

Contamination of soil, medicinal, and fodder plants with lead and cadmium present in mine-affected areas, Northern Pakistan

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Abstract This study aimed to investigate the lead (Pb) and cadmium (Cd) concentrations in the soil and plants (medicinal and fodder) grown in chromite mining-affected areas, Northern Pakistan. Soil and plant samples were collected and analyzed for Pb and Cd concentrations using atomic absorption spectrometer. Soil pollution load indices (PLIs) were greater than 2 for both Cd and Pb, indicating high level of contamination in the study area. Furthermore, Cd concentrations in the soil surrounding the mining sites exceeded the maximum

allowable limit (MAL) (0.6 mg kg^{-1}), while the concentrations of Pb were lower than the MAL (350 mg kg^{-1}) set by State Environmental Protection Administration (SEPA) for agriculture soil. The concentrations of Cd and Pb were significantly higher ($P < 0.001$) in the soil of the mining-contaminated sites as compared to the reference site, which can be attributed to the dispersion of toxic heavy metals, present in the bed rocks and waste of the mines. The concentrations of Pb and Cd in majority of medicinal and fodder plant species grown in surrounding areas of mines were higher than their MALs set by World Health Organization/Food Agriculture Organization (WHO/FAO) for herbal (10 and 0.3 mg kg^{-1} , respectively) and edible (0.3 and 0.2 mg kg^{-1} , respectively) plants. The high concentrations of Cd and Pb may cause contamination of the food chain and health risk.

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Introduction

The local flora is an important source of traditional medicines that are used for the treatment of various diseases. More than 50,000 plant species have been used for medicinal purposes (Bako et al. 2005). Local people get knowledge from their ancestors about the utilization of medicinal plants. A major part of Pakistan is quite rich in medicinal herbs due to its healthy climate (Abbasi et al. 2010a, b). Heavy metals such as lead (Pb), cadmium (Cd), zinc (Zn), and copper (Cu) are considered as global

environmental contaminants. These metals have both natural (weathering of parent rocks and ore deposits) and man-made (mining, industrial emission; smelting, waste water irrigation and application of fertilizers) sources (Nawab et al. 2015; Khan et al. 2013a, 2013b; Wei and Yang 2010). Mining activities cause serious adverse effects on plants and soil (Jung 2008) which are mostly connected with open dumping of mine tailings that usually affect the condition of natural plants growing on it (Nawab et al. 2015; Nouri et al. 2011). Native plant species growing on mine-affected sites have greater potential for Pb and Cd accumulation considering the combine pollution of most contaminated sites (Zhao et al. 2014). Plant species can tolerate these severe conditions up to some level. However, high heavy metal concentrations in medicinal plants can cause stress and stomata resistance (Shah et al. 2009; Fayiega et al. 2004). Heavy metals play an important role in food chain contamination (Waqas et al. 2015); ruminants such as cattle, sheep, and goats feed on fodder grasses and accumulate heavy metals over time. It is now well understood that there is transfer of toxic heavy metals from contaminated soil to plants and from plants to livestock (Oskaron et al. 1992). The toxicity of heavy metals in the food chain increases as the heavy metal concentrations exceed the maximum allowable limits (MALs) in the soil. Mostly, the health risk is associated with toxic metal-contaminated soil or dust deposited on plants (Holmgren et al. 1993; Carrington and Bolger 1992). Therefore, grazing animals are more at risk as fodder grasses are consumed directly without being washed.

Pb and Cd are toxic pollutants, and they are not known for any significant biological functions. They rather produce different toxic effects in animals and human beings, which may result in undesirable biochemical and physiological functions (Khan et al. 2014). Abnormal liver disorders and plasma hormonal changes have been reported in cows that were exposed to Pb and Cd in industrial areas (Ogundiran et al. 2012; Swarup et al. 2007).

Plants play an important role for the transformation of heavy metals such as Cd and Pb from contaminated soils to humans (Khan et al. 2015). In polluted soil, the grown plants can uptake more heavy metals. Cd and Pb were chosen for risk assessment because of their high toxicity or comparatively high levels in all of the collected rice samples in previous research. Cd is toxic to the kidney and has a long biological half-life in human. Cd also affects lungs, reproductive system, kidneys, and bones (Godt et al. 2006). Pb has shown to be associated

with densification of central nervous system and can cause various health problems like anemia, neurological disorders, hyperactivity, and enzyme changes in humans (Muhammad et al. 2011; Marsden 2003; Chamannejadian 2011). Previous studies indicated that overdose or prolonged ingestion of medicinal plants leads to the chronic accumulation of different elements and hence creates various health problems (Sharma et al. 2009; WHO 1992). Therefore, it is important to screen out the elemental concentrations in medicinal plants for their quality control insurance (Kunle et al. 2012; Arceusz et al. 2010; Liang et al. 2004). Assessment of toxic metals in raw herbal material and finished products must be ensured. Recently, several research works have been focused on heavy metal concentrations in different medicinal plants (Hussain et al. 2011; Baye and Hymete 2010).

In the view of the previous research work, the current study is aimed to evaluate the Cr associated toxic metals like Pb and Cd levels in mine-affected soil, medicinal herbs, and fodder grasses that are locally used in the study area. The pollution load index and transfer factor of selected elements were also investigated.

Materials and method

Study area

The study area is mainly comprised of mining-affected sites (MAS) that are Shangla District (MAS1), Malakand Agency (MAS2), and Mohmand Agency (MAS3) in northern parts of Pakistan. Geographically, MAS1 lies between latitude 34, 31, to 33°, 08° N and longitude 72, 33, to 73°, 01° E with a total area of 1586 km² and elevation of 3164 m above from mean sea level. MAS1 is bound on the east by district Batagram and tribal area of Kala Dhaka along which the river Indus flows for about 75 km, on the west by district Swat, on the south by district Buner and tribal area on Kala Dahka, and on the north by district Kohistan.

MAS2 is located between latitude 34° 30' N and longitude 71° 45' E and stretches over an area of about 95,200 ha (Fig. 1). The area is bounded on the north by mountainous range of Swat and lower Dir, on the west by mountainous range of Bajaur and Mohmand Agency, on the east by Buner, southeast by Mardan District, and southwest by Charsada District. MAS3 is located between latitude 34° 22' 20" N and longitude 71° 27' 26" E. Bajaur Agency is in the north, Khyber Agency is in

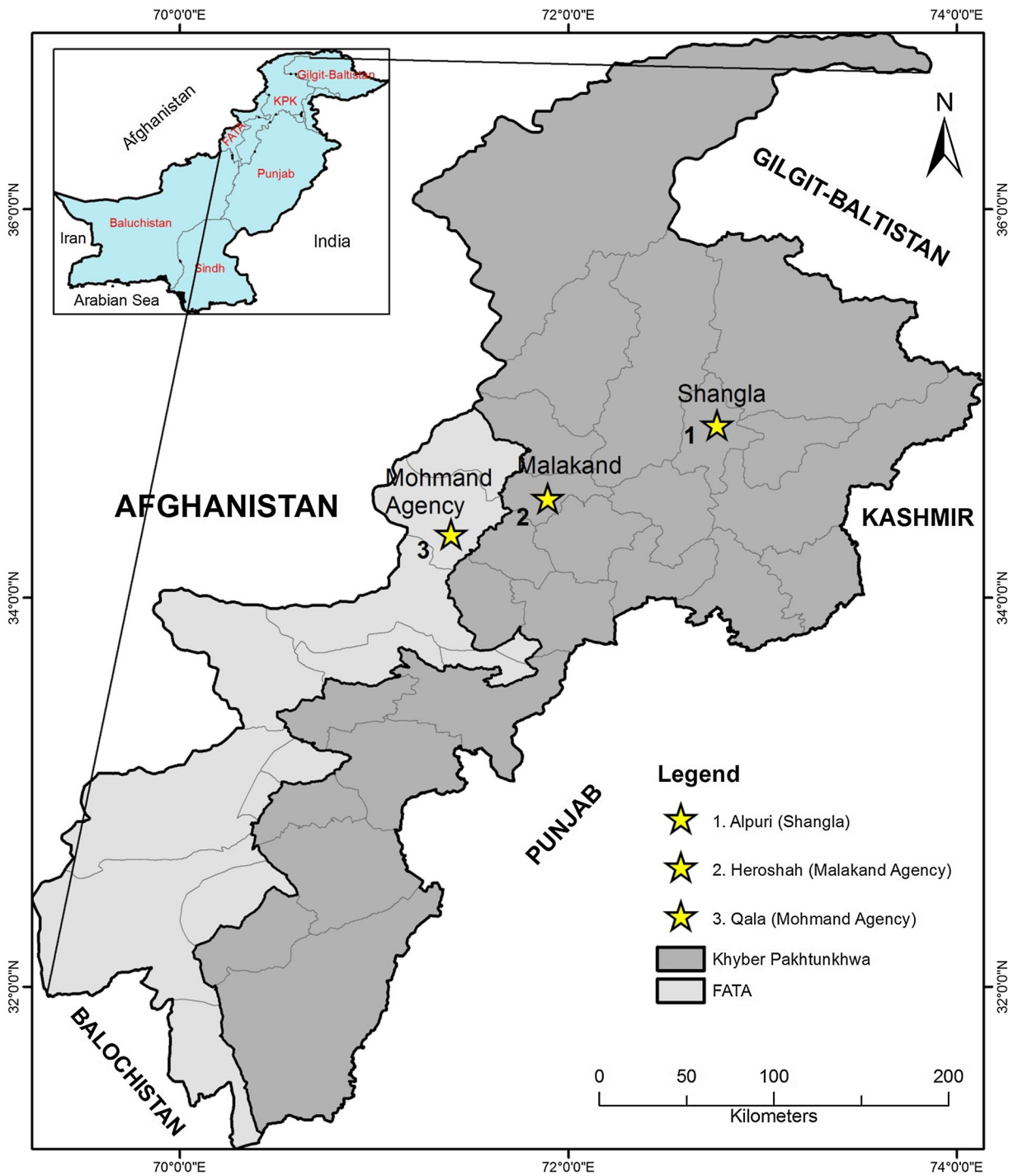


Fig. 1 Map of the study area showing the chromite mine sampling sites

the south, Malakand Agency and Charsada District are in the east, and Peshawar District is in the southeast of Mohmand Agency. Geologically, the MAS1, MAS2, and MAS3 are mainly comprised of mafic and ultramafic rocks (Searle and Khan 1996). These rocks are hosting chromite deposits and at some places lead-zinc, copper, and manganese ores are present where small-scale minings are in progress. The study was conducted in these areas because the flora is enriched with medicinal and fodder plants and majority of the people use the medicinal plants for various health problems, while the fodder plants consume by the cows, goats, and sheep in the study area.

Background sampling area

For background sampling, Charsadda area was selected which has lacustrine soil deposits with no contamination from the mafic and ultramafic rocks (Shah and Tariq 2007). It is 20–170 km away from contaminated sampling site. Therefore, for comparison purposes, soil and plant samples were collected for background heavy metal concentrations.

Soil and plant sampling

The soil samples were collected with auger (from where the plant samples uprooted, $n=115$) from the upper horizon (0–20 cm). Organic matter, gravels, pebbles, and twigs were removed with hands from the collected soil samples and were kept in the kraft papers. Plants samples were randomly collected, properly marked, and stored in polythene bags. Herbarium sheets for each plant species were prepared, and plants were identified with the help of taxonomist. Names of the collected plants with their common uses are given in Table 1. Both soil and plant species were transported to the Department of Environmental Sciences, University of Peshawar, Pakistan for further analysis.

Preparation and digestion of soil samples

Soil samples were air dried at room temperature and passed through ≤ 2 mm mesh for further analysis. The subsamples were analyzed for the physicochemical parameters like soil texture, pH, electrical conductivity (EC), and soil organic matters (SOMs) according to the standard procedures. For heavy metal extraction, wet digestion method (WDM) was used (Khan et al. 2013a; Macalalad et al. 1988). Dried soil samples of

0.5 g were taken in Teflon beakers, and 15 ml of aqua regia (1:3 HNO₃ and HCl) was added to each beaker. The samples were kept overnight and then gently heated on hot plate at 80 °C until the appearance of transparent extract was observed. The digested extracts were filtered into clean volumetric flasks and diluted up to 100 ml with double deionized water for further analyses. Digestion was performed in triplicates.

Preparation and digestion of plant samples

Plants were washed with double deionized water, separated into roots and shoots and oven dried at 70 °C. The samples were ground into powder with electric grinder. About 1 g of each sample was taken in beaker digested with 15 ml mixture of perchloric (HClO₄) and nitric acid (HNO₃) in the ratio of 1:4 (Khan et al. 2013a). The samples were kept overnight. Each sample was heated on hot plate at low temperature until transparent solution was observed. After cooling, the solution was filtered and diluted with double deionized water up to 100 ml for further interpretation. Like soil, digestion of plants was also performed in triplicates

Analytical procedure and quality control

Analytical grade chemicals were used for sample preparation and analysis. Standards for each heavy metal were prepared according to the procedure adopted by Khan et al. (2008). The concentrations of Cd and Pb in the plant and soil samples were analyzed using (AAS Perkin-Elmer A-700). The standard reference soil and plant materials (GBW-07406 (GSS-6) and GBW-07602 (GSV-1), respectively, were purchased from National Research Centre for Certified Reference Materials, China) and blank samples were included in each sample batch to verify the accuracy and precision of the digestion procedure and subsequent analyses. The accuracy and precision were found within 90–95 % confidence limit. All analyses were performed in triplicates at the National Center for Excellence in Geology (NCEG), University of Peshawar, Pakistan.

Contamination factor

Contamination factor (CF) is a ratio obtained from heavy metal concentrations in contaminated and background sites, as adopted by Mmolawa et al. (2010).

Table 1 Common uses of medicinal and fodder plants in “traditional system” Northern Pakistan

Plant specie name	Family	Abbreviations	Local name	Uses	Ethnobotanical uses	References
<i>Aerva javanica</i>	Amaranthaceae	<i>A. javanica</i>	Kaharbotey	Medicinal	Hemorrhoids, skin dryness, and self cracking of skin	Murad et al. 2010
<i>Solanum surattense</i>	Solanaceae	<i>S. surattense</i>	Maraghoney	Medicinal	Expectorant, digestive, astringent, diuretic, asthma, cough, fever, chest pain, and jaundice	Haq et al. 2011
<i>Cymbopgon jwarancusa</i>	Poaceae	<i>C. jwarancusa</i>	Sargara	Medicinal	Used for chicken pox	Ahmad et al. 2010
<i>Calotropis procera</i>	Asclepiadaceae	<i>C. procera</i>	Spalmay	Medicinal	Paste of leaves in oil is used as pain killer, to cure skin itch, and scabies. The root bark is used for the treatment of cholera and constipation.	Ahmad et al. 2011
<i>Artemisia scoparia</i>	Asteraceae	<i>A. scoparia</i>	Jokey	Medicinal	Respiratory stimulant, anthelmintic purgative and against earache	Ahmad et al. 2011
<i>Razia stracta</i>	Apocynaceae	<i>R. stracta</i>	Gandechar	Medicinal	The leaves extract is used as anticancer. Extract of root is used for toothache	Murad et al. 2010
<i>Lotus corniculatus</i>	Fabaceae	<i>L. corniculatus</i>	Fathkhaney	Medicinal	Decoction of dried powdered plant with ghee or boiled water against sexual debility and backache	Akhtar et al. 2013
<i>Solanum xanthocarpum</i>	Solanaceae	<i>S. xanthocarpum</i>	Markundai	Medicinal	Respiratory disorder, stomach disorder, throat sore, paste of roots applied on gums, in snake bite and scorpion bite.	Malik et al. 2011
<i>Sisymbrium irio linn</i>	Solanaceae	<i>S. irio</i>	Khob Kalan	Medicinal	Leaves are used for stomach problems. Leaves are used to as antipyretic.	Marwat et al. 2008
<i>Impatiens bicolor royle</i>	Balsaminaceae	<i>I. bicolor royle</i>	Bantil	Medicinal	It is diuretic, tonic, and has cooling effect.	Haq et al. 2011
<i>Plantago lanceolata</i>	Plantaginaceae	<i>P. lanceolata</i>	Jabey	Medicinal	Fresh crushed leaves are applied to treat wounds sores and inflamed surfaces, particularly in feet.	Sher et al. 2010
<i>Nepeta cartia L.</i>	Lamiaceae	<i>N. cataria</i>	Pisho botai	Medicinal	Dried leaves and flowering tops carminative	Akhtar et al. 2013
<i>Rumex hastatus</i>	Polygonaceae	<i>R. hastatus</i>	Tharukay	Medicinal	Leaves used as local vegetable which enhances digestion	Sher et al. 2010
<i>Isodon roguses</i>	Labiatae	<i>I. roguses</i>	Sperkey	Medicinal	Dried leaves put in mouth as remedy for toothache	Haq et al. 2011
<i>Amaranthus spinosus</i>	Amaranthaceae	<i>A. spinosus</i>	Ghano chalwae	Medicinal	Leaves are cooked and used to kill thread worms within the body, for kidney and low blood pressure	Mushtaq et al. 2012
<i>Amaranthus viridis L.</i>	Amaranthaceae	<i>A. viridis</i>	Ganhar	Medicinal	Leaf extract is emollient, also used for curing cough and asthma.	Akhtar et al. 2013
<i>Ajuga bracteosa</i>	Lamiaceae	<i>A. bracteosa</i>	Booti	Medicinal	Fresh leaf extract is taken orally before breakfast for curing earache, eye ache, boils, mouth gums, and throat pain	Sher et al. 2010
<i>Xanthoxylem armatum</i>	Rutaceae	<i>X. armatum</i>	Dambara	Medicinal	Body tonic, aromatic, fever, cholera, dyspepsia stomachache, and toothache	Haq et al. 2011
<i>Tribulus terrestris L.</i>	Zygophyllaceae	<i>T. terrestris</i>	Markundai	Medicinal	It is used as a folk medicine for increased muscle	Hashim et al. 2014

Table 1 (continued)

Plant specie name	Family	Abbreviations	Local name	Uses	Ethnobotanical uses	References
<i>Tagetes minuta</i>	Asteraceae	<i>T. minuta</i>	Hamesha	Medicinal	strength, sexual potency, and in treatments of urinary infections, heart diseases, and cough. Fresh leaf paste is applied on wounds twice a day for 3–4 days to kill germs in wounds.	Abbasi et al. 2010a, b
<i>Verbiscum thapsus</i>	Scrophulariaceae	<i>V. thapsus</i>	Khardag	Medicinal	Cough, pulmonary diseases, bleeding of bowels, and other skin diseases	Haq et al. 2011
<i>Cynodon dactylon</i>	Poaceae	<i>C. dactylon</i>	Kabal	Medicinal	Fresh plant paste is applied on bleeding wounds twice a day. Decoction as blood purifier	Abbasi et al. 2010a, b
<i>Bromus japonicus</i>	Poaceae	<i>B. japonicus</i>	Jawkey	Medicinal	Young shoots are used for diarrhea. Also used for broom making	Islam et al. 2006
<i>Indigofera heterantha</i>	Papilionaceae	<i>I. heterantha</i>	Ghoreja	Medicinal	Dried powdered root taken with glass of water against scabies; leaves against stomach problems	Sher et al. 2010
<i>Convolvulus arvensis</i>	Convolvulaceae	<i>C. arvensis</i>	Prewatkey	Medicinal	Purgative, also applied in skin disorders	Akhtar et al. 2013
<i>Dedonia viscosa</i>	Sapindaceae	<i>D. viscosa</i>	Ghwaraskey	Medicinal	Ash is used to treat burns and skin infections. Water extracts of leaves is used as antihelminthic.	Ahmad et al. 2011
<i>Dichantum annulatum</i>	Poaceae	<i>D. annulatum</i>	Wakha	Fodder	Fodder for cattles	Present study
<i>Heteropogon contortus</i>	Poaceae	<i>H. contortus</i>	Soormal wakha	Fodder	Fodder for cattles	Present study
<i>Cenchrus ciliaris</i> L.	Poaceae	<i>C. ciliaris</i>	Barwaz, Wakha	Fodder	Fodder for cattles	Present study
<i>Sacharum griffithi</i>	Poaceae	<i>S. griffithi</i>	Bogara	Fodder	Fodder for cattles	Present study
<i>Apluda mutica</i> L.	Poaceae	<i>A. mutica</i>	Wakha	Fodder	Fodder for cattles	Present study
<i>Aristida cyanantha</i>	Poaceae	<i>A. cyanantha</i>	Mashkeeza	Fodder	Fresh and dry fodder for cattles	Present study

$$CF = \frac{Cm \text{ Sample}}{Cm \text{ Background}}$$

where *Cm* represents the concentrations of metals in contaminated and background sites, respectively

Pollution load index

In this study, the pollution load index (PLI) was calculated by the following formula adopted from Tomlinson et al. (1980).

$$PLI = n\sqrt{Cf1 \times Cf2 \times Cf3 \dots \dots \dots Cf n}$$

where *n* is the number of metals and *Cf* is the contamination factor value.

Bioaccumulation factors

Heavy metal concentrations were calculated in the extracts of roots, shoots, and soils on dry weight (d.w) basis. Root concentration factors (RCFs) and shoot concentration factors (SCFs) are often used for contaminant concentrations in plants because heavy metal accumulation by plants is one of the main source for pollutants to enter in to food chain. RCFs and SCFs were calculated according to formula adopted from (Khan and Cao 2012).

$$RCF = C_{\text{root}}/C_{\text{soil}}$$

$$SCF = C_{\text{shoot}}/C_{\text{soil}}$$

where C_{root} , C_{shoot} , and C_{soil} represent heavy metal concentrations in root, shoot, and soil on dry d.w, respectively.

Translocation factor

Translocation factor (TF) is a ratio, calculated to determine relative translocation of metals from root to shoot of the plant species (Gupta et al. 2008).

$$TF = \frac{C(\text{Shoots})}{C(\text{Roots})}$$

where C is the contaminant concentrations in shoot and root samples of the selected plant species.

Statistical analyses

The data was statistically analyzed using the one-way analysis of variance (ANOVA) and Pearson correlation provided in SPSS (version 21). Mean and standard deviation were calculated using Statistix (version 8.1)

Results and discussion

Soil contamination

Table 2 summarizes the physicochemical properties of the soil samples collected from Shangla District, Mohmand Agency, Malakand Agency, and the reference sites. The soil pH values ranged from 5.5 to 8.2, EC ranged from 0.014 to 1.3 mS cm^{-1} , and the SOMs ranged from 1.75 to 3.8 %. Almost all the soil properties

varied in the study area except the soil class that was loamy sand. This variation in the soil physical properties may be due to the different environmental conditions (rainfall, altitude, and climate) of the study area.

Figure 2a represents the Pb and Cd concentrations in the soil samples collected from different locations. Pb concentrations in the study area ranged from 7 to 82 mg kg^{-1} and the Cd from 0.5 to 8 mg kg^{-1} . Highest mean Pb concentration (49.9 mg kg^{-1}) was reported in Malakand Agency, while the lowest (34.2 mg kg^{-1}) in Shangla. Highest Cd mean concentration was found in Malakand Agency (4.28 mg kg^{-1}), while the lowest in Shangla (3.39 mg kg^{-1}). The Pb and Cd concentrations in the soil samples of Shangla District, and Mohmand and Malakand agencies were significantly higher ($P \leq 0.001$) than the reference soil. However, the results showed that Pb concentrations were below the permissible limit (350 mg kg^{-1}) set for agriculture soils by (SEPA 1995), while Cd concentrations were observed multifold higher than the permissible limit (0.6 mg kg^{-1}) as shown in Table 3.

Contamination factor and pollution load index

Figure 2b summarizes the values of Pb and Cd CF and PLI. The PLI values were >2 in the soil samples of Shangla District and Mohmand and Malakand agencies. The CF and PLI values for Pb and Cd were in order of Malakand Agency $>$ Mohmand Agency $>$ Shangla District. The PLI showed that the soil of MAS2 site was highly contaminated as compared to MAS1 and MAS3. This high contamination could be due to the bed rocks, mining activities, and fossil fuel combustion in the study area.

Table 2 Selected physiochemical properties of soils collected from the study area

Locations	pH (1:5)	Electrical conductivity (ms/cm) (1:5)	Soil organic matter (%)	Soil texture			Class
				Clay (%) (<0.02 mm)	Silt (%) (0.02–0.05 mm)	Sand (%) (0.05–2 mm)	
MAS1	5.5–7	0.1–1.3	1.8–2.5				
	6.3±0.6	0.5±0.5	2.1±0.3	18	16	65.2	Loamy sand
MAS2	7–8.2	0.09–0.17	3.0–3.8				
	7.6±0.47	0.13±0.03	3.3±0.3	6.8	36	57.2	Loamy sand
MAS3	6.5–7.8	0.05–0.11	1.5–2.1				
	7±0.41	0.08±0.02	1.8±0.27	10.8	12	77.2	Loamy sand
Reference site	6.8	0.014	1.75	4.8	28	67.2	Loamy sand

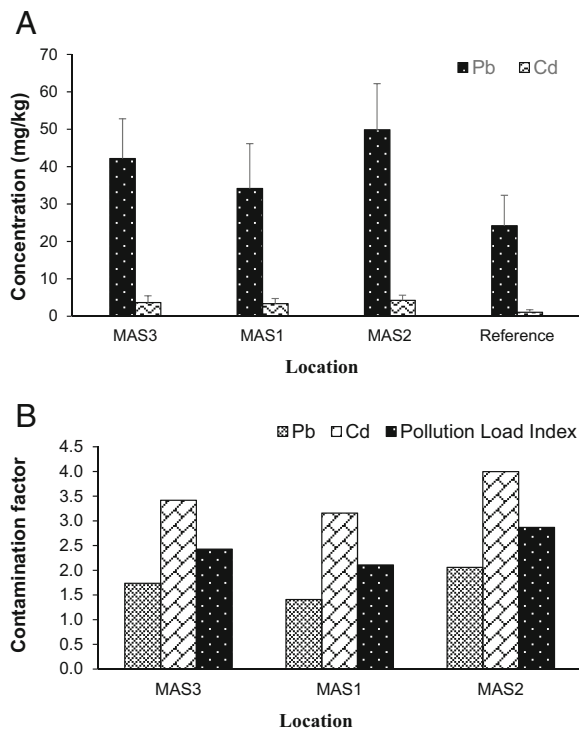


Fig. 2 Pb and Cd concentrations **a** in soil samples and **b** mean values of CF and PLI of Pb and Cd in the soil of the study area

Pb and Cd concentration in plant species

Medicinal plant species

Table 3 represents the concentration of Pb and Cd in plant roots and shoots collected from the study area. The concentration of Pb in the roots of the plant species ranged from below detection limit (BDL) to 45 mg kg⁻¹. The highest mean concentration (39.7 mg kg⁻¹) was reported in *Zanthoxylum armatum* and the lowest (BDL) in *Solanum xanthocarpum*. The concentration of Pb in the shoots of the plant species ranged from BDL to 28.4 mg kg⁻¹. The highest mean concentration (20.6 mg kg⁻¹) was reported in *Z. armatum* and lowest (BDL) in *S. xanthocarpum* and *Pantago lenceolata*. The concentrations of Pb in 50 % medicinal plant samples exceeded safe limits (10 mg kg⁻¹) set by WHO (2007). Pb can cause various health risks like anemia, neurological disorders, hyperactivity, and enzyme changes in humans (Marsden 2003). In this study, the concentrations of Pb in medicinal plants were higher than those reported by Baye and Hymete (2010) in medicinal plants collected from environmentally different sites in Ethiopia.

The concentrations of Cd in the roots of the selected medicinal plant species ranged from 0.07 to 6 mg kg⁻¹ with highest mean concentration (4.63 mg kg⁻¹) reported in *Razia stracta* and lowest (0.3 mg kg⁻¹) in *Tribulus terrestris*. The concentrations of Cd in the shoots ranged from 0.08 to 7.4 mg kg⁻¹, with the highest mean concentration (5 mg kg⁻¹) in *R. stracta* and lowest (0.2 mg kg⁻¹) in *T. terrestris*. Cd toxicity affect plant cells, even at low concentrations (Dai et al. 2012). High concentration of Cd is toxic to plants for a number of reasons, which inhibit germination, chlorosis of leaf, and plant biomass reduction (Wang et al. 2008). In 100 % plant samples, the Cd concentration exceeded the safe limits (0.3 mg kg⁻¹) set by WHO (2007). Cd accumulates in the human body and has a long biological life, which affects the lungs, kidneys, liver, bone, cardiovascular system, and immune system (Dai et al. 2011).

Almost all the medicinal plant species showed detectable concentrations of Pb and Cd in samples. The one-way ANOVA result showed that the Pb and Cd concentrations in roots and shoots of the selected plant species were significantly ($P < 0.001$) higher than the reference plant species. Considering the MAL of Pb (10 mg kg⁻¹) and Cd (0.3 mg kg⁻¹) set by WHO (2007) for medicinal herb, majority of the plant species exceeded these limits and showed high enrichment for the selected metals. The concentrations of Cd and Pb in medicinal plants also exceeded from the study conducted by Ebrahim et al. (2012) in Sudan. The heavy metal concentrations were within the permissible limit set by WHO (2007). Similar study was also conducted by Kulhari et al. (2013) in environmentally diverse locations in Northwest India. They also found Cd and Pb within the permissible limit set by WHO (2007). High concentration of heavy metals may pose serious problem on quality of medicinal plants and their products (Baye and Hymete 2010, 2013). To improve quality standards for herbal medicines, it is necessary to examine the concentrations of toxic heavy metals in medicinal plants (Cao et al. 2009). The present results indicate that there is a serious potential health risk associated with Cd and Pb in medicinal plants that threatens inhabitants of the study area as majority of the local people uses these species for various medicinal purposes.

Fodder plant species

Table 4 represents the concentrations of Pb and Cd in fodder plant species collected from the study area. The

Table 3 The concentrations (mg kg⁻¹) of Cd and Pb soil, medicinal, and fodder plants from the study area

Plant species	Cd																	
	Pb			Soil			Shoot			Root			Shoot					
	Range	Mean ± SD	Ref	Range	Mean ± SD	Ref	Range	Mean ± SD	Ref	Range	Mean ± SD	Ref	Range	Mean ± SD	Ref			
<i>A. javanica</i> (n=3)	42-62.2	52.13±10.1	21	21-30	25.66±4.5	5	7.0-16	11.66±4.42	2	3.0-6	4.52±1.52	2	3.0-5.8	4.31±1.41	1.5	2.0-4	3±1	1.3
<i>S. suratense</i> (n=4)	37-58	47.97±10.51	18	10.0-29.2	18.79±10.58	8	2.0-7.6	5.53±3.07	5	0.5-2	1.16±0.76	0.4	0.9-2.8	1.63±1.39	0.2	1.5-7.4	4.96±3.08	0.6
<i>C. javanica</i> (n=3)	30-42.8	38.66±5.39	29	16.0-26	21±5	3.8	9.0-13	10.41±2	3.38	2.5-2	3.71±1.62	0.7	3.5-5.2	4.56±1.21	0.1	1.0-5	2.82±1.82	0.7
<i>C. procera</i> (n=4)	45-73	60.33±14.18	20	24-35	29.66±5.5	13.5	5.0-20	12.76±7.39	9	1.0-3.8	2.53±1.41	1.1	0.5-3	2.16±1.44	1	2.0-6	4.6±2.25	1.9
<i>A. scoparia</i> (n=3)	34-54.7	44.46±10.35	25	4.6-22	12.06±8.95	6	6.0-18	12.66±6.11	4	3.5-5.9	4.76±1.2	2	2.0-3	2.66±0.57	1	3.0-4	3.33±0.55	0.48
<i>R. stracta</i> (n=5)	28-49	41.23±9.62	18	12.4-28	18.93±8.1	9	5.0-11	8.32±2.43	5	3.0-8	6±2.64	0.5	3.5-6	4.63±1.05	0.3	4.0-6	5±1	0.5
<i>L. corniculatus</i> (n=3)	37-58	47±10	14	4.0-13	8±4.5	2	2.5-8.8	4.86±3.52	0.8	1.5-3.5	2.5±1	1	1.0-3	2±1	0.1	1.5-3.5	2.5±1.05	0.3
<i>S. xanthocarpum</i> (n=4)	24-44	31±10.85	10	BDL	BDL	0.004	BDL	BDL	BDL	2.8-5.6	4.46±1.52	2	1.0-2.4	1.8±0.72	0.12	1.0-2	1.5±0.5	0.1
<i>S. iriolim</i> (n=5)	7.0-37.8	24.93±16.01	13	3.0-9.2	5.46±3.28	2.8	2.0-3.4	2.93±0.8	1.5	0.65-3	1.98±1.2	0.4	1.0-2.7	2±1.3	0.15	1.0-3.6	2.51±1.36	0.14
<i>I. bicolor royle</i> (n=4)	18.4-58.4	34.26±21.24	32	5.0-17.6	8.73±7.78	5	1.5-6	4±2	2	2-3.5	2.83±0.76	1	2.0-3	2.4±0.46	0.5	1.0-2.2	1.73±0.64	0.3
<i>P. lanceolata</i> (n=3)	21-40	31.93±9.81	37	0.6-3.7	2.43±1.62	8.5	BDL	BDL	1.2	3.0-5	4.16±1.04	1.4	1.0-4.2	2.4±1.63	0.8	0.1-2	1.2±0.98	0.43
<i>N. cataria</i> (n=4)	19-28	24.27±5.02	30	11.0-14	12.33±1.52	18	4.0-6	5.33±1.15	12	1.0-4	2.66±1.52	0.7	1.0-3.4	2.26±1.20	0.3	1.0-3.1	2.06±1.05	0.15
<i>R. hastatus</i> (n=3)	31-46.7	39.4±7.92	28	17-28	22.66±5.5	10	10.0-18	13.66±4.04	5	2.0-4	3.1±1.01	0.8	1.0-4	2.66±1.52	0.3	1.0-2.8	1.93±0.9	0.2
<i>I. roguses</i> (n=3)	26-56.5	38.07±16.2	35	11.0-15	13±2	12	7.0-8	7.33±0.57	7	2.5-5	4.09±1.38	1	2.0-4	3.12±1	0.6	1.8-3.9	2.91±1.02	0.5
<i>A. spinosus</i> (n=3)	40.5-60	50.8±10	26	10.0-21	15.66±5.5	8	8.0-15	11.21±3.51	5	3.0-5	4.1±1.02	BDL	2.0-3	2.33±0.57	BDL	2.0-2.5	2.16±0.28	BDL
<i>A. viridis</i> (n=3)	23-55.4	42.13±16.97	29	19-30	25.42±6.34	11	12.0-21	15.37±5.87	8	2.0-3.4	2.46±0.8	0.09	1.0-3	1.81±1.05	0.3	1.5-2.2	2.06±0.23	0.4
<i>A. bracteosa</i> (n=4)	21-32	28.21±6.35	41	0.8-4	2.26±1.61	4	0.1-3	1.10±1.64	2	1.5-4	3.29±1.04	2	0.8-2	1.2±0.69	0.8	0.5-2.2	1.64±0.98	1
<i>Z. armatum</i> (n=3)	38-68.7	52.13±15.24	18	32-45	39.66±6.8	5	15-28.4	20.56±7.05	3	2.9-4	3.3±0.6	2	2.0-5.6	4.06±2.3	1	2.8-5	4.2±1.21	1.3
<i>T. terrestris</i> (n=3)	20-43	29.31±11.79	25	5.0-17	11.54±5.31	6	3.0-5	3.90±1.07	2	1.0-2.7	1.88±0.79	1	0.8-0.5	0.3±0.16	0.08	0.1-0.4	0.2±0.1	0.13
<i>T. minuta</i> (n=4)	30-51	40.66±10.5	11	6.0-15	10.33±4.5	3	3.0-8	5±2.64	1	3.0-5.1	4.06±1.05	1	3.0-4.6	3.73±0.8	0.6	2.0-3	2.33±0.57	0.87
<i>V. thapsus</i> (n=4)	48-68.3	58.2±10.15	23	15-26	20.8±5.61	5	8.0-15	11.26±3.52	2	4.0-7	5.66±1.52	1	2.0-4.6	3.41±1.34	0.72	3.0-6.4	4.93±1.74	0.98
<i>C. dactylon</i> (n=3)	21-45	35.46±13.48	16	14-27	21±6.55	4	10.0-18	14.66±4.16	2.2	4.0-6.8	5.53±1.41	0.2	0.28-1	0.7±0.37	0.08	0.15-0.67	0.38±0.25	0.06
<i>B. japonicus</i> (n=4)	50-82	68±16.37	19	16.5-32	25.63±8.15	7	6-23.6	15.2±8.82	3	2.2-3.6	2.93±0.70	0.5	0.9-2	1.3±0.6	0.25	0.5-1.62	1.10±0.56	0.14
<i>I. heterantha</i> (n=3)	24-41	33.56±8.69	30	10.0-25	18.33±7.63	8	4.0-15	9.73±5.50	3	1.0-3	2.12±0.85	2	0.32-1	0.60±0.35	1.5	0.08-1	0.49±0.46	0.8
<i>C. arvensis</i> (n=5)	28-47	40.2±10.15	27	5.0-8	6.66±1.52	BDL	2.0-4	3±1	BDL	3.2-6.1	4.8±1.47	0.9	3.0-5	3.82±1	0.5	2.0-4	3±1	0.3
<i>D. viscosa</i> (n=3)	16-35	24.98±9.50	22	7.0-13	9.66±3.05	7	2.0-7	4.21±2.54	3	1.0-5	3.11±1.94	2	0.7-3	1.78±1.15	0.6	1.0-2.4	1.93±0.80	0.51
WHO/FAO (2007) limits					10													
herbal plants																		
Fodder plants																		
<i>D. annulatum</i> (n=3)	28.5-35	30.83±3.61	34	5.0-11	7.33±3.21	6	1.0-5	2.66±2.08	2	1.0-3	2±1	0.1	1.2-3	2.3±0.90	0.1	1.5-3	2.26±0.86	0.2

Table 3 (continued)

Plant species	Pb						Cd											
	Soil			Shoot			Root			Soil			Shoot			Root		
	Range	Mean ± SD	Ref	Range	Mean ± SD	Ref	Range	Mean ± SD	Ref	Range	Mean ± SD	Ref	Range	Mean ± SD	Ref	Range	Mean ± SD	Ref
<i>H. contortus</i> (n=4)	20–40	31.66±10.40	28.5	7.0–15	10.26±5.02	8	1.6–7	4.25±2.27	2.28	2.0–4	2.93±1.06	2	1.8–3.4	2.73±0.83	1.3	2.0–4	3.33±1.15	1.51
<i>C. ciliaris</i> (n=3)	32–53	41.32±10.67	20	3.0–8	4.79±3.3	3	2.0–4.5	3.12±1.33	1.64	3.0–5.5	4.34±1.02	1	1.1–2.5	1.76±0.88	0.5	1.0–2	1.41±0.68	1
<i>S. griffithii</i> (n=3)	31–51.2	41.1±10.1	18	6.0–17	11.6±5.5	5	4.0–11	7±3.6	2.6	0.9–2.8	1.83±0.95	1.4	2.0–4	3±1	0.005	3.0–4.2	3.53±0.61	0.6
<i>A. mutica</i> (n=4)	38–56.2	46.8±9.11	22	7.0–12	9.33±2.51	4	3.0–5	4.33±1.15	2	4.0–6	5±1	2	3.0–5.9	4.43±1.45	1.1	4.0–7	5.66±1.53	1.2
<i>A. cyanantha</i> (n=5)	36–52	42.96±8.13	19	2.0–6	3.66±2.08	BDL	1.0–3	2.0±1	BDL	2.3–4.4	3.53±1.09	1.8	3.2–5.2	3.96±1.07	0.8	2.0–4	2.66±1.15	1
Sig		0.001			0.001			0.001		0.001				0.001				0.001
SEPA limits		350																
WHO/FAO (2007) limits edible plants					0.3													0.2

SD standard deviation, Ref reference value, BDL below deduction limit

Pb concentrations in roots ranged from 2 to 17 mg kg⁻¹, while in shoots ranged from 1 to 11 mg kg⁻¹. The highest mean concentration in root (11.6 mg kg⁻¹) and shoot (7 mg kg⁻¹) was found in *Sonneratia griffithii*, while the lowest in root (3.66 mg kg⁻¹) and shoot (2 mg kg⁻¹) of *Aristida cyanantha*, respectively. Cd concentrations in roots ranged from 1.1 to 5.9 mg kg⁻¹, while in shoots, it ranged from 1 to 7 mg kg⁻¹. The highest mean concentrations in root (4.43 mg kg⁻¹) and shoot (5.66 mg kg⁻¹) was reported in *Apluda mutica*, and lowest was found in root (1.76 mg kg⁻¹) and shoot (1.41 mg kg⁻¹) of *Cenchrus ciliaris* (Table 3). Almost all fodder species exceeded the safe limit of WHO (2007) for Pb (0.31 mg kg⁻¹) and Cd (0.2 mg kg⁻¹) in edible plants. Pb is highly toxic to animals at a level of 30 mg kg⁻¹ in diet on a d.w basis (FAO 1992) and leads to different problems in red blood cell synthesis (NRCC 1978). The Pb and Cd concentrations in fodder plant species were significantly (P<0.001) higher than the reference site. Previously, Pb contamination has been reported from natural and anthropogenic sources (Rozso et al. 2003). Pb concentrations in fodder plants grown in agriculture areas, industrial area, and busy highways showed that Pb contamination was higher in industrial and highway sites as compared to agriculture sites. Mor (2005) also reported high Pb concentrations in fodder plants grown in agriculture areas of Busra, Turkey. Cd rapidly accumulates in kidney and other organs of animals, therefore, can create potential hazards for human health after consumption (Canadian Council of Resource and Environment Ministers (CCREM), 1995). The United State Environmental Protection Agency (USEPA) reported that the continuous intake of Pb in diet from 2 to 8 mg kg⁻¹ of body weight day⁻¹ can cause death in most animals (USEPA 1993). Our present study highlights that Pb and Cd concentrations in majority of fodder grasses were above the threshold level (10 and 0.3 mg kg⁻¹, respectively) in mining areas. Such high concentrations of these metals may pose health problem for grazing animals (cows, sheeps, and goats) and local community in the study area.

Contamination and translocation factors of Pb and Cd in medicinal and fodder plant species

Table 4 represents the CF and TF values for plant species in MAS1, MAS2, and MAS3 sites. The CF value for Pb and Cd highly varied among the selected

Table 4 The values of RCF, SCF, TF, and CF in plant species collected from the study area

Plant species	Pb				Cd				Plant species	Pb				Cd			
	RCF	SCF	TF	CF	RCF	SCF	TF	CF		RCF	SCF	TF	CF	RCF	SCF	TF	CF
<i>A. javanica</i>	0.5	0.22	0.5	6	1	0.66	0.7	2.3	<i>A. bracteosa</i>	0.1	0.03	0.5	0.6	0.4	0.49	1.36	1.6
<i>S. surattense</i>	0.4	0.1	0.3	1	1.4	4.27	3.04	8.2	<i>D. viscosa</i>	0.4	0.16	0.4	1.4	0.6	0.62	1.08	3.8
<i>C. jwarancusa</i>	0.5	0.26	0.5	3	1.2	0.76	0.62	4	<i>T. terrestris</i>	0.4	0.13	0.3	2	0.2	0.1	0.67	1.5
<i>C. procera</i>	0.5	0.21	0.4	1	0.9	1.81	2.13	2.4	<i>T. minuta</i>	0.3	0.12	0.5	5	0.9	0.57	0.62	2.7
<i>A. scoparia</i>	0.3	0.28	1	3	0.6	0.69	1.25	6.9	<i>V. thapsus</i>	0.4	0.19	0.5	5.6	0.6	0.87	1.45	5
<i>R. stracta</i>	0.5	0.2	0.4	2	0.8	0.83	1.08	10	<i>C. dactylon</i>	0.6	0.41	0.7	6.7	0.1	0.06	0.54	6.3
<i>L. corniculatus</i>	0.2	0.1	0.6	6	0.8	1	1.25	8.3	<i>B. japonicus</i>	0.4	0.22	0.6	5.1	0.4	0.37	0.85	7.9
<i>S. xanthocarpum</i>	0	0	0	0	0.4	0.33	0.83	15	<i>I. heterantha</i>	0.5	0.28	0.5	3.2	0.3	0.23	0.82	0.6
<i>S. irio linn</i>	0.2	0.11	0.5	2	1	1.27	1.27	18	<i>C. arvense</i>	0.2	0.07	0.5	3	0.8	0.62	0.79	10
<i>I. bicolor royle</i>	0.3	0.11	0.5	2	0.8	0.61	0.72	5.8	<i>X. armatum</i>	0.8	0.39	0.5	6.9	1.4	1.27	0.9	3.2
<i>P. lanceolata</i>	0.1	0	0	0	0.6	0.28	0.5	2.8	<i>D. annulatum</i>	0.2	0.08	0.4	1.3	1.2	1.13	0.98	11
<i>N. cataria</i>	0.5	0.21	0.4	0.4	0.8	0.77	0.91	14	<i>H. contortus</i>	0.3	0.13	0.4	1.9	0.9	1.21	1.22	2.2
<i>R. hastatus</i>	0.6	0.34	0.6	3	0.9	0.62	0.73	9.7	<i>C. ciliaris</i>	0.1	0.07	0.7	1.9	0.4	0.32	0.8	1.4
<i>I. roguses</i>	0.3	0.19	0.6	1	0.8	0.71	0.95	5.9	<i>S. griffithi</i>	0.3	0.17	0.6	2.7	1.6	1.92	1.18	5.9
<i>A. spinosus</i>	0.3	0.22	0.7	2	0.6	0.52	0.93	2.1	<i>A. mutica</i>	0.2	0.09	0.5	2.2	0.9	1.13	1.28	4.7
<i>A. viridis</i>	0.6	0.36	0.6	2	0.7	0.83	1.14	6.5	<i>A. cyanantha</i>	0.1	0.02	0.5	2	1.1	0.75	0.67	2.7

RCF root concentration factor, SCF shoot concentration factor, TF transfer factor, CF contamination factor

plant species. Highest CF value ($\times 6.9$) for Pb was reported in *Z. armatum* and lowest (BDL) in medicinal plants such as *S. xanthocarpum* and *P. lanceolata*. The highest CF ($\times 18$) for Cd was reported in medicinal plant (*Sisymbrium irio*), while the lowest ($\times 0.6$) was reported in medicinal (*Indigofera heterantha*). The CF values of Pb and Cd were reported greater than 1 in 88 and 97 % plant species, respectively.

The TF indicates internal metal transportation from root to shoots. The present study revealed that Pb accumulated by the medicinal herbs and fodder plants growing near chromite mines was largely retained in roots that have TF values < 1 (Table 4). The highest TF value (1.00) for Pb was reported in medicinal plant (*Artemisia scoparia*), while the lowest (BDL) in *S. xanthocarpum* and *P. lanceolata*.

The TF values for Cd were greatly varied among the selected medicinal and fodder plant species. Highest TF value (3.04) for Cd was reported in *Solanum surattense*, while lowest (0.5) in *P. lanceolata*. Out of 32 plant species, 13 plants showed TF values > 1 , while 19 plant species showed TF value < 1 . Unlike medicinal plants, all the fodder plant species showed CF and TF values at medium level for both Pb and Cd.

Bioaccumulation factor of Pb and Cd in medicinal and fodder plant species

Bioaccumulation factor for Pb and Cd was calculated for roots and shoots (Table 4). The RCF values for Pb in the study area ranged from BDL to 0.8. The highest concentration was reported in medicinal plant *Z. armatum* and the lowest in *S. xanthocarpum*. No plant species in the study area showed RCF value greater than 1 for Pb. Similarly, the RCF values for Cd ranged from 0.1 to 1.6 with the highest value reported in fodder plant *S. griffithii* and lowest in medicinal plant *Cynodon dactylon*. The SCF values for Pb ranged from BDL to 0.41, with highest value in medicinal plant *C. dactylon* and lowest in medicinal plant *S. xanthocarpum* and *P. lanceolata*. In almost all plant species, SCF values for Pb were found less than 1. The SCF values for Cd ranged from 0.06 to 4.27 with the highest SCF value in medicinal plant *S. surattense* and lowest in *C. dactylon*. The medicinal plant species showed SCF value greater than 1 for Cd in *S. surattense* (4.27), *Calotropis procera* (1.81), *Lotus corniculatus* (1), *S. irio* (1.27), and *Z. armatum* (1.27) and the fodder plant species such as *Dichanthum*

annulatum (1.13), *Heteropogon contortus* (1.21), *S. griffithii* (1.92), and *A. mutica* (1.13). These plant species are generally used for medicinal and fodder purposes in the study area (Table 1). Therefore, such high concentrations in the shoots of these plant species can create health problems in the study area for both human and animals.

Pearson's correlation

Pearson's correlation was used to correlate the concentrations of Pb and Cd in the (soil–plant roots) and (soil–plant shoots) samples of the study area. The correlation matrix for Pb between soil and plant roots was strong ($r=0.610$), followed by correlation between soil and plant shoots ($r=0.593$). Similarly, the correlation matrix for Cd between soil and plant roots was slightly weak ($r=0.458$) than the correlation between soil and plant shoots ($r=0.52$). The results indicate that these metals originate from the same polluting source. The significant correlation coefficients $P<0.01$ between Pb and Cd (soil–plant roots) and (soil–plant shoots) can be attributed to anthropogenic (chromite mining and fossil fuel combustion) activities in the study area.

Conclusion

The current research work has shown high levels of Pb and Cd contamination in the soil, medicinal, and fodder plants species along MAS1, MAS2, and MAS3 in Northern Pakistan. The results clearly showed that PLI values for Pb and Cd in soil of study area were greater than 2. MAS2 was highly contaminated as compared to MAS3 and MAS1. The concentrations of Pb and Cd in soil, medicinal, and fodder plant species surrounding the mine areas were significantly ($P<0.001$) higher than the reference site and showed multifold enrichment. Cd concentrations exceeded its MAL in soil, while Pb was observed within its MAL. The Pb and Cd concentrations in the majority of medicinal and fodder plants exceeded the safe limits set by FAO/WHO for herbal and edible plants of the study area. Such high contamination with toxic metals may be a threat to the local community; therefore, this study suggests that both medicinal and fodder plant species should be analyzed for heavy metal concentrations in the contaminated sites before utilization.

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Conflict of interest The authors declare that they have no conflict of interest.

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