

Effect and Risk Assessment of Animal Manure Pollution on Huaihe River Basin, China

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Abstract: Currently the deteriorated water quality for Huaihe River Basin (HRB) in China was still serious because of the negative influence multiple pollution sources including animal manure. However, little attention was paid to the potential risk of animal manure for farmland and water quality of HRB. This study was quantified and forecasted animal manure risk and its spatiotemporal variations in HRB from 2008 to 2018, through pollution discharge coefficient method and pollution load calculation, combined with kriging interpolation method of ArcGIS technology, based on statistics principle. All the data were originated from livestock and poultry breeding in HRB from 2008 to 2018. The future risk of farmland and water environment in HRB was further forecasted. The results indicated that the livestock and poultry manure has become a key pollution source causing a negative influence on farmland and water quality owing to a large amount of animal manure production without efficient recycle utilization. The chemical oxygen demand (COD) and total nitrogen (TN) discharge of animal manure in HRB almost accounted for 17.00% and 39.00% of the whole COD and TN discharge in China. The diffusion concentration of TN and TP in those regions of Shangqiu, Zhoukou, Heze, Zhumadian, Luohe, Jining, Xuchang, Kaifeng, Taian and Zhengzhou of HRB has exceeded the threshold value 10.00 mg/L of TN and 0.08 mg/L of TP, causing water eutrophication and cancer villages. The assessment of farmland and water quality risk revealed that Zhumadian, Zhoukou, Shangqiu, Taian, Jining, Heze, Linyi and Rizhao belonged to high risk areas in HRB, which were still obtained high farmland and water quality risk index in 2030. The results provided insight into an important significance of sustainable balance of livestock and poultry development and ecosystem in HRB.

Keywords: Huaihe River Basin (HRB); China; animal manure; farmland load; diffusion concentration; risk assessment

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1 Introduction

Huaihe River Basin (HRB), with complex meteorologic-

al condition, complicated water systems and unique geography and topography, plays an essential role in China (Jiang et al., 2014; Wang et al., 2014). In the past,

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floods and droughts often occurred in HRB from 1949 to 1978, the water quality deteriorated seriously there except floods and droughts from 1979 to 2005, but the accelerated deteriorative water quality was still serious from 2006 to present, as one of among the major seven large river systems in China (Song et al., 2018; Yu et al., 2020). The water quality from HRB has been observed worse than the Grade IV under the National Surface Water Quality Standards in China (Jiang et al., 2011), led to low drinking water security for approximately ten million local residents (Xu et al., 2018) as well as high cancer morbidity and mortality exceeding national average level (Lu et al., 2015; Han et al., 2016). The agricultural pollution, especially animal manure emissions, has outweighed municipal and industrial sources in HRB (Zhang et al., 2015; Han et al., 2016; Liu et al., 2019). The intensive livestock and poultry farms have prevailed in HRB without an effective treatment pathway targeted at animal manure (Zhou et al., 2013; Herrero et al., 2015; Hu et al., 2017; Song et al., 2018). Organic contaminants regarding to chemical oxygen demand (COD), biological oxygen demand (BOD), nitrate (NO_3^- -N), nitrite (NO_2^- -N) and ammonia (NH_4^+ -N) and phosphorus (P) from the animal manure discharged into farmland directly, further affected water ecosystems through running off and leaching in HRB (Liu et al., 2019; Zhang et al., 2019). Relevant statistics showed that 2.10×10^6 t total nitrogen (TN) production from animal manure in HRB in 2015, approximately occupied 24.00% of the agricultural non-point sources (Song et al., 2017). The massive livestock manure in HRB (Bao et al., 2019; Post et al., 2020) has caused soil mineralization, water eutrophication, greenhouse effect and ultimately posed a threat of ecological environment and human health (Wang et al., 2003; Bao et al., 2019; Li et al., 2020a, b; Zhou et al., 2020).

Despite composting fermentation technology and anaerobic digestion method have been used to deal with livestock manure (Hwang et al., 2020; Li et al., 2020b), it is still subsistent that potential pollution risk for ecological environment and public health, due to lack of labor and fund support leading to low recycled utilization rate of livestock manure as organic fertilizer (Zhang et al., 2015; Hu et al., 2017; Bao et al., 2019). Based on risk assessment method, negative effects of livestock manure on soil and water environment can be quantified, further to guide public debate and policy development

in maintaining sustainable livestock production (Post et al., 2020; Chen et al., 2021). Utilizing interpolation methodologies such as kriging interpolation method with the aid of ArcGIS technology, spatial and temporal variability of pollution is capable of direct and clear visualization (Venkatesan et al., 2020; Arkoc, 2021). At present, most researchers pay close attention to livestock excrement adverse impact on environment and make an effort to estimate animal manure risk at national and administrative area scale (Song et al., 2012; Sun et al., 2015; Bao et al., 2018a, b). Specifically, researches into simulation of N and P diffusion pollution (Feng et al., 2019), assessment of soil erosion (Li et al., 2019), evaluation of water pollution (Li et al., 2017) as well as appraisal of historical drought (Jiang et al., 2014) have been stressed for HRB. However, little attention has been paid to the spatial and temporal variations analysis and the present and future risk assessment in terms of animal manure at watershed scale such as HRB through pollution discharge coefficient method and pollution load calculation, combined with kriging interpolation method of ArcGIS technology.

Therefore, the potential risk of animal manure for farmland and water quality of HRB in recent decade was investigated. The specific objectives of this study were: 1) to quantify the animal manure production as well as the amount of COD, BOD, NH_4^+ -N, TN and TP produced in animal manure among 2008, 2013 and 2018 in HRB, based on pollution discharge coefficient method; 2) to analyze the animal manure load on farmland and diffusion concentration of COD, BOD, NH_4^+ -N, TN and TP contained in animal manure in terms of the HRB, according to the calculation method of pollution load; 3) to predict and evaluate the spatiotemporal variation of animal manure risk to farmland and water environment from HRB, by means of kriging interpolation method of ArcGIS technology.

2 Materials and Methods

2.1 Study area and data source

The HRB is one of the most important rivers in the eastern China, spanning between $30^\circ 55' \text{N}$ to $38^\circ 20' \text{N}$ and $111^\circ 55' \text{E}$ to $122^\circ 45' \text{E}$ (Song et al., 2017), originating from Tongbai Mountain in the south of Henan Province (Xu et al., 2018) and flowing to the confluence at Sanjiang camp, eventually inflowing into the Yangtze River,

with the length of 1000 km and the catchment area of $27.47 \times 10^4 \text{ km}^2$ (Zhai et al., 2017). The HRB is between Yangtze and Yellow rivers, China. Its east is near to the Yellow sea, its south connects with Dabie and Wanshan mountains, and its north beginning to the southern embankment of the Yellow River and Yimeng Mountain (Song et al., 2017). The HRB mainly covers Henan, Anhui, Shandong and Jiangsu provinces from west to east (Fig. 1a), including thirty-five cities (Fig. 1b).

The population of livestock and poultry, farmland area and the gross amount of water resource from 2008 to 2018 was originated from the statistical yearbook in local bureau of statistics website from the provinces of Anhui, Henan, Shandong, Jiangsu and relevant literatures from the bibliographical database Web of Science (<http://apps.webofknowledge.com/>) and China National Knowledge Infrastructure (<http://www.cnki.net/>).

2.2 Animal manure volume calculation for Huaihe River Basin

2.2.1 Calculating amount of pig dung equivalent

According to the excrement coefficient of livestock and poultry and raising cycle (Table 1) (Song et al., 2012; Bao et al., 2018a; b), the amount of animal manure could be calculated in line with Equation (1).

$$Q_i = N_i \times k_i \times T_i \quad (1)$$

where Q_i is the annual amount of livestock manure (t/yr); N_i is amount of livestock and poultry on hand, head or capita; k_i is the excreta coefficient of animal manure (kg/d); T_i is the raising cycle days of livestock and poultry per year; i is denoted as the kind of cow, pig, sheep or poultry.

Based on the conversion coefficient of pig dung equivalent of animal manure (Table 2) (Song et al., 2012; Bao et al., 2018a; b), as well as the animal manure pro-

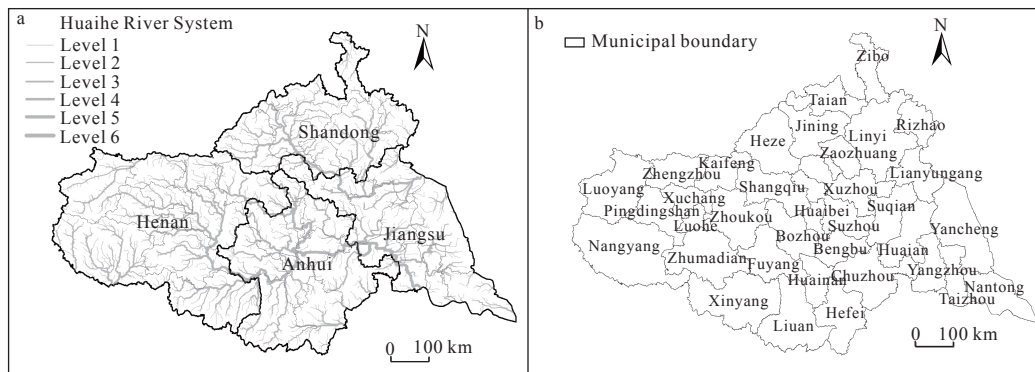


Fig. 1 Main river system (a) and administrative division (b) in Huaihe River Basin

Table 1 Excretion coefficient of animal manure and raising cycle in Huaihe River Basin

Animal	Excretion coefficient / (kg/d)		Raising cycle / (d/a)
	Feces	Urine	
Cow / per head	20.00	10.00	365
Pig / per head	2.00	3.30	199
Sheep / per head	2.60	0.00	365
Poultry / per capita	0.13	0.00	210

Note: data derived from relative references (Song et al., 2012; Bao et al., 2018a; b).

Table 2 Conversion coefficient of pig dung equivalent for animal manure

Coefficient	Pig manure	Pig urine	Cow manure	Cow urine	Sheep manure	Poultry manure
Nitrogen / (%)	0.70	0.33	0.45	0.80	0.80	1.37
μ_i	1.00	0.51	0.69	1.23	1.23	2.10

Note: data originated from relative references (Song et al., 2012; Bao et al., 2018a, b)

duction (Equation (1)), the pig dung equivalent was able to be calculated as follows:

$$Q'_i = Q_i \times \mu_i \quad (2)$$

where Q'_i is the pig dung equivalent (t/yr); μ_i is the conversion coefficient of pig dung equivalent for animal manure.

2.2.2 Calculating pollutant content of animal manure

Animal manure was referred to complex component containing COD, BOD, $\text{NH}_4^+\text{-N}$, TN and TP. On the basis of concentration coefficient of pollutants with respect to COD, BOD, $\text{NH}_4^+\text{-N}$, TN and TP in animal manure (Table 3) (Song et al., 2012; Bao et al., 2018a; b), the amount of COD, BOD, $\text{NH}_4^+\text{-N}$, TN and TP in animal manure was capable of being calculated, respectively. The relevant calculation function was shown in Equation (3).

$$M_j = Q_{ij} \quad (3)$$

where M_j is on behalf of the amount of pollutants in animal manure per year (t/yr); δ_j is concentration coefficient of pollutant in animal manure (kg/t); j is referred to COD, BOD, $\text{NH}_4^+\text{-N}$, TN and TP, respectively.

2.3 Animal manure load on farmland and risk value calculation

2.3.1 Calculating animal manure load on farmland

Feces and urine released from livestock and poultry breeding can be used as organic fertilizers for agricultural production (Li and Liu, 2020), but superabundant waste generated by the livestock and poultry breeding caused a serious pollution for farmland because of the limited land capacity. According to animal manure production (Equation (2)) and farmland area, the animal manure load on farmland was able to be obtained as shown in Equation (4).

$$q = \frac{\sum_{i=1}^4 Q'_i}{S} \quad (4)$$

where q is the animal manure load on farmland (t/(ha·yr)); S is the farmland area (ha).

2.3.2 Risk value and risk level of animal manure load on farmland

The farmland risk value of animal could be calculated, based on Equation (5). The magnitude of animal manure risk for farmland was divided into five levels based on the farmland risk value (Song et al., 2012; Bao et al., 2018a; b), as shown in Table 4.

$$r = \frac{q}{e} \quad (5)$$

where r is the farmland risk value of animal manure; e is the maximum permissible load of animal manure per hectare of farmland, 45.00 t/(ha·yr) (Sun et al., 2015).

2.3.3 Risk forecast of animal manure on farmland

On the basis of farmland risk index in HRB in recent years, the mean year risk increase or decrease rate could be calculated and used to evaluate the prospective farmland risk of animal manure. The risk evaluation of animal manure for farmland was shown in Equation (6), based on relative reference (Bao et al., 2019).

$$r_{\text{future-value}} = r_{\text{mean-value}} \left\{ 1 + \left[\left(\frac{r_{\text{mean-value}}}{r_{\text{initial-value}}} \right)^{\frac{1}{n_{\text{final-year}} - n_{\text{initial-year}}}} - 1 \right] \right\}^{\frac{1}{n_{\text{future-year}} - n_{\text{final-year}}}} \quad (6)$$

where $r_{\text{future-year}}$ is the risk value of animal manure for farmland in future year; $r_{\text{mean-value}}$ is the mean risk value of animal manure for farmland between initial year and final year; $r_{\text{initial-value}}$ is the risk value of animal manure for farmland in initial year; $r_{\text{final-value}}$ is the risk value of

Table 3 Concentration coefficient of pollutants in animal manure / (kg/t)

Faceces	COD	BOD	$\text{NH}_4^+\text{-N}$	TN	TP
Pig manure	52.00	57.03	3.08	5.88	3.41
Pig urine	9.00	5.00	1.43	3.30	0.52
Cow manure	31.00	25.53	1.71	4.37	1.18
Cow urine	6.00	4.00	3.47	8.00	0.40
Sheep manure	4.60	4.10	0.80	7.50	2.60
Poultry manure	45.70	38.90	2.80	10.40	5.80

Note: all data derived from relative references (Song et al., 2012; Bao et al., 2018a; b)

Table 4 Risk value (r) and risk level of animal manure on farmland

r	≤ 0.40	$0.40 < r \leq 0.70$	$0.70 < r \leq 1.00$	$1.00 < r \leq 1.50$	> 1.50
Level	I	II	III	IV	V
Degree of pollution	none	slightly	moderately	extremely	very

animal manure for farmland in final year; $n_{\text{initial-year}}$ is on behalf of the initial year; $n_{\text{final-year}}$ is represented the final year; $n_{\text{future-year}}$ is in the future year.

2.4 Diffusion concentration and water pollution risk from animal manure

2.4.1 Diffusion concentration of animal manure

Animal manure by the way of leaching or surface runoff posed a threat to water eco-environment (Bao et al., 2019). The loss rate of animal manure was approximately at 30.00% according to previous study (Peng et al., 2010). Based on the pollutant content of animal manure (Equation (3)) and the gross of water resource volume the diffusion concentration of pollutants of animal manure in water system was calculated, as shown in Equation (7).

$$C_j = \frac{M_j \times l}{V} \quad (7)$$

where C_j is diffusion concentration of COD, BOD, $\text{NH}_4^+\text{-N}$, TN and TP from animal manure in water, respectively (mg/L); V is the gross of water resource in Huaihe River basin (m^3); l is loss rate of animal manure (30.00%).

2.4.2 Water pollution risk index of animal manure

According to water quality standard level III (MEEPRC, 2002), as well as diffusion concentration of pollutants in animal manure (Equation (7)), water pollution risk index of animal manure could be calculated, as shown in Equation (8). Based on the Equation (8), the magnitude of water pollution risk on water was further divided into five levels, as shown in Table 5.

$$I = \sum_{j=1}^5 \frac{C_j}{C_{0j}} \quad (8)$$

where I is the water pollution risk index of animal manure; C_{0j} is the water quality level III (MEEPRC, 2002)

Table 5 Risk index (I) and risk level of animal manure in water

I	≤ 0.50	$5.00 < I \leq 10.00$	$10.00 < I \leq 15.00$	$15.00 < I \leq 20.00$	> 20.00
Level	I	II	III	IV	V
Degree of pollution	none	slightly	moderately	extremely	very

(mg/L).

2.4.3 Risk forecast of animal manure on water pollution

Based on water risk index in HRB in recent years, the mean year risk increase or decrease rate was obtained and utilized to assess the prospective water risk of animal manure. The risk evaluation of animal manure for water pollution was shown in Equation (9), according to the relative reference (Bao et al., 2019).

$$I_{\text{future-value}} = I_{\text{mean-value}} \left\{ 1 + \left[\left(\frac{I_{\text{mean-value}}}{I_{\text{initial-value}}} \right)^{\frac{1}{n_{\text{final-year}} - n_{\text{initial-year}}}} - 1 \right] \right\}^{\frac{1}{n_{\text{future-year}} - n_{\text{final-year}}}} \quad (9)$$

where $I_{\text{future-year}}$ is the risk value of animal manure for water pollution in future year; $I_{\text{mean-value}}$ is the mean risk value of animal manure for water pollution between initial year and final year; $I_{\text{initial-value}}$ is the risk value of animal manure for water pollution in initial year; $I_{\text{final-value}}$ is the risk value of animal manure for water pollution in final year; $n_{\text{initial-year}}$ is on behalf of the initial year; $n_{\text{final-year}}$ is represented the final year; $n_{\text{future-year}}$ is present in the future year.

2.5 Data processing

Kriging interpolation method based on ArcGIS 10.2 software was adopted to analyze the spatiotemporal distribution of animal manure risk to farmlands and water systems from HRB. The HRB was divided into 178 grids in total with the same size, each grid was approximately 52.25×66.67 km from west to east and from north to south (Fig. 2). Each grid point was attached parameter of risk value from each region location of HRB, of which average risk value in the intersection of

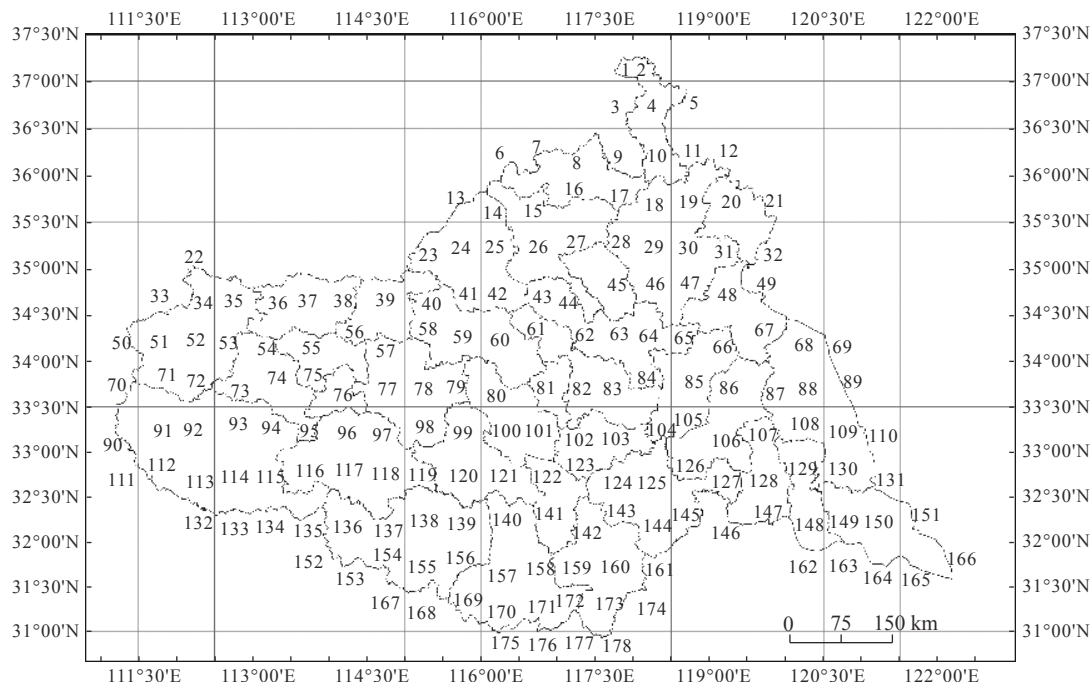


Fig. 2 Grid distribution points for the Huaihe River Basin

region was considered. All original statistical data were analyzed and depicted using Microsoft Excel 2010 software and Origin 9.0 software.

3 Results

3.1 Amount of animal manure in the Huaihe River Basin

3.1.1 Pig dung equivalent production

The pig dung equivalent in HRB from 2008 to 2018 was shown in Fig. 3a. The pig dung equivalent in HRB increased with years firstly and the maximum pig dung equivalent was achieved to 5.34×10^8 t/yr in 2013. Then

the pig dung equivalent gradually decreased in HRB with years. In 2017, the pig dung equivalent was at the minimum value of 3.03×10^8 t/yr. The average pig dung equivalent was at 3.67×10^8 t/yr in HRB from 2008 to 2018. The pig dung equivalent from Henan Province was higher than that from Shandong, Anhui and Jiangsu provinces, in the whole HRB. The result indicated a large amount of animal manure was produced in HRB.

The spatio-temporal variation of pig dung equivalent in HRB every five year during the decade was further emphasized and analyzed, as shown in Fig. 3b. In 2018, the total pig dung equivalent production in HRB was

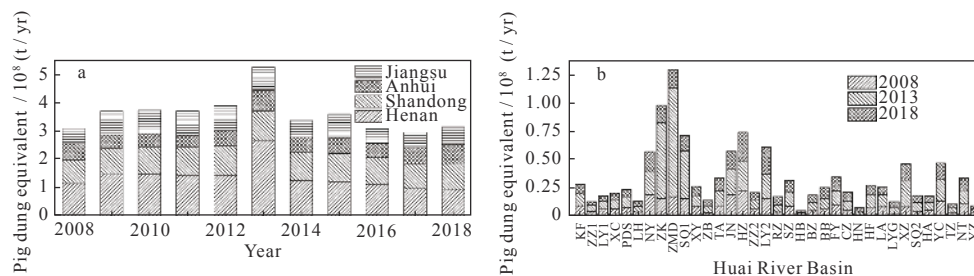


Fig. 3 Pig dung equivalent in the Huaihe River Basin (HRB). The cities in HRB were from Henan Province including Kaifeng (KF), Zhengzhou (ZZ1), Luoyang (LY1), Xuchang (XC), Pingdingshan (PDS), Luohe (LH), Nanyang (NY), Zhoukou (ZK), Zhumadian (ZMD), Shangqiu (SQ1) and Xinyang (XY), Shandong Province including Zibo (ZB), Taian (TA), Jining (JN), Heze (HZ), Zaozhuang (ZZ2), Linyi (LY2) and Rizhao (RZ), Anhui Province including Suzhou (SZ), Huaibei (HB), Bozhou (BZ), Bengbu (BB), Fuyang (FY), Chuzhou (CZ), Huainan (HN), Hefei (HF) and Liuan (LA), and Jiangsu Province including Lianyungang (LYG), Xuzhou (XZ), Suqian (SQ2), Huaian (HA), Yancheng (YC), Taizhou (TZ), Nantong (NT) and Yangzhou (YZ)

achieved to 3.21×10^8 t, which was obviously less than that of 5.34×10^8 t in 2013, but close to that of 3.17×10^8 t in 2008. The pig dung equivalent production in those regions of Zhumadian, Zhoukou, Heze, Shangqiu, Linyi, Jining, Nanyang, Yancheng and Xuzhou reached to 1.28×10^8 t, 0.97×10^8 t, 0.73×10^8 t, 0.71×10^8 t, 0.61×10^8 t, 0.57×10^8 t, 0.47×10^8 t and 0.46×10^8 t among 2008, 2013 and 2018, respectively. It was evident that the pig dung equivalent production in those regions was higher compared with other regions of HRB (Fig. 3b). The total pig dung equivalent production in those regions contrib-

uted to approximately 54.00% in the whole HRB, among 2008, 2013 and 2018.

3.1.2 Pollutants from animal manure

The spatio-temporal variation of pollutants with regard to COD, BOD, $\text{NH}_4^+\text{-N}$, TN and TP from animal manure in HRB was shown in Fig. 4. COD and BOD content of animal manure in HRB in 2018 was achieved to 7.39×10^6 t and 6.67×10^6 t, respectively, which was separately more than 4.00% and 5.00% compared with that content in 2008, but separately less than 42.00% and 38.00% compared with that content in 2013. The

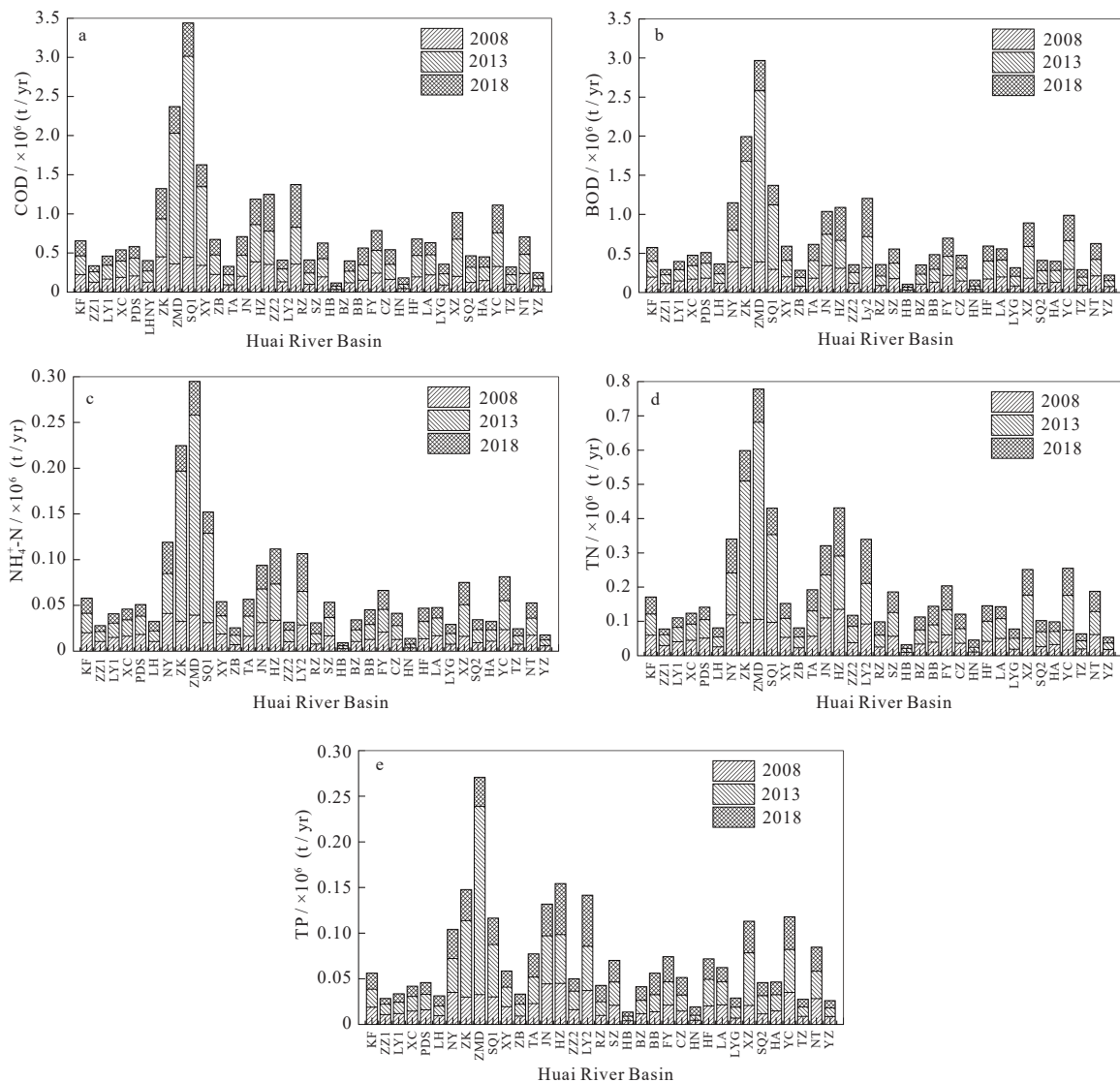


Fig. 4 The content of pollution from animal manure in the Huaihe River Basin (HRB). The cities in HRB were from Henan Province including Kaifeng (KF), Zhengzhou (ZZ1), Luoyang (LY1), Xuchang (XC), Pingdingshan (PDS), Luohe (LH), Nanyang (NY), Zhoukou (ZK), Zhumadian (ZMD), Shangqiu (SQ1) and Xinyang (XY); Shandong Province including Zibo (ZB), Taian (TA), Jining (JN), Heze (HZ), Zaozhuang (ZZ2), Linyi (LY2) and Rizhao (RZ); Anhui Province including Suzhou (SZ), Huaibei (HB), Bozhou (BZ), Bengbu (BB), Fuyang (FY), Chuzhou (CZ), Huainan (HN), Hefei (HF) and Liuan (LA); and Jiangsu Province including Lianyungang (LYG), Xuzhou (XZ), Suqian (SQ2), Huaian (HA), Yancheng (YC), Taizhou (TZ), Nantong (NT) and Yangzhou (YZ)

COD and BOD content in Zhumadian, Zhoukou, Shangqiu, Linyi, Nanyang, Heze, Jining, Yancheng and Xuzhou was obviously higher as compared to other regions of HRB (Figs. 4a, b). The total COD and BOD content in those regions occupied 54.00% in HRB, among 2008, 2013 and 2018.

There were 0.58×10^6 t $\text{NH}_4^+\text{-N}$ and 1.82×10^6 t TN derived from animal manure in HRB in 2018. The $\text{NH}_4^+\text{-N}$ and TN content was close to the content in 2008, but separately decreased by 46.00% and 42.00% compared to 2013. The quantity of $\text{NH}_4^+\text{-N}$ and TN was higher in Zhumadian, Zhoukou, Shangqiu, Linyi, Nanyang, Heze, Jining, Yancheng and Xuzhou, compared with other regions of HRB (Figs. 4c, d). The total content of $\text{NH}_4^+\text{-N}$ and TN accounted for 56.00% in HRB, among 2008, 2013 and 2018.

TP content of animal manure in 2018 was achieved to 0.74×10^6 t, increased by 9.00% compared with 2008, but decreased by 33.00% compared with 2013. The TP content in those regions of Zhumadian, Zhoukou, Shangqiu, Linyi, Nanyang, Heze, Jining, Yancheng and Xuzhou was obviously higher than other regions of HRB (Fig. 4e). The total content of TP nearly took up 52.00% in the whole HRB, among 2008, 2013 and 2018.

3.2 Effect of animal manure on farmland in the Huaihe River Basin

3.2.1 Animal manure load on farmland

The spatio-temporal change for animal manure load on farmland in HRB was presented in Fig. 5. In 2018, the average animal manure load on farmland for HRB was reached to 18.60 t/(ha·yr) and dropped by 12.00% and 44.00%, respectively, compared with 2008 and 2013. It was found that the animal manure load on farmland in those regions of Zhumadian, Zhoukou, Shangqiu, Taian, Jining, Heze, Zaozhuang, Bengbu, Hefei, Xuzhou and Nantong was extremely higher than other regions of HRB (Fig. 5). Especially in 2013, the farmland load in Zhumadian, Zhoukou, Shangqiu, and Xuzhou was achieved to 99.09, 76.61, 58.18 and 46.76 t/(ha·yr), respectively, which has been exceeded the land carrying capacity of 45.00 t/(ha·yr) (Sun et al., 2015).

3.2.2 Farmland risk evaluation of animal manure

The spatial and temporal distribution of animal manure risk to farmland in HRB was shown in Fig. 6. In 2008, the average risk index (r) of HRB was achieved to 0.46.

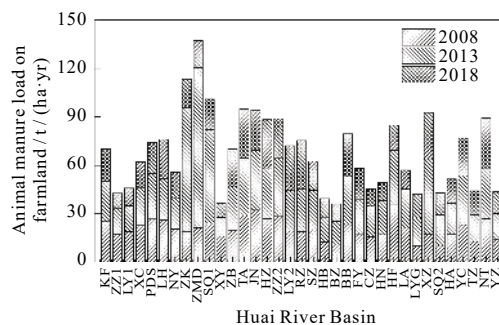


Fig. 5 Animal manure load on farmland in Huaihe River Basin (HRB). The cities in HRB were from Henan Province including Kaifeng (KF), Zhengzhou (ZZ1), Luoyang (LY1), Xuchang (XC), Pingdingshan (PDS), Luohe (LH), Nanyang (NY), Zhoukou (ZK), Zhumadian (ZMD), Shangqiu (SQ1) and Xinyang (XY); Shandong Province including Zibo (ZB), Taian (TA), Jining (JN), Heze (HZ), Zaozhuang (ZZ2), Linyi (LY2) and Rizhao (RZ); Anhui Province including Suzhou (SZ), Huaibei (HB), Bozhou (BZ), Bengbu (BB), Fuyang (FY), Chuzhou (CZ), Huainan (HN), Hefei (HF) and Liuan (LA); and Jiangsu Province including Lianyungang (LYG), Xuzhou (XZ), Suqian (SQ2), Huai-an (HA), Yancheng (YC), Taizhou (TZ), Nantong (NT) and Yangzhou (YZ)

The result indicated that there was a slight pollution due to animal manure emission for the whole HRB (Table 4). In 2008, those regions of Taian, Jining, Zaozhuang, Heze, Hefei, Liuan and Nantong belonged to high risk pollution districts as depicted in Fig. 6a, where the risk level was between II and III with a slight and moderate pollution degree (Table 4). In 2013, the average risk index of HRB was reached to 0.74. It was demonstrated that the farmland of HRB was polluted moderately because of animal manure emissions (Table 4). The risk index has exceeded 1.00 in those regions of Zhumadian, Zhoukou, Shangqiu and Xuzhou (Fig. 6b), where were suffered an extremely serious pollution of animal manure. Additionally, the farmland from the regions of Jining, Zaozhuang, Taian, Heze, Suzhou, Hefei, Yanzhou and Nantong were subject to a moderate extent of animal manure pollution (Fig. 6b, Table 4). In 2018, the farmland risk index was achieved to 0.41 in HRB. The result illustrated that the farmland from the whole HRB exhibited a slight pollution risk on account of animal manure emissions. There was a high risk of farmland pollution in the northeast and southeast of HRB as described in Fig. 6c.

The farmland risk of animal manure in HRB in 2030 was displayed in Fig. 6d, according to Equation (6). The average farmland risk of animal manure in HRB would

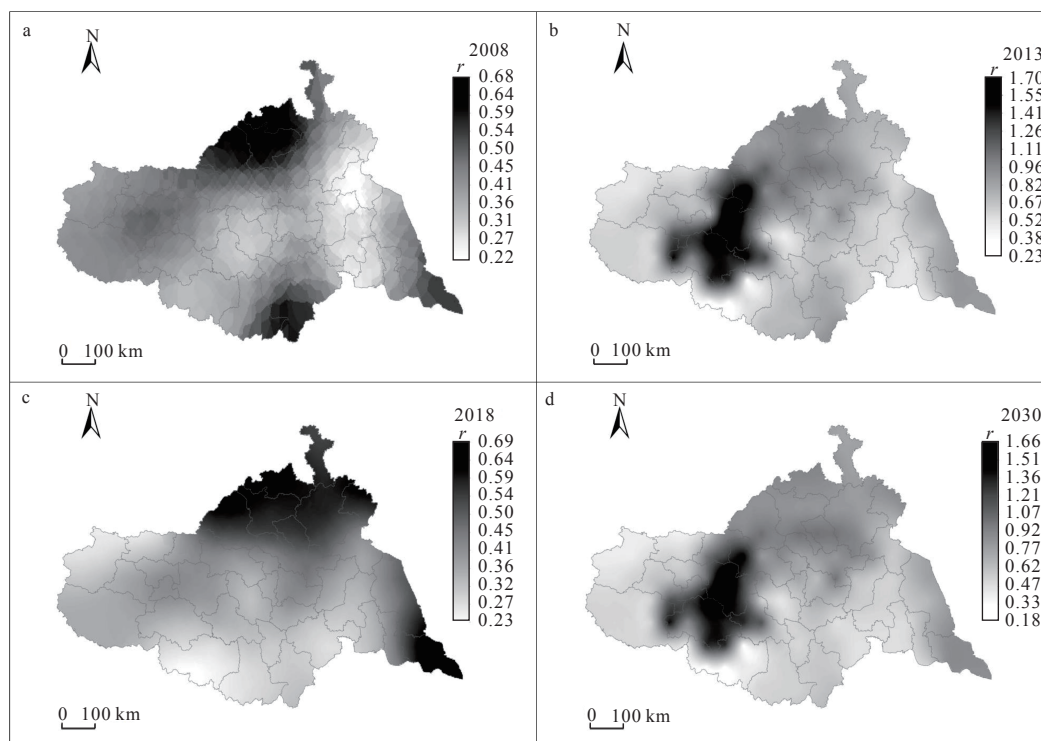


Fig. 6 Risk index (r) of animal manure on farmland in the Huaihe River Basin

be achieved to 0.62 in 2030. It was revealed that there was slight farmland pollution in the whole HRB, due to animal manure emissions. The farmland from those regions of Zhumadian, Zhoukou, Shangqiu and Xuzhou was suffered an extremely serious pollution of animal manure, because the farmland risk index in those regions enough exceeded 1.00 (Table 4). Besides, those regions of Zibo, Taian, Jining, Heze, Zaozhuang, Linyi and Rizhao in the northeast of HRB, as well as Yancheng and Nantong in the southeast of HRB belonged to farmland risk pollution of animal manure (Fig. 6d, Table 4).

3.3 Effect of animal manure on water in Huaihe River Basin

3.3.1 Diffusion concentration of pollutants from animal manure

The spatio-temporal concentration variations of pollutants referring to COD, BOD, $\text{NH}_4^+\text{-N}$, TN and TP from animal manure in HRB was shown in Fig. 7. The COD and BOD concentration in HRB in 2018 was achieved to 24.76 and 22.42 mg/L, respectively, which was lower than 7.00% and 5.00% compared with those concentration in 2008, as well as lower than 49.00% and 45.00% compared with those content in 2013. The concentra-

tion of COD and BOD in Shangqiu, Zhoukou, Luohe, Jining, Heze, Zhumadian, Xuchang, Taian, Kaifeng and Zhengzhou was obviously higher as compared to other regions of HRB (Figs. 7a, b). Among 2008, 2013 and 2018, the average concentration of COD and BOD in those regions of Shangqiu, Zhoukou, Luohe, Jining, Heze, Zhumadian, Xuchang, Taian, Kaifeng and Zhengzhou was significantly higher than the average COD and BOD concentration of 33.18 and 28.87 mg/L in HRB.

The concentration of $\text{NH}_4^+\text{-N}$ and TN derived from animal manure in HRB was achieved to 1.97 and 6.11 mg/L, respectively, in 2018. The $\text{NH}_4^+\text{-N}$ and TN concentration was separately lower than 11.00% and 11.00% of those in 2008, and separately decreased by 52.00% and 50.00% compared to 2013. It was evident that the $\text{NH}_4^+\text{-N}$ and TN concentration was higher in Shangqiu, Zhoukou, Heze, Zhumadian, Luohe, Jining, Xuchang, Kaifeng, Taian and Zhengzhou, compared with other regions of HRB (Figs. 6c, d). In those regions among 2008, 2013 and 2018, the concentration of $\text{NH}_4^+\text{-N}$ and TN was an obvious more than the average 2.75 mg/L $\text{NH}_4^+\text{-N}$ and 8.37 mg/L TN, in HBR.

In 2018, the TP concentration of animal manure was reached to 2.45 mg/L, which was close to that in 2008, but declined by 43.00% compared with 2013. In those

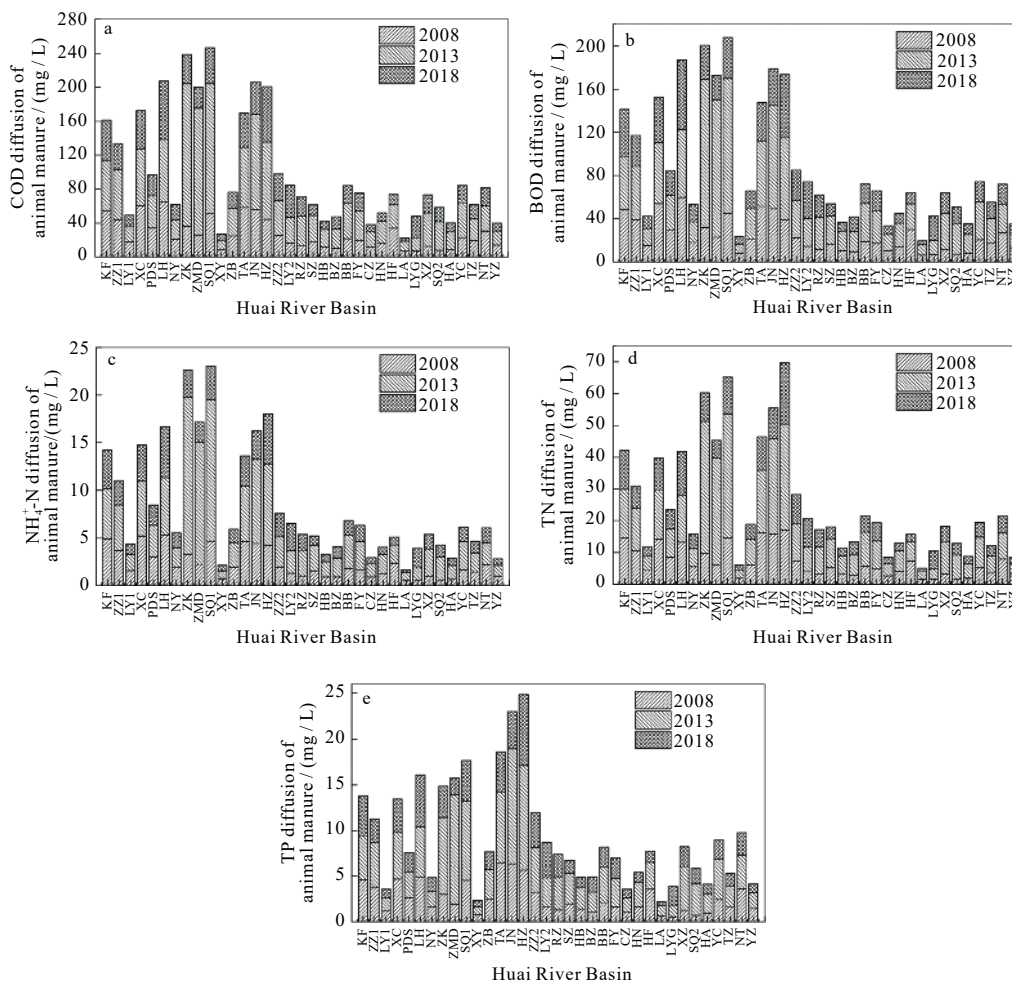


Fig. 7 Diffusion concentration of pollutants from animal manure in Huaihe River Basin (HRB). The cities in HRB were from Henan Province including Kaifeng (KF), Zhengzhou (ZZ1), Luoyang (LY1), Xuchang (XC), Pingdingshan (PDS), Luohe (LH), Nanyang (NY), Zhoukou (ZK), Zhumadian (ZMD), Shangqiu (SQ1) and Xinyang (XY); Shandong Province including Zibo (ZB), Taian (TA), Jining (JN), Heze (HZ), Zaozhuang (ZZ2), Linyi (LY2) and Rizhao (RZ); Anhui Province including Suzhou (SZ), Huaibei (HB), Bozhou (BZ), Bengbu (BB), Fuyang (FY), Chuzhou (CZ), Huainan (HN), Hefei (HF) and Luan (LA); and Jiangsu Province including Lianyungang (LYG), Xuzhou (XZ), Suqian (SQ2), Huaian (HA), Yancheng (YC), Taizhou (TZ), Nantong (NT) and Yangzhou (YZ)

regions of Heze, Jining, Taian, Shangqiu, Luohe, Zhumadian, Zhoukou, Kaifeng, Xuchang and Zhengzhou exhibited higher TP concentration than that of other regions in HRB (Fig. 7e). Among 2008, 2013 and 2018, the TP concentration in those regions markedly exceeded the average TP concentration of 3.09 mg/L in HRB.

3.3.2 Water pollution risk index of animal manure

The spatial and temporal distribution of animal manure risk for water environment in HRB was shown in Fig. 8. In 2008, the average risk index (I) of HRB was achieved to 28.91. It was demonstrated that there was extreme serious water pollution due to animal manure emission for the whole HRB (Table 5). In 2008, Taian, Jining, Heze, Kaifeng, Zhengzhou, Pingdingshan, Xuchang,

Zhoukou, Shangqiu, Zibo and Nantong belonged to very high risk pollution districts as described in Fig. 8a, where the risk level exceeded V along with an extreme pollution degree (Table 5). In 2013, the average risk index of HRB was achieved to 50.26. The result indicated that the water environment was suffered a quite serious pollution due to animal manure emissions (Table 5). The risk index in those regions of Taian, Jining, Heze, Kaifeng, Shangqiu, Xuchang, Zhoukou, Pingdingshan and Zhumadian was far beyond 20.00 (Fig. 8b), owing to the negative influence of animal manure emissions. In 2018, the water pollution risk index of animal manure was achieved to 27.71 in HRB, which was more than 20.00 (Table 5). The result revealed that the water environment was still suffered a severe pollution because of

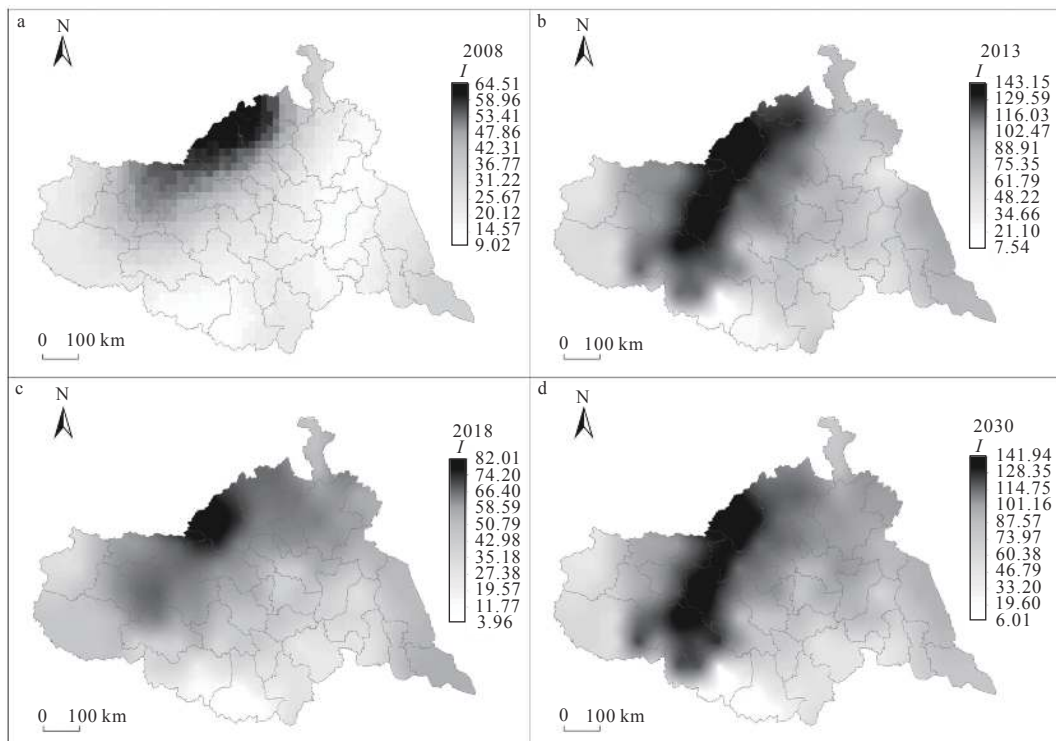


Fig. 8 Risk predication of water pollution (I) from animal manure in the Huaihe River Basin

animal manure emissions. There was very serious water environment pollution in those regions of Heze, Jining, Taian, Kaifeng, Xuchang, Luohe, Zhoukou, Shangqiu, Zhengzhou and Linyi as depicted in Fig. 8c.

The water pollution risk of animal manure in HRB in 2030 was exhibited Fig. 8d. Average water pollution risk of animal manure in HRB was achieved to 47.82 in 2030, on the basis of Equation (9). The predicted result indicated that the water environment in the whole HRB was encountered with a serious pollution due to animal manure emissions. Especially in those regions of Heze, Kaifeng, Shangqiu, Zhoukou, Zhumadian, Jining, Taian, Kaifeng, Luohe, Xuchang, Pingdiangshan, Xinyang, Nantong, Linyi and Rizhao, there was a serious water pollution of animal manure (Fig. 8d, Table 5).

4 Discussion

The pig dung equivalent production in HRB has been achieved to 3.21×10^8 t in 2018 (Equation (2) and Fig. 3), as well as 7.39×10^6 t of COD, 6.67×10^6 t of BOD, 0.58×10^6 t of $\text{NH}_4^+\text{-N}$, 1.82×10^6 t TN and 0.74×10^6 t TP in animal manure (Equation (3) and Fig. 4). The result indicated that there was a massive animal manure production in HRB that was prone to pose

a highly detrimental influence on water quality for HRB. Actually, the water quality in HRB was still a serious deteriorative status currently (Song et al., 2017; Xu et al., 2018; Liu et al., 2019; Yu et al., 2020). Based on the State of the Environment Report of China, 2.30×10^7 t COD and 2.40×10^6 t TN were discharged in 2013 (Lu et al., 2015). The COD and TN discharge of animal manure were reached to 3.83×10^6 t and 0.94×10^6 t, respectively, in line with 30.00% of loss rate of animal manure (Peng et al., 2010). As a result, the COD and TN discharge of animal manure for HRB almost accounted for 17.00% and 39.00% of the whole COD and TN discharge in China (Peng et al., 2010; Lu et al., 2015).

The farmland has been suffered an adverse effect in HRB according to the result of animal manure load, particularly in Zhumadian, Zhoukou, Shangqiu and Xuzhou regions where the animal manure load on farmland has been far beyond the environmental capacity (Sun et al., 2015). Those regions also belonged to high risk farmland districts on account of animal manure emissions, which was agreement with Song et al.'s (2017) study. The farmland risk predication indicated that Zhumadian, Zhoukou, Shangqiu, and Xuzhou in HRB were still a serious farmland risk districts due to animal manure pol-

lution in 2030. Therefore, an efficient control measure should be offered urgently targeting at those regions in order to clear animal manure harm for farmland.

The diffusion concentration of COD, BOD, $\text{NH}_4^+\text{-N}$, TN and TP from animal manure was quite high in HRB. Previous study indicated that the groundwater concentration of $\text{NO}_3^-\text{-N}$ over 10.00 mg/L of the World Health Organization (WHO) standard could induce chronic illness including cancers (Han et al., 2016; He et al., 2019). Research of He et al. (2019) indicated that the concentration more than WHO standard was mainly distributed around the cities of Pingdingshan, Xuchang, Zhoukou, Luohe and Fuyang in Shaying River Basin, as one of the HRB tributaries. This study in terms of TN concentration from animal manure emissions in those regions of Shangqiu, Zhoukou, Heze, Zhumadian, Luohe, Jining, Xuchang, Kaifeng, Taian and Zhengzhou was significantly more than 10.00 mg/L (Fig. 7d) resulting in cancer villages in HRB (Lu et al., 2015). Additionally, the eutrophication thresholds of TN and TP for rivers were separately at 1.20 and 0.08 mg/L (Smith et al., 1999) and TN and TP concentration thresholds should be targeted at below 0.80 and 0.05 mg/L, respectively, whether it is easy to appear *Mircrocystis* dominated bloom (Xu et al., 2015). However, the TN and TP concentration of animal manure emissions in HRB was considerably beyond the eutrophication thresholds as well as intrinsic growth rate of *Mircrocystis* dominated blooms.

Utilizing the quantitative risk evaluation analysis and spatiotemporal distribution change by kriging interpolation method of ArcGIS technology, the results indicated that the water systems from Heze, Jining, Taian, Kaifeng, Xuchang, Luohe, Zhoukou, Shangqiu, Zhengzhou and Linyi in HRB were suffered a severe pollution at present causing by animal manure runoff and leaching. In 2030, those regions above were still faced a serious water pollution risk of animal manure, except Zhumadian, Pingdiangshan, Xinyang, Nantong and Rizhao, almost occupied half regions of the whole HRB.

On the basis of animal manure adverse effect on farmland and water systems with its risk evaluation, we could conclude that livestock and poultry manure has become a key pollution source in HRB. To avoid the animal manure pollution and decrease its harm risk, livestock excrements including feces and urine should be recycled utilization as an effective resource. Compost-

ing and anaerobic digestion were supposed to a potential methods to solve animal manure pollution problems (Hwang et al., 2020; Li et al., 2020b). Although the composting could reduce the quantity of the manure wastes through mineralization process to produce organic fertility, it inevitably emitted malodorous gases and greenhouse gases such as carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) (Hwang et al., 2020). Therefore, composting aiming at animal manure in HRB should consider malodorous gas and greenhouse gas emission. Anaerobic digestion could recycle livestock excrements and produce renewable energy such as biogas, there was created approximately $1.42 \times 10^8 \text{ m}^3$ of biogas when $3.21 \times 10^8 \text{ t}$ of livestock excrements production in HRB in 2018 from the equation offered by Bao et al. (2019). It was worth mentioning that more labor and fund support was demanded through anaerobic digestion to remove animal manure pollution in HRB. In future, drawbacks of composting and anaerobic digestion method could be conquered to realize sustainable and ecological livestock and poultry breeding with harmless and recycle utilization of animal manure in HRB.

5 Conclusions

This study focused on the negative effect of animal manure on farmland and water quality with its risk evaluation in HRB through quantitative analysis including pollution discharge coefficient method and pollution load calculation, combined with kriging interpolation method of ArcGIS technology. The results indicated that the water quality in HRB was still at a serious pollution status at present and the livestock and poultry manure has become an important pollution source posing a negative influence on farmland and water quality due to a large amount of animal manure production without efficient recycle utilization. The COD and TN discharge of animal manure for HRB almost accounted for 17.00% and 39.00% of the whole COD and TN discharge in China. The farmland has been suffered animal manure pollution in HRB, particularly in Zhumadian, Zhoukou, Shangqiu, and Xuzhou regions. The diffusion concentrations of COD, BOD, $\text{NH}_4^+\text{-N}$, TN and TP from animal manure were very high in HRB. Especially, the concentrations of TN and TP from animal manure in Shangqiu, Zhoukou, Heze, Zhumadian, Luohe, Jining,

Xuchang, Kaifeng, Taian and Zhengzhou regions were far beyond WHO standard value, resulting in water eutrophication and cancer disease. Based on the quantitative risk evaluation and the present and future spatiotemporal variations by kriging interpolation method of ArcGIS technology, the results indicated that those regions in Zhumadian, Zhoukou, Shangqiu, Xuzhou, Heze, Jin- ing, Taian, Kaifeng, Xuchang, Luohe, Zhengzhou and Linyi were suffered a serious animal manure pollution risk on farmland or water quality from HRB in 2030. The results provide a quantitative animal manure risk and spatiotemporal variations of animal manure pollution in HRB that will help to guide public debate and policy development in maintaining sustainable live- stock production to improve water quality of HRB. The proposed risk evaluation combined with kriging inter- polation method of ArcGIS technology could be further considered application in other similar field research pollution of risk forecast.

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