Spatial evaluation of complex non-point source pollution in urban–rural watershed using fuzzy system

Haijun Wang, Wenting Zhang, Song Hong, Yanhua Zhuang, Hongyan Lin and Zhen Wang

ABSTRACT

Non-point source (NPS) pollution has become the major reason for water quality deterioration. Due to the differences in the generation and transportation mechanisms between urban areas and rural areas, different models are needed in rural and urban places. Since land use has been rapidly changing, it is difficult to define the study area as city or country absolutely and the complex NPS pollution in these urban–rural mixed places are difficult to evaluate using an urban or rural model. To address this issue, a fuzzy system-based approach of modeling complex NPS pollutant is proposed concerning the fuzziness of each land use and the ratio of belonging to an urban or rural place. The characteristic of land use, impact of city center and traffic condition were used to describe spatial membership of belonging to an urban or rural place. According to the spatial membership of belonging to an urban or rural place, the NPS distributions calculated by the urban model and rural model respectively were combined. To validate the method, Donghu Lake, which is undergoing rapid urbanization, was selected as the case study area. The results showed that the urban NPS pollutant load was significantly higher than that of the rural area. The land usage influenced the pollution more than other factors such as slope or precipitation. It also suggested that the impact of the urbanization process on water quality is noteworthy.

Key words | Donghu Lake, fuzzy system, non-point source pollution, urban–rural watershed

Haijun Wang
Song Hong (corresponding author)
Yanhua Zhuang
Hongyan Lin
Zhen Wang
School of Resource and Environmental Science,
Wuhan University,
Wuhan,
China
E-mail: *environmentalanalytics@gmail.com*

Wenting Zhang
Department of Geography and Resource Management,
The Chinese University of Hong Kong,
Hong Kong

INTRODUCTION

Excessive loads of pollution into rivers, lakes, reservoirs and estuaries are now becoming a major concern to water resource managers across the world (Shrestha *et al.* 2008; Jing & Chen 2011; Liu & Tong 2011). Non-point source (NPS) pollution significantly contributes to the deterioration of water quality (Leone *et al.* 2009; Ouyang *et al.* 2010) due to the difficulty in identifying, assessing and controlling the sources of this type of pollution. The major NPS pollutants are nitrogen (N) and phosphorus (P). Recently, numerous research efforts have been made to discover the process and spatial quality of NPS N and P pollutants to support prevention and mitigation measures (Zhang & Huang 2011). In particular, the NPS pollution can be classified into two types: agricultural/rural NPS and urban NPS (Edwin *et al.* 2010). As the generation and transportation process of NPS pollutants in urban and rural places are different, the models and the factors as well as the corresponding parameters must be different (William *et al.* 1985; Kim *et al.* 1993; Zhang *et al.* 2006; Phillips *et al.* 2007) to ensure accurate results. To begin with, close attention was paid to rural NPS pollution, as agricultural chemicals contributed to the NPS pollution a great deal. For example, the empirical quantitative approach, namely, the universal soil loss equation (USLE), is developed to predict large scale soil erosion and the designation of potential risk zones for agricultural plots (Pandey *et al.* 2007). Thanks to its low data and parameter requirements, in contrast to physically-based models, as well as its scale-independent geometric resolution (Renard *et al.* 1997; Bahadur 2009; Dumas *et al.* 2010; Volk *et al.* 2010), USLE is widely used in the evaluation of

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the rural NPS pollutant loads by providing average annual soil erosion (Fistikoglu & Harmancioglu 2002; Haregeweyn & Yohannes 2003). Additionally, the other rural NPS evaluation model, the export coefficient model (ECM), is well-developed in determining NPS pollution (Do *et al.* 2011) with the simple model format for agricultural areas (Johnes & Heathwaite 1997) at the same time. In short, the model for rural NPS evaluation is fully developed and widely applied. Yet, as land use is changing from agricultural to urban, the natural soil surface is replaced with impermeable surfaces (Chris *et al.* 2004) which suffer from higher population density and more intensive human activities (Shon *et al.* 2012). This will influence the generation and transportation process of NPS pollutants. Recently, due to the wide process of urbanization all over the world, the urban NPS pollution research has became more popular. For instance, Shon *et al.* (2012) used a storm water management model to estimate the NPS pollutant loads; Bhaduri *et al.* (2000) proposed a Geographical Information System (GIS)–NPS model to assess the NPS pollutant loads under urbanization by using the Long-Term Hydrologic Impact Assessment (L-THIA) model.

Even if the NPS evaluation models for urban or rural areas are well-developed, all these models concentrate on one aspect, urban or rural pollutant loads, which is insufficient to evaluate the NPS pollutant loads in the urban-rural mixed areas. In this study, we identify the NPS pollution in urban–rural mixed areas and caused by various pollutants in rural and urban surface runoff together as the complex NPS pollution. Since more and more urban–rural mixed areas are emerging, as the result of rapid economic development, some efforts have concentrated on the NPS pollution in mixed urban and rural watershed and have mentioned that the process of urbanization impacted the water quality greatly (Wang *et al.* 2008; Zheng *et al.* 2011). For example, Chris *et al.* (2004) measured the water quality in agricultural, urban and mixed land, and determined the water quality from these three places, but Chris *et al.* just compared the measured samples to confirm that the water quality varied in different areas and did not address the problem of how to evaluate the NPS pollutant loads in different areas. Shields *et al.* (2008) pointed out that the urbanizing study area is different from the traditional urban or rural catchment. Thus, Shields *et al.* employed the Baltimore Ecosystem Study method to explore the impact of urbanization on the magnitude and export flow distribution of nitrogen, concluding that they are highly correlated. However, their studies still did not separate the rural region from the urban region for their study area and the problem of modeling complex NPS pollution at urban–rural mixed place has not yet been solved.

To address the problem of evaluating the NPS pollutant loads in an urban–rural mixed area, Zhuang *et al.* (2013) proposed a CA-AUNPS model to asses the spatial and temporal variation of complex NPS pollution for a lake watershed of central China. In this model, Focal Neighborhood method was used as the coupling model to combine the export empirical model and L-THIA model. In our study, a fuzzy membership-based approach is proposed. To our knowledge, it is difficult to classify an area into urban or rural absolutely due to the fact that the multiple or fuzzy characteristics of non-urban, partly-urban and urban states in the process of urban development are not solved (Liu & Phinn 2003). Conventionally, the land use can be classified into '0' meaning non-urban or rural and '1' meaning urban. According to this classification, urban land which is surrounded by rural land and the land use at the boundary of rural-urban areas may be misclassified and then result in mistakes in the evaluation of the NPS pollutant loads. The fuzzy membership can express the ratio of the land cell belonging to urban or rural, which ranges from 0 to 1. The fuzzy expression may be suitable for use in the NPS evaluation and have been employed to assist in the calculation of water quality (Yang *et al.* 2012). For example, Dixon (2005) incorporated GIS, global position system, remote sensing (RS) and the fuzzy rule-based model to generate groundwater sensitivity maps. Besides the traditional calculation for groundwater quality, his methodology was further refined through fuzzy rule-based model to incorporate land-use/pesticide application and soil structure information. Gemitzi *et al.* (2006) combined GIS with fuzzy logic and multi-criteria evaluation techniques for data acquisition and the production of factor images. Then, he created the intermediate and final ground water vulnerability map based on factor images. In accordance with previous studies and the aims of this study, the fuzzy membership-based approach is employed to describe the fuzziness in land usage and then to express the complex NPS pollutants in rural and urban mixed areas.

In this paper, the universal soil loss equation model (Wischmeier & Smith 1978), the export coefficient model and the long-term hydrologic impact assessment model (Harbor 1994; Lim *et al.* 2006), are employed to evaluate the spatial distribution and quality of rural and urban NPS pollutant loads respectively. In particular, the USLE model and the ECM are integrated to calculate the NPS pollutant loads with the hypothesis that the study area is rural, yet the L-THIA model is used to achieve the urban NPS pollutant loads.

Generally, this study aims to develop an integrated method to assess complex NPS pollution under the process of urbanization, which accounts for the fuzziness of the real world. It involves the following objectives: (1) calculating rural and urban NPS pollutant loads by using well-developed NPS evaluation models, USLE, ECM and L-THIA models respectively; (2) classifying the study area into rural and urban by fuzzy membership function of the characteristic of land use, impact of city center and traffic condition; (3) combining the results of rural and urban pollutant loads calculating models, according to the fuzzy membership; and (4) carrying out the case study of a rapid developing watershed, Donghu watershed in central China, to confirm the proposed methods.

STUDY AREA

The study area (Figure 1, 114°18′~114°30′ E, 30°30′~30°38′ N, 18,075 ha.), the Donghu watershed, is located in the eastern portion of the city of Wuhan (Gao *et al.* 2009). The study site is one of the largest downtown lakes in China. In addition to the general functions of a lake, such as regulating climate, degrading pollution, providing living space for aquatic life and preventing flooding, the Donghu watershed has a significant impact on the ecological environmental safety of Wuhan. Due to the radial effects of urban centers, the impact of intense anthropogenic activities and urbanization on the watershed water quality is profound.

The land use classification is extracted from the LANDSAT TM images in 1991, 2002 and 2005 by the ERDAS software package, and the resolution of the RS images is 30 m × 30 m. Then the results are revised by the land usage pattern provided by '*The Earth System Science Data Sharing Nets*'.

Figure 2 shows the temporal land use patterns of the site in 1991, 2002 and 2005, respectively. From the view of land use pattern, the north-western portion of the area functions completely as a city under the urban expansion of Wuhan, while the obstruction of the lake still allows for rural properties. In 1991, the agricultural land accounted for 48.16% of the whole land area (the total area of built-up, forest, agricultural land). Therefore, the agricultural land area was larger than the built-up land area, which was still more scattered, with no centralized developing tendency at that time. By 2002, under the background of economic development of China, the western basin exhibited features of a city, as the western development rate was significantly higher than that of the eastern area. Accompanied with the significant process of urban expansion, the built-up land increased to 93.13 km^2 in 2005, while the eastern area was still rural because of the obstruction of the lake. The built-up area of Donghu watershed increased from 51.44 km^2 in 1991 to 93.13 km^2 in 2005, and the agriculture and forest were reduced to provide space for urban development (according to the statistic data of land use map). Generally, we can conclude that the Donghu watershed was a typical urban–rural mixed area in 2005.

METHODS

Firstly, assuming that the watershed is rural primarily, this study calculates the particulate pollutant loads by using the USLE model, considering the factors of slope, normalized difference vegetation index (NDVI), land use type, soil type and rainfall. Subsequently, the dissolved pollutant loads were determined by the ECM which uses the export coefficient and the corresponding land use pattern to establish the relationship of land use type and pollutant loads. In particular, NDVI is a simple graphical indicator to assess whether the target that has been observed contains live green vegetation or not (Rulinda *et al.* 2011). Secondly, assuming that the watershed is urban, the L-THIA model is used to generate the spatial distribution of NPS pollutant loads in terms of total phosphorus and nitrogen. Finally, fuzzy membership functions are established to define the rural and urban weights for each land use cell. As opposed to binary weight, the weights defined here are used to combine the results of the rural and urban NPS pollutant loads calculating models.

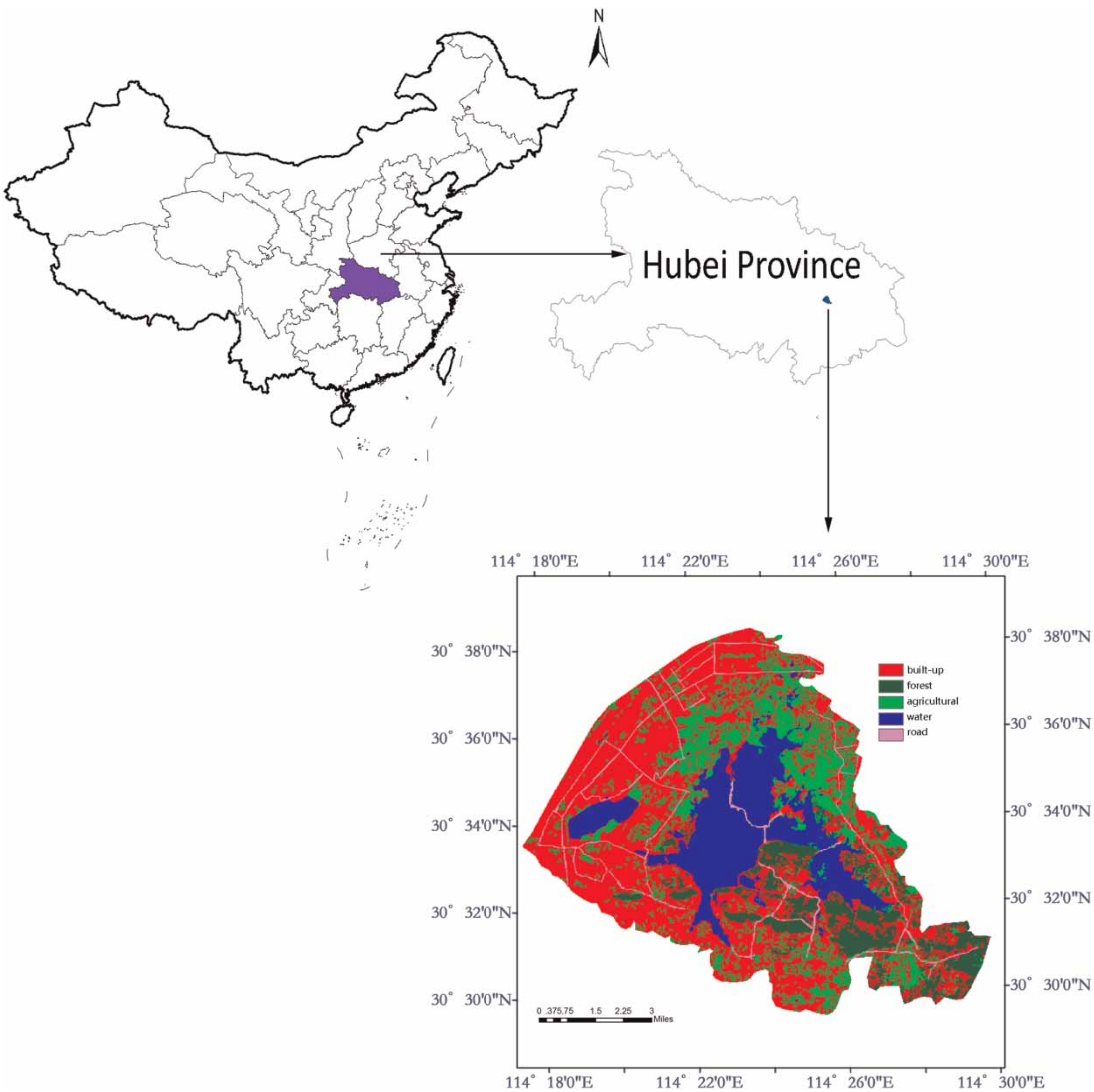

Figure 1 | Location and land use pattern of the study watershed.

The spatial data used in our study, like NDVI, slope, distance to road, distance to city center and land use pattern, are retrieved from LANDSAT TM imageries with the resolution of 30 m × 30 m. The non-spatial data, including rainfall capacity and the soil type, are obtained from the *Wuhan Statistical Year Book* and other statistical sources.

Evaluating NPS pollutant loads in rural areas

In this section, the classic USLE model and the ECM are used to acquire the particulate and dissolved pollutant loads of N and P, respectively.

Particulate N and P loads based on USLE model

The USLE was proposed by Wischmeier & Smith (1978), and has since been widely used at a watershed scale. It is an empirical model allowing the average annual soil loss based on the product of five erosion risk indicators (Meusburger *et al.* 2000). The empirical model to obtain particulate loads is represented in Equation (1):

$$W_{xp} = \beta \cdot C \cdot A \cdot \eta \cdot S_d \qquad (1)$$

where, W_{xp} is the particulate pollutant load (kg/(hm^2.a)); β is the dimensionless unit conversion constant; C_{N} and

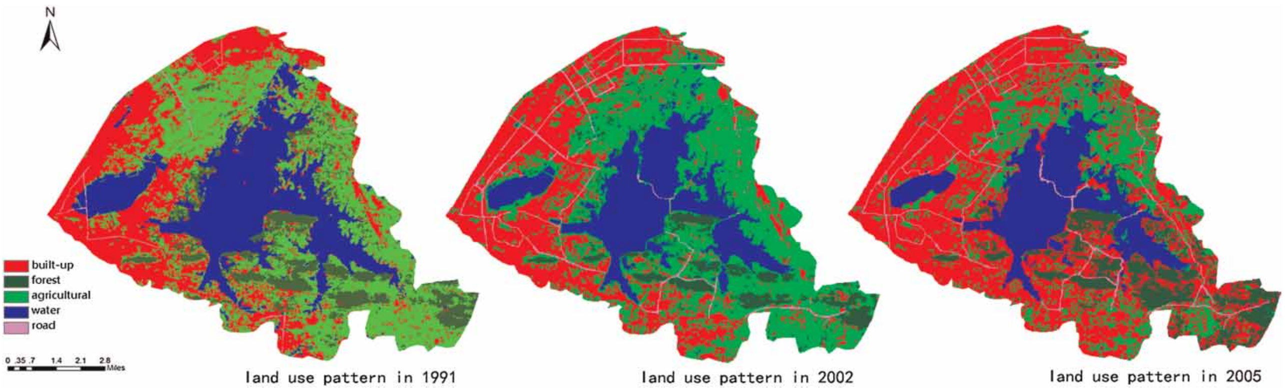

Figure 2 | Temporal and historical landscape patterns of study area.

C_P are the concentrations of particulate N and P, respectively, which are available from the almanac of soil in the Hubei and Henan provinces and the soil database of China provided by the Institute of Soil Science, Chinese Academy of Science, Nanjing; A is the amount of the soil loss (t/(hm^2.a)); η is the non-dimensional concentration coefficient; S_d is the ratio of the final pollutant loading into the lake to the original load generated in each cell. The specific value of each parameter is shown in Table 1 (Shi *et al.* 2002; Xu *et al.* 2006; Xue 2006).

From the USLE model, the amount of the soil loss can be obtained as Equation (2) shows, where K is the soil erodibility factor (t hm^2 h)/(hm^2 MJ mm), P is the support practice factor (non-dimensional), C is the cover management factor (non-dimensional), R is the rainfall erosivity factor ((MJ mm)/(hm^2 h a)) and LS is the slope steepness factor (non-dimensional).

$$A = K \cdot P \cdot C \cdot R \cdot LS \tag{2}$$

The soil erodibility factor (K) is related to the integrated effects of rainfall, runoff and infiltration on soil loss and can reflect the process of soil loss during storm events on upland areas (Renard *et al.* 1997). In our study, the soil type is general red earth and according to experimental data (Deng *et al.* 2003; Wang 2005; Zhang *et al.* 2007), the K value is denoted as 0.299.

The support practice factor (P) reflects the effects of soil conservation operations or other measures that will reduce the amount of water runoff and, thus, reduce the erosion rate (Volk *et al.* 2010), ranging from 0 to 1. The support practice factor can be obtained through considering the variation of the land use pattern. In this paper, by referencing soil conservation operations and relevant research (Bu *et al.* 1997; Cai *et al.* 2000; Xu & Shao 2006) on the study area, the P values are identified according to the land use type: the built-up land has a P value of 0.35; forest is 0.5; agriculture is 0.66; water body is 0.

Table 1 | The description of parameters in Equation (1)

Parameter	Scale	Description
β	1,000	Converting t/hm^2 a to kg/hm^2 a
C_N, C_P	Adsorption P concentration is 0.048%; Adsorption N concentration is 0.084%	In red earth
A	0–139.05 t/hm^2 a	–
η	2	Referring to the relevant research, in this paper the η was identified as 2
S_d	0.1–0.4	Referring to the S_d of Changjiang basin provided by Changjiang Water Resources Committee and the feature of our study site, S_d was identified varying from 0.1 to 0.4 with the 0.25 as the average value for all the cells and then the S_d of grid (i, j) could be calculated depending on the distance from the grid to the lake

The cover management factor (C) is a weighted index, which takes the effect of land use on soil erosion into account (Dumas *et al.* 2010). It is measured as the ratio of soil loss to land cropped under continuously fallow conditions (Wischmeier & Smith 1978). By definition, C equals 1 under standard fallow conditions. As vegetative cover approaches 100%, the C factor value approaches the minimal value. The C value of each cell is obtained by Equation (3) (Cai *et al.* 2000; Zhao *et al.* 2007).

$$C = \begin{cases} 1 & lc = 0 \\ 0.6805 - 0.3436 \lg lc & 0 \le lc < 78.3\% \\ 0 & 78.3\% \le lc \end{cases} \tag{3}$$

$$lc = \begin{cases} 0 & -1 \le \mathrm{NDVI} \le -0.0675 \\ \dfrac{\mathrm{NDVI} + 0.0675}{0.47} & -0.0675 < \mathrm{NDVI} \le 0.4025 \\ 1 & 0.4025 < \mathrm{NDVI} \le 1 \end{cases} \tag{4}$$

where lc is the vegetation coverage, non-dimensional. lc in Equation (3) can be obtained through the function of NDVI (see Equation (4)). An NDVI approaching a value of 1 means the associated area is fully covered by vegetation. Using NDVI retrieved from RS data, C values for our site can be calculated ranging from 0 to 1, with the average value of 0.3316 (see Figure 3(a)).

The rainfall factor (R) represents two characteristics of a storm that determine its erosivity: the amount of rainfall and the peak intensity sustained over an extended period. R is computed by using the function of monthly precipitation (Dumas *et al.* 2010) (see Equation (5)):

$$R = \sum_{i=1}^{12} (-2.6398 + 0.3046 P_i) \tag{5}$$

where R is in $\mathrm{MJ\,mm/(hm^2\,h\,a)}$, and P_i is the precipitation in the ith month which is obtained from the statistics yearbook.

The slope length and steepness factor (LS) represent the effect of topography on erosion, as an increase in slope length and steepness will produce higher overland flow velocities, thus, stronger erosion. LS is derived from Equation (6) (Wischmeier & Smith 1978; Dumas *et al.* 2010):

$$LS = \left(\frac{l}{22.13}\right)^m (0.085 + 0.045\theta + 0.0025\theta^2) \tag{6}$$

$$m = \begin{cases} 0.3 & 22.5^0 \le \theta \\ 0.25 & 17.5^0 \le \theta < 22.5^0 \\ 0.2 & 12.5^0 \le \theta < 17.5^0 \\ 0.15 & 7.5^0 \le \theta < 12.5^0 \\ 0.10 & \theta < 7.5^0 \end{cases} \tag{7}$$

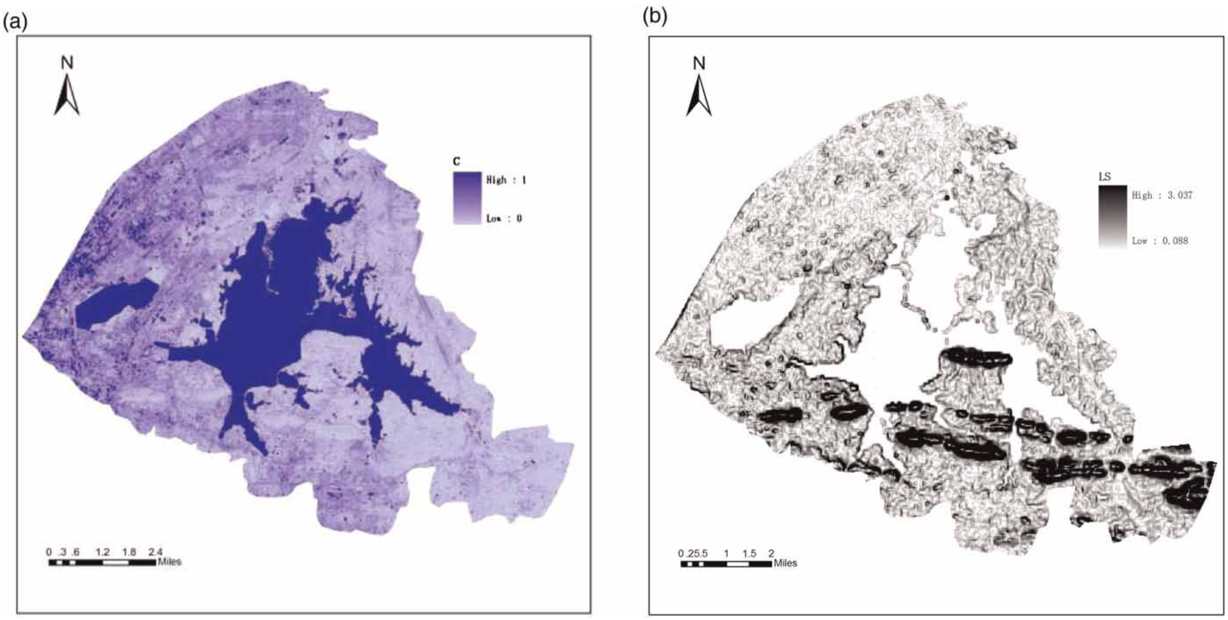

Figure 3 | Distributions of USLE factors: (a) the cover management factor C; (b) the slope length and steepness factor LS.

where l is the slope length in meters, θ is the slope angle in degrees, and m is the slope angle contingent variable ranging from 0.1 to 0.3 (McCool *et al.* 1987) computed by the function of slope (see Equation (7)). l and θ are calculated applying a 30 m resolution DEM and ArcGIS software package, followed by the values of m for each cell. *LS* values vary from 0.088 to 3.037, with an average of 0.152 (Figure 3(b)).

Dissolved N and P loads based on the export coefficient model

The ECM is a well-developed method that has been widely employed in NPS pollution studies (Kay *et al.* 2008) which avoids the difficulty of the physical models. This eliminates the difficulties associated with the complex formation of NPS pollution, thereby reducing the requirements of monitoring the processes of the migration and transformation of the pollutants. Thus, the ECM is available for estimating the NPS pollution for the medium or the large-scale watershed. This model is commonly represented in the form of Equation (8):

$$W_{xd} = \sum_{i}^{n} \sum_{j}^{m} E \times \alpha \tag{8}$$

where W_{xd} is the output quantity of the dissolved pollutant (kg/hm^2 a), E is the export coefficient of the pollutant (t/km^2 a) on different land usages, and α is the conversion factor with the value of 10. The value of E is identified according to the literature review of NPS studies on the Yangtze River and city of Chongqing (Liu *et al.* 2006; Cao *et al.* 2007) and characteristic of our site (see Table 2).

NPS pollutant loads in rural areas

Based on the USLE which integrated with the empirical model and the ECM, the particulate pollutant load, W_{xp}, and the dissolved pollutant load, W_{xd}, are achieved respectively. Then, the NPS pollutant loads assuming the study area is rural, NPS$_{\text{rural_model}}$, can be calculated by adding the particulate and dissolved NPS pollutant loads (see Equation (9)).

$$\text{NPS}_{\text{rural\_model}} = W_{xp} + W_{xd} \tag{9}$$

Table 2 | Export coefficient of the pollutant (E) under hypothesis of rural area

Land use type	The concentration of dissolved N t/km^2 a	The concentration of dissolved p t/km^2 a
Built-up	0.6	0.011
Forest	0.119	0.007
Agricultural	1.2	0.04
Water	0	0

Evaluating NPS pollutant loads in urban areas

A distributed hydrological-water quality model based on hydrological response units, the L-THIA model (Phillips *et al.* 2007), is selected to simulate the urban NPS pollutant loads. It takes long-term hydrological impacts on land use change into consideration, so it can be useful in researching the relationships between urbanization, surface runoff and urban NPS pollution (Yang *et al.* 2008). L-THIA was developed as an effective approach to estimate the NPS pollution resulting from past or proposed land use changes (Zhang *et al.* 2011).

Based on the L-THIA model, the NPS pollutant loads can be acquired through Equation (10):

$$\text{NPS}_{\text{urban\_model}} = AR * AE * UR \tag{10}$$

$$AR = \frac{(RP - 0.2S)^2}{RP + 0.8S} \quad (P \geq 0.2S) \tag{11}$$

$$S = 25.4\left(\frac{1000}{CN} - 10\right) \tag{12}$$

in which NPS$_{\text{urban_model}}$ is the NPS pollutant load (kg/hm^2 a); *UR* is the unit conversion constant, 10^{-2}; and *AE* is the concentration of pollutant in the surface runoff for each land use type (mg/L). Due to the difficulty in collecting data, we identify the concentration of pollutant, *AE*, in different land use types by literature review and the detailed information is represented in Table 3. *AR* is the quantity of actual runoff, in mm which can be retrieved from the function of total annual precipitation, *RP*, and potential maximum precipitation, *S* (see Equation (11) and Equation (12)). According to the precipitation data measured by the

Table 3 | Concentrations of pollutants (AE) under the hypothesis of urban area

Land use type	Concentration of N (mg/L)	Concentration of P (mg/L)
Built-up	3.92	0.4
Forest	1.9	0.42
Agricultural	5.7	1.6
Water	0	0

monitoring station and the approach of interpolation, the annual precipitation of each cell is obtained (see Figure 4). The maximum precipitation can be identified by the *CN* value which is obtained from literature review. In Yang's study (2008), the *CN* value of the Hanyang district, which is approximately 15 kilometers away from Donghu Lake, experiencing similar temperatures and rainfall as our site, were proposed. In addition, the *CN* value used in Yang's paper had been modified by the antecedent moisture condition (AMC) already (Zhao 2008; Li *et al*. 2009; Wang *et al*. 2009) and the detailed values are presented in Table 4.

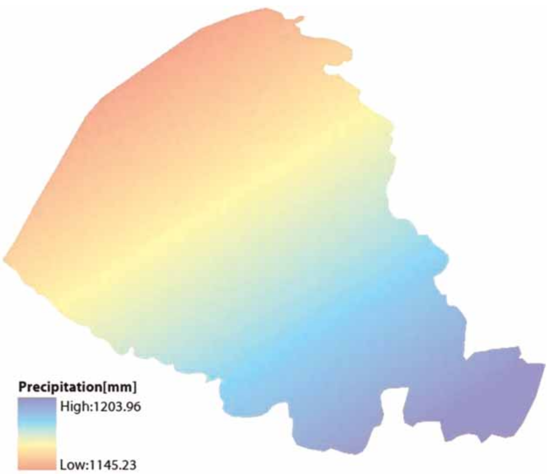

Figure 4 | Spatial distribution of summed annual precipitation in 2005.

Table 4 | CN value in each land use type in the L-THIA model

Land use type	*CN*
Built-up	98.81
Forest	92.51
Agricultural	96.02
Water	0.00

Complex NPS loads in urban and rural mixed places

Due to the fact that generations and properties of NPS pollutants in rural and urban areas are totally different, it is essential to discriminate the rural and urban areas and use various models and parameters to evaluate the NPS in these two places. However, it is difficult to discriminate the rural and urban cells in a rapid developing area where the rural and urban places are mixed and coexisting. Conventionally, an administrative boundary is employed to distinguish the characteristic of the study area. In China, a city administratively contains a built up area, suburbs, and counties under city administration. Usually the built-up area is urban, and the suburbs are a mix of urban and rural. In fact, it is hard to distinguish, using an administrative boundary, between urban place and rural place which suffer totally different process of NPS pollutant generation and transportation. Especially in some rapid developing cities, rural places are continually changing into urban areas to satisfy the requirement of economic development and population growth.

Conventional classification methods divided the land into classic clusters, for example '0' meaning rural or '1' meaning urban. Fuzziness exists in the real world, especially in the boundaries where it is hard to judge or classify. Hence, the classic classification method would be unsuitable. A fuzzy membership function-based approach is proposed to define a cell as urban or rural which not only classifies the cell into an urban or rural cell, but also provides the membership of belonging to a rural or an urban place which can reflect the degree of belonging to a certain cluster and can be used to combine the urban and rural NPS evaluation results.

In our work, three factors are defined to evaluate whether a cell belongs to urban place: (1) characteristic of surrounding land use, (2) influence of city center, (3) traffic condition. The density of built-up land within the land use cell is used to express the characteristic of surrounding land use. The grids which are within 150 m are taken into consideration to calculate the density. The city center is defined as the CBD (Central Business District) of Wuhan, and the distance from the city center is employed to measure the influence of the city center. Finally, the distance to the nearest road line is proposed to reflect the traffic condition, and a smaller distance a more convenient traffic condition.

Particularly, the factors stated above can be denoted as $X = \{x_1, x_2, x_3, \ldots, x_m\}$ where m is the number of attribution, and the fuzzy mapping can be established: $f \sim : X \to \vartheta(r)$. In other words, by function f the attribute of a certain land use cell, x_i, can be mapped to the membership of belonging to a certain cluster j, which can be written as r_{ij}. The function f is determined by the suggestions of experts and the characteristic of the study area.

According to the membership of single attribution, the comprehensive evaluation can be conducted. The membership of belonging to a certain cluster j can be calculated according to the fuzzy operator of r_{ij} (see Equation (13)). In our study, the multiple product of all memberships of single attribution is employed (see Equation (14)).

$$R_j = \otimes r_{ij} \tag{13}$$

$$R_j = \prod_{i=1}^{m} r_{ij} \tag{14}$$

Generally, a cell with a large density of built-up land, small distance to the city center and small distance to a road tends to be an urban cell. Inversely, the cell should be a rural cell. And then the bell-shaped function, the Gaussian curve function, and sigmf function, which is a function composed of the difference between two sigmoidal membership functions can be used as fuzzy function (see Equations (15)–(17)) to express the relationship between attributions and the membership functions. The parameters of the fuzzy function are feasible to be generated according to the opinion of experts. The Analytic Hierarchy Process (AHP) (Saaty 1980) which is a pairwise comparison approach has been used to extract the experts' opinion.

$$f_{\mathrm{bell}}(x, a, b, c) = \frac{1}{1 + |(x-c)/a|^{2b}} \tag{15}$$

$$f_{\mathrm{sigmf}}(x, a1, c1, a2, c2) = \frac{1}{1 + \mathrm{e}^{-a1(x-c1)}} - \frac{1}{1 + \mathrm{e}^{-a2(x-c2)}} \tag{16}$$

$$f_{\mathrm{gaussian}}(x, a, b, c) = a \cdot \mathrm{e}^{-(x-b)^2/c^2} \tag{17}$$

By suggestion of experts, the fuzzy functions are defined and Figure 5 represents the tendency line between membership and the value of factors.

The fuzzy functions of belonging to a rural place are defined as $f_{\mathrm{rural}} = 1 – f_{\mathrm{urban}}$. Then, the membership of belonging to an urban place is defined as Equation (18) shows.

$$f_{\mathrm{urban}}(X) = f_{\mathrm{urban1}}(\mathrm{DeBuilt}, a, b, c) * f_{\mathrm{urban2}}(\mathrm{DisCen}, a, b, c) * f_{\mathrm{urban3}}(\mathrm{DisRoad}, a, b, c) \tag{18}$$

Finally, the NPS in land use cell can be calculated as Equation (19) shows. Otherwise, the integrated NPS would be calculated according to binary weight as Equation (20) shows.

$$\mathrm{NPS} = f_{\mathrm{urban}}(X) * \mathrm{NPS}_{\mathrm{urban\_model}} + f_{\mathrm{rural}}(X) * \mathrm{NPS}_{\mathrm{urban\_model}} \tag{19}$$

$$\mathrm{NPS} = \begin{cases} \mathrm{NPS}_{\mathrm{urban\_model}} * 1 + \mathrm{NPS}_{\mathrm{rural\_model}} * 0 \\ \qquad \text{cell} \in \text{urban place} \\ \mathrm{NPS}_{\mathrm{urban\_model}} * 0 + \mathrm{NPS}_{\mathrm{rural\_model}} * 1 \\ \qquad \text{cell} \in \text{rural place} \end{cases} \tag{20}$$

RESULTS AND DISCUSSION

Urban NPS

Urban NPS pollutant loads calculated by the L-THIA model are shown in Figures 6(a) and 7(a), assuming that the study area is totally urban. The L-THIA model determines the urban NPS pollutant loads through rainfall-runoff and concentration of pollutants within each land use type because in an urban system the land is covered by impervious areas and the influence of natural factors such as slope or soil type is less while the impact of human activities is larger. In the L-THIA model, the intensity of human activities is indirectly reflected by land use. As a result, the total nitrogen (TN) in the built-up land area is around 42 to 47 km/hm^2 a, and that of the forest area is about 17 to 22 km/hm^2 a. Meanwhile the agricultural land area undergoes the highest TN load, achieving about 60 km/hm^2 a, and the total phosphorus (TP) load suffers similar spatial distribution to that of TN.

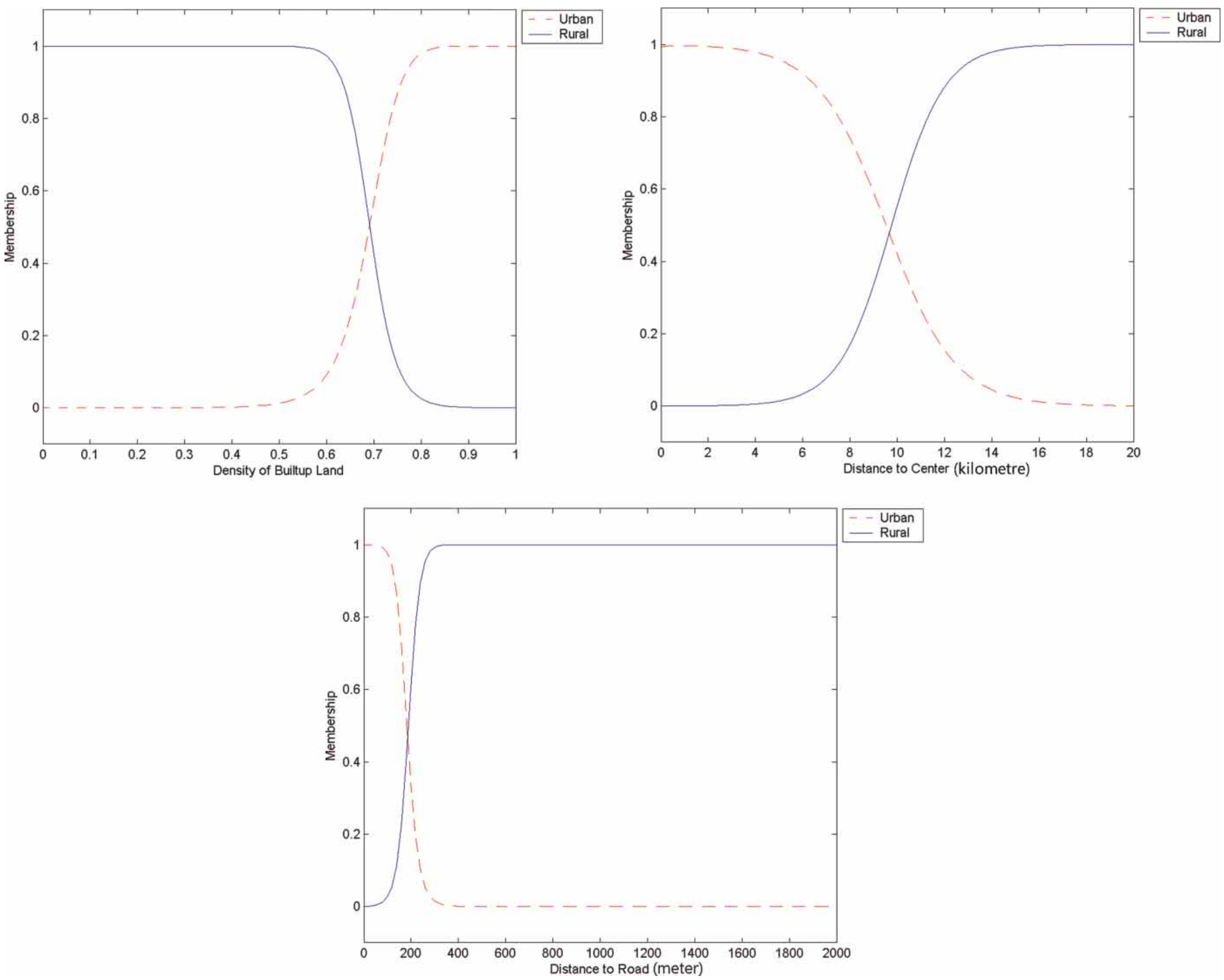

Figure 5 | The membership functions of three factors.

Rural NPS

As for the rural NPS pollutant loads (Figures 6(b) and 7(b)) calculated by the USLE model, the TN and the TP range from 0 to 30 km/hm^2 a and 1 to 10 km/hm^2 a, respectively, which do not correspond to the land use pattern but relate much more to the nature factors like slope, the ration of vegetation (which is measured by NDVI), soil type and so on. In Figures 8(a) and 9(a), TP and TN highlights are both concentrated in the southern region, and the analysis reveals that these highlights usually corresponded with difficult terrain, such as a larger slope which tends to be associated with hard runoff and large soil erodibility, resulting in higher NPS loads. Since the USLE model combines rainfall, topography, management factors, soil types, cover management factors and other factors to simulate NPS pollutant loads, the distributions tend to be gentle and gradual.

Division of urban and rural areas

By the fuzzy membership functions and three factors, the land use cells can be classified into rural and urban at the same time. Figure 10 displays three factors in x, y and z axis, with the position of the point denoting the value of three factors and the color denoting the membership of belonging to an urban place. In plot A (see Figure 10) where the density of built-up is close to 1 (the maximum) and the distance to center is short, the cells undergo higher membership of belonging to an urban place. In plot B, the cells with medium value of three factors are hard to

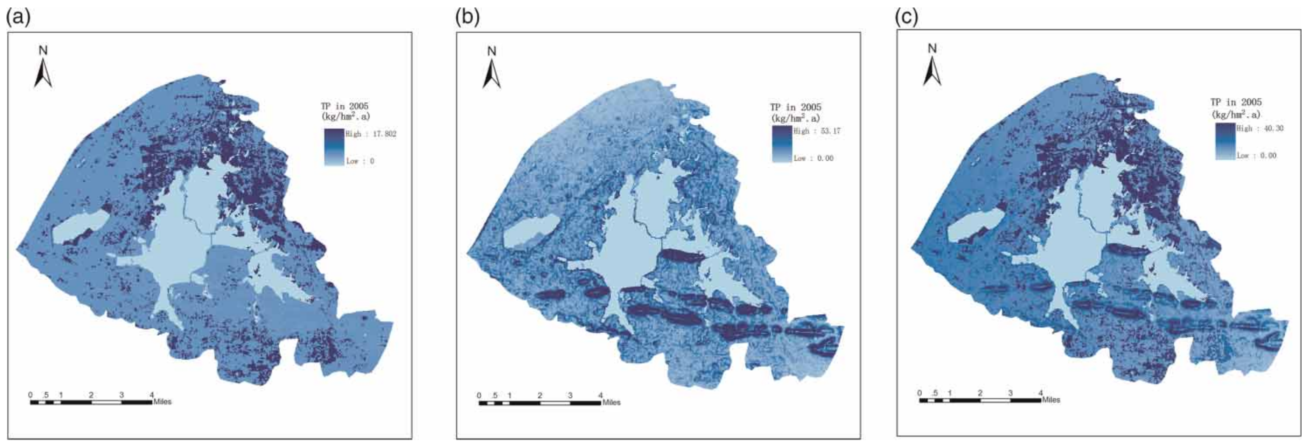

Figure 6 | Results of TP load by different methods: (a) by urban method, (b) by rural method, and (c) integrated result.

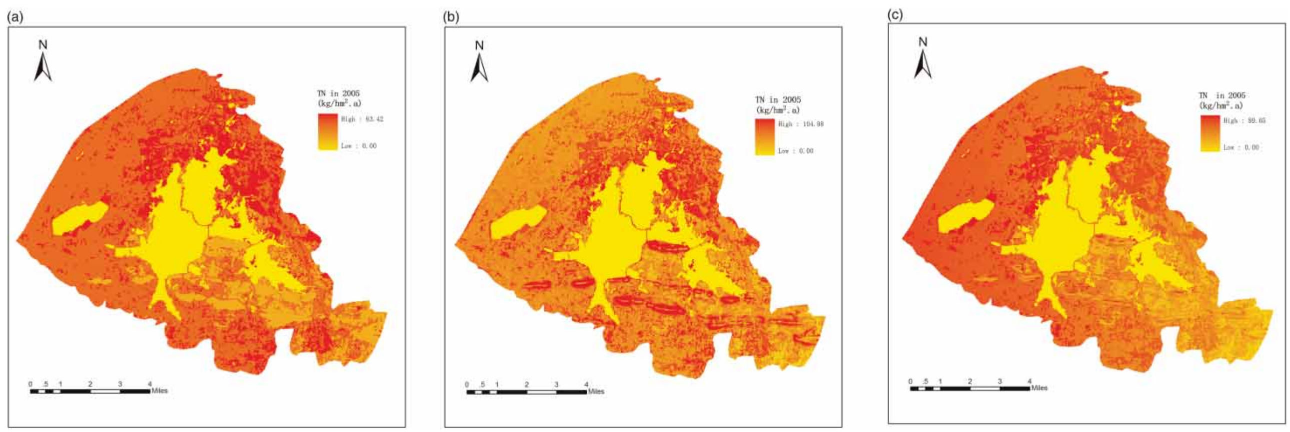

Figure 7 | Results of TN load by different methods: (a) by urban method, (b) by rural method, and (c) integrated result.

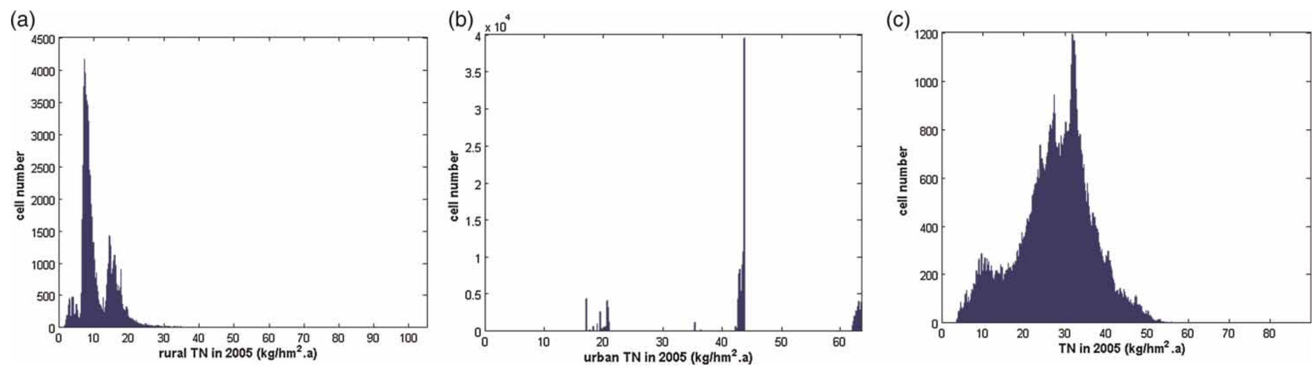

Figure 8 | Histograms of TP load: (a) by USLE model, (b) by L-THIA model, and (c) integrated result.

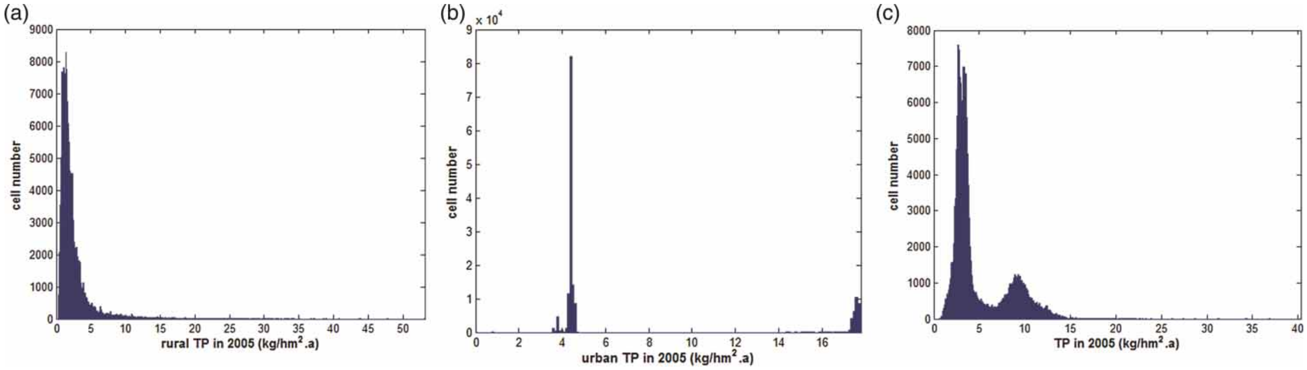

Figure 9 | Histograms of TN load: (a) by USLE model, (b) by L-THIA model, and (c) integrated result.

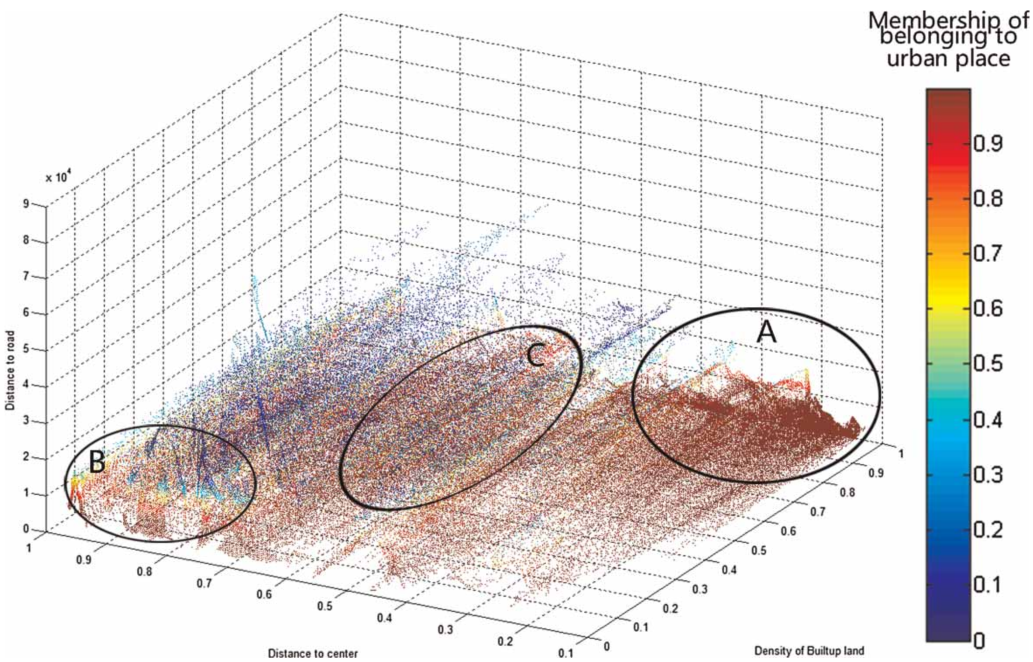

Figure 10 | Three-dimensional distribution of the membership with three factors denoted by x, y and z axis.

classify and the memberships of these cells are around 0.4 to 0.6. Additionally, we find that as the distance from the road increases, the cell tends to undergo small membership of belonging to urban places. Besides the cells in plot B, the cells in the center of this three-dimensional space (Plot C) also had membership near to 0.5.

Correspondingly, Figure 11(a) represents the membership based on spatial distribution of the land use cells. We find out the cells with the highest fuzziness (around 0.5) are concentrated around the boundary of the urban and rural places.

Additionally, to justify the advantage of using the fuzzy approach, the conventional results with binary values obtained by the k-means classification method are displayed in Figure 11(b). By comparison, we find that the membership ranging from 0 to 1 can reflect the distribution and characteristic of land use better than the classic results where only two integers are denoted, as 'rural' and 'urban'. The fuzzy approach generates generally similar results to that of the classic method where the rural and urban distribution is generally the same. But in the boundary, the fuzzy approach

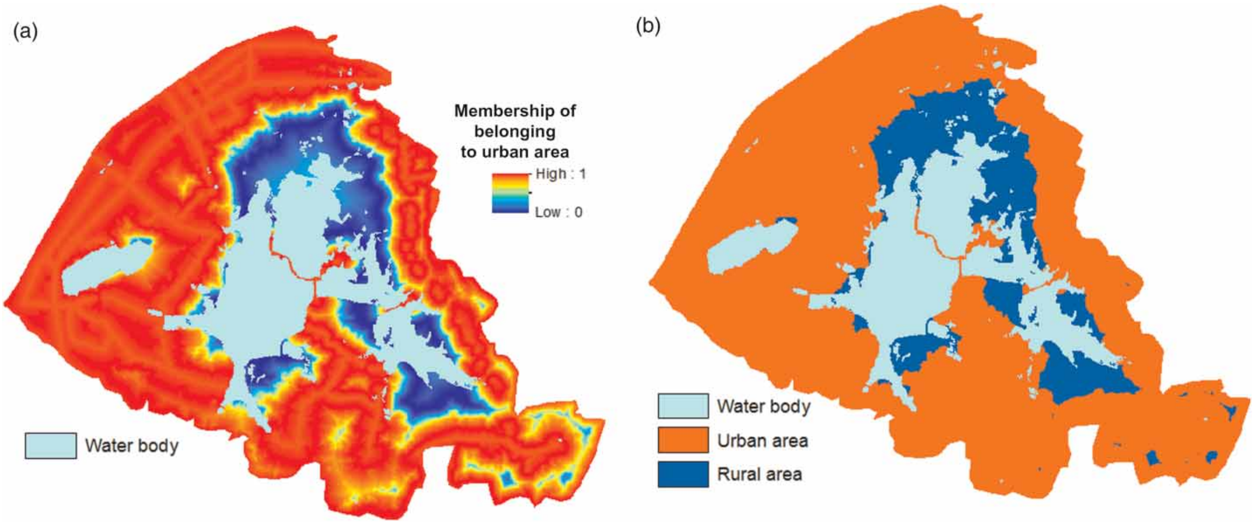

Figure 11 | The classification of land use cells to urban and rural in two-dimensional space: (a) the results by fuzzy approach; (b) the results by conventional k-means method.

uses a gradual value to express the change from urban to rural however the classic results provide a sharpened break.

Complex NPS

Ren *et al.* (2003) investigated and analyzed the urbanization level and the water quality of Shanghai from 1947 to 1996, showing that the faster the rate of urbanization increased, the poorer the water quality became in his case study; Sartor *et al.* (1972) focused on the pollutant loads on urban streets, and pointed out that pollutant loads of nutrients in urban runoff were much higher than that of rural areas in his case study; Shon *et al.* (2012) argued that the amount of NPS pollutant loads discharged into rivers was larger in urban regions than in forests and farmlands, because of the high population and greater impermeable areas, and then used a storm water management model (SWMM) to simulate NPS pollutant loads in the target area. Similarly, this study revealed that NPS pollutant loads in the scope of urban land are larger than that of the rural model (Table 5) in Donghu watershed. The integrated results (see Figures 6(c) and 7(c)) better reflects the complex NPS pollution distribution because it assumed that the study area is rural and urban mixed. According to the membership of belonging to urban and rural, the urban and rural NPSs are integrated, and there is no sharpened break between rural and urban areas, meaning the NPSs in rural and urban areas are interactive even if they have different generations and characteristics. Especially in the area of boundary, the urban NPS and rural NPS are mixed and interactive. Hence, in the integrated NPS distribution maps, there is no sharpened break, at the same time the different NPS evaluation models are applied for different places.

Table 5 | Pollutant loads of each land use type by different methods

		Built-up (kg/hm^2 a)	Forest (kg/hm^2 a)	Agriculture (kg/hm^2 a)	Water (kg/hm^2 a)
Average P	Urban	3.451	5.6545	11.921	0
	Rural	2.7952	4.8293	8.8148	0
	Over all	3.1443	4.888	9.4385	0
Average N	Urban	30.804	17.386	45.156	0
	Rural	22.381	12.809	35.667	0
	Over all	26.865	13.135	37.573	0

CONCLUSION

Computation and analysis of NPS pollutants according to land use changes, precipitations, topography, soil type, vegetation and others in urban–rural mixed places were presented. In this study, we established a comprehensive model that successfully calculates rural and urban NPS pollutant loads respectively by USLE, ECM and L-THIA. Then, we introduced the fuzzy membership function based approach to integrate the rural and urban NPS pollutant loads through the evaluation of land use characteristic. Afterwards, the results were successfully obtained regarding complex NPS pollution in an urban–rural mixed watershed, Donghu watershed.

Even if numerous studies are concerned with the variation of NPS pollutant loads under the rapid urbanization process, there is no applicative model that alludes to the increasingly urban–rural mixed watershed and that considers the difference in generation and characteristic of pollution between the urban and rural areas. To address this issue, we firstly identify the NPS pollution in urban–rural mixed areas and caused by various pollutants in rural and urban surface runoff together as the complex NPS pollution, and then employ the fuzzy membership function to classify the urban areas and rural areas so as to integrate the well-developed urban NPS model and rural NPS model. The results are proven to be consistent with existing research conclusions and with the characteristics of our site.

To our knowledge, urbanization is popular worldwide. Take China as an example: the national urbanization level stood at 11% in 1949 and sharply increased to 29% in 1996 (Wang *et al*. 2004). Although the rapid urbanization process has boosted the economy and led to a higher quality of life, some adverse effects have been brought along with it. For example, in addition to detrimental water quality, the descent of indoor-air-quality (Wang *et al*. 2004), damage of ecosystem, climate change (Grimm *et al*. 2008), threat to biodiversity (Pompeu *et al*. 2005), effect on tree growth (Gregg *et al*. 2003), promotion of asthma (Lin *et al*. 2001) and so on are associated with rapid urbanization. Hence, it is significant to evaluate, analyze and understand the relationship between urbanization and the corresponding detrimental effects. Within this context, the model proposed in this study which focused on the fuzziness in the rural-urban mixed places is innovative and applicable for the above mentioned problems. In particular, the model proposed is capable of being employed in any other rural and urban mixed regions, which undergo rapid urbanization, with corresponding classification factors. For example, in Donghu watershed the distance to the city center, the density of built-up land, and the distance to the nearest road are employed to describe the characteristics of each land use cell by fuzzy membership function, while in other case study areas the factors may be various. Then, according to fuzziness in terms of land usage, the degree of being urban or rural can be identified. Afterward, the relationship between urbanization and the above mentioned problems can be exactly assessed.

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