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Modeling of non-point source nitrogen pollution from 1979 to 2008 in Jiaodong Peninsula, China

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Abstract:

Efforts to reduce land-based non-point source (NPS) pollutions from watersheds to coastal waters are ongoing all around the world. In this study, annual yield of NPS nitrogen (NPS-N) pollution in Jiaodong Peninsula, China from 1979 to 2008 was estimated. The results showed that: from 1979 to 2008, NPS-N yields exhibited significant inter-annual variations and an increasing trend on decadal scale. High NPS-N yield was mainly found in east and south parts, as well as the urbanized coastal regions in Jiaodong Peninsula. Among the 32 river basins, the three largest basins yielded more than 41.16% of the NPS-N. However, some small coastal watersheds along the South Yellow Sea and Jiaozhou Bay had higher per unit area yield. Most of the small watersheds characterized by seasonal runoff had coastal waters pertain to mild and moderate pollution levels. The ratio of watershed area to shoreline length and the up-stream land use had significant impacts on NPS-N flux through the shoreline. Among the four adjacent coastal areas of Jiaodong Peninsula, Jiaozhou Bay was the most noteworthy one not only because of high levels of land-based NPS-N pollution but also because of its nearly enclosed structure. The combination between integrated coastal zone management and integrated river basin management, land use planning and landscape designing in Jiaodong Peninsula is recommended. Copyright © 2013 John Wiley & Sons, Ltd.

KEY WORDS coastal environment; Jiaodong Peninsula; watershed; land use; nitrogen; non-point source pollution

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INTRODUCTION

Non-point source (NPS) pollution usually refers to pollutions that come from diffuse runoff moving over and through land surface, picking up and carrying various natural or anthropogenic pollutants and finally depositing them into receiving waters (Nikolaidis *et al*., 1998; Shen *et al*., 2012). NPS pollution is diffuse in character and plays an important role in water quality degradation; therefore, it has become an active subject of water environment research since 1970s partly because of the eutrophication 'crisis' of the Great Lakes in North America and the promulgation of the Clean Water Act in the USA. In the past three decades, more and more literatures that talked about NPS pollution at local, regional, national and global scales have emerged (Schreiber *et al*., 2001; Norse, 2005; Ulen *et al*., 2007; Yuan *et al*., 2007; Dowd *et al*., 2008; Lee *et al*., 2009; Badruzzaman *et al*., 2012). In China, the rapid social-economic development since the late 1970s has contributed to serious NPS pollution, and hence studies on NPS pollution have gradually attracted attentions in the past three decades. Earlier researches on NPS pollution in China in late 1980s were mainly focused on urban runoff pollution in Beijing and some important water bodies such as Dianchi and Taihu Lake (Guo *et al*., 2004; Kuang *et al*., 2004; Ongley *et al*., 2010). The main progress had been made in this period was the recognition of the problems and preliminary understandings on mechanisms and characteristics of NPS pollution. Among the following studies, a great deal of efforts have been put on agricultural NPS pollution because China is one of the largest producers and consumers of chemical fertilizers and pesticides in the world, and the excessive nutrient loads from agricultural watersheds has been considered as the primary source of NPS pollution (Chen *et al*., 2011; Shen *et al*., 2012; Wu *et al*., 2012). Specifically, nutrients from excessive fertilizer utilization and pesticide spraying were deposited into rivers, lakes and reservoirs, resulting in deterioration of water quality, eutrophication and reduction of biodiversity in the

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aquatic ecosystems (Maxted *et al*., 2009; Meixler and Bain, 2010; Yen *et al*., 2012).

Land-based pollution, especially NPS pollution that is closely related, with land use changes, is becoming a hotspot of multi-disciplinary research in coastal zone throughout the world (Nikolaidis *et al*., 1998; Blanco *et al*., 2010; Xu *et al*., 2012). As a matter of fact, land-based NPS pollution has become the fundamental reason of coastal water degradation in many places, which boosts phytoplankton biomass, reduces water clarity, increases frequency of toxic algae blooms and aggravates risks in coastal zone (Goolsby and Battaglin, 2001; Bradley and David, 2003; Islam and Tanaka, 2004; Houk *et al*., 2005; Wit *et al*., 2005; Selig *et al*., 2006; Howarth, 2008; Rollo and Robin, 2010; Nagy *et al*., 2012; Williams *et al*., 2012). For recent years, very extensive land use changes have happened in China. As a result, excessive nutrients and heavy metals closely related with NPSs have led to serious pollution and algae blooms, and disturbed wetland hydrology in both inland and coastal waters (Guo *et al*., 2004; Li *et al*., 2009a; Ongley *et al*., 2010; Xu *et al*., 2012). With the abatement of point source pollution, NPS pollution has become an increasingly important environmental concern of aquatic ecosystem and human beings (Shamshad *et al*., 2008; Ma *et al*., 2011; Miller *et al*., 2011). Efforts to reduce NPS pollutants loadings from watersheds to coastal waters have been ongoing for a long time in the developed countries (Pierobon *et al*., 2012). However, up to now, limited studies have been found with a focus on land-based NPS pollution in China's coastal zone (Yuan *et al*., 2007; Huang and Hong, 2010; Chen *et al*., 2011), and thereby, knowledge about this issue is lagging far behind that has been achieved in inland water bodies in China.

Jiaodong Peninsula, as the largest peninsula in China, has experienced rapid population growth, social and economic development, and dramatic land use and land cover changes in the past three decades. As a result, nutrient over-enrichment and serious environmental problems caused by increased point source and NPS pollution have been found in inland and coastal waters of Jiaodong Peninsula (Wei *et al*., 2008), such as repeated red tide outbreaks offshore of Yantai city from 2004 to 2009 (Hao *et al*., 2011). However, present studies in this region mainly put emphasis on estuary nutrient flux and its environmental impacts on a number of critical areas of coastal water for short time periods (Liu *et al*., 2007; Zhang, 2007; Liu *et al*., 2010; Han *et al*., 2011). Long-term characteristics of NPS pollution and its impacts on coastal water are scarcely talked about, which restricted our deep understanding of the land-ocean interactions in coastal zone (LOICZ) and the effective implementation of the integrated coastal zone management (ICZM) in Jiaodong Peninsula.

In this study, by taking NPS nitrogen (NPS-N) pollution as an example, the primary objectives are the spatial-temporal characteristics of long-term NPS-N yield in land area and the spatial patterns of land-based NPS-N pollution risks at estuaries that have permanent runoff and shoreline segments that have storm water runoff only in Jiaodong Peninsula. In specific, we establish a multi-factor database and develop evaluation models in GIS to estimate the long-term NPS-N pollution and to assess the risks of land-based NPS-N pollution in coastal waters in Jiaodong Peninsula. The section 'Materials and methods' briefly introduces the Jiaodong Peninsula and then describes the datasets and methods used to evaluate long-term characteristics of NPS-N pollution. The section 'Results and discussion' includes the spatiotemporal variations of NPS-N yield, NPS-N fluxes from land to coastal waters through estuaries and shoreline segments, and levels of land-based NPS-N pollution in coastal waters. The last section is the 'Conclusion'.

MATERIALS AND METHODS

Study area

Jiaodong Peninsula usually refers to the spatial area that lies the east of Jiaolai River in Shandong province, China. It locates between 35.8–38.0º N and 119.8–122.9º E, facing Bohai Sea to the northwest and Yellow Sea to the south and northeast. In this paper, based on the shuttle radar topography mission digital elevation model (SRTM DEM), the study area is redefined to the spatial domain of river basins (Figure 1) delineated by hydro-analysis techniques in ArcGIS 9.3, which has very limited difference with Jiaodong Peninsula in the usual sense. In specific, three big river basins that have perennial river runoff had been delineated, which are Dagu River basin, Wulong River basin and Dagujia River basin. In addition, 29 small watersheds, which are mostly seasonal rivers that have storm water runoff only in recent decades, had been delineated also. As the largest peninsula in China, Jiaodong Peninsula covers cities of Yantai and Weihai and most part of Qingdao, with an area about 28 230 km^2. It is characterized by warm-temperate humid monsoon climate, with the highest average monthly temperature of 25 °C in July and the lowest monthly average temperature of −3 to −1 °C in January. The average annual precipitation ranges from 650 to 850 mm in different parts of the peninsula. Agriculture land is the main land use type, which accounts for more than 57.30% of the whole peninsula; the following is grassland, which accounts for 14.80%; and the third is forest, which accounts for 11.45%; urban area, rural settlement and independent industrial-mining area account for 13.40% in total, and the rest of the land are inland water body and unused land, which accounts for

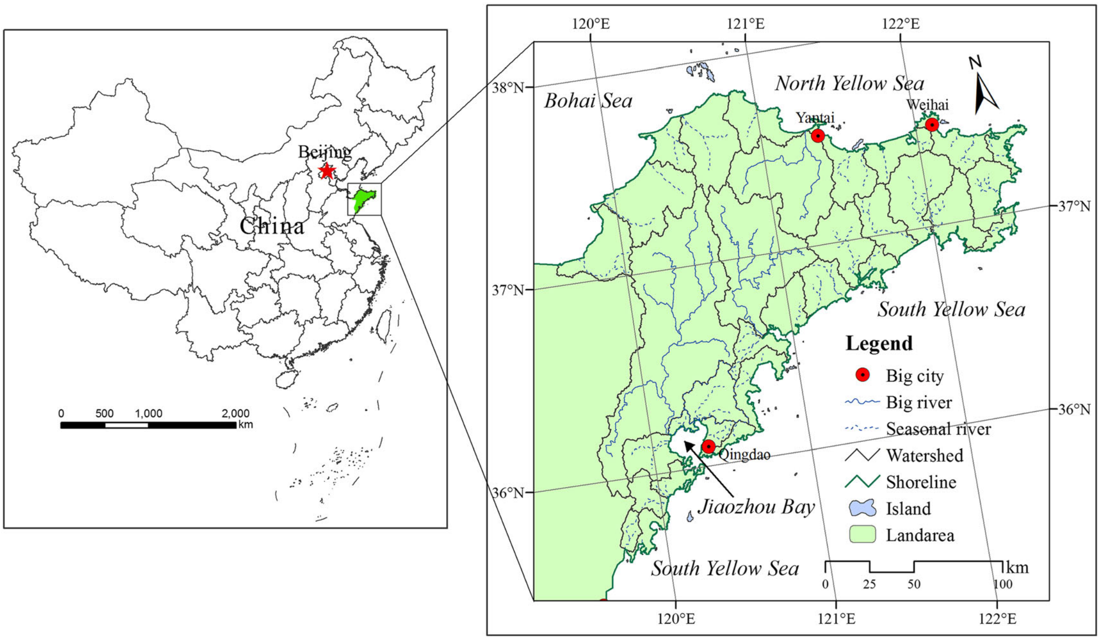

Figure 1. Location map of the study area and major rivers

3.05% in total. Both rapid urbanization and intensive coastal zone development have resulted in remarkable land use and land cover changes in the past 30 years, among which urban expansion and sea reclamation are the two most significant characteristics.

Data

A multi-source spatial database was prepared, including remote sensing images and land use maps in multiple years, DEM, soil maps and soil survey data, precipitation data of meteorological stations, and runoff data of hydrological stations, pollutant coefficients and a variety of ancillary data. Essential data and fundamental processing are as follows.

Predominantly cloud-free images of Landsat MSS/TM/ETM + that captured in 1979, 1990, 1995, 2000 and 2005, respectively, were acquired mainly from USGS EROS Data Center (http://eros.usgs.gov/) and GLCF Data Center (http://glcf.umiacs.umd. edu/). Multi-period land use maps were created by the visual interpretation method based on these images, photos and GPS data collected during field surveys, and a variety of ancillary data, such as topographical maps, existing land use/cover maps, road network maps, river maps, settlement maps and so on. The hierarchy of land use was established based on image capabilities, research goals and regional land use characters, which include agriculture (AGRL), forest (FRST), grass/pasture (PAST), water body (WATR), urban (URAN), rural settlement (RURL) and independent industrial-mining (INDL).

The DEM data was downloaded from the SRTM 90 m digital elevation database distributed by the GLCF Data Center (http://glcf.umiacs.umd.edu/). DEM data was used to delineate watersheds, extract river networks and calculate surface flow variables by using the spatial analysis techniques and hydrological analysis tools in ArcGIS 9.3. Overall, 32 watersheds were delineated, among which there are three largest watersheds with permanent rivers flow into coastal waters, and other 29 small coastal watersheds with mostly seasonal rivers in recent decades.

Precipitation is the fundamental source of surface runoff and main impetus for NPS pollutant transportation. Monthly precipitation data from 1979 to 2008 at 15 meteorological stations were available from the China Meteorological Data Sharing Service System provided by the Chinese Meteorological Administration. Annual precipitation series were first synthesized, and then, station-gauged series were expanded into gridded spatial data by Kriging method in ArcGIS 9.3.

The annual runoff data from 1972 to 1985 were collected from five hydrologic stations (Tuanwang (S1), Menlou (S2), Fushan (S3), Muyu (S4) and Mishan (S5)), which are distributed evenly in the study area. Runoff data was used to calibrate and validate the Soil Conservation Service Curve Number (SCS-CN) model.

Soil data were extracted from the 1:500 000 soil maps and soil-profile property data of Shandong Province, China. The data were generated during the Second Soil Survey in the mid 1980s. According to the guidelines of the SCS-CN model, soils were classified into hydrologic soil groups (HSGs) to determine runoff CN (Shamshad *et al.*, 2008). Specifically, the HSGs, which are A, B, C and D assigned to each soil type, are elements used in defining the minimum rate of infiltration. From A to D, soils have high, moderate, slow and very slow infiltration rates, respectively, even when thoroughly wetted (Table I).

Methods

SCS-CN method was used at 100 m spatial scale to simulate surface runoff, and the pollutant coefficients are combined to quantify NPS-N yields.

Surface runoff yield can be estimated by the SCS-CN method which is a widely used empirical model developed by the U.S. Department of Agriculture Natural Resources Conservation Service. A CN is assigned primarily based on soil permeability. CN is used to calculate the potential maximum retention S (Equation (1)), and then, runoff is calculated by combining S and precipitation (Mishra *et al*., 2006; Xiao *et al*., 2011):

$$S_i = 254\left(\frac{100}{CN_i}\text{-}1\right) \tag{1}$$

$$Q = \begin{cases} {}^{(P-0.2S)^2}\Big/_{(P+0.8S)} & P \geq 0.2S \\ 0 & P < 0.2S \end{cases} \tag{2}$$

where CN_i is a given CN value and S_i is the corresponding potential maximum retention (mm), P is the rainfall (mm), Q is the runoff quantity (mm) and S is the potential maximum retention (mm).

The initial CN values of agriculture land, forest and grass/pasture were cited from a literature (Zou *et al*., 2008) by which the SCS-CN model was developed for a small sub-basin of Dagu watershed in Jiaodong Peninsula. However, the study area of the article failed to cover the other kind of land use; therefore, two more literatures were cited (Huang *et al*., 2005; Xiao *et al*., 2011) to obtain the initial CN values of the other kind of land use.

Pollutant coefficients represent the concentration of a specific pollutant in runoff coming from a particular land use type within a watershed. Pollutant coefficients are reported as a mass of pollutant per unit volume of water (mg l^{-1}). In this study, an intensive literature review (Ying *et al*., 2010) and an in-depth comparison between Jiaodong Peninsula and four hotspot areas of China, including Miyun reservoir basin (Bao *et al*., 1997; Wang *et al*., 2002, 2003, 2004), Taihu lake basin (Li *et al*., 2004, 2006a, b, c; Li *et al*., 2007, 2009a, 2009b), Upper-middle Yangtze River basin (Liang *et al*., 2007; Long *et al*., 2008; Liu *et al*., 2009) and Jiulong River basin (Huang *et al*., 2004, 2005), had been carried out. The comparability in terms of geographical locations, climate conditions, topographic characters, vegetation, soil, land use and so on (Table II) were analyzed in order to pick out coefficients suitable for Jiaodong Peninsula. The coefficients of nitrogen pollutant for agriculture land, forest, grass/pasture, urban area, rural settlement and independent industrial-mining land in this study are selected as 3.95, 3.09, 2.91, 3.22, 3.58 and 1.26 mg .l^{-1}, respectively.

Table I. Hydrological soil groups in Jiaodong Peninsula

Hydrological soil groups	Soil texture	Infiltration rates
A	Sand, sandy loam	High
B	Silt loam, loam	Moderate
C	Sandy clam loam	Slow
D	Clay loam, silty clay, sandy clay, clay	Very slow

Calibration and validation

Surface runoff is the elementary factor to determine the transport of NPS pollutants. Therefore, it is important to calibrate and validate the SCS-CN model based on runoff records of hydrological stations. In detail, the trials and errors method is adopted to calibrate and validate the SCS-CN model until acceptable results are obtained. The assessing indicators of the closeness of the predicted to the observed values include coefficient of determination (R^2), relative error (RE) and Nash–Sutcliffe efficiency (NSE). R^2 refers to the degree of consistency between the simulated and the observed data and shows the proportion of the variance in observed data explained by the model. It varies from zero to one with higher values indicating higher consistency, and generally, values greater than 0.5 are acceptable (Santhi *et al*., 2001; Van Liew *et al*., 2003). RE describes the bias of the model and proves better modeling performance if it is close to zero. NSE is a statistical indicator that determines the relative magnitude of the residual variance compared to the observed data variance (Nash and Sutcliffe, 1970). It shows how well the plot of simulated data *versus* observed fits the 1:1 line. It ranges from ($-\infty$ to 1] with low values indicating high deviations between observed and simulated data. If the measured variable is predicted exactly for all observations, the NSE is 1. Negative NSE indicates unacceptable model performance. NSE and RE are defined as follows:

$$NSE = 1 - \left[\frac{\sum_{i=1}^{n}\left(Y_i^{obs} - Y_i^{sim}\right)^2}{\sum_{i=1}^{n}\left(Y_i^{obs} - Y_i^{mean}\right)^2}\right] \tag{3}$$

$$RE = \frac{\left(V - V^{`}\right)}{V^{`}} \times 100\% \tag{4}$$

where Y_i^{obs} and Y_i^{sim} are the ith observed and simulated value for the constituent being evaluated, respectively,

Table II. Comparison between Jiaodong Peninsula and four hotspot areas

Hot spot area	Geographical location	Topographic characters	Climate conditions	Vegetation, soil and land use	N concentration	Literatures cited
Miyun reservoir basin	40°19′–41°31′ N and 115°25′–117°33′ E, the total area is about $1.58 \times 10^4\,km^2$	Covered by highland and hills mainly, accounting for 79.7% of the whole basin	Warm-temperate semi-humid monsoon climate, annual precipitation amounts to 488.9 mm, and are mainly rainstorms	Temperate deciduous broad-leaved forest zone; mainly cinnamon soil; forest covers 25% of the area, grassland distributes widely, farmland is restricted to very limited area for water source conservation	Varies from $3.38\,mg.l^{-1}$ to $27.50\,mg.l^{-1}$ for different kinds of land use	Bao *et al.*, 1997; Wang *et al.*, 2002, 2003, 2004
Taihu lake basin	Located in the Yangtze River Delta hinterland, the total area is about $3.69 \times 10^4\,km^2$	Area of hills, plains and lakes accounts for 19.89%, 71.46%, and 8.65%, respectively	Subtropical monsoon climate, annual precipitation amounts to 1177 mm	Subtropical vegetation area; mainly paddy soil; paddy field, urban area and water area distributes widely, farmland covers 60% of the whole basin; a land of fish and rice	Varies from $0.27\,mg.l^{-1}$ to $6.04\,mg.l^{-1}$ for different kind of land use	Li *et al.*, 2004, 2006a, b, c; Li *et al.*, 2007, 2009b
Upper-middle Yangtze River basin	A macro spatial domain includes the Three Gorges Reservoir, Jialing river basin and so on	Mountainous area characterized by acute topographic	Subtropical monsoon climate, most area have annual precipitation more than 1000 mm	Evergreen broad-leaf forest, evergreen deciduous broad-leaved mixed forest; red soil, yellow soil, yellow brown soil and purple soil distributes widely; Grassland and forest cover most areas, very limited farmland could be found	Varies from $1.00\,mg.l^{-1}$ to $6.50\,mg.l^{-1}$ for different kind of land use	Liang *et al.*, 2007; Long *et al.*, 2008; Liu *et al.*, 2009
Jiulong River basin	24°23′–25°53′ N and 116°46′–118°2′E, the total area is about $1.47 \times 10^4\,km^2$	Mainly mountainous area with small plain in the lower reaches of the river	South subtropical maritime monsoon climate, annual precipitation varies from 1400 mm to 1800 mm	Subtropical vegetation zone; red soil, yellow soil and paddy soil distribute widely; forest covers more than 60% of the basin	Varies from $1.25\,mg.l^{-1}$ to $4.73\,mg.l^{-1}$ for different kind of land use	Huang *et al.*, 2004, 2005
Jiaodong Peninsula	35°48′–38° N and 119°48′–122°54′ E, the total area is about $2.82 \times 10^4\,km^2$	Covered by highland and hills mainly, accounting for more than 60% of the whole peninsula	Warm-temperate humid monsoon climate, the annual precipitation varies from 650 to 850 mm in different parts of the peninsula	Warm temperate deciduous broad-leaved forest zone; brown soil distributes widely; farmland covers more than 57.3% of the whole peninsula, forests mainly distribute on highland and hills		

Y_i^{mean} is the mean value of observed data and n is the total number of observation; V is the simulated values, while V' is the corresponding observed values.

Observed runoff records from 1972 to 1985 were split into two groups, 1972–1981 and 1982–1985 for calibration and validation purpose, respectively. In specific, parameters in SCS-CN model were calibrated based on the observed surface runoff at five hydrological stations from 1972 to 1981. Calibration was carried out through repeated trials and errors method until satisfactory results were obtained. Afterwards, the calibrated model was verified to estimate annual surface runoff from 1982 to 1985.

RESULTS AND DISCUSSION

Validation of the surface runoff estimation

CN values are the most important parameters of SCS model; after a lot of trials and errors, the acceptable SCS-CN model has been obtained, in which the final CN values accepted for Jiaodong Peninsula are shown in Table III. The estimated runoff by SCS-CN model *versus* the observed runoff of the two time segments was presented in Figure 2, and three variables for the measurement of the closeness between them were shown in Table IV. The R^2 reached 0.89 and 0.77 in the calibration period and the validation period, respectively, which were far beyond the acceptable levels. The values of RE were much lower in two periods, which shows that the bias of the model were lower. The NSE reached to 0.86 and 0.69 in the two periods, respectively, which further confirmed that acceptable performance had been achieved by the model, according to Moriasi *et al.* (2007).

The observed data of nitrogen is unavailable. However, we compared our result with the simulated result by Zou *et al.* (2008) who developed the AnnAGNPS model to simulate the NPS pollution in upper-middle part of Dagu river basin, the largest river basin in Jiaodong Peninsula. The simulated yield of NPS-N from 2000 to 2001 was demonstrated as 10.48 kg.hm^{-2}.a^{-1} by Zou *et al.* (2008), and our estimated result (10.18 kg.hm^{-2}.a^{-1}) was similar with it, which indicated that both the calibrated SCS-CN model and the pollutant coefficients are acceptable for NPS-N pollution simulation.

Table III. Final curve number assigned for Jiaodong Peninsula

Land use	Hydrological soil group			
	A	B	C	D
Agricultural	60	72	79	87
Forest	30	55	67	77
Grass/Pasture	30	48	60	71
Urban	89	92	94	95
Rural settlement	59	74	82	86
Independent industrial-mining	81	88	91	93

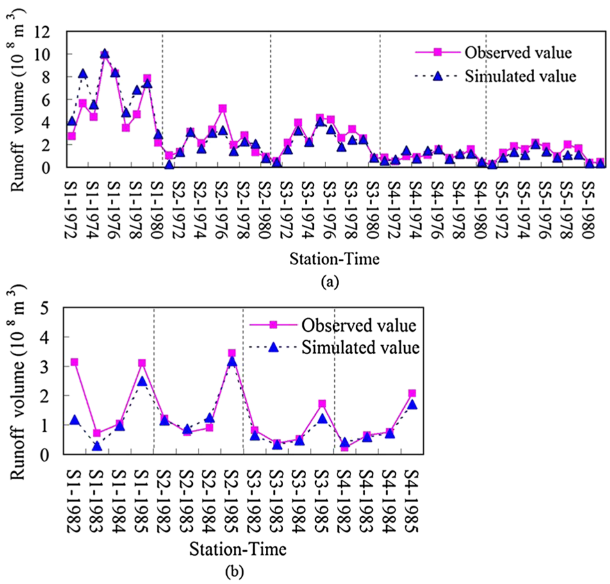

Figure 2. Comparisons of observed and simulated surface runoff for calibration period (a) and validation period (b)

Table IV. Statistical parameters of the surface runoff modeling

	R^2	RE	NSE
Calibration period (1972–1981)	0.89	22.96%	0.86
Validation period (1982–1985)	0.77	25.08%	0.69

Temporal variations of NPS-N yield

The annual NPS-N yield in Jiaodong Peninsula from 1979 to 2008 exhibited sharp inter-annual variations (Figure 3-a). It varied from $7.81 \times 10^6\, kg \cdot a^{-1}$ in 1981 to $51.30 \times 10^6\, kg \cdot a^{-1}$ in 2007, with average annual value of $20.35 \times 10^6\, kg \cdot a^{-1}$. A decadal increasing trend was apparent when dividing the time period into three 10-year periods (Table V). The increases of fertilizer use and poultry farm, as well as the intensification of human activities, are the fundamental reasons of the overall increasing trend of NPS-N yield.

Dagu watershed, Wulong watershed and Dagujia watershed are the three largest watersheds in Jiaodong Peninsula. They have long rivers, large spatial domain, perennial runoff regimes, and overall, they dominate the primary spatial-temporal characters of the annual NPS-N yield in the whole peninsula. The estimated NPS-N yield from 1979 to 2008 in the three watersheds also showed significant inter-annual fluctuations (Figure 3-b). Dagu

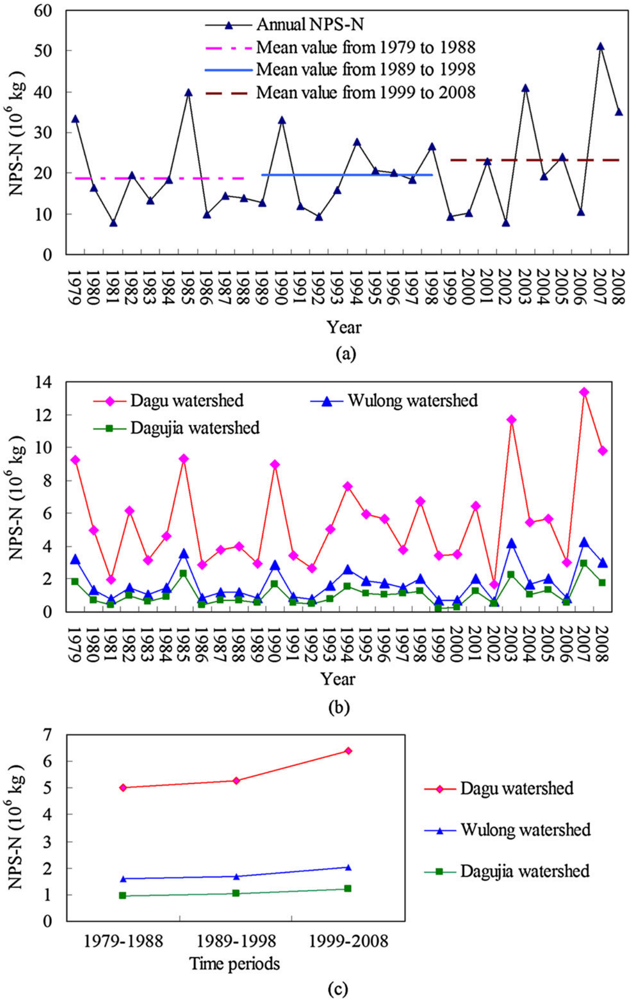

Figure 3. Temporal changes of NPS-N yield in Jiaodong Peninsula (a) and the three big watersheds (b, c)

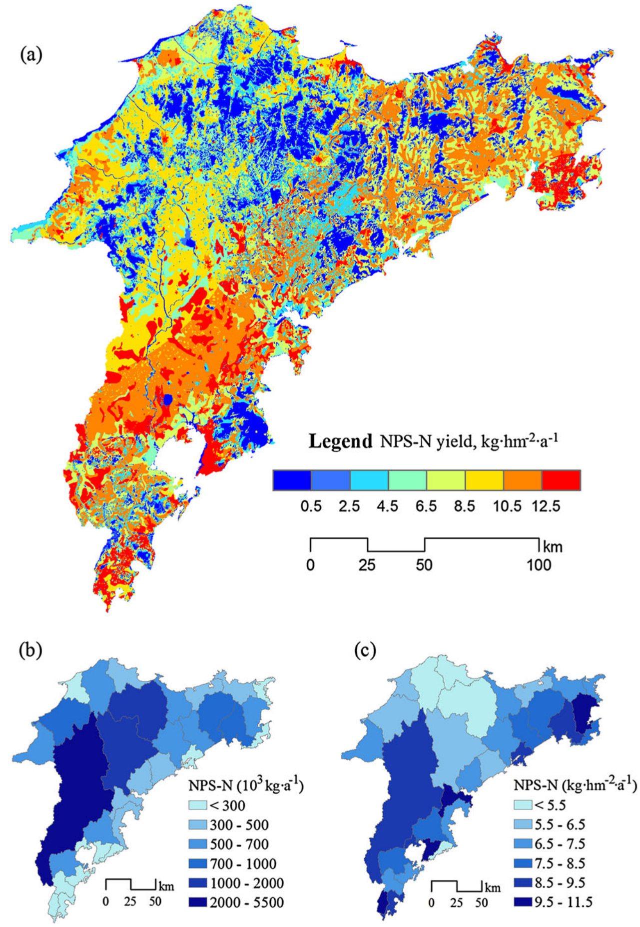

Figure 4. Spatial patterns of NPS-N yield from 1979 to 2008: average annual yield at pixel scale (a) and watershed scale (b), and per unit area average annual yield at watershed scale (c), respectively

Table V. NPS-N yield in three 10-year periods in Jiaodong Peninsula

Time range	1979–1988	1989–1998	1999–2008
Average annual NPS-N loads, 10^6 kg	18.69	19.64	23.16
Per unit area average annual NPS-N loads, $kg \cdot hm^{-2} \cdot a^{-1}$	6.69	7.04	8.29

watershed had the highest NPS-N yield among the three watersheds, with an annual average yield of 5.50×10^6 kg. Estimated annual average NPS-N yield in Wulong watershed and Dagujia watershed were 1.81×10^6 kg and 1.07×10^6 kg, respectively. The per unit area NPS-N yield in Dagu watershed, Wulong watershed, and Dagujia watershed were $9.05\,kg \cdot hm^{-2} \cdot a^{-1}$, $6.37\,kg \cdot hm^{-1} \cdot a^{-1}$, and $4.69\,kg \cdot hm^{-2} \cdot a^{-1}$, respectively. On decadal scale, the increase trend of NPS-N yield in all of the three watersheds was apparent (Figure 3-c). Differences in terms of agricultural intensification among the three largest watersheds determines the differences of NPS-N yield; in detail, Dagu watershed possess more preferable agricultural resource and therefore possess higher agricultural intensification than Wulong watershed and Dagujia watershed. Consequently, the most severe environmental impacts of land use are founded in Dagu watershed.

Spatial patterns of NPS-N yield

The average annual NPS-N yield for the 30 years was estimated and classified into eight levels for each grid cell over the Jiaodong Peninsula, which were < 0.5, 0.5–2.5, 2.5–4.5, 4.5–6.5, 6.5–8.5, 8.5–10.5, 10.5–12.5, and $>12.5\,kg \cdot hm^{-2} \cdot a^{-1}$ respectively (Figure 4-a). The per unit area annual average yield of the entire peninsula was $7.37\,kg \cdot hm^{-2} \cdot a^{-1}$. It was the main characters of the spatial pattern that the lower the altitude and the nearer to the sea, the higher the NPS-N yield. Low NPS-N yield was mainly found in high-altitude mountain regions where vegetation coverage (mainly forest, shrubbery and grassland) is intact. The area proportions of the three levels with higher NPS-N yield (8.5–10.5, 10.5–12.5, and >12.5 $kg \cdot hm^{-2} \cdot a^{-1}$) amounted to 14.88%, 22.07% and 9.52% respectively, and approximately 46.47% in total. These areas were mainly piedmont plains and valleys that widely distributed in coastal zones and low-altitude areas of Jiaodong Peninsula. The highest NPS-N yield mainly distributed in the southern and eastern, as well as urbanized coastal areas in Jiaodong Peninsula.

Average annual NPS-N yield (Figure 4-b) and its per unit area value (Figure 4-c) in each river basin in the past 30 years showed remarkable spatial differences among the river basins. For annual average yield, watershed size determined the spatial patterns of NPS-N yield in Jiaodong Peninsula, the larger the river basin, the higher the NPS-N yield. Among the 32 watersheds, the three largest watersheds had the highest average annual yields. For per unit area value of average annual yield, high yields were mostly found in the east and south part of the peninsula.

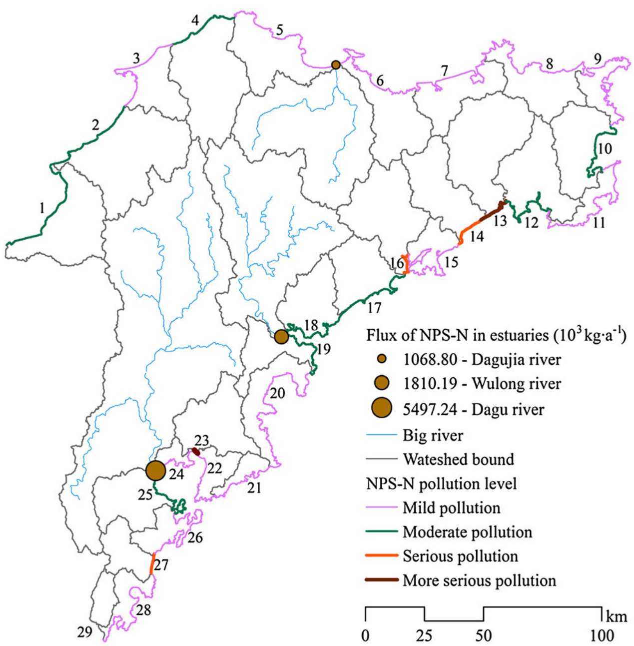

Figure 5. Spatial patterns of NPS-N flux to coastal waters through estuaries and shoreline segments

NPS-N flux to coastal waters through estuaries and shoreline segments

We divided the 32 watersheds into two groups according to their hydrological characters, one was the three largest watersheds that still have perennial runoff regimes, and the other was the 29 mesoscale and small-scale watersheds that were characterized by intermittent runoff (storm-water runoff mainly) in recent decades. Then, in the three largest watersheds, we defined the estuary flux of NPS-N as the basin-wide overall yields of NPS-N ($kg \cdot a^{-1}$). While in the 29 mesoscale and small-scale watersheds, we defined the flux of NPS-N as amounts of NPS-N that flows into coastal waters through shoreline along with the storm-water runoff, which was calculated approximatively as the basin-wide overall yields of NPS-N divided by the shoreline length ($kg \cdot km^{-1} \cdot a^{-1}$). Therefore, the average annual NPS-N fluxes to coastal waters of the 32 watersheds were ranked (Figure 5). As expected, NPS-N flux was positively associated with watershed size. The average annual NPS-N flux through shoreline segments of the 29 mesoscale and small-scale watersheds was further divided into four levels (Table VI). It shows that, most shoreline segments had mild and moderate pollution levels, while only a few shoreline segments had serious and more serious pollution levels. In specific, 14 segments whose length totally amount to 1458.35 km had mild pollution level, nine segments whose length totally amount to 557.08 km had moderate pollution level, three segments whose length totally amount to 49.80 km had serious pollution level and the three segments whose length totally amount to 25.12 km had more serious pollution level.

Table VI. Ranks of NPS-N flux to coastal waters through shoreline segments in Jiaodong Peninsula

Ranks of NPS-N pollution	NPS-N flux	Shoreline segments			Total area of upstream source area	Ratio of watershed area to shoreline length ($R_{W/S}$)
		Amounts	Total length	Length ratio		
	$10^3 kg \cdot km^{-1} \cdot a^{-1}$		km	%	km^2	$km^2 \cdot km^{-1}$
Mid pollution	<5	14	1458.35	69.77	5152.42	3.53
Moderate pollution	5–20	9	557.08	26.65	7107.61	12.76
Serious pollution	20–40	3	49.80	2.38	2075.15	41.67
More serious pollution	>40	3	25.12	1.20	2089.83	83.20
Summary	—	29	2090.34	100.00	16 425.01	7.86

The ratio of watershed area to shoreline length ($R_{W/S}$) is a very basic shape index of coastal watershed. It depicts the extensity of the up-stream area for a certain shoreline segment. Average value of $R_{W/S}$ in watersheds with the same NPS-N pollution level has been calculated (Table VI). It shows that the larger the $R_{W/S}$, the higher the NPS-N pollution levels. The scatter plots of NPS-N flux against $R_{W/S}$ for the 29 watersheds individually (Figure 6) further confirm a strong linear (positive correlation) relationship between these two variables. This character proves that the extensity of the up-stream area for a certain shoreline segment has very significant impacts on the levels of land-based diffuse pollution in the shoreline's adjacent coastal waters. We infer that, most probably, the rule revealed by these results is universally valid if there are limited differences of physical conditions and social-economic status among the up-stream area of different shoreline segments. At the same time, we think that more case studies are necessary to test the rule.

Levels of land-based NPS-N pollution in different sea areas

The coastal waters around Jiaodong Peninsula are spatially divided into four sea areas, including Bohai Bay, North-Yellow Sea, Jiaozhou Bay and South-Yellow Sea. Total NPS-N yields in their adjacent coastal watersheds had been summed up (Table VII), which depict the status of the land-based NPS-N pollutions in the four sea areas. It shows that, Bohai Bay had the lowest average annual loads of land-based NPS-N, but its land-based NPS-N load per unit area was higher than that in North-Yellow Sea. South-Yellow Sea had the largest average annual loads of land-based NPS-N, but its land-based NPS-N load per unit area was lower than that in Jiaozhou Bay. Overall, among the four sea areas adjacent to Jiaodong Peninsula, Jiaozhou Bay was the most noteworthy area not only because it had very serious land-based NPS-N pollution, but also because it was a nearly enclosed sea.

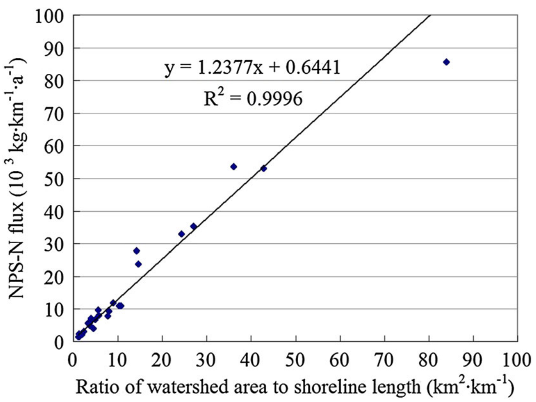

Figure 6. Scatter plots of NPS-N flux and Ratio of watershed area to shoreline length

Effects of land use structures on land-based NPS-N pollution of coastal waters

Land use structures had strong impacts on NPS-N pollution. Generally, watersheds with high proportions of farmland, rural settlement and urban built-up area had higher NPS-N yields, while watersheds with high proportions of forest and grassland had lower NPS-N yields. Jiaodong Peninsula has been one of the few areas in China with a high productivity in fruit and grains, and with a widespread distribution of farmland and orchard. The results of this study confirm that the spatial patterns of land use had significant impacts on watershed NPS-N yield and land-based NPS pollutions of coastal waters in Jiaodong Peninsula. Farmland accounts for 34.03%, 49.47% and 73.68% of the total area in Dagujia river basin, Wulong river basin and Dagu river basin, respectively. Accordingly, among the three river basins, Dagu river basin had the highest average annual NPS-N yield while the Dagujia river basin had the lowest (Figure 7-a). Furthermore, variation of land use structure among the 32 watersheds was one of the key control factors of the inconsistency between spatial patterns of NPS-N yield (Figure 4-b) and that of per unit area NPS-N yield (Figure 4-c). Among the four levels of NPS-N flux through the 29 shoreline segments (Figure 5, Figure 7-b), the NPS-N pollution levels and land use structure in its

Table VII. Levels of land-based NPS-N pollution in different sea areas around Jiaodong Peninsula

Sea area	Shoreline segment, estuary	Shoreline length, km	Catchment area, km^2	Load of land-based NPS-N, $10^3\,kg \cdot a^{-1}$	Land-based NPS-N load per unit area, $kg \cdot km^{-2} \cdot a^{-1}$
Bohai bay	1–4	256.60	3943.08	2385.46	604.97
North-yellow Sea	5–9, Dagujia estuary	571.99	5075.50	2915.00	574.33
South-yellow Sea	10–21, 26–29, Wulong estuary	1089.53	10 864.61	8111.48	746.60
Jiaozhou bay	22–25, Dagu estuary	172.22	7730.50	6939.04	897.62

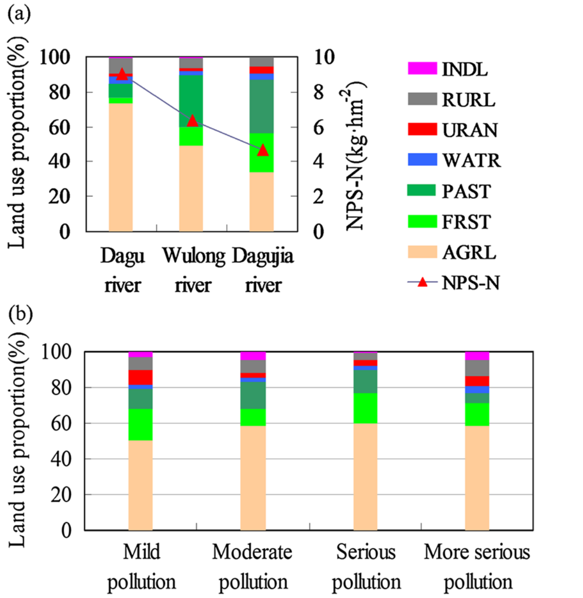

Figure 7. Correlations between land use structures and NPS-N pollution

watershed area were strongly correlated. Low proportion of farmland and high proportion of forest- grassland had mild pollution, high proportion of farmland and low proportion of forest-grassland had moderate pollution, highest proportion of farmland had serious pollution, while lowest proportion of forest-grassland and highest proportion of rural settlements had extreme serious pollution.

Moreover, data in some watersheds do confirm that more intensive watershed land use activities, mainly characterized by much higher proportion of built-up area and much lower proportion of forest-grassland had boosted the NPS-N pollution levels. For example, in the case of shoreline segment 13 and 16 whose $R_{W/S}$ was 53.11 and 53.67, respectively, the NPS-N pollution level ranked 'More serious pollution' and 'Serious pollution', respectively (Figure 5), which were inconsistent with the liner (positive correlation) relationship between NPS-N flux and $R_{W/S}$ (Figure 6). This inconsistency was mainly ascribed to the irrational land use activities in watershed area of shoreline segment 13. In detail, the proportion of farmland in watershed area of shoreline segment 13 was far more than that of shoreline segment 16, while the proportions of forest and grassland in the former were far less than that of the latter.

CONCLUSIONS

In this study, NPS-N pollution in Jiaodong Peninsula, China from 1979 to 2008 was estimated at 100 m spatial scale based on multi-source database and using spatial analysis techniques, SCS-CN method and pollutant coefficient method. The main conclusions are as follows:.

(1) The average annual NPS-N yield was about $20.35 \times 10^6\,\mathrm{kg \cdot a^{-1}}$ in the past 30 years in Jiaodong Peninsula. The annual yield exhibited a strong inter-annual variation and an increasing trend on a decadal scale. Overall, peninsula's mountainous regions with high vegetation cover had low NPS-N yields, while southern and eastern parts of the Peninsula, as well as urbanized coastal areas, contributed to most of the NPS-N yields.
(2) At the watershed scale, spatial patterns of total NPS-N yield were mainly determined by the watershed size. Larger river basins usually have a higher total NPS-N yield. However, per unit area NPS-N yield in each watershed showed a more complicated spatial pattern because land use structures become one of the main impact factors. Watersheds with high proportions of farmland and urbanized areas and low proportions of forest and grassland were characterized by much higher NPS-N yield in Jiaodong Peninsula.
(3) Large river basins usually have large NPS-N fluxes to estuaries. The NPS-N flux from land to coastal waters through shoreline segments in the 29 mesoscale and small-scale watersheds exhibited complicated spatial patterns and temporal variations. The ratio of watershed area to shoreline length ($R_{W/S}$) was a fundamental controlling factor of the shoreline NPS-N flux. In large-scale coastal areas lack of observed water quality data, the rule revealed by the scatter plots of NPS-N flux against $R_{W/S}$ will support us to find coastal waters with high-risk levels of land-based NPS pollutions. Furthermore, land use structure affected or modified the levels and spatial patterns of shoreline NPS-N flux. Watersheds with intense land use tend to yield large NPS-N fluxes.

The implications or inspirations from this study are as follows. (1) Watershed- or river-basin-based evaluations on long-term NPS pollution in coastal areas are critical for both LOICZ studies and ICZM practices because differences of NPS pollution and their controlling factors among watersheds are very significant. As a result, the combination between ICZM and integrated river basin management (IRBM) in coastal zone is highly recommended. (2) NPS pollutants fluxes through shoreline into coastal waters in mesoscale and small-scale coastal watersheds with seasonal rivers are mainly affected by storm water pollutions. Furthermore, watersheds with a high $R_{W/S}$ (ratio of watershed area to shoreline length) should be paid more attentions in terms of hydrological and environmental monitoring and IRBM practices. (3) The main source areas of NPS pollutants should be taken as the key targets of 'pollution source control practices', and on the other hand, watershed land use planning and landscape designing should be considered as primary approaches of prevention and mitigation of water pollutions in coastal zones.

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