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Comprehensive assessment of heavy metal pollution in river and lake sediments using face graph and index of geoaccumulation

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Abstract——Surface sedimental samples were collected from (1) Le An River in Jiangxi Province, south of China, and (2) several lakes located in southwest of Germany. Concentrations of heavy metals in these samples were determined. Moreover, the situations of heavy metal pollution in two regions were comprehensively assessed by combined index of geoaccumulation with visualized multivariate graphical method——modified Chernoff Face Graph. Face graphs intuitively demonstrated strong heavy metal contamination in Le An River, especially copper pollution. However, in lakes' sedimental samples, contamination of other heavy metals showed mild or clean except cadmium and chromium.

Keywords: heavy metals; sediment; assessment; index of geoaccumulation; face graph.

1 Introduction

Dexing Copper Mine is the largest opencast mine in China. It situates close to Le An River in Jiangxi Province, which joins with a few rivers and finally flows into Po Yang Lake, the largest freshwater lake in China. At present, its ore production is over sixty thousand tons per day, and will expand to one hundred thousand tons or more by the end of this century (Tang, 1994). Owing to exceeding the load of local waste water treatment plant, a great number of various pollutants, mainly heavy metals, have been discharged into Le An River and downstream more than two hundred kilometers. On the other hand, near Mannheim and Worms in southwest of Germany, several lakes have been polluted by some industrial contaminative sources nearby, such as wastewater discharging from electroplate plants, however, because of strict protection regulations and measures, the pollution extent in this region is much weaker than that in De Xing District. Since sediment is extremely significant to study history and current status of heavy metal pollution in aquatic ecosystem (Förstner, 1979), the comparative investigation on sediment polluted by heavy metals between different areas can provide basic information for restoration engineering and can supply databank for foundation of relevant sediment quality criteria (Wen, 1993).

Because quantitative evaluation of heavy metal pollution inevitably involves multi-factors, but normal 2 dimension plane or 3 dimension solid figures are very difficult to describe high-dimensional data exactly by tracing, so some particular multivariate graphical methods become more popular and attractive, such as radar chart, linked vector plot, constellation graph, face graph , non-linear mapping and so on (Schmid, 1983; MacEachren, 1994). The Chernoff Face Graph (Chernoff, 1973; 1975) is the most elaborate type of icon plot: cases are visualized by schematic faces such that the relative values of variables selected for the graph are represented by the variations of specific facial features. Although this means has been used widely (Bruckner, 1978; Riedwyl, 1981; Fang, 1989; Zhao, 1994), report on application of face graph to aquatic environmental chemistry is considerably fewer (Zhao, 1994).

In this paper, based on principle of sedimentology and current developments in research of heavy metal pollution, we carry out a comprehensive assessment of heavy metal contamination in sedimental samples mentioned above, taking advantage of index of geoaccumulation introduced by G. Müller (Müller, 1981), combined with modified multivariate graphical method-face graph initially devised by H. Chernoff(Chernoff, 1973).

2 Materials and methods

2.1 Sampling and experiments

In order to survey recent developments of heavy metal contamination, surface sedimental samples (0—10 cm depth) of lakes neighboring Mannheim and Worms in southwest of Germany were collected by two-jaw grab in September 1994. Material of fraction less than 63μm was wet sieved and dried within 30—40℃. After grinding with mortar made by agate and mixing with automatic maxer, 1 g of each sample was digested by HNO_3-HCl mixture. Cr, Cu, Pb, Zn, Cd, Ni and Mn were determined by AAS, meanwhile Hg was measured by mercury analyser.

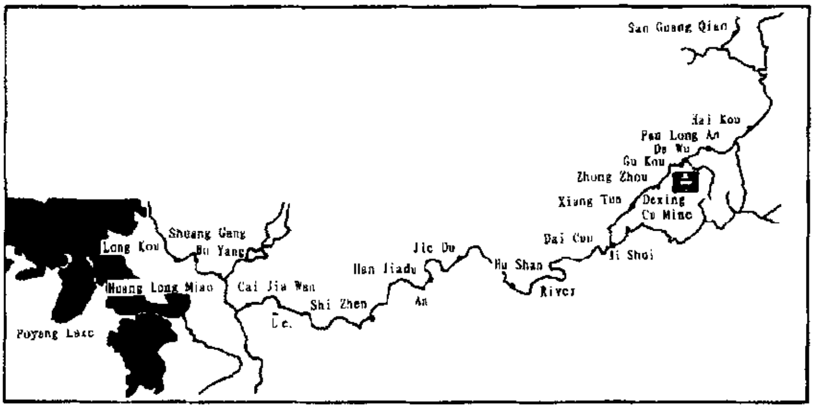

Fig. 1 Sketch map of Le An River

The Cooperative Ecological Research Project(CERP) was launched in 1987 within the

International Man and Biosphere (MAB) Program with a fund in trust from Federal Ministry for Research and Technology (BMFT), Germany. As one of the most important CERP subprojects, it was anticipated to assess the pollution situation and potential ecological impacts in Le An River and Po Yang Lake aquatic ecosystem. Along Le An River from upstream to downstream, 15 sampling sites were established in 1993(Fig. 1), less than 63μm fraction in each sample was selected as material in order to compare results each other. Pretreatment and digestion procedures of samples were similar to lakes' samples mentioned previously. Cr, Cu, Pb, Zn, As, Ni, Mn and Cd were determined by AAS, ICP and X-ray fluorescence spectrograph. Some of raw data quoted in this paper were gained from CERP-DBMS(also a sub-project of CERP)after permission.

2.2 Data transformation and statistical drawing

Since raw data would lose practical significance on face graph, it was necessary to conduct a linear transformation for each variable into a predetermined range. The transformation formula for raw data of heavy metal concentration was as follows:

$$X_{ij} = a_j + (b_j - a_j)\frac{\log_2 Y_{ij} - \log_2 Y_{minj}}{\log_2 Y_{maxj} - \log_2 Y_{minj}}; i = 1,2,\cdots n; j = 1,2\cdots p. \qquad (1)$$

Here X_{ij} was the data after transformation; Y_{ij} was the raw content of No. i sample and No. j heavy metal; Y_{minj} and Y_{maxj} were the minimum and maximum concentration of all samples for No. j heavy metal , respectively; a_j and b_j were upper and lower limit of range separately in linear transformation. In this report, the geochemical background average value of various heavy metals in general shale of the earth lithosphere was treated as one of sedimental samples.

Index of geoaccumulation for heavy metals proposed by G. Müller (Müller, 1981) was calculated by Formula(2):

$$I_{geo} = \log 2(C_n/(K \times B_n)) . \qquad (2)$$

Here C_n was the heavy metal concentration measured in experiment; B_n was the geochemical background average value for each heavy metal in general shale of the earth lithosphere (Müller, 1981; Rosler, 1972); $K = 1.5$ was a constant, considered the variation of geochemical background value caused by diagenesis. Based on I_{geo}, the heavy metal pollution of sediment could be classified into 7 grades presented in Table 1.

Table 1 Index of geoaccumulation and classification of heavy metal pollution in sediment

I_{geo}	Rank	Pollution grade
>5	6	Extra strong
4—5	5	Strong
3—4	4	Partial strong
2—3	3	Moderate
1—2	2	Partial moderate
0—1	1	Mild
<0	0	Clean

Drawing Chernoff Face Graph, it usually needed 18 to 20 variables for position of the face, if less than the standard number of variables, some positions would be fixed, i. e. their values became default or fixed by user or computer software (Statistical analysis & drawing software in this thesis was: STATISTICA™ for Windows™, Vol. 4. 5). By modified the traditional procedure, we selected 12 variables to draw face graph virtually, involved details are tabulated in Table 2.

Table 2 Description of facial features and ranges

Variable	Element	Range	Facial feature	Implication of definition
$X1$	Cr	5—9	Face width (horizontal distance between two ears)	The wider the face, the higher the concentration of Cr
$X2$	Rank-SUM(8)	5—9	Ear levitation (vertical distance from ear to centre of face)	The higher the ear levitation, the higher the Rank-SUM(8)
$X3$	Cu	5—9	Height of half face	The longer the half face, the higher the concentration of Cu
$X4$	Ni	5—9	Eccentricity of upper ellipse of face	The bigger the eccentricity, the higher the concentration of Ni
$X5$	Mn	5—9	Eccentricity of lower ellipse of face	The bigger the eccentricity , the higher the concentration of Mn
$X6$	As	5—9	Length of nose	The longer the nose, the higher the concentration of As in river(in lake, the value was default)
$X7$	Pb	5—9	Position of centre of mouth curve (Vertical distance from centre of mouth curve to centre of face)	The farther the distance, the higher the concentration of Pb
$X8$	I_{geo}-AVG(6)	5—9	Curvature of mouth curve (protruding down meant positive; protruding up meant negative)	The greater the positive curvature the higher the I_{geo}-AVG(6); the greater the negative curvature, the lower the I_{geo}-AVG(6)
$X9$	Hg	5—9	Length of mouth	The longer the mouth, the higher the concentration of Hg in lake (in river, the value was default)
$X10$	Zn	5—9	Height of eyes (vertical distance from eyes centre to face centre)	The higher the eyes' location, the higher the concentration of Zn
$X11$	None	5 (Constant)	Separation level of two eyes	Fixed default meaning
$X12$	Cd	5—9	Slant angle of two eyes and brows	The greater the slant degree of eyes and brows down to the face centre, the higher the concentration of Cd

In Table 2, I_{geo}-AVG(6) was the average of six typical heavy metals' I_{geo}, including Cr, Cu, Hg (or As), Pb, Zn and Cd. Rank-Sum(8) was the sum of rank values of I_{geo} for all 8 heavy metals. To I_{geo}-AVG(6) and Rank-Sum (8) of each sample, the transformation formula was: (definition of each item in this formula was similar to Formula(1)).

$$X_{ij} = a_j + (b_j - a_j)\frac{Y_{ij} - Y_{minj}}{Y_{maxj} - Y_{minj}}\ ;\ i = 1,2,\ldots,n;\ j = 1,2,\ldots,p. \qquad (3)$$

3 Results and discussion

3.1 Concentrations, I_{geo} and classification of heavy metal pollution in two regions

Table 3 Concentrations of heavy metals in sedimental samples from Le An River and several lakes

(Unit: mg/kg)

Samples No.	Samples name	Cr	Cu	Pb	Hg	As	Zn	Cd	Ni	Mn
1	HK	69	35	58	—	17	245	1.7	33	665
2	PLA	60	831	58	—	16	102	2.0	19	1152
3	GK	138	3500	100	—	41	165	1.7	30	370
4	ZZ	104	2700	66	—	43	170	2.4	45	953
5	XT	118	1650	54	—	19	184	3.0	40	1392
6	DC	102	1200	320	—	126	1500	2.3	32	1810
7	HS	115	1100	110	—	55	1100	2.1	36	1830
8	JD	127	450	78	—	36	780	2.8	43	1100
9	HJD	97	600	116	—	46	608	2.2	43	1600
10	SZ	107	780	77	—	39	490	2.1	44	2000
11	CJW	111	910	78	—	43	760	2.5	44	1200
12	HLM	119	310	80	—	29	340	2.2	33	1510
13	SG	96	280	81	—	27	540	2.1	35	1370
14	LK	93	320	83	—	30	390	1.9	35	1380
15	BY	102	350	77	—	27	1200	1.9	35	1380
16	VA1	138	80	62	0.7	—	93	2.5	42	524
17	VA2	141	39	40	0.2	—	83	2.0	49	519
18	VA3	155	51	44	0.3	—	85	2.5	54	507
19	SS1	164	41	51	0.2	—	85	2.5	59	526
20	SS2	173	42	48	0.2	—	82	2.0	66	525
21	SS3	161	33	37	0.2	—	78	2.5	63	530
22	STW	154	31	33	0.2	—	81	2.5	50	532
23	SPW	148	31	31	0.2	—	78	2.0	48	547
24	BK	62	45	34	0.35	13	118	0.4	68	850

HK——Haikou; PLA——Panlongan; GK——Gukou; ZZ——Zhongzhou; XT——Xiangtun; DC——Daicun; HS——Hushan; JD——Jiedu; HJD——Hanjiadu; SZ——Shizhen; CJW——Caijiawan; HLM——Huanglongmiao; SG——Shuanggang; LK——Longkou; BY——Boyang; VA 1 to VA 3——Vorderev Altrhein 1 to Vorderev Altrhein 3; SS 1 to SS 3——Silbersee 1 to Silbersee 3; STW——Standerweiher; SPW——Spaitplatzweiher; BK——Geochemical background average value in general shale of the earth lithoshere (Müller, 1981; Rosler, 1972).

Raw data of heavy metal pollution in sedimental samples from Le An River and several lakes are shown in Table 3. Then, according to Formula (2) and Table 1, related I_{geo} and rank of pollution are listed in Table 4.

Table 4 I_{geo} and rank of heavy metal pollution in sedimental samples from Le An River and several lakes

No.	Name	Cr	Cu	Pb	Hg	As	Zn	Cd	Ni	Mn	I_{geo}—	Rank—
		I/R*	I/R	I/R	I/R	I/R	I/R	I/R	I/R	I/R	AVG(6)	SUM(8)
1	HK	-0.43/0	-0.95/0	0.19/1	—	-0.20/0	0.47/1	1.50/2	-1.63/0	-0.94/0	0.10	4
2	PLA	-0.63/0	3.62/4	0.19/1	—	-0.29/0	-0.80/0	1.74/2	-2.42/0	-0.15/0	0.64	7
3	GK	0.57/1	5.70/6	0.97/1	—	1.07/2	-0.10/0	1.50/2	-1.77/0	-1.78/0	1.62	12
4	ZZ	0.16/1	5.32/6	0.37/1	—	1.14/2	-0.06/0	2.00/3	-1.18/0	-0.42/0	1.49	13
5	XT	0.34/1	4.61/5	0.08/1	—	-0.04/0	0.06/1	2.32/3	-1.35/0	0.13/0	1.23	12
6	DC	0.13/1	4.15/5	2.65/3	—	2.69/3	3.08/4	1.94/2	-1.67/0	0.51/1	2.44	19
7	HS	0.31/1	4.03/5	1.11/2	—	1.50/2	2.64/3	1.81/2	-1.50/0	0.52/1	1.90	16
8	JD	0.45/1	2.74/3	0.61/1	—	0.88/1	2.14/3	2.22/3	-1.25/0	-0.21/0	1.51	12
9	HJD	0.06/1	3.15/4	1.19/2	—	1.24/2	1.78/2	1.87/2	-125/0	0.33/1	1.55	14
10	SZ	0.20/1	3.53/4	0.59/1	—	1.00/2	1.47/2	1.81/2	-1.21/0	0.65/1	1.43	13
11	CJW	0.26/1	3.75/4	0.61/1	—	1.14/2	2.10/3	2.06/3	-1.21/0	-0.09	1.65	14
12	HLM	0.36/1	2.20/3	0.65/1	—	0.57/1	0.94/1	1.87/2	-1.63/0	0.24/1	1.10	10
13	SG	0.05/1	2.05/3	0.67/1	—	0.47/1	1.61/2	1.81/2	-1.54/0	0.01/1	1.11	11
14	LK	0.00/1	2.25/3	0.70/1		0.62/1	1.14/2	1.66/2	-1.50/0	0.32/1	1.06	11
15	BY	0.13/1	2.37/3	0.59/1	—	0.47/1	2.76/3	1.66/2	-1.54/0	0.11/1	1.33	12
16	VA1	0.57/1	0.24/1	0.28/1	0.42/1	—	-0.92/0	2.06/3	-1.28/0	-1.28/0	0.44	7
17	VA2	0.60/1	-0.81/0	-0.34/0	-1.39/0	—	-1.09/0	1.74/2	-1.05/0	-1.30/0	-0.22	3
18	VA3	0.74/1	-0.39/0	-0.21/0	-0.81/0	—	-1.05/0	2.06/3	-0.92/0	-1.33/0	0.06	4
19	SS1	0.82/1	-0.72/0	0.01/1	-1.39/0	—	-1.14/0	2.06/3	-0.78/0	-1.28/0	-0.06	5
20	SS2	0.89/1	-0.68/0	-0.08/0	-1.39/0	—	-1.11/0	1.74/2	-0.62/0	-1.28/0	-0.11	3
21	SS3	0.79/1	-1.02/0	-0.48/0	-1.39/0	—	-1.18/0	2.06/3	-0.70/0	-1.27/0	-0.20	4
22	STW	0.73/1	-1.14/0	-0.63/0	-1.39/0	—	-1.13/0	2.06/3	-1.02/0	-1.26/0	-0.25	4
23	SPW	0.67/1	-1.14/0	-0.70/0	-1.39/0	—	-1.17/0	1.74/2	-1.08/0	-1.22/0	-0.33	3
24	BK	-0.58/0	-0.58/0	-0.58/0	-0.58/0	-0.58/0	-0.58/0	-0.58/0	-0.58/0	-0.58	-0.58	0

* I —— I_{geo}; R —— Rank of I_{geo} classification

3.2 Chernoff face graph and sedimental quality assessment of heavy metal pollution in two regions

All samples, including geochemical background average value (BK), are integrated into one specimen set. According to Formula (1) and (3), Fig. 2 represents the Chernoff Face Graph for each sample.

The last graph in Fig. 2 is the standard graph, which reflects the geochemical back-

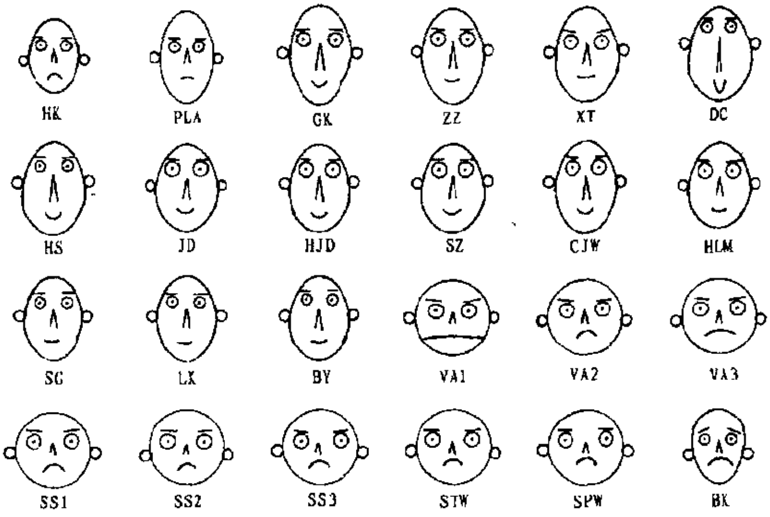

Fig. 2 Chernoff Face Graphs of each sampling site

ground average value in generall shale of the earth lithosphere. Others manifest the heavy metal contaminative situation in samples of Le An River (From HK to BY sequentially) and several lakes (from VA to SPW in order) located in southwest of Germany.

As for Le An River near Dexing Copper Mine, the relative pollution degrees of 15 samples are described with visualized face graphs vividly. The first one——Haikou can act as a reference site because of its closest shape and feature of face graph to the standard graph (BK). Undoubtedly, Cu plays the most dominant role in this mine region, and with expression on face graph, the half face heights of most sampling sites in Le An River are much longer than BK. From Panlongan in upstream site to Caijiawan in downstream site, the ranks of I_{geo} classification for Cu pollution almost come up to 4 or more. Furthermore, in Gukou and Zhongzhou strongly affected by a waste water discharging tributary through Dexing Copper Mine-Dawu River, their half face heights indicate that the level of Cu pollution may reach the maximum. Besides this area, from Xiangtun to Hushan suffer severe Cu pollution as well, resulting from another discharging tributary-Jishui River.

The intensity of Cd and Zn contamination is secondary and displayed by the slant and height of the eyes on face graph respectively. In Fig. 2, since the distribution model for Cd along Le An River is rather even, and Cd concentrations of all river samples are commonly higher than BK, so all face graph's eyes apparently slope towards the centre of face graph compared with BK. In contrast, Zn approximately has a two-peak distribution pattern which kurtosis areas concentrate in Daicun-Hushan section and in Boyang Lake, thus , their eye's heights are superior to others. The distributions of I_{geo} for As and Pb are similar along Le An River, and the corresponding pollution grades are lower except moderate pollution in Daicun, which possesses the longest nose and the lowest position of mouth centre among all samples of Le An River. Comparatively, the pollution levels of Cr, Mn and Ni are mild or clean.

More important, indices of Rank-SUM(8) and I_{geo}-AVG(6) can comprehensively show the overall situation of sediment contaminated by various heavy metals. On face graph, they are illustrated by the levitation of ears and the curvature of mouth curve respectively. According to I_{geo}-AVG(6), the distribution pattern of heavy metal pollution along Le An River can be divided into five sections: section I is a relative clean range, including upstream samples No. 1 and No. 2; section II is a strong polluted range where the intensity of pollution decreases gradually, containing samples No. 3—No. 5; Section III is a peak range of pollutants accumulation, such as samples No. 6 and No. 7; section IV is a stable polluted range where all samples have similar features, consisting of samples No. 8—No. 11; the last section V is a smooth declining range from sample No. 12 to No. 15. Mirrored on face graphs, samples in section III have the greatest positive curvature of mouth curve, so their accumulation and contamination of heavy metals are most vident. From section IV to V, although there are some small fluctuations, the degree of positive curvature on face graph reduces gradually up to near level-off. Meanwhile, the curvature of mouth curve in section II also decreases by degrees, reflecting the actual pollution status. And within the first section, the curvature of mouth has already changed from positive to negative, this means mild pollution in this range. In light of the whole tendency, the Rank-SUM(8) is in agreement with the I_{geo}-AVG (6).

In contrast to Le An River intensively, the polluted situation of heavy metals for several lakes in southwest of Germany is fairly weak except Cd and Cr, this can be observed from basic characteristics of their face graphs compared with BK. In most part of lake's samples, Cd concentrations fall within the range from partial moderate to moderate contaminative grade, so that their eyes incline towards centre of face evidently(referring to BK). Due to all of face graphs for these samples are wider than BK in some degree, therefore, Cr may be an universal pollutant probably in this industrial district. As a whole , the appearances and traits of face graphs for these lake's sedimental samples are similar, only different at Vorderev Altrhein 1 and Vorderev Altrhein 3. The mouth curvature of these two samples is relative gentle as their I_{geo}-AVG(6) slightly greater than 0. In addition, Hg concentration of Vorderev Altrhein 1 is somewhat higher than others causing its different length of mouth. However, the contamination of other heavy metals is much faint commonly.

4 Conclusions

In this report, by the aid of index of geoaccumulation combined with modified Chernoff Face Graph, we carry out a comprehensive assessment of heavy metal pollution in sediment from two different areas. By comparing concentrations of heavy metals with their visualized schematic face graphs, much more valuable information can be obtained, and it is very beneficial to the following treatment and restoration works in these polluted areas.

As for Le An River, the significant longer faces in most for samples demonstrate the extreme serious Cu pollution along this river, especially close to the discharging points of waste water from Dexing Copper Mine, such as Gukou and Daicun. The contamination grades of Cd and Zn belong to moderate, moreover, the spatial distribution model of Cd is throughout uniform along Le An River, but Zn content approximately has a two-peak distribution pattern from upstream to downstream. From the relevant face graphs, kurtosis areas for As and Pb contamination are alike in Le An River and their polluted levels become weaker. However, the polluted degrees of other heavy metals are relative mild. Based on average of I_{geo}, the contamination of various heavy metals in Le An River can be divided into five groups: (1) a relative clean area in upstream; (2) an areain the up-middle stream where pollution strength decreases gradually; (3) an extra strong polluted area in middle-stream; (4) a stable contamination area in middle-down stream; and (5) a smooth declining area in downstream.

Generally, owing to their similar shape and features of face graph, the polluted situation of heavy metals in several lakes in southwest of Germany is rather slight except Cd and Cr. The distribution model for Cd is quite even in this region, and the pollution grades of Cd for these lake's samples commonly reach moderate. Meanwhile, to certain degree, Cr also leads to a widespread pollution caused by industrial discharging nearby. However, other heavy metals' contamination is considerably weak.

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