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ASSESSING THE RISK OF HEAVY ELEMENT DISTRIBUTION IN THE SURROUNDING SOILS OF FARAVARI ZINC PROCESSING INDUSTRY

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ABSTRACT

Heavy elements in agricultural soil pose a risk to human health. Assessing distribution of such elements in agricultural soils is an approach to determine contamination levels. In order to assess the risk of soil contamination, 19 points were selected in the lands surrounding the Faravari Zinc processing factory in the northwest of Iran. The samples were obtained from the depths of 15 to 25 cm using grade system sampling. Concentrations of lead and zinc were determined after acid digestion and using an atomic absorption instrument. For assessing soil contamination levels of lead, cadmium, and zinc in the activity zone of a single factory, the single-factor index, Nemero comprehensive pollution index and geoaccumulation index (I_{geo}), were used. The mean lead, cadmium, and zinc levels were found to be 153.4, 12.3, and 953.28 mg/kg, respectively, which were 2.05, 2.46, and 1.9 times greater than the corresponding Iranian standards for those elements. Assessment by I_{geo} and single-factor index in soil listed the elements, in increasing order of pollution levels, as Zn > Pb, Cd, and assessment by single-factor index alone returned similar results. Assessing soil quality by a comprehensive method showed that the samples S1, S7, S11, S15, and S16 were contaminated by high levels of heavy metals. Comparing the

average metal concentrations at a depth of 60 cm around the Faravai zinc factory with soil surfaces at 0 – 25 cm showed that the distribution of heavy metals is not due to anthropogenic sources. The results indicated that the zinc industry's activities constitute one of the most important factors in heavy metal distribution and diffusion in the area's soil.

Keywords: Heavy metals, Zinc industry, Geo-accumulation, Single factorindex, comprehensive pollution index.

INTRODUCTION

The highest amount of zinc ore used in Iran's zinc industry is obtained from Angouran Lead and Zinc Mine, 110 km west of the city of Zanjan. Due to the high lead and zinc yields from the mine, zinc-related industrial facilities have been concentrated in this province [1].

The majority of factories in Zanjan Province utilize hydrometallurgy techniques to obtain zinc from mineral ores. The ores are first transported into the factory, crushed, and processed by leaching. All impurities in the ore are separated as three type of separate filter cake and eventually exit as a disposal cake. The filter cake contains metals such as cadmium, cobalt, nickel, and lead, which are regarded as heavy, environmentally toxic elements (Table 1). The primary proportions of elements in these cakes are recycled due to their economic value.

Table 1 Average concentration of the elements in the zinc industry waste[1]

Cake type	Analysis (percentage) %				
	Zn	Co	Ni	Pb	Cd
Leach filter cake	5.03	0.010	0.02	4.710	0.05
Hot filter cake	5.21	0.020	0.51	0.040	0.06
Cold filter cake	19.88	0.001	3.00	0.493	4.85

A high percentage of the heavy elements in these filter cakes are in the forms of zinc sulfate, with a solubility coefficient of 57.7 g in 100 mL; cadmium sulfate, with solubility coefficient of 76.7 g in 100 mL; and lead sulfate, with solubility coefficient of 0.0404 g in 100 mL, at a temperature of 25°C[2][12]. The disposed cake filters are metal salts with acidic sulfate bases that are easily dissolved in water, and can thereby accumulate in water sources and permeate into agricultural soils [3][15]. Excessive accumulation of these elements in soil may not only change the quality of agricultural soils but can also be a factor for contamination diffusion into superficial and subterranean water bodies. The solubility of heavy metals in aqueous solutions permits their mobility and is regarded as a source of water and soil contamination [4][14]. Factors such as organic matter in soil, the cationic exchange capacity, and pH of the soil play important roles in heavy element displacement in soil [5][9]. The salt forms of the elements, such as carbonate, sulfate, and oxide salts in the environment also affect these elements' ionic strength and bio-availability[6][2].

Results obtained from studies on grain crops cultivated in agricultural soil irrigated with sewage polluted by heavy elements resulted in plants with alarmingly high concentrations of those elements [7] [10]. Most heavy metals are toxic and detrimental to human health. Lead and cadmium are among the top 10 most harmful elements according to a publication issued by the Agency for Toxic Substances and Disease Registry [8][6].

Dissimilar methods have been utilized to assess soils for heavy metal contamination. In order to evaluate the soil quality on rice cultivation farms in Exiang, China, 99 soil samples contaminated by heavy metals were obtained from depths of 0–20 cm. After analyzing the heavy element concentrations, the results were assessed using a single-factor index and comprehensive pollution index. Those results were then categorized based on levels of contamination [9] [11]. To do so, a region in Tianjin, a center of industrial facilities and mineral processing activities, was selected, and its sources of pollution were studied. The regional distribution of contamination and ecological risks were analyzed using an index developed from the Nemero pollution index and I_{geo} index. The results showed that zinc contamination levels were higher than the levels of the other 7 elements and that industrial activities and fossil fuels were the greatest contamination sources. Moreover, the research revealed that spatial analysis methods could be used as an approach to study human environmental perils in the future [10] [8]. Another viable approach for studying these risks is the heavy elements assessment model [11-12] [4, 6].

Using statistical methods in geo-chemistry such as factorial analysis and cluster analysis comprises one of the decision-making approaches for determining the similarities of major metal and element groups. This is done to identify joint sources that lie in the depth of the sediments and classify the regions geographically [13] [16].

The present research aims to determine the extent of heavy metal pollution in a 3-km radius of soil surrounding the Faravari zinc industry. The objectives of the study include distribution mapping of the contaminants; a qualitative pollution severity assessment of the soil using single-factor index, I_{geo}, and the Nemero comprehensive pollution index; and developing related data by using a statistical correlation coefficient and cluster analysis.

Materials and Method

The region considered in the present study consisted of a 3-km radius of land surrounding the Faravari zinc factory, with the geographical coordinates of 36°31'52” N 47°39'50” E.

Nineteen points were selected for sampling. In order to create a composite sample, the sampling area was divided into squares of 20 × 20 m², and 5 samples were obtained from each of the squares at their angles and centers at depths ranging between 0 and 25 cm deep. The 5 samples obtained from every single square were amalgamated, and one mixed sample was taken from every site. Sampling was conducted using a hand soil auger through grade system sampling [14][7]. A proper amount of soil in each sample was taken and put in an oven for two hours at 110°C. Once the samples were completely dehydrated, they were crushed, passed through a 2-mm sieve, and prepared for digestion. In order to determine metals in soil, extraction method was employed [15] [17].The ultimate concentration of the elements was measured using an atomic absorption set (AA110 VARIAN)[15] [17].In order to assess laboratory analysis data, a guide for soil quality by the Department of Environment of Iran was used.

Preparing dispersion maps of heavy elements (Geo-chemical map)

Maps of element dispersion in the lands surrounding the Faravari Zinc Factory were obtained using Arc GIS software and laboratory data obtained from sampling points.

Results of heavy metal pollution in the area of study

In order to determine the concentrations of elements in soil, 12 samples were obtained from depths of 50 to 60 cm from arable soils within 30 km east of the factory. The mean concentrations of all heavy elements, including lead, cadmium, and zinc, in these samples were 26.82, 0.78, and 32.45 mg/kg, respectively, which were used as context concentrations of these elements in estimations.

The results obtained from heavy metal concentration measurements in the soil surrounding the Faravari Zinc Factory are shown in Table 3.

Table 2 Soil contamination standard (Environment Preservation Organization in terms of mg/kg in soil)

	Concentration of the element in the environment in terms of (mg/kg)					Agrarian soils' standard		Environmental contamination standards
	Element In agrarian soils	The Earth's crust	In soil sample Ave	Max	Min	pH>7	pH<7	pH>7
Pb	3-189	16	131.25	251	20	75	50	300
Cd	0.01-2.5	0.2	11.2	5.38	2	5	1	3.9
Zn	17-125	80	1141.58	2347	68	500	200	200

Table 3: Results of soils analysis surrounding the Faravari Zinc factory

Sample	Pb	Cd	Zn	pH	Om	Clay
S1	251	10.5	1589.0	7.85	0.87	30.0
S2	39	3.4	138.0	7.73	0.70	44.0
S3	68	3.5	143.0	7.77	1.13	34.0
S4	143	3.8	750.0	7.81	0.70	44.0
S5	197	6.5	850.0	7.99	0.59	46.0
S6	120	3.6	350.0	7.77	0.68	42.0
S7	210	11.2	2310.0	7.88	1.01	12.0
S8	27	2.4	151.0	7.82	0.97	10.0
S9	155	2.3	844.0	7.41	0.87	14.0
S10	168	10.1	1561.0	7.53	0.63	18.0
S11	110	2.0	2241.0	7.65	0.67	16.0
S12	100	4.0	670.0	7.28	0.98	12.0
S13	64	5.4	388.0	7.32	0.74	13.8
S14	267	11.0	500.0	7.35	0.92	28.0
S15	240	11.3	2280.0	7.23	0.84	14.0
S16	310	12.3	450.0	7.62	0.64	37.0
S17	57	3.5	140.5	7.82	0.86	33.0
S18	197	7.3	1160.0	7.68	0.78	36.0
S19	209	10.1	1578.0	7.48	0.93	24.0
Min	27	2	138	7.23	0.59	10
Max	310	12.3	2310	7.99	0.13	46
Ave	154.3	6.53	952.28	7.63	0.81	26.72
StD	±83.11	±3.7	±759.2	±0.22	±0.15	±12.69

Based on the results, the mean concentration of lead in the soil surrounding the company was 154.3 mg/kg. The lowest lead concentration, 27 mg/kg, was observed at point S8, and the highest concentration, 310 mg/kg was observed at the S16. The mean concentration of lead at the sample points was 9.64 times greater than the concentration in the Earth's crust, but only 2 times greater than the concentration in agricultural soils standard.

Heavy elements such as lead are dispersed in the environment and eventually end up in the human body by entering the human food cycle. Once in the body, the lead inhibits cell reproduction activities and can cause encephalopathy, nervous system deficits, anemia, kidney degeneration, hypertension, and poisoning. One of the greatest concerns regarding the lead is related to its inhibitory of heme synthesis [16][13] .

The average concentration of cadmium in soil was found to be 6.53 mg/kg. The lowest concentration of cadmium was 2 mg/kg at S11, and the highest concentration, 11.2 mg/kg, was at point S7. The mean cadmium concentration was 32.6 times greater than the concentration in the Earth's crust and approximately 1.3 times greater than that of agricultural soils standard.

Cadmium is one of the most elements that occurs in relatively high amounts in disposed cakes at zinc processing facilities. Kidneys are target organs in cadmium toxicity; cadmium concentrations in the top layer of the kidney induces reduced cellular activity levels, along with protein, amino acids, and glucose depletion via urine. Cadmium toxicity leads to reproductive disorder [16][13].

The mean zinc concentration for the soil in this study was 958.28 mg/kg. The lowest concentration of zinc was at S2, 138 mg/kg, and the highest concentration was found at S7, which was 2310 mg/kg. The average concentration of zinc in the soil surrounding the factory was 11.9 times higher than the concentration found in the Earth's crust, and 1.9 times higher than that found in agricultural soils.

Geochemical map

Geochemical maps for lead, zinc, and cadmium for the area under investigation were prepared. The results for lead distribution suggest that the concentration of lead was high between the northwest and southeast points, with the highest concentration levels observed at points S1, S14, S15, and S16; S16 having the highest concentration of lead. The lowest concentrations were observed at points S2, S3, S12, and S17, with the lowest concentration obtained at point S8.

The results suggest that the lead distribution in arable soils surrounding the activity area of the factory was regular and its intensity range varied by the distance from the dispersion source. Figure 1 shows the dispersion expansion of lead in the activity area of the factory.

Cadmium concentrations were high between the northwest and central parts, and to the southeast. Concentrations at the points S7, S10, S14, and S15 were high and S16 had the highest concentration of cadmium. Points S1, S2, S3, S4, S6, S8, S11, S12, and S17 had low concentrations, and point S11 contained the lowest concentration of cadmium.

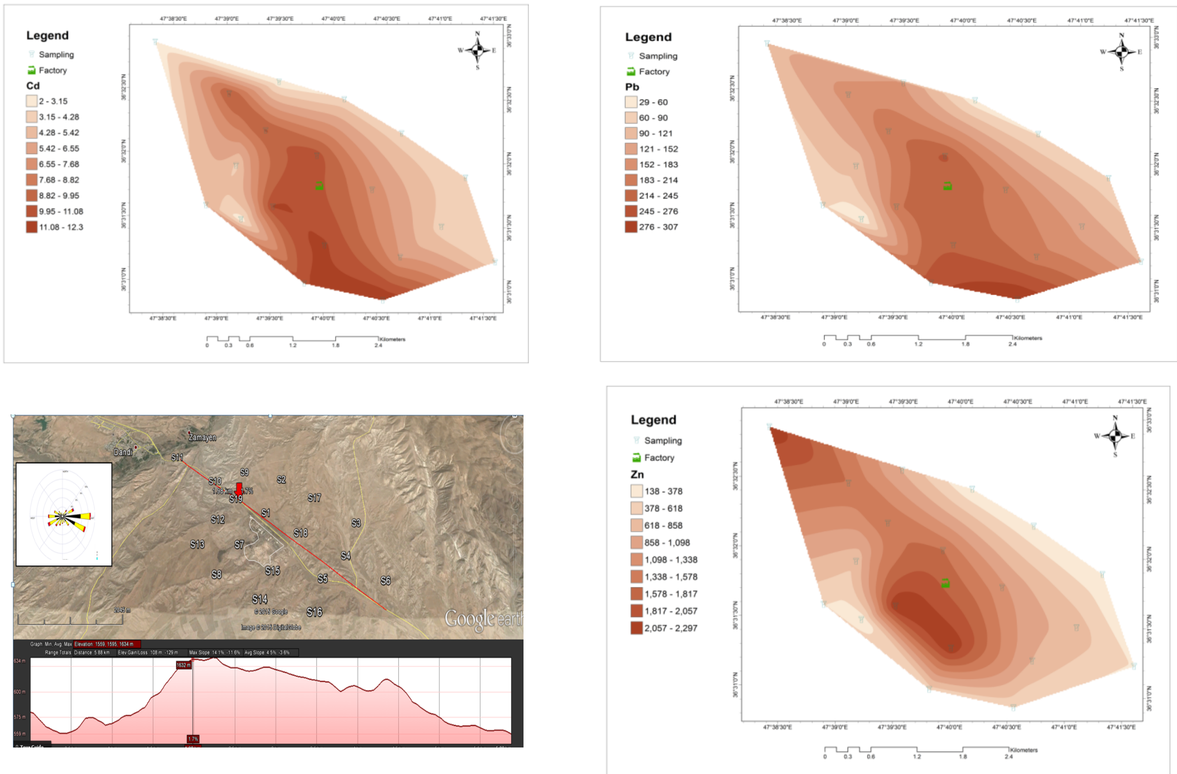

Figure 1 Geochemical map for Pb, Cd and Zn with direction of dominant winds, land slope, factory waste dumping site.

Zinc had a high concentration between the southwest and south points, and was concentrated at points S1, S7, S10, S11, S15, and S18. Point 7 had the maximum concentration of zinc. Points S2, S3, S7, S8, and S17 had low concentrations of this metal, and point 2 showed the lowest concentration of any point. Based of Figure 1 it showed thatclimate factors such as wind speed, wind duration, rainfall, also the size and nature of particles emitted are the main factors influencing environmental distribution of these elements.

Risk Assessment and Environment Quality Evaluation

In order to evaluate the environment quality, mono-element index was utilized [17] [5].

Equation 1

$$(P_i = \frac{C_i}{S_i})$$

Where, P_i is environment quality index, C_i is heavy elements concentration in soil and S_i is the allowed standard amount of the same element in soil in terms of mg/kg of soil. This is one of the highly used methods for assessing heavy element contamination grades in soil [18][6].

In order to assess the risks of heavy elements including lead, zinc, and cadmium in the soil surrounding the factory via contamination grading, the Nemero comprehensive index method was utilized in 5 categories (equation 2) [19][14]. This method is used, based on multi-criteria weighting, not only for an array of values but also for determining an environment quality index.

Equation 2 $$P=\sqrt{[(P_{ijmax})^2 + P_{ijave})^2]/2}$$

Where, P_i is comprehensive contamination index for each soil sample

Table 4 Standardized values for comprehensive index of pollution factor

Grade	1	2	3	4	5
P_i	$P_i \leq 0.7$	$1 < P_i \leq 2$	$1 < P_i \leq 2$	$2 < P_i \leq 3$	$P_i > 3$
Pollution Grade	Clean	Warning limit	Slight pollution	Moderate pollution	Heavy pollution

Based on the results from Table 3, mono-element indices of the elements were determined. The results are shown in Table 5

Table 5 Conclusion of single-factor and comprehensive pollution index for Pb, Cd and Zn

Sample	P_i *(Pb)*	P_i *(Cd)*	P_i *(Zn)*	P_{ijave}	P_{ijmax}	P	*PDegree*
S1	3.347	2.1	3.178	2.875	3.347	3.120	Heavy
S2	0.520	0.68	0.276	0.492	0.68	0.593	Clean
S3	0.907	0.7	0.286	0.631	0.907	0.781	Warning
S4	1.907	0.76	1.5	1.389	1.907	1.668	Slight
S5	2.627	1.3	1.7	1.876	2.627	2.283	Moderate
S6	1.600	0.72	0.7	1.007	1.600	1.337	Slight
S7	2.800	2.24	4.62	3.220	4.62	3.982	Heavy
S8	0.360	0.48	0.302	0.381	0.48	0.433	Clean
S9	2.067	0.46	1.688	1.405	2.067	1.767	Slight
S10	2.240	2.02	3.122	2.461	3.122	2.811	Moderate
S11	1.467	0.4	4.482	2.116	4.482	3.505	Heavy
S12	1.333	0.8	1.34	1.158	1.34	1.252	Slight
S13	0.853	1.08	0.776	0.903	1.08	0.995	Warning
S14	3.560	2.2	1	2.253	3.560	2.979	Moderate
S15	3.200	2.26	4.56	3.340	4.56	3.997	Heavy
S16	4.133	2.46	0.9	2.498	4.133	3.415	Heavy
S17	0.760	0.7	0.281	0.580	0.760	0.676	Clean
S18	2.627	1.46	2.32	2.136	2.627	2.394	Moderate
S19	2.787	2.02	2.627	2.478	2.627	2.554	Moderate

By using Table 4 and results presented in Table 5 it show that the pollution quality grade for lead at points S1, S14, S15, and S16 are at an heavy pollution level, and points S5, S7, S9, S18, and S19 are moderate pollution. As for cadmium, the pollution are moderate at points S1, S7, S10, S15, S16, and S19 and S11. Points S1, S7, S10, and S15 were heavy polluted and points S18, S19 were moderate polluted with zinc.

The pollution degree of the soil was attained based on the results from the mono-element method using equation 2 for a pollution accumulation index.

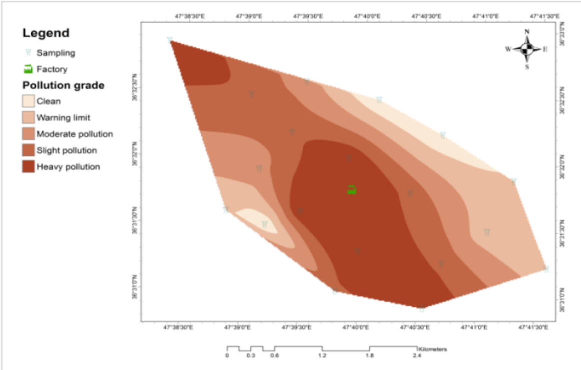

Figure 2 Result of soil pollution intensity evaluation by comprehensive pollution index method

Conclusion of evaluation by comprehensive pollution index indicate that the area between S1, S7, S11, S15, and S16 were highly contaminated; points S5, S10, S14, S18, and S19 were moderately contaminated; point S4, S6, S9, S12 were slightly contaminated; points S2, S8, and S17 were almost clean and points S3, S13 found to have an warning level of pollution.

Geo-accumulation (I_{geo})

The Muller index is a method to determine metal sediment contaminations, provided in equation (3), which was first defined in 1979[20] [18]. Existing concentration comparisons and context concentrations for industrial regions are used in this method [21][19]

Equation 3

$$log_2[Cn/Bn * 1.5]$$

Where I_{geo} is geo-accumulation index, log_2 is logarithm based on 2, ‹C_n is the concentration of tested element in soil, ‹Bn is the concentration of geo-chemical context in similar metal. The 1.5 factor is the matrix correction factor from lithologic effects [20] [18].

According to the Muller index, soils could be divided into seven groups by degrees of contamination. Table 6.

Table 6: Standardized values for index of geo-accumulation (I_{geo})

Index of geo-accumulation (Igeo) of contamination level						
Clean	Uncontaminated /moderately contaminated	Moderately/ contaminated	Moderately /strongly contaminated	Strongly/ contaminated	Strongly/ extremely contaminated	Extremely/ contaminated
0≤	0 – 1	1 - 2	2 - 3	3 - 4	4 - 5	> 5

The index of geo-accumulation (I_{geo}) has been calculated for the soil samples under study using equation (3) and the results have been presented in table (7).

Table 7 Comparison and correlation pollution index for the soil around of Faravari Zinc factory

Element	Geo-accumulation Index				Index of Geo-accumulation	Single factor Index
	Min	Max	Mean	SD		
Pb	0.575	2.945	1.669	0.997	Moderately contaminated	Slight pollution
Cd	0.358	2.979	1.791	0.896	Moderately contaminated	Slight pollution
Zn	1.503	5.018	3.730	1.426	Strongly contaminated	Moderate pollution

Statistical analysis

Correlation coefficient and cluster analysis.

Using the context data correlations, the results can be assessed to identify the sources of the heavy metals [22]. To determine the sources and correlation rates of the metals, Minitab Software (ver. 17) was used. The coefficient of the heavy metals in soils surrounding the mineral processing factory was calculated using Pearson's coefficient test to interpret statistical correlations in relation to each other and determine their sources. The correlation coefficient results have been presented in Table 8, which were used to obtain similarity coefficients and draw a dendogram. According to the results, the elements whose correlation coefficients are over 0.7 have high correlations and those with correlation coefficients between 0.4 and 0.7 have average correlations.

As shown in Table 8, there is a strong positive correlation between lead and cadmium concentrations, which suggests contamination from the same source, such as waste dumping from the factory within the activity area. The average correlation of zinc with lead and cadmium could be partly affected by context concentrations of the elements within the aforementioned area. During the production process, crushing, melting, and casting zinc bullions can result in zinc dispersion within the area. There is a negative correlation among heavy elements and organic compounds concentrations, pH, and clay soil rate.

Table 8: Correlation coefficient of the measured factors in surrounding soils of Dandi Zinc Processing Factory

	Zn	Cd	Pb	pH	Om	Clay
Zn	1.00					
Cd	0.472	1.00				
Pb	0.480	0.856	1.00			
pH	0.128-	0.195-	0.151-	1.00		
Om	-0.032	-0.013	-0.143	0.124-	1.00	
Clay	-0.420	0.058-	0.090	0.540	-0.426	1.00

For samples with low population using cluster analysis elements is a good way to identify different sources [23].

Conclusion of cluster analysis is shown in figure (3)

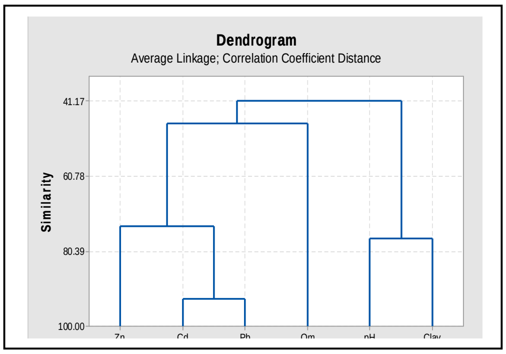

Figure 3 Cluster tree metals distribution in soil around of Faravari Zinc industry

CONCLUSION

The process of changing zinc ore into zinc concentrate is accompanied by the creation of disposal waste, which is one of the primary sources of heavy element dispersion into the environment.

In this study, the total amount of context heavy elements including lead, zinc, and cadmium was calculated. Contamination coefficients were identified using I_{geo} and the comprehensive pollution index to determine and assess heavy metal concentrations in the soils surrounding a mineral processing factory. The results revealed that soil in the vicinity of the factory was polluted by lead, zinc, and cadmium. The I_{geo} findings showed that the soils surrounding the factory were heavily polluted by zinc and moderately contaminated by lead and cadmium. The results of the mono-element pollution index indicated slight lead and cadmium pollution while the mean for zinc indicated moderate pollution. The pH of the surrounding soils was over 7. According to standards determined by the Environment Preservation Organization of Iran, shown in Table 2, allowed concentrations for lead, cadmium, and zinc in arable soils have been determined to be 75, 5, and 500 mg/kg, respectively. In this study, mean lead, cadmium, and zinc levels were found to be 153.4, 12.3, and 953.28 mg/kg, respectively, which were 2.05, 2.46, and 1.9 times greater than the corresponding Iranian standards for those compounds. The slope direction of the lands surrounding the factory, as well as the dominant wind direction, was from the southwest to the northwest, which played an important role in pollution dispersion over the region, as surface sewage caused by precipitation could permeate into lower lands.

It must be mentioned that at least 5% of the ore weight in the mine consists of lead. The results of the comprehensive pollution index for all three elements revealed that the area within the points S1, S7, S11, S15, and S16 were highly polluted. Factors such as crushing minerals outdoors, melting and casting, cake transit, the spillage caused by the waste, and storing the waste outdoors are all important factors that may be linked to the observed pollution.

The point S11 is closer than the other points to the Angoran lead and zinc company and is affected by the factory activities. In addition, points S6 to S11 are on a linear path along a road along which all of the trucks porting lead-containing soil move. Road transit could also play a part in the soil pollution of the area. According to the dendogram results in figure 3, lead and cadmium had a resemblance coefficient of 92.78% in group one. Lead and cadmium had a resemblance coefficient of 73.83% with zinc, and these compounds had a negative correlation with organic soil compounds, which suggests that heavy elements in the area do not have an organic basis, and have been dispersed over the soils surrounding the factory as minerals with a sulfate base. In the following branches of clay soil, pH had a resemblance coefficient of 76.98%, which had a resemblance of 46.87% with organic compounds in the soil. This factor also had a negative correlation with lead, cadmium, and zinc; that is, dispersion of these elements over the soil of the area are not related to anthropogenic sources, but are instead caused by human activities. In addition, comparing the context concentration of the elements from the depth of 60 cm with element concentrations from a depth of 25 cm showed that the concentration increased from the depth to the surface. Contamination levels that are higher on the surface of the land indicate a non-anthropogenic origin. The initial analysis of three different dumping cakes contained a percentage of lead, cadmium, zinc, nickel, and cobalt, whose arbitrary storage could be an important source of pollution dispersion in the environment because much of the soil surrounding the mineral processing facility is agricultural, the presence of these elements in the soil could give rise to their accumulation in plant tissues through transition process inside plants. Plants absorb these heavy elements which occur in the soil and accumulate them in the tissues. This is the first path through which heavy elements are introduced to food cycles in the environment.

The results of the present study could lead to the formulation of a program requiring that all zinc-related industries are obligated to install control systems in their industrial units, complying with environmental health standards, and replacing the polluted soil surrounding the facility with clean and healthy soil. Another option to reduce pollutant accumulation might be to change the plants that are cultivated nearby to inedible species with low element absorption to promote environmental preservation and renewal.

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