

## Investigating and Compiling a Map of the Severity of Heavy Metal Pollution in the Soil around the Landfill of Sabzevar Municipal Waste with Different Indicators

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### ABSTRACT

**Introduction:** Uncontrolled and improper landfilling of solid waste increases the concentration of heavy metals in the soil. Therefore, this study aimed to investigate soil contamination with heavy metals in the landfill of municipal and hospital waste in Sabzevar city by compiling a map of the severity of heavy metal pollution in the soil.

**Materials and Methods:** Concentrations of heavy metals were analyzed by ICP-OES at distances of 10, 100 m from the burial site and at depths of 10 and 30 cm. To quantitatively evaluate the severity of pollution and the environmental effects of heavy metals in the soils around the Sabzevar landfill, enrichment factor (EF), index of geoaccumulation ( $I_{geo}$ ), and investigating carcinogenic and noncarcinogenic hazards of heavy metals were used. Then a general map of soil pollution severity was prepared using the limitation scores (LS) and potential ecological risk index (RI) method.

**Results:** Mean concentrations of As, Zn, Pb, Cr, and Cu in topsoil were 6.01, 41.4, 6.31, 26.77, and 31.45 mg/kg, respectively, as well as Hg and Cd were 60.79, and 61.60  $\mu$ g/kg, respectively. However, mean concentrations of As, Zn, Pb, Cr, and Cu in the soil at a depth of 30 cm were 5.75, 38.33, 6.25, 22.68, and 31.04 mg/kg, respectively, as well as Hg and Cd were 66.57, and 59.98  $\mu$ g/kg, respectively.

**Conclusion:** According to the estimates of  $I_{geo}$  and FE indices, only Hg and Cd showed severe contamination. The noncarcinogenic risks of heavy metals were estimated to be less than 1.

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### Introduction

Population growth and lifestyle changes are two common factors in most countries that have caused a keen growth in solid waste<sup>1</sup>. As a result, municipal solid waste management is one of the health and environmental issues in developing countries, such as Iran. In cities of Iran,

landfilling is used for solid waste disposal, since it is more inexpensive, simpler, and more economical with fewer technology barriers compared to other methods of waste management<sup>2, 3</sup>. Uncontrolled and improper landfilling of solid waste is a major source of soil

pollution and degradation that adversely affects the environment and human health<sup>4, 5</sup>. In the past, soil or land pollution was not as important as air and water pollution, since soil pollution is not tangible and soil pollution is more difficult to be controlled than air and water pollution. However, the risk of soil pollution is not less than other pollutants and can reduce the quality of the environment<sup>4, 6</sup>. Landfill leachate generation cannot be prevented even in best-engineered landfills in developed countries. Landfill leachate contains different types of heavy metals, so landfills are one of the main sources of the increased level of heavy metal in the soil. The concentration of heavy metals varies according to the type and source of solid waste placed in the landfill<sup>3, 4</sup>. Heavy metals are natural components of the soil and their high concentrations in the soil can have a geological origin, such as parent material and various soil formation processes. They may be caused due to harmful human activities, such as iron and steel industries, mining, road transportation, disposal of industrial wastewater, use of chemical substances and fertilizers in agriculture, and unsanitary disposal of waste<sup>4, 7-9</sup>. The adverse effects of heavy metals on soil become most apparent when their concentration exceeds a certain level, which depends on the type of metal, soil type, various human activities, and time of heavy metal accumulation<sup>4, 10</sup>. Due to the lack of biodegradability, very high stability, toxicity, and the ability to gradually accumulate in the body tissue of animals and plants in the food chain, heavy metals have destructive effects on human and animal health<sup>2, 11-12</sup>. These metals greatly threaten the health of citizens by causing several symptoms and complaints, such as headache, dizziness, insomnia, forgetfulness, neurological disorders, joint pain and stones, and even cancers, such as liver, stomach, colon, bladder, breast, and prostate cancers<sup>13</sup>. Today, cancer is the leading cause of death in developed and developing countries worldwide. Many researchers have investigated the environmental effects and health risks of heavy metals in soils due to the

environmental and human health concerns associated with heavy metals. Moreover, soil and its quality play a fundamental role in ecosystem health, and knowledge regarding the concentration of these elements is an important indicator in predicting the risks and diseases caused by these metals<sup>14, 15</sup>.

Sanitary landfills are the oldest and most common method of solid waste disposal in many cities in Iran, such as Sabzevar<sup>1, 11</sup>, and do not have a landfill leachate collection and treatment system. Comprehensive research studies have not been done on the contamination of potentially toxic elements in the soils surrounding the landfill of Sabzevar city. In this study, enrichment factor (EF) and index of geoaccumulation ( $I_{geo}$ ) were used to quantitatively assess the pollution intensity and environmental effects of heavy metals, including arsenic (As), mercury (Hg), lead (Pb), zinc (Zn), copper (Cu), cadmium (Cd), and chromium (Cr) on the soils of Sabzevar waste landfills. Also, risk assessment of the mentioned heavy metals was performed for soil samples of Sabzevar municipal waste landfills (MWLs). Health risk assessment was used to determine carcinogenic and non-carcinogenic effects in adults and children. The types of exposure to important pollutants, including dermal contact, inhalation, and oral consumption in the soil of Sabzevar MWLs were also investigated. Finally, the zoning and general map of the soil pollution intensity surrounding the study area with heavy metals were plotted using the potential ecological risk index (RI) and cumulative limitation scores (LS).

## Materials and Methods

### Area of study

The waste produced in Sabzevar city since 2008 in a place called Nakhbar 25 km from Sabzevar city - is buried by soil dumping. Geological study of Sabzevar zone shows that it includes one of the largest ophiolite assemblages in Iran, a large part of which is composed of perpetuity rocks and serpentine rocks. Serpentine soils are considered as a major natural source of heavy metal accumulation<sup>16, 17</sup>.

### Sampling and analysis

Because of the geological structure and slope direction in the study area and the possibility of spreading toxic pollutants, sampling points were determined randomly at distances of 10, 100 meters from the burial site at 18 sampling stations (A to R). The location of the sampling points was recorded using the geographic information system (GPS). To avoid surface traces, as well as organic matter, sampling was performed at two depths of 10 and 30 cm. Figure 1 shows the location of soil sampling stations around the Sabzevar landfill. These samples were then mixed and a composite sample was prepared. Geological characteristics and background materials of the samples were taken as the natural background of the area, which was the same as the soil samples around the landfill.

After preparing the soil samples, they were air-dried for 48 hours in the laboratory, passed through mesh sieve No. 10, and placed in an oven at 110°C for 24 hours<sup>18</sup>.

The total concentrations of As, Hg, Zn, Pb, Cd, Cr, Cu, and Fe as reference metals were determined using microwave digestion with nitric acid (ISO 11465: 1993 (E) - Soil quality) and ICP-OES Agilent (Model 5100)<sup>3, 11-12</sup>.

Quality assurance (QA) and quality control (QC) were performed by measuring control samples and duplicate samples (with less than 10% error). To confirm the accuracy of the measurement, 5.5% of the samples (N = 18) were randomly tested. The consistency of repeated measurements of EC, pH, and heavy metals was determined to be 89.99%, 90.76%, and 90.87%, respectively<sup>13</sup>.

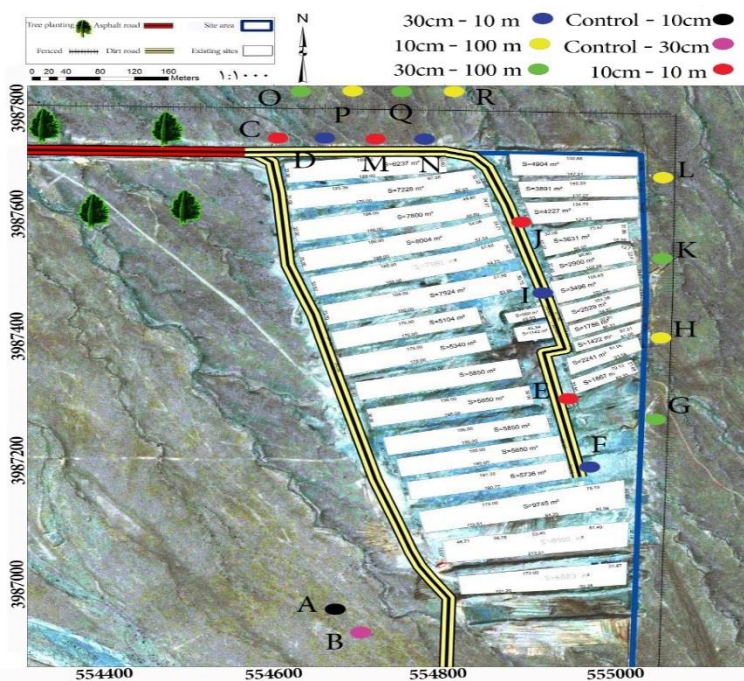


Figure 1: Location of soil sampling stations around the landfill of Sabzevar city

### Calculation of heavy metal pollution indices

In this study, EF,  $I_{geo}$ , and ecological risk potential (Er) were assessed for quantitative evaluation of pollution intensity and environmental effects of As, Hg, Pb, Zn, Cu, Cd, and Cr in soils around the Sabzevar landfill.

EF: It can distinguish heavy metal resources

from natural or human resources and can be calculated by Equation 1<sup>12, 19-20</sup>.

Equation 1:

$$EF = \frac{\left(\frac{C}{Fe}\right)_{\text{sample}}}{\left(\frac{C}{Fe}\right)_{\text{background}}}$$

Where EF is the enrichment factor,  $C_{\text{sample}}$  is the concentration of an element in the soil sample,  $C_{\text{background}}$  is the concentration of an element in the earth's crust,  $Fe_{\text{sample}}$  is the concentration of Fe in the soil sample, and  $Fe_{\text{background}}$  is the concentration of Fe in the earth's crust<sup>12, 19-20</sup>.

The basis of this pollution index is based on comparing the concentration of heavy metal in the samples with the concentration of the same heavy metal in the non-contaminated area. These concentrations are normalized based on the concentration of the reference element, which can be aluminium (Al), manganese (Mn), scandium (Sc), or iron (Fe). Given that the natural concentration of Fe in the earth's crust is high and its distribution in the earth's crust is not related to other metals and also its concentration is not dependent on human activities, Fe was used as a reference element in this study. Based on this factor, the enrichment intensity of the desired metal can be divided into five categories: > 2 (low), 5-2 (medium), 5-20 (high), 40-20 (very high), and < 40 (extremely high)<sup>12, 19-20</sup>.

Index of geoaccumulation: The  $I_{\text{geo}}$  was introduced by Muller and can determine soil contamination with heavy metals according to Equation 2<sup>12, 19-20</sup>.

Equation 2:

$$I_{\text{geo}} = \text{Log}_2 \left( \frac{C_n}{1.5B_n} \right)$$

In which,  $I_{\text{geo}}$  is the accumulation index,  $C_n$  is the concentration of metal in the sample, and  $B_n$  is the concentration of the desired metal (mean shale).

A constant coefficient of 1.5 minimizes the

effect of changes in background concentrations, which is usually because of changes in the soil lithology.

The basis of this index is the comparison of the measured concentration of each heavy metal in the sample with the concentration of its geochemical background in the soil.

The term geochemical background refers to the normal abundance of an element in a barren land or soil without the effects of human activities. This index helps to divide soils into seven groups in terms of pollution < 0 (uncontaminated), 0-1 (uncontaminated or moderately contaminated), 1-2 (moderately contaminated), 2-3 (moderately to heavily contaminate), 3-4 (heavily contaminated), 4-5 (heavily extremely contaminated) and 5 > (extremely contaminated)<sup>12, 19-20</sup>.

Er and potential ecological RI: The Er index was introduced by Hakanson to assess the potential environmental hazards of metals in soil. It can be calculated based on Equation 3<sup>19-21</sup>:

Equation 3:

$$RI = \sum Er = \sum Tr \times \frac{C_s}{C_b}$$

In which,  $C_s$  represents the amount of metal concentration measured in each sample and  $C_b$  shows the amount of heavy metals in unpolluted soil, Tr is the toxic reaction agent for heavy metals, Er is the ecological risk potential of each element, and RI is the ecological risk of all elements. Tr for heavy metals, such as As, Hg, Pb, Zn, Cu, Cd, and Cr were 10, 40, 5, 1, 5, 30, and 2, respectively<sup>19-21</sup>. According to Table 1, the pollution levels are classified into five and four levels based on Er and RI indices, respectively<sup>19-21</sup>.



**Table 1:** Values of Er and RI indices used in determining soil pollution<sup>21</sup>

Er	The degree of environmental risks of each metal	RI	Potential environmental risks to the environment
Er < 40	Low risk	RI < 150	Low potential ecological risk
40 < Er < 80	Moderate risk	150 < RI < 300	Moderate potential ecological risk
80 < Er < 160	Considerable risk	300 < RI < 600	Considerable potential ecological risk
160 < Er < 320	High risk	RI > 600	High potential ecological risk
ER > 320	Very high risk		

To better identify the contaminated areas or areas exposed to heavy metal contamination, an overview of soil contamination was presented using the LS to integrate one-element maps of metal concentration. To this end, first, the values of the total concentration of the measured elements, the allowable and hazardous limits of each element were converted into LS using an exponential equation. Then, the sum of the constraint points of the elements at the observation points was calculated, finally, it was zoned in ArcGIS software with the help of an ordinary kriging estimator<sup>1, 22</sup>. The total concentration values measured for each element based on the allowable limit (pollution standard) and hazard limit (cleaning standard) of the elements according to the standards presented in Table 2, were converted into LS according to the Equations 4 and 5:

Equation 4:

$$\text{if: } HMC > x_1 \rightarrow LS = b_0 \times HMC^{b_1} - 1$$

Equation 5:

$$\text{if: } HMC < x_1 \rightarrow LS = 0$$

Where LS is limitation scores,  $b_0$  and  $b_1$  are the coefficients of the equation, HMC is the heavy metal concentration and  $x_1$  is the allowable heavy metal concentration.  $b_0$  and  $b_1$  are the allowable coefficients of heavy metals. These coefficients can be obtained by solving the regression model of Equation 6<sup>22</sup>:

Equation 6:

$$\ln(LS + 1) = \ln b_0 + b_1 \times \ln(HMC)$$

In which, three known scores are used to find the values of two unknowns in the equation. For example, for As,  $LS + 1 = 0$  for zero concentration,  $LS + 1 = 1$  for 5 mg/kg,  $LS + 1 = 5$  for 50 mg/kg.

Concentrations of 5 and 50 mg/kg, respectively, are the maximum permissible and hazardous limits for As<sup>23</sup>. After solving the equation and determining its coefficients for each of the studied heavy elements, the relevant LS were calculated by placing different values of the concentrations of each element in Equations 4 and 5. After calculating the LS for all the studied elements, their sum was calculated at each sampling point<sup>22</sup>.

Health risk assessment: In this study, the assessment of heavy metal hazards of both carcinogenic and non-carcinogenic hazards was performed based on the health risk assessment method provided by the US Environmental Protection Agency (USEPA). In the study of human exposure to metals from the three paths of ingestion, inhalation, and dermal contact were considered and the average daily dose (ADD) of metals in each path was calculated using the Equations 7 to 13<sup>2, 12-13</sup>.

Equation 7:

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

Equation 8:

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

Equation 9:

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

Equation 10:

$$LADD = \frac{C \times \text{EF}}{\text{AT}} \times \left[ \left( \frac{\text{CR} \times \text{ED}}{\text{BW}} \right)_{\text{child}} + \left( \frac{\text{CR} \times \text{ED}}{\text{BW}} \right)_{\text{adult}} \right]$$

Equation 11:

$$CR_{ing} = IngR \times 10^{-6}$$

Equation 12:

$$CR_{inh} = \frac{InhR}{PEF}$$

Equation 13:

$$CR_{dermal} = SA \times SL \times ABS \times 10^{-6}$$

Where  $ADD_{ing}$ ,  $ADD_{inh}$ ,  $ADD_{dermal}$  are the mean daily exposure (mg/kg.day) through dermal contact, inhalation, and ingestion, respectively,  $LADD_{inh}$  is daily exposure to carcinogenic heavy metals (mg/kg.day) by inhalation.  $C$  is soil metal concentration (mg/kg),  $IngR$  is ingestion rate (mg/day) and dust inhalation rate ( $m^3/day$ ),  $EF$  is frequency of metal exposure (day/year),  $ED$  is duration of metal exposure (year),  $BW$  is body weight of the person exposed to metals (kg),  $AT$  is duration of exposure to any amount of metals on average (day) and  $PEF$  is metal-to-air diffusion factor ( $m^3/kg$ ). As well as  $SA$  is area of skin surface exposed to metals ( $cm^2$ ),  $SL$  is soil-to-skin adhesion factor ( $mg/cm^2.day$ ),  $ABS$  is dermal contact factor (without units), and  $CR$  is the rate of ingestion or inhalation or dermal contact<sup>2, 12-13</sup>.

Non-carcinogenic risk assessment: As, Cu, Hg, Zn, Cd, Pb, and Cr all have non-carcinogenic risks. Thus, after calculating the daily metal uptake for each route, the hazard quotient (HQ) of the total routes for children and adults was determined from the total ADD of each route to the reference value of the metal toxicity<sup>2, 12-13</sup>.

Equation 14:

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

HQ is the non-carcinogenic risk of metals in each route, and ADD is the daily uptake of metals in each route of metal exposure (mg/kg.day). If it is  $HQ < 1$ , it is not incompatible with human health, and if it is  $HQ > 1$ , it has adverse effects on human health. The value of the total non-carcinogenic RI of total metals for both adults and

children is obtained according to Equation 9<sup>2, 12-13</sup>.

Equation 15:

$$HI = \sum HQ_i = HQ_{ing} + HQ_{inh} + HQ_{dermal}$$

Hazard index (HI) is the sum of HQ, indicating the total risk of a non-carcinogenic element through three routes of exposure to the element. If the value is  $1 > HI$ , non-carcinogenic risk effects do not occur, the value  $1 < HI$  indicates the possibility of adverse health effects. It is possible that by increasing HI values, the risk of carcinogenicity of an individual for any type of cancer increases throughout life<sup>2, 12-13</sup>.

Carcinogenic risk assessment: Heavy metals, such as As, Cd, and Cr pose a potential genetic cancer risk through inhalation. Carcinogenic risk assessment of each of the three pathways for these metals was performed with a ratio of 10<sup>2, 12-13</sup>.

Equation 16:

$$RI = \sum LADD_i \times SF$$

In this equation, RI is cancer risk,  $ADD_i$  is the daily uptake of metals in each of the metal exposure pathways (mg/kg.day) and the SF is the risk factor for cancer per unit of metal exposure (mg/kg.day). RI indicates the risk of developing cancer, which is usually seen as the percentage of people with cancer in a given unit of the population. The cancer slope factors (CSF) indicates the highest risk of cancer for the body when exposed to a certain amount of contaminant<sup>2, 12-13</sup>. If the value is  $10^{-6} > IR$ , the risk of carcinogenicity for soil health can be neglected, and  $IR > 10^{-4}$  indicates the high risk and progression of cancer in humans. Values in the range of  $10^{-6} < IR < 10^{-4}$  indicate acceptable or tolerable risk and human health<sup>2, 12-13</sup>. Details of each parameter and its values used in the risk assessment equations are given in Tables 2, 3 and 4.

**Table 2:** The maximum standards of permissible limits, hazards, and coefficients b0 and b1 of the studied heavy metals<sup>23</sup>

Heavy metal	As(p)	Hg	Cd	Cr	Cu	Pb	Zn
Permissible limits (mg / kg) (e)	5	0.5	1	100	100	60	200
Hazard limits (mg / kg)	100	5	20	300	200	750	400
Ln (b <sub>0</sub> )	-1.12	0.80	0	-10.69	-18.28	-5.47	-38.21
b <sub>1</sub>	0.70	1.16	0.70	2.32	3.70	1.34	7.219

e: Guidance values are defined based on environmental hazards.

p: The risk of groundwater contamination at concentrations below the guidance value is lower than usual.

**Table 3:** Guide to the parameters of the equations for assessing the risk of carcinogenicity and non-carcinogenicity of heavy metals in the soil<sup>13,24</sup>

Parameter	Unit of measurement	Adult	Children
IngR	mg/day	100	200
InhR	m <sup>3</sup> /day	12.8	7.63
EF	day/year	350	350
ED	Year	24	6
BW	Kg	70	15
AT	Days	ED × 365 Non-carcinogenic effect 70 × 365 Carcinogenic effect	ED × 365 Non-carcinogenic effect 70 × 365 Carcinogenic effect
PEF	m <sup>3</sup> /kg	1.36 × 10 <sup>9</sup>	1.36 × 10 <sup>9</sup>
SA	cm <sup>2</sup>	4350	1600
SL	mg/cm <sup>2</sup> .day	0.07	0.2
ABS	-	0.001	0.001

**Table 4:** Reference value of its toxicity of heavy metals (RfDs) for health risk assessment<sup>13-15</sup>

Element	As	Cd	Cr	Pb	Hg	Cu	Zn
SF derm	3.66	-	2 × 10 <sup>1</sup>	-	-	-	-
Ing	1.5	-	5.01 × 10 <sup>-1</sup>	8.5 × 10 <sup>-3</sup>	-	-	-
Inh	4.3 × 10 <sup>-3</sup>	8.4 × 10 <sup>-1</sup>	4.2 × 10 <sup>1</sup>	-	-	-	-
RfD <sub>ing</sub>	3 × 10 <sup>-4</sup>	1 × 10 <sup>-6</sup>	3 × 10 <sup>-3</sup>	1.4 × 10 <sup>-3</sup>	3 × 10 <sup>-3</sup>	4 × 10 <sup>-2</sup>	3 × 10 <sup>-1</sup>
RfD <sub>inh</sub>	1.23 × 10 <sup>-4</sup>	1 × 10 <sup>-3</sup>	2.86 × 10 <sup>-5</sup>	3.52 × 10 <sup>-3</sup>	8.57 × 10 <sup>-5</sup>	4 × 10 <sup>-2</sup>	3 × 10 <sup>-1</sup>
RfD <sub>derm</sub>	1.23 × 10 <sup>-4</sup>	5 × 10 <sup>-5</sup>	3 × 10 <sup>-4</sup>	5.24 × 10 <sup>-4</sup>	2.1 × 10 <sup>-5</sup>	1.2 × 10 <sup>-2</sup>	6 × 10 <sup>-2</sup>

### Data analysis

After performing laboratory analyses, contamination indices were analyzed using Excel software 2016.

### Ethical issue

This study was authorized by Sabzevar University of Medical Sciences ethics committee IR.MEDSAB.REC.1398.053.

### Results

#### Concentrations of metals

The distribution of sampling concentrations of As, Hg, Pb, Zn, Cd, Cr, and Cu in the sampling stations are separately shown in Figures 2 and 3. The studied heavy metals revealed a wide range of

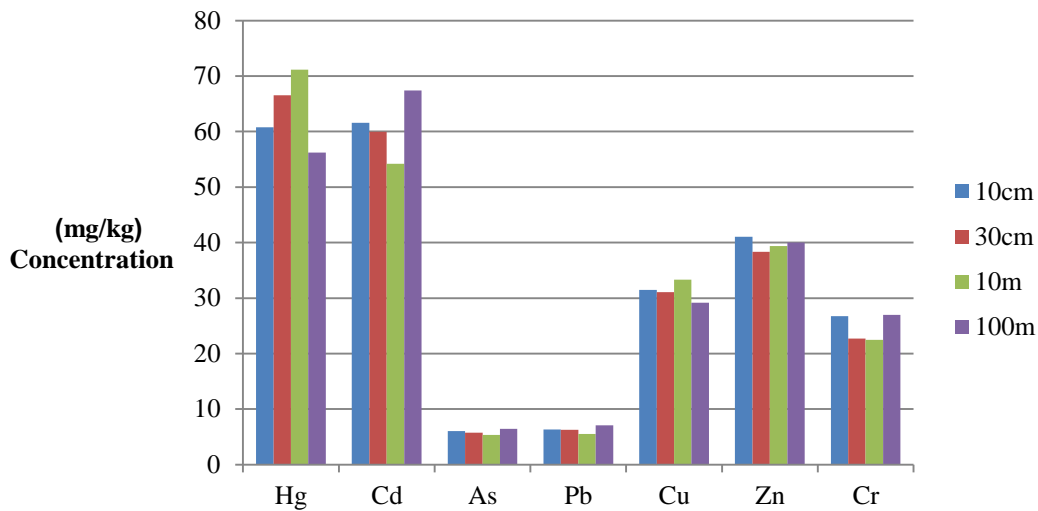
concentrations. The mean concentration of all heavy metals in the soil samples around the MWLs and hospital waste of Sabzevar city was higher than the background control. In the present study, the mean concentrations of As, Fe, Zn, Pb, Cr, and Cu in the surface soil were 6.013, 20854.49, 41.04, 6.31, 26.77, and 31.45 mg/kg, respectively. Besides, levels of Hg and Cd were 60.79 and 61.60 µg/kg, respectively. However, the mean concentrations of As, Fe, Zn, Pb, Cr, and Cu in the deep soil were 5.75, 21088.90, 38.33, 6.25, 22.68, and 31.04 mg/kg, respectively, whereas the levels of Hg and Cd in deep soil were, respectively, 66.57 and 59.98 µg/kg.

#### Enrichment factor

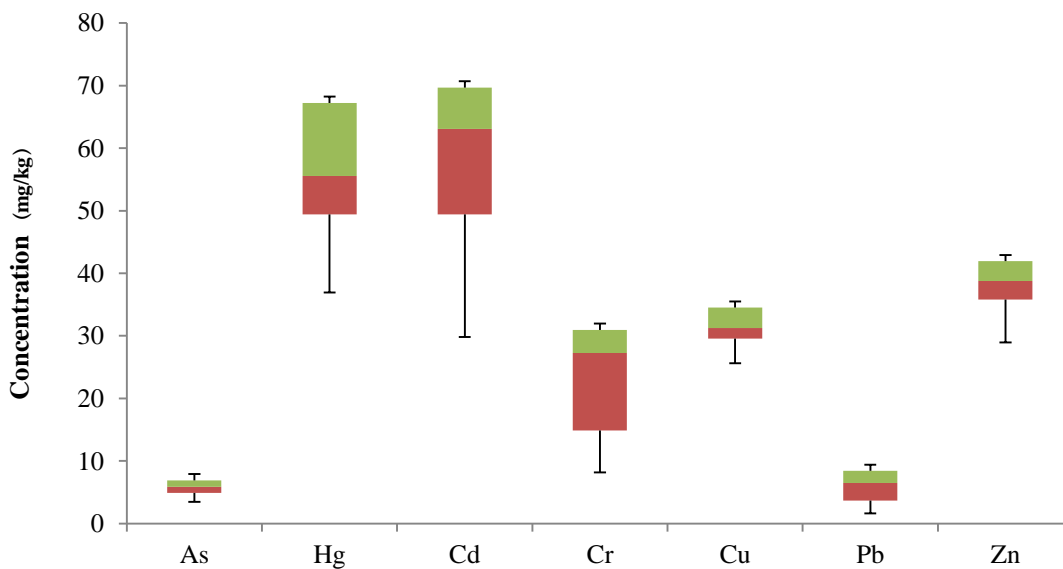
The mean EF for Cd and Hg was found to be 1818.88 and 390.82, respectively, indicating extremely high EF with human source, while the value of EF was moderate for As (2.13) and low for other metals (< 1). The mean EF pattern for the study heavy metals according to Table 5 was as follows: Cd > Hg > As > Cr > Cu > Zn > Pb.

**Geoaccumulation index**

The distribution pattern of heavy metals based on the mean  $I_{geo}$  was Hg > Cd > As > Cu > Zn > Pb > Cr. The mean  $I_{geo}$  values for Hg and Cd were 7.03 and 9.29, respectively, which was classified as highly polluted. However, the  $I_{geo}$  for As, Pb, Zn, Cr, and Cu in the target study soil were 0.41, 1.44, 0.98, -2.78, and -0.50, respectively, indicating no heavy metal pollution in the target soil.



**Figure 2:** Relationship between concentrations of heavy metals, Hg, Cd, As, Pb, Cu, Zn, and Cr with soil depth and the distance from the waste landfill (values for Hg and Cd are presented as  $\mu\text{g}/\text{kg}$ )



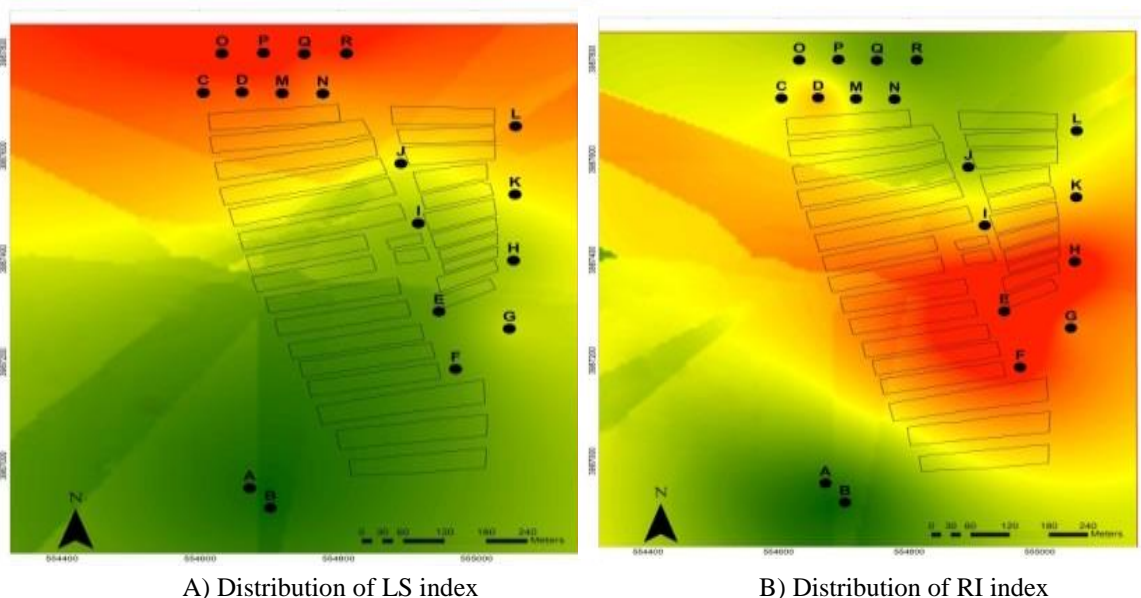
**Figure 3:** Distribution of heavy metal sampling concentration in waste sampling stations C to R (concentration for Hg and Cd are presented as  $\mu\text{g}/\text{kg}$ )



**Er index and total ecological risk elements**

As shown in Table 5, the mean distribution pattern of the Er was  $Hg > Cd > As > Cu > Pb > Zn > Cr$ , respectively. The mean values of Er for Hg, and Cd were 82278.62 and 29551.46, respectively, which was very high. However, mean values of As, Pb, Zn, Cr, and Cu in the target soil was 11.19, 3.11, 0.77, 0.48, and 4.97, respectively, indicating no heavy metal pollution. Despite the low mean Er for As, Pb, Zn, Cr, and Cu, the mean total ecological RI was shown to be very high for all seven elements, while the mean RI was lower for As, Pb, Zn, Cr, and Cu. By calculating RI, which simultaneously takes into account the

pollution status of several metals in estimating the pollution level, the soil of the study area was estimated highly polluted. Zoning of RI distribution for all the heavy metals of As, Hg, Pb, Zn, Cd, Cr, and Cu in the soil of the target area by conventional kriging method using ArcGIS software showed that E, F, G, and H stations were the most polluted parts of the study area. Evaluation of the spatial variation pattern using cumulative LS of As, Hg, Pb, Zn, Cd, Cr, and Cu through conventional kriging estimator in ArcGIS showed that by increasing the distance from the waste landfill, the severity of pollution gradually increased (Figure 4).



**Figure 4:** Zoning of RI distribution and spatial variations of cumulative LS of heavy metals in the soils surrounding Sabzevar MWLs. This figure shows RI and spatial variations of cumulative LS for As, Hg, Pb, Zn, Cd, Cr, and Cu in the soils around the Sabzevar MWLs. The typical kriging estimator by ArcGIS software was applied to measure these indicators.

**Table 5:** The values of EF, Igeo, and Er for heavy metals, As, Hg, Pb, Zn, Cd, Cr, and Cu in samples taken around MWLs of Sabzevar city in 2019

	As	Hg	Pb	Cr	Cd	Cu	Zn
EF	2.13	390.82	0.01	0.46	1818.88	0.02	0.01
I <sub>geo</sub>	0.41	7.03	1.44	-2.78	9.29	-0.50	0.98
Er	11.19	82278.62	3.11	0.48	29551.46	4.97	0.77

**Non-carcinogenic risk assessment**

HQ was used as a risk criterion to calculate the mean values of selected elements in the soil samples surrounding the Sabzevar MWLs through ingestion, dermal contact, and inhalation (Table

3). Table 6 shows the average HI and HC for soil samples collected from Sabzevar MWLs in 2019. The highest and lowest non-carcinogenic risk in children age group for all heavy metals As, Cd, Cr, Cu, Hg, Pb, and Zn were ingestion pathway,

dermal contact, and inhalation, respectively. The non-carcinogenicity risk of heavy metals in adults was as high as in the children. The non-carcinogenicity risk through dermal contact, inhalation, and ingestion was higher among the children. The non-carcinogenicity risk through ingestion and dermal contact in all the samples was  $4.52 \times 10^{-5}$  to  $1.39 \times 10^{-1}$  and  $2.26 \times 10^{-6}$  to  $2.81 \times 10^{-4}$ , respectively. However, the non-carcinogenicity risk through the respiratory tract was  $3.65 \times 10^{-15}$  to  $5.17 \times 10^{-11}$ . Among the examined heavy metals, Cd had the highest non-carcinogenic potential through ingestion among both adults and children, and Cr had the highest risk for both age groups through dermal contact and inhalation pathways. The lowest non-carcinogenicity risk in children and adults was caused by Hg through ingestion. Besides, the lowest risk through dermal contact and inhalation pathways was related to Zn and Cd for both age groups, respectively. The HQ for the heavy metals through all the three pathways was estimated less than 1, thus they will not have harmful impacts on human health.

The HI values in the MWLs samples were calculated by obtaining the total HI through ingestion, dermal contact, and inhalation. Table 3 reveals that the values of the non-carcinogenic HI of the total absorption pathways among children

were higher than adults. The HI of all the heavy metals in the target soil was found to be  $2.04 \times 10^{-1}$  for children and  $5.48 \times 10^{-3}$  for adults, indicating higher heavy metal exposure in children than in adults, which was supported by the results of previous studies. The total HI of all the absorption pathways in the two age groups of children and adults were as follows: Hg < Zn < Cu < Pb < Cr < As < Cd. Total amounts of heavy metals were less than 1.

#### Carcinogenic risk assessment

Due to the lack of cancer slope factors (CSF) for different elements in this study, the carcinogenic RI value was calculated only for As, Cd, Cr, and Pb (Table 5). RI values for As, Cr, and Cd by inhalation were in an observable range ( $7.78 \times 10^{-8}$ ). The carcinogenic risk for As through ingestion and Cr through ingestion and dermal contact was more than  $1 \times 10^{-6}$ . This suggests that exposure to soil polluted with As and Cr may have adverse health effects, especially among children. In terms of carcinogenic risk, the soil of the target region was in the range of tolerable rate to human health. Cr was identified as the main threatening element to human health, thus, more attention should be paid to the region due to the level of carcinogenic hazards.

**Table 6:** Mean HI and HC for the soil samples collected from Sabzevars MWLs in 2019

Elements	HQ <sub>ing</sub>		HQ <sub>inh</sub>		HQ <sub>dem</sub>		HI		RI		
	Adult	Child	Adult	Child	Adult	Child	Adult	Child	Ingestion	Inhalation	Dermal Contact
As	$1.12 \times 10^{-3}$	$4.18 \times 10^{-2}$	$2.57 \times 10^{-13}$	$2.86 \times 10^{-12}$	$8.31 \times 10^{-6}$	$1.63 \times 10^{-4}$	$1.13 \times 10^{-3}$	$4.19 \times 10^{-2}$	$1.38 \times 10^{-5}$	$1.89 \times 10^{-12}$	$6.85 \times 10^{-8}$
Cd	$3.47 \times 10^{-3}$	$1.29 \times 10^{-1}$	$3.27 \times 10^{-16}$	$3.63 \times 10^{-15}$	$2.11 \times 10^{-7}$	$4.14 \times 10^{-6}$	$3.47 \times 10^{-3}$	$1.29 \times 10^{-1}$		$3.83 \times 10^{-12}$	
Cr	$4.7 \times 10^{-4}$	$1.75 \times 10^{-2}$	$4.64 \times 10^{-12}$	$5.17 \times 10^{-11}$	$1.43 \times 10^{-5}$	$2.81 \times 10^{-4}$	$4.85 \times 10^{-4}$	$1.78 \times 10^{-2}$	$1.94 \times 10^{-5}$	$7.78 \times 10^{-8}$	$1.57 \times 10^{-6}$
Cu	$4.46 \times 10^{-5}$	$1.66 \times 10^{-2}$	$4.2 \times 10^{-15}$	$4.70 \times 10^{-14}$	$4.53 \times 10^{-7}$	$8.88 \times 10^{-6}$	$4.5 \times 10^{-5}$	$1.67 \times 10^{-3}$			
Hg	$1.21 \times 10^{-6}$	$4.52 \times 10^{-5}$	$3.99 \times 10^{-15}$	$4.44 \times 10^{-14}$	$5.27 \times 10^{-7}$	$1.03 \times 10^{-5}$	$1.74 \times 10^{-6}$	$5.56 \times 10^{-5}$			
Pb	$3.45 \times 10^{-4}$	$1.29 \times 10^{-2}$	$9.58 \times 10^{-15}$	$1.07 \times 10^{-13}$	$2.08 \times 10^{-6}$	$4.09 \times 10^{-5}$	$3.47 \times 10^{-4}$	$1.29 \times 10^{-2}$	$8.36 \times 10^{-8}$		
Zn	$7.55 \times 10^{-6}$	$2.82 \times 10^{-4}$	$7.11 \times 10^{-16}$	$7.91 \times 10^{-15}$	$1.15 \times 10^{-7}$	$2.25 \times 10^{-6}$	$7.67 \times 10^{-6}$	$2.84 \times 10^{-4}$			

#### Discussion

In the present study, the amount of soil pollution in the study area was determined, using EF,  $I_{geo}$ , and Er. Also, to better identify the polluted areas or areas exposed to these pollutants, the general scheme of soil pollution was drawn using the total

ecological risk values of the total elements of heavy metals in the surface soils of the region and the LS method for integrating single maps of heavy metals concentrations, such as AS, Hg, Pb, Zn, Cd, Cr and Cu using a conventional kriging estimator in ArcGIS software.

According to the results of heavy metal concentrations, by increasing sampling depth from 10 to 30 cm, the heavy metals concentrations decreased except for Hg. This result is consistent with the results of heavy metal contamination in soil and groundwater in Nigeria conducted by Saheed et al.<sup>8</sup>. They found that heavy metals in landfill leachate move along deeper layers and around the soil<sup>25</sup>. Comparing the mean total concentration of all the samples except Mn in the whole study area with threshold values, it was observed that the mean concentration of all the heavy metals except As in the Sabzevar MWLs was less than the threshold values. Nevertheless, to determine the amount of soil pollution exposure to heavy metals and to determine the origin of elements in the soil, the concentration of elements in the region could be compared with the amount of elements in the earth's crust and the global mean levels of heavy metals in soils. Comparison of the amounts of elements in the study area with their values in the earth's crust and global values of these elements showed that the mean concentration of As, Hg, and Cd in the soil was higher than the global mean concentration. The mean concentrations of all the mentioned heavy metals were higher than the natural background values for each heavy metal in the Sabzevar MWL soils, indicating the role of soil pollution by human activity of non-engineered waste landfills, which is consistent with the results of Egwu et al.<sup>26</sup>.

According to the results of the present study, the mean values of EF,  $I_{geo}$ , and ER index for As, Pb, Zn, Cr, and Cu in the soil of the study area, showed no pollution. However, due to disposal of medical waste in trenches near stations E, F, G, and H, the study area was estimated to be very contaminated with Hg and Cd heavy metals, due to the high man-made origin of these two heavy metals in non-ferrous plastics. This issue was investigated by Jafari et al. in the study of heavy metals downstream of Ardabil MWLs<sup>27</sup>. The study of spatial variation pattern of cumulative LS of all the studied heavy metals, including As, Hg, Pb, Zn, Cd, Cr, and Cu in the soil of the study area showed that due to the slope of the landfill to Sabzevar

city, by increasing the distance from the waste landfill, the severity of pollution gradually increased.

In all the elements, the distribution of HQ and HI for both groups of adults and children, from different pathways were as follows: ingestion > dermal contact > inhalation. Therefore, the direct pathway "soil ingestion" had the highest level of HI and HQ for children and adults and for all the studied heavy metals, which is consistent with the results of previous studies<sup>15, 28-29</sup>. Non-food or sucking on hands and fingers by children was mentioned in a study by Wei et al. who assessed the health risks of heavy metals in dust in China's industrial areas<sup>30</sup>, the results of which were consistent with the results of Qing et al. in China<sup>31</sup>. In general, the carcinogenic RI in all the three pathways in the soils around the Sabzevar MWLs indicated good human health, while the carcinogenic risk in a review study in India for Cr and As was within a relatively unacceptable range<sup>14</sup>.

The results obtained in this study can be compared with other similar studies in this field.

Mukhopadhyay et al. examined the soil of the Kolkata landfill in India to assess heavy metal pollution. According to the  $I_{geo}$ , the intensity of soil pollution was different for each heavy metal, so that the severity of pollution was unpolluted for Hg, slightly polluted for Mn and As, slightly polluted for Cr, Zn, and Cu, and slightly polluted to severely polluted for Pb. The EF index showed that the severity of pollution was low for heavy metals of Hg, Mn, and As, while moderate for Cr and high for Zn, Cu, and Pb. Also, the evaluation of RI of the elements showed a moderate risk of pollution for all the elements in the soil around the landfill, and the level of ER for Zn, Mn, Cr, As, Cu, and Pb was estimated low but moderate risk for Pb and Hg, and the most important heavy metals were Hg and Pb, respectively<sup>19</sup>.

Thongyuan et al. conducted a study in the area around MWLs in Thailand, by examining pollution indices and calculating the concentration of heavy metals in the region. They found that the mean concentrations of heavy metals were as follows: Al

> Fe > Mg > Mn > Zn > Cu > Bi > Cr > Pb > Li > Ni > Co > Ga > Cd. The severity of soil pollution with heavy metals was also confirmed using the  $I_{geo}$  for Pb, Ni, Co, and Cr ranging from unpolluted to slightly polluted, whereas for Zn, Cd, and Cu, it was highly polluted to severely polluted and severely polluted for bismuth. In the study of soil pollution risk with Er index, contamination for Mn, Pb, and Cr showed low hazard, while for Ni, Co, Zn, and Cu, the considerable hazard was estimated and Cd was the most important ecological risk factor. However, according to the cancer RI, there was no carcinogenic risk to human health in the two age groups of children and adults for the Cd and Ni, but the dominant role in soil pollution related to cancer risk was first dependent on Cr followed by Pb. The carcinogenic risk for these two metals was acceptable. The main pathway led to cancer risk was Cr ingestion, but the highest risk for the Pb was through dermal contact. It was estimated that exposure to Pb through the dermal contact could be identified as an occupational risk associated with cancer risk in landfill workers<sup>24</sup>. Alam et al. examined the soil of the Mogla Bazar landfill in Bangladesh to assess the pollution of heavy metals in water, soil, and plants around an open landfill. Based on the  $I_{geo}$  for Cu, Mn, Zn, and Fe around the landfill, there was no pollution, while for Pb and Cd, the severity of soil pollution was estimated from unpolluted to slightly polluted<sup>11</sup>.

Akoto et al. evaluated various heavy metals at two non-engineered landfills in Aboabo and Santasi in Kumasi, Ghana, and found that ingestion of soil particles was a major source of getting polluted with Pb, Cr, Co, Zn, and Fe. The inhalation and dermal contact of heavy metals had little or no effect on the health of children living surrounding the landfills. Pb and Fe had the highest risk of non-carcinogenicity, while Zn had the lowest risk, and Pb toxicity was reported to cause abdominal pain, loss of appetite, seizures, brain damage, and death in children. However, Fe toxicity may lead to vomiting, fever, diarrhea, and stomach pain in children<sup>28</sup>. Given that there are limited number of studies on metal levels in human

body management and tissue sampling, it is recommended that tissue research be conducted on the level of metals in the human body.

### Conclusion

The results from calculating EF, land accumulation, and Er indices showed that according to the proposed classifications, pollution in the soils of Sabzevar municipal and medical landfills is very high. Examination of the distribution map of the total ecological RI of the soils around Sabzevar landfills revealed that the highest severity of contamination in the region was related to sampling stations near landfills with high concentrations of Hg and Cd. The pattern of spatial changes with cumulative LS of As, Hg, Pb, Zn, Cd, Cr, and Cu in the study soil area showed that by increasing distance from the landfill because of alignment with the slope of the land to Sabzevar, the severity of the infection increased. Health risk assessment showed that heavy metal contamination in the soils around the landfill of Sabzevar city is below the acceptable threshold for carcinogenic and non-carcinogenic hazards. The HQ and HI values for metals through ingestion, inhalation, and dermal contact were less than one, indicating that there is no health risk in the current situation. In all the elements, IR and HQ values for both groups of adults and children were from different pathways, such as ingestion > dermal contact > inhalation. Therefore, the most important pathway of exposure to heavy metals for children and adults is ingestion. According to the findings of this study, it can be concluded that there is heavy metal contamination caused by municipal waste in the soil of the Sabzevar landfill.

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## Conflict of interest

The authors declare that there is no conflict of interest.

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