



Assessment of Heavy Metal Contamination in Soils around of Khash Cement Plant, SE Iran

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Abstract

20 soil samples collected from the vicinity of the Khash Cement plant, Iran, were analyzed for As, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb and Zn using ICP-OES. The results revealed that the metal distribution were in a fluctuating manner considering various distances and direction from the cement facility. However, it was observed that the mean concentration of the soil samples has no special trend with respect to distance and direction from the facility for most metals. However, the cement plant is a major source responsible for metal distribution, but it seems that uniformity in topography and vegetation is major factor to control this kind of distribution pattern. According to the index of geoaccumulation, the soils of the study area are considered to uncontaminated with respect to As, Co, Cr, Cu, Mn, Mo, Pb and Zn. Cd and Ni shows uncontaminated to moderated contaminated characteristics. The result of enrichment factor show that, with the exception of Cd and Ni enrichment, all the metals were deficiency to minimal enriched in all the distances and directions considered for the study. The results of the metal analysis indicated that the environment under study is not at risk seriously.

Keywords: Geochemistry, Cement Plant, Heavy Metals, Contamination, Geoaccumulation Index, Khash.

1. Introduction

Air pollution has long been recognized as a lethal form of pollution. Much of the problems of societal concern today are the heavy metals associated with air pollution. Heavy metal mobilization in the biosphere by human activities has become an important process in the geochemical cycling of these metals [1]. This is evident in industrial areas where stationery and mobile sources release large quantities of heavy metals into the atmosphere, soil and plants exceeding the natural emission levels [2, 3]. Pollution of natural environment by heavy metals is a worldwide problem because these metals are indestructible and most of them have toxic effects on living organisms where they exceed a certain concentration [4, 5, 6 and 7]. Metal distribution between soils is a key issue in assessing environmental effect of metals in the environment [8]. Soil is not only a medium for plant growth or pool to dispose of undesirable materials, but also a transmitter of many pollutants to surface water, groundwater, atmosphere and food [9].

The determination of the metals in soils, dusts, plants and sediments are very important in monitoring environmental pollution. The contribution of metals to environmental pollution from industrial, agricultural and mining processes besides automobile emission, have been the main subject of many studies and research in recent years [10].

Onder et al. [11] has observed that soil sampling is the most economical and reasonable method for monitoring heavy metals in the atmosphere. Hence, soil has been widely used as cumulative matrices of long term exposure to environmental pollutants [12-15]. Atmospheric emissions from industrial establishments are one of the major sources of environmental pollution. One type of industry that causes particle pollution is cement industry [16]. The main inputs of cement activity on the environment are the broadcasts of dust and gases [17]. Cement dust spreads along large areas through wind rain etc, and are accumulated in and on soils, plants and animals can affect human health badly [18]. Heavy metals are among the most relevant substances emitted during the process of cement manufacture [14, 16, and 19]. The influence of cement dust as a major cause of heavy metal contamination in soils has been observed by several researchers [14, 16, 17, 20, 21, and 22]. Metals especially known to have toxic effect in environmental studies are arsenic, cadmium, lead, mercury and thallium [23, 24]. Aluminum, beryllium, chromium, copper, manganese, nickel and zinc, among others, have been identified in the emission from cement plants [14]. The main purpose of the present study was to evaluate the environmental impact caused by dust emissions from a cement plant in the Khash Region of Iran within adjacent area with no other air pollution source in the area.

Khash city, with 60,000 inhabitants, lies about 45 km south of the dormant stratovolcano of Taftan, within latitudes 61° 4.8' N and 62° 20.9' N and

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longitudes 28° 4.8' E and 28° 21.6' E, in the core of an eroded anticline. Except the report of 1:100,000-scale geological map of Khash (Figure.1) [25], there is no integrated study to define the valley geometry and geology. The older rocks in the zone are of Eocene age. They are composed of limestone, marl, calcareous shale, and sandstone. The rocks exposed in the area appear to be entirely Tertiary and Quaternary in age with the exception of blocks of ultramafic rocks and pelagic limestone within areas of Cretaceous "Coloured Melange". The Eocene limestones and their associated calcareous sediments form prominent ridges and inselbergs throughout the area. On the road to cement plant, 15 km east-southeast of Kash, purplish and grayish coarse clastic sediments, with volcanogenic fragments are interbedded with the shales. Black recrystallized Nummulitic limestone appears to underly these sediments in this locality. The massive, usually foraminiferal limestone (En1) forms prominent ridges throughout the area. Overlying the foraminiferal limestone is a rather variable sequence of calcareous sediments (Elm). Generally, these are orange, brown or variegated purple and green marls, shales and limestones with little or no fossils content. They appear to represent a change to pelagic, calcareous sedimentation. This sequence is highly variable in colour and thickness. In the area immediately east of Khash these rocks pass upwards into red, purple and green calcareous shale (Elsh).

2. Samples and methodology

The sampling sites were selected in such a manner to cover the entire vicinity of the cement plant. To provide a satisfactory environmental representation of the study area, concentric circles of radii 500m, 1250m, 2000m around the main stack of the cement facility was taken into consideration. On first and second circles five sampling sites and in third circle ten sampling sites representing the north-east, east, south, north-west (prevalent wind direction) and west directions were created on a map (Figure 2). On the field, these pre-determined points were located by means of a GPS.

In April, 2012, 20 soil samples were collected in the designated sampling points. Soil specimens were taken with a small plastic shovel from the upper 10 cm of the soil and stored into labeled plastic bag. Any large stones or foreign objects were removed. In the laboratory, the soils were dried for three continuous days. All samples were sieved and after sieving, particles below 63 micron (mud fraction) were being dried, crushed and homogenized by agate mortar and stored in darkness until analytical procedures were carried out [26].

Analysis of metals was carried out using ICP-OES. The samples were digested to dryness using a four-acid digestion with 10 ml HF, 5 ml HClO₄, 2.5 ml HCl, and

2.5 ml HNO₃, then dissolved in 20% aqua regia and made up to 10 ml for ICP-OES analysis in GSI (Geological Survey of Iran). Toxic elements studied include As, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, and Zn.

3. Results and Discussion

Tables 1 summarize the concentrations of 10 heavy metals, in 20 soil samples collected in the vicinity of the Khash Cement plant. All the 10 elements display their presence in all the soil samples used for the study. The highest level corresponded to Mn and Ni followed by Cr. The lowest level of metal was recorded for Cd and Mo. Bergmann [27] has shown that the toxic levels of Cr in soil is around 2-50 ppm. The critical level for Ni in soil has been investigated by many researchers [27-30] and estimated to be in the range of 2-50 ppm. According to the present study, the range of Ni (91-127 ppm) is alarming suggesting that Ni pollution is critical in the investigated area. Meanwhile, the elemental concentrations in soil samples are in the decreasing order of Mn>Ni>Cr>Zn>Cu>Pb>Co>As>Mo>Cd.

To assess the influence of the cement facility on the area directly within the vicinity, various radii distances (500, 1250 and 2000 m) as well as different directions (north-east, east, south, north-west and west) with the facility as the centre were considered. Tables 2 and 3 summarize the mean elemental concentrations of soil with respect to distance and direction from the cement facility. A close observation from the available data reveals that the highest levels of some metals occur closer from the plant. For instance, the highest level of Cr and Ni is observed at a radius distance (rd) of 500m (Figure 3). However, a close inspection of Table 2 suggests that the mean concentration of the soil samples has no special trend with respect to distance from the facility for most metals. In addition according to table 3, there is no relationship between prevalent wind direction (north-west) and distribution pattern of heavy metals.

Table 1. Heavy metal concentration in soil samples from Khash Cement plant

Metal	Heavy metal concentration(ppm)				
	Min	Max	Mean	Median	Std
As	4.11	21.70	10.67	9.98	3.88
Cd	0.67	1.22	0.90	0.88	0.19
Co	11.95	25.73	18.52	18.50	3.29
Cr	53.03	176.17	83.24	81.34	27.42
Cu	22.91	46.50	33.17	32.72	6.09
Mn	611.81	804.98	726.02	722.83	56.01
Ni	91.07	127.10	103.61	102.52	8.34
Pb	8.45	27.41	18.59	19.81	5.53
Zn	55.84	90.25	67.66	65.72	7.99
Mo	0.34	3.72	2.15	2.20	0.85

Table 2. Concentration of heavy metals (ppm) in soil samples at varying distance from the Khash Cement plant

Distance(m)	As	Cd	Co	Cr	Cu	Mn	Ni	Pb	Zn	Mo
500	12.41	0.81	19.38	98.04	30.23	690.61	108.63	20.71	65.74	2.01
1250	12.39	0.87	19.07	81.87	32.30	699.84	106.52	17.74	66.66	2.23
2000	10.83	0.90	18.59	84.01	32.99	723.22	103.97	18.65	67.53	2.15

Table 3. Concentration of heavy metals (ppm) in soil samples at varying direction from the Khash Cement plant

Metal	Geographical directions from cement factory				
	NE	NW	SW	SE	S
As	0.73	0.96	0.79	0.76	0.85
Cd	2.81	2.99	3.43	2.86	2.92
Co	1.09	0.94	0.97	1.00	0.85
Cr	1.21	0.79	0.86	0.84	0.90
Cu	0.78	0.59	0.77	0.75	0.78
Mn	0.87	0.80	0.84	0.86	0.87
Ni	2.18	1.94	2.08	1.99	2.14
Pb	0.85	0.89	0.94	1.00	0.95
Zn	0.70	0.66	0.71	0.69	0.78
Mo	0.91	0.97	0.83	0.71	0.69

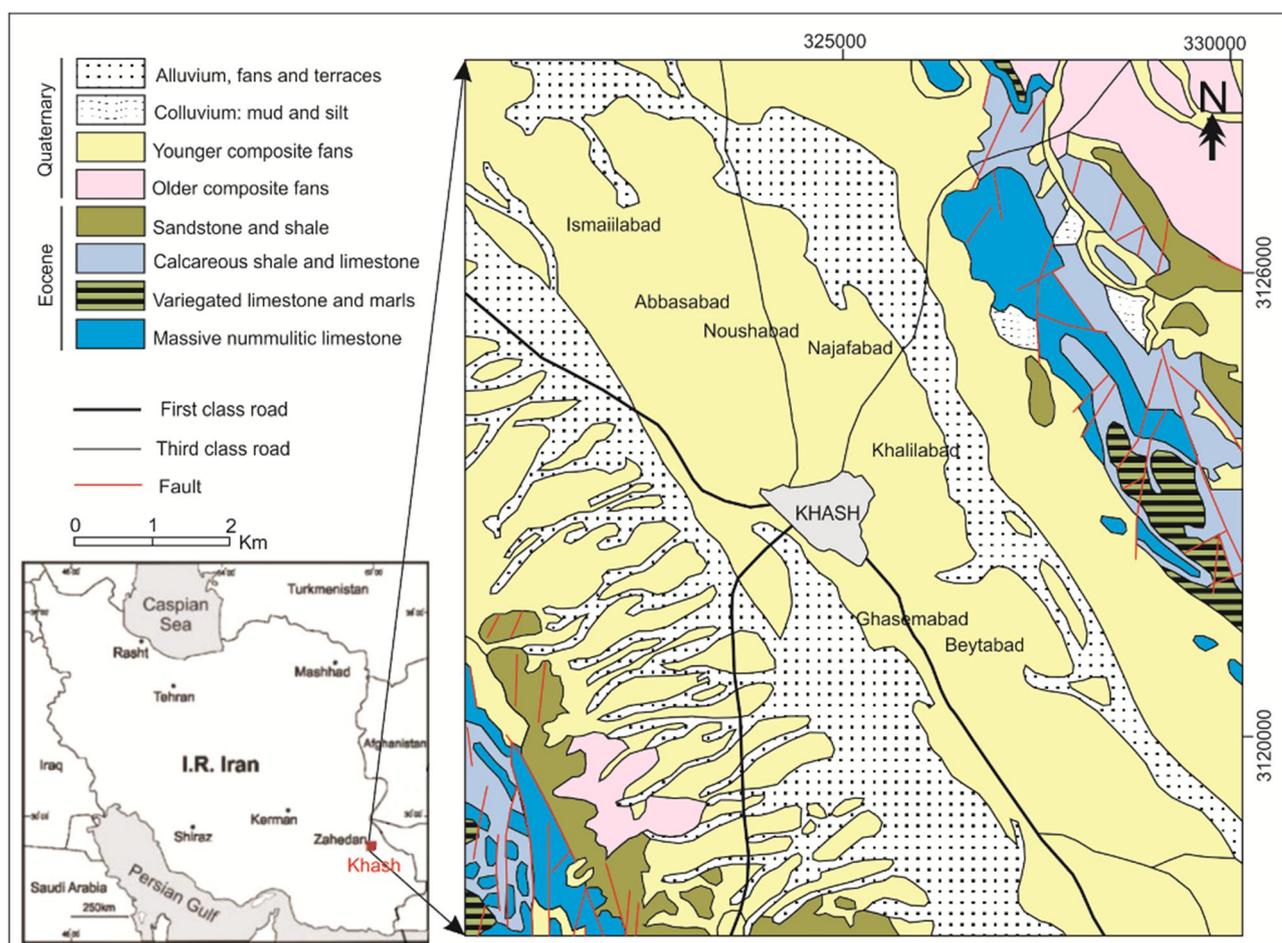


Fig. 1. Geological map of Khash area (redrawn from [25]).

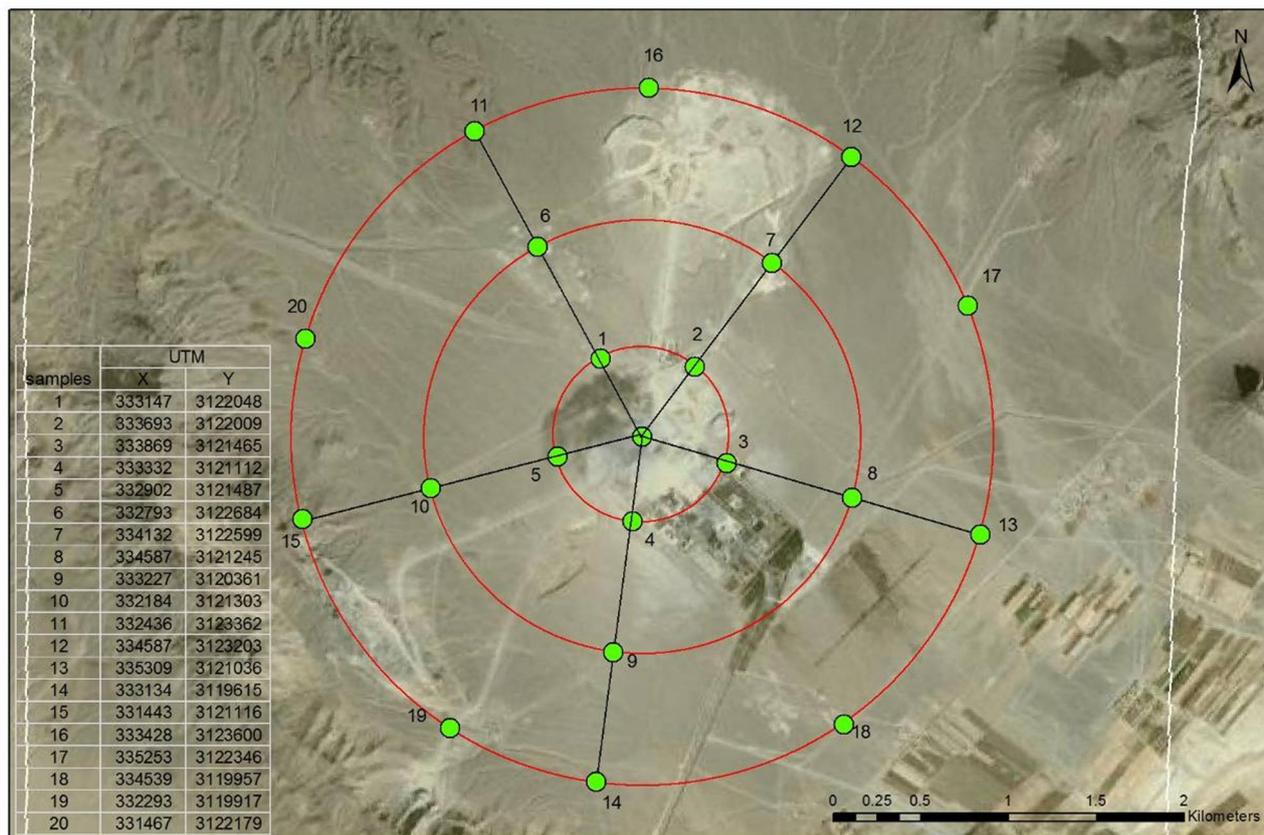


Fig. 2. Location of sampling points around Khash Cement plant.

As a result of the complicated relationship in levels of individual element in the different set of samples, focusing on their distribution in terms of distance and direction will be futile; instead we find a way of discussing contamination status of the sampling sites. Surface soil is considered as major sink of airborne metals. Consequently, the measurement of metals in soil can be useful to establish trends and abundance and their consequences as a result of natural changes and those caused by man [31].

In this study, three contamination indices have been used to express the extent of soil pollution in the area. The assessment of soil or sediment enrichment can be carried out in many ways. The most common ones are the index of geoaccumulation and enrichment factors [32]. In this work, the index of geoaccumulation (Igeo), Enrichment Factor (EF) and Pollution Load Index (PLI) have been applied to assess heavy metals (i.e., As, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb and Zn) distribution and contamination in surface soils in the vicinity of the Khash Cement facility. The index of geoaccumulation (Igeo) was originally used with bottom sediment by Muller [33]. It is computed by the following equation:

$$[I_{geo} = \log_2 \left[\frac{C_n}{1.5 B_n} \right]] \quad (1)$$

Where, C_n is the measured concentration of the element in the tested sediment (surface soils) and B_n is the geochemical background value of the element in fossil argillaceous sediment (continental crusted average or average shale). The constant 1.5 is introduced to minimize the effect of possible variations in the background values which may be attributed to lithologic variations in the sediments [32]. Odewande and Abimbola [34] gave the following interpretation for the geoaccumulation index: $I_{geo} < 0$ = uncontaminated; $0 < I_{geo} < 1$ = uncontaminated to moderate contaminated; $1 < I_{geo} < 2$ = moderately contaminated; $2 < I_{geo} < 3$ = moderately to strongly contaminated; $3 < I_{geo} < 4$ = strongly contaminated; $4 < I_{geo} < 5$ = strongly to extremely contaminated; and $I_{geo} > 5$ = extremely contaminated.

Enrichment factor (EF) has been employed for the assessment of contamination in various environmental media by several researchers [32, 35, 36, and 37]. Its version adapted to assess the contamination of various environmental media is as follows:

$$[EF = \left[\frac{\left[\frac{C_x}{C_{ref}} \right]_{sample}}{\left[\frac{B_x}{B_{ref}} \right]_{background}} \right]] \quad (2)$$

Where C_x is content of the examined element in the examined environment; C_{ref} is content of the

examined element in the reference environment; Bx is content of the reference element in the examined environment, and Bref is content of the reference element in the reference environment.

An element is regarded as a reference element if it is of low occurrence variability and is present in the sample in trace amounts. It is also possible to apply an element of geochemical nature whose substantial amounts occur in the environment but has no characteristic effects i.e. synergism or antagonism towards an examined element. Usually, Al is selected as normalization element. But in the surface living environment, the geochemical activity of Sc is less active than that of Al. So in view of the geochemical behavior peculiarity, it is reasonable to select Sc as normalization element. Sc as normalization element has been thought in surveying the soil background value in China. The result of survey proves it is successful [38- 40].

Five contamination categories are recognized on the basis of the enrichment factor: EF < 2 states deficiency to minimal enrichment; EF = 2-5 moderate enrichment; EF = 5-20 severe enrichment; EF = 20-40 very high enrichment; and EF > 40 extremely high enrichment [35]. Each sampling site was evaluated for the extent of metal pollution by employing the method based on the pollution load index (PLI) developed by Thomilson et al. [41] as follows:

$$[PLI = n\sqrt{CF1 \times CF2 \times CF3 \times \dots CFn}] \quad (3)$$

Where n is the number of metals studied (ten in this study) and CF is the contamination factor defined by $CF = C_{metal} / C_{background}$. C_{metal} is the concentration of pollutant in sediment and $C_{background}$ is the background value for the metal. The PLI provides simple but comparative means for assessing a site quality, where a value of $PLI < 1$ denote perfection; $PLI = 1$ present that only baseline levels of pollutants are present and $PLI > 1$ would indicate deterioration of site quality [36].

The EF and Igeo of heavy metals under the current study was computed for each element for each radii distance and direction relative to the background value of the element in average shale value of the elements were taken from Turekian and Wedepohl [42]. In addition, the PLI was calculated for the overall distances and directions. Tables 4 and 5 display a summary of the EF and Igeo values for each heavy metal in terms of distance and direction respectively, in addition to PLI of the environments at each distance and direction.

According to Zhang and Liu [43], EF values between 0.5 and 1.5 indicate that a metal is entirely from crusted material or natural processes, whereas EF values greater than 1.5 suggests that the source are more likely to be anthropogenic. The result of the present study show that, with the exception of Cd and

Ni enrichment, all the metals were deficiency to minimal enriched in all distances and directions considered for the study (Table 4 and 5) since the EF values of the metals are smaller than 1.5. Fortunately these results indicate that the environment under study is not at risk seriously. The differences in the EF values may be due to the difference in the magnitude of input for each metal in the soil and/or differences in the removal rate of each metal from the soil [44].

The pollution or the contamination levels in the environment under consideration was further expressed in terms of geoaccumulation index. The index indicated that the environment is uncontaminated in all distance and direction with respect to As, Co, Cr, Cu, Mn, Mo, Pb and Zn (Table 4 and 5). Cd and Ni shows uncontaminated to moderated contaminated characteristics at all distances and direction. Banat et al. [45] revealed that air emission from cement kilns is possible sources of Cd. To effectively compare whether the vicinity of the plant is contaminated or not, the PLI described earlier was used. The PLI is aimed at providing a measure of the degree of overall contamination at the sampling sites in terms of distance and direction. Based on the results presented in Table 4 there is a very minor difference in the amount of PLI. Based on the results presented in Table 6, the overall degree of contamination by the 10 metals in distance terms is of the order $500m > 1250m > 2000m$. The analysis of the distance results indicate that the content of contaminants have almost uniform pattern in whole of the study area. Probably this situation is due to uniform topography and vegetation in the vicinity of Khash Cement plant. In addition, vehicular traffic activity is another responsible factor that controls this pattern, because trucks are active in entire area. These trucks may contribute to the distribution of dust from processed cement and cement materials in the study area. The distribution of the metal concentration of the soil in the study area indicated that this area has not been affected by anthropogenic activity seriously.

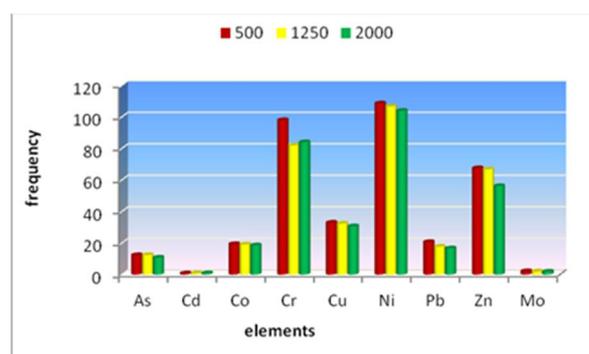


Fig. 3. Bar chart of concentration of heavy metals (ppm) in soil samples at varying distance from the Khash Cement plant

Table 4. Values of the enrichment factor and geoaccumulation index and PLI for heavy metals at varying distances from the cement facility

Distance(m)	Index	Heavy metals										
		As	Cd	Co	Cu	Cr	Mn	Mo	Ni	Pb	Zn	PLI
500	I _{geo}	-0.69	0.83	-0.57	-1.19	-0.57	-0.89	-0.96	0.53	-0.56	-1.14	1.06
	EF	1.10	3.10	1.19	0.77	1.27	0.94	0.91	2.51	1.21	0.79	
1250	I _{geo}	-0.72	0.91	-0.60	-1.10	-0.83	-0.87	-0.91	0.50	-0.81	-1.11	1.03
	EF	1.10	3.27	1.14	0.81	1.04	0.94	0.97	2.42	1.01	0.79	
2000	I _{geo}	-0.94	0.96	-0.64	-1.06	-0.75	-0.82	-1.02	0.47	-0.75	-1.09	1.02
	EF	0.93	3.27	1.07	0.80	1.02	0.93	0.91	2.28	1.02	0.78	

Table 5. Values of the enrichment factor (EF) and geoaccumulation index (I_{geo}) for heavy metals at various directions from the cement facility

Heavy metals	Index	Geographical directions around Khash Cement plant				
		NE	SE	S	SW	NW
As	I _{geo}	-1.21	-1.07	-0.85	-0.93	-0.75
	EF	0.82	0.80	0.91	0.85	1.17
Cd	I _{geo}	0.87	0.93	0.93	1.17	0.97
	EF	3.01	3.00	3.12	3.65	3.62
Co	I _{geo}	-0.46	-0.61	-0.84	-0.64	-0.67
	EF	1.19	1.03	0.90	1.04	1.15
Cu	I _{geo}	-0.94	-1.01	-0.96	-0.99	-1.35
	EF	0.85	0.78	0.83	0.82	0.71
Cr	I _{geo}	-0.43	-0.87	-0.76	-0.80	-0.94
	EF	1.34	0.87	0.96	0.93	0.95
Mn	I _{geo}	-0.79	-0.80	-0.78	-0.83	-0.90
	EF	0.94	0.90	0.94	0.91	0.97
Mo	I _{geo}	-0.77	-1.21	-1.22	-1.28	-0.64
	EF	0.99	0.74	0.73	0.89	1.17
Ni	I _{geo}	0.54	0.41	0.51	0.47	0.38
	EF	2.36	2.08	2.29	2.25	2.35
Pb	I _{geo}	-0.88	-0.64	-0.71	-0.71	-0.86
	EF	0.93	1.03	1.01	1.01	1.08
Zn	I _{geo}	-1.10	-1.12	-0.96	-1.07	-1.18
	EF	0.76	0.72	0.83	0.77	0.80
PLI		1.08	1.01	1.03	1.058	1.059

These results revealed that majority of the sampling points were not enriched or contaminated with all heavy metals except Ni and Cd. Fortunately these results indicate that the environment under study is not at risk seriously. It seems that cement facility together with the attendant vehicular traffic and emissions were implicated as responsible for some metal pollution (i.e., Ni and Cd) in the study area. The PLI values for all sites are not significantly different. It seems that the prevailing wind (Northwest) does not affect to dispersion of pollutants. By way of monitoring the operational influence of the cement facility on the environment, this study underlines the need for replicating periodic studies (two years duration) on the facility on the environment in addition to the evaluation of the facility on human health.

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