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Multifractal characteristic-based comparison of elements in soils within the Daxing and Yicheng areas of Hefei, Anhui Province, China

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Abstract

Industrial and agricultural activities can generate heavy metal pollution that can have a number of negative environmental and health impacts. This means that identifying areas with heavy metal pollution and the sources of these pollutants, especially in urban or developed areas with multiple possible sources of pollution, is an important first step in mitigating the effects of these contaminating but necessary economic activities. Here, we present the results of a heavy metal (Cu, Pb, Zn, Cd, As and Hg) soil geochemical survey and outline a new multifractal characteristic-based comparison method that allows deeper interrogation of soil geochemistry in urban or developed areas. This survey focuses on Hefei, the provincial capital of Anhui Province, China, an area that contains a number of individual towns within a large

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municipal area. This study focuses on the towns of Daxing and Yicheng both of which are incorporated within Hefei and are are economically focused on industry and agriculture, respectively. Here, we use a multifractal spectral technique to identify differences in the geochemistry of soils within the Daxing and Yicheng areas. The height difference between the two ends of the multifractal spectrum of the geochemical data ($\Delta f(a)$) for soils in the Daxing area decreases as follows: Pb>As>Cd>Cu>Zn>Hg, whereas $\Delta f(a)$ values of the geochemical data for soils in the Yicheng town areas decreases as follows: Hg>Zn>As>Cd>Pb>Cu. These differences indicate that the soils in these areas have differing multifractal geochemical characteristics, suggesting that the differing economic activities in these areas generate distinctly different heavy metal pollutant loads (e.g. Hg dominated agricultural pollution vs. Pb dominated industrial pollution). In addition, all of the elements barring Hg have larger $\Delta f(a)$ values in the Daxing area compared to the Yicheng area. These larger ranges indicate that the higher concentrations of heavy metals present in soils within the Daxing area (compared to the Yicheng area) are more likely to be related to industrial activities than agriculture. The industrial Daxing area contains significant Pb and As soil contamination, whereas Hg is the main heavy metal present in soils within the Yicheng area, indicating that differing clean-up procedures and approaches to remediating these polluted areas are needed, rather than a single approach to heavy metal pollution. The research presented here also highlights that the soils in these areas (and the source of these pollutants) need to be remedied in order to avoid further health and environmental impacts.

Keywords: soil geochemistry; multifractal modelling, heavy metal pollution, Hefei**.**

1. Introduction

Multifractal based analytical techniques have recently been used in a number of differing fields, including geophysics (Schertzer et al., 2011), medicine (Jennane et al., 2001), computer science (Wendt et al., 2009), geology (Deng et al., 2011;Zuo et al., 2012; Cheng, 1995; Yuan et al., 2012), environmental science (Lima et al., 2003;

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Albanese et al., 2007; Guillén et al., 2011; Salvadori et al., 1997), and ecology (Scheuring and Riedi, 1994; Pascual et al., 1995) among others. The advantages of these multifractal techniques include the fact that these approaches can identify non-linear characteristics, yielding new information that can be used to understand the controls on the distribution of key elements within the objects or data being studied (Gonçalves, 2000;Zuo et al., 2012). Multifractal techniques can also be used to analyze soil characteristics, including the identification of porous structures and the spatial variability in the characteristics of soils (Dathe et al., 2006; Caniego et al., 2005). These techniques and can also enable the characterization of complex phenomena in the spatial distribution of elements within soils, improving our knowledge of the controls on the geochemistry of soils and the regolith (Gonçalves, 2000). This means that these approaches can not only be used in mineral exploration (Yuan et al., 2012; Yuan et al., 2015; Zuo, 2014; Nazarpour et al., 2014) but can also be used in the analysis of pollutants such as heavy metals within soils (Guillén et al., 2011; Salvadori et al., 1997). Heavy metal pollution poses a serious risk for human health and the environment, meaning that soil pollution caused by anthropogenic activities (including industry and agriculture) has been the focus of a significant amount of research in recent years (McGrath et al., 2004; Wang et al., 2007; Leyval et al., 1997; Thomas and Stefan, 2002; Chunling et al., 2011). This in turn indicates that multifractal techniques enable the more precise identification of areas of contamination and the degree of contamination in polluted areas. Multifractal techniques, such as singularity mapping and multifractal interpolation, also enable more detailed analysis of the spatial distribution of heavy metals by the use of C–A models to define threshold values between background (i.e. geological) and anthropogenic anomalies, S–A modeling that uses these thresholds to spatially separate anomalies (i.e., anthropogenically derived heavy metal concentrations in this case) from background concentrations (i.e., geologically derived heavy metal concentrations), and using multifractal spectra to highlight non-linear characteristics and identify anomalous behavior that reflects the characteristics of some multifractal

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sets (Gonçalves, 2000; Albanese et al., 2007; Guillén et al., 2011; Lima et al., 2003; Cheng, 2001).

Hefei is the provincial capital of Anhui Province, China, and has an urban area that includes the towns of Daxing and Yicheng, areas that focus on industrial and agricultural activity, respectively. These towns provide an ideal location to compare and contrast the degree and characteristics of any heavy metal contamination of soils associated with these anthropogenic activities. This study focuses on these areas, and the results presented here further our understanding of any heavy metal pollution that is likely associated with these differing activities, both enabling and informing future planning for any necessary remediation of these soils. Our study uses multifractal techniques to determine the multifractal characteristics of the distribution of heavy metals in soils in these areas, enabling the characterization and contrasting of the heavy metal pollution of soils in these two towns.

2. Study area and geochemical data

2.1 Study area

The city of Hefei is situated in central–eastern China (Fig. 1(a)), has approximately 7.7 million inhabitants and covers an area of around 11,408 km^2. This paper focuses on the towns of Daxing and Yicheng (Fig. 1(b)), with the former representing one of the traditional industrial bases of the Hefei area and containing numerous industrial factories that are involved in the steel industry, chemical industry, paper making, and the production of furniture and construction materials, amongst others. In contrast, the town of Yicheng is agricultural, with the economy of the town focused on agricultural production, byproduct processing, livestock and poultry breeding, flower planting, and other enterprises related to agricultural activity.

2.2 Sampling and analysis

The study areas are covered by Quaternary sedimentary soils and are free of both natural mineralization and mining activities. A total of 169 surface (<20 cm below surface) soil samples were taken from the towns of Daxing and Yicheng on 1 × 1 km grids, yielding 78 samples from Daxing and 91 samples from Yicheng (Fig. 1(c–d)).

Sampling errors were minimized by splitting each sample into 3–5 sub-samples, each of which weighed more than 500 g. Each of these sub-samples was dried in air before being broken up using a wooden roller and then sieved to pass through a 0.85 mm mesh. The concentrations of 6 heavy metal elements (Cu, Pb, Zn, Cd, As and Hg) in the soil samples described above were determined during this study. Cd, Cu, Pb and Zn concentrations were determined by inductively coupled plasma–mass spectrometry (ICP–MS), with Hg and As concentrations determined by hydride generation–atomic fluorescence spectrometry (AFS). These techniques have detection limits of 1 ppm for Cu, 2 ppm for Pb and Zn, 30 ppb for Cd, 0.5 ppm for As and 5 ppb for Hg. The accuracy of these analyses was monitored by repeat analysis of standards and replicate analyses of sub-sets of samples using instrumental neutron activation analysis (INAA). Analytical precision was monitored using analysis of variance of the results obtained from duplicate analyses.

2.3 Results

The results of a statistical analysis of the resulting soil geochemical data are given in Table 1. Samples from Daxing have higher Cu, Pb, Zn, Cd and As maximum, standard deviation, skewness, and kurtosis values than soil samples from the Yicheng area. In addition, the soil samples from Daxing have much higher coefficient of variation (CV) values for Cu, Pb, Zn, Cd and As than the samples from the Yicheng area, indicating that soils in the Daxing area contain much higher and more variable concentrations of these elements. This suggests that samples from the Daxing area with elevated concentrations of heavy metals beyond the natural background variations in these areas were probably contaminated by anthropogenic activity.

All of the elements (barring Cu in the Yicheng area) in both the Yicheng and Daxing areas yield histograms that are positively skewed and contain some outliers, indicating that these data have non-normal, fractal-, or multifractal-type distributions. This means that multifractal techniques may be more suitable for the characterization of the geochemistry of the contaminated soils in these areas (Fig. 2).

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3. Multifractal spectrum analysis

Multifractal formalisms can decompose self-similar measures into intertwined fractal sets that are characterized by singularity strength and fractal dimensions (Cheng, 1999). Using multifractal techniques allows non-linear characteristics within datasets to be identified, enabling the extraction of information that can be used to understand the controls on the distribution of key elements within data. Fractal spectra ($f(a)$) are multifractal formalisms that can be used to describe the multifractal characteristics of a dataset and can be estimated using box-counting based moment, gliding box, histogram and wavelet methods, among others (Cheng, 1999; Lopes and Betrouni, 2009). The most widely used of these methods of estimating $f(a)$ values are the box-counting and gliding box methods, both of which are based on the moment method.

The initial step of the box-counting method estimates mass exponent function τ(q) values using a partition function as follows (Halsey et al., 1986):

$$\tau(q) = \lim_{\varepsilon \to 0}\left(\frac{\log(\chi^{q}(\varepsilon))}{\log(\varepsilon)}\right) = \lim_{\varepsilon \to 0}\left(\frac{\log \sum_{i=1}^{N(\varepsilon)} \mu_i^{q}(\varepsilon)}{\log(\varepsilon)}\right)$$

where $\mu_i(\varepsilon)$ denotes a measure with the i_{th} box of size ε and $N(\varepsilon)$ indicates the total number of boxes of size ε with $\mu_i(\varepsilon)$ values that $\neq 0$.

The calculation of the mass exponent function $\tau(q)$ for the gliding box method is different from the box-counting method, with the gliding box method providing a useful approach that can increase the number of samples within a dataset that are available for statistical estimation (Tarquis et al., 2006; Xie et al., 2010; Buczkowski et al., 1998). This means that the gliding box approach often provides better results with lower uncertainties than the box-counting method (Cheng, 1999). As such, we have used the gliding box approach during this study.

The calculation of the mass exponent function $\tau(q)$ for the gliding box method uses a partition function as follows (Cheng, 1999):

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$$\langle \tau(q) \rangle + D = \lim_{\varepsilon \to 0}\left(\frac{\log(\mu^q(\varepsilon))}{\log(\varepsilon)}\right) = \lim_{\varepsilon \to 0}\left(\frac{\log\left(1/N^*(\varepsilon)\right)\sum_{i=1}^{N^*(\varepsilon)}\mu_i^q(\varepsilon)}{\log(\varepsilon)}\right)$$

where $\mu_i(\varepsilon)$ denotes a measure with the i_{th} cell of a gliding box of size ε, <> indicates the statistical moment, and $N^*(\varepsilon)$ indicates the total number of gliding boxes of size ε with $\mu_i(\varepsilon)$ values that $\neq 0$.

The values of $\tau(q)$ derived using this equation can be then used to determine a and $f(a)$ values using a Legendre transformation, as expressed below:

$$\alpha(q) = \frac{d\tau(q)}{dq}$$

$$f(q) = q\alpha(q) - \tau(q) = q\frac{d\tau(q)}{dq} - \tau(q)$$

where Δα and Δf are essential parameters required to analyze the multifractal characteristics of the dataset in question. The widths of the left and right branches within the multifractal spectra are then defined using the following equations:

$$\Delta\alpha_L = \alpha_0 - \alpha_{\min}$$

$$\Delta\alpha_R = \alpha_{\max} - \alpha_0$$

$$\Delta\alpha = \alpha_{\max} - \alpha_{\min}$$

and the height difference $\Delta f(a)$ between the two ends of the multifractal spectrum are then extracted using:

$$\Delta f(\alpha) = f(\alpha_{\max}) - f(\alpha_{\min})$$

Higher Δa and $\Delta f(a)$ values are generally indicative of datasets with heterogeneous distribution patterns and higher levels of multifractality (Cheng, 1999; Kravchenko et al., 1999). In addition, multifractality associated with ordinary spatial analysis parameters, as represented by the $\tau''(1)$ parameter, can also be used to quantitatively characterize the multifractality of a dataset (Cheng, 2006) using the following equation:

$$\tau''(1) = \tau(2) - 2\tau(1) + \tau(0)$$

4. Calculation processes and discussion

The gliding box method used during this study can increase the number of samples that can be used in statistical estimations and provides results with lower uncertainties than the box-counting method. This, combined with the relatively sparse sample locations used in this study, means that we used the gliding box method to calculate multifractal spectra values for the geochemical data from the study area. This calculation used a range of q values from −10 to 10 with an interval of 1, yielding the multifractal analytical results given in Table 2 and the multifractal spectra (in the form of an α–$f\,(\alpha)$ diagram) shown in Fig. 3.

The multifractal data shown in Table 2 indicate that all of the elements barring Cu and Pb in the Yicheng area are characterized by a wide range of α values (i.e. have high Δa values) and have $\tau''(1)$ values less than –0.01. In addition, these data have a wider range of $\Delta f(\alpha)$ values compared to the Δa and $\tau''(1)$ values shown in Table 2. This means that the $\Delta f(\alpha)$ values obtained from these data may be the best measure to determine the multifractal characteristics of the distribution of these elements in soils within the study area.

The range of $f(\alpha)$ values for the geochemical data for soils within the Daxing area decreases in the order: Pb>As>Cd>Cu>Zn>Hg, whereas the values for these elements in soils within the Yicheng area decreases in the order: Hg>Zn>As>Cd>Pb>Cu, indicating a significant difference in the geochemical characteristics (and heavy metal pollution) in the soils within these two areas. These variations are linked to multifractal spectra (shown as an α–$f(\alpha)$ plot in Fig. 3), where combining the singularity exponent α and the corresponding fractal dimension $f(\alpha)$ generates a multifractal spectrum with an inverse bell shape. All of these spectra (barring the data for Cu in soils from the Yicheng area) show a steep increase (i.e. have a good positive correlation between the values) followed by a shorter section of the curve where these values negatively correlate (Fig. 3). All of these data are also asymmetric with respect to α for all elements, indicating that soils containing low and high concentrations of these elements are not evenly distributed within the study area (as is expected for

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areas containing point source pollutants like factories or individual farms).

All of the heavy metals analyzed during this study barring Hg have higher $\Delta f(\alpha)$ values in soils from the Daxing area, with Hg having higher values in soils from the Yicheng area (Table 2). This suggests that the industrial activities in the Daxing area generate multi-element heavy metal contamination soil contamination, whereas the only significant heavy metal pollution associated with the agricultural activity in the Yicheng area is Hg contamination. However, the Hg $\Delta f(\alpha)$ values in Yicheng area are larger than all of the other elements in this area as well as some of the elements in the Daxing area, indicating both the prevalence and significant degree of agricultural Hg contamination in the Yicheng area. This is important, primarily as Hg pollution can seriously impact human health as this element is concentrated upward in the food chain (e.g. (Jiang et al., 2006)), meaning that this contamination needs to be evaluated further and remediated to avoid any deleterious effects.

We further analyzed the spatial distribution of contamination within soils from the Daxing and Yicheng areas by examining the elements with the highest $\Delta f(\alpha)$ values, namely Pb and Hg, respectively. We used an approach focused on filled contour maps showing the distribution of Pb in the Daxing area and Hg in the Yicheng area using inverse distance weighted interpolation (Fig. 4–5). These maps indicate that areas with elevated levels of Pb contamination within the Daxing area are directly correlated to the location of industrial factories, whereas the Hg contamination in the Yicheng area is spatially correlated with the location of agricultural breeding facilities. This strongly suggests that the larger $\Delta f(\alpha)$ values for these elements within the geochemical data are related to the industrial and agricultural activities in the Daxing and Yicheng areas, respectively. The Hg contamination in the Yicheng area is of significance, especially as this form of contamination can cause serious health issues (e.g. Minamata disease). As such, the soils in this area may well require remediation, especially as Hg can be concentrated up the food chain and the Yicheng area is heavily agricultural, indicating that this activity may both be concentrating Hg as well as contaminating soils in this area. This distribution of soils with elevated concentrations of Hg also contrasts with the symmetrical distribution and relatively low $\Delta f(\alpha)$ values for Cu within the Yicheng area (Fig. 3). Comparing the distribution of Cu and Hg in the filled contours maps for the Yicheng area (Fig. 5–6) indicates an anti-correlation in terms of the spatial location of anomalously high concentrations of Cu and breeding facilities. This indicates that little Cu has been anthropogenically added (or removed) from the soils in the Yicheng area, suggesting that these soils maybe contain only natural background concentrations of Cu and that the agricultural activity in this area does not produce any significant Cu contamination. These data indicate that differing clean-up procedures and approaches to remediating these polluted areas are needed, rather than a single approach to heavy metal pollution. The results also indicate that multifractal modeling and the associated generation of multifractal parameters, such as $\Delta f(\alpha)$ values, are a useful approach in the evaluation of heavy metal pollution in soils and the identification of major element of heavy metal contamination.

5. Conclusions

Our data indicate that the soils from the Daxing area have a larger range of $f(\alpha)$ values for Cu, Pb, Zn, Cd and As than soils from the Yicheng area, although have a larger range in $f(\alpha)$ values for Hg. The range of $f(\alpha)$ values for the soil geochemical data in the Daxing area decreases in the order Pb>As>Cd>Cu>Zn>Hg, whereas soils in the Yicheng area have $f(\alpha)$ value ranges that decrease in the order Hg>Zn>As>Cd>Pb>Cu. In addition, Cu concentrations in soils in the Yicheng area may still have their original (i.e. natural) distribution and may not have been influenced by human activities. These data indicate that the industrial activity concentrated in the Daxing area generates multi-element heavy metal soil contamination whereas the agricultural activity concentrated in the Yicheng area generates Hg dominated heavy metal soil contamination. The latter is important, as Hg contamination can cause serious health issues (e.g. Minamata disease) and the soils in this area may well require remediation, especially as Hg can be concentrated up the food chain and the Yicheng area is heavily agricultural, indicating that this activity may both be concentrating Hg as well as contaminating soils in this area.

The initial results presented here indicate that multifractal modeling and the associated generation of multifractal parameters may be a useful approach in the evaluation of heavy metal pollution in soils and the identification of major sources of of heavy metal contamination. Finally, the fact that $\Delta f(\alpha)$ yield larger differences than compared with the Δa and $\tau''(1)$ value means that $f(\alpha)$ values may be more useful than Δa and $\tau''(1)$ values during the determination of the multifractal characteristics of datasets analyzed using this method.

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Table 1. Statistical analysis of soil geochemical data from the towns of Daxing and Yicheng.

Town	Element	Concentrations						CV*
		Min	Max	Mean	Standard deviation	Skewness	Kurtosis	(%)
Daxing	Cu (mg/kg)	19.00	111.50	33.87	13.26	3.20	14.93	39.16
	Pb (mg/kg)	18.90	291.30	39.57	35.03	5.37	35.41	88.51
	Zn (mg/kg)	40.90	526.10	105.8	94.40	2.91	8.59	89.19
	Cd (mg/kg)	0.045	1.48	0.23	0.24	3.45	13.81	108.23
	As (mg/kg)	4.93	308.20	13.97	33.89	8.72	76.64	242.56
	Hg (mg/kg)	0.03	0.60	0.11	0.11	2.68	7.78	107.29
Yicheng	Cu (mg/kg)	9.60	37.80	24.34	5.77	-0.38	0.41	23.71
	Pb (mg/kg)	10.40	46.30	22.77	4.91	0.87	5.51	21.56
	Zn (mg/kg)	20.80	194.80	54.70	21.43	3.45	20.27	39.17
	Cd (mg/kg)	0.054	0.43	0.15	0.08	1.84	3.49	51.85
	As (mg/kg)	2.30	44.20	7.29	4.39	6.68	56.55	60.24
	Hg (mg/kg)	0.02	0.62	0.06	0.07	5.75	41.26	113.09

*CV: coefficient of variation.

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Table 2. Multifractal parameters that describing the multifractality of all of the elements within the soil samples analyzed during this study.

Town	**Element**	a_{min}	a_{max}	Δa_L	Δa_R	Δa	$\Delta f(a)$	$\tau''(1)$
Daxing	Cu	1.733	2.057	0.280	0.044	0.324	1.270	-0.015
	Pb	1.439	2.050	0.567	0.044	0.611	1.659	-0.068
	Zn	1.733	2.109	0.288	0.088	0.376	0.841	-0.066
	Cd	1.482	2.285	0.499	0.304	0.803	1.358	-0.066
	As	1.285	2.094	0.739	0.070	0.809	1.490	-0.243
	Hg	1.780	2.191	0.248	0.163	0.411	0.656	-0.079
Yicheng	Cu	1.971	2.067	0.036	0.060	0.096	0.168	-0.007
	Pb	1.900	2.062	0.104	0.058	0.162	0.646	-0.005
	Zn	1.729	2.112	0.275	0.108	0.383	1.275	-0.016
	Cd	1.800	2.103	0.201	0.102	0.303	0.829	-0.023
	As	1.659	2.076	0.343	0.075	0.418	1.224	-0.036
	Hg	1.507	2.084	0.497	0.080	0.577	1.243	-0.096

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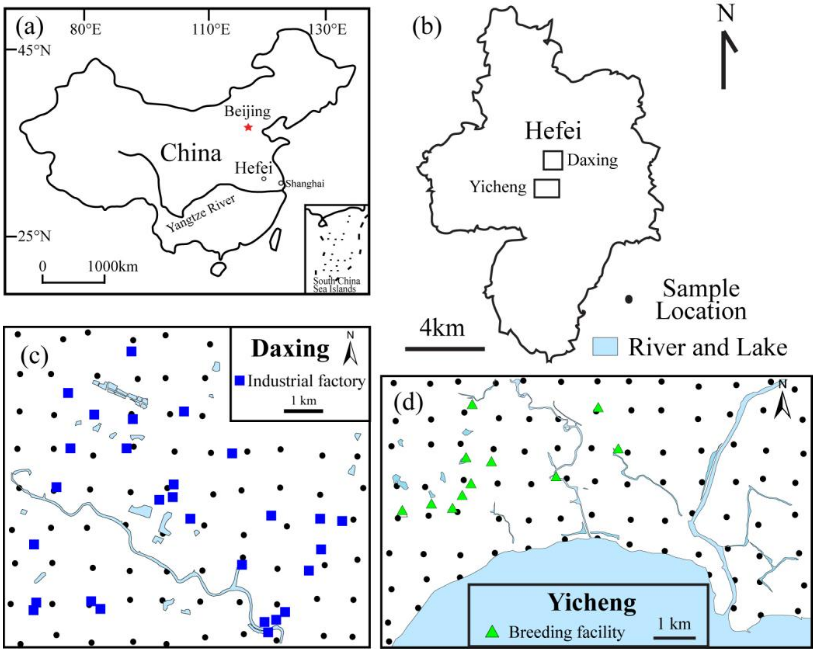

Fig.1. (a) Map showing the location of Hefei in central-eastern China; (b) Map showing the location of the study areas within Hefei; (c) Map showing the location of soil samples taken in a 1 x 1 km grid in the town of Daxing; (d) Map showing the location of soil samples taken in a 1 x 1 km grid in the town of Yicheng.

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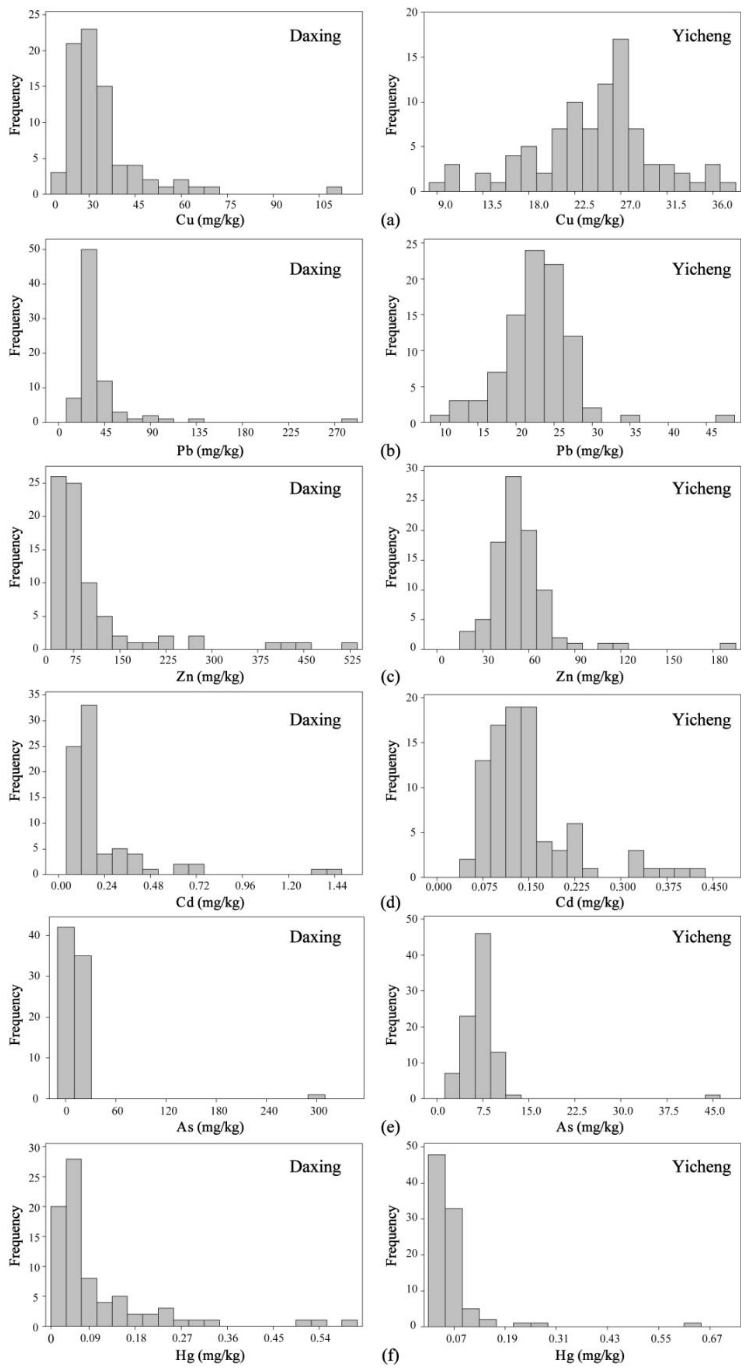

Fig. 2. Histograms showing the distribution of Cu (a), Pb (b), Zn (c), Cd (d), As (e) and Hg (f) concentrations within soils from the towns of Daxing and Yicheng.

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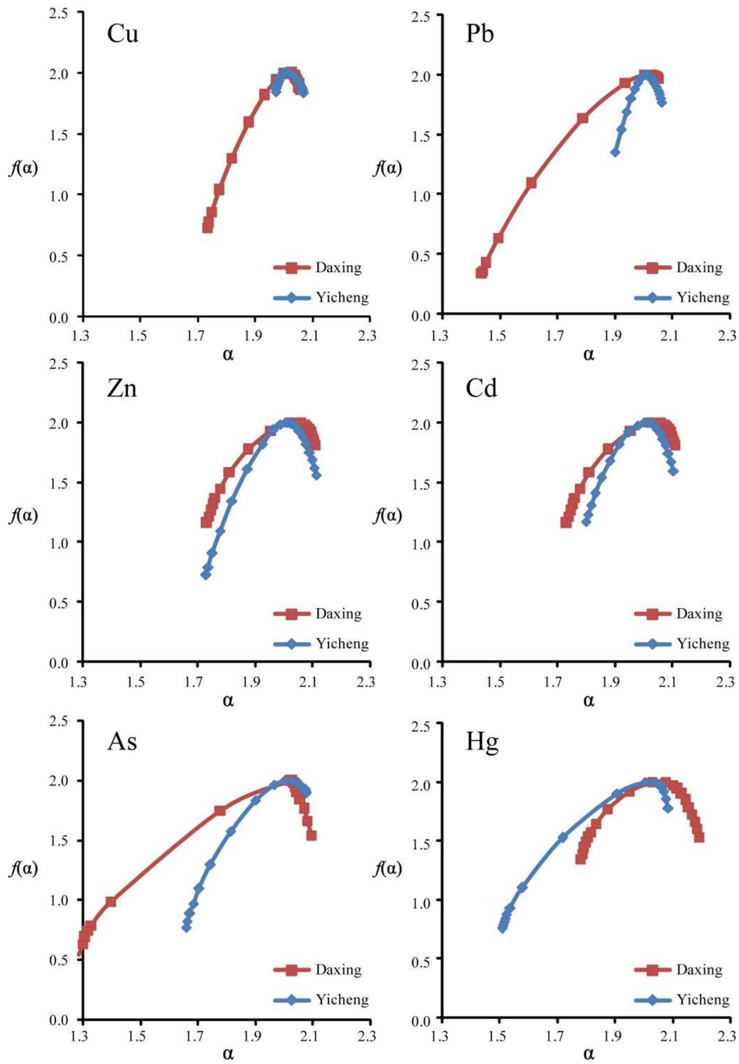

Fig. 3. Multifractal spectra ($f(\alpha)$ vs α) for soil samples analyzed during this study, showing the multifractal characteristics within all datasets barring the Cu data from the Yichen area, which gives a good indication of the behavior of a metal with typical (i.e. non-anthropogenic) background concentrations.

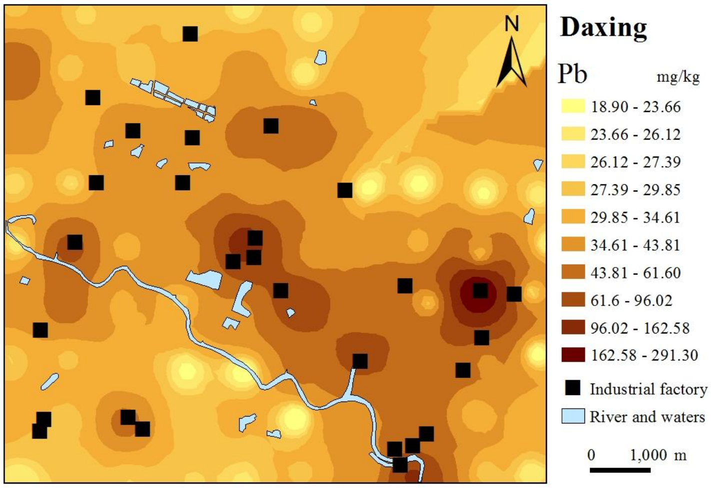

Fig. 4. Filled contour maps produced by inverse distance weighted interpolation showing the spatial distribution of Pb in the Daxing area and the clear correlation between Pb contamination and the location of heavy industry.

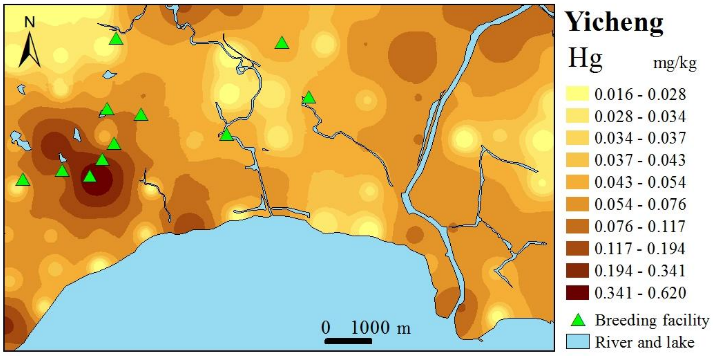

Fig. 5. Filled contour maps produced by inverse distance weighted interpolation showing the spatial distribution of Hg and the clear spatial relationship with the location of breeding facilities in the Yicheng area.

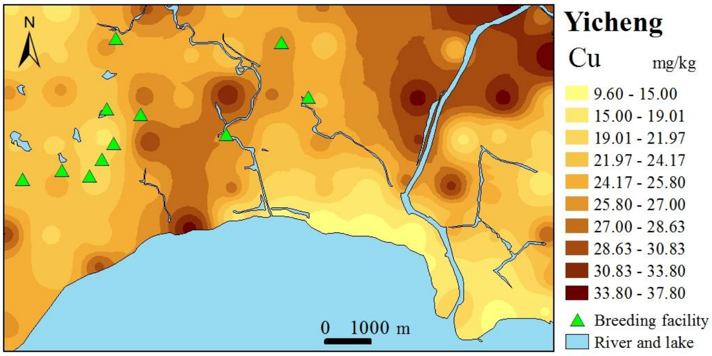

Fig. 6. Filled contour maps produced by inverse distance weighted interpolation showing the spatial distribution of Cu and the location of breeding facilities in the Yicheng area; this distribution shows that the distribution of Cu in soils in this region is unlike the Hg contamination in this area.