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# Status and fuzzy comprehensive assessment of metals and arsenic contamination in farmland soils along the Yanghe River, China

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Concentrations of metals and arsenic in farmland soils along the Yanghe River, upstream of Guanting Reservoir, were evaluated using fuzzy comprehensive assessment (FCA). FCA is an emerging methodology for assessing environmental status based on fuzzy logic which can model linguistic data and represent chaotic conditions. The ratio of concentrations of metals and arsenic (As) in surface soils of farmland along the Yanghe River to the corresponding reference values decreased in the order Cd > Zn > Cr > Ni > Pb > Cu > As. Based on the FCA, 86% of surface soils along the Yanghe River were classified as grade C, which is suitable for forestry with a greater absorption capacity, suggesting that most farmland along the Yanghe River has to be abandoned as farmland. In order to improve the quality of soil along the Yanghe River and avoid degeneration of water quality in the river and the Guanting Reservoir, remediation of farmland soils along the river is suggested.

Keywords: fuzzy assessment; heavy metal; contamination; drinking water; cluster assessment

### 1. Introduction

Because of their persistence and long half-lives for elimination from the body, some metals in soils have attracted worldwide attention [1,2]. Concentrations of metals in soils are influenced by natural and anthropogenic factors, including parent materials, mining, land use, application of pesticides and fertilisers, effluent from metallurgical industries, use of paint pigments, and irrigation with wastewater. In China, nearly  $2.0 \times 10^5$  km<sup>2</sup> of cultivated land has been contaminated by metals [3]. Cadmium (Cd) is the most widespread contaminant, present in  $2.8 \times 10^3$  km<sup>2</sup> arable land in 25 regions in 11 provinces, and 50 million kg of Cd-contaminated rice was produced in 1999 [4]. Metals can accumulate in crops and, through consumption, damage animal and human tissues

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[5]. Soil is a significant interface environment which can be a source of pollution to surface and ground waters, living organisms, sediments and oceans [6].

Four well-known pollution events along the Yanghe River took place in 1988, 1992, 1993 and 2009, respectively. The total amount of farmland damaged in the first three events was  $\sim 68 \text{ km}^2$ , and direct economic losses were estimated to be US\$4.6 million. In 2009, the total area of farmland damaged was  $\sim 12-18$  km<sup>2</sup>. Crops, especially rice, but also vegetables, grapes and corn, were affected. Most farmland was irrigated with polluted water from the Yanghe River, which is the main tributary flowing into Guangting Reservoir. Historically, Guanting Reservoir has been used as one of the two sources of water for agriculture, industry and municipalities in Beijing. Since the 1970s, the Yanghe River watershed has undergone significant agricultural and industrial development and consequently water in Guanting Reservoir has become contaminated. This is a serious issue because Beijing currently has a severe shortage of water [7]. The Yanghe River watershed is a natural and ecological shelter zone for Beijing, as well as an important source of water. Farmland can be polluted by irrigation with wastewater, which may then, through run-off from farmland, be a source of metals to aquatic ecosystems [8]. The contamination of farmland therefore has the potential to deteriorate water quality in the Yanghe River and Guanting Reservoir. Assessments of soil quality and the protection of farmland along the Yanghe River are therefore important for safeguarding water quality in Guanting Reservoir, which will ultimately serve as one of the main sources of drinking water for Beijing for the foreseeable future.

In recent years, several investigations into metals in soils around Guanting Reservoir have been conducted [7,9,10]. However, there is limited information about the concentrations of metals in and the environmental quality of farmland along the Yanghe River, upstream of Guanting Reservoir. The results presented here provide critical data for policy makers to improve the quality of the soil along the Yanghe River and drinking water supplies for Beijing.

One of several methods that have been used in environmental quality assessment [11], is fuzzy comprehensive assessment (FCA) which is based on fuzzy logic [6]. Fuzzy set theory, introduced by Zadeh in 1965, reduces inconsistency in data and vagueness in interpretation, and is useful in assessing and classifying soils [12]. It is useful in solving problems of fuzzy boundaries and controlling for the effects of monitoring errors on assessment results [13]. Fuzzy evaluation methods are used to decrease fuzziness when comprehensively evaluating the contributions of various pollutants according to pre-established weights based on membership functions [14]. Thus, its sensitivity is greater than other indices that have been used in the evaluation of contamination [15].

The objectives of this study were to: (1) determine the concentrations of the metals cadmium (Cd), copper (Cu), nickel (Ni), lead (Pb), chromium (Cr) and zinc (Zn) and the metalloid arsenic (As) in the surface soils of farmland along the Yanghe River; (2) use multivariate analysis to determine the relative proportions coming from natural or anthropogenic sources; and (3) apply FCA to assess the current overall soil environment quality along the Yanghe River to guide the development of management and land-use practices.

#### 2. Materials and methods

#### 2.1. Study area

The Yanghe River is located in Hebei Province, China (latitude, 40.33–40.74°; longitude 114.71– 115.55°), where it flows through valleys and enters Guanting Reservoir. The climate of the region is cool temperature, continental monsoons, with an average annual precipitation of 394.4 mm and an annual average temperature of 9.95 °C. The Yanghe River is 106 km long with a total catchment area of 9762 km<sup>2</sup>. The primary rock types along the Yanghe River are intermediate and acidic igneous; and the soil types are fluvo-aquic (FAS), calcareous-cinnamon (CCS), fluvo-cinnamon (FCS) and meadow-wind sandy (MWSS) [16]. Land use in the region includes farms and orchards, as well as fallow land. Most areas are dedicated to agricultural cultivation, predominated by corn and a variety of vegetables. Agrochemicals were applied to the farmland along the Yanghe River, with an average chemical fertiliser application of  $\sim 107 \text{ kg} \cdot \text{ha}^{-1}$  and pesticide usage of  $\sim 4.5 \text{ kg} \cdot \text{ha}^{-1}$ . There is also industrial development, such as the Xuanhua iron and steel plant which was established 80 years ago, along the river.

## 2.2. Soil sampling

Sampling sites were selected based on the distribution of farmland along the Yanghe River, with more sampling locations in areas with more intensive farming. Throughout the survey, a global position system (GPS) was used to locate the sampling locations (Figure 1). Thirty-six surface soil sampling locations were identified. Five subsamples of topsoil (0–10 cm) were collected at each location and mixed thoroughly to form a representative composite sample. This resulted in a total of 36 representative surface soil samples. Most soil samples were collected from corn fields. Samples were extracted using an automatic core drill.

# 2.3. Metal quantification

Soil was air-dried, crushed in an agate mortar, passed through a nylon 100-mesh sieve and digested with HNO<sub>3</sub> (15.8 mol·L<sup>-1</sup>) and H<sub>2</sub>O<sub>2</sub> (30%) using Method 3050B [17]. Samples were carefully handled to avoid the introduction or loss of trace elements during preparation and analysis. All materials used during analytical determinations were kept in Teflon or other metal-free containers. The concentrations of the metals, Cd, Cu, Ni, Pb, Cr and Zn, and the metalloid As in the digestion solution were determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES). The method detection limit (MDL), which is defined as the minimum concentration of



element that can be measured and reported with 99% confidence, was determined using EPA 40CFR Part 136, Appendix B. Standard reference materials, GSS-1 soils, obtained from the Center of National Standard Reference material of China were analysed as part of the quality assurance and quality control (QA/QC) procedures. Satisfactory recoveries were obtained for Cr (107–113%), Cd (94–102%), Zn (108–114%), Cu (94–98%), Pb (102–106%), Ni (102–103%) and As (105–106%). Concentrations were expressed on a dry weight (dw) basis. The pH values were measured by a standard method [18]. Soil pH was measured at a soil/water ratio of 1:2.5 using a potentiometric glass electrode.

#### 2.4. Statistical analyses

Statistical analyses were conducted by use of Microsoft Excel and SPSS 13.0 statistical software on a personal computer. The distribution of concentrations was tested with the Kolmogorov–Smirnov method to determine whether they approximated the normal probability distribution. Differences between mean concentrations of measured parameters and the corresponding reference value were tested using a one-sample *t*test. The level of significance was set at p < 0.05 based on two-tailed tests.

To evaluate the relationships among concentrations of individual elements, correlation analysis and cluster analysis (CA) were used. Pearson's correlation coefficient (for normally distributed concentrations), r, was used to measure the strength of a linear relationship between metals and arsenic. CA was used to elucidate the latent relationships between metal and arsenic in farmland soils along the Yanghe River, and to investigate metal sources. Hierarchical CA was performed using the following settings: the linkage type used was furthest neighbor and the distance method was Pearson's correlation.

#### 2.5. Fuzzy comprehensive assessment

The FCA was conducted using previously described methods [11,12,13,19,20]. Briefly, the methods consisted of the following steps. Assessment parameters were selected that were representative, rational and accurate to form an assessment factor set U, which is based on the actual local situation (Equation 1).

$$U = \{\mathbf{u}_1, \mathbf{u}_2, \dots \mathbf{u}_n\} \tag{1}$$

Where n is the number of selected assessment parameters (7 in the current assessment). The assessment criteria set V was established from the National Environmental Quality Standards of China (Equation 2).

$$V = \{\mathbf{v}_1, \mathbf{v}_2, \dots \mathbf{v}_m\} \tag{2}$$

Where *m* is the number of assessment criteria categories. The Environmental Quality Standard for Each Element in Soil (China, GB15618-1995) (Table 1) was used as the assessment criterion. In the Environmental Quality Standard for Soil in China (GB15618-1995) soils can be classified into three grades (A, B and C). Grade A soils are those in natural conservation areas, tea gardens or soils in which metals occur at natural background concentrations. Grade B soils consist of farmland, vegetable, tea, fruit and grazing land. Grade C soils are suitable for forestry or areas with greater absorption capacity. In conclusion, grades A, B and C are defined as pristine, moderately enriched and extremely impacted, respectively [12].

The next step was to formulate the membership function, which represents the degree to which the specified concentration belongs to the fuzzy set. The membership degree of assessment parameters at each level can be described quantitatively by a set of formulae of membership functions

	Soil grade							
	В							
Metal	А	pH < 6.5	рН 6.5–7.5	pH > 7.5	С			
Cd	0.2	0.3	0.3	0.6	1			
As	15	40	30	25	40			
Cu	35	50	100	100	400			
Pb	35	250	300	350	500			
Cr	90	150	200	250	300			
Zn	100	200	250	300	500			
Ni	40	40	50	60	200			

Table 1. Maximum metal concentrations prescribed by the Environmental Quality Standard for Soil in China (GB15618-1995) (mg·kg<sup>-1</sup>, dw).

Note: Soil grade A, natural conservation area, drinking water catchments, tea garden. Metals in soil at natural background values. Soil grade B, farmland, vegetables, tea, fruit and grazing land. Soil grade C, forestry and land with a higher absorption capacity.

(Equations 3–5).

$$u_1(x) = \begin{cases} 1 & x \le v_1 \\ (v_2 - x)/(v_2 - v_1) & v_1 < x < v_2 \\ 0 & x \ge v_2 \end{cases}$$
(3)

$$u_{2}(x) = \begin{cases} 0 & x \le v_{1}, x \ge v_{3} \\ (x - v_{1})/(v_{2} - v_{1}) & v_{1} < x \le v_{2} \\ (v_{3} - x)/(v_{3} - v_{2}) & v_{2} < x < v_{3} \end{cases}$$
(4)

$$u_{3}(x) = \begin{cases} 0 & x \le v_{2} \\ (x - v_{2})/(v_{3} - v_{2}) & v_{2} < x \le v_{3} \\ 1 & x \ge v_{3} \end{cases}$$
(5)

Where x is the actual monitoring data (raw metals value) of any assessment parameters, grades A, B and C correspond to limits  $v_1$ ,  $v_2$  and  $v_3$ , respectively.  $u_1(x)$ ,  $u_2(x)$ ,  $u_3(x)$  are the membership degrees of assessment parameter (x) for classification grades A, B or C, respectively.

The next step was calculation of the membership function and matrix formation. This was done by substituting the monitoring data of each assessment parameter at each location and the national standards into the membership functions. From this, the fuzzy matrix  $\tilde{R}$  was expressed (Equation 6).

$$\tilde{R} = \begin{bmatrix} u_{11} & u_{12} & \cdots & u_{1m} \\ u_{21} & u_{22} & \cdots & u_{2m} \\ \vdots & \vdots & & & \\ u_{n1} & u_{n2} & \cdots & u_{nm} \end{bmatrix}$$
(6)

Where  $r_{ij}$  (i = 1, 2...n; j = 1, 2, ...m) is the membership degree of the *i*th assessment parameter at the *j*th level. The weight matrix was then developed by allocating weights of each assessment parameter at each monitoring site to generate matrix B (Equations 7 and 8).

$$W_i(k) = a_{i(k)} / \sum_{i=1}^n a_{i(k)}$$
 (7)

$$a_{i(k)} = x_{i(k)}/s_i \tag{8}$$

Where k is the monitoring site,  $a_{i(k)}$  is the monitored concentration of the *i*th assessment parameter at the kth monitoring site,  $s_i$  is the average assessment criterion of the *i*th assessment parameter,  $W_{i(k)}$  represents the weight of the *i*th assessment parameter at the kth monitoring site.  $\tilde{B}(k)$ , weight matrix  $\tilde{B}$  at the monitoring site k, can be expressed (Equation 9).

$$B(k) = [W_{1(k)}, W_{2(k)}, \dots W_{n(k)}]$$
(9)

Where *n* is the number of selected assessment parameters.

It is important to choose an appropriate weight and several methods are available to do so. For example, some researchers have determined weight according to soil environmental quality standards. Here the weights were based on not only soil quality assessment criteria, but also the above-standard principles. Here  $s_i$  is an average assessment criterion,  $a_{i(k)}$  which specifies the magnitude of exceeding the average assessment criterion. Thus, this parameter includes not only the difference between each element, and the criterion, but the magnitude of contamination.

Determination of the fuzzy algorithm of  $\tilde{B} \times \tilde{R}$  can be computed by matrix multiplication. The fuzzy matrix (Equation 10) can be combined with the weight matrix (Equation 11) and rearranged to obtain (Equation 12).

$$R = (u_{ij})_{n \times m} \tag{10}$$

$$\tilde{B} = (W_i)_{1 \times n} \tag{11}$$

$$\tilde{B} \times \tilde{R} = (\mathbf{b}_1, \mathbf{b}_2, \dots, \mathbf{b}_m), \text{ where } \mathbf{b}_j = \sum_{i=1}^n w_i u_{ij}, \quad (j = 1, 2, \dots, m)$$
 (12)

#### 3. Results and discussion

## 3.1. Concentrations of metals and arsenic in soils

The descriptive statistics for concentrations of metals, As and pH value in farmland soils along the Yanghe River are presented (Table 2). The pH was generally slightly alkaline. Because the probability of the raw datasets of Zn was skewed, it was necessary to transform data prior to further statistical analyses. The Kolmogorov–Smirnov test indicated that concentrations of Cr, Cd, Pb, Ni, As and Cu followed normal distributions, and Zn was ln-normally distributed (K–S p > 0.05) Considering the concentrations of metals in the dominant soil and rock types along the Yanghe River and its geography, geology, landscape and climate, which are close to those of Beijing, heavy metal background levels in Beijing soils [21] were used as reference values for unpolluted soils along the Yanghe River.

The results of the summary statistics (Table 2) indicated that mean concentrations of most metals, except Zn, were significantly different using a one-sample *t*-test (p < 0.05) from their corresponding reference values. The mean concentration of Cd (1.56 mg·kg<sup>-1</sup>, dw) was greater than its background value (0.15 mg·kg<sup>-1</sup>, dw) and also significantly greater than its critical value (1 mg·kg<sup>-1</sup>g, dw) [22] associated with serious ecological effects. It can thus be concluded that surface soils of farmland along the Yanghe River were contaminated with Cd. Cd is relatively mobile in soils [23], and thus could migrate to the Yanghe River, or even Guanting Reservoir.

The mean concentration of Zn was  $2.9 \times 10^2 \text{ mg} \cdot \text{kg}^{-1}$ , dw, which was significantly greater than the corresponding reference value of 59.6 mg $\cdot$ kg<sup>-1</sup>, dw. Twenty-four samples significantly exceeded 59.6 mg $\cdot$ kg<sup>-1</sup>, dw. The range of Zn (4179 mg $\cdot$ kg<sup>-1</sup>, dw) between the minimum and maximum values was the greatest of the elements studied and had a coefficient of variation of 249% and skewness of 4.74, which were also the greatest of the elements studied. This observation

Heavy metal	$\begin{array}{c} \text{Range} \\ (\text{mg}{\cdot}\text{kg}^{-1}) \end{array}$	$\underset{(mg\cdot kg^{-1})}{Min}$	$\begin{array}{c} Max \\ (mg \cdot kg^{-1}) \end{array}$	$\frac{\text{Mean} \pm \text{SD}}{(\text{mg} \cdot \text{kg}^{-1})}$	Coefficient of variation %	Skewness (mg·kg <sup>-1</sup> )	Kurtosis (mg·kg <sup>-1</sup> )	K–S P	Reference value $\pm$ SD <sup>b</sup>	Number exceeding reference values <sup>c</sup>
Cr	92.53	1.98	94.51	$61.92 \pm 17.41^*$	28.13	1.19	3.48	0.58	$31.3 \pm 9.29$	34
Cd	4.75	ND	4.75	$1.56 \pm 0.90^{*}$	57.62	0.91	3.69	0.47	$0.15 \pm 0.11$	33
Zn	4178.72	ND	4178.72	$291.61 \pm 726.73$ NS	249.21	4.74	24.71	0.12 <sup>a</sup>	$59.6 \pm 16.29$	24
Cu	39.91	ND	39.91	$8.54 \pm 9.70^{*}$	113.63	1.39	1.84	0.15	$19.7\pm6.33$	6
Pb	38.45	ND	38.45	$13.4 \pm 8.42^{*}$	62.80	0.68	0.86	0.93	$25.1\pm5.08$	2
Ni	19.92	15.45	35.37	$24.09 \pm 5.54^{*}$	23.01	0.20	-0.98	0.84	$27.9 \pm 7.90$	10
As	10.06	ND	10.06	$1.99 \pm 2.44^{*}$	122.92	1.65	2.51	0.09	$7.81 \pm 3.22$	1
pН	0.80	8.05	8.85	$8.46\pm0.24$	0.01	-0.05	-0.91			

Table 2. Summary statistics of heavy metald in soils along the Yanghe river (mg  $kg^{-1}$ , dw).

Note: Significant at the 0.05 (\*) level of probability when compared with reference value. NS, not significant, comparing with reference value. ND, not detectable. <sup>a</sup>Dataset is In-transformed. <sup>b</sup>Chen et al. [21]. <sup>c</sup>The number whose value is significantly greater than the corresponding reference value.

is the result of the heterogeneity of concentrations of Zn with a few locations having greater concentrations. The mean concentration of Cr was  $61.92 \text{ mg} \cdot \text{kg}^{-1}$ , dw, which was greater than the corresponding reference value of  $31.3 \text{ mg} \cdot \text{kg}^{-1}$ , dw. Concentrations of Cr in 34 of the samples significantly exceeded the reference value. Thus, Zn and Cr contamination in farmland soils along the Yanghe River should also be given attention.

Mean concentrations of Ni, Cu and Pb were 24, 8.5 and  $13 \text{ mg} \cdot \text{kg}^{-1}$ , dw, respectively. The number of samples significantly exceeding the corresponding reference value for Ni, Cu and Pb were 10, 6 and 2, respectively. The mean concentration of As was  $1.99 \text{ mg} \cdot \text{kg}^{-1}$ , dw) and the concentration of As exceeded the reference value in only one sample Thus, it can be concluded that As contamination in farmland soils along the Yanghe River is minimal. Ratios of the mean concentrations in farmland soils along the Yanghe River divided by the corresponding reference value decreased in the order Cd > Zn > Cr > Ni > Pb > Cu > As. Multiplying the ratios by the number of values significantly exceeding the respective reference provides a combined estimation of both the magnitude and extent of contamination along the Yanghe River. This weighted result suggests that the magnitude of heavy metal pollution in farmland soils along the Yanghe River decreases in the order Cd > Zn > Cr > Ni > Pb > As.

#### 3.2. Relationships and sources of metals and arsenic in surface soil

Correlation analysis was conducted to determine the extent of association among metals in farmland soils along the Yanghe River (Table 3). The results from correlative analysis showed that combined pollution exists in the soil. Concentrations of the metalloid As were weakly correlated with concentrations of metals in farmland soils along the Yanghe River, which suggests different sources from other metals. Concentrations of Cd were significantly correlated (p < 0.01) with concentrations of Cr and Pb, and less correlated (p < 0.05) with concentrations of Zn. There were significant correlations between concentrations of Cu and Cr, Cu and Zn, Ni and Cr, Ni and Cu in soils along Yanghe River.

The correlation analysis provides little information about the sources of metals and As. Thus, a cluster analysis was performed using the furthest neighbour linkage method based on Pearson's coefficients (Figure 2). The distance cluster represents the degree of association between elements. The smaller the value on the distance cluster the more significant the association. A criterion for distance clusters requiring that they be between 10 and 15 was adopted.

In farmland soils along the Yanghe River, four distinct clusters were identified (Figure 2). Cluster I contains Cd, Cr and Pb. The soils along the Yanghe River are deficient in phosphate. Phosphate fertiliser has been applied in large quantities to increase agricultural production, the local pyrite and phosphate rocks used by the phosphate fertiliser plant contained large concentrations of Cd (1.94 and 0.258 mg·kg<sup>-1</sup> dw for pyrite and phosphate rocks, respectively) [24]. In addition,

			-				
	Cr	Cd	Zn	Cu	Pb	Ni	As
Cr	1						
Cd	0.55**	1					
Zn	0.22	0.42*	1				
Cu	$0.40^{*}$	0.05	$0.44^{*}$	1			
Pb	0.22	0.43**	0.20	0.18	1		
Ni	0.33*	0.17	-0.16	0.34*	0.24	1	
As	-0.11	0.07	0.13	-0.16	-0.19	0.02	1

Table 3. Correlation coefficients of heavy metal contents

Notes: \*Correlation is significant at the 0.05 level (two-tailed). \*\*Correlation is significant at the 0.01 level (two-tailed).



Figure 2. Dendrogram of the cluster analysis of metals and arsenic contents.

wastewater irrigation and land application of sludge are likely important sources of Cd and Cr in soils along the Yanghe River [25]. Also, the elevated Pb concentrations in farmland soils have been shown to be most likely due to the use of Pb-based insecticides, manure and sewage sludge [26]. Thus, it can be deduced that Cd, Cr and Pb probably came from the application of fertiliser and pesticides, wastewater irrigation and sewage sludge.

Cluster II contains Cu and Ni. Cu concentrations are enriched in the predominant rocks of the Yanghe River catchment [27]. Thus, natural weathering of rocks is likely to be the major source of Cu in soils along the Yanghe River. The relatively small concentrations of Ni in soils were similar to those of the predominant minerals along the Yanghe River, which had Ni concentrations of 22.6 and 19.9 mg·kg<sup>-1</sup>, dw, respectively [27]. This means that both Cu and Ni may originate from the natural parent materials of the soils.

Cluster III contains As. The concentration of As in two major rock types along the Yanghe River were 8.1 and 7.7 mg·kg<sup>-1</sup>, dw [27], approaching or exceeding its reference value (7.81 mg·kg<sup>-1</sup>, dw) (Table 2). This result suggests that As pollution in some soils along the Yanghe River is due to natural causes.

Cluster IV contains Zn. Zn is an ingredient in insecticides, especially fungicides [28], and fertiliser use has been implicated in elevated Zn concentrations in soils [29]. However, the two predominant rock types, intermediate igneous rock and acidic igneous rock, in the catchment of Yanghe River had Zn concentrations of 86.6 and  $73.8 \text{ mg} \cdot \text{kg}^{-1}$ , dw, respectively [27], both of which were greater than the reference of Zn,  $59.6 \text{ mg} \cdot \text{kg}^{-1}$ , dw. It can be concluded that both the natural and anthropogenic sources have contributed to the observed concentration of Zn in farmland soils of the Yanghe River.

#### 3.3. Fuzzy comprehensive assessment of metals in farmland soils

Copper, Zn, Pb, Cr, Cd, Ni and As were selected as assessment parameters to form an assessment factor set 'U', based on concentrations in soil along the Yanghe River. An assessment criteria set 'V' was also established based upon the Environmental Quality Standards for Soil in China (Table 1).

According to the formulae of the membership function, the membership degrees of each assessment parameter can be calculated at grade A, B and C. After substitution of actual monitoring data and standards at each level, fuzzy matrices of  $\tilde{R}_1 \sim \tilde{R}_{36}$  and weight matrices  $\tilde{B}_1 \sim \tilde{B}_{36}$  were obtained. The standards of grade B are variable for different soil pH values, so during calculation, it was important to substitute relevant standards. Since all of the pH values in surface soils of farmland along Yanghe River were >7.5, the appropriate grade B of assessment criteria for Cr, Cd, Zn, Cu, Pb, Ni and As were 250, 0.6, 300, 100, 350, 60 and 20 mg·kg<sup>-1</sup>, respectively.

Predominant contaminants can be identified from the corresponding weight matrices. For example, at location 1, the average assessment criteria for Cr, Cd, Zn, Cu, Pb, Ni and As were 213.33, 0.60, 300, 178.33, 295, 100 and 21.67 mg·kg<sup>-1</sup>, respectively. Monitored concentrations of these metals at site 1 were 71, 2.1,  $1.7 \times 10^2$ , 11, 24, 29 and  $6.2 \times 10^{-1}$  mg·kg<sup>-1</sup>, respectively. From these, the values of  $\tilde{B}_1 = (0.07 \ 0.72 \ 0.12 \ 0.01 \ 0.02 \ 0.06 \ 0.01)$ . Thus, it is concluded that Cd was the element that exceeded the criterion by the greatest magnitude. The results of the fuzzy assessment for location 1 are given as an example to explain the calculation process. The first step in the assessment was to calculate the membership function matrix, the  $\tilde{R}_1$  of site 1 (Equation 13).

$$\tilde{R}_{1} = \begin{vmatrix} 0.96 & 0.04 & 0.00 \\ 0.00 & 0.00 & 1.00 \\ 0.54 & 0.46 & 0.00 \\ 1.00 & 0.00 & 0.00 \\ 0.99 & 0.01 & 0.00 \\ 1.00 & 0.00 & 0.00 \\ 1.00 & 0.00 & 0.00 \end{vmatrix}$$
(13)

The next step was the calculation of the weights matrix  $\tilde{B}_1$ . Each row of the  $\tilde{R}_1$  and the column of  $\tilde{B}_1$  represents Cr, Cd, Zn, Cu, Pb, Ni and As, respectively. Finally, the fuzzy algorithm of  $\tilde{B} * \tilde{R}$  was computed by matrix multiplication, fuzzy algorithms of B\*R for site 1 were derived (Equation 14).

$$\tilde{B}_1 \times \tilde{R}_1 = (0.24\ 0.04\ 0.72) \tag{14}$$

Because the membership degree of grade C (0.72) is greater than that of grade A (0.24) and grade B (0.04), the soil environmental quality at location 1 is classified as grade C. Using this approach, FCA results were calculated for all of 36 locations:

Site 1 $\tilde{B}_1 \times \tilde{R}_1 = (0.24\ 0.04\ 0.72)$ Site 2 $\tilde{B}_2 \times \tilde{R}_2 = (0.16\ 0.00\ 0.84)$
Site 3 $\tilde{B}_3 \times \tilde{R}_3 = (0.10\ 0.00\ 0.90)$ Site 4 $\tilde{B}_4 \times \tilde{R}_4 = (0.25\ 0.06\ 0.75)$
Site 5 $\tilde{B}_5 \times \tilde{R}_5 = (0.22\ 0.00\ 0.78)$ Site 6 $\tilde{B}_6 \times \tilde{R}_6 = (0.28\ 0.00\ 0.72)$
Site 7 $\tilde{B}_7 \times \tilde{R}_7 = (0.24\ 0.00\ 0.76)$ Site 8 $\tilde{B}_8 \times \tilde{R}_8 = (0.21\ 0.01\ 0.79)$
Site 9 $\tilde{B}_9 \times \tilde{R}_9 = (1.00\ 0.00\ 0.00)$ Site 10 $\tilde{B}_{10} \times \tilde{R}_{10} = (0.85\ 0.15\ 0.00)$
Site 11 $\tilde{B}_{11} \times \tilde{R}_{11} = (0.27\ 0.00\ 0.73)$ Site 12 $\tilde{B}_{12} \times \tilde{R}_{12} = (0.20\ 0.00\ 0.79)$
Site 13 $\tilde{B}_{13} \times \tilde{R}_{13} = (0.20\ 0.12\ 0.68)$ Site 14 $\tilde{B}_{14} \times \tilde{R}_{14} = (0.29\ 0.00\ 0.71)$
Site 15 $\tilde{B}_{15} \times \tilde{R}_{15} = (0.04\ 0.00\ 0.96)$ Site 16 $\tilde{B}_{16} \times \tilde{R}_{16} = (0.16\ 0.06\ 0.84)$
Site 17 $\tilde{B}_{17} \times \tilde{R}_{17} = (0.22\ 0.00\ 0.78)$ Site 18 $\tilde{B}_{18} \times \tilde{R}_{18} = (0.30\ 0.01\ 0.69)$
Site 19 $\tilde{B}_{19} \times \tilde{R}_{19} = (0.26\ 0.00\ 0.74)$ Site 20 $\tilde{B}_{20} \times \tilde{R}_{20} = (0.07\ 0.00\ 0.93)$
Site 21 $\tilde{B}_{21} \times \tilde{R}_{21} = (1.00\ 0.00\ 0.00)$ Site 22 $\tilde{B}_{22} \times \tilde{R}_{22} = (0.28\ 0.00\ 0.72)$
Site 23 $\tilde{B}_{23} \times \tilde{R}_{23} = (0.07\ 0.00\ 0.93)$ Site 24 $\tilde{B}_{24} \times \tilde{R}_{24} = (0.28\ 0.00\ 0.72)$
Site 25 $\tilde{B}_{25} \times \tilde{R}_{25} = (0.24\ 0.01\ 0.75)$ Site 26 $\tilde{B}_{26} \times \tilde{R}_{26} = (0.28\ 0.01\ 0.71)$
Site 27 $\tilde{B}_{27} \times \tilde{R}_{27} = (0.39\ 0.01\ 0.60)$ Site 28 $\tilde{B}_{28} \times \tilde{R}_{28} = (0.37\ 0.03\ 0.60)$
Site 29 $\tilde{B}_{29} \times \tilde{R}_{29} = (1.00\ 0.00\ 0.00)$ Site 30 $\tilde{B}_{30} \times \tilde{R}_{30} = (0.31\ 0.00\ 0.69)$

Site 31 
$$\tilde{B}_{31} \times \tilde{R}_{31} = (0.20\ 0.08\ 0.72)$$
 Site 32  $\tilde{B}_{32} \times \tilde{R}_{32} = (0.29\ 0.00\ 0.71)$   
Site 33  $\tilde{B}_{33} \times \tilde{R}_{33} = (1.00\ 0.00\ 0.00)$  Site 34  $\tilde{B}_{34} \times \tilde{R}_{34} = (0.27\ 0.00\ 0.73)$   
Site 35  $\tilde{B}_{35} \times \tilde{R}_{35} = (0.32\ 0.00\ 0.68)$  Site 36  $\tilde{B}_{36} \times \tilde{R}_{36} = (0.24\ 0.00\ 0.76)$ 

Interpreting these results as we did for site 1, we can conclude that the environmental quality of 31 soils along Yanghe River would be classified as grade C which is defined as extremely impacted; while only five soils (9, 10, 21, 29 and 33) were classified as grade A, which is defined as pristine. Sites 9 and 10 were in corn fields, and also close to each other. This result might be due to irrigation with groundwater that was not polluted. In the north of China, farmers often irrigate their land with groundwater because of a lack of surface water. Site 21 was in a vegetable field, whereas sites 29 and 33 were collected from orchards. Different land-use types can cause different contamination statuses of metals because of different metal application quantity, metal remediation capacities, cultivation pattern and soil properties [10]. Thus, it could be hypothesised that land use affected the concentration of metals and As in farmland soils. FCA of the overall soil environmental quality along the Yanghe River gives a clear picture of the current situation, and provides a scientific basis for further environmental planning and comprehensive management.

# 4. Conclusion

The metals of greatest concern in farmland soils along the Yanghe River were Cd, Zn and Cr. In order to improve the quality of the soil along the Yanghe River and drinking water supplies for Beijing from the Guanting Reservoir, concentrations of these metals in surface soils should not be allowed to increase and if possible should be decreased. Thus, national and local governments need to set as a priority the continuous monitoring and control of metal contamination along the Yanghe River, for example, the introduction of reasonable application of phosphate fertiliser, cleaner technology for treatment or control of domestic waste discharge, not allow activities that would add any additional Cd, Zn and Cr. On account of the chaotic conditions of soil environmental measurements, the FCA method provides a scientific basis for analysing and evaluating the environmental quality of soils along the Yanghe River. Consequently, most of the land along the Yanghe River will have to be abandoned as farmland, to prevent contamination of the food chain and any associated hazards to human health. Other feasible alternative land uses include hydroponic cultivation of vegetables in greenhouses and/or forestry land could also be trialled.

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