Dear Prof. Schertzer and Dr. Miras-Avalos,

We thank anonymous referee #1 and Jose Miranda for their 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, and italics for text that is included in the revised manuscript. We hope that after these revisions our manuscript will be considered suitable for publication in Nonlinear Processes in Geophysics.

RC1: Anonymous Referee #1

Introduction:

I did not like this way of introducing because you did not set the basis for performing your study. Anyway, this a personnal opinion. However, certain objective remarks need to be addressed:

Line 54: I would remove "in recent years". You cited here works from 1997, that is 20 years ago, therefore, I do not think is very recent. By the way, in what order did you put your citations? It is neither chronological nor alphabetical! Please, edit according to journal's instructions.

We have removed "in recent years" and have listed all of the citations in chronological order.

Lines 55-59: Please, try to reduce this sentence because it is too long.

We have reorganized this sentence and divided it to two sentences to make it clearer.

Lines 61-62: I would remove "using multifractal techniques to" and substitute "determine" for "determining".

We have removed "using multifractal techniques to" and substituted "determine" for "determining".

Line 63: I would use other term instead of "further". Maybe "enhance" or "improve".

We have used "improve" instead of "further".

Line 64: What activities are you referring to?

We have used "anthropogenic" to restrict the "activities".

Lines 65-74: This is somewhat confusing since you did not define any threshold, separated anomalies or identified behaviours in your study. At least I did not understand it that way.

This section introduces the potential uses of several different multifractal techniques and indicate that multifractal techniques are useful techniques for understanding the distribution of heavy metals in soils. As such, we state that "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".

Line 76: Remove “and so on”.

We have removed “and so on”.

Line 78: “polluntation”??? Do you mean “pollution”?

This was a spelling error and we have corrected this.

Line 80: Remove “be used”.

We have removed “be used”.

Lines 74-84: What order did you follow for citations? It is neither chronological nor alphabetical.

We have put all of our citations in chronological order.

Lines 87-90: In this sentence the word “multifractal” is repeated four times, please, re-phrase.

We have rephrased this sentence.

Line 91: Remove “the” before “anthropogenic”.

We have removed “the” before “anthropogenic”.

Study area and geochemical data:

Line 114: Remove “during this study, with”and use a dot to separate the sentence after “determined”. Then begin the new sentence as “The concentrations of”.

We have revised these sentences as suggested by the reviewer.

Line 115: Remove “concentrations” and use “were” before “determined”.

We have removed “concentrations” and have included “were” before “determined”.

Line 116: Use “were” before “determined”.

We have used “were” before “determined”.

Lines 114-117: Please, add references for the methods used for heavy metal determination.

We have added references to provide information on the methods used.

Lines 119-122: This is not clear, please, re-phrase it.

We have rephrased these sentences.

Multifractal spectrum analysis

Lines 148-149: Again, order of citations!

We have put all of our citations in chronological order.

Line 151: Since you used the gliding box method, why explaining the calculation of the boxcounting method in lines 140-144?

The boxcounting method is the basis of the gliding box method used during this study. As such, we originally explained this method as well as the gliding box method; however, in order to more clearly explain the methods used we have removed this paragraph as suggested by the reviewer and have renumbered our equations appropriately.

Line 154: Please, improve the readability of this equation.

We have updated this equation.

Line 159: "$f(\alpha)$" does not appear in equation 4.

We have revised this equation.

Lines 160-161: What is "q" in these equations?

q is the order moment of the measure $\mu_i(\varepsilon)$. We have this description to in Line 134.

Lines 162-163: These symbols do not appear in the equations 3 and 4, why beginning with "where"? You must specify the meaning of the symbols in each equation, otherwise, readers will not know what are you describing mathematically.

We have deleted the "where" in the begining of this sentence.

Lines 173-174: "multifractality associated with ordinary spatial analysis parameters", what parameters? What is the relation?

We have changed this sentence to " In addition, local multifractality $\tau''(1)$, can also be used as a measure to quantitatively characterize the multifractality of a dataset using the equation 8, where ordinary spatial analysis functions (autocorrelation and semivariogram) are related to the low order statistical moments (0 to 2nd) that may determine $\tau''(1)$ (Cheng, 2006)".

Line 178: "is the box-counting dimension", but you were using the gliding-box approach. I am lost.

The gliding box method is derived from the box-counting method, meaning that the D involved in the box-counting box method is as same that used in the gliding-box approach. The fact that we have removed the equation in question from the revised manuscript means that we have also changed this text from "the box-counting dimension" to "the gliding-box dimension".

Line 179: "smaller values". Not clear, smaller than what? Positive or negative?

The fact that $-D < \tau''(1) < 0$ means that the value of $\tau''(1)$ will be negative. As such, we have changed changed "smaller values" to "more negative values" to clarify this.

Line 181: "used" instead of "use".

We have used "used" instead of "use".

Line 182: “heterogeneous patterns”, of what?

Heterogeneous patterns are ordered, complex, clustered patterns; to clarify this we have added. We have added this to the manuscript.

Lines 183-184: “as well as enabling the comparison of the distribution of differing elements in the soils in this region”. If you say so, but I am not so sure, in fact, you performed this using inverse distance weighting interpolation.

We have changed this sentence to “as well as enabling the comparison of the multifractality of differing elements in the soils in this region”.

Geochemical analysis results

Line 187: You do not indicate that means were also higher for the Daxing area. Besides, they are also higher in this area for Hg.

We have added "mean" to this line.

Line 196: “yielded concentration histograms” instead of “yield histograms”.

We have used “yielded concentration histograms” instead of “yield histograms”.

Lines 198-200: And also that these concentrations depended on the type of human activities developed within each area.

We have changed this sentence to "This means that multifractal techniques are highly suited for the characterization of the geochemistry of the soils discrimination of the differing types of human activities ongoing in each area.".

Calculation processes of multifractal spectrum and discussion

Lines 210-211: This is already explained in the description of the multifractal analysis that has been carried out.

We have rewritten lines 210-212 to clarify this

Lines 211-212: “used a range of *q* values from –10 to 10”, did you select this range? Besides, you did not explain what “*q*” is.

We did select *q* values from –10 to 10 and have clarified this in the text. As described above, *q* is the order moment of the measure, as described in Line 134. We have also rewritten lines 210-212.

Line 216: I would remove “showing the multifratal characteristics of all”.

We have removed “showing the multifractal characteristics of all”.

Line 217: I would remove “(barring Cu)”and add “Daxing” to the figure caption.

We have removed “(barring Cu)”and added “Daxing” to the figure caption.

Lines 219-220: I would remove "combine the singularity exponent and the corresponding fractal dimension $f(\alpha)$ to generate a multifractal spectrum with" and use just "showed".

We have used "showed" to make this sentence clearer.

Line 221: "are also" should be substituted for "were" and "is" for "was".

We have used "were" instead of "are also" and "was" instead of "is".

Line 223: "samples" instead of "the soils".

We have used "samples" instead of "the soils"

Line 226: Remove "analytical" and use "indicated" instead of "indicate".

We have removed "analytical" and used "indicated" instead of "indicate".

Line 227: "were characterized" instead of "are characterized".

We have used "were characterized" instead of "are characterized".

Lines 227-230: Please, re-phrase. Use the past tense and remove innecessary words.

We have re-phrased this sentence.

Line 232: "had" instead of "have".

We have used "had" instead of "have".

Lines 232-233: Are these differences significant?

We think these differences are significant; the Daxing area has $\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$ values that are double the values for the Yicheng area for the majority of the elements analysed during this study.

Lines 235-236: "the significant heavy metal pollution associated with agriculture"; however, concentrations of Hg were greater in the industrial area.

We thank the reviewer for pointing this out. As such, we have added the following section to the manuscript: "Although the mean concentrations of Hg in soils are greater in 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 concentrations of Hg in the Yicheng area are lower than in the soils in the Daxing area, both are of significance. Indeed, the lower concentrations in the Yicheng area may be of more concern than the higher concentrations in the Daxing area, as the agricultural activity in this 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."

Lines 237-240: I do not agree with this explanation. The high values for the multifractal indices used in this study just mean that in your data series high concentration values were very different from low concentrations for a given element.

We thank the reviewer for their suggestion and have made our discussion more rigorous, as evidenced by the revision of this sentence as follows: " The $\Delta f(\alpha)$ and α 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".

Lines 241-243: I do not see why you only concentrate on Hg pollution in your discussion, what about the other elements?

The main aim of this paper is to explain that the three multifractal indices used during this study are useful tools for the evaluation of the degree of influence on the heavy metals in soils caused by human activities. We focus on Hg in the Yicheng area rather than the other elements as Hg has higher $\Delta f(\alpha)$ and $\Delta\alpha$ and lower $\tau''(1)$ values than all of other elements in the Yicheng area and some of the elements in the Daxing area, even though the mean Hg concentrations in the Yicheng area are lower than in the Daxing area. These characteristics mean we have focused on the Hg contamination of the soils in the Yicheng area.

Lines 243-244: “deleterious effects”, on what?

We mean the heavy metal pollution of people, crops and animals, and have stated this in the text.

Line 246: I would remove “within the soil samples”.

We have removed “within the soil samples” in the title of Table 2.

Lines 249-251: This should be explained in the materials and methods. Besides, you should indicate that data were sorted within each area and not on both at the same time.

The standard of the order is described in lines 151 and 160 and we have revised some of this text to make it clearer. We have also changed this sentence to "In order to compare variations in multifractality,

different elements within Daxing and Yicheng area were sorted by Δα, Δf(α) and τ″(1) parameters respectively, in addition to sorting by basic statistics such as standard deviation and coefficient of variation values (Table 3)".

Line 260: I would remove "the analyses afforded by classic".

We have removed "the analyses afforded by classic".

Lines 263-265: This is not clear, please, re-phrase it.

We have rephrased this sentence to "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".

Line 267: I would change "parameters and coefficient of variation values" to "and basic statistic indices".

We have changed "parameters and coefficient of variation values" to "and basic statistic indices" in the title of Table 3.

Table 3: Using standard deviation is of not very much use here since its effect would depend on the magnitude of your data. For instance Zn has a very high standard deviation compared to Hg, so logically, this index would give Zn always the first order. I suggest only using coefficient of variation, since this index is normalized for all elements.

We thank the reviewer for their suggestion and have revised Table 3, which is now only sorted by coefficient of variation values.

Line 273: According to table 3, it should be "Hg>As>Zn>Cd>Pb>Cu".

We have corrected this in the text to "Hg>As>Zn>Cd>Pb>Cu" .

Line 280: Why not performing this inverse distance weighting interpolation for the rest of the elements? Besides, be careful since here you are not using multifractals; however, your discussion is oriented as if this technique was multifractal.

We wanted to use inverse distance weighting (IDW) interpolation to show that the multifractal spectrum technique and associated parameters can used to evaluate the degree of multifractality caused by human activities. The IDW interpolation is not the key focus of this paper and we have only used IDW to compare the spatial distribution of the elements with highest and lowest degrees of multifractality in the Daxing and Yicheng areas.

Lines 284-285: Not exactly, only in the area where is a bunch of breeding facilities. In the case of Pb, this concentration would depend on the type of industry involved.

We have calculated a correlation matrix for the density map of breeding factories in the Yicheng area (Fig. 7), and a filled contour map for Hg (Fig. 5). The result indicates a strong spatial correlation between the Hg contamination and the location of breeding facilities in the Yicheng area.

Lines 287-290: What are they using as Hg source? It must appear from somewhere!!!

We think fertilizer, fodder, pesticides and water could all be Hg sources in the Yicheng area, although further work is needed to identify the main source of contamination; this is beyond the scope of the current paper although we have mentioned this in the text.

Lines 293-295: Please, re-phrase. This is not clear.

We have rewritten this as follows "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 (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 (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 activity in this area does not produce significant Cu contamination"

Line 296: "a significant" instead of "an significant".

We have used "a significant" instead of "an significant"

Lines 297-299: I do not see this from your figure. In fact, the evolution of both elements is very similar at lower classes.

We have accepted the reviewer's suggestion and have used a correlation matrix to show the correlation instead of a figure.

Lines 301-302: I do not totally agree. You can see that the shape of the curve is similar for Cu and Hg, only different for the greater classes.

Although the curves are similar, the new correlation matrix obtained during this study indicates a significant correlation between the location of agricultural facilities and high concentrations of Hg (correlation value = 0.434), whereas there is an independent or a negative correlation between agricultural breeding facilities and areas of high Cu concentrations (correlation value = -0.064).

Line 304: "can efficiently reflect the multifractality", of course, they are designed to do this.

We have removed this sentence to make this paragraph more brief.

Line 305: Remove “by”.

We have removed "by".

Lines 317-319: This is unclear. Please, re-phrase it.

We have re-phrased this as follows: "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).".

Line 320: Remove “especially in areas with significant heavy metal pollution”. From my viewpoint, it is not needed.

We have removed “especially in areas with significant heavy metal pollution”.

Line 324: Remove “in this area”.

We have removed “in this area”.

Figure 4: In the upper left-hand side of the map there are some industries and the Pb concentration is rather low. Similar values are observed in the lower left-hand side of the map. This may indicate that Pb concentrations in soils depend more on the type of industry than on the fact that there is an industry, as you imply in your discussion.

Figure 4 shows that only 4 factories are located in the area with low Pb concentrations in the upper and lower left hand sides of the maps, with all of the other factories located within areas with relatively high Pb concentrations (>34.61 mg/kg). As such, we agree with the reviewer although we still think that our data support our conclusion that areas with elevated Pb concentrations within the Daxing area are correlated to the location of industrial factories. However, we have also added a comment that reflects this variability as suggested by the reviewer.

Figure 5: In contrast with the former figure, in the right-hand side of this map, we can observe high concentrations of Hg in the soil, but there are no breeding facilities on this part of the map… Then, why do these high Hg concentrations appear? According to your discussion, there is a direct relationship between the existence of a breeding facility and the high concentrations of Hg observed.

Figure 6: Coinciding with the former map, there are very high Cu concentrations in the right-hand side of this map, why? What is over there?

Figure 5 indicates that some areas with the highest concentrations of Hg are spatially correlated with the breeding facilities, whereas other areas with slightly elevated concentrations of Hg are spatially correlated with the river in the right-hand side of the figure. In comparison, Figure 6 shows that a significant number of areas with elevated concentrations of Cu are located beside the river. We have discussed these variations within the text.

Figure 7: To me, this graph is difficult to interprete. Both lines show correlations between number of facilities and Hg or Cu concentrations in soils. Besides, the caption is not clear. You talk about an anti-correlation and this is not observed in the graph.

We have used a correlation matrix (Table 4) to show the relationship between the spatial density of breeding facilities in the Yicheng area and filled contours maps for the heavy metal concentrations in this area to show the correlation between these facilities and Hg concentrations and the lack of a correlation between these facilities and Cu concentrations in this region.

Conclusions:

Line 355: According to table 3, the overal multifractality in Yicheng decreased as Hg>As>Zn>Cd>Pb>Cu and not as Hg>Zn>As>Cd>Pb>Cu as is reported here.

We have revised this.

Lines 356-364: What about other problems caused by the other heavy metals? People in Daxing are inmune to heavy metal pollution?

The multi-element heavy metal pollution within the Daxing area is strongly influenced by human activities. However, the heavily industrial Daxing region contrasts with the highly agricultural nature of the Yicheng region, an area that supplies significant amounts of food to the city of Hefei. The contamination (especially Hg) in this area can more easily make its way into the human food chain (as well as being concentrated up the food chain), thereby having a more rapid direct impact on human health. As such, we have highlighted this in our paper to show that although both areas require remediation, the Hg contamination in the Yicheng area is especially worrisome.

Line 365: "The initial results", I do not understand why you termed your results as "initial".

We think that further research in this area is needed to identify the sources of pollution as mentioned by the reviewer earlier in their comments. As such, this paper is based in initial research that highlights areas for future research as well as demonstrating the validity of this multispectral approach. However, we acknowledge that the term "initial" may be confusing, and we have removed it.

Lines 366-367: "multifractal parameters can efficiently reflect the multifractality caused by industrial and agricultural activities", well, of course, multifractal indices are designed to do this.

We have revised this sentence to "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".

Lines 369-370: "and the identification of major sources of heavy metal contamination". I do not agree, the identification of these sources was made through inverse distance weigthing interpolation, which is not a multifractal technique.

We have revised this sentence to make it clearer as follows "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.".

RC2: Miranda, Jose vivasm@gmail.com

Lines 234-240

The authors suggest a causal association between f(alfa) spectrum properties and contaminations. But the only real outcome of f(alfa) properties are related to heterogeneity and, as far as I know, there is no direct association between heterogeneity and contamination. The suggestion proposed by authors has no meaning. Unless the authors prove a correlation between heterogeneity and contamination.

We thank the reviewer for their comment. The fact that the association between heterogeneity and contamination is hard to express means that we have removed the word "heterogeneity" from the paper.

Lines 293-298

The authors answer:

Curves in figure 7 seems to be wrong, once in the map there are only 11 facilities and the graph shows 12. I can't understand how the graphs in Figure 7 can prove the existence of correlations between the location of industries and contamination. In my opinion, the fact that the number of facilities increase with rank is an obvious result, since a lower level contain all facilities of the highest levels. Thus, Figure 7 does not support the hypothesis of correlation neither causality between factors. I still think that statistical correlations could help to understand this relation.

One of the breeding facilities is close to the margin of the figure and was clipped; however, we have revised this figure and more clearly demonstrated the correlation between the location of these facilities and heavy metal contamination using a statistical correlation (a correlation matrix) rather than using a figure.

Technical Comments

Format problems in many equations (ex.: 2, 5, 7 and 8)

We have revised equation 2.

We thank the anonymous referee and J. Miranda for their positive comments and have improved the written English and revised 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~~causes~~ 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 and use ~~these data to evaluate and compare the characteristics of heavy metal pollution in soils within 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 municipal area. This study uses~~ a multifractal spectral technique to identify and compare the multifractality of heavy metal concentrations~~in the geochemistry~~ of soils within the industrial Daxing and agricultural Yicheng areas. This paper uses~~ of Anhui Province. Determining~~ three multifractal parameters ($\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$) for these soil geochemical data to indicate the~~indicates that~~ 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 ~~for the~~ Daxing area compared to~~decreases as follows: Pb>Cd>As>Zn>Hg>Cu, whereas the overall amount of multifractality within the soil geochemical data for~~ the Yicheng area. The ~~decreases as follows: Hg>Zn>As>Cd>Pb>Cu. These~~ differences in the degree of multifractality between Daxing and Yicheng areas indicate~~indicates~~ that the soils in these areas have differing multifractal geochemical characteristics, suggesting that the differing economic activities in these areas generate very different heavy metal pollutant loads. In addition, the~~ (e.g. Hg dominated agricultural pollution vs. Pb dominated industrial pollution). In addition, 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. These larger values 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 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.~~ and the identification of major sources of heavy metal contamination.~~

Keywords: soil geochemistry; multifractal modelling;~~modelling,~~ heavy metal pollution;~~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., ~~in recent years (McGrath et al., 2004; Wang et al., 2007;~~ 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 ~~has a lot of advantages, including the fact that these approaches~~ can investigate many of the problems of nonlinear variability which commonly arise when dealing with pollutants and as well as enabling the identification of~~identify~~ non-linear characteristics within datasets. This approach can yield~~, yielding~~ 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~~using multifractal techniques to determine~~ the multifractal characteristics of the distribution of heavy metals in soils can improve~~further~~ 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~~identification~~ of porous structures and ~~the~~ spatial variations~~variability~~ in soil properties (~~and so on (Dathe et al., 2006;~~ Caniego et al., 2005; Dathe et al., 2006).~~2005).~~ This means that multifractal techniques can be ~~provide a lot of~~ useful tools for the ~~the~~ analysis of heavy metal pollutions ~~pollutantion~~

within soils (e.g., Salvadori et al., 1997; (Lima et al., 2003; Albanese et al., 2007; Guillén et al., 2011).2011; Salvadori et al., 1997). These multifractal techniques are not only used in environmental science, but also be 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 (Cheng, 1995; (Deng et al., 2011; Zuo et al., 2012, 2014; Cheng, 1995; Nazarpour et al., 2014; Yuan et al., 2012, 2015; Nazarpour et al., 2014)2015) 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 threetechniques to determine the multifractal characteristics of the distribution of heavy metals in soils in these areas, using three multifractal 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 the anthropogenic activities in this region. The results will further enable and inform future planning for any necessary remediation of these 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 ~~and~~ Hg and As concentrations were determined by hydride generation–atomic fluorescence spectrometry (AFS; Armstrong et al., 1999; Gómez-Ariza et al., 2000).~~(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 data was monitored by repeat ~~determinations of standards~~ and replicate determinations ~~of sub-sets of samples~~ using instrumental neutron activation analysis (INAA), with analytical~~(INAA). Analytical~~ precision was monitored using ~~determinations of~~ variance of the results obtained from duplicate analyses.

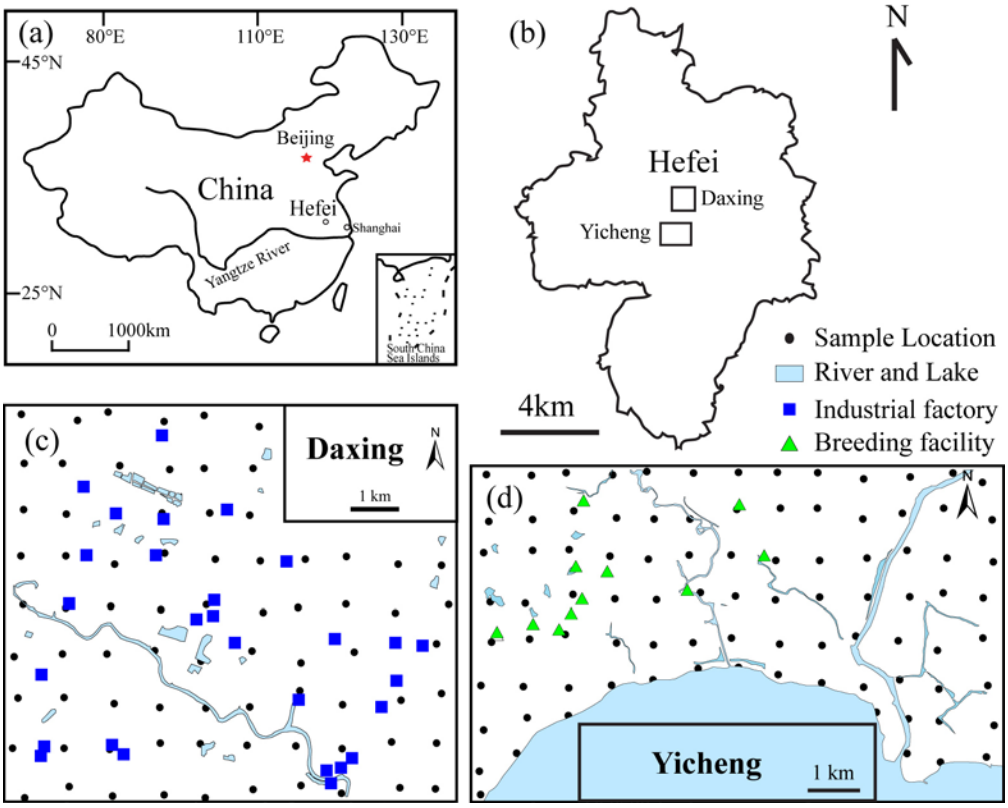

Fig. 1. Location of Hefei in central-eastern China (a); location of the study areas within Hefei (b); the 1 × ~~x~~ 1 km grids used for soil sampling in the towns of Daxing (c) and Yicheng (d)

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

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

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

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

$$\langle \tau(q) \rangle + D = \lim_{\varepsilon \to 0}\left(\frac{\log(\mu^{q}(\varepsilon))}{\log(\varepsilon)}\right) = \lim_{\varepsilon \to 0}\left(\frac{\log\left(1/N^{*}(\varepsilon)\right)\sum_{i=1}^{N^{*}(\varepsilon)} \mu_i^{q}(\varepsilon)}{\log(\varepsilon)}\right) \quad (1)(2)$$

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, <> 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 a and $f(\alpha)$ values using a Legendre transformation, as expressed below:

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

(2)(3)

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

(3)(4)

where $\Delta\alpha$ 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:

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

(4)(5)

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

$$\Delta\alpha_R = \alpha_{\max} - \alpha_0 \quad (6)$$

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

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

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

(7)(8)

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 associated with ordinary spatial analysis parameters, as represented by the $\tau''(1)$, parameter, 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 $\tau''(1)$ (Cheng, 2006): using the following equation:

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

(8)(9)

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

~~——~~Here, we use the three multifractal parameters described above ($\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$) to better identify ~~heterogeneous patterns and~~ the degrees of multifractality within the soil geochemical data for the study area as well as enabling the comparison of the multifractality~~distribution~~ 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~~are~~ 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.~~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 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.~~contaminated soils in these areas.~~

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.

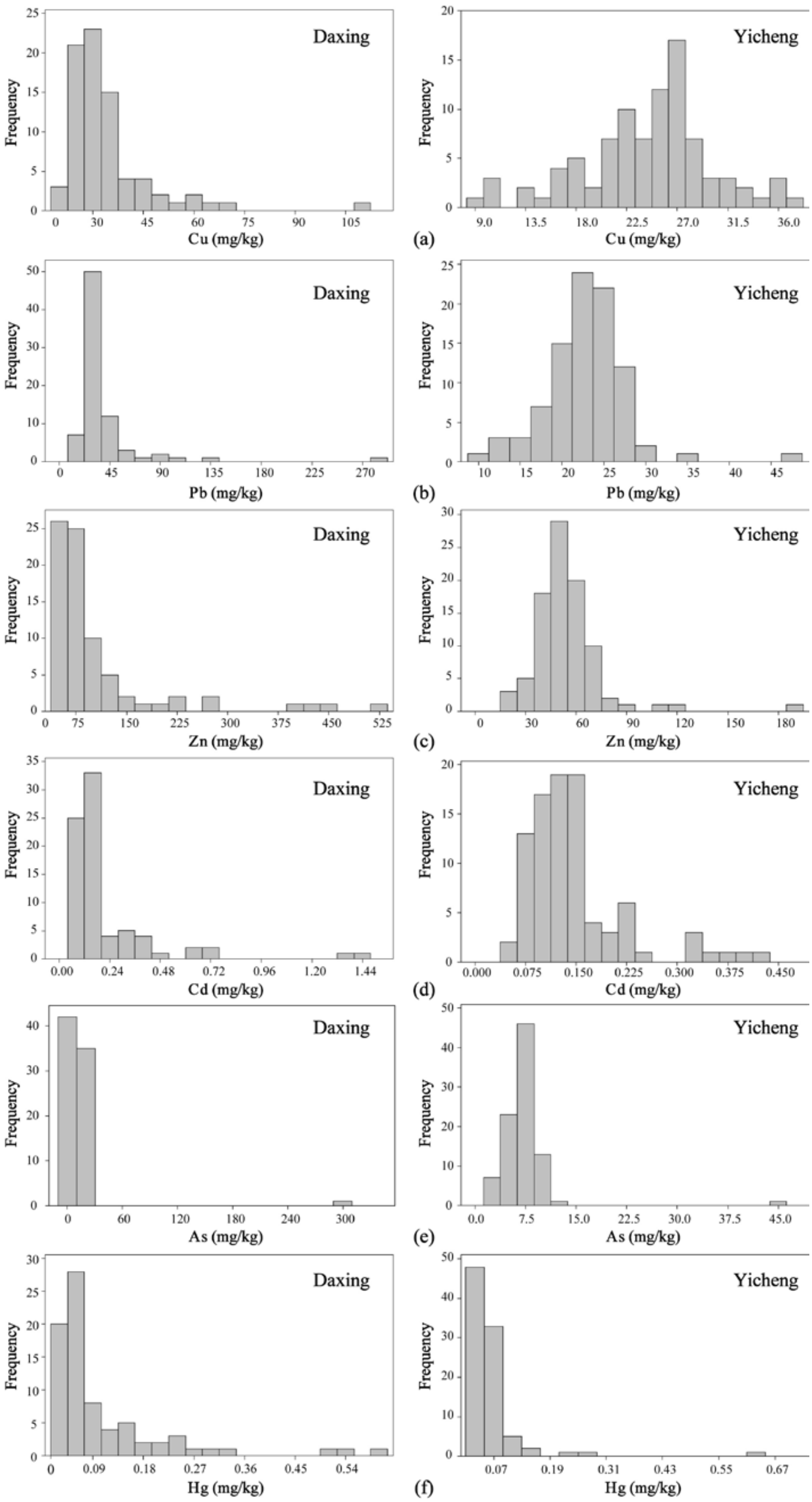

Fig. 2. Histograms showing the distribution of Cu (a), Pb (b), Zn (c), Cd (d), As (e) and Hg (f) concentrations within soils from the towns of Daxing and Yicheng.

5. Calculation processes of multifractal spectrum and discussion

The ~~Here, we use the gliding box method to calculate~~ multifractal spectra (in the form of an α–$f(\alpha)$ diagram) for the geochemical data are shown in Fig. 3 using~~from the study area. This calculation used~~ a range of q values from −10 to 10 with an interval of 1. ~~1, yielding the multifractal spectra (in the form of an α–$f(\alpha)$ diagram) shown in Fig. 3.~~

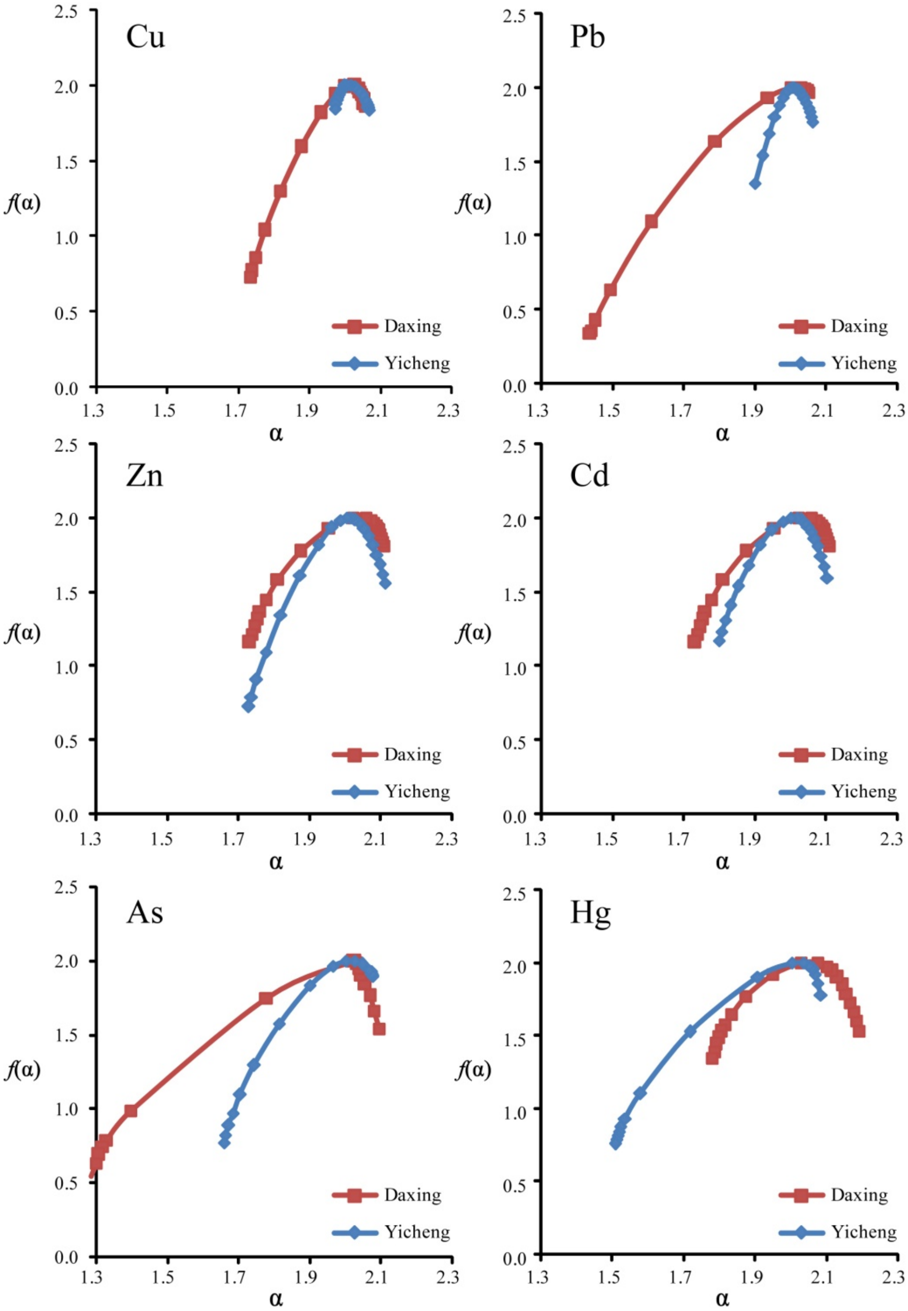

Fig. 3. Multifractal spectra ($f(\alpha)$ vs α) ~~showing the multifractal characteristics of all~~ of the soil

geochemical data (barring Cu) from the Daxing and Yichen area.

These multifractal spectra have— Multifractal spectra combine the singularity exponent α and the corresponding fractal dimension $f(\alpha)$ to generate a multifractal spectrum with an inverse bell shapes (Fig. 3) and are. All of these multifractal spectra are also asymmetric (i.e. $\Delta\alpha_L$ valuesis significantly different from $\Delta\alpha_R$, equations 5-6) with the exception of(barring the Cu data for soils from the Yicheng area,area), indicating that the samplessoils 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 analytical results givenshown 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(i.e. have high $\Delta\alpha$ values), have $\tau''(1)$ values less than –0.01 (barring Cu and Pb in the Yicheng area) and have $\Delta f(\alpha)$ values larger than 0.5, (barring Cu in the Yicheng area), all of which indicate that these elements have 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 α 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 iswould be Hg contamination. The $\Delta f(\alpha)$ and α values of Hg in the Yicheng area are larger than the values for 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, even considering the lower overall (but not maximum) concentrations of Hg within the Yicheng area compared to the Daxing area.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 is important, primarily as Hg pollution couldcan seriously impact human health, as Hg because this element is preferentially concentrated upward in the food chain (e.g. (Jiang et al., 2006)). This means~~2006)), meaning~~ that although~~this~~ 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~~ within the soil samples~~ 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, different~~ Different~~ elements within Daxing and Yicheng area were sorted by $\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$ parameters, respectively,~~ in order to compare variations in multifractality,~~ 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 Zn data within the Daxing area has largest standard deviation value but only a moderate coefficient of variation, but the $\Delta\alpha$ and $\Delta f(\alpha)$ values for these Zn data are indicative of only weak multifractality compared to the other heavy metals in the soils within~~of~~ the Daxing area. In comparison, the Hg data for soils in the Yicheng area yielded the lowest standard deviation value but the largest –$\Delta\alpha$ and $\tau''(1)$ values, indicating these Hg data have 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 ~~analyses afforded by classic~~ 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,~~first averaged~~ 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~~and $\tau''(1)$ before sorting~~ again to compare the overall multifractality of these data.

Table 3. Elements sorted by multifractal parameters and basic statistic indices.~~coefficient of variation values.~~

Town	**Element**	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.~~Hg>Zn>As>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~~maps indicate~~ that areas with elevated levels of Pb contamination within the Daxing area are ~~directly~~ correlated with~~to~~ 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 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,~~whereas~~ 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 ~~in the Yicheng area is of significance, especially as this form of~~ 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~~can cause~~ serious health issues (e.g. Minamata disease) caused by the potential concentration of Hg~~. As such, the soils in this area may well require remediation, especially as Hg can be concentrated~~ up the food chain. This indicates that soils in both areas may well require control and remediation.~~ 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 weakest multifractality for Cu within the Yicheng area (Fig. 3, 5-6). Here, we generated~~We used~~ a correlation matrix that compares the relationship between the spatial density of breeding locations in the Yicheng area (Fig. 7) and filled contours maps~~plot~~ showing the distribution of Hg (Fig. 5) and Cu (Fig. 6) in this region~~rank of concentration contour vs number of agricultural facilities within the same rank of concentration contour~~ to identify whether there are any~~demonstrate the~~ spatial correlations between the location of agricultural facilities and areas containing soils with elevated heavy metal concentrations (Table 4). The correlation matrix~~in soils (Fig. 7). This diagram~~ shows a~~n~~ significant correlation between agricultural facilities and high concentrations of Hg (correlation value = 0.434),~~,~~ whereas the location of these~~re is an anti-correlation when comparing~~ agricultural breeding facilities and areas of high Cu concentrations either have no relationship or are negatively correlated (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 does not produce significant Cu contamination. The correlation matrix, symmetrical distribution and weakest multifractality 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.be contain only natural background concentrations of Cu and that the agricultural activity in this area does not produce significant Cu contamination.

These results— All of the above suggests that the multifractal parameters for the heavy metal concentrations within soil geochemical data can efficiently reflect the multifractality associated with by industrial and agricultural activities in the Daxing and Yicheng areas, respectively. These results also 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 procedures and 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 numberamount 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 beyond2014), with the scope of results presented in this study, the multi-element nature of the contamination in the Daxing area means suggesting that physical and chemical approaches to remediation (i.e., (soil removal, soil vitrification, soil consolidation, electroremediation, or soil washing) are probably well suitedmore appropriate for the remediation of heavy metal contaminated soil in this region (the Daxing area, especially Pb).in areas with significant heavy metal pollution. 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, 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.~~in this area (Mulligan et al., 2001;Wang et al., 2006).~~

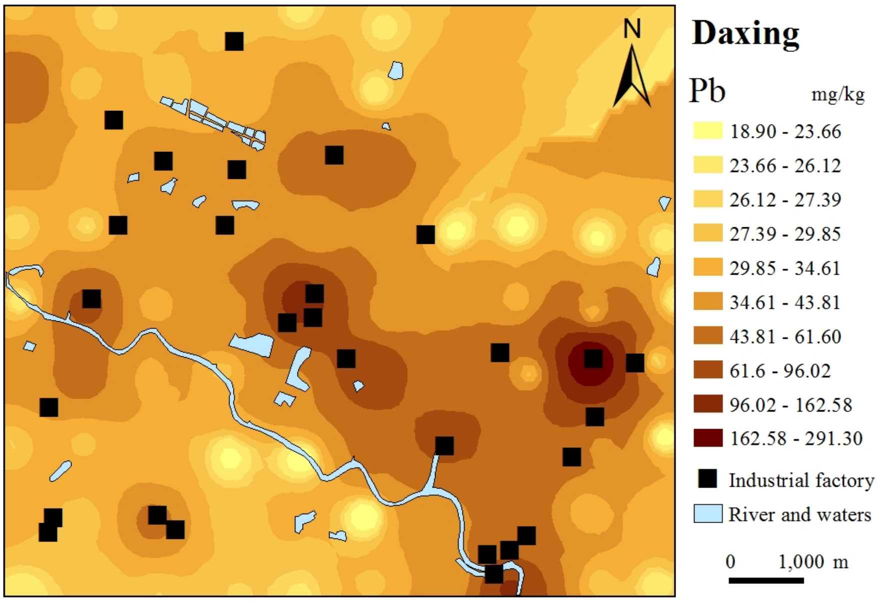

Fig. 4. Filled contour map generated by inverse distance weighted interpolation showing the spatial distribution of soil Pb concentrations in the Daxing area.

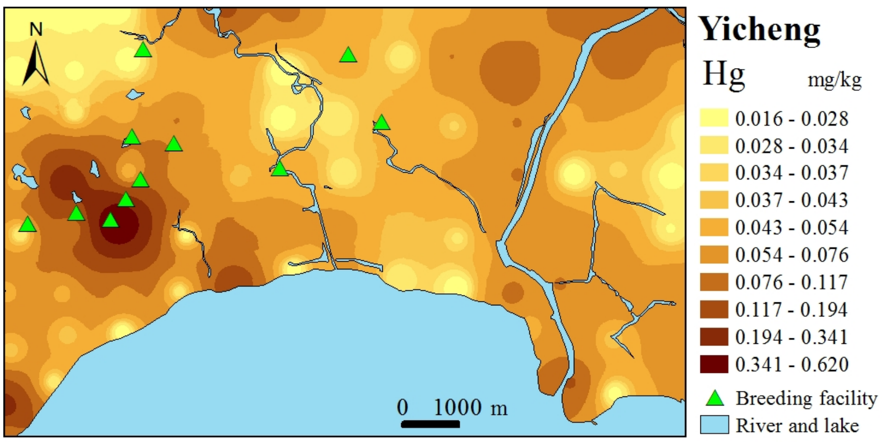

Fig. 5. Filled contour map generated by inverse distance weighted interpolation showing the spatial distribution of soil Hg concentrations in the Yicheng area.

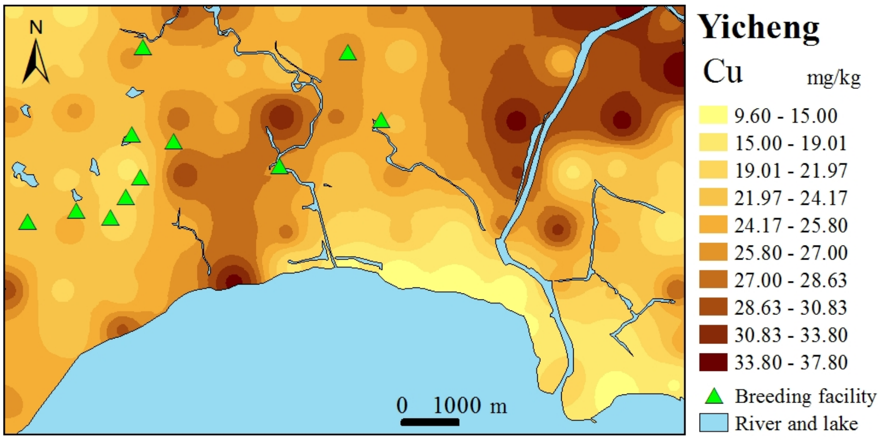

Fig. 6. Filled contour map generated by inverse distance weighted interpolation showing the spatial distribution of soil Cu concentrations and the location of breeding facilities in the Yicheng area

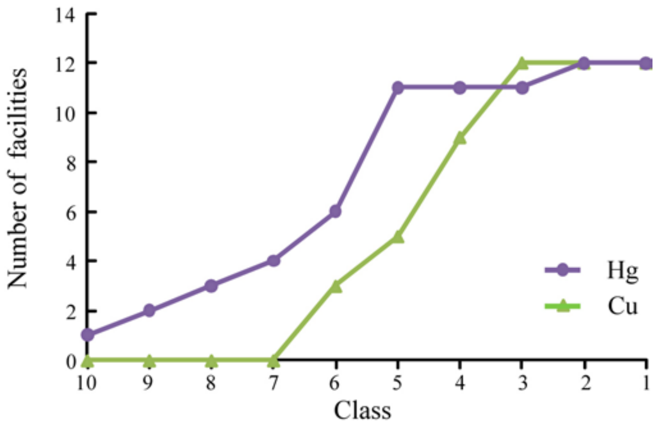

Fig. 7. Density map of breeding facilities in Yicheng area (generated using the kernel density tool within the ArcGIS software package).~~Plot of number of agricultural facilities in Yicheng area within the same rank of Hg and Cu concentration contour showing a positive spatial correlation between location of agricultural facilities and Hg concentrations but an anti-correlation between the location of agricultural facilities and Cu concentrations.~~

Table 4. Correlation matrix comparing the breeding facility density map and the filled contour maps for Hg and Cu data for the Yicheng area.

Layers	Layer 1	Layer 2	Layer 3
Layer 1	1.00000	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. 8);
Layer 2: Filled contour map of Hg concentrations of Yicheng area (Fig. 8);
Layer 3: Filled contour map of Cu concentrations of Yicheng area (Fig. 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 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.~~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 three~~generation of~~ 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~~major sources of~~ heavy metals that need remediation in the research area. contamination.

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