

Application of modified export coefficient method on the load estimation of non-point source nitrogen and phosphorus pollution of soil and water loss in semiarid regions

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Abstract Chinese Loess Plateau is considered as one of the most serious soil loss regions in the world, its annual sediment output accounts for 90 % of the total sediment loads of the Yellow River, and most of the Loess Plateau has a very typical characteristic of “soil and water flow together”, and water flow in this area performs with a high sand content. Serious soil loss results in nitrogen and phosphorus loss of soil. Special processes of water and soil in the Loess Plateau lead to the loss mechanisms of water, sediment, nitrogen, and phosphorus are different from each other, which are greatly different from other areas of China. In this study, the modified export coefficient method considering the rainfall erosivity factor was proposed to simulate and evaluate non-point source (NPS) nitrogen and phosphorus loss load caused by soil and water loss in the Yanhe River basin of the hilly and gully area, Loess Plateau. The results indicate that (1) compared with the traditional export coefficient method, annual differences of NPS total nitrogen (TN) and total phosphorus (TP) load after considering the rainfall erosivity factor are obvious; it is more in line with the general law of NPS pollution formation in a

watershed, and it can reflect the annual variability of NPS pollution more accurately. (2) Under the traditional and modified conditions, annual changes of NPS TN and TP load in four counties (districts) took on the similar trends from 1999 to 2008; the load emission intensity not only is closely related to rainfall intensity but also to the regional distribution of land use and other pollution sources. (3) The output structure, source composition, and contribution rate of NPS pollution load under the modified method are basically the same with the traditional method. The average output structure of TN from land use and rural life is about 66.5 and 17.1 %, the TP is about 53.8 and 32.7 %; the maximum source composition of TN (59 %) is farmland; the maximum source composition of TP (38.1 %) is rural life; the maximum contribution rates of TN and TP in Baota district are 36.26 and 39.26 %, respectively. Results may provide data support for NPS pollution prevention and control in the loess hilly and gully region and also provide scientific reference for the protection of ecological environment of the Loess Plateau in northern Shaanxi.

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Introduction

Non-point source (NPS) pollution refers to the pollution from diffuse sources such as farmland runoff, rural life, decentralized livestock breeding, soil erosion, and urban runoff, etc. (Cheng et al. 2006; Li and Li 2010). It affects the receiving waters (rivers, lakes, reservoirs, oceans, and underground water, etc.) under the leaching and erosion action of rainfall runoff (Kang 2010). The generation of NPS pollution is caused by natural processes and strengthened by human activities (Fu and Kang 2012). Generally, the rainfall runoff

is the main natural reason of NPS pollution; the anthropogenic land use activity is the most fundamental reason of NPS pollution (Li et al. 2008). Nitrogen and phosphorus are nutrient factors for crop growth (Giles 2005); In addition to parts of the plain region, the nitrogen and phosphorus in most areas of the Loess Plateau are deficient (Jia et al. 1994); extensive use of fertilizers has become necessary means for increasing crop yields, but it also leads to serious water environment problems (Hart et al. 2004; Ongley et al. 2010; Sun et al. 2012). The average annual precipitation in the Loess Plateau is about 200–700 mm; the inter-annual variability of precipitation is large; the intra-annual precipitation is highly concentrated; the precipitation in 7–9 months accounts for about 60–70 % throughout the year (Li et al. 2010b); the amount of soil erosion and sediment transport in this seasons accounts for about 80 % throughout the year (Xu 1999); the spatial distribution is also very uneven; it basically presents a decreasing trend from the southeast to the northwest (Jiao et al. 1999). Most of the concentrated precipitation in the flood season is heavy rainfall; 85 % of the sediment throughout the year is from the heavy storms in the flood season (Ren 2006).

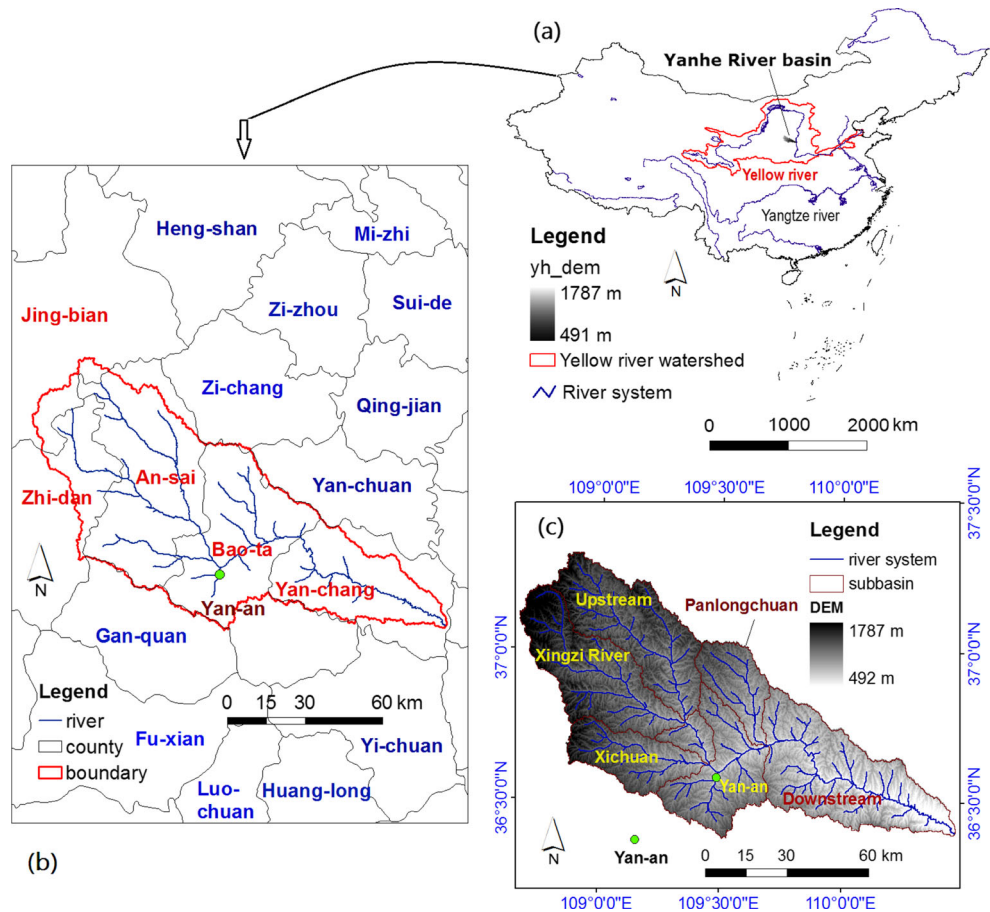
Agricultural production is an important source of NPS pollution in the Loess Plateau (Liu et al. 1995). Soil and nutrient loss in the Loess Plateau is mainly caused by few rainstorms (Zhang et al. 2004); nutrient loss accompanied by transient rainfall events plays a decisive role in annual total NPS pollution load (Austin et al. 2004). NPS pollution in this region shows seasonal regularities in time and geographical features in space (Cheng et al. 2006). Rainfall runoff is one of the main driving forces for the output of NPS pollution; soil and water loss is an important way of pollutant migration (Li 2009). Soil and water loss especially sediment loss is an important reason for soil nitrogen and phosphorus loss of the loess region (Shi 1997; Hu and MA 2008). Yanhe River is a primary tributary of the Yellow River; the River basin is located in the loess hilly and gully region with serious soil erosion, is one of the important sediment source areas of the Yellow River; serious soil and water loss caused loss of nitrogen and phosphorus in soil (Wang 2010). Therefore, the research of NPS nitrogen and phosphorus load in the Yanhe River watershed of the loess hilly and gully region is of great scientific significance for the protection of the ecological environment in Northern Shaanxi and regional economic development of the Loess plateau.

On river basin scale, models are the necessary means for the extension research of NPS nitrogen and phosphorus pollution load from multipoint monitoring to the whole river basin (Wu et al. 2010). Among them, the export coefficient model is widely used in the study of NPS pollution at large-scale basins due to small data demand and simple application (Li and Zhuang 2003; Huang et al. 2012; Zhang et al. 2014; Zhou et al. 2014). Agricultural land use has strong influence on nitrogen and phosphorus pollution load in river water (Bu et al. 2014). In the 70s of twentieth Century, the concept of

export coefficient method (Reckhow and Simpson 1980) was first proposed from the relationship between land use and eutrophication of lakes by Canada and USA; it is also known as the load method of unit area. This method provided a new way for the study of NPS pollution; its biggest advantage was to directly establish the relationship between land use and NPS pollution load of receiving water using land use data which is relatively easy to get (Long et al. 2008), but the early export coefficient model also exist some problems, such as land use classification was relatively simple and not further subdivided into various agricultural crop types; the pollutant output of various land use types showed a linear relationship with the land area, etc.. For the initial deficient of export coefficient method, Johnes (1996) comprehensively considered effects of crops, livestock and poultry species, population, and other factors based on land use types; to a large extent, it enriched the content of export coefficient model and improved the model sensitivity to land use changes. With the wide application of export coefficient model, many scholars have comprehensively taken into account the pollution sources closely related to classification of the export coefficient (Nandish and Keith 1996; Worrall and Burt 1999; Wickham et al. 2000) and also considered transport losses of pollutant and inter-annual changes of rainfall subsequently (Wickham and Wade 2002). Based on the export coefficient method, Wickham analyzed and evaluated NPS pollution output risk by stochastic simulation (Wickham and Wade 2002). Liu et al. (2009) estimated output risk of NPS pollution in Liaohe River basin from administrative divisions. Through the spatial and temporal comparison, Tian et al. (2011) analyzed NPS pollution output risk of Daning River watershed in the Three Gorges Reservoir area from administrative divisions and proved the relationship between the output risk and land use types.

Export coefficient method can estimate watershed pollution load output using land use information relatively easy to obtain (Cai et al. 2004); it greatly decreases the complex process of traditional NPS pollution research and largely reduces the cost of experimental monitoring and modeling; it provides a certain accuracy application for the estimation of NPS pollution load in large-scale watershed (Wu et al. 2012; Ke et al. 2014). Therefore, the modified export efficient model considering hydrological factors was presented to effectively simulate and predict the annual variability of pollution loadings in the Yanhe River watershed. Based on this, the objectives of this study are to (1) establish environmental database of the Yanhe River watershed for NPS pollution load estimation, (2) estimate NPS TN and total phosphorus (TP) loss load of the Yanhe River watershed in the loess hilly and gully region using the modified export coefficient method, (3) evaluate annual variations, spatial distributions, output structures, source compositions, and contribution rates of TN and TP in the Yan River watershed, and propose control strategies for erosion-type NPS pollution.

Fig. 1 The study area. **a** The relative location between the study area and the Yellow River/ Yellow River Basin, **b** the geographical location sketch of the Yanhe River watershed in the county-level administrative division of Shaanxi province, China, **c** longitude and latitude coordinates of the study area, digital elevation model (DEM) data, and the delineation of sub-basin and river systems within the Yanhe River watershed



Material and methods

Study area

The Yanhe River, which originates from the Baiyu Mountain of Jingbian county, is one of the main tributaries in the right shore of the Yellow River (Fig. 1a). The mainstream full length of Yanhe River is 286.9 km; it mainly includes tributaries of Xing-zi River, Xi-chuan River, Pan-longchuan River, and Nan-chuan River. Yanhe River flows through five counties including Jing-bian, Zhi-dan, An-sai, Baota, Yangchang from northwest to southeast in Shaanxi province, and finally, it empties into the Yellow River in Nanhe ditch, Yan-chang county (Fig. 1b). The Yanhe River watershed is located between 108°38' to 110°29' E longitude and 36°21' to 37°19' N latitude (Fig. 1c); it crosses three bioclimatic zones (forest, forest steppe, and steppe) and has criss-cross ravines and gullies, broken terrain, and low vegetation coverage; it is the most serious and typical soil erosion basin in the hilly and gully loess region of loess plateau (Zhao et al. 2008). The area of the watershed is 7725 km²; the climate of the watershed is sub-tropical monsoon type with an average annual

temperature of 8.8–10.2 °C; the average annual precipitation is about 495.6 mm and almost 60 % of the total annual

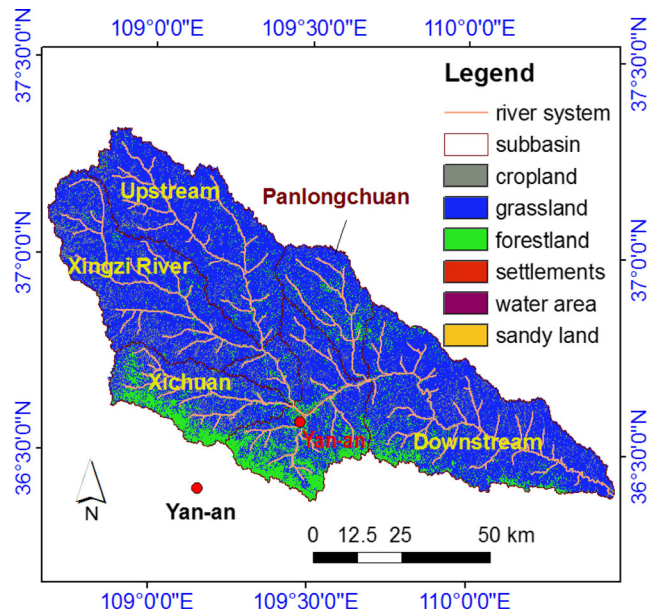


Fig. 2 Reclassified land use types in each sub-basin of the Yanhe River watershed

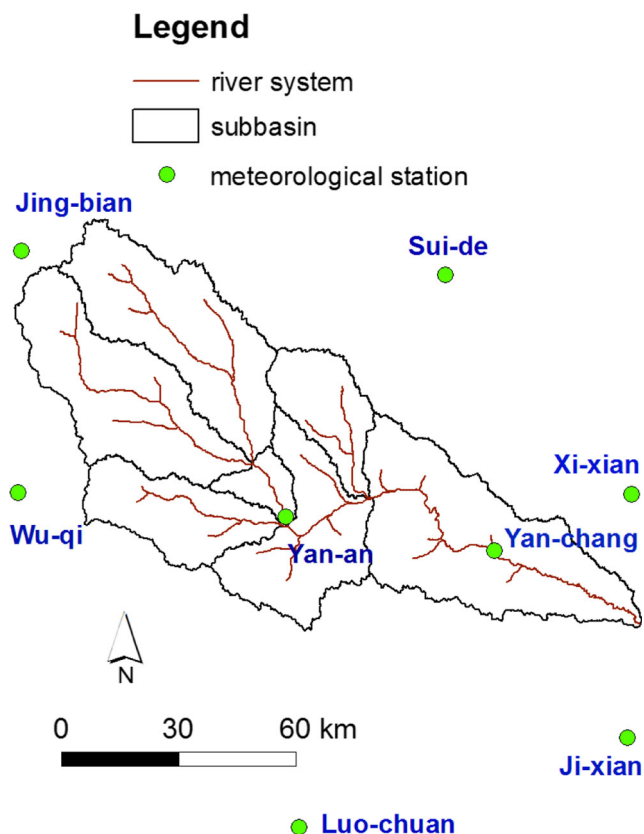


Fig. 3 Spatial distribution of meteorological stations and sub-basin division of the Yanhe River watershed

rainfall occurs during the monsoon period (July–September). The average annual runoff amount is 0.293 billion m^3 . The long-term average sediment transport capacity of the

Yanhe River watershed is 8.756 million tons, and the sediment transport modulus is $1.12 \times 10^4 t/(km^2 a)$. Soils in this basin mainly consist of alluvial soil, clay soil, and Heilu soil, where the most widely distributed soil is alluvial soil. Land use types in the study are reclassified as cropland, grassland, forest land, residential area, water area, and sandy land (Fig. 2). The daily meteorological data from 8 stations were used to generate surface rainfall data and estimate the spatiotemporal variations of rainfall erosivity in the Yanhe River watershed during the period 1957–2013. There are two stations in the watershed area and additional six stations adjacent to the area (Fig. 3).

Watershed environmental database

The parameters included in this study include digital elevation model (DEM), meteorology data (dew point temperature, relative humidity, precipitation, wind speed, sun hours), water quality, runoff, soil properties, land use types; socio-economic conditions include population, livestock and poultry breeding, fertilizer application, and administrative divisions (Table 1).

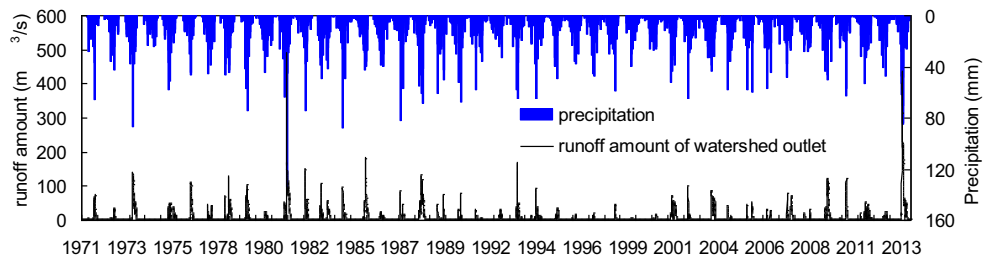
1. Rainfall runoff data

The semi-distributed land use-based runoff process (SLURP) hydrological model (Kite 2001, 2002) is selected to simulate the runoff data of the watershed outlet. The model is a basin model which takes day as a time step and simulates the hydrological cycle from precipitation to runoff including the effects of reservoirs, dams, regulators, water extractions, and irrigation schemes. The model uses

Table 1 Descriptions and sources of the environmental database for the Yanhe River watershed

Data layer	Format	Description	Source
DEM	Raster	30 m spatial resolution DEM data of the Yanhe River watershed	Computer Network Information Center, Chinese Academy of Sciences http://datamirror.csdb.cn/index.jsp
Land use	Raster	Farmland, grassland, forest land, residential area, water area, sandy land (30 m spatial resolution)	Data Center for Cold and Arid Region Sciences http://westdc.westgis.ac.cn/
Soil	DBF	TN and TP background content	National Science and Technology Infrastructure Center—Data Sharing Infrastructure of Earth System Science (http://loess.geodata.cn/)
Meteorological data	DBF	Daily values of precipitation, temperature, sun hours, wind speed, and relative humidity (1957–2013)	China Meteorological Data Sharing Service Network http://www.cdc.sciencedata.cn
Administrative division	VECTOR	Jingbian, Zhidan, Ansai, Baota, Yanchang	Data Sharing Network of Earth System Science http://www.geodata.cn/
Runoff	Excel	Time series of daily observed values of runoff amount in Ansai and Zaoyuan hydrological stations (1971–1980)	National Science and Technology Infrastructure Center—Data Sharing Infrastructure of Earth System Science (http://loess.geodata.cn/)
Economic conditions	DBF	Population, livestock and poultry breeding, fertilizer application (1990–2007)	Statistical yearbooks and reports of Shaanxi Province

Fig. 4 Rainfall and runoff values of the Yanhe River watershed outlet from 1971 to 2013



the topographic analysis package TOPAZ and SLURPAZ to derive and process the topographic and land cover inputs (Garbrecht and Campbell 1997; Lacroix et al. 2002). The model simulates the vertical water balance at each element of the sub-basin/land cover matrix and transforms discrete point meteorological data into surface meteorological data using spatial interpolation method (Linsley et al. 1975) using daily meteorological data which include precipitation, temperature, relative humidity, wind speed, and sun hours. Finally, runoffs from each matrix element are then routed through each sub-basin to the basin outlet taking account of reservoir regulation, diversions, groundwater extractions, and water exports from the basin.

In this study, the measured values of streamflow at Ansai and Zaoyuan hydrological stations from 1 January 1971 to 31 December 1975 were applied to calibrate hydrological model parameters. Furthermore, the optimization results of model parameters were verified by the measured runoff amount from 1 January 1976 to 31 December 1980; the Nash-Sutcliffe efficiency coefficient of average monthly runoff amount was 0.67 during the verification period; the maximum relative error was 23.6 %; it indicated that the SLURP hydrological model had good simulation results in the Yanhe River watershed. Therefore, the annual runoff values of the watershed outlet were simulated using the daily meteorological data from 1957 to

2013; the simulated runoff values from 1971 to 2013 were shown in Fig. 4.

2. Land use data

The land use information is also obtained by the SLURP hydrological model. Firstly, the DEM and land use/cover of the study area are pretreated by IDRISI GIS and Image Processing Software to generate the corresponding data file; secondly, the terrain analysis software TOPAZ was used to determine the basin boundary, generate river network, and extract the area information of various land use types. In this study, the land use types of the Yanhe River watershed were reclassified into farmland, grassland, forest land, residential area, water area, and sandy land. The area matrix statistics of different land use types in each sub-basin (Fig. 5) are shown in Table 2.

Besides, the main land use types of counties or districts in the Yanhe River watershed are comparatively analyzed by statistics in Fig. 6.

3. Agricultural population data

According to the statistical yearbook and report of Shaanxi Province, the permanent agricultural population data of the Yanhe River watershed from 1999 to 2008 were listed in Table 3 by statistics.

4. Livestock and poultry breeding statistics

According to the Shaanxi statistical yearbooks from 1999 to 2008, the livestock and poultry are mainly divided into four types including big livestock, pig, poultry, and sheep in this study. Specifically, livestock and poultry statistics of different districts or counties are shown in Table 4.



Fig. 5 Spatial distribution of 7 sub-basins within the Yanhe River watershed

Methodology

Export coefficient method is a kind of method to estimate watershed NPS pollution load by the export coefficient of pollutant (Yang and Xue 2009; Liao et al. 2014). Export coefficient model is the specific performance of the export coefficient method; it is a mathematical weighted equation to calculate the annual average pollution load by a semi-distributed pathway; its essence is a sort of semi-distributed lumped model (Ying et al. 2010). The traditional method directly established the relationship between land use types and NPS

pollution output load using the corresponding export coefficients; the basic expression for the method is:

$$L = \sum_{i=1}^m E_i A_i \tag{1}$$

where L is the total output of a certain kind of pollutant in various land use types, kg/a; m is the number of land use types; E_i is the pollutant export coefficient of the i -th land use type, kg/(ha•a); A_i is the area of the i -th land use type, ha.

Aiming at the deficiency of the initial export coefficient method, Johnes (1996) established a more perfect model for the export coefficient method in 1996. The basic equation of the model is:

$$L = \sum_{i=1}^n E_i [A_i(I_i)] + P \tag{2}$$

where L is the total loss load of nutrients; n is the number of the year; E_i is the export coefficient of the i -th nutrient source; A_i is the area of different land use types, or the number of livestock and poultry, or the number of population; I_i is the nutrient input for the i -th nutrient source; P is the nutrient (nitrogen and phosphorus) input from rainwater.

The above export coefficient models in consideration of hydrological factors are insufficient; it is mainly manifested in (1) the output coefficient of the same pollution source in different years uses the same value; (2) it is lack of consideration in the effect of rainfall and runoff processes which are directly related to the generation of NPS pollution; it only considers pollutants carried by rainwater (Li and Zhuang 2003). In order to predict watershed NPS pollution load for different years more accurately, Cai et al. (2004) improved Johnes’s export coefficient model, based on the relationship between annual NPS pollution load and annual rainfall; they presented the export coefficient model considering the effect

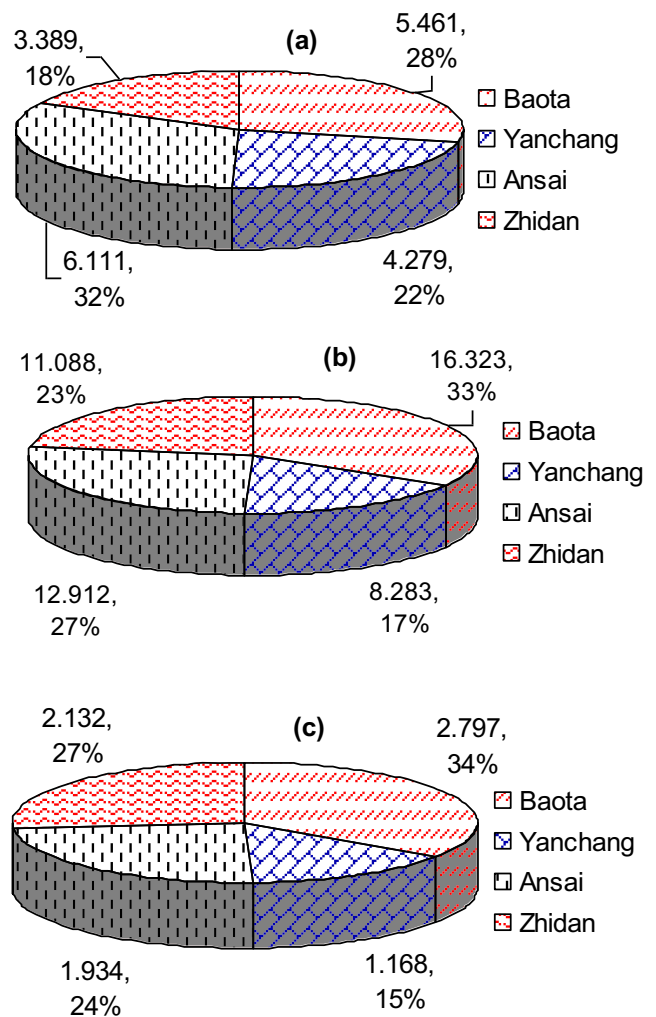


Fig. 6 The average area ratio (10⁴hm², %) of different land use types in each county from 1999 to 2008. **a** farmland, **b** grassland, **c** forestland

of rainfall. This study area is located in the hilly and gully region of the Loess Plateau; soil erosion in this region is serious, and annual soil and nutrient loss are closely related to annual rainfall erosivity; therefore, based on the existing research (Cai et al. 2004; Li and Mao 2008; Yang and Xue 2009;

Table 2 The area matrix statistics of sub-basins and different land use types in the Yanhe River watershed

Sub-basin	Area/km ²	Farmland	Grassland	Forest land	Residential area	Water area	Sandy land
1	1484	434.812	991.312	54.908	0	1.484	1.484
2	1511	350.552	1092.453	63.462	0	1.511	3.022
3	212.2	62.599	129.8664	18.2492	0.4244	1.061	0
4	804.2	238.0432	384.4076	180.945	0.8042	0	0
5	1038	224.208	543.912	260.538	8.304	1.038	0
6	587.1	116.2458	403.9248	66.9294	0	0	0
7	1974	497.448	1314.684	157.92	0	3.948	0
Total/km ²	7610.5	1923.908	4860.5598	802.9516	9.5326	9.042	4.506

Table 3 Agricultural population of each county or district in Yanhe River watershed (ten thousand)

Area	Year									
	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008
Baota	33.46	34.23	34.85	35.67	37.46	39.9	40.55	40.59	40.98	44.20
Yanchang	13.78	13.96	14.1	14.44	14.61	14.71	14.41	14.42	14.42	15.03
Ansai	15.02	15.11	15.19	15.31	15.51	16.16	16.56	16.57	16.62	17.10
Zhidan	11.75	11.86	11.97	12.01	12.79	13.03	13.44	13.45	13.54	14.54

Mao et al. 2012), the modified export coefficient method was established by means of the inter-annual variation factor of rainfall erosivity,

$$L = \mu \sum_{i=1}^n E_i [A_i(I_i)] + P \tag{3}$$

$$\mu = R_a / R \tag{4}$$

$$P = cA_p B_r \tag{5}$$

where μ is the inter-annual variation factor of rainfall erosivity, R_a represents the annual rainfall erosivity of the watershed, R is the average rainfall erosivity for many years, c is the

nutrient concentration in rainwater (g/m^3), A_p is annual rainfall amount (mm), and B_r is the runoff coefficient.

In this study, a half-month simple algorithm of rainfall erosivity established by Zhang et al. (2002) was used to estimate the annual rainfall erosivity values; the half-month algorithm of rainfall erosivity estimated by daily rainfall is as follows:

$$R_i = \alpha \sum_{j=1}^k (P_j)^\beta \tag{6}$$

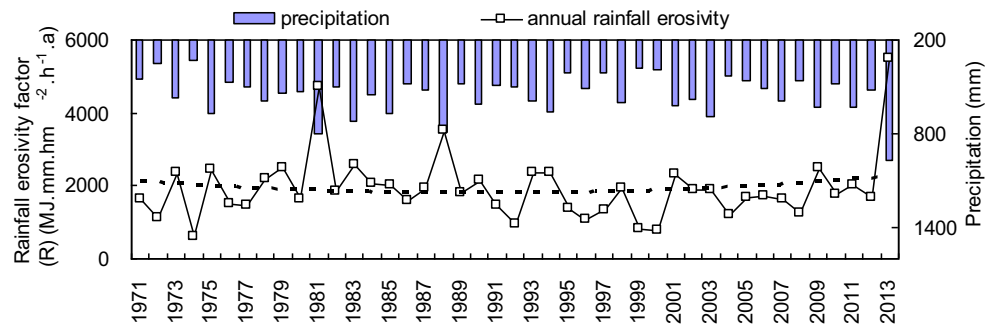
$$\beta = 0.8363 + \frac{18.144}{P_{d12}} + \frac{24.455}{P_{y12}} \tag{7}$$

$$\alpha = 21.586\beta^{-7.1891} \tag{8}$$

Table 4 Livestock and poultry statistics of different districts or counties in the Yanhe River watershed

Sorts	Counties or districts	Year									
		1999	2000	2001	2002	2003	2004	2005	2006	2007	2008
Big livestock (10^4 head)	Baota	4.32	4.28	4.20	3.42	2.77	3.52	3.65	3.68	3.71	1.72
	Yangchang	3.10	2.86	2.11	2.04	2.00	2.01	2.02	2.02	1.98	1.40
	Ansai	2.52	2.34	2.18	2.11	2.09	1.92	1.70	1.69	1.83	1.26
	Zhidan	4.95	5.01	5.09	5.14	5.81	8.52	7.61	7.83	7.98	2.72
Pig (10^4 head)	Baota	8.76	7.88	7.17	7.09	6.60	6.90	6.45	6.42	3.22	3.43
	Yangchang	2.40	2.32	1.87	1.66	1.45	1.40	1.39	1.39	0.71	1.02
	Ansai	2.98	3.03	3.87	4.58	5.22	5.40	4.89	4.89	2.45	2.63
	Zhidan	4.22	4.25	4.72	4.99	5.13	6.28	6.28	6.91	2.15	2.60
Poultry (10^4 poultry unit)	Baota	55.01	54.85	49.73	50.23	48.60	53.32	55.70	53.22	37.44	41.10
	Yangchang	13.88	12.90	12.58	10.95	11.23	13.84	14.20	14.20	10.93	12.20
	Ansai	20.00	19.70	18.99	19.87	18.95	19.80	20.83	21.50	18.38	20.00
	Zhidan	18.32	17.65	15.78	16.76	15.43	17.89	22.45	25.52	14.92	17.20
Sheep (10^4 sheep unit)	Baota	5.05	8.65	10.09	9.87	8.21	2.08	1.35	1.43	1.26	1.30
	Yangchang	3.03	3.55	4.18	8.21	9.95	8.01	5.57	6.02	2.86	3.39
	Ansai	2.31	3.09	4.74	8.93	16.81	12.14	2.24	3.35	1.87	1.89
	Zhidan	15.27	17.64	18.76	18.08	18.10	18.20	18.21	18.23	5.73	6.71

Fig. 7 Comparison of annual precipitation and annual rainfall erosivity from 1971 to 2013 in the Yanhe River watershed



where R_i represents the rainfall erosivity during i -th half-month period ($\text{MJ}\cdot\text{mm}\cdot\text{hm}^{-2}\cdot\text{h}^{-1}\cdot\text{a}$), k represents the number of days within the half-month period, P_j is the rainfall amount in the j -th day during the half-month period; the requirement of daily rainfall is greater than or equal to 12 mm; otherwise, it is calculated by 0; 12 mm is just correspondence with the erosive rainfall standard (Liu et al. 2005), α and β are parameters, P_{d12} represents the average daily rainfall when the daily rainfall is greater than 12 mm, P_{y12} represents the average annual rainfall when the daily rainfall is greater than 12 mm. In this method, half-month is determined as the calculation period; the half-month algorithm was established using daily rainfall; the division standard of half-month period is that the first 15 days of each month are used as the former half-month period, and the remaining part of the month as another half-month period (Zhang et al. 2003). The estimated results of annual rainfall erosivity are shown in Fig. 7.

Determination of the export coefficient

The application key of export coefficient model is to determine the export coefficients reasonably (Li et al. 2009; Ying et al. 2010; Lu et al. 2012). TN and TP were considered as the main pollutant types of the Yanhe River watershed; three kinds of nitrogen and phosphorus pollution sources include land use, rural residential areas, and livestock and poultry breeding. In this study, the farmland, forest land, and grassland are used as the main land use sources to choose the export coefficients; the human excrement and sewage from rural residential areas are used as the main rural sources; the amounts of big livestock, pig, sheep, and poultry are used as the main livestock and poultry breeding sources to determine the export coefficients.

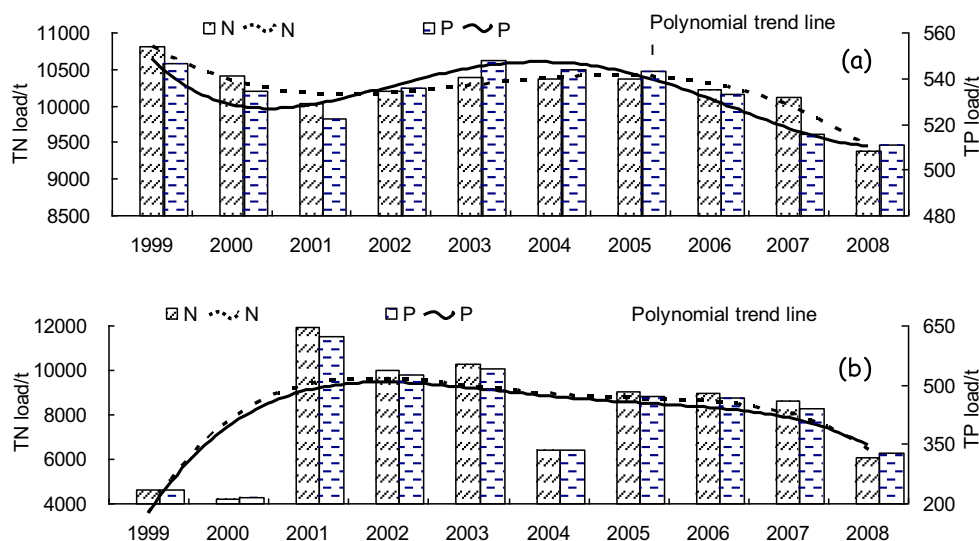
For the export coefficients, a lot of research results have been reported in China (Liu et al. 2011; Zhao et al. 2012; Ma et al. 2012). For example, Li et al. (2004a, b; 2006a, b) monitored and determined the average export coefficient of polluted surface runoff from agricultural land use types (forest land and cultivated land) and urban lands (commercial area, residential area, roads, suburban, and rural areas) in the Taihu River Basin; Li et al. (2007) established the quantitative relationship between area of different land use types and nutrient concentrations through the water quality monitoring of 11 typical sub-watershed outlets in Xitiaoqi small watershed of Taihu region and then calculated the TN and TP export coefficients of cropland and forestland. Ding et al. (2006) estimated the export coefficients of each nutrient source by means of the historical hydrology and water quality data and the principle of mass conservation. In addition, Liang et al. (2005) and Li et al. (2008) determined the export coefficients of NPS pollution in different land use types by the method of field-scale monitoring and rainfall simulation, respectively.

Considering land use types and underlying surface conditions of the Yan River watershed, the suitable export coefficient of the Yanhe River watershed were determined as follows. The farmland export coefficients of TN and TP are 29 and $0.9\text{ kg}/\text{hm}^2\cdot\text{a}$, respectively (Li et al. 2007); the approximate forestland export coefficients of TN and TP were selected as 3.27 and $0.10\text{ kg}/\text{hm}^2\cdot\text{a}$ which can represent the entire forestland region of Shaanxi province (Wang 2004); the optimum export coefficients of TN and TP for grassland are 1.578 and $0.20\text{ kg}/\text{hm}^2\cdot\text{a}$, respectively (Li et al. 2010a). The TN export coefficients of excreta from big livestock, pig, and

Table 5 Classification and values for export coefficients of the Yanhe River watershed

Type	Land use/ $\text{kg}\cdot(\text{hm}^2\cdot\text{a})^{-1}$			Livestock and poultry / $\text{kg}\cdot(\text{ca}\cdot\text{a})^{-1}$				Population/ $\text{kg}\cdot(\text{ca}\cdot\text{a})^{-1}$
	Farmland	Grassland	Forest land	Big livestock	Pig	Sheep	Poultry	
TN	29	1.578	3.27	10.21	0.74	0.40	0.04	2.14
TP	0.9	0.20	0.10	0.2179	0.1417	0.045	0.0054	0.2142

Fig. 8 Inter-annual changes of TN and TP load in the Yanhe River watershed from 1999 to 2008: **a** the traditional export coefficient method, **b** the modified export coefficient method



sheep are 10.21 kg/ca•a, 0.74 kg/ca•a, and 0.40 kg/ca•a, respectively; The TP export coefficients of excreta from big livestock, pig, and sheep are 0.2179 kg/ca•a, 0.1417 kg/ca•a, and 0.045 kg/ca•a, respectively (Bian 2000); the poultry export coefficients of TN and TP are 0.04 and 0.0054 kg/ca•a, respectively; the TN export coefficient of human feces is 2.14 kg/ca•a; and the TP export coefficient of human feces is 0.2142 kg/ca•a (Liu et al. 2008).

Through the above literature review, the TN and TP export coefficients of different land use types, agricultural population, livestock and poultry breeding in the Yanhe River watershed are shown in Table 5.

Results and discussion

Based on the traditional and modified export coefficient models, the NPS TN and TP pollution load in the Yanhe River watershed were estimated from 1999 to 2008. The pollution characteristics were comparatively analyzed from 5 aspects including the annual variation, spatial distribution, output structure, source composition, and contribution rate, respectively.

The annual changes of NPS pollution load

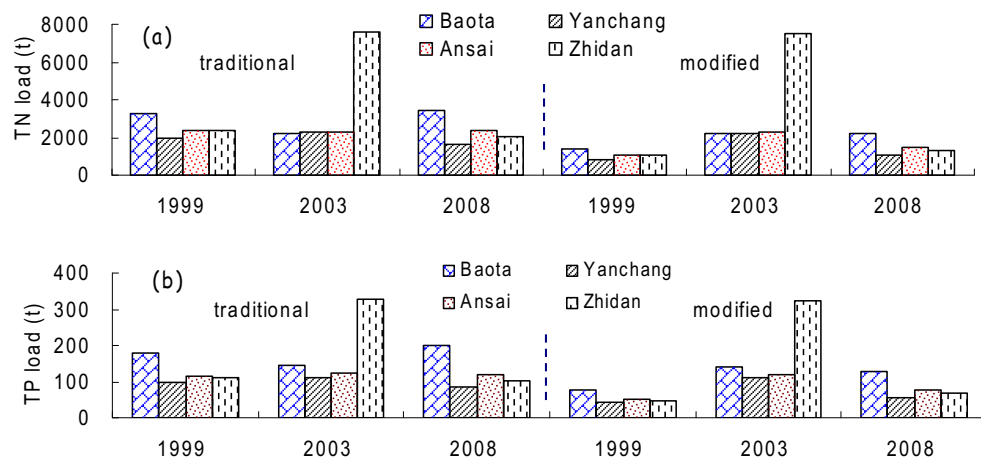
Figure 8a indicates that the changing trends of NPS TN and TP pollution from 1999 to 2008 in Yan River watershed show a trend of first decline, second rising, and then decline in overall. TN output decreased from 10804.89 t in 1999 to 10041.53 t in 2001, then rose to 10196.44 t in 2003, and then dropped to 9390.92 t in 2008; TP output decreased from 546.86 t in 1999 to 522.32 t in 2001, then rose to 548.19 t in 2003, and then dropped to 511.05 t in 2008. In short, annual changes have slight fluctuations under the traditional export coefficient method; the main reason can be attributed to the lack of consideration of the hydrological factors.

Figure 8b shows the changing trends of NPS TN and TP pollution from 1999 to 2008 under the modified export coefficient method; the annual changes of NPS pollution load have closely linear correlation with annual rainfall erosivity. Because the rainfall erosivity in 2001 was significantly higher than the average for many years, the output loads of TN and TP were also greater than other years, and the sudden decrease in 2004 was particularly related to the less rainfall amount and rainfall intensity. Under the modified export coefficient method, TN output increased from 4641.83 t in 1999 to 11956.41 t in 2001, then dropped to 6034.96 t in 2008; TP output also

Table 6 Variations of NPS TN and TP emission intensity in the Yanhe River watershed from 1999 to 2008 (brackets for the modified method)

County or district	TN emission intensity (t)			TP emission intensity (t)		
	1999	2003	2008	1999	2003	2008
Baota	3238.8 (1391.4)	2219.5 (2189.9)	3422.7 (2199.5)	179.5 (77.1)	144.2 (142.3)	198.3 (127.4)
Yanchang	1973.6 (847.8)	2253.5 (2223.6)	1621.1 (1041.8)	96.1 (41.3)	111.6 (110.1)	85.1 (54.7)
Ansai	2401.4 (1031.6)	2314.3 (2283.5)	2330.1 (1497.4)	116.9 (50.3)	121.5 (119.9)	118.7 (76.3)
Zhidan	2380.7 (1022.7)	7592.3 (7491.3)	2065.5 (1327.4)	112.2 (48.2)	326.7 (322.3)	102.9 (66.2)

Fig. 9 The output load changing trends of 1999, 2003, and 2008 in the Yanhe River watershed (the left side of the dotted line in the figure is the traditional method, the right side is the modified method): **a** TN load, **b** TP load



increased from 234.93 t in 1999 to 621.92 t in 2001, then dropped to 328.42 t in 2008. Therefore, the hydrological factor is the main reason for the inter-annual variations of pollution load. Besides, the increase of source strength about TN and TP pollution in the watershed was also an important reason for the increase of TN and TP pollution load; for instance, a sudden increase of pollution load may be also related with the increase of chemical fertilizer application and the expansion of livestock and poultry breeding.

The spatial distribution of NPS pollution load

As a result of the broadly similar distribution in each year, this study only listed the years of 1999, 2003, and 2008 in Table 6 and Fig. 9. Under the traditional and modified conditions, Table 6 and Fig. 9 both indicate that annual changes of TN and TP emission intensity in four counties or districts took on the similar trends from 1999 to 2008; TN and TP emissions in Baota district and Ansai county both show a trend of first decrease and then increase; TN and TP emissions in

Yanchang and Zhidan counties both present a trend of first increase and then decrease. There are also differences in TN and TP emission intensity of each county during the 10 years: under the traditional method, the maximum change of nitrogen emission intensity is Zhidan county, nitrogen emissions reduced by 5526.8 t, the minimum change is Ansai county, nitrogen emissions increased by 15.88 t; the maximum change of phosphorus emission intensity is Zhidan county, phosphorus emissions decreased by 223.7 t, the minimum change is Ansai county, phosphorus emissions decreased by 2.8 t; under the modified method, the maximum change of nitrogen emission intensity is Zhidan county, nitrogen emissions reduced by 6468.57 t, the minimum change is Baota district, nitrogen emissions increased by 9.61 t; the maximum change of phosphorus emission intensity is Zhidan county, phosphorus emissions decreased by 256.16 t, the minimum change is Baota district, phosphorus emissions decreased by 14.88 t. The maximum and minimum load values both occurred between 2003 and 2008. The results indicate that the load emission intensity not only has great relationship with rainfall intensity but also

Table 7 Output structures of NPS pollution load for three different pollution sources under the modified export coefficient method

Year	TN (tons and %)			TP (tons and %)		
	Land use	Livestock and poultry breeding	Rural life	Land use	Livestock and poultry breeding	Rural life
1999	3187.4 (68.67 %)	774 (16.67 %)	680.4 (14.66 %)	134.3 (57.15 %)	32.6 (13.86 %)	68.1 (28.99 %)
2000	2817 (67.41 %)	714.8 (17.10 %)	647.3 (15.49 %)	118.7 (55.38 %)	30.8 (14.38 %)	64.8 (30.24 %)
2001	7984.7 (66.78 %)	2032.4 (17.00 %)	1939.3 (16.22 %)	336.3 (54.08 %)	91.5 (14.71 %)	194.1 (31.21 %)
2002	6738.5 (67.40 %)	1620.6 (16.21 %)	1639 (16.39 %)	283.9 (54.01 %)	77.7 (14.78 %)	164.1 (31.21 %)
2003	6903.7 (67.38 %)	1645.5 (16.06 %)	1697 (16.56 %)	290.8 (53.77 %)	80.2 (14.83 %)	169.9 (31.40 %)
2004	4084.8 (63.57 %)	1228.9 (19.13 %)	1111.9 (17.30 %)	172.1 (51.02 %)	53.9 (15.99 %)	111.3 (33.0 %)
2005	5849.3 (64.85 %)	1588.5 (17.61 %)	1582.6 (17.54 %)	246.4 (52.12 %)	67.9 (14.36 %)	158.4 (33.51 %)
2006	5819.8 (64.66 %)	1577.8 (17.53 %)	1602.4 (17.80 %)	245.1 (52.24 %)	63.8 (13.59 %)	160.4 (34.18 %)
2007	5595.4 (64.87 %)	1469.9 (17.04 %)	1560 (18.09 %)	235.7 (53.67 %)	47.3 (10.78 %)	156.1 (35.55 %)
2008	4216 (69.86 %)	569.3 (9.43 %)	1249.7 (20.71 %)	177.6 (54.08 %)	25.7 (7.84 %)	125.1 (38.09 %)

Table 8 Comparison of traditional and modified NPS TN and TP loads from different sources in the Yanhe River watershed in 2008

Pollution source classification	TN/t		TP/t	
	Traditional	Modified	Traditional	Modified
Forestland (79,700 hm ²)	260.64	167.50	7.97	5.12
Farmland (191,000 hm ²)	5538.39	3559.19	171.88	110.46
Grassland (482,500 hm ²)	761.37	489.29	96.50	62.01
Big livestock (7.1 × 10 ⁴ head)	724.91	465.86	15.47	9.94
Pig (9.68 × 10 ⁴ head)	71.63	46.03	13.72	8.81
Sheep (13.29 × 10 ⁴ sheep unit)	53.16	34.16	5.98	3.84
Poultry (90.5 × 10 ⁴ poultry unit)	36.20	23.26	4.89	3.14
Population (90.87 × 10 ⁴ people)	1944.62	1249.69	194.64	125.09

with the regional distribution of land use and other pollution sources.

As far as the year of 2008 is concerned, under the traditional method, the nitrogen emission intensity of Baota District within the Yanhe River watershed reaches the maximum value of 3422.66 t; it accounted for 36.26 % of total emissions; Yanchang county is the minimum value of 1621.13 t; it accounted for 17.17 % of total emissions; the phosphorus emission intensity of Baota District also reaches the maximum value of 198.30 t; it accounted for 39.26 % of total emissions; Yanchang county is still the minimum value of 85.12 t; it accounted for 16.85 % of total emissions. Although the nitrogen emission intensity under the modified method changed a lot, the proportion in each year stayed largely the same. The results indicated that the spatial distribution of NPS pollution load was mainly related to the distribution of farmland, livestock and poultry breeding, agricultural population density, and the situation of local soil loss.

The output structure of NPS pollution load

Compared with the traditional export coefficient model, the corresponding output structure proportion based on the improved model changes little, the annual variations of the

output load are obvious. The output estimation results of TN and TP by the modified model are shown in Table 7. Table 7 demonstrates that land use and rural life are the main TN and TP pollution source of the Yanhe River watershed; the livestock and poultry breeding is also an important pollution source for the watershed. It also could be seen that although the annual variations of NPS pollution load were large, the output structure of all sources was relatively stable in 1 year.

Firstly, land use is the main pollution source of nitrogen within the watershed; the proportion of nitrogen from land use between 1999 and 2003 was relatively large and accounted for about 67.53 % in average; the proportion from 2004 to 2007 was relatively small with around 64.49 % in average; and the proportion increased in 2008 with a value of 69.86 %. Secondly, land use is also the main source of phosphorus output within the watershed; the ratio of phosphorus from land use decreased from 57.15 % in 1999 to 51.02 % in 2004, then increased to 54.08 % in 2008. Thirdly, the rural life is the important source of phosphorus output within the watershed; the ratio of phosphorus from rural life increased from 28.99 % in 1999 to 38.09 % in 2008.

As far as the average case was concerned, the output structure ratio of TN in descending order was land use (66.5 %), rural life (17.1 %), and livestock and poultry (16.4 %); while TP was land use (53.8 %), rural life (32.7 %), and livestock and poultry (13.5 %). The composition ratio indicated that the NPS load was mostly from land use, where TN pollution loads of land use accounted for about 2/3, and TP pollution loads of land use roughly accounted for more than 1/2; rural life was also an important pollution source (1/3) for TP load. Thus, strengthening soil and water conservation in different land use types and lowering the pollutants discharge from rural settlements was the major measures to control the generation of NPS pollution.

The source composition of NPS pollution load

Table 8 and Fig. 10 show the NPS pollution load and its source composition based on the traditional and modified methods in 2008, respectively. Table 8 indicates that annual changes of the output load by the modified method are obvious compared with the traditional method, but the corresponding source

Fig. 10 Source compositions of TN and TP for different pollution sources in 2008

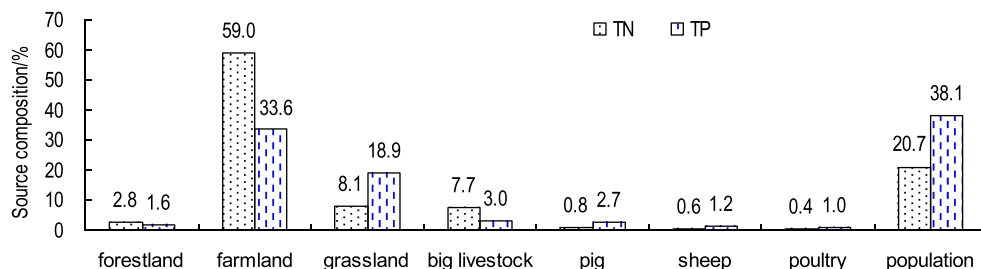
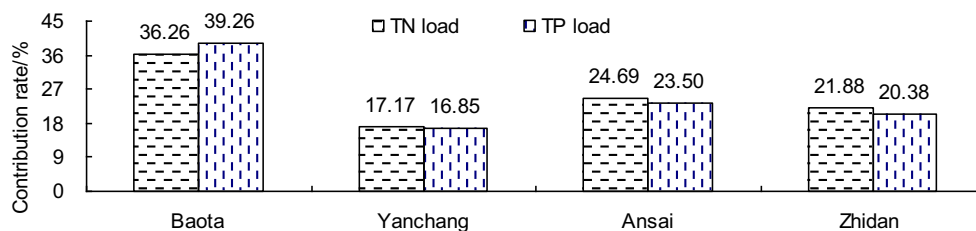


Fig. 11 TN and TP contribution rates of each county in the Yanhe River watershed



composition based on the modified model changes little. Figure 10 shows that NPS TN and TP pollution sources in the Yanhe River watershed include farmland, grassland, big livestock, poultry, pig, sheep, and agricultural population, where nitrogen emission from farmland is the largest and accounts for 59 % of the total emissions, and the rural life contributes to the largest phosphorus emission and accounts for 38.1 % of the total emissions; TN and TP emissions from grassland, big livestock, and forestland are also relatively large. So, farmland, rural life, and big livestock breeding in this watershed are the main NPS pollution sources. Therefore, the NPS pollution control in the Yan River watershed should take small watershed as a centralized unit and implement the comprehensive management policy of rational fertilization, water and soil conservation tillage, and rural sewage treatment to abate total pollution load.

The contribution rates of NPS pollution load in each administrative district

Compared with the traditional export coefficient model, the contribution rates of NPS pollution load in each administrative district have no obvious change. Taking the year of 2008 as an example, Fig. 11 shows that the largest contribution rates of TN and TP pollution in the Yanhe River watershed both appeared in the Baota district; the TN and TP contribution rates in this area were 36.26 and 39.26 %, respectively. The contribution rates of NPS TN pollution load in descending order were as follows: Baota district (36.26 %), Ansai county (24.69 %), Zhidan county (21.88 %), and Yanchang county (17.17 %). Furthermore, the contribution rates of NPS TP pollution load were as follows: Baota district (39.26 %), Ansai county (23.5 %), Zhidan county (20.38 %), and Yanchang county (16.85 %). These data indicate that (1) TN and TP pollution are mainly from the Baota district; other three counties also play an important role in the pollution contribution; (2) the contribution rate of NPS load in each administrative district is not only correlated to the distribution of different land use types but also associated with the number of population and the amount of livestock and poultry breeding within the Yanhe River watershed.

Conclusions

In this study, the modified export coefficient method was proposed to simulate and evaluate NPS TN and TP pollution load caused by soil and water loss in the Yanhe River watershed of loess hilly and gully area, Chinese Loess Plateau. Based on the results of the study, the following major conclusions were drawn:

1. Annual changes of NPS TN and TP load from 1999 to 2008 in the Yanhe River watershed have slight fluctuations under the traditional export coefficient method; annual changes of NPS TN and TP pollution load have closely linear correlation with annual rainfall erosivity under the modified export coefficient method. Therefore, after considering the rainfall erosivity factor, the annual differences of NPS load become obvious; this is more in line with the general law of NPS pollution formation in a watershed; it can reflect the annual variability of NPS pollution more accurately.
2. Under the traditional and modified conditions, annual changes of TN and TP emission intensity in four counties or districts took on the similar trends from 1999 to 2008. Under the traditional and modified methods, the maximum changes of nitrogen emission intensity are both Zhidan county, nitrogen emissions reduced by 5526.8 and 6468.57 t, respectively; the minimum change is Ansai county and Baota district; nitrogen emissions increased by 15.88 and 9.61 t, respectively; the maximum change of phosphorus emission intensity are both Zhidan county; phosphorus emissions decreased by 223.7 and 256.16 t; the minimum change is Ansai county and Baota district; phosphorus emissions decreased by 2.8 and 14.88 t, respectively; the results indicate that the load emission intensity not only has great relationship with rainfall intensity but also with the regional distribution of land use and other pollution sources.
3. The output structure, source composition, and contribution rate of NPS pollution load under the modified method are basically the same with the traditional method. For the output structure, the pollutions from land use and rural life are the main sources of NPS TN and TP pollution in the Yan River watershed, where the average nitrogen output is about 66.5 and 17.1 %, and the average phosphorus

output is about 53.8 and 32.7 %. For the source composition, the maximum output of nitrogen load is farmland; it accounts for 59 %; the maximum output of phosphorus load is rural life; it accounts for 38.1 %. For the contribution rate, the TN and TP load in Baota district are both the biggest; the contribution rates of TN and TP load are 36.26 and 39.26 %, respectively.

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