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ORIGINAL ARTICLE

Investigation and Identification of Types and Amounts of Heavy Metals in Soil of an Industrial Area

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KEYWORDS

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Sampling

Soil contamination

Standard

ABSTRACT: This study was mainly designed to investigate and identify the amounts and types of heavy metals in the soils of National Iranian Oil Refining & Distribution Company in Shahrood region and tried to establish a logical relation between the presence of heavy metals and their damage on vegetation. In addition, considering the power of drained soil and due to the proximity of ground water in Shahrood region, conducting this study provides a better insight into recognition of the possible contamination centers of drinking waters. The gridding and selective method was used for sampling step. Accordingly, five sub-samples were taken from each grid and finally after mixing all of the sub-samples, the final samples were obtained with an average weight of 400 grams prior to sending to the laboratory. To determine the total concentration of heavy metals in soil, extraction was done using concentrated solutions of HCl and HNO_3. The total concentration of the heavy metals of chromium, cobalt and nickel were measured using an ICP-MS instrument, and the rest of the elements using an XRF device. The results explicitly indicated that the quantities of nickel, lead, zinc and strontium in patient samples exceeded the standard, and the other elements were lower than their standard limits. More specifically, the contents of lead and strontium in both normal and patient samples were higher than their standard contents. Moreover, the majority of the vegetation loss across the affected areas was caused by heavy metal accumulation, particularly nickel, lead, zinc, and strontium.

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INTRODUCTION

Environmental pollution, including soil contamination, is one of the important factors influencing the harmony of the nature [1-9]. Soil along with air and water are considered as the major components of the environment [5, 6 and 9]. Soil not only houses the xenophile, but also serves as a very unique environment for various living organisms, especially plants [10]. Nowadays, increase in human activities on the earth has led to the impaired functioning of the soil which is one of the main parts of the earth's crust [11]. Soil pollution can generally be divided into two categories. The first one consists of different pesticides that are used for agricultural purposes while the second and more important category involves industrial pollutants, including heavy metals like lead, nickel, zinc and cadmium. Sometimes, milligrams per kilogram of these elements endanger the soil health, and ultimately human beings and other creatures [12]. Heavy metals are known as metals which have relatively a high density and are toxic at low concentrations [13]. Contamination of heavy metals not only affects the physical and chemical features of the soil, but also decreases the biologic activities as well as the access of nutrition in the soil, directly. More specifically, it is considered as a big danger to human beings through entering to the food chain and to the environmental safety by penetrating into underground waters [14].

Soil is called non-contaminated when the concentration of different elements in it, especially heavy elements, is less than or maximum equal to that of world standards introduced for soil – a concentration called background concentration [6, 7]. Researchers classify soils according to their heavy metals contamination under three global groups [15]. The first group includes soils with low pollution that usually includes non-polluted agricultural soils and areas, which are away from cities. Group II covers soils with low to moderate pollution where soil should not be used for farming. Due to low to moderate levels of contamination, the best way to clean this type of soils is phytoremediation. Finally, group III are soils existing around industrial areas, industrial and domestic sewage and sludge disposal sites and roads with heavy traffic.

The fate of heavy metals and metal complexes discharged into soils and water varies greatly depending on environmental conditions. There are so many factors influencing the uptake of metals [16]. In this regard, parameters like colloid type and amount, soil pH, ion concentration, metal cation concentration, competitive metal cations, and organic and inorganic ligands play a key role [17]. Heavy metals can cause cancer malignant tumors; some of these elements do not have contamination threshold and for any given concentration, although very small, are harmful that in long term will lead to adverse effects [16, 18-20]. Lead (Pb), cadmium (Cd), and mercury (Hg) are heavy metals which do not have contamination threshold [21].

Biochemically speaking, the mechanism of toxic effect of heavy metals is induced because of the extreme tendency of their cations to react with sulfur. Heavy metal cations enter the body through swallowing molecules containing these metals. They easily attach to sulfuhydrils (-SH) found abundantly in the human body. The resulting metal-sulfur combination usually affects enzymes which control the rate of the important metabolic reactions in the human body, so these enzymes cannot do their usual function, leading to human health loss and even death [22].

Despite the importance of heavy metals, a vast and exhaustive study on the quantity and distribution of these elements in the soil of the country has not been done up to present. However, some sparse researches can be pointed out in the literature. Amini et al. provided the map of cadmium and lead contamination in the soils of Isfahan regions [23]. Furthermore, Khosravi et al. studied the distribution pattern of heavy metals in urban, industrial and agricultural soils of Isfahan city [24]. Delijani et al. analyzed the enrichment and distribution of heavy metals in soils of Pars Special Economic Energy Zone [25]. In addition, Baghaie et al. studied the resulting contamination of lead and nickel from two foundries in Isfahan region [26].

This study was conducted to investigate and identify the amount and types of heavy metals in National Iranian Oil Refining & Distribution Company, Shahrood region, Semnan Province, Iran. Some of the plantation in the study area suffers from problems in their leaves and aerial components most probably due to the existence of some toxic elements. Therefore, this study aimed at recognizing the relation between the presence of heavy elements and their possible damages on plant covering. Furthermore, because of the drainage capability of the soil in the area to the adjacency of the underground water, conducting this study would help have a better recognition of the prospective contamination resources regarding the city drinkable water.

MATERIALS AND METHODS

Study area

The study area, National Iranian Oil Refining & Distribution Company in Shahrood region, the storage and distribution facility of petroleum products (oil storage) of Shahrood, one of the oldest oil storages in Iran, is located in an area of 55,000 square meters. It comprises 15 fuel tanks, 6 gasoline tanks, 6 oil gas tanks and 3 tanks of kerosene involving 57.7, 21, 25 and 2.6 million liters respectively. At the Fifteenth National Congress on the Green Industrial Units and Services, the company was ranked among the four green service units in Iran. Figure 1 shows the location map of the study area.

Figure 1. Location of study sites in the Shahrood area

Research methodology

This research is of applied type. Its data collection was based on the literature review and complimentary lab experimentations. Finally, to make conclusions, the outcome was compared with authentic and reliable standards. In order to have a better study on pollutants, two types of samples were prepared: The first one from the regions where there were no sick plant species and the second from where sick and damaged vegetation had been observed. The soil test consists of three main steps:

- Sample collection;
- Sample analysis;
- Interpretation of results.

The sampling devices used in this study consisted of a bucket, a shovel or lift, plastic bags, a tape measure, brushes, and cloth or sacks for mixing the gathered soil. As mentioned earlier, the sampling method included grid and selective sampling. Number of samples and sampling method in both cases (patients and controls) were similar. In the next step, five sub-samples were taken from each grid (Figure 2). Finally, after mixing all of the sub-samples, the final samples, weighing 400 g involving separately patients and controls, were sent to the laboratory. The samples were dug from 25-30 cm deep. At first, 5 cm of topsoil was pushed aside. In addition, in accordance with sampling standards, sampling was carried out under the canopy of the trees. The soil samples were transferred to a trusted lab (Jahad Building Materials and Minerals Laboratory, Semnan Province, Iran). At first, the soil samples were dried in an Edwards lyophilizer at 50 ˚ C and 10^{-1} mmHg pressure, grounded with a porcelain pestle and mortar with the help of a pistil, both in porcelain, immediately after being dried and sieved in a 2-mm nylon sieve. Then, the digestion and extraction process of the heavy metals were performed using concentrated solutions of HCl and HNO_3. In the next step, these sub-samples obtained were stored in closed plastic flasks, closed with plastic film and stored at 4 ˚ C until chemical analyses [27, 28]. The weighing of the sub-samples was anticipated to avoid error in the determination of the dry sediment mass due to humidity absorption.

Finally, the contents of the heavy metals present in each sample were determined [29]. Accordingly, the total concentrations of chromium, cobalt and nickel were measured with an ICP-MS (HP, QUADIAPOLEDUTECTORMS, Italy), while other elements were quantified with an XRF device (Philips RT1123). The Limit of detection (LOD) of XRF for the direct determination of common heavy metals occurring in soils samples is as ppm.

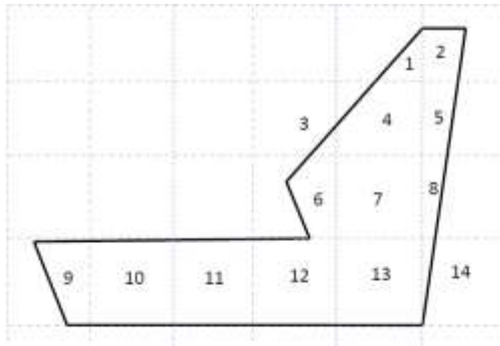

Figure 2. Gridding the region to sample

RESULTS

The results of the measurement of the common heavy metals encountered in regions with sick soil-plant samples are presented in Table 1.

Table 1. Results of soil analysis, sample No. 1: patient sample

(ppm) Co	(ppm) As	(ppm) Cl	(ppm) Ba	(ppm) Sr	(ppm) Cu	(ppm) Zn	(ppm) Pb	(ppm) Ni	(ppm) U	(ppm) Cr
2	1	405	198	455	22	98	39	59	1	49
(ppm) Th	**(ppm) Mo**	**(ppm) Ga**	**(ppm) Nb**	**(ppm) V**	**(ppm) Ce**	**(ppm) La**	**(ppm) W**	**(ppm) Zr**	**(ppm) Y**	**(ppm) Rb**
1	1	12	1	73	9	2	1	1	25	52

Furthermore, the results of the assessment of heavy metals in the soils of some regions containing no sick plant samples are presented in Table 2.

Table 2. Results of soil analysis, sample No. 2: normal sample

Element	Ag (ppm)	Al (ppm)	As (ppm)	Ba (ppm)	Be (ppm)	Ca (ppm)	Cd (ppm)	Ce (ppm)	Co (ppm)
DL[a]	0.1	100	0.5	5	0.2	100	0.1	1	1
Sample No. 2	0.21	34758	4.7	257	1.1	114144	0.26	38	10
Element	**Cr (ppm)**	**Cu (ppm)**	**Fe (ppm)**	**K (ppm)**	**La (ppm)**	**Li (ppm)**	**Mg (ppm)**	**Mn (ppm)**	**Mo (ppm)**
DL	1	1	100	100	1	1	100	5	0.5
Sample No. 2	92	18	22993	10944	20	25	8758	477	1.23
Element	**Na (ppm)**	**Ni (ppm)**	**P (ppm)**	**Pb (ppm)**	**S (ppm)**	**Sb (ppm)**	**Sc (ppm)**	**Sr (ppm)**	**Th (ppm)**
DL	100	1	10	1	50	0.5	0.5	2	0.5
Sample No. 2	129	30	614	24	631	1.06	6.5	308	*9*

[a] : Detection limit

Figures 3 and 4 represent the heavy metals measured in patients and normal samples under the identical conditions,respectively.

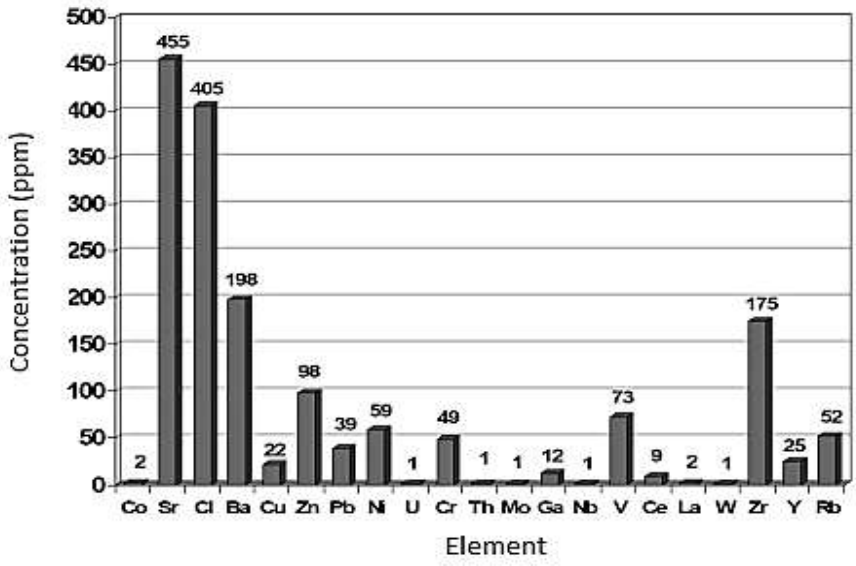

Figure 3. Heavy metals in patient samples (ppm)

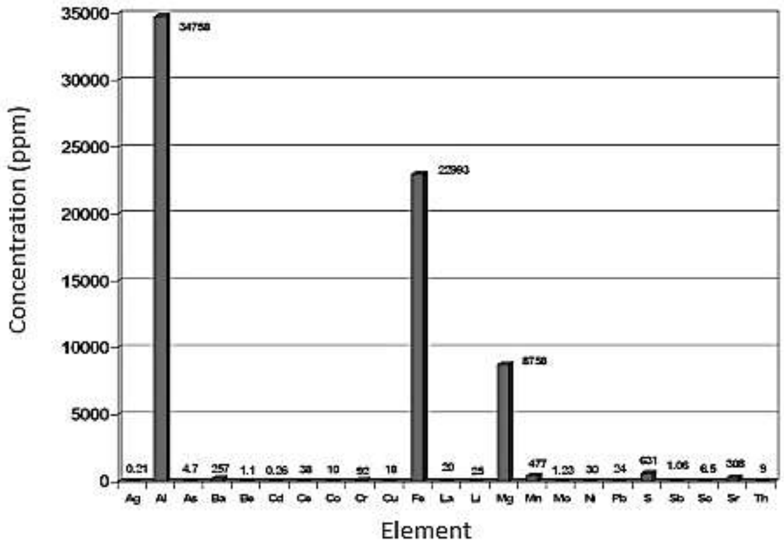

Figure 4. Heavy metals in normal samples (ppm)

DISCUSSION

Since, concerning soil and limits of polluting elements, there is not a certain standard or reference in Iran, some standards from globally reliable sources and previously published papers in the literature are presented (Table 3) and afterwards the status quo is compared with them (Table 4).

Table 3. Heavy metals limits and standards in soil

Element	Standard							
	Extent that is necessary to improve soil	(mg/kg) Permitted limit to human health and the environment	USEPA [a] Appropriate limit of soil	Global average (ppm)	Holmgeren et al, 1993 [31] (mg/kg)	Boemgen & Shackette, 1981 [30] (mg/kg)	GLC (ppm)	USEPA (ppm)
Mn	-	-	-	850	-	-	600	600
Cd	20	5	1	0.3	0.294	N/A	-	0.06
Fe	-	-	-	23000			NR	NR
Ni	500	100	50	33.7	27.4	16	20	4
Cr	800	250	100	80	-	-	10	100
Co	300	50	20	19	-	-	NR	8
Pb	600	150	50	20	8.6	16	10	10
Cu	500	100	50	45	-		NR	30
As	50	30	20	-	N/A	6.4	-	5
Zr	-	-	-	-	-	-	-	300
V	-	-	-	108	-	-	-	100
Sr	-	-	-	208	-	-	-	200
Zn	3000	500	200	59.8	-	-	-	50
Ba	2000	400	200	568	-	-	-	430

Continue of Table 3

La	-	-	-	41.2	-	-	-	30
Mo	200	40	10	40787	-	-	-	2
Hg	10	2	0.	-	N/A	0.11	-	-
Sn	300	50	20	-	-	-	-	-

[a] United States Environmental Protection Agency

Table 4. Evaluation of prevalent element values in soil

Element	Permitted levels of health (mg/Kg)			Permitted levels of ecological (mg/Kg)
	Commercial and industrial uses	Parks, open areas, areas for playing	Urban and garden uses	
Metals/Metalloids				
Antimony, Sb	820	-	30	20
Arsenic, As	500	200	100	20
Barium, Ba	100000	-	5370	400
Beryllium, Be	100	40	20	-
Cadmium, Cd	100	40	20	3
Chromium (III)	60%	24%	12%	-
Chromium (VI)	500	200	100	-
Chromium (Total), Cr	-	-	210	50
Cobalt, Co	500	200	100	50
Copper, Cu	5000	2000	1000	60
Lead, Pb	1500	600	300	300
Manganese, Mn	7500	3000	1500	500
Methyl mercury	50	20	10	-
Mercury, Hg	75	30	15	1
Molybdenum, Mo	10220	-	390	40
Nickel, Ni	3000	600	600	60
Tin, Sn	100000	-	46900	50
Zinc, Zn	35000	14000	700	200

Source: USEPA, 2005 [32]; WHO 1998 [16]

As seen in Figure 5, the rate of the four elements Ni, Pb, Zn, and Sr in sick samples exceeded the standards, while the other elements are lower than those of standards are. In addition, regarding the normal samples (Figure 6), it can be said that Sr, Pb, Co, and Cd concentrations are more than their standard limits. It was also observed that the amounts of Pb and Sr were much higher than the standard levels in both normal and sick samples.

Figure 7 shows the measured lead in the soil from study area. Furthermore, in this figure, a comparison was made with the international standards available from some authentic studies like environmental protection agency (EPA)[5-9, 16, 18-20, 33 and 34].

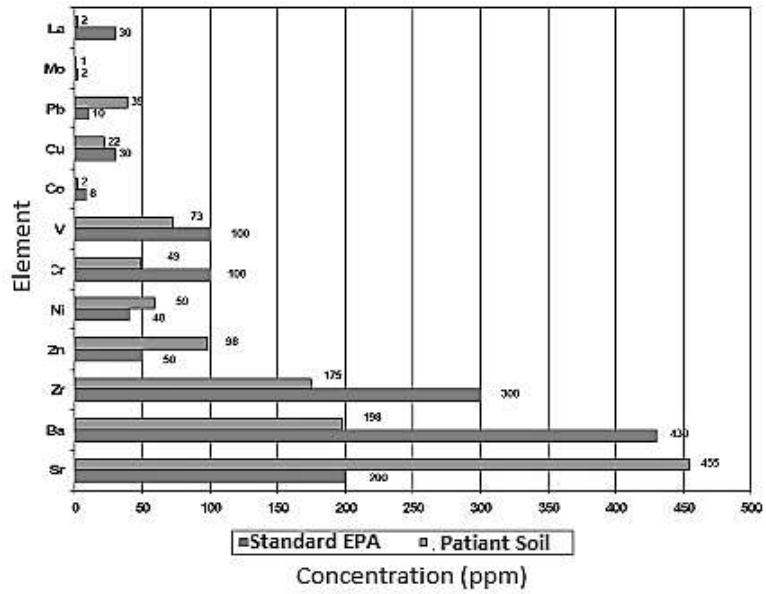

Figure 5. Comparison between the elements in patient samples and the standard (ppm)

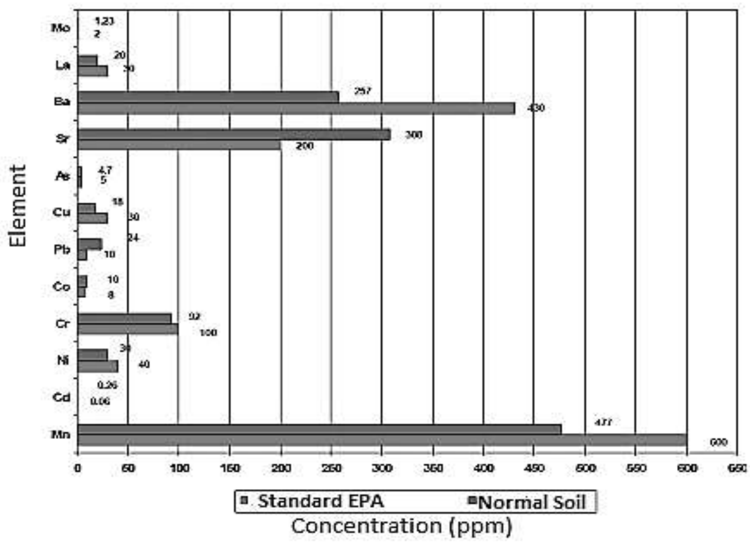

Figure 6. Comparison chart between the elements in the normal sample and the standard (ppm)

The obtained results indicate that much of the vegetation destruction in sick soil occurs because of the heavy metal accumulation, especially Ni, Pb, Zn, and Sr. In addition, the excessive quantities of Cd in those parts of the study area where no vegetation damage has been reported, does not necessarily mean that certain environmental and health problems have not happened.

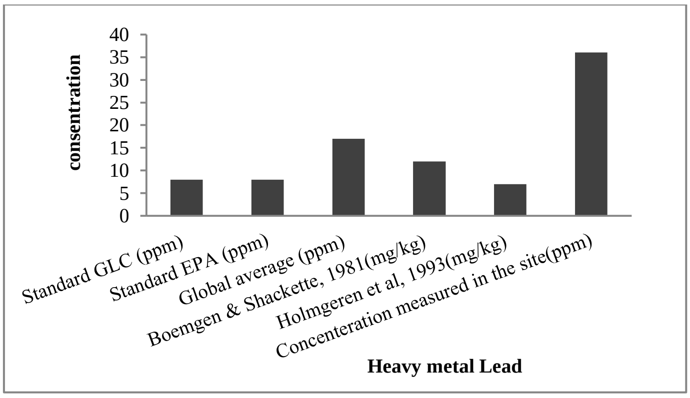

Figure 7. Comparison between the highest lead levels measured, standards, and previous reports

By studying the zoning of the vegetation damage (Figure 8), the dispersion of heavy metals in the soil of the site (Figure 9), and the overlap between zones, it can be concluded that the damage inflicted on the site vegetation (trees and shrubs) directly correlates with the type and amount of heavy metals in the soil of the region.

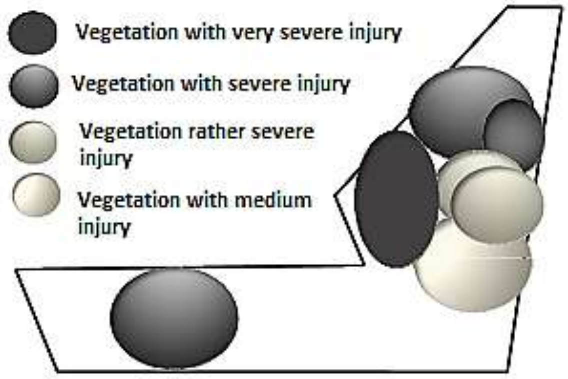

Figure 8. Vegetation damage zoning of the study site

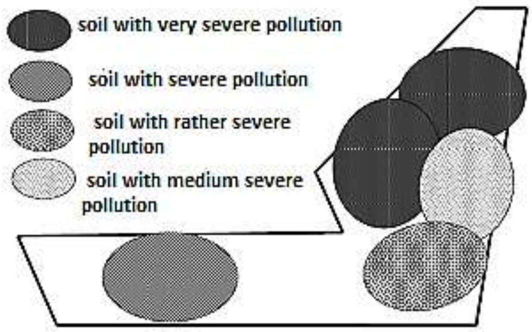

Figure 9. Distribution of heavy metals in the study site

Cd disturbs the relative distribution of zinc in the body [35]. In case of its acute toxicity, some symptoms such as nausea, diarrhea, severe headaches, muscle and abdominal pain, increased salivation, shock, liver damages, kidney failure occur [36, 37]. According to Food and Agriculture Organization of the United Nations (FAO), the allowable amount of Cd input to the body for each person lays over the range 0.4–0.6 mg, weekly [38]. Furthermore, Pb is a dangerous heavy metal entering the environment by human beings in different ways [33]. The half-life of Pb in blood, soft tissues, and bone is about 2-4 weeks, 4 weeks, and 27.5 years, respectively. It is known as a metabolic poison since long [39]. Some symptoms of lead poisoning include severe fatigue, lethargy, mild abdominal discomfort, and anemia. FAO and WHO expert committees have pointed out that the weekly transient absorption of lead for each person is about 4 mg [16].

Ni toxicity in humans appears when 250 mg of dissolved nickel enters the body [29]. Nickel is considered as a carcinogen, and its maximum allowable concentration in the water comes to 0.05 mg per liter. Maximum allowable daily absorption of this material through food is expressed as 5 mg per kg of the body weight per day [40].

Because of chemical reactions, insoluble compounds of Sr in water can become much more soluble. Soluble compounds in water are more harmful to human health than the insoluble ones owning to their easier uptake by the leaving tissues. Therefore, Sr compounds serve as serious contaminators of drinking water [34, 41]. Fortunately, Sr concentration in drinking water is very low. It is evident that breathing air or dust, eating food, drinking water, or contact with contaminated soil to strontium let a small amount of strontium enter the human's body. Strontium entrance into the body is likelier by eating than drinking. Sr in food adds to the strontium in the body. Grains, leafy vegetables and dairy products have high amounts of strontium. The amount of strontium in most people's body is about its average acceptable value. Among the strontium compounds, it is only strontium chromate that even in small quantities is harmful for human health. Absorption of strontium in children can cause growth disorders. Strontium salts do not cause skin rashes or other skin problems. When strontium absorption is extremely high, bone growth will be impaired. However, this problem arises only when the absorption of strontium is more than thousands of ppm [38, 40].

CONCLUSIONS

The currents work aims at measuring of the contents of the heavy metals present in two types of the soil samples, namely patient and normal samples. These samples were collected from the National Iranian Oil Refining & Distribution Company in Shahrood region and subsequently their elemental compositions were compared with each other as a case study. After preparation of the samples and performing some preliminary steps, their analyses revealed that amounts of the metals involving nickel, lead, zinc and strontium in patient samples exceeded the standard levels while the other elements were lower than their standard limits. It was also observed that the contents of lead and strontium in both normal and patient samples were higher than their standard contents. In view of the obtained results, the majority of the vegetation loss across affecting the study areas can be attributed to the heavy metal accumulation, particularly nickel, lead, zinc, and strontium.

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