

Article

Grain size fraction of heavy metals in soil and their relationship with land use

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Abstract

The aim of the present study was to investigate the distribution of heavy metals (Pb, Ni, Cr and Cd) in different grain-size fractions of the surface soils. The soil samples of different land uses were taken from 20cm depth at 12 stations where is located at Amir Abad of Birjand city, Iran. The air-dried samples were passed through sieves of different sizes to collect the fractions lesser than 63 μm , 63-125 μm , 125-250 μm , 250-500 μm , 500-1000 μm and 1000-2000 μm . The samples were digested by perchloric acid and nitric acid; and the concentrations of heavy metals were determined using Atomic Absorption Spectrophotometer (model AAcontr700). The highest concentrations for Pb, Cr, Ni and Cd (139.5, 195.4, 98.4 and 3.15 mg/kg respectively) were obtained in the fractions less than 63 μm from the road-side soils. The contamination factor values for the fractions less than 63 μm were higher than other fractions. The comparison of different toxic metals concentration from various grain-size fractions demonstrated higher Cr levels, especially in the 63-125 μm size fractions from the road-side lands. Thus, the comparison of grain size factors from different soils revealed that Cr exhibited highest values in relation to the other toxic elements studied.

Keywords contamination factor; natural soil; pollution load index; residential soil; road-side soil.

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1 Introduction

Soil signifying living bed has more importance as compared to air and water and hence soil protection gains paramount importance as an environmental political duty. Although heavy metals are natural components of the Earth's crust, soils are the major sinks for heavy metals in the terrestrial environment (Ghaleno et al., 2015; Sayadi et al., 2015a). The problem of heavy metal pollution in soils has been focused in the past few years due to the human actions that have severely changed the balance between biochemical and geochemical cycles of some heavy metals (Kumar et al., 2008). Because of the potentially harmful nature and long residence time of heavy metals in soils to human health, an estimation of the environmental risk arising out of soil pollution in

agricultural and non-agricultural areas are of special importance as they may enter the food chain in significantly high quantities (Patel et al., 2006). In the municipal environments also the soils are deeply contaminated by heavy metals (Miguel et al., 2007) posing challenging and difficult situation to the scientific community. The distribution of heavy metals increased with decrease of particle size (Sarлак, 2015) and the mobility, immobility and thus toxicity of heavy metals in soil depend largely on their type of binding forms (Ahmadipour et al., 2014). As toxic heavy metals can bio-accumulate in plants and animals it is essential to know the fate of these metallic pollutants and therefore frequent case studies have been undertaken in developed countries to assess the intensity of heavy metal pollution in soils. However, in developing countries this type of realization is seen only in recent times leading not only in the investigation of heavy metals in soil, but also assessment of complete environmental quality of soils (Sarлак, 2015; Zojaji et al., 2014; Zojaji et al., 2015; Shabani and Sayadi, 2012). Numerous studies have considered on heavy metal concentrations in soils all around the world. In order to assess the environmental risk, it is important to know in which scale particles the heavy metals are preferred to distribute. However, studies of heavy metal in soils are rare in Iran (Rafiei et al., 2010). The present study was carried out as a preliminary survey on soil contamination of Amir Abad area. There are only a few studies on level of soil pollution in this area (Sayadi and Rezaei, 2014; Sayadi et al., 2015b). The main aim of this study was to identify and analyze the status of toxic metals in the soils and influence of grain-size on their distribution in the areas of different land-use by employing different geochemical indices.

2 Study area and Methodology

2.1 Study area

Amir Abad is a relatively small city (village) in the Birjand county having a population of 1000 individuals which is located 5 km towards west of Birjand city on the Birjand-Kerman road in the southern Khorasan province. This area enjoys a special status due to agricultural talent involved in various agricultural activities and multiple animal husbandries. At the same time urban activities, small repair centres, workshops and one of the main highways of South Khorasan province adjoining this area are the potential pollutant resources threatening the natural environment. Similarly, human induced pollutants from an industrial township have considerable influence on the natural environment of this area (Fig. 1). Therefore, in the present study three types of land-use patterns viz. residential, natural and road-side areas have been selected for characterizing toxic heavy metal concentrations and assessment.

2.2 Soil sampling and analysis of samples

Soils from three different land-use patterns were sampled at 20 cm depths in 12 stations. From each station 5 samples were collected with three replications. After the integration of 5 samples from each station a single composite sample of 1.5 kg was prepared. The geographical locations of each sampling station were determined using Global Positioning System, the details of which are tabulated in Table 1.

The collected samples after drying in an open air were passed through sieves of different sizes and the fractions of less than 63 μm , 63-125 μm , 125-250 μm , 250-500 μm , 500-1000 μm and 1000-2000 μm was collected (Sutherland et al., 2012). Later the samples were digested by per chloric acid and nitric acid at 120°C (Burt, 2004). Atomic Absorption Spectrophotometer (model AA contr 700) was used to determine heavy metal concentrations.

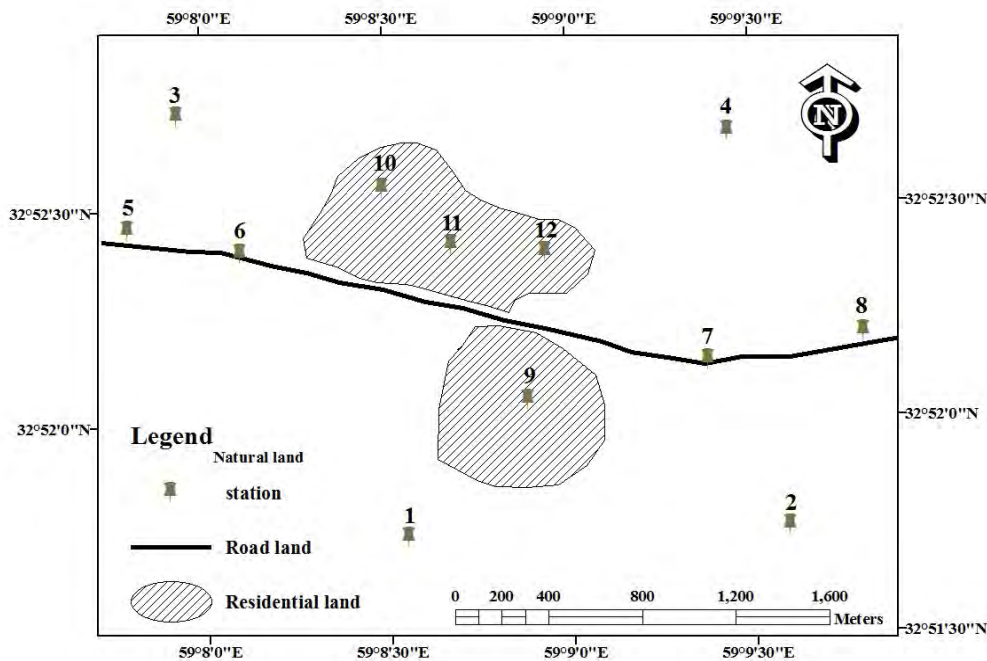


Fig. 1 Sampling map of the study area.

Table 1 The geographical co-ordinate and users of sampling stations.

Station	Land-use	Latitude	Longitude	Station	Land-use	Latitude	Longitude
1	Natural	32.86oN	59.14oE	7	Road	32.87oN	59.15oE
2	Natural	32.86oN	59.15oE	8	Road	32.87oN	59.14oE
3	Natural	32.86oN	59.16oE	9	Residential	32.87oN	59.15oE
4	Natural	32.87oN	59.14oE	10	Residential	32.87oN	59.16oE
5	Road	32.87oN	59.14oE	11	Residential	32.88oN	59.14oE
6	Road	32.86oN	59.16oE	12	Residential	32.87oN	59.14oE

2.3 Indices

The collected geochemical data were processed via Contamination Factor (CF), Pollution Load Index (PLI), and Grain Size Factor (GSF) to characterize and assess the concentrations of toxic heavy metals in different grain-size fractions of the soils. The CF is the percent concentration of a specified contaminant element in a sediment or soil relative to the background concentration of the same contaminant element into the natural environment. This factor can be calculated using the following formula given by Hakanson (1980).

$$CF = C_{\text{heavy metal}} / C_{\text{background}}$$

It is required to mention, that the background concentration of heavy metals are calculated by Sayedi and Rezaei (2014).

The PLI was calculated using the formula given by Tomlinson et al. (1980)

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n}$$

The Grain Size Factor was obtained using the following formula (Zhu et al. 2008; Sutherland 2003)

$$GSF_{Load} = \frac{C_i \times GS_i}{\sum_{i=1}^m C_i \times GS_i}$$

In this formula, C_i is the heavy metal concentration in a specific grain-size fraction and GS_i is the weight percent of existing grains in that grain-size fraction. This factor provides a percentage heavy metal load estimate in each of the size fractions, and the sum of all GSF loads for a given sample and element amounts to 100%.

3 Results and Discussion

3.1 Concentrations of heavy metals

The heavy metals concentrations in different grain-size fractions of the soils collected from different land-use patterns i.e. residential, road-side and natural lands are shown in Table 2. Geochemical data revealed that Pb had the highest concentration (139.5 mg/kg) in the size fraction less than 63 μ m followed by 63–125 μ m fraction (138.7mg/kg) of the soils adjacent to the road. At the same time, the lowest Pb concentration was recorded in 1000-2000 μ m fractions of soils from the natural lands. The highest concentrations of Cr (195.4 mg/kg), Ni (98.4 mg/kg) and Cd (3.15 mg/kg) were observed in the grain sizes less than 63 μ m from the lands surrounding road. Cr showed lowest concentration in 500-1000 μ m fraction of the soils from natural lands (18.2 mg/kg) while Cd showed minimum values in the 250-500 μ m fractions of soils from residential areas (0.19 mg/kg) and Ni in 500-1000 μ m fractions of soils from natural lands (10.9 mg/kg). In an essence, the heavy metal contents in the soils showed quite high values in the road-side lands (which can be attributed to the entry of contaminants from the vehicular traffic) followed by soils in residential areas. Surprisingly, in natural lands, their values were lowest attributing to the least interference of humans. The comparison of heavy metal contents in different size fractions revealed highest concentrations in the fractions of less than 63 μ m in all the three land-use patterns which can be ascribed to the larger surface area of soil particles which in-effect render higher metal adsorbance. The present observations are in concurrence with the previous findings (Seshan et al., 2010; Zhao et al., 2010).

Lowest Pb concentrations were found in the 1000-2000 μ m soil fractions from road-side (value) and natural (value) while in residential areas the lowest concentrations (value) were in 500-1000 μ m fractions. However, Cr and Ni showed lowest concentrations in the 500-1000 μ m fractions of soils from natural and road-side lands and 1000-2000 μ m fractions of the soils from residential areas. Likewise, Cd showed lowest concentrations in 1000-2000 μ m fractions of the soils from the road-side and natural lands and 250-500 μ m fractions of the soils from residential areas. The road-side lands demonstrated highest concentrations of Cr and Pb and lowest abundance of Cd. In case of the soils from residential and natural areas, the abundances of heavy metals were as follows Cr>Ni>Pb>Cd.

3.2 Mass loading and leach ability

In order to estimate the fractions contribution to the total contents of trace metals in soil, mass loading was calculated (Fig. 2). The element percentages of natural land clay size particle <63 μ m contributed the total contents of 23% for Pb, 21% for Cr, 26% for Ni and 35% for Cd respectively. The element percentages of road-side land clay size particle <63 μ m were 17% for Pb, 24% for Cr, 23% for Ni and 28% for Cd respectively. The element percentages of residential land clay size particle < 63 were 18% for Pb, 23% for Cr, 25% for Ni and 34% for Cd respectively. It should be noted that the clay size fraction especially <63 were easily transferred and could induce further contamination (Lijung et al., 2006).

Table 2 Mean \pm SD of heavy metal concentration (mg/kg) in different grain-size fractions.

Heavy metals	Pb (mg/kg)			Cr (mg/kg)			Ni (mg/kg)			Cd (mg/kg)		
	NA	RD	RS	NA	RD	RS	NA	RD	RS	NA	RD	RS
bulk	8.6 \pm 3.7	124.3 \pm 86	34.4 \pm 18	16.8 \pm 9.4	131.2 \pm 114	36.6 \pm 23	13.2 \pm 8.3	69 \pm 41	23.2 \pm 16	0.6 \pm 0.04	1.8 \pm 0.94	0.7 \pm 0.09
< 63 μ m	16.7 \pm 4.2	139.5 \pm 102	45.3 \pm 31	27.7 \pm 15.8	195.4 \pm 132	57.6 \pm 27	26.6 \pm 11	98.4 \pm 47	47.6 \pm 26	1.15 \pm 0.85	3.15 \pm 1.86	1.66 \pm 1.02
63-125 μ m	16.3 \pm 3.7	138.7 \pm 98	42.8 \pm 27	23.9 \pm 14.5	168.3 \pm 127	39.4 \pm 19	24.1 \pm 9.8	90.7 \pm 51	32.2 \pm 21	0.58 \pm 0.33	2.84 \pm 1.77	1.17 \pm 0.87
125-250 μ m	14.7 \pm 3.1	138.1 \pm 111	43.1 \pm 23	24.4 \pm 15.4	189.1 \pm 141	48.2 \pm 28	20.7 \pm 8.3	85.8 \pm 38	29.9 \pm 18	0.46 \pm 0.29	2.25 \pm 1.43	0.84 \pm 0.34
250-500 μ m	13.6 \pm 2.8	137.7 \pm 106	37.4 \pm 19	20.6 \pm 13.6	101.5 \pm 83	46 \pm 23	14.3 \pm 4.9	60.5 \pm 25	22.1 \pm 13	0.38 \pm 0.17	2.16 \pm 1.06	0.19 \pm 0.11
500-1000 μ m	13.5 \pm 3.2	132.6 \pm 89	36.9 \pm 21	18.2 \pm 13.9	77.8 \pm 37	32.4 \pm 16	10.9 \pm 3.7	54.2 \pm 23	21.7 \pm 15	0.42 \pm 0.19	0.27 \pm 0.20	0.23 \pm 0.17
1000-2000 μ m	12.9 \pm 2.6	131.4 \pm 107	37 \pm 26	22.1 \pm 15.7	91 \pm 48	31.9 \pm 21	18.6 \pm 6.4	60.3 \pm 37	21.4 \pm 12	0.34 \pm 0.11	0.22 \pm 0.19	0.27 \pm 0.22
Mean	14.62 \pm 7.3	136.3 \pm 112	40.42 \pm 32	22.8 \pm 17	137.2 \pm 98	42.58 \pm 28	19.2 \pm 9.5	74.98 \pm 51	29.15 \pm 16	0.56 \pm 0.13	1.82 \pm 0.77	0.73 \pm 0.36

NA: Natural Land; RD: Road-side Land; RS: Residential Land.

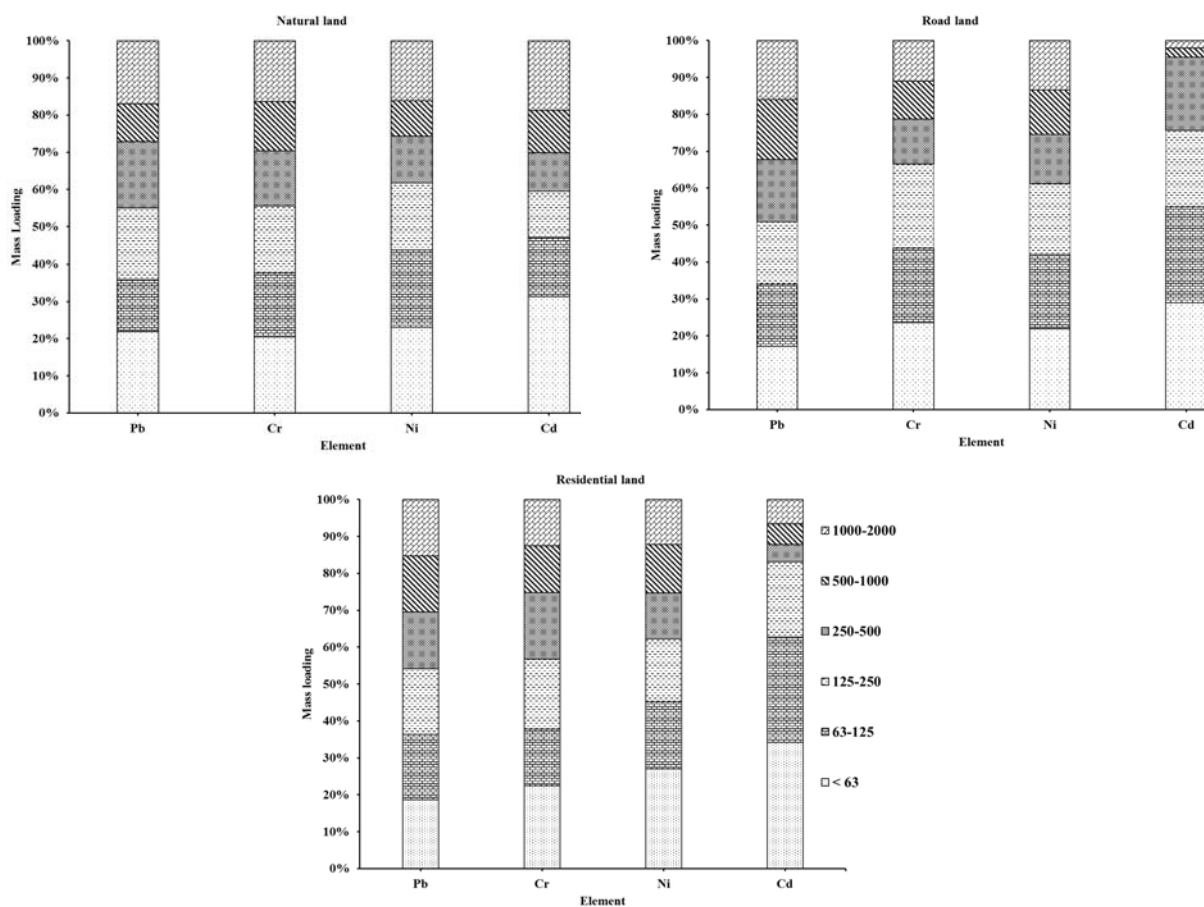


Fig. 2 The heavy metal mass loading (%) of soil particle size fractions at different land use.

In the second mass loading of the natural land fine size particle (< 250 μm) the total contents were: 32 % for Pb, 36% for Cr, 38% for Ni and 27% for Cd respectively. The second mass loading of road land fine size particle (< 250 μm) was: 35% for Pb, 46% for Cr, 38% for Ni and 47% for Cd respectively. The second mass loading of the residential land fine size particle (< 250 μm) was: 36 % for Pb, 33 % for Cr, 37% for Ni and 49 % for Cd respectively. The third mass loading of natural land coarse size particle (250-1000) was: 29 % for Pb, 29 % for Cr, 21 % for Ni and 20 % for Cd respectively. The third mass loading of road land coarse size particle (250-1000) was: 33 % for Pb, 19 % for Cr, 26 % for Ni and 23 % for Cd respectively. The third mass loading of residential land coarse size particle (250-1000) was: 30 % for Pb, 31 % for Cr, 26 % for Ni and 11 % for Cd respectively.

The lowest loading of Natural land was (16 % for Pb, 14 % for Cr, 15 % for Ni and 18 % for Cd respectively), Road land was (15% for Pb, 11% for Cr, 13% for Ni and 2% for Cd respectively) and Residential land was (16% for Pb, 13% for Cr, 12% Ni and 6% for Cd respectively) resulting from the coarse aggregates (>1000). Undoubtedly, the tendency of heavy metals adsorbed in clay and finer fractions should be taken into account during the environmental risk assessment and soil remediation.

Besides, weathering of rocks and minerals results in Ni entering the natural environment essentially via human activities like used batteries and tanks storing petroleum products (Sayadi and Sayyed, 2011). While Ni can replace important elements from the enzymes leading to the breakage of metabolism route in high concentrations it is also carcinogenic and even leads to heart and liver diseases (Sayadi and Torabi, 2009).

Chromium enters the soil environment by humans via tanning, pharmaceutical and metals industries. Usage of pigments containing chromium, petroleum products, oil and grease also contribute chromium to the natural environment (Kumar et al. 2008). Although chromium is an essential element for fats and protein metabolism, in high concentrations, it harms liver and kidneys and could lead to cancer (Sayadi and Torabi, 2009).

Cadmium enters the natural environment from battery, pigment, jewellery and plating materials producing factories. The plastic, ceramic and glass factories are also other sources of cadmium pollution in a natural environment (Srinivasa and Gowd Govil, 2007). The noteworthy health hazards due to cadmium pollutions are chronic renal and cardiovascular diseases, anaemia, respiratory system disorders, prostate cancer, lung infection, skin, rickets and tooth decay, etc. (Sayadi and Torabi, 2009). Lead in blood is found in three Free states i.e. ionized, connected to red corpuscles and adjoined to proteins. This element can replace calcium in the bones and is capable to create blood, enzyme, bone and nerve disorders and lead to the general weakness, anorexia, muscle relaxation, neurotic disorders, anaemia, insomnia and skin discoloration (Sayadi and Torabi, 2009). Although benzene and automobile manufacturing industries account for nearly half of the global lead consumption, it is also utilized in the production of different alloys, paint, potteries, ceramics, tiles, pesticides, plastics, etc. resulting its access to environment via different routes (Sayadi et al., 2009).

3.3 Contamination factor

The CF comparative values (Fig. 3) indicated highest concentrations of heavy metals in road-side soils while the natural lands demonstrated lowest CF. Considering the mean of the total data, Pb exhibited highest concentration while Ni lowest, while the lowest enrichment level for Ni (CF: 0.22) was observed in 500-1000 μm fraction of natural domains. The average value of the metals in the grain size fraction road-side land was in the order of $\text{Pb} > \text{Cd} > \text{Cr} > \text{Ni}$.

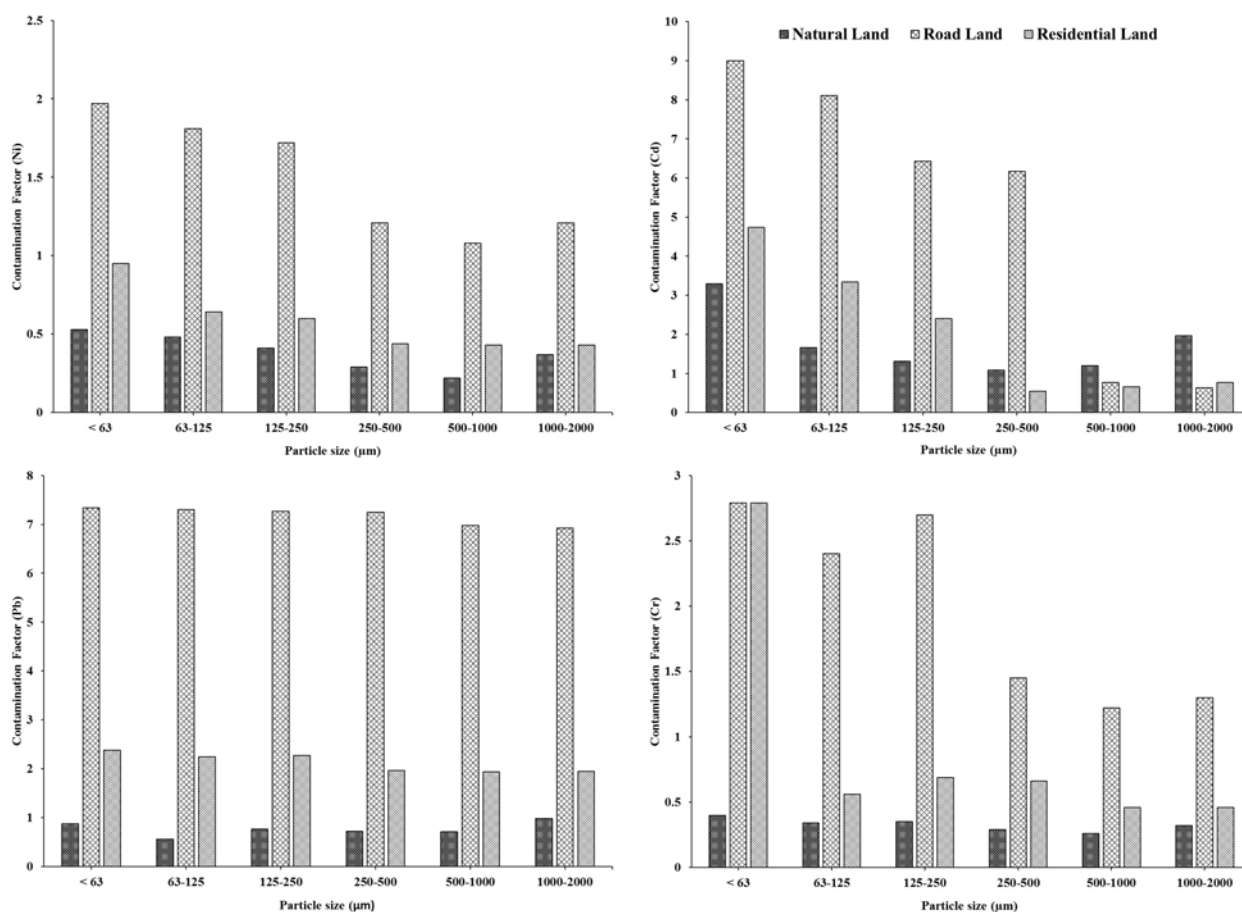


Fig. 3 Contamination Factor for soil particle size fractions at different land use.

The CF values were dissimilar in different size fractions; Pb (CF: 7.34) was found to be highest in less than the 63μm fraction from the road-side. The size fraction below 63μm from the road-side lands showed highest enrichment of Cd (CF: 9.00) and lowest enrichment of Cr (CF: 0.40) in the natural lands. Contamination factor also was higher in the size fraction less than 63μm and barring a few exceptions was the lowest in 500-1000 μm fraction. It was earlier reported that the soil samples of road-side land were considerably contaminated by trace metals Pb, Cr and Cd (Sayadi and Rezaei, 2014). However, in the present research, high contamination factor values were reported in the soil samples of residential land where all the grain sizes were considerably contaminated by Pb where as Cd was demonstrated in the grain size fractions (<63, 63-125 and 125-250), and Cr in the 63μm fraction respectively.

3.4 Pollution Load Index

The numeral value of PLI in the soils more than 1 showed a distinct pollution with the considered metal and value less than 1 signified nil pollution. Pollution load index values (Fig. 4) suggested that the Amir Abad soils are not much polluted with Cr and Ni metals since they demonstrated lower PLI values. As far as Cd is concerned, the soil fractions less than 500 μm were polluted whereas fractions more than 500 μm were devoid of heavy metal pollution. Further, it was noted that the entire size fractions were highly polluted with Pb which could be due to lead present in gasoline that after gasoline burning entered the air and settled in soils, especially the road-side lands causing severe pollution (Sayadi and Sayyed, 2010).

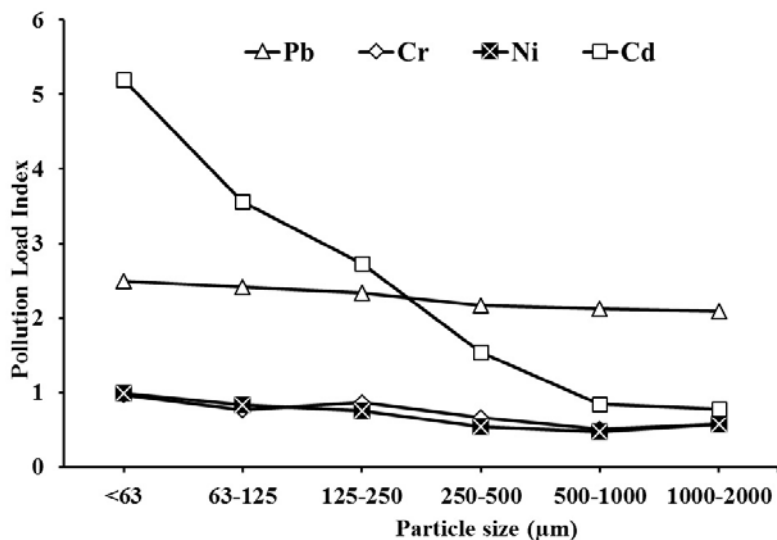


Fig. 4 Pollution load index of the heavy metals at soil particle size fractions.

3.5 Grain size factor

The GSF plays key role in mobility of heavy metals in different sized particles in relation to pollutant levels (Bian and Zhu 2009). GSF values of heavy metals in different grain-size fractions (Fig. 5) from the soils in the present study were compared to attain certain conclusions.

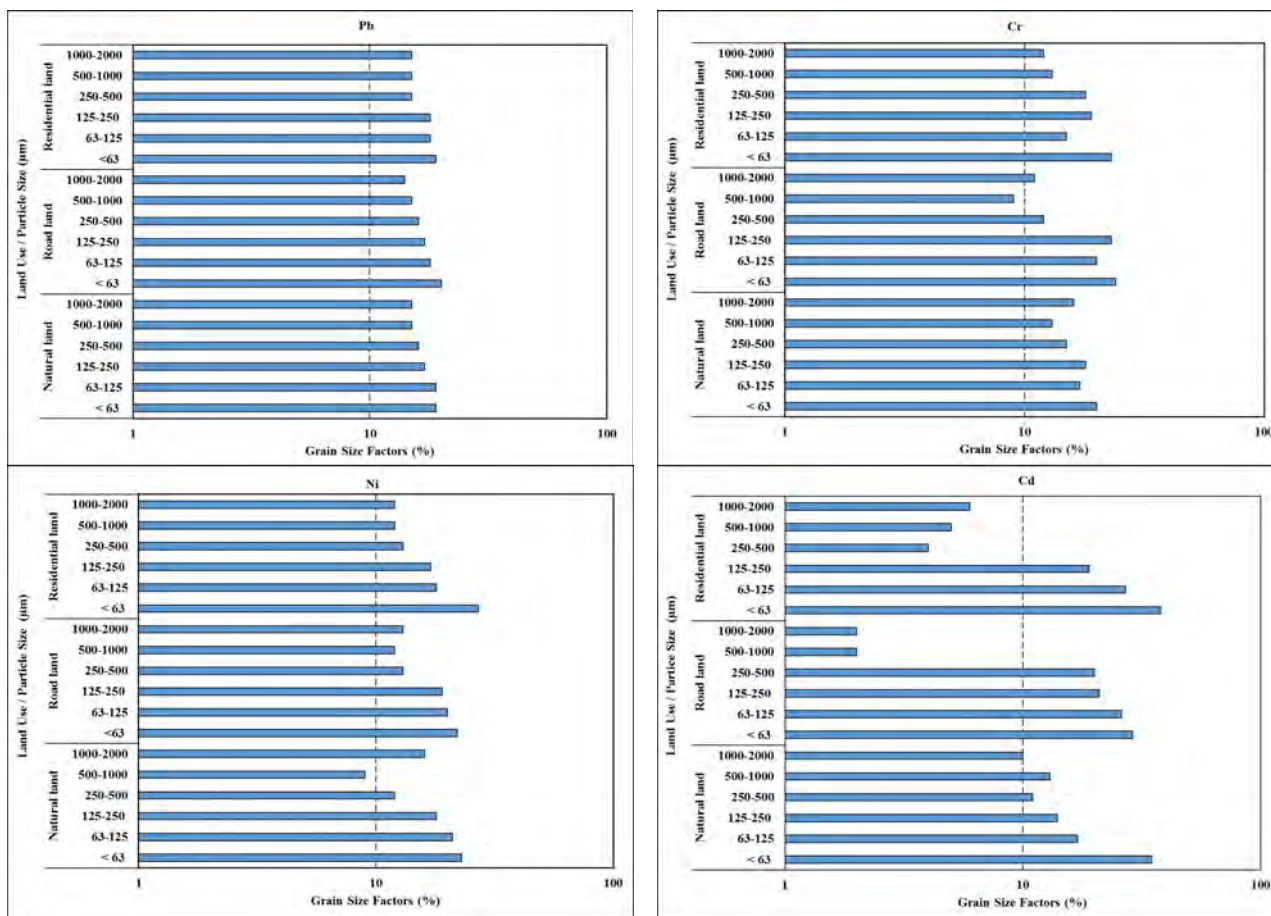


Fig. 5 Grain size factors values of heavy metals at different land use and grain size fractions.

However, the highest GSF value was recorded for Cr in road-side lands and the lowest level for Pb in natural lands. For Pb, Cr and Ni the highest GSF value was recorded in 63-125 μ m fractions from road-side soils while fractions less than 125 μ m from natural land soils revealed the lowest GSF values. Cd showed the highest and lowest values of GSF in 250-500 μ m grains in road-side and residential lands respectively. Nevertheless, with comparison of GSF values in different soil fractions it can be safely stated that Cr showed the maximum value in 63-125 μ m fractions from road-side lands while Pb exhibited minimum value in size fraction of below 125 μ m in soils from natural lands. Zhao and colleagues showed that particles with the smallest grain size (<44 μ m) had the highest metal concentration in most areas (Zhao et al., 2010). Sutherland reported that particles with a grain size <63 μ m were the single most important mass component, with 38% Pb of the total sediment being stored in this fraction (Sutherland, 2003). Sutherland and colleagues reported that particles with a grain size <63 μ m had the highest loading for all metals examined for each of the four geochemical fractions (Sutherland et al., 2012). In the present study, we found that the smallest fraction had the greatest influence on the overall contamination of the soils.

4 Conclusions

The distribution of most heavy metals in different size particles increased with decreasing particle size. The results showed that the contamination levels, distribution characteristics and primary controlling factors were different in the natural land, road land and residential land grains in the roadside soil rotation field. However, the mass loading of adsorbed heavy metals in the finer fractions should be taken into account during the environmental risk assessment and soil remediation. The mean concentrations of Pb, Cr and Cd in soils of road-side land were higher as compared to the equivalent soil from natural land and residential land. The comparison of the concentrations factor indicated highest concentrations of the studied heavy metals in road-side soils, whereas the natural lands exhibited the lowest. The concentrations of trace metals in different grain size classes' demonstrated higher accumulation in clay fractions, and the trace metals, mainly accumulated in the fractions of size < 63 and 63-125. Therefore, it could be safely concluded that the highest GSF value was recorded for Cr in road-side lands and the lowest level for Pb in natural lands.

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