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Spatial distribution and source identification of heavy metal pollution in roadside surface soil: a study of Dhaka Aricha highway, Bangladesh

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Abstract

Introduction: In this study, metal pollution and their sources in surface soils were evaluated by pollution indices and multivariate statistical techniques in association with a geographical information system (GIS).

Methods: Surface soil samples were collected in dry season from different locations of Dhaka Aricha highway and analyzed by energy dispersive X-ray fluorescence (EDXRF).

Results: Thirteen different metals were found in the tested samples. Pollution indices are determined by enrichment factor in an order of Zr > Sn > P > Mn > Zn > Rb > Fe > Ba > Sr > Ti > K > Ca > Al. The resulting geoaccumulation index (I_{geo}) value shows the following order: Sn > Zr > P > Mn > Zn > Rb > Fe > Ba > Ti > Sr > K > Ca > Al. Contamination factors (CFs) of the metals range from 1.422 to 3.979 (Fe); 0.213 to 1.089 (Al); 0.489 to 3.484 (Ca); 1.496 to 2.372 (K); 1.287 to 3.870 (Ti); 2.200 to 14.588 (Mn); 5.938 to 56.750 (Zr); 0.980 to 3.500 (Sr); 2.321 to 4.857 (Rb); 2.737 to 6.526 (Zn); 16.667 to 27.333 (Sn); 3.157 to 16.286 (P); and 0.741 to 3.328 (Ba). Pollution load index calculated from the CFs indicates that soils are strongly contaminated by Zr and Sn. Principal component analysis (PCA) of parameters exhibits three major components. R-mode cluster analysis reveals three distinct groups in both site and metal basis clustering that shows a similar pattern with the PCA.

Conclusions: These results might be helpful for future monitoring of further increase of heavy metal concentrations in surface soils along highways.

Keywords: Heavy metals, Geoaccumulation index (I_{geo}), Contamination factor (CF), Pollution load index (PLI), Principal component analysis (PCA), Dhaka Aricha highway

Introduction

Rapid urbanization and industrialization in Bangladesh leads to economic growth and gives the chance of thinking of being a developed country in the future; on the other side, this process changes the whole environment drastically. Our planet, or in a small sense the environment, has the capability to minimize the adverse effects, but now, it is alarming for us to think about it. Dhaka, a megacity in the world, is in the worst situation in terms of environmental perspective. Environmental pollution has crossed its line, degrading the whole environment day by day. High population and large density of vehicle emissions are industrial pollutions that are circulated everywhere, the main culprits to degrade the system (Chowdhury 2006; Islam 2014). The situation is worst in the transition point of the city like bus stands. The major highway like Dhaka Aricha highway is being polluted for vehicle emissions, an industrial pollution that causes the disturbance of the environment. Heavy metal contamination of aquatic ecosystems has received much attention because of its toxicity, abundance, and persistence (Arnason and Fletcher 2003). Elevated levels of

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heavy metals in environmental compartments, such as aquatic soils, may pose a risk to human health due to their transfer in aquatic media and uptake by living organisms, thereby entering the food chain (Sin et al. 2001; Varol and Sen 2012). Soils are ecologically sensitive components of the aquatic ecosystems and are also a reservoir of the contaminants, which take part considerably in maintaining the trophic status for any water reservoir (Singh et al. 2005a, b).

Roads play a major role in stimulating social and economic progress, and road construction has also resulted in heavy environment pollution in this region (Liu et al. 2006). Road traffic is an important deleterious factor concerning air quality, noise, and land consumption (Zechmeister et al. 2005). The contribution of cars and road transports to the global emission of atmospheric pollutants is regularly increasing (Viard et al. 2004). The road transports also induce the contamination of nearer soils by a pollutant transfer via the atmospheric fallouts (Viard et al. 2004; Nabuloa et al. 2006) or road runoff (Mitsch and Gosselink 1993; Nabuloa et al. 2006). Maximum researchers have stated the influence of the traffic load on heavy metal contents in topsoils and their variability with distance (Ward et al. 1977; Rodriguez and Rodriguez 1982; Garcia and Milla´n 1998; Zhang et al. 1999; Turer and Maynard 2003). Nabuloa et al. (2006) also showed total trace metal concentrations in roadside soils decreased exponentially with increasing distance from roadways. Although the concentrations of metals in the roadside soil were influenced by meteorological conditions (Othman et al. 1997; Sezgin et al. 2003), traffic density (Garcia and Milla´n 1998; Nabuloa et al. 2006), the kind of vehicle in traffic (Sezgin et al. 2003; Nabuloa et al. 2006), and soil parameters (Viard et al. 2004) were also verified in some studies; little information was known about the heavy metal accumulation in roadside soils along the roads with different transportation periods.

Dhaka Aricha highway plays a vital role in inter-district and inter-regional transports as it links the northwestern and northern region of Bangladesh with Dhaka. It originates from Amin Bazar Bridge and ends at Aricha Ghat, covering a length of 75.4 km (Hoque et al. 2007). Huge vehicle loads and industrial activities make a pollutant hotspot zone around these highway areas. Emissions from high transportation density disperse around the agricultural field, water body, and livestock areas which are alongside the highway areas. Huge contamination loads especially heavy metals accumulate in the biotic components and enter into the food chain. Concentration of these heavy metals in soils is associated with geometrical cycles and biological processes and could be greatly influenced by high traffic load and transportation activities. In the food chain, primary producers, i.e., plants, are capable of absorbing these metals from the soil (Kakulu and Abdullahi 2004; Rajaram and Das 2008). These metals each contaminate into the soil when they undergo chemical reactions and could come in direct contact with roots of plants (Udosen et al. 1990). When these plants in the form of vegetables are consumed by man, trace metals become bioaccumulated and eventually result in several ailments which may subsequently cause death (Odiette 1999). In some cases, plants accumulate some of these metals which are not injurious to them but may be poisonous to animals grazing on the plants (Raven and Evert 1976). Nabuloa et al. (2006) reported that leaves of roadside crops can accumulate trace metals at high concentrations, causing a serious health risk to consumers.

Monitoring of anthropogenic release of heavy metals is usually done to determine the distribution of pollutants and apportionment of sources (Kelepertsis et al. 2006). Among the statistical techniques, both principal component analysis (PCA) and cluster analysis (CA) are useful methods to discover common patterns in data distribution, leading to initial dimension reduction of datasets and helping its interpretation (Franco-Uría et al. 2009). PCA and CA assist to set up analyzed parameters in different factors/groups on the basis of contribution from their possible sources. FA and PCA have been widely used to expose variable redundancy and combine variables into single factors (Wilcke et al. 1998; Chen et al. 1999; Kumru and Bakac 2003; Navas and Machin 2002; Bretzel and Calderisi 2006). CA is often coupled to FA and PCA to provide groupings of individual variables according to distances or similarity indices (Facchinelli et al. 2001; Granero and Domingo 2002; Manta et al. 2002; Wang et al. 2005; Han et al. 2006). The explanation of the above data processing helps to identify pollution sources and allocate natural vs. anthropic contribution. The geographical information system (GIS) software is increasingly used in environmental studies because of its capability to expose non-point source contaminants (Sultan 2007; Wang et al. 2006) and as a visual support in interpreting heavy metal spatial distribution.

In Bangladesh, determination of the heavy metals along the roadside is now a growing demand because of metal biomagnification in the food chain and their potential health impact. This study focuses on the identification of heavy metals in the roadside surface soils of Dhaka Aricha highway which will be served as a baseline study in Bangladesh for future monitoring of heavy metals and their levels around the roadside areas. Major objectives of the present study were (i) to measure the concentrations of metals (Fe, Al, Ca, K, Ti, Mn, Zr, Sr, Rb, Zn, Sn, P, and Ba) in surface soils of Dhaka Aricha highway; (ii) to determine potential pollution indices using enrichment factor (EF), geoaccumulation index (I_{geo}), contamination factor (CF), and pollution load index (PLI); and (iii) to define their natural/anthropogenic contributions using multivariate statistical methods. It is anticipated that the study would provide a baseline data regarding the distribution, accumulation, and sources of heavy metals in the roadside surface soils of Dhaka Aricha highway.

Study area

The area selected for the study was along the Dhaka Aricha road which lies between latitudes from 23° 47′ 45.84′′ N to 23° 47′ 40.08′′ N and longitude 90° 16′ 36.04′′ E to 90° 19′ 33.80′′ E which is 5.04 km long. The study area situated near Savar which is 17 km north from the Dhaka center runs northward. The study area was selected because it links with Dhaka City with comparatively high traffic density and has industrial influence. It carries, on an average, 9000 motor vehicles per day. The study area is surrounded by numerous brick fields and near the Amin Bazar landfill area. The Gabtoli Amin Bazar area is the transition point of Dhaka City, the largest bus stand acting as the entry and exit points of the city. Average elevation is 26.5 ft above sea level. This area is perennially inundated by monsoon flood (June to August) and roadside runoff. The geology of this area is the uplifted Madhupur area which is covered by dark reddish-brown to brownish-red, mottled, sticky, and compact Madhupur Clay Residuum of the Pleistocene age, underlain by Plio-Pleistocene Dupi Tila sandstone formation (Maitra and Akhter 2011).

Methods

Sample collection

A total of 19 soil samples (prefixed S) was collected January, 2014, during dry seasons from roadside surface soils of Hemayetpur to Gabtoli area, Savar of Dhaka Aricha highway (Fig. 1). The soil samples were collected manually with a stainless steel spatula, cleaned after each sampling for foreign matter and carried within zip-mouthed PVC packages. All the soil samples were collected from the upper layer of the soil (about 0–5 cm). The soil samples were tightly packed and transferred to Institute of Food Science and Technology (IFST), Bangladesh Council of Scientific and Industrial Research (BCSIR), Dhaka, for metal determination in energy dispersive X-ray fluorescence (EDXRF). The samples were properly labeled and kept in room temperature.

Sample preparation

The collected soil samples were homogeneously mixed up. Unwanted portions like plant roots, stones, or other debris were removed. Then the samples were kept in a microwave oven about 48 h (at 60 °C). The soil samples were kept in room temperature and grinded with mortar and pestle. Fifteen grams of the grinded samples was taken for pellet formation. In the VANEOX pressing machine, a 15-ton pressure was used to form the pellet. After the pellet formation, the samples were ready for the analysis in EDXRF.

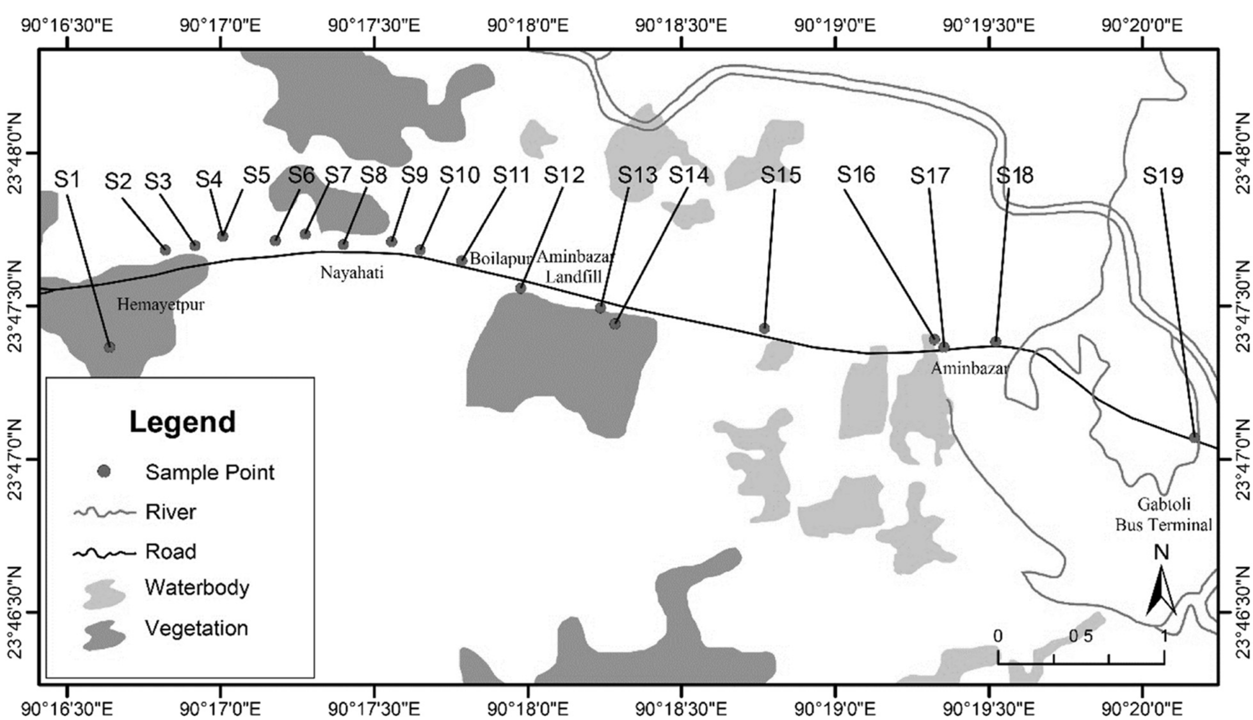

Fig. 1 Sampling location map of the study area

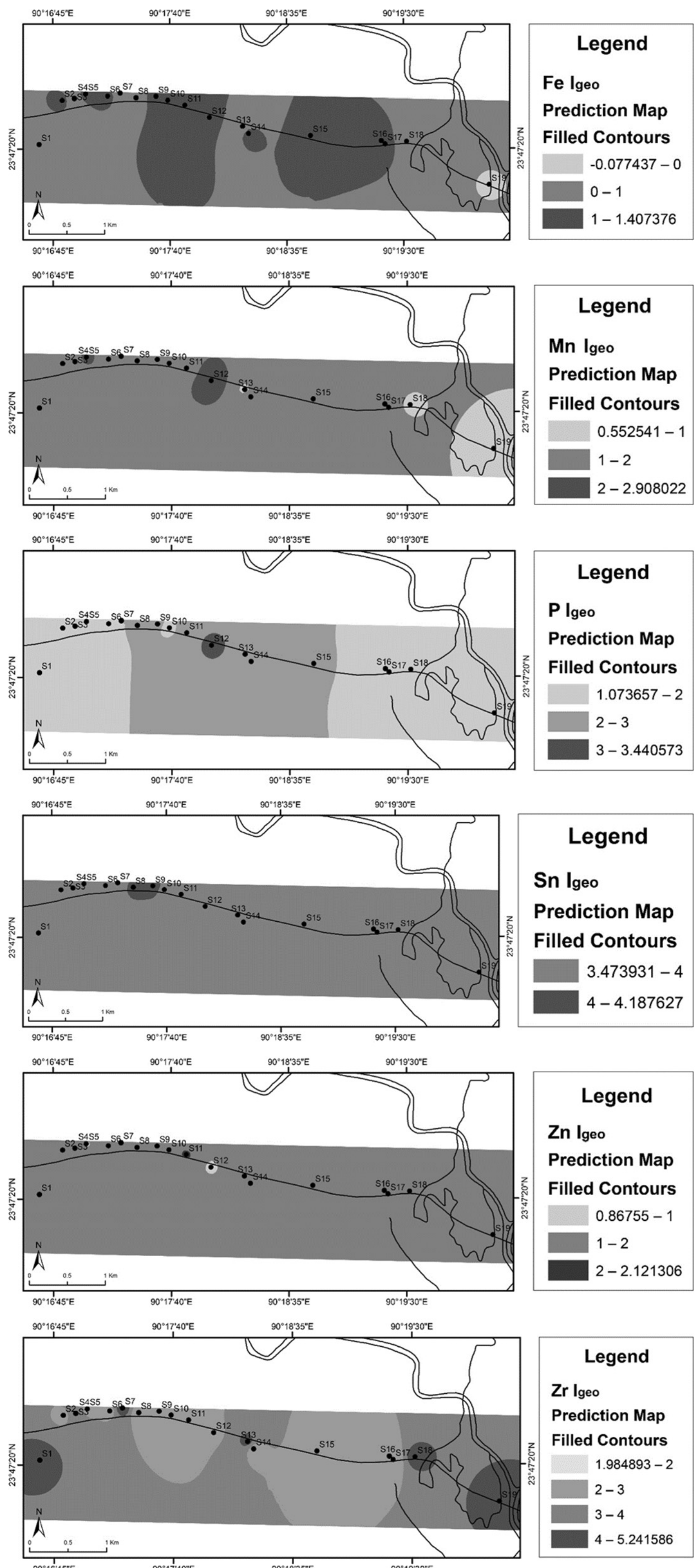

Fig. 2 Spatial distribution showing I_{geo} value of metals in different sampling sites

Analysis of elements and data acquisition by EDXRF

The elemental analysis was performed by ARL QUANT'X EDXRF, Thermo Scientific, USA, a spectrometer at IFST, BCSIR, Dhaka. EDXRF is equipped with a rhodium (Rh) anode along with an assembly of eight filters (Al, cellulose, Cu thick, Cu thin, No, Pd medium, Pd thick, Pd thin), and a Si (Li) detector (with a 15-mm^2 area and less than an equal 76-μm beryllium window) was used for the determination of elements of the samples (Adyel et al. 2012). The sample is positioned in the Teflon cup assemblies. In the present work, the measurements were carried out in air. UniQUANT ED is the main system software to run the analysis in this EDXRF. The acquired data were processed with the help of a connected computer. The data is generated in percentage value. It can be converted to ppm value by multiplying by 10,000 (conversion process described in the software system). This instrument shows the >5-ppm value (instrument setup process). The value is generated by the average values of three time running value commands by the operator. The value is the average value of three-time running values in the instrument.

Assessment of soil pollution

EF

The enrichment factor can be calculated by dividing its ratio to the normalizing element by the same ratio found in the chosen baseline (Turekian and Wedepohl 1961). EF is calculated by the following equation:

$$\mathrm{EF} = (\mathrm{Metal}/\mathrm{Fe})_{\mathrm{Sample}} / (\mathrm{Metal}/\mathrm{Fe})_{\mathrm{Background}}$$

The EF values close to unity indicate crusted origin; those less than 1.0 suggest a possible mobilization or depletion of metals (Zsefer et al. 1996).

EFs >1.0 suggest possible anthropogenic origin. EFs >10 are suggest to be a non-crusted source. For geochemical normalization, iron (Fe) was used as the reference element (Daskalakis and O'Connor 1995).

I_{geo}

I_{geo} is calculated to estimate the enrichment of metal concentrations above the background level which was proposed by Muller (1969). I_{geo} is calculated using following equation:

$$I_{\mathrm{geo}} = \mathrm{Log}_2(C_{\mathrm{n}}/1.5B_{\mathrm{n}})$$

where

C_{n} = concentration of the element in the enriched samples

B_{n} = background value of the element

The factor 1.5 is introduced to minimize the effect of possible variations in the background values which may be attributed to lithologic variations in the soils (Stoffers et al. 1986). Muller (1969) proposed the descriptive classes for increasing I_{geo} value (Table 1).

CF

The CF is the ratio obtained by dividing the concentration of each metal in the soil by the baseline or background value (Turekian and Wedepohl 1961):

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

The contamination levels can be classified based on their intensities on a scale ranging from 1 to 6. They were classified as 0 = none, 1 = none to medium, 2 = moderate, 3 = moderately to strong, 4 = strongly polluted, 5 = strong to very strong, and 6 = very strong (Muller 1969).

PLI

For the entire sampling site, PLI has been determined as the *n*th root of the product of the *n* CF (Usero et al. 2000):

$$\mathrm{PLI} = (\mathrm{CF}_1 \times \mathrm{CF}_2 \times \mathrm{CF}_3 \times \cdots \times \mathrm{CF}_n)^{1/n}$$

Statistical analysis

Analyzed data were subjected to multivariate analysis: PCA and FA and CA using SPSS version 22.0 and Microsoft Excel 2013.

PCA and FA

PCA is designed to transform the original variables into new, uncorrelated variables (axes), called the principal components, which are linear combinations of the original variables. The new axes lie along the directions of maximum variance (Sarbu and Pop 2005). The principal component (PC) can be expressed as the following:

$$z_{ij} = ai_1x_1j + ai_2x_2j + ai_3x_3j + \cdots \ldots\ldots\ldots\ldots\ldots.. + a_{im}x_{mj}$$

where z is the component score, a is the component loading, x is the measured value of a variable, i is the component number, j is the sample number, and m is the total number of variables.

Table 1 Muller's classification for the geoaccumulation index

I_{geo} value	Class	Soil quality
≤0	0	Unpolluted
0–1	1	From unpolluted to moderately polluted
1–2	2	Moderately polluted
2–3	3	From moderately to strongly polluted
3–4	4	Strongly polluted
4–5	5	From strongly to extremely polluted
>6	6	Extremely polluted

Table 2 Concentration, enrichment factor, geoaccumulation index, contamination factor, and pollution load index of metals for surface soil

Sampling site	Fe					Al					Ca					K				
	Conc. (ppm)	EF	I_{geo}	CF	PLI	Conc. (ppm)	EF	I_{geo}	CF	PLI	Conc. (ppm)	EF	I_{geo}	CF	PLI	Conc. (ppm)	EF	I_{geo}	CF	PLI
S1	115,000	1	0.700	2.436	2.877	25,500	0.131	−2.234	0.319	0.597	52,200	0.969	0.655	2.362	1.247	49,900	0.770	0.323	1.876	2.033
S2	156,200	1	1.142	3.309		80,100	0.303	−0.583	1.001		11,000	0.150	−1.592	0.498		46,100	0.524	0.208	1.733	
S3	106,200	1	0.585	2.250		31,600	0.176	−1.925	0.395		77,000	1.549	1.216	3.484		54,600	0.912	0.453	2.053	
S4	151,800	1	1.100	3.216		80,200	0.312	−0.581	1.003		11,300	0.159	−1.553	0.511		39,800	0.465	−0.004	1.496	
S5	171,200	1	1.274	3.627		84,600	0.292	−0.504	1.058		10,800	0.135	−1.618	0.489		50,000	0.518	0.326	1.880	
S6	163,400	1	1.207	3.462		76,100	0.275	−0.657	0.951		19,800	0.259	−0.744	0.896		57,600	0.626	0.530	2.165	
S7	103,200	1	0.544	2.186		20,300	0.116	−2.563	0.254		38,700	0.801	0.223	1.751		48,500	0.834	0.282	1.823	
S8	108,400	1	0.615	2.297		38,400	0.209	−1.644	0.480		36,100	0.711	0.123	1.633		63,100	1.033	0.661	2.372	
S9	166,600	1	1.235	3.530		72,900	0.258	−0.719	0.911		17,200	0.220	−0.947	0.778		57,900	0.617	0.537	2.177	
S10	182,400	1	1.365	3.864		55,400	0.179	−1.115	0.693		30,000	0.351	−0.144	1.357		63,000	0.613	0.659	2.368	
S11	187,800	1	1.407	3.979		78,600	0.247	−0.610	0.983		21,100	0.240	−0.652	0.955		58,600	0.554	0.555	2.203	
S12	169,000	1	1.255	3.581		45,700	0.160	−1.393	0.571		61,100	0.772	0.882	2.765		45,500	0.478	0.189	1.711	
S13	78,900	1	0.156	1.672		20,700	0.155	−2.535	0.259		38,200	1.034	0.205	1.729		56,800	1.277	0.510	2.135	
S14	177,600	1	1.327	3.763		76,000	0.252	−0.659	0.950		31,800	0.382	−0.060	1.439		62,900	0.628	0.657	2.365	
S15	156,400	1	1.143	3.314		62,600	0.236	−0.939	0.783		27,700	0.378	−0.259	1.253		55,500	0.630	0.476	2.086	
S16	161,000	1	1.185	3.411		87,100	0.319	−0.462	1.089		22,800	0.302	−0.540	1.032		60,700	0.669	0.605	2.282	
S17	170,300	1	1.266	3.608		72,700	0.252	−0.723	0.909		23,200	0.291	−0.515	1.050		60,700	0.632	0.605	2.282	
S18	94,900	1	0.423	2.011		20,400	0.127	−2.556	0.255		38,300	0.862	0.208	1.733		46,900	0.877	0.233	1.763	
S19	67,100	1	−0.077	1.422		17,000	0.149	−2.819	0.213		34,300	1.092	0.049	1.552		58,100	1.536	0.542	2.184	
Sampling site	Ti					Mn					Zr					Sr				
	Conc. (ppm)	EF	I_{geo}	CF	PLI	Conc. (ppm)	EF	I_{geo}	CF	PLI	Conc. (ppm)	EF	I_{geo}	CF	PLI	Conc. (ppm)	EF	I_{geo}	CF	PLI
S1	9200	0.821	0.415	2.000	2.395	3020	1.458	1.244	3.553	4.351	7300	18.726	4.927	45.625	13.515	813	1.112	0.853	2.710	2.157
S2	12,100	0.795	0.810	2.630		3960	1.408	1.635	4.659		1520	2.871	2.663	9.500		371	0.374	−0.279	1.237	
S3	9100	0.879	0.399	1.978		2680	1.401	1.072	3.153		3430	9.528	3.837	21.438		942	1.396	1.066	3.140	
S4	12,400	0.838	0.846	2.696		12,400	4.536	3.282	14.588		1400	2.721	2.544	8.750		294	0.305	−0.614	0.980	
S5	13,200	0.791	0.936	2.870		3910	1.268	1.617	4.600		1650	2.843	2.781	10.313		334	0.307	−0.430	1.113	
S6	12,100	0.760	0.810	2.630		3450	1.172	1.436	4.059		1340	2.419	2.481	8.375		558	0.537	0.310	1.860	
S7	11,500	1.143	0.737	2.500		2540	1.367	0.994	2.988		9080	25.955	5.242	56.750		702	1.070	0.642	2.340	
S8	9200	0.871	0.415	2.000		3020	1.547	1.244	3.553		950	2.585	1.985	5.938		902	1.309	1.003	3.007	

Table 2 Concentration, enrichment factor, geoaccumulation index, contamination factor, and pollution load index of metals for surface soil *(Continued)*

S9	11,700	0.721	0.762	2.543		3860	1.287	1.598	4.541		1700	3.010	2.824	10.625		515	0.486	0.195	1.717	
S10	17,800	1.001	1.367	3.870		4860	1.480	1.930	5.718		1320	2.135	2.459	8.250		773	0.667	0.781	2.577	
S11	12,000	0.656	0.798	2.609		4030	1.192	1.660	4.741		1480	2.325	2.624	9.250		552	0.462	0.295	1.840	
S12	15,300	0.929	1.149	3.326		9570	3.144	2.908	11.259		1700	2.967	2.824	10.625		1050	0.978	1.222	3.500	
S13	7800	1.014	0.177	1.696		2240	1.577	0.813	2.635		5710	21.349	4.572	35.688		905	1.805	1.008	3.017	
S14	13,000	0.751	0.914	2.826		4070	1.273	1.675	4.788		1180	1.960	2.298	7.375		779	0.690	0.792	2.597	
S15	12,200	0.800	0.822	2.652		3460	1.228	1.440	4.071		1390	2.622	2.534	8.688		644	0.648	0.517	2.147	
S16	12,100	0.771	0.810	2.630		4200	1.449	1.720	4.941		1000	1.832	2.059	6.250		686	0.670	0.608	2.287	
S17	11,700	0.705	0.762	2.543		3960	1.291	1.635	4.659		1100	1.905	2.196	6.875		637	0.588	0.501	2.123	
S18	7500	0.811	0.120	1.630		2130	1.246	0.740	2.506		7190	22.350	4.905	44.938		731	1.212	0.700	2.437	
S19	5920	0.905	−0.221	1.287		1870	1.548	0.553	2.200		5440	23.917	4.503	34.000		776	1.820	0.786	2.587	
Sampling site	Rb					Zn					Sn					P				
	Conc. (ppm)	EF	I_{geo}	CF	PLI	Conc. (ppm)	EF	I_{geo}	CF	PLI	Conc. (ppm)	EF	I_{geo}	CF	PLI	Conc. (ppm)	EF	I_{geo}	CF	PLI
S1	430	1.261	1.034	3.071	3.561	350	1.512	1.296	3.684	3.993	123	8.414	3.773	20.500	20.866	2210	1.296	1.074	3.157	5.71
S2	430	0.928	1.034	3.071		320	1.018	1.167	3.368		126	6.346	3.807	21.000		2310	0.997	1.138	3.300	
S3	473	1.502	1.171	3.379		363	1.698	1.349	3.821		124	9.185	3.784	20.667		4560	2.895	2.119	6.514	
S4	427	0.948	1.024	3.050		287	0.939	1.010	3.021		123	6.374	3.773	20.500		3010	1.337	1.519	4.300	
S5	520	1.024	1.308	3.714		264	0.766	0.890	2.779		102	4.687	3.503	17.000		2590	1.020	1.303	3.700	
S6	580	1.197	1.466	4.143		370	1.125	1.377	3.895		128	6.162	3.830	21.333		3850	1.589	1.874	5.500	
S7	391	1.277	0.897	2.793		423	2.036	1.570	4.453		105	8.004	3.544	17.500		4205	2.747	2.002	6.007	
S8	542	1.686	1.368	3.871		431.5	1.978	1.598	4.542		164	11.902	4.188	27.333		4560	2.836	2.119	6.514	
S9	500	1.012	1.252	3.571		440	1.312	1.627	4.632		158	7.461	4.134	26.333		4410	1.785	2.070	6.300	
S10	620	1.146	1.562	4.429		330	0.899	1.212	3.474		136	5.865	3.918	22.667		3630	1.342	1.790	5.186	
S11	640	1.149	1.608	4.571		620	1.640	2.121	6.526		132	5.529	3.874	22.000		6070	2.179	2.531	8.671	
S12	660	1.317	1.652	4.714		260	0.764	0.868	2.737		100	4.655	3.474	16.667		11400	4.548	3.441	16.286	
S13	372	1.590	0.825	2.657		415	2.613	1.542	4.368		112	11.167	3.637	18.667		8065	6.892	2.941	11.521	
S14	650	1.234	1.630	4.643		570	1.595	2.000	6.000		143	6.334	3.990	23.833		4730	1.796	2.171	6.757	
S15	538	1.160	1.357	3.843		310	0.985	1.121	3.263		120	6.036	3.737	20.000		4680	2.018	2.156	6.686	
S16	680	1.424	1.695	4.857		290	0.895	1.025	3.053		118	5.766	3.713	19.667		2400	1.005	1.193	3.429	
S17	600	1.188	1.515	4.286		420	1.225	1.559	4.421		148	6.837	4.040	24.667		2960	1.172	1.495	4.229	
S18	335	1.190	0.674	2.393		470	2.461	1.722	4.947		132	10.942	3.874	22.000		4000	2.842	1.930	5.714	
S19	325	1.633	0.630	2.321		490	3.628	1.782	5.158		107	12.544	3.572	17.833		3600	3.618	1.778	5.143	

Table 3 Concentration, enrichment factor, geoaccumulation index, contamination factor, and pollution load index of metals for surface soil

Sampling site	Ba				
	Concentration (ppm)	EF	I_{geo}	CF	PLI
S1	1520	1.076	0.805	2.621	2.589
S2	1670	0.870	0.941	2.879	
S3	430	0.330	−1.017	0.741	
S4	1500	0.804	0.786	2.586	
S5	1350	0.642	0.634	2.328	
S6	1780	0.887	1.033	3.069	
S7	1440	1.136	0.727	2.483	
S8	1680	1.261	0.949	2.897	
S9	1610	0.786	0.888	2.776	
S10	1680	0.750	0.949	2.897	
S11	1680	0.728	0.949	2.897	
S12	1880	0.905	1.112	3.241	
S13	1370	1.413	0.655	2.362	
S14	1930	0.884	1.150	3.328	
S15	1600	0.833	0.879	2.759	
S16	1650	0.834	0.923	2.845	
S17	1910	0.913	1.134	3.293	
S18	1550	1.329	0.833	2.672	
S19	1350	1.637	0.634	2.328	

PCA of the normalized variables was performed to extract significant PCs and to further reduce the contribution of variables with minor significance; these PCs were subjected to varimax rotation with Kaiser Normalization generating VFs (Brumelis et al. 2000; Singh et al. 2004, 2005a, b; Love et al. 2004; Abdul-Wahab et al. 2005). The factor analysis can be shown by the following equation:

$$z_{ji} = a_{f_1} f_{1i} + a_{f_2} f_{2i} + a_{f_3} f_{3i} + \cdots \ldots\ldots\ldots\ldots\ldots \\ + a_{f_m} f_{mi} + e_{fi}$$

where

z = measured variable

a = factor loading

f = factor score

e = residual term accounting for errors or other sources of variation

i = sample number

m = total number of factors

CA

The purpose of CA is to identify groups or clusters of similar sites on the basis of similarities within a class and dissimilarities between different classes (Sparks 2000). CA is a group of multivariate techniques whose primary purpose is to assemble objects based on the characteristics they possess. CA classifies objects so that each object is similar to the others. The resulting clusters of objects should then exhibit high internal (within cluster) homogeneity and high external (between clusters) heterogeneity.

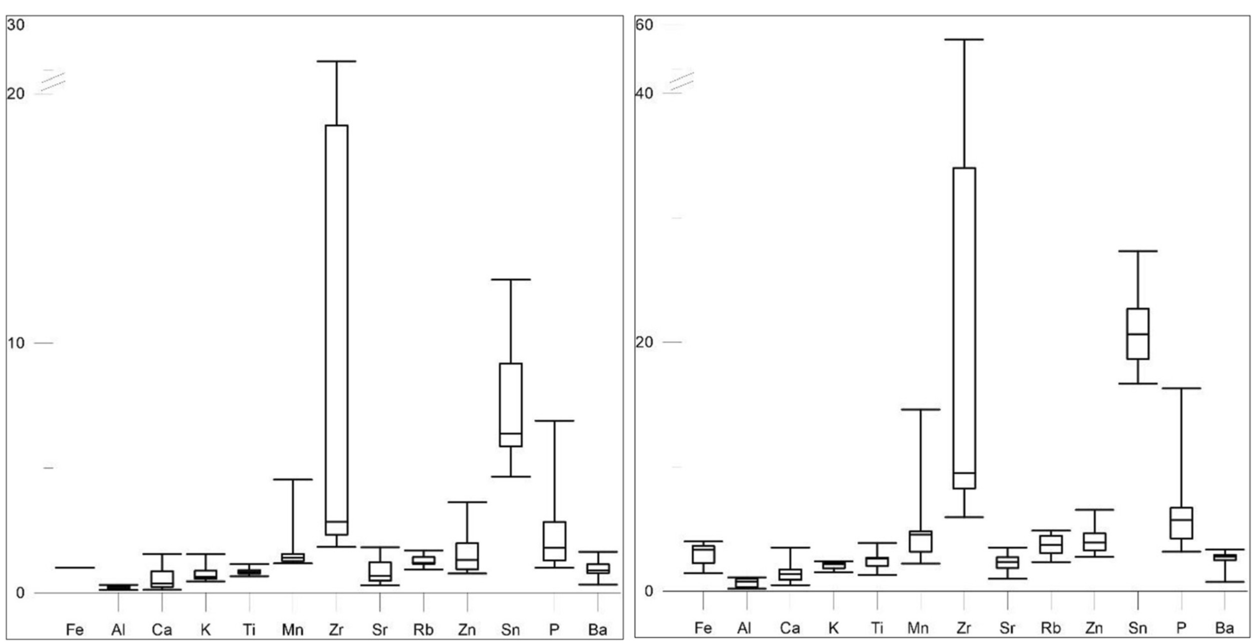

Fig. 3 Box-whisker plots of the EF (*left*) and CF (*right*) of metals in soils (the *whisker* shows the minimum and maximum values and the *line* of each plot is the median value)

In this study, hierarchical agglomerative CA was performed on the normalized dataset by means of the Ward's method, using squared Euclidean distances as a measure of similarity. CA was applied on experimental data standardized through z-scale transformation in order to avoid misclassification due to wide differences in data dimensionality (Liu et al. 2003).

Table 4 Matrix of three principal components

Elements	Component		
	PC1	PC2	PC3
Fe	**0.927**	−0.221	−0.059
Al	**0.789**	**−0.541**	−0.053
Ca	−0.355	**0.846**	−0.044
K	0.277	0.148	**0.853**
Ti	**0.840**	0.015	−0.287
Mn	0.507	0.004	**−0.663**
Zr	**−0.854**	0.175	−0.085
Sr	−0.189	**0.927**	0.237
Rb	**0.911**	0.211	0.174
Zn	−0.161	0.031	**0.745**
Sn	0.280	−0.185	**0.701**
P	0.174	**0.789**	−0.162
Ba	**0.600**	−0.122	0.172
Eigenvalue (total)	5.149	2.473	2.253
% of total variance	39.610	19.020	17.331
Cumulative % of variance	39.610	58.630	75.961

Moderate to strong loadings are in boldface

Inverse distance weighting

The factor scores from the R-mode PCA and I_{geo} values were used with ArcGIS 10.1 to determine the spatial variations of the dominant processes and soil pollution level using the inverse distance weighting (IDW) method. The IDW method estimates the values of an attribute at unsampled points using a linear combination of values at sampled points weighted by an inverse function of the distance from the point of interest to the sampled points. The weights can be expressed as follows:

$$\lambda_{\mathrm{i}} = \frac{1/{d_i}^{\mathrm{P}}}{\sum_{i=1}^{n} 1/{d_i}^{\mathrm{P}}}$$

where

d_i = the distance between x_0 and x_i

p = power parameter

n = the number of sampled points used for the estimation

The main factor affecting the accuracy of IDW is the value of the power parameter (Isaaks and Srivastava 1989). The most popular choice of p is 2, and the resulting method is often called inverse square distance.

Table 5 Component matrix showing three factor models for sampling sites

Elements	PC1	PC2	PC3
S1	**−1.112**	0.205	−0.218
S2	0.009	**−1.544**	−0.685
S3	**−1.046**	**1.295**	−0.125
S4	0.265	**−1.428**	**−2.137**
S5	0.228	**−1.310**	**−1.138**
S6	0.583	−0.505	0.256
S7	**−1.301**	0.151	−0.555
S8	0.031	0.496	**1.546**
S9	0.410	**−0.745**	**0.816**
S10	**1.192**	0.417	0.230
S11	**0.860**	−0.114	**0.992**
S12	**1.364**	**2.730**	**−1.984**
S13	**−1.183**	**1.002**	0.117
S14	**1.071**	0.337	**1.380**
S15	0.349	−0.050	−0.237
S16	**0.816**	−0.283	0.060
S17	**0.750**	−0.442	**0.952**
S18	**−1.536**	−0.157	0.194
S19	**−1.751**	−0.054	0.535

Moderate to strong factor loadings are boldface

Results and discussion

Pollution indices

The EF values for Fe is 1 in all sampling sites; Al ranges from 0.116 to 0.319; Ca from 0.135 to 1.549; K from 0.465 to 1.536; Ti from 0.656 to 1.143; Mn from 1.172 to 4.536; Zr from 1.832 to 25.955; Sr from 0.305 to 1.820; Rb from 0.928 to 1.686; Zn from 0.764 to 3.628; Sn from 4.655 to 12.544; P from 0.997 to 6.892; and Ba from 0.330 to 1.637 (Tables 2 and 3). The average order of the EF values for the metals is Zr (8.106) > Sn (7.590) > P (2.311) > Mn (1.625) > Zn (1.531) > Rb (1.256) > Fe (1) > Ba (0.948) > Sr (0.865) > Ti (0.840) > K (0.747) > Ca (0.561) > Al (0.218). The EF values between 0.05 and 1.5 indicate that the metal is entirely from crustal materials or natural processes; on the other hand, the EF values higher than 1.5 indicate that the sources are likely to be anthropogenic (Zhang and Liu 2002). According to Han et al. (2006), EF ≤2 suggests deficiency to minimal metal enrichment, whereas EF >2 suggests higher degrees of metal enrichment.

The I_{geo} brought in by Muller (1969) is used as a reference of calculating the level of metal pollution.

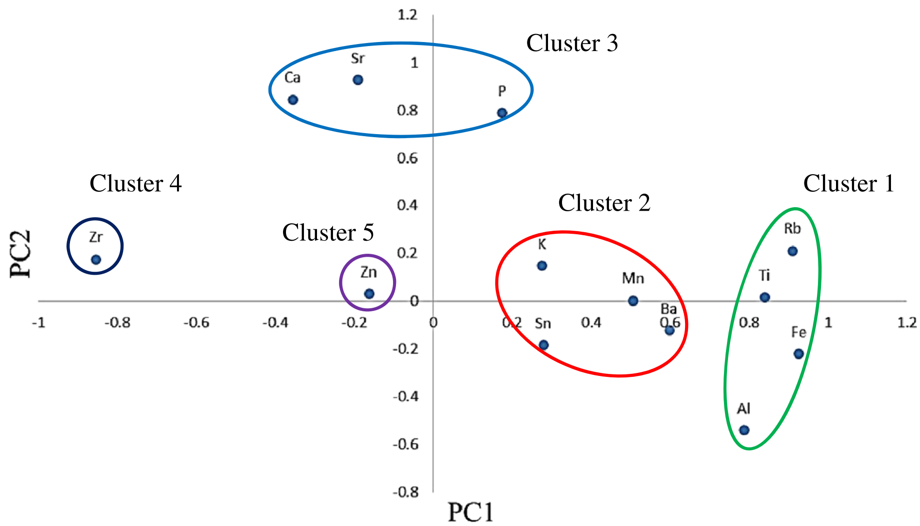

Fig. 4 Plots of PC1 vs. PC2 showing all metal dataset

From Tables 2 and 3, the I_{geo} value of Fe is 0.939 ± 0.444; K is 0.439 ± 0.190; Ti is 0.675 ± 0.375; Mn is 1.537 ± 0.667; Zr is 3.172 ± 1.096; Sr is 0.524 ± 0.509; Rb is 1.247 ± 0.337; Zn is 1.412 ± 0.358; Sn is 3.798 ± 0.201; P is 1.929 ± 0.604; and Ba is 0.788 ± 0.464. According to Table 1, overall sampling site is unpolluted by Al and Ca; unpolluted to moderately polluted by Fe, K, Ti, Sr, and Ba; moderately polluted by Mn, Rb, Zn, and P; and strongly polluted by Sn and Zr. The distributions of I_{geo} in different sites are shown in Fig. 2. The CFs of the metals range from 1.422 to 3.979 (Fe); 0.213 to 1.089 (Al); 0.489 to 3.484 (Ca); 1.496 to 2.372 (K); 1.287 to 3.870 (Ti); 2.200 to 14.588 (Mn); 5.938 to 56.750 (Zr); 0.980 to 3.500 (Sr); 2.321 to 4.857 (Rb); 2.737 to 6.526 (Zn); 16.667 to 27.333 (Sn); 3.157 to 16.286 (P); and 0.741 to 3.328 (Ba) (Tables 2 and 3). PLI calculated from CF depicts that the soils are strongly contaminated by Zr and Sn (Tables 2 and 3). Figure 3 shows the Box-whisker plots of Enrichment Factor (EF) and Contamination Factor (CF) of metals in soil of the area. CF, EF, and I_{geo} show minor similarity with Jayaprakash et al.'s (2009) study in the Indian coast area.

PCA and FA

Using varimax rotation with Kaiser Normalization, PCA was performed on the metal data maximizing the sum of the variance of the factor coefficients. This technique clusters variables into different groups. The PCA results obtained for the elements are shown in Table 4. Three principal components having eigenvalues greater than 1

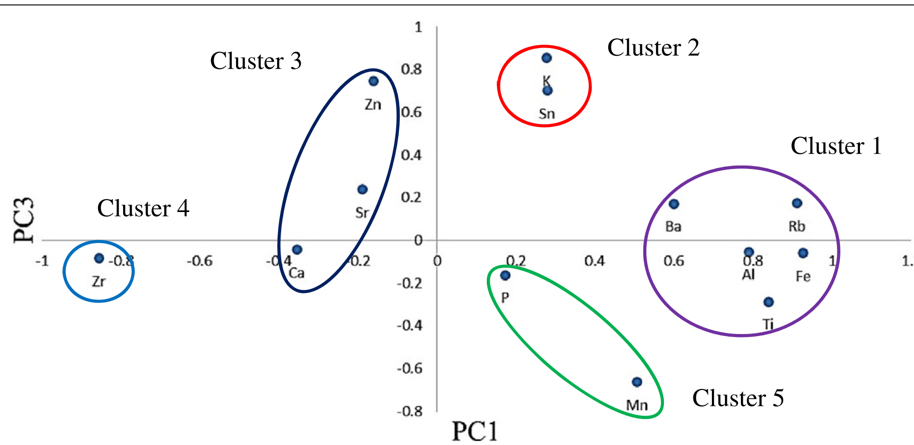

Fig. 5 Plots of PC1 vs. PC3 showing all metal dataset

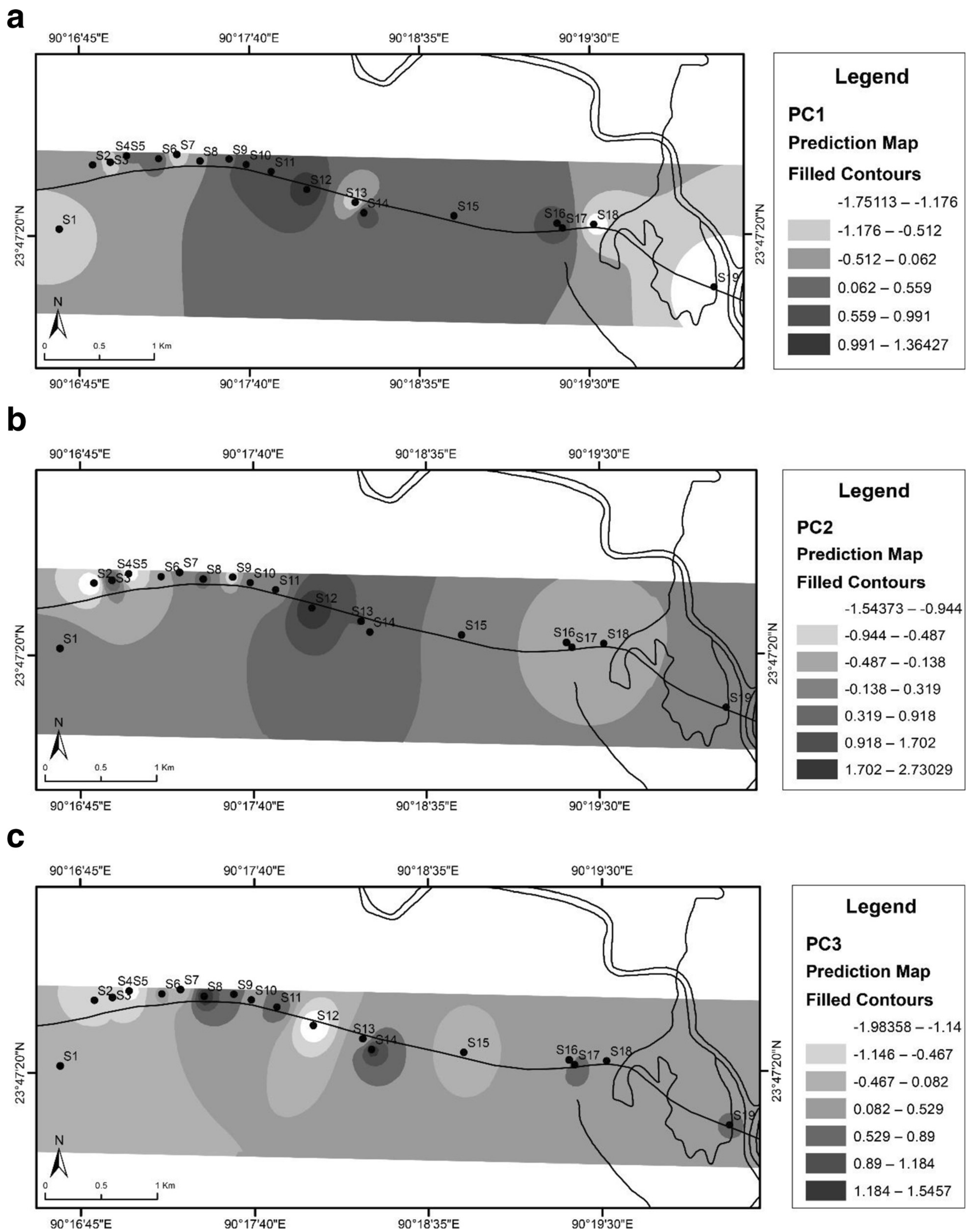

Fig. 6 Factor score map of Principle Components (PC1, PC2 and PC3) as **a** for PC1, **b** for PC2 and **c** for PC3

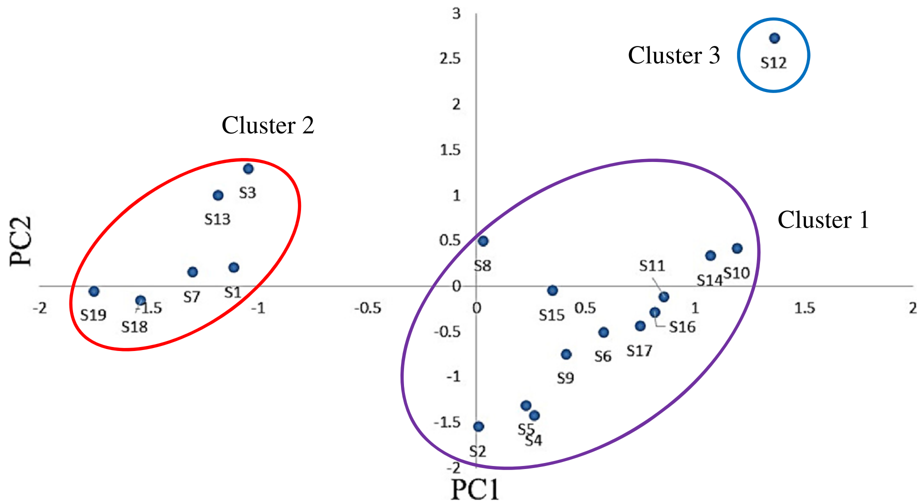

Fig. 7 PC1vs. PC2 plot of sampling site grouping

were considered. According to Liu et al. (2003), strong, moderate, and weak factor loadings range from >0.75, 0.75 to 0.5, and 0.5 to 0.3, respectively.

The first principal component (PC1) in the datasets explains 39.610 % of total variance and is strongly positively loaded with Fe, Al, Ti, and Rb and moderately positively loaded with Ba, indicating both natural and anthropogenic sources. The dominant factor loading of Fe in the first PC1 strongly suggests that the origin of Fe could be associated to the local emission sources such as metallurgical plant (Mmolawa et al. 2011). Al correlates with Fe in weathered materials and can be an indicator of mafic rocks. Anthropogenic sources of Ti and Rb include paint pigments and glass dust, but mainly natural sources are more important than anthropogenic sources (Reimann and de Caritat 1998). Major sources of Ba include manufacture of rubber, paper, fabrics, glass, plastics, and enamels. These parameters retain high positive scores in S10, S11, S12, S14, S16, and S17 and negative scores in S1, S3, S7, S13, S18, and S19 (Table 5).

The PC2 in the datasets explains 19.020 % of variance. PC2 is strongly positively loaded with Ca, Sr, and P and moderately negatively loaded with Al, indicating anthropogenic sources. The long-established agricultural practice and liming are the sources of Ca and P. Cement factories, fertilizers, and dust can also be regarded as anthropogenic sources of Ca. Sr can be released from industrial waste, disposal of coal ash, and incinerator ash

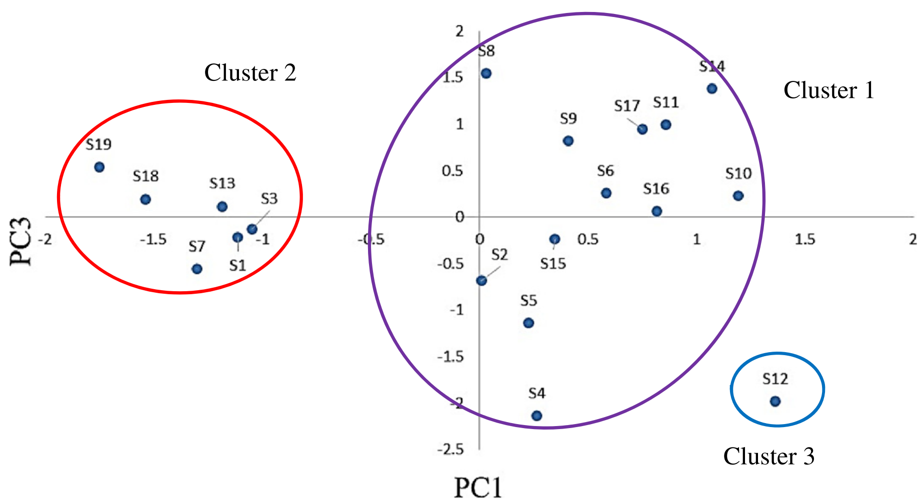

Fig. 8 PC1 vs. PC3 plot of sampling site grouping

(Reimann and de Caritat 1998). These parameters retain high positive scores in S3, S12, and S13 and negative scores in S2, S4, S5, and S9 (Table 5).

PC3 represents 17.331 % of variance and is positively loaded with K, Zn, and Sn and moderately negatively loaded with Mn, indicating anthropogenic sources. Zn is dispersed in the environment from high traffic density (tire wear particles) (Callender and Rice, 2000). High positive loading for K indicated their sources related with soil parent material (Ali and Malik, 2011). Zinc is readily adsorbed by clay minerals and carbonates (Krishna and Govit 2004). Possible reason for Zn concentration being higher is due to its association with sewage pollution (Muniz et al. 2003).

Sn is released from waste incineration and coal and wood combustion in the surrounding Brickfield area. These parameters retain high positive scores in S8, S9, S11, S14, and S17 and negative scores in S4, S5, and S12 (Table 5).

For all the elemental dataset, five clusters are found in the PC1 vs. PC2 plot (Fig. 4). Cluster 1 incorporates Rb, Ti, Fe, and Al, and cluster 2 consists of K, Mn, Ba, and Sn. Cluster 3 includes Ca, Sr, and P. Clusters 4 and 5 include Zr and Zn, respectively. For the PC1 vs. PC3 plot (Fig. 5), similarly five main clusters are obtained. Cluster 4 of both plots shows similar grouping.

Spatial similarities and site grouping

Using GIS, factor score maps were generated following the IDW method for three principal components. Interpolation surfaces are created using the coordination data and site-based factor scores. The power value was set to 2; standard neighborhood was used instead of smooth neighborhood, and sector type was sector 4 with 45° offset. Obtained interpolation surface explains three dominant processes in the study area. Figure 6a, b, and c represents factor score maps for PC1, PC2, and PC3, respectively.

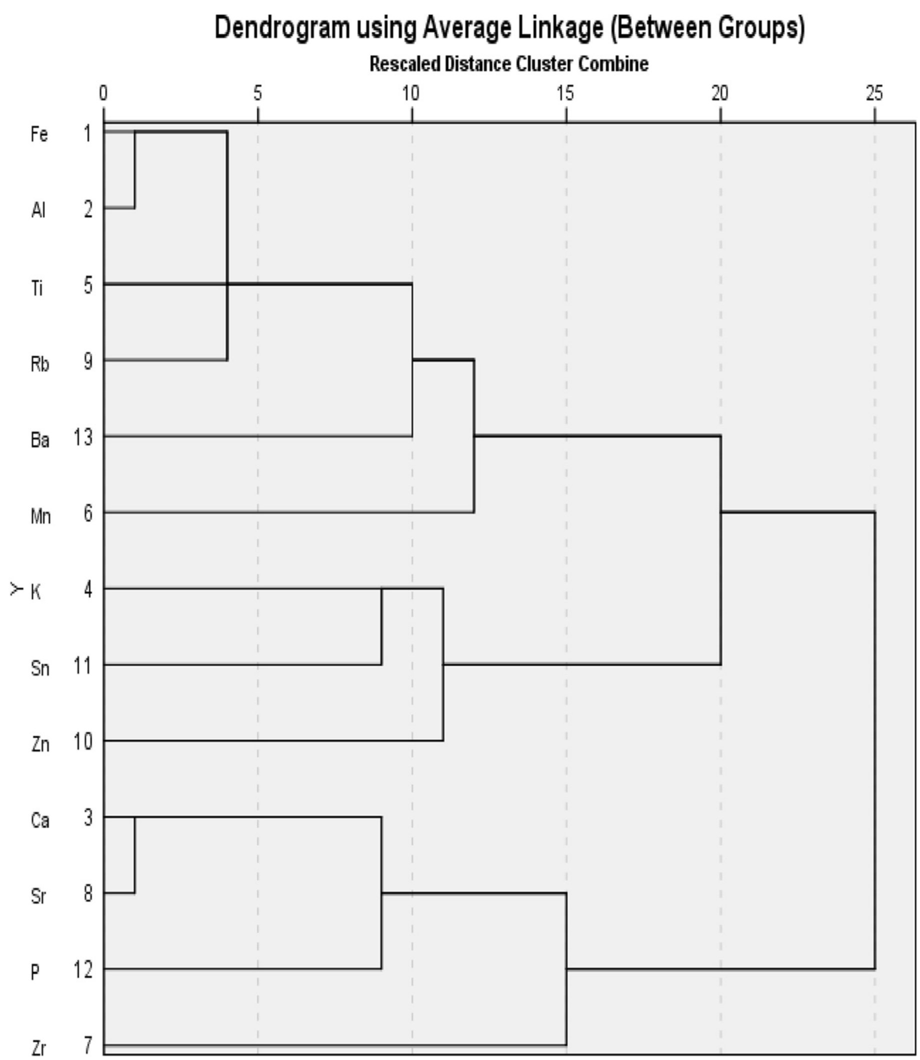

Fig. 9 Hierarchical clusters formed among analyzed metals

Within the −1.75113 to 1.36427 range of scores, about 49.501 % of the study area lies within positive factor loading, and about 50.498 % of the area lies within the range of −1.75113 to 0.062 (Fig. 6a). This indicates the processes related to PC1. The loading of PC1 increases from the western to the eastern parts then decreases at the eastern side of the study area. S12, S16, and S17 show the highest positive impact of PC1.

In Fig. 6b, the factor scores of PC2 range from −1.54373 to 2.73029. About 21.289 % of the study area bears positive factor loading, and about 78.710 % of the area has a loading in the range of −1.54373 to 0.319. The highest positive impact of PC2 occurs in S12 which is near the Amin Bazar landfill area.

The PC3 factor score map (Fig. 6c) ranges from −1.98358 to 1.5457. About 68.477 % of the area covers the positive factor loadings. The highest positive impact of PC3 occurs in S8 and S14.

In order to identify sample site clustering, factor scores obtained from PCA are used and PC1 vs. PC2 and PC1 vs. PC3 plots are generated (Figs. 7 and 8). On the PC1 vs. PC2 plot, three main clusters are obtained. Cluster 1 includes S2, S4, S5, S6, S8, S9, S10, S11, S14, S15, S16, and S17. Cluster 2 contains S1, S3, S7, S13, S18, and S19. Cluster 3 contains only S12. For the PC1 vs. PC3 plot (Fig. 7), three main clusters are obtained. All the three clusters are similar to PC1 vs. PC2.

CA

CA performed on the elemental data reveals three major clusters (Fig. 9). Cluster 1 comprises Fe, Al, Ti, Rb, Ba, and Mn. The interrelated association among these metals shows similar positive loadings in PC1. Cluster 2 includes K, Sn, and Zn. The interrelated association shows similar positive loadings in PC3. Cluster 3 contains Ca, Sr, P, and Zr, and its positive loadings are similar to PC2.

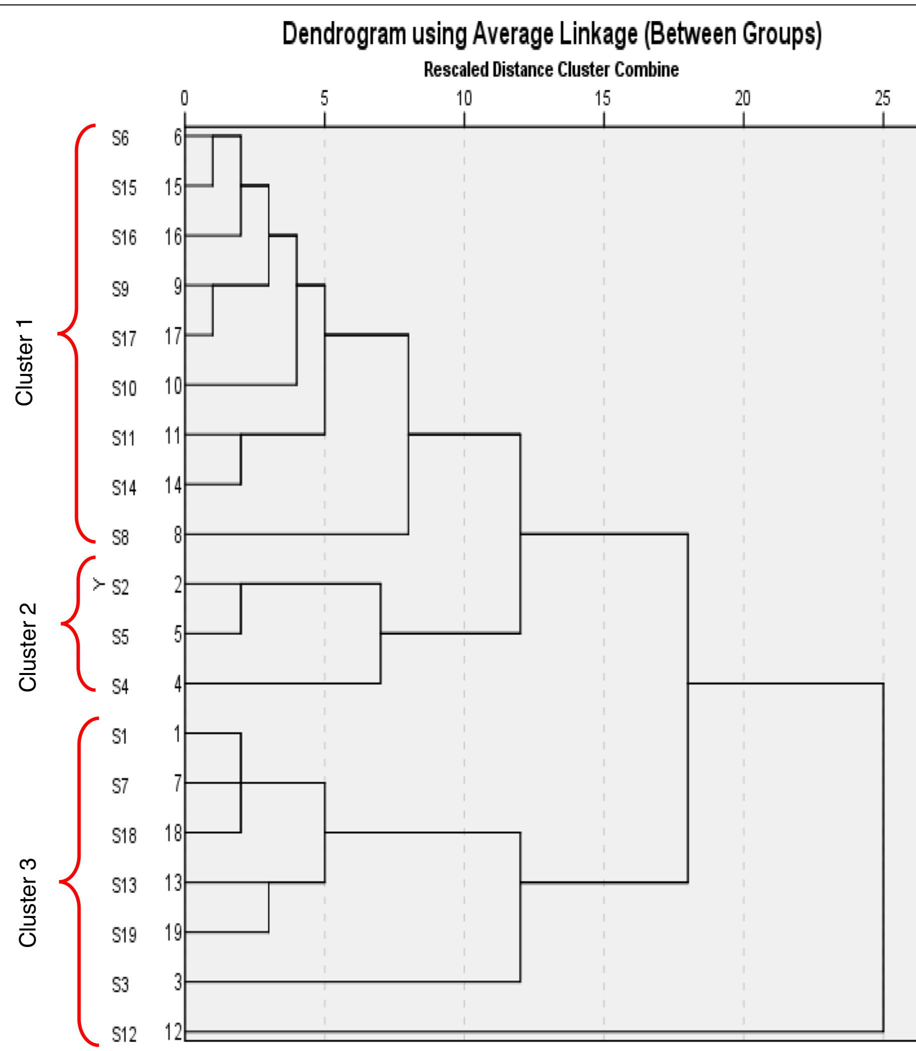

Fig. 10 Hierarchical clusters formed among sampling sites

Spatial similarities are discovered by R-mode CA. Nineteen sampling sites form three major clusters (Fig. 10). In cluster 1, the similarities among sampling sites S6, S8, S9, S10, S11, S14, S15, S16, and S17 are also observed in the factor score map of PC3. Cluster 2 represents the similarities among sampling sites S2, S4, and S5 which are noticed in the factor score map of PC2. The same observation is found in cluster 3 which represents the similarities among sampling sites S1, S3, S7, S12, S13, S18, and S19 in the factor score map of PC1.

Pearson's correlation matrix (CM)

Pearson's CM brings out some interconnection between the parameters (Table 5). Strong positive relationship is found among Fe-Al, Fe-Ti, Fe-Rb, Al-Ca, and Ca-Sr ($P < 0.01$) which is also observed in PC1. Negative correlation is found among Fe-Zr, Al-Ca, Al-Zr, and Zr-Rb ($P < 0.01$) which is described in PC1 and PC2.

Multivariate analysis and management implication

PCA, FA, and CA will be excellently used in future studies to find inter-parameter associations existing between different pollutants. This data-mining technique will further help in reducing the number of pollution parameters to be tested and subsequent cost of analysis. The result of this study supports the fact that multivariate statistical methods including CA and PCA/FA can be applied to interpret complex datasets of heavy metals in soil, understand spatial variation in of heavy metals along roadside areas, and identify latent pollution sources/factors. Therefore, this evaluation study can help managers identify the main sources of pollution in different regions so as to determine their priorities for pollution minimization and source reduction. Since multivariate statistical methods are easily applied to heavy metal data, using them can be a practical approach to environmental impact assessment. The Dhaka Aricha highway is a pollution hotspot, dispersing the toxic metals in the environment. For source identification, important heavy metals, and their hotspot location, we can easily use multivariate tools for pollution source zonation and to reveal the main harbor of contamination of heavy metals in this area. In this study, for PCA, FA, and CA metal datasets, three major principal components and three major clusters were formed. Major metals like Fe, Rb, Ti, and Al are found in PC1 and cluster 1. A quite similar pattern is also shown in PC2, cluster 2, and PC3, cluster 3. So we can easily identify the major metals in the study area and their sources. We can reduce their point and non-point sources of pollution and reduce their concentration in soil. Thus, we can easily manage or handle the pollution reduction strategy and also give priority to those sites where close monitoring is needed.

Conclusions

This work was undertaken to evaluate the surface soil state of the Hemayetpur-Gabtoli region. CF, EF, PLI, and I_{geo} indicated the pollution state and their associated anthropogenic sources. Zr and Sn show high loading, and Al and Ca show low pollution load in CF, EF, and I_{geo}. From PCA, three major principal components were extracted which perfectly reduced the data dimension and also indicated possible anthropogenic sources. These components explain 75.961 % of the total variance. From the factor score map, high positive loading is found near the Boilapur-Amin Bazar landfill site (PC1), near Boilapur (PC2), and near the Noyahati-Amin Bazar landfill site (PC3). CA formed three major clusters for both water parameters and sampling sites. This result regarding sources showed similarities among PCA and CA. The present investigation clearly indicates that the soils from freshwater reservoir are contaminated with some toxic heavy metals. Consequently, there is a dire need to reduce/regulate the anthropogenic sources of pollution in the study area.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

FA generate the idea and write the manuscript. ANMF supervised the work, MdTI analyzed the statistical part and helps in writing. NK, TAK, MdMR helped in laboratory analysis. ATMA supervises the laboratory works and helps in writing. All authors read and approved the final manuscript.

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