SYNTHESIS AND EMERGING IDEAS

Nitrogen sources and exports in an agricultural watershed in Southeast China

Nengwang Chen · Huasheng Hong · Luoping Zhang · Wenzhi Cao

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Abstract The nitrogen (N) budget was developed for Jiulong River Watershed (JRW), an agricultural watershed in a warm and humid area of southeast China. Water quality monitoring, field surveys, modelling and GIS techniques were applied to estimate N flux of atmospheric deposition, mineralization, runoff, denitrification, and ammonia volatilization. Over the whole watershed, fertilizers, import of animal feeds, biotic fixation, mineralization and atmospheric deposition contributed 67.1%, 16.5%, 2.1%, 4.9% and 9.5%, respectively, of total N input (129.3 kg N ha⁻¹ year⁻¹). Runoff, sale of production, denitrification, and ammonia volatilization contributed 7.3%, 24.4%, 10.5% and 57.8% of total N output $(72.9 \text{ kg N ha}^{-1} \text{ year}^{-1})$, respectively. The N budget for the JRW suggested that more than 50% of the N input was lost to the environment, and about 14% was discharged as riverine N, which indicated that agricultural and human activities in the watershed substantially impacted the estuary and coastal water quality, and so altered the N biogeochemistry process.

Keywords Nitrogen · Sources and exports · Southeast China · Watershed budget

Abbreviation

JRW Jiulong River Watershed

Introduction

Human induced input of reactive nitrogen (N) into the global biosphere has markedly altered N cycling in nature (Galloway et al. 1995; Vitousek et al. 1997). The estimation of watershed fluxes of pollutants provides a powerful tool for understanding changes associated with human activities. Several examples of watershed-scale N budgets include studies of N cycling in the northeastern U.S.A and its watershed (Breemen et al. 2002) and in China's major watersheds (Xing and Zhu 2002). The main anthropogenic sources of N pollution are related to fertilizer application, waste production and fossil fuel burning. The excessive use of commercial inorganic fertilizers for raising crop yield and meeting the demand of population growth in China has led to increased nutrient additions. In southeast China, anthropogenic N inputs far exceeded N outputs in products shipped out of agricultural ecosystems (Cao and Zhu 2000). However, the sources and fate of this excess N at

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watershed level under humid temperate climatic conditions of Asia is poorly understood. Research on nitrogen-water quality linkages should be broadened to address issues of N losses and watershed-scale N balance.

In this study, the N sources and exports were quantified for the Jiulong River Watershed (JRW), an agricultural hilly watershed in the coastal area of southeast China. Water quality monitoring, field surveys and GIS techniques were linked to estimate N flux of atmospheric deposition, runoff, denitrification, and ammonia volatilization. A pilot study of N sources and exports has been finished at village-scale in the catchment of the JRW (Cao et al. 2006). The current work developed an N budget and provides a further understanding of the sources of N to the land-scape and the associated N fluxes in the exports, and highlights research showing how anthropogenic activities impact N cycling in this coastal watershed.

Materials and methods

Study area

The JRW is a mesoscale watershed (14,700 km²) located in southeast China (Fig. 1). The main stem of the Jiulong River is formed by the confluence of two major tributaries and it discharges about 14 billion m³ year⁻¹ of water into the Xiamen sea. Annual precipitation varies from 1400 to 1800 mm, 75% of which occurs between April and November. The watershed is mainly level (21% with altitude <200 m) but with hillslopes in upstream regions, comprising 67% forest, 17.5% arable land, 3% residential land, 5% bare land, and 7.5% water body. 81% of watershed is plain with slope less than 30°. Red earth, lateritic red earth and yellow earth are the main soil types, contributing about 86% of total area with organic matter content ranging from 0.7% to 5.2%. Banana (79,380 ha), Litchi (3890 ha), longan (4410 ha), citrus (2940 ha), rice (14,500 ha), and vegetables (36,750 ha) are the main crops in the watershed. Crops are generally grown in the wet season. Typically, about 300–1100 kg ha⁻¹ of N (<10% is manure) are applied annually as fertilizer on crops lands. For JRW, there is increasing impact on water quality due to extensive soil erosion and nutrient discharges from intensive agricultural activities and animal feeding.



A biogeochemical budget was established for the JRW. N input included atmospheric deposition, mineral fertilizers, import of feeds, biological fixation, and potential mineralizable N from the soil organic matter pool. Planted seeds were not considered in this study since they often account for a very small portion of N input. Output items included sale of crops and animal production, river discharge, denitrification and ammonia volatilization. The present study is primarily give N inputs and outputs pattern rather than biogeochemical behavior within watershed, e.g., animal consumption of crops and manure N recycled on arable lands. Although the average N leaching loss below the rooting zone was 13.4 kg N ha⁻¹ year⁻¹ over a 9.4 km² catchment, representing about 3.8% of the total N inputs (Cao et al. 2006), for a large scale area, e.g., 14,700 km² in this study, the N leaching is assumed to be equal to the return to streamflow through baseflow, and the seepage N into estuary and sea is minimum. In N output items, river discharge N calculation based on water quality data and the relevant flows in the river outlet (Fig. 1) can be assumed involving both N in surface runoff and leaching N from upstream regions.

All calculations were conducted for 2004. The basic data concerned in N budget calculation were determined from long term surveys and monitoring, experiments, statistics, as well as literature values.

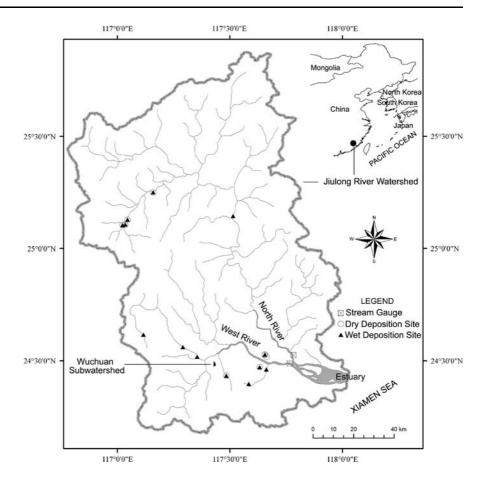
Sources of N input

Fertilizers

The town scale statistical data of mineral N fertilizers, i.e., ammonium bicarbonate, urea and mixed fertilizer, was obtained from the Statistics Bureau of Fujian Province (SBF 2005). These data were then multiplied by the percentages of N in each type of fertilizer used. The total amount of N input from fertilizers to the JRW was calculated by summation of the above town data. Transboundary towns were excluded in calculation since they cover small area and most are forest land. Input N flux from the application of mineral fertilizers averaged 86.7 kg N ha⁻¹ year⁻¹ on an areal basis over the JRW.



Fig. 1 Location of Jiulong River watershed and sampling site of atmospheric N and precipitation. Stream gauging station located in the outlet of North River and West River



Import of animal feeds

Animal feedlots were expanded in recent years to meet the increasing demand for animal production by the growing population of China. As of 2004, a total of 3.6 million swine were present in the JRW (Fig. 2) and 108 million ton of animal feeds (mainly soybean and maize) were imported to the watershed. N inputs from feeds considered swine only, since other animals consume mainly crop harvests from the watershed rather than from imports.

Biological fixation

The main source of biological N fixation is through the growing of legumes, such as soybean (*Glycine max*) and peanut (*Arachis hypogaea*). N inputs from biological fixation considered both symbiotic N-fixation crops (peanut and soybean only, since the area under other symbiotic N-fixation crops was very small in the JRW) and nonsymbiotic N-fixation crops in rice plantations and other cropland areas. Data on the area of various crops were extracted from the Statistics Yearbook (SBF 2005). Assumed average N-fixation rates of each crop are listed in Table 1. Total N inputs to agricultural lands from biological fixation were then estimated by multiplying the area of each crop by the relevant N-fixation rates. The actual N input of biological fixation could be higher if the forest fixation was included, although the fixation rate in forest lands is very low. An assumed fixation rate of 1 kg N ha⁻¹ year⁻¹ was used in the regional N budget studies in South Korean agricultural areas (Bashkin et al. 2002). The area of forest in JRW is 984,900 ha, then the fixation in forest equals to 984.9 t year⁻¹. Inputs from total biological fixation averaged 2.7 kg N ha⁻¹ year⁻¹ in the JRW.

Mineralizable N

Potentially mineralizable N from the soil organic matter pool constitutes a substantial source of N (Zech



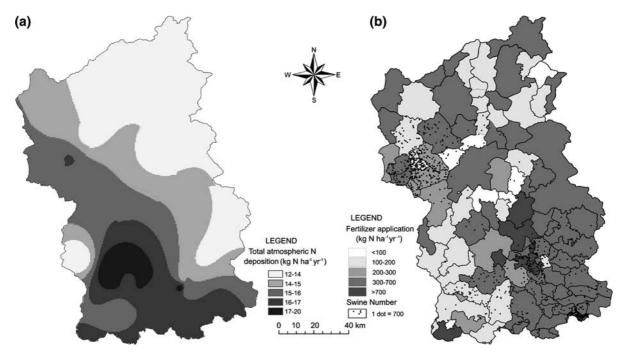


Fig. 2 (a) Spatial distribution of deposition flux of atmospheric nitrogen in Jiulong River Watershed, (b) Fertilizer application and swine feeding over Jiulong River Watershed. Data on

fertilizer application rate on crops land was calculated but visualized at county basis in GIS map

et al. 1997). It is particularly true in places where soil organic matter content is high and climate is favorable. Previous study estimate the mineralizable N in JRW to be 9230 ton in a year, based on a statistical model and a GIS soil map layer with soil properties (i.e., bulk density and organic matter content of soil). Details of modeling approach can be seen in Cao et al. (2005). It is noted that only arable and horticultural lands were used for mineralization estimate due to frequent tillage, and continuous additions of fresh organic matter (crop residue). Mineralization and denitrification over nonagricultural lands were thought to be equivalent.

Atmospheric deposition

Input of N from total atmospheric deposition (dry and wet) was calculated using GIS interpolation technique based on recorded data from nine dry deposition sites and 13 wet deposition sites over the JRW (Fig. 1). ArcView GIS 3.2 (ESRI, California) was used for surface analysis, interpolation and statistical work. After cross validation, Universal Kriging method and

IDW (Inverse distance weighted technique) was selected to estimate precipitation and N, respectively. During 2004–2005, monthly dry samples (mainly particulate N, ammonium and nitric vapor) were continuously collected using a water surface method (Balestrini et al. 2000). On the day after collection, aliquots of water samples containing dry depositions were taken to the laboratory where different N species were measured. Based on the sum of monthly data in each site, the annual dry atmospheric N deposition was interpolated and quantified for the JRW. As of 2004, a total of 642 rain samples were collected in thirteen sites during storm events using a 35 cm polyethylene funnel fitted on a 151 polyethylene bucket, and analyzed for ammonium N, nitrate N and dissolved total N (DTN) followed by filtration through 0.45 µm nucleopore membranes. Wet deposition flux was calculated using selected GIS interpolation technique based on monthly mean DTN concentrations and precipitation records from eight weather stations within the JRW. In order to maintain comparability with N flux estimates, only the results for 2004 are included in this article.



Table 1 Coefficients for estimation of nitrogen budget

N sources/exports	Items	N contents	Calculation basis	References
Biotic fixation	Peanuts	86 kg N ha ⁻¹ ·year ⁻¹	Land area	Barry et al. (1993)
	Soybean	$78 \text{ kg N ha}^{-1} \text{ year}^{-1}$	Land area	Barry et al. (1993)
	Paddy	$45 \text{ kg N ha}^{-1} \text{ year}^{-1}$	Land area	Zhu (1997)
	Other crops	$15 \text{ kg N ha}^{-1} \text{ year}^{-1}$	Land area	Zhu (1997)
Feeds import	Swine	29 g kg^{-1}	Fresh weight	Personal communication
Products sale	Swine	25 g kg^{-1}	Fresh weight	Fortin and Elliot (1985)
	Banana	2.24 g kg^{-1}	Fresh weight	Chen (2002)
	Citrus	1.28 g kg^{-1}	Fresh weight	Chen (2002)
	Litchi	1.44 g kg^{-1}	Fresh weight	Chen (2002)
	Vegetable	2.88 g kg^{-1}	Fresh weight	Chen (2002)
	Sugarcane	$1.0 \; \mathrm{g \; kg^{-1}}$	Fresh weight	Cao and Zhu (2000)
	Rice	$31 \text{ kg N ha}^{-1} \text{ year}^{-1}$	Land area	Huang et al. (1995)
	Peanut	$154 \text{ kg N ha}^{-1} \text{ year}^{-1}$	Land area	Huang et al. (1995)
	Bean	$89 \text{ kg N ha}^{-1} \text{ year}^{-1}$	Land area	Huang et al (1995)
Denitrification	Banana	$17.3 \text{ kg N ha}^{-1} \text{ year}^{-1}$	Land area	DNDC Simulated data
	Vegetable	$41.0 \text{ kg N ha}^{-1} \text{ year}^{-1}$	Land area	DNDC Simulated data
	Fallow	16.1 kg N ha ⁻¹ year ⁻¹	Land area	DNDC Simulated data
	Paddy	29.3 kg N ha ⁻¹ year ⁻¹	Land area	Xu et al (1997); Xing (1998)
	Forest	$2.0 \text{ kg N ha}^{-1} \text{ year}^{-1}$	Land area	Brumme et al. (1999); Hahn et al. (2000); Henrich and Haselwandter (1997)
	Ponds/water	$10 \text{ kg N ha}^{-1} \text{ year}^{-1}$	Land area	Sjodin et al. (1997)

Exports of N output

Sale of crops and animal production

The total N output in terms of crop harvests and animal production was estimated using N contents and yield values which were shipped out of the system. Harvest yield and shipped value was extracted from statistical yearbook (SBF 2005). The total N content in crops, fruits and animal production was collected from different sources (Fortin and Elliot 1985; Huang et al. 1995; Cao and Zhu 2000; Chen 2002, see Table 1). The total N in vegetation was assumed to be 2.88 g kg⁻¹, because production from diverse vegetable species was summed as an overall production rather than itemized in the yearbook. Animal production including swine, cattle, and mutton have a wide range of N content, but 0.25 g kg⁻¹ was assumed for the N harvest calculation in animal production (Fortin and Elliot 1985). Other crops and animals products were not considered in the calculation because of their small sales.

Runoff

The amount of riverine N output was flow-weighted, and estimated by multiplying measured concentration of DIN (nitrate N + nitrite N + ammonium N) in two river outlet by the relevant flows. As of 2004, bimonthly water quality data and monthly flow data obtained from local government were used to calculate annual DIN discharge. Total discharged N was scaled-up according to the finding that DIN comprises approximately 75% of the total N exported from the Wuchuan subwatershed, a small representative agricultural watershed set up for nutrient pollutant research in the late 1990s (Cao et al. 2003; Chen et al. 2006) as well as other five subwatersheds in the JRW (Chen 2006).

Denitrification

N losses due to denitrification was calculated using land use map and DNDC model (Li et al. 1992a, b). Before this calculation, the DNDC model was



calibrated and validated using plot experiment data (i.e., denitrification rate, texture of soils, pH, bulk density). The plot experiment was carried out in the Wuchuan subwatershed. The denitrification rate under banana, vegetable, fallow, forest were examined monthly using acetylene-inhibition technique (Ryden et al. 1987). Traditional crops management practices including irrigation and fertilization (mineral fertilizer and some animal waste) carried through the whole plot experiment period. Denitrification rates measured on soil cores were expressed on an areal basis, and extrapolated over the period between sampling date to estimate annual N loss by denitrification. The Nash-Sutcliffe coefficient E (Nash and Suttcliffe 1970) values were used to determine goodness of fit between predictions and the observed values. The predicted denitrification rates under banana, vegetable, fallow well matched the observed with E values >0.90 during calibration, and equal to 0.77, 0.72 and 0.87 during validation, respectively. A very low E value of 0.4 was found under forest, so the denitrification rate under forest and paddy, ponds/ water were estimated based on the reported data (Xu et al. 1997; Xing 1998; Henrich and Haselwandter 1997; Sjodin et al. 1997; Brumme et al. 1999). The amount of gaseous N output was estimated by multiplying the area of land use by the relevant denitrification rates (Table 1).

Ammonia volatilization

Ammonia volatilization is an important pathway of N loss. Fertilizer application, confined animal feeding operations, and manure application on fields contribute most to ammonia emission (Goebes et al. 2003). Gaseous N loss rate through fertilizer application primarily varies with fertilizer types and application method (Burkart and James 1999). Ammonium bicarbonate, urea, and mixed fertilizer were the main N fertilizers applied in the JRW (Chen et al. 2006). Recent field experiments showed that the ammonia loss rate through volatilization from urea in a paddy field in eastern China varied from 5% to 37% of total applied N, depending on when fertilizer was added and crop-growing periods (Li et al. 2001; Tian et al. 2001). Our plot experiments under banana and fallow in the Wuchuan subwatershed showed that ammonia N losses ranged between 20% and 44% of total applied ammonium bicarbonate N, and about 20% of total applied mixed fertilizer N. In view of the above, an annual average loss rate of 25% was assumed for ammonia volatilization from ammonium bicarbonate and urea, and 18% for mixed fertilizer, according to land use component, actual conditions of soil, climate, tillage practices and application method (most surface spreading) in the JRW. Ammonia volatilization from confined animal farm was estimated using the emission factor corresponding to each type of animal, i.e., swine 3.8, cattle 24.1, sheep 3.1, and poultry 0.24 kg N capita⁻¹ year⁻¹ (Sun and Wang 1997). Rate of ammonia volatilization after manure application to the field primarily varies with manure type and application method (Sommer and Hutchings 2001). The average amount of swine manure applied to agricultural land was investigated using statistical data from farmers and local government. About 30% of the applied ammonium N in manure was assumed to be lost through volatilization according to previous studies (Chambers et al. 1997; Huijsmans et al. 2003). The total N output from ammonia volatilization was then obtained by summation of the above three sources of gaseous N loss.

Results and discussion

Sources of N input

The N budget was established as shown in Table 2. Overall, on an annual basis, the area-weighted, mean input rate of N over the JRW was 129.3 kg ha⁻¹. Inputs from the application of mineral fertilizers averaged 86.7 kg ha⁻¹, which was the maximum input source in the budget (67%). The actual N application rate in agricultural lands was 495.5 kg ha⁻¹, approximately 409 kg N ha⁻¹ higher than the previous estimate for the whole watershed. Fertilizers application varied spatially and temporally among the different land uses. Most of the inorganic fertilizers were applied to cash crops such as bananas and longan, and growing season (between April and September) received the highest application rates of fertilizers. Historically, the mean N application rate for the whole of China was very low (Zhu 1997). In the Taihu Lake region in east China, the application rate of N only from manure ranged between 70 and 100 kg N ha⁻¹ year⁻¹ in the 1950s. However, since the 1980's, mineral fertilizers have been widely used,



Table 2 Nitrogen budget for the Jiulong River Watershed

N budget	Ton N year ⁻¹	N Flux (kg N ha ⁻¹ year ⁻¹)	Percent of input (%)
Inputs (I)			
Fertilizers	127,460	86.7	67.1
Feed	31,296	21.3	16.5
Biological fixation	3,961	2.7	2.1
Mineralizable N	9,230	6.3	4.9
Deposition	18,089	12.3	9.5
Subtotal	190,036	129.3	100
Outputs (O)			
Production sale	7,831	5.3	4.1
Runoff	26,138	17.8	13.8
Denitrification	11,258	7.7	5.9
Ammonia volatilization	61,937	42.1	32.6
Subtotal	107,164	72.9	56.4
Budget (I–O)	82,872	56.4	43.6

and so N application rates have increased greatly and recently varied from 500 to 800 kg N ha⁻¹ on different crops (Ellis and Wang 1997). In JRW, fertilizer application is a dominant source of N inputs to agricultural ecosystem.

Followed by fertilizer application, animal feed import and atmospheric deposition accounted for 16.5% and 9.5% of total N inputs, with annual N fluxes of 21.3 kg N ha⁻¹ and 12.3 kg N ha⁻¹, respectively. The growing number of confined animal feedlots required more feed import, and subsequently, larger amounts of manure and excrement were produced, which resulted in gaseous N losses and environmental problems. Study of atmospheric N deposition indicated that higher ammonia volatilization from fertilizer application in the growing season, and animal feeding, together provided the largest N source to atmospheric deposition (Fig. 2).

The annual deposition of atmospheric N flux amounted to $12.3 \text{ kg N ha}^{-1}$, of which dry deposition and wet deposition accounted for 34% and 66%, respectively. Approximately 80% of atmospheric deposition occurred in spring and summer, indicating that large gaseous N losses were highly correlated with intensive fertilization during the growing season, and ammonia volatilization from animal manure induced by high temperature during this period. The annual wet deposition ratio of about 8.1 kg N ha^{-1} in

the JRW is fairly higher than 6.23 kg N ha⁻¹ in the Chesapeake Bay watershed (Sheeder et al. 2002), 3.9–5.2 kg N ha⁻¹ in the Delaware Inland Bays (Scudlark et al. 2005), and 5.2 kg N ha⁻¹ in the southwest France (Rimmelin et al. 1999). However, this estimated value is consistent with 7.5 kg N ha⁻¹ over rice field in Jiangshu porvince in east China (Su et al. 2003), but less than the field measurement results elsewhere in China, 4–23 kg N ha⁻¹ in 16 national experimental sites (Shen 1998), 6.3–26.6 kg N ha⁻¹ during 1990–1994 in westnorth China (Li and Li 1999), and 35.57 kg N ha⁻¹ over forest in Guangdong province. This may be explained by the great spatial-temporal variation of atomospheric N deposition.

Estimated mineralizable N flux $(6.3 \text{ kg N ha}^{-1})$ contributed approximately 4.9% of the N sources. N fixation $(2.7 \text{ kg N ha}^{-1})$ also contributed a small proportion (2.1%) of the overall N input due to the minimal sown areas of legume crops in the watershed. Estimated N fixation in agricultural lands and in forests shared 75% and 25%, respectively.

The total annual N input in the JRW was lower than the rate of 350 kg N ha⁻¹ over a small scale watershed (9.4 km²) within the JRW (Cao et al. 2006). It was also lower than the input rate of 534 kg N ha⁻¹ in another small agricultural watershed, Liuchahe watershed, with an area of 7.32 km² in mid China (Yan et al. 1999). The N inputs flux may be varied significantly due to land use component, agricultural practices and human activities. However, this fertilizer-dominated input pattern can be found among watersheds at different scales in different regions (Bashkin et al. 2002; Breemen et al. 2002; Xing and Zhu 2002).

N outputs

The mean output rate of N over the JRW was 72.9 kg ha⁻¹. Among total N outputs from the JRW, ammonia volatilization was the maximum (57.8%), followed by runoff (24.4%), denitrification (10.5%), and sale of crops/animal production (7.3%).

The major output from the watershed is estimated to be ammonia volatilization to the atmosphere. Fertilizer application, confined animal feeding operations, and manure application to arable land accounted for 50.2%, 45.4%, and 4.4%, respectively, of total volatilize N (61.9×10^3 ton N) in the JRW. Although manure application on fields results in



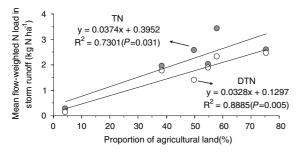


Fig. 3 Relationship between mean flow-weighted N load in stormflow and proportion of agricultural land within the watershed. Six circles in figure represented six subwatersheds with different land use component

ammonia emissions much lower than from commercial fertilizer due to lower rates of manure application to fields, manure from confined animal feeding operations can be significant. Ammonia gas is pretty reactive during its transport in the atmosphere, and most reacts primarily with acidic species to form ammonium sulfate, ammonium nitrate or ammonium chloride, or it may be deposited to the earth's surface by either dry or wet deposition processes (Aneja et al. 2001), hence the N output in ammonia volatilization was not really a loss from the JRW. The exact amount of volatilized ammonia deposited back to the watershed as part of N input depends on the distance from the site of emission to the boundary of the JRW.

A high riverine N export of 17.8 kg N ha⁻¹ year⁻¹ for the Jiulong River can be in response to the great N input over the watershed. Study on N losses and concentrations in Finland during the period 1981–1997 showed that total N loss in three agricultural catchments (average 15 kg N ha⁻¹ year⁻¹) was higher than in nine forested catchments (average 2.5 kg N ha⁻¹ year⁻¹) (Vuorenmaa et al. 2002). On a per area basis, the export of total N varied according to land use and was significantly correlated to the net input of anthropogenic N (Filoso et al. 2003). Mean N exports during storm event in six experimental subwatersheds within JRW showed that riverine N export may be significantly correlated to the proportion of agricultural lands (Chen 2006, see Fig. 3).

The current estimated denitrification rate was $7.7 \text{ kg N ha}^{-1} \text{ year}^{-1}$ over the whole watershed whereas the N loss by denitrification in agricultural lands accounts for about 6.5% of the applied N fertilizer. The DNDC model simulated denitrification under vegetable $(41.0 \text{ kg N ha}^{-1})$ and banana (17.3 kg)

N ha⁻¹) in this study are fairly higher than the large amount of field experimental data elsewhere in China (Xing 1998). This can be explained as great denitrification variation among the different land uses, soils as well as climate conditions.

Total N input to the JRW was 129.3 kg N ha year⁻¹ of which 4.1% was accounted for production (sales only), 13.8% was discharged in runoff, 5.9% was denitrified, 32.6% was lost through ammonia volatilization, and the remaining 43.6% accumulated in the landscape (Table 2).

N budget

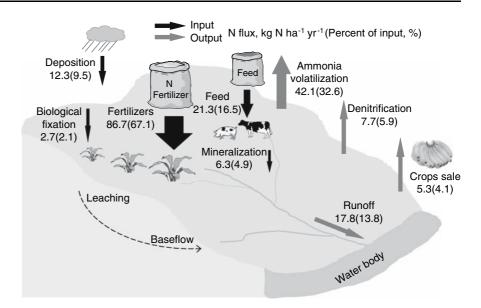
The N budget for the JRW was positive, with inputs exceeding outputs by 56.4 kg N ha⁻¹, with a high fraction (43.6%) of total N input (Table 2). The accumulation of N in the landscape was due to the rates of fertilization, atmospheric deposition, and N imports that were not matched by output rates in river discharge, gaseous losses, and sale of crops/animal production. The N inputs/outputs patterns of each watershed vary widely, but inputs were consistently higher than outputs in agricultural watersheds (Vitousek et al. 1997).

The N budget for the JRW suggested that more than half of the N input was lost to the environment (air and water). As to the fate of the N input, previous studies indicated that the flux of N in large rivers is well explained as a function of the net anthropogenic inputs of N to the landscape (Howarth et al. 2006). Riverine N export estimated in this study was a fairly low fraction (about 14% of input N) compared to the findings that average 20–25% of the anthropogenic N inputs are exported by surface runoff in large rivers in North America and Europe (Howarth et al. 1996; Boyer et al. 2002; Breemen et al. 2002), and 40% to 45% of the N inputs are exported in the most wet watersheds in northeastern United States (Howarth et al. 2006). For the low fraction of riverine N export compared to the other regions, authors argued that there should be a considerable deposition to sediment, assimilation to plants as well as other biogeochemistry process in such a warm climate zone which resulted in a decreasing actual N output through runoff in the JRW.

The riverine N discharge calculated (26,138 ton N year⁻¹) in this study were apparently greater than the two previous estimates, 6000 ton N year⁻¹ by



Fig. 4 Nitrogen cycling in the Jiulong River Watershed



Chen et al. (1993) and 12,600 ton N year⁻¹ by Zhang (1996). Although these estimations are subject to considerable uncertainties and weak comparability because of complexity of N cycles, authors suggest a need to better consider the influence of humanderived N export as well as climate change on riverine N fluxes as part of management efforts to control coastal N pollution and excessive eutrophication. While the fate and accumulation rate of this excess N in the environment is poorly understood for many regions of the world (Galloway 1998), the current study indicated that the excess anthropogenic N inputs increased N storage in landscape and gradually increased riverine N discharge to coastal areas.

N crop uptake efficiency in China is less than in European countries (Zhu 1997). The JRW is very low with <15% efficiency, even if all crop harvests (e.g., seeds, stocks and economic parts) were included in the calculation (data not shown). The large amount of surplus N in the budget supports the idea that an increasing non-sustainability has occurred within agriculture, and the nutrient management complex. N accumulated in arable land at a high rate caused soil concretion, acidification, and resource waste, and subsequently led to a disturbance of the nutrient cycle in the agroecosystems.

As a graphical representation, the N input/output pattern illustrated in Fig. 4 can be a useful tool for easily identifying pollution pathways and finding nutrient management strategy at the watershed scale.

In general, fertilizer dominated the anthropogenic N input pattern. Ammonia volatilization and riverine N exports were the main pathways of excess N loss, and the main factor affecting environmental quality as well as sustainability of agriculture. Reducing N inputs through proper management of fertilization and tillage practices in the growing season, and intercepting storm runoff in the wet season by the installation of Best Management Practices (wetland, multiponds, terrace, buffer strips and riparian vegetation, etc.) would be useful to mitigate N pollution in the JRW and coastal marine ecosystem.

Uncertainty analyses

There can be unavoidable uncertainty associated with calculations of N flux arising from uncertainty in the variables used in the calculation. In this study, careful data checking and meticulous model validation decrease the uncertainty of N budget. Mean values (i.e., N contents in products, gasous N loss from fertilization, etc.) from different reported data were used to find a balance between the watershed complexity and the quality of the data available needed during N budget estimation. Whereas fertilizers and ammonia volatilization contributed the largest percent of N input/output, and obviously was a sensitive variable in N budget calculation, the fact of human influence on N sources/exports pattern was exactly revealed in the JRW.



Conclusions

In the JRW, fertilizers, import of animal feeds, biotic fixation, mineralization and atmospheric deposition contributed 67.1%, 16.5%, 2.1%, 4.9% and 9.5%, respectively, of total N input $(190 \times 10^3 \text{ ton N})$ 29.3 kg N ha⁻¹ year⁻¹). Riverine discharge, sale of crops and animal production, denitrification, and ammonia volatilization contributed 7.3%, 24.4%, 10.5% and 57.8% of total N output $(107 \times 10^3 \text{ ton N})$, 72.9 kg N ha⁻¹ year⁻¹), respectively. The N budget for the JRW suggested that more than 50% of the N input was lost to the environment, and about 14% was discharged as riverine N, which indicated that agricultural and human activities in the watershed substantially impacted the estuary and coastal water quality, and so altered the N biogeochemistry cycle. Understanding N sources and exports at the watershed scale can provide a knowledge base for remediation of diffuse agricultural pollution, but further studies are needed still on biogeochemical behavior within the watershed, and the retention of N in the soil and groundwater.

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