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Temporal and spatial characteristics of the water pollutant concentration in Huaihe River Basin from 2003 to 2012, China

Ming Dou · Yan Zhang · Guiqiu Li

Abstract Based on the monitoring data of 78 monitoring stations from 2003 to 2012, five key water quality indexes (biochemical oxygen demand: BOD_5, permanganate index: COD_{Mn}, dissolved oxygen: DO, ammonium nitrogen: NH_3-N, and total phosphorus: TP) were selected to analyze their temporal and spatial characteristics in the highly disturbed Huaihe River Basin via Mann-Kendall trend analysis and boxplot analysis. The temporal and spatial variations of water pollutant concentrations in the Huaihe River Basin were investigated and analyzed to provide a scientific basis for water pollution control, water environment protection, and ecological restoration. The results indicated that the Yinghe River, Quanhe River, Honghe River, Guohe River, and Baohe River were the most seriously polluted rivers, followed by Hongze Lake, Luoma Lake, Yishuhe River, and Nansi Lake. BOD_5, COD_{Mn}, and NH_3-N were the major pollution indexes, for which the monitoring stations reported that more than 40 % of the water quality concentrations exceeded the class IV level. There were 21, 50, 36, and 21 monitoring stations that recorded significantly decreasing trends for BOD_5, COD_{Mn}, NH_3-N, and TP, respectively, and 39 monitoring stations showed a significantly increasing trend for DO. Moreover, the water quality concentrations had a certain concentricity and volatility according to boxplot analysis for the 20 monitoring stations. The majority of monitoring stations recorded a large fluctuation for the monitoring indexes in 2003 and 2004, which indicated that the water quality concentrations were unstable. According to the seasonal variations of the water quality concentrations in the mainstream of Huaihe River, the monthly variation trends of the BOD_5, COD_{Mn}, DO, NH_3-N, and TP concentrations were basically consistent among the seven monitoring stations. The BOD_5, COD_{Mn}, NH_3-N, and TP concentrations were affected by the change of the stream discharge; changes in DO and NH_3-N concentrations were influenced by the regional environmental temperature, and the DO and NH_3-N concentrations decreased when the water temperature increased.

Keywords Temporal and spatial characteristic · Mann-Kendall trend analysis · Boxplot analysis · Water pollutant concentration · Huaihe River Basin

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Introduction

The Huaihe River Basin is one of the main water systems in China, and it is an agriculturally developed and densely populated district. In recent years, the deteriorating water quality seriously damaged the ecological environment, and large-scale water pollution accidents caused a serious impact on people's productivity and

lives because of the population growth and rapid development of industrial and agricultural production.

Huaihe River is the first comprehensive-governance river in China. In order to solve the problems of flood and water shortage, there had been approximately 11,000 water storage reservoirs and control sluices built in the Huaihe River Basin by the end of the year 2000, which caused substantial changes in the natural runoff process, and the Huaihe River Basin has the characteristics of a wide area, large geographic and geomorphic diversity, and different production and living standards; thus, different external conditions result in a large disparity in the water quality and water quantity of the rivers. There is a strong influence on the river's ecological environment by the large quantity of untreated industrial and domestic sewage discharged directly into the river, and the pollutant discharge totals are far more than the water environment capacity (Jiang et al. 2011).

Some relevant research results indicated that the water quality has exhibited a gradually worsening trend in the Huaihe River Basin since the 1980s (Tan et al. 2005; Ge et al. 2000; Cheng et al. 2001). Owing to the impacts of human activities and water pollution, there are large differences among different regions in terms of the natural hydrochemical characteristics and the water pollution has changed to ammonia nitrogen pollution (Cheng et al. 2005; Wang 2004; Liu 2009; Mao et al. 2003). Temporal and spatial research on the water environment has successfully characterized water quality by the change of ion concentration (Li et al. 2004; Zhu 2004; Fan and Li 2004). There are some researches about Hongze Lake, the results showed that water quality in the Hongze Lake has significant volatility and temporal-spatial difference (Li and Pu 2003), and the water physicochemical parameters had significant seasonal variations except water depth and secchi depth, TP and COD_{Mn} were the highest in winter, while nitrogen content was the highest in spring (Li et al. 2013). The temporal-spatial characteristic of Lancangjiang River water quality was investigated along the middle reaches through river section monitoring, and sample analyses were made with several representative indexes, including metals such as Cu, Zn, Pb, and Cd—for example, NH_3-N and PO_4-P—over more than 10 years (Yao et al. 2005). By monitoring the main contamination parameters of NH_3-N, BOD_5, COD_{Cr}, TP, arsenic, and fluoride in the Guohe River stem,

the results showed that the mainstream of the Guohe River featured the prominent temporal-spatial variety characteristic of water contamination (Fu et al. 2007). As compared with historical data from the past 10 years, the results indicated that the water quality of rivers around Taihu Lake may be showing changes for better tendency, while the inflow river pollution was still severe (Lu et al. 2011). Twenty-one sub-watersheds in the Jiulong River watershed were chosen for in situ water quality monitoring of the baseflow during flood season, dry season, and average season in 2010; the results show that the water quality in dry season is the worst, whereas the water quality in flood season is comparatively good (Huang et al. 2012).

There have been a series of research on spatial-temporal characteristics of water pollutant concentration via different statistical analysis techniques. In recent years, multivariate analyses have been widely applied to evaluate the temporal and spatial characteristics of water quality. Several studies (Singh et al. 2005; Ozbay et al. 2009; Kamble and Vijay 2011; Yab et al. 2011) have classified monitoring stations based on selected water quality parameters to determine and identify the stations most affected by pollution in a water system. Cun and Vilagines (1997) revealed the varying pattern of DO, chloride, and nitrogen in the Seine River over the past 90 years. Wang et al. (2010) analyzed the space and seasonal changes of water pollutants of the Grand Canal, and Varol et al. (2012) evaluated the spatial-temporal variations of water quality datasets for Kralkızı, Dicle, and Batman dam reservoirs in the Tigris River basin via statistical analysis methods. The spatial and temporal patterns of water quality in Kuwait Bay were investigated using data from six stations between 2009 and 2011, and the results showed that most water quality parameters such as phosphorus, nitrate, dissolved oxygen, and total suspended solids fluctuated over time and space (Al-Mutairi et al. 2014). Three key water quality elements at 18 monitoring stations were selected to analyze their spatial-temporal variations in the highly disturbed the Huaihe River Basin using a seasonal Mann-Kendall test and Moran's I method (Zhai et al. 2014). Hayakawa et al. (2015) evaluated the spatial and temporal variations in dissolved inorganic nitrogen and soluble reactive phosphorus concentrations and the processes that control them in water and

riverbed sediments at the mouths of five main rivers that enter Lake Hachiro, a shallow eutrophic lake in Japan, and used the Kruskal-Wallis test to identify significant differences among rivers.

Overall, there are many studies on the spatial-temporal variations of water quality for drainage basins, but there is a lack of comprehensive analysis of the spatial-temporal variations of the water pollutant concentration in the Huaihe River Basin. To meet the economic and social water demand, the channel flow was controlled by the reservoirs and sluices, and water resource utilization has exceeded its carrying capacity, which has caused water environment problems in the Huaihe River Basin. Thus, the key and foundation of water environmental management and ecological restoration for the Huaihe River are to understand the current situation of the water ecological environment and the variation trend. In this study, to analyze the characteristics of water pollutant concentration in the Huaihe River Basin from 2003 to 2012, we used the Huaihe River Basin as a research object and adopted water quality monitoring data of 78 monitoring sites. The spatial-temporal characteristics, variation trend, and seasonal variability were analyzed via the methods of statistical analysis, and the results could provide guidance for pollution prevention and sluice operation in the Huaihe River Basin.

Materials and methods

Study area

The Huaihe River Basin (111° 55′ E–121° 25′ E, 30° 55′~36° 36′ N; Fig. 1) is located in the east of China, between the Yangtze and Yellow Rivers, with a total length of 1000 km for main stream and a drainage area of 270,000 km^2. The area of Huaihe River Basin accounts for 3.5 % of China's territorial area, and the water quantity accounts for 3.4 % of the total amount of water resources in China. In addition, the farmland accounts for 15.2 % of China's cultivated land resources, the population accounts for 16.2 % of the Chinese population, the utilization factor of water resources is more than 60 %, and the degree of rational development and utilization is more than the mean level of the inland rivers, which is 30 % (Jiang et al. 2011). The water environment pollution

began in the 1970s in the Huaihe River Basin; the main sources of pollutants were industrial sewage, domestic sewage, and agricultural nonpoint source pollution, which are closely related to the rapid development of industry and agriculture, the rapid growth of the population, and the acceleration of urbanization. After 2000, the Chinese government invested significant financial and material resources into the prevention and disposal of water pollution in the Huaihe River Basin, and the water quality has been significantly improved. However, water environment problems are still not solved in terms of the large spatial-temporal difference of water pollution load and the sudden pollution accidents, and the water pollution situation is thus still very serious (Wang 2014).

Data description

The five water quality elements in this study were selected because they were the most serious water quality indices in the Huaihe River Basin: biochemical oxygen demand (BOD_5), permanganate index (COD_{Mn}), dissolved oxygen (DO), ammonium nitrogen (NH_3-N), and total phosphorus (TP). All water quality monitoring data were collected from the monitoring center of the Huaihe River Water Resources Protection Bureau. All elements were tested following the national standard of water quality testing (SEPA and GAQSIQ 1989, 2009a, 2009b). Monthly water quality concentrations were collected from 2003 to 2012 at 78 monitoring stations (Table 1) according to the actual monitoring data of monitoring stations. There are 76 monitoring stations for BOD_5; 78 monitoring stations for COD_{Mn}, DO and NH_3-N; and 73 monitoring stations for TP. The 78 monitoring stations are mainly located in the mainstream of the Huaihe River, Honghe River, Shihe River, Yinghe River, Quanhe River, Pihe River, Guohe River, Huihe River, Baohe River, Tuohe River, Hongze Lake, downstream of the Huaihe River, Yishuhe River, Luoma Lake, and Nansi Lake (Fig.1).

Methods

In this study, the data of five water quality indexes (i.e., BOD_5, COD_{Mn}, DO, NH_3-N, and TP) were first analyzed via statistical analysis methods, which
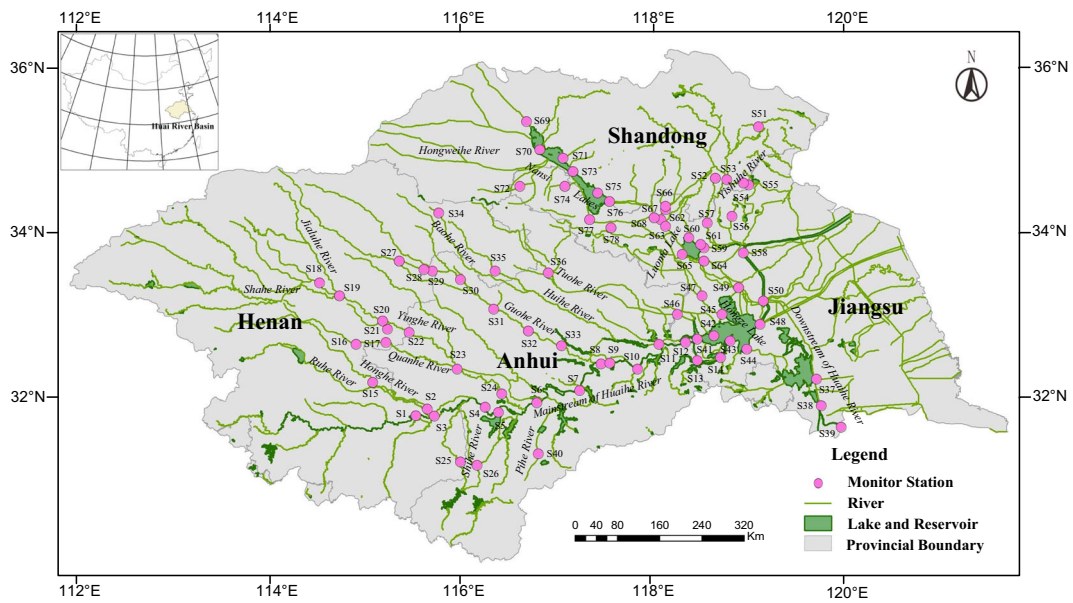
Fig. 1 Location of study area, water quality monitoring stations

could determine the average concentration values of the water quality indexes in each monitoring year and were classified according to the environmental quality standards (GB 3838–2002). Second, the variation trend of the water pollutant concentrations for the monitoring stations was analyzed by Mann-Kendall trend analysis. Third, the temporal variation characteristics of water quality concentrations for 20 monitoring stations were analyzed by the boxplot analysis. Finally, seasonal variability characteristics of five water quality indexes for seven monitoring stations in the mainstream of Huaihe River were explored.

Mann-Kendall trend analysis

The Mann-Kendall trend analysis is a method of nonparametric tests based on rank whose superiority is the ability to test a linear or nonlinear trend (Hisdal et al. 2001; Wu et al. 2008). This extension of the Mann-Kendall test is robust and exact, and has been widely used in water quality trend detection (Larned et al. 2003; Bouza-Deaño et al. 2008). In the Mann-Kendall test, the null hypothesis (H_0) and alternative hypothesis (H_1) are equal in the problem of whether there is a time series trend in the external observation data. The statistic (S) and standardized test statistics (Z_{MK}) are as follows:

$$S = \sum_{i=1}^{n=1} \sum_{j=i+1}^{n} \text{sgn}(X_j - X_i) \tag{1}$$

$$\text{sgn}(X_j - X_i) = \begin{cases} +1 & (X_j - X_i) > 0 \\ 0 & (X_j - X_i) = 0 \\ -1 & (X_j - X_i) < 0 \end{cases} \tag{2}$$

$$Var(S) = \frac{1}{18}\left[n(n-1)(2n=5) - \sum_{p=1}^{q} t_p(t_p-1)(2t_p+5)\right] \tag{3}$$

$$Z_{MK} = \begin{cases} \dfrac{S-1}{\sqrt{Var(S)}} & S > 0 \\ 0 & S = 0 \\ \dfrac{S+1}{\sqrt{Var(S)}} & S < 0 \end{cases} \tag{4}$$

where X_i and Y_i are the corresponding values for the years i and j in the time series, n is the length of the time series data, t_p is the corresponding bound value for the p-number, and Z_{MK} is the variation trend for the time series data, which exhibits an increasing trend if Z_{MK} is greater than zero and a decreasing trend if Z_{MK} is less than zero. When

Table 1 The contrast for number and name of station

Station number	River	Station name	Station number	River	Station name	Station number	River	Station name
S1	Mainstream of Huaihe River	Huaibin	S27	Guohe River	Xuanwu Sluice	S53	Yishuhe River	Daguanzhuang Sluice
S2		Diliyu	S28		Luyi	S54		Daxingzhen
S3		Wangjiaba	S29		Huangzhuang	S55		Shilianghe Reservoir
S4		Runheji	S30		Boxian Sluice	S56		Ahu Reservoir
S5		Xinhekou	S31		Guoyang	S57		Xinan
S6		Lutaizi	S32		Mengcheng Sluice	S58	Luoma Lake	Shuyang Sluice
S7		Huainan	S33		Longkangji	S59		Zhangshan Sluice
S8		Bengbu Sluice	S34	Baohe River	Sunzhuang	S60		Miaowei
S9		Wujiadu	S35	Huihe River	Heshun Sluice	S61		Yanghetan Sluice
S10		Linhuaiguan	S36	Tuohe River	Shangheli	S62		Tanshangji
S11		Wuhe	S37	Downstream of Huaihe River	Gaoyou	S63		Yunhe
S12		Xiaoliuxiang	S38		Shaobo Sluice	S64		Suqian Sluice
S13		Huayuanju	S39		Sanjiangying	S65		Zaohe Sluice
S14		Xuyi	S40	Pihe River	Xiepu	S66		Linzizong
S15	Honghe River	Xincai	S41	Hongze Lake	Xinhetou	S67		Linzixihong
S16	Quanhe River	Xiangcheng	S42		Linhuaitou	S68		Liushan Sluice
S17		Shenqiu Sluice	S43		Laozishan	S69	Nansi Lakes	Houying
S18	Yinghe River	Huangqiao	S44		Jiangba	S70		Nanyangnan
S19		Zhoukou	S45		Xiangchengzhuang	S71		Makou
S20		Huaidian Sluice	S46		Tuanjie Sluice	S72		Zhaozhuang
S21		Huzhai	S47		Jinsuozhen	S73		Erjihu Sluice
S22		Jieshou	S48		Erhe Sluice	S74		Peicheng Sluice
S23		Fuyang Sluice	S49		Siyang Sluice	S75		Daguanzhuang
S24		Yingshang Sluice	S50		Huaiyin Sluice	S76		Xinhetou Sluice
S25	Shihe River	Liji Sluice	S51	Yishuhe River	Doushan Reservoir	S77		Linjiaba Sluice
S26		Sanyuandian	S52		Liuji Sluice	S78		Xietai Sluice

$|Z_{MK}| > Z_{(1 - \alpha/2)}$, the null hypothesis is rejected, and there is a significant trend in the time series data. Values of $Z_{(1 - \alpha/2)}$ can be found by the standard normal distribution table; when the level of $\alpha = 5$ % is significant, the corresponding value is 1.96 (Wei and Deng 2014).

Boxplot analysis

The boxplot analysis assesses temporal and spatial variability in analyte concentrations as well as one-way analysis of variance using a Kruskal-Wallis test for differences among station medians (Whitall et al. 2010). The boxplot is a simple tool for describing statistical analyses; the shape of the data can be directly presented with a simple combination of graphics, which is used to reflect the center position and distribution range of one or more groups of continuous quantitative data (Zhuang 2003).

A boxplot can reflect the data trends of the focus and dispersion and can directly show the shape and

distribution structure and identify the outliers in the water quality data. A boxplot can also conduct spatial-temporal data comparison and analysis, which has been widely used (Shrestha and Kazama 2007; Naddeo et al. 2007). The US Environmental Protection Agency regarded the boxplot as the main statistical method of water quality standards for rivers and lakes; Australia also regarded the boxplot as the main statistical method for the river health evaluation (Zhou et al. 2007; Razmkhah et al. 2010; Bu et al. 2010). The primary parameters of a boxplot are the median, minimum, maximum, and 25th and 75th percentile values. Using this method, the pollution degree of water quality can be compared by analyzing the section of the box for each monitoring station, and the length of the box can be used to analyze the stability of the monitoring data.

Results and discussion

The spatial distribution of water quality Pollution

The data of five water quality indexes (i.e., BOD_5, COD_{Mn}, DO, NH_3-N, and TP) were analyzed via statistical analysis methods, which could determine the average concentration value of the water quality indexes from 2003 to 2012 for each monitoring station and were classified according to Chinese Environmental quality standards for surface water (GB 3838–2002) (China's State Environmental Protection Administration 2002). The specific classification for the five water quality indexes is shown in Table 2 and Fig. 2.

The water quality classification of BOD_5 for 76 monitoring stations is shown in Fig. 2a. BOD_5 at the monitoring stations belonged to classes I, II, III, IV, and V and inferior class V, accounting for 14.47, 25.00, 19.74, 22.37, 10.53, and 7.89 %, respectively, which showed that 40.79 % of the monitoring stations detected poor water quality (i.e., class IV, V, and inferior class V). There were six monitoring stations at which the water pollutant concentrations reached the inferior class V level: Xinan, Liushan Sluice, Doushan Reservoir, Jinsuozhen, Yanghetan Sluice and Houying. The average concentrations of BOD_5 in those six monitoring stations reached 37.62, 31.10, 13.81, 12.86, 11.86, and 10.45 mg/L, respectively. The highest concentration value of BOD_5 exceeded ten times the maximum value of class III (4 mg/L). On the whole, there were some rivers in which the water pollutant concentrations of BOD_5 belonged to class III or below, such as the main-stream of the Huaihe River, Shihe River, Pihe River, downstream of the Huaihe River, Huihe River, and Tuohe River. The water pollutant concentrations of BOD_5 belonged to class IV or above in the Yinghe River, Honghe River, Quanhe River, Guohe River, and Baohe River. Most of the monitoring stations of BOD_5 pollutant concentrations in the Hongze Lake, Luoma Lake, Yishuhe River, and Nansi Lake detected class III or below, but there were several monitoring stations at which the BOD_5 pollutant concentrations belonged to class IV or above.

In Fig. 2b, the water quality classification of COD_{Mn} for the 78 monitoring stations is presented. COD_{Mn} concentrations of the monitoring stations belonged to class II, III, IV, and V and inferior class V, accounting for 24.36, 29.49, 34.62, 6.41, and 5.12 %, respectively. The monitoring stations at which COD_{Mn} pollutant concentrations exceeded the class IV level accounted for 46.15 %. There were four monitoring stations (i.e., Xinan, Liushan Sluice, Xuanwu Sluice, and Jinsuozhen) at which the water pollutant concentrations reached the inferior class V level, where average concentrations of COD_{Mn} reached 37.62, 31.1, 13.81, 12.86, 11.86, and 10.45 mg/L, respectively; these monitoring stations are mainly distributed in the Yishuhe River, Luoma Lake, Hongze Lake and Guohe River. The highest concentration value of COD_{Mn} exceeded ten times the maximum value of class III (6 mg/L). On the whole, there were some rivers (i.e., the mainstream of Huaihe River, Shihe River, Pihe River, downstream of the Huaihe River and Tuohe River) in which the water pollutant concentrations of COD_{Mn} belonged to class III or below. The water pollutant concentrations of COD_{Mn} belonged to class IV or above in the Yinghe River, Honghe River, Quanhe River, Guohe River, Huihe River, and Baohe River. At most of the monitoring stations (i.e., Hongze Lake, Luoma Lake, Yishuhe River, and Nansi Lake), COD_{Mn} pollutant concentrations basically belonged to class III or below, but there were several monitoring stations at which COD_{Mn} pollutant concentrations belonged to class IV or above.

In Fig. 2c, in terms of the water quality classification of DO in the 78 monitoring stations, the class I, II, III, and IV water accounted for 51.28, 41.03, 6.41, and 1.28 of the total, respectively, and those that exceeded the class IV level accounted for only 1.28 %. There was only one monitoring station (i.e., Xinan) at which the water pollutant concentrations reached the inferior class

Table 2 Standard limiting values of surface water environment quality (mg/L)

Water quality index	Class I	Class II	Class III	Class IV	Class V
$BOD_5 \leq$	3	3	4	6	10
$COD_{Mn} \leq$	2	4	6	10	15
$DO \geq$	7.5	6	5	3	2
$NH_3\text{-}N \leq$	0.15	0.5	1	1.5	2
$TP \leq$	0.02	0.1	0.2	0.3	0.4

If the water quality concentration exceeded the maximum value of class V, we considered the water quality level to be the inferior class V

V level, and the average concentration value of DO reached 3.93 mg/L. Overall, the water quality of DO was preferably in the examination area of the Huaihe River Basin and basically belonged to class III or below.
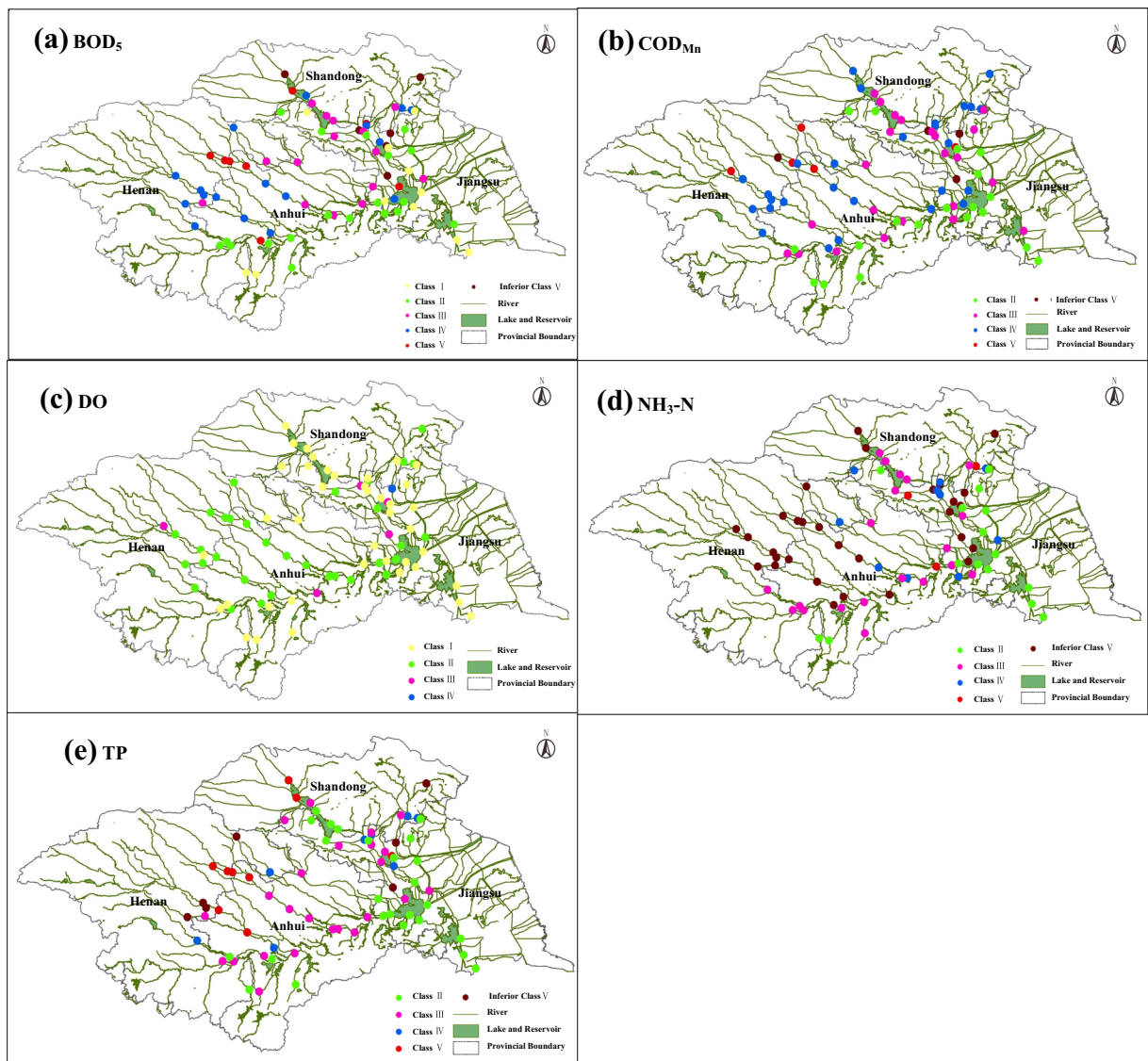
Fig. 2 The classification of the water quality indexes for each monitoring station

The water quality classification of NH_3-N for the 78 monitoring stations is shown in Fig. 2d. NH_3-N concentrations at the monitoring stations belonged to class II, III, IV, V and inferior class V, accounting for 19.23, 25.64, 12.82, 3.85, and 38.46 %, respectively, which showed that the monitoring stations with poor water quality (i.e., class IV, V, and inferior class V) accounted for 55.13 % of the total. There were 30 monitoring stations at which the water pollutant concentrations for NH_3-N reached the inferior class V level, which are mainly distributed in the Yinghe River, Quanhe River, Guohe River and Baohe River, plus some of the monitoring stations in the mainstream of the Huaihe River, Hongze Lake, Yishuhe River, Luoma Lake, and Nansi Lake. The average concentration value of NH_3-N (i.e., Doushan Reservoir, and Xinan) reached 22.28 and 21.63 mg/L respectively, and the highest concentration value of NH_3-N exceeded 22 times the maximum value of class III (1 mg/L). There were some rivers in which the water pollutant concentrations of NH_3-N belonged to class III or below, such as the downstream of the Huaihe River, Shihe River, Pihe River, and Tuohe River, plus some of the monitoring stations in the mainstream of the Huaihe River, Hongze Lake, Yishuhe River, Luoma Lake, and Nansi Lake.

In Fig. 2e, the water quality classification of TP for 73 monitoring stations is presented. The water in classes II, III, IV, and V and inferior class V accounted for 34.24, 32.88, 10.96, 12.33, and 9.59 % of the TP concentrations, respectively, and the monitoring stations with TP pollutant concentrations that exceeded the class IV level accounted for 32.88 %. There were seven monitoring stations (i.e., Xinan, Jinsuozhen, Sunzhuang, Doushan Reservoir, Huaidian Sluice, Huzhai, and Xiangcheng) at which the water pollutant concentrations reached the inferior class V level, and the average concentrations of TP reached 1.386 mg/L, 1.005 mg/L, 0.704 mg/L, 0.572 mg/L, 0.459 mg/L, 0.458 mg/L, and 0.402 mg/L, respectively; the highest concentration value of TP exceeded seven times the maximum value of class III (0.2 mg/L). There were some rivers in which the water pollutant concentrations of TP belonged to class III or below, such as the mainstream of the Huaihe River, Shihe River, Pihe River, Tuohe River, and Hongze Lake and the downstream of the Huaihe River, plus some of the monitoring stations in the Guohe River, Yishuhe River, Luoma Lake, and Nansi Lake; however, there were several monitoring stations at which the TP pollutant concentrations belonged to class IV or above.

Overall, NH_3-N was the most serious pollutant in the Huai River Basin, followed by COD_{Mn}, BOD_5, and TP; the water quality in terms of DO concentration was the best. The Yinghe River, Quanhe River, Honghe River, Guohe River, and Baohe River were the most seriously polluted rivers, followed by Hongze Lake, Luoma Lake, Yishuhe River, and Nansi Lake.

Temporal-spatial variation trend of water quality from 2003 to 2012

The variation trend of the water pollutant concentrations (i.e., BOD_5, COD_{Mn}, DO, NH_3-N, and TP) at the monitoring stations was analyzed by Mann-Kendall trend analysis, as shown in Fig. 3. The quantities of wastewater effluent of the provinces (i.e., Henan, Anhui, Shandong, and Jiangsu) in the Huai River Basin from 2003 to 2012 are shown in Fig. 4.

In Fig. 3a, there were ten monitoring stations at which the BOD_5 concentrations presented a significant increasing trend—Liji Sluice, Sanyuandian, Shangheli, Shaobo Sluice, Shuyang Sluice, Zhangshan Sluice, Houying, Nanyangnan, Makou, and Peicheng Sluice—which were mainly distributed in the Shihe River, Tuohe River, the downstream of the Huaihe River, Luoma Lake, and Nansi Lake; there were 21 monitoring stations at which the BOD_5 concentrations showed a significant decreasing trend, which were mainly distributed in Yinghe River, the mainstream of the Huaihe River, Hongze Lake, and Yishuhe River.

The variation trend of COD_{Mn} concentration is shown in Fig. 3b; six monitoring stations (i.e., Liji Sluice, Shenqiu Sluice, Shangheli, Xinhetou, Daguanzhuang, and Makou) showed a significant increasing trend, which were mainly distributed in the Shihe River, Quanhe River, Tuohe River, Hongze Lake, and Nansi Lake. There were 50 monitoring stations at which the COD_{Mn} concentrations presented a significant decreasing trend, which indicated that the effect of COD_{Mn} pollution in the Huai River Basin was weakening year by year, and the variation trend was basically consistent for the quantity of wastewater effluent of COD (as shown in Fig. 4a).

In Fig. 3c, there were eight monitoring stations at which DO concentrations presented a significant decreasing trend—Bengbu Sluice, Xiepu, Shangheli, Sanjiangying, Shaobo Sluice, Shuyang Sluice, Zhangshan Sluice, and Ahu Reservoir—which indicated that DO concentration decreased year by year and that the water quality was deteriorating. There were 39
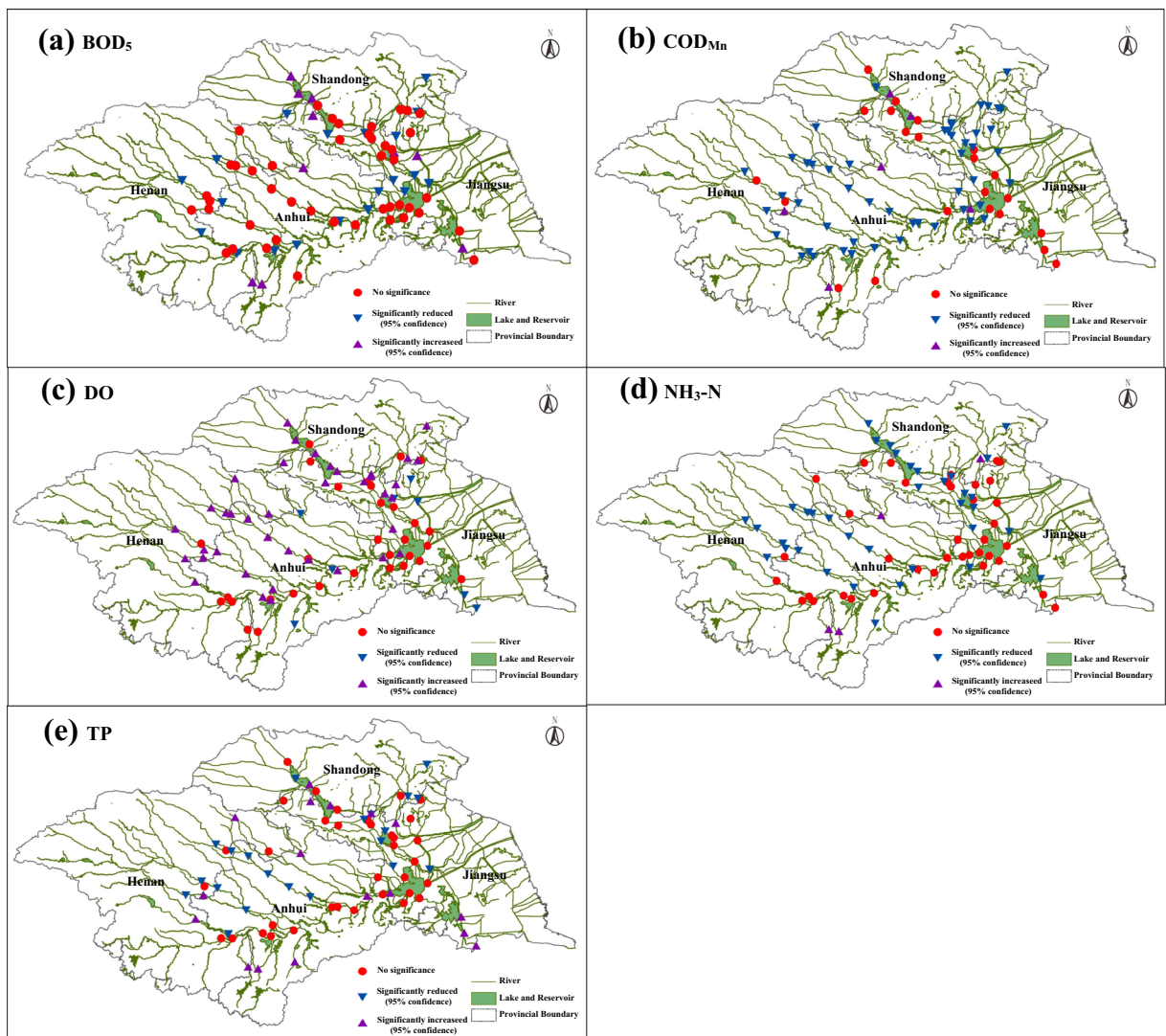
Fig. 3 The variation trend of water quality concentration for each monitoring station
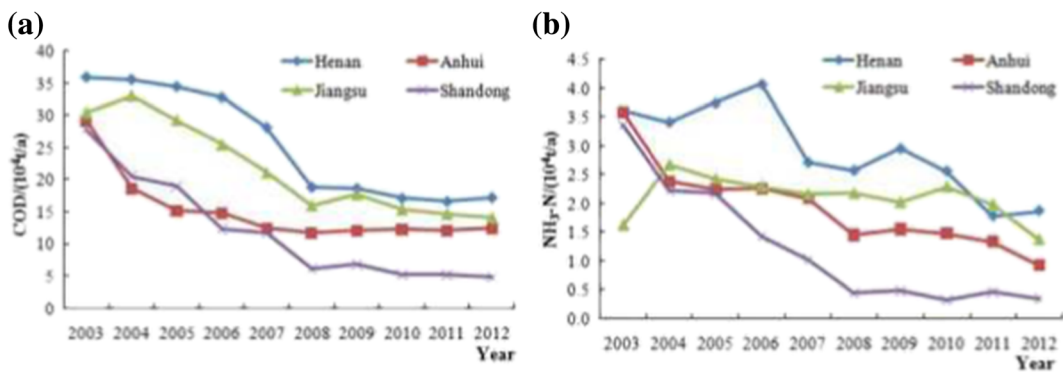

Fig. 4 The quantity of wastewater effluent of the province in the Huaihe River Basin from 2003 to 2012

monitoring stations at which DO concentrations showed a significant increasing trend, which were mainly distributed in the Yinghe River, Quanhe River, Honghe River, Guohe River, Huihe River, and Baohe River, plus some of the monitoring stations in the Yishuhe River, Luoma Lake, Nansi Lake, and the downstream of the Huaihe River.

The variation trend of NH_3-N concentration is shown in Fig. 3d, there were only four monitoring stations (i.e., Liji Sluice, Sanyuandian, Shangheli, and Liuji Sluice) that showed a significant increasing trend. However, almost half the monitoring stations recorded a significant decreasing trend, which were mainly distributed in the Yinghe River, Shihe River, Guohe River, Luoma Lake, and Nansi Lake, indicating that the effect of NH_3-N pollution in the Huai River Basin was weakening year by year; the variation trend was basically consistent for the quantity of wastewater effluent of NH_3-N (as shown in Fig. 4b).

In Fig. 3e, there were 17 monitoring stations at which the TP concentrations presented a significant increasing trend, which were mainly distributed in the Shihe River, Honghe River, Pihe River, Baohe River, Tuohe River, Nansi Lake, and the downstream of the Huaihe River. There were 21 monitoring stations at which the TP concentrations showed a significant decreasing trend, which were mainly distributed in the Yinghe River, Guohe River, Yishuhe River, Luoma Lake, and Hongze Lake.

The boxplot analysis for the water quality concentrations

According to the spatial distribution of water quality pollution, temporal-spatial variation trend of water quality, and the distribution of monitoring stations, the 20 stations were selected to analyze the variation characteristics of the water quality concentrations via boxplot analysis.

The boxplot analysis for BOD$_5$ concentrations

According to the temporal variations of BOD$_5$ concentration in the 20 monitoring stations between 2003 and 2012 shown in Fig. 5, the medians of some monitoring stations were obviously closer to the 75th percentile values for BOD$_5$ and presented a strong left-skewed distribution, which indicated that the smaller BOD$_5$ concentration values were relatively concentrated for

the monitoring stations in the corresponding years, such as Wangjiaba (i.e., 2004 and 2007), Diliyu (i.e., 2004 and 2006), Bengbu Sluice (i.e., 2003, 2004 and 2008), Xincai (i.e., 2003 and 2004), Liji Sluice (i.e., 2004 and 2011), Huaidian Sluice (i.e., 2003 and 2007), Xiangcheng (i.e., 2003 and 2004), Xiepu (i.e., 2004, 2009, 2010, 2011 and 2012), Xuanwu Sluice (i.e., 2003 and 2004), Mengcheng Sluice (i.e., 2005), Heshun Sluice (i.e., 2003 and 2010), Sunzhuang (i.e., 2004, 2006 and 2008), Shangheli (i.e., 2004 and 2005), Shaobo Sluice (i.e., 2003 and 2004), Zhangshan Sluice (i.e., 2007), Xinan (i.e., 2003 and 2006), Makou (i.e., 2006 and 2010), and Liushan Sluice (i.e., 2005). The median of some monitoring stations were obviously closer to the 25th percentile values and presented a strong right-skewed distribution, which indicated that the larger BOD$_5$ concentration values were relatively concentrated for the monitoring stations in the corresponding years, such as Wangjiaba (i.e., 2011), Bengbu Sluice (i.e., 2010), Liji Sluice (i.e., 2007 and 2012), Xiepu (i.e., 2006 and 2007), Heshun Sluice (i.e., 2009), Sunzhuang (i.e., 2005), Shangheli (i.e., 2003, 2007, 2010, 2011, and 2012), Jinsuozhen (i.e., 2005), Shaobo Sluice (i.e., 2005, 2008, and 2011), and Zhangshan Sluice (i.e., 2005 and 2010).

The fluctuations of BOD$_5$ concentration were relatively serious according to the length of the box for the monitoring stations (i.e., Bengbu Sluice, Xiepu, Shangheli, and Shaobo Sluice), which indicated that BOD$_5$ concentrations were not stable in each monitoring year. There was a wide range of fluctuation in the earlier stage of the monitoring period, and the fluctuation was weakened in the later stage of the monitoring period in Xincai, Huaidian Sluice, Xiangcheng, Xuanwu Sluice, Mengcheng Sluice, Heshun Sluice, Jinsuozhen, Xinan, Houying, and Liushan Sluice, which indicated that BOD$_5$ concentrations tended to be stable in the later stage of the monitoring period. However, there were many fluctuations in BOD$_5$ concentration in individual monitoring years, such as Wangjiaba (i.e., 2003, 2004, 2005, and 2007), Diliyu (i.e., 2004, 2005, and 2006), Liji Sluice (i.e., 2006 and 2009), Sunzhuang (i.e., 2004, 2005, and 2006), Zhangshan Sluice (i.e., 2007 and 2010) and Makou (i.e., 2006 and 2007).

The boxplot analysis for COD$_{Mn}$ concentrations

In Fig. 6, the medians of some monitoring stations were obviously closer to the 75th percentile values for COD$_{Mn}$
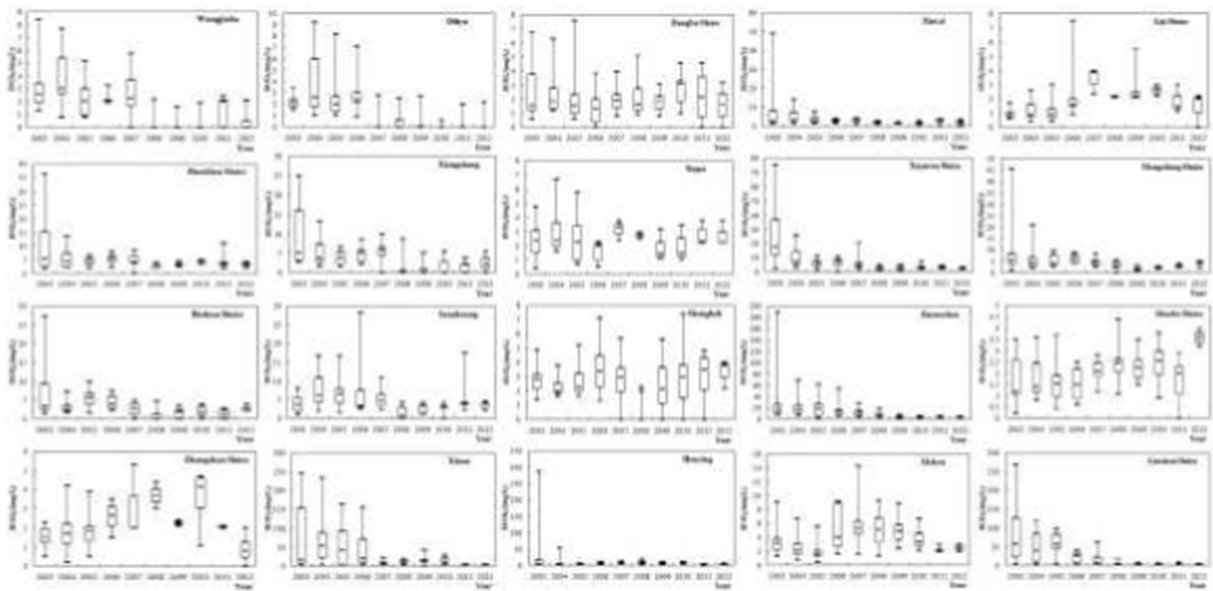
Fig. 5 The boxplots of BOD$_5$ concentrations at 20 stations in the Huaihe River Basin between 2003 and 2012. Maximum and minimum values indicate extremes; *horizontal lines* represent the median, and bars represent the 25th and 75th percentiles

and presented a strong left-skewed distribution for Xincai, Huaidian Sulice, Xiangcheng, and Xinan in 2003, which indicated that the smaller COD$_{Mn}$ concentration values were relatively concentrated for the monitoring stations in the corresponding years. This phenomenon was also reflected in other monitoring stations, such as Diliyu (i.e., 2005), Bengbu Sluice (i.e., 2004, 2005, and 2008), Liji Sluice (i.e., 2005, 2007, and 2009), Xipu (i.e., 2007), Mengcheng Sluice (i.e., 2004), Sunzhuang (i.e., 2003, 2004, and 2006), Shangheli (i.e., 2008), Jinsuozhen (i.e., 2004 and 2008), Shaobo Sluice (i.e., 2008 and 2011) and Zhangshan Sluice (i.e., 2005 and 2007). The medians of some monitoring stations were obviously closer to the 25th percentile values and presented a strong right-skewed distribution, which indicated that the larger COD$_{Mn}$ concentration values were relatively concentrated for the monitoring stations in the corresponding years, such as Bengbu Sluice (i.e., 2003), Xuanwu Sluice (i.e., 2003), Heshun Sluice (i.e., 2003), Diliyu (i.e., 2006), Xiepu (i.e., 2011), Sunzhuang (i.e., 2012), Shangheli (i.e., 2012), Shaobo Sluice (i.e., 2003, and 2005), and Liushan Sluice (i.e., 2003 and 2004).

The fluctuations of COD$_{Mn}$ concentration were relatively serious according to the length of the box for the monitoring stations (i.e., Diliyu, Bengbu Sluice, Liji Sluice, Xiepu, Heshun Sluice, Sunzhuang, Shangheli, Shaobo Sluice, and Makou), which indicated that COD$_{Mn}$ concentrations were not stable in most

monitoring years. There was a wide range of fluctuation in the earlier stage of the monitoring period, and the fluctuation was weakened in the later stage of the monitoring period in Wangjiaba, Xincai, Huaidian Sluice, Xiangcheng, Xuanwu Sluice, Mengcheng Sluice, Jinsuozhen, Xinan, Houying, and Liushan Sluice, which indicated that COD$_{Mn}$ concentrations tended to be stable in the later stage of the monitoring period. However, there were many fluctuations for COD$_{Mn}$ concentration at Zhangshan Sluice in 2007.

The boxplot analysis for DO concentrations

According to the temporal variations of DO concentration at the 20 monitoring stations between 2003 and 2012 shown in Fig. 7, the medians of some monitoring stations were obviously closer to the 75th percentile values for DO and presented a strong left-skewed distribution, which indicated that the smaller DO concentration values were relatively concentrated for the monitoring stations in the corresponding years, such as Wangjiaba (i.e., 2010 and 2012), Diliyu (i.e., 2009 and 2012), Xincai (i.e., 2003), Liji Sluice (i.e., 2009), Huaidian Sluice (i.e., 2010 and 2011), Xiangcheng (i.e., 2012), Xuanwu Sluice (i.e., 2012), Mengcheng Sluice (i.e., 2005 and 2010), Heshun Sluice (i.e., 2004 and 2010), Sunzhuang (i.e., 2003 and 2004), Shangheli (i.e., 2004, 2008, 2009, and 2011), Jinsuozhen (i.e.,
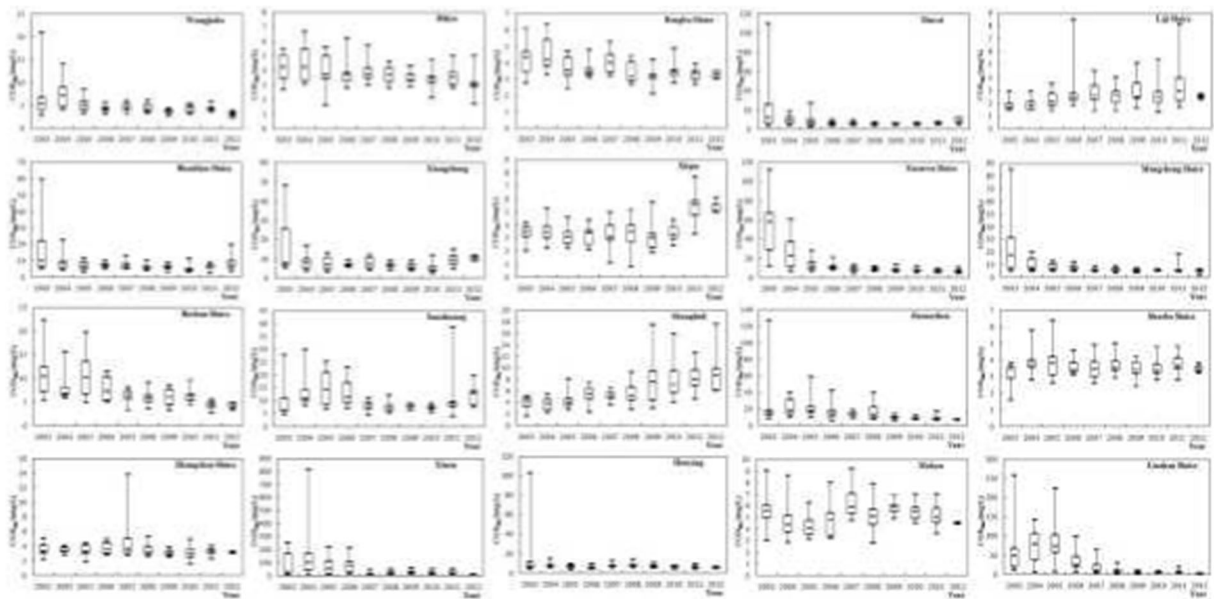
Fig. 6 The boxplots of COD$_{Mn}$ concentrations at 20 stations in the Huaihe River Basin between 2003 and 2012. Maximum and minimum values indicate extremes; *horizontal lines* represent the median, and *bars* represent the 25th and 75th percentiles

2006, 2011, and 2012), Shaobo Sluice (i.e., 2007, 2009, and 2010), Zhangshan Sluice (i.e., 2003, 2009, and 2011), Xinan (i.e., 2003 and 2004), Makou (i.e., 2003, 2005, 2007, and 2011), and Liushan Sluice (i.e., 2011). The medians of some monitoring stations were obviously closer to the 25th percentile values and presented a strong right-skewed distribution, which indicated that the larger DO concentration values were relatively concentrated for the monitoring stations in the corresponding years, such as Wangjiaba (i.e., 2011), Bengbu Sluice (i.e., 2007 and 2010), Liji Sluice (i.e., 2005 and 2012), Xuanwu Sluice (i.e., 2003, 2006, and 2010), Mengcheng Sluice (i.e., 2003 and 2012), Heshun Sluice (i.e., 2012), Sunzhuang (i.e., 2006, 2011, and 2012), Shangheli (i.e., 2005 and 2012), Jinsuozhen (i.e., 2003, 2004, and 2009), Shaobo Sluice (i.e., 2006), Zhangshan Sluice (i.e., 2010 and 2012), Xinan (i.e., 2007 and 2008), Houying (i.e., 2012), Makou (i.e., 2010 and 2012) and Liushan Sluice (i.e., 2006 and 2012).

On the whole, the fluctuations of DO concentration were relatively serious according to the length of the box for the most monitoring stations after 2009, which indicated that DO concentrations were not stable. However, Xinan appeared to have a larger fluctuation from 2003 to 2010, and Makou presented a large fluctuation from 2006 to 2011.

The boxplot analysis for NH$_3$-N concentrations

In Fig. 8, the medians of some monitoring stations were obviously closer to the 75th percentile values for NH$_3$-N and presented a strong left-skewed distribution for Wangjiaba, Diliyu, Bengbu Sluice and Xincai in most monitoring years, which indicated that the smaller NH$_3$-N concentration values were relatively concentrated for the monitoring stations in the corresponding years. This phenomenon was also reflected in other monitoring stations, such as Liji Sluice (i.e., 2005 and 2007), Huaidian Sluice (i.e., 2005 and 2007), Xiangcheng (i.e., 2003, 2006, 2008 and 2009), Xipu (i.e., 2005 and 2008), Xuanwu Sluice (i.e., 2006 and 2009), Mengcheng Sluice (i.e., 2007 and 2008), Sunzhuang (i.e., 2003), Sunzhuang (i.e., 2003 and 2009), Shangheli (i.e., 2009 and 2012), Jinsuozhen (i.e., 2009, 2011 and 2012), Shaobo Sluice (i.e., 2009), Zhangshan Sluice (i.e., 2003, 2008, 2010 and 2011), Xinan (i.e., 2003, 2004, 2005 and 2007), Houying (i.e., 2003) and Liushan Sluice (i.e., 2004 and 2007). The medians of some monitoring stations were obviously closer to the 25th percentile values and presented a strong right-skewed distribution, which indicated that the larger NH$_3$-N concentration values were relatively concentrated for the monitoring stations in the
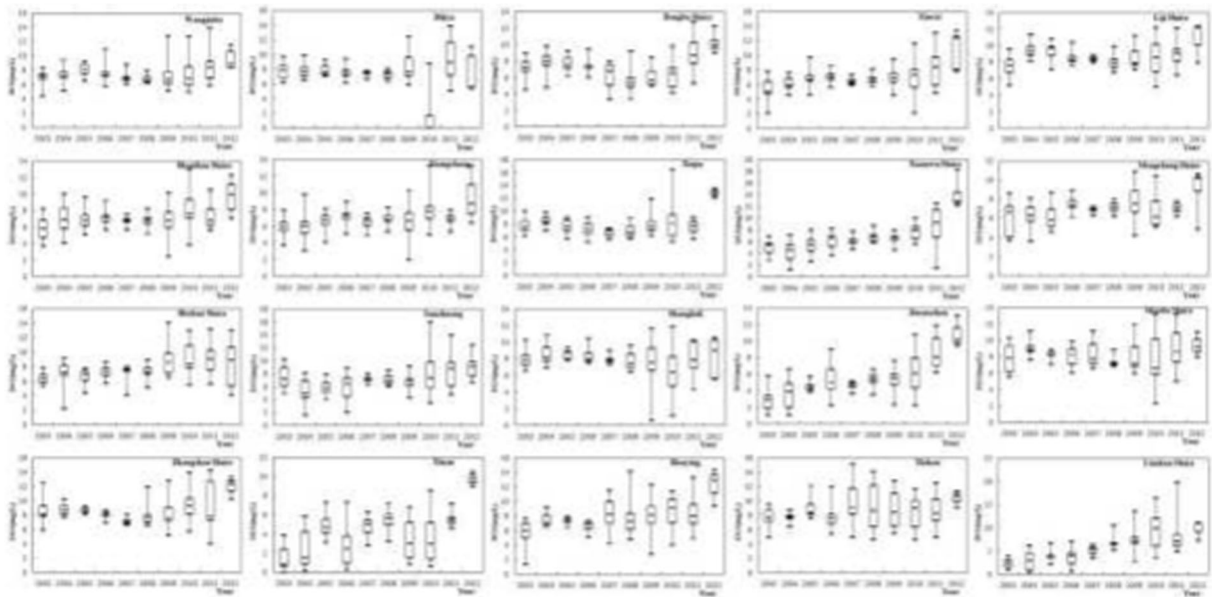
Fig. 7 The boxplots of DO concentrations at 20 stations in the Huaihe River Basin Between 2003 and 2012. Maximum and minimum values indicate extremes; horizontal lines represent the median, and bars represent the 25th and 75th percentiles

corresponding years, such as Diliyu (i.e., 2003), Bengbu Sluice (i.e., 2003), Xincai (i.e., 2010 and 2012), Liji Sluice (i.e., 2006 and 2010), Huaidian Sluice (i.e., 2004), Xiangcheng (i.e., 2004), Xuanwu Sluice (i.e., 2004, 2005, and 2010), Sunzhuang (i.e., 2012), Shaobo Sluice (i.e., 2012), Zhangshan Sluice (i.e., 2005, 2007, and 2012), Xinan (i.e., 2006, 2009, and 2011) and Liushan Sluice (i.e., 2005).

The fluctuations of NH$_3$-N concentration were relatively serious according to the length of the box for the monitoring stations (i.e., Wangjiaba, Diliyu, Bengbu Sluice, Xincai, Xiangcheng, Xuanwu Sluice, Sunzhuang, Jinsuozhen, Zhangshan Sluice, and Xinan) in most monitoring years, which indicated that NH$_3$-N concentrations were not stable. There was a wide range of fluctuation in the earlier stage of the monitoring period, and the fluctuation was weakened in the later stage of the monitoring period in Huaidian Sluice, Mengcheng Sluice, and Shaobo Sluice, which indicated that NH$_3$-N concentrations tended to be stable in the later stage of the monitoring period. However, there were many fluctuations for NH$_3$-N concentration in the individual monitoring years, such as Heshun Sluice (i.e., 2003), Houying (i.e., 2003), Makou (i.e., 2003), Liji Sluice (i.e., 2006 and 2009), Xiepu (i.e., 2003 and 2005), Liushan Sluice (i.e., 2005 and 2007), and Shangheli (i.e., 2009, 2010, 2011, and 2012).

The boxplot analysis for TP concentrations

According to the temporal variations of TP concentration in the 20 monitoring stations between 2003 and 2012 shown in Fig. 9, the medians of some monitoring stations were obviously closer to the 75th percentile values for TP and presented a strong left-skewed distribution, which indicated that the smaller TP concentration values were relatively concentrated for the monitoring stations in the corresponding years, such as Wangjiaba (i.e., 2005 and 2012), Diliyu (i.e., 2003 and 2007), Bengbu Sluice (i.e., 2004 and 2010), Xincai (i.e., 2006, 2009, and 2012), Liji Sluice (i.e., 2004, 2005, 2008, and 2009), Huaidian Sluice (i.e., 2003 and 2006), Xiangcheng (i.e., 2003 and 2009), Xiepu (i.e., 2005 and 2008), Xuanwu Sluice (i.e., 2003, 2004, and 2012), Mengcheng Sluice (i.e., 2005), Heshun Sluice (i.e., 2011), Sunzhuang (i.e., 2003, 2006, and 2012), Jinsuozhen (i.e., 2004 and 2006), Shaobo Sluice (i.e., 2004, 2007, 2008, and 2011), Zhangshan Sluice (i.e., 2004), Xinan (i.e., 2003, 2007, 2008, 2010, and 2011), and Liushan Sluice (i.e., 2004, 2007, 2008, 2011, and 2012). The medians of some monitoring stations were obviously closer to the 25th percentile values and presented a strong right-skewed distribution, which indicated that the larger TP concentration values were relatively concentrated for the monitoring
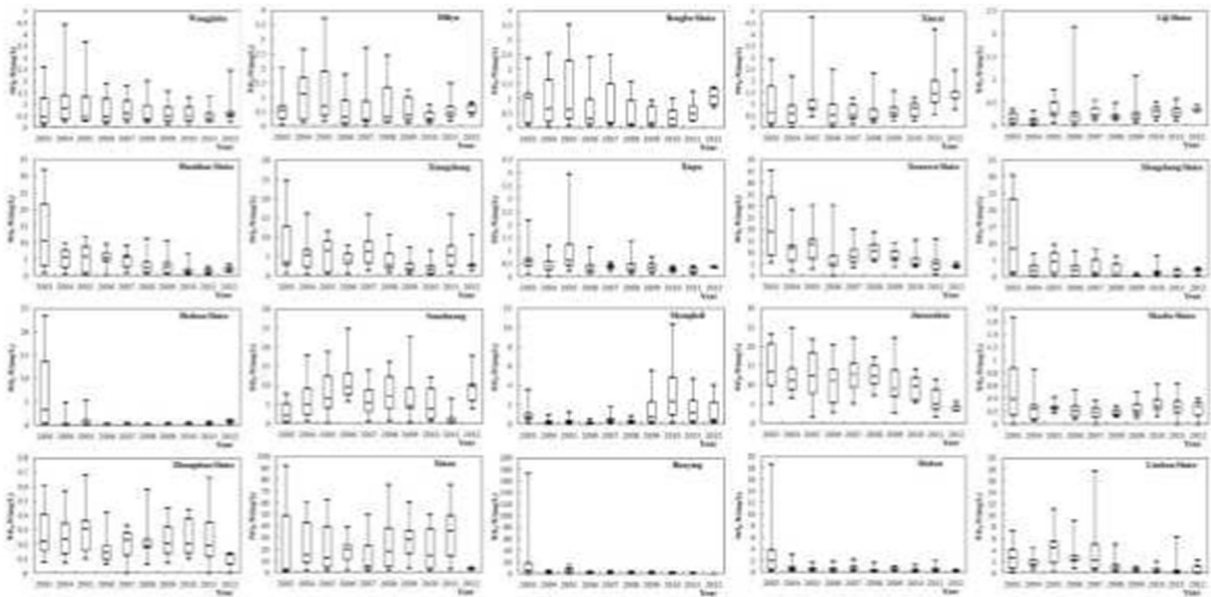
Fig. 8 The boxplots of NH$_3$-N concentrations at 20 stations in the Huaihe River Basin between 2003 and 2012. Maximum and minimum values indicate extremes; *horizontal lines* represent the median, and *bars* represent the 25th and 75th percentiles

stations in the corresponding years, such as Wangjiaba (i.e., 2011), Diliyu (i.e., 2004, 2005, and 2009), Huaidian Sluice (i.e., 2004), Liji Sluice (i.e., 2011), Xiepu (i.e., 2009), Heshun Sluice (i.e., 2003), Sunzhuang (i.e., 2004, 2007, and 2009), Shangheli (i.e., 2009, 2010, 2011, and 2012), and Shaobo Sluice (i.e., 2012).

The fluctuations of TP concentration were relatively serious according to the length of the box for the monitoring stations (i.e., Xiangcheng, Sunzhuang, Xinan, Shaobo Sluice, and Liushan Sluice) in most monitoring years, which indicated that TP concentrations were not stable. However, there were many fluctuations for TP concentration
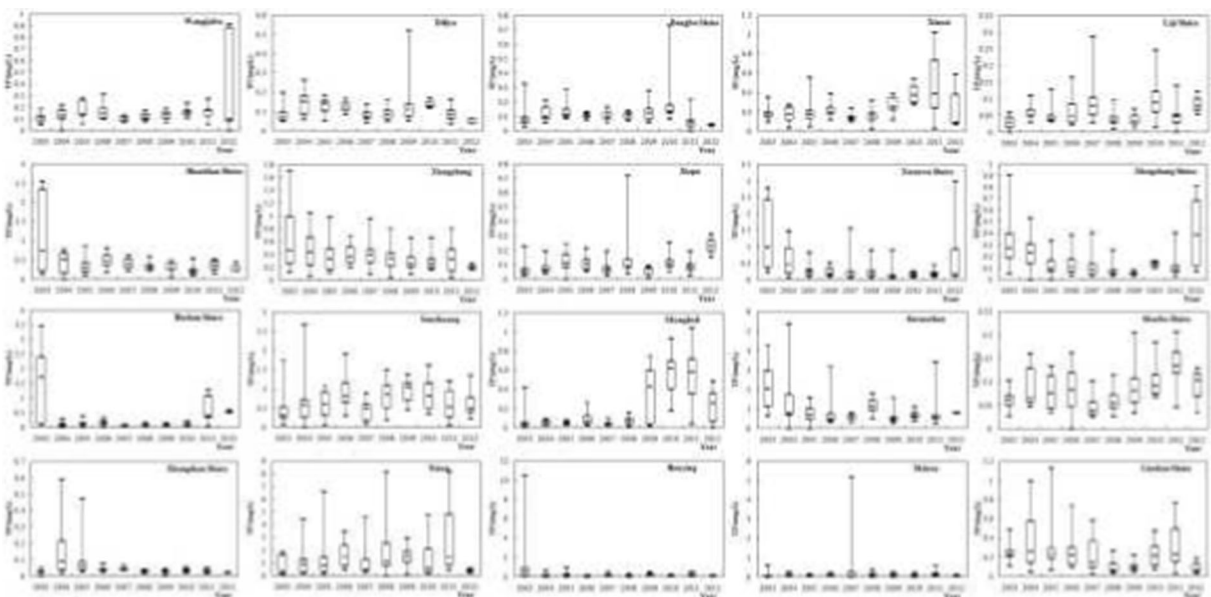

Fig. 9 The boxplots of TP concentrations at 20 stations in the Huaihe River Basin between 2003 and 2012. Maximum and minimum values indicate extremes; *horizontal lines* represent the median, and *bars* represent the 25th and 75th percentiles
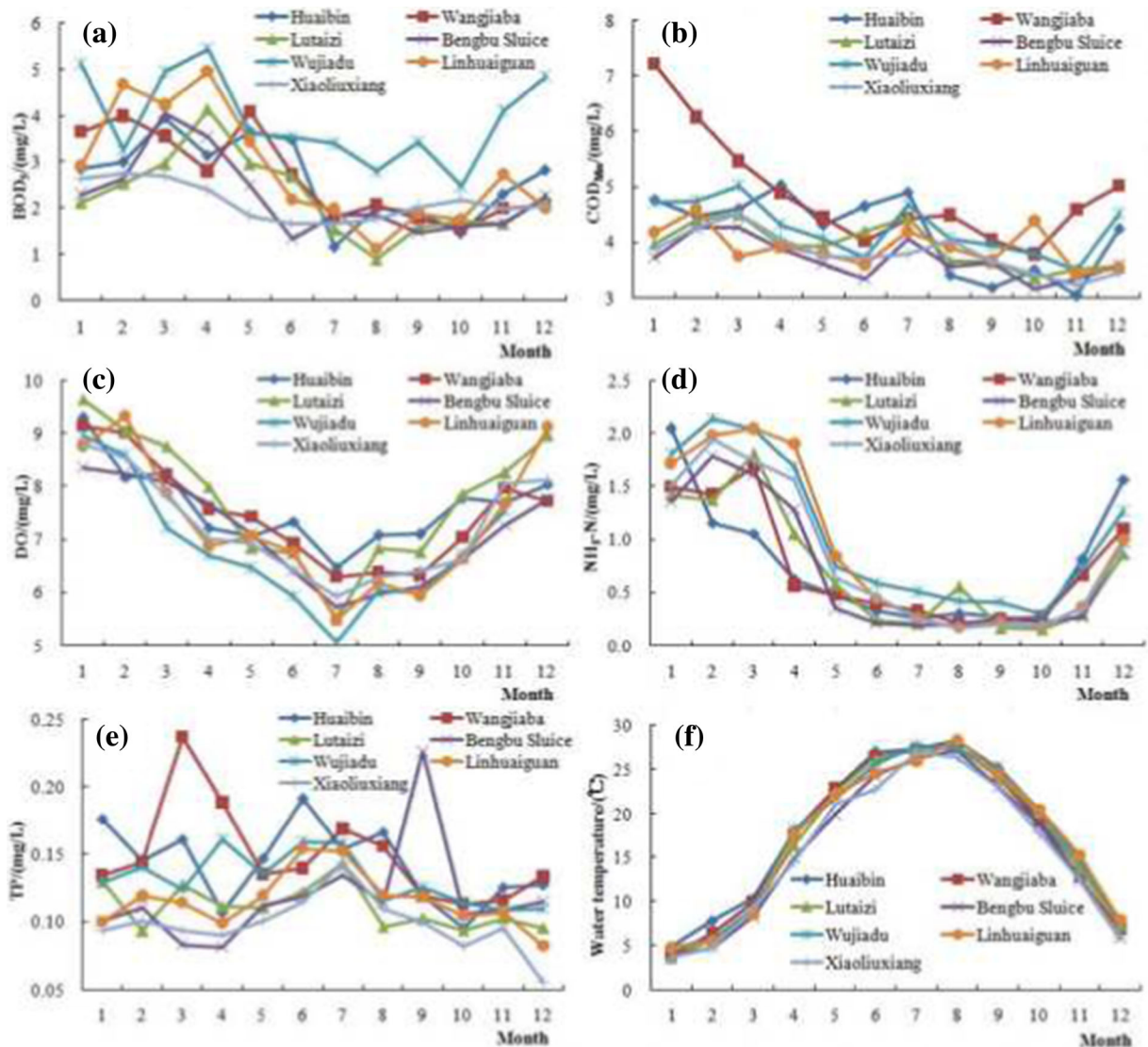
Fig. 10 The seasonal variations of the water quality concentrations and the water temperatures

in individual monitoring years, such as Huaidian Sluice (i.e., 2003), Houying (i.e., 2003), Makou (i.e., 2007, 2009, 2010, and 2012), Diliyu (i.e., 2007, 2009, 2010, and 2012), Bengbu Sluice (i.e., 2007, 2009, 2010, and 2012), Wangjiaba (i.e., 2007, 2009, 2010, and 2012), Xincai (i.e., 2005, 2011, and 2012), Liji Sluice (i.e., 2007 and 2010), Xuanwu Sluice (i.e., 2003, 2004, and 2012), Mengcheng Sluice (i.e., 2003, 2004, and 2012), Heshun Sluice (i.e., 2003 and 2011), Jinsuozhen (i.e., 2003, 2004, 2006, and 2011), Zhangshan Sluice (i.e., 2004 and 2005), and Shangheli (i.e., 2009, 2010, 2011, and 2012).

Seasonal variations of the water quality concentrations

To analyze the seasonal change rules of water quality concentrations in the mainstream of the Huaihe River, seven monitoring stations (i.e., Huaibin, Wangjiaba, Lutaizi, Bengbu Sluice, Wujiadu, Linhuaiguan, and Xiaoliuxiang) were selected in this study. The seasonal variations of the water quality concentrations and the water temperatures are shown in Fig. 10.

According to Fig. 10, the monthly variation trends of the BOD_5, COD_{Mn}, DO, NH_3-N, TP concentrations, and the water temperatures were basically consistent among the seven monitoring stations, which showed a

significant negative correlation between the variation trends of DO, NH_3-N, and the variation trend of the water temperatures, but the variation trends of BOD_5, COD_{Mn}, and TP were not to significantly for the variation trend of the water temperatures. The seasonal variation trend of BOD_5 concentrations for the seven monitoring stations is shown in Fig. 10a. The BOD_5 concentrations reached the maximum in March and April, and a decreasing trend followed. The BOD_5 concentrations reached their minimum in July and August, followed a slightly increasing trend, which indicated that the BOD_5 concentrations would increase in the dry season because the river flows were slow, and the BOD_5 concentrations would rapidly decrease in the wet season with the increasing stream discharge. The BOD_5 concentrations at Wujiadu were significantly higher than the BOD_5 concentration values at the other monitoring stations after June, which may be due to the impact of sewage discharge.

In Fig. 10b, the COD_{Mn} concentrations presented fluctuation changes for the seven monitoring stations. There were obvious fluctuations in July and November, which were affected by the change of the inflow water quantity in the flood and non-flood season in the rivers. The COD_{Mn} concentrations at Wangjiaba were significantly higher than the COD_{Mn} concentration values at the other monitoring stations because the river flow decreased in January, February, and March. The DO concentrations showed the tendency of decreasing first and then increasing for the seven monitoring stations in Fig. 10c, and the DO concentrations reached their minimum in July, but the water temperatures showed the tendency of increasing first and then decreasing for the seven monitoring stations, and the water temperatures reached their maximum in July, which indicated that the change of DO concentrations is significantly influenced by the regional environmental temperature and the DO concentrations would decrease when the water temperature increased.

The seasonal variation trend of NH_3-N concentrations for the seven monitoring stations is shown in Fig. 10d. The seven monitoring stations had higher NH_3-N concentration values in January, February, and March but showed an obvious decreasing tendency after April. The NH_3-N concentrations were in a state of lower concentration values from May to October, whereas the NH_3-N concentrations showed a rebound phenomenon in November and December; which indicated that the NH_3-N concentrations were affected by

the change of the inflow quantity in the flood and non-flood season in the rivers. On the whole, the variation trend of NH_3-N concentrations was obvious influence by the water temperatures. In Fig. 10e, the TP concentrations presented fluctuation changes for the seven monitoring stations, and there were obvious fluctuations in July. The TP concentrations had a larger fluctuation at Wangjiaba and Bengbu Sluice in March and September, respectively, which may be caused by a sudden increase in the sewage discharge.

Conclusions

Using water quality monitoring data from 2003 to 2012, this study analyzed the temporal and spatial characteristics of water pollutant concentrations in the Huaihe River Basin using Mann-Kendall trend analysis and boxplot analysis. Several concluding remarks from this research are as follows. (1) The Yinghe River, Quanhe River, Honghe River, Guohe River, and Baohe River were the most seriously polluted rivers, followed by Hongze Lake, Luoma Lake, and Yishuhe River and Nansi Lake. BOD_5, COD_{Mn}, and NH_3-N were the major pollution indicators—more than 40 % of the monitoring stations reported that the water quality concentrations exceeded the class IV level—followed by the TP concentrations, and the water quality in terms of DO concentrations was the best. (2) There were 21, 50, 36, and 21 monitoring stations that recorded significantly decreasing trend for BOD_5, COD_{Mn}, NH_3-N, and TP, respectively; 39 monitoring stations showed a significantly increasing trend for DO, which indicated that the water quality had gradually improved in the Huaihe River Basin. However, a few of the monitoring stations showed a significantly increasing trend for BOD_5, COD_{Mn}, NH_3-N, and TP, which were mainly distributed in the Shihe River, Tuohe River, Nansi Lake, and the downstream of the Huaihe River, which become the main area of water pollution control. (3) The water quality concentrations had a certain concentricity and volatility according to boxplot analysis for the 20 monitoring stations. The majority of monitoring stations (i.e., Huaidian Sluice, Xuanwu Sluice, Mengcheng Sluice, Heshun Sluice, Xinan, and Houying) recorded a large fluctuation for the monitoring indexes in 2003 and 2004, which indicated that the water quality concentrations were unstable. The fluctuation was obvious from 2009 to 2012 at Shangheli. (4) According to the

seasonal variations of the water quality concentrations in the mainstream of the Huaihe River, the monthly variation trends of the BOD_5, COD_{Mn}, DO, NH_3-N, and TP concentrations were basically consistent among the seven monitoring stations. The BOD_5, COD_{Mn}, NH_3-N, and TP concentrations were affected by changes in the stream discharge; the change of DO and NH_3-N concentrations are influenced by the water temperatures, and the DO and NH_3-N concentrations would decrease when the water temperature increased.

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