Net anthropogenic nitrogen inputs (NANI) index application in Mainland China

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ABSTRACT

This study provides a new understanding to sources of nitrogen (N), and may serve as a foundation for further exploration of anthropogenic effects on N inputs. Estimation of net anthropogenic nitrogen inputs (NANI) was based on an inventory of atmospheric N deposition, N fertilizer use, N in human food and animal feed, seeding N and N fixation. This study took a step forward to calculate NANI in detail on a regional scale, and analyzed its temporal variations and geographic differences. Over the past 28 years, NANI increased significantly in Mainland China, from 2360 kg N km⁻² yr⁻¹ to 5013 kg N km⁻² yr⁻¹. On a geographical basis, NANI was higher in southeast where China's eight major watersheds are located than in northwest, and the largest NANI, 26160 kg N km⁻² yr⁻¹, appeared in Shanghai. The administrative regions corresponding to Haihe watershed, Huaihe watershed and Tai lake watershed have the largest NANI. N input of fertilizer is the largest source of NANI, followed by atmospheric N deposition and N fixation. The primary factor in relation to the change in NANI is total population density, followed by cultivated land area and total grain yield. In those densely populated large cities and watersheds, reasonably allocating the social resources to reduce the existing population density is the most effective way to address the problem of high N inputs, while in those agriculture-dominated regions and watersheds, the most effective way for reducing NANI is to improve fertilizer utilization efficiency in agriculture.

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1. Introduction

China is one of the countries in the world with rapidest population and economy growth. In the past thirty years, population and economy growth bring about many environmental problems (Hu et al., 2011; Wu et al., 2011). The nutrient pollution is the most serious one. Even though some technology has been used to control nutrient pollution, the effect is imperceptible because the control is merely focused on end-control engineering projects, such as establishment of sewage farm and artificially-reclaim of wetland at bayou or watercourse (Guan et al., 2011; Liu et al., 2010). Therefore, it is essential to investigate the pollutant sources and find strategies to control these sources.

Eutrophication is a kind of water pollution caused by excessive nutrients. Nitrogen (N) is one of the main pollution elements of eutrophication. N emission from different pathways and sources is a key issue concerning the protection of water quality and sustainable watershed

management practices (Lowrance et al., 1985; Neset et al., 2008; Rivera et al., 2007).

The nutrient input to the watershed is mainly caused by human activities (Leeben et al., 2008), such as crop cultivation, livestock breeding, human excretion, household garbage and so on. Many researchers have drawn their attention to the evaluation of pollutant loadings by human activities. Jordan and Weller (1996) put forth the concept of net anthropogenic nitrogen inputs (NANI), which refers to the sum of the total amount of N derived from applied fertilizer, N fixation and atmospheric deposition and the total amount of N exported from food and feed in the watershed ecosystem. The study of McIsaac et al. (2001) shows that NANI is more sensitive to the N flux in the watershed, i.e. a slight change in NANI may result in a great change in riverine N content. Russell et al. (2008) also conducted a relevant investigation on net anthropogenic phosphorus inputs (NAPI), pointing out that NAPI is an important indicator of P pollution in watershed. The theories provide useful research methods for investigation on nutrient inputs (Bristow et al., 2008; Han et al., 2011). However, few of them pay sufficient attention to N in seed in their calculation of anthropogenic nitrogen. Therefore, we

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introduced a comparatively reasonable way of studying N inputs by taking N in seed into consideration, that is, $NANI = \text{atmospheric N deposition} + \text{N fertilizer use} + \text{seeding N} + \text{N in human food and animal feed} + \text{N fixation}$. [Ti et al. \(2012\)](#) studied N budget in mainland China and its impact on environment, but analysis on N sources and their change was insufficient, so they lacked deep understanding to the fate of accumulated N and did not mention the solution to high level of N. With NANI as a starting point, we conducted the following studies: (a) researching the spatial distribution and temporal variation of net N input; (b) analyzing the main sources of net N inputs and their change, and proposing the strategies for reducing NANI in different areas (those densely populated urban areas and those rural areas with more fertilizer application, etc.); and (c) discussing the relationship between typical socioeconomic factors and net N input. To some extent, this paper overcomes the difficulties in quantifying N sources and introduces a more reasonable way for studying N inputs.

2. Methods

2.1. Study sites

China is situated at 4° to 53°30' North Latitude and 73°40' to 135°05' East Longitude and composed of 23 provinces, 5 autonomous regions, 4 municipalities and 2 special administrative regions. The study region in this paper is Mainland China, namely, Taiwan province, Hongkong and Macao special administrative regions are excluded. Therefore, with each administrative region as a research unit, there are 31 research units in total as shown in [Fig. 1](#) and [Table 1](#).

We obtained the data from the National Bureau of Statistics of China. Land cover was derived from satellite images of Ministry of Land and Resources of China. We calculated net N input values for years 1981, 1990, 2000 and 2009.

2.2. NANI estimation

NANI was estimated by accounting for the anthropogenic N inputs in each unit. We quantified N inputs in each unit, most of which is derived from human activities: Net atmospheric N deposition, fertilizer use, seeding N, human food and animal feed, and N fixation (see [Fig. 2](#)).

2.2.1. Atmospheric N deposition

Atmospheric deposition includes dry deposition and wet deposition. Atmospheric N deposition in this paper includes both of them. [Ti \(2011\)](#) calculated the value of atmospheric N deposition in different districts of Mainland China between 1985 and 2007. Based on [Ti Chaopu's](#) conclusion, values of atmospheric N deposition were calculated in this paper by mean deviation for years 1981, 1990, 2000 and 2009 (see [Table 1](#)).

Atmospheric N deposition is associated with fertilizer use and human and animal N excretion. To avoid duplicating N inputs, all emitted and re-deposited N has been excluded ([Asman et al., 1998; Hutchings et al., 2001](#)). Thus, we estimated volatilization losses as a percentage of each applied fertilizer as (see 'input: nitrogenous fertilizer use'): 15% of urea, 2% of ammonium nitrate, 2.5% of ammonium bicarbonate, 0.1% of anhydrous ammonia, and 2% of other combined fertilizers ([Battye et al., 1994](#)). Human and animal N excretion is 2% ([Battye et al., 1994; Su et al., 2005; Yang et al., 2010](#)).

Fig. 1. Geographic location of Mainland China and distribution of 31 research units.

Table 1
Characteristics of Mainland China in 2010 and atmospheric N deposition in difference years ($\text{kg N km}^{-2} \text{yr}^{-1}$).

	Characteristics of Mainland China in 2010		Atmospheric N deposition in difference years ($\text{kg N km}^{-2} \text{yr}^{-1}$)			
	Area (ten thousands km^2)	Population (ten thousands)	1981	1990	2000	2009
Beijing	1.68	1755.00	1584	2133	2742	3291
Tianjin	1.13	1228.16	1327	2117	2994	3784
Hebei	19.00	7034.40	954	1386	1866	2298
Shanxi	15.60	3427.36	1443	2072	2772	3401
Inner Mongolia	118.30	2422.07	262	385	522	645
Liaoning	14.57	4319.00	1657	2077	2544	2964
Jilin	18.70	2739.55	1046	1353	1693	1999
Heilongjiang	46.90	3826.00	412	549	700	837
Shanghai	0.62	1921.00	3785	4827	5986	7029
Jiangsu	10.26	7725.00	2541	3296	4135	4890
Zhejiang	10.18	5180.00	2536	2850	3200	3514
Anhui	13.90	6131.00	1359	1802	2295	2739
Fujian	12.00	3627.00	1302	1675	2089	2462
Jiangxi	16.66	4432.16	1327	1712	2139	2524
Shandong	15.30	9470.30	2016	2592	3231	3806
Henan	16.70	9487.00	1406	2130	2935	3659
Hubei	18.74	5720.00	1183	1559	1978	2354
Hunan	21.00	6406.00	900	1244	1626	1969
Guangdong	18.60	9638.00	2008	2558	3170	3721
Guangxi	23.63	4856.00	998	1470	1995	2466
Hainan	3.40	864.07	820	1329	1895	2404
Chongqing	8.20	2859.00	1081	1658	2299	2875
Sichuan	48.80	8185.00	1458	1766	2109	2418
Guizhou	17.00	3798.00	864	1168	1506	1810
Yunnan	39.40	4571.00	648	946	1277	1575
Tibet	122.00	290.03	25	50	77	102
Shaanxi	20.50	3772.00	1537	2070	2662	3194
Gansu	45.00	2635.46	604	787	991	1174
Qinghai	72.00	557.30	478	491	506	519
Ningxia	6.64	625.20	793	1218	1690	2116
Xinjiang	160.00	2158.63	95	147	205	257
Mainland China	956.41	131660.69	677	898	1142	1363

2.2.2. N fertilizer use

Current management practices in agricultural production are highly dependent on intensive fertilizer use, so fertilizer use is one of the important sources of NANI. Estimates of fertilizer application are available for each subunit (data from National Bureau of Statistics of China), broken down by type: ammonium nitrate, anhydrous ammonia, ammonium bicarbonate, urea, and miscellaneous forms. Then we estimated volatilization losses as a percentage of applied fertilizers and aggregated the

Fig. 2. Flow of NANI.

subunit-level fertilizer data using GIS software by weighting each subunit's data. N content in different fertilizers is: 46% in urea, 35% in ammonium nitrate, 17% in ammonium bicarbonate, 82.3% in anhydrous ammonia, and 12.8% in other combined fertilizers (Liang, 1999).

2.2.3. Seeding N

We chose vegetable and 7 main agricultural crops to estimate seeding N in each research unit. The vegetable was represented by cabbage because the value of seeding N was slightly different between different vegetable species. Seeding N per unit area for each crop type has been reported by Agricultural Technology Promotion Center of China (1999) as shown in Table 2. The data on the cultivated area of each crop type were obtained from the National Bureau of Statistics of China. Seeding N was estimated by multiplying the seeding N input per unit area for each crop type by the cultivated area of the research unit.

2.2.4. Net N input in human food and animal feed

Human and animal require a great deal of food and feed during their lives, so net N in human food and animal feed is also an important source of NANI. Human food and animal feed are associated with input/output N of agricultural products. We used the method introduced by Jordan and Weller (1996) (Han et al., 2013; Russell et al., 2008), who quantified the net input of N as: *net input in food and feed = food and feed consumption of human and animal – animal production for human consumption – crop production*.

We obtained data on human population as well as crop products and animal products from the National Bureau of Statistics of China. Each item, with the final result, in this calculation is presented by research unit data.

2.2.4.1. Food and feed consumption. Human consumption of N in food was estimated by multiplying the number of inhabitants by the N consumption per capita. N consumption per capita per year was calculated based on food consumption per capita that was obtained from the National Bureau of Statistics. By calculating N content in main human food, we obtained that N consumption per capita per year was 3.92 kg, 4.67 kg, 4.75 kg and 4.58 kg in 1981, 1990, 2000, and 2009, respectively.

Animal consumption of N was obtained by multiplying the number of each animal species by N consumption of individual. We chose the consumption values as reported by Wu (2005) and the values of the percentage N excreted as reported by Van (1998) (see Table 3). We assumed all animals were completely formula fed, i.e., 50% from corn and 50% from pasture (Li, 2007).

2.2.4.2. Crop production. We chose vegetable and 14 main agricultural crops to estimate N production in each research unit. The vegetable was represented by cabbage because nitrogen content was slightly different between different vegetable species. We calculated N content in the crop harvest in each research unit. N content ($\text{g} \cdot \text{kg}^{-1}$) in each crop type has been reported by Wang (2003) as shown in Table 4. We assumed that pests, spoilage, and processing caused 10% loss for all crops (Da, 2013; Hu et al., 2012; Zheng, 2011).

Table 2
Input of seeding N.

Seed	Seeding N ($\text{g} \cdot \text{kg}^{-1}$)	Seeding N ($\text{kg} \cdot \text{km}^{-2}$)
Paddy	19.1	69.2
Wheat	28.2	227.1
Corn	14.3	25.9
Soybeans	59.2	107.5
Broomcorn	16.64	30.1
Peanuts	19.36	35.2
Cotton	30.4	5.8
Vegetables	33.6	2.8

Table 3

Animal N consumption and excretion (Van, 1998; Wu, 2005).

Animal species	Waste production (N excretion, kg N individual ⁻¹ yr ⁻¹)	Percent excreted	Consumption (N intake rate, kg N individual ⁻¹ yr ⁻¹)	Animal production (kg N individual ⁻¹ yr ⁻¹)
Hogs and pigs	11.51	69	16.68	5.17
Horses and cattle	48.79	89	54.82	6.03
Chickens	0.37	65	0.57	0.20
Ducks	0.41	65	0.63	0.22
Sheep and lambs	5.75	84	6.85	1.10

2.2.4.3. Animal production. Animal N production includes meat, milk, eggs, etc. We estimated animal N production as the difference between animal feed consumption (intake) and animal excretion (waste production). We assumed that spoilage and inedible components caused 10% loss of animal products available for consumption.

2.2.5. N fixation

Depletion of soil N in agriculture occurs primarily through plant N uptake. This is often replenished through fertilizer N additions, or, at least in part through natural processes, the most important of which is biological N fixation (BNF). It has been estimated that on a global scale, BNF may contribute some 90 million tons ha⁻¹ yr⁻¹ to agricultural systems (Bohloul et al., 1992). Burns and Hardy (1975) estimated BNF based on different land types (Dou, 1989) as 500 kg N km⁻² yr⁻¹, 1000 kg N km⁻² yr⁻¹, 1500 kg N km⁻² yr⁻¹, 0 kg N km⁻² yr⁻¹ and 100 kg N km⁻² yr⁻¹ for agricultural land, forest land, grass land, developed land and other land, respectively. Total N fixation in each subunit was derived by multiplying the value of fixation for each land cover type by its area. The data on the areas of agricultural land, forest land, grass land, developed land and other land came from the National Bureau of Statistics of China and satellite images of Ministry of Land and Resources of China.

2.3. Data analysis

Based on atmospheric N deposition, N fertilizer use, N in human food and animal feed, seeding N and N fixation, NANI was calculated by summing the values of these N sources in all research units within Mainland China. Geographic and spatial analysis of NANI was conducted using average NANI values of the 31 research units in years 1981, 1990, 2000 and 2009. The land cover data was analyzed with the ArcInfo 9.3 (ESRI Inc.) geographic information system to calculate the proportion of row crops and forest in each unit, and the number of animals in each unit was calculated from agricultural census data. We also investigated the change in NANI and in the sources of NANI between different

years. Grey relational grade analysis was conducted between the socio-economic factors (gross domestic product, gross agricultural output value, cultivated land area, total population density, agricultural population density, the total fertilizer, total population and total grain yield) and the NANI calculation.

3. Results

3.1. Temporal variation of NANI

Table 5 shows NANI in different years in Mainland China. Across Mainland China, the largest NANI value (5013 kg N km⁻² yr⁻¹) appeared in 2009. Over the past 28 years, NANI had an upward trend in all of these 31 research units, implying a rising trend in population growth and economic development. Cultivated area in Mainland China increased from 9.9×10^5 km² to 12.2×10^5 km², fertilizer use quantity increased from 133.49×10^5 ton to 539.06×10^5 ton and meanwhile population grew from 9.96×10^8 to 13.17×10^8 . Therefore, NANI was also rising. As for NANI in different research units, Henan (15941 kg N km⁻² yr⁻¹) had the greatest change over the last

Table 5

NANI of different research units in different years.

Name	NANI (kg N km ⁻² yr ⁻¹)				
	1981	1990	2000	2009	2009–1981
Beijing	7148	9836	13062	12668	5520
Tianjin	5437	7915	14726	19568	14131
Hebei	4487	7258	11406	12256	7769
Shanxi	3681	5307	6916	7581	3900
Inner Mongolia	1091	1310	1608	2117	1026
Liaoning	5384	6940	8716	9414	4030
Jilin	2732	4442	5919	6834	4102
Heilongjiang	1472	1650	2148	2306	834
Shanghai	21125	27919	30734	24896	3771
Jiangsu	10236	17457	23419	24219	13983
Zhejiang	7044	9943	10110	10964	3920
Anhui	5108	8777	12284	13121	8013
Fujian	4123	6241	8217	8501	4378
Jiangxi	3462	5145	6073	6608	3146
Shandong	8712	13241	18285	18072	9360
Henan	5149	11054	16610	21090	15941
Hubei	3882	7055	10292	12756	8874
Hunan	4162	5484	7163	8511	4349
Guangdong	6221	9210	10508	13115	6894
Guangxi	3122	4530	5981	7196	4074
Hainan	3057	4753	7289	9793	6736
Chongqing	4448	7016	8797	10103	5655
Sichuan	3360	4582	5548	6227	2867
Guizhou	2821	3984	5214	6035	3214
Yunnan	2016	2940	4160	5161	3145
Tibet	760	788	823	870	110
Shaanxi	3266	5257	7539	8855	5589
Gansu	1689	2245	2716	3173	1484
Qinghai	1292	1323	1329	1351	59
Ningxia	2236	3684	5495	6765	4529
Xinjiang	851	995	1195	1405	554
Mainland China	2360	3388	4401	5013	2653

Table 4

N content in agricultural crop products (Wang, 2003).

Crop	N (g kg ⁻¹)
Corn	14.08
Wheat	17.92
Soybean	56.16
Paddy	11.84
Millet	14.40
Broomcorn	16.64
Peanut	19.36
Potato	3.20
Chestnut	6.72
Apple	0.32
Grape	0.80
Peach	0.80
Pear	0.48
Watermelon	2.46
Vegetable	2.72

Fig. 3. Average NANI in each research unit in 1981, 1990, 2000 and 2009; eight major watersheds; and land use type in each research unit in 2000. The black outlines indicate the boundary of the units. Eight major watersheds are Songhuajiang, Liaohe, Haihe, Huanghe, Huaihe, Changjiang, Zhujiang and Taihu. The land use type is classified as forest land, agriculture land, grass land, developed land, river and other land.

28 years, followed by Tianjin ($14131 \text{ kg N km}^{-2} \text{ yr}^{-1}$) and Jiangsu ($13983 \text{ kg N km}^{-2} \text{ yr}^{-1}$).

3.2. Spatial variation of NANI

Fig. 3a shows the geographic difference of NANI average in 1981, 1990, 2000 and 2009. NANI is higher in southeast Mainland China than in northwest Mainland China. The southeast is mainly covered by agricultural and developed land, while the northwest is mainly covered by forest and desert (see Fig. 3c). Economy develops rapidly in the southeast, so it has a greater population density and a higher gross domestic product (GDP). Thus, NANI in the southeast is higher. Fig. 3a also shows that Shanghai has the largest NANI of $26160 \text{ kg N km}^{-2} \text{ yr}^{-1}$. Over the past 28 years, the average NANI was $3791 \text{ kg N km}^{-2} \text{ yr}^{-1}$ in Mainland China.

Fig. 3b shows China's eight major watersheds. Compared with Fig. 3a and c, these watersheds almost cover all those areas that have greater NANI, and these areas are covered by cultivated lands and developed lands. Therefore, these eight major watersheds have the most serious water pollution in China. The administrative regions of watershed 3 (Haihe watershed), watershed 5 (Huaihe watershed) and watershed 8 (Tai lake watershed) have greater NANI.

3.3. Change in NANI sources

The sources of NANI in Mainland China in different years were analyzed (see Table 6). N input of fertilizer is the largest source of NANI, followed by atmospheric N deposition and N fixation. N fertilizer use had an upward trend from 42.4% ($1000 \text{ kg N km}^{-2} \text{ yr}^{-1}$) to 60.6% ($3040.4 \text{ kg N km}^{-2} \text{ yr}^{-1}$) of NANI from 1981 to 2009, but the cultivated area increased merely from $9.90 \times 10^5 \text{ km}^2$ to $12.17 \times 10^5 \text{ km}^2$. As can be seen, fertilizer use per unit area had a sharp increase.

Over the past 28 years, value of atmospheric N deposition increased (from $677 \text{ kg N km}^{-2} \text{ yr}^{-1}$ to $1363 \text{ kg N km}^{-2} \text{ yr}^{-1}$), indicating the

atmospheric environmental deterioration that may be caused by fertilizer volatilization, automobile exhaust and so on. However, its percentage changed slightly due to more rapid increase of NANI.

Net N in human food and animal feed is proportional to the human population and animal number. In Table 6, the negative percentage values of N for human food and animal feed indicate that N output occurs.

China's population growth over the past 28 years leads to increase of N in human food from $306 \text{ kg N km}^{-2} \text{ yr}^{-1}$ to $451 \text{ kg N km}^{-2} \text{ yr}^{-1}$. However, the percentage values do not have significant change though N consumption per capita per year and total N consumption have a remarkable increase. This is because the agricultural food products and animal products also increased.

N fixation and N content in seed are related to land use. Over the past 28 years, there were some change in land use, but the value of N fixation and N content in seed changed insignificantly.

Change in NANI sources in the 31 research units over the past 28 years is shown in Table 7. N fertilizer use has the biggest change in these research units except Beijing, Shanghai, Tibet and Qinghai. Beijing and Shanghai are the most densely populated cities in China, and population change in these two cities is the most important factor for the change in NANI. Tibet and Qinghai have low population density and fewer areas of cultivated land but larger areas of desert, so the change in NANI in these two research units is mainly affected by atmospheric deposition.

4. Discussion

4.1. Relational analysis between socioeconomic factors and NANI

Grey relational grade analysis was conducted between the socioeconomic factors (gross domestic product, gross agricultural output value, cultivated land area, total population density, agricultural population density, total fertilizer, total population and total grain yield) and

Table 6

Sources of N inputs and their change ($\text{kg N km}^{-2} \text{ yr}^{-1}$). The symbol “–” means contribution to NANI reduction.

Year	Atmospheric N deposition	N fertilizer use	N in human food and animal feed					N fixation	N content of the seed	NANI
			Food consumption of human	Feed consumption of animal	Animal production for human consumption	Crop production	Total			
1981	677(28.7%)	1000(42.4%)	306	395	243	495	–37(–1.6%)	711(30.1%)	9(0.4%)	2360
1990	897(26.5%)	1842(54.4%)	437	500	304	705	–71(–2.1%)	709(20.9%)	10(0.3%)	3388
2000	1142(26.0%)	2592(58.9%)	509	607	373	817	–74(–1.7%)	727(16.5%)	13(0.3%)	4401
2009	1363(27.2%)	3040(60.6%)	553	528	337	869	–125(–2.5%)	723(14.4%)	13(0.3%)	5013
Average	1020(26.9%)	2119(55.9%)	451	508	314	722	–77(–2.0%)	718(18.9%)	11(0.3%)	3791

Table 7
Change in NANI sources ($\text{kg N km}^{-2} \text{ yr}^{-1}$). These values were obtained by the values of 2009 minus the values of 1981. The alphabet after the value indicates the ranking of change in the same row, wherein (a) indicates the greatest change, followed by (b) and (c). The symbol “–” means contribution to NANI reduction.

Name	Atmospheric N deposition	N fertilizer use	N in human food and animal feed					N fixation	N content of tsathe seed
			Food consumption of human	Feed consumption of animal	Animal production for human consumption	Crop production	Total		
Beijing	1706(b)	932(c)	2661(a)	–125	90	329	2954	–59	–13
Tianjin	2456(b)	10228(a)	2241(c)	542	–450	–885	1448	–9	8
Hebei	1344(b)	6960(a)	674(c)	498	–315	–1397	–540	–9	13
Shanxi	1957(b)	2038(a)	405(c)	–92	23	–438	–102	5	2
Inner Mongolia	382(b)	788(a)	36	89(c)	–44	–235	–154	9	1
Liaoning	1307(b)	2450(a)	466	583(c)	–335	–455	259	13	2
Jilin	952(b)	3814(a)	231	463(c)	–242	–1160	–708	39	5
Heilongjiang	425(b)	1435(a)	128	243(c)	–141	–1293	–1063	34	3
Shanghai	3244(b)	2155(c)	5177(a)	–629	587	620	5755	–66	–7
Jiangsu	2349(b)	11862(a)	1242(c)	–224	134	–1401	–249	6	16
Zhejiang	978(b)	1486(a)	887(c)	–204	112	656	1452	5	–1
Anhui	1381(b)	7466(a)	712(c)	–18	–107	–1490	–904	46	24
Fujian	1160(b)	2458(a)	564(c)	228	–245	211	759	2	0
Jiangxi	1197(b)	1978(a)	481(c)	253	–199	–574	–39	9	1
Shandong	1790(b)	8278(a)	1029(c)	623	–415	–1980	–743	9	26
Henan	2253(b)	15157(a)	981	1663(c)	–1215	–2965	–1536	24	43
Hubei	1171(b)	7678(a)	482(c)	360	–312	–536	–5	25	5
Hunan	1069(b)	3162(a)	467	676(c)	–562	–473	108	9	1
Guangdong	1713(b)	3365(a)	1182(c)	–775	621	800	1827	–10	–1
Guangxi	1468(b)	2674(a)	371(c)	–526	310	–260	–105	33	5
Hainan	1584(b)	4825(a)	508(c)	–720	393	102	282	39	6
Chongqing	1795(b)	4079(a)	432(c)	220	–213	–646	–206	–10	–3
Sichuan	960(b)	1812(a)	238	290(c)	–198	–233	97	–1	0
Guizhou	946(b)	2151(a)	391	406(c)	–274	–492	31	75	11
Yunnan	927(b)	2259(a)	230	305(c)	–223	–400	–88	42	6
Tibet	77(a)	17(c)	5	19(b)	–6	–3	15	1	0
Shaanxi	1657(b)	4010(a)	324(c)	29	–52	–387	–86	6	1
Gansu	570(b)	908(a)	120	131(c)	–62	–198	–10	14	2
Qinghai	41(a)	31(b)	15(c)	–23	6	–11	–12	0	0
Ningxia	1323(b)	3423(a)	265	266(c)	–127	–647	–243	22	4
Xinjiang	162(b)	459(a)	29(c)	19	–11	–109	–71	3	1
Mainland China	685(b)	2041(a)	246(c)	133	–93	–374	–88	12	4

NANI calculation with a discrimination coefficient (ρ) of 0.5. The results are as follows (see Table 8):

$$X_4 > X_9 > X_3 > X_6 > X_7 > X_5 > X_1 > X_8 > X_2.$$

The primary factor in relation to the change in NANI is total population density, followed by total grain yield and cultivated land area. Among all of the factors, total population density, total population and agricultural population density are main social factors and their relational grades are 0.9235, 0.8098 and 0.8027, respectively. Total population density has the greatest relational grade, indicating that change in NANI is mainly determined by the total population density. Cultivated land area, total grain yield and total fertilizer use are main agricultural factors and their relational grades are 0.8706, 0.8710 and 0.8426, respectively. As can be seen, cultivated land area has the greatest relational grade, indicating that among the agricultural factors, cultivated land area is a main factor in relation to the change in NANI. Gross domestic product and gross agricultural output value are main economic factors and their relational grades are 0.7985 and 0.6984, respectively. Gross domestic product has a greater relational

grade, so it is the closest economic factor in relation to NANI change in this study.

4.2. Vary of per capita food consumption

As shown in Table 9, from 1980 to 2009, the grain and vegetable consumption increased firstly and then decreased, while the consumption of meat, egg, milk and aquatic product showed an increasing trend, indicating that the diet of Chinese people was gradually changing from a simply adequate meal type to a nutritious meal type, which leads to NANI change. In the densely populated major cities, such as Beijing and Shanghai, and their corresponding watersheds, population growth and diet habit change are the most importance factors for rapid increase of NANI (see Table 7). A large amount of N input leads to regional eutrophication. Therefore, the most effective way for solving the problem of high level of N input is to reasonably allocate the social resources to reduce the existing population density. Meanwhile, the decline in population density is also helpful for reduction in non-food N input, such as vehicle exhaust, feces evaporation, etc., and therefore objectively reduces atmospheric N deposition. In addition, more reasonable and nutrient diet may also contribute to the reduction of N input.

Table 8
Grey relational grade analysis.

	Gross domestic product	Gross agricultural output value	Cultivated land area	Total population density	Agricultural population density	Total fertilizer	Total population	Total area	Total grain yield
	X ₁	X ₂	X ₃	X ₄	X ₅	X ₆	X ₇	X ₈	X ₉
Relational grade	0.7985	0.6984	0.8706	0.9135	0.8027	0.8426	0.8098	0.7274	0.8710

Table 9
Per capita consumption food (kg).

	1980	1990	2000	2009
Crop	219.00	262.08	250.23	232.00
Vegetable	83.00	134.00	106.74	98.44
Meat	11.08	12.59	18.30	21.53
Egg	1.03	2.41	4.77	5.32
Milk	0.52	1.10	1.06	3.60
Aquatic product	0.89	2.13	3.92	5.27

4.3. Grain yield

World-wide agricultural development has proved that fertilizer application is the most effective and important means for increasing production. China is an intensive-agriculture country, and agricultural production is highly dependent on fertilizer application (Chen et al., 2007; Li et al., 1998). The cultivated land area in China is 1/7 of the total of the world, but its fertilizer application accounts for 1/3 of the total, ranking first in the world. In this study, the total grain yield in 2009 was 1.62 times that in 1981, while the amount of applied fertilizer was 4.04 times (see Table 10). Grain yield per unit mass of fertilizer showed an obvious downward trend, indicating that the amount of applied fertilizer per unit weight of grain was gradually increasing, i.e., the fertilizer utilization efficiency was gradually reducing. Low fertilizer utilization efficiency may decline soil fertility, degrade crop quality and lead to serious environmental pollution. Therefore, in those agriculture-dominated research units and watersheds, it is significant to improve fertilizer utilization efficiency in agriculture because in these units it is the most effective way for reducing NANI and is one of the most important means for mitigating nutrient point source pollution.

4.4. Method for improving fertilizer utilization efficiency

China is facing harsh problems of growing population, shrinking arable land and increasing food demand. Maximization of fertilizer utilization efficiency is extremely important for food security and ecological environment protection (Hu and Li, 2005; Yan et al., 2008). At present, the main techniques and methods are: 1. fertilizing accurately in accordance with the farmland nutrients and promoting formulated fertilization technology; 2. developing organic fertilizers; and 3. improving the methods of fertilizing. Firstly, based on the rule of fertilizer requirement of crops, soil testing results and fertilizer utilization efficiency, the amount of fertilizer is reasonably adjusted to supply the crop with comprehensive and reasonable nutrients so as to maximize the crop's potential in increasing yield, thereby improving economic efficiency (Gao and Lü, 2006; He et al., 2007; Liu et al., 2006; Peng et al., 1996, 2002). Secondly, the majority of nitrogen-supplying organic fertilizers contain insoluble nitrogen and act as a slow-release fertilizer, so they can increase physical and biological nutrient storage mechanisms in soils and mitigate risks of over-fertilization. Organic fertilizer nutrient content, solubility, and nutrient release rates are typically much lower than mineral (inorganic) fertilizers (Du et al., 2005). A University of North Carolina study found that potential mineralizable nitrogen (PMN) in the soil was 182–285% higher in organic mulched systems than in the

synthetics control (Do et al., 2013; Tu et al., 2006). Thirdly, according to the specific fertilizer demand, crop should be fertilized reasonably in different growth stages to satisfy the nutrient demand at each stage so as to achieve the purpose of economic fertilization (Jess, 2004; Lu and Wu, 2004; Wang, 2002; Yu and Zhang, 2006). Deep application of fertilizer also needs to be promoted to minimize the loss of fertilizer and improve fertilizer utilization efficiency.

4.5. Fate of NANI

There is a high level of N input in Mainland China every year. But, where has it gone? Part of it is deposited in plants and animals. N is essential for animals and plants as it is an important component of nucleic acid in animals and is indispensable for photosynthesis and respiration in plants. Part of it is deposited in soil. After a fertilizer is applied into soil, plant retains no more than 50% of the fertilizer (Wang et al., 2010), and the rest of the fertilizer is deposited in soil, resulting in many environmental problems such as soil compaction (Gregorich et al., 2011; Jordan et al., 2003; Ruser et al., 2006). Part of it is input in groundwater. N accumulated in the soil infiltrates into the groundwater or flows in runoff to rivers and streams. The other part N in human excretion flows through sewers into rivers. N in groundwater and rivers leads to eutrophication and causes loss of ecological functions of water, thereby resulting in serious environmental problems such as the death or even extinction of aquatic animals and plants. For example, 56% of the Yangtze River Estuary and its subsidiaries have been heavily polluted due to high N concentration (Quan et al., 2005). Algal blooms frequently occurred in recent years in Taihu Lake, located at the lower reaches of the Yangtze River (Qin et al., 2007). These environmental problems are thought to be directly related to the increased N export to water bodies (Xu et al., 2009).

By the end of 2009, 53.2% of household sewage in Mainland China could be treated by sewage treatment plants (Zhou, 2010). After the treatment, 90% of sewage N was left in the sludge and would eventually be landfilled. In recent years, researches on re-use of sewage sludge are being conducted (e.g., used as agricultural fertilizer or used for biogas production), but they have not yet been applied on a large scale (Xiong and Zhang, 2007).

5. Conclusion

In mainland China, NANI has increased with time over the past 28 years, and it is higher in the southeast than in the northwest. N input of fertilizer is the largest source of NANI, followed by atmospheric N deposition and N fixation. China's eight major watersheds are located in the densely populated areas and cultivated areas, and these areas coincide with those areas that have higher NANI. Haihe watershed, Huaihe watershed and Tai lake watershed have the largest NANI. The primary factor in relation to the change in NANI is total population density, followed by cultivated land area and total grain yield. In those densely populated large cities, human N consumption contributes the most to the NANI change, so the most effective way to address the problem of high level of N input is to rationally allocate the social resources to reduce the existing population density. In those agriculture-dominated regions, increase in the amount of applied fertilizers is the most important factor causing increase in NANI, the most effective way for reducing NANI in these regions is to improve fertilizer utilization efficiency in agriculture.

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Table 10
Grain yield per unit mass of fertilizer.

Year	Total fertilizer (10 ⁷ kg)	Fertilizer N (10 ⁷ kg)	Total grain yield (10 ⁷ kg)	Grain yield per unit mass of fertilizer (kg·kg ⁻¹)	Grain yield per unit mass of fertilizer N (kg·kg ⁻¹)
1981	1334.9	950.3	32502.0	24.3	34.2
1990	2590.3	1751.1	44624.3	17.2	25.5
2000	4146.4	2464.5	46217.5	11.1	18.8
2009	5390.6	2890.4	52657.9	9.8	18.2

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