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# Pollution assessment, spatial distribution and exposure of Cd and Pb in surface soils of abandoned landfill site in Gorgan, north of Iran

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### Abstract

The purpose of the present research was Cadmium (Cd) and Lead (Pb) pollution assessment and monitoring the spatial distribution of soil contamination around the municipal landfill site in southwest of Gorgan, Iran. Thus, in a systematic network with a distance of 250m, from 32 stations three samples of surface soil (0 to 20cm) were obtained and mixed together. In addition, three samples away from the landfill with similar characteristics and geology were used as control. The concentration of heavy metals for the samples was determined using nitric acid and hydrochloric acid and atomic absorption spectrometry. Mean concentrations of Cd and Pb in the soil around the landfill site were 0.37 and 17.31 mg/kg respectively. The total concentration of Cd and Pb was higher than the control areas. There was a positive correlation between Cd and Pb showing these elements in the soil are from the same origin. Based on our research, soil in the landfill area has a moderate ecological risk index, which is about 120. The central parts of the study area have higher concentrations of Cd and Pb than southern parts due to the slope and runoff of the waste leachate. In general, there was an increase in the concentration of Cd and Pb in the sampling sites compared to the control site.

Keywords: Soil pollution, Cd and Pb, Pollution assessment criteria

## Introduction

The Gorgan landfill site is located in the west of the city in Hezarpich hill which is also a tourist destination due to its panoramic view of the city and the surrounding landscapes. The cool climate is always welcomed by tourists and even residents of different cities of the province. Apparently, there has been a conflict between tourism and waste deposition in the area leading to pollution and downgrading of the area for other uses. Studying and resolving these problems is hampered more because of the budget and time constraints in most management decisions (Ebrahimi et al., 2011). In the last decades, an increase in the population, industrialization and urbanization has led to the degradation of the quality of environment and soil in the area. The soil is polluted heavily by metals causing a major concern because of pollution effects on flora, fauna and the humans (Shaheen and Iqbal, 2018; Khaledian et al., 2011). Human can cause activities air and soil contamination and solid waste can be regarded as one of the major sources of pollution. For instance, in Asian developing countries, the promotion of solid waste management is difficult and there is a shortage of areas pinpointed for final disposal (Forti et al., 2019). The spread of disease and environmental pollutions from the landfill area is rampant in most mismanaged areas. Leachate containing heavy metals can easily migrate to belowground further spreading of the pollution. The consumption of polluted agricultural produce with heavy metals is a means of transferring metals to food.

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Cadmium, Chromium, Copper, Lead, Nickel, and Zinc are among the most hazardous heavy metals in these areas (Vongdala et al., 2019). The leachates that are produced from man-made waste can pollute the environment contributing to diseases such as asthma, cancer, skeletal deformities and convulsion (Boateng et al., 2019). Phosphorous (P) and Potassium (K) are essential nutrients for living organisms while Cadmium (Cd) and Lead (Pb) can exhibit harmful effects during uptaking. Cd is responsible for causing cancer in humans while Pb is regarded as neuro-toxicant and teratogen (Ackah, 2019). Metals in the sediment and soils are originated from Earth's crust and human activities and can be measured with the aid of a diverse range of pollution indices. It is also possible to determine if their source is natural or anthropogenic. In addition to analysis, processing, and conveying the environmental information to relevant bodies, pollution indices are also useful and necessary for the technicians and analysts (Martínez-Guijarro et al., 2019). Presently, Geographical Information Systems (GIS) and geo-statistics are used in studies of spatial variation and risk assessment of soils heavy metals (Zhao et al., 2019). Kriging is one of the popular methods to interpolate and identify the polluted areas (Zhao et al., 2020; Khaledian et al., 2018). The study area has been used for domestic landfill for many years.

The purpose of this research was: 1) to investigate the current status of the study site by monitoring the changes of the pollutants in the landfill site, 2) to evaluate the commination rate by Cadmium and Lead and its severity using criteria such as geo-accumulation index ( $I_{geo}$ ), Enrichment factor (*Er*), pollution load index (*PLI*) and Potential Ecological Risk Index (*PERI or RI*), and generate pollution maps.

# Materials and methods *Study area*

The research was conducted in Hazarpich area which is in the west of Gorgan, Iran (Figure 1).

# Soil sampling

Soil sampling applied was systematically for 32 locations. At each location 3 surface soils sampling were considered. The total amount of soil for each location was 1.5kg, which was mixed. For this purpose, a systematic 25×25m grid with 250×250m intervals in the soil surrounding the landfill site was designed using ArcGIS software and the grid crossing sites were considered as sampling points. The geographical coordinates of each sample were recorded by the Global Positioning System (GPS). In addition, we took three samples away from the landfill with similar characteristics and geology as reference or control samples.



Figure 1. The study area and soil sampling spots

## Soil analysis

To measure the total concentration of heavy metals, 0.25g soil samples obtained from 0.50 mm sieve were transferred to digestion tubes. Hydrochloric acid and nitric acid were used to digest the samples. The contents of the digestive tubes were heated at 105°C for 1 hour and at 140°C for digestion of the samples (McGrath and Cunliffe, 1985). The total concentration of heavy metals was measured by atomic absorption spectrometry (VARIAN-AA240), and the physical-chemical parameters of the soil including reaction (pH) (McLean, 1982), electrical conductivity (EC) (Page et al., 1992), soil texture (Gee & Bauder, 1990), organic carbon (OC) (Nelson & Sommers, 1996) and calcium carbonate (CaCO<sub>3</sub>) (Richards, 1954) were measured.

# Pollution Assessment Methods for Heavy Metals

Proper indices and indicators of contamination should be used for

Table 1. Classes of Geo-Accumulation Inde	Х
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effective assessment of soil contamination with heavy metals as a guide and reference for geochemical assessments of soils. Indices are useful for the estimation of environmental risk and soil degradation. In addition, the indices help to find the origins of heavy metals be it natural or humanmade (Weissmannová and Pavlovský, 2017).

One of the indices that is used for the assessment of soil pollution is geoaccumulation index as the ratio of the polluted soils with heavy metals to its natural state. To detect anthropogenic effects and the natural fluctuations of metals, a constant 1.5 is multiplied with the index (Muller, 1969; Loska et al., 2003; Ji et al., 2008; Lu and Bai., 2010). The geoaccumulation index has seven grades (Table 1).

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

C<sub>n</sub>: Concentration of individual heavy metal B<sub>n</sub>: Value of geochemical background and 1.5-constant

Index	Value	Soil quality	
Igeo	$I_{geo} \leq 0$	Uncontaminated	
	$0 \le I_{geo} < 1$	Uncontaminated to moderately contaminated	
	$1 \leq I_{geo} < 2$	Moderately contaminated	
	$2 \le I_{geo} < 3$	Moderately to strongly contaminated	
	$3 \leq I_{geo} < 4$	Strongly contaminated	
	$4 \le I_{geo} < 5$	Strongly to extremely contaminated	
	$I_{geo} > 5$	Extremely high contaminated	

The Enrichment Factor (FE) as a ratio of tested metal to a natural background soil consists of five classes (Table 2), (Sutherland, 2000), and is calculated based on the following formula:

 $EF = \frac{\binom{C_n}{C_{Ref}}sample}{\binom{B_n}{B_{Ref}}background}$ Where C<sub>n</sub>: content of the examined element in the examined environment,

 $C_{ref}$ : content of the reference element in the examined environment,

 $B_n$  (background): the amount of the examined element in the reference environment, and

 $B_{ref}$  (background): the amount of the reference element in the reference environment (Loska et al., 2004).

calculated as follows:

single pollution index.

 $\mathbf{PLI} = \sqrt[n]{\mathbf{PI}_1 \times \mathbf{PI}_2 \times \mathbf{PI}_3 \cdots \mathbf{PI}_n}$ 

Index	Value	Soil quality
EF	EF < 2	Deficiency to minimal mineral enrichment
	EF = 2 - 5	Moderate enrichment
	EF = 5 - 20	Significant enrichment
	EF = 20 - 40	Very high enrichment
	EF > 40	Extremely high enrichment

Table 2. Classes of Enrichment Factor

Pollution Load Index (PLI) is used to describe the heavy metals concentration of soil. If the PLI is almost 1, it means the heavy metal continuation is similar to its background; however, if the PLI value is greater than 1, it denotes soil pollution (Table 3), (Liu et al., 2005). To asses the site quality, the PLI

Table 3. Classes of Pollution Load Index

Index	Value	Soil quality
PLI	PLI < 1	Not polluted
	PLI = 1	Baseline levels of pollution
	PLI > 1	Polluted

Based on 1) single index of ecological risk factor  $(E_r^i)$ , 2) pollution coefficient of a single element  $(C_f^i)$ , and 3) toxic response factor of a metal  $(T_r^i)$  the Potential Ecological Risk Index (*PERI*, or *RI*) can be calculated. The toxic response factors for Pb and Cd are 5 and 30 respectively (Yuan et al., 2014). The RI is calculated using the following formula:

$$RI = \sum_{i=1}^{n} E_r^i$$

where, n: the number of heavy metal,  $E_r^i$ : ecological risk factor of single index,  $\mathbf{E}_r^i = \mathbf{T}_r^i \times \mathbf{C}_r^i$ 

provides a comparative means and is

where n-the number of analyzed heavy

metals, and PI-calculated values for the

where,  $T_{r}^{i}$  toxicity coefficient response of a metal,  $C_{f}^{i}$  factor of contamination.

According to the potential ecological risk index, soils can be classified into five classes (Table 4).

Table 4	<b>.</b> Cl	lasses	of	Potenti	al	Ecol	log	ica	l R	isk	Inc	lex
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Index	Value	Index	Value	Ecological Risk
Er	Er < 40	RI	RI < 90	Low potential ecological risk
	40 < Er < 80		90 < RI < 180	Moderate potential ecological risk
	80 < Er < 160		180 < RI < 360	Considerable potential ecological risk
	160 < Er < 320		360 < RI < 720	Very high potential ecological risk
	Er > 320		$RI \geq 720$	Extremely potential ecological risk

## **Data Analysis**

We used SPSS software for descriptive statistics of heavy metals in soil and other chemical properties including mean, median, standard deviation, min, max, range, coefficient of variation, kurtosis and skewness, and coefficient of correlation. The distribution pattern of all parameters of soil was estimated based upon the standard deviation and coefficient variation values. We considered skewness in the range -1 and 1 showing a normally distributed dataset (Shaheen and Iqbal, 2018).

### **Pattern Analysis**

A number of research papers and books can be found in the literature that explain why and how to conduct geostatistics and its various methods such as semi-variogram and kriging (Goovaerts, 1997; Webster and Oliver, 2001). In this research, kriging was used to interpolate heavy metal values in the study area. The kriging is a special method for calculating the spatial variation characteristics more effectively from known data and semi-variance function (Zhao et al., 2020) and then to interpolate the point data to across space.

## Results and discussion Descriptive Statistics

According to Table 5, the percentage of calcium carbonate ranged from 4.5 to 35% with a mean 15.9%. Calcium carbonate of soil affects the adsorption of heavy metals indirectly and at the same time it affects the soil reaction as well (Smith, 1968; McBride, 1980; Martin et al., 2006). Organic carbon is one of the most important soil quality indicators which plays an important role in the soil nutrient cycle (Rattan et al., 2005). Organic carbon in the area was found to be affected by the landfill

in the range 0.23 to 2.77 percent with a mean 1.3 percent. Usually the amount of organic carbon is directly proportional to the contamination of heavy metals in the soil, so if there is an increase in the amount of organic carbon, there will be an increase in the concentration of heavy metals as well (Camobreco et al., 1996; Mirsal, 2008). The electrical conductivity was between 0.54 to 3.78 dS/m with a mean 1.1 dS/m. In soils with low electrical conductivity, the plant absorbs less heavy elements (Mirsal, 2008). The soil reaction was in the range 7.52 to 8pH with a mean 7.8pH. The risk of heavy metal contamination in high reaction soils is low (Mirsal, 2008). Mean percentages of clay, silt, and sand particles were 34.7, 45.9 and 19.4 respectively. Clays generally have a high concentration of heavy metals due to their ability to absorb metal ions (Alloway, 2013).

Table 5. Statistical summary of the basic soil properties and element concentrations

Variable	Lime	OC	EC	pH	Clay	Silt	Sand	Cadmium	Lead
Minimum	4.50	0.23	0.54	7.52	5.60	36.80	13.60	0.10	7.00
Maximum	35.00	2.77	3.78	8.00	45.60	66.80	31.60	0.80	86.00
Mean	15.90	1.32	1.11	7.81	34.66	45.86	19.48	0.37	17.31
Range	30.50	2.54	3.24	0.48	40.00	30.00	18.00	0.70	79.00
Median	13.40	1.26	0.83	7.85	35.60	44.80	18.60	0.40	14.50
Std. Deviation	7.67	0.67	0.75	0.11	7.47	6.20	4.00	0.16	13.97
CV	48%	51%	68%	1%	22%	14%	21%	43%	81%
Skewness	0.79	0.35	2.41	-1.19	-2.08	1.50	1.56	0.18	4.20
Kurtosis	-0.05	-0.46	5.37	1.34	6.79	3.78	2.71	0.86	19.90
Background value								0.10	9.00

The average concentrations around the landfill site for Lead and Cadmium were 17.31 mg/kg and 0.37 mg/kg respectively and Lead had much higher concentration than Cadmium. Cadmium has the lowest standard deviation and range of variation among heavy metals due to its low concentration and low dispersion (Sollitto et al., 2010). The global degree of variability for variables can be described through Coefficient of Variation (CV). If the CV value is smaller than 10%, it is considered as low variability; however, if the CV is greater than 90%, it is considered as extensive variability (Zhao et al., 2020). It can be seen from table 5 that Lime, OC, clay, silt, sand, and Cd have a moderate variability while the Pb shows an extensive variability. pH has a week variability in the study area which is just 1%. It was found that the CVs for Cd and Pb were higher than 10% showing the effects of human activities (Liu et al., 2013).

#### Correlation

According to Table 6, we can see that there are significant relationships between Cd and Pb (r=580, p<0.01) indicating Cd and Pb are derived from the same origin. However, Cd and Pb were not significantly correlated to Lime, OC, EC, pH, clay, silt, or sand (p<0.01 and p<0.05).

Variable	Lime	OC	EC	pН	Clay	Silt	Sand	Cadmium	Lead
Lime	1								
OC	-0.423*	1							
EC	$0.362^{*}$	-0.126	1						
pН	-0.347	0.076	-0.858**	1					
Clay	-0.471**	0.160	-0.557**	$0.541^{**}$	1				
Silt	0.233	-0.067	0.332	-0.385*	-0.845**	1			
Sand	$0.518^{**}$	-0.196	$0.526^{**}$	-0.413*	-0.557**	0.026	1		
Cadmium	-0.216	0.265	-0.264	0.135	0.080	-0.145	0.077	1	
Lead	-0.130	0.186	-0.091	-0.030	0.118	-0.142	0.000	$0.580^{**}$	1

Table 6. Pearson's correlation between the parameters

\*. Correlation is significant at the 0.05 level, \*\*. Correlation is significant at the 0.01 level.

#### **Pollution Assessment**

In Table 7, the percentages of class distribution for pollution assessment of Cd and Pb are shown using  $I_{geo}$ .  $I_{geo}$  has different values for Cd and Pb with various levels of contamination such as uncontaminated, moderate, and highly contaminated. Moreover, it is noticed that 87.5% of the landfill site has  $I_{geo}$  greater

than 1 for Cd which suggests a moderate level of pollution (Muller, 1969). From Table 1, it can be understood that the  $I_{geo}$  values for Cd and Pb lay above the moderate level of concentration in 59.4% and 9.4% of samples respectively. There is a consistency between these findings and other research (Fonge et al., 2017).

Table 7. Evalua	ation results of	Geo-Accumu	lation Index
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$I = alogg(\theta/)$	Elements			
Igeo Class (70)	Cd	Pb		
Uncontaminated	12.5	46.8		
Uncontaminated to moderately contaminated	28.1	43.8		
Moderately contaminated	56.3	6.3		
Moderately to strongly contaminated	3.1	3.1		
Strongly contaminated	-	-		
Strongly to extremely contaminated	-	-		
Extremely high contaminated	-	-		
Mean	1.13	0.15		
Minimum	-0.58	-0.95		
Maximum	2.42	2.67		

#### Table 8. Evaluation results of Enrichment Factor

EE aloss (9/)	Elements			
ET Class (70)	Cd	Pb		
Deficiency to minimal mineral enrichment	-	25		
Moderate enrichment	18.8	68.8		
Significant enrichment	81.2	6.2		
Very high enrichment	-	-		
Extremely high enrichment	-	-		
Mean	7.38	2.99		
Minimum	2.00	1.21		
Maximum	16.00	14.86		

The Enrichment Factor (EF) index of the soils depicts the degree of human activities. Based on Table 8, we see that the mean EF values of Cd and Pb have been 7.38 and 2.99 respectively. The mean EF for Cadmium and Lead were higher than 2, which implies human activities have caused these metal levels in the soils of the study area (Desaules, 2012). It is seen that 75% of soil's EF for Cd and Pb is higher than 2 connoting moderate to high pollution that is consistent with other research (Adelopo et al., 2018; Essien et al., 2019). Cd and Pb with higher EF values indicate that the metals are from human activities and most

of them come from waste disposal in the landfill site.

The pollution load index of Cd and Pb for soils were considered in a combined form using Table 9. The pollution load index indicates the extent and infiltration of heavy metals in the soil samples (Fonge et

Table 9. Pollution Load Index for the study area

PLI class (%)ElementsNot polluted3.1Baseline levels of pollution-Polluted96.9Mean2.58Minimum0.88Maximum8.74

According to Table 10, the mean value of  $E_r$  for Cd was 110.63 which belongs to the considerable potential ecological risk level. Most of the samples obtained from points for Cd were at considerable potential ecological risk level that was consistent with the results of other studies (Essien et

al., 2019; Zhao et al., 2020). The mean  $E_r$  for Pb is less than 40 which is regarded as low potential ecological risk level. The mean *RI* was 120.24 which demonstrates that the soil in the study area is at moderate ecological risk.

al., 2017). This can be explained by the

high concentrations of Cd and Pb resulting

from the decomposition of assorted waste in the landfill. This finding is consistent

with the results of other researchers (Liu et

al., 2013; Rocco et al., 2016).

Table 10. Evaluation results of Potential Ecological Risk Index

$\operatorname{Er}$ along $(0/)$	Elem	DEDI/D	
EI Class (76)	Cd	Pb	PEKI/K
Low potential ecological risk	12.5	96.9	
Moderate potential ecological risk	6.3	3.1	
Considerable potential ecological risk	75	-	
Very high potential ecological risk	6.2	-	
Extremely potential ecological risk	-	-	
Mean	110.63	9.62	120.24
Minimum	30.00	3.89	33.89
Maximum	240.00	47.78	287.78



Figure 2. Spatial distribution of Cd and Pb in soils of the study area

#### **Distribution Pattern**

Kriging was conducted for Cd and Pb in the soils of the study area (Figure 1). The high contamination of Cd in soils were found from the center to the northwest and northeast. The low concentration was seen at the southwest in the studied area. Generally speaking, the high concentrations of Cd and Pb in soils are noticed in the center of the study area due to the slope and runoff of the waste leachate while low concentrations of these elements were recorded at the southern parts of the site.

## Conclusion

Hezarpich area in Gorgan has been a landfill for more than three decades, and is polluted with construction debris and domestic sewage. It seems that the widespread distribution of various pollutants for a long time has caused the spread of pollution in the region and the surrounding ecosystem. Considering the climatic conditions of the region and due to the fact that the landfill site is located in the highlands with relatively steep slopes, the need for studies and efficient management is essential. This study was designed to assess heavy metals pollution and monitor the spatial distribution of soil pollution around Gorgan (Hazarpich area) urban landfill

We found that the concentration of Pb was 17.31mg/kg while for the Cd it was 0.37mg/kg in the landfill area, which means that Pb has much higher concentration than Cd. Based on previous research, if the CV

is greater than 10% for Cd and Pb, we may as well postulate that the effects originate from human activities. It is seen that there are significant relationships between Cd and Pb (r = 580, p < 0.01), which means Cd and Pb are derived from the same source.

More than 75% of soils around the research site show a moderate to high level of concentration. In short, it can be said that the landfill site is highly polluted with Cd and Pb through waste decomposition. Based on the samples obtained from the study area, ecological risk factor for Cd was higher than 40, which is regarded as considerable potential ecological risk level, while Pb concentration showed low level of potential ecological risk. The study area almost reached a moderate ecological risk level with an average risk index of 120.24. The results showed that Cd and Pb in soils had moderate spatial autocorrelation mostly controlled by extrinsic factors. To sum up, the high concentrations of Cd and Pb in soils are noticed in the center of the study area due to the slope and runoff of the waste leachate whereas low concentrations of these elements were recorded at the southern parts of the site.

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