

## How do nitrogen inputs to the Changjiang basin impact the Changjiang River nitrate: A temporal analysis for 1968–1997

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[1] We present estimates of nitrogen (N) inputs to the Changjiang River basin for the period 1968–1997. The total N input is approximately  $7.8 \times 10^9$  kg in 1997, which is a threefold increase over 1968 levels. N fixation was often a dominant input before 1978, providing about  $2.2 \times 10^9$  kg year<sup>-1</sup>, while N fertilizer dominated N input after 1983, supplying an additional input of some  $4.4 \times 10^9$  kg year<sup>-1</sup>. More than 40% of total N inputs is converted into manure N, and half of total manure N is returned to agricultural soil. We estimate that the river nitrate concentration and flux have increased about tenfold from 1968 to 1997. Our study suggests that the percent of N inputs to the basin that are exported by the river as NO<sub>3</sub>-N has increased steadily over the 30-year period and that about 30% of total N input is transported through the river. The integrated N input, budget, and storage have been linked to the increasing temporal trends of Changjiang River nitrate. N fertilizer application and human population density, as well as manure N production in the basin, are good predictors of the river's nitrate concentration and flux. Therefore, how N balance is kept (especially for effective application of N fertilizer) is a crucial problem to the sustainable development of the basin.

**INDEX TERMS:** 1615 Global Change: Biogeochemical processes (4805); 1803 Hydrology: Anthropogenic effects; 1871 Hydrology: Surface water quality; 9320 Information Related to Geographic Region: Asia; **KEYWORDS:** anthropogenic effects, Changjiang River, nitrate

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### 1. Introduction

[2] Nitrogen is a key, but also limiting, element for terrestrial and marine production. The global rate of terrestrial N fixation has roughly doubled over the past few decades as a result of human activities, such as fertilizer application, population growth, and combustion [Galloway *et al.*, 1995]. These activities have severely impacted the balance of N biogeochemical cycles, resulting in a number of environmental problems both on a regional and global scale: from acidification of freshwater ecosystems [Brimblecombe and Steadman, 1982; Grennfelt and Hultberg, 1986] to coastal eutrophication [Nixon, 1995]; from climate change [Matthews, 1994] to effects on human health and loss of biodiversity [Wedin and Tilman, 1996]. Recently, SCOPE has identified global N overload as a main emerging envi-

ronmental issue in the coming 21st century [Munn *et al.*, 1999]. Therefore the global N overload problem grows critical [Moffat, 1998], and N is and will continue to be a present and a future threat to the environment.

[3] Rivers are an important link between terrestrial and oceanic ecosystems; almost 30% of all new N input is transported to the ocean by the world's rivers [Galloway *et al.*, 1995]. Hence N riverine flux may constitute a sensitive predictor for land and ocean interaction. N is transported by rivers in several forms, and nitrate is not the only human impact on N export, but nitrate is an indicator of these impacts [Caraco and Cole, 1999]. Recent analyses of the global N export by rivers suggest that human activities play a significant role in river nitrate export over spatial scales [Peierls *et al.*, 1991; Turner and Rabalais, 1991; Jordan and Weller, 1996; Howarth *et al.*, 1996; Zhang *et al.*, 1995, 1999]. However, historical data on nitrate in larger rivers relating to N inputs are extremely rare. A critical question, therefore, is how human activities impact river nitrate export

on a temporal scale. It is crucial to understand the mechanisms controlling river N export. For this purpose, we analyzed historical data of N inputs in China's Changjiang River basin for the period 1968–1997. Previous studies reported a shorter term record of nitrate export from the Changjiang River (mainly concentrating on 1980s period) [Duan *et al.*, 2000; Zhang *et al.*, 1995]. However, Zhang's samples were collected at Nantong Station, which is near the river mouth, therefore, the data would be greatly influenced by the tidal effects. Furthermore, both Duan *et al.* [2000] and Zhang *et al.* [1995] did not quantitatively relate the nitrate export to the N inputs within the Changjiang basin. There are several factors controlling N inputs to the basin: N fertilizer application, population growth, N fixation, and atmospheric N deposition. However, estimating the amount of N deposition into the basin is inherently uncertain due to the lack of systematic annual measurements of N content in precipitation through the period of 1968–1997, we will focus on the temporal trends of riverine nitrate discharge in relation to N fertilizer application, N fixation and population growth, in order to explore the human influence on N biogeochemical cycles on a regional scale.

## 2. Methods

### 2.1. Study Area

[4] The Changjiang River is the largest in China, draining almost one fifth of the total area of the nation. In terms of length (6300 km), suspended sediment load ( $500 \times 10^6$  ton  $\text{yr}^{-1}$ ), and discharge ( $900 \text{ km}^3 \text{ yr}^{-1}$ ), the river channel is the third, fourth, and fifth, respectively, largest river in the world [Milliman *et al.*, 1984]. The population density in the Changjiang River basin is higher than many other river basins of the world (Table 1). The period 1968–1997 witnessed rapid development in China. Therefore, during the modernization process, the basin has been and continues to be significantly affected by human activities. As such, the Changjiang River basin might serve as the best region for assessing the impact of those activities on land and ocean interactions and regional biogeochemical cycles in eastern Asia. In order to quantify regional N budgets and riverine N flux, we chose the Datong Hydrological Station (DHS). DHS ( $117^\circ 37' \text{E}$ ,  $30^\circ 46' \text{N}$ ), located on the lower reaches of the river, is free from tidal effects and the industrial pollution of cities [Chen and Shen, 1987]. It drains a wide area of nearly  $1.71 \times 10^6 \text{ km}^2$ , representing more than 95% of the area of the basin. For a more detailed description of the Changjiang basin and DHS, see Yan and Zhang [2003].

### 2.2. Data Calculation

[5] In this study, we consider three factors: N fertilizer application, N fixation, and population growth. All the data were obtained at the provincial level within the basin due to the unavailability of the county-based data, and we converted the provincial data into basin scale data based on the area. The province composition and absolute area and area percentage in the basin are listed in Table 2. The area of the Hubei, Hunan, Jiangxi, and Sichuan provinces, which are totally located within the basin, accounted for about 65% of the total basin area, while the area of the other 13 provinces

**Table 1.** Comparison of Regional Data Between the Changjiang Basin and Other Major River Basins in the World

	Drainage Area, $10^6 \text{ km}^2$	Water Discharge, $10^9 \text{ m}^3 \text{ year}^{-1}$	Population Density, Individual $\text{km}^{-2}$
Changjiang basin	1.81	900	197
Mississippi basin <sup>a</sup>	3.23	546	20
Amazon basin	6.49	7000	2
Baltic Sea	1.50	475	47
North Sea	0.84	380	185

<sup>a</sup>Values for other major river basins except the Changjiang basin are from Howarth *et al.* [1996].

accounted for 35% of the total basin area. In this case, we assume that the input to the province is equally introduced over the province, and the potential error does not significantly impact the results.

[6] Data on N fertilizer application and form in the basin are available in the annual reports of China [China State Statistical Bureau, 1998]. The most commonly used N fertilizers were urea and ammonium bicarbonate. The data of all N fertilizer forms were converted into element N to estimate N fertilizer input.

[7] N fixation refers to the sum of symbiotic N fixation by cultivation of legume crops, and non-symbiotic N fixation by microorganisms in both managed and natural ecosystems. For managed ecosystems, we estimated N fixation inputs by multiplying the rate of fixation for each crop type by the crop area within the basin. Data on cultivated areas of these crops were available for the period 1968–1997 in the annual reports of China [China State Statistical Bureau, 1998]. Legume crops are generally grouped as soybeans, peanuts, and green-manure crops (i.e., red clovers) in the basin. The rate of symbiotic N fixation varies considerably among legume crops. For example, Keyser and Li [1992] reported the range of the median rates as 72 to 201  $\text{kg N ha}^{-1}$ . Galloway *et al.* [1995] estimated a range of legume fixation rate of 70 to 140  $\text{kg N ha}^{-1}$ . Smil [1999] summarized the ranges of published N fixation rates, and used specific ranges for the three different kinds of legumes to estimate N fixation (Table 3). We believe that Smil's data may provide the best possible estimates of N fixation. Therefore we use mean values to estimate N fixation with low and high values in the bracket for reference. These values are 80(15, 450), 80(37, 206), and 150(28, 300)  $\text{kg N ha}^{-1} \text{ year}^{-1}$  fixation rates for soybeans, peanuts, and green-manure, respectively. The cultivation of rice and other non-legume crops supplies an additional source of N fixation by microorganisms [Watanabe, 1986]. We use values of 30(15.8, 53.8) and 15  $\text{kg N ha}^{-1} \text{ year}^{-1}$  for non-symbiotic N fixation rates of rice and other non-legume crops, respectively [Burns and Hardy, 1975; Zhu and Wen, 1990; Zhu *et al.*, 1997; Yan *et al.*, 1999]. Natural ecosystems in the basin mainly include sub-tropical and temperate forest, grassland, and aquatic ecosystems. We estimated N fixation by multiplying the rate of fixation for each natural ecosystem by its area within the basin. Forests accounted for about 20% of total basin area for 1978–1997. Grassland area accounted for 5% of total basin area, and aquatic ecosystem area was 3.7% of total basin area. N fixation rates are scarce in these natural ecosystems, and we use values of 16(1, 160), 2.7(0.1, 10), and 28.5  $\text{kg N ha}^{-1} \text{ year}^{-1}$  of N fixation

**Table 2.** Province Composition and Area Percentage at the DHS in the Changjiang Basin

Province	Provincial Area, 10 <sup>3</sup> km <sup>2</sup>	Area Within the Basin, 10 <sup>3</sup> km <sup>2</sup>	Percentage, <sup>a</sup> %
Hubei	187	187	100
Hunan	210	210	100
Jiangxi	163	163	100
Sichuan <sup>b</sup>	560.5	560.5	100
Guizhou	170	117	69
Shaanxi	190	70.6	37
Yunnan	380	120	31
Qinghai	720	160	22
Anhui	130	23	17
Henan	160	27.3	17
Gansu	530	38	7
Guangxi	230	8.0	4
Tibet	1200	23.5	2

<sup>a</sup>Percent of province in Changjiang basin.<sup>b</sup>Including Chongqing City.

rate for forest, grass and aquatic ecosystems, respectively, reported by *Cleveland et al.* [1999] (Table 3).

[8] Population directly influences N cycles by producing manure. Manure N production by the excretion of humans and livestock in the basin is the product of the number of humans and livestock and the individual waste production rate [*Yan et al.*, 1999]. Data on numbers of humans and livestock in the basin are available in the annual reports of China [*China State Statistical Bureau*, 1998].

[9] We estimated annual riverine N discharge based on the month discharge-weighted data, by multiplying the month concentration of nitrate by the month river discharge rate. N concentration and river discharge from the basin were calculated based on the 30-year unpublished data at DHS from the annual hydrologic reports of China [*Anonymous*, 1998]. Water discharge was recorded at DHS twice each day. Water samples were taken two times per month for nitrate measurement. At each sampling time, a same sampling profile was used with three sampling points at fixed positions from the right bank of the river (260, 1050, and 1590 m, respectively). At each point, water was collected from three different depths (0.2, 0.6, 0.8 times

of the depth). A total of nine samples (3 points × 3 depths) were taken and then mixed thoroughly as one composite sample. Then duplicate water samples were collected from the composite, and filtered through filter papers (before 1987) and 0.45- $\mu$ m filter membranes (after 1987) prior to analysis. The filtrates were measured using a spectrophotometric method with phenol disulfonic acid before 1987, and UV spectrophotometric method after 1987 [*China EPA*, 1989]. The “phenol” method is suitable for the concentration ranges from 0.02–2.0 mg L<sup>-1</sup> with the standard deviation 6.7%, while the “UV” method is suitable for the concentration ranges from 0.08–4.0 mg L<sup>-1</sup> with the standard deviation 1.1%, but both the methods are China standard methods for nitrate. Therefore the change of the method should not impact on the precision of the data.

### 3. Results and Discussion

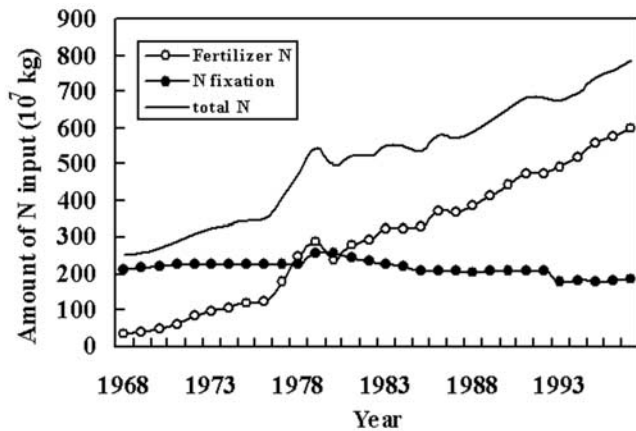
[10] Our analysis shows significant temporal trends in N inputs (Figure 1). N fertilizer input is the major contributor to the basin, and varies greatly. N fertilizer application in the basin began in the early 1950s and climbed dramatically through 1997. The total amount of N fertilizer application was about 600 × 10<sup>7</sup> kg N in 1997, corresponding to approximately 35 kg ha<sup>-1</sup> N fertilizer applied to the basin, while only 2.3 kg ha<sup>-1</sup> was applied in the late 1960s and 15 kg ha<sup>-1</sup> in the late 1970s. N fertilizer application grew by about 11% annually from 1968 to 1997.

[11] N fixation in managed and natural ecosystems within the basin is the second source of N inputs. The amount of N fixation varied only slightly during the period 1968–1997. According to our estimates, the amount of N fixation was about 215 (92, 849) × 10<sup>7</sup> kg N in 1968, corresponding to 12.5 kg N ha<sup>-1</sup> total area, and climbed to a peak of 258 × 10<sup>7</sup> kg N in 1978, and then decreased annually through 1997 to 184 (93, 807) × 10<sup>7</sup> kg N. N fixation decreased about 14% between 1968 and 1997. This decrease should be attributed to the land use changes during the period, particularly, the dramatic decrease in the cultivation area of green manure (Figure 2). This striking changes in the cropland areas resulted from the adjustment of agricultural planting structure, because green manure

**Table 3.** Summary of the Ranges of Published N Fixation Estimates and the Mean Values Used for Calculating Regional N Fixation in the Changjiang Basin

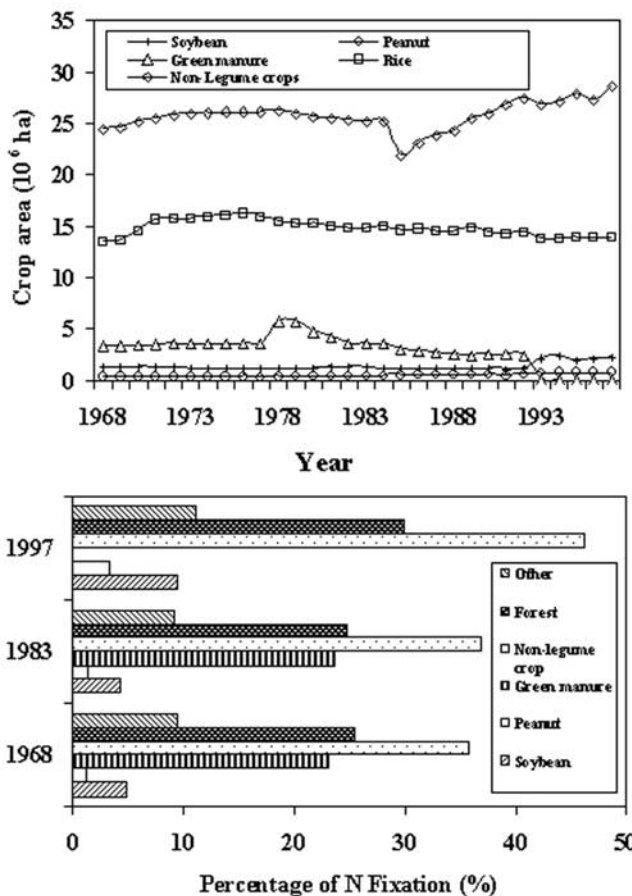
Vegetation Type	Area, × 10 <sup>6</sup> ha			Ranges of Published Estimates, <sup>a</sup> kg N ha <sup>-1</sup>	Values Used in Calculation, <sup>a</sup> kg N ha <sup>-1</sup>	Mean Estimate of N Fixation, <sup>b</sup> 10 <sup>7</sup> kg N		
	1968	1983	1997			1968	1983	1997
1. Leguminous crops	4.93	5.08	2.93			62.6 (12.4, 165)	65.1 (13.0, 166.8)	23.5 (6.1, 113.6)
Soybean	1.32	1.2	2.17	15–450	80	10.6 (2.0, 59.6)	9.6 (1.8, 54)	17.4 (3.3, 97.9)
Peanut	0.31	0.39	0.76	37–206	80	2.5 (1.2, 6.4)	3.1 (1.4, 8.0)	6.1 (2.8, 15.7)
Green manure	3.30	3.49	0.00	28–300	150	49.5 (9.2, 99)	52.4 (9.8, 104.8)	0.00
2. Rice	13.4	14.8	13.9	15.8–53.8	30	40.2 (21.2, 72.1)	44.4 (23.4, 79.6)	41.8 (22.0, 75.0)
3. Nonlegume crops	24.4	25.1	28.6		15	36.6	37.7	43.0
4. Forest	34.2	34.2	34.2	1.0–160	16.0	54.9 (3.4, 549)	54.9 (3.4, 549)	54.9 (3.4, 549)
5. Grass	8.6	8.6	8.6	0.1–10.0	2.7	2.3 (0.09, 8.5)	2.3 (0.09, 8.5)	2.3 (0.09, 8.5)
6. Aquatic vegetation	6.3	6.3	6.3		28.5	18.0	18.0	18.0
7. Total	91.8	94.1	94.5			214.6 (91.7, 849.2)	222.4 (95.6, 859.6)	183.5 (92.6, 807.1)

<sup>a</sup>Data are from *Watanabe* [1986], *Zhu and Wen* [1990], *Zhu et al.* [1997], *Cleveland et al.* [1999], and *Smil* [1999].<sup>b</sup>With low and high values in the bracket.



**Figure 1.** Temporal trends in annual N inputs in Changjiang basin for the period 1968–1997.

became less important due to the rapidly increasing use of chemical fertilizers during the period. The variation in the amount of N fixation has a positive relation with the change in the cultivation area of green manure ( $r^2 = 0.97$ ). Although N fixation by green manure accounted for only about 20%



**Figure 2.** (top) Temporal variation of crop area (land use) within the basin. (bottom) N fixation for different ecosystems as a percentage of total N fixation.

of total N fixation before 1992, the cultivation of green manure is one of the main factors influencing N fixation in the basin.

[12] Atmospheric N deposition is one of the main sources of N inputs. Ideally, N input through atmospheric deposition should be considered in the study, but estimating the amount of N deposition into the basin is inherently uncertain due to the lack of systematic annual measurements of N content in precipitation through the period of 1968–1997. Therefore, the total N input (N fertilizer + N fixation) to the basin is certainly an underestimate as it does not consider the term of atmospheric deposition.

[13] The total N input amount (N fertilizer + N fixation) was about  $784.4 \times 10^7$  kg, corresponding to approximately  $45.8 \text{ kg N ha}^{-1}$  total area in 1997, while only  $14.3 \text{ kg N ha}^{-1}$  was input in 1968, and  $32.0 \text{ kg N ha}^{-1}$  in 1983. Thus the N load increased about threefold in the Changjiang basin as a result of regional human activities during the period 1968–1997. Our estimate of the total N input amount (N fertilizer + N fixation) in 1997 is similar to the value of  $860 \times 10^7$  kg estimated by Xing and Zhu [2002]. If considering the fact that the area we calculated is 95% of the total basin, our estimate is almost the same with the value estimated by Xing and Zhu. We compared N inputs in the Changjiang basin with four other basins in the world (Table 4). Our findings indicate that total N inputs in the Changjiang basin were among the highest in the world basins when inputs are expressed on a per unit basin area except for the North Sea. This reflects the fact that the intensity of human disturbance has become especially strong in the Changjiang basin.

[14] The relative contribution of different N inputs varied substantially for the period 1968–1997. N fertilizer input accounted for more than 50% of total-N inputs as in 1983 and 1997 (Figure 3). This suggests that N fertilizer application has dominated N inputs in the basin since the mid-1980s. The percentage of N fixation against total N inputs varied substantially. N fixation accounted for about 85% of total N inputs in 1968, and decreased to about 20% of total N in 1997. Our results suggest that N fixation was a dominant input to the basin over the first 10-year period (1968–1977), accounting for about 50% of total-N inputs, while the percentage of N fixation against total-N inputs has been decreasing, the estimated rate of N fixation has remained relatively constant over the 30-year period indicating that N fixation would be a stable source of N inputs to the basin.

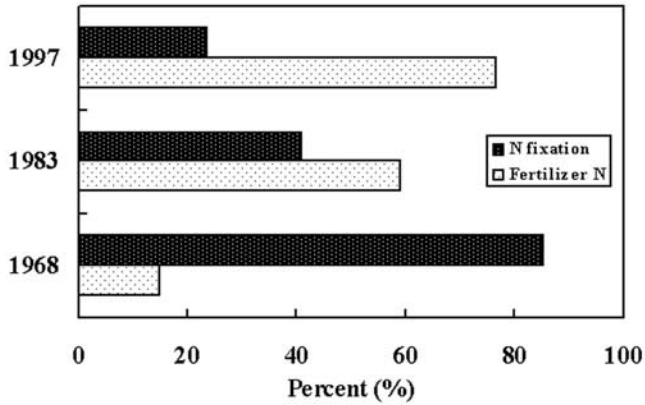
**Table 4.** Comparison of N Inputs Between the Changjiang Basin and Other Major River Basins in the World<sup>a</sup>

	N Fertilizer	N Fixation	Total Inputs
Changjiang basin <sup>b</sup>	35.1	10.7	45.8
Mississippi basin <sup>c</sup>	18.4	10.6	29.0
Amazon basin	0.63		0.63
Baltic Sea	17.3	0.27	17.6
North Sea	59.6	0.05	59.7

<sup>a</sup>Values are in  $\text{kg N ha}^{-1}$  total area year<sup>-1</sup>.

<sup>b</sup>Values for the Changjiang basin are for the year 1997.

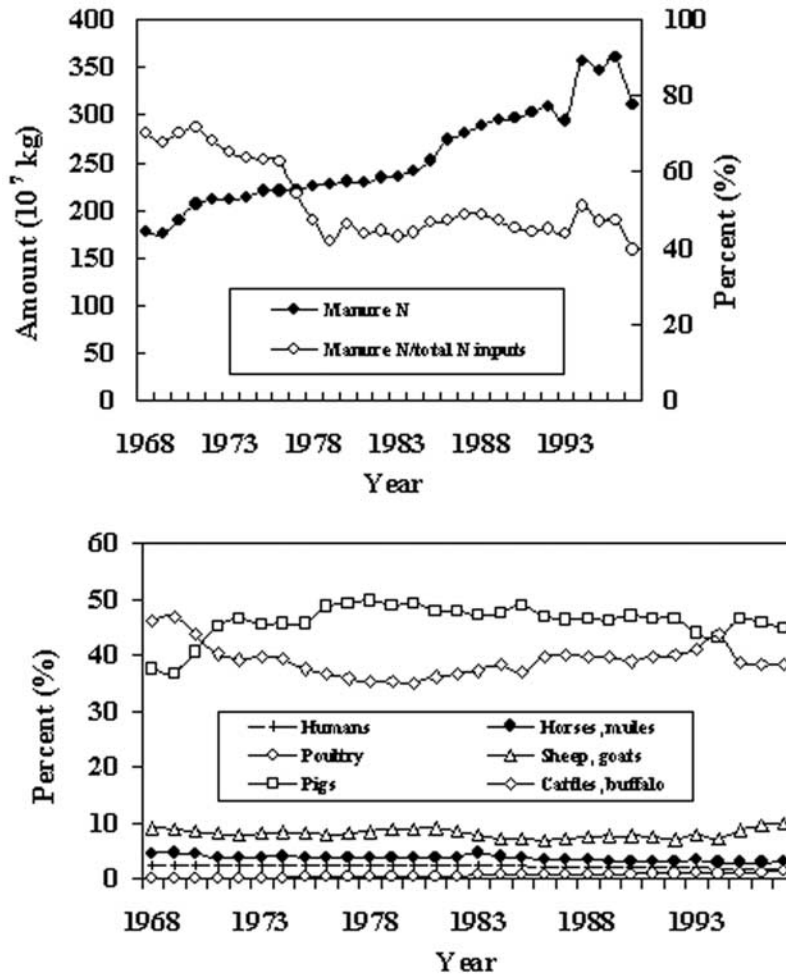
<sup>c</sup>Values for other major river basins except the Changjiang basin are from Howarth et al. [1996].



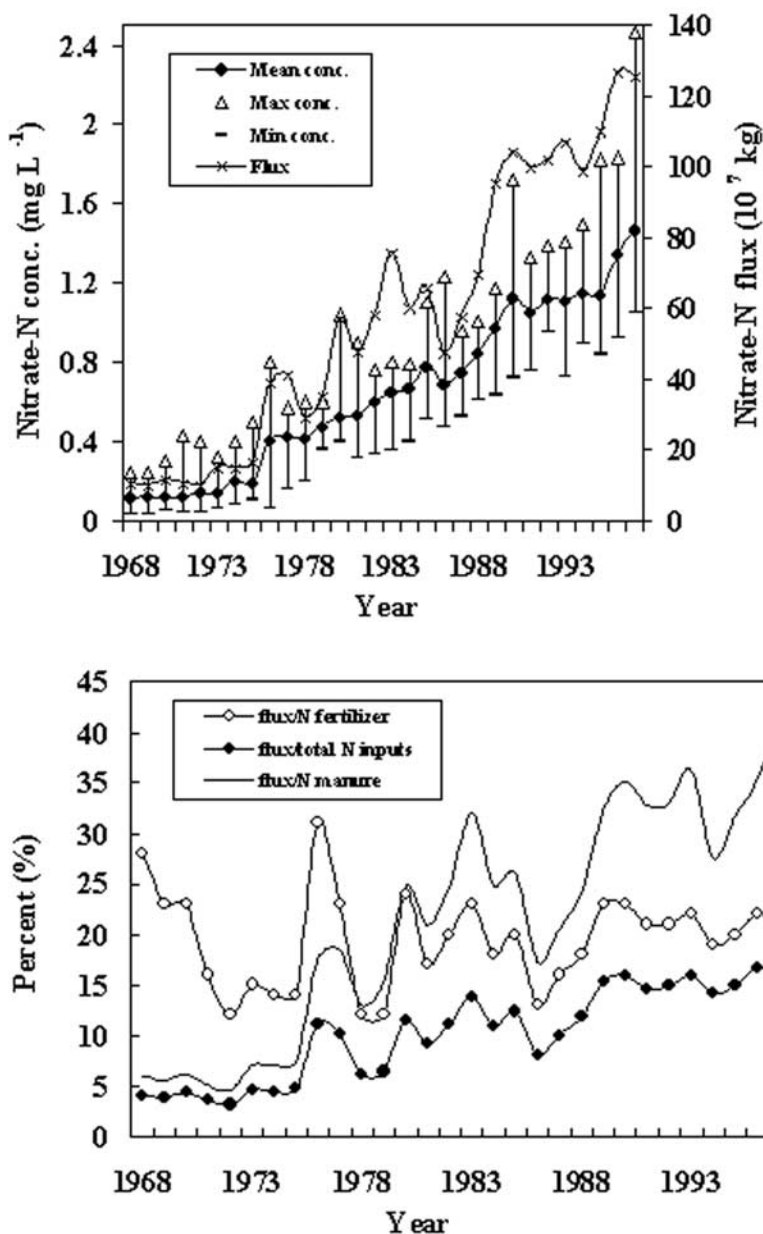
**Figure 3.** Different N inputs as a percentage of total N inputs, illustrating that N fertilizer input dominated total N inputs after 1983, while N fixation was a dominant input before 1978.

[15] Manure N is not a new input to the basin, but rather it is stored and joins the internal N cycling in the basin. Therefore manure N has a great influence on river nitrate. The amount of manure N was  $177 \times 10^7$  kg in 1968, equivalent to 70% of the total N inputs to the basin, and increased to  $311 \times 10^7$  kg, or 40% of the N inputs in 1997 (Figure 4). Over the 30-year period, our estimate shows that an average of more than 50% of total-N inputs to the basin was converted into manure N (Figure 4). Humans produced only 2% of total manure N in the basin during the period; almost 98% of total manure N was produced by livestock, of which an average of 46% of total manure N came from pig manure, while 39%, 8%, 4% came from cattle (including buffalo), sheep (including goats), and horse (including mules, etc.) manure, respectively. Poultry produced the least manure N, accounting for about 0.6% of total manure N in the basin (Figure 4). Quantifying the amount of manure N that is internal cycled within the basin is more difficult.

[16] There was a temporal increase in Changjiang River nitrate-N concentration over time between 1968 and 1997



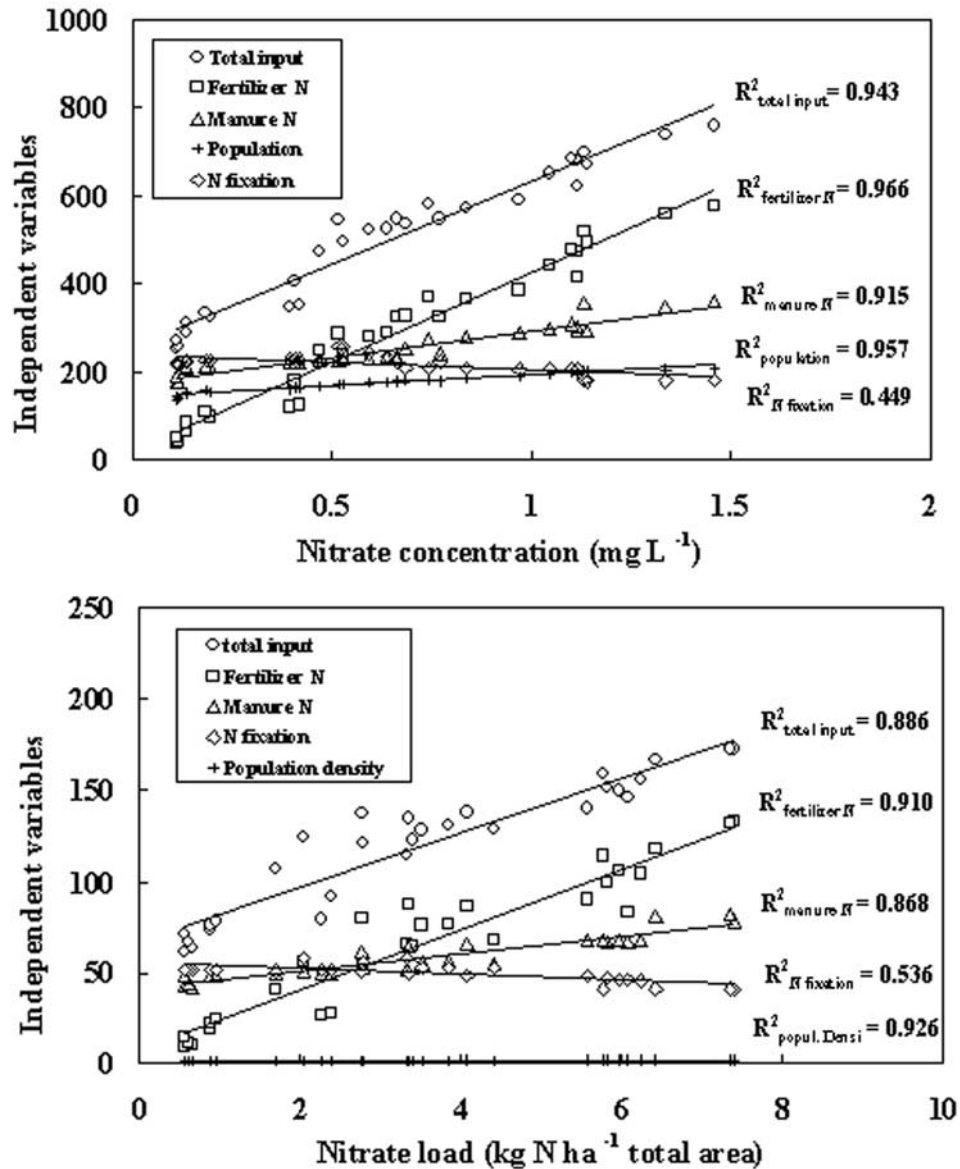
**Figure 4.** (top) Temporal trends of manure N production, and manure N production as a percentage of total N inputs in the basin. (bottom) Percentage of different kind of manure N against total N input, showing 85% of manure N came from pig and cattle (including buffalo) manure during the period of 1968–1997.



**Figure 5.** (top) Temporal trends of nitrate-N concentration and flux through the Changjiang River at DHS from 1968–1997. (bottom) Temporal variation of the ratio of the river nitrate-N flux to different variables of N inputs.

(Figure 5). The annual nitrate-N concentration was more than  $1.4 \text{ mg L}^{-1}$  in 1997, compared to only about  $0.1 \text{ mg L}^{-1}$  in 1968, increasing almost 14-fold for the period 1968–1997. By comparing three different 10-year periods (1968–1977, 1978–1987, and 1988–1997), we found that the river NO<sub>3</sub>-N concentration increased slightly in 1968–1977, with annual average concentration being approximately  $0.2 \text{ mg L}^{-1}$ . However, it has doubled from 1968–1977 to 1978–1987, and again from 1978–1987 to 1988–1997, respectively. Several investigations on nitrate concentrations of the Changjiang River have been reported during the 1980s [Duan *et al.*, 2000; Meybeck, 1982; Shen, 1997; Zhang *et al.*, 1995]. Comparing our results of nitrate concentrations during 1980s with these studies, our results (mean value of

$0.69 \text{ mg L}^{-1}$ ) are similar to  $0.721 \text{ mg L}^{-1}$  [Duan *et al.*, 2000], and  $0.731 \text{ mg L}^{-1}$  [Shen, 1997], but higher than  $0.24 \text{ mg L}^{-1}$  [Meybeck, 1982], and  $0.46 \text{ mg L}^{-1}$  [Zhang *et al.*, 1995]. Our study showed that nitrate concentrations have a similar increasing trend to Mississippi River study [Turner and Rabalais, 1991], but mean annual nitrate concentrations at DHS are  $0.399 \text{ mg L}^{-1}$  from 1975–1980 and  $0.69 \text{ mg L}^{-1}$  from 1980–1989, which are much lower than these (about  $0.842$  and  $1.19 \text{ mg L}^{-1}$ ) at St. Francisville for the corresponding periods. Nitrate-N flux through the river discharge reflected the increasing trend of the concentration. The annual river nitrate-N flux was only  $10 \times 10^7 \text{ kg}$  in 1968, and  $120 \times 10^7 \text{ kg}$  in 1997. The percentages of riverine nitrate-N flux against total N inputs (N fertilizer + N



**Figure 6.** Correlations of the Changjiang River nitrate with different variables for the period 1968–1997. (top) Relationship between nitrate-N concentration ( $\text{mg L}^{-1}$ ) and variables (expressed as  $10^7$  kg N or total individual). (bottom) Relationship between Nitrate-N load (expressed as  $\text{kg N year}^{-1} \text{ha}^{-1}$  total area) and variables (expressed as  $\text{kg N year}^{-1} \text{ha}^{-1}$  agricultural land or individual  $\text{year}^{-1} \text{ha}^{-1}$  total area).

fixation) and manure N were 4 and 6%, respectively, in 1968, and increased to 16 and 40%, respectively, in 1997 (Figure 5), suggesting that the increase of the river nitrate-N flux coincides with these of total N inputs and manure N. The fertilizer N was a fairly constant percentage of riverine nitrate-N flux, with a mean value of 20%. The mean percentage of riverine nitrate-N flux against total N inputs was about 14% after 1983. Estimating the amount of total-N (TN) export trend through the river is difficult due to the lack of systematic annual measurements of TN through the period of 1968–1997. However, in order to give a rough estimate of TN flux, we use data measured in 1998–1999 and 2001, respectively, as a reference. Accord-

ing to Duan [2000], the amounts of nitrate-N and TN fluxes were 113.7 and  $198.6 \times 10^7$  kg, respectively, in 1999. The ratio of nitrate-N against TN was 57%. Our measurements of nitrate-N and TN fluxes in 2001 are 94.6 and  $149.6 \times 10^7$  kg, respectively, in 2001. The ratio of nitrate-N against total-N is 63.2%. Both values of the ratio are similar to that (60%) in Dunn's [1996] study. If assuming that the ratio of the river nitrate-N against TN remains constant (60%), we estimate that the average river TN flux is approximately  $200 \times 10^7$  kg, 30% of total N inputs for the period 1983–1997.

[17] To analyze which factors are most closely related to the Changjiang River N, we compared our estimates of different variables of N factors to the annual average river

nitrate-N concentration (Figure 6). Our analysis shows a highly significant linear relationship between the river nitrate-N concentration and several variables. We repeated the analysis for the calculated nitrate-N flux, and found similar significant linear relationships. Fertilizer N input ( $r^2 = 0.966$ ), population ( $r^2 = 0.957$ ), total N inputs ( $r^2 = 0.943$ ) and manure N ( $r^2 = 0.915$ ) are closely related to the Changjiang River nitrate-N concentration, while population density (expressed as individual year<sup>-1</sup> ha<sup>-1</sup> total area) ( $r^2 = 0.926$ ), fertilizer N input (expressed as kg N year<sup>-1</sup> ha<sup>-1</sup> agricultural land) ( $r^2 = 0.910$ ), total N inputs (expressed as kg N year<sup>-1</sup> ha<sup>-1</sup> agricultural land) ( $r^2 = 0.886$ ), and manure N load (expressed as kg N year<sup>-1</sup> ha<sup>-1</sup> agricultural land) ( $r^2 = 0.868$ ) are closely related to the Changjiang River nitrate-N load (expressed as kg N year<sup>-1</sup> ha<sup>-1</sup> total area). This suggests that human activities, especially fertilizer N input and population, are good predictors of the river's nitrate-N concentration and flux. In contrast, N fixation is least related to the river's nitrate-N concentration ( $r^2 = 0.449$ ) and load ( $r^2 = 0.536$ ), reflecting that this anthropogenic N input has become less important in contributing to the river N during the period 1968–1997.

[18] Careful study of N budgets is critical to predict river N flux, and studies in small watersheds suggest that the capacity to store N can ultimately saturate, as N inputs to the watersheds increase [Aber, 1992; Kahl et al., 1993]. Therefore it can be inferred that N storage within the basin can be expected to saturate rapidly as the region develops due to the replacement of natural vegetation with more impervious surface, for percent of N inputs to Changjiang basin that are exported as NO<sub>3</sub>-N has increased steadily from about 4% in 1968 to 16% in 1997. However, in order to improve our understanding of what should eventually be responsible for higher rates of the river N flux, research needs to focus on integrated N biogeochemical budgets at regional scales. This includes: (1) N input to the basin by deposition; (2) N outputs from the basin, such as N emissions both through denitrification and volatilization, exports of N-containing products; and (3) N storage within the basin, such as N storage in manure, agricultural soil, and surface water.

#### 4. Conclusions

[19] The total N input is approximately  $7.8 \times 10^9$  kg in 1997, which is a threefold increase over 1968 levels. N fixation was often a dominant input before 1978, providing about  $2.2 \times 10^9$  kg year<sup>-1</sup>, while N fertilizer dominated N input after 1983, supplying an additional input of some  $4.4 \times 10^9$  kg year<sup>-1</sup>. This demonstrates that agriculture became extremely intensive by using more and more fertilizer to achieve sufficient yields after 1983. More than 40% of total N inputs is converted into manure N, and half of total manure N is returned to agricultural soil. Normally, all fertilizers and manure are top-dressed and liable to be lost from agricultural soils by runoff in the basin. As a result, most lakes and other surface waters in the basin are eutrophic [Jin, 1995], and agricultural nonpoint source pollution of nutrients accounts for more than half of the total load [Tu et al., 1990]. Therefore we arrive at a critical

question: How is N fertilizer (both synthetic fertilizer and manure) effectively applied on a regional scale? We estimate that the river nitrate concentration and flux have increased by about tenfold from 1968 to 1997. Our study suggests that about 30% of total N input is transported through the Changjiang River. The integrated N inputs have been linked to the increasing temporal trends in Changjiang River nitrate. Our study demonstrates that integrated N input, especially N fertilizer application (synthetic plus manure), as well as human population density in the basin, are good predictors of the river's nitrate concentration and flux. Further studies are required to focus on the dynamic linkages and spatial patterns of human activities on N integrated biogeochemical budgets at many scales (subwatershed, watershed, regional).

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