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Analysis and assessment of nickel and chromium pollution in soils around Baghejar Chromite Mine of Sabzevar Ophiolite Belt, Northeastern Iran

Eisa SOLGI, Javad PARMAH

Department of Environment, Faculty of Natural Resources and Environment, Malayer University, Malayer, Hamedan 65719-95863, Iran

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Abstract: The key objective of this research was to estimate the Ni and Cr contents of soil around the Baghjar Chromite Mine (BCM) of Sabzevar Ophiolite Belt, Northeastern Iran, and assess the degree of soil pollution using the pollution indices. Soil samples (0–20 cm depth) were collected at various distances from the BCM. In the present research, heavy metals (Cr and Ni) in soil samples were analyzed by atomic absorption spectrometry to detect their concentrations and contour maps were produced to explain the metal spatial distribution. Also, the degree of metal pollution was quantified. The results indicate that the soils in the studied area are contaminated by Cr and Ni. The corresponding concentrations for Cr and Ni are (156.19±24.45) and (321.7±133.27) mg/kg, respectively, which exceed the corresponding maximum allowable concentrations in soils. The different indices demonstrate that soils around chromite mine are significantly contaminated with Cr and Ni, suggesting several times higher levels of toxic metals than normal ranges. The above results revealed that the heavy metal concentrations increase with increasing the distance from the mine and mining pollutants can be transported to long distances from their sources.

Key words: ophiolite belt; chromite mine; spatial pattern; geoaccumulation index; pollution load index

1 Introduction

Increasing environmental pollution by heavy metals is one of the most significant environmental problems facing human. Heavy metals occur either naturally or through anthropogenic sources introduced into terrestrial ecosystems. The presence of heavy metals in soil is as a result of human activities such as urbanization, agriculture activities, industrialization and mining activities [1]. Mining and its activities are usually related to a negative impact on the environment and local residents if proper planning is not done. All of the components of the environment such as soils, sediments, air, water, flora and fauna can be greatly impacted. Release and contamination of heavy metals in the environment occur during mining activities [2]. Mining activities not only release heavy metal but also result in deterioration in the quality of soil, decrease in bulk density, organic matter, total nitrogen and available phosphorus of the soil [3] and also destruction of the ecological function of the system [4]. In recent years, many scientific studies have been carried out to examine heavy metal contamination in soils, plants, waters and sediments in the vicinity of mines throughout the world [1,5–11]. Among these researches, only very few studies focused on chromite mining [1,9,12–14].

Chromite, an iron chromium oxide ($FeCr_2O_4$), is a mineral that is the most important ore of chromium. Although many minerals have low concentrations of chromium, chromite ore is the only commercial source of chromium. Chromite is observed in peridotite from the Earth's mantle and also occurs in lavered ultramafic intrusive rocks and in metamorphic rocks such as serpentine and corundum [15]. The most important use of chromite ore is the production of ferrochrome (FeCr), an iron-chromium alloy, which is applied to producing stainless steel. Other uses of chromite are in the production of stainless steel, chemicals, pigments, refractories and foundry sands [16]. Mining of chromite has significant environmental impacts on the nearby surrounding with Cr (VI) contamination. The increase in the amount of Cr (VI) in water, soil and air sources is considered to be an important source of environmental pollution [17]. In this regard, pollution studies must be carried out on the contamination of chromite pollution in

Corresponding author: Eisa SOLGI; Tel: +98-8133339841; Fax: +98-8133339844; E-mail: e.solgi@malayeru.ac.ir; e.solgi@yahoo.com DOI: 10.1016/S1003-6326(15)63853-5

soil near a mining area. Iran is one of the countries that is ranked as a producer country in terms of chromite resources. This research is preliminary study to examine the influence of chromite mining activities on the adjacent areas by assessing heavy metal (Cr and Ni) concentrations in the surrounding surface soils of the Baghejar Chromite Mine (BCM), Iran. The other aim of this study was to use contamination factor (C_f) and pollution load index (PLI) to evaluate the degree of contamination due to mining activities. Finally, the purpose of this research was to assess spatial changes of Cr and Ni in surface soils around the chromite mine.

2 Experimental

2.1 Studied area

Sabzevar ophiolite is located in the north of Sabzevar City, which is situated in the northeast of Iran. The Sabzevar ophiolite forms an outcrop belt, 200 km in length and 3 to 22 km in minimum and maximum width, respectively [18]. The present research was carried out in Baghejar Chromite Mine (BCM). BCM (36°19′23.59″ N, 57°49′43.70″ E) is one of the most important chromite mining sites in Iran. The BCM is located 4 km of the Baghejar Village and 20 km northeast of Sabzevar County in Khorasan Razavi Province, Iran. The mine site covers an area of approximately 20 km². The climate of the studied area is cold arid desert climate, with annual mean temperature and rainfall levels of 17.3 °C and 189.1 mm, respectively. It has an average elevation of approximately 977 m above mean sea level.

2.2 Soil sampling and analysis

In May 2013, a total 21 soil samples (0-20 cm in depth) were collected at various distances in the range of 200-3000 m from the BCM. Each soil sample comprised a composite of five subsamples collected from a square of 20 cm \times 20 cm with the aid of spade and completely homogenized and reserved in polyethylene bags for laboratory analysis. The scheme map showing the locations of all soil samples is presented in Fig. 1. Soil samples were air-dried at room temperature for 72 h or longer if necessary. Later, they were milled using porcelain mortar and pestle and sieved through screens with pore sizes of 2.00 and 0.147 mm, respectively. All the chemicals applied in this experiment were of analytical grade. To measure heavy metal in soil samples, 12 mL aqua regia (V(HCL): $V(HNO_3)=3:1$) was added to 1 g soil sample in a digester tube, then heated at 140 °C in a heating block. After total digestion and cooling, the solution was filtered with a whatman No. 42 filter paper and diluted to 25 mL. Each solution was performed in three replicates. For analytical precision, the soil samples were analyzed in three replicates for each sampling point and reagent blanks were also used for each batch of digestion. The precisions of analytical, measured as relative standard deviation (RSD) which ranged from 3% to 6% total contents of Ni and Cr in the soils were analyzed by flame atomic absorption spectrometry (FLAAS). The limits of detection (LOD) were 0.25 and 1.5 mg/L for Cr and Ni, respectively. Soil pH and electrical conductivity (EC) were determined using standard procedure (m(soil): m(distilled water)=1:5). Soil



Fig 1 Distribution of sampling sites and location map of studied area

and water solution in mass ratio of 1:5 were homogenized together (by mechanically shaking for 2 h) and then soil pH and EC were measured by means of pH and EC meters, respectively.

2.3 Pollution assessment

Several methods of calculation have been proposed for quantifying the degree of metal pollution. Among these methods, the pollution is classified from low level to high level contamination. In this research, three methods have been used as follows.

The geoaccumulation index (I_{geo}) , proposed by MULLER [19] for assessment of metal pollution by comparing the concentrations of metals obtained to a background concentrations. Firstly, it was used for bottom sediments [19]. This index can also be used to the assessment of soil contamination [20]. It is mathematically calculated using the following equation: $I_{\text{geo}} = \log_2(C_n/1.5 \times B_n)$, where C_n is the measured concentration of metal in mine soil, B_n is geochemical background level of metal n and constant 1.5 is a correction factor due to lithogenic effects [21]. The geoaccumulation index was classified into seven classes (Table 1). The assessment of the soil contamination was also carried out using the contamination factor, degree of contamination and pollution load index. Contamination factor $(C_{\rm f})$ and degree of contamination $(C_{\rm deg})$ were calculated based on suggestion by HAKANSON [22]. The contamination factor can be calculated from the following relation:

$$C_{\rm f} = C_0 / C_n \tag{1}$$

where C_0 and C_n respectively refer to the mean contents of metals of at least five samples in the contaminated site and the baseline concentration in reference environment like Earth's crust in this study. HAKANSON [22] suggested four categories of C_f to assess the metal contamination levels as shown in Table 2. In this survey, the contamination degree (C_{deg}) was computed based on the sum of all contamination factors. The degree of contamination in soil by metals was calculated based on the method proposed by HAKANSON [22], by applying the following formula:

$$C_{\text{deg}=}\sum_{i=1}^{n} C_{\text{f}}$$
(2)

where *n* is the number of analyzed metals. The contamination degree of soil is divided into four groups: low ($C_{deg} < 8$), moderate ($8 \le C_{deg} < 16$), considerable ($16 \le C_{deg} < 32$), and very high contamination degree ($C_{deg} \ge 32$).

A modified form of the contamination degree equation for the calculation of the overall degree of contamination was presented by ABRAHIM and PARKER [23]. The modified degree of contamination

$I_{\rm geo}$ value	I_{geo} class	Pollution intensity	
< 0	Ι	Unpolluted	
0-1	Π	Unpolluted to moderately polluted	
1-2	III	Moderately polluted	
2-3	IV	Moderately to strongly polluted	
3-4	V	Strongly polluted	
4-5	VI	Strongly to very strongly polluted	
> 5	VII	Very strongly polluted	

Factor range	Description
$C_{\rm f} < 1$	Low contamination factor
$1 < C_{f} < 3$	Moderate contamination factor
$3 < C_{\rm f} < 6$	Considerable contamination factor
6 <c<sub>f</c<sub>	Very high contamination factor

 $(C_{\text{deg,m}})$ was calculated by the sum of all the contamination factor (C_{f}) for a given set of soil pollutants divided by the number of analyzed pollutants. This was calculated by the following formula:

$$C_{\deg,\mathrm{m}} = \sum_{i=1}^{n} C_{\mathrm{f}}/n \tag{3}$$

The classifications of the modified degree of contamination ($C_{deg,m}$) in soil are as follows: $C_{deg,m} < 15$, very low degree of contamination; $15 < C_{deg,m} < 2$, low degree of contamination; $2 < C_{deg,m} < 4$, moderate degree of contamination; $4 < C_{deg,m} < 8$, high degree of contamination; $8 < C_{deg,m} < 16$, very high degree of contamination; $16 < C_{deg,m} < 32$, extremely high degree of contamination; $C_{deg,m} < 32$, ultra high degree of contamination spatial.

Pollution load index (P_{LI}) has been largely used to assess contamination level and pollution in soils and sediments. The pollution load index (P_{LI}) proposed by TOMLINSON et al [24] was used to measure P_{LI} in mine soils. This parameter is expressed as

$$P_{\rm LI} = \sqrt[n]{C_{f_1} C_{f_2} C_{f_3} \cdots C_{f_n}}$$
(4)

Based on TOMLINSON et al [24], the following degrees are obtained: no pollution if $P_{LI}=0$, baseline levels if $P_{LI}=1$ and progressive deterioration if $P_{LI}>1$.

2.4 Maps of spatial distribution

Several methods of spatial interpolation were applied in soil investigations of spatial distribution of physicochemical soil properties. One of the most commonly techniques used for interpolation of scatter points is inverse distance weighted (IDW) interpolation. This is a statistical procedure used to estimate general values between sampling points. IDW method is based on the assumption that objects that are close to one another are more similar than those are farther apart. In fact, IDW creates a continuous surface from sampled point values. In current research, using ArcGIS software (V.9.2), the spatial distribution map of variables was produced by IDW technique. It is calculated according to the following equation:

$$Z(s_0) = \sum_{i=1}^{N} \lambda_i Z(s_i)$$
⁽⁵⁾

where $Z(s_0)$ is the estimated value for location (s_0) , N is the total number of measured points surrounding the prediction, λ_i is the weight allocated to each observed point, and $Z(s_i)$ is the observed value at the *i*th location. The weight factor λ_i was calculated according to the following formula:

$$\lambda_i = \left(\frac{1}{d_i}\right)^p / \sum_{i=1}^N \left(\frac{1}{d_i}\right)^p \tag{6}$$

where d_i is the distance between point *i* and the unknown point, and *p* is the power ten of weight.

2.5 Statistical methods

The normality distribution of all data was tested with the Shapiro–Wilk test. In order to certify the association among soil properties (pH and EC) and heavy metal content, a Pearson's correlation analysis was used to dataset analyses. All statistical data analyses were performed with SPSS software (version 20.0 for windows).

3 Results and discussion

The statistical summary showing the mean, standard deviation, coefficient of variation, minimum and maximum, Kurtosis and Skewness values for each of the soil properties determined is presented in Table 3. The pH of the samples from chromite mine ranged from 8.26 to 9.95 (Table 3). The lowest pH value has been found in approximately 2100 m from mine and the highest pH value in nearest point to mine. Soil pH is considered one of the most significant factors affecting the concentration of metals in the soil solution, their mobilization and available for plant uptake [25]. pH of mine soil is higher than 7, therefore, mine soil can be regarded as alkaline soil. The EC values also varied between samples, in a range from 0.11 to 0.82 dS/m (Table 3). The ranges of the concentrations were found from 115 mg/kg for Cr to 550 mg/kg for Ni. Based on the mean and median concentrations of all samples analyzed here (Table 3), the metal abundance could be ordered as follows: Ni>Cr. According to the calculated values of coefficient of variation (CV) (Table 3), Ni content in soils showed a higher variability than Cr, being 41.4% and 15.65%, respectively. Chemical analysis indicated that soils around chromite mine have been considerably polluted by Cr and Ni, with the mean concentrations of these two metals exceeding of the EPA's guideline for soil quality (25 mg/kg Cr and 16 mg/kg Ni). The average concentrations of Cr and Ni are higher than world average soil concentration values (125 and 23 mg/kg, respectively [26]). The mean concentration of Cr did not exceed and Ni exceeded the ecotoxicological limit [27]. The natural concentrations of Ni in soil range within 10-50 mg/kg, and the permissible limit of Ni concentration in soil is 100 mg/kg [28]. However, the nickel levels in all of the soil samples were above the natural and maximum permissible concentration. The mean concentration of Cr in the studied area is 156.19 mg/kg and this finding is greater than the maximum concentration of Cr (100 mg/kg) stated hv KABATA-PENDIAS and PENDIAS [28] in soil used in agriculture. These results are above the risk reduction standard of 100 mg/kg as legislated for Cr and 50 mg/kg for Ni [29]. This shows that the studied area has high values of Cr and Ni that can pose a significant health threat for residential and agricultural uses.

Table 3 Descriptive statistics of soil properties and metalcontents around chromite mine area at N of 21

Parameter	$w(Ni)/(mg \cdot kg^{-1})$	$w(Cr)/(mg\cdot kg^{-1})$	pН	EC/ (dS·m ⁻¹)
Min	132.36	115.00	8.26	0.11
Max	550	194.61	9.95	0.82
Average	321.7	156.19	9.02	0.29
Median	346.80	150	9.00	0.24
SD	133.27	24.45	0.38	0.17
CV	41.4*	15.65*	4.20^{*}	58.62
Variance	17759.04	597.74	0.15	0.03
Skewness	0.25	0.242	0.367	1.70
Kurtosis	-1.24	-1.12	0.340	2.95

* represents values in %

Comparison of the present results with those found in previous studies for the heavy metals in the chromite mine soils [1,9] indicates that the soils evaluated here were less contaminated with Cr than those from India and Vietnam, while Ni had higher values here than in India. Research of KIEN et al [9] into chromite mines in Vietnam indicated that metals of primary concern at this mine are Ni, Cu and Cr. Chromite ore mine provides obvious source of contamination of several metals such as Cr. Various metals typically occur along with Cr in chromite mine soil and water [14]. A Pearson correlation matrix was used to establish the relationships of the studied parameters in soils around the mine (Table 4). Soil pH and EC correlated negatively with Ni due to the fact that soil pH is the important agent which controls Ni 2384

level in soils [30]. GIL et al [31] found that pH was negatively correlated to Ni. Also, no correlation existed among Cr, pH and EC. Soil pH was significantly positively correlated with soil EC.

 Table 4 Pearson correlation coefficients among metals and soil properties

Item	Ni	Cr	pН	EC
Ni	1	0.20	-0.62**	-0.49*
Cr	0.20	1	-0.28	-0.31
pН	-0.69**	-0.28	1	0.70^{**}
EC	-0.49*	-0.31	0.70^{**}	1

** Significant at 0.01 level and * significant at 0.05 level

The heavy metal contamination of the soils around the chromite mine was evaluated based on contamination factor, degree of contamination, modified degree of contamination ($C_{deg,m}$) and pollution load index (P_{LI}). The results indicate that the I_{geo} values for Cr and Ni are all in the range of moderately to extremely contaminated.

The calculated geoaccumulation index (I_{geo}) values of heavy metals from 21 sites are presented in Fig. 2. The I_{geo} obtained for Cr changes from 1.1 to 2.07 (Fig. 2). The average I_{geo} for Cr is 1.69, which shows moderately contaminated soil, while maximum values classify soil as moderately to heavily contaminated. The I_{geo} values for Ni range from 2.14 to 4.2 with an average of 3.3, which denotes heavily contaminated and maximum values classify that the soil is heavily to extremely contaminated. It can be observed from Fig. 2 that geoaccumulation indexes for Cr and Ni are high, which implies the existence of considerable contribution of anthropogenic sources. The most probable source of the addition of these metals to the natural soil is mining activities.



Fig. 2 Geoaccumulation index (I_{geo}) for contamination levels in soil samples

In order to characterize the general contamination in mine soils, the contamination factor $(C_{\rm f})$, contamination

degree (C_{deg}) and modified degree of contamination $(C_{\text{deg,m}})$ were calculated and presented in Table 5. The minimum, maximum and average values of $C_{\rm f}$, $C_{\rm deg}$ and $C_{\text{deg,m}}$ of the soil are also presented in Table 5. The overall contamination of soils at the site, based on the average $C_{\rm f}$ values indicates that soils are considerably contaminated with Cr and very high contamination with Ni. The contamination degree (C_{deg}) was calculated as the sum of all contamination factor values of a particular point (see Table 5). Calculation of the average contamination degree reveals that the studied area is considerably contaminated (C_{deg} =20.54), indicating loading from anthropogenic sources. The modified degree of contamination ($C_{deg,m}$) procedure can supply a cumulative assessment of the overall enrichment and contamination impact of groups of contaminants in the soil [32]. The modified degree of contamination $(C_{deg,m})$ for the soils of the studied area is shown in Table 5. It is found that the $C_{deg,m}$ values ranging from 5.80-16.43 reflect high to extremely high degree of contamination. Overall, the mean value of $C_{deg,m}$ is 10.27, indicating very high degree of contamination. Also, in the studied area, pollution load index (P_{IJ}) was estimated to better realize the pollution level. In addition, it also provides useful data to the decision makers on the pollution level of the area. The $P_{\rm LI}$ values calculated for soil samples are displayed in Fig. 3. Levels of P_{LI} at all studied sites were

Table 5 Descriptive statistics of $C_{\rm f}$, $C_{\rm deg}$ and $C_{\rm deg,m}$ in soils around chromite mine area at N of 21

Parameter	$C_{\rm f}({\rm Cr})$	$C_{\rm f}({\rm Ni})$	C_{deg}	$C_{\text{deg,m}}$
Min	3.29	6.62	11.60	5.80
Max	5.56	27.50	32.86	16.43
Average	4.46	16.08	20.54	10.27
SD	0.70	6.66	6.83	3.42
Skewness	0.24	0.248	0.367	0.367
Kurtosis	-1.19	-1.24	-1.17	-1.17



Fig. 3 Pollution load index (P_{LI}) for soil collected around mine area

found to be polluted ($P_{LI}>1$), indicating an input of anthropogenic. P_{LI} values of the analyzed samples range from 5.46 to 12.13 with an average value of 8.26, which shows that the soils are polluted and the environment is deteriorated in their quality. This shows that the chromite region is strongly affected by mining activities and soils in this region are seriously contaminated by Ni and Cr. Highest pollution load index value is obtained at 2100 m from chromite mine. These results reveal that the soils around the chromite mine are severely contaminated with Cr and Ni.

Mapping is a useful approach for studying spatial and temporal variabilities of soil properties [33]. Also, mapping metal concentrations is often a primary stage in the decision-making process, such as description of contaminated sites or identification of zones suitable for crop production and farming [34]. Spatial variation maps with IDW method for Cr, Ni, pH and EC are shown in Fig. 4. All the properties have distinct geographical distribution. The spatial distribution maps show similar geographical trends, for Cr and Ni, with high contents both in the southeast and center areas. The spatial variability of soil properties may be attributed to intrinsic characters (natural factors, such as soil parent materials and soil formation factors) and extrinsic factors (anthropogenic factors, such as fertilization, agricultural practices and industrial effluent discharge) [35]. The spatial distributions of pH and EC showed similar patterns. The extensive variation of EC of the soils might be because of different concentrations of basic cations in the soils [36]. Also, soil pH variation range indicates the variation in soil metal concentrations across the sampling sites.

The distribution of Cr (Fig. 4) showed the maximum value in the center and southern parts of the studied area, while the low values were mainly located in

the west and north area. High concentration of Cr was distributed over the whole studied area. The high concentration areas of Ni (Fig. 4) were situated in the middle and southeast of the studied area, which was the area with intensive human activity, due to waste disposal activities of Baghjar Village and proximity to Sabzevar-Khoshab-Ghochan transit road. The low content values of Ni were mainly located in the north and near to center area. Overall, similar spatial patterns were found for these metals, suggesting that these were from the same sources such as mining activities. High concentrations of Cr and Ni were distributed over the whole studied area. No remarkable differences of Ni and Cr concentrations with distance from the chromite mine site were observed. However, high Cr and Ni concentrations were found in soil samples collected within 2 km distance from the mine, which may suggest that these two metals can be transported considerably to long distances from the source. Many reasons can be explained for high concentration of Cr and Ni even at long distances from chromite mine. Firstly, for some metals, such as lead, most of their emissions to the air are deposited close to the source, but some particulate matters ($<2 \mu m$) can be transported over much longer distances and lead to contamination of remote areas [37]. Also, it is known that mercury released from contaminated areas can be deposited locally or can move over long distances and deposited even at the most remote sites far from the sources [38]. HU et al [39] found that the Cr concentrations increased gradually with the increase in the distance from the mine. In Humberside, UK, RAWLINS et al [40] measured Pb and Sn contents in areas around the former smelter and indicated that significant levels of elements were deposited up to 24 km from the smelter by the prevailing wind. Secondly, this area was located within ophiolite belts.



Fig. 4 Spatial distribution of Cr content (a), Ni content (b), pH (c) and EC (d) in soils using inverse distance weighting (IDW) method

HAJIZADEH NAMAGHI et al [41] found that the higher concentrations of Cr and Ni in Firuzabad, Shahrood, Iran, are due to development of soil in ultramafic rock. Thirdly, there are many mines in this region where soils near mining sites have mainly large amounts of heavy metals such as Cr and Ni.

4 Conclusions

Soil is a major pool for contaminants as it encompasses ability to bond with various chemical materials and media for transportation of forms of various pollutants in the atmosphere, hydrosphere, and biomass. Therefore, it is the most specific component of the biosphere. The results obtained in the present research of Cr and Ni in soil sample collected around chromite mine showed that soil quality in the mine and areas around the mine chromite is degrading. The studied area is extremely contaminated by Cr and Ni due to many years of mining activities. Our data disclose that Cr and Ni concentrations in soil samples are higher compared with the worldwide average, ecotoxicological limit, maximum allowable concentration and EPA's guideline for soil quality. The pollution assessment methods showed that soils in the studied area are significantly contaminated by Cr and Ni. Thus, in the future, based on the environmental quality criteria for soils, the site would need remediation. This research revealed that the heavy metal concentrations increase with increasing distance from the mine and mining pollutants can be transported to long distances from their sources. The introduction of Cr and Ni into the food chain via soil may affect human health, thus Cr and Ni accumulation in vegetables must be studied.

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伊朗东北部萨卜泽瓦尔蛇绿岩带 Baghejar 铬铁矿周边土壤的镍、铬污染分析与评估

Eisa SOLGI, Javad PARMAH

Department of Environment, Faculty of Natural Resources and Environment, Malayer University, Malayer, Hamedan 65719-95863, Iran

摘 要: 主要分析伊朗东北部萨卜泽瓦尔蛇绿岩带 Baghejar 铬铁矿(BCM)周边土壤的镍、铬浓度,采用污染指数 评估土壤污染程度。土壤样品(深度为 0~20 cm)采于距 BCM 矿不同距离的地点。采用原子吸收光谱测定土壤样品 中重金属(铬和镍)含量。绘制等高线图描述重金属的空间分布,同时对金属污染程度进行了量化。结果表明,采 样地土壤受到铬和镍污染。铬和镍的相应浓度分别达到(156.19±24.45)和(321.7±133.27) mg/kg,超出对应的土壤最 大允许浓度。各种指数显示出铬铁矿周边土壤显著受到铬和镍污染,超出正常范围数倍之多。研究表明,重金属 浓度随着与矿区距离增加而增加。采矿产生的污染可以迁移到距源头较远的地方。

关键词: 蛇绿岩带; 铬铁矿; 空间特征; 地质累积指数; 污染负荷指数