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Pollution intensity of nickel in agricultural soil of Hamedan region

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ABSTRACT

In the recent years, agricultural soils have received more chemicals in various forms for higher yields. This may result in the accumulation of metals in soils which subsequently may be transferred into the agricultural crops. In the present investigation, nickel content of 58 composite soil samples and also agricultural crops, is studied. Ni in samples of soil and agricultural products were extracted and determined using atomic absorption spectrometer. The spatial distribution map of Ni was drawn using Kriging method. Mean concentration of nickel in the soil of the study area is about 62 mg.kg^{-1}. It was also found that among the harvested crops, corn has a higher concentration of nickel than the standard limit. According to chemical fractionation results of nickel in soil, anthropogenic and natural shares of this metal are 19%, and 81% of the bulk concentration, respectively. Results of comparison of heavy metal pollution intensity in the agricultural soil by I_{geo} index as well as I_{POLL} index showed that Ni is in the non-polluted to moderately polluted class. Analysis of zoning map of pollution index showed that Ni is mainly of geological sources.

INTRODUCTION

Organic and inorganic pollutants find their way to the soil by many human activities (Shetty & Rajkumar 2009). Most of heavy metals including nickel are toxic or poisonous even at low concentration. These toxic metals can find their way into agricultural crops through soil (Mohammadpour Roudposhti *et al.* 2016). The poisoning effects of heavy metals depend on many factors. It is very well known that heavy metals are extremely persistent in the environment. Due to their resistance, they can be accumulated and reach to toxic levels (Khan *et al.* 2009). The toxicity level is different among metals. For the above mentioned reasons, more attention has been paid to the nature of metal toxicity in the recent years (Lado *et al.* 2008). Soil properties and quality can be adversely affected by the over-concentration of agricultural and industrial activities. On the other hand, preserving soil quality and preventing its deterioration are essential to the sustainable development (Karbassi *et al.* 2008). In general, wastewater contains substantial amounts of beneficial nutrients and toxic heavy metals, which are creating opportunities and problems for agricultural production, respectively (Singh *et al.* 2010). Heavy metal accumulation in plants depends upon plant species and the efficiency of different plants in absorbing metals is evaluated by either plant uptake or soil-to-plant transfer factors of the metals. Metals concentration in vegetables mainly depends on the texture of soil or media on which they grow but this also depends on the type and nature of plant (Kabata-Pendias & Pendias 2000). Therefore it is very important to know about the health risk assessment of metals. There are several studies in different countries for interpolation and determination of spatial distribution of heavy metals concentration in

soil (Mohammadpour Roudposhti *et al.* 2016). European topsoil was evaluated using the cross validation method (Lado *et al.* 2008). The spatial distribution of metals by Kriging method has received ample attention (Juang *el al.,* 2001; Rodriguez *et al.* 2009). Therefore, due to the importance of Hamedan City as one of the main centers of agricultural activities in Iran, it is of utmost importance to determine the pollution of topsoil and agricultural crops of this area. Thus in the present study, we have tried to bring out the sources of Ni in agricultural soils of Hamedan. The transfer factor of this metal

from soil to crops was subsequently determined. Finally, the health risk of this metal as a result of consumption of various agricultural yields was computed.

MATERIALS AND METHODS

Hamedan Province occupies an area of 2831 km². Samples were collected from depth of 0-20 cm in accordance with the systematic method. A total of 58 composite samples of topsoil and crops of wheat, barley, corn, alfalfa and potatoes were collected. Fig. 1 shows the location map of the study area and sampling points.
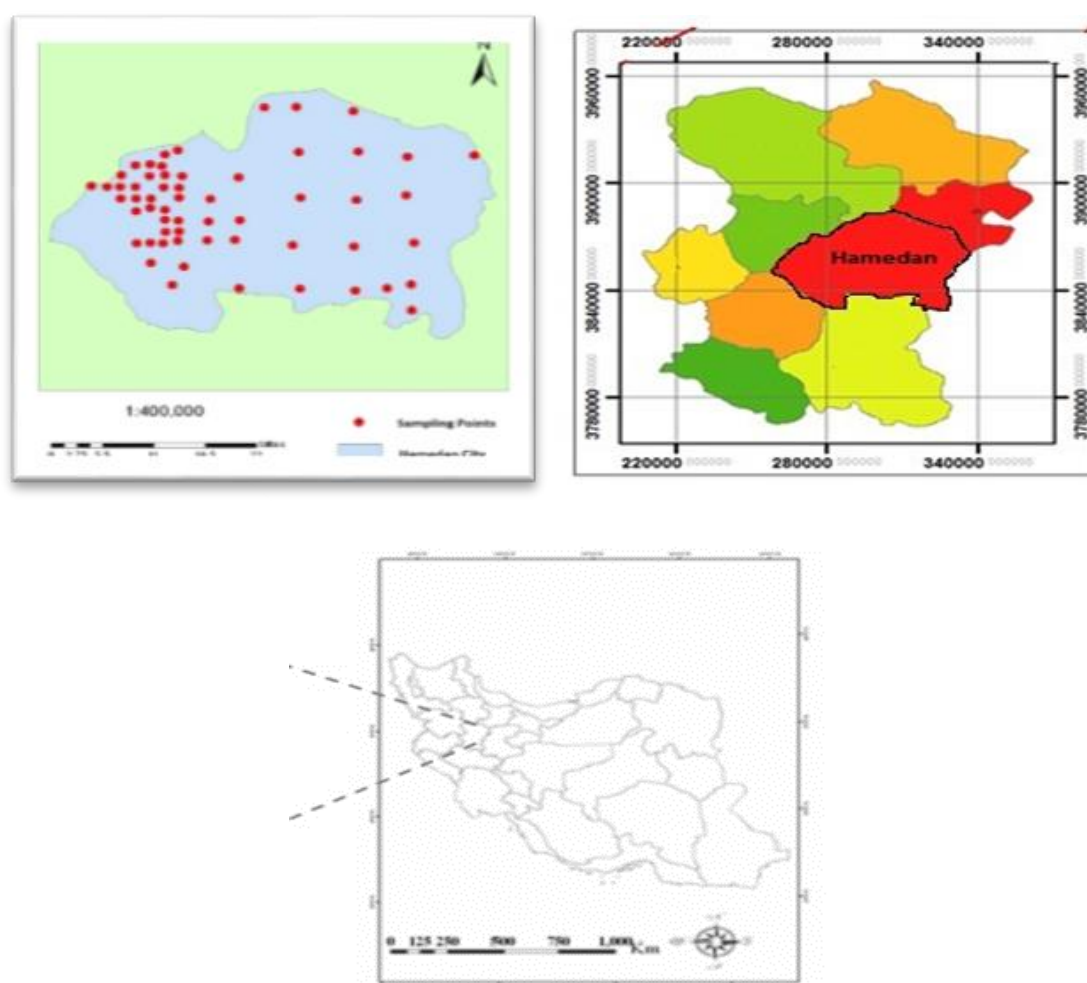

Fig. 1. Location of soil samples in Hamedan.

Soil samples were dried and passed through 63 micron sieve. Phosphorus was measured using Olsen P extracting solution (0.5 M NaHCO₃, pH 8.5); total nitrogen by Kjeldahl digestion; pH was analyzed by glass electrode using a 1:1 soil: water ratio; and EC and salinity were measured using conductivity meter in a soil-

water extract (1 : 2 soil : water ratio) (Dewis & Freitas.1984; ASTM 2000; USDA 2004). Titration method was used to determine lime. Sodium and potassium were measured using flame detector (AOAC 2005). Soil texture was assessed in accordance with (Gee & Bauder., 1986). Total C was measured as described by

Allison (1965) and Supaphol *et al.* (2006). Cation exchange capacity was measured according to the standard methods (APHA, 1998; Aparna *et al.* 2010). Total organic carbon was computed as per method reported by Karbassi *et al.* (2008). Measurement of calcium and magnesium of soil was carried out by EDTA solution using complexometric titration (AOAC 2005). About 2 gr of dried and sieved soil was poured into a capped container and 15 ml of 4N nitric acid was added. Then the flasks were left for 12 hours in a hot water bath at 80°C. The samples were filtered and concentration of Ni was measured using atomic absorption spectrometer. The dry ash extraction method was used for digestion of plant samples (Shaw 1989). Then concentration of Ni was determined using atomic absorption spectrometer (Varian model AA-400). The accuracy and precision of the overall procedure were determined and estimated to be around 3% for Ni. The quality assurance of the analytical results was controlled using certified reference marine organism IAEA-407 provided by International Atomic Energy Agency. The soil - to - plant metal transfer was computed as transfer factor (TF), which was calculated using the equation $TF = C_{Plant}/C_{Soil}$ where, C_{Plant} is the concentration of heavy metals in plants and C_{Soil} is the concentration of heavy metals in soil (Mahmood & Malik 2014). Daily intake of vegetables in adult was calculated by data obtained during the study through a questionnaire. DIM was calculated by the following equation:

$DIM = C_{metal} \times C_{factor} \times D_{food\ intake} / B_{average\ weight}$ (Chary *et al.*, 2008) Where, C_{metal}, C_{factor}, $D_{food\ intake}$ & $B_{average\ weight}$ represent the heavy metals

concentration in plants (mg.kg⁻¹), conversion factor (0.085), daily intake of vegetables and average body weight, respectively. In the present study, the vegetables grown at the soils were collected from the study area and their metal concentration was used to calculate the health risk index (HRI). Value of HRI depends upon the daily intake of metals (DIM) and oral reference dose (RfD). It should be noted that RfD is an estimated per day exposure of metal to the human body that has no hazardous effect during life time (US-EPA IRIS 2006). The health risk index for Sb, Mn & Fe by consumption of contaminated vegetables was calculated by the following equation:

$$HRI = \frac{DIM}{RFD}$$

Where DIM represents the daily intake of metals and RfD represents reference oral dose. RfD value for Sb, Mn and Fe is 0.0004, 0.14 (mg.kg⁻¹ bw.day⁻¹) and 0.7 % respectively (WHO, 1993; EPA, 2007). To quantify the degree of the heavy metal pollution in soil, Igeo was calculated according to Muller and is given in Eq. (1) (Muller 1979; Praveena *et al.* 2008). The results were interpreted using Igeo classes given in Table 1.

$$I_{geo} = Log_2\left(\frac{C_n}{1.5\ B_n}\right)$$

Where Cn is the concentration of the examined metal in the soil, Bn is the geochemical background value of a given metal in the soil (Turekian & Wedepohl 1961) and the factor 1.5 is used to account the possible variations in the background values.

Table 1. Igeo classes in relation to soil quality (Serbaji *et al.* 2012).

Igeo	Igeo class	Soil quality
0-0	0	Unpolluted
0-1	1	Unpolluted to moderately polluted
1-2	2	Moderately polluted
2-3	3	Moderately to highly polluted
3-4	4	Highly polluted
4-5	5	Highly to very highly polluted
5-6	>5	Very highly polluted

To assess the intensity of metal contamination in Hamedan soils, the pollution index (Karbassi *et al.* 2008) was calculated using:

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

Where, Cn is the total elemental content in soils and Bn is the geochemical background of element.

Chemical fractionation method suggests that heavy metals form five bonds with soil and sediments including loosely bond, sulfide bond, organometallic bond, most resistant bond and within lattice bond among which loosely bond, sulfide bond and organometallic bond indicate anthropogenic elements of the environment and the other two bonds, namely, most resistant bond and within lattice bond indicate natural part of heavy metals in soil (Spencer & Macleod 2002; Karbassi & Shankar 2005). The Five-stage Chemical Fractionation method was applied in this study in order to specify the anthropogenic part of heavy metals and the natural part of heavy metals. To understand the relationship amongst various metals and environmental indicators, the Multi

Variable Statistical Program (MVSP) was used. This analytical software is frequently used by various researchers (Karbassi *et al.* 2004, 2008). The results of clustering are displayed in the form of a Dendrogram. Kriging interpolation method was applied to show changes in concentration distribution of the investigated heavy metals (Johnston *et al.* 2001).

RESULTS AND DISCUSSION

Investigation of physicochemical parameters of soil showed that the parameters of electrical conductivity (EC), sodium, calcium carbonate (CaCO₃) and salinity have coefficients of variation above 50% which indicates the large variations of concentration of these variables in soil of Hamedan City. The rest of soil physicochemical parameters have coefficients of variation less than 50% which indicates its low variation in soil of Hamedan City. The average acidity of soil is 7.72 which indicate the soil of the study area is of alkaline nature. The dominant soil texture in the area is silty clay loam soil. A summary of statistical status of some physicochemical properties of the soil of study area is presented in Table 2.

Table 2. Physicochemical characteristics of the soils used in the study.

Factors	Unit	Minimum	Maximum	Mean	Standard deviation	Coefficient of variation
pH	-	6.80	8.90	7.7224	0.52249	0.068
Organic	%	0.90	2.00	1.4200	0.25782	0.182
CEC	Cmolc.kg⁻¹	9.40	39.40	23.1112	7.72925	0.334
Clay	%	8.50	54.30	27.2512	12.16787	0.447
Sand	%	8.90	69.40	30.9724	14.25832	0.460
Silt	%	7.90	77.40	46.0547	17.81106	0.387
EC	(ds.m⁻¹)	0.05	3.70	1.6533	1.14063	0.691
CaCO₃	%	1.00	43.00	12.3793	6.75856	0.546
N	%	0.01	0.09	0.0526	0.02283	0.457
P	(mg.kg⁻¹)	3.40	39.40	21.9807	8.21287	0.374
Ca	(mg.kg⁻¹)	4.40	49.40	22.9752	10.42741	0.454
Mg	(mg.kg⁻¹)	69.40	199.50	143.6884	29.98786	0.209
Na	(mg.kg⁻¹)	8.50	71.60	32.2821	17.76203	0.550
K	(mg.kg⁻¹)	10.40	79.40	28.6617	12.34805	0.431
Salinity	(ds.m⁻¹)	0.10	2.24	0.7386	0.40293	0.545

According to the Kolmogorov-Smirnov test, all data of soil and harvested yields were normal. Table 3 provides a summary of the statistical characteristics of Ni concentration in all analyzed samples in this study. The mean

concentration of Ni in soil of Hamedan region was about 62 mg.kg⁻¹.

Table 4 compares the concentration of Ni in the soil of studied area with those of Europe and of the world. As shown in the Table, the mean

concentration of Ni in the present study is higher than in the world. Table 5 shows the mean concentration of Ni in the harvested agricultural yields of Hamedan County.

The standard limit of Ni in agricultural yields is 1.5 mg.kg^{-1} (Pandey & Pandey 2009). In this study, only the concentration of Ni in corn was lower than the standard limit.

Table 3. Mean concentration of Ni in the soils.

Heavy metal	Minimum	Maximum	Mean	Standard deviation	Standard level
Ni	37	99	61.93	13.92	European Union Standards (EU 2002)

Table 4. Comparison of the concentrations of nickel in the studied area with the values from the region, Europe, and the world (Facchinelli *et al.* 2001; Franco-Uria *et al.*2009).

Heavy metal	Europe mean	Area mean	World median	Area median
Ni	73.3	61.93	50	60.5

Table 6 shows the transfer factor from soil to agricultural crops. The Table shows that Ni has a high transfer factor (about 61).

This may be due to the low retention rate of the metal in soil. The difference in transfer factor among the various yields may be due to the differences in the soil metal concentrations and the differences in intake of the element by different yields (Cui *et al.* 2004).

The degree of toxicity of heavy metals to human being depends upon their daily intake. The population will pose no risk, if the ratio is less than 1 but if the ratio is equal or greater than 1, then population will experience health risk (Sajjad *et al.* 2009).

Table 7 shows the daily intake and health risk of Ni in children and adults. Considering the daily intake of metals from agricultural yields and since health risk index of Ni is less than 1, thus children and adults will be safe to use agricultural yields. Comparing the health risk index of this study with those of Khan *et al.* (2010) and Jan *et al*. showed that HRI is less than those of these authors (Khan *et al.* 2010; Jan *et al.* 2010).

Ni is an essential nutrient for some mammalian species and has been suggested to be essential for human nutrition. By extrapolation from animal data, it is estimated that a 70-kg person would have a Ni daily requirement of 50 µg per kg diet (ATSDR, 1997).

Table 8 presents the intensity of soil pollution with Ni using three indices including Muller geo-chemical index, Karbassi modified geomechanical accumulation index and contamination factor. Results of comparison of Ni pollution in agricultural soil with Muller's and also Karbassi indices showed that Ni is placed in non-polluted to moderately polluted class.

According to the results, the rates of contamination factor of Ni in soil of Hamedan City, 8.62% are placed in non-polluted class, 81.03% in non-polluted to moderately polluted class and 10.35% in moderately polluted class.

Table 9 shows the chemical fractionation of Ni in soil of Hamedan City.

The results showed that the highest concentration of Ni in soil are associated with the loosely bounds, sulfide bounds and also organic bounds, while the lowest ones are related to the most resistant bounds and also within lattice bounds.

The obtained results showed that about 19 and 81% of bulk content of Ni are respectively derived from anthropogenic and lithogenic sources. Thus, Ni is mainly controlled by the chemistry of parent rocks.

Table 5. Mean concentration nickel in the crops (mg.kg-1).

Crops	Ni
Wheat	0.4653
Barley	1.3174
Potato	0.9602
Alfalfa	1.5213
Corn	2.0230

Table 6. Transfer factor (TF) of Ni in agriculture crops grown in Hamedan City.

Factor	Range	Minimum	Maximum	Mean	Std. Deviation
Ni	62	37	99	61.93	13.92

Table 7. DIM and HRI of Ni in adults and children.

Heavy metal	Heavy metal in crops	DIM in adults	DIM in children	HRI in adults	HRI in children
Ni	1.257	6.594×10^{-4}	7.580×10^{-4}	0.033	0.0379

Table 8. Muller geochemical index, Karbassi modified geochemical accumulation index and contamination factor of the studied soils.

Heavy metal	I_{poll}	I_{geo}
Ni	0.630	0.046

Krigging method is applied to evaluate Ni in this study. Since the threshold concentration of Nickel is reported to be 75 mg.kg-1, therefore some parts of the north, south and center of the study area located on shale, sandstone, igneous and sedimentary bedrocks, had strip-shaped concentrations above the threshold concentration. Fig. 3 shows the spatial distribution of Ni in the soil of the study area. Ni concentration is naturally high in shale and sandstone bedrocks. By overlaying the resulted zoning and geological maps and also the land use map of the study area, it was found that this part of Hamedan City has mainly agricultural and pasture use. In fact, it may be mentioned that Ni in Hamedan soil is of geological source, however anthropogenic activities related to the use of fertilizers and sewage in the agricultural lands may lead to increased amount of this metal in the soil of Hamedan City. Since the anthropogenic activities which enter Ni into the environment, include chemical fertilizers, steel, mint and metal containers industries, detergents and burning of fossil fuels and because only the chemical fertilizers were applied in the study area, it can be concluded that the factor influencing the increased concentration of this element is of the geological structure origin (shale, limestone and sandstone). Furthermore, the agricultural activities (excessive use of chemical fertilizers such as urea, phosphate, and potash due to presence of Ni in their chemical structure) may cause the increased concentration of Ni in the agricultural lands. Facchinelli *et al.* (2006), Luis *et al.* (2008), Luo *et al.* (2007) and Mico *et al.* (2006) in their studies related to the sources of heavy metals in the soil, concluded that the concentration of Ni in soil is controlled by the bed rock.

Table 9. Chemical bonds of Ni in the soils of Hamedan City.

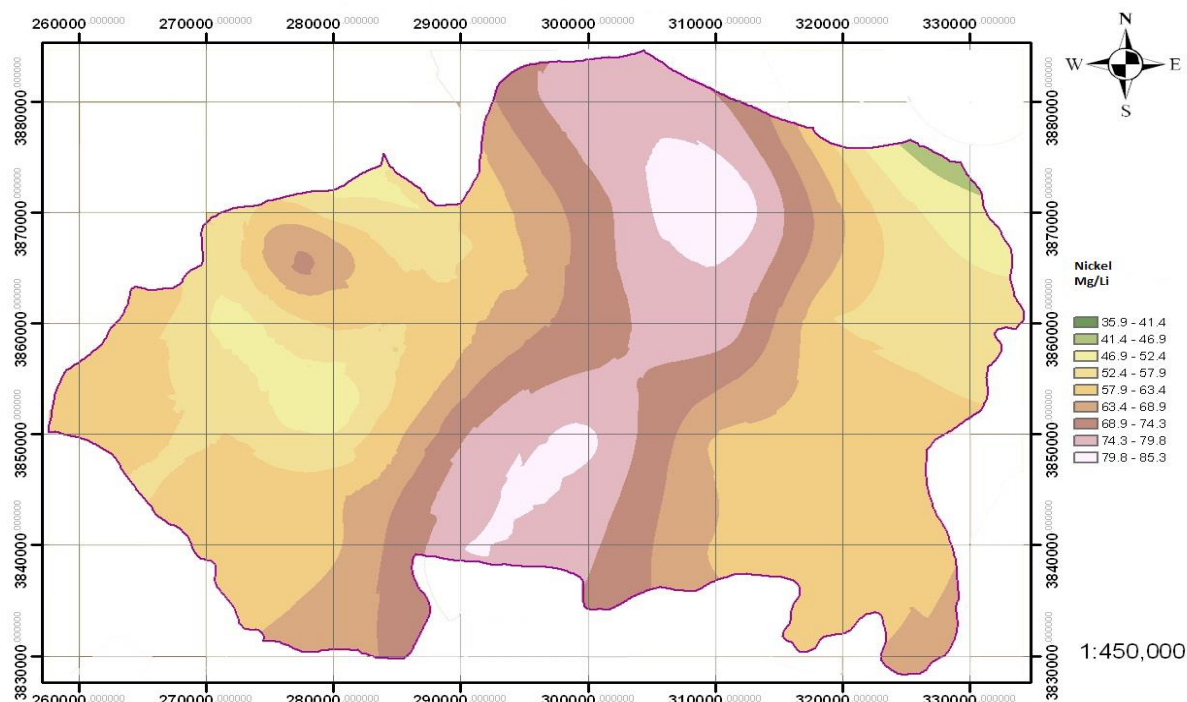| Heavy metals	Bulk concentration	Fractional Steps					Anthropogenic portion	Lithogenic portion
		Step 1	Step 2	Step 3	Step 4	Step 5		
Ni(mg.kg-1)	62	3.4	5.9	2.4	48	2	12	50

Fig. 3. Spatial distribution of Ni in the soil of Hamedan region.

CONCLUSION

The spatial distribution maps of heavy metals and land use and also the geological maps of the study area suggest that the main cause of the high Ni concentration in the area is the parent material of soil. However, the possible increase in concentration of Ni in the agricultural land is not far from expectation due to presence of heavy metals in chemical fertilizers, pesticides and also sewage and their excessive application in agriculture.

Therefore, given the hazards of heavy metals intake especially in the existing agricultural lands, it is recommended to prevent the further distribution of heavy metals by managing the application of chemical fertilizers and pesticides and also not to apply sewage for irrigation of lands as well as to avoid planting of those plants with high potential of heavy metals intake in the polluted areas.

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شدت آلودگی نیکل در خاک کشاورزی

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چکیده

در سال‌های اخیر مواد شیمیایی با فرم‌های مختلف در زمین‌های کشاورزی برای افزایش محصول، مورد استفاده قرار گرفته است. بنابراین احتمال انباشت این مواد و انتقال آنها به محصولات کشاورزی وجود دارد. در تحقیق حاضر، غلظت نیکل در ۵۸ نمونه خاک ترکیبی و انواع محصولات کشاورزی مورد مطالعه قرار گرفته است. برای این منظور نیکل از خاک و نمونه‌های محصولات کشاورزی استخراج و توسط دستگاه جذب اتمی اندازه‌گیری شد. توزیع جغرافیایی عنصر نیکل با روش کریجینگ رسم شد. میانگین غلظت عنصر نیکل در خاک کشاورزی همدان حدود ۶۲ میلی گرم بر کیلوگرم است. در بین محصولات کشاورزی مورد مطالعه، ذرت حاوی بیشترین مقدار نیکل است و غلظت آن از حدود استاندارد تجاوز می‌کند. بر اساس نتایج حاصل از تفکیک شیمیایی مشخص شد که ۱۹ و ۸۱ درصد از غلظت اولیه نیکل به ترتیب دارای منشاء انسان‌ساخت و زمینی می‌باشد. نتایج فرمول‌های شدت آلودگی شامل Igeo و Ipoll نشان‌دهنده قرارگیری نیکل در محدوده فاقد آلودگی تا آلودگی متوسط در خاک کشاورزی همدان است. نقشه‌های درون‌یابی نیز نشان دادند که منشاء اصلی نیکل در خاک کشاورزی، زمینی است.

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