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ORIGINAL ARTICLE

Spatial distribution and source apportionment of heavy metals in soils of Gebeng industrial city, Malaysia

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Abstract Heavy metal concentrations were examined in 30 soil samples from Gebeng industrial city, Malaysia using inductively coupled plasma–mass spectrometry for As, Ba, Cd, Co, Cr, Cu, Ni, Pb and Zn, and direct mercury analyzer (DMA-80) for Hg. Multivariate statistical techniques including hierarchical cluster analysis (CA), principal component analysis (PCA), correlation analysis and analysis of variance were used to identify the spatial distribution and potential sources of heavy metals. The mean concentrations of heavy metals in the soil samples are in decreasing order as follows: Co > Ba > Zn > As > Pb > Cr > Cu > Ni > Hg > Cd. The Gebeng soils are characterized by high mean relative concentration of As, Ba, Cd, Co, Cr, Cu, Hg, Ni, Pb and Zn in the industrial zone (IZ) than the Kampung-Balok residential area (KB) and submerged area (SA), indicating inputs from industrial activities. Geochemical results suggested that Gebeng soils are primarily polluted by As, Co, Hg, Pb, and Cu subsequently derived from anthropogenic sources. PCA and CA in the heavy metals indicate both anthropogenic and natural origin. However, the geoaccumulation index and pollution load index further confirm the high contamination levels of the heavy metals in IZ and low to uncontaminated in KB and SA regions.

Keywords Heavy metal · Geoaccumulation index · Pollution load index · Principal components · Cluster analysis · Malaysia

Introduction

Metals are common natural components of all soils in the earth's crust (Kabata-Pendias and Mukherjee 2007). Heavy metal pollution of soil is a major environmental problem in the last few decades owing to the rapid increase in urbanization and industrialization (Gowd et al. 2010; Purushotham et al. 2012). Urban soil, which is strongly influenced by anthropogenic activities, differs largely from natural soils and receives a major proportion of trace metal emissions from industrial, commercial, and domestic activities (Chen et al. 1997; Bullock and Gregory 2009; Cheng et al. 2014). Heavy metals in soils largely depend on complex biological and geochemical cycles, which may be influenced by industrial activities, treatment of wastes, vehicles' exhaust and agricultural practices (Smith 2009; Fabietti et al. 2010; Ramos-Miras et al. 2011; D'Emilio et al. 2013). The heavy metals such as Cu, Fe, Zn, Mo and Mn are micronutrients and are considered to be essential to sustaining life in biological systems (Asrari 2014). Soil can act as a sink for both anthropogenic and naturally released heavy metals in surface environment (Birkefeld et al. 2005; Uria et al. 2008; Bai et al. 2011; Iqbal and Shah 2011; Solgi et al. 2012). Lalah et al. (2008) reported that spatial distribution and abundances of heavy metals in sediments are largely controlled by both anthropogenic and natural/geogenic factors. Natural factors include orogenesis, volcanism and crustal erosion, whereas anthropogenic factors include sewage discharge, industrial waste/waste water discharge and agricultural fertilizer leaching and so on,

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which directly influence the quality of the atmosphere, soil and water bodies, and threaten the ecosystems, food safety, health and life of organisms/animals and human beings (Cheng 2003; Oyedele et al. 2006; Bai et al. 2011; Ye et al. 2011; Dheeba and Sampathkumar 2012; Solgi et al. 2012; Li et al. 2013). In general, the concentration of metals in the soils ranges from traces levels to as high as 100,000 mg kg^{-1} depending on the type of element and its location (Blaylock and Huang 2000; Asrari 2014).

Gebeng is one of the largest industrial cities of peninsular Malaysia having a wide range of industries such as metal, chemical and petrochemical, polypropylene, gas and power, food and beverage, manufacturing and mining (Hossain et al. 2012; Nasly et al. 2013). These industries produce glasses, plastic containers, aluminum profiles, food processing, PVC pipes, furniture, paint, insecticide, disinfectant, herbicide, detergent, metal skeleton, car spare parts, electrical and electronics equipment, refrigerators and freezers, oven, ethanol, electroplating, etc. Heavy metals such as Cd, Cu, Cr, Ni, Pb, and Zn have been widely used in metal industries to produce alloys and steels (Li et al. 2009a, b). Therefore, the contamination of soils by heavy metals from industrial sources has become a potential threat to soil ecosystems, surface environments and human health in and around the city.

Spatial distribution and pollution of soils were measured by principal component analysis (PCA), hierarchical cluster analysis (CA), contamination factors (CF), geoaccumulation index (I_{geo}) and pollution load index (PLI). The present study assesses the heavy metal contents in soils from Gebeng industrial city, Pahang, Malaysia. Pollution indices, multivariate and geostatistical approaches (e.g., PCA, CA, etc.) were used to evaluate sources of heavy metals and degree of pollution materials in the studied soil samples.

Study area

Gebeng is a big city of Malaysia with rapid expansion of urbanization and industrialization over the last few decades. The Gebeng industrial city is located in the east coast of peninsular Malaysia, which lies between latitudes 3°55′0″N to 4°01′0″N and longitudes 103°22′0″E to

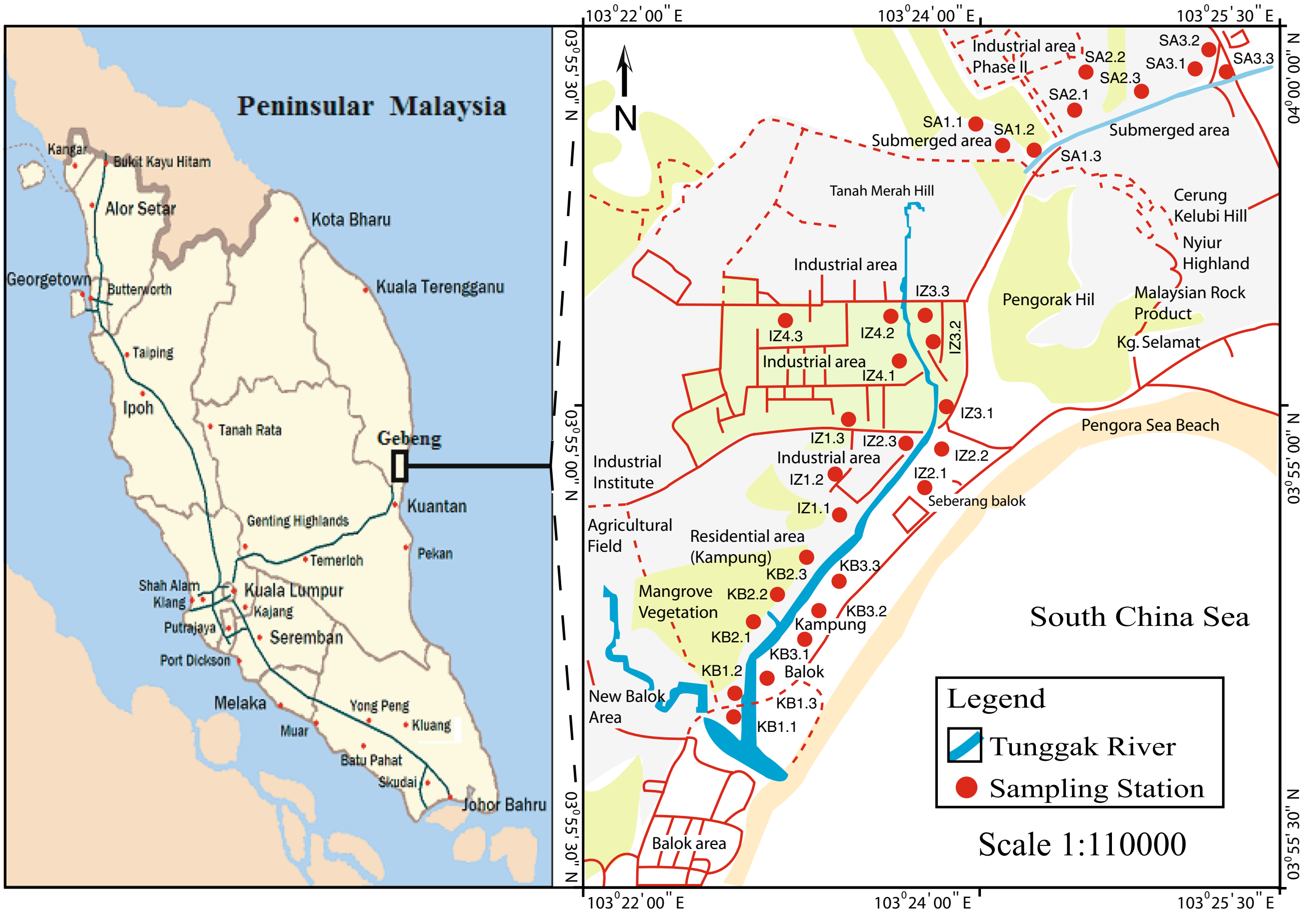

Fig. 1 Map of Gebeng industrial city and surrounding areas, showing location of sampling sites

103°27′0″E (Fig. 1). The city comprises an industrial park namely Gebeng industrial estate, sea-port and residential area (Kampung). A large number of industries are currently active in Gebeng city such as petrochemicals, chemicals, oil and gas, mining and metallurgy, food and wood processing, detergent and manufacturing industries (Hossain et al. 2013; Islam et al. 2013; Nasly et al. 2013).

Sediments in the study area were derived from Gebeng hilly region and were transported by the Balok and Tunggak rivers (Islam et al. 2013). Balok River flows from western part and Tunggak River from central part of the Gebeng industrial city (Fig. 1). Both rivers joined together near the Angler marine center at distance of ~6 km from the Gebeng city and plunges into the South China Sea (Islam et al. 2013; Nasly et al. 2013). The majority of soils in upland areas of Peninsular Malaysia are highly weathered (Shamshuddin and Anda 2008). These soils are developed from a range of parent materials, and are classified as Oxisols which are dominated by kaolinite, gibbsite, goethite and hematite in its clay fraction (Tessens and Shamshuddin 1983; Shamshuddin and Anda 2008). Soil samples in the study area contained relatively high concentration of clay, silt and organic carbon. The Gebeng basin fill contains ~38 m thick Quaternary sediments underlain by granitic and basaltic rocks of Cretaceous in age. Stratigraphically, the Gebeng sedimentary succession is subdivided into the Beruas and Simpang formations, in ascending order from oldest to youngest.

Materials and methods

Sample collection

Thirty soil samples were collected from three major sites namely Kampung-Balok residential area (KB, $n = 9$), industrial zone (IZ, $n = 12$) and submerged area (SA, $n = 9$) in the Gebeng industrial city, Malaysia. Each of the sample sites consisted of 3–4 sub-sites. Sampling sites along with sample numbers (within bracket) are: KB-1 (1.1, 1.2, 1.3), KB-2 (2.1, 2.2, 2.3) and KB-3 (3.1, 3.2, 3.3) are Kampung-Balok residential sites; IZ-1 (1.1, 1.2, 1.3), IZ-2 (2.1, 2.2, 2.3), IZ-3 (3.1, 3.2, 3.3) and IZ-4 (4.1, 4.2, 4.3) are industrial sites; SA-1 (1.1, 1.2, 1.3), SA-2 (2.1, 2.2, 2.3) and SA-3 (3.1, 3.2, 3.3) are submerged sites. Soil sample locations are shown in Fig. 1. The samples were collected using Dutch auger top soil (0–15 cm depth) and subsequently taken into ziploc polythene bags for transport and storage. The soil samples were air dried at room temperature and sieved through a <2 mm mesh, and then stored in sealed polythene bags until instrumental analysis.

Analytical methods

A total of 10 heavy metals (As, Ba, Cd, Co, Cr, Cu, Ni, Pb, Zn and Hg) were identified in the soil samples from the Gebeng industrial city, Malaysia. Nine heavy metals (As, Ba, Cd, Co, Cr, Cu, Ni, Pb and Zn) concentrations were identified by inductively coupled plasma–mass spectrometry (ICP–MS). Before analyzing the heavy metals by ICP–MS, the soil samples were first digested. Concentrated nitric acid (5 ml) was added with 0.5 g dried soil sample into a 50-ml Folin digestion tube. The mixture was heated at 130 °C for 15 h and then treated with hydrogen peroxide. After digestion, the sample was diluted to 50 ml with 2 % nitric acid. This solution was further diluted 1:9 (solution:nitric acid) for the analysis by ICP–MS. The analysis was done following the 'method 6020A: ICP-MS' (EPA 2007a). Hg was analyzed by 'method 7471B: Mercury in solid or semisolid' waste using a direct mercury analyzer (DMA-80) (EPA 2007b). Soil organic matter (OM) was determined by weight loss ignition method (Schulte and Hopkins 1996; Combs and Nathan 1998). Soil pH and electrical conductivity (EC) measurements were done by soil survey standard test method (Piper 1942; Rayment and Higginson 1992).

Geoaccumulation index (I_{geo})

The I_{geo} indexes allow the evaluation of contamination by correlating the obtained current concentration of metals with their pre-industrial concentrations. I_{geo} index for the metals is determined using the following equation (Muller 1969):

$$I_{geo} = \log_2 \frac{C_n}{1.5B_n}$$

where, C_n is the concentration of metals in soil samples and B_n is the geochemical background concentration of the metal (n). Factor (1.5) is the background matrix correction factor due to lithological variations. In the present study, I_{geo} index was calculated using the modified equation proposed by Loska et al. (2004). Where, C_n is the concentration of metals in soil samples and B_n is the geochemical background value in the earth's crust (Taylor and McLennan 1995). The I_{geo} index consists of seven grades/classes (Muller 1969; Bhuiyan et al. 2010; Solgi et al. 2012). Grade 0 (practically uncontaminated): $I_{\text{geo}} \leq 0$ (grade 0); Grade 1 (uncontaminated to moderately contaminated): $0 < I_{\text{geo}} < 1$; Grade 2 (moderately contaminated): $1 < I_{\text{geo}} < 2$; Grade 3 (moderately to highly contaminated): $2 < I_{\text{geo}} < 3$; Grade 4 (heavily contaminated): $3 < I_{\text{geo}} < 4$; Grade 5 (heavily to extremely contaminated): $4 < I_{\text{geo}} < 5$; Grade 6 (extremely contaminated): $I_{\text{geo}} > 5$. Notably, Grade 6 is an open class and comprises all the I_{geo} index values greater than Grade 5 (Muller 1969; Bhuiyan et al. 2010; Solgi et al. 2012).

Pollution load index (PLI)

The PLI is an empirical index that provides a simple and comparative way to evaluate the level of heavy metal pollution (Tomlinson et al. 1980; Usero et al. 2000; Bhuiyan et al. 2010; Bentum et al. 2011). The CF was developed by Muller (1969), and it has been used to calculate the PLI value. The CF ratio was estimated by dividing the concentration of each metal in the soil by the baseline/background value (Bhuiyan et al. 2010):

$$\mathrm{CF} = \frac{C_{\mathrm{metal}}}{C_{\mathrm{background\ value}}}$$

However, PLI is calculated using the equation:

$$\mathrm{PLI} = \sqrt{[n]}(CF_1 \times CF_2 \times CF_3 \times \cdots \times CF_n)$$

where, C_{metal} = metal concentration obtained from sample, $C_{\mathrm{background\ value}}$ = geochemical background/baseline value of the metal and n = number of metals. The $\mathrm{PLI} > 1$ indicates heavy metal pollution and $\mathrm{PLI} < 1$ indicates no pollution (Tomlinson et al. 1980; Harikumar et al. 2009).

Multivariate statistical analysis

Multivariate analyses of heavy metal contents in soils were carried out using Pearson's correlation analysis, factor analysis (FA) and CA by SPSS software package version 17.0 for windows. Pearson's correlation analysis is also used to measure correlations among elements and/or heavy metals (Simeonova and Simeonov 2006; Astel et al. 2008; Zhao et al. 2012; Li et al. 2013). PCA is commonly used to identify pollutant sources of heavy metals in both sediments and waters, and to differentiate between natural and anthropogenic source materials (Zhou et al. 2008; Bhuiyan et al. 2010; Lu et al. 2010; Li and Zhang 2010; Yuan et al. 2011; Chen et al. 2012; Li et al. 2013). FA is the component of PCA and is performed by Varimax rotation to minimize some complex variables to a few latent factors for analyzing relationship among the observed variables (Li et al. 2013). PCA was conducted using factor extraction after Varimax rotation with Kaiser Normalization with an eigenvalue >1 (Bengraine and Marhaba 2003). CA is used to evaluate similarity of monitoring stations with respect to the concentration of heavy metals in soils. CA was performed based on Ward's method, and the Euclidean distance was applied for calculating the distance between clusters of comparable metal contents (Bhuiyan et al. 2010). Finally, the obtained results from clustering analysis were presented in Dendrogram.

Results and discussion

Ten heavy metal concentrations (As, Ba, Cd, Co, Cr, Cu, Hg, Ni, Pb and Zn) together OM, pH and EC in the Gebeng soil samples are listed in Table 1. Heavy metal concentrations (ppm) were found in the range of 2.75–85.04 for As, 6.08–65.53 for Ba, 0.01–0.81 for Cd, <0.000012–1,015.17 for Co, 7.00–17.10 for Cr, 0.49–41.43 for Cu, 0.03–18.31 for Hg, 0.51–6.97 for Ni, 3.82–69.14 for Pb, and <0.000012–70.27 for Zn, respectively (Table 1). Mean pH value was relatively high in SA (6.12, 5.16–7.82), whereas quite low in IZ (5.83, 4.82–7.92) and KB (5.47, 3.55–7.54) sites. However, relatively high EC (up to 22,550 µS/cm) was recorded in KB site, inferring the influence of saline water intrusion from the South China Sea to the area (Hossain et al. 2012). The concentration of OM in IZ is relatively high (4.25–14.81 %) than that of SA (0.96–9.23 %) and KB (1.84–9.98 %). Spatial distributions of heavy metals show the increases in As, Ba, Cd, Co, Cr, Cu, Hg, Ni, Pb and Zn for IZ, and the concentrations of most metals in the SA and KB tend to have decreases, reflecting both natural and anthropogenic source material inputs (Pekey et al. 2004; Li et al. 2009a, b; Li and Zhang 2010). The mean concentrations of heavy metals in soil samples are in decreasing order as follows: Co > Ba > Zn > As > Pb > Cr > Cu > Ni > Hg > Cd (Table 1). The mean contents of heavy metals in IZ are relatively higher than in KB and SA, and values are in decreasing order as follows: Co > As > Zn > Ba > Pb > Cu > Cr > Ni > Hg > Cd, Ba > Cr > Pb > Zn > As > Cu > Ni > Hg > Cd > Co and Ba > Cr > Pb > Zn > As > Cu > Ni > Co > Hg > Cd, respectively. High concentration of Co (1,015.17 ppm) was recorded in the IZ sample, whereas very low in the SA (0.81 ppm) and KB (< 0.000012 ppm) samples. Similarly, mean values of As, Zn, Pb, Cu and Cr were also high in the IZ samples. Abundances of As, Co and Hg in the Gebeng soils fall within the standard limit of soil metals contamination in most of the developed countries considered to be contaminated (Chen 1998). However, high contents of these heavy metals in the soil samples indicate an anthropogenic source input to the investigated area. Abundances of Ba in the KB, IZ and SA sample range from 23.73 to 43.46, 19.58 to 60.75 and 6.08 to 65.53 ppm, respectively (Table 1). Ba is commonly associated with clay minerals due to weathering of felsic source materials (Hossain et al. 2010). High abundance of Ba in the IZ and SA soil samples suggests industrial effluent as well as clay mineral control.

The I_{geo} is widely used to evaluate the degree of metal contamination in geologic samples. The statistical values of I_{geo} in the soil samples are shown in Table 2. The mean I_{geo} values for Hg, As and Co are 2.96, 2.25 and 2.03, respectively, suggesting moderate to heavily

Table 1 Concentrations (ppm) of soil heavy metals with pH, EC (μS/cm) and OM (%) in the Gebeng industrial city, Malaysia

Sample	pH	EC	OM	Cr	Co	Ni	Cu	Zn	As	Cd	Ba	Pb	Hg
Kampung-Balok													
KB1.1	3.58	22,550	3.88	7.00	b.d.l.	0.94	0.72	7.29	2.90	0.04	37.38	6.00	0.12
KB1.2	3.80	4,515	1.84	7.34	b.d.l.	1.02	0.65	9.34	3.78	0.03	37.12	6.76	0.07
KB1.3	3.55	22,450	2.36	7.67	b.d.l.	0.90	0.87	8.69	3.31	0.04	38.31	5.46	0.14
KB2.1	5.83	140	1.89	9.31	b.d.l.	0.79	1.18	14.98	5.32	0.03	23.73	6.45	0.07
KB2.2	6.00	123	1.88	8.56	b.d.l.	0.71	0.49	8.94	4.80	0.01	25.49	6.47	0.03
KB2.3	7.54	955	2.52	8.04	b.d.l.	0.72	0.79	b.d.l.	3.68	0.01	43.46	9.18	0.06
KB3.1	6.48	1,317	9.98	10.32	b.d.l.	0.91	3.00	4.60	4.60	0.08	26.88	10.28	0.71
KB3.2	5.46	2,565	3.35	8.15	b.d.l.	1.56	2.01	4.65	4.65	0.05	32.18	9.08	0.09
KB3.3	7.03	286	2.63	11.85	b.d.l.	1.28	2.83	4.28	4.90	0.34	25.73	10.76	0.08
Max	7.54	22,550	9.98	11.85	b.d.l.	1.56	3.00	14.98	5.32	0.34	43.46	10.76	0.71
Min	3.55	123	1.84	7.00	b.d.l.	0.71	0.49	b.d.l.	2.90	0.01	23.73	5.46	0.03
Mean	5.47	6,100	3.37	8.69	b.d.l.	0.98	1.39	6.97	4.22	0.07	32.25	7.83	0.15
STD	1.51	9,403	2.57	1.56	0.00	0.28	0.97	4.24	0.82	0.10	7.09	1.99	0.21
Industrial zone													
IZ1.1	7.92	994	7.06	9.27	b.d.l.	3.38	21.92	40.97	41.64	0.38	45.93	49.25	3.08
IZ1.2	4.82	3,365	4.86	10.62	b.d.l.	4.55	37.96	55.91	64.88	0.39	46.77	61.89	2.05
IZ1.3	6.04	359	6.41	9.29	b.d.l.	4.34	29.06	55.72	53.69	0.39	60.75	69.14	0.15
IZ2.1	6.12	809	6.75	10.19	319.68	3.32	32.54	42.31	60.80	0.43	39.57	5.29	3.17
IZ2.2	5.70	369	4.25	11.17	341.49	3.96	5.73	22.35	73.28	0.17	19.58	6.34	0.87
IZ2.3	6.27	643	8.79	12.64	339.29	6.97	41.43	70.27	40.13	0.73	32.60	5.01	18.31
IZ3.1	6.09	258	6.03	10.82	1,015.17	2.89	6.80	49.14	7.77	0.26	49.23	6.48	0.48
IZ3.2	5.78	538	8.25	10.19	609.63	2.68	10.45	39.99	19.01	0.28	35.78	48.35	0.35
IZ3.3	5.85	427	6.89	10.87	802.40	3.01	24.42	44.87	50.52	0.41	32.66	48.08	0.35
IZ4.1	5.25	514	14.81	14.49	0.88	2.10	35.10	39.98	33.37	0.20	54.46	15.12	1.25
IZ4.2	5.34	721	8.79	7.37	0.69	1.30	38.86	39.98	85.04	0.81	22.57	38.73	0.65
IZ4.3	4.88	1,000	10.14	11.56	0.56	1.67	17.94	35.81	32.44	0.28	23.78	39.83	0.92
Max	7.92	3,365	14.81	14.49	1,015.17	6.97	41.43	70.27	85.04	0.81	60.75	69.14	18.31
Min	4.82	258	4.25	7.37	b.d.l.	1.30	5.73	22.35	7.77	0.17	19.58	5.01	0.15
Mean	5.83	833	7.75	10.71	285.82	3.35	25.18	44.78	46.88	0.39	38.64	32.79	2.64
STD	0.81	833	2.79	1.78	356.72	1.52	12.72	12.01	22.36	0.19	13.12	23.80	5.04
Submerged area													
SA1.1	5.49	262	9.23	12.10	0.04	1.16	1.52	4.87	2.75	0.05	24.43	6.52	0.32
SA1.2	5.76	593	5.87	14.38	0.27	1.85	1.94	7.44	3.17	0.05	16.26	4.87	0.13
SA1.3	5.16	233	6.78	12.39	0.75	1.14	1.43	6.36	3.25	0.04	20.25	5.82	0.32
SA2.1	5.51	138	3.14	17.10	b.d.l.	0.51	0.62	5.06	4.49	0.02	6.08	3.82	0.34
SA2.2	5.43	117	4.08	15.18	b.d.l.	0.55	0.69	4.24	4.28	0.03	6.13	4.61	0.20
SA2.3	5.17	89	3.20	11.94	0.20	0.88	1.64	5.70	3.42	0.05	13.94	4.10	0.23
SA3.1	6.98	1,482	0.96	14.17	0.24	0.64	0.86	4.52	3.70	0.02	15.23	4.06	0.06
SA3.2	7.79	847	2.27	10.70	0.81	1.41	0.95	1.98	8.28	0.20	65.53	12.12	0.14
SA3.2	7.82	814	2.84	11.89	0.36	0.85	2.09	7.44	4.39	0.06	47.77	6.33	0.11
Max	7.82	1,482	9.23	17.10	0.81	1.85	2.09	7.44	8.28	0.20	65.53	12.12	0.34
Min	5.16	89	0.96	10.70	b.d.l.	0.51	0.62	1.98	2.75	0.02	6.08	3.82	0.06
Mean	6.12	508	4.26	13.32	0.30	1.00	1.31	5.29	4.19	0.06	23.96	5.80	0.21
STD	1.10	470	2.57	2.02	0.30	0.44	0.54	1.71	1.65	0.06	19.95	2.57	0.10
Overall mean	5.81	2,316	5.39	10.89	114.42	1.93	10.88	21.59	21.27	0.20	32.32	17.21	1.16

b.d.l. below detection limits (<0.000012)

Table 2 Geoaccumulation index (I_{geo}) values of soil heavy metals in the Gebeng industrial city, Malaysia

Sample	Cr	Co	Ni	Cu	Zn	As	Cd	Ba	Pb	Hg
Kampung-Balok										
KB1.1	−2.02	−14.04	−3.46	−3.95	−2.68	0.25	−8.15	−3.09	−1.61	0.69
KB1.2	−1.97	−14.04	−3.38	−4.05	−2.43	0.52	−8.51	−3.10	−1.49	0.20
KB1.3	−1.92	−14.04	−3.51	−3.77	−2.51	0.39	−8.33	−3.07	−1.70	0.88
KB2.1	−1.73	−14.04	−3.63	−3.45	−1.96	0.86	−8.59	−3.55	−1.54	0.11
KB2.2	−1.81	−14.04	−3.75	−4.33	−2.48	0.76	−9.23	−3.48	−1.53	−0.61
KB2.3	−1.88	−14.04	−3.74	−3.86	−16.00	0.49	−9.30	−2.94	−1.18	−0.06
KB3.1	−1.63	−14.04	−3.50	−2.53	−3.14	0.71	−7.53	−3.42	−1.07	2.46
KB3.2	−1.86	−14.04	−2.96	−2.93	−3.13	0.73	−7.96	−3.24	−1.20	0.45
KB3.3	−1.49	−14.04	−3.15	−2.58	−3.21	0.78	−6.07	−3.47	−1.03	0.24
Max	−1.49	−14.04	−2.96	−2.53	−1.96	0.86	−6.07	−2.94	−1.03	2.46
Min	−2.02	−14.04	−3.75	−4.33	−16.00	0.25	−9.30	−3.55	−1.70	−0.61
Mean	−1.81	−14.04	−3.45	−3.49	−4.17	0.61	−8.19	−3.26	−1.37	0.48
STD	0.17	0.00	0.26	0.66	4.45	0.21	0.97	0.22	0.25	0.86
Industrial zone										
IZ1.1	−1.73	−14.04	−2.18	−0.54	−0.96	2.92	−5.95	−2.89	0.50	3.94
IZ1.2	−1.60	−14.04	−1.89	0.01	−0.64	3.36	−5.93	−2.87	0.72	3.53
IZ1.3	−1.73	−14.04	−1.93	−0.26	−0.65	3.17	−5.94	−2.61	0.83	0.92
IZ2.1	−1.64	3.06	−2.20	−0.14	−0.92	3.30	−5.84	−3.04	−1.73	3.97
IZ2.2	−1.55	3.13	−2.02	−1.88	−1.56	3.48	−6.75	−3.74	−1.55	2.67
IZ2.3	−1.42	3.12	−1.46	0.10	−0.42	2.88	−5.31	−3.23	−1.79	5.72
IZ3.1	−1.58	4.21	−2.34	−1.71	−0.77	1.24	−6.35	−2.82	−1.53	2.07
IZ3.2	−1.64	3.70	−2.42	−1.28	−0.98	2.13	−6.26	−3.14	0.48	1.78
IZ3.3	−1.57	3.98	−2.30	−0.43	−0.86	3.11	−5.89	−3.23	0.47	1.77
IZ4.1	−1.29	−2.83	−2.66	−0.07	−0.98	2.70	−6.59	−2.72	−0.69	3.04
IZ4.2	−1.96	−3.08	−3.14	0.04	−0.98	3.63	−5.20	−3.60	0.26	2.39
IZ4.3	−1.51	−3.29	−2.89	−0.74	−1.09	2.67	−6.26	−3.55	0.28	2.74
Max	−1.29	4.21	−1.46	0.10	−0.42	3.63	−5.20	−2.61	0.83	5.72
Min	−1.96	−14.04	−3.14	−1.88	−1.56	1.24	−6.75	−3.74	−1.79	0.92
Mean	−1.60	−2.51	−2.29	−0.57	−0.90	2.88	−6.02	−3.12	−0.31	2.88
STD	0.17	7.51	0.46	0.69	0.28	0.66	0.46	0.36	1.06	1.28
Submerged area										
SA1.1	−1.47	−5.91	−3.26	−3.20	−3.08	0.20	−8.07	−3.52	−1.53	1.68
SA1.2	−1.30	−4.03	−2.78	−2.96	−2.66	0.34	−8.02	−3.93	−1.82	0.80
SA1.3	−1.44	−3.00	−3.27	−3.27	−2.82	0.37	−8.28	−3.71	−1.64	1.67
SA2.1	−1.12	−14.04	−4.07	−4.10	−3.05	0.69	−8.77	−4.91	−2.06	1.74
SA2.2	−1.24	−14.04	−4.00	−4.00	−3.22	0.64	−8.62	−4.90	−1.87	1.20
SA2.3	−1.48	−4.33	−3.53	−3.13	−2.93	0.42	−8.00	−4.08	−1.99	1.34
SA3.1	−1.31	−4.13	−3.84	−3.77	−3.16	0.50	−8.86	−3.99	−2.00	−0.06
SA3.2	−1.59	−2.92	−3.06	−3.68	−3.98	1.30	−6.60	−2.53	−0.91	0.86
SA3.3	−1.49	−3.72	−3.56	−2.89	−2.66	0.67	−7.80	−2.85	−1.56	0.61
Max	−1.12	−2.92	−2.78	−2.89	−2.66	1.30	−6.60	−2.53	−0.91	1.74
Min	−1.59	−14.04	−4.07	−4.10	−3.98	0.20	−8.86	−4.91	−2.06	−0.06
Mean	−1.38	−6.24	−3.49	−3.44	−3.06	0.57	−8.11	−3.82	−1.71	1.09
STD	0.15	4.51	0.44	0.45	0.40	0.32	0.68	0.80	0.36	0.60
Overall mean	−1.57	2.03	−2.74	−1.24	−1.60	2.25	−6.62	−6.62	−0.56	2.96

contamination of the metals (Muller 1969). Consequently, mean I_{geo} values are <0 for Pb (−0.56), Cu (−1.24), Cr (−1.57), Zn (−1.60), Ni (−2.74), Cd (−6.62) and Ba (−6.62) reflecting that investigated soil samples are practically uncontaminated/less contaminated. However, group-wise mean I_{geo} values for Hg (2.88) and As (2.88) in

Table 3 Contamination factors (CFs) and pollution load indices (PLIs) of soil heavy metals in the Gebeng industrial city, Malaysia

Sample	CFs										PLI
	Cr	Co	Ni	Cu	Zn	As	Cd	Ba	Pb	Hg	
Kampung-Balok											
KB1.1	0.20	b.d.l.	0.05	0.03	0.10	1.93	0.0004	0.07	0.30	2.99	0.03
KB1.2	0.21	b.d.l.	0.05	0.03	0.13	2.52	0.0003	0.07	0.34	1.82	0.29
KB1.3	0.22	b.d.l.	0.05	0.03	0.12	2.21	0.0004	0.07	0.27	3.61	0.30
KB2.1	0.27	b.d.l.	0.04	0.05	0.21	3.55	0.0003	0.04	0.32	1.68	0.35
KB2.2	0.24	b.d.l.	0.04	0.02	0.13	3.20	0.0001	0.05	0.32	0.81	0.25
KB2.3	0.23	b.d.l.	0.04	0.03	0.00	2.45	0.0001	0.08	0.46	1.41	0.08
KB3.1	0.29	b.d.l.	0.05	0.12	0.06	3.07	0.0008	0.05	0.51	17.64	0.41
KB3.2	0.23	b.d.l.	0.08	0.08	0.07	3.10	0.0005	0.06	0.45	2.34	0.36
KB3.3	0.34	b.d.l.	0.06	0.11	0.06	3.27	0.0035	0.05	0.54	1.91	0.34
Max	0.34	b.d.l.	0.08	0.12	0.21	3.55	0.0035	0.08	0.54	17.64	0.56
Min	0.20	b.d.l.	0.04	0.02	0.00	1.93	0.0001	0.04	0.27	0.81	0.06
Mean	0.25	b.d.l.	0.05	0.06	0.10	2.81	0.0007	0.06	0.39	3.80	0.36
STD	0.04	0.00	0.01	0.04	0.06	0.55	0.0010	0.01	0.10	5.25	0.15
Industrial zone											
IZ1.1	0.26	b.d.l.	0.17	0.88	0.58	27.76	0.0039	0.08	2.46	77.05	1.26
IZ1.2	0.30	b.d.l.	0.23	1.52	0.79	43.25	0.0040	0.09	3.09	51.35	1.47
IZ1.3	0.27	b.d.l.	0.22	1.16	0.78	35.79	0.0039	0.11	3.46	3.75	1.10
IZ2.1	0.29	31.968	0.17	1.30	0.60	40.53	0.0043	0.07	0.26	79.23	1.09
IZ2.2	0.32	34.149	0.20	0.23	0.31	48.85	0.0018	0.04	0.32	21.70	0.75
IZ2.3	0.36	33.929	0.35	1.66	0.99	26.75	0.0074	0.06	0.25	457.82	1.44
IZ3.1	0.31	101.517	0.14	0.27	0.69	5.18	0.0026	0.09	0.32	11.90	0.66
IZ3.2	0.29	60.963	0.13	0.42	0.56	12.67	0.0029	0.07	2.42	8.85	0.83
IZ3.3	0.31	80.240	0.15	0.98	0.63	33.68	0.0042	0.06	2.40	8.79	1.02
IZ4.1	0.41	0.088	0.10	1.40	0.56	22.25	0.0021	0.10	0.76	31.35	1.06
IZ4.2	0.21	0.069	0.06	1.55	0.56	56.69	0.0083	0.04	1.94	16.37	0.99
IZ4.3	0.33	0.056	0.08	0.72	0.50	21.62	0.0029	0.04	1.99	23.12	0.92
Max	0.41	101.517	0.35	1.66	0.99	56.69	0.0083	0.11	3.46	457.82	2.17
Min	0.21	b.d.l.	0.06	0.23	0.31	5.18	0.0018	0.04	0.25	3.75	0.42
Mean	0.31	28.582	0.17	1.01	0.63	31.25	0.0040	0.07	1.64	65.94	1.23
STD	0.05	35.67	0.08	0.51	0.17	14.91	0.0020	0.02	1.19	126.07	0.67
Submerged area											
SA1.1	0.35	0.004	0.06	0.06	0.07	1.83	0.0005	0.04	0.33	8.03	0.33
SA1.2	0.41	0.027	0.09	0.08	0.10	2.11	0.0005	0.03	0.24	3.34	0.33
SA1.3	0.35	0.075	0.06	0.06	0.09	2.17	0.0004	0.04	0.29	7.97	0.34
SA2.1	0.49	b.d.l.	0.03	0.02	0.07	2.99	0.0002	0.01	0.19	8.55	0.26
SA2.2	0.43	b.d.l.	0.03	0.03	0.06	2.85	0.0003	0.01	0.23	5.00	0.24
SA2.3	0.34	0.020	0.04	0.07	0.08	2.28	0.0005	0.03	0.20	5.73	0.30
SA3.1	0.40	0.024	0.03	0.03	0.06	2.47	0.0002	0.03	0.20	1.42	0.24
SA3.2	0.31	0.081	0.07	0.04	0.03	5.52	0.0020	0.12	0.61	3.53	0.36
SA3.3	0.34	0.036	0.04	0.08	0.10	2.92	0.0006	0.09	0.32	2.75	0.35
Max	0.49	0.081	0.09	0.08	0.10	5.52	0.0020	0.12	0.61	8.55	0.52
Min	0.31	b.d.l.	0.03	0.02	0.03	1.83	0.0002	0.01	0.19	1.42	0.18
Mean	0.38	0.030	0.05	0.05	0.07	2.79	0.0006	0.04	0.29	5.14	0.33
STD	0.06	0.030	0.02	0.02	0.02	1.10	0.0006	0.04	0.13	2.59	0.16
Overall mean	0.31	11.44	0.10	0.44	0.30	14.18	0.0020	0.06	0.86	29.06	0.78

b.d.l. below detection limits (<0.000012) the IZ soils are relatively higher than KB and SA soils (Table 2), demonstrating that IZ samples are highly contaminated due to anthropogenic source materials input, confirmed by high industrial activity of the area. The mean I_{geo} values for Ba, Cd, Cr, Cu, Ni and Zn are all < 0, therefore assume to be relatively very low polluted regions by the metals (Muller 1969), and consistent to predominantly natural sources.

The CF and PLI are widely used to evaluate the degree of heavy metal pollution in the soils (Bhuiyan et al. 2010). The calculated CF and PLI are listed in Table 3. The CF values for As (1.83–56.69) and Hg (0.81–457.82) are relatively high in the samples studied, indicating that the Gebeng soils are highly polluted by these two metals. The mean CF values for the metals in the IZ follow the decreasing in order Hg > As > Co > Pb > Cu > Zn > Cr > Ni > Ba > Cd. However, incredibly high mean PLI value (1.23) in the IZ is ascribed to be polluted. Relatively low CF (< 1) and low PLI (< 0.1) in the KB and SA suggest less polluted and/or unpolluted (Muller 1969; Tomlinson et al. 1980; Harikumar et al. 2009). This result is consistent with the study of Bhuiyan et al. (2010), Jordanova et al. (2013) and Banerjee and Gupta (2013), and stated that soil pollution could have been responsible for high influx of heavy metals in the soils due to large-scale industrial activities.

The hierarchical clustering was carried out in standardized data applying Ward's method (Gotelli and Ellison 2004; Li et al. 2013), and the squared Euclidean distance as a similarity measure (Bhuiyan et al. 2010; Li et al. 2013). CA is commonly used to assess metal variables to show a spatial sampling strategy (Li and Zhang 2010; Li et al. 2013). However, CA performed by sampling sites was also organized and the dendrogram obtained showed five statistically significant clusters, i.e., cluster 1 (KB1.2, KB2.1–2.3, KB3.2 and SA3.2–3.3), cluster 2 (SA1.1–1.3, SA2.1–2.3, KB3.1, KB3.3 and SA3.1), cluster 3 (KB1.1 and KB1.3), cluster 4 (IZ1.1–1.3, IZ2.1–2.2, IZ3.1–3.3 and IZ4.1–4.3) and cluster 5 (IZ2.3) (Fig. 2). The clusters show variable degree of pollutions derived from anthropogenic sources. Most of the sampling sites in the KB and SA belonged to the clusters 1, 2 and 3 corresponding to relatively lower pollution regions and/or uncontaminated. All samples in the IZ belonged to the cluster 4, except sample site IZ2.3 which also belonged to the cluster 5. The clusters 4 and 5, which contain relatively high Co, As, Zn, Ba, Pb, Cu, Cr, Ni, Hg and Cd ascribed to be highly polluted confirming that these heavy metals were probably derived from industrial effluents.

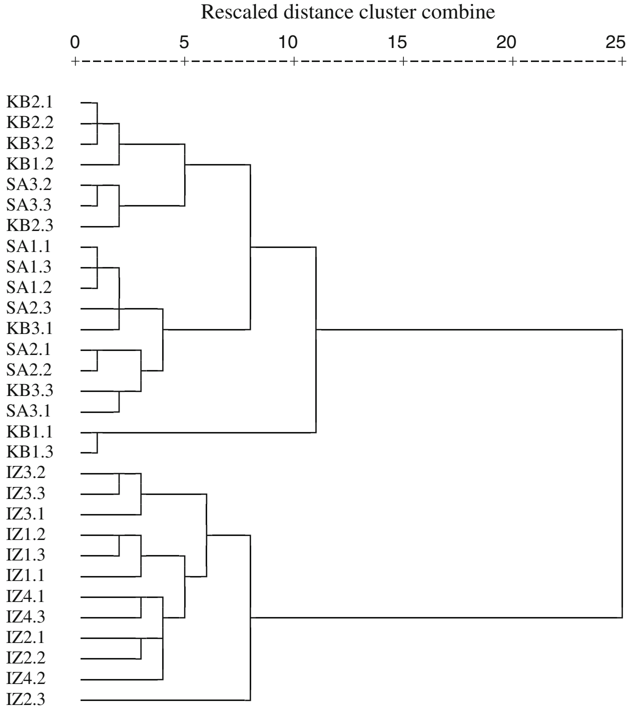

Fig. 2 Dendrogram showing clustering of sampling sites according to Ward's method using squared Euclidean distance

Pearson correlation coefficient of the heavy metals and p values (probability of no correlation) for statistical hypothesis testing are listed in Table 4. Zn shows a marked positive correlation with Cu ($r = 0.89$, $p = < 0.01$), Ni ($r = 0.86$, $p = < 0.01$) and Cd ($r = 0.81$, $p = < 0.01$). Similarly, As shows strong positive correlation with Cu ($r = 0.83$, $p = < 0.01$), Cd ($r = 0.79$, $p = < 0.01$) and Zn ($r = 0.74$, $p = < 0.01$), and weak positive correlation with Ni ($r = 0.66$, $p = < 0.01$) and Pb ($r = 0.61$, $p = < 0.01$). Cd also shows marked positive correlation with Cu ($r = 0.87$, $p = < 0.01$). Cd is closely associated with industrial sources (Yang et al. 2011). These positive correlations indicate that the contents of heavy metals in the Gebeng soils probably originated from similar source minerals. OM is positively correlated with Cu ($r = 0.61$, $p = < 0.01$), Zn ($r = 0.54$, $p = < 0.01$), Cd ($r = 0.46$, $p = < 0.01$), As ($r = 0.41$, $p = < 0.01$) and Ni ($r = 0.37$, $p = < 0.01$) suggesting that these elements are largely controlled by soil OM (Gao et al. 1997; Guo et al. 2005; Zhang et al. 2009). Tume et al. (2011) reported that soil OM positively correlated with Cr, Ni, Pb and Zn, implying that OM has a high adsorption capacity towards these heavy metals (Yin et al. 2002; Quenea et al. 2009). Dragović et al. (2008) reported that heavy metals (Cd, Cr, Cu, Ni, Pb and Zn) are positively correlated with OM inferring a common affinity for clay minerals. EC negatively correlated with Cr ($r = -0.44$, $p = < 0.01$) suggests a low amount of soluble salts in aqueous solution of the soil (Tume et al. 2011). Ba shows a negative correlation with Cr ($r = -0.42$, $p = < 0.05$) suggesting that Ba may be originated from different sources. Ba is

Table 4 Pearson correlation matrix for heavy metal concentration in soils from the Gebeng industrial city, Malaysia

Parameter	pH	EC	OM	As	Ba	Cd	Co	Cr	Cu	Hg	Ni	Pb	Zn
pH	1												
EC	**−0.58****	1											
OM	−0.08	−0.22	1										
As	−0.01	−0.18	**0.41***	1									
Ba	0.24	0.14	0.15	0.20	1								
Cd	0.14	−0.20	**0.46****	**0.79****	0.26	1							
Co	0.06	−0.15	0.20	0.24	0.15	0.33	1						
Cr	0.19	**−0.44***	0.19	−0.15	**−0.42***	−0.14	−0.001	1					
Cu	−0.01	−0.18	**0.61****	**0.83****	0.35	**0.87****	0.20	−0.04	1				
Hg	0.11	−0.08	0.29	0.28	0.06	**0.57****	0.18	0.12	**0.54****	1			
Ni	0.09	−0.17	**0.37***	**0.66****	0.36	**0.70****	**0.44***	−0.004	**0.74****	**0.72****	1		
Pb	0.04	−0.13	0.34	**0.61****	**0.39***	**0.55****	0.14	−0.23	**0.60****	−0.03	**0.46****	1	
Zn	−0.02	−0.17	**0.54****	**0.74****	**0.39***	**0.81****	**0.52****	−0.07	**0.89****	**0.56****	**0.86****	**0.64****	1

Bold values represent correlation with significance

* Significance at the 0.05 probability level ($p < 0.05$)

** Significance at the 0.01 probability level ($p < 0.01$)

Table 5 Varimax rotated principal component analysis for heavy metals in soils from the Gebeng industrial city, Malaysia

Parameter	Principal component			
	PC1	PC2	PC3	PC4
pH	−0.114	0.129	0.143	0.915
EC	−0.246	−0.017	0.362	−0.812
OM	0.683	0.151	−0.27	−0.013
As	0.841	0.226	0.125	0.01
Ba	0.197	0.19	0.788	0.172
Cd	0.736	0.492	0.127	0.087
Co	0.127	0.537	0.12	0.134
Cr	−0.033	0.068	−0.801	0.338
Cu	0.859	0.412	0.062	−0.005
Hg	0.163	0.88	−0.171	−0.036
Ni	0.533	0.749	0.13	0.073
Pb	0.801	−0.122	0.371	0.12
Zn	0.759	0.573	0.157	0.022
Eigenvalue	5.604	1.947	1.371	1.131
Total variance (%)	43.108	14.976	10.545	8.703
Cumulative variance (%)	43.108	58.084	68.63	77.333

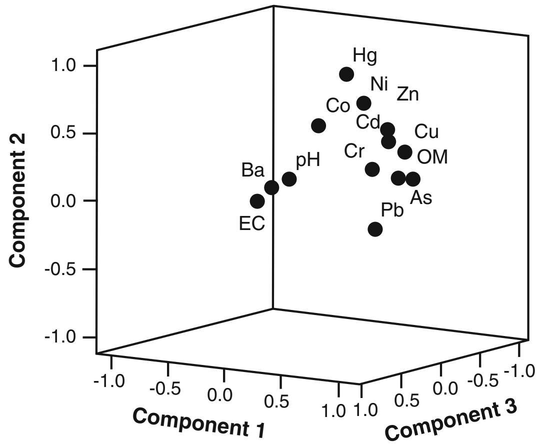

Fig. 3 Component plot in rotated space for heavy metals (factor loadings, factor 1 vs. factor 2 vs. factor 3, rotation: varimax normalized, extraction: principal components) in soils from the Gebeng industrial city, Malaysia

strongly associated with clays (Kabata-Pendias and Mukherjee 2007). The high relative abundance of Ba in the IZ samples implies an anthropogenic addition associated with natural sources.

PCA is widely used as a tool for evaluation of metal contamination as well as source identification in soils (Rubio et al. 2000; Dragović et al. 2008; Franco-Uría et al. 2009; Chabukdhara and Nema 2013). PCA and CA distinguish factor of anthropogenic and natural sources of heavy metals. Varimax rotation with Kaiser Normalization is applied to maximize the sum of the variance of the factor coefficients (Gotelli and Ellison 2004). Four principal components (PCs) with eigenvalues (>1) were extracted which explain 77.33 % of the total variance (Table 5). The scores and loadings of the first three PCs for the heavy metals are shown in Fig. 3. PC1 is positively loaded on OM, As, Cd, Cu, Ni, Pb, and Zn, which explain 43.11 % of the total variance. Based on factor loading classification (Liu et al. 2003), PC1 exhibits high loading for As, Cu, Pb and Zn, whereas moderate loading for OM, Cd and Ni. These metals are positively correlated with each other (Table 4) and subsequently belonged to the cluster 4

(Fig. 2), pointing to anthropogenic source affinity such as industrial processes. However, frequently adding phosphatic fertilizers to the agricultural soils has resulted in a concomitant increase in As, Cu, Cd, and Zn (Zarcinas et al. 2004). The contamination of soils occurs mainly in IZ zone where factories are located (Fig. 1). Similar results for source identification of soils were also reported by Zhou et al. (2007) and Banerjee and Gupta (2013). A high loading of Hg and moderate loading of Co, Ni, and Zn on PC2 accounting for 14.98 % of the total variance indicating the possibly influence from both industrial activity and agricultural runoff (Li et al. 2013). PC3 shows high positive loading of Ba and negative loading of Cr and accounts for 10.55 % of the total variance, suggesting a mixed source from both natural and anthropogenic inputs. PC4 accounts for 8.70 % of the total variance with marked positive loading on pH and negative loading on EC. This inverse relation is ascribed to saline water intrusion in the KB residential areas and pH loading. Bhuiyan et al. (2010) presented that PC4 is positively loaded with both pH and EC and negatively loaded with Ca, indicating agricultural influence which release some ionic substances from chemical fertilizer to the soils. In the KB area, agricultural field is located at the western part and South China Sea in the eastern part (Fig. 1). The influx of agricultural effluents and sea water intrusion in the KB region could be changed the pH and EC values to the soils (Bhuiyan et al. 2010; Hossain et al. 2012).

Conclusions

Heavy metal concentrations (As, Ba, Cd, Co, Cr, Cu, Hg, Ni, Pb, and Zn) were examined in 30 soil samples from Gebeng industrial city, Pahang, Malaysia. The mean concentrations of heavy metals in the soil samples are in decreasing order as follows: Co > Ba > Zn > As > Pb > Cr > Cu > Ni > Hg > Cd. The Gebeng soils are characterized by high mean relative concentration of As, Ba, Cd, Co, Cr, Cu, Hg, Ni, Pb and Zn in the IZ than the KB and SA, indicating inputs from anthropogenic sources. The I_{geo} and PLI of the heavy metals show that the IZ site is highly polluted and low to uncontaminated in the KB and SA sites. PCA, CA and correlation matrix suggest that the soils are mainly polluted by As, Co, Hg, Pb, and Cu. However, high abundances of these heavy metals in the studied soil samples reflect an influx from anthropogenic sources.

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