Dear Prof. Schertzer and Dr. Miras-Avalos,

We thank these careful and constructive reviews of our manuscript. We have uploaded our response as a supplement to the comments and have incorporated these changes to our revised manuscript. For clarity, we have used a blue font for the reviewer's text, a black font for our text. We hope that after these revisions our manuscript will be considered suitable for publication in Nonlinear Processes in Geophysics.

Abstract:

Line 22: "city. We used a multifractal" instead of "city and use a multifractal".

We have used "city. We used a multifractal" instead of "city and use a multifractal".

Line 25: Remove "for these soil geochemical data".

We have removed "for these soil geochemical data".

Lines 28-31: Please, consider re-phrasing this sentence. For instance, I propose to reduce it to "The degree of multifractality suggests that the differing economic activities in Daxing and Yicheng generate very different heavy metal pollution loads".

We agree to this suggestion and re-phrasing this sentence.

Introduction and overview of the study area:

Line 47: "may allow for assessing" instead of "can investigate".

We have used "may allow for assessing" instead of "can investigate".

Line 48: "with pollutants, as well as" instead of "with pollutants and as well as".

We have used "with pollutants, as well as" instead of "with pollutants and as well a".

Line 61: Remove "in this case".

We have removed "in this case".

Lines 72-73: Please, check the order of citations here. Besides, please, consider reducing the number of citations seems I think there are far too many than necessary.

We have reduced some citations here.

Study area and geochemical data:

Line 109: Remove "was" from "was monitored".

We have removed "was" from "was monitored".

Multifractal spectrum analysis:

Check the readability of equation 1. Parenthesis and symbols are overlapping.

It is a format conversion error. We have used Adobe acrobat to convert the ms-doc file to pdf file again. Now it is correct.

Lines 141-142: You should indicate what are α and $f(\alpha)$.

We have updated this sentence to "The values of $\tau(q)$ derived using this equation can be then used to determine singularity α and fractal spectra $f(\alpha)$ values using a Legendre transformation".

Line 146: "a given dataset" instead of "the dataset in question". Indicate the symbols that you used for "left and right branches".

We have used "a given dataset" instead of "the dataset in question", and indicated $\Delta\alpha_L$ and $\Delta\alpha_R$ in this sentence.

Lines 148-150: Check the readability of equations 4 to 6. The "min" is not visible.

It is a format conversion error. We used Adobe acrobat to convert the ms-doc file to pdf file again. Now it is correct.

Lines 156-160: Re-phrase, not clear.

We have rephrased this sentence to "In addition, local multifractality $\tau''(1)$, which may determined by ordinary spatial analysis functions (autocorrelations and semivariograms), can also be used as a measure to quantitatively characterize the multifractality of a dataset using equation 8".

Line 164: "monofractal" instead of "single fractal".

We have used "monofractal" instead of "single fractal".

Lines 165-168: Consider removing, already stated in lines 77-80.

We have removed the sentence between 77-80.

Geochemical analysis results

Lines 173-174: Ok, but the minimum and the mean were higher in the Daxing area.

The minimum of Cd in Yicheng area is a little bit higher than Daxing area, so in this sentence we didn't include the minimum in this sentence.

Line 185: I do not think that you need multifractals to discriminate this.

We have updated this sentence to "This means that multifractal techniques are highly suited for the characterization of the geochemistry of the soils".

Calculation processes of multifractal spectrum and discussion

Lines 196-197: "using a range of q values from -10 to 10 with and interval of 1" should be stated in the "multifractal spectrum analysis" section.

We have moved these words to the "multifractal spectrum analysis" section.

Line 212: "have a high multifractality" instead of "have highly multifractality".

We have used "have a high multifractality" instead of "have highly multifractality"

Line 215: It should be $\Delta\alpha$ instead of α.

We have revised this, used $\Delta\alpha$ instead of α.

Line 222: "but not maximum", if you look at your histogram (figure 2) you will see that most of the samples from Yicheng were below 0.31 mg/kg of Hg, whereas in Daxing there are more samples above 0.5 mg/kg of Hg.

Thanks for this comment, we have removed the "but not maximum".

Line 229: What do you mean by "deleterious effects such as the heavy metal pollution of people, crops and animals"?

In order to make this point more clear, we have rephrase this sentence to "This means that although contamination in both areas needs to be evaluated further and should be remediated to avoid any deleterious effects, the fact that the Hg contamination in the Yicheng area may be more bioavailable and may have a larger effect on the population of this region (as a result of the agricultural activity in this area) means it should be considered a priority".

Line 236: "the elements within the samples from" instead of "different elements within".

We have used "the elements within the samples from" instead of "different elements within".

Lines 238-245: Please, remove all references to standard deviation because it is not used for sorting the elements according to table 3.

We have revised these lines as follows: "The data shown in Table 3 indicates that the Pb data within the Daxing area has close to the lowest coefficient of variation, but largest the $\Delta f(\alpha)$ and $\tau''(1)$ values for these Pb data are indicative of strongest multifractality compared to the other heavy metals in the soils within the Daxing area. In comparison, the As data for soils in the Daxing area yielded the largest coefficient of variation but the moderate $\Delta f(\alpha)$ and $\tau''(1)$ values, indicating these As data only have moderate multifractality. These differences indicate that the multifractal parameters reveal new information about the nonlinear variability and the characteristics of these geochemical data compared to the basic statistics for these samples. ".

Line 262: “indicate” instead of “indicates”.

We have used “indicate” instead of “indicates”.

Line 276: “polluting more than others” instead of “more polluting than others”.

We have used “polluting more than others” instead of “more polluting than others”.

Lines 296-297: How was spatial density assessed? It is not explained anywhere.

We have explained it in the title of Fig. 7 as follows: "generated using the Kernel Density method within spatial analyst tools of the ArcGIS software package)".

Lines 302-305: When you are talking about correlation value, do you refer to coefficient of correlation? Could you indicate the p-values for these correlations, please?

Yes, we have used "coefficient of correlation" instead of "correlation value" in these lines and the title of Table. 4. We are sorry that ArcGIS software can not provide p-values for the correlation matrix, but we think the coefficient of correlation can be used to compare the correlation between two layers very well.

Line 308: “do” instead of “does”.

We have used “do” instead of “does”.

Lines 308-311: Please, re-phrase, not clear.

We have rephrased this sentence as follows: "The negative correlation coefficient, symmetrical distribution and weakest multifractality of Cu give one clue to the spatial relationship between Cu contamination and the river in the right hand side of Fig. 6."

Line 339: Include a parenthesis after “water”.

We have revised this sentence to " In addition, the source of the Hg contamination (e.g. fertilizer, fodder, pesticides, water, or some other source) remains unclear."

Line 340: Remove “or other source”.

We have removed “or other source”.

Line 346: Inverse distance weighted interpolation is not mentioned or explained in the methodology.

The Inverse distance weighted interpolation is an well known interpolation method. In this paper, we used this method provided by ArcGIS software to interpolate the data. Because this method is not the key point of this paper, we revised the title of Fig. 4~ Fig. 6 to show the method and software we have used. If the reader want to know the detail about this method, they can find it in the manual of the ArcGIS software package.

Line 360: How was this density map created? This should be explained in the methods.

We have revised the title of Fig. 7 to show the method and software we have used.

Table 4: How was this correlation matrix made? It is not explained in the methodology.

We have revised the title of Table 4 to show the method and software we have used.

Lines 365-367: There is no figure 8.

We have revised it.

Lines 368-371: Is this needed?

We have removed these sentences.

Conclusions:

Lines 374-376: Consider removing the first sentence.

We have removed the first sentence.

Line 382: Remove "the overall order in soils".

We have removed "the overall order in soils".

Line 390: Remove the word "well".

We have removed "well" from this sentence.

Lines 393-396: I think that this sentence can be removed.

We have removed this sentence.

References:

Lines 417-418: Caniego et al. 2005 are not cited in the text. Please, remove.

Caniego et al. 2005 are cited in Line 64.

Lines 429-431: Dathe et al. 2006 are not cited in the text. Please, remove.

Dathe et al. 2006 are cited in Line 64.

Lines 445-447: Hasley et al. 1986 are not cited in the text. Please, remove.

We have removed this reference.

Lines 469-474: McGrath et al. 2004 should come before Mulligan et al. 2001

We have moved the McGrath et al. 2004 before Mulligan et al. 2001.

We thank again for these positive comments and helping us revising the confusing sentences within our paper. We hope that this manuscript is now acceptable for publication with the corrections and edits noted above. Please do not hesitate to contact me if you need any more information on or clarification of these revisions.

Yours faithfully,
Feng Yuan

Comparison of the multifractal characteristics of heavy metals in soils within two areas of contrasting economic activities in China

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Abstract

Industrial and agricultural activities can generate heavy metal pollution that can cause a number of negative environmental and health impacts. This means that evaluating heavy metal pollution and identifying the sources of these pollutants, especially in urban or developed areas, 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 in Hefei city. We ~~and~~ use a multifractal spectral technique to identify and compare the multifractality of heavy metal concentrations of soils within the industrial Daxing and agricultural Yicheng areas. This paper uses three multifractal parameters ($\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$) ~~for these soil geochemical data~~ to indicate the overall amount of multifractality within the soil geochemical data. The results show all of the elements barring Hg have larger $\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$ values in the Daxing area compared to the Yicheng area. The ~~differences in the~~ degree of multifractality suggests~~between Daxing and Yicheng areas indicate that the soils in these areas have differing multifractal geochemical characteristics, suggesting~~ that the differing economic activities in Daxing and Yicheng ~~these areas~~ generate very different heavy metal pollution~~pollutant~~ loads. In addition, the industrial Daxing area contains significant Pb and Cd 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. The results also indicate that multifractal modeling and the associated generation of multifractal parameters can be a useful approach in the evaluation of heavy metal pollution in soils.

Keywords: soil geochemistry; multifractal modelling; heavy metal pollution; Hefei

1. Introduction and overview of the study area

Heavy metal pollution within soil poses a serious risk for human health and the environment, and thus soil pollution caused by anthropogenic activities (including industry and agriculture) has been the focus of a significant amount of research (e.g., Leyval et al., 1997; Thomas and Stefan, 2002; McGrath et al., 2004; Wang et al., 2007; Luo et al., 2011). Analyzing soil geochemistry and pollution using multifractal techniques may allow for assessing~~can investigate~~ many of the problems of nonlinear variability which commonly arise when dealing with pollutants,~~, and~~ as well as enabling the identification of non-linear characteristics within datasets. This approach can yield new information that can be used to understand the factors controlling the distribution of key elements within the objects or data being studied (Salvadori, 1997; Gonçalves, 2000; Zuo et al., 2012). This in turn means that determining the multifractal characteristics of the distribution of heavy metals in soils can improve our understanding of any heavy metal pollution that is associated with these differing anthropogenic activities.

Multifractal techniques include singularity mapping and multifractal interpolation that enable more detailed analysis of the spatial distribution of heavy metals, concentration-area modeling that can be used to define threshold values between background (i.e. geological) and anthropogenic anomalies (Lima et al., 2003), spectral density-area modeling that can be used to define thresholds to separate anomalies (i.e., anthropogenically derived heavy metal concentrations in this case) from background concentrations (i.e., geologically derived heavy metal concentrations; Cheng, 2001), and multifractal spectra that highlights non-linear characteristics and identifies anomalous behavior that reflects the characteristics of some multifractal sets (Gonçalves, 2000; Albanese et al., 2007; Guillén et al., 2011), such as the presence of porous structures and spatial variations in soil properties (Caniego et al., 2005; Dathe et al., 2006). This means that multifractal techniques can be useful tools for the analysis of heavy metal pollution within soils (e.g., Salvadori et al., 1997; Lima et al., 2003; Albanese et al., 2007; Guillén et al., 2011). These multifractal techniques are not only used in environmental science, but also in a number of differing fields, including geophysics (Schertzer et al., 2011), medicine (Jennane et al., 2001), computer science (Wendt et al., 2009), geology (Cheng, 1995; Deng et al., 2011; ~~Zuo et al., 2012, 2014;~~ Yuan et al., 2012, 2015)~~2015; Nazarpour et al., 2014)~~ and ecology ~~((Scheuring and Riedi, 1994;~~ Pascual et al., 1995), among others.

Hefei is the capital of Anhui Province, China, and has an urban area that includes the towns of Daxing and Yicheng, which focus on industrial and agricultural activities, respectively. Here, we use multifractal spectra techniques and three parameters ($\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$) to analyze and compare the degree and characteristics of the multifractality of heavy metal contamination in soils associated with anthropogenic activities in this region. The results will further enable and inform future planning for any necessary remediation of the soils in the Daxing and Yicheng areas.

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 areas of Hefei and containing numerous factories that are involved in the steel industry, chemical industry, paper making, and the production of furniture and construction materials, among others. In contrast, the town of Yicheng focuses its economic activities on agricultural production, byproduct processing, livestock and poultry breeding, ornamentals, 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-related contamination. A total of 169 surface (<20 cm depth) 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 air-dried 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) were determined during this study, with Cd, Cu, Pb and Zn concentrations determined by inductively coupled plasma–mass spectrometry (ICP–MS), whereas Hg and As concentrations were determined by hydride generation–atomic fluorescence spectrometry (AFS; Armstrong et al., 1999; Gómez-Ariza et al., 2000). 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 data was monitored by repeat and replicate determinations using instrumental neutron activation analysis (INAA), with analytical precision ~~was~~ monitored using variance of the results obtained from duplicate analyses.

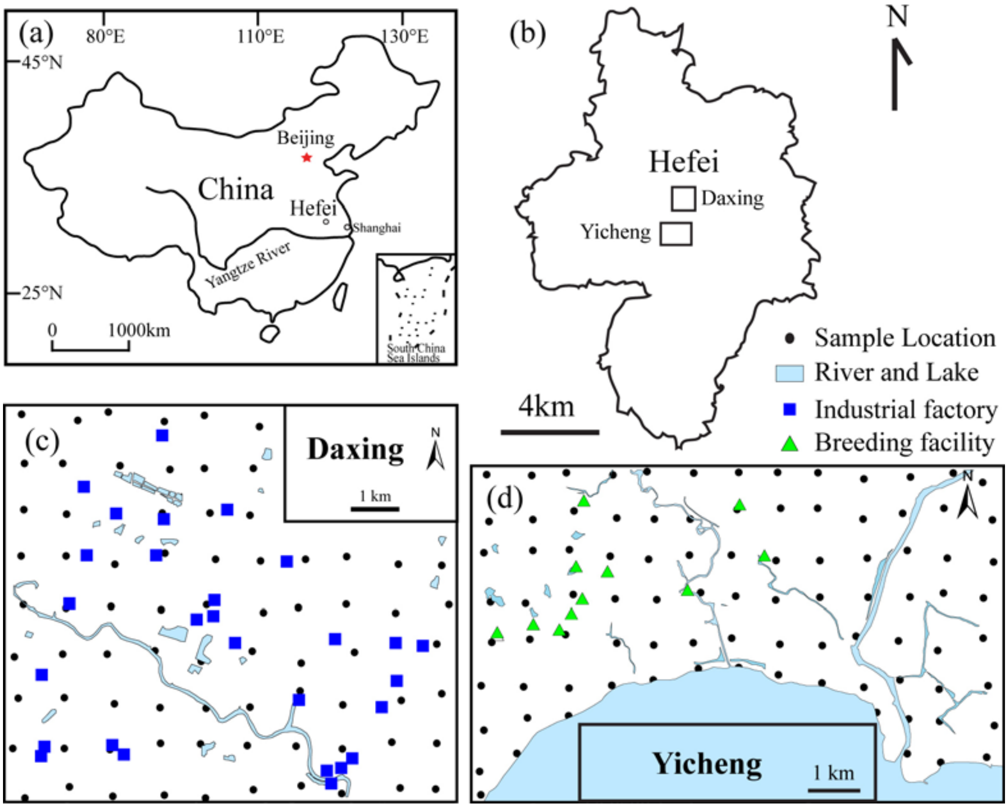

Fig. 1. Location of Hefei in central-eastern China (a); location of the study areas within Hefei (b); the 1 × 1 km grids used for soil sampling in the towns of Daxing (c) and Yicheng (d)

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 factors controlling the distribution of key elements within the data. Fractal spectra ($f(\alpha)$) are 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 are the box-counting and gliding box methods, both of which are based on the moment method.

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 that are available for statistical estimation within a dataset (Buczkowski et al., 1998; Tarquis et al., 2006; Xie et al., 2010). 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):

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

where $\mu_i(\varepsilon)$ denotes a measure with the i_{th} cell of a gliding box of size ε, q is the order moment of this measure (this paper used a range of q values from −10 to 10 with an interval of 1), ~~;~~ <> indicates the statistical moment, and $N^*(\varepsilon)$ indicates the total number of gliding boxes of size ε with $\mu_i(\varepsilon)$ values different from 0.

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

$$\alpha(q) = \frac{d\tau(q)}{dq} \quad (2)$$

$$f(\alpha) = q\alpha(q) - \tau(q) = q\frac{d\tau(q)}{dq} - \tau(q) \quad (3)$$

$\Delta\alpha$ and Δf are essential parameters required to analyze the multifractal characteristics of a given ~~the~~ dataset. ~~in question.~~ The widths of the left ($\Delta\alpha_L$) and right ($\Delta\alpha_R$) branches within the multifractal spectra are then defined using the following equations:

$$\Delta\alpha_L = \alpha_0 - \alpha_{\min} \quad (4)$$

$$\Delta\alpha_R = \alpha_{\max} - \alpha_0 \quad (5)$$

$$\Delta\alpha = \alpha_{\max} - \alpha_{\min} \quad (6)$$

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

$$\Delta f(\alpha) = f(\alpha_{max}) - f(\alpha_{min}) \tag{7}$$

Higher $\Delta\alpha$ and $\Delta f(\alpha)$ values are generally indicative of datasets with more heterogeneous patterns (ordered, complex, clustered) and higher levels of multifractality (Cheng, 1999; Kravchenko et al., 1999). In addition, local multifractality $\tau''(1)$, which may determined by ordinary spatial analysis functions (autocorrelations and semivariograms), can also be used as a measure to quantitatively characterize the multifractality of a dataset using equation 8~~, where ordinary spatial analysis functions (autocorrelations and semivariograms) are related to low order statistical moments (0 to 2nd) that may determine τ″(1)~~ (Cheng, 2006):

$$\tau''(1) = \tau(2) - 2\tau(1) + \tau(0) \tag{8}$$

If μ is a multifractal and $-D < \tau''(1) < 0$, where D is the gliding-box dimension, then more negative values of $\tau''(1)$ are indicative of higher degrees of multifractality, whereas otherwise $\tau''(1) = 0$ for mono~~a single~~ fractal.

~~Here, we use the three multifractal parameters described above (Δα, Δf(α) and τ″(1)) to better identify the degrees of multifractality within the soil geochemical data for the study area as well as enabling the comparison of the multifractality of differing elements in the soils in this region.~~

4. Geochemical analysis results

A statistical summary of the soil geochemical data for the study area is given in Table 1. Samples from the Daxing area have higher Cu, Pb, Zn, Cd and As maximum, mean, standard deviation, skewness, and kurtosis values than soil samples from the Yicheng area, whereas the Yicheng area has a higher maximum Hg concentration value than the Daxing 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 higher and more variable concentrations of these elements. This also suggests that samples from the Daxing area containing elevated concentrations of heavy metals were probably contaminated by anthropogenic activity.

All of the elements (barring Pb and Cu in the Yicheng area) in both the Yicheng and Daxing areas yielded concentration histograms that are positively skewed and contain some outliers (Fig. 2), indicating that these data have non-normal and potentially fractal- or multifractal-type distributions. This means that multifractal techniques are highly suited for the characterization of the geochemistry of the soils. ~~and discrimination of the differing types of human activities ongoing in each area.~~

Table 1. Summary statistics of soil heavy metal concentrations within samples from the Daxing and Yicheng areas.

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

*CV: coefficient of variation.

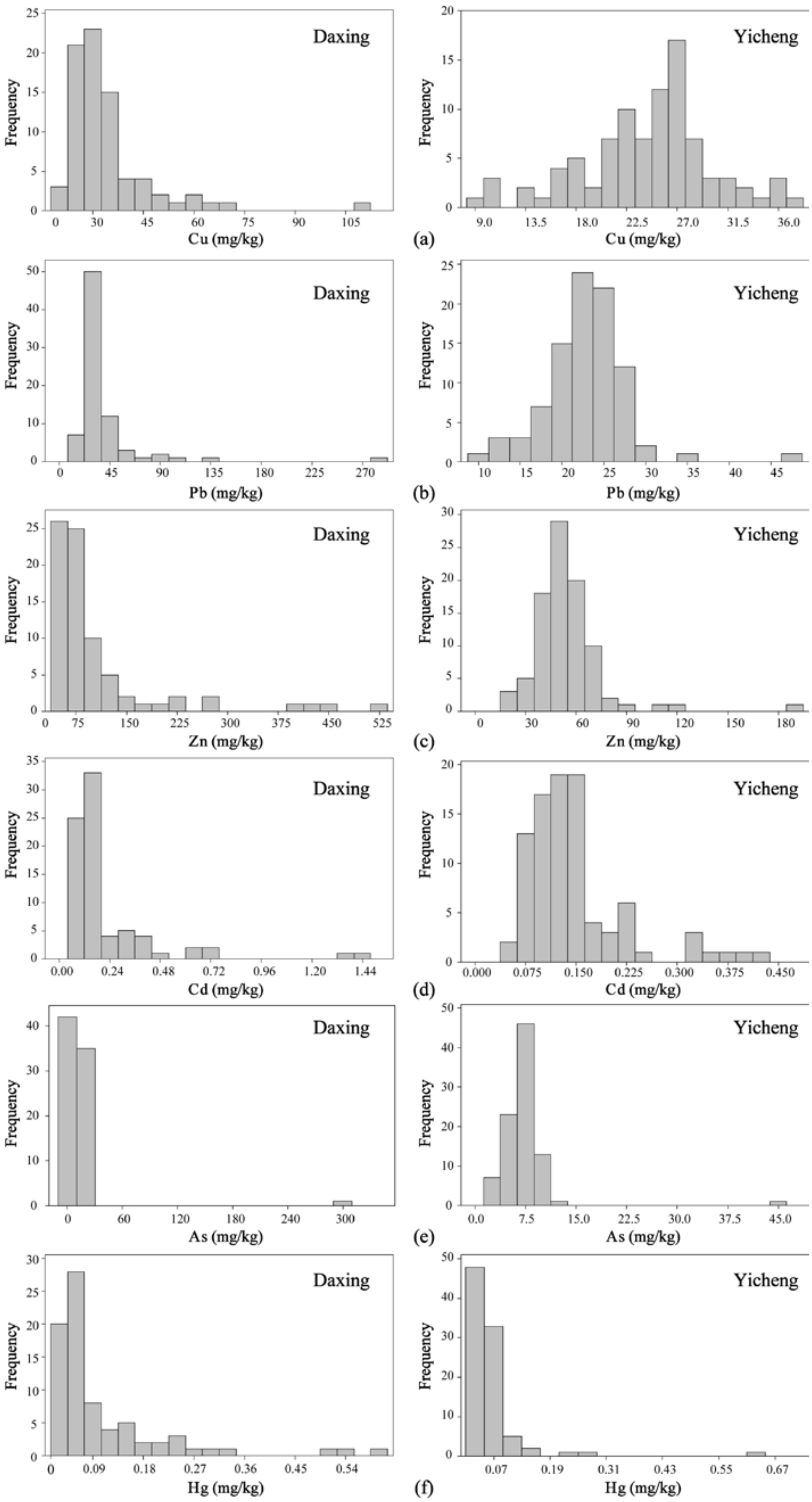

Fig. 2. Histograms showing the distribution of Cu (a), Pb (b), Zn (c), Cd (d), As (e) and Hg (f) concentrations within soils from the towns of Daxing and Yicheng.

5. Calculation processes of multifractal spectrum and discussion

The multifractal spectra (in the form of an α–$f(\alpha)$ diagram) for the geochemical data are shown in Fig. 3.~~3 using a range of q values from −10 to 10 with an interval of 1.~~

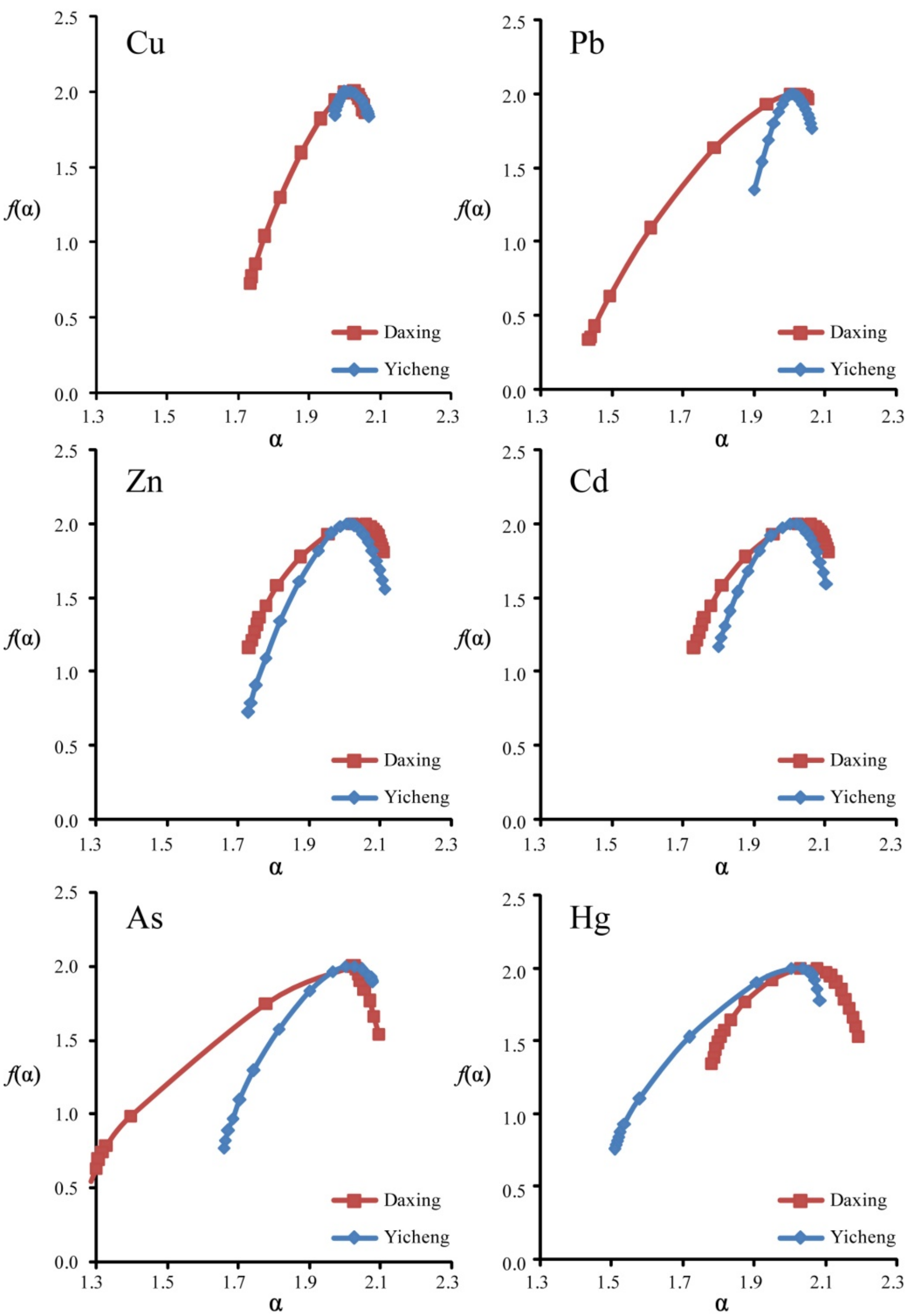

Fig. 3. Multifractal spectra ($f(\alpha)$ vs α) of the soil geochemical data from the Daxing and Yichen area.

These multifractal spectra have inverse bell shapes (Fig. 3) and are asymmetric (i.e. $\Delta\alpha_L$ values significantly differ from $\Delta\alpha_R$, equations 5-6) with the exception of the Cu data for soils from the Yicheng area, indicating that the samples containing low and high concentrations of these elements are not evenly distributed within the study area (as is expected for areas containing point source pollutants like factories or animal breeding facilities).

The multifractal results given 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 with $\tau''(1)$ values less than –0.01 and $\Delta f(\alpha)$ values larger than 0.5, all of which indicate that these elements have a high~~highly~~ multifractality within the soils in these two areas. All of the elements analyzed during this study (barring Hg) have higher $\Delta f(\alpha)$ and α values (except Zn) and lower $\tau''(1)$ values in soils from the Daxing area, with Hg having higher $\Delta f(\alpha)$ and $\Delta\alpha$ and lower $\tau''(1)$ values in soils from the Yicheng area (Table 2). This suggests that the industrial activities in the Daxing area generate multi-element heavy metal soil contamination, whereas the most significant heavy metal pollution associated with the agricultural activity in the Yicheng area is Hg contamination. The $\Delta f(\alpha)$ and $\Delta\alpha$ values of Hg in the Yicheng area are larger than the values for all 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, even considering the lower overall ~~(but not maximum)~~ concentrations of Hg within the Yicheng area compared to the Daxing area. This contamination should be considered a priority in terms of remediation, because the interaction between the agricultural activity in the Yicheng area and this Hg pollution could seriously impact human health, as Hg is preferentially concentrated upward in the food chain (e.g. (Jiang et al., 2006)). This means that although contamination in both areas needs to be evaluated further and should be remediated to avoid any deleterious effects~~, such as the heavy metal pollution of people, crops and animals,~~ the fact that the Hg contamination in the Yicheng area may be more bioavailable and may have a larger effect on the population of this region (as a result of the agricultural activity in this area) means it should be considered a priority.

Table 2. Multifractal parameters of the elements analyzed during this study.

Town	Element	α_{min}	α_{max}	$\Delta\alpha_L$	$\Delta\alpha_R$	$\Delta\alpha$	$\Delta f(\alpha)$	$\tau''(1)$
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

In order to compare variations in multifractality, the~~different~~ elements within the samples from Daxing and Yicheng area were sorted by $\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$ parameters, respectively, in addition to sorting by ~~basic statistics such as standard deviation and~~ coefficient of variation values (Table 3). The data shown in Table 3 indicates that the Pb~~Zn~~ data within the Daxing area has close to the lowest~~largest standard deviation value but only a moderate~~ coefficient of variation, but largest the~~ $\Delta\alpha$ and~~ $\Delta f(\alpha)$ and $\tau''(1)$ values for these Pb~~Zn~~ data are indicative of strongest~~only weak~~ multifractality compared to the other heavy metals in the soils within the Daxing area. In comparison, the As~~Hg~~ data for soils in the Daxing~~Yicheng~~ area yielded the largest coefficient of variation~~lowest standard deviation value~~ but the moderate $\Delta f(\alpha)$~~largest $\Delta\alpha$~~ and $\tau''(1)$ values, indicating these As~~Hg~~ data only have moderate~~strong~~ multifractality. These differences indicate that the multifractal parameters~~ $\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$~~ reveal new information about the nonlinear variability and the characteristics of these geochemical data compared to the basic statistics for these samples. In addition, the data given in Table 3 indicates that these elements have different orders depending on whether they are sorted by $\Delta\alpha$, $\Delta f(\alpha)$ or by $\tau''(1)$ values, all of which reflects differing aspects of the multifractality of these data. Here we consider that $\Delta\alpha$, $\Delta f(\alpha)$ or by $\tau''(1)$ have equal weightings that reflect the overall multifractality of the data from the study area. As such, the ordering of these elements by $\Delta\alpha$, $\Delta f(\alpha)$ or by $\tau''(1)$ involved the summation of these values with the summed ordering then sorted again to compare the overall multifractality of these data.

Table 3. Elements sorted by multifractal parameters and basic statistic indices.

Town	Element	Order				
		Basic statistics	Multifractal parameters			
		Coefficient of variation	$\Delta\alpha$	$\Delta f(\alpha)$	$\tau''(1)$	Overall*
Daxing	Cu	6	6	4	6	6
	Pb	5	3	1	1	1
	Zn	4	5	5	2	4
	Cd	2	2	3	3	2
	As	1	1	2	5	3
	Hg	3	4	6	4	5
Yicheng	Cu	5	6	6	5	6
	Pb	6	5	5	6	5
	Zn	4	3	1	4	3
	Cd	3	4	4	3	4
	As	2	2	3	2	2
	Hg	1	1	2	1	1

Overall: the overall order of $\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$.

The overall amount of multifractality within the soil geochemical data for the Daxing area decreases as follows: Pb>Cd>As>Zn>Hg>Cu, whereas the overall amount of multifractality within the soil geochemical data for the Yicheng area decreases as follows: Hg>As>Zn>Cd>Pb>Cu. The overall orders indicates that the Pb and Hg soil data have the highest degree of multifractality in the Daxing and Yicheng areas, respectively, whereas Cu has the weakest multifractality irrespective of the area.

We further analyzed the spatial distribution of contamination within soils from the Daxing and Yicheng areas and evaluated whether there is any significant correlation between multifractality and anthropogenic activity. Filled contour maps showing the distribution of Pb in the Daxing area and Hg and Cu in the Yicheng area were calculated using inverse distance weighted interpolation (Fig. 4–6). These figures show that areas with elevated levels of Pb contamination within the Daxing area are correlated with the location of industrial factories, although interestingly the areas in the upper and lower left hand side of Fig. 4 contain factories but not elevated concentrations of Pb. This indicates that the Pb concentrations in these soils may be dependent on both the presence and type of industry in this area, with some industries ~~more~~ polluting more than others, either as a direct result of the differing industries present in this area or as a result of differing (or a lack of in some areas) approaches to lessening environmental impacts. In comparison, the Hg contamination in the Yicheng area is definitely spatially correlated with the location of agricultural breeding facilities. Although the mean concentrations of Hg in soils are greater in the Daxing area, all of the multifractal parameters determined during this study ($\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$) indicate that the Hg data in the Daxing area has a lower multifracticality than the Hg data in the Yicheng area. The Yicheng area is heavily agricultural, meaning that the agricultural activities in this area may be both concentrating Hg as well as contaminating soils. In addition, although the mean concentrations of Hg in the Yicheng area are lower than in the soils in the Daxing area, the former has a higher maximum concentration than the latter, and both areas have significant Hg contamination. Indeed, the contamination in the Yicheng area may be of more concern than the contamination in the Daxing area, as the agricultural activity in the Yicheng area may lead to greater human intake of Hg than from the soils in the mainly industrial Daxing area, a factor that could lead to serious health issues (e.g. Minamata disease) caused by the potential concentration of Hg up the food chain. This indicates that soils in both areas may well require control and remediation.

This distribution of soils with elevated concentrations of Hg also contrasts with the symmetrical distribution and weakest multifractality for Cu within the Yicheng area (Fig. 3, 5-6). Here, we generated a correlation matrix that compares the relationship between the spatial density of breeding locations in the Yicheng area (Fig. 7) and filled contours maps showing the distribution of Hg (Fig. 5) and Cu (Fig. 6) in this region to identify whether there are any spatial correlations between the location of agricultural facilities and areas containing soils with elevated heavy metal concentrations (Table 4). The correlation matrix shows a significant correlation between agricultural facilities and high concentrations of Hg (coefficient of

~~(~~correlation~~ value~~ = 0.434), whereas the location of these agricultural breeding facilities and areas of high Cu concentrations either have no relationship or are negatively correlated (coefficient of ~~(~~correlation~~ value~~ = –0.064). This indicates that very little Cu has been anthropogenically added (or removed) from the soils in the Yicheng area, suggesting that these soils may contain only natural background concentrations of Cu and that the breeding facilities in this area do~~es~~ not produce significant Cu contamination. The negative correlation coefficient,~~matrix,~~ symmetrical distribution and weakest multifractality of~~for~~ Cu give one clue to~~ the derivation of the Cu contamination in this area is~~ the spatial relationship between Cu contamination and the river in the right hand side of Fig. 6. This may suggest a non-anthropogenic source (e.g. flooding causing the deposition of Cu or some other relationship between water and Cu contamination) for some of the slightly elevated Cu concentrations in this region. In addition, the fact that some breeding facilities are not associated with significant Hg contamination (Fig. 5) suggests again that although there is a relationship between the presence of these facilities and contamination, it may be that the Hg contamination in this area reflects differing types of breeding facilities or differing (or a lack of) approaches to lessening environmental impacts.

These results indicate that multifractal modeling and the associated generation of multifractal parameters are a useful approach in the evaluation of heavy metal pollution in soils and the identification of major element of heavy metal contamination. In addition, the differing orders of the multifractality of the geochemical data for soils within the Daxing area and Yicheng area are indicative of a significant difference in the geochemical characteristics (and heavy metal pollution) in the soils within these two areas. This indicates that differing treatment strategy and clean-up approaches to remediating these two polluted areas are needed, rather than a single cover-all strategy and approach to the remediation of heavy metal pollution. A significant number of different remediation approaches can be used to resolve the issues of heavy metal soil contamination (e.g., Bech et al., 2014; Koptsik, 2014). Although somewhat beyond the scope of this study, the multi-element nature of the contamination in the Daxing area means that physical and chemical approaches to remediation (i.e., soil removal, soil vitrification, soil consolidation, electroremediation, or soil washing) are probably well suited for the remediation of heavy metal contaminated soil in this region (especially Pb). In comparison, the differing (i.e. Hg-dominated) type of soil contamination in the Yicheng area could be more efficiently treated using microremediation and phytoremediation, primarily as the agriculture in this area requires a rapid reduction in the mobility and biological availability of heavy metals in the soils (Mulligan et al., 2001; Wang et al., 2006). In addition, the source of the Hg contamination (e.g. fertilizer, fodder, pesticides, water)~~water, or some other source~~ remains unclear. Identifying this source is also beyond the scope of this paper although it is also clearly an area for future research, as the identification of the source or sources of this contamination may prevent the future heavy metal pollution of soils in this region.

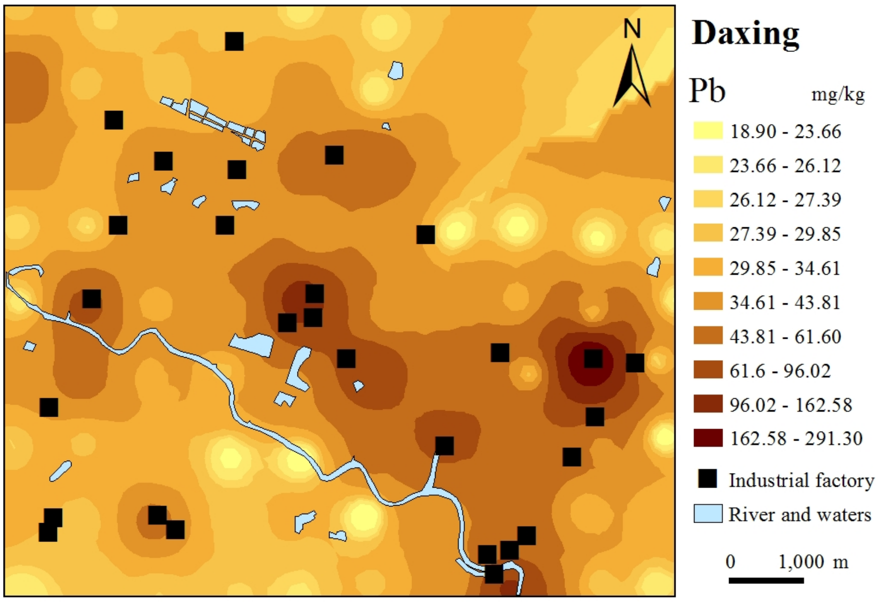

Fig. 4. Filled contour map generated by inverse distance weighted interpolation showing the spatial distribution of soil Pb concentrations in the Daxing area (generated using Inverse Distance Weighted Interpolation method within spatial analyst tools of the ArcGIS software package).

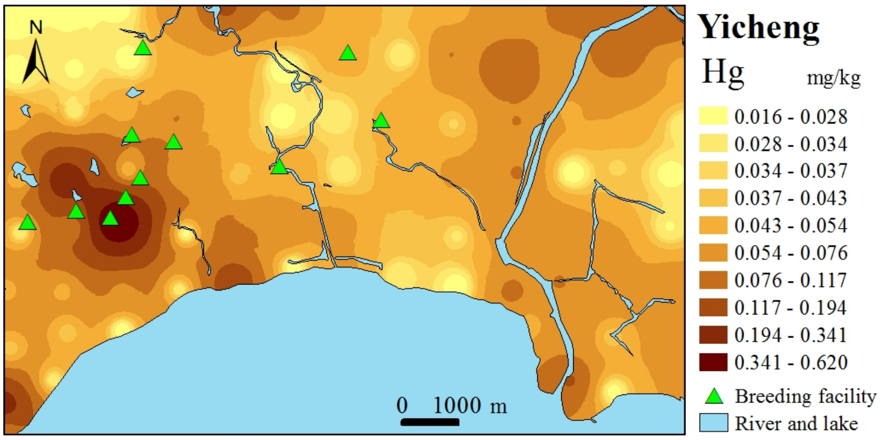

Fig. 5. Filled contour map generated by inverse distance weighted interpolation showing the spatial distribution of soil Hg concentrations in the Yicheng area (generated using Inverse Distance Weighted Interpolation method within spatial analyst tools of the ArcGIS software package).

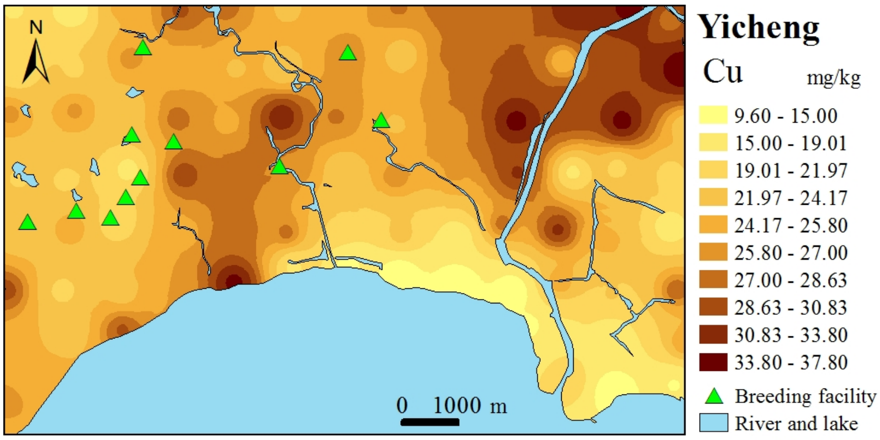

Fig. 6. Filled contour map generated by inverse distance weighted interpolation showing the spatial distribution of soil Cu concentrations and the location of breeding facilities in the Yicheng area (generated using Inverse Distance Weighted Interpolation method within spatial analyst tools of the ArcGIS software package).

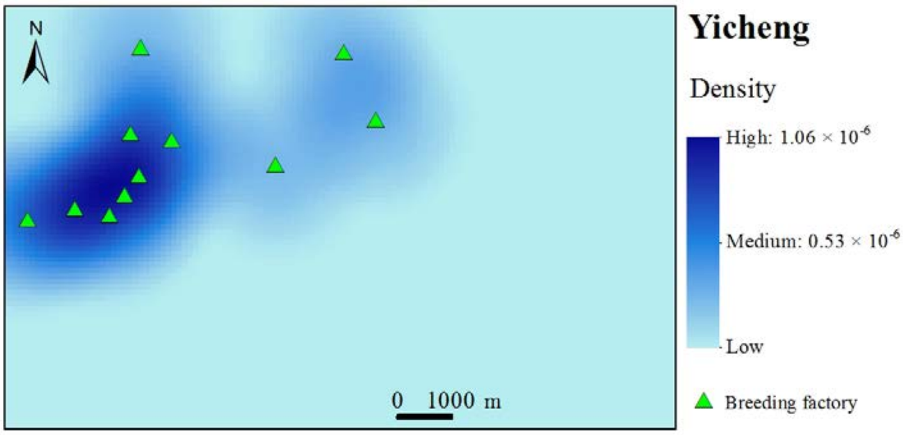

Fig. 7. Density map of breeding facilities in Yicheng area (generated using the Kernel

Density method~~kernel density tool~~ within spatial analyst tools of the ArcGIS software package).

Table 4. Correlation matrix comparing the breeding facility density map and the filled contour maps for Hg and Cu data for the Yicheng area (calculated using the Band Collection Statistics within spatial analyst tools of the ArcGIS software package).~~.~~

Layers	Layer 1	Layer 2	Layer 3
Layer 1	1.000~~00~~	0.434	–0.064
Layer 2	0.434	1.000	–0.464
Layer 3	–0.064	–0.464	1.000

Layer 1: Density map of breeding factories of Yicheng area (Fig. 7);~~8);~~
Layer 2: Filled contour map of Hg concentrations of Yicheng area (Fig. 5);~~8);~~
Layer 3: Filled contour map of Cu concentrations of Yicheng area (Fig. 6).~~8).~~
~~The correlations range from +1 to –1, where a positive correlation indicates a direct relationship between the two layers and a negative correlation means that one variable is negatively correlated with the other. A correlation of zero means that two layers are independent of one another.~~

5. Conclusions

~~This study focuses on the geochemistry of heavy metal contaminated soils from the Daxing and Yicheng areas, both of which are located close to the city of Hefei, in Anhui Province, China.~~ Multifractal modelling and the resulting multifractal parameters in this paper indicate that the soils from the Daxing area have stronger multifractality for Cu, Pb, Zn, Cd and As than soils from the Yicheng area, although the latter have relatively strong multifractality for Hg. The ordering of values for the multifractal parameters $\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$ indicate the degree of multifractality for the geochemical data for soils within the Daxing area descends as follows: Pb>Cd>As>Zn>Hg>Cu, whereas ~~the overall order in soils~~ within the Yicheng area descends as follows: Hg>As>Zn>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 results presented here indicate~~The results presented here indicate that multifractal modeling and the associated three multifractal parameters ($\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$) can efficiently reflect the multifractality caused by industrial and agricultural activities in the Daxing and Yicheng areas, respectively. This in turn indicates~~ that multifractal modeling can be a useful approach in the evaluation of heavy metal pollution in soils and the identification of problematic heavy metals that need remediation in the research area.

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