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Defining the geochemical baseline: a case of Hong Kong soils

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Abstract A geochemical baseline provides the means to distinguish between the pedogenic origin and the anthropogenic origin of the trace element in the environmental compartments. We collected 271 soil samples representative of different parent rocks and soil types from the whole territory of Hong Kong and analyzed the composition of clay mineralogy and the contents of 15 chemical elements (Fe, Cd, As, etc.) for these samples. The baseline was predicted with the method of the normalization procedure combined with the relative cumulative frequency curve. The result indicated that Fe was the best reference element for the normalization procedure among the five potential reference elements (Fe, Al, Sc, Ti, and Mn), followed by Sc and Ti. A poor correlation was found between Sc, Ti, and Cu. The predicted baseline was much lower than the *A*-value of the Dutch List used usually in screening the polluted soil of Hong Kong, implying that the extent of heavy metal pollution might have been underestimated with respect to local lands. We also applied the cluster analysis to distinguish the geochemical associations of the trace elements due to its importance to the baseline. Approximately three major associations including the Fe–Mn-oxides related, Al oxides or Al-bearing-clay-mineralogy related and sulfide- related associations were observed from the dendrogram.

Keywords Geochemical baseline · Normalization procedure · Trace element · Soil

Introduction

A geochemical baseline of trace elements provides the means of defining the natural spatial variations in the geochemistry of the earth's surface materials like soil and sediment. Meanwhile, it can distinguish between the pedogenic origin and the anthropogenic origin of trace elements in the environmental compartments (Darnley 1997; Salminen and Tarvainen 1997). However, the term 'geochemical baseline' has not yet been defined clearly. Since the first definition proposed by Tidball et al. (1974), many ways of defining the geochemical baseline have been suggested (Beus and Grigorian 1977; Rose et al. 1979; Ebens and Connor 1980). Among them, the most commonly used was the natural abundance of an element in a particular material such as soil, sediment, and rock with respect to a particular area or dataset. However, this definition was used mainly in exploration rather than environment because it generally refers to a single value

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(Salminena and Gregorauskienè 2000). Prior to a definition of geochemical baseline in environment, it is crucial to acknowledge the natural variability of any analyte in a given matrix under the aspect of the migration of chemical compounds and elements within and between individual environmental compartments. Therefore, a geochemical baseline or geochemical background should be characterized by regional variability and it is a function of time (Matschullat et al. 1999).

The normalization procedure is a widely applied method to obtain the regional geochemical baseline particularly in sedimentology recently (Covelli and Fontolan 1997; Donoghue et al. 1997; Tam and Yao 1998; Hamon et al. 2004). In this approach, the measured element concentrations have to be fitted with the reference element that is not influenced by anthropogenic activities. This is done by linear regression so that the baseline can be estimated for every single point that confirms to the regression conditions. The samples that lie beyond the confidence interval (95%) will be posted as anthropogenically influenced. However, the selection of the reference element is not universal but mostly depends on local geological and mineralogical features (Tam and Yao 1998; Hamon et al. 2004). Aluminum, a major constituent of the aluminosilicate mineral fraction, is often used as the reference element (Covelli and Fontolan 1997; Donoghue et al. 1997; Miko et al. 1999). Iron, a clay mineral indicator element, has been used as an alternative to Al by Tam and Yao (1998) and Tsai et al. (2003). In addition, Ti and Sc have also been used as reference elements due to their characteristic of geochemical stability in the environmental compartments (Teng et al. 2003). Another approach of calculating the baseline is relative cumulative frequency (RCF), which is widely used as well (Bauer et al. 1992; Bauer and Bor 1995). This technique does not require any assumption concerning the distribution function and simply applies the curves of the individual elements to display the RCF linearly. The first turning point of the curve can therefore be defined as the upper limit of any background data collective (Matschullat et al. 1999).

Since the geochemical baseline for the soils of Hong Kong was not investigated in the past, the target value of the Dutch List that was used to distinguish between clean and polluted soil in The Netherlands was usually referred in screening the polluted soil (Wong et al. 1996; Chen et al. 1997; Tam and Yao 1998). The calculation of the mean value for the heavy metals that have been collected from the countryside is an alternative way to determine the background of the soils (Li 2001; Li et al. 2001). However, such methods seem to be inappropriate in determining the soil pollution precisely. The main objective of this study is to establish a geochemical baseline for the soils of Hong Kong by application of the normalization procedure. Furthermore, it was assumed to be useful in defining the geochemical baseline for the soils under a similar environment.

Materials and methods

Study area descriptions

Hong Kong is located between 22°08′–22°35′ north latitude and 113°49′–114°31′ east longitude, with a total land area of 1,076 km^2 in the southeastern tip of the Chinese mainland. It covers Hong Kong Island, Kowloon, New Territories and a number of outlying islands and Lantau Island is the biggest one. Three–quarters of the land areas is covered by countryside where most of the areas are managed as country parks and natural reserve areas. The limited agricultural land is scattered irregularly in the marginal land of New Territories (AFCD 2002). Geologically, Hong Kong is located in the extension to the South China Caledonian folding-orogenic belt and belongs to the part of west Pacific magmatic mobile belt of Mesozoic. The pre-Devonian basement is unexposed in the district and only a few strata of middle Devonian, lower Carboniferous, Permian, Jurassic, and Cretaceous are exposed (Lee et al. 1999). The middle and later Jurassic volcanic rocks in particular of the tuffs are the most widely distributed in Hong Kong, and Granite, Granodiorite, adamellite, feldspathic, porphyry of middle-late Yanshanian phase are well developed (Peng 1986). There is an anticlinorium with the axial direction of northeast–southwest in the regional tectonics. The mineral resources are composed of metallic and nonmetallic deposits, with the former found in Hong Kong that includes pyrometasomic, pegmatite, hydrothermal replacement, and fissure filling, while the latter includes graphite, kaolin, clay, feldspar quartz, building stone, and sand (Peng 1978).

Major soil types of Hong Kong consist of Ferralisols, Ferrisols, Luvisols, Cambisols, Gleysols, Entisols, and Anthrosols according to the classification of Chinese soil taxonomy (CASNNFC 2001). The climate of Hong Kong is generally described as subtropical monsoon. The mean monthly temperature ranges from 15.9°C in January to 28.9°C in July, with the extreme minimum and the maximum of 0 and 36.2°C, respectively, based on the records of the Hong Kong Observatory (2001). Rainfall is also highly seasonal and 77% falls between

May and September, with the mean annual rainfall of 2,214 mm.

Soil sampling and chemical analyses

Soil profiles were collected based on the parent material, land using, vegetation types, and elevations from the countryside of Hong Kong including the New Territories, Hong Kong Island and Lantau Island. Soil layers were sampled according to the soil genesis sequence in each profile and a total of 271 horizon samples representative of 51 soil profiles were collected (Fig. 1). The collected samples were air-dried at room temperature for a few days, screened through a 2-mm sieve and then stored in glass bottles prior to analysis. Samples were digested with 60% perchloric acid, 40% hydrofluoric acid, concentrated nitric acid and concentrated hydrochloric acid (Page et al. 1986). Concentrations of Al, Co, Cr, Cu, Fe, Mn, Ni, Pb, Ti, Sc, and Zn were analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES; Perkin-Elmer 3300 DV), while concentrations of As, Cd, Hg, and Se were analyzed either by a graphite furnace atomic absorption spectroscopy (GFAAS; Varian 220Z) or cold vapor atomic absorption spectroscopy (CVAAS; CETAC M-6000A). For quality control, reagent blanks, replicates, and standard reference material (GSS-4 soil purchased from the National Research Center for Geoanalysis of China) were incorporated to detect contamination and assess precision and bias. The results showed no sign of contamination and revealed that the precision and bias of the analysis were generally below 10%. The recovery rates for the elements in the standard reference material (GSS-4) ranged from 75 to 115%. Mineralogical analyses of the selected soil samples were performed by X-ray diffraction (XRD) (D/max-IIIC).

Statistical analysis

Statistical analysis such as cluster analysis and cumulative frequency distribution was performed by the SPSS 11.0 for Windows. The Kolmogorov–Smirnov (K–S) test was applied to test the normal distribution for both raw and log-transformed data. The non-normal distributed data were log-transformed prior to further data analyses. Since the concentration of elements varied to a great extent, the raw data were standardized to the *Z* score (with a mean of 0 and a standard deviation of 1) before execution of the cluster analysis. Cluster was performed with the method of weighted average linkage between the groups. Pearson correlation for the cluster intervals and the elements that showed a close correlation were identified and grouped for further analysis.

Results and discussion

Mineral composition of Hong Kong soils and the reference element selection

Particle size analysis revealed that the texture of Hong Kong soil varied to a great extent from clay to sand most of which was loam. Kaolinite dominated the clay mineralogy in the soils, other clay minerals such as vermiculite, micas, gibbsite, quartz, goethite, montmorillonite, and mixed-layer clay minerals were also

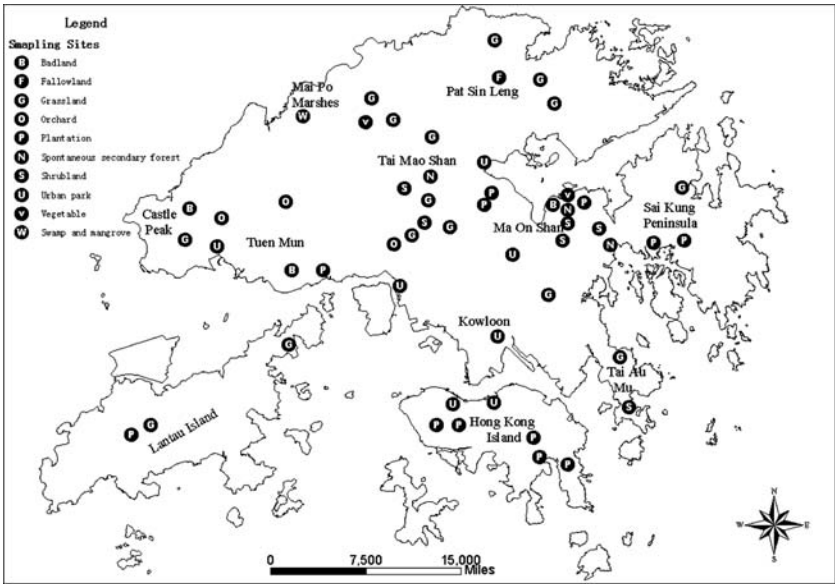

Fig. 1 Map of soil samples locations

identified by XRD. It indicated that a high degree of weathering had occurred in the soils of Hong Kong (Duzgoren-Aydin et al. 2002; Duzgoren-Aydin 2003).

In general, as the intensity of weathering increases, Ca, Na, K, and Si decrease, whereas Fe, Ti, and Al together with some trace elements increase. The relative enrichment of these elements can be mostly attributed to the formation of secondary phases, particularly sesquioxides and clay minerals, upon which most major elements and some trace elements can be fixed during weathering (Malpas et al. 2001). In addition, the geochemical behavior of trace elements was also controlled by the type and abundance of clay minerals. Duzgoren-Aydin and Aydin (2003) observed the enrichment of Cu, Ni, and Pb in the clay minerals with a higher degree of weathering in Hong Kong as a result of the relative abundance of Mn-oxides, which hold a high-sorption affinity. Since the reference element for the determination of the geochemical baseline must be an important constituent of one or more of the major trace element carries, Fe, Ti, Al, and Mn were therefore selected to be the potential reference elements based on the result of clay mineralogy analysis. Meanwhile, Sc was also selected as the potential reference element due to its relative immobility. Correlation analysis between the potential reference elements and elements of concern was performed as an aid to making the correct choice (Table 1). On the whole, Fe seems to be the best reference element as it has a significant positive relationship with all the elements of concern and would be used in the following normalization procedure.

Geochemical baseline of Hong Kong soil

The linear relationship between the contents of each element of concern and Fe contents was determined in the form of $y = ax + b$, where y is the value of the element, x is the Fe content in the samples. As proposed by Covelli and Fontolan (1997), the data lying out of the 95% confidence band were eliminated, and then build a new linear equation with the updated dataset until all the data were within the 95% confidence band. Table 2 shows the final equations for predicting the geochemical baseline of Hong Kong soil. Based on the equations and the measured Fe contents, a predicated value for each element at each sample location was computed. It was therefore defined that the predicated mean value as the critical value and mean ± 2δ as the range of the geochemical baseline for each element (Covelli and Fontolan 1997; Reimann and Filzmoser 1999), where δ is the standard deviation of predicated dataset. Table 3 is the predicted geochemical baseline of Hong Kong soil. It could be observed that the predicted baseline was close to the measured data according to the mean values for most of the elements, as all the samples were collected from the countryside areas, where the effect of industrial activities was relatively low to the soils on the whole. However, an exception was found with As, the measured mean value was over two times higher than the predicted data. This may probably result from the agricultural activities in the past as the inorganic forms of As were used extensively in agriculture such as insecticides, herbicides, fungicides, desiccants, defoliants and as additives to animal feeds prior to 1970s in Hong Kong (AFCD 2002). Chen et al. (1997) also found a relative higher concentration of As in the soils of agricultural lands and forestlands in Hong Kong. However, the upper limit derived from the normalization procedure for each element was much lower than the counterpart of the measured data. It implied that the impact of anthropogenic activities on the trace element contents of the soil could be eliminated by the normalization procedure, whereas the natural variability caused by the pedogenic process was not covered up. It could be represented by the range of the baseline with such a method.

Table 1 Correlation matrix for potential reference elements with concerned elements

Concerned elements	Potential reference elements				
	Al	Fe	Mn	Sc	Ti
lgAs	0.101	0.380**	–0.129	0.186*	0.404**
lgCd	0.446**	0.876**	0.458**	0.640**	0.792**
lgCo	0.208*	0.451**	0.435**	0.486**	0.742**
lgCr	0.089	0.454**	0.207*	0.199*	0.538**
lgCu	0.376**	0.227*	–0.139	0.109	0.062
lgHg	0.438**	0.599**	0.420**	0.447**	0.580**
Ni	0.446**	0.570**	0.453**	0.488**	0.578**
lgPb	0.198*	0.247**	0.431**	0.263**	0.205*
lgSe	0.585**	0.633**	0.068	0.348**	0.288**
lgZn	0.302**	0.385**	0.523**	0.412**	0.370**

**Correlation is significant at the 0.01 level (two-tailed)

*Correlation is significant at the 0.05 level (two-tailed)

Table 2 Linear relationship between trace metals and Fe

Equations	Selected statistical results		
	SEE[a]	*r*	*p*
$\lg[\mathrm{As}] = 0.113\,[\mathrm{Fe}] + 0.415$	0.279	0.461	<0.05
$\lg[\mathrm{Cd}] = 0.225\,[\mathrm{Fe}] - 0.957$	0.120	0.876	<0.01
$\lg[\mathrm{Co}] = 0.157\,[\mathrm{Fe}] + 0.234$	0.165	0.696	<0.01
$\lg[\mathrm{Cr}] = 0.071\,[\mathrm{Fe}] + 1.048$	0.141	0.449	<0.01
$\lg[\mathrm{Cu}] = 0.045\,[\mathrm{Fe}] + 0.936$	0.184	0.252	<0.01
$\lg[\mathrm{Hg}] = 0.120\,[\mathrm{Fe}] - 1.669$	0.158	0.629	<0.01
$[\mathrm{Ni}] = 1.563\,[\mathrm{Fe}] + 1.653$	2.046	0.502	<0.01
$\lg[\mathrm{Pb}] = 0.050\,[\mathrm{Fe}] + 1.408$	0.178	0.274	<0.05
$\lg[\mathrm{Se}] = 0.186\,[\mathrm{Fe}] - 0.600$	0.200	0.705	<0.01
$\lg[\mathrm{Zn}] = 0.086\,[\mathrm{Fe}] + 1.681$	0.188	0.357	<0.01

[a] SEE indicates the standard error of Y estimation

Geochemical baseline obtained from relative cumulative frequency method as a comparison

The technique of RCF curve was conducted in order to make a validation for the results of the normalization procedure. Figure 2 shows the RCF curves for each element. As shown in the curves, at least one turning point was indicated in each curve. The mean value of the first band of the slope was regarded as the critical value of the baseline as proposed by Matschullat et al. (1999). The result is shown in the Table 4. It could be observed that the baseline obtained from the RCF method was much identical to that of the normalization procedure based on the mean value. This implied that it is feasible to use the element of iron as the reference element in estimating the geochemical baseline of Hong Kong soil.

In addition, the reference background value applied usually to the soils of Hong Kong in the past was provided as a comparison in Table 4. It is obvious that the predicted geochemical baseline is much lower than the *A*-value of the Dutch List (VROM 2000), which means that heavy metal pollution might be underestimated for the Hong Kong soil when the *A*-value was referred to. The predicted geochemical baseline was also different from the background of Guangdong soils for most of the elements, although the two areas are related geographically and geologically. As for the background value of Guangdong soils, a mean value was computed from the dataset covering the whole territory in which the land uses and parent rock were much more complex than those of Hong Kong (Xia et al. 1987). Therefore, it could vary greatly between the two areas in geochemistry of the trace elements as shown in Table 4. However, the background (or baseline) would not be estimated appropriately without a proper sampling scheme or data processing even if the dataset were derived from the local soils. Li et al. (2001) applied the mean value of the heavy metal from the countryside as a background. In this case, the soil pollution for Cd, Cu, and Pb might be overestimated since the samples collected were not distributed as extensively as the whole area of Hong Kong. On the other hand, log-normal distribution occurred for most of the elements in the soils, hence, the mean value would be affected by the extreme of the dataset. This might also result in the over- or under-estimation of the background values (Reimann and Filzmoser 1999).

Table 3 Geochemical baselines computed by the normalization procedure and measured results

Trace elements	Geochemical baseline (mg kg^{-1})		Measured contents (mg kg^{-1})	
	Mean	Range	Mean	Range
As	4.69	2.08–7.31	11.5	0.83–147
Cd	0.39	0–0.82	0.41	0.05–1.17
Co	3.94	0.96–6.92	4.79	0.13–24.1
Cr	16.1	10.8–21.4	17.1	6.14–35.3
Cu	10.2	7.8–12.7	14.7	2.69–83.5
Hg	0.041	0.017–0.064	0.045	0.013–0.141
Ni	5.03	1.99–8.07	5.09	0.19–13.3
Pb	33.0	25.5–40.5	39.7	10.2–128
Se	0.69	0.06–1.32	0.76	0.07–2.26
Zn	76.5	48.5–104	92.1	22.0–280

Geochemical association of the trace elements in Hong Kong soil

Geochemical association of trace element in the soil is mainly determined by the regional geological features, pedogenesis and the characteristics of trace element. It would influence the value of regional geochemical baseline (Baize and Sterckeman 2001). Figure 3 is the result of the cluster analysis for the 15 elements of the Hong Kong soil and three major geochemical associations were distinguished in the dendrogram. The first group is composed of Cd, Fe, Ti, Sc, Co, Ni, Zn, Pb, Mn, and Cr. This indicated that trace elements such as Cd, Ni, and Zn were mainly associated with Fe–Mn oxides and were relatively immobile in geochemistry. Similar geochemical association was also found in the stream sediment of Hong Kong in which Cd and Co were related to Fe, while Pb was correlated to Mn significantly (Parry 2000). This implied that the first association was mainly influenced by local geological features. The element of chromium was also incorporated into the first group although it showed a weak correlation to the other elements. Chromium is frequently found to be associated with Fe in natural lateritic Fe-oxide crust. The trivalent form chromium (Cr^{3+}) probably accumulated during weathering through the way of incorporation into the octahedral

Fig. 2 Relative cumulative frequency curves indicates the upper limits of the geochemical baseline for the trace elements of Hong Kong

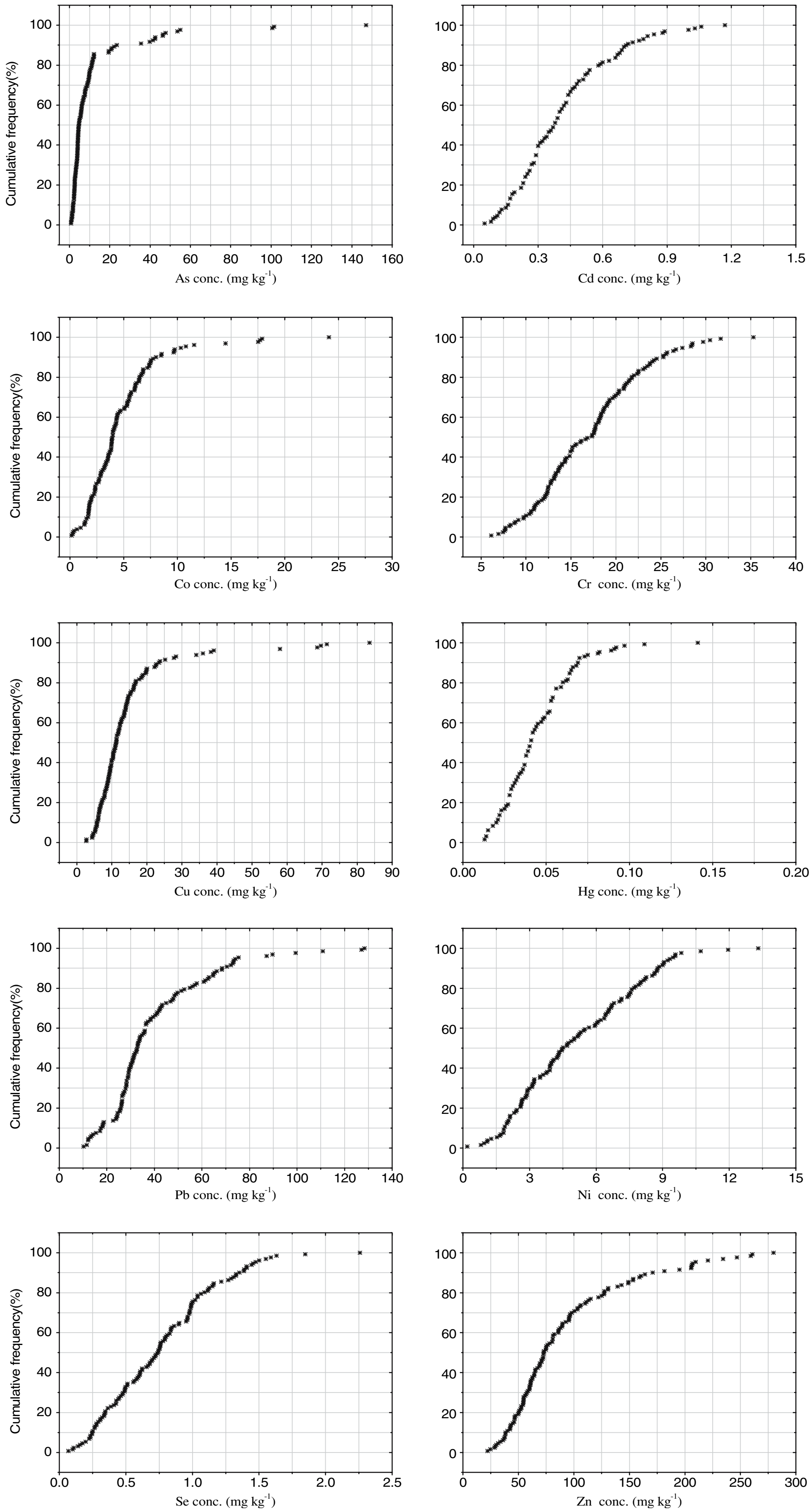

Table 4 Geochemical baseline estimated by relative cumulative frequency and the reference background applied for the Hong Kong soil

Trace elements	Geochemical baseline[a]		Guang-dong soil[b]	Dutch List *A* value[c]	Li et al. (2001)	
	Mean	Range			Mean	Range
As	4.69	0.83–9.58	11.9	29	NA	NA
Cd	0.37	0.01–0.49	0.046	0.8	0.15	0.02–0.37
Co	3.88	0.13–7.53	6.97	20	NA	NA
Cr	15.7	6.14–24.3	66.9	100	NA	NA
Cu	10.2	2.69–16.9	14.7	36	5.17	2.57–13.8
Hg	0.041	0.013–0.07	0.025	0.3	NA	NA
Ni	4.86	0.19–9.07	34.4	35	NA	NA
Pb	26.7	10.2–36.6	23.9	85	8.66	3.58–28.2
Se	0.63	0.07–1.16	0.26	NA	NA	NA
Zn	63.4	22–103.2	62.2	140	76.6	23.8–164

δ standard deviation, *NA* data is not available

[a] Estimated by the method of relative cumulative frequency

[b] Data sourced from Xia et al. (1987), it means the background value

[c] Data sourced from HSPE (2000)

sites in the structures of kaolinite, gibbsite, hematite, and goethite, the minerals that make up most of the clay fraction (Marques et al. 2004). The second group contained Hg, Se, Al, and Cu, indicating this association was connected with the Al oxides or with the Al-bearing clay minerals mostly. In addition, Hg and Cu were always presented in the form of sulfides such as HgS and $CuFeS_2$ in geochemistry, while Se occurred among these sulfides as an associated mineral (Peng 1986). The last group was the element of arsenic, which occurs as a sulfide generally in geochemistry. However, it was not associated with Hg, Se, Cu, and Al of the second group in the dendrogram. This might be attributed to the anthropogenic sources of arsenic in the soil as arsenic-bearing pesticides were applied in the past (Chen et al. 1997). Figure 4 shows the relationship between the content of As and that of S in the soil with the raw dataset and the dataset within the baseline range, respectively. For the former, no relationship was found between them, which was reflected by the low value of r^2 (0.0009), whereas, a significant

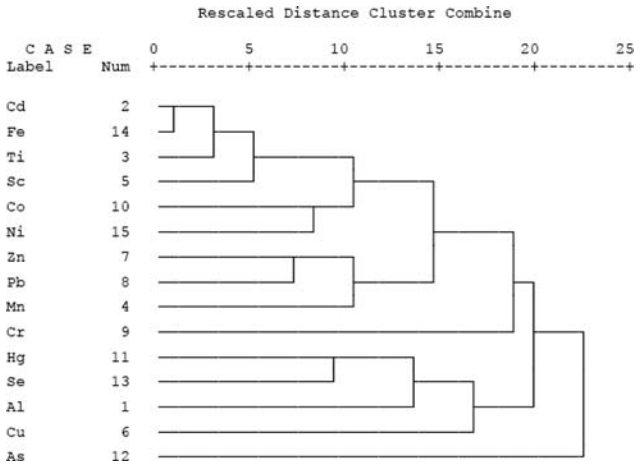

Fig. 3 Dendrogram using average linkage (between groups) to display the elements association in the soil of Hong Kong

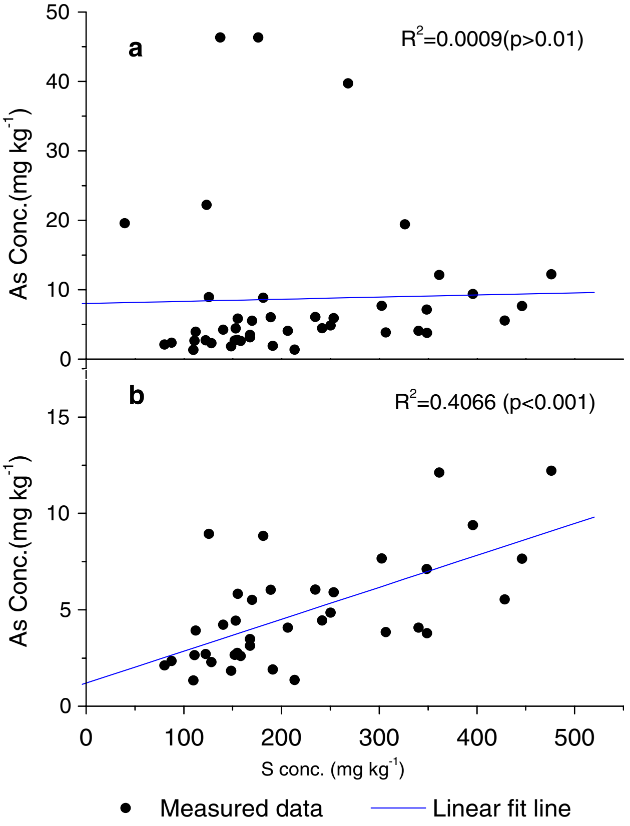

Fig. 4 Correlations between As contents and S contents in the soils. **a** and **b** represents raw dataset and dataset within the geochemical baseline, respectively

($r^2 = 0.4066$, $p < 0.01$) positive correlation was observed when only the data within the baseline range were focused on.

Conclusions

This study indicates that the element of Fe is a good reference element in the determination of the geochemical baseline for the soil of Hong Kong as when compared to the other four elements Al, Ti, Sc, and Mn. The baselines of ten trace elements that were of concern in the survey of soil pollution were predicted through the linear regression between iron and each element. The predicted values were much closer to those obtained from the RCF curve. All the estimated baselines were much lower than the *A*-values of the Dutch List, which was used as an alternative background in screening the contaminated soil of Hong Kong. This implied that the trace elements in pollution might have been underestimated for the Hong Kong soils. A geochemical association of the trace element would play an important role in the geochemical baseline. Approximately three types of geochemical associations could be identified in Hong Kong soils including associations with Fe–Mn oxides, Al oxides or Al-bearing clay minerals and sulfides.

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