Heavy Metal Contamination and Risk Assessment of Surface Soils of Babol in Northern Iran

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Received 2016 August 03; Revised 2016 December 09; Accepted 2017 January 13.

Abstract

Background: Currently there is much concern in the field of soil contamination with heavy metals due to rapid urbanization. Soil contamination significantly reduces environmental quality and affects human health.

Objectives: This study was performed to determine the level of As, Cd, Co, Cr, Ni, Pb, and Zn in the soils of Babol city in Mazandaran province and to evaluate the pollution indices of heavy metals.

Methods: Concentrations of heavy metals in 50 samples of surface soils (5 to 15 cm) in urban and rural areas of Babol city were measured using inductively coupled plasma optical emission spectrometry (ICP-OES). In this study, various indices including contamination factor (Cfi), degree of contamination (Cdeg), Nemerow integrated pollution index (NIPI) and the potential ecological risk (ERi) with heavy metals were assessed.

Results: Mean content of As, Pb, Cd, Zn, Cr, Ni, and Co in soil samples of different locations were 6.90 ± 2.60, 31 ± 5.70, 0.32 ± 0.1, 82.80 ± 15, 32.40 ± 8.30, 34.50 ± 7.50, and 22.6 ± 6 mg/kg, respectively. The average content of these metals is ranked in the following descending order: Zn > Cr > Ni > Pb > Co > As > Cd. The order of contribution of the potential ecological risk by metals was Cd (51.91%) > As (23.75%) > Pb (7.61%) > Cr (5.57%) > Co (5.24%) > Zn (3.04%) > Ni (2.88%).

Conclusions: This study indicated that the risk assessments of the soil samples with heavy metals in the studied locations are categorized from low to moderate degree of contamination.

Keywords: Heavy Metals, Surface Soil, Pollution Index, Ecological Risk Index.

1. Introduction

Global industrialization and unsystematically agricultural activities have been effected natural environments and ecosystems (1, 2). Contamination of environments with heavy metals has been increased significantly after the start of the industrial revolution (3). Soil contamination with heavy metals has been reported in both developing and developed countries (4).

The metals are categorized as “heavy metals” that have a specific gravity of more than 5 g/cm³. There are well-known 60 heavy metals around the world (2). Overall, every 1000 kg of normal soil theoretically contains 200 g of chromium, 80 g nickel, 16 g lead, 0.5 g mercury, and 0.2 g cadmium (5).

Heavy metals enter human bodies by direct pathways, such as ingestion, inhalation, dermal contacts, and indirect ways or through the food chain (6). These elements may be accumulated in soils, plants, and animals, and eventually reach human beings (7, 8). Heavy metals that are accumulated in plants could have a contradictory effect on physiological functions of plants such as photosynthesis, gaseous exchange, and nutrient absorption (6).

According to previous research, heavy metals are highly hazardous to human body and could cause pathological change of organs and diseases related to the cardiovascular system, kidneys, bones, etc, and could even cause cancer owing to excessive heavy metal accumulation in human bodies (9, 10). These elements are also known to be potential inhibitors of ATPases (11). These compounds enter an organism and reach the target organ by passing through living membranes and in the process, adversely effect their permeability and degrade normal enzyme transport (12). The most dangerous heavy metals are the compounds that bind to cell membranes affecting the intracellular transport process in living forms (13).

Although heavy metals are naturally present in soil, there is metal pollution due to local sources, such as power plants and iron (14, 15), steel and chemical industries (16), agriculture activities, including irrigation with polluted waters (17), sewage sludge and fertilizers (18), contaminated manure and pesticide containing heavy metals (19), waste incineration (20), combustion of fossil fuels, and road traffic (21, 22). Long-range transport of atmospheric pollutants adds to the contamination of metals in the nat-
ural environment (23).

Contamination of soils with heavy metals, especially in agricultural soils, is more important because of their potential transfer from plants to animals (feed crops) and animals to humans (food crops and vegetables) (24, 25). The use of the contaminated sludge with heavy metals, fertilizers, and animal manures could cause high contents of metals in the soils of urban and rural areas (26, 27). On the other hand, little information is available regarding human exposure and risk assessment of the soil contaminated with heavy metals in urban and rural areas in developing cities (28).

In one study by Azimzadeh et al. concentrations of heavy metals and the contamination of farmland soil samples, in 60 and 48 soil samples from urban and natural areas in center of Mazandaran province in Iran were evaluated (29). Soleimannejad et al. compared the concentrations of cadmium, zinc, copper, lead, arsenic, and iron in the soil of industrial sites and landfills of Ghaemshahr city in Mazandaran (30). Ravankhah et al. evaluated soil contamination with heavy metals in Aran and Bidgol districts of Kashan in Iran. They found that soil contamination with lead and cadmium originated from human activities, and nickel, zinc and copper were affected by natural and human factors (31).

Gong et al. studied soil pollution in rural and urban areas with potentially dangerous heavy metals (Cd, Co, Cr, Cu, Hg, Mn, Pb, Ni and Z) in the state of Wuhan, China (26). Dantu studied the situation of soil contamination in Ranga Reddy district of the southeastern part of the state of Andhra Pradesh in India to heavy metals (As, Ba, Co, Cr, Mo, Ni, Pb, Rb, Sr, V, Y, Zn, and Zr) with X-ray fluorescence spectrometry (2). This investigation was performed to determine the level of As, Cd, Co, Cr, Ni, Pb, and Zn in collected soil samples from agricultural areas of Babol district in center of Mazandaran province and to evaluate pollution indices such as contamination factor (Cf) and degree of contamination (Cdeg), Nemerow integrated pollution index (NIPI), and potential ecological risk (RI) in the studied locations.

2. Methods

2.1. Study Area

Babol city with a population of 520,000 people is the most populated city in Mazandaran province and is located in northern Iran. The city of central Mazandaran in southern Caspian Sea (15 km from Caspian Sea) is located at a longitude of 52 ° 44′ and latitude of 36° 24′ 15″. Average annual precipitation and temperature is 17.1°C and 799 mm, respectively. This region is one of the most important agricultural areas of Iran (29). Another characteristic of this area is the establishment of various industries and factories. The city has several rivers, especially Babolrood River that comes into the Caspian Sea.

2.2. Sample Collection and Preservation

Fifty soil samples in depth of 5 to 15 cm were collected from different locations of urban and rural places of Babol city (Figure 1). These composite soil samples were collected in polyethylene bags and transferred to the laboratory. The soil samples were air-dried, grounded, passed through a standard 2-mm sieve and sealed in clean bags and then stored in a refrigerator (18).

2.3. Sample Preparation and Analysis

One gram of dried soil samples was digested in a digestion system (21-mL HNO3 + 7-mL HCL). The digested samples were filtered through Whatman paper (No.42) and deionized water was added to reach 50 mL and stored at 4°C. The heavy metal contents in soil samples were determined by inductively coupled plasma atomic emission spectrometry (ICP-AES), SPECTRO model, under optimum conditions. The instrument calibration was used for the quantification of selected metals and the samples (32, 33). Preparation of the reagents and calibration standards, and double distilled water was performed. The standard solutions were prepared from stock solution of 1000 mg/L by dilutions. All the soil samples were gathered with 3 replications and blank samples were analyzed simultaneously.

2.4. Determination of Contamination Factor (Cf) and Degree of Contamination (Cdeg)

The assessment of soil contamination is performed by Contamination factor (Cf) and degree of contamination indices. The contamination factor of a metal is determined by the following equation:

\[
C_f = \frac{C_{mi}}{C_{ni}}
\]

That:

- \(C_{mi}\) is the mean concentration of metals.
- \(C_{ni}\) is the natural concentration of individual elements (background level).

Hakanson (34) defines four categories (Cf), as follows:

- a. \(C_f < 1\) low contamination factor => low contamination
- b. \(1 < C_f < 3\) moderate contamination factor => mean contamination
- c. \(3 < C_f < 6\) considerable contamination factor => high contamination
- d. \(C_f > 6\) very high contamination factor => very high contamination
The $C_i$ is a single element index, and the sum of contamination factors for all metals is indicated by the contamination degree ($C_{deg}$) of the location. The degree of contamination is classified in four classes (24):

- $C_{deg} < 8$ => low degree of contamination
- $8 < C_{deg} < 16$ => moderate degree of contamination
- $16 < C_{deg} < 32$ => considerable degree of contamination
- $C_{deg} > 32$ => very high degree of contamination

2.5. Evaluation of Pollution Index (PI)

The pollution level of a metal was determined by a single pollution index ($PI_i$). The $PI_i$ was calculated with the following equation:

$$PI_i = \frac{C_i}{S_i}$$  \hspace{1cm} (2)

Where $C_i$ = Metal concentration in a soil sample
$S_i$ = Reference value

The total potential pollution of soil samples by heavy metals was evaluated by the Nemerow integrated pollution index (NIPI).

$$NIPI = \sqrt{\frac{PI_{ave}^2 + PI_{imax}^2}{2}}$$  \hspace{1cm} (3)

Where Plave and Pimax are the average and maximum of the pollution indices for individual metals, respectively. The NIPI is categorized as follows: 1. NIPI < 0.7 => non-pollution; 2. 0.7 < NIPI < 1 => warning line of pollution; 3. 1 < NIPI < 2 => low level of pollution; 4. 2 < NIPI < 3 => moderate level of pollution; 5. NIPI > 3 => high level pollution (26).
Estimation of the Potential Ecological Risk

The potential ecological risk of heavy metal in soil samples was evaluated using the ecological risk index (RI) (27). The RI index was determined by the sum of risk factors of the metals:

\[ RI = \sum_{i=1}^{7} Er^2 \]  

Where \( Er^i \) is the risk factor of any metal, and is determined as:

\[ Er^i = Tr^i \times C_{if}^i = Tr^i \times C_i/B_i \]

\( Ti \) = the toxic-response factor for any metal.

The Toxic- response values of As, Pb, Cd, Zn, Cr, Ni, and Co are 10, 5, 30, 1, 2, 2, and 2, respectively.

\( C_{if} \) = the metal contamination factor calculated from the measured concentration (\( C_i \)) and the background concentration (\( B_i \)) of the metal.

The potential ecological risk of heavy metals in soils was classified to four categories:

a. RI < 150 => Low ecological risk
b. 150 < RI < 300 => Moderate ecological risk
c. 200 < RI < 600 => Considerable ecological risk
d. RI > 600 => Very high ecological risk

Statistical Analysis

Data analysis was done with the Excel software, using one-way Analysis of Variance (ANOVA) and the t-test.

Results

Heavy metals content in soils of selected locations

The range of As, Pb, Cd, Zn, Cr, Ni, and Co in the soil samples were 3.1 to 12.6, 10.5 to 62.5, 0.11 to 0.5, 25.8 to 143, 20.5 to 92.6, 17.1 to 79, and 5.8 to 37.5 mg/kg, respectively (Table 1). The total concentration in the soil samples was as follows, Zn > Cr > Ni > Pb > Co > As > Cd. Overall, 80% for all samples had a content of As below 7.5 mg/kg, Pb below 36.2 mg/kg, Cd below 0.35 mg/kg, Zn below 89 mg/kg, Cr below 31 mg/kg, Ni below 35.1 mg/kg, and Co below 25.4 mg/kg. The background levels of As, Pb, Cd, Zn, Cr, Ni, and cobalt in natural and unpolluted soils of these locations were 5.2, 36.5, 0.3, 48.5, 20.8, 41.6, and 15.4, respectively. The mean contents of As, Cr, Co, and Zn in all soil samples was higher than their background levels. The mean concentrations of Cd and Pb in 40% and 20% of soil samples were higher than their background contents, respectively (Table 2).

Determination of contamination factor (CF), degree of contamination (Cdeg) and Nemerow Integrated Pollution Index (NIPI) of Heavy Metals in Soil Samples of Selected Locations

Average contamination factors of selected metals in the soil samples are shown in Table 3. Among the selected metals, mean CF of As, Pb, Cd, Zn, Cr, Ni, and Co were determined as 1.32, 0.85, 0.97, 1.70, 1.42, 0.84, and 1.34, respectively. The degree of contamination (Cdeg) of different locations containing L1, L2, L3, L4, and L5 was 7.08, 7.82, 8.43, 9.06, and 9.57, respectively. The mean and range of Cdeg in the soil samples from different locations were 8.41 and 3.57 to 17.53, respectively.

In Table 4, the range of pollution index of Plave, Plmax, and NIPI in the soil samples of the studied locations was 0.51 to 2.50, 0.99 to 4.45 and 0.79 to 3.61, respectively. The NIPI for most of the samples (80%) showed a low level of pollution. The analysis of results showed that the average amount of the elements was in the following order, Zn (1.70) > Cr (1.42) > Co (1.34) > As (1.32) > Co (0.97) > Pb (0.85) > Ni (0.81).

Estimation of Ecological Risk of Heavy Metals in Soils of the Selected Locations

Table 5 shows the mean ecological risk factors of different metals in the soil of locations and their participation in the total potential ecological risk. Order of ecological risk of heavy metals was Cd > As > Cr > Pb > Co > Ni (Figure 2). The average and range of contents of the potential ecological risk for soils of urban and rural areas was 58.48 and 22.47 to 103.31, respectively.

Discussion

Mean content of As, Pb, Cd, Zn, Cr, Ni, and Co in soil samples of different locations were 6.9 ± 2.60, 31 ± 5.70,
Table 1. Heavy Metals Concentrations in the Surface Soils from the Selected Locations

| Location Sample | As   | Cd   | Pb   | Zn   | Cr   | Ni   | Co   | TotalHM |
|-----------------|------|------|------|------|------|------|------|---------|
| L1              | 54   | 7.4  | 24.6 | 107  | 6.3  | 0.23 | 0.02 | 170     |
| L2              | 34   | 36.8 | 15   | 12.6 | 5.1  | 2.5  | 0.1  | 89      |
| L3              | 44   | 36.8 | 15   | 12.6 | 5.1  | 2.5  | 0.1  | 89      |
| L4              | 44   | 36.8 | 15   | 12.6 | 5.1  | 2.5  | 0.1  | 89      |
| L5              | 44   | 36.8 | 15   | 12.6 | 5.1  | 2.5  | 0.1  | 89      |

Table 2. Comparison of the Heavy Metals Content in the Studied Soil Samples and Maximum Allowable Contamination Based on European Union Standards

| Substance (name) | Mean ± SD, mg.kg⁻¹ | MAC, mg.kg⁻¹ |
|------------------|---------------------|--------------|
| Arsenic          | 6.9 ± 2.4           | 5            |
| Cadmium          | 0.32 ± 0.1          | 1            |
| Chrome           | 32.4 ± 8.3          | 100          |
| Cobalt           | 22.6 ± 6.3          | 20           |
| Lead             | 31 ± 5.7            | 60           |
| Nickel           | 32.5 ± 7.5          | 10           |
| Zinc             | 82.8 ± 15           | 200          |

Abbreviation: SD, standard deviation.

0.32 ± 0.10, 82.80 ± 15, 32.40 ± 8.30, 34.50 ± 7.50, and 22.6 ± 6 mg.kg⁻¹, respectively, and were larger than background levels. Total metal contents in the soil samples of different locations follow this descending order: L5 > L4 > L3 > L2 > L1. Amouei et al. determined Pb, Cd, and Zn concentrations in agricultural soils of Babol area. In this study, contents of Pb, Cd, and Zn was 19.75 ± 10.5, 0.11 ± 0.1, and 2.5 ± 3.1 mg/kg, respectively (2). Gong et al. studied the concentration of heavy metals in urban and rural top soils of Wuhan, central China. They concluded that the mean contents of Cd, Co, Cr, Pb, Ni, and Zn were 0.20, 16.5, 33, 34, and 85 mg/kg, respectively (12). Modrzewska and Wyszkowski determined that the mean concentrations of Pb, Cd, Cr, and Ni in soils along state Road 51 leading from Olsztyn to Olszynek, northeastern Poland, were 24.3 to 70.3, 0.31 to 0.85, 9.9 to 11.6, and 67.5 to 120 mg/kg, respectively (21). In this study, the background levels of As, Pb, Cd, Zn, Cr, Ni, and cobalt in natural and unpolluted soils of these locations were 5.2, 36.5, 0.3, 48.5, 20.8, 41.6, and 15.4, respectively.

Among the selected metals, mean contamination factors of As, Pb, Cd, Zn, Cr, Ni, and Co were determined as 1.32, 0.85, 0.97, 1.42, 0.84, and 1.34, respectively. The degree of contamination (C<sub>deg</sub>) of the different locations containing L1, L2, L3, L4, and L5 were 7.08, 7.82, 8.43, 9.06, and 9.57, respectively. In the current study, 40% of soil samples (L1, L2), based on contamination factor (C<sub>f</sub>) and degree of contamination (C<sub>deg</sub>), were found at low levels and the others (L3, L4, L5) were categorized at a medium degree. Sayadi et al. studied the surface soils of Amir-Abad of Birjand, based on pollution and ecological indices. The results showed that the ecological risk of surface soil for the users of road-residential areas was high and the agricultural land use had moderate ecological risk and dairy farm had low ecological risk (35). IQbal et al. showed that the average C<sub>deg</sub> values for the selected metal contents in urban soils from Islamabad, Pakistan during summer and winter were 16.67 and 20.50, respectively, indicating a considerable degree of contamination (33). The NPI for most of the
### Table 3. Contamination Factor (Cf) and Degree for Heavy Metals on the Surface of Soils

| Location | Sample | As | Pb | Cd | Zn | Cr | Ni | Co | Cdeg |
|----------|--------|----|----|----|----|----|----|----|------|
| L1       | 10     | 1.52 | 0.58 | 0.60 | 1.50 | 1.59 | 0.76 | 0.49 | 7.08<sup>a</sup> |
| L2       | 10     | 1.04 | 0.92 | 0.77 | 1.58 | 1.47 | 0.94 | 1.28 | 7.02<sup>a</sup> |
| L3       | 10     | 1.21 | 0.67 | 0.93 | 1.72 | 1.56 | 0.88 | 1.53 | 6.84<sup>a</sup> |
| L4       | 10     | 1.38 | 1.08 | 1.07 | 1.84 | 1.52 | 0.70 | 1.75 | 9.06<sup>b</sup> |
| L5       | 10     | 1.44 | 0.99 | 1.33 | 1.98 | 1.63 | 0.81 | 1.65 | 9.86<sup>b</sup> |
| Max      | -      | 2.42 | 1.71 | 1.67 | 2.95 | 4.45 | 1.90 | 2.43 | 17.53<sup>a</sup> |
| Min      | -      | 0.60 | 0.29 | 0.37 | 0.53 | 0.99 | 0.46 | 0.89 | 3.57<sup>b</sup> |
| Mean     | -      | 1.32 | 0.85 | 0.97 | 1.70 | 1.55 | 0.83 | 1.47 | 8.70<sup>b</sup> |
| SD       | -      | 0.52 | 0.35 | 0.38 | 0.6 | 0.55 | 0.33 | 0.46 | 1.2 |

Abbreviation: SD, standard deviation.
<sup>a</sup>Low degree of contamination.
<sup>b</sup>Medium degree of contamination.

### Table 4. The Pollution Indices of Heavy Metals in Soils from Selected Locations

| Location | PI<sub>As</sub> | PI<sub>Pb</sub> | PI<sub>Cd</sub> | PI<sub>Zn</sub> | PI<sub>Cr</sub> | PI<sub>Ni</sub> | PI<sub>Co</sub> | PIave | PI<sub>max</sub> | NIPI |
|----------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|-------|----------------|------|
| L1       | 1.04           | 0.92           | 0.77           | 1.58           | 1.59           | 0.76           | 0.49           | 1.04  | 1.63           | 1.36<sup>a</sup> |
| L2       | 1.04           | 1.08           | 1.27           | 1.99           | 1.72           | 1.72           | 1.20           | 1.20  | 1.58           | 1.58<sup>b</sup> |
| L3       | 1.04           | 0.99           | 1.33           | 1.98           | 1.72           | 1.72           | 1.20           | 1.20  | 1.58           | 1.58<sup>b</sup> |
| L4       | 1.38           | 1.08           | 1.27           | 1.99           | 1.72           | 1.72           | 1.20           | 1.20  | 1.58           | 1.58<sup>b</sup> |
| L5       | 2.42           | 1.71           | 1.67           | 2.95           | 4.45           | 1.90           | 2.43           | 17.53 | 3.61           | 3.61 |
| Max      | -              | 2.42           | 1.71           | 1.67           | 2.95           | 4.45           | 1.90           | 2.43  | 17.53           |      |
| Mean     | -              | 1.32           | 0.85           | 0.97           | 1.70           | 1.55           | 0.83           | 1.47  | 8.70           |      |
| SD       | -              | 0.52           | 0.35           | 0.38           | 0.6           | 0.55           | 0.33           | 0.46  | 1.2            |      |

Abbreviation: SD, standard deviation.
<sup>a</sup>Low level of pollution.

### Table 5. The Potential of Ecological Risk of Metals in the Soils of Selected Locations

| Location | Er<sup>1</sup><sub>As</sub> | Er<sup>1</sup><sub>Pb</sub> | Er<sup>1</sup><sub>Cd</sub> | Er<sup>1</sup><sub>Zn</sub> | Er<sup>1</sup><sub>Cr</sub> | Er<sup>1</sup><sub>Ni</sub> | Er<sup>1</sup><sub>Co</sub> | RI    | Risk Degree |
|----------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-------|--------------|
| L1       | 15.19           | 2.91            | 18              | 1.50            | 3.18            | 1.53            | 2.27            | 44.67 | Low          |
| L2       | 10.38           | 4.60            | 23              | 1.58            | 2.94            | 1.69            | 2.56            | 46.60 | Low          |
| L3       | 12.12           | 3.34            | 1.72            | 2.95            | 4.45            | 1.96            | 2.43            | 2.50  | 4.45         |
| L4       | 13.85           | 5.41            | 35              | 1.84            | 3.04            | 1.55            | 3.49            | 63.23 | Low          |
| L5       | 14.42           | 4.96            | 40              | 1.98            | 3.26            | 1.55            | 3.30            | 60.85 | Low          |
| Max      | 24.23           | 8.56            | 50              | 2.95            | 8.90            | 3.8             | 4.87            | 103.31|              |
| Min      | 5.96            | 1.44            | 11              | 0.53            | 1.67            | 0.62            | 0.75            | 22.47 |              |
| Mean     | 13.27           | 4.25            | 29              | 1.70            | 3.08            | 1.84            | 2.93            | 55.8  |              |
| SD       | 6.52            | 1.35            | 5.35            | 0.52            | 1.03            | 0.47            | 1.89            | 5.26  |              |

Abbreviation: SD, standard deviation.

4.1. Conclusions

Soil contamination and the potential risk assessment with heavy metals in Babol city of Mazandaran province samples (80%) showed a low level of pollution. The analysis results showed that the NIPI average for the elements was as follows, Zn (1.70) > Cr (1.42) > Co (1.34) > As (1.32) > Co (0.97) > Pb (0.85) > Ni (0.81). The maximum NIPI value (3.61) belonged to soil samples from urban areas and waste disposal/treatment sites (26). The average and range of the potential ecological risk, including for soils of agricultural locations, calculated as the sum of the mean risk factors of the metals, was 58.48 and 22.47 to 103.31, respectively indicating an overall low ecological risk posed by heavy metals. In the study of Jiang et al., the potential ecological risk in the order of Cd > Pb > Cu > Cr > Zn was obtained, which showed that Cd was the most important factor leading to risk (27).
was investigated and 50 surface samples (5 to 15 cm) were taken from a wide range of exposed either bare soil or vegetated locations. Topsoil samples collected from urban areas (L1, L4, and L2) had high As, Cd, Pb, and Zn concentrations coupled with high coefficients of variation, indicating the dominance of anthropogenic sources, where Co, Cr, and Ni exhibited generally low coefficients of variation and quite homogenous distributions across the study area, suggesting a major natural origin. Mean content of As, Pb, Cd, Zn, Cr, Ni, and Co in soil samples of different locations were 6.9 ± 2.60, 31 ± 5.70, 0.32 ± 0.10, 82.80 ± 15, 32.40 ± 8.30, 34.50 ± 7.50, and 22.6 ± 6 mg.kg⁻¹ respectively, and were larger than their background levels. Application of the index of contamination factor (Cᵢ), degree of contamination (C₀), Nemerow integrated potential index (NIPI), and the potential ecological risk (ER) clearly indicates that all of the soil samples were categorized in low to moderate degree of contamination and the potential ecological risk by heavy metals. The contribution order of ecological risk of heavy metals was as follows, Cd (51.91%) > As (23.75%) > Pb (7.61%) > Cr (5.57%) > Co (5.24%) > Zn (3.04%) > Ni (2.88%).

Acknowledgments

The authors gratefully acknowledge food health and chemistry laboratory personnel of Guilan University of Medical Sciences for carrying out the heavy metals analysis.

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