas CF values of

the Pb ranged from 0 to 0.91 which showed low contamination in the study area. This classification of soils contaminated with heavy metals is based on CF values, i.e., CF < 1 = low contamination factor (indicating low sediment contamination of the substance in question), $1 \leq CF < 3$ = moderate contamination factor, $3 \leq CF < 6$ = considerable contamination factor, and $CF \geq 6$ = very high contamination factor (Hakanson 1980). The maximum value of pollution load index (PLI) was 0.98 which showed that the soils in the study area were not polluted with respect to Cd and Pb. PLI reported in the present study were less than the values reported by Muhammad et al. (2019) and Mehmood et al. (2019). The reason might be the use of different background values of heavy metals as a reference for the calculation of CF, PLI, and Igeo; thereby, the use of different values may cause some discrepancies in the risk assessment (Hakanson 1980; Wedepohl 1995). For example, both

Table 4 Hazard quotients (HQ) of Cd in children and adults through different exposure routes (ingestion, inhalation and dermal routes)

Sr. No.	Village name	Children			Adults		
		HQing	HQinh	HQderm	HQing	HQinh	HQderm
1	Bijlighar Thall	0.004119	1.56012E-05	0.161454	0.000441	6.68624E-06	0.040775
2		0.004776	1.80919E-05	0.187229	0.000512	7.75366E-06	0.047285
3		0.00442	1.67428E-05	0.173268	0.000474	7.17547E-06	0.043759
4		0.003132	1.18652E-05	0.122791	0.000336	5.0851E-06	0.031011
5	Kalan Bala Thall	0.003014	1.14155E-05	0.118137	0.000323	4.89237E-06	0.029836
6		0.002822	1.06891E-05	0.110619	0.000302	4.58104E-06	0.027937
7		0.00295	1.11734E-05	0.115631	0.000316	4.78859E-06	0.029203
8		0.00295	1.11734E-05	0.115631	0.000316	4.78859E-06	0.029203
9		0.001525	5.77695E-06	0.059784	0.000163	2.47584E-06	0.015099
10		0.001571	5.94991E-06	0.061574	0.000168	2.54996E-06	0.015551
11		0.001699	6.43421E-06	0.066586	0.000182	2.75752E-06	0.016816
12		0.001671	6.33043E-06	0.065512	0.000179	2.71304E-06	0.016545
13		0.001963	7.43739E-06	0.076968	0.00021	3.18745E-06	0.019438
14		0.002037	7.71413E-06	0.079832	0.000218	3.30606E-06	0.020162
15		0.002119	8.02546E-06	0.083054	0.000227	3.43948E-06	0.020975
16	Kalan Payeen Thall	0.001936	7.33361E-06	0.075894	0.000207	3.14298E-06	0.019167
17		0.001498	5.67317E-06	0.058711	0.00016	2.43136E-06	0.014827
18		0.001589	6.0191E-06	0.06229	0.00017	2.57961E-06	0.015731
19		0.001388	5.25806E-06	0.054415	0.000149	2.25345E-06	0.013742
20		0.001498	5.67317E-06	0.058711	0.00016	2.43136E-06	0.014827
21		0.001635	6.19206E-06	0.06408	0.000175	2.65374E-06	0.016184
22		0.002886	1.09312E-05	0.113125	0.000309	4.68481E-06	0.02857
23	Thall Khas	0.002913	1.1035E-05	0.114199	0.000312	4.72929E-06	0.028841
24		0.002639	9.99723E-06	0.103459	0.000283	4.28453E-06	0.026129
25		0.002685	1.01702E-05	0.105249	0.000288	4.35866E-06	0.026581
26		0.002247	8.50976E-06	0.088066	0.000241	3.64704E-06	0.022241
27	Darwazo Thall	0.002329	8.82109E-06	0.091288	0.00025	3.78047E-06	0.023055
28		0.001973	7.47198E-06	0.077326	0.000211	3.20228E-06	0.019529
29		0.002219	8.40598E-06	0.086992	0.000238	3.60256E-06	0.02197
30		0.002831	1.07237E-05	0.110977	0.000303	4.59586E-06	0.028027
31	Haji Shai Payeen Thall	0.003005	1.13809E-05	0.117779	0.000322	4.87754E-06	0.029745
32		0.002904	1.10004E-05	0.113841	0.000311	4.71446E-06	0.028751
33		0.002968	1.12426E-05	0.116347	0.000318	4.81824E-06	0.029384
34		0.002913	1.1035E-05	0.114199	0.000312	4.72929E-06	0.028841
35		0	0	0	0	0	0
36		0	0	0	0	0	0
37		0	0	0	0	0	0
38		0	0	0	0	0	0
39	Haji Shai Bala Thall	0	0	0	0	0	0
40		0	0	0	0	0	0
41		0	0	0	0	0	0
42		0	0	0	0	0	0
43		0	0	0	0	0	0

Table 4 (continued)

Sr. No.	Village name	Children			Adults		
		HQing	HQinh	HQderm	HQing	HQinh	HQderm
44		0	0	0	0	0	0
45		0	0	0	0	0	0
46		0	0	0	0	0	0
47		0	0	0	0	0	0
48		0	0	0	0	0	0
49		0	0	0	0	0	0
50		0	0	0	0	0	0
51		0	0	0	0	0	0
52		0	0	0	0	0	0

Mehmood et al. (2019) and Muhammad et al. (2019) used the background values of Cd and Pb as 6.0 and 1.0 adopted from Harikumar et al. (2009). However, in the present study, background values (Bn) of Cd and Pb used for the calculation of CF were 0.3 and 20 mg kg^{-1}, respectively, adopted from Martin and Meybeck (1979). Geo-accumulation indices (Igeo) of Cd and Pb were less than 1 (Table 3) and showed moderate soil pollution in the study area. The mean CF, PLI, and Igeo values of Cd and Pb in different soil samples of the study area are presented in Fig. 3.

Ecological risk factors and potential ecological risk index

The data regarding ecological risk factors (Eri) of Cd showed that only three samples (highlighted as bold) had the values of Eri greater than 40, indicated moderate whereas for Pb, none of the samples had Eri value greater than 40, thereby posing low potential ecological risks (Fig. 4).

Potential ecological risk index (RI) helps in the characterization of the sensitivity of the local system to the toxic metals and represents the ecological risks associated with overall contamination. RI of all the samples was less than 150 which showed that there were low ecological risks associated with the overall contamination of heavy metals tested (Table 3). Our results are in line with various researchers, who have found that Cd was one of the most contributing factors towards ecological risks associated with the overall contamination of heavy metals (Yuanan et al. 2013; Ajah et al. 2015).

Average daily dose

Table S1 shows the data regarding the average daily dose (ADD, mg kg^{-1} day^{-1}) of Cd in children and adults through ingestion, inhalation, and dermal exposure routes. The data showed that the ingestion was the main route of heavy metal exposure in both children and adults, followed by dermal and inhalation exposure pathways. The values of the reference dose (RfD) have not been established globally; therefore, the available values of RfDs were used in the calculation of ADD through different exposure routes. A similar trend in the values of ADD of Pb was observed in children and adults through ingestion, inhalation, and dermal exposure routes (Table S2) as in the case of Cd was noted. Ingestion of heavy metals was the primary exposure route for both children and adults. It was followed by dermal and inhalation exposure pathways. Earlier, various researchers around the world demonstrated that heavy metals could be dangerous to human through the abovementioned exposure routes (Hamad et al. 2014; Golia et al. 2019; Askari et al. 2020) and found a similar trend in the values of ADD through ingestion, inhalation, and dermal exposure routes (Gu et al. 2016; Liu et al. 2016). Moreover, the values of ADD through various exposure routes in the case of children were more compared to the adults who might be due to their low body weight and hand-to-mouth practices (nail-biting, thumb sucking, soil playing, etc.) (Hamad et al. 2014; Kelepertzis 2014).

Hazard quotients

The hazard quotients (HQ) were found in children and adults following this order: dermal > ingestion >

Table 5 Hazard quotients (HQ) of Lead (Pb) in children and adults through different exposure routes (ingestion, inhalation and dermal routes)

Sr. No.	Village name	Children			Adults		
		HQing	HQinh	HQderm	HQing	HQinh	HQderm
1	Bijlighar Thall	1.1637E-02	4.4081E-07	0.013034	0.001247	1.89E-07	0.003292
2		1.2055E-02	4.5662E-07	0.013501	0.001292	1.96E-07	0.00341
3		1.3673E-02	5.1790E-07	0.015313	0.001465	2.22E-07	0.003867
4		1.2107E-02	4.5860E-07	0.01356	0.001297	1.97E-07	0.003425
5	Kalan Bala Thall	1.5342E-02	5.8115E-07	0.017184	0.001644	2.49E-07	0.00434
6		1.5055E-02	5.7028E-07	0.016862	0.001613	2.44E-07	0.004259
7		1.7926E-02	6.7900E-07	0.020077	0.001921	2.91E-07	0.00507
8		1.6256E-02	6.1575E-07	0.018206	0.001742	2.64E-07	0.004598
9		1.0855E-02	4.1116E-07	0.012157	0.001163	1.76E-07	0.00307
10		1.2707E-02	4.8133E-07	0.014232	0.001361	2.06E-07	0.003594
11		1.1455E-02	4.3389E-07	0.012829	0.001227	1.86E-07	0.00324
12		1.3803E-02	5.2284E-07	0.015459	0.001479	2.24E-07	0.003904
13		3.0633E-02	1.1603E-06	0.034309	0.003282	4.97E-07	0.008665
14		4.0809E-02	1.5458E-06	0.045706	0.004372	6.62E-07	0.011543
15		3.8147E-02	1.4450E-06	0.042725	0.004087	6.19E-07	0.01079
16	Kalan Payeen Thall	4.0705E-02	1.5418E-06	0.045589	0.004361	6.61E-07	0.011514
17		0	0	0	0	0	0
18		0	0	0	0	0	0
19		0	0	0	0	0	0
20		0	0	0	0	0	0
21		3.9870E-02	1.5102E-06	0.044654	0.004272	6.47E-07	0.011277
22		4.6106E-02	1.7464E-06	0.051638	0.00494	7.48E-07	0.013041
23	Thall Khas	4.7332E-02	1.7929E-06	0.053012	0.005071	7.68E-07	0.013388
24		4.5532E-02	1.7247E-06	0.050995	0.004878	7.39E-07	0.012879
25		0	0	0	0	0	0
26		3.3920E-04	1.2849E-08	0.00038	3.63E-05	5.51E-09	9.59E-05
27	Darwazo Thall	1.3046E-04	4.9418E-09	0.000146	1.4E-05	2.12E-09	3.69E-05
28		1.3568E-03	5.1395E-08	0.00152	0.000145	2.2E-08	0.000384
29		0	0	0	0	0	0
30		0	0	0	0	0	0
31	Haji Shai Payeen Thall	5.7404E-04	2.1744E-08	0.000643	6.15E-05	9.32E-09	0.000162
32		0	0	0	0	0	0
33		0	0	0	0	0	0
34		5.7404E-04	2.1744E-08	0.000643	6.15E-05	9.32E-09	0.000162
35		1.3568E-03	5.1395E-08	0.00152	0.000145	2.2E-08	0.000384
36		0	0	0	0	0	0
37		2.3483E-04	8.8952E-09	0.000263	2.52E-05	3.81E-09	6.64E-05
38		1.8265E-03	6.9185E-08	0.002046	0.000196	2.97E-08	0.000517
39	Haji Shai Bala Thall	0	0	0	0	0	0
40		0	0	0	0	0	0
41		0	0	0	0	0	0
42		0	0	0	0	0	0

Table 5 (continued)

Sr. No.	Village name	Children			Adults		
		HQing	HQinh	HQderm	HQing	HQinh	HQderm
43		0	0	0	0	0	0
44		0	0	0	0	0	0
45		0	0	0	0	0	0
46		0	0	0	0	0	0
47		0	0	0	0	0	0
48		0	0	0	0	0	0
49		8.7410E-03	3.3110E-07	0.00979	0.000937	1.42E-07	0.002472
50		9.0541E-03	3.4296E-07	0.010141	0.00097	1.47E-07	0.002561
51		7.3059E-03	2.7674E-07	0.008183	0.000783	1.19E-07	0.002067
52		5.0620E-03	1.9174E-07	0.005669	0.000542	8.22E-08	0.001432

inhalation (Table 4). The inhalation of soil particles through mouth and nose is negligible compared to the other exposure routes, thereby not causing any significant risk. A similar trend in HQ of Pb was noted in children and adults through different exposure routes that show HQ of Pb in children and adults through ingestion, inhalation, and dermal exposure route. The data in Table 5 revealed that the HQ of Pb was found in children and adults following this order: dermal > ingestion > inhalation. The values of HQ through various exposure routes were higher in children as compared to that of the adults, which showed that children were at a higher level of risk associated with the contamination of heavy metals. Earlier, other researchers have also found dermal exposure as the main route posing potential health risks associated with heavy metal-contaminated

Table 6 Rotated component matrix using varimax with Kaiser normalization rotation for heavy metals and physicochemical properties of samples soils

	PC1	PC2	PC3
TOC	*0.985*	0.157	0.037
OOC	*0.985*	0.158	0.038
OM	*0.985*	0.158	0.037
Cd	0.161	*0.836*	− 0.058
Pb	0.163	*0.741*	0.076
EC	0.154	− 0.265	*0.819*
pH	0.074	− 0.306	− *0.804*
Cumulative variance (%)	42.743	63.973	83.002

The italicized components contributed more towards the explanation of total variance

soils (Mehmood et al. 2019). The reason behind might be due to more outdoor activities by the children as compared to adults, thereby becoming more susceptible to the risks associated with heavy metal contamination through ingestion and dermal exposure routes (Karim and Qureshi 2013; Mehmood et al. 2019). Other activities like hand-to-mouth practices may also be responsible for higher values of HQ in children as compared to the adults (Hamad et al. 2014; Kelepertzis 2014).

Total hazard index

Total hazard index (THI) values observed for Cd and Pb in children (0.00 to 0.22) and adults (0.00 to 0.05) were generally less than 1 (USEPA 1989), indicating that these metals were unlikely to have any significant additive or toxic interactions (Table S3). THI represents the cumulative health risks associated with heavy metal contamination in the collected soil samples from different villages. The overall less value might be due to a smaller number of samples with heavy metal contamination above than permissible limits. The values of THI observed in the present study were higher than the values observed in children (1.92E-03 to 9.19E-02) and adults (5.94E-04 to 1.04E-02) during the health risk assessment of heavy metal contamination in the soil of a municipal solid waste dump in Uyo, Nigeria (Golia et al. 2019; Askari et al. 2020). However, higher values of THI in children (1.16–1.36) and adults (0.166–0.195) as compared to the present study have also been reported during the health risk assessment of soil from an abandoned open dumpsite (Ying et al. 2015). Another study

Table 7 Pearson correlation between different heavy metals detected in soil samples and different soil properties (*OOC*, oxidizable organic carbon; *TOC*, total organic carbon; *OM*, organic matter; *pH*, acidity/alkalinity; *EC*, electrical conductivity)

		Pb	Cd	OOC	TOC	OM	pH	EC
Pb	Pearson correlation	1						
	Sig. (2-tailed)							
Cd	Pearson correlation	0.379**	1					
	Sig. (2-tailed)	0.006						
OOC	Pearson correlation	0.261	0.283*	1				
	Sig. (2-tailed)	0.064	0.044					
TOC	Pearson correlation	0.260	0.283*	1.000**	1			
	Sig. (2-tailed)	0.065	0.045	0.000				
OM	Pearson correlation	0.261	0.283*	1.000**	1.000**	1		
	Sig. (2-tailed)	0.064	0.044	0.000	0.000			
pH	Pearson correlation	− 0.104	− 0.163	− 0.035	− 0.035	− 0.035	1	
	Sig. (2-tailed)	0.468	0.253	0.805	0.809	0.806		
EC	Pearson correlation	0.004	− 0.167	0.113	0.112	0.112	− 0.327*	1
	Sig. (2-tailed)	0.976	0.242	0.432	0.436	0.435	0.019	

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

* Correlation is significant at the 0.05 level (2-tailed); $N = 51$

(Peng et al. 2014), during health risk assessment of agricultural soils disturbed by pipeline construction in China, has also reported similar results like THI values noted in children (0.5–1.2), and adults (0.2–0.4) were higher as compared to the present study.

Principal component analysis

A multivariate principal component analysis (PCA) was performed using varimax with Kaiser normalization rotation (Table 6 and Fig. S1). The value of Kaiser-Meyer-Olkin Measure of Sampling Adequacy was 0.723 which was quite good to perform this analysis. The data in Table 6 revealed that three components were extracted based on the initial eigenvalues > 1 and these components were responsible for 83% of the total variance in seven variables tested during PCA analysis. Principal component 1 (PC1) explained 42.7% of the total variance and was dominated by TOC, OOM, and OM. It was considered to be lithogenic because it is controlled by lithogenic and pedogenic soil processes having high organic matter contents. Also, most of the attributes tested were controlled by these factors as this premise has also been supported by strong correlations between these attributes conferred from Pearson correlation analysis given in Table 7. PC2 included Cd and Pb and these accounted for 21.23% of the total variance.

It was defined as an anthropogenic component related to different agronomic practices such as fertilizer addition and pesticide application. Earlier, it has been found that long-term use of phosphatic fertilizers and pesticides in agricultural as well as orchard soils results in the accumulation of these heavy metals (Nogueirol et al. 2010; Khan et al. 2013; Liang et al. 2015). PC3 explained only 19% of the total variance, accounting for pH and EC of the soil samples tested. This may be defined as a lithogenic and anthropogenic component as the soils in the study area contained a high amount of organic matter, and there was also the usage of different phosphatic fertilizers as well as pesticides. The component plot in the rotated space observed through PCA analysis is presented in Fig. S1.

Pearson correlation analysis

Pearson correlation analysis among all the parameters tested during the health risk assessment of different soil samples collected from the study area was performed (Table 7). Two levels of significance ($P < 0.05$ and $P < 0.01$) for multi-element correlation were calculated. The r values (> 0.7) observed during the analysis showed a high degree positive and significant correlation between various parameter pairs tested. A highly significant and positive correlation ($P < 0.01$) was

Ⓐ Springer

observed between Cd and Pb concentration in different soil samples tested. A similar type of highly significant and positive correlation ($P < 0.01$) between oxidizable organic carbon (OOC) and organic matter (OM), OOC, and total organic carbon (TOC) and OM and TOC as clear from the data is presented in Table 7. However, a statistically significant positive but weak correlation ($P < 0.05$) was observed between Cd concentration and OOC, OM, and TOC. Only one significant but negative correlation ($P < 0.05$) between pH and EC was observed.

Conclusions

From the results, it was found that concentrations of Cd and Pb in all the soil samples were under permissible limits set by the World Health Organization (WHO) and Canadian Council of Ministers of the Environment (CCME) except in three soil samples collected from Bijlighar Thall, which were higher Cd than the permissible limits. Moreover, the low pH and high organic matter contents recorded in the present study might have resulted in increased leaching and contamination to the surrounding areas. The soils in the study area were classified as uncontaminated to moderately contaminated soils conferred from the contamination factor (CF) values of different soil samples. The human health risk assessment through hazard quotient (HQ) values showed that dermal was the main exposure route for heavy metal contamination in both adults and children. The findings of this study could be helpful for government agencies involved in environmental protection such as Pakistan Environmental Protection Agency (Pak-EPA) which can suggest control measures for Cd and Pb contamination in the agricultural soils.

References

Adimalla, N., Qian, H., & Wang, H. (2019). Assessment of heavy metal (HM) contamination in agricultural soil lands in northern Telangana, India: an approach of spatial distribution and multivariate statistical analysis. *Environmental Monitoring and Assessment, 191*, 246. https://doi.org/10.1007/s10661-019-7408-1.

Ahmad, I., Akhtar, M. J., Mehmood, S., Akhter, K., Tahir, M., Saeed, M. F., Hussain, M. B., & Hussain, S. (2018). Combined application of compost and Bacillus sp. CIK-512 ameliorated the lead toxicity in radish by regulating the homeostasis of antioxidants and lead. *Ecotoxicology Environmental Safety, 148*, 805–812.

Ahmad, I., Tahir, M., Daraz, U., Ditta, A., Hussain, M. B., & Khan, Z. U. H. (2020). Responses and tolerance of cereal crops to metals and metalloids toxicity. In H. Hasanuzzaman (Ed.), *Agronomic crops*. Singapore: Springer. https://doi.org/10.1007/978-981-15-0025-1_14.

Ajah, K. C., Ademiluyi, J., & Nnaji, C. C. (2015). Spatiality, seasonality and ecological risks of heavy metals in the vicinity of a degenerate municipal central dumpsite in Enugu, Nigeria. *Journal of Environmental Health Science & Engineering, 13*(1), 15. https://doi.org/10.1186/s40201-015-0168-0.

Askari, M. S., Alamdari, P., Chahardoli, S., & Afshari, A. (2020). Quantification of heavy metal pollution for environmental assessment of soil condition. *Environmental Monitoring and Assessment, 192*, 162. https://doi.org/10.1007/s10661-020-8116-6.

Askarova, M. A., & Mussagaliyeva, A. N. (2014). The ecological situation in contaminated areas of oil & gas exploration in Atyrau Region. *The Procedia - Social and Behavioral Sciences, 120*, 455–459.

Caeiro, S., Costa, M. H., & Ramos, T. B. (2005). Assessing heavy metal contamination in Sado Estuary sediment: an index analysis approach. *Ecological Indicators, 5*, 151–169.

Cao, C., Zhang, Q., Ma, Z.-B., Wang, X.-M., Chen, H., & Wang, J.-J. (2018). Fractionation and mobility risks of heavy metals and metalloids in wastewater-irrigated agricultural soils from greenhouses and fields in Gansu, China. *Geoderma, 328*, 1–9.

CCME (Canadian Council of Ministers of the Environment). (1991). *Interim Canadian environmental quality criteria for contaminated sites*. Winnipeg: CCME.

Chen, H. Y., Teng, Y. G., Lu, S. J., Wang, Y. Y., & Wang, J. S. (2015). Contamination features and health risk of soil heavy metals in China. *Science of the Total Environment, 512–513*, 143–153.

Chuan, M. C., Shu, G. Y., & Liu, J. C. (1996). Solubility of heavy metals in a contaminated soil: effects of redox potential and pH. *Water Air & Soil Pollution, 90*, 543–556. https://doi.org/10.1007/BF00282668.

Dzul-Caamal, R., Vega-López, A., & Osten, J. R. (2020). Distribution of heavy metals in crop soils from an agricultural region of the Yucatan Peninsula and biochemical changes in earthworm Eisenia foetida exposed experimentally. *Environmental Monitoring and Assessment, 192*, 338. https://doi.org/10.1007/s10661-020-08273-7.

Elbehiry, F., Elbasiouny, H., El-Ramady, H., & Brevik, E. C. (2019). Mobility, distribution, and potential risk assessment of selected trace elements in soils of the Nile Delta, Egypt. *Environmental Monitoring and Assessment, 191*, 713. https://doi.org/10.1007/s10661-019-7892-3.

Ferreira-Baptista, L., & De Miguel, E. (2005). Geochemistry and risk assessment of street dust in Luanda, Angola: a tropical

urban environment. *Atmospheric Environment, 39*(25), 4501–4512.

Fowler, B. A. (2009). Monitoring of human populations for early markers of cadmium toxicity: a review. *Toxicology and Applied Pharmacology, 238*, 294–300.

Fu, G., Zhang, L., Cui, W., Wang, Y., Shen, W., Ren, Y., & Zheng, T. (2011). Induction of heme oxygenase-1 with β-CD-hemin complex mitigates cadmium-induced oxidative damage in the roots of Medicago sativa. *Plant and Soil, 345*(1–2), 271–285.

Githaiga, K. B., Njuguna, S. M., Makokha, V. A., Wang, J., Gituru, R. W., & Yan, X. (2020). Assessment of Cu, Zn, Mn, and Fe enrichment in Mt. Kenya soils: evidence for atmospheric deposition and contamination. *Environmental Monitoring and Assessment, 192*, 167. https://doi.org/10.1007/s10661-020-8123-7.

Golia, E. E., Tsiropoulos, G. N., Füleky, G., Floras, S., & Vleioras, S. (2019). Pollution assessment of potentially toxic elements in soils of different taxonomy orders in central Greece. *Environmental Monitoring and Assessment, 191*, 106. https://doi.org/10.1007/s10661-019-7201-1.

Gu, Y. G., Gao, Y. P., & Lin, Q. (2016). Contamination, bioaccessibility and human health risk of heavy metals in exposed-lawn soils from 28 urban parks in southern China's largest city, Guangzhou. *Applied Geochemistry, 67*, 52–58. https://doi.org/10.1016/j.apgeochem.2016.02.004.

Gul, N., Shah, M. T., Khan, S., Khattak, N. U., & Muhammad, S. (2015). Arsenic and heavy metals contamination, risk assessment and their source in drinking water of the Mardan District, Khyber Pakhtunkhwa, Pakistan. *Journal of Water and Health, 13*(4), 1073–1084.

Guo, H., Yang, L., Han, X., Dai, J., Pang, X., Ren, M., & Zhang, W. (2019). Distribution characteristics of heavy metals in surface soils from the western area of Nansi Lake, China. *Environmental Monitoring and Assessment, 191*, 262. https://doi.org/10.1007/s10661-019-7390-7.

Hakanson, L. L. (1980). Ecological risk index for aquatic pollution control, a sedimentological approach. *Water Research, 14*(8), 975–1001.

Hamad, S. H., Schauer, J. J., Shafer, M. M., Al-Rheem, E. A., Skaar, P. S., Heo, J., & Tejedor-Tejedor, I. (2014). Risk assessment of total and bioavailable potentially toxic elements (PTEs) in urban soils of Baghdad–Iraq. *Science of the Total Environment, 494*, 39–48.

Harikumar, P., Nasir, U., & Rahman, M. M. (2009). Distribution of heavy metals in the core sediments of a tropical wetland system. *International journal of Environmental Science and Technology, 6*, 225–232.

HC. (2004). *Federal Contaminated Site Risk Assessment in Canada-Part II: Health Canada Toxicological Reference Values (TRVs) and Chemical-Specific Factors*. Ottawa, Canada: HC.

Hou, S., Zheng, N., Tang, L., Ji, X., & Li, Y. (2019). Effect of soil pH and organic matter content on heavy metals availability in maize (Zea mays L.) rhizospheric soil of non-ferrous metals smelting area. *Environmental Monitoring and Assessment, 191*, 634. https://doi.org/10.1007/s10661-019-7793-5.

Ihedioha, J. N., Ukoha, P. O., & Ekere, N. R. (2017). Ecological and human health risk assessment of heavy metal contamination in soil of a municipal solid waste dump in Uyo.

Nigeria. *Environmental Geochemistry and Health, 39*(3), 497–515.

Jan, A. T., Azam, M., Siddiqui, K., Ali, A., Choi, I., & Haq, Q. M. R. (2015). Heavy metals and human health: mechanistic insight into toxicity and counter defense system of antioxidants. *International Journal of Molecular Sciences, 16*(12), 29592–29630.

Karim, Z., & Qureshi, B. A. (2013). Health risk assessment of heavy metals in urban soil of Karachi, Pakistan. *Human and Ecological Risk Assessment, 20*, 658–667.

Kelepertzis, E. (2014). Investigating the sources and potential health risks of environmental contaminants in the soils and drinking waters from the rural clusters in Thiva area (Greece). *Ecotoxicology and Environmental Safety, 100*, 258–265.

Khan, K., Lu, Y., Khan, H., Ishtiaq, M., Khan, S., Waqas, M., Wei, L., & Wang, T. (2013). Heavy metals in agricultural soils and crops and their health risks in Swat District, northern Pakistan. *Food and Chemical Toxicology, 58*, 449–458.

Lacatusu, R. (2000). Appraising levels of soil contamination and pollution with heavy metals. In H. J. Heinike, W. Eckselman, A. J. Thomasson, R. J. A. Jones, L. Montanarella, & B. Buckeley (Eds.), *Land information systems for planning the sustainable use of land resources. European Soil Bureau Research Report No. 4* (pp. 393–402). Luxembourg: Office of Official Publication of the European Communities.

Latif, J., Akhtar, J., Ahmad, I., Mahmood-ur-Rehman, M., Shah, G. M., Zaman, Q., et al. (2020). Unraveling the effects of cadmium on growth, physiology and associated health risks of leafy vegetables. *Brazilian Journal of Botany, 43,* 799–811. https://doi.org/10.1007/s40415-020-00653-0.

Leblebici, Z., Kar, M., & Başaran, L. (2020). Assessment of the heavy metal accumulation of various green vegetables grown in Nevşehir and their risks human health. *Environmental Monitoring and Assessment, 192*, 483. https://doi.org/10.1007/s10661-020-08459-z.

Liang, Q., Xue, Z., Wang, F., Sun, Z., Yang, Z., & Liu, S. (2015). Contamination and health risks from heavy metals in cultivated soil in Zhangjiakou City of Hebei Province, China. *Environmental Monitoring and Assessment, 187*(12), 754.

Liu, Y., Ma, J., Yan, H., Ren, Y., Wang, B., Lin, C., & Liu, X. (2016). Bioaccessibility and health risk assessment of arsenic in soil and indoor dust in rural and urban areas of Hubei province, China. *Ecotoxicology and Environmental Safety, 126*, 14–22. https://doi.org/10.1016/j.ecoenv.2015.11.037.

Manahan, S. E. (2001). *Fundamentals of environmental chemistry* (2nd ed.pp. 853–861). LLC Boca Ralton, Florida: CRC Press.

Martin, J., & Meybeck, M. (1979). Elemental mass-balance of material carried by major world rivers. *Marine Chemistry, 7*, 178–206.

Mehmood, K., Ahmad, H. R., Abbas, R., & Saifullah, & Murtaza, G. (2019). Heavy metals in urban and peri-urban soils of a heavily-populated and industrialized city: assessment of ecological risks and human health repercussions. *Human and Ecological Risk Assessment: An International Journal, 26*(6), 1705–1722. https://doi.org/10.1080/10807039.2019.1601004.

Muhammad, S., Ullah, R., & Jadoon, I. A. K. (2019). Heavy metals contamination in soil and food and their evaluation for risk assessment in the Zhob and Loralai valleys,

Baluchistan province, Pakistan. *Microchemical Journal, 149*, 103971. https://doi.org/10.1016/j.microc.2019.103971.

Muller, G. (1969). Index of geoaccumulation in sediments of the Rhine River. *GeoJournal, 2*(3), 108–118.

Nickson, R. T., McArthur, J. M., Shrestha, B., Kyaw-Myint, T. O., & Lowry, D. (2005). Arsenic and other drinking water quality issues, Muzaffargarh District, Pakistan. *Applied Geochemisrty, 20*, 55–68.

Nogueirol, R. C., Alleoni, L. R. F., Nachtigall, G. R., & de Melo, G. W. (2010). Sequential extraction and availability of copper in Cu fungicide-amended vineyard soils from southern Brazil. *Journal of Hazardous Materials, 181*, 931–937.

Nwajei, P. E., & Gagophein, P. O. (2000). Distribution of heavy metals in the sediments of Lagos lagoon. *Pakistan Journal of Scientific and Industrial Research, 43*, 338–340.

Odum, H. T. (2000). Back ground of published studies on lead and wetland. In T. O. Howard (Ed.), *Heavy metals in the environment using wetlands for their removal* (p. 32). New York: Lewis publishers.

Parente, C. E. T., Lino, A. S., Junior, E. R. A., Zonta, E., Dorneles, P. R., Torres, J. P. M., Meire, R. O., & Malm, O. (2019). Multi-temporal accumulation and risk assessment of available heavy metals in poultry litter fertilized soils from Rio de Janeiro upland region. *Environmental Monitoring and Assessment, 191*, 28. https://doi.org/10.1007/s10661-018-7156-7.

Peng, S., Jun, X., Yafeng, W., & Liding, C. (2014). Assessment of ecological and human health risks of heavy metal contamination in agriculture soils disturbed by pipeline construction. *International Journal of Environmental Research and Public Health, 11*, 2504–2520.

Shah, M. T., Kifyattullah, Q., & Irfan, M. (2004). Pedo and biogeochemical study of zinc–lead deposits of the Besham area, northern Pakistan; its implication in mineral exploration and environmental degradation. *Environmental Geology, 45*, 544–549.

Stoffers, P., Glasby, G. P., Wilson, C. J., Davis, K. R., & Walter, P. (1986). Heavy metal pollution in Wellington Harbour. *New Zealand Journal of Marine and Freshwater Research, 20*(3), 495–512.

Teng, Y. G., Ni, S. J., Wang, J. S., Zuo, R., & Yang, J. (2010). A geochemical survey of trace elements in agricultural and nonagricultural topsoil in Dexing area, China. *Journal of Geochemical Explorations, 104*(3), 118–127.

Tomlinson, D. L., Wilson, J. G., Harris, C. R., & Jeffrey, D. W. (1980). Problems in the assessment of heavy-metal levels in estuaries and the formation of a pollution index. *Helgoländer Meeresuntersuchungen, 33*(1), 566–575.

USEPA. (1989). Risk assessment guidance for superfund. *Human Health Evaluation Manual (Part A), I*, EPA/540/1–89/002.

Washington, DC: Office of Emergency and Remedial Response.

USEPA. (1996). *Water quality criteria and standards newsletter, Jan/Feb 1996*. Washington, DC: USEPA Office of Science and Technology.

USEPA. (2010). User's guide (EB/OL). http://www.epa.gov/. Accessed 26 Sept 2019

US-EPA. (2001). *Supplemental guidance for developing soil screening levels for superfund sites*. Washington DC: U.S. Environmental Protection Agency, Office of Solid Waste and Emergency Response.

Van den Berg, R. (1995). Human exposure to soil contamination: a qualitative and quantitative analysis towards proposals for human toxicological intervention values. RIVM Report no. 725201011. Bilthoven, The Netherlands: National Institute of Public Health and Environmental Protection (RIVM).

Walkley, A. (1947). A critical examination of a rapid method for determining organic carbon in soils: Effect of variations in digestion conditions and of organic soil constituents. *Soil Science, 63*, 251–263.

Wang, Y., Qiao, M., Liu, Y., & Zhu, Y. (2012). Health risk assessment of heavy metals in soils and vegetables from wastewater irrigated area, Beijing-Tianjin city cluster, China. *Journal of Environmental Science, 24*(4), 690–698.

Waseem, A., Arshad, J., Iqbal, F., Sajjad, A., Mehmood, Z., & Murtaza, G. (2014). Pollution status of Pakistan: a retrospective review on heavy metal contamination of water, soil, and vegetables. *BioMed Research International., 2014*, 1–29. https://doi.org/10.1155/2014/813206.

Wedepohl, K. H. (1995). The composition of the continental crust. *Geochimica et Cosmochimica Acta, 59*, 1217–1232.

World Health Organization (WHO). (2004). *Guidelines for drinking water quality. World Health Organization (WHO)* (Vol. 1, 3rd ed.). Geneva: World Health Organization.

Ying, H., YongXia, L., Jian, Y., MinMin, X., Bo, S., FuWei, G., & Ning, W. (2015). Harmful chemicals in soil and risk assessment of an abandoned open dumpsite in Eastern China. *Journal of Chemistry, 2015*, 1–10. https://doi.org/10.1155/2015/297686.

Yuanan, H., Xueping, L., Jinmei, B., Kaimin, S., Eddy, Y. Z., & Hefa, C. (2013). Assessing heavy metal pollution in the surface soils of a region that had undergone three decades of intense industrialization and urbanization. *Environmental Science and Pollution Research, 20*, 6150–6159.