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Anthropogenic nitrogen sources and exports in a village-scale catchment in Southeast China

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Abstract

An experimental village-scale catchment was selected for investigation of nitrogen (N) sources and exports. The mean N application rate over the catchment was 350.2 kg N ha^{-1}, but this rate varied spatially and temporally. The N leaching loss rate varied from 8.1 to 52.7 kg N ha^{-1} under different land use regimes. The average N leaching loss rate was 13.4 kg N ha^{-1} over the whole catchment, representing about 3.8% of the total N inputs. The N export rate through stormflows was 28.8 kg N ha^{-1}, about 8.2% of the total N inputs. Seasonal patterns showed that 95% of N exports through stormflows occurred during July to September in 2002. Overall, the maximum riverine N exports were 12.1% of total N inputs and 15.5% of the inorganic fertilizer N applied. Understanding N sources and exports in a village-scale catchment can provide a knowledge base for amelioration of diffuse agricultural pollution.

Introduction

The excessive use of commercial inorganic fertilizers and animal manures for raising crop yield and meeting the demand of population growth in China has resulted in nutrient losses from agricultural catchments (Li & Zhang 1999; Yan *et al.* 1999; Cao *et al.* 2003). Agricultural nutrient losses can accelerate freshwater and coastal water eutrophication and lead to chronic hypoxia, reductions in species diversity, and stressed fisheries resources (Nixon *et al.* 1995; Vitousek *et al.* 1997; Carpenter *et al.* 1998). Nitrogen can also be of concern in drinking water supplies due to methemoglobinemia in infants and concern about possible links to non-Hodgkin's lymphoma (Ward *et al.* 1996). In Southeast China, anthropogenic N inputs far exceed N outputs in regional agricultural ecosystems, and N imbalance has resulted in N losses (Cao & Zhu 2000). Research has shown that water quality in the Jiulong River estuary and coastline has deteriorated with eutrophication and excessive growth of benthic algae occurring since about 1985 (Chen *et al.* 1993). Reversal of eutrophication and contamination caused by agricultural nutrient losses and development of remedial strategies require the identification of nutrient sources and assessing exports from sources to waterbodies (e.g. Cirmo & McDonnell 1997; Mitchell 2001; Worrall & Burt 2001). The objectives of the present study were to investigate anthropogenic N sources in an experimental village-scale catchment and to characterize N exports in leaching and stormflows.

Materials and methods

Study area

An experimental village-scale catchment called Wuchuan, with an area of 9.4 km^2 and a population of 4509, was selected for investigating N sources and exports. The stream is a tributary of the upper Jiulong River in Fujian province, Southeast China. Catchment elevation varies between 7.5 and 130 m. Land uses are very complex (Figure 1a) and can be combined into forestry, including planted forest (31%) and bamboo (7%); horticulture, including bananas (15%), longan (10%), sugarcane (8%), lychee (5%) and mango (3%); and rice paddies (3%); vegetables (3%); residential areas (5%); fishponds (3%) and the remaining 7% (other uses).

Rainfall is strongly influenced by the monsoon system. Total recorded rainfall in 2002 was 1624 mm, less than the annual average of 1720 mm. The rainfall occurring between July and September (the wet season) was 1078 mm, representing about 66% of the 2002 total.

The topography is rolling, undulating hills with their heavily weathered granite bases dissected by small streams. The incised narrow valleys on the hillslopes often open out, join together at the hill foot, and form a wide flat alluvial valley bottom where intensive agriculture and horticulture have developed. Red earth and lateritic red earth are the main soil types in the catchment, with pH values ranging from 4.0 to 4.8 and a mean value of 4.5.

N source estimates

The anthropogenic N sources in the catchment include inorganic fertilizers, animal excreta, domestic wastes, atmospheric deposition and fishpond water discharge. Although the N in animal excreta, domestic wastes and fishpond water was internally recycled and therefore not a new N input to the area, a significant part of N from the above sources has been applied traditionally to agricultural land. This relocated N, together with inorganic fertilizer N, may have been washed out by rainfall-runoff and thus contributed to N exports in rivers.

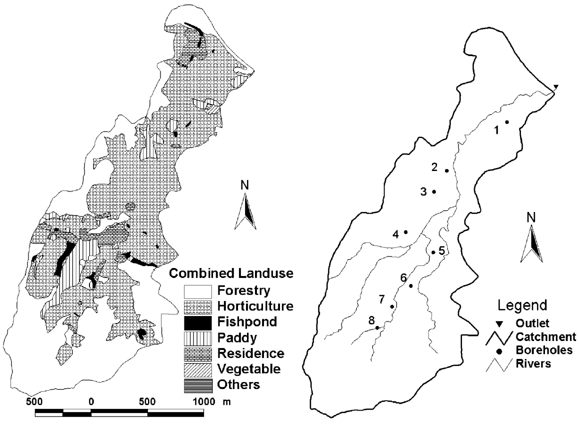

Fig. 1. Combined land uses (a) and borehole location (b) in the Wuchuan catchment.

Data on inorganic N fertilizers were obtained from regular visits and questionnaires. Ammonium bicarbonate and urea were the main N fertilizers applied in this area (Cao *et al.* 2003). Estimates of the amount of elemental N produced in manures and wastes were made using human and animal population censuses and typical elemental N concentrations in manures and wastes (Lu *et al.* 1996). It was assumed that 25% of total human and animal excreta were applied as fertilizers in the catchment (Yan *et al.* 1999). Atmospheric N deposition in the river catchment was estimated based on recorded rainfall from a meteorological station and N concentrations in rainfall from a comprehensive survey on acid rain conducted for more than 10 years (Tang *et al.* 1996; Yu *et al.* 1998).

Fishponds were cleared once a year. Water was pumped out for irrigation and the sediment was removed and applied to crops. Nitrogen from fishpond removal was estimated using the product of the water volume, the amount of sediment and the mean concentrations of N in 21 samples of discharge water ($9.02\ \mathrm{mg\ l^{-1}}$) and sediment ($2.31\ \mathrm{g\ kg^{-1}}$). The water volume and amount of sediment were estimated using the product of the surface areas of the fishponds and their water and sediment depths.

N export measurements

Pathways of N losses include gaseous emission, leaching losses and losses in streamflow. However, the N losses in leaching and streamflow were of more environmental concern at a local scale. Stormflow was measured every 0.5 h using a flowmeter installed at the outlet. Water quality was monitored for each storm at the outlet. Typically 8–10 water quality samples were taken manually for each storm and each water sample was analyzed for total N. The sampling interval varied from 0.5 to 4 h depending on rainfall duration.

Eight boreholes were drilled within the study area and monitoring pipes with a diameter of 110 mm and a slot at the bottom were installed in 2002 for water quality analysis in the shallow aquifer (Figure 1b). The boreholes were established under areas of different land use with a depth varying from 1.5 to 2.2 m (Table 1). When sampling, exposed water (old water) in the monitoring pipes was first pumped out, and then fresh water was used for analysis. Nitrate nitrogen (NO_3-N) was analyzed in samples from shallow aquifers below the crop-rooting zone.

The amount of NO_3-N exported in leaching was determined as the product of the monthly monitored concentration of NO_3-N and the estimate of monthly water flux reaching the shallow groundwater table under different land uses. A simple water balance model was used to estimate the monthly water flux to shallow aquifers under each land use. The model calculated monthly water balance using rainfall minus streamflow at the outlet and estimated evaporation for different land uses (Zhang 2003). The evaporation estimate used the potential evapotranspiration method recommended by the UN Food and Agricultural Organization (Allen *et al.* 1998). The mean NO_3-N concentrations of April and December were used to overcome missing measurements during January to March. When occasional monthly NO_3-N measurements under any particular land use were missing the monitored concentration from the preceding month from the same land use was used for calculation of the monthly NO_3-N export in leaching. Standard colorimetric techniques were used to analyze total N and NO_3-N. All samples were taken to the laboratory and analyzed within 24 h of collection.

Results

Anthropogenic N sources

Over the catchment, inorganic fertilizers were the largest single source N input (77.9%, Table 2), followed by manure application (11.1%), domestic wastes (4.6%), atmospheric wet deposition (4.1%), and fishpond removal (2.3%). Overall, the area-weighted mean application rate of N over the catchment was 350 kg N ha^{-1}, but this rate varied spatially and temporally among the different land uses (Figure 2). Inorganic N fertilizers were applied to crops at rates ranging from 100 to 810 kg N ha^{-1}, with maximum rates on vegetables and minimum on bamboo. However, most of the inorganic fertilizers were applied to cash crops such as bananas and longan, with roughly 43% applied to bananas and about 13% each to longan and sugarcane. Vegetable growing areas were small but received the highest application rates of inorganic fertilizers.

Historically, the mean N application rate for the whole of China was very low (Zhu 1997). In Lake Taihu region, east China, the N application rate varied from 70 to 100 kg N ha^{-1} and N was applied only in manures in the 1950s. However, since the 1980's N application rates have increased greatly and recently varied from 500 to 800 kg N ha^{-1} on different crops (Ellis & Wang 1997). In the Wuchuan catchment, N fertilizers were not applied to forests that cover 33% of the total area. Thus, the actual N application rate in arable areas and

Table 1. Land use, management and depth of boreholes across the catchment.

No.	Land use	Fertilizers applied	Depth (m)
1*	Fallow	None	1.5
2	Double rice cropping	Ammonium hydrogen carbonate	1.7
3	Vegetables and rice rotation	Manure compost, Ca–Mg phosphate	1.7
4	Sugarcane	Ammonium hydrogen carbonate, Ca–Mg phosphate	2.5
5	Bananas	Urea and ammonium hydrogen carbonate	2.5
6	Vegetables	Manure compost, urea and ammonium hydrogen carbonate	2.0
7	Bananas	Manure compost, urea and ammonium hydrogen carbonate, potassium phosphate	2.0
8	Vegetables and fallow	Manure compost	1.5

* Number of boreholes in first column shown in Figure 1b.

Table 2. Anthropogenic N sources for three villages: Fengtian (A), Tuwei (B), and Shanbian (C).

Anthropogenic N sources			Amount (t $year^{-1}$)	Rate (kg N ha^{-1})	Contribution (%)
Animal manures	Cattle	A	7.4	8.0	2.3
		B	4.9	5.3	1.5
		C	4.9	5.3	1.5
	Pigs	A	1.8	2.0	0.6
		B	2.2	2.4	0.7
		C	7.6	8.3	2.4
	Poultry	A	3.9	4.3	1.2
		B	1.8	2.0	0.6
		C	1.2	1.3	0.4
	Subtotal		35.6	38.9	11.1
Domestic wastes	Excreta	A	2.2	2.4	0.7
		B	2.8	3.1	0.9
		C	7.1	7.7	2.2
	Sewage	A	0.5	0.5	0.2
		B	0.6	0.7	0.2
		C	1.5	1.6	0.5
	Subtotal		14.7	16.1	4.6
Fishpond sources	Discharges		4.6	5.0	1.4
	Sediments		2.8	3.1	0.9
	Subtotal		7.4	8.1	2.3
Atmospheric deposition			13.2	14.4	4.1
Inorganic N fertilizers			250.1	272.7	77.9
Total			321.1	350.2	100.0

orchards was 522 kg N ha^{-1} when forests were excluded from the calculations, about 172 kg N ha^{-1} higher than the previous estimate in which forests were included.

Nitrate leaching

Nitrate-N is highly mobile and retention of NO_3-N by soil is minimal so that NO_3-N is very susceptible to leaching. Water quality measurements showed that NO_3-N concentrations in shallow aquifers were highly variable under different land uses, and had a great temporal variability. The NO_3-N concentrations were highest under banana and vegetables and the peak NO_3-N concentrations occurred during July to August (Figure 3). Rainfall and fertilizer application are the major factors influencing NO_3-N leaching. Rainfall intensity during July to August was very high and comprised 50% of the total rainfall in 2002. Intensive rainfall accelerated the soil infiltration and the processes that lead to water flow that bypasses the soil matrix, leading to a buildup of NO_3-N in the shallow aquifer during this period. Thus, the timing and application rate of fertilizer also

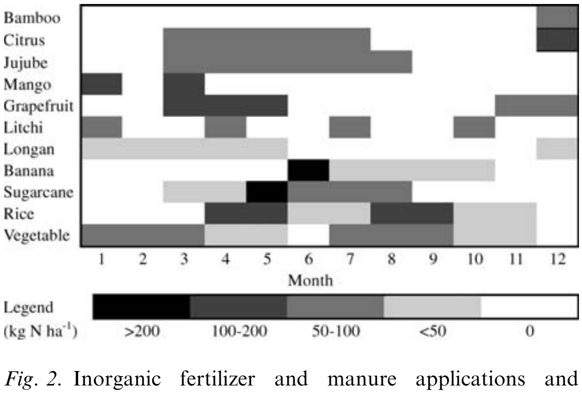

Fig. 2. Inorganic fertilizer and manure applications and growing seasons of different crops.

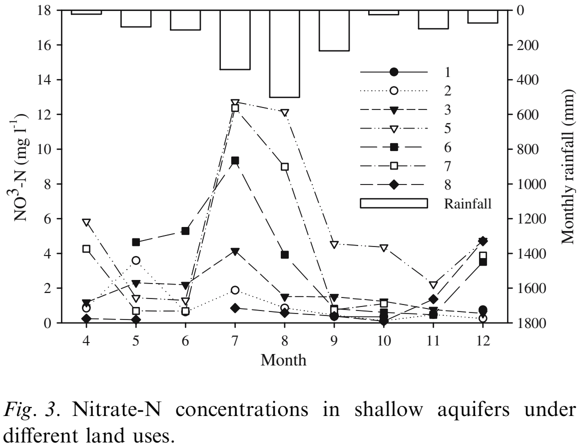

Fig. 3. Nitrate-N concentrations in shallow aquifers under different land uses.

exerted an influence on NO_3-N concentrations in the shallow aquifers. Fertilizers were applied to banana fields from June to October and NO_3-N concentrations increased over the same period (Figure 2). These factors clearly determined the seasonal patterns of NO_3-N concentrations in the local shallow aquifers.

The NO_3-N export rate in leachate differed substantially with land use and varied from 8.1 to 52.7 kg N ha^{-1}, with a mean value of 13.4 kg N ha^{-1}. The maximum leaching loss occurred under bananas and the minimum under fallow. There was a strong correlation between leaching N loss and the amount of fertilizer N applied to each land use. The mean NO_3-N export rate in leaching was equivalent to approximately 3.8% of the total N inputs and about 4.9% of the inorganic fertilizer N inputs.

N export in stormflows

The N export in each stormflow was quantified by multiplying measured N concentrations by the flow rates during storm events. The N export rate in stormflows was aggregated to a monthly scale (Figure 4), owing to difficulties in distinguishing flows between intermittent rainfall events. The monthly N export rate in stormflows varied from almost zero to 17.6 kg N ha^{-1}, with an aggregate of 28.8 kg N ha^{-1} in 2002. Seasonal N export patterns showed that approximately 95% of N exports in stormflows occurred during July to September when rainfall intensity was very large and the rainfall was about 66% of the total rainfall in 2002. These results are in agreement with the findings of Novotny and Chesters (1989) that overall nutrient yield is generally accounted for by the few extreme flood events in a given year. The mean N exports in stormflows accounted for approximately 8.2% of the total N inputs to the catchment and about 10.6% of the inorganic fertilizer N inputs in 2002.

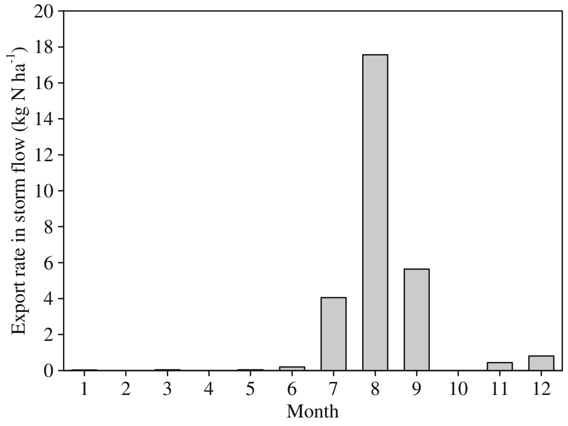

Fig. 4. Monthly N export rate in stormflows in the Wuchuan catchment.

Discussion

The overall anthropogenic N inputs over the Wuchuan catchment were 350 kg N ha^{-1}. Nitrate-N was the predominant N form in groundwater owing to the strong retention of ammonium-N by soil. The N exports in stormflow and leaching therefore amounted to 42.2 kg N ha^{-1}, which accounted for only 12.1% of the total anthropogenic N inputs to the catchment and 15.5% of the inorganic fertilizer N applied. This is consistent with the general conclusion that N inputs to terrestrial systems exceed riverine exports (Howarth *et al.* 1996; David & Gentry 2000). Of the N exported from the Wuchuan catchment, 32% was found in leachate and 68% was lost through stormflows. However, percolation can partially return as stream baseflow or move vertically to deep groundwater. Therefore, the maximum riverine N exports were 12.1% of the total anthropogenic N inputs to the catchment and 15.5% of the inorganic fertilizer N applied only if no further recharges to deep groundwater occurred and the N in shallow aquifers was not re-used by deep-rooted crops during the dry seasons. Consequently, approximately 88% of the anthropogenic N inputs were taken up by crops or stored, denitrified, or volatilized in the catchment. These results are generally consistent with the finding that less than 30% of the anthropogenic N inputs to catchments are exported to the oceans with surface runoff in rivers and streams (Howarth *et al.*, 1996; Boyer *et al.* 2002; Breemen *et al.* 2002) and the remainder (> 70%) is stored, denitrified or volatilized. However, the export rate of streamflow (maximum 12.1%) in the Wuchuan catchment is even less than the value of 25% found by Boyer *et al.* (2002) in the northeastern USA.

Surveys have shown that the predominant inorganic fertilizers applied in this catchment were ammonium hydrocarbonate and urea (Cao *et al.* 2003), compounds that are very susceptible to loss to the atmosphere by ammonia volatilization (Chen & Zhu 1981). Strong volatilization and denitrification processes in the catchment were likely responsible for the higher N losses to the atmosphere, and there is also evidence that gaseous N losses through volatilization and denitrification over the whole of China accounted for as much as 22% to 32% of the total N output (Xing & Zhu 2002). However, sediment N deposition in rivers, ponds, and wetlands might account for another significant portion of the N inputs.

Conclusions

The environmental implications of this study are that the total N inputs to the catchment were much larger than the exports through streamflow and were dominated by inorganic fertilizer N inputs. The mean N export rates in leaching and stormflows were about 13.4 and 28.8 kg N ha^{-1}, respectively. The maximum riverine N export rate (42.2 kg N ha^{-1}) accounted for about 12.1% of the total N inputs and 15.5% of the inorganic fertilizer N applied in 2002. Consequently, approximately 88% of the N inputs were taken up by crops or stored, denitrified or volatilized in the catchment. Nitrogen emission rate to the atmosphere through volatilization and denitrification was likely responsible for the higher N loss rate in the catchment, and sediment N deposition in rivers, ponds, and wetland might account for another significant portion of the N inputs. Seasonal patterns showed that about 95% of the total N exports through stormflows occurred during July to September. Of the riverine N export in 2002, 68% was through stormflow but accounted for only 12% of the flow time (stormflow days over the year), and baseflow was assumed to account for the remaining 32% of riverine N export and 88% of the flow time. Understanding N sources and exports in a village-scale catchment can provide a knowledge base for remediation of diffuse agricultural pollution.

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References cited

Allen RG, Pereira LS, Raes D, Smith M. 1998 Crop evapotranspiration – Guidelines for computing crop water requirements – FAO Irrigation and drainage paper 56. FAO – Food and Agriculture Organization of the United Nations, Rome.

Boyer EW, Goodale CL, Jaworski NA, Howarth RW. 2002 Anthropogenic nitrogen sources and relationships to riverine nitrogen export in the northeastern USA. *Biogeochemistry* **57/58**, 137–169.

Breemen NV, Boyer EW, Goodale CL, Jaworski NA, Paustian K, Seitzinger SP, Lajtha K, Mayer B, Vandam D, Howarth RW, Nadelhoffer KJ, Eve M, Billen G. 2002 Where did all the nitrogen go? Fate of nitrogen inputs to large watersheds in the northeastern USA. *Biogeochemistry* **57/58**, 267–293.

Cao W, Hong H, Yue S, Ding Y, Zhang Y. 2003 Nutrient loss from an agricultural catchment and soil landscape modelling in Southeast China. *Bull Environ Contam Toxicol* **71**, 761–767.

Cao W, Zhu H. 2000 *Characteristics and Control of Regional Agricultural Ecosystems in Fujian Province*, Beijing: China Agriculture Press.

Carpenter SR, Caraco NF, Correl DL, Howarth RW, Sharpley AN, Smith VH. 1998 Nonpoint pollution of surface waters with phosphorus and nitrogen. *Ecol Appl* **8**, 559–568.

Chen RY, Zhu ZL. 1981 The fate of nitrogen fertilizers in paddy soils. In: Institute of Soil Science, eds. Proceedings of Symposium of Paddy Soil, Springer-Verlag, Berlin, pp. 597–602.

Chen ST, Ruan WQ, Zhen RZ. 1993 The biogeochemical study of phosphorus in the Jiulong River estuary and Western Sea (I). *Acta Oceanologica Sinica* **15**(1), 62–70.

Cirmo CP, McDonnell JJ. 1997 The hydrological and biogeochemical controls of nitrogen transport in near-stream riparian zones of temperate forested watersheds. *J Hydrol* **199**, 88–120.

David MB, Gentry LE. 2000 Anthropogenic inputs of nitrogen and phosphorus and riverine export for Illinois, USA. *J Environ Qual* **29**, 494–508.

Ellis EC, Wang SM. 1997 Sustainable traditional agriculture in the Tai lake region of China. *Agric Ecosyst Environ* **61**, 1777–1193.

Howarth RW, Billen G, Swaney D, Townsend. A, Jaworski N, Lajtha K, Downing JA, Elmgren R, Caraco N, Jordan T, Berendse F, Freney J, Kudeyarov V, Murdoch P, Zhao-Liang Z. 1996 Regional nitrogen budgets and riverine N and P fluxes for the drainages to the North Atlantic Ocean: natural and human influences. *Biogeochemistry* **35**, 75–139.

Li Y, Zhang JB. 1999 Agricultural diffuse pollution from fertilizers and pesticides in China. *Water Sci Technol* **39**, 25–32.

Lu RK, Liu HX, Weng DZ, Qin SW, Zhang JY, Wang ZQ. 1996 Nutrient cycle and balance in some agricultural ecosystems in China I: output parameters. *Chin J Soil Sci* **27**(4), 145–151.

Mitchell MJ. 2001 Linkages of nitrate losses in watersheds to hydrological processes. *Hydrol Process* **15**, 3305–3307.

Nixon SW, Granger SL, Nowicki BL. 1995 An assessment of the annual mass balance of carbon, nitrogen and phosphorus in Narragansett Bay. *Biogeochemistry* **35**, 15–61.

Novotny V, Chesters G. 1989 Delivery sediments and pollutants from nonpoint sources: a water quality perspective. *J Water Soil Conserv* **44**, 568–576.

Tang D, Wang W, Pang Y, Liu H, Wang S, Wang R. 1996 Contribution of NO_x to acid rain in Minnan Area. *Res Environ Sci* **9**, 38–40.

Vitousek PM, Aber JD, Howarth RW, Likens GE, Matson PA, Schindler DW, Schlesinger WH, Tilman DG. 1997 Human alteration of the global nitrogen cycle: sources and consequences. *Ecol Appl* **7**, 737–750.

Ward MH, Mark SD, Cantor KP, Weisenbuger DD, Correa-Villasenor A, Sahm SH. 1996 Drinking water nitrate and the risk of non-Hodgkin Lymphoma. *Epidemiology* **7**, 465–471.

Worrall F, Burt TP. 2001 Inter-annual controls on nitrate export from an agricultural catchment – how much land-use change is safe?. *J Hydrol* **243**, 228–241.

Xing GX, Zhu ZL. 2002 Regional nitrogen budgets for China and its major watersheds. *Biogeochemistry* **57/58**, 405–427.

Yan WJ, Yin CQ, Zhang S. 1999 Nutrient budgets and biogeochemistry in an experimental agricultural watershed in Southeastern China. *Biogeochemistry* **45**, 1–19.

Yu S, Gao C, Cheng Z, Cheng X, Cheng S, Xiao J, Ye W. 1998 An analysis of chemical composition of different rain types in 'Minnan Golden Triangle' region in the southeastern coast of China. *Atmos Res* **47/48**, 245–269.

Zhang Y. 2003 *Modeling Agricultural Diffuse Pollution in the Wuchuan Catchment*. *Ph.D. thesis*, Xiamen: Xiamen University.

Zhu ZL 1997. Nitrogen balance and cycling in agroecosystem of China. In Zhu ZL, ed.*Soil Nitrogen in China.* London: Kluwer, pp. 323–338.