



Spatial Variability and Pollution Status of Lead and in Nickel the Street Dust of Zanjan City, Iran

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ABSTRACT

Background: Focus on environmental heavy metals is important due to their adverse impact on the human health and environment. This study aimed to determine the lead and nickel concentrations and their spatial patterns in the street dust of Zanjan city, Iran, which is enclosed with metal production (especially lead and zinc industries).

Methods: Fifty urban street dust samples were collected from Zanjan and analyzed for lead and nickel after Aqua Regia digestion via flame atomic-absorption spectrophotometry. The sediment contamination degree and origin of the pollutants were assessed using the geo-accumulation index. Pollution status was assessed using the enrichment factor and potential ecological risk index (Hakanson index).

Results: The mean, minimum, and maximum lead were 745, 30, and 4610 mg/kg and 72, 32, and 154 mg/kg for nickel, respectively. The mean lead concentration was higher than the local background value. The element spatial distribution showed higher lead concentration in the east, west, and southwest, main streets, and Qazvin-Tabriz highways. Higher potential ecological risk was observed for lead, confirming regional pollution.

Conclusion: High lead concentrations could be attributed to vehicle emissions and industrial activities (lead and zinc companies in Zanjan). Traffic and frequent braking of vehicles could increase nickel concentrations.

1. Introduction

Heavy metals are found in water, air, soil, and sediments, as well as street dust at trace levels. Evidently, the growth of the population, industries, and transportation systems contribute to the increased heavy metal concentrations in urban dust and the environment, signifying pollution. These pollutants have accumulative and carcinogenic properties and may give rise to numerous health and environmental issues [1].

In 2016, the United Nations (United Nations, 2016) announced that 54.5% of the world's population inhabited urban areas, and the number of urban citizens is on the rise.

Furthermore, it has been claimed that 60% of the population will reside in urban areas by 2030. Therefore, the quality of the urban environment affects a high percentage of the population's life, and proper public health risk assessments are required to be formulated and applied [2].

Dust is a particle in the form of a solid powder, which is found on the ground or surface of objects and could be displaced by wind. According to the definitions provided by the World Health Organization (WHO, 1999) and Turner Broadcasting System (TBS, 2005), dust is a small solid particle that is located on various surfaces or scattered in the air, conventionally with the particle diameters of less than 75 micrometers, which settle under their own weight



and may remain suspended for some time. Heavy metals in street dust are major pollutants in urban environments, which may spread through heavy traffic, industries, building erosion, transportation systems, mining activities, and combustion of fossil fuels [3, 4].

Street dust is considered to be the major vehicle for heavy metals to easily enter soil, plants, and the human body. Accordingly, heavy metals in urban dust and soil are considered to a significant threat to the human health since they could easily enter the body through inhalation, ingestion, and dermal contact. Children may be exposed to dust frequently as they tend to play on the ground, pick up dust and soil by hand, and make the dust contact with the mouth. Adults are also at the same risk as inhalation is a key route for toxic heavy metals to enter the human body [5, 6]. The release of heavy metals from street dust by runoff and drainage is an important daily issue, which may cause problems in various treatment processes and exert adverse health effects on humans and animals [7, 8].

Combustion of fossil fuels produces large amounts of heavy metals, such as nickel, chromium, lead, and manganese, which have accumulative and carcinogenic properties and may cause numerous health and environmental issues. Exposure to these elements could also lead to low levels of consciousness, renal disorders, and even death due to long-term exposure [9,10]. In general, the degradability and aggregation of these elements in the dust particles that cause toxicity in some cases (e.g., cancer) are considered to be a severe issue. Several studies have shown that urban soil is exposed to lead more than rural areas due to industrial activities [11]. However, the previous studies in this regard have mainly been focused on the levels of these elements, pollution assessment, and evaluation of the potential ecological risk of heavy metals in urban dust [12-14]. Due to the contamination potential of these metals, street dust is considered to be a significant source of urban area pollution, which also enables the measurement of numerous contaminants.

The present study aimed to determine the concentrations of lead and nickel, as well as their spatial distribution patterns, using ArcGIS for the mapping of the spatial patterns of their levels in street dust samples.

2. Materials and Methods

2.1. Study Area

Zanjan city (48°31' E, 36°39' N) is located in the northwest of Iran at the average latitude of 1,638 meters above the sea level. The average annual precipitation of this city is approximately 1,130 millimeters, and the average temperature has been estimated at 11.48°C. Zanjan city covers an area of 81 square kilometers. According to the population census in 2016, Zanjan has 433,475 habitants [15].

2.2. Dust Sample Collection

In total, 52 dust samples were collected from Zanjan city in 2018-2019 with full-covered distribution with the aim of covering the entire area of Zanjan city with an acceptable number of samples. Figure 1 shows the position of the sampling points for the mapping of the spatial distribution

of lead and nickel. During the sample collection, the surface physical debris (e.g., thorns, shavings, roots, sand) was removed from the dust samples. Following that, the samples were collected using a rigid plastic brush. In addition, large impurities (e.g., rocks and roots) were removed using a stainless sieve (mesh: 2 mm).

At each sampling point, five roadside dust samples were collected (minimum distance: 100 m) and mixed thoroughly in order to obtain a representative bulk sample. The collected samples were preserved in polyethylene plastic bags.

In order to record the geographical location of the samples (longitude and latitude), we used a hand-held GPS locator (model: Garmin GPS map 629sc, China). Data on the sampling date, number of the samples, weather conditions, dust texture, and color were recorded, as well as the data on the surrounding traffic and visitor flow rates from each sampling site. Afterwards, the samples were transferred to the Environmental Science Research Laboratory at the Department of Environmental Sciences of the University of Zanjan. After dust sampling, vegetation debris was reduced for higher data quality.

2.3. Analytical Procedure

After transferring the dust samples to the laboratory, they were expanded on paper to air-dry for five days. The dust samples were ground using an agate mortar and sieved sequentially through a two-millimeter mesh sieve to remove the remaining impurities. At the next stage, the prepared dust samples were placed in polyoxyethylene sample bags again until the digestion procedure. All the experiments were performed at the Environmental Science Research Laboratory of the University of Zanjan.

The concentrations of lead and nickel in the dust samples were assessed. Initially, 0.5 gram of the dust sample was added to the contents of a digestion tube. Following that, six milliliters of concentrated hydrochloric acid (37% HCl; Merck, Germany) and three milliliters of concentrated nitric acid (69% HNO₃; Merck, Germany) was added to each sample. The mixture was pre-digested at room temperature for 16 hours, and the sample tube was digested at the temperature of 140°C until the brown fumes ceased. Afterwards, the temperature was raised to 180°C and maintained until no further brown fumes were emitted. The obtained solution was cooled to room temperature and filtered through an ash-less Whatman 41 filter paper, diluted to 50 milliliters with distilled water, and stored in polyethylene bottles at room temperature. Finally, a flame atomic-absorption spectroscopic device (FAAS; model: Variant 220) was used to measure the lead and nickel contents in each sample.

2.4. Pollution Indexes

In order to recognize the dust quality and metal contamination in Zanjan city, we considered other approaches for environmental assessment with various advantages. Some international evaluation methods are used to study heavy metals in the sediments, such as the enrichment factor (EF), geo-accumulation index (I_{geo}), potential ecological risk index (RI), and pollution load index (PLI) [16-19]. In the present study, these indices were used for the quality assessment of the dust samples.

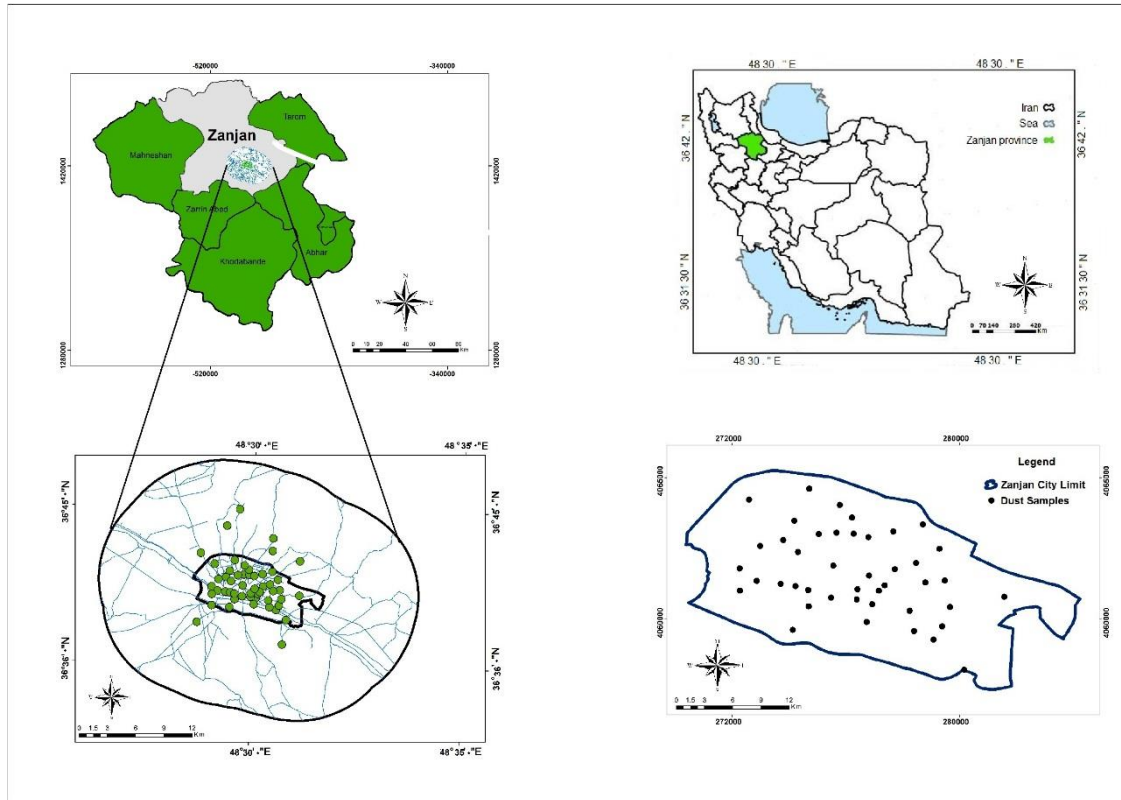


Figure 1: Spot Distribution of Dust Sampling Stations in Zanjan City

2.4.1. Enrichment Factor

EF (Eq. 1) was used to identify the heavy metal sources. EF could be used to evaluate sediment quality and distinguish metal pollution. In Equation 3, C shows concentration, s and Fe represent the mean studied elements and iron, respectively, and sample and background are the mean samples and background samples, respectively. It is notable that iron and aluminum were utilized as the normalization elements to diminish the variations generated by the heterogeneous sediments [20, 21]. In addition, the geochemical average shale values were used in the EF calculation, which were estimated at 68.0 and 20.0 mg/kg-1 for nickel and lead, respectively [22].

$$EF = \left(\frac{C_s}{C_{Fe}} \right)_{Sample} / \left(\frac{C_s}{C_{Fe}} \right)_{Background} \tag{1}$$

EF values indicate the origination of elements (natural sources such as the weathering processes of rocks or anthropogenic sources) and environmental contamination status of the elements. In this context, lower EF values than 1.5 confirm that heavy metals move to sediments from the materials found in the earth crust or through natural weathering processes. Higher EF values than 1.5 show that the considerable source of the metal is rendered from anthropogenic sources, which could be an important contributor to crustal materials [23-25]. Therefore, the following categories could be obtained from EF values [26]: EF < 1: no enrichment; 1 ≤ EF < 3: minor enrichment;

3 ≤ EF < 5: moderate enrichment; 5 ≤ EF < 10: moderately severe enrichment; 10 ≤ EF < 25: severe enrichment; 25 ≤ EF < 50: very severe enrichment; EF > 50: extremely severe enrichment. In the present study, EF% was assessed using Equation 2 [27], as follows:

$$EF(\%) = \frac{C - C_{min}}{C_{max} - C_{min}} \times 100 \tag{2}$$

In the equation above, C shows the mean heavy metal concentration in the sediment, C_{max} is the maximum concentration, and C_{min} represents the minimum measured concentration.

2.4.2. Geo-Accumulation Index (I_{geo})

In the present study, I_{geo} was used to recognize the sediment contamination extent in the studied area, which was assessed using Equation 3 [28]:

$$I_{geo} = \text{Log}_2 \left(\frac{C_n}{1.5 B_n} \right) \tag{3}$$

In which C_n shows the number of the heavy metals found in sediment samples, and B_n represents the background value of the heavy metals (geochemical/original source). Factor 1.5 was the correction factor for the background matrix, which was considered for lithospheric effects [16-19]. Moreover, seven classes of geo-accumulations were distinguished with the following index values: I_{geo} ≤ 0: class

zero (practically uncontaminated); $0 < I_{geo} \leq 1$: class I (uncontaminated to moderately contaminated); $1 < I_{geo} \leq 2$: class II (moderately contaminated); $2 < I_{geo} \leq 3$: class III (moderately to heavily contaminated); $3 < I_{geo} \leq 4$: class IV (heavily contaminated); $4 < I_{geo} \leq 5$: class V (heavily to extremely contaminated) and $I_{geo} > 5$: class VI (extremely contaminated).

2.4.3. Potential Ecological Risk Index Method

In the present study, the potential ecological risk index (RI) was applied for the contamination rating of the heavy metals. In addition, the Hakanson potential ecological RI was calculated using the following Equations 4-7, as follows:

$$C_f^i = \frac{C_s^i}{C_n^i} \tag{4}$$

$$E_r^i = T_r^i \times C_f^i \tag{5}$$

$$RI = \sum_{i=1}^m E_r^i \tag{6}$$

$$C_d = \sum_{i=1}^m C_f^i \tag{7}$$

In these equations, " C_f^i " is the contamination factor for the i th heavy metal, " C_s^i " shows the i th heavy metal concentration in the selected sediment sample, " C_n^i " is the reference value of the heavy metal (background value of each heavy metal in soil), C_d represents the monomial and polynomial contamination factors [18, 29], " E_r^i " represents the potential ecological risk factor for a given contaminant (i), and " T_r^i " is the toxic response factor of each element (Pb = 5, Ni = 6) [30-32]. The C_d , " E_r^i " and RI values were denoted in the following terms [18, 33, 34]: $C_d < 8$, low ecological risk; $8 < C_d \leq 16$, moderate ecological risk; $16 < C_d \leq 32$, high ecological risk; $16 < C_d$, very high ecological risk; " $E_r^i < 40$, low ecological risk; $40 < E_r^i \leq 80$, moderate ecological risk; $80 < E_r^i \leq 160$, significant ecological risk; $160 < E_r^i \leq 320$, high ecological risk and " $E_r^i > 320$, severe ecological risk; $RI < 150$, low ecological risk; $150 \leq RI < 300$, moderate ecological risk; $300 \leq RI < 600$, significant ecological risk and $RI > 600$: very high ecological risk.

2.4.4. Pollution Load Index

The pollution load index (PLI) is a potent tool for the evaluation of heavy metal pollution. In the current research, the PLI values were used to determine the integrated pollution status of the combined toxicant groups at the sampling stations by calculating the n th root of the product of n CF for the studied heavy metals using Equation 8, as follows [35, 36]:

$$PLI = (CF_1 \times CF_2 \times \dots \times CF_n)^{\frac{1}{n}} \tag{8}$$

In the equation above, the PLI values of >1 indicated pollution, while the values of <1 indicated no pollution [26, 37].

2.4.5. Statistical Analysis

Data analysis was performed in SPSS version 22.0 and Microsoft Excel (2010; Armonk, NY, USA). The homogeneity and normality of the collected data were assessed using the Kolmogorov-Smirnov and Levene's test. The P -value was considered as the probability for obtaining the test statistics (e.g., Kolmogorov-Smirnov statistic), which was at least as extreme as the value calculated from the samples in the case of the normal data. Larger values of the Kolmogorov-Smirnov statistic indicated that the data did not have normal distribution, and could not be confirmed ($P < 0.05$), requiring nonparametric tests. In addition, the analysis of variance (ANOVA) with line charts were used to determine whether there was a significant difference in the heavy metal concentrations (mg/kg) at the studied sites (city center, along the highways, and away from the highway). All the significant statistical tests were performed, and the results were expressed with lower probability (P) than 0.05. For the spatial distribution description of the studied elements, map patterns were provided using the ArcGIS 10.3 software (ESRI, Redlands, CA, USA) and inverse distance weighted (IDW) interpolation method.

3. Results and Discussion

3.1. Lead And Nickel Concentrations in Dust Samples

Table 1 shows the data on the heavy metal concentrations (mean, maximum, minimum, and standard deviation) in the dust samples. According to the obtained results, the ranges of the mean concentrations of nickel and lead in Zanjan were 72 (32-154) and 745 (30-4610) mg/kg, respectively. The highest and lowest lead and nickel concentrations were determined to be 4,610 and 745 mg/kg in in Jade-Bijar area, respectively and 30 mg/kg in Painkoh area. Therefore, there was an evident tendency toward the accumulation of heavy metals in the dust of the studied regions.

According to the findings of the current research, the lead and nickel concentrations in the street dust of Zanjan city exceeded the respective background values. The main causes of such high concentrations were the lead and zinc industrial plants, vehicles, local traffic, and geochemistry of the area. Some important variables in the study (e.g., traffic, industrial towns, and wind direction) were also considered to be the major potential sources of air pollution with heavy metals in the region. In a study in this regard, Farahmandkia et al. (2017) reported that 25-40% of the suspended particles in the air of Zanjan originated from resuspended solids and dust, confirming that in the downtown of Zanjan, the influential factors producing particulate matter were soil particles (40.36%), fuel combustion and traffic (26.8%), tailing soils (lead and zinc) (21.32%), and nickel and industrial emissions (5.7%) [38].

Table 1: Summary of statistical concentration values (mg/kg) in the street dust samples of the study area

Heavy metals	Minimum (mg/kg)	Maximum (mg/kg)	Average value (mg/kg)	Average shale (mg/kg)*	SD**
Ni	32	154	72	68	22
Pb	30	4610	745	20	855

* Average shale, World geochemical background concentration
 ** Represents standard deviation

3.2. Spatial Distribution of Lead and Nickel

For the proper presentation and analysis of the data, geographical information systems and remote sensing have recently been used as digital and computerized technologies for the interpretation and presentation of spatial data and geochemical modeling. Furthermore, the spatial distribution of the concentrations of the studied contaminants and materials is an effective visual method to assess the probable sources of enrichment and hotspots identifying with high heavy metal concentrations [16,17].

In the present study, the spatial distribution of lead and nickel in the dust obtained from the sampling sites were described using spatial distribution pattern maps in the ArcMap software version 10.3. In addition, the spatial distribution patterns of lead and nickel were analyzed using the Kriging tool method in the ArcGIS software after the validation of the method. A spatial database was also prepared in the ArcGIS software, and the lead and nickel concentrations were linked to the sampling stations. Figure 2-a depicts the spatial distribution pattern of nickel in the dust samples collected from the studied areas. Correspondingly, the spatial distribution of nickel had lower variability in the studied area.

Due to the normal distribution and spatial pattern of nickel, it could be claimed that nickel had background values or geochemistry sources. In other words, nickel was not strongly influenced by anthropogenic activities. Since there were no significant direct industries and the main activities in the research area were represented, the nickel content of the dust samples might have been slightly influenced by anthropogenic sources (e.g., lead). The distribution of nickel was similar to lead distribution in the study area (Figure 2-b). Therefore, it could be concluded that lead and nickel had the same sources in the area. Due to the lead, zinc, and edible oil factories in the east, southeast, and south of the study area, the elements had several hotspots.

Figure 2-b shows the lead concentration in the dust samples collected from the entire urban area of Zanjan. Accordingly, the minimum lead concentrations were observed in the central, northern, and southeast regions of Zanjan. With some exceptions, the lead concentration patterns in the dust samples were similar in a wide area, especially in the central part of the studied area, and only three areas of the analyzed samples were not normal in this regard.

According to the map of lead spatial distribution, the hotspots (areas with the highest concentration of lead) were located in the northeast, west, and southwest of the study area, which were in the vicinity of industrial areas (Koyeh Farhang, Koye Samin, Khayyam, and Bijar roads). In addition, high lead concentrations were detected near the areas affected by human activities and a major road with significant traffic. Similar to other studies [1-4], these findings confirmed that the high lead concentrations in the urban dust were in the areas with numerous vehicular exhausts or in the vicinity of the local industries. The contribution of lead and zinc industries of Zanjan to the emission of carcinogenic metals has been reported to be 42.3% higher compared to the other sources [39]. The other studies in this regard have also confirmed that the children in Zanjan city may be high-risk for cancer, non-cancer diseases, and heavy metal pollution [39-41].

3.3. EF Values of Lead and Nickel in Street Dust

The calculated EF values for the dust samples showed a wide range of lead and nickel enrichment (Table 2).

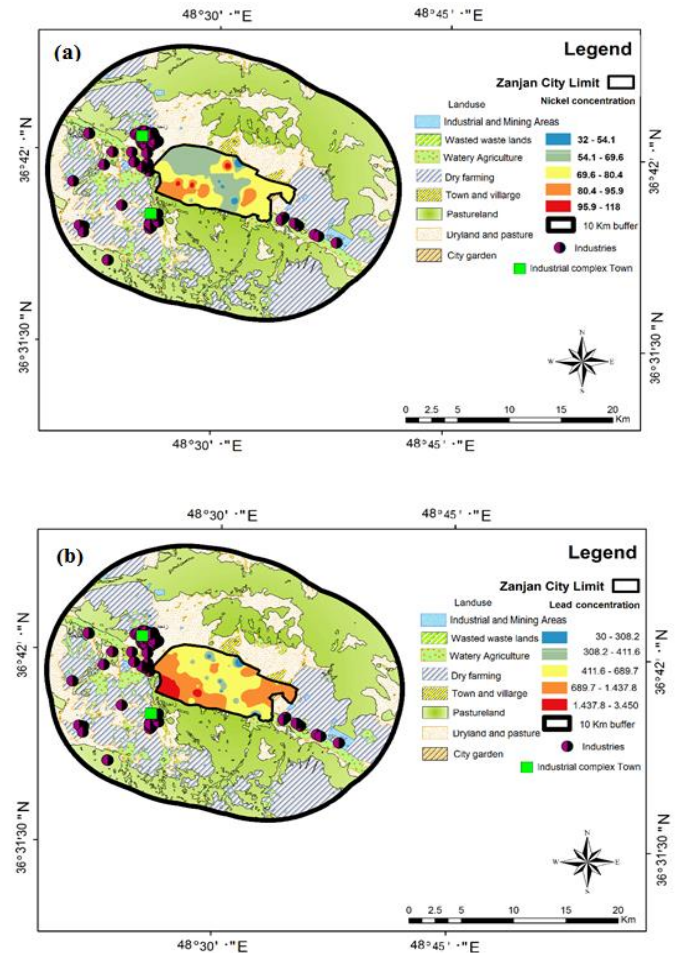


Figure 2: Spatial Distribution Maps of a) Nickel and b) Lead in Zanjan Roads

In general, the mean values indicated no enrichment for nickel and severe enrichment for lead. Moreover, 11% of the sample exhibited extremely severe lead enrichment, while 25% showed minor enrichment in this regard (Figure 3).

3.4. Geoaccumulation Index Values of Lead and Nickel in the Dust Samples

The Igeo values of nickel and lead in the street dust samples are presented in Table 2. Accordingly, the mean Igeo for nickel and lead were confirmed in class I (uncontaminated to moderately contaminated) and class V stations (heavily to extremely contaminated), respectively. In the case of nickel, 48% of the dust samples were in class zero (practically uncontaminated), 50% were in class I (uncontaminated to moderately contaminated), and 2% were in class II (moderately contaminated).

Therefore, the concentration of lead was higher in the dust samples compared to nickel. In terms of the lead concentrations, the dust samples were categorized as class I (uncontaminated to moderately contaminated; 2%), class III (moderately to heavily contaminated; 2%), class IV (heavily contaminated; 28%), class V (heavily to extremely contaminated (48%), class VI (extremely contaminated; 32%). Figure 4 shows the Igeo value mapping of nickel and lead.

Table 2: Summary of statistical values EF , I_{geo} , E_r^i , C_f^i , C_d and RI in the street dust of in Zanjan City roads

	Ni				Pb						
	EF	I_{geo}	C_f^i	E_r^i	EF	I_{geo}	C_f^i	E_r^i	C_d	RI	PLI
Min	0.30	-1.09	0.47	2.82	2.10	0.58	1.50	7.50	1.97	10.32	0.84
Max	2.73	1.18	2.26	13.59	277.56	7.85	230.50	1152.50	232.76	1166.09	22.85
Mean	0.90	0.03	1.06	6.38	31.67	4.63	37.27	186.35	38.33	192.72	5.83

3.5. Hakanson Potential Ecological Risk Index Values of Lead and Nickel in the Dust Samples

The Hakanson potential ecological risk index is shown in Table 2, including the calculated C_f^i , E_r^i , C_d and RI values for nickel and lead. The obtained contamination factor (C_f^i) indicated that the concentrations of nickel and lead in the dust samples of the study area were 0.58-7.84 and 1.50-230.50 times higher than the reported background values, respectively.

According to the information in Table 2, the scopes of the minimum, mean, and maximum potential ecological risk indices of nickel and lead were E_r^i : 2.82, 6.38, and 13.59 and 7.50, 186.35, and 1,152.50, respectively. Due to E_r^i (Ni), all the sampling stations had low ecological risk, while due to the lead content, they are classified as low ecological risk (4%), moderate ecological risk (29%), significant ecological

risk (35%), high ecological risk (19%), and severe ecological risk (13%).

Cadmium levels are considered to be a polynomial contamination factor, confirming that the sampling stations in the present study were classified as low (4% of dust samples), moderate (30%), high (35%), and very high levels (36%). The obtained RI level also indicated that the stations were at low (62%), moderate (23%), high (10%), and very high ecological risk (6%) (Figure 5-a).

3.6. PLI of Lead and Nickel in the Dust Samples

The estimated PLI for the sampling stations is presented in Table 2. Accordingly, the mean PLI in all the station ranged from 0.84 (minimum) to 22.85 (maximum). the collected dust samples from Zanjan city were considered polluted, with the mean PLI value estimated at 5.83. Moreover, higher PLI values were observed in the southwest of Zanjan (Figure 5-b).

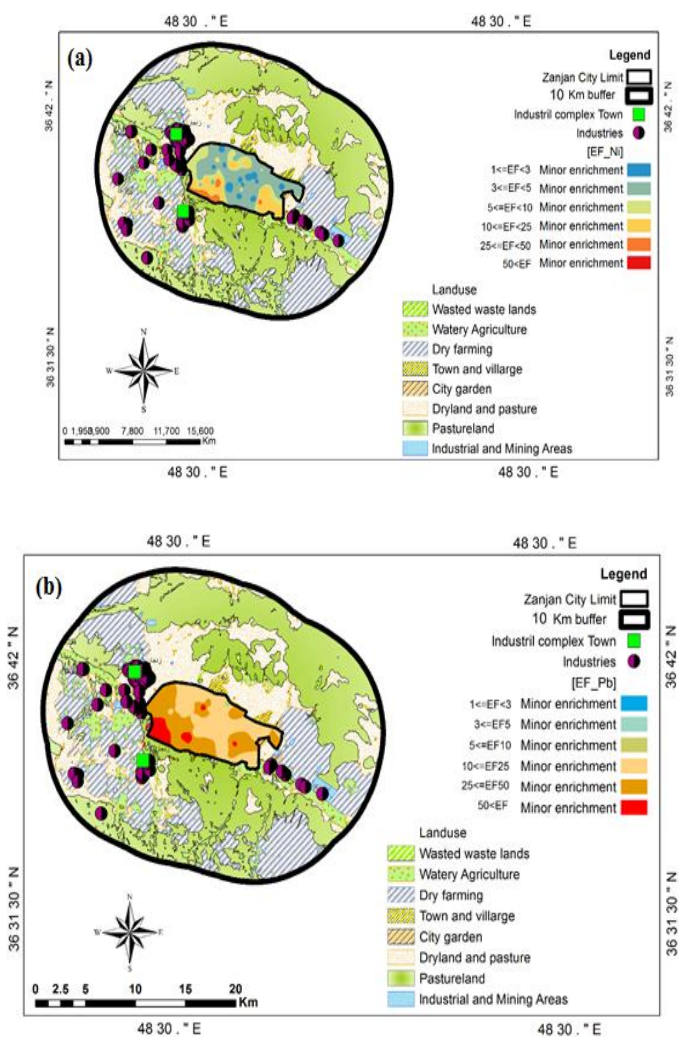


Figure 3: Spatial Distribution Maps of EF for a) Nickel and b) Lead in Zanjan Roads

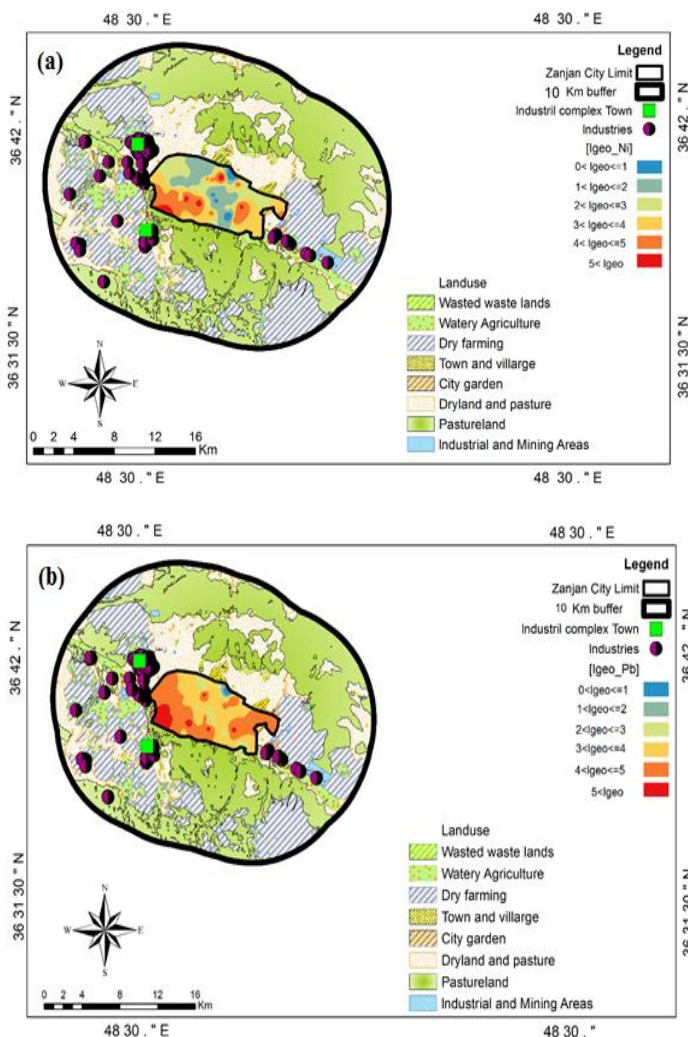


Figure 4: Spatial Distribution Maps of Geoaccumulation Index Values for a) Nickel and b) Lead in Zanjan Roads

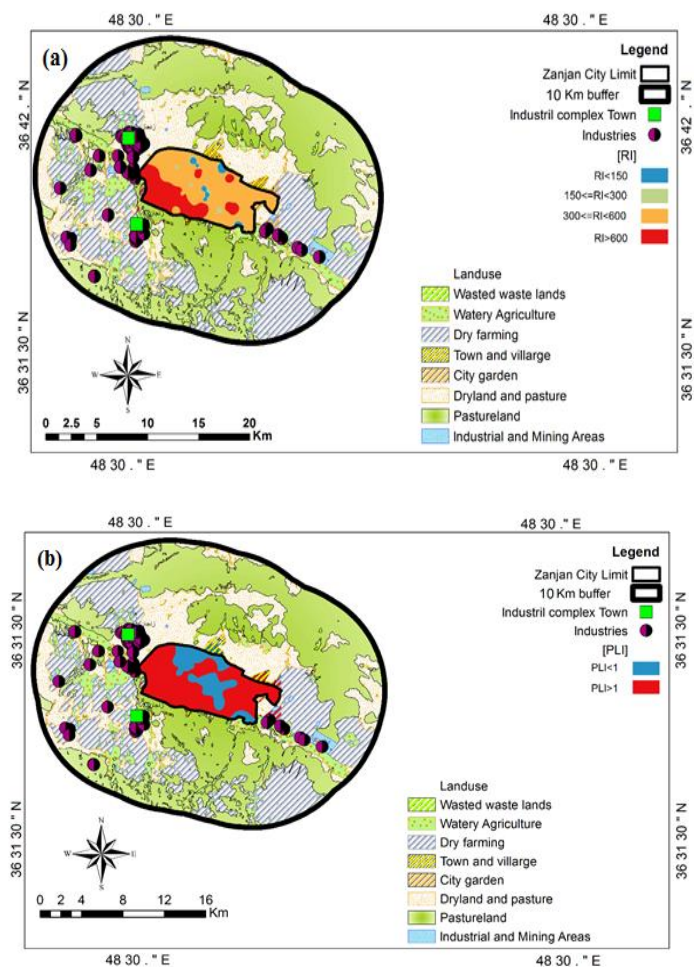


Figure 5: Spatial Distribution of a) RI and b) PLI in Dust Samples in Zanjan Roads

4. Conclusion

Street dust is a major source of the dispersion of heavy metal contamination in urban environments, and the issue is becoming increasingly problematic. The heavy metals produced by human activity significantly affect human health in residential areas. In this study, the street dust of Zanajn city was analyzed to determine the effects of traffic density and industrial centers on the heavy metal concentrations in an urban area. A flame atomic-absorption device was used to collect 52 street dust samples from Zanjan city, which were assessed in terms of lead and nickel concentrations, with the values estimated at 745 and 72 mg/kg, respectively.

The permissible levels of heavy metals in street dust have not been established so far, and it is inappropriate to compare the obtained values directly with such standards for soil or air. In general, it could be concluded that the mean content value of nickel was lower than which was indicated for agriculturally used soils in Poland (Ni: 100 mg/kg, Pb: 100 mg/kg). It is also notable that in the case of soil, heavy metals are immobilized to some extent and may be absorbed in the human body mainly through food, such as the plants that absorb these elements from soil.

In the case of street dust, heavy metals could be released via air mass movement and absorbed directly by the

respiratory system. Therefore, even low levels of heavy metals may be harmful [42]. In this study, the concentration of lead was generally higher than the background values. The interpolation maps of the heavy metals in the ArcGIS software identified various sources of heavy metals by presenting the individual distribution of these elements (e.g., lead), which may be attributed to industrial and human activities. Therefore, it could be inferred that the highest concentration of nickel was in the north, east, and southwest of Zanjan city.

In the case of nickel, this element is mainly controlled by geogenic and pedogenic sources. In addition, fertilizers could easily influence nickel concentrations. In this study, the Hakanson potential ecological risk index values indicated that the dust samples had a high ecological risk, while the PLI in Zanjan showed pollution, which requires special attention. Therefore, awareness of the spatial distribution of the dust contaminated by heavy metals would play a key role in evaluating the pollution potential of a region.

Authors' Contributions

This article was carried out by all the authors. A.Z., Y.Kh., and A.P., conceived, designed and planned the study and the experiments. M.Sh. and Z.Sh., carried out the experiments (sampling, sample preparation, digestion, heavy metal determination and initial analysis). All authors provided critical feedback and helped shape the research, analysis and contributed to preparation the manuscript.

Conflict of Interest

The Authors declare that there is no conflict of interest.

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ABBREVIATIONS

<i>Pb</i>	Lead
<i>Ni</i>	Nickel
<i>ppm</i>	parts-per-million
<i>GIS</i>	Geographic Information System
<i>IDW</i>	Inverse Distance Weighted
<i>mg/kg</i>	Milligram per Kilogram
<i>WHO</i>	World health organization
<i>GPS</i>	Global Positioning System

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