Dear Prof. Schertzer and Dr. Miras-Avalos,

We thank anonymous referee #1, J. Miranda and Miras Avalos 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

This small note describes a good experimental dataset collected in two contracted landuse areas. The manuscript has to be amended with hypothetical explanation of processes causing the observed multifractal properties. Otherwise it is purely descriptive narrative that does add to science. The recommendations of soil reclamation have to be substantiated with suggestions of reclamation techniques and purposes of reclamation, otherwise these recommendations are superfluous. Note that references are mostly old, all older than 5 years.

We thank the reviewer for their suggestions. Our analysis indicates that different clean-up and remediation approaches are needed to resolve the issues relating to the differing heavy metal pollution in these areas, rather than a single approach to resolving heavy metal pollution. We have amended the text to reflect this as follows:

"*A significant amount of different remediation approaches can be used to resolve the issues of heavy metal soil contamination (e.g., Bech et al., 2014; Koptsik, 2014). The results presented in this study suggest that physical and chemical approaches (soil removal, soil vitrification, soil consolidation, electroremediation, soil washing) are more appropriate for the remediation of heavy metal contaminated soil in the Daxing area, especially in areas with significant heavy metal pollution, whereas the differing 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 in this area (Mulligan et al., 2001;Wang et al., 2006)*".

RC2: J. Miranda

GENERAL COMMENTS

Interesting article that uses the multifractal spectrum to assess possible soil contamination by industrial and agricultural activity in two regions of China. An excellent data collection work and proper use of the chosen methods. The proposed use of the distribution of alpha singularity exponents to evaluate diffusion of contaminants in the soil is valid, but the conclusions require more robust criteria of causality. The main conclusions outlined by the authors are based on correlations and comparisons that were not carefully evaluated. The authors use visual and inaccurate comparisons to validate important statements in the paper argument. The following is a detailed description of suggestions to improve the work.

SPECIFIC COMMENTS

Line 138: I would say it suggests a non-normal distribution a priori, only. The possible fractal/multifractal pattern is something to be evaluated a posteriori.

Thanks for the comments provided by J. Miranda.

We have revised this sentence as follows:

"All of the elements (barring Pb and Cu in the Yicheng area) in both the Yicheng and Daxing areas yield 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."

210: Why did you choose these values of *q*? Is there any argument (e.g., when the D*q*x*q* curve stabilises)?

A range of q values between −10 and 10 with an interval of 1 is commonly used in these types of studies (Gonçalves et al., 2001; Xie et al., 2004; Dathe et al.,2006). To ensure that the results are reproducible we also replicated this analysis using a range of q values from −1 to 1 with an interval of 0.1, which yielded the same conclusions to the original range of q values.

Line 227: A comparison between the $\Delta f(\alpha)$ of the locations is considered here. The authors claim significant differences comparing only the order of the metals, sorted by $\Delta f(\alpha)$. Here a paired comparison statistic could prove the significant difference between areas.

We have added some text at lines Line 246-259 to compare the differences in heavy metal pollutions in the Daxing and Yicheng areas.

232,234: In my point of view, Figure 3 shows no sufficient evidence to conclude about correlations between the spectra of the two regions. A correlation test between Δα (left and right) in Daxing and Δα (left and right) in Yicheng could give more support to the argument.

We want to use Fig. 3 to show the differences shapes of the spectra between the two different parts of the study area as well as demonstrating the different multifractal characteristics of the heavy metal pollution in these areas. However, we have revised the manuscript using three parameters ($\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$) to compare the heterogeneous patterns and degree of multifractality of the different heavy metals and areas, and we have rephrased the text to ensure this approach is described clearly.

255-257: A logistic correlation could substantiate the statement of significant correlation between the location of industrial/agricultural facilities and metals concentrations.

258-260: A very interesting hypothesis, associating the asymmetry of the spectra with the presence of anthropic actions. Whereas the single symmetric spectrum found was the Cu in the Yicheng area, we would expect a not significant logistics correlation between the presence of agricultural facilities and concentration of Cu in Yicheng (map in Figure 6) and significant correlation in the asymmetry cases. These tests would substantiate the argument of using multifractal for evaluation of anthropogenic changes.

We thank the reviewer for their suggestions, and we have added a new figure (Fig. 7) to show the relationship between heavy metal concentrations and the number of facilities in each area. This figure demonstrates a very good positive spatial correlation between the agricultural facilities in the Yicheng area and the high Hg concentration areas in this region and an even better positive spatial correlation between agricultural facilities in the Yicheng area and the high Cu concentration areas in this region. However, although this figure cannot show the degree of heavy metal pollution, it does demonstrate the spatial correlation between the location of industrial/agricultural facilities and areas of high metal concentrations, indicating a significant logistical correlation between the multifractality of the datasets and the industrial and agricultural activities in this area.

TECHNICAL COMMENTS

143: Just a suggestion: Make the legend a bit clear. The legend information is spread in the figure.

We have edited the legend in Fig. 1 as suggested.

197: I would say more heterogeneous patterns, given the non-binary feature of heterogeneity.

We changed the words "heterogeneous distribution patterns" to "more heterogeneous patterns".

214: "that describing the multifractality" - Unnecessary text.

We have removed "that describing the multifractality" from the table caption.

221: The *f* spectrum is only another way to characterise your set. I am not sure if 'best measure' is the most suitable term.

We have used three indexing methods ($\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$) in the revised paper to allow a better analysis of the multifractal characteristics of the heavy metal pollution in soil in urban or developed areas.

234: Asymmetry concept could be better explained, it is presented in a way which might lead to misunderstandings. I would suggest an explanation based on the equations of the lines 191, 192 and 193.

We have added an additional comment in brackets as per the equation between lines 162-164 as follows: ($\Delta\alpha_L$ is significantly different from $\Delta\alpha_R$, equations 5-6).

238: Just two missed commas – "All of the heavy metals analyzed during this study, barring Hg, have higher $\Delta f(\alpha)$ values in soils from the Daxing area, with Hg having higher values in soils from the Yicheng area (Table 2)."

We have added the two missing commas.

241: "The only significant heavy metal pollution associated with the agricultural activity in the Yicheng area WOULD BE the Hg contamination

We have revised this sentence according to the suggestions provided by both Miranda and Miras Avalos as follows:

"***This suggests that the industrial activities in the Daxing area generate multi-element heavy metal soil contamination, whereas the only significant heavy metal pollution associated with the agricultural activity in the Yicheng area is Hg contamination.***"

EC1: J. M. Miras Avalos

Specific comments to the authors:

Please, organize the manuscript in Introduction, Materials and Methods, Results and Discussion, and Conclusions.

Title:

I suggest the authors to slightly modify the title of their manuscript to: "Comparison of the multifractal characteristics of heavy metals in soils within two areas of contrasting economic activities in China".

We thank the reviewer for their suggestion and have modified the title of our manuscript as follows: "*Comparison of the multifractal characteristics of heavy metals in soils within two areas of contrasting economic activities in China*".

Abstract:

The abstract is too long from my point of view.

We have shortened the abstract to highlight the main findings of our research.

Lines 16-17: "causing" instead of "that can have".

We have used "causing" instead of "that can have".

Line 23; "allows deeper interrogation", this expression is not clear. Please, re-phrase it.

We have revised this sentence to "Here, we present the results of a heavy metal (Cu, Pb, Zn, Cd, As and Hg) soil geochemical survey and use these data to evaluate and compare the characteristics of heavy metal pollution in soil in urban or developed areas."

Lines 26-28: "This study focuses…", this sentence can be removed since its information is reported in the next one.

We have removed this sentence.

Line 29: Include "(industrial)" after "Daxing" and "(agricultural)" after "Yicheng".

We have changed this sentence to "*This study uses a multifractal spectral technique to identify the multifractality in the geochemistry of soils within the industrial Daxing and agricultural Yicheng areas of Anhui Province* ".

Lines 31, 32 and 38: Use α instead of a in $\Delta f(\alpha)$, please.

We have used α instead of a in $\Delta f(\alpha)$.

Line 33: There is a mistake here; according to table 2, the $\Delta f(\alpha)$ in Yicheng decreased as Zn>Hg>As>Cd>Pb>Cu instead of Hg>Zn>As>Cd>Pb>Cu as is reported here.

We thank the reviewer for pointing out this mistake and for providing us with a new idea. Previously we thought that using f(α) to study contamination was sufficient, but we now realise that this is not sufficient and that $\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$ values also reflect different aspects of multifractality. As such, we have used $\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$ together in the revised manuscript to study and evaluate the multifractality of heavy metal contamination in the study area.

Line 34: I would remove the word "geochemical".

We have removed the word "geochemical".

Line 36: "clearly different" instead of "distinctly different".

We have changed "distinctly different" to "clearly different".

Lines 44-45: I would remove "rather than a single approach to heavy metal pollution" since it is not needed.

We have removed these words.

Introduction:

This section is not clear, the state-of-the-art is not put into context and thus the introduction seems out of focus. Moreover, this section begins with a list of references because in the first 6 lines, authors cited 13 references.

We have reorganized the introduction as recommended by the reviewer.

Line 51: I would remove "recently".

We have removed "recently".

Lines 58-59: "the factors controlling the distribution" instead of "the controls on the distribution".

We have changed "the controls on the distribution" to "the factors controlling the distribution".

Line 62: "in soil properties" instead of "in the characteristics of soils".

We have changed "in the characteristics of soils" to "in soil properties".

Line 63: Remove "and".

We have removed "and".

Lines 63-65: Please, check English, this sentence is unclear.

We have reorganized this sentence.

Lines 67-68: "but also in the analysis of" instead of "but can also be used in the analysis of".

We have changed "but can also be used in the analysis of" to "but also in the analysis of".

Line 70: “and thus” instead of “meaning that”.

We have changed “meaning that” to “and thus”.

Lines 73-75: Please, re-phrase this sentence. It is not clear what you mean and must be put in context with the former sentence.

We have reorganized this sentence.

Line 77: Please, define “C-A” when first used.

We have used "Concentration-Area" instead of "C-A".

Line 79: Please, define “S-A” when first used.

We have used the " Spectral density-Area" instead of "S-A".

Lines 78-83: This is not clear, please, revise it.

We have reorganized this sentence.

Line 86: Remove “provincial”.

We have removed this word.

Line 87: Remove “areas”.

We have removed this word.

Line 88: “activities” instead of “activity”.

We have changed “activity” to “activities”.

Lines 88-96: This portion of the text is repetitive and unclear. Please, revise it and state clearly the aims of your study.

We have reorganized these sentences to make the text more clear and more concise as follows:

"Here, we use multifractal techniques 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 analyse 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."

Study area and geochemical data:

Line 99: Include “it” before “has”. Line 102: “industrial areas of Hefei” instead of “industrial bases of the Hefei area”.

We have added “it” before “has” and used “industrial areas of Hefei” instead of “industrial bases of the Hefei area”.

Line 103: Remove the word “industrial”.

We have removed this word.

Line 105-106: “In contrast, the town of Yicheng focuses its economic activities on agricultural production” instead of “In contrast, the town of Yicheng is agricultural, with the economy of the town focused on agricultural production”.

We have used “In contrast, the town of Yicheng focuses its economic activities on agricultural production” instead of “In contrast, the town of Yicheng is agricultural, with the economy of the town focused on agricultural production”.

Line 107: “ornamentals” instead of “flower planting”.

We have used “ornamentals” instead of “flower planting”.

Line 110: I do not understand what you mean by “natural mineralization”.

Here, we want to show that the soil in these areas are not influenced by mineralization or deposits.

Lines 110-111: “(< 20 cm depth)” instead of “(<20 cm below surface)”.

We have used “(<20 cm depth)” instead of “(<20 cm below surface)”.

Line 114: “was air-dried” instead of “was dried in air”.

We have used “was air-dried” instead of “was dried in air”.

Line 117: Remove “in the soil samples described above”, remove also “during this study”.

We have removed these words.

Line 119: “whereas Hg and As concentrations were determined” instead of “”with Hg and As concentrations determined”.

We have used “whereas Hg and As concentrations were determined” instead of “with Hg and As concentrations determined”.

Lines 122-125: You repeat too many times the word “analysis”, sometimes you can use the synonym “determinations”.

We thank the reviewer for this suggestion and have revised this sentence to "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). Analytical precision was monitored using determinations of variance of the results obtained from duplicate analyses."

Lines 116-125: Have you got references for the analytical methods? If so, please, add them to this portion of the text.

We think these analytical methods are well known as they have been used for a significant period of time and we do not want to make the reference list longer; as such, we have not made any specific reference to these techniques in the manuscript.

Line 126: "2.3. Results", this should be a section after the explanations of the materials and methods used.

We have moved this paragraph to Section 4.

Line 127: "A statistical summary" instead of "The results of a statistical analysis".

We have used "A statistical summary" instead of "The results of a statistical analysis".

Line 134: "the natural background". Maybe you should indicate what was the natural background.

Our original phrasing was not accurate; as such, we have changed this sentence to "*This also suggests that samples from the Daxing area containing elevated concentrations of heavy metals were probably contaminated by anthropogenic activity.*"

Line 136: I would include Pb with Cu for the Yicheng area since the distribution of its concentrations in soils seems to follow a normal distribution as well.

We thank the reviewer for their suggestion and have revised the manuscript appropriately.

Line 137: I would include "(Fig. 2)" after "outliers".

We have moved "(Fig. 2)" after "outliers".

Line 138: I am not sure, I agree that they are non-normal but how can you tell from the histograms that they are fractal?

We are only speculating that these data have fractal distributions in this section; as such, we have changed the text to reflect this as follows: "*indicating that these data have non-normal and potentially fractal- or multifractal-type distributions.*"

Line140: Remove "(Fig. 2)" from here.

We have removed "(Fig. 2)" from here.

Lines 143-146: I would rephrase this figure caption to "Location of Hefei in central-eastern China (a); location of the study areas within Hefei (b); 1 x 1 km grid for soil sampling in the towns of Daxin (c) and Yicheng (d)".

We have changed this figure caption.

Line 148: Re-phrase the title of this table to "Summary statistics of soil heavy metal concentrations from the Daxing and Yicheng samples".

We have changed this figure caption.

Table 1: Skewness and kurtosis are not concentrations and they are dimensionless. I would put the units below each column title, I mean below "minimum", "maximum", "mean" and "standard deviation". I would remove "Concentrations" from the table.

We thank the reviewer for this suggestion and have revised this table accordingly.

Mutifractal spectrum analysis:

Equations should be numbered.

We have numbered all of the equations in the text.

Line 159: "the factors controlling the distribution" instead of "the controls on the distribution". What do you mean by "of key elements within data".

We have changed the "the controls on the distribution" to "the factors controlling the distribution".

The key elements we want to express are the important study objects within the data, such as the heavy metals, nutrition component, porosity of soil, and so on.

Line 160: Remove the word "multifractal". "$f(\alpha)$" instead of "$f(a)$".

We have remove the word "multifractal" and now use "$f(\alpha)$" instead of "$f(a)$".

Line 163: Remove "of estimating f(a) values" since it is not needed.

We have remove the words "of estimating $f(a)$ values".

Line 170: "different from 0" instead of "that $\neq$ 0".

We have changed to this to "different from 0" instead of "that $\neq$ 0" as suggested by the reviewer.

Lines 173-174: Move "within a dataset" to after "statistical estimation".

We have moved "within a dataset" to after "statistical estimation"

Line 183: "different from 0" instead of "that $\neq$ 0".

We have changed this to "different from 0" from "that $\neq$ 0"

Lines 184-185: Use alpha (α) instead of a when referring to the multifractal spectra.

We have changed this throughout the manuscript.

Line 194: "spectrum is" instead of "spectrum are".

We have used "spectrum is" instead of "spectrum are".

Line 197: Use alpha (α) instead of a when referring to the multifractal spectra.

We have changed this throughout the manuscript.

Line 201: "by the following" instead of "using the following".

We have used "by the following" instead of "using the following".

Calculation processes and discussion:

Lines 205-209: This has already been said in the former section.

We have removed these sentences from this section.

Line 214: Remove "that" and "all of".

We have removed these words.

Lines 217-223: This description should be greatly improved. Check English, please. Only Cu and Pb for Yicheng area have $\tau''(1)$ values lower than -0.01.

We have revised this sentence as follows as per the reviewer's comments:

"*The multifractal analytical results shown in Table 2 indicate that all of the elements (barring Cu in the Yicheng area) are characterized by a wide range of α values (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*".

Line 225: Use "indices" instead of "elements". You are not talking about the elements but the multifractal indices that you obtained.

The revised version of this manuscript uses three multifractal parameters to study the multifractality of the heavy metal distribution in soils in the study area. We have amended the text to reflect this as follows:

"*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>Zn>As>Cd>Pb>Cu*".

Line 226: "decrease" instead of "decreases". There is a mistake here, Zn have a greater $f(\alpha)$ value than Hg for the Yicheng samples.

We have corrected the text and have discussed all three of the multifractal parameters within the text.

Line 227: This has already been observed in the statistical summary.

We have removed this sentence. However, we have also compared the differences between the statistical summary and the results of our multifractal analysis as follows:

"*Table 3 indicates that the Zn data has largest standard deviation and a moderate coefficient of variation within the Daxing area, but the $\Delta\alpha$ and $\Delta f(\alpha)$ values for these Zn data indicate only weak multifractality compared with other heavy metals. In comparison, the Hg data for soils in the Yicheng area yields the lowest standard deviation 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*".**

Line 229-231: In fact, you are plotting these data.

We have deleted this sentence to make the text more logical.

Lines 232-234: This is not clear. Please, re-phrase it.

We have revised this paragraph, as follows:

"*Multifractal spectra combine the singularity exponent α and the corresponding fractal dimension f(α) to generate a multifractal spectrum with an inverse bell shape (Fig. 3). All of these multifractal spectra are also asymmetric (Δ αL is significantly different from Δ αR, equations 5-6) (barring the Cu data for soils from the Yicheng area), indicating that the soils 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).*"

Line 235: Remove "for all elements".

We have removed "for all elements".

Lines 235-240: I am not sure about understanding this. Please, re-phrase it.

We have revised this sentence as follows:

"*All of these multifractal spectra are also asymmetric (Δ αL is significantly different from Δ αR, equations 5-6) (barring the Cu data for soils from the Yicheng area), indicating that the soils 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).*"

Line 241: "heavy metal contamination of soil" instead of "heavy metal contamination soil contamination".

We have used "heavy metal contamination of soil" instead of "heavy metal contamination soil contamination".

Line 243: "Yicheng area is caused by Hg" instead of "Yicheng area is Hg contamination".

We have used "would be mainly caused by Hg" instead of "is Hg contamination".

Lines 243-244: This is not true. According to table 2, As has a very similar $\Delta f(\alpha)$ value than that of Hg and the value for Zn is even greater than that of Hg.

We have updated this and now use three multifractal parameters to discuss the results of our study.

Line 247: "because this element" instead of "as this element".

We have used "because this element" instead of "as this element".

Line 252: Well, this is not exact. The element from Yicheng samples that showed the highest $\Delta f(\alpha)$ values was Zn, according to table 2.

We have updated this and now use three multifractal parameters together to discuss the results of our study.

Line 253: Remove "showing the distribution of Pb in the Daxing area and Hg in the Yicheng area" since it is already said in the former sentence.

We have removed "showing the distribution of Pb in the Daxing area and Hg in the Yicheng area" from the sentence.

Lines 255-279: This portion of text is a very poor discussion of your results. You did not discuss anything about Daxing contamination. It is also funny that you talk about Hg contamination in Yicheng but the concentrations of this element were greater in the samples of Daxing (see table 1). I am also not sure about the need of performing a multifractal analysis for obtaining these results; a simple geostatistical approach would be enough.

We have added Table 3 and Fig. 5 as well as associated text to enhance our discussion of our results, including comparing the differences between the results of purely statistical summaries and multifractal analysis. Our study indicates that multifractal modeling and the associated generation of multifractal parameters is a useful approach for the evaluation of heavy metal pollution in soils and the identification of major sources of heavy metal contamination.

Lines 283-286: Please, re-phrase this caption, it is not clear.

We have rephrased and simplified this caption to make it more clear.

Lines 289-291: I would change the caption of this figure to "Filled contour map obtained by inverse distance weighted interpolation showing the spatial distribution of soil Pb concentrations in the Daxing area".

We thank the reviewer for their suggestion and have amended the caption for Fig. 4 appropriately.

Lines 294-296: I would modify the caption of this figure to "Filled contour map obtained by inverse distance weighted interpolation showing the spatial distribution of soil Hg concentrations in the Yicheng area".

We have changed the caption of Fig. 5.

Lines 299-302: I would change the caption of this figure to "Filled contour map obtained by inverse distance weighted interpolation showing the spatial distribution of soil Cu concentrations and the location of breeding facilities in the Yicheng area".

We have changed the caption of Fig. 6.

Conclusions:

Line 306: Include “the latter” after “although”.

We have included “the latter” after “although”.

Line 307: Remove “for the soil geochemical data”.

We have removed these words.

Line 309: Remove the word “value” before “changes”.

We have removed the word “value” before “ranges”.

Line 310: There is a mistake here; according to table 2, the $\Delta f(\alpha)$ in Yicheng decreased as Zn>Hg>As>Cd>Pb>Cu instead of Hg>Zn>As>Cd>Pb>Cu as is reported here.

We have updated the conclusions to include this.

Lines 314-319: However, the Hg concentrations in soils from the Daxing area were greater than in Yicheng.

We have updated the conclusions to include this.

Lines 320-326: I am not sure about this conclusion. Further explanations are needed in the discussion section to state this.

We have rewritten the conclusions to make them more clear and to reflect our enlarged discussion section.

References:

Line 343: Use the full name of the journal; in this case it should be “Computers and Geosciences” instead of “Comput. Geosci.”.

We have used the full name of the journal.

Lines 346-347: It should be spelled with a capital letter: “University of Geosciences”.

We have revised this to use a capital letter.

Line 353: It should be spelled with a capital letter: “Journal of Hazardous Materials”

We have revised this to use a capital letter.

We thank Drs Miras-Avalos, J. Miranda and an anonymous referee 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~~Multifractal characteristic-based comparison~~ of the multifractal characteristics of heavy metals~~elements~~ in soils within two~~the Daxing and Yicheng~~ areas of contrasting economic activities in~~Hefei, Anhui Province,~~ China

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Abstract

Industrial and agricultural activities can generate heavy metal pollution that causes~~can have~~ a number of negative environmental and health impacts. This means that evaluating~~identifying areas with~~ heavy metal pollution and identifying the sources of these pollutants, especially in urban or developed areas~~, with multiple possible sources of pollution,~~ is an important first step in mitigating the effects of these contaminating but necessary economic activities. Here, we present the results of a heavy metal (Cu, Pb, Zn, Cd, As and Hg) soil geochemical survey and use these data to evaluate and compare the characteristics of heavy metal pollution in soils with~~outline a new multifractal characteristic-based comparison method that allows~~

~~deeper interrogation of soil geochemistry~~ in urban or developed areas. This survey focuses on Hefei, the provincial capital of Anhui Province, China, an area that contains a number of individual towns within a large municipal area. This study uses~~focuses on the towns of Daxing and Yicheng both of which are incorporated within Hefei and are are economically focused on industry and agriculture, respectively. Here, we use~~ a multifractal spectral technique to identify the multifractality~~differences~~ in the geochemistry of soils within the industrial Daxing and agricultural Yicheng areas of Anhui Province. Determining three ~~. The height difference between the two ends of the~~ multifractal parameters ($\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$) for these soil ~~spectrum of the~~ geochemical data indicates that overall amount of multifractality within the soil geochemical data for the ~~($\Delta f(\alpha)$) for soils in the~~ Daxing area decreases as follows: Pb>Cd>As>Zn>Hg>Cu,~~Pb>As>Cd>Cu>Zn>Hg,~~ whereas the overall amount of multifractality within the soil geochemical data for~~$\Delta f(\alpha)$ values of~~ the ~~geochemical data for soils in the~~ Yicheng area~~town areas~~ decreases as follows: Hg>Zn>As>Cd>Pb>Cu. These differences in the degree of multifractality indicates~~dicate~~ that the soils in these areas have differing multifractal geochemical characteristics, suggesting that the differing economic activities in these areas generate very~~distinctly~~ different heavy metal pollutant loads (e.g. Hg dominated agricultural pollution vs. Pb dominated industrial pollution). In addition, all of the elements barring Hg have larger $\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$~~$\Delta f(\alpha)$~~ values in the Daxing area compared to the Yicheng area. These larger values~~ranges~~ indicate that the higher concentrations of heavy metals present in soils within the Daxing area (compared to the Yicheng area) are more likely to be related to industrial activities than agriculture. The industrial Daxing area contains significant Pb and Cd~~As~~ soil contamination, whereas Hg is the main heavy metal present in soils within the Yicheng area, indicating that differing clean-up procedures and approaches to remediating these polluted areas are needed. The results also indicate that multifractal modeling and the associated generation of multifractal parameters can be ~~needed, rather than~~ a useful~~single~~ approach in the evaluation of~~to~~ heavy metal pollution in ~~. The research presented here also highlights that the~~ soils ~~in these areas (and the source of these~~

~~pollutants) need to be remedied in order to avoid further health~~ and the identification of major sources of heavy metal contamination. ~~environmental impacts.~~

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

1. Introduction

~~Multifractal based analytical techniques have recently been used in a number of differing fields, including geophysics (Schertzer et al., 2011), medicine (Jennane et al., 2001), computer science (Wendt et al., 2009), geology (Deng et al., 2011;Zuo et al., 2012; Cheng, 1995; Yuan et al., 2012), environmental science (Lima et al., 2003; Albanese et al., 2007; Guillén et al., 2011; Salvadori et al., 1997),~~ **and overview of** ~~ecology (Scheuring and Riedi, 1994; Pascual et al., 1995) among others. The advantages of these multifractal techniques include~~ **the study area**

Heavy metal pollution within soil poses a serious risk for human health and the environment, and thus~~fact that these approaches can identify non-linear characteristics, yielding new information that can be used to understand the controls on the distribution of key elements within the objects or data being studied (Gonçalves, 2000;Zuo et al., 2012). Multifractal techniques can also be used to analyze soil characteristics, including the identification of porous structures and the spatial variability in the characteristics of soils (Dathe et al., 2006; Caniego et al., 2005). These techniques and can also enable the characterization of complex phenomena in the spatial distribution of elements within soils, improving our knowledge of the controls on the geochemistry of soils and the regolith (Gonçalves, 2000). This means that these approaches can not only be used in mineral exploration (Yuan et al., 2012; Yuan et al., 2015; Zuo, 2014; Nazarpour et al., 2014) but can also be used in the analysis of pollutants such as heavy metals within soils (Guillén et al., 2011; Salvadori et al., 1997). Heavy metal pollution poses a serious risk for human health and the environment, meaning that~~ soil pollution caused by anthropogenic activities (including industry and agriculture) has been the focus of a significant amount of research in recent years (McGrath et al., 2004; Wang et al., 2007; Leyval et al., 1997; Thomas and Stefan, 2002; Luo~~Chunling~~ et al., 2011). Analyzing soil geochemistry and pollution using ~~This in turn indicates that~~ multifractal techniques has a lot~~enable the more precise identification~~ of advantages, including the fact that these approaches can investigate many~~areas~~ of ~~contamination and~~ the problems~~degree~~ of nonlinear variability which commonly arise when dealing with pollutants and identify non-linear characteristics, 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 using multifractal techniques to determine the multifractal characteristics of the distribution of heavy metals~~contamination~~ in soils can further our understanding of any heavy metal pollution that is associated with these differing activities.

~~polluted areas.~~ Multifractal techniques include~~, such as~~ singularity mapping and multifractal interpolation that~~, also~~ enable more detailed analysis of the spatial distribution of heavy metals, concentration-area modeling that can be used~~ by the use of C–A models~~ to define threshold values between background (i.e. geological) and anthropogenic anomalies (Lima et al., 2003), spectral density-area~~, S–A~~ modeling that can be used to define ~~uses these~~ thresholds to~~ spatially~~ separate anomalies (i.e., anthropogenically derived heavy metal concentrations in this case) from background concentrations (i.e., geologically derived heavy metal concentrations; Cheng, 2001),~~concentrations),~~ and ~~using~~ multifractal spectra that highlights~~to highlight~~ non-linear characteristics and identifies~~identify~~ anomalous behavior that reflects the characteristics of some multifractal sets (Gonçalves, 2000; Albanese et al., 2007; Guillén et al., 2011), such as identification of porous structures and the spatial variability in soil properties and so on (Dathe et al., 2006; Caniego et al., 2005). This means that multifractal techniques provide a lot of useful tools for the the analysis of heavy metals pollutantion within soils (Lima et al., 2003; Albanese et al., 2007; Guillén et al., 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 (Deng et al., 2011; Zuo et al., 2012, 2014; Cheng, 1995; Nazarpour et al., 2014; Yuan et al., 2012, 2015) and ecology (Scheuring and Riedi, 1994; Pascual et al., 1995), among others.

~~2007; Guillén et al., 2011; Lima et al., 2003; Cheng, 2001).~~

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

2. Study area and geochemical data

2.1 Study area

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

2.2 Sampling and analysis

The study areas are covered by Quaternary sedimentary soils and are free of both natural mineralization and mining-related contamination.~~ activities.~~ A total of 169 surface (<20 cm depth)~~below surface)~~ soil samples were taken from the towns of Daxing and Yicheng on 1 × 1 km grids, yielding 78 samples from Daxing and 91 samples from Yicheng (Fig. 1(c–d)). Sampling errors were minimized by splitting each sample into 3–5 sub-samples, each of which weighed more than 500 g. Each of these sub-samples was air-dried~~ in air~~ before being broken up using a wooden roller and then sieved to pass through a 0.85 mm mesh. The concentrations of 6 heavy metal elements (Cu, Pb, Zn, Cd, As and Hg) ~~in the soil samples described above ~~were determined during this study, with~~study.~~ Cd, Cu, Pb and Zn concentrations ~~were~~ determined by inductively coupled plasma–mass spectrometry (ICP–MS) and~~, with~~ Hg and As concentrations determined by hydride generation–atomic fluorescence spectrometry (AFS). These techniques have detection limits of 1 ppm for Cu, 2 ppm for Pb and Zn, 30 ppb for Cd, 0.5 ppm for As and 5 ppb for Hg. The accuracy of these data~~analyses~~ was monitored by repeat determinations~~analysis~~ of standards and replicate determinations~~analyses~~ of sub-sets of samples using instrumental neutron activation analysis (INAA). Analytical precision was monitored using determinations~~analysis~~ of variance of the results obtained from duplicate analyses.

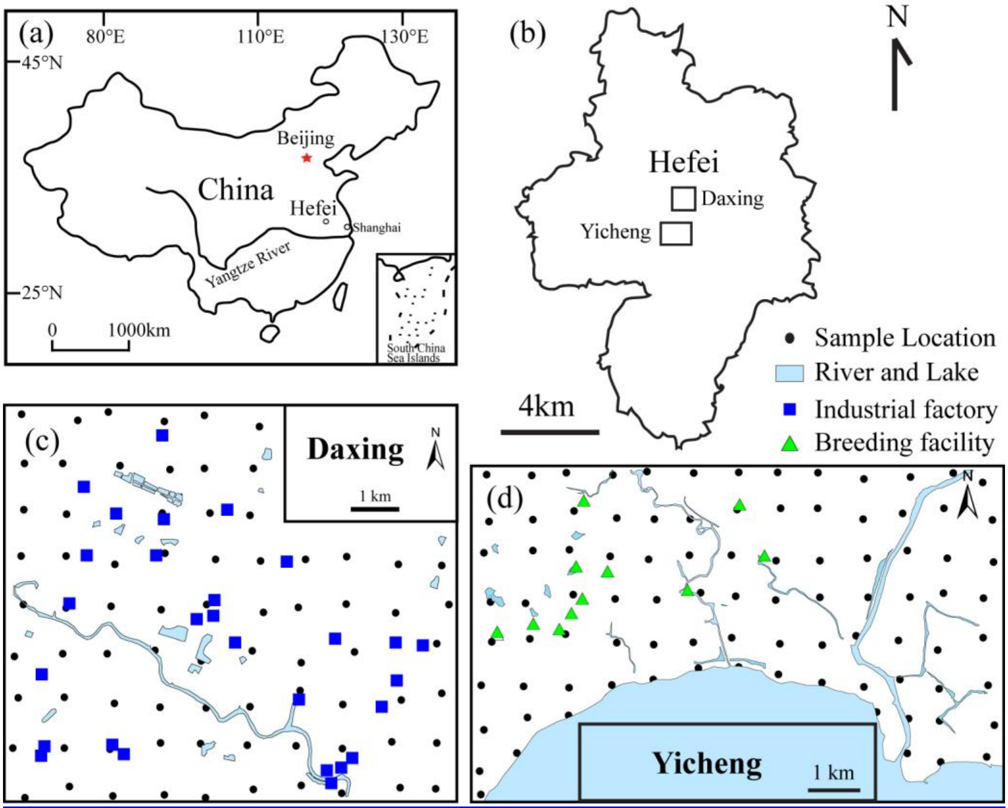
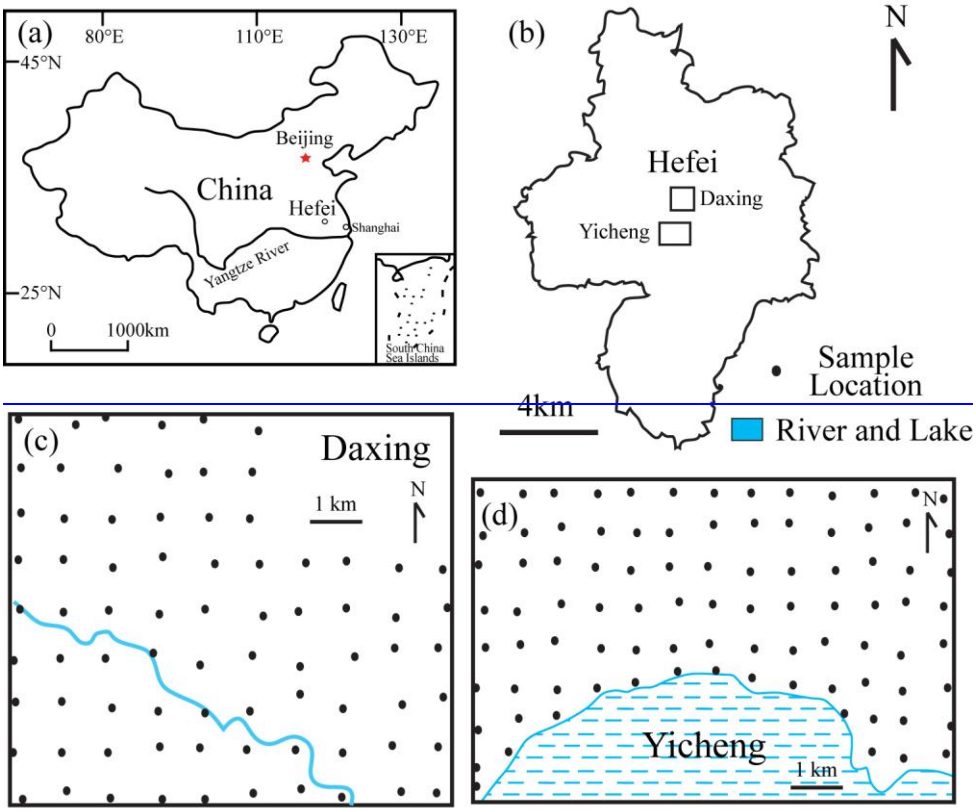


Fig.1. Location~~**2.3 Results**~~

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

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

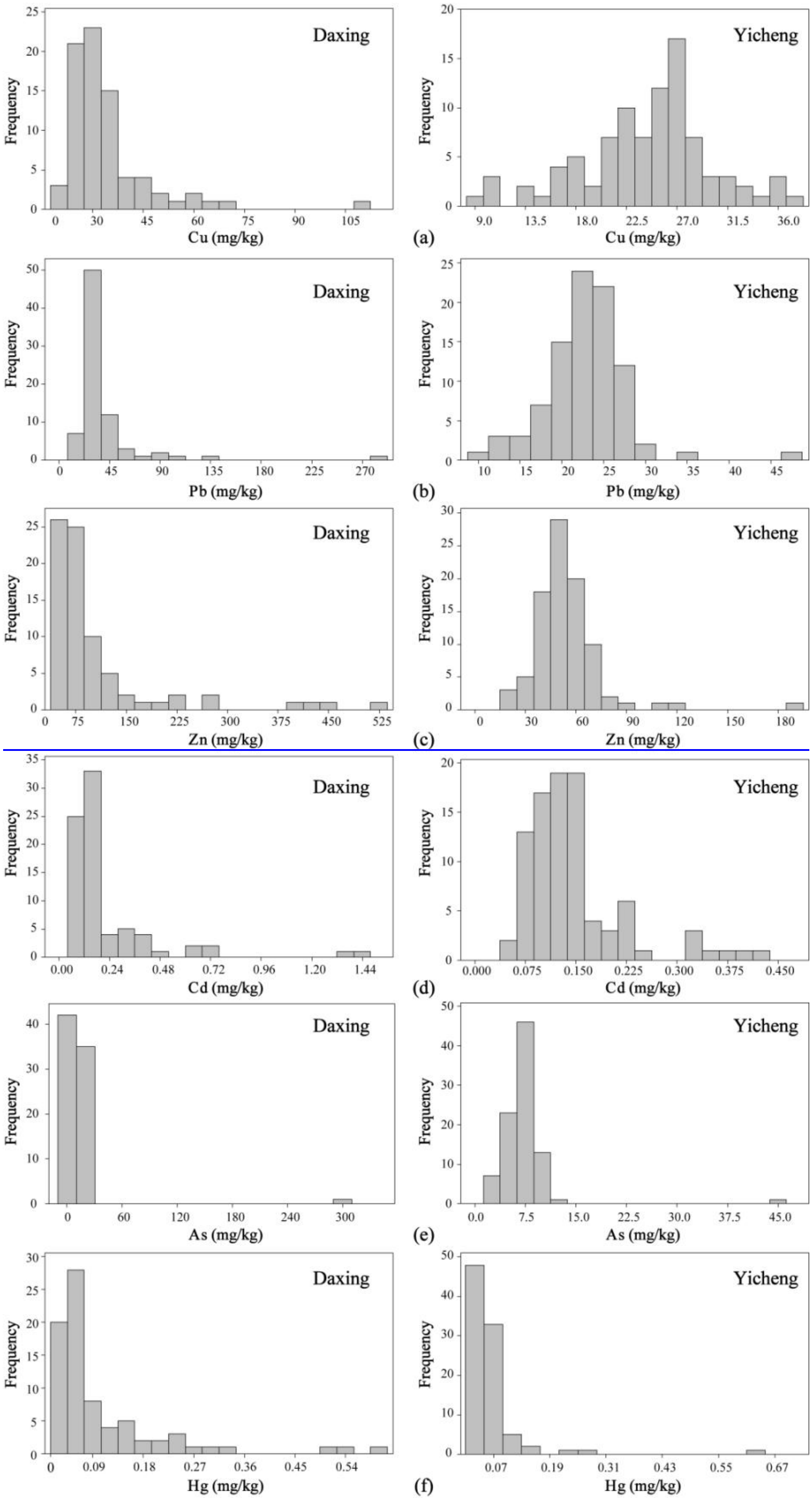


~~Fig.1. (a) Map showing the location of~~ Hefei in central-eastern China (a);~~; (b) Map showing the~~ location of the study areas within Hefei (b); the~~; (c) Map showing the location of soil samples taken in a~~ 1 x 1 km grids used for soil sampling in~~ in the town of Daxing; (d) Map showing the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

~~Table 1. Statistical analysis of soil geochemical data from~~ the towns of Daxing (c) and Yicheng (d)~~.~~

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

~~*CV: coefficient of variation.~~



~~**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.~~

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~~controls on~~ the distribution of key elements within the data. Fractal spectra ($f(\alpha)$)~~($f(a)$)~~ are~~ multifractal~~ formalisms that can be used to describe the multifractal characteristics of a dataset and can be estimated using box-counting based moment, gliding box, histogram and wavelet methods, among others (Cheng, 1999; Lopes and Betrouni, 2009). The most widely used of these methods ~~of estimating $f(a)$ values~~ are the box-counting and gliding box methods, both of which are based on the moment method.

The initial step of the box-counting method estimates mass exponent function $\tau(q)$ 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~~that $\neq$~~ 0.

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

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

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

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

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

$$\alpha(q) = \frac{d\tau(q)}{dq} \tag{3}$$

$$f(q) = q\alpha(q) - \tau(q) = q\frac{d\tau(q)}{dq} - \tau(q) \tag{4}$$

where $\Delta\alpha$~~Δa~~ 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:

$$\Delta\alpha_L = \alpha_0 - \alpha_{min} \tag{5}$$

$$\Delta\alpha_R = \alpha_{max} - \alpha_0 \tag{6}$$

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

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

$$\Delta f(\alpha) = f(\alpha_{max}) - f(\alpha_{min}) \tag{8}$$

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

$$\tau''(1) = \tau(2) - 2\tau(1) + \tau(0) \tag{9}$$

If μ is a multifractal and $-D < \tau''(1) < 0$, where D is~~**4. Calculation processes and discussion**~~

~~The gliding box method used during this study can increase the number of samples that can be used in statistical estimations and provides results with lower uncertainties than~~ the box-counting dimension, then 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~~method. This, combined with~~ the degrees of multifractality within the soil geochemical data for the study area as well as enabling the comparison of the distribution of differing elements in the soils~~relatively sparse sample locations used~~ in this region.

4. Geochemical analysis results

A statistical summary of the soil geochemical data for the study area are given in Table 1. Samples from Daxing have higher Cu, Pb, Zn, Cd and As maximum, standard deviation, skewness, and kurtosis values than soil samples from the Yicheng area. In addition, the soil samples from Daxing have much higher coefficient of variation (CV) values for Cu, Pb, Zn, Cd and As than the samples from the Yicheng area, indicating that soils in the Daxing area contain much higher and more variable concentrations of these elements. This 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 yield 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 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 (mg/kg)	Max (mg/kg)	Mean (mg/kg)	Standard deviation -	Skewness -	Kurtosis -	CV* (%)
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.

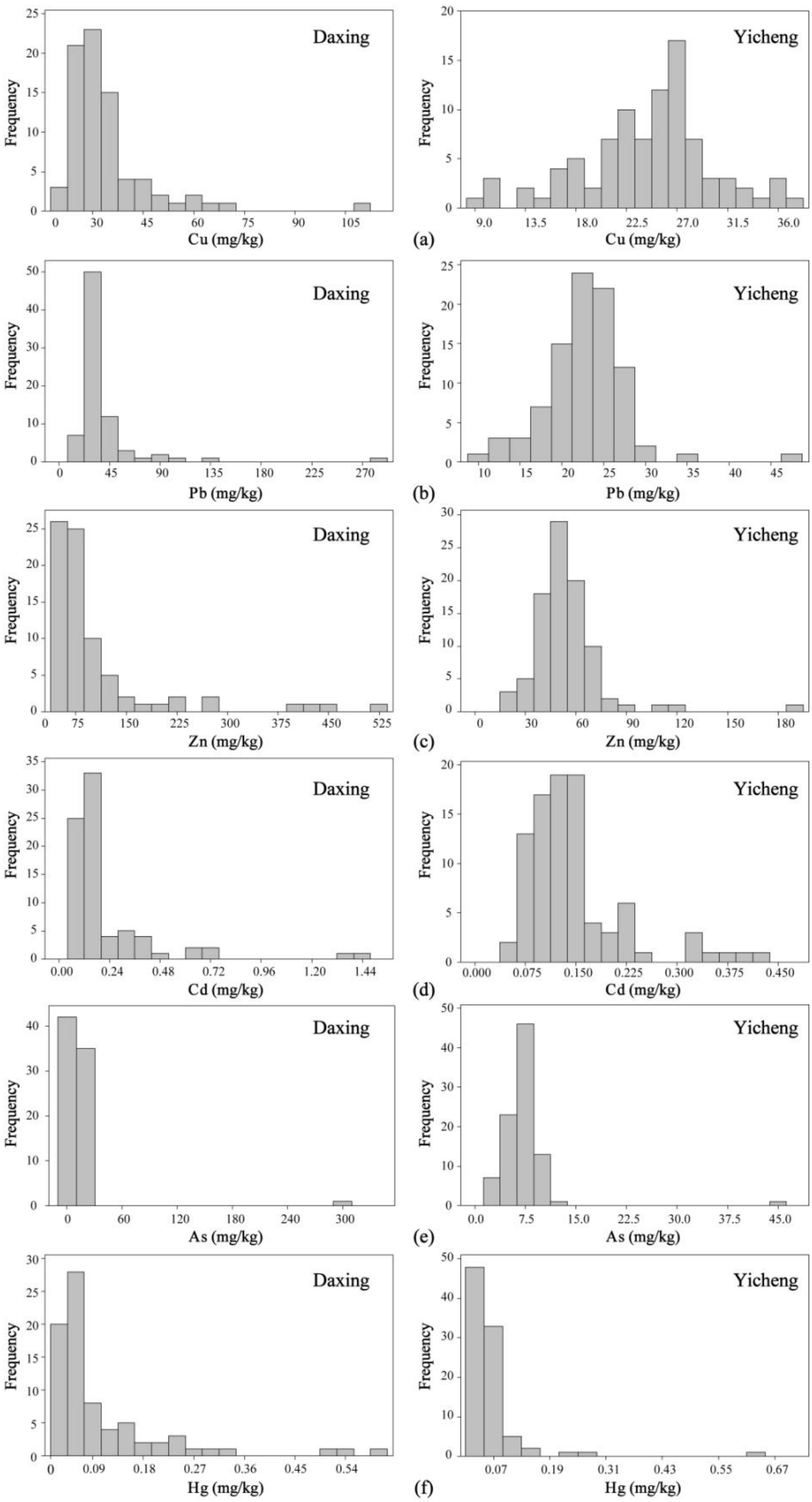


Fig. 2. Histograms showing the distribution of Cu (a), Pb (b), Zn (c), Cd (d), As (e) and Hg (f) concentrations within soils from the towns of Daxing and Yicheng.

5. Calculation processes of multifractal spectrum and discussion

Here, we use~~d~~ the gliding box method to calculate multifractal spectra ~~values~~ for the geochemical data from the study area. This calculation used a range of q values from −10 to 10 with an interval of 1, yielding the multifractal ~~analytical results given in Table 2 and the multifractal~~ spectra (in the form of an α–~~$f\alpha$~~ $f(\alpha)$~~(α)~~ diagram) shown in Fig. 3.

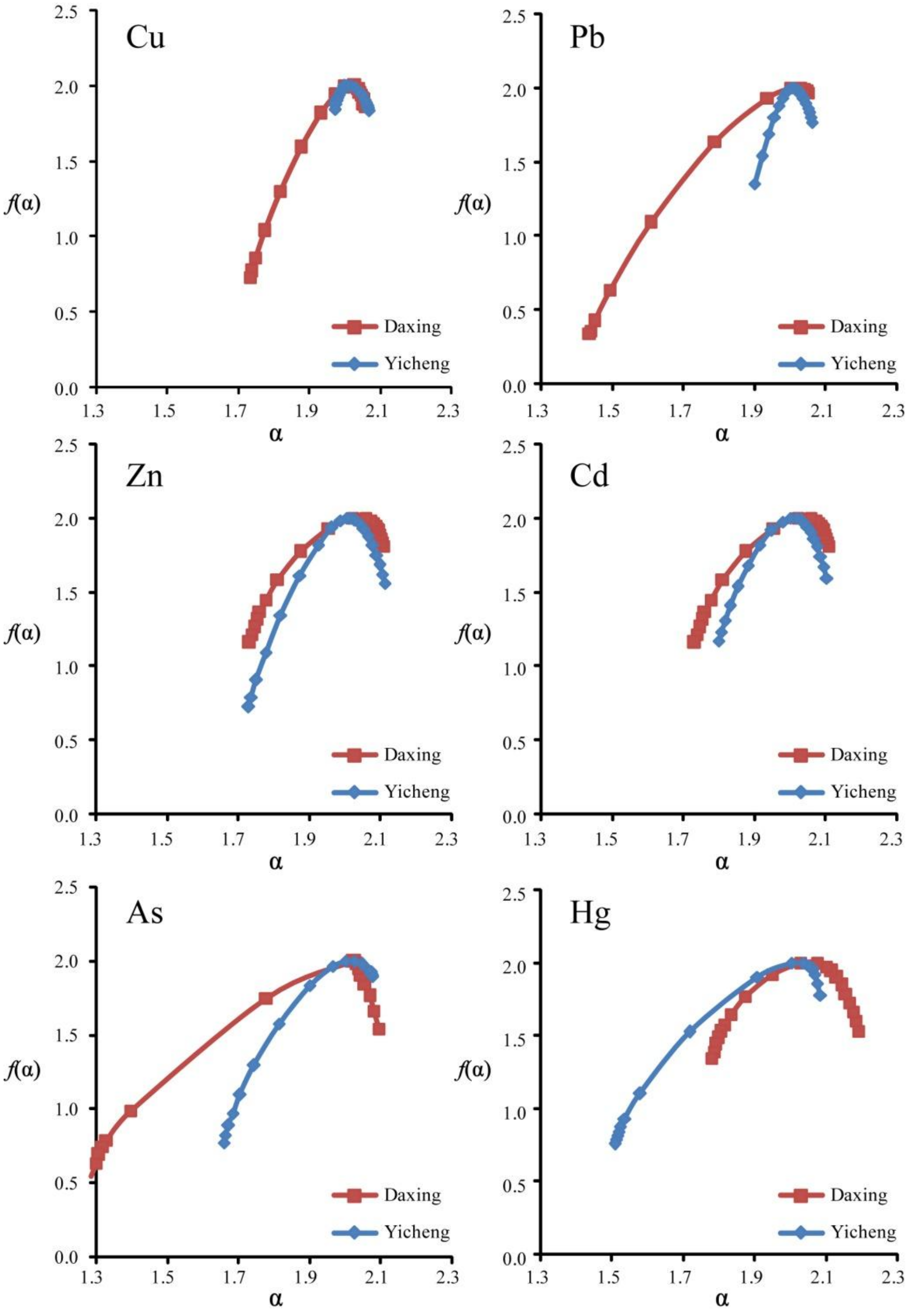


Fig. 3.—

~~**Table 2.**~~ Multifractal spectra (*f* ~~(parameters that describing the multifractality of all 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~~

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

~~The range of $f(\alpha)$ values for the~~ geochemical data (barring Cu) from the Yichen area.

Multifractal spectra combine~~for soils within the Daxing area decreases in the order: Pb>As>Cd>Cu>Zn>Hg, whereas the values for these elements in soils within the Yicheng area decreases in the order: Hg>Zn>As>Cd>Pb>Cu, indicating a significant difference in the geochemical characteristics (and heavy metal pollution) in the soils within these two areas. These variations are linked to multifractal spectra (shown as an α–$f(\alpha)$ plot in Fig. 3), where combining~~ the singularity exponent α and the corresponding fractal dimension $f(\alpha)$ to generate~~generates~~ a multifractal spectrum with an inverse bell shape~~. All of these spectra (barring the data for Cu in soils from the Yicheng area) show a steep increase (i.e. have a good positive correlation between the values) followed by a shorter section of the curve where these values negatively~~

~~correlate~~ (Fig. 3). All of these multifractal spectra ~~data~~ are also asymmetric ($\Delta\alpha_L$ is significantly different from $\Delta\alpha_R$, equations 5-6) (barring the Cu data~~with respect to α~~ for soils from the Yicheng area),~~all elements,~~ indicating that the soils 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).~~individual farms).~~

The multifractal analytical results shown in Table 2 indicate that all of the elements (barring Cu in the Yicheng area) are characterized by a wide range of α values (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~~heavy metals~~ 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 ~~contamination~~ soil contamination, whereas the ~~only~~ significant heavy metal pollution associated with the agricultural activity in the Yicheng area would be~~is~~ Hg contamination. The~~However, the Hg~~ $\Delta f(\alpha)$ and α values of Hg in Yicheng area are larger than all of the other elements in this area as well as some of the elements in the Daxing area, indicating both the prevalence and significant degree of agricultural Hg contamination in the Yicheng area. This is important, primarily as Hg pollution can seriously impact human health because~~as~~ this element is preferentially concentrated upward in the food chain (e.g. (Jiang et al., 2006)), meaning that this contamination needs to be evaluated further and remediated to avoid any deleterious effects.

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

Different elements were sorted by $\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$ parameters 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 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. 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 first averaged the ordering of these elements by $\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$ before sorting again to compare the overall multifractality of these data.

Table 3. Elements sorted by multifractal parameters and coefficient of variation values.

Town	**Element**	**Order**					
		Basic statistics		Multifractal parameters			
		Standard deviation	Coefficient of variation	$\Delta\alpha$	$\Delta f(\alpha)$	$\tau''(1)$	Overall*

Daxing	Cu	4	6	6	4	6	6
	Pb	2	5	3	1	1	1
	Zn	1	4	5	5	2	4
	Cd	5	2	2	3	3	2
	As	3	1	1	2	5	3
	Hg	6	3	4	6	4	5
Yicheng	Cu	2	5	6	6	5	6
	Pb	3	6	5	5	6	5
	Zn	1	4	3	1	4	3
	Cd	5	3	4	4	3	4
	As	4	2	2	3	2	2
	Hg	6	1	1	2	1	1

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

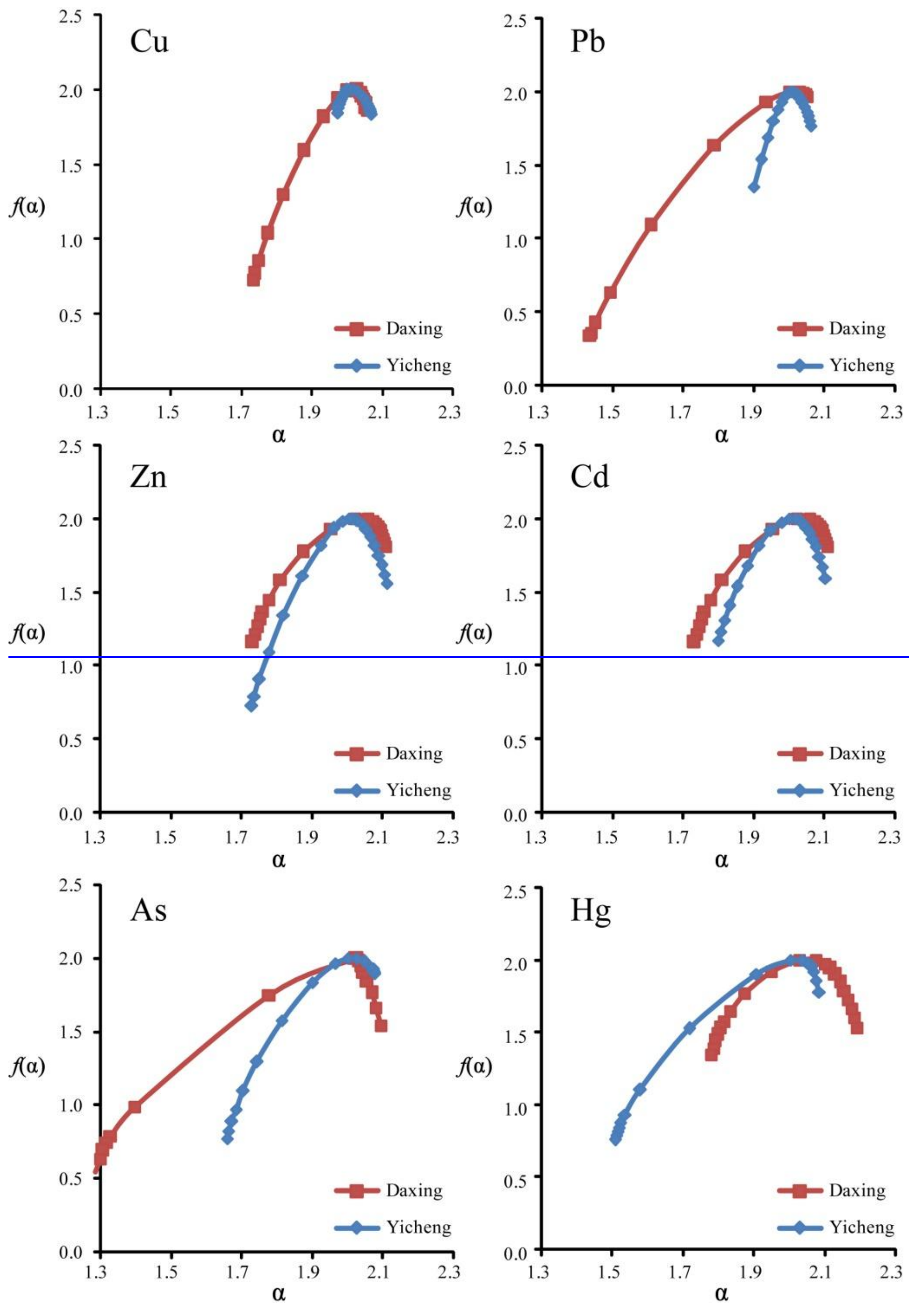
The overall amount of multifractality within the soil geochemical data for the Daxing area decreases as follows: Pb>Cd>As>Zn>Hg>Cu, whereas the overall amount of multifractality within the soil geochemical data for the Yicheng area decreases as follows: Hg>Zn>As>Cd>Pb>Cu. The overall orders indicates 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~~by examining the elements with the highest $\Delta f(\alpha)$ values, namely Pb and Hg, respectively. We used an approach focused on filled~~ contour maps showing the distribution of Pb in the Daxing area and Hg and Cu in the Yicheng area were calculated using inverse distance weighted interpolation (Fig. 4–6).~~4–5).~~ These maps indicate that areas with elevated levels of Pb contamination within the Daxing area are directly correlated to the location of industrial factories, whereas the Hg contamination in the Yicheng area is spatially correlated with the location of agricultural breeding facilities. ~~This strongly suggests that the larger $\Delta f(\alpha)$ values for these elements within the geochemical data are related to the industrial and agricultural activities in the Daxing and Yicheng areas,~~

~~respectively.~~ The Hg contamination in the Yicheng area is of significance, especially as this form of contamination can cause serious health issues (e.g. Minamata disease). As such, the soils in this area may well require remediation, especially as Hg can be concentrated up the food chain and the Yicheng area is heavily agricultural, indicating that this activity may both be concentrating Hg as well as contaminating soils in this area.

This distribution of soils with elevated concentrations of Hg also contrasts with the symmetrical distribution and weakest multifractality~~relatively low $\Delta f(\alpha)$ values~~ for Cu within the Yicheng area (Fig. 3, 5-6). We used a plot showing~~3). Comparing~~ the rank~~distribution~~ of concentration contour vs number of agricultural facilities within ~~Cu and Hg in~~ the same rank of concentration contour to demonstrate~~filled contours maps for~~ the ~~Yicheng area (Fig. 5–6) indicates an anti-correlation in terms of the~~ spatial correlation between the location of agricultural facilities and heavy metal concentrations in soils (Fig. 7). This diagram shows an significant correlation between agricultural facilities and ~~anomalously~~ high concentrations of Hg, whereas there is an anti-correlation when comparing agricultural ~~Cu and~~ breeding facilities and areas of high Cu concentrations.~~.~~ This indicates that very little Cu has been anthropogenically added (or removed) from the soils in the Yicheng area, suggesting that these soils maybe contain only natural background concentrations of Cu and that the agricultural activity in this area does not produce ~~any~~ significant Cu contamination.

All of the above suggests~~These data indicate~~ that the multifractal parameters for the~~differing clean-up procedures and approaches to remediating these polluted areas are needed, rather than a single approach to~~ 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~~pollution. The~~ results also indicate that multifractal modeling and the associated generation of multifractal parameters~~, such as $\Delta f(\alpha)$ values,~~ are a useful approach in the evaluation of heavy metal pollution in soils and the identification of major element of heavy metal contamination. In addition, the differing orders 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 clean-up procedures and approaches to remediating these polluted areas are needed, rather than a single cover-all approach to the remediation of heavy metal pollution. A significant amount of different remediation approaches can be used to resolve the issues of heavy metal soil contamination (e.g., Bech et al., 2014; Koptsik, 2014), with the results presented in this study suggesting that physical and chemical approaches (soil removal, soil vitrification, soil consolidation, electroremediation, soil washing) are more appropriate for the remediation of heavy metal contaminated soil in the Daxing area, especially 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 in this area (Mulligan et al., 2001;Wang et al., 2006).



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

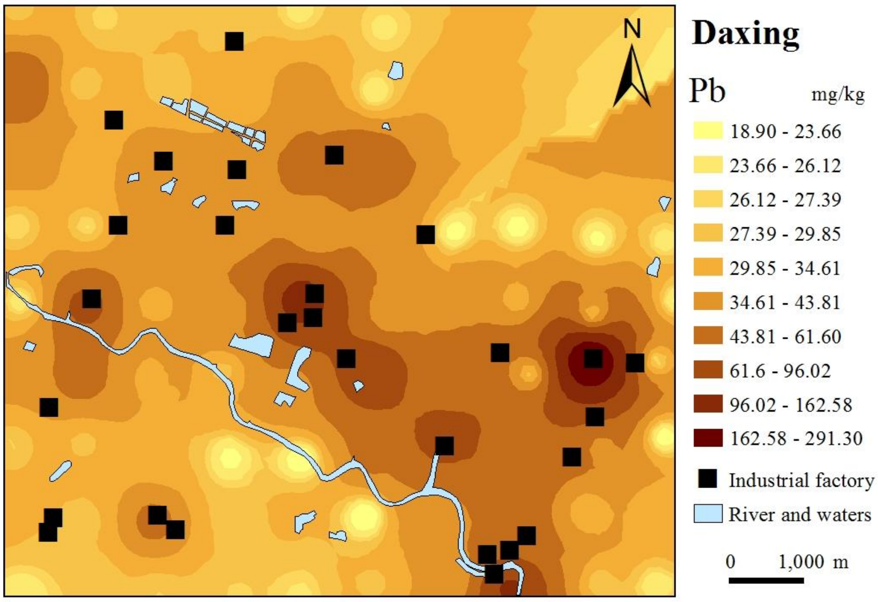


Fig. 4. Filled contour map generated~~s produced~~ by inverse distance weighted interpolation showing the spatial distribution of soil Pb concentrations~~Pb~~ in the Daxing area.~~ and the clear correlation between Pb contamination and the location of heavy industry.~~

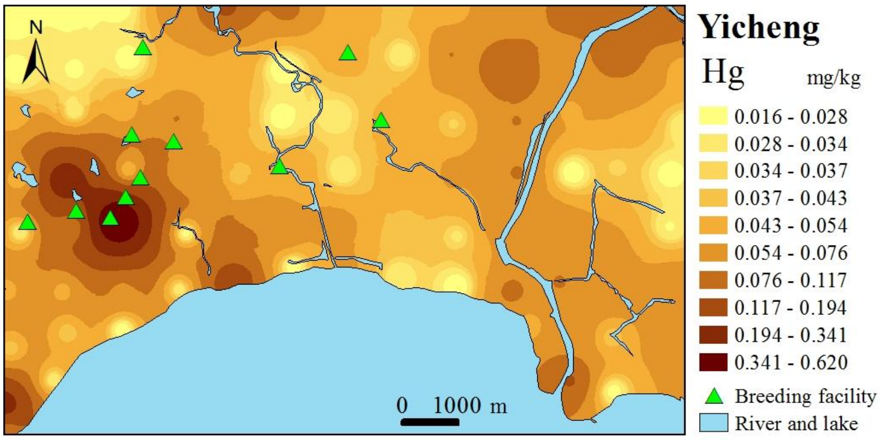


Fig. 5. Filled contour map generated~~s produced~~ by inverse distance weighted interpolation showing the spatial distribution of soil Hg concentrations~~Hg and the clear spatial relationship with the location of breeding facilities~~ in the Yicheng area.

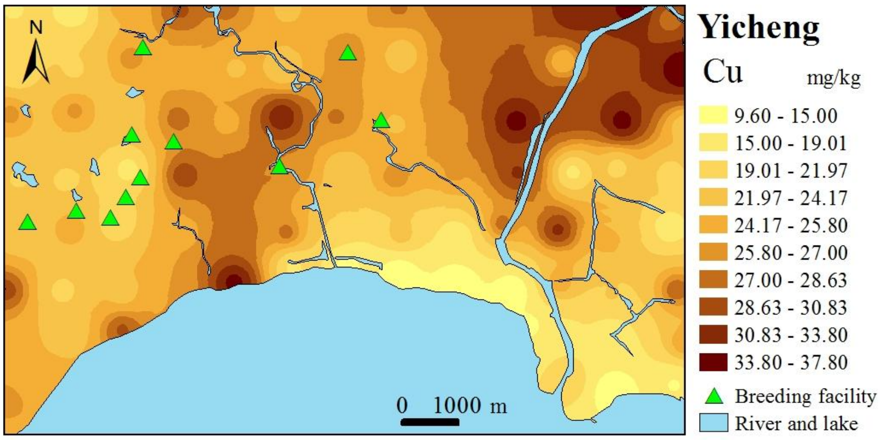


Fig. 6. Filled contour map generated~~s produced~~ by inverse distance weighted interpolation showing the spatial distribution of soil Cu concentrations~~Cu~~ and the location of breeding facilities in the Yicheng area

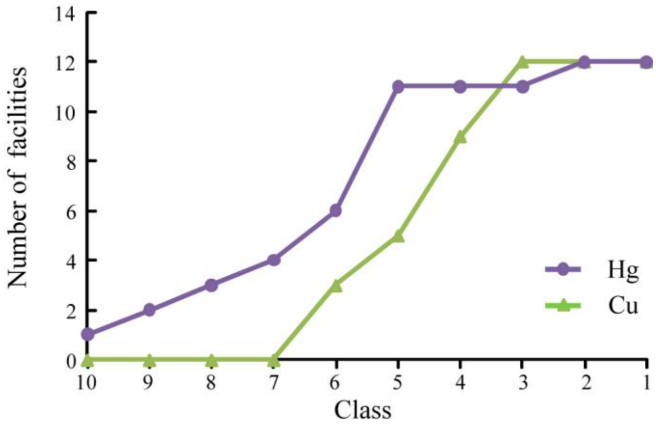


Fig. 7. Plot of number of agricultural facilities in Yicheng area within the same rank of Hg and Cu concentration contour showing a positive spatial correlation between location of agricultural facilities and Hg concentrations but an anti-correlation between the location~~; this distribution shows that the distribution~~ of agricultural facilities and Cu concentrations.~~in soils in this region is unlike the Hg contamination in this area.~~

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 ~~Our data~~ indicate that the soils from the Daxing area have stronger multifractality~~a larger range of $f(\alpha)$ values~~ for Cu, Pb, Zn, Cd and As than soils from the Yicheng area, although the latter have relatively strong multifractality~~have a larger range in $f(\alpha)$ values~~ for Hg. The ordering~~range~~ of~~ $f(\alpha)$~~ values for the multifractal parameters $\Delta\alpha$, $\Delta f(\alpha)$ and $\tau''(1)$ indicate the degree of multifractality for the~~soil~~ geochemical data for soils within the Daxing area descends as follows: Pb>Cd>As>Zn>Hg>Cu,~~decreases in the order Pb>As>Cd>Cu>Zn>Hg,~~ whereas the overall order in soils within the Yicheng area descends as follows:~~have $f(\alpha)$ value ranges that decrease in the order~~ Hg>Zn>As>Cd>Pb>Cu. In addition, Cu concentrations in soils in the Yicheng area may still have their original (i.e. natural) distribution and may not have been influenced by human activities. These data indicate that the industrial activity concentrated in the Daxing area generates multi-element heavy metal soil contamination whereas the agricultural activity concentrated in the Yicheng area generates Hg-~~—~~dominated heavy metal soil contamination. The latter is important, as Hg contamination can cause serious health issues (e.g. Minamata disease) and the soils in this area may well require remediation, especially as Hg can be concentrated up the food chain and the Yicheng area is heavily agricultural, indicating that this activity may both be concentrating Hg as well as contaminating soils in this area.

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

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