



Assessment of potentially toxic metal(loid)s contamination in soil near the industrial landfill and impact on human health: an evaluation of risk

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Abstract The generation of solid waste is increasing with each passing day due to rapid urbanization and industrialization and has become a matter of concern for the international community. Leachate leakages from landfills pollute the soil and can potentially harm the human health. In this paper, inductively coupled plasma-optical emission spectrometric studies were employed to assess and analyze the

composition of metals (Ba, Cd, Pb, Hg, Cu, Cr and Mn) and metalloid (As) in soil samples. Results of Cr, Mn, Cu, As, Ba, Cd, Pb and Hg from CRM (certified reference material, SRM 2709a) of San Joaquin soil were evaluated and reported in terms of percent recoveries which were in the range of 97.6–102.9% and show outstanding extraction efficiency. Other than copper, where the permitted limit set by the EU is specified as 50–140 mg/kg in soil, the average amount of all the metals in soil was found within the permissible limits provided by WHO, the European Community (EU) and US EPA. Soil contaminated with Hg (PERI=100) and Cd (PERI=145.50) posed an ecological risk significantly. Pollution load index (PLI) value is greater than 1, while degree of contamination (C_{deg}) value is less than 32 which indicated that the soil is polluted and considerably contaminated with metals and metalloid, respectively. In terms of the average daily dosage (ADD) of soil, children received the highest doses of all metals ($ADD_{ing} = 1.315 \times 10^{-7} - 2.470 \times 10^{-3}$ and $ADD_{derm} = 9.939 \times 10^{-7} - 5.292 \times 10^{-11}$), whereas ADD_{ing} ($1.409 \times 10^{-8} - 2.646 \times 10^{-4}$) was found greater in adults. For all metals except for Ba, the hazard quotient (HQ) trend in both children and adults was observed to be $HQ_{ing} > HQ_{derm} > HQ_{inh}$ of soil. Children who are at the lower edge of cancer risk had a lifetime cancer risk (LCR) of 2.039×10^{-4} for Cr from various paths of soil exposure.

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Introduction

The solid waste is growing on daily basis due to anthropogenic activities all over the world despite widespread information. Such a rapid growth of solid waste can potentially harm the environment and human health and thus has become a global concern (Naveen et al., 2017). The world community is putting a lot of efforts in the waste management (Kassim, 2012). Landfilling is a traditional form of waste management and is popular among masses due to its convenience, economic advantages and less technological hurdles (Adamcová et al., 2017; Gonzalez-Valencia et al., 2016; Samadder et al., 2017). Water contents in soil produce leachate from landfills under favorable environmental conditions that can ultimately enter into the aquatic environment and the soil (Edokpayi et al., 2018; Grugnaletti et al., 2016). Landfill leachate contains a number of pollutants such as suspended particles (inorganic and organic), metalloids and metals including heavy metals (HMs) and toxic metals (TMs). The leachate contents can have detrimental effects on both terrestrial and aquatic ecosystems and the general public's health (Boateng et al., 2019; Mavakala et al., 2016; Naveen et al., 2017). Ultimately, soil is the sink for metalloids, HMs and TMs, which is a prime component of the terrestrial ecosystem and functions as a conduit for their dispersion into the environment, living things and water bodies. Metalloids, HMs and TMs are accumulative and persistent and interact with both organic and inorganic debris in soil to make it more hazardous.

The mobility of various elements in the soil environment depends on a number of different properties of soil. In particular, the metals distribution in the solid and liquid phases is determined by the nature of metals, pH of the soil, their loading rate, soil structure, clay and organic contents, and the redox potential. Above properties cause chemical combinations of dissimilar degrees of solubility, and thus, the bioavailability of metals varies with the soil colloids. Metals are present in the soil solution as single ions or ion pairs, as chelated with organic compounds, sorbed onto colloids as exchangeable ions, as co-precipitated with or occluded into oxides, phosphates, carbonates

and secondary minerals, and in combinations with crystal lattices of primary minerals to account for residual fractions. In addition to causing alteration in the agricultural soil composition, structure and function, metals contamination also inhibits growth of the crop root and thus reduces the crop yields (Alloway, 2012; Kabata-Pendias, 2000).

As a result, these metals can build up in the chain of food and through dietary intake can ultimately enter the human body. Direct ingestion, cutaneous contact absorption and inhalation are some of the other routes through which the mentioned metals can also enter the human body (Mohammadi et al., 2019; Wu et al., 2020). Two main routes of exposure to hazardous metals in human body are drinking water and breathing in soil particles (Alexander et al., 2006; Zhu et al., 2011). Therefore, the issues related to the soil metal pollution are gaining much importance and hence the focus of scientists. The humans are exposed to these hazardous metals through 3 main routes: inhalation, ingestion and skin contact. Thus, they pose a serious threat to the human health. Hence, it is imperative and critically important to monitor the soil pollution caused by the toxic metals because of their direct impact on the quality of environment and human health (Golia et al., 2019).

The effect of chromium on human health depends on its oxidation state. Chromium (III) is an essential component of the human food and plays a prime part in the metabolism, while chromium (VI) is extremely carcinogenic and causes liver and cardiovascular problems (Bahadır et al., 2014). Mn is a crucial component for the metabolism of amino acids, cholesterol and carbohydrates. However, higher amounts of Mn can cause neurological problems, infertility in males, birth malformations and defects in bones (Santamaria, 2008). Copper plays a key part in the physiological functions of the human bodies, yet consuming too much of it can be harmful to public health. The gut, liver and stomach are all negatively affected by excessive amounts of Cu (Gaetke & Chow, 2003; Harmanescu et al., 2011; Rahman et al., 2014), and its ingestion in greater amounts can cause vascular disease, angiosarcoma, skin cancer, cutaneous lesions and peripheral neuropathy (Hartley & Lepp, 2008; Jia et al., 2018). Likewise, excessive amount of Cd can harm the lungs, liver, bones, kidneys, pulmonary adenocarcinomas, the body's immune system and prostatic proliferative lesions (Klaassen et al., 2009;

Lin et al., 2013). Humans who consume Ba may experience heart rhythm abnormalities or paralysis, which may ultimately lead to death if prompt medical attention is not acquired. Brief exposures to Ba may result into abnormal blood pressure, breathing issues, diarrhea, muscle weakness and vomiting (Oskarsson, 2015).

Depletion of cellular antioxidants occurs as a result of formation of a covalent bond between Hg (II) and cysteine residue of protein. So, oxidative damage due to Hg gives rise to reactive oxygen species (ROS). Molecular mechanism of toxicity is described by oxidative damage. ROS plays a prime role in the cellular responses induced by metals causing a damage to the DNA which initiates the carcinogenic process (Tchounwou et al., 2012; Valko et al., 2006). A higher Pb amount in the blood can lead to hypertension, skeletal, endocrine and immune system damages, abnormal kidney and heart functioning, and lower IQ potential in children (Ekong et al., 2006; Navas-Acien et al., 2007; Nieboer et al., 2013). Thus, exposure to metal(loid)s (Mn, Cr, Cu, As, Ba, Cd, Pb and Hg) through soil can have an extremely detrimental effect on human health. Proximity to industrial dumps causes contamination of soil with TMs and HMs. Due to the persistence, toxicity and bioaccumulation of metal(loid)s, industrial dumps have become one of the most difficult environmental and health challenges.

In this context, the goal of current research is to analyze the composition of metal(loid)s in the soil by employing ICP-OES (inductively coupled plasma-optical emission spectroscopy) and to examine the levels of metal pollution in the soil close to the industrial dump sites in Gujranwala (an industrial region situated in the Punjab province of Pakistan).

The potential ecological and human health risks (carcinogenic and non-carcinogenic) of metals in soil samples are preferably evaluated through risk assessment models. The risk assessment of the discharge sources is considered as an important measurement of perniciousness index and is helpful to monitor the contamination sources. The PMF (positive matrix factorization) is a widely used receptor model to quantify the sources of accumulation of heavy metals in the soil. This model has an obvious advantage of non-negative constrain condition and weighs each species by using uncertainty. Hence, the models try to model abnormal data correctly to make sources to be

reliable physically (Cai et al., 2019; Jiang et al., 2020, 2021).

CF (contamination factor), I_{geo} (geo-accumulation index), C_{deg} (degree of contamination), PERI (potential ecological risk index) and pollution load index (PLI) were calculated to evaluate pollution, assess the overall contamination picture and determine the potential risk because of toxic concentrations of metalloid, HMs and TMs in soil and an exposure to ecological sensitivity. Background values are a common evaluation standard to assess the results of above-mentioned quantitative indices for pollution evaluation. The ADD (average daily dose), non-carcinogenic THQ (target hazard quotient), HI (hazard index) and lifetime CR (carcinogenic risk) coefficients were also calculated to assess the risk to human health because information on these metals is equally necessary to examine the possible risk to health of humans.

To our knowledge, there has never been a research done to estimate the levels of metal(loid)s in the soil close to an industrial waste site in Gujranwala. In order to determine the possible health impact of these metals on ecosystem as well as on human health, various regions close to the dump site were chosen for the research of the composition of metal(loid)s in soil. By calculating numerous contamination parameters, this study also attempted to furnish useful information to control pollution through scientific and systematic management of industrial projects related to human health risk and ecosystems.

Experimental

Study area

The current studies were performed in the industrial metropolitan of Gujranwala (located at 32° 9' 58.8636" N and 74° 11' 45.2400" E) in the Punjab province of Pakistan (Fig. 1). Gujranwala is well known for producing a variety of household goods, such as fans, sanitary wares and melamine and steel-made kitchenware. The selected area for this study was 3198 km². In 2019, the city's total population was 2,169,000 and the study area's mean annual temperature was 30.8 °C. In April, the temperature ranged from 18.4 to 33.7 °C, whereas in October, it ranged from 17.8 to 33.1 °C, with humidity levels averaging between 50 and 69%. Households in small

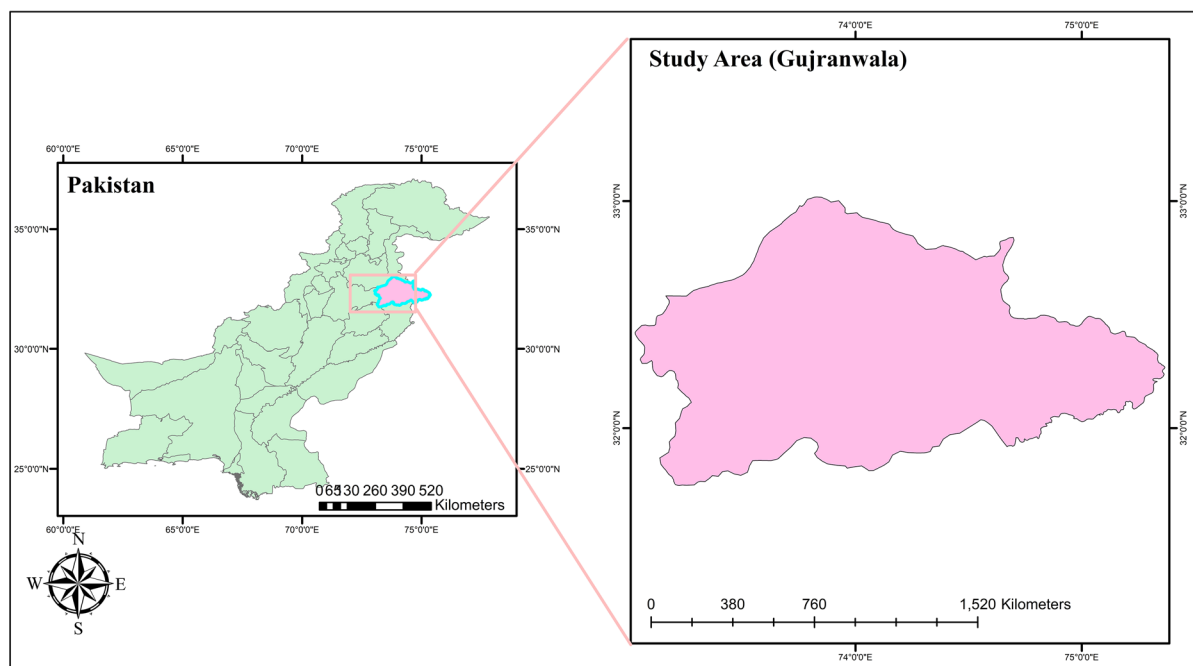


Fig. 1 Location of the study area

industries generate almost 50% of the waste, whereas rest of the waste comes from major industries. Industrial solid waste is dumped in vacant lots or yards; these landfills are not lined, but their bases are compacted with clay to reduce leachate seepage into the subsurface ecosystem.

Solutions and reagents

65% m/m HNO_3 (nitric acid) and 48% m/m HF (hydrofluoric acid) were procured from Lahore-Pakistan (Hajvery Scientific Store, originate to Merck, Germany). HNO_3 and HF were used for the solution preparation to digest soil. A number of dilutions with 2% v/v HNO_3 of the certified 1000 mg/L reference material of each element (Merck, Germany) were used to generate calibration curves. Ultrapure water having 18 M Ω .cm resistivity from GenPure water system (Thermo Scientific, USA) was employed to prepare different dilutions of the solutions. All the glassware and equipment used in the study was kept in 20% v/v HNO_3 overnight and then was thoroughly cleaned with ultrapure H_2O . San Joaquin's soil (SRM 2709a), the certified reference materials were employed as references.

Collection, preparation and analysis of soil samples

In order to collect soil samples for systematic analysis of soil, the study area was divided into 4 areas: region I (east), region II (west), region III (north) and region IV (south). Twenty samples were taken in total, five from each location having a 10 to 50 m distance (following a subsequent distance of 10 m) from the dump site (5 samples $\times$ 4 regions). A 0.5-cm top layer of the soil surface was erased prior to sampling, and each sample point was sampled at a depth of 0.5–20, 20–40 and 40–60 cm yielding approximately 500 g of material. Three subsamples from each sample point were combined and homogenized to generate the representative sample. The typical samples were collected and stored in polyethylene bags and were delivered to the laboratory later. The samples were coded as GJES1-GJES5, GJWS1-GJWS5, GJNS1-GJNS5 and GJSS1-GJSS5, respectively, and represent five samples from each region. The samples were crushed, sieved through a filter with a mesh size of no. 10, stored in polyethylene bottles for acid digestion and were then air-dried in an oven having air circulation (Vision Scientific, Korea) for 72 h at 35 °C. A typical sample of 1 g of soil was weighed and

moved into a porcelain crucible that contained ten milliliter 1:1 mixture of HNO₃ and HF already; then, it was completely digested. Whatman filter paper no. 41 was employed to eliminate any suspended particles by filtering the resultant solution. The solution was then moved to a measuring flask of 100 mL volume, and the volume was then increased to the mark with ultrapure water (Epa, 1996). ICP-OES (Thermo Fisher Scientific, iCAP 7400) was used to measure Mn, Cr, Cu, Cd, As, Ba, Pb and Hg in soil samples. The optimized operating settings are presented in Table 1S, and all measurements were taken in triplicate. To manage the system, Thermo iteva software was employed.

Quality assurance

For soil analysis, the precision of the acid digestion method was evaluated by repeating the procedure in replicates of 10 ($n=10$) under identical conditions, with the repeatability results demonstrated in percent RSD. The accuracy of the method was ensured by CRM analysis, and the recovery was calculated in percent (%). LOD=3 σ /S and LOQ=10 σ /S, respectively, were calculated as the detection limits. In these equations, S is the slope of the calibration curve ($y=mx+b$) and σ is the standard deviation of the analytical blank measurement ($n=18$) (Ahmad et al., 2021; Ahmed et al., 2016; Costa et al., 2020).

TMs and HMs contamination evaluation in soil

Researchers have developed the I_{geo} (geoaccumulation index), CF (contamination factor), PLI (pollution load index), PERI (potential ecological risk index) and C_{deg} (degree of contamination) coefficients to evaluate the contamination level of a soil. These parameters measure pollution, analyze the contamination pattern and assess the potential risk from exposure to the ecological sensitivity, toxicity and amount of HMs and TMs in soil. I_{geo} was determined using a formula given below in equation a (Mohammadi et al., 2019).

$$I_{geo} = \log_2 \left(\frac{C_n}{1.5B_n} \right) \quad (1)$$

where B_n is the unaffected soil's geochemical background value at the city site and C_n in the soil

sample is the element's concentration. We were able to reduce the effect of variation in background concentration caused by lithogenic influences by using the constant value equal to 1.5 (Chonokhuu et al., 2019). I_{geo} index categorization can be used to differentiate between different types of soil., $I_{geo} \leq 0$ (uncontaminated), $0 < I_{geo} < 1$ (between unpolluted and moderately polluted), $1 < I_{geo} < 2$ (moderately polluted), $2 < I_{geo} < 3$ (between moderately and heavily polluted), $3 < I_{geo} < 4$ (heavily polluted), $4 < I_{geo} < 5$ (between heavily and extremely polluted), $5 < I_{geo}$ (extremely polluted). An estimation of CF value was done by using following equation (Chen et al., 2022; Mohammadi et al., 2019).

$$CF = \frac{C_n}{C_b} \quad (2)$$

where C_b is value of geochemical background of unaffected soil at the city site and C_n in the soil sample is the element's concentration. By using the CF value, soil quality can be categorized as follows: $CF < 1$ (low contamination), $1 \leq CF < 3$ (moderate contamination), $3 \leq CF < 6$ (considerable contamination) and $CF \geq 6$ (very high contamination) (Ferreira et al., 2022; Mohammadi et al., 2019).

Equation (c) can be used to determine the potential ecological risk of each metal element (Hanfi & Yarmoshenko, 2020).

$$E_r^i = C_r^i \times T_r^i = \left(\frac{C_s^i}{C_n^i} \right) \times T_r^i \quad (3)$$

where C_s^i =concentration of an element in the soil sample, C_n^i =geochemical background value of non-affected soil, T_r^i =toxic response factor for each metal.

Toxic response factors were taken at 2, 5, 10, 20, 40 and 50 for Cr, Cu, Pb, As and Hg, respectively (Zhang & Liu, 2014). The quality of soil can be categorized by E_r value as follows: $E_r \geq 320$ (very high risk), $160 \leq E_r < 320$ (high risk), $80 \leq E_r < 160$ (considerable risk), $40 \leq E_r < 80$ (moderate risk) and $E_r < 40$ (low risk) (Ferreira et al., 2022).

PLI was determined using a formula

$$PLI = (CF_1 \times CF_2 \times \dots \times CF_n)^{\frac{1}{n}} \quad (4)$$

where CF_1 , CF_2 and CF_n are the factors of contamination of the elements 1, 2 and n, respectively. This index divides the soil into 3 different categories:

$PLI < 1$ (soil is not polluted), $PLI = 1$ (pollution of baseline levels) and $PLI > 1$ (soil is polluted) (Ferreira et al., 2022).

Following equation is used to calcite C_{deg} .

$$C_{deg} = \sum_{i=1}^n CF_i \quad (5)$$

For each investigated element (i), CF_i is the contamination factor in above equation. Soil is classified into four categories according to this index, i.e., $32 \leq C_{deg}$ (contamination of very high degree), $16 \leq C_{deg} \leq 32$ (contamination of considerable degree), $8 \leq C_{deg} \leq 16$ (contamination of moderate degree) and $C_{deg} < 8$ (contamination of low degree) (Ferreira et al., 2022).

Human health risk assessment

ADD (average daily dose), non-carcinogenic THQ (target hazard quotient), hazard index (HI) and CR (lifetime carcinogenic risk) coefficients were used to estimate the likelihood of human exposure to HMs and TMs in soil as well as in water (Boateng et al., 2019; Kamunda et al., 2016; Mohammadi et al., 2019).

Average daily dose

The method was employed to calculate the ADD of three soil exposure routes, which include ADD_{ing} (ingestion), ADD_{inh} (inhalation) and ADD_{derm} (dermal contact), for metals such as Cr, Cu, Mn, As, Cd, Pb, Hg and Ba.

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

$$ADD_{inh} = C \times \frac{InhR \times EF \times ED}{PEF \times BW \times AT}$$

$$ADD_{derm} = C \times \frac{SA \times AF \times ABF \times EF \times ED}{BW \times AT} \times 10^{-6}$$

where C represents the concentrations (in mg/kg) of TMs and HMs, $IngR$ represents the rate of ingestion in mg per day ($IngR = 100$ for adults and 200 for children), EF represents exposure frequency in days per year (180), ED represents the duration of exposure (24 years for adults & for children is 6 years), BW

represents the average weight of body (adult = 70 kg and child = 15 kg), AT represents the average time ($365 \times ED$), $InhR$ represents the rate of inhalation (1.36×10^9 for both adults and children), PEF is particle emission factor in $m^3 \text{ kg}^{-1}$ (1.36×10^9 for both children and adults), AF is the adherence factor of skin (in mg/cm^2) for the soil (0.2 for adults and 0.07 for children), SA is the exposed surface area of skin in cm^2 (2145 for adults and 1150 for children), and ABF is the dermal absorption factor (0.03 for arsenic and 0.001 for other metals) by using the formula (Dehghani et al., 2019; Hanfi & Yarmoshenko, 2020; Karimi et al., 2020; Mohammadi et al., 2019).

Total hazard quotient and index

THQ is a ratio that is commonly employed to quantify the non-carcinogenic risk potential to humans of exposure to metals through 3 separate pathways. The ratio of ADD (from three exposure pathways) and RfD is called THQ, whereas RfD is the chronic reference dose in mg/kg BW/day for each metal.

$$THQ = \frac{ADD(\text{ingestion, inhalation or dermal})}{RfD}$$

Table 1 shows the RfD for Cr, Cu, As, Mn, Ba, Cd, Pb and Hg in mg/kg BW/day, whereas THQ 1 assumes that there is no major danger to the exposed population's health.

While the hazard index (HI), which is employed to calculate the overall possible non-carcinogenic risks of various pollutants through three different exposure routes stated above, is equal to the sum of all expected HQs (non-carcinogenic risks) through inhalation, oral and dermal routes.

$$HI = \sum HQ = HQ_{ing} + HQ_{inh} + HQ_{derm}$$

If the HI score is lesser than one, then there is no appreciable danger of non-carcinogenic consequences. However, there is a chance that non-carcinogenic consequences will manifest themselves when $HI > 1$, and the chance grows as HI rises.

Carcinogenic risk assessment

For each exposure pathway, the rating of life cancer risk cumulatively was calculated using the formula given below to predict the LCR value, i.e., cancer risk

Table 1 The values of RfD and CSF used in the current studies

Metal	RfD _{ing}	RfD _{inh}	RfD _{derm}	CSF _{ing}	CSF _{inh}	CSF _{derm}
Cr	3×10^{-3}	2.86×10^{-5}	6×10^{-5}	0.5	–	41.0
Mn	140×10^{-3}	140×10^{-3}	1.8×10^{-3}	–	–	–
Cu	4×10^{-2}	4.02×10^{-2}	1.2×10^{-2}	–	–	–
As	3×10^{-4}	3×10^{-4}	1.23×10^{-4}	1.5	1.5	1.5
Cd	1×10^{-4}	1×10^{-4}	1×10^{-5}	0.38	–	6.3
Ba	7×10^{-2}	1.43×10^{-4}	4.9×10^{-3}	–	–	–
Pb	3.5×10^{-3}	3.25×10^{-3}	5.25×10^{-4}	0.0085	–	0.042
Hg	3×10^{-4}	3×10^{-4}	2.1×10^{-5}	–	–	–

for lifetime subjection or exposure due to Pb, Cr, As, Hg and Cd.

$$LCR = ADD(\text{ingestion, inhalation or dermal}) \times CSF$$

$$= \sum \text{cancer risk} = \text{cancer risk}_{\text{ing}} + \text{cancer risk}_{\text{inh}} + \text{cancer risk}_{\text{derm}}$$

In above equation, CSF represents cancer slope factor of each metal for 3 exposure paths (please see Table 2). As opposed to LCR 10^{-6} , LCR $> 1 \times 10^{-4}$ and LCR 1×10^{-6} to 1×10^{-4} , which, respectively, denote zero carcinogenic risk, a high risk to develop cancer and a tolerable level of risk for humans.

Statistical analysis and PMF model

The descriptive statistics including minimum, maximum, mean, median, variation coefficient, standard deviation (SD), kurtosis and skewness were employed to assess metals. To make sure and analyze whether the metalloid and metals (TMs and HMs) concentrations were normally distributed or otherwise, the Kolmogorov–Smirnov (K-S) test was applied. The SPSS 20 (SPSS Inc., Chicago, IL, USA) for windows was employed to perform Pearson correlation. The PMF is a multivariate receptor factor model to analyze source apportionment. PMF measures the contribution of each sample and determines the source profile.

Table 2 TMs & HMs concentration (mg/kg $\pm$ SEM) in soil ($n=20$)^a

Region	Concentration (mg/kg)	Cr	Mn	Cu	As	Cd	Ba	Hg	Pb
I	Min	22.31	350.01	83.81	2.04	1.01	65.01	0.050	30.01
	Max	44.01	455.03	170.04	2.43	1.16	80.04	0.061	60.01
	Mean	34.88 ± 3.88	398.26 ± 20.97	122.78 ± 19.46	2.14 ± 0.07	1.07 ± 0.03	70.62 ± 2.53	0.056 ± 0.0002	44.10 ± 5.52
II	Min	70.01	370.03	200.01	1.75	0.92	96.01	0.025	56.71
	Max	125.01	410.01	260.01	2.62	1.24	120.02	0.035	110.01
	Mean	90.62 ± 12.66	388.84 ± 8.09	231.56 ± 11.38	2.21 ± 0.17	1.08 ± 0.05	110.74 ± 2.47	0.028 ± 0.0019	76.35 ± 8.95
III	Min	45.01	340.01	205.03	2.51	0.92	110.01	0.010	101.02
	Max	65.01	380.03	255.04	3.51	1.24	125.05	0.020	165.03
	Mean	50.84 ± 3.77	362.38 ± 6.61	231.68 ± 8.77	2.87 ± 0.19	1.05 ± 0.06	115.25 ± 3.10	0.017 ± 0.0020	132.88 ± 10.64
IV	Min	36.01	310.01	110.01	2.05	0.91	89.05	0.010	30.02
	Max	101.01	405.01	260.01	2.73	1.14	135.01	0.046	72.02
	Mean	70.82 ± 12.31	353.01 ± 15.13	200.01 ± 25.54	2.25 ± 0.12	1.02 ± 0.04	103.43 ± 8.52	0.030 ± 0.0058	57.62 ± 7.25
Total	Min	22.31	310.01	83.81	1.75	0.91	65.01	0.005	30.01
	Max	125.01	455.03	260.01	3.51	1.24	135.01	0.046	165.03
	Mean	61.79 ± 6.41	375.62 ± 7.68	196.51 ± 13.01	2.37 ± 0.09	1.05 ± 0.02	100.01 ± 4.67	0.020 ± 0.0027	77.74 ± 8.65
Skewness		0.94	0.49	−0.91	1.12	0.39	−0.34	0.49	0.87
Kurtosis		0.15	0.23	−0.41	1.45	−0.61	−0.96	−0.89	−0.07
K-S p		0.84	0.74	0.78	0.76	0.93	0.81	0.75	1.02

^a Five samples of soil from the selected regions, SEM: standard error mean, K-S p is the significance level of Kolmogorov–Smirnov test for normality

In the current studies, EPA PMF 5.0 was used to identify quantitatively different sources of TMs, HMs and metalloids in the soil samples.

Results and discussion

Quality assurance

Accuracy is a crucial and primary aspect because analytical results are susceptible to inaccuracies that cause them to differ from the actual determinants' concentrations. The ability to make decisions based on the collected results is affected by these results. The results may depend on numerous variables including the laboratory's environmental conditions, standards, the strength of matrices' effects, the stability of the instruments and the reagent purity. Thus, SRM 2709a San Joaquin soil samples were studied in order to confirm accuracy during the examination of Mn, Cr, Cu, As, Cd, Hg, Pb and Ba in actual samples. Results were then provided in terms of percent recovery studies (Tables 2S, 3S). The recovery rates that were attained, which were in the range of 97.6–102.9%, show outstanding extraction efficiency.

Each concentration's peak height (y) was plotted versus the nominal concentration in triplicate to obtain the linear calibration curve in $y = mx + b$ form. This yielded a dynamic linear range of 0.2–1.0 $\mu\text{g/mL}$ for Cu and Mn, 0.01–500 $\mu\text{g/mL}$ for Cr, Cd, Ba and As, and 0.02–1000 $\mu\text{g/mL}$ for Hg and Pb. In $y = mx + b$, " m " stands for the calibration curve's slope and " b " for its intercept. In Table 4S, the linear regression equation has been illustrated and the required parameters are tabulated. According to the suggested approaches, LOD and LOQ are as indicated in Table 4S. The lower detection limit numbers show that the method's sensitivity was sufficient. The percent RSD was obtained after the precision investigation in terms of repetition ($n = 10$, Table 4S) was completed. In the examination of SRM 2709a of San Joaquin soil, the percent RSD values varied from 0.20 to 6.40 (Table 4S), respectively.

Application to real samples

On soil samples, the well-known acid extraction process was used to extract Cr, Cu, Mn, As, Cd, Ba, Pb and Hg. The results are shown in Table 2 and data

from Table 2 demonstrated that the concentration of As is highly skewed, while the Cr, Cu and Pb contents are moderately skewed, whereas the Mn, Cd, Ba and Hg contents are symmetrical distributed. Kurtosis is higher than 1 for As which indicated the data are highly peaked. The K-S test demonstrated that the contents of Mn, Cu, As and Hg are normally distributed.

The metals were divided into metalloid, HMs and TMs, and their amount in the soil was determined to be in the following order: $\text{Mn} > \text{Cu} > \text{Ba} > \text{Pb} > \text{Cr} > \text{As} > \text{Cd} > \text{Hg}$. It was hypothesized that the point closest to the landfill site is the most polluted and contaminated because the concentration of both TMs and HMs dropped as sample point distance from the garbage site increased (Table 6S supplementary data).

The average Cr concentration was found lower than the regulatory threshold imposed by organizations like the EU (100–150 mg/kg) (Waseem et al., 2014) and WHO (0.05 mg/L), but this soil is not fit for agricultural purposes because Cr amount was found greater than the concentration limit set by US EPA, i.e., 11 mg/kg (Kinuthia et al., 2020). Since soil and Cr are closely related, the main source of Cr in this study region is the movement of landfill leachate into the soil of surrounding area, where the majority of the landfill is made up of waste from industries such as tanneries and chemical dyeing. The harmful effects of Cr on the biota of Chernozem have a significant impact on the biological activities of the soil and decrease the biota's catalytic activity.

Cu and Mn both metals are utilized in the production of a large number of objects such as automotive parts, wires, pipes and alloys that end up in the landfill; their quantities among the TMs were greater in soil samples taken from the surroundings of garbage points. Their high concentration can be a sign that leachate with a high Cu and Mn content has moved into the nearby soils. Mn levels in soil were found to be within the ranges of reference advised by the WHO (FAO/WHO: not more than 600 mg/kg); however, Cu levels were found to be greater than the allowed limit established by the EU (50–140 mg/kg) (Waseem et al., 2014) and FAO/WHO (36–75 mg/kg) (Onyedikachi et al., 2018). Although Cu is a necessary nutrient, too much of it in the soil can be poisonous to some plants and helpful microbes, limit the mineralization of phosphorus and nitrogen, have negative effects on human health and be harmful to

fish and other aquatic life (Romić et al., 2014). More than 300 mg/kg of Cu in the soil affects wheat germination, lowers the biomass in cabbage shoots and roots and lowers rice yield (Chiou & Hsu, 2019). The average amount of As in soil samples did not go above the levels advised by the EU (20 mg/kg) (Kabir et al., 2016). Because of a linear dependence of plant uptake on the concentration of As in soil, cultivation in As-contaminated soil has a negative impact on plant development and yield (Waseem et al., 2014).

The average Cd concentration was lower than the regulatory threshold imposed by organizations like the EU (1–3 mg/kg). (Waseem et al., 2014), US EPA (1.4 mg/kg) (Mekonnen et al., 2020); however, due to the Cd concentration above the limits established by the WHO (0.003 mg/kg), the investigated soil is unsuitable for farming and gardening (Kinuthia et al., 2020). Cd is strongly linked to soil, and in the research region, its main source is the leachate from landfills, where the majority of the trash is industrial waste that is found as impurities in solders, body care items, alloys, galvanized pipes and plant colors. The harmful effects of the high Cd concentration on helpful bacteria disrupt their metabolism and prevent their growth. In humans, Cd has a long biological half-life (10–35 years) (Agoro et al., 2020; Liao et al., 2005).

Ba and Pb are utilized in the production of numerous commodities, such as Pb-based paints, Pb batteries, rubber goods and glass that end up in landfills, exalted Pb and Ba amounts and concentrations among the HMs were discovered in soil samples taken from nearby dump sites. The average Ba concentration in soil was below the US EPA's standard (70–3000 mg/kg) (US-EPA, 2017). Ba overexposure in soil may interfere with the calcium metabolism (Vodyanitskii, 2016). The average Pb concentration was determined to be within the regulatory threshold imposed by organizations like the EU (50–300 mg/kg) (Waseem et al., 2014), US EPA (200 mg/kg) (Kinuthia et al., 2020). The greater Pb concentration in soil has a significant impact on its carbon and nitrogen-containing microbial mass and worsens fertility by slowing down the rate of nutrient cycling (Zeng et al., 2006).

The average Hg content was determined to be within the WHO standard limit (0.08 mg/kg) (Kinuthia et al., 2020). Nematodes, which reside in the interstitial spaces of soils and are among the metazoan phyla with the greatest diversity of species, are crucial to the soil's ability to cycle nutrients. The

high concentration of Hg harms these nematodes. An exposure to humans affects immune system, organs, and nervous system, and results in reproductive abnormalities (Martinez et al., 2019).

Leaching of TMs and HMs into soil is also significantly influenced by the degradation of biodegradable materials in landfills. The aquifer matrix contains a significant amount of oxygen as an oxidant, which produces reducing conditions and disturbs the metals' natural states in groundwater. Due to the high concentrations of non-activated and activated colloids, small and large dissolved particles, and TMs and HMs, these metals were able to bind to the aquifer's pores (Boateng et al., 2019). The fact that As, Cr, Pb and Cd have been discovered to play a major role to the risk of metal pollution in the soil caused by human activity. They were classified as human carcinogens, and placed in the groups of priority metals which are crucial for human health, and could harm many organs (Kusin et al., 2018).

Source apportionment of soil metalloid, TMs and HMs

The Pearson correlation coefficients between metalloid, TMs and HMs in soil samples collected from the landfill are presented in Table 3. As shown in Table 3, a significantly positive correlation at $P < 0.01$ was found between the metal pairs Cr–Cu ($r = 0.732$), Cr–Cd ($r = 0.470$), Cr–Ba ($r = 0.700$), Mn–Cd ($r = 0.698$), Mn–Hg ($r = 0.647$), Cu–Ba ($r = 0.909$), Cu–Pb ($r = 0.712$), Ba–Hg ($r = 0.583$) and Ba–Pb ($r = 0.518$). In addition, significantly positive correlation coefficients between the metalloid and metal pairs As–Cd ($r = 0.609$), As–Ba ($r = 0.667$), As–Pb ($r = 0.869$), Cu–As ($r = 0.596$) were found at the level of 0.01. Cd had poor correlations with other HMs (Ba, Hg and Pb), whereas Hg was found to have significantly positive correlation at $P < 0.05$ with Pb. The above relationship indicated that the metalloid and metals (TMs and HMs) had the same source as leachate of industrial landfill.

For the PMF model, the input files comprised of content data and the corresponding uncertainty data of HMs (Cd, Ba, Hg and Pb), TMs (Mn, Cr and Cu) and metalloid (As). The selection of the most appropriate number was made depending on the stable and minimum Q value. In this study, the suitable number of factor was found to be 4, whereas the residual value

Table 3 Correlation matrix of the metalloid, TMs and HMs concentrations

	Cr	Mn	Cu	As	Cd	Ba	Hg	Pb
Cr	1	0.281	0.732**	0.235	0.470*	0.700**	−0.111	0.231
Mn		1	0.218	0.177	0.698**	0.013	0.647**	0.058
Cu			1	0.596**	0.409	0.909**	−0.433	0.712**
As				1	0.609**	0.667**	−0.283	0.869**
Cd					1	0.315	0.380	0.381
Ba						1	−0.583**	0.718**
Hg							1	−0.516*
Pb								1

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

*Correlation is significant at the 0.05 level (2-tailed)

of most soil samples was determined between −3 and 3. The coefficient of determination between the predicted and observed value (r^2) exhibits a strong correlation among the investigated metals. The r^2 value determined for Pd was 0.998, and the r^2 value for Mn was found to be the minimum. For the rest of the metals, the r^2 value was found to be greater than 0.844 and the results were appropriate. Thus, the PMF model used sufficient number of factors to completely elaborate the information given in the real data. Figure 2 shows the run results of the PMF model.

Factor 1 had high factor loading values of 74.4% and 49.0% for Pb and Cu, respectively (Fig. 2a). The results show that Pb and its byproducts are discharged into the soil because of various industrial activities like printing, and preparation of photographic materials, pigments, storage batteries and television tubes (Kumar et al., 2020). So, the factor indicates that Pb present in the studied soil samples likely comes from the landfill of above-mentioned industries.

As (32.7%), Ba (32.0%), Cd (30.9%) and Mn (30.7%) characterize the factor 2 (Fig. 2b). The sources of As are the electronic devices and the gallium arsenide wafers. In addition to above sources, As can also be found in the operations of silicon semiconductors that utilize As implants of much high doses. A further impact is created by the applications of herbicides, pesticides and crop desiccants, and the use of additives of As in livestock feed (Malsiu et al., 2020). Ba is employed in industry in a number of different forms. For example, it finds applications in the preparation of paper, soaps, alloys, rubber, valves and linoleum. Ba is also found in fossil fuels and coal. Besides, the compounds of Ba are used in specialty arc welding, glass industries, the cement, cosmetics, electronics, pharmaceuticals, insecticides and paints (Corrêa Nogueiro* & Ferracciu Alleoni, 2013).

Some of the anthropogenic sources of Cd contamination are electroplating, batteries and metallurgical industries (Tanong et al., 2017). Mn is used in different electrical equipments and mechanical devices. Thus, the second factor is mainly because of welding, electronic, batteries and glass industries sources. A number of other reported studies indicate the similar results (Tanong et al., 2017).

Hg with a percentage of 72.6% dominates the factor 3. Paints industry and the dental practice wastes, which can likely cause 50% of the crude Hg contents, are the major sources of this metal in the landfill. Thus, the factor 3 is attributed to the combined effect of paint manufacturing and dental implant-related activities.

The factor 4 was dominated by Cr and Cu with the percentages of 72.1% and 57.9%, respectively (Fig. 2c). It was noticed that the average amounts of both these metals were higher than their background values. Cr is used on a wider scale in metallurgy, electroplating and in the manufacturing of pigments, paints, pulp, papers and preservatives among others (Kinuthia et al., 2020). Cu finds applications in brass production, electric goods industry, manufacturing of printed circuit board, wire drawing, electronics plating, paint production, copper polishing, printing operations and wood preservatives (Wieczorek et al., 2021). So, it can be concluded that electroplating, metallurgy, paint and pigment manufacturing industry are the main sources of Cr and Cu metals.

Assessment of TMs and HMs contamination in soil

To examine pollution, assess the pattern of contamination and identify the potential risks associated with the toxicity and concentration of HMs and TMs in soil and the exposure to ecological

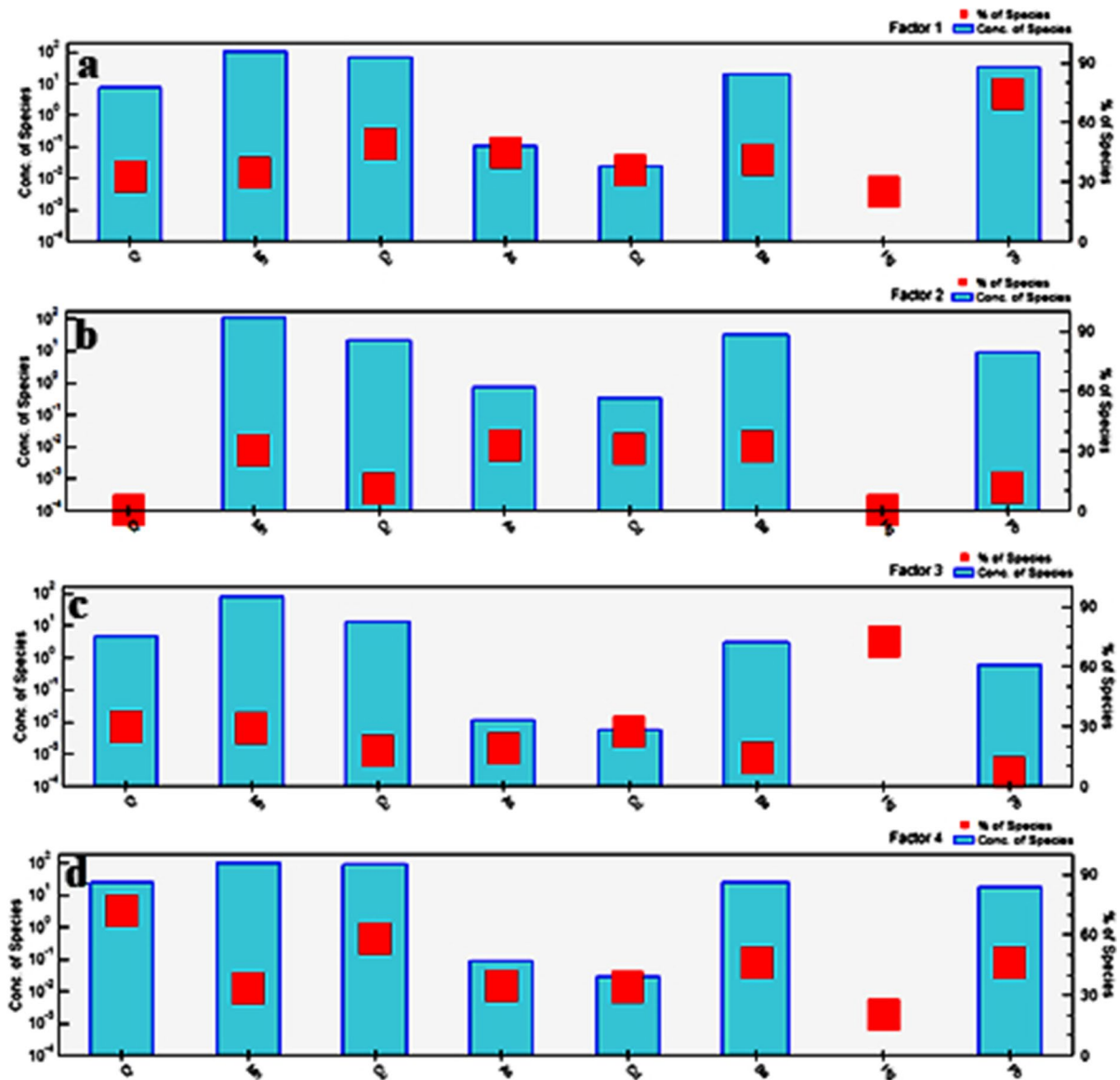


Fig. 2 Factor profiles and contribution percentage of metalloid, TMs and HMs in soil

sensitivity, the geoaccumulation index (I_{geo}), contamination factor (CF), and potential ecological risk index (PERI), degree of contamination (C_{deg}) and pollution load index (PLI) were developed. For all elements except Pb, which were classed as moderately contaminated, I_{geo} revealed (Table 4) that soil is unpolluted to moderately polluted. Similar to the distribution of I_{geo} , the soil was moderately contaminated and significantly contaminated for Pb (3.01). According to data (Table 4), soil

contaminated with Pb posed a significant ecological risk, but there is a low ecological risk due to rest of metals investigated. Data from Table 3 showed that PLI value is greater than 1, while C_{deg} value is less than 32 ($16 \leq C_{deg} \leq 32$) which indicated that the soil is polluted and considerably contaminated with metals, respectively.

Furthermore, the significant environmental damage posed by Cd and Hg has long been understood. According to a report on the composition of HMs

Table 4 I_{geo} , CF, PERI, PLI and C_{deg} calculation for soil Unaffected soil's geochemical background value (mg/kg) at the city site: Cr=35.68, Mn=140.92, Cu=65.68, As=0.95, Cd=0.35, Ba=35.84, Hg=0.008, Pb=25.82

Index	TMs				HMs			
	Cr	Mn	Cu	As	Cd	Ba	Hg	Pb
I_{geo}	0.21	0.83	0.99	0.70	0.96	0.90	0.74	1.01
CF	1.73	2.65	2.99	2.44	2.91	2.79	2.50	3.01
PERI	3.46	–	14.95	24.40	145.50	–	100	15.05
PLI	2.59							
C_{deg}	21.02							

in water in China for the assessment of ecological risk, there was a moderate to high risk as a result of the metal contamination. When using the contaminated soils for agricultural purposes in the current region of study, humans may be exposed to HMs and TMs, and their accumulation in the soil increases phyto-accumulation in the agricultural products that are grown.

Assessment of human health risk

Average daily dose

For metals such as Cr, Cu, Mn (TMs), Ba, Cd, Pb and Hg, (HMs), and As (metalloid), ADD in mg/kg/day was estimated using three exposure pathways: ADD_{ing} , ADD_{inh} and ADD_{derm} . The ADD tendency

(Table 5) was detected in both children and adults in the following order: $\text{ADD}_{\text{ing}} > \text{ADD}_{\text{derm}} > \text{ADD}_{\text{inh}}$. The highest dose of all metals in the ADD of soil for children was in ADD_{ing} and ADD_{derm} , whereas ADD_{ing} was greater in adults. It is possible that this is a result of children's unique behavior through skin contact, notably hand-to-mouth contact. Therefore, it is important to teach kids to wash their hands often to lower health hazards. In a nut shell, children are more likely to be exposed to TMs, HMs and metalloid as compared to adults.

Total hazard quotient and index

For all metals, except for Ba, the HQ trend (Table 5) was observed in children as well as in adults in the same order: $\text{HQ}_{\text{ing}} > \text{HQ}_{\text{derm}} > \text{HQ}_{\text{inh}}$. Both in children

Table 5 Results of ADD and HQ values for adults and children

Metal	ADD_{ing}	ADD_{inh}	ADD_{derm}	HQ_{ing}	HQ_{inh}	HQ_{derm}
Soil						
Adults						
Cr	4.355×10^{-5}	6.404×10^{-9}	1.866×10^{-7}	1.452×10^{-2}	2.239×10^{-4}	3.110×10^{-3}
Mn	2.646×10^{-4}	3.891×10^{-8}	1.134×10^{-6}	1.890×10^{-3}	2.779×10^{-7}	6.300×10^{-4}
Cu	1.384×10^{-4}	2.035×10^{-8}	5.934×10^{-7}	3.460×10^{-3}	5.062×10^{-7}	4.945×10^{-5}
As	1.634×10^{-6}	2.403×10^{-10}	2.103×10^{-7}	5.446×10^{-3}	8.010×10^{-7}	1.709×10^{-3}
Cd	7.185×10^{-7}	1.056×10^{-10}	3.080×10^{-9}	7.185×10^{-3}	1.056×10^{-6}	3.080×10^{-4}
Ba	7.049×10^{-5}	1.036×10^{-8}	3.022×10^{-7}	1.007×10^{-3}	7.244×10^{-5}	6.167×10^{-5}
Hg	1.409×10^{-8}	2.072×10^{-12}	6.040×10^{-11}	4.696×10^{-5}	6.906×10^{-9}	2.876×10^{-6}
Pb	5.476×10^{-5}	8.054×10^{-9}	2.347×10^{-7}	1.564×10^{-2}	2.478×10^{-6}	4.470×10^{-4}
Children						
Cr	4.064×10^{-4}	2.988×10^{-8}	1.636×10^{-7}	1.355×10^{-1}	1.044×10^{-3}	2.726×10^{-3}
Mn	2.470×10^{-3}	1.816×10^{-7}	9.939×10^{-7}	1.764×10^{-2}	1.297×10^{-6}	5.521×10^{-4}
Cu	1.292×10^{-3}	9.501×10^{-8}	5.199×10^{-7}	3.230×10^{-2}	2.363×10^{-6}	4.332×10^{-5}
As	1.525×10^{-5}	1.122×10^{-9}	1.842×10^{-7}	5.083×10^{-2}	3.740×10^{-6}	1.497×10^{-3}
Cd	6.706×10^{-6}	4.931×10^{-10}	2.699×10^{-9}	6.706×10^{-2}	4.931×10^{-6}	2.699×10^{-4}
Ba	6.579×10^{-4}	4.837×10^{-8}	2.647×10^{-7}	9.398×10^{-3}	3.382×10^{-4}	5.402×10^{-5}
Hg	1.315×10^{-7}	9.669×10^{-12}	5.292×10^{-11}	4.383×10^{-4}	3.223×10^{-8}	2.520×10^{-6}
Pb	5.111×10^{-4}	3.758×10^{-8}	2.057×10^{-7}	1.460×10^{-1}	1.156×10^{-5}	3.918×10^{-4}

and adults, the HI values for any given metal through all soil exposure pathways were less than 1, indicating no major health risk according to data from Table 6.

Carcinogenic risk assessment

A lifespan exposure to TMs (Cr, As, Cu, Mn) and HMs (Cd, Pb, Hg, Ba) was evaluated through the carcinogenic slope factor using diverse exposure routes such as inhalation, ingestion and skin contact. $LCR < 10^{-6}$, $LCR > 1 \times 10^{-4}$ and $LCR \ 1 \times 10^{-6}$ to 1×10^{-4} , denote no risk of human carcinogenesis, elevated risk of acquiring cancer and low carcinogenic risk, respectively. For children who are at the lower edge of cancer risk, the LCR value for Cr by various soil exposure routes was 2.039×10^{-4} (Table 6). Children are therefore at greater risk than adults in the current region of study where cutaneous and inhalation channels are the next significant contributors to access LCR.

Sometimes, the use of indices such as I_{geo} , CF, PERI, C_{deg} and PLI cannot answer the following two important questions despite their wide use and importance. Firstly, the background value of metals and metalloids is required from the studied region to calculate the I_{geo} for the metals and metalloids.

However, this parameter is mostly unknown for the area or region being studied. In such cases, the background value of the earth's crust is utilized which can produce conclusions with false negatives. Secondly, the CF and PLI are the instruments used to assess the contamination degree by one or several metals present in the region under observation. But, these indices are measured as a function of the background values of the respective metals or elements in the region under examination. Again, the difficulty can arise because the background values of the respective elements are not known for that region. Thus, the background value of the crust of earth is employed in the calculations. This research on soil around landfill sites is limited by the remediation method, which aims to develop a final solution that is protective of human health and the environment.

Conclusion

In this investigation, ICP-OES was used to measure the content of metal(loid)s (Cr, Mn, Cu, As, Cd, Ba, Hg and Pb) in soil specimens taken from the surrounding areas of industrial waste landfill in Gujranwala city of Punjab province of Pakistan. CF, I_{geo} ,

Table 6 HI & LCR values from water and soil analysis for children & adults

Metal	HI	Interpretation	LCR	Interpretation
Soil				
Adults				
Cr	1.785×10^{-2}	No significant risk	2.942×10^{-5}	No carcinogenic risk
Mn	2.520×10^{-3}	No significant risk	–	–
Cu	3.509×10^{-3}	No significant risk	–	–
As	7.155×10^{-3}	No significant risk	2.767×10^{-6}	No carcinogenic risk
Cd	7.494×10^{-3}	No significant risk	2.924×10^{-7}	No carcinogenic risk
Ba	1.141×10^{-3}	No significant risk	–	–
Hg	4.984×10^{-5}	No significant risk	–	–
Pb	1.608×10^{-2}	No significant risk	4.663×10^{-7}	No carcinogenic risk
Children				
Cr	1.392×10^{-1}	No significant risk	2.039×10^{-4}	Carcinogenic risk
Mn	1.819×10^{-2}	No significant risk	–	–
Cu	3.234×10^{-2}	No significant risk	–	–
As	5.233×10^{-2}	No significant risk	2.289×10^{-5}	No carcinogenic risk
Cd	6.733×10^{-2}	No significant risk	2.549×10^{-6}	No carcinogenic risk
Ba	9.790×10^{-3}	No significant risk	–	–
Hg	4.408×10^{-4}	No significant risk	–	–
Pb	1.464×10^{-1}	No significant risk	4.352×10^{-6}	No carcinogenic risk

C_{deg} , PERI and PLI were calculated to assess the overall picture of soil contamination pollution and ecological sensitivity due to presence of metal(loid)s in the soil under investigation. Human health risk was also evaluated by considering the ADD, THQ, HI and CR values.

The amount of metal(loid)s in soil was found in the following order: $Mn > Cu > Ba > Pb > Cr > As > Cd > Hg$. The average Cr concentration was found greater than the concentration limit set by US EPA, i.e., 11 mg/kg. Thus, this soil was found not fit for the agricultural purposes. Although Cu is a necessary nutrient, a higher than the allowed amount established by the EU (50–140 mg/kg) and FAO/WHO (36–75 mg/kg) can be poisonous to some plants and helpful microbes. The investigated soil is also unsuitable for farming and gardening because of the Cd concentrations above the limits established by the WHO (0.003 mg/kg). Similar to the distribution of Igeo, the soil was moderately or significantly polluted and contaminated with Pb (CF=3.01). The findings showed that among the soil ADD, ADD_{ing} ($Mn = 2.470 \times 10^{-3}$) and ADD_{derm} ($Mn = 9.939 \times 10^{-7}$) for children had the highest doses among all the other metals, whereas ADD_{ing} ($Mn = 2.646 \times 10^{-4}$) was also found greater in adults as compared to the other metals. Additionally, the HI value in soil for all metals was found lesser than 1, indicating no major harm to human health. Children are relatively at greater risk than adults in the areas studied. Metals in soil present low non-carcinogenic and carcinogenic risks to children and adults residing in the research area, according to the findings of human health risk assessments.

However, there is a need for awareness and action in order to reduce and prevent contamination with metal(loid)s in soil by slowing down the movement of leachate through the suitable design of new dump sites with suitable foundations. The current observations and results provide significant information about the hazardous and heavy metal contamination and pollution of the soil in an industrial area. They are also very important in the region where improper solid waste management methods are present. Government organizations should work to reduce the quantity of these hazardous and heavy metals in soil and water since they can bioaccumulate, and this research can help with preventive measures to preserve urban environments. To avoid the release of

heavy metals into soil, government agencies should take into account new technologies with greater capacity for extracting them from landfills. To stop the leaching of these metals into water and soil, vertical engineered barriers (VEB), liners and caps can be constructed. Therefore, the fact that humans have been exposed to soil shows the need for further interventions and legislative changes.

Author contributions WA helped in, methodology, formal analysis, MZ, Supervision, performed review, MA helped in review & editing, writing—original draft, MA and AH validated the study, SL was involved in investigation, formal analysis, QK contributed to formal analysis, review, DNI performed review, analysis.

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Declarations

Conflict of interest The authors declare no conflict of interest.

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