

Spatio-Temporal Characteristics and Source Apportionment of Water Pollutants in Upper Reaches of Maotiao River, Southwest of China, from 2003 to 2015

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ABSTRACT. The Maotiao River is playing an indispensable role in protecting water quality of the Yangtze River of China. Its hydro-power development also provides adequate power and clean resources for the local areas. To understand the water quality of the upper reaches (i.e., Maijia River), seven indices such as dissolved oxygen (DO), chemical oxygen demand (COD_{Cr}), biochemical oxygen demanded (BOD₅), ammonia nitrogen (NH₃-N), total nitrogen (TN), total phosphorus (TP) and fluoride of samples collected from 4 sites from 2003 to 2015 were studied using multiple analysis approaches. For winter-spring and summer-autumn seasons, pictures of spatio-temporal characteristics were presented and the reasons behind their variation trend were elaborated. The Canadian Council of Ministers of the Environment Water Quality Index (CCME WQI) was evaluated to concisely mark the water quality. Principal component analysis (PCA) was applied to identify the source of pollutants. The results showed that the water quality status in Maijia River was poor from 2008 to 2011 and acceptable from 2003 to 2007, and 2012 to 2015, respectively. The COD_{Cr}, NH₄-N and TN were considered to be the primary pollutants during winter-spring and summer-autumn seasons. The quality of Maijia River was influenced strongly by human activities. Environmental treatment and pollution sources of the middle and lower reaches of the river need to be focused. This study paves a way to improve the ecological environment of Maotiao River and overall water quality management of the middle and upper reaches of Yangtze River.

Keywords: Upper reaches of Maotiao River, CCME WQI, principal component analysis, spatio-temporal characteristics, source apportionment of water pollutants

1. Introduction

Water shortages and deterioration of water quality become more serious because of industrial development and urbanization (Zheng et al., 2011). Particularly, increasing industrialization leads to ever increasing pollution of rivers in developing countries (Jonnalagadda and Mhere, 2001). Water resources have become a constraint on China's sustainable development (Jiang, 2015). Moreover, surface water quality affects human health.

The Maotiao River located in the center of Guizhou Province, Southwest of China is a branch of the Wujiang distributary of the Yangtze River. Being an important part of upper reaches of the Yangtze River, the Maotiao River plays an indispensable role in protecting water quality of the Yangtze River. However, the Yangtze River basin is facing severe issues such as degradation of water quality (Wu et al., 2013). Due to its pe-

culiar geological environment and shallow depth of groundwater of karst regions, ground water is vulnerable to pollution from human activities, and once contaminated its remediation is a difficult and time consuming project (Lang et al., 2006). In the light of high interchange between surface and ground water hydrologic systems, identification of the cause of pollution in order to alleviate the increasingly serious problem of water quality deterioration in karstic areas is highly significant. However, data that characterize the sources and spatial and temporal distribution in this area are still very limited. To our knowledge, no study has ever systematically addressed the pollution characteristics and trends in the upper reaches of Maotiao River.

Recently, water quality assessment has become a critical issue (Tsakiris et al., 2015) because it can effectively distinguish the factors of potential threats to human health and help governments formulate relevant policies (Wang et al., 2017). The surface-groundwater system is a typical hydrogeological system in Guizhou Province with the utilization of karst water resources accounts for 80% of the total water resources, which is extremely sensitive to human activities (Li et al., 2018). It is essential to find adaptive methods to assess spatial-temporal patterns and trends in water quality of the area. To the best of

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our knowledge, in China, there are some typical methods for evaluating water quality i.e., single factor assessment method (Yan et al., 2016a), pollution index method (Li et al., 2015), fuzzy method (Jia and Zhang, 2011), grey system method (Ip et al., 2009), analytic hierarchy process method (Zhang et al., 2015) and artificial neural network method (Bo et al., 2018). However, these methods have their disadvantages. For instance, a typical problem of fuzzy method is that the difficulty of determining the weights of experts' evaluations, which induces inaccurate judgment of quality of the water (Baranyi et al., 2005; Zhou et al., 2016). Artificial neural network method requires greater computational resources and leads to overfitting a training data set with poor performance in external test data sets (Tu, 1996), which is not suited to the explicit computational object in the present study. Although, other methods mentioned above could depict the status of water quality in section, they are unable to explain the relationship between sections and characterize spatio-temporal characteristics of water pollutants (Mei et al., 2014). In contrast, the Canadian Council of Ministers of the Environment Water Quality Index (CCME WQI) is an important technique in water quality assessment (Yan et al., 2016b), which has been used in many countries and received great assessments because of its several merits (Tunc Dede et al., 2013; Venkatraman et al., 2016; Li et al., 2017a; Bilgin, 2018). Compared with different forms of water quality indices, CCME WQI has several advantages including flexibility in selection of input parameters and tolerance to missing data (Terrado et al., 2010). Moreover, the CCME WQI provides a convenient means of summarizing complex water quality data that can be easily understood by people from various fields such as the public, water distributors, planners, managers and policy makers (Lumb et al., 2006). In terms of main weaknesses, the WQI is affected by the insensitivity towards a particular parameter in the process of aggregation (Lumb et al., 2011). Above all, the index method provides significant information about a particular water-body and describes trends in the results in an accessible, clear and simple manner, which avoids considering assignment of relative weights to each parameter by subjective judgments (Salcedo-Sánchez et al., 2016). The CCME WQI as an effective tool has been used to characterize the quality of drinking source (Hurley et al., 2012) and its application is widespread in the environmental management of rivers, lakes and reservoirs (Akkoyunlu and Akiner, 2012; Gao et al., 2016). It has also been applied to compare the water quality parameters between the shrimp farm water supply lagoon and coastal environments (Ferreira et al., 2011). In order to directly evaluate strategies by control agencies to improve water quality and use by authorities, decision-makers and evaluators of water quality, CCME WQI was applied in the case study.

Defining the rules of spatio-temporal change of water quality has been a major focus of water hydrology (Chang, 2008), which would allow water management authorities to take adequate measures (Abaurrea et al., 2011). Multivariate statistical techniques such as correlation analysis, cluster analysis (CA), principal component analysis (PCA) and factor analysis (FA), are usually used for the evaluation of both spatio-temporal

variations and the interpretation of large and complex water quality datasets. These can serve as powerful tools for surface/ground water resources on a local or even on a regional scale (Singh et al., 2013). Although multivariate analysis has been used to assess surface water quality in a number of studies from different regions of China (Wang et al., 2006; Li et al., 2007, 2017b; Vadde et al., 2018; Yang et al., 2018; Zhang et al., 2018). However, no such assessment has so far been made in the karst hydrological system. The strong interchange between surface water and groundwater is frequent in the karstic area, and the development or contamination of one commonly affects the other (Tao et al., 2011). Amidst this general scenario, the spatio-temporal variation of water quality and source apportionment of pollutants affect the quality of groundwater in the region that if understood clearly, can lead to appropriate strategies and determine priorities for river pollution control and effective water resources management of the karstic area.

Previous study reported that nutrient concentrations and fluxes through river network vary strongly over time and nutrient residence times can be on the order of decades in ambient medium, which lead to quantify the effectiveness of changes in land management and requires both long-term and high-frequency monitoring (Abbott et al., 2018). Most studies only used a limited number of monitoring periods which offered a limited value for understanding the dynamics of changing water quality, and did not explicitly consider the issues of scale over time. For example, limited length of data (5 years) precludes the assessment of the actual effects on groundwater recharge in Trier-Petrisberg, southwestern Germany (Kessler et al., 2012). Konrad and Booth (2002) demonstrated that 10 years' datasets were not adequate to represent long-term changes in hydrologic analysis and were likely to involve other factors such as climate variability and vegetation water uptake. Thus, the study of water quality assessment model necessitates long-time monitoring of stream flow in a catchment to discard any climate effects (Hamel et al., 2013). It is imperative to select more appropriate time periods (> 10 years) and more representative sites in the upper reaches of the Maotiao River to analyze the spatio-temporal distribution characteristics of the water quality parameters.

In this study, a large data matrix of 13 years (2003 ~ 2015) was obtained from four monitoring sites during winter-spring and summer-autumn seasons. The aims of the study were to: (1) assess the four sites (S1, S2, S3 and S4, detailed in the following section) with seven water quality parameters of upper reaches of the Maotiao River; (2) determine the classification of water quality; and (3) identify water quality variables responsible for spatio-temporal variations in river water quality. In addition, the study combined the usefulness of multivariate statistical techniques (CA, DA and PCA) with CCME WQI in improving our understanding of pollution characteristics of surface water in the karst region. The results will offer great significance for improving the ecological environment of the Maotiao River and can be beneficial to the understanding and management of groundwater in the region.

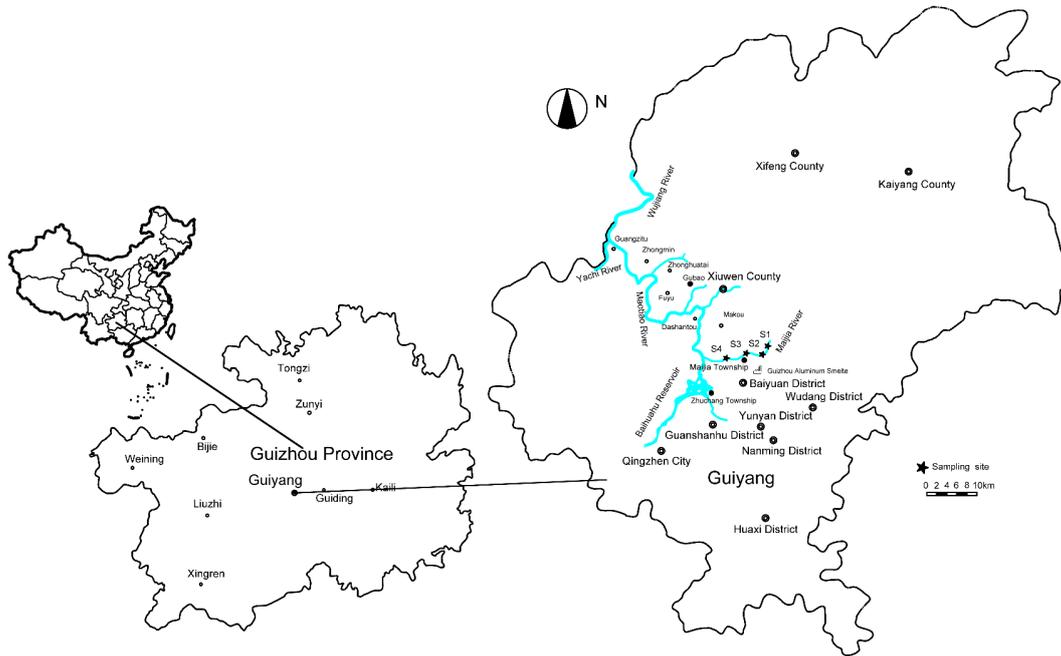


Figure 1. Sampling sites of the Maijia River with the location in Baiyun District of Guiyang City.

2. Materials and Methods

2.1. Study Area

The Maijia River is located in Guiyang, Guizhou Province of China. It is the upper reaches of the Maotiao River and the biggest river in Baiyun District of Guiyang, originating in the west of Zhouwu Mountain of Xiuwen County, flowing through the middle and southwest low-laying mountainous basin and valleys, leading to the Maotiao River. The region has an annual average temperature of 12.5 to 14.5 °C and receives an average annual precipitation of 1,147 to 1,191 mm. The Maijia River is 26.15 km long, with a total catchment area of 150.2 km² having annual mean runoff of 2.70 m³/s. With the objective of exploring water pollution problems in the Maijia river of Guiyang, sampling campaigns were conducted during the winterspring and summer-autumn seasons from 2003 to 2015. Four sites along the Maijia River were selected as study sites (Figure 1), which were determined by the Environmental Protection Bureau of Guizhou Province according to the procedure of combining river mixing length, human activities, and geographic information systems. Sites in the Maijia River included S1 (106°40'48" E, 26°43'48" N), S2 (106°40'12" E, 26°44'24" N), S3 (106°37'48" E, 26°43'12" N), and S4 (106°35'59" E, 26°42' 36" N).

2.2. Sample Analysis

According to China's national environmental quality standard for surface water (GB 3838-2002), the selected detection indices, including dissolved oxygen (DO), chemical oxygen demand (COD_{Cr}), biochemical oxygen demanded (BOD₅), ammonia nitrogen (NH₃-N), total nitrogen (TN), total phosphorus (TP), and fluoride, were analyzed semi-annually, according to water and

wastewater monitoring analysis method (SEPA, 2002) in the laboratory except DO, which was determined in situ. The instrument detection limits of DO, COD_{Cr}, BOD₅, NH₃-N, TN, TP and fluoride are 0.2, 5, 0.5, 0.025, 0.05, 0.01, and 0.001 mg/L, respectively with recovery rate between 80 to 120%, relative standard deviation (RSD) of < 5%.

2.3. The CCME WQI Model

CCME WQI is established based on the British Columbia Ministry of Environment, Lands and Parks (Zandbergen and Hall, 1998), comprising 3 factors in calculation: scope (f_1), frequency (f_2) and amplitude (f_3) (CCME, 2002; Terrado et al., 2010; Mostafaei, 2014). The formula is described in Equation (1):

$$CCME_{WQI} = 100 - \left(\frac{\sqrt{f_1^2 + f_2^2 + f_3^2}}{1.732} \right) \quad (1)$$

where f_1, f_2, f_3 represent the percentage of variables that do not meet their objectives at least once (failed variables), the percentage of individual tests that do not meet their objectives (failed tests), and the amount by which failed tests do not meet their objectives, respectively, the divisor 1.732 normalizes the result to a range 0 ~ 100 (CCME, 2002; Terrado et al., 2010; Mostafaei, 2014)

CCME WQI describes the water quality index from worst to best water quality (Tunc Dede et al., 2013), which is illustrated in Table 1 (CCME, 2002).

(i) Calculation method of f_1 and f_2 :

$$f_1 = \left(\frac{\text{number of failed variables}}{\text{total number of variables}} \right) \times 100 \quad (2)$$

$$f_2 = \left(\frac{\text{number of failed tests}}{\text{total number of tests}} \right) \times 100 \quad (3)$$

(ii) Calculation method of f_3 .

When the test value is the larger the better, such as DO, the calculation method is following:

$$\text{excursion}_i = \text{objective}_i / \text{failed test value}_i - 1 \quad (4)$$

When the test value is the smaller the better, such as COD_{Cr} , the calculation method is following:

$$\text{excursion}_i = \text{failed test value}_i / \text{objective}_i - 1 \quad (5)$$

The sub-factor “*nse*” is defined by the CCME, referred to as the normalized sum of excursions (Terrado et al., 2010; Mostafaei, 2014):

$$nse = \sum_{i=1}^n \text{excursion}_i / \text{total number of tests} \quad (6)$$

The f_3 is then calculated by the formula:

$$f_3 = nse / (0.01 \times nse + 0.01) \quad (7)$$

2.4. Statistical Analysis

2.4.1. Cluster Analysis

Cluster analysis (CA) helps in grouping objects into classes according to similarities with a class and dissimilarities between different classes, with the results helping in interpreting the data (Vega et al., 1998). The dendrogram resulted from CA provides a visual summary of the clustering processes, which presents a drawing of the groups and their proximity with the low-dimensional data. Euclidean distance is most commonly used in CA due to its simplicity (Shrestha et al., 2008). In this paper, hierarchical agglomerative CA was performed on the normalized data set by between-groups linkage, using Euclidean distances as a measure of similarity. In hierarchical clustering, each object (here each year’s pollution concentration of indices) initially constituted its own cluster. Between-groups linkage was calculated by the with cluster sum of squares (WCSS) for every cluster, as in Equation (8). The two nearest cluster were then combined and this process was continued until all objects belonged to one cluster (Mallye et al., 2014):

$$WCSS = \sum_{j=1}^m (x_j - \bar{x})^2 \quad (8)$$

where WCSS is the squared Euclidean distance between an object in the cluster (x_i) and the mean of that cluster (\bar{x}), m is all objects in that cluster.

2.4.2. Discriminant analysis

Discriminant analysis (DA) is used to discriminate the results of CA whether effective and classify the significance of pollutant indices (Singh et al., 2004):

$$f(G_i) = k_i + \sum_{j=1}^n w_{ij} p_{ij} \quad (9)$$

where i is the number of groups (G), n is the number of indices used to classify a set of data into a given group, w_{ij} is the weight coefficient, p_{ij} is the pollution concentration of indices, and k_i is the constant inherent to each group.

In this study, two groups for temporal evaluations have been selected during winter-spring and summer-autumn seasons. DA was performed on each raw data matrix using standard, enter independents together mode in constructing discriminant functions to evaluate temporal variations in river water quality. The temporal were the grouping variables, whereas all the measured indices constituted the independent variables.

2.4.3. Principal Component Analysis

It is difficult to assess the river water quality with a lot of factors. Hence, reducing the number of monitoring factors instead of original large amounts of data simplifies the analysis. Principal component analysis (PCA) is a multivariate statistical method that can be used for reducing complexity of input variables instead of a large volume of information and it is intended to have a better interpretation of variables (Pires et al., 2008). In this study, PCA was performed using SPSS 23.0 for Windows, which was performed to extract significant principal components and to further reduce the contribution of variables with minor significance. These principal components were subjected to varimax rotation generating varifactors (Singh et al., 2004). Furthermore, feature vectors and standardized variables performed by PCA produced the absolute principal component scores. Then to analyze and calculate the average pollution contribution by multiple linear regression, using pollutant concentration as dependent variable, which should determine the influence degree of each water evaluation index by principal component (Zhou et al., 2007).

3. Results and Discussions

3.1. Characteristics of Water Pollutants in Upper Reaches of Maotiao River

As shown in Table 2, seven monitoring indices were analyzed during winter-spring and summer-autumn seasons from 2003 to 2015. The maximal value of COD_{Cr} was 87 mg/L, which was 2.9 times higher than the Standard IV grade of China’s national environmental quality standard for surface water (GB 3838-2002), meaning that the river was subjected to organic pollutants. Nitrogen pollution was most serious in the Maijia River with 3.655 ~ 4.468 mg/L, below standard V grade (i.e., 2.0 mg/L) of surface water. Furthermore, 77.9% samples of TN and 32.7% samples of $\text{NH}_3\text{-N}$ were inferior to water quality standard V grade of surface water. The maximal values of TN

Table 1. Category of CCME WQI

Category	CCME WQI value	Characteristic
Poor	0 ~ 44.9	The water quality condition has been threatened and destroyed continuously, usually departing from natural or desirable levels.
Marginal	45.0 ~ 64.9	The water quality condition has been threatened and destroyed frequently, often departing from natural or desirable levels.
Fair	65.0 ~ 79.9	The water quality condition has been threatened and destroyed occasionally, sometimes departing from natural or desirable levels.
Good	80.0 ~ 94.9	The water quality condition is protected with only a minor degree of threat and destruction, rarely departing from natural or desirable levels.
Excellent	95.0 ~ 100	The water quality condition is protected with a virtual absence of threat or destruction, very close to natural or desirable levels.

Table 2. Statistical Water Quality Data Monitored during Winter-Spring and Summer-Autumn Seasons at Cross-Sections in Maijia River, Upper Reaches of Maotiao River (2003 ~ 2015)

Evaluation index	Total number	Winter-spring season					Summer-autumn season					Standard level IV
		Min	Max	mean	SD	CV(%)	Min	Max	mean	SD	CV(%)	
DO (mg/L)	104	2.4	8.7	6.2	1.5	23.9	3.3	8.3	6.2	1.3	21.1	3
COD _{Cr} (mg/L)	104	5	87	24.2	15.0	62.0	5	35	17.3	8.1	46.8	30
BOD ₅ (mg/L)	104	1.0	14.0	4.6	3.2	68.9	1.0	9.5	3.5	2.1	61.1	6
NH ₃ -N (mg/L)	104	0.087	10.000	2.475	2.372	95.8	0.062	4.220	1.169	1.256	107.4	1.5
TP (mg/L)	104	0.07	1.90	0.23	0.33	143.1	0.01	8.00	0.37	1.28	347.3	0.3
TN (mg/L)	104	0.83	11.00	4.47	2.47	55.3	1.06	14.00	3.66	2.33	63.8	1.5
Fluoride (mg/L)	104	0.157	5.220	1.379	1.212	87.9	0.075	9.300	1.629	1.795	110.2	1.5

and NH₃-N were 9.3 and 6.7 times higher than the standard IV grade (2.0 mg/L) of surface water, respectively.

Except TP and fluoride, the average values of monitoring indices during winter-spring season were higher than summer-autumn season. Since the Maijia River is originated from the precipitation, pollutants in the river were diluted due to rainfall during summer-autumn season. However, the average values of TP and fluoride during summer-autumn season (i.e., 0.368 and 1.692 mg/L, respectively) were higher than winter-spring season. The reason might be the source of the excessive use of water for household purposes during summer-autumn season and then the discharge of used water directly into water bodies. Additionally, with higher water temperature during summer-autumn season (Jensen and Andersen, 1992), phosphorus released from sediments from Maijia River was also an important cause of pollution. As shown in Table 2, NH₃-N, TP, TN and fluoride did not reach the standard IV grade of surface water.

3.2. Comprehensive Evaluation of Water Quality of CCME WQI

Water quality of CCME WQI was evaluated (Table 3), which showed that the range of CCME WQI was from 35.8 to 86.0, categorized as poor, marginal, fair, and good. During the winter-spring season, with the exception of S1, each of CCME WQI was poor, while water quality of all sites (S1, S2, S3, and S4) had undergone a drastic change from fair to poor in summer-autumn season. In fact, during the winter-spring season the CCME WQI was lower than the summer-autumn, which meant that the water quality was different between winter-spring and summer-autumn seasons, showing that water quality was worse

in winter-spring season.

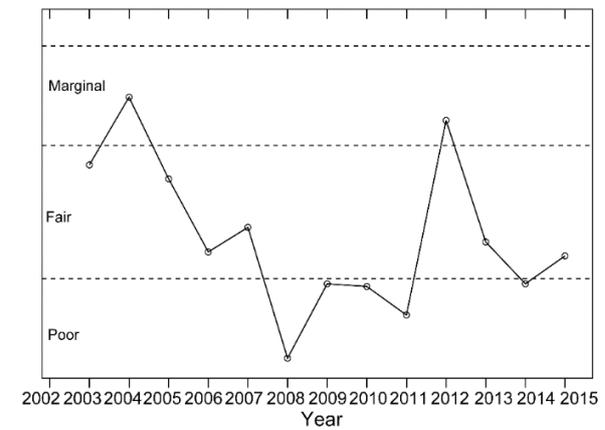


Figure 2. Annual water quality index (CCME WQI) of the Maijia River.

For the spatial variation, it showed the water quality was different from 2003 to 2015 (Figure 2). The X-axis shows the years, while the Y-axis is divided into the four different index categories, ranging from poor to marginal. Even though from 2003 to 2004 water quality mainly ranged from fair to marginal, it returned again marginal from 2005 to 2007, while it was poor from 2008 to 2011. It was remarkable that the highest and lowest CCME WQI values occurred in 2004 and 2008, respectively. The poor category was the one assigned to most years during 2003 ~ 2011. Marginal water quality was appeared in 2012, which became fair from 2013 to 2015, especially in 2014. Since

Table 3. CCME WQI and Classification of Pollution Sites

Site	Coordinate	Time	f_1	f_2	f_3	CCME WQI	Category
S1	106°40'48" E, 26°43'48" N	Winter-spring	71.4	17.6	14.3	56.7	Marginal
		Summer-autumn	42.9	16.5	21.5	72.4	Fair
		Annual	71.4	34.1	24.3	86.0	Good
S2	106°40'12" E, 26°44'24" N	Winter-spring	85.7	42.9	44.3	39.0	Poor
		Summer-autumn	71.4	27.5	21.5	54.1	Marginal
		Annual	85.7	70.3	54.3	68.7	Fair
S3	106°37'48" E, 26°43'12" N	Winter-spring	100	33.0	21.8	37.9	Poor
		Summer-autumn	57.1	24.2	17.2	62.8	Marginal
		Annual	100	57.1	32.7	81.1	Good
S4	106°35'59" E, 26°42'36" N	Winter-spring	85.7	52.7	47.4	35.8	Poor
		Summer-autumn	85.7	38.5	33.8	42.4	Poor
		Annual	85.7	91.3	58.6	66.2	Fair

burgeoning urbanization was proposed in the documents of the 16th National Congress of the Communist Party of China in 2003, the tertiary industry had achieved great development (Dai, 2003), which increased the emissions of pollutants to surface water. During 2003 ~ 2014, the urbanization rate of Guiyang had increased by nearly 15 to 72.1% (Figure 3). Likewise the amount of waste generation increased from 5 million tons to 20 million tons over time (Fang et al., 2017), which probably increased emissions of pollutants to surface water.

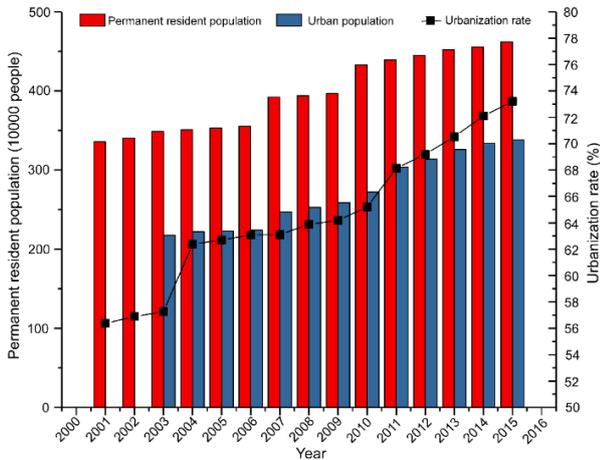


Figure 3. Population change in Guiyang from 2001 to 2015. Data in this figure were collected from Guiyang Municipal Bureau of Statistics (Guiyang Statistical Yearbook Committee, 2016).

In general, the water quality of the Maijia River was influenced by human activities from 2003 to 2015, such as urbanization, Gross National Product (GNP)/capita and the use of chemical fertilizers (Figure 4). WQI decreased from 2004 to 2008, which were successfully simulated with line equation by the three factors, respectively. It indicated that the three factors increased during 2004 ~ 2008, which caused water quality to deteriorate severely. Fortunately, Guiyang was selected as a national Low-Carbon Pilot City in 2011 and an Ecological Civilization Pilot City in 2014. Moreover, several projects towards wa-

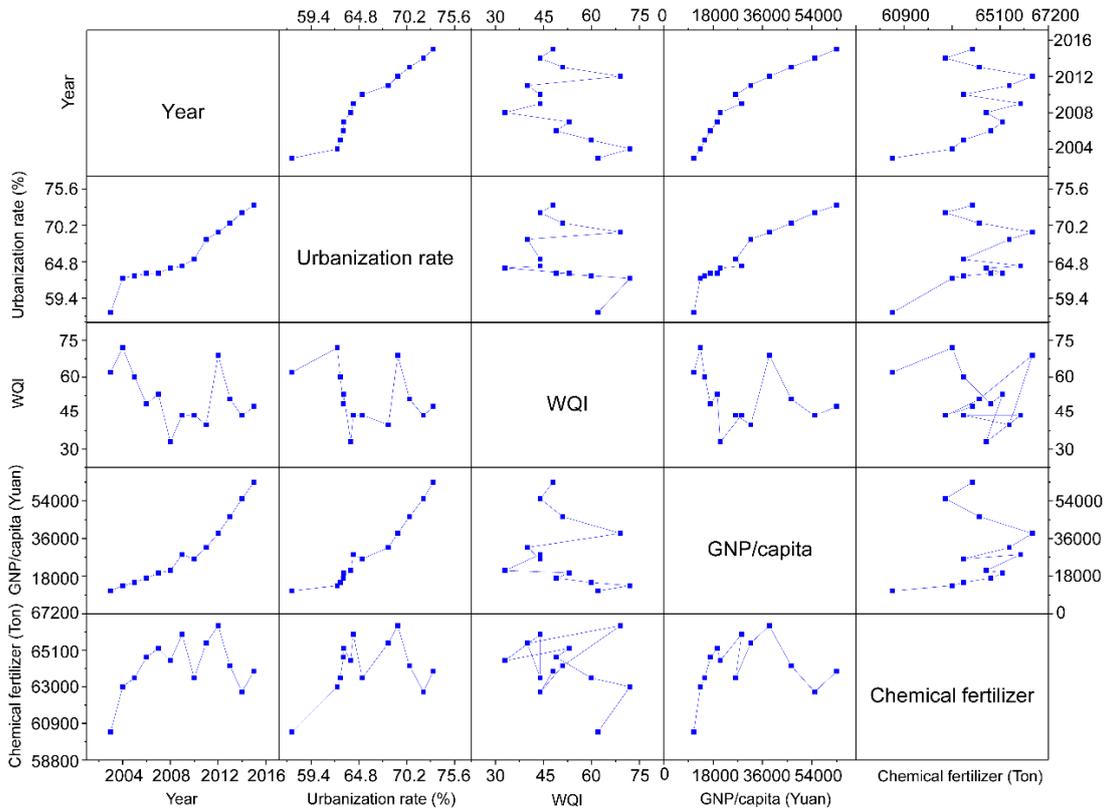
ter governance and green belt rebuilt were launched in Guiyang after 2008, such as the “river chief mechanism”, which were meaningful for the protection and management of water resources. These measures gradually improved the water quality in the river after 2009. However, factors influencing water quality still existed and were more complex, which were not explained very well by the regression equation after the year.

3.3. Characteristics of Spatio-Temporal Distribution of Pollutants

3.3.1. Similarity and Difference of Temporal Characteristics of the Pollutants

Temporal cluster analysis and discriminate analysis of pollutants showed that during winter-spring and summer-autumn seasons the characteristics of pollutants were divided into two periods i.e., period I, with low concentration of pollutants, and period II, with high concentration of pollutants (Figures 5 and 6). The results of cross validation showed that the accuracy of temporal cluster analysis reached to 92.3% during winter-spring and 84.6% in summer-autumn season (Table 4). However, there were differences during the winter-spring and summer-autumn seasons. The degree of pollution of the water in winter-spring season of 2008 was higher (Figure 5(a)), while the degree of pollution of the water in summer-autumn season of 2008, 2011, 2013, 2014 and 2015 was higher (Figure 5(b)). Temporal discriminate analysis of indices i.e., DO, COD_{Cr}, BOD₅, NH₃-N, TP, TN and Fluoride, reflected the difference laws in the temporal diversity of the water quality of the Maijia River (Table 5). As shown in the temporal variation of pollutants in the Maijia River (Figure 6(a)), the concentration of COD_{Cr} declined dramatically from 58 mg/L in 2008 (period I) to 18 mg/L in 2015 (period II) during the winter-spring season. On the other hand, the concentration of COD_{Cr} and TN increased from 25 to 27 mg/L and 3.198 to 7.975 mg/L during 2008 (period I) to 2015 (period II) in the summer-autumn season (Figure 6(b)). In general, the degree of water pollution was mostly severe in 2008 and slightly improved from 2012 to 2015.

In addition, since the planning of ecological restoration, functional optimization and intensively land use through conceptual top-level design was proposed by local government



- (1) Regression equation between WQI (Y_1) and urbanization rate (X_1) (from 2004 to 2008):
 $Y_1 = -2513 \cdot X_1 + 1637.2, R = 0.9871$
 (2) Regression equation between WQI (Y_2) and GNP/capita (X_2) (from 2004 to 2008):
 $Y_2 = -0.0043 \cdot X_2 + 126.29, R = 0.8968$
 (3) Regression equation between WQI (Y_3) and chemical fertilizer (X_3) (from 2004 to 2008):
 $Y_1 = -0.0109 \cdot X_3 + 753.72, R = 0.6867$

Figure 4. The relationships among WQI and urbanization rate, GNP/capita and chemical fertilizer and regression equations. Data in this figure were collected from Guiyang Municipal Bureau of Statistics (Guiyang Statistical Yearbook Committee, 2016).

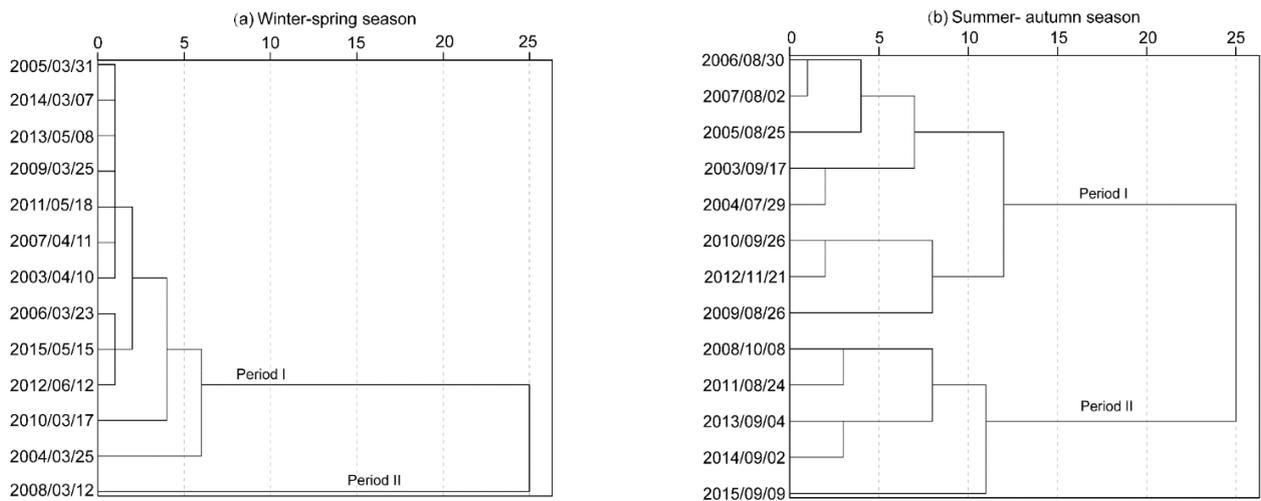


Figure 5. Temporal cluster analysis of pollutants during (a) winter-spring and (b) summer-autumn seasons in the Majjia River.

(Yang and Yang, 2016), lots of measures had been taken to protect water quality such as reinforcement of domestic wastewater control and improvement of water resource management level. Taken altogether, the concentration of pollutants during summer-autumn season was lower, because the amount of rainfall was heavier, which diluted the concentration of pollutants (Tang and Liu, 2008) and improved the water quality to some extent.

Table 4. Temporal Discriminate Analysis of Pollutants in the Maijia River

Time	Group	Number of sample		
		Accuracy (%)	First group	Second group
Winter-spring	First group	100	12	0
	Second group	0	1	0
	Subtotals	92.3	13	0
Summer-autumn	First group	75	6	2
	Second group	100	0	5
	Subtotals	84.6	6	7

Table 5. Classification Functions for Discriminant Analysis of Pollutants in the Maijia River

Index	Winter-spring season		Summer-autumn season	
	First group	Second group	First group	Second group
DO	21.286	31.911	31.225	23.472
COD _{Cr}	3.404	6.282	-4.076	0.370
BOD ₅	0.171	-1.752	-9.913	-6.484
NH ₃ -N	-8.336	-15.533	-7.094	4.666
TP	2.933	18.653	612.321	394.456
TN	1.974	1.485	5.199	-0.843
Fluoride	15.320	18.867	6.328	2.557
Constant	-109.541	-265.158	-96.505	-100.882

3.3.2. Similarities and Differences of Spatial Characteristics of the Pollutants

As shown in Figure 7, the concentration of pollutants at S1 site was lower than other sites (S2, S3, and S4), because the headstream of the Maijia River (S1) was influenced slightly by human activities in every season. Along the river, the mass concentration of COD_{Cr}, BOD₅, TP and TN had roughly trended up during winter-spring season, while the mass concentration of COD_{Cr}, NH₃-N, TP and TN had roughly trended up during summer-autumn season. The main reason was that human activities become more frequent in the middle and lower reaches of the river, such as discharging wastewater directly into the river as well as agricultural pollution. The mass concentration of COD_{Cr} was 87 mg/L in the site of S2 during the winter-spring season, with the highest concentration among the four sites (Figure 7(a)), because population in the areas close to the site of S2 was increased during 2008 ~ 2011. It was likely that new development or agricultural activities intensified as a result of population increase. Figure 7(a) described the upward trends for NH₃-N and TN during 2008 ~ 2011, with similar trends for TN of S2 showed in Figure 7(b). The downward trends for other water quality parameters of S2 during 2011 ~ 2015 can be seen in Figure 7(a), while Figure 7(b) exhibited opposite trends.

Generally, the mass concentration of pollutants at the site of S3 was lower than S2 and S4 during winter-spring season or summer-autumn season (Figure 7). Since there were not agricultural activities with unused land and forested areas around S3, so the spatial heterogeneity in land use influenced the spatial variability of pollutants concentrations (Ahearn et al., 2005). These results suggested that local land cover and vegetation extent were the primary driving forces behind the variations in pollutant concentrations (Tang and Liu, 2008). To protect surface water resources, it has the vital significance to establish a national eco-civilization zone in Guizhou Province, having more than 50% of the province's land areas covered with forests by 2020 (Chuai et al., 2013).

For the site S4, the lower reaches of the Maijia River, the pollutant concentration were highest especially during winter-spring season from 2008 to 2011, which declined from 2012 to 2015 because the effect of point-source pollution control and other local water quality management practices influenced in this regard. However, variations in pollutants concentration had increased quite significantly in four years (2012 ~ 2015) during the summer-autumn season. A lot of humus and other agricultural pollutants were mixed with the Maijia River with heavy rainfall during the summer-autumn season, leading to widespread organic pollution of water (Tang and Liu, 2008; Lin et al., 2011).

3.4. Source Apportionment of Pollution

The original data were standardized and then applied the method of factor analysis to discriminate the main pollution factors and source. Referring to absolute loading values of greater than 0.75, the factor loadings included reasonably most information of original variables (Liu et al., 2003). Moreover, the rotation of the factor axis was executed to yield a 'sample structure' in order to facilitate interpretation in terms of original variables.

Firstly, the methods of Kaiser-Meyer-Olkin (KMO) and Bartlett's test of sphericity were used to test correlation matrix for the original data during winter-spring and summer-autumn season, and then four principal components were extracted. The values of KMO were calculated as 0.769 and 0.700, after the exploratory factor analysis, and the Chi-square of Bartlett spherical test were 140.560, 96.096 having the significance level coefficient of $p < 0.05$, which meant that the method of factor analysis was reasonable.

Table 6 presented the eigenvalues, contribution rates and cumulative contribution rates of principal components for the factor analysis of original data during the winter-spring and summer-autumn seasons. It revealed that the four components explained approximately 86.841 and 83.126% of total variance during winter-spring and summer-autumn seasons, respectively. Table 6 also showed the loading of vaimax rotated factor matrix for four-component model. Evidently, the first component was generally more correlated with the variables than the other components. It was expected because these components were extracted successively, each one accounting for as much of the remaining variance as possible.

During the winter-spring season, the meanings represent-

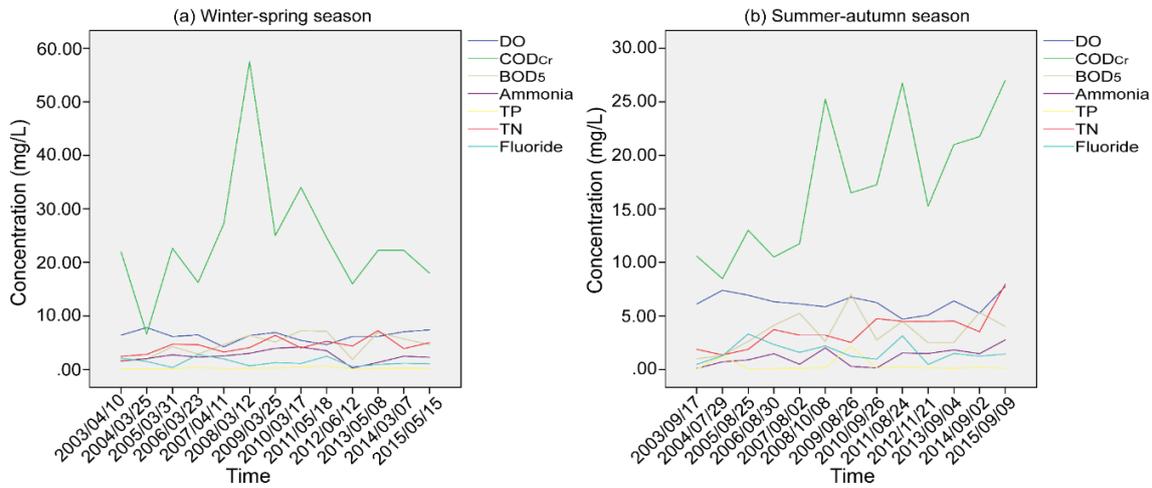


Figure 6. Temporal variation of pollutants during (a) winter-spring and (b) summer-autumn seasons in the Maijia River.

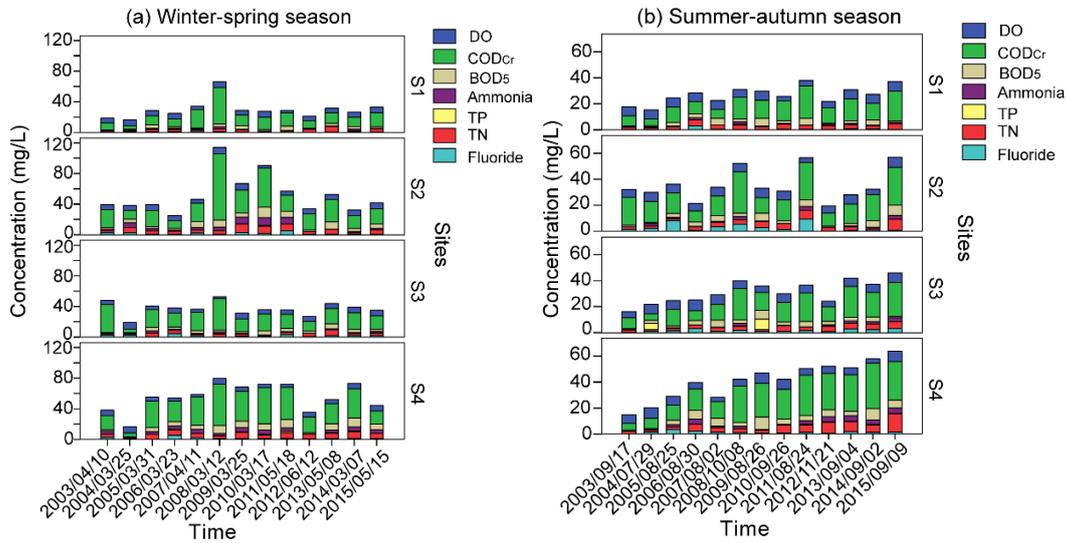


Figure 7. Spatial variation of pollutants during (a) winter-spring and (b) summer-autumn seasons in the Maijia River.

ed by the main factors were explained as follows: PC1, which explained 33.9% of the total variance, had strong positive loading on NH₃-N and TN, and moderate loadings on BOD₅ and TP. During the winter-spring season, there was less rainfall, which was a significant factor in the non-point source pollution yield (Yang et al., 2011), less non-point source pollution and mass point source pollution in dry season. NH₃-N was mainly from the relatively constant industrial and municipal point source pollution loads (Wu and Chen, 2013). So NH₃-N, TN, BOD₅, and TP grew out of domestic, and food processing wastewater. In addition, NH₃-N included in PC1 was total-quantity control of pollutant, so the PC1 was considered as a potential factor, which grows significantly with economic development.

PC2, which explained 18.6% of the total variance (Table 6), had strong positive loading on COD_{Cr}, and moderate loadings on BOD₅. The indices in connection with biochemical fac-

tors reflected the basic water quality of river. Discharges of domestic and industrial wastewater resulted in serious deterioration of water quality (Mojahedi and Attari, 2009; Wu and Chen, 2013).

PC3, which explained 18.6% of the total variance (Table 6), had strong positive loading on DO, and moderate loadings on TP. It possibly revealed that domestic wastewater was the main pollution source. Besides, there was significant negative correlation between DO concentration and TP, which meant the phosphate concentration decreased at increasing DO. This phenomenon was consistent with the common biophosphorus removal processes in sequential anaerobic and aerobic condition, where the phenomenon was attributed to the activity of phosphorus-accumulating organisms (PAOs) (Mahendraker et al., 2005).

PC4 accounted for 15.8% of the total variance (Table 6), had strong positive loading on fluoride. As shown in Figure 1,

Table 6. Loading Matrix of Rotated Factors

Time	Monitored item	PC1	PC2	PC3	PC4
Winter-spring season	DO	-0.024	-0.202	-0.918	-0.100
	COD _{Cr}	0.260	0.916	0.164	-0.030
	BOD ₅	0.721	0.513	0.188	-0.048
	NH ₃ -N	0.746	0.367	0.088	0.365
	TP	0.644	-0.009	0.618	-0.029
	TN	0.889	0.149	0.013	0.165
	Fluoride	0.153	-0.040	0.073	0.964
Contribution (%)		33.901	18.585	18.575	15.780
Cumulative contribution (%)		33.901	52.486	71.061	86.841
Summer-autumn season	DO	-0.093	-0.068	0.986	-0.014
	COD _{Cr}	0.826	-0.001	-0.190	0.160
	BOD ₅	0.579	0.560	-0.078	-0.095
	NH ₃ -N	0.851	-0.077	-0.110	0.209
	TP	-0.184	0.898	-0.045	0.023
	TN	0.896	-0.080	0.106	-0.020
	Fluoride	0.150	-0.008	-0.014	0.976
Contribution (%)		37.267	16.249	14.861	14.749
Cumulative contribution (%)		37.267	53.516	68.377	83.126

Table 7. Comprehensive Evaluation for the Water Pollution of the Maijia River

Time	Site	PC1 score	PC2 score	PC3 score	PC4 score	Comprehensive evaluation score	Pollution rank
Winter-spring season	S4	0.701	0.200	0.367	-0.116	0.325	1
	S2	0.491	0.283	-0.420	0.220	0.175	2
	S3	-0.674	-0.054	0.116	0.451	-0.145	3
	S1	-0.518	-0.429	-0.062	-0.555	-0.355	4
Summer-autumn season	S4	0.818	0.064	-0.093	-0.247	0.265	1
	S2	-0.059	-0.163	-0.004	0.673	0.051	2
	S3	-0.355	0.401	0.110	0.111	-0.034	3
	S1	-0.404	-0.302	-0.013	-0.537	-0.282	4

Guizhou aluminum factory is 4 km from the river, with the product of alumina and aluminum being 1.2 and 0.43 Mt/year, respectively. It indicated that the factory in the area was likely the main pollution source of industrial fluorine. On the other hand, fluoride derives mainly from the lithological sources (Hem, 1985). Besides, the rocks absorbed fluoride in the soil are also the source (Ayooob and Gupta, 2006). Since the special geological characteristics of karst in Guizhou Province, fluoride is not surprised to be found in the Maijia River.

Like winter-spring season, PC4 accounted for 14.7% of the total variance during summer-autumn season. PC1, which explained 37.3% of the total variance, had strong positive loading on COD_{Cr}, NH₃-N, TN and moderate loadings on BOD₅, and compared with winter-spring season the contribution of COD_{Cr} was upward. These are the dominant indices for water quality assessment of river which indicate the pollution status of organics and nutrients, closely relating with acceptance of daily sewerage discharge and non-point pollution. PC2 accounted for 16.2% of the total variance, had strong positive loading on TP, and moderate loadings on BOD₅. That possibly revealed that the non-point pollution was substantial during summer-autumn season. PC3 explained 14.9% of the total variance with

strong positive loading on DO, which revealed that the discharge of daily sewerage and non-point pollution reduced the concentration of DO (Chang, 2008). Generally, the principal influent factors were not identical, but the four principal components suggested the impact of human activities on water during the different time.

According to the coefficient matrix of factor scores and the monitoring data of all indices, calculating equations were established as follow:

During the winter-spring season:

$$F_1 = 0.278x_1 - 0.279x_2 + 0.262x_3 + 0.257x_4 + 0.376x_5 + 0.538x_6 - 0.145x_7 \tag{10}$$

$$F_2 = -0.278x_1 + 0.939x_2 + 0.221x_3 + 0.116x_4 + 0.445x_5 - 0.233x_6 + 0.014x_7 \tag{11}$$

$$F_3 = -0.814x_1 - 0.061x_2 - 0.049x_3 - 0.137x_4 + 0.463x_5 - 0.186x_6 + 0.008x_7 \tag{12}$$

$$F_4 = -0.091x_1 + 0.035x_2 - 0.174x_3 + 0.224x_4 - 0.233x_5 - 0.052x_6 + 0.937x_7 \tag{13}$$

During the summer-autumn season:

$$F_1 = 0.065x_1 + 0.300x_2 + 0.245x_3 + 0.312x_4 - 0.087x_5 + 0.387x_6 - 0.081x_7 \quad (14)$$

$$F_2 = 0.071x_1 - 0.019x_2 + 0.486x_3 - 0.074x_4 + 0.804x_5 - 0.061x_6 + 0.047x_7 \quad (15)$$

$$F_3 = 0.979x_1 - 0.100x_2 + 0.052x_3 - 0.024x_4 + 0.057x_5 + 0.189x_6 + 0.043x_7 \quad (16)$$

$$F_4 = 0.039x_1 + 0.036x_2 - 0.153x_3 + 0.082x_4 + 0.101x_5 - 0.151x_6 + 0.981x_7 \quad (17)$$

The normalized monitoring data during winter-spring and winter-spring seasons were put into Equations (10) ~ (13), (14) ~ (17), respectively in order to obtain the score of four components. The comprehensive evaluation score of the sites explained the degree of water pollution, which were calculated by combining the score of four components with corresponding contribution rate as follow:

$$F = 33.901\%F_1 + 18.585\%F_2 + 18.575\%F_3 + 15.780\%F_4 \quad (18)$$

$$F = 37.267\%F_1 + 16.249\%F_2 + 14.861\%F_3 + 14.749\%F_4 \quad (19)$$

According to the comprehensive evaluation score pollution rank of sites were obtained (Table 7), the higher comprehensive evaluation scores the more serious pollution. During both winter-spring and summer-autumn seasons, the pollution rank was $S4 > S2 > S3 > S1$. The result was in accordance with the spatial characteristics of the pollutants, i.e., the mass concentration of pollutants of the site of S3 was lower than S2 and S4. Hydrological process related to water flow-paths exert significant influence on runoff and contaminant transport (Jones et al., 2001; Molenat et al., 2008); further research is needed for more detailed information about hydrological process.

4. Conclusions

A holistic picture of the water quality of the Maijia River was conducted by investigating seven water quality parameters through various analysis methods. The main conclusions were as follows:

(1) Owing to diffusion influences, the degree of environmental pollution sources such as $\text{NH}_3\text{-N}$, TP and fluoride were very unevenly distributed in the Maijia River. Because of frequent human activities in the middle and lower reaches of the river, the mass concentration of COD_{Cr} , BOD_5 , TP, TN had roughly trended up during winter-spring season, while COD_{Cr} , $\text{NH}_3\text{-N}$, TP, and TN had roughly trended up respectively during summer-autumn season.

(2) Seven water quality parameters were identified by PCA, showing that contribution of the first principal component accounted for more than one third of the total variances which played a dominant role in the pollution of the river. It had strong positive loading on $\text{NH}_3\text{-N}$ and TN during the winter-spring

season as well as COD_{Cr} , $\text{NH}_3\text{-N}$ and TN during the winter-spring season, suggesting that $\text{NH}_3\text{-N}$ and TN were main pollutants throughout the year. Moreover, the degree of pollution at monitoring sites by comprehensive evaluation score was $S4 > S2 > S3 > S1$. The results of CCME WQI, which were applied to the Maijia River showed that the water quality was poor from 2008 to 2011 and acceptable from 2003 to 2007 and 2012 to 2015, respectively but slightly improved from 2012 to 2015, suggesting that the measures to protect in-stream quality has begun to work. In general, organic matter and nitrogen are still the main pollutants in the Maijia River. The government should take specific measures to reduce the pollutants.

(3) The quality of the Maijia River was influenced strongly by human activities and non-point pollution sources, such as urbanization, Gross National Product (GNP)/capita and the use of chemical fertilizers, which were explained very well by linear regression equations before 2009. Although local government took some measurements to protect the water quality in recent years, the factors influencing water quality still existed and were more complex. It is necessary to consider how to utilize systematical approach to control pollutants and meanwhile, keep economic growth. Additionally, based on its peculiar geological environment and shallow depth of groundwater of karst regions, in order to manage water quality in the Maijia River and in future work the scalar complexity of biogeochemical mechanisms will have to be focused.

This study will provide indicators in developing optimal strategies and determining priorities for upper reaches of Maotiao River pollution control and effective water resources management.

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