

Description and Application of a Model for Simulating Regional Nitrogen Cycling and Calculating Nitrogen Flux

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(Received 11 December 2006; revised 30 April 2007)

ABSTRACT

A regional nitrogen cycle model, named IAP-N, was designed for simulating regional nitrogen (N) cycling and calculating N fluxes flowing among cultivated soils, crops, and livestock, as well as human, atmospheric and other systems. The conceptual structure and calculation methods and procedures of this model are described in detail. All equations of the model are presented. In addition, definitions of all the involved variables and parameters are given. An application of the model in China at the national scale is presented. In this example, annual surpluses of consumed synthetic N fertilizer; emissions of nitrous oxide (N_2O), ammonia (NH_3) and nitrogen oxide (NO_x); N loss from agricultural lands due to leaching and runoff; and sources and sinks of anthropogenic reactive N (Nr) were estimated for the period 1961–2004. The model estimates show that surpluses of N fertilizer started to occur in the mid 1990s and amounted to 5.7 Tg N yr^{-1} in the early 2000s. N_2O emissions related to agriculture were estimated as 0.69 Tg N yr^{-1} in 2004, of which 58% was released directly from N added to agricultural soils. Total NH_3 and NO_x emissions in 2004 amounted to 4.7 and 4.9 Tg N yr^{-1} , respectively. About 3.9 Tg N yr^{-1} of N was estimated to have flowed out of the cultivated soil layer in 2004, which accounted for 33% of applied synthetic N fertilizer. Anthropogenic Nr sources changed from 2.8 (1961) to 28.1 Tg N yr^{-1} (2004), while removal (sinks) changed from 2.1 to 8.4 Tg N yr^{-1} . The ratio of anthropogenic Nr sources to sinks was only 1.4 in 1961 but 3.3 in 2004. Further development of the IAP-N model is suggested to focus upon: (a) inter-comparison with other regional N models; (b) overcoming the limitations of the current model version, such as adaptation to other regions, high-resolution database, and so on; and (c) developing the capacity to estimate the safe threshold of anthropogenic Nr source to sink ratios.

Key words: nitrogen cycle, nitrous oxide, reactive nitrogen, model, agriculture, China

DOI: 10.1007/s00376-008-0181-7

1. Introduction

In the pre-industrial era, nitrogen (N) flowed steadily across the globe at ca. 140 Tg (1 Tg = 10^{12} g) N yr^{-1} (Socolow, 1999; Galloway et al., 2004), through the loop of dinitrogen (N_2) fixation by biological processes (BNF) and N_2 release by denitrification of reactive N (Nr, including all forms of N, except N_2). In the last century, human creation of Nr went from being of minor importance to becoming the dominant force in the transformation of N_2 to Nr (Galloway et al., 2004). By the early 1990s, for instance, the amount of Nr created by natural terrestrial processes had decreased by ca. 15%, while Nr created by anthropogenic processes

increased by ca. tenfold, as compared to 1860 (Galloway et al., 2004). Consequently, the conversion rate of N_2 to Nr due to natural and anthropogenic processes on the global scale amounted totally to 268 Tg N yr^{-1} in the early 1990s, of which 156 Tg N yr^{-1} was due to anthropogenic activity, while 112 Tg N yr^{-1} was due to natural terrestrial processes and lightning (Galloway et al., 2004).

Much of the new Nr created is dispersed throughout the continents' various environments. Currently, about 30% is transferred (via atmospheric and riverine transport) to the marine environment, where most is further converted to N_2 by denitrification, about 4% is emitted to the atmosphere as nitrous oxide (N_2O),

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about 43% is converted to N_2 , and about 23% is accumulated in biomass and soils of terrestrial systems (Galloway et al., 2004). It is predicted that Nr creation by anthropogenic activity will be further intensified this century (Galloway et al., 2004). During dispersion, redistribution and conversion processes of new Nr created intensively by human activity, negative cascade effects have widely occurred and will further occur, consequently threatening environmental quality, biodiversity and even climate (e.g., Galloway et al., 2003; Zheng et al., 2002, 2006). These negative cascade effects are realistically or potentially caused by not only further transference and transformation of long-lived Nr stored in terrestrial, aquatic and atmospheric systems, but also by fast conversion and turnover of short-lived Nr in ecosystems and environmental systems (e.g., Galloway et al., 2003; Ju et al., 2004). Therefore, both the public and decision-makers are urgently anticipating having to reduce or avoid negative cascade effects of anthropogenic Nr, created intentionally or involuntarily.

Establishment of any countermeasures to effectively reduce or even eliminate the negative impacts of anthropogenic Nr has to rely on a sound scientific basis. This requires quantitative understanding of N transference and conversion processes at different temporal, or spatial, scales. As potential robust tools, regional N cycle models may meet this requirement and provide such a basis for decision-making. We have recently designed a regional N cycle model, named IAP-N, to quantitatively assess the potential influences of human activity on regional N cycles, with the purposes of providing a firm scientific grounding for N management in terms of protecting food security and environmental health. In the present paper, we describe for the first time the model's conceptual structure and modeling processes, as well as detailed specification of its parameterization schemes. In addition, an applied example at the national scale in China is presented for a period covering the last four and a half decades.

2. Key nitrogen cycling processes: a basis for the conceptual structure of IAP-N

The objective for designing IAP-N was to assess quantitatively the dynamics of transferring fluxes or conversion rates of anthropogenic Nr among the linked components of regional N cycles. For this purpose, the following human activities relevant to creation, redistribution and conversion processes of Nr are involved: N fertilizer application, cultivation of legume crops and paddy rice, livestock production, manure management, and energy production and consumption. To reach the above objective, the expectation is for IAP-N

to be capable of estimating the following amounts on regional scales at annual or decadal timescales: (a) the surplus of consumed synthetic N fertilizer relative to the demands of maintaining crop yields; (b) emissions of individual gaseous Nr species from agricultural activities and energy consumption; (c) discharges of Nr into aquatic systems through runoff and leaching; and (d) environmental Nr enrichment. Therefore, IAP-N was designed to simulate the whole regional N cycle in association with agricultural activities and energy consumption. The simulation is realized by describing the Nr flowing among the agricultural, environmental and other components. Figure 1 illustrates the N linkages among different components, and details are stated below.

2.1 *Agricultural components*

The agricultural components include crops, humans and livestock, and the cultivated soil layer. They contribute to atmospheric Nr through direct emissions from cultivated soils, biomass burning of crop residues, manure management, and indirect emissions from water bodies, including ponds, reservoirs, lakes, rivers, and oceans (IPCC, 1997, 2000). Crops take up the N nutrients from the cultivated soil layer. Humans and livestock acquire N in protein and plant tissues from crop products. Nr is also discharged from the cultivated soil layer by leaching and runoff processes.

2.1.1 *Cultivated soil layer*

The cultivated soil layer directly provides N nutrients for crops, while it receives Nr through the amendment of synthetic N fertilizers, BNF, application of human and livestock manure, deposition of Nr from the atmosphere, direct return of crop residues, and remnant particulate N in ashes of residue burning (PNS). It also loses Nr through (a) moving nitrate to deep-layer sediments, groundwater, and/or surface water by leaching; (b) moving nitrate, ammonium, and particulate N to surface waters by runoff; and (c) releasing gaseous N through denitrification, nitrification and ammonia (NH_3) volatilization. Nitrogen incorporated into the cultivated soil layer is assumed to remain there for no more than two years. Nr remnants from the previous year are assumed to be lost through leaching from the cultivated soil layer.

2.1.2 *Crops*

Crops take up Nr from cultivated soils. At the end of the growing season, Nr taken up by crops is partitioned into human food, animal feed, residues for bio-fuel and/or wastes, organic matter returned to the cultivated soil layer in the form of roots, stubble and litter, particulate N returned to the soil in the ashes from residue burning in the fields, and residues used

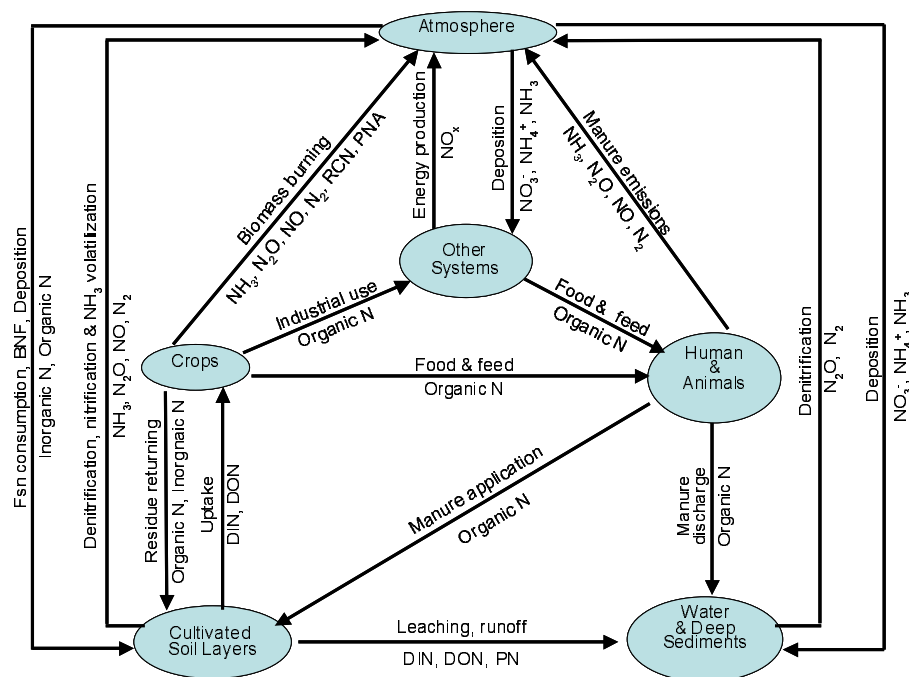


Fig. 1. Key N cycling processes and conceptual structure of IAP-N.

as industrial materials.

2.1.3 Humans and livestock

Humans and livestock are the consumers of crop products. They primarily receive organic Nr from crop products in the form of human food and animal feed. They also receive minor organic Nr from other systems (e.g., natural grassland, international/inter-regional trade markets, oceanic fisheries, and forests). While the Nr in manure produced by humans and livestock is stored in various management systems, part of it is lost in the form of NH_3 , N_2O , nitric oxide (NO), and N_2 . Most of the Nr in manure in agricultural areas is returned to cultivated soils or grasslands, while a minor amount is directly discharged into aquatic and other systems or burned as fuel. All of the Nr produced in urban areas by humans is discharged into the environment.

2.2 Environmental systems

2.2.1 Atmosphere

Dinitrogen, which is biologically unavailable, makes up 78% of the atmosphere where there are various Nr species. The Nr species in the atmosphere include N_2O , NO_y (including NO , NO_2 , NO_3^- , NO_2^- , HNO_3 , N_2O_3 , N_2O_5 and N_2O_4), NH_x (including NH_3 and NH_4^+), and ON (organic N, including reductive, oxidized, and organism organic N (Graedel et al., 1986). Nitrous oxide is a greenhouse gas (GHG) and is one of the major GHGs to be controlled under the

Kyoto Protocol of the United Nations Framework Convention on Climate Change. Nitric oxide and nitrogen oxide (NO_2), which catalyze key photochemical reactions in the atmosphere, are the most important atmospheric NO_y species. They play an indirect role in the greenhouse effect, as they contribute to tropospheric ozone production and stratospheric ozone destruction (IPCC, 2000). They also act as precursors of HNO_3 , which in turn contributes to acid rain. PAN (i.e., $\text{CH}_3\text{COO}_2\text{NO}_2$) is a major component of oxidized organic N, which contributes to the photochemical haze in polluted atmospheres. Nitric oxide and nitrogen oxide are removed from the atmosphere through dry or wet deposition after transformation into HNO_3 , NO_3^- and PAN. Nitrate usually stays for a long time in the atmosphere and may be transported great distances. Therefore, atmospheric transportation of NO_3^- by advection is a major pathway for the transfer of Nr from land to the open ocean. When ammonia is transformed into ammonium nitrate (NH_4NO_3), which is a very important aerosol component, it may then be quickly removed from the atmosphere through dry/wet deposition. Thus, ammonia usually stays in the atmosphere for at most a few days before deposition within a short distance from its source (Wang, 1999). The atmosphere receives anthropogenic Nr due to emissions from soils, biomass burning, manure management, and waters, while it loses Nr through chemical/photochemical transformation and dry/wet deposition. Deposition of atmospheric Nr also provides N

nutrients for downwind ecosystems.

2.2.2 *Water and deep sediments*

Water systems here include continental ground and surface water bodies, continental shelves and open ocean. Deep sediments indicate the part of soil profiles lower than the cultivated layers. In the current version of IAP-N, the N exchanges between water bodies and the beds of rivers, lakes and oceans are not taken into account. Both water and deep sediments receive Nr mainly from cultivated soil layers through leaching and runoff. Water bodies also receive Nr due to discharge of human/animal manure/sewage. Ocean receives Nr from continents via atmospheric advection and deposition. Meanwhile, water and deep sediments lose Nr through denitrification, whereby Nr is transformed into N_2O and N_2 and released finally to the atmosphere.

2.2.3 *Other systems*

These mainly include non-crop land, international/inter-regional trade markets and industries. They receive Nr from humans and livestock in the form of manure, and from crops as industrial materials, while they provide food and feed for humans and livestock. Non-crop land also receives Nr from atmospheric deposition.

3. Description of the model calculation procedures

The calculation procedures of IAP-N start from estimating the surplus of consumed synthetic N fertilizer, which in turn is defined as the N residual of actual consumption and potential demand of synthetic N fertilizer needed in order to sustain harvested or expected crop production [Eq. (A1) in Appendix A].

Quantification of all the N flows illustrated in Fig. 1 is involved in the surplus calculation. To calculate the regional N surplus as well as all associated regional N flows on the annual timescale, the model is designed to drive by annual regional statistical data on crops (type, harvest area, yield), livestock (type, population) and fossil energy (type, consumption). There are 13 crop types involved, which are: maize; wheat; rice; sorghum; barley and rapeseed, soybeans; pulses (excluding soybean); seed-cotton; root and tuber vegetables; sugar cane; sugar beet; and vegetables (including melons). The livestock types are divided into six categories: swine; sheep; dairy cattle; non-dairy cattle; poultry, including chicken, turkey, duck, and geese; and other animals, including asses, camels, goats, horses, and mules. The fossil energy consumption is categorized into three types: petroleum, coal,

and dry natural gas.

All formulae for the calculation procedures are given in Appendix A. Any equation cited below refers to Appendix A, with its variables defined in Appendix B, and its parameters defined and evaluated, with citation sources for the values, in Appendix C. When the variable, parameter, or name of the formulae is given in the text, their subscripts such as i (animal-type code), j (crop-type code) and k (year code) are not included. Further, detailed parameters, including N excretion rates of livestock animals and humans, harvest index, root-to-shoot ratio, dry fraction of harvested product, and N contents in harvested products and residues of crops, as well as emission factors of individual Nr species from biomass burning, are provided in Appendices D, E and F, which are also linked with Appendix C.

It should be noted that the current version of the IAP-N model does not include Nr created by the natural processes of non-agricultural BNF and lightning. Also not included are the processes of N_2O emission from fossil fuel consumption and industrial processes. The calculation procedures will now be described in detail.

3.1 *Surplus of consumed synthetic nitrogen fertilizer*

To estimate the surplus of synthetic N fertilizer using Eq. (A1) (Appendix A), the key process is to estimate synthetic N fertilizer demand while consumed synthetic N fertilizer is usually inputted as annual statistical data or given scenarios.

As Eq. (A2) in Appendix A describes, synthetic N fertilizer demand is estimated by taking into account N uptake by crops (N_{Up}), BNF by rice (N_{Bnfr}) and legume cultivation (N_{Bnfl}), N remnants in the cultivated soil layer from the previous year (N_{Sg}), organic manure application and/or residue incorporation (N_{Om}), and atmospheric deposition (N_{Dm}). The use efficacies of these N supplies are the key parameters for estimating synthetic N fertilizer demands. China-specific N use efficiencies, $27\% \pm 11\%$ and $36\% \pm 15\%$ (Zhu and Wen, 1992), are adopted for organic manure and mineral N, respectively. Considering that the remnant soil N from the previous year and the deposited N from the atmosphere are available not only in the crop growing seasons but also during other periods, their use efficiency by crops is assumed to be the mineral N use efficiency. As BNF in submerged soils is mainly available during the rice-growing season, its use efficiency is assumed to be the same as that of minimal N fertilizer. The BNF by legumes is assumed to fully take up, i.e., with a use efficiency of 100%. The specification of all these N use efficiencies is also given in

Eq. (A2).

To estimate the N uptake of all crops and BNF in legume products and residues (including roots), the N storage in harvested products (N_Hp), straw and stubbles (N_Rs), and roots (N_Rr) of individual crops are estimated separately with harvested yields (P_Hp) and relevant parameters [Eqs. (A3)–(A7) in Appendix A]. The harvested yields are inputted as annual statistical data or given scenarios. The parameters involved are: dry matter fraction of harvest yields (f_Dr); harvest index (LHi, i.e., the ratio of economic yield to the sum of economic yield and harvested residue or straw); the shoot-to-root ratio (R_Rs); and the N content in residues (f_Nr) harvested products (f_Np). As shown by Eq. (A8) in Appendix A, the N biologically fixed in paddies during the rice season is estimated as the product of a BNF rate on an area basis and the rice harvest area (A_Ra), which in turn is inputted as statistical, or remote sensing, data or given scenarios.

The applied or incorporated organic N is estimated as the sum of the N in returned, or remnant, crop residues, and the N in manure produced by livestock and humans in rural areas (N_Mah) [Eq. (A9) in Appendix A]. The fraction of returned straw to total non-root residue is a key parameter for estimating the organic N input in the form of crop residue. The N in manure is calculated using statistical populations of livestock and humans in rural areas (P_Ah); the parameters of the N excretion rate (f_Wah); excreta partitioning fractions into management systems, excluding grazing and fuel usage; and the emission factor of NH₃ plus NO_x (f_Enn) during storage [Eq. (A10) in Appendix A].

The N deposited on croplands from the atmosphere originates from particulate N (PNA) emitted from residue burning [Eq. (A11) in Appendix A] and emissions of reactive gaseous N (N_Ern) from crop residue burning (N_Erb), from soils of dry croplands and paddy rice fields (N_Ers), from manure storage in various management systems for animal and rural human wastes (N_Erm), and from fossil energy consumption (N_Ere) [Eq. (A12) in Appendix A]. The deposited PNA is calculated with the fraction of burned residues (f_Rb), the emission factor (f_Pna), and total N in non-root residue production. Emitted reactive gaseous N to the atmosphere is assumed to deposit partially back to croplands, while the remainder is assumed to be deposited in non-crop systems on land as well as in the ocean. These two parts are defined with the ratio of cropland acreage to total land area (r_Cl) and the fraction of deposition to continental area (r_Dl). The reactive gaseous N released from soils of dry croplands (NH₃ and NO) and paddy rice fields (NH₃) is calculated with Eq. (A13) in Appendix

A, which integrates the emissions from N sources of atmospheric deposition to croplands in the previous year, amended/incorporated organic manure and/or crop residues, synthetic fertilizers, and BNF by rice and legume crop cultivation. Specific NH₃ factors are used for dry croplands (f_Nh3sfu) and paddy rice fields (f_Nh3sfp). For the first simulated year, the contribution of N deposited from the atmosphere into soils of either dry croplands or paddy rice fields in the previous year is regarded to be the same as that of the first year [Eq. (A13) in Appendix A]. The reactive gaseous N released from crop residue burning is estimated by taking into account the emission factors of NH₃ (f_Nh3), NO_x (f_Nox), and gaseous organic N (f_Rcn) and the amount of burned residues [Eq. (A14) in Appendix A]. The reactive gaseous N emissions from management of livestock and rural human manure [Eq. (A15) in Appendix A] is due to both the emission factor of NH₃ plus NO (f_Enn) and excreta N [Eq. (A16) in Appendix A]. The NO_x emission from fossil energy consumption is estimated with consumed coal (C_Ct), dry natural gas (C_Gt) and petroleum (C_Pt), which could be inputted as annual statistical data or given scenarios, and corresponding emission factors [Eq. (A17) in Appendix A]. The NO_x emissions from petroleum combusted in industries, road transportation, residential consumption and other consumption are quantified with partitioning fractions and specific emission factors [Eq. (A18) in Appendix A].

The N remnants or gains in the cultivated soil layer are defined as the difference between annual N input (N_In) and output (N_Ot) [Eq. (A19) in Appendix A]. Nitrogen is annually inputted into the cultivated soil layer through synthetic fertilizer application, organic matter incorporation or amendment, BNF by rice and legume cultivation, and atmospheric deposition. Meanwhile, the N gain in the previous year is also considered as one of the input items [Eq. (A20) in Appendix A]. For the first simulated year, however, the input item of N gain in the previous year is simply assumed as zero. The N input from BNF by legume crops is quantified with Eq. (A4) in Appendix A.

As Eq. (A21) in Appendix A describes, the N annually flowing out of the cultivated soil layer occurs via crop uptake (N_Up), leaching plus runoff (N_Lr), and emissions of NH₃ plus NO (N_Ers), N₂O (N_En2os), and N₂ (N_En2s). The fractions for N loss due to leaching and runoff are different among the areas of non-legume upland crops, legume upland crops, and paddy rice fields [Eq. (A22) in Appendix A]. As Eq. (A23) in Appendix A means, emissions of N₂O are counted as the product of the total N input into the cultivated soil layer and the direct N₂O emission factor (f_n2os). Due to different soil moisture conditions,

different emission factors are used to calculate the denitrification loss of N_2 from paddy rice fields and non-rice croplands [Eq. (A24) in Appendix A].

3.2 Gaseous Nr emissions from agricultural activity and energy consumption

Gaseous Nr (N_{Gnr}) emitted from agricultural activity and energy consumption to the atmosphere includes N_2O (N_{N2o}) and other reactive gaseous N [Eq. (A25) in Appendix A]. The latter consists of NH_3 , NO and organic N compounds. Their emissions (N_{Ern}) are quantified with Eqs. (A12)–(A17) in Appendix A, with details having been described above. On regional/landscape scales, N_2O emissions due to agricultural activity and agriculture-related energy production [Eq. (A26) in Appendix A] consist of the release from the N input into cultivated soils (N_{En2os}), the N removed from the cultivated soil layer to deep sediments and water bodies due to leaching and runoff (N_{En2olr}), manure management systems (N_{N2om}), atmospheric deposition of N to non-agricultural land and water bodies (N_{En2od}), and crop residue burning (N_{N2ob}). As stated above, N_2O emissions from the cultivated soil layer are estimated with Eq. (A23) in Appendix A, of which the N source from atmospheric deposition [Eqs. (A19) and (A29) in Appendix A] is included. N_2O emissions from the N removed from the cultivated soil layer by leaching and runoff (N_{N2olr}) are calculated as the product of the N amount and the emission factor [Eq. (A27) in Appendix A]. N_2O emissions from the N deposited to non-agricultural land and water bodies, which originates from the reactive N releases due to agricultural activity as well as energy consumption, are estimated with Eq. (A28) in Appendix A. N_2O emissions from manure storage are estimated as the sum of the releases from the manure partitioned in individual management systems [Eqs. (A30)–(A37) in Appendix A]. The N_2O emitted from the burning of crop residues is estimated with Eq. (A38) in Appendix A. It should be noted that the current version of IAP-N does not include N_2O emissions created by the combustion of fossil fuels.

3.3 Nr enrichment

The amount of Nr enriched annually in an ecosystem or environment due to human activity in a certain region or on a global scale is indicated simply by the residual of the annual source and sink rates of Nr, or by the ratio of Nr source to sink (Zheng et al., 2002).

Human activity creates Nr via three pathways: (a) the Haber-Bosch process of synthesizing ammonia for the production of N fertilizers and other industrial purposes; (b) the cultivation of N-fixing crops; and (c) the burning of fossil fuels (Smil, 2001). Accordingly, the

Nr source rate on a regional scale due to anthropogenic creation (N_{Nrc}) is estimated as the sum of consumed synthetic N fertilizer, biological fixation N by cultivation of legume crops and paddy rice, and released N in the form of NO_x by fossil fuel consumption [Eq. (A39) in Appendix A]. The N_2O created from the consumption of fossil fuels is neglected in the Nr source rate estimation, since it is only equivalent to approximately 2.5%–4% of the simultaneously created NO_x -N.

The sink of Nr is mainly due to transformation of Nr to N_2 in biological denitrification and pryodenitrification in biomass burning (Sanhueza and Crutzen, 1998; and references therein). Biological denitrification occurs in cultivated soils, aquatic ecosystems (rivers, estuaries, shelves), and anaerobic lagoons and liquid systems for manure storage. Pryodenitrification occurs during the burning of crop residues and in animal manure. To estimate the gross Nr rate of sink of these processes (N_{Nrs}) with Eq. (A40) in Appendix A, the N_2 loss rates from the N in the cultivated soil layer (N_{En2s}); the N transported into water bodies by leaching and runoff (N_{En2lr}); the N in wet, or anaerobic, manure management systems (N_{En2m}); and the N in burned crop residues and manure (N_{En2b}) are separately calculated with Eqs. (A24), (A41), (A42) and (A43) (Appendix A), respectively.

In the estimation of N_2 loss from cultivated soils, separate loss factors are applied for dry croplands and paddy rice fields [Eq. (A24) in Appendix A]. Considering the N_2O emission factor of 1.25% (IPCC, 1997), the denitrification rate of $31\% \pm 17\%$, and the ratio of N_2O (12%–18%) to denitrification products (Ryden et al., 1979), the N_2 loss factor of dry croplands (f_{N2up}) is determined as 7.1%. Considering the denitrification rate of $36\% \pm 3\%$ for N incorporated into paddy rice fields (Zhu and Wen, 1992), and assuming the same N_2O ratio in denitrification products as stated above, a N_2 loss factor of 30.9% is applied for paddy rice fields (f_{N2pr}). As Galloway et al. (2003) report, about 50%, 23% and more than 22% of the N originating from agricultural lands due to leaching and runoff were lost in the form of N_2 due to denitrification in rivers, estuaries and shelves, respectively. Thus, only about 5% may finally arrive in open Ocean. Accordingly, a N_2 loss factor of 95% could be obtained by summing up these rates (f_{En2lr}) and applied for the anthropogenic Nr flowing out of cultivated soils by leaching and runoff [Eq. (A41) in Appendix A]. Only anaerobic conditions are favorable for biological denitrification. Thus, N_2 loss due to denitrification during manure storage is assumed to occur only in anaerobic lagoons and liquid systems; and the N_2 loss factor is assumed to be the same as that for submerged paddy soils [Eq. (A42) in Appendix A]. According to Sanhueza and Crutzen

(1998), about 46.7% of burned biomass N is released in the form of N_2 . This factor is applied to calculate the N_2 loss during the burning of either crop residues or animal manure [Eq. (A43) in Appendix A].

3.4 Model inputs

The parameter values given in Appendices C–F have been fixed in the model programmed with FORTRAN. This can be applied to estimate the annual N fluxes mentioned above for various spatial scales, from county to country or even global. In applying the current model, required input data include: (a) consumed synthetic N fertilizer (N_Sf); (b) crop yields of individual crops (P_Hp); (c) population of individual animal types and humans in rural areas (P_Ah); (d) harvest area of paddy rice (A_Ra) and other individual crops (A_Cr); (e) consumption amounts of coal (C_Ct), dry natural gas (C_Gt) and petroleum (C_Pt); (f) arable land area (A_Al); and (g) total land area (A_Land). These data are usually collected through surveys or directly provided by annual statistics.

4. An example of the model applied in China at a national scale

An application of the model described above is considered by estimating the surplus of consumed synthetic N fertilizers, and gaseous Nr emissions due to human activity, losses of Nr from agricultural soils by leaching and runoff, and the source-to-sink ratio of anthropogenic Nr. The example is conducted at the national scale in China for the period 1961–2004. Due to a lack of statistical data, however, the estimation does not cover the regions of Hong Kong, Macao and Taiwan.

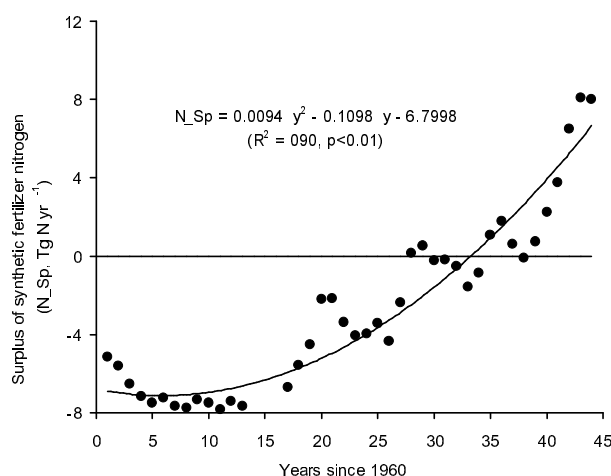


Fig. 2. Dynamics of surpluses of synthetic N fertilizer in China.

4.1 Data sources

The data used as the input variables are available at the national scale from statistical yearbooks in China. They are also available from the databases of the Food and Agriculture Organization of the United Nations (FAO) (<http://faostat.fao.org/faostat/>) and the Energy Information Administration, USA (EIA) (<http://www.eia.dep.gov>).

4.2 Nitrogen fluxes resulting from IAP-N calculations

4.2.1 Surplus of synthetic nitrogen fertilizer

The results of estimating the annual surplus of synthetic N fertilizer during the period 1961–2004 are illustrated in Fig. 2. In obtaining these estimates with the model, constant use efficiencies of mineral N and organic N were assumed for the entire period. As Fig. 2 shows, there was an under-application of synthetic N fertilizers for the crop yields before the late 1980s, with mean deficits of $-7.1 \text{ Tg N yr}^{-1}$ in the 1960s and 1970s, and $-2.5 \text{ Tg N yr}^{-1}$ in the 1980s. Following an increase in fertilizer application, over-application has occurred since the mid 1990s, with surpluses of 0.8 Tg N yr^{-1} on average in the late 1990s and 5.7 Tg N yr^{-1} in the early 2000s. These annual estimates are consistent with a former report in which the surplus or over-application of synthetic N fertilizer in China was first estimated using IAP-N, but with estimations conducted at 5-year intervals (Zheng et al., 2002). Figure 2 also shows that the surplus dynamics during the investigated period can be described with a non-linear function. The function is given as Eq. (1) listed in Table 1 (hereafter, all equations numbered (1)–(19) consecutively are listed in Table 1).

4.2.2 N_2O emissions

Figure 3a illustrates the dynamics of (a) overall estimated N_2O emissions in China associated with agricultural activity, as well as N deposition derived from fossil energy consumption; and (b) direct estimates of N_2O emissions from cultivated soils. Clearly, either the overall or direct N_2O emissions increased linearly during the investigated period. Overall emissions varied from 0.12 (1961) to 0.69 (2004) Tg N yr^{-1} , while direct emissions varied from 0.055 ± 0.050 (1961) to 0.399 ± 0.361 (2004) Tg N yr^{-1} . The linear changes for the years after 1960 can be described by Eq. (2) for overall emissions and by Eq. (3) for direct emissions from cultivated soils in Table 1. Annual increases of 0.0138 and $0.0086 \text{ Tg N yr}^{-1}$ on average are indicated for overall and direct N_2O emissions, respectively, as per the slopes of the regression curves described by Eqs. (2) and (3) in Table 1, respectively.

The effort required to meet the food and energy de-

Table 1. Empirical functions for describing the dynamics of N fluxes during 1961–2004.

No.	Equation detail	R^2	P^*	Related figures
Eq. (1)	$N_Sp=0.0094y^2-0.1098y-6.7998$	0.90	<0.01	Fig. 2
Eq. (2)	$N_N2o=0.0138y+0.0631$	0.98	<0.01	Fig. 3a
Eq. (3)	$N_En2os=0.0086y+0.0108$	0.98	<0.01	Fig. 3a
Eq. (4)	$N_N2o=0.3211 P^{2.6467}$	0.99	<0.01	Fig. 4
Eq. (5)	$N2O_{per\ capita}=0.0084y+0.1545$	0.97	<0.01	Fig. 5
Eq. (6)	$Fraction_{soil}=-0.0001y^2+0.0085y+0.3929$	0.86	<0.01	Fig. 5
Eq. (7)	$N_En2os=0.0159N_{sf}+0.0395$	0.995	<0.01	Fig. 3b
Eq. (8)	$N_N2o=0.0255N_{sf}+0.1111$	0.99	<0.01	Fig. 3b
Eq. (9)	$N_ (NH_3+NO_x)=0.1871y+0.9304$	0.99	<0.01	Fig. 6
Eq. (10)	$N_NH_3=0.0899y+0.7232$	0.99	<0.01	Fig. 6
Eq. (11)	$N_NO_x=0.0973y+0.2072$	0.97	<0.01	Fig. 6
Eq. (12)	$N_Lr=0.0693y+0.8086$	0.98	<0.01	Fig. 7
Eq. (13)	$So=-0.0004y^3+0.030y^2+0.059y+2.9426$	0.99	<0.01	Fig. 8a
Eq. (14)	$Share_{BNF}=0.0552y^2-3.7346y+72.075$	0.98	<0.01	Fig. 8b
Eq. (15)	$Share_{sf}=-0.0495y^2+3.4675y+16.527$	0.96	<0.01	Fig. 8b
Eq. (16)	$Si=0.156y+1.7448$	0.98	<0.01	Fig. 8a
Eq. (17)	$Ratio=-0.0008y^2+0.0841y+1.2243$	0.98	<0.01	Fig. 8a
Eq. (18)	$N_{st}=-0.0003y^3+0.0235y^2-0.009y+0.722$	0.98	<0.01	Fig. 9
Eq. (19)	$N_{sf}=-0.0005y^3+0.0342y^2-0.0927y+0.8078$	0.98	<0.01	Fig. 9

Note: Applies to the whole China; y is years since 1960, $0 < y < 45$, which applies for all equations in this table. *Significance level indicated by P (probability) values of the F -test. N_Sp , surplus of synthetic N fertilizer using; N_N2o , overall N_2O emissions; N_En2os , direct N_2O emissions from cultivated soils; P , overall population (10^9 capita); $Fraction_{soil}$, fraction of N_2O emissions from cultivated soil to overall N_2O emissions (no dimension); $N2O_{per\ capita}$, overall N_2O emissions on a population basis (kg N yr^{-1} by per person); $N_ (NH_3+NO_x)$, overall emissions of NH_3 and NO_x ; N_NH_3 , NH_3 emissions; N_NO_x , NO_x emissions; N_Lr , N loss from cultivated soils due to leaching and runoff; So , Nr source; Si , Nr sink; $Ratio$, ratio of Nr source to sink (no dimension); N_{sf} , synthetic N fertilizer; N_{st} , N enrichment, i.e., storage in agricultural products and the environment; $Share_{BNF}$, percentage of Nr created by BNF to the total anthropogenic Nr source; $Share_{sf}$, percentage of Nr created by energy production through fossil fuel combustion to the total anthropogenic Nr source. Variables without specified dimensions are measured in 10^{12} g N yr^{-1} .

mands of a dramatically increasing population, which had almost doubled in 2004 compared to 1961, is thought to account for the increases in N_2O emissions. This may be supported by the significant dependence of overall N_2O emissions upon population (Fig. 4), which can be described with a power function, as in Eq. (4) of Table 1. Overall emissions on a population basis also increased linearly during the investigated period [Eq. (5) in Table 1], from 0.183 (1961) to 0.520 (2004) kg N yr^{-1} by per person, with an annual increase of 0.0084 kg N yr^{-1} on average (Fig. 5). During the 44 years investigated, annual overall N_2O emissions were intensified by a factor of about six, while annual overall emissions per capita only increased by a factor of around three.

Of the overall N_2O emissions associated with agricultural activity and N deposition due to NO_x emissions from fossil energy consumption, the direct soil emissions from cultivated soils accounted for 45%–49% in the 1960s–1970s and ca. 56% since the 1980s (Fig. 5). The dynamics of the ratio of direct to overall N_2O emissions can be described with Eq. (6) in Table 1. The increase in direct N_2O emissions from cultivated

soils could be attributed to the increase in consumed synthetic N fertilizers, which in turn could explain 99.5% of the change in direct N_2O emissions during the 4.5 decades investigated [Fig. 3b; Eq. (7) in Table 1].

As one of the most important efforts needed to feed a large and increasing population, the increased consumption of synthetic N fertilizers not only stimulates direct N_2O emission, but also explains the 99% of increase in overall N_2O emissions during 1961–2004 [Fig. 3b; Eq. (8) in Table 1]. The direct N_2O emission from agricultural soils in the early 1990s is estimated as 0.294 Tg N yr^{-1} , which accounts for 9.2% of global soil emissions (Galloway et al., 2004).

4.2.3 Emissions of NH_3 and NO_x

The estimated emissions of either the sum or individual species of NH_3 and NO_x increased significantly and linearly during 1961–2004 (Fig. 6). The dynamics of these linear changes after 1960 can be described with Eqs. (9)–(11) in Table 1. The amount of released NH_3+NO_x was estimated at 1.4 Tg N yr^{-1} in 1961, of which NH_3 accounted for 73% and NO_x for 27%.

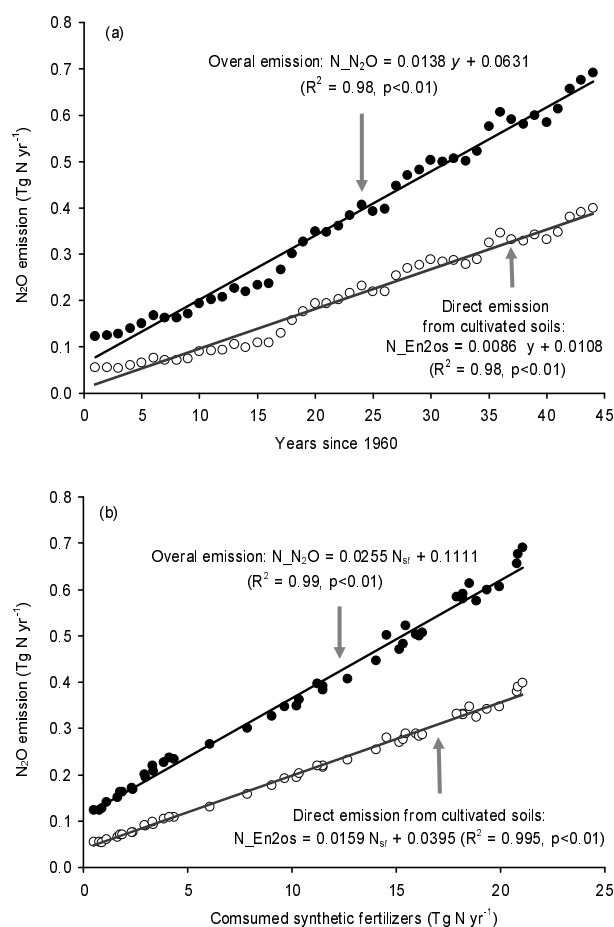


Fig. 3. Overall nitrogen oxide (N_2O) emissions (N_{N_2O}) from agricultural activity and energy consumption in China, plus direct N_2O emissions from cultivated soils (N_{En2os}) during 1961–2004. (a) dynamics during 1961–2004; (b) correlation between N_2O emissions and consumed synthetic N fertilizer.

With an annual increase of $0.187\ Tg\ N\ yr^{-1}$, total emissions rose to $9.6\ Tg\ N\ yr^{-1}$ in 2004, of which 49% was in the form of NH_3 ($4.7\ Tg\ N$) and 51% in the form of NO_x ($4.9\ Tg\ N$). The increase in the share of NO_x was mainly caused by the dramatic intensification of fossil fuel consumption, which was estimated to account for, on average, only ca. $0.47\ Tg\ N\ yr^{-1}$ in the 1960s, but $4.2\ Tg\ N\ yr^{-1}$ in 2004 (an enlargement by a factor of approximately 13). In the early 2000s, about 14% of the total anthropogenic Nr in China originated from fossil fuel combustion, which is clearly higher than the percentage (9%) in the early 1960s. The estimates of NH_3 and NO_x emissions for 1993 amount to 3.6 and $3.4\ Tg\ N\ yr^{-1}$, respectively, which account for 7.6% and 9.5% of the global estimates, respectively, for the same years (Galloway et al., 2004).

4.2.4 Nitrogen loss from cultivated soils by leaching and runoff

The N loss from cultivated soils due to leaching and runoff in 1961 was estimated to be ca. $1.1\ Tg\ N\ yr^{-1}$. This loss rate accounted for ca. 51% of BNF by all legume crops and rice paddies and ca. 38% of the total Nr source estimate. Since more and more Nr was introduced into, or created in, cultivated soils, the annual N loss due to leaching and runoff increased on average by $0.0693\ Tg\ N\ yr^{-1}$ during 1961–2004 (Fig. 7), and thus amounted to ca. $3.9\ Tg\ N\ yr^{-1}$ in 2004. This loss rate in 2004 accounted for 19% of consumed synthetic N fertilizer but only 14% of the total Nr source estimate of that year. The dynamics of N loss due to leaching and runoff is described by Eq. (12) in Table 1.

Because leaching and runoff are the major pathways for N loss from the cultivated soil horizon, the reduced share of N loss by these pathways, as compared to four decades ago, may suggest N accumulation in the cultivated soils. The N accumulation in cultivated soils may be partially proven by the long-term fertilization experiments conducted nationwide in China during the 1980s–1990s (Lin et al., 1994). These experiments showed that the total N content in cultivated soils of the dominant cropping systems in China increased on average by ca. 7.5% after continuous amendment of synthetic N fertilizer, as well as phosphorous (P) alone, or together with potassium (K) for 10 years (Lin et al., 1994). These experiments also showed that amendments of N, N+P, N+P+K and N+P+K+M (M indicating organic manure or straw) caused an annual change in N storage in the cultivated soils by -0.4% , 0.2% , 1.2% and 1.6% per year, respectively (Lin et al., 1994). A further implication of this report by Lin et al. (1994) is that improving supplies of synthetic K fertilizers may intensively stimulate N accumulation in cultivated soils. According to the statistical data of China provided by the FAO database (<http://faostat.fao.org/faostat/>), the ratios of consumed synthetic P and K fertilizer to N on average during 1990–2002 were 0.37 and 0.13, respectively, which were clearly higher than the mean ratios during 1961–1989 (being 0.28 and 0.04, respectively). The P:N ratio increased by a factor of 1.3, while the K:N ratio increased by a factor of 3.3. This implies that the estimated reduction in the ratio of N loss to applied synthetic N or total Nr source due to leaching and runoff might be partially attributed to the increased K:N ratios, which might have led to N accumulation in cultivated soils. As Huang and Sun (2006) report, the organic carbon storage in the cultivated soils of China increased by 359 to $463\ Tg\ C\ yr^{-1}$ over the last two decades. Because soil total N content is approximately

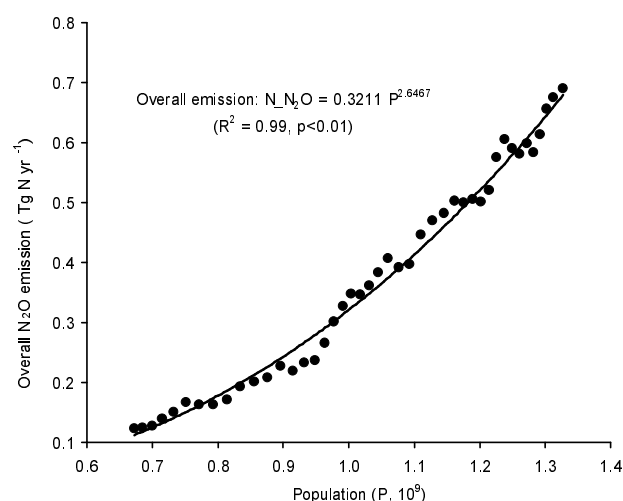


Fig. 4. Relationship between overall N_2O emissions in China and population during 1961–2004.

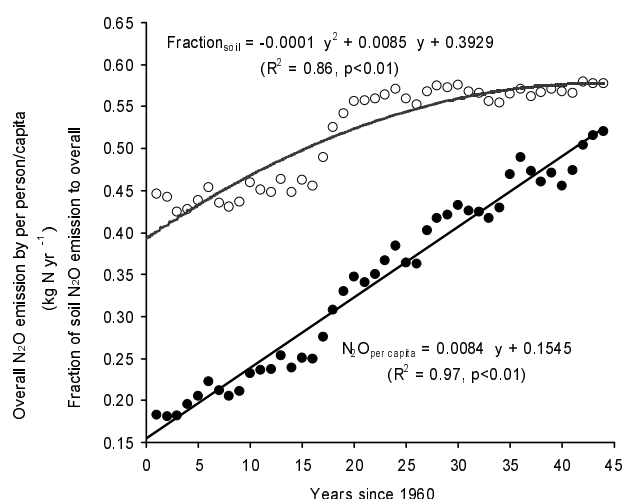


Fig. 5. Overall N_2O emissions per person from agricultural activity and deposited atmospheric N due to fossil energy consumption, plus the fraction of cultivated soils' contribution to the overall N_2O emissions in China during 1961 and 2004.

in proportion to organic carbon content, this report by Huang and Sun (2006) may suggest indirectly a nitrogen accumulation in cultivated soils. However, further direct experimental evidence is still required to confirm the fact of N accumulation in the cultivated soil layer.

4.2.5 *Nr* source and sink

The Nr source, i.e., the process of transforming N_2 into Nr , integrates consumed synthetic N fertilizer, BNF and released N from fossil fuel combustion. As Fig. 8a shows, the estimated annual Nr source of

China increased by a factor of approximately 10 during the investigated period, from ca. 2.8 Tg N yr^{-1} in 1961 to ca. $28.1 \text{ Tg N yr}^{-1}$ in 2004. Its dynamics can be described with Eq. (13) in Table 1. Of the total Nr source, consumption of synthetic fertilizers, energy production with fossil fuels, and BNF accounted for 18%, 9% and 73%, respectively, in 1961, but 75%, 15% and 10%, respectively, in 2004 (Fig. 8b). The dynamics for the share of synthetic N consumption and BNF to the overall anthropogenic Nr source can be described with Eqs. (14) and (15) in Table 1, respectively. Clearly, the overwhelming contributor to the overall Nr source was BNF in the early 1960s, but it was taken over in the latter decade by synthetic N fertilizers. In the 1970s, synthetic N fertilizers, fossil fuel combustion and BNF accounted for 54%, 18% and

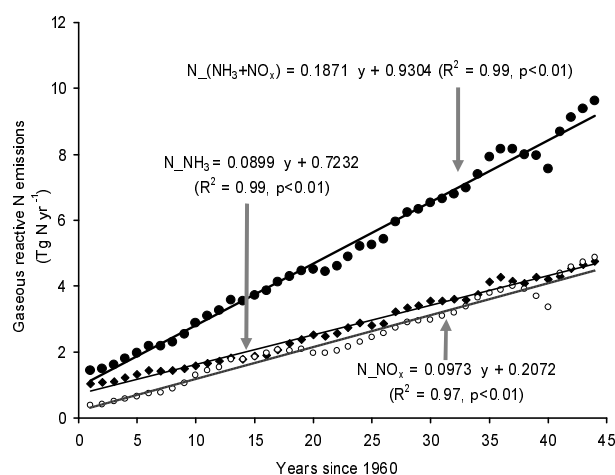


Fig. 6. Emissions of reactive gaseous N from agricultural activity and energy consumption in China during 1961 and 2004.

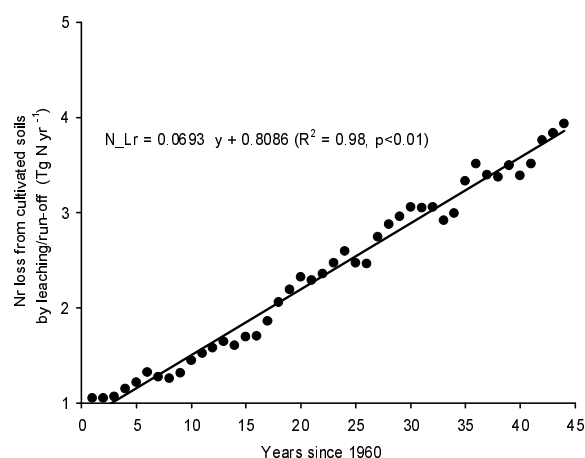


Fig. 7. Nr loss from cultivated soils by leaching and runoff during 1961–2004.

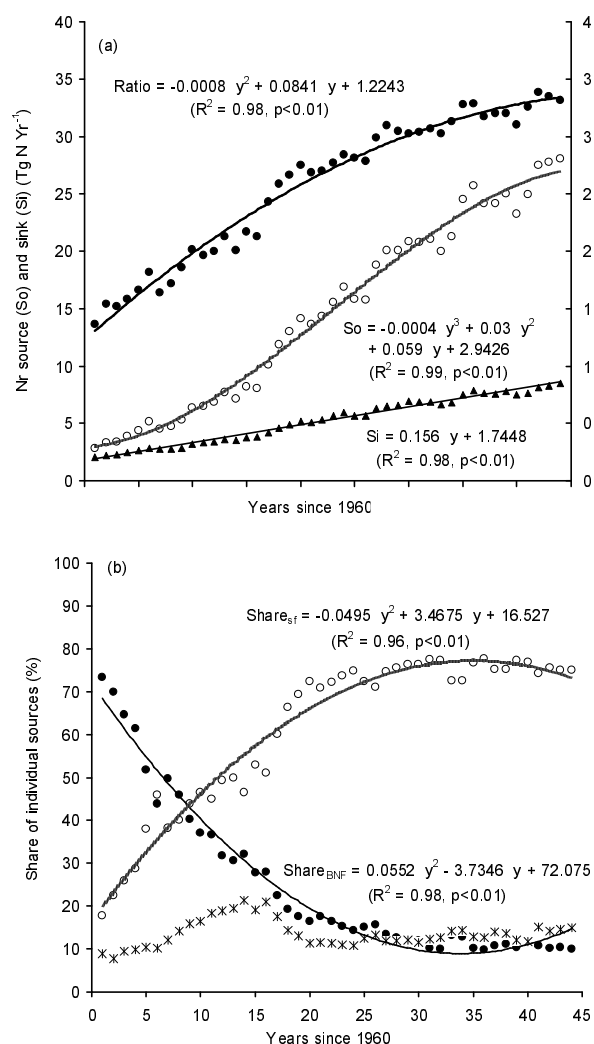


Fig. 8. Nr source and sink (N_2 emissions) during 1961–2004. (a) source and sink fluxes and source-to-sink ratios; (b) share (percentage) of individual source to the overall anthropogenic Nr source.

28%, respectively. Since the late 1980s, the contribution of synthetic fertilizers has been stable at a high level of approximately 75%; meanwhile the contribution of BNF gradually and slowly decreased to a level lower than fossil fuel combustion (Fig. 8b). The great reduction in the share of BNF was mainly due to the dramatic decrease in the cultivation of legume crops (Zheng et al., 2002).

As Fig. 8a illustrates, the estimated annual sink (i.e., the process of transforming Nr into N_2) of anthropogenic Nr created in China increased linearly from 2.1 Tg N yr^{-1} in 1961 to 8.4 Tg N yr^{-1} in 2004.

It was equivalent to about 69% of the anthropogenic Nr source in the early 1960s, but only about 30% in the early 2000s. Its dynamics can be described

with Eq. (16) in Table 1. As Fig. 8a shows, the source-to-sink ratio was estimated to be 1.4 in 1961 and 3.3 in 2004 [Eq. (17) in Table 1]. The source-to-sink ratios of anthropogenic Nr may be an indicator of Nr enrichment within ecosystems as well as environmental components. The annual Nr enrichment rates during 1961–2004 were estimated as the residuals of annual sources and sinks and the results are illustrated in Fig. 9. The dynamics of annual Nr enrichment rate are described with Eq. (18) in Table 1. As Fig. 9 shows, the estimated annual Nr enrichment rate was equivalent to 120%, 102% and 90% of consumed synthetic N fertilizer in the 1960s, 1970s and 1980s–2000s, respectively. The dynamics of synthetic N fertilizer consumption is also shown in Fig. 9 and Eq. (19) in Table 1. During the investigated period, the dynamics and magnitude were similar between the annual consumption rates of synthetic N fertilizer and the annual enrichment rate of anthropogenic Nr. This suggested that consumption of N fertilizers was the principal driving force for Nr enrichment. A certain proportion of the anthropogenic Nr was stored on purpose in the ecosystems at timescales from a few years to a shorter term. The storage Nr in the forms of food, feed, fabric and other materials was the direct goal of Nr creation by synthetic N fertilizer production, as well as BNF through the cultivation of legume crops. Humans have benefited from such an enrichment of anthropogenic Nr and, consequently, have been experiencing an enhanced standard of living and an enlarging population. Simultaneously, however, the remaining anthropogenic Nr is stored unexpectedly in enviro-

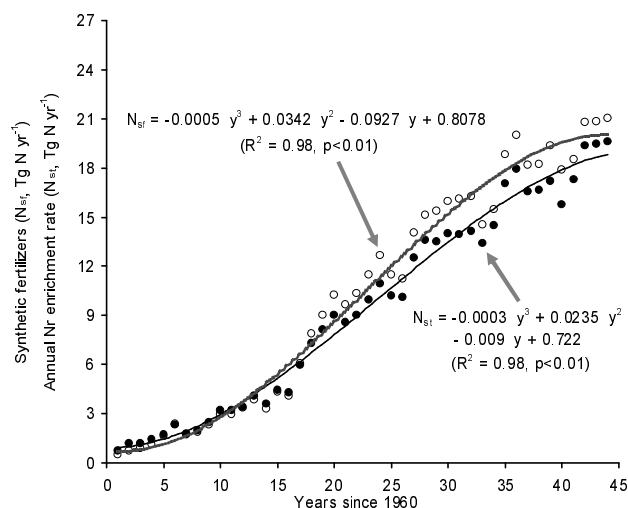


Fig. 9. Dynamical consumption of synthetic N fertilizer and Nr enrichment (i.e., storage in agricultural products and the environment).

omental components. Some evidence has already proven the facts of Nr enrichment in environmental components. For instance, N_2O accumulates in the global atmosphere at a rate of ca. 3.8 Tg N yr^{-1} ; nitrate accumulation was observed in the profile beneath the cultivated horizon of the irrigated croplands in the Northern China Plain (Ju et al., 2004). The Nr enriched in the environment usually causes negative cascade effects, which in turn may threaten human health, biodiversity, ecosystem sustainability and climate stability (Galloway et al., 2003).

5. Discussion

As stated above, IAP-N is a regional model. It is designed to simulate N cycles in association mainly with agriculture at annual, or decadal timescales and at regional, national, or even global spatial scales. Since it is usually hard to observe directly the Nr fluxes concerned at these time or spatial scales, the simulation results of this model are still hard to validate directly. However, in applying the model to simulating nitrogen fluxes associated with agricultural activity and fossil energy consumption in the Asian region at 5-year intervals, results comparable with other studies (Zheng et al., 2002) were yielded. Nevertheless, inter-comparison of different models may provide better opportunities to assess the uncertainties of the estimates by this model. Using the revised 1996 IPCC guidelines (IPCC, 1997) with the same database in this paper to estimate direct emissions of N_2O from cropland soils, we obtain 0.041 Tg N in 1961 and 0.388 Tg N in 2004, which are similar to the results in this paper. Using the latest IPCC method (IPCC, 2006) to estimate direct emissions of N_2O , we get slightly lower results (0.032 Tg N in 1961 and 0.223 Tg N in 2004) than those through using the older IPCC method and results in this paper. This is because N_2O emission factors in IPCC (2006) are different for flooded rice field (0.003) and glebe fields (0.01), and they are much lower than default N_2O emission factors of the revised 1996 IPCC guidelines (IPCC, 1997). Therefore, one focus of IAP-N work in the near future is to include comparisons with other models.

In addition, the current version of IAP-N still has the following limitations, which obstruct substantially its wider application. N_2O emissions from fossil fuel combustion are not yet included. And tea gardens, orchards and crops other than the 13 types already considered may consume a considerable amount of N fertilizer, but are ignored in the current model version—a shortcoming that may lead to considerable uncertainty. So it causes methodologies for uncertainty assessment and optimization of parameters have not yet

been established. Another, most of the model parameters is determined by mainly referring to China-specific data. To apply the model to other countries or continents, localization of the parameters is necessary. Otherwise, only the N fluxes associated with agricultural activity and NO_x emissions from fossil fuel consumption are involved in this IAP-N version. This is, however, not enough to understand the entire nature of nitrogen cycling because features of other ecosystems, such as forests, grasslands etc. are not yet involved in the modeling process. Furthermore, databases to drive IAP-N for simulation at various timescales (annual or decadal) and spatial scales (county, provincial, national, continental, or even global) have not yet been well established. To widely apply this model, such databases containing full data on consumed synthetic N fertilizer; yields of individual crops; populations of individual animal types and humans living in rural areas; harvest areas of paddy rice and other individual crops; consumption amounts of coal, dry natural gas and petroleum; arable land area; and total land area of the region concerned are exclusively required. The immediate goal of further developing IAP-N is to overcome these limitations.

As the example above demonstrated, the ratio of anthropogenic Nr to sink is usually greater than one. This is because Nr is purposefully enriched in order to provide enough protein for the increasing human population. However, unexpected Nr enrichment also occurs simultaneously and induces negative cascade impacts on human beings, as well as wildlife, and in the long-run thereby threatens the living environment for humans. Therefore, how to manage effectively anthropogenic Nr for the purposes of reducing unexpected enrichment while increasing the expected enrichment is still a challenge facing humans. In order to manage anthropogenic Nr in terms of environmental safety, it is of great importance to identify the threshold of the Nr source-to-sink ratio. The threshold ratio is regarded as an indicator of whether the unexpected Nr enrichment is still safe. Understandably, further developments of IAP-N may attempt to build up its capacity for identifying the safe threshold of the Nr source-to-sink ratio, though this is a hard task due to many difficulties in data and methodology availability.

6. Conclusions

The IAP-N model, used to simulate N cycling at annual and regional scales, was designed to be driven by statistical or survey data (including consumed synthetic N fertilizer; yields and harvest areas of individual crops; populations of livestock and rural people; consumption amounts of coal, dry natural gas and

petroleum; arable land area; and total land area) of the regions concerned and to estimate annual N fluxes flowing among cultivated soils, crops, livestock, humans, aquatic systems and the atmosphere.

An application of IAP-N in China at the national scale during the period 1961–2004 was described, which simulated the dynamics of N surpluses, gaseous N emissions, N loss from agricultural land due to leaching and runoff, and anthropogenic Nr source and sink ratios. These estimates may provide a sound scientific basis for N management. However, it is still hard to validate directly these model estimates due to no directly measured data of the N fluxes concerned at regional scales. Thus, inter-comparison of IAP-N with other models is still required to access the model outputs. In addition, the current version of IAP-N still

has some limitations in terms of uncertainty assessment and its wider application to other countries or at continental/global scales. This suggests that further development of this model is required to focus on overcoming the identified limitations. Moreover, further development of IAP-N is also required to build its capacity for determining the safe threshold of Nr source-to-sink ratio, which in turn is essential for N management.

Acknowledgements. This study was jointly financed by the National Natural Science Foundation of China (NSFC) (Grant Nos. 40605030, 40331014, and 40425010) and the Chinese Academy of Sciences (KZCX2-YW-204).

APPENDIX A

Formulae involved in the calculations*

$$N_Sp = N_Sf - N_Cr \quad (A1)$$

$$N_Cr_k = (N_Up_k - N_Bnfr_k \times f_Mn - N_Lc_k - N_Sg_{k-1} \times f_Mn - N_Om_k \times f_Om - N_Dm_k \times f_Mn) / f_Mn \quad (A2)$$

$$N_Up = \sum_{j=1}^{13} (N_Rs_j + N_Rr_j + N_Hp_j) \quad (A3)$$

$$N_Lc = \sum_{j=7}^8 (N_Rs_j + N_Rr_j + N_Hp_j) \quad (A4)$$

$$N_Hp_j = P_Hp_j \times f_Dr_j \times f_Np_j \quad (A5)$$

$$N_Rs_j = P_Hp_j \times f_Dr_j \times (1 / I_Hi_j - 1) \times f_Nr_j \quad (A6)$$

$$N_Rr_j = P_Hp_j \times f_Dr_j \times (R_Rs_j / I_Hi_j) \times f_Nr_j \quad (A7)$$

$$N_Bnfr = A_Ra \times f_bnfr \quad (A8)$$

$$N_Om = \sum_{j=1}^{13} (N_Rr_j + N_Rs_j \times f_Ri_j) + N_Mah \quad (A9)$$

$$N_Mah = (1 - f_Enn) \times \sum_{i=1}^7 [P_Ah_i \times f_Wah_i \times (f_Wahan_i + f_Wahlq_i + f_Wahsd_i)] \quad (A10)$$

$$N_Dm = f_Pna \times \sum_{j=1}^{13} (N_Rs_j \times f_Rb_j) + N_Ern \times r_Dl \times (A_Al / A_Land) \quad (A11)$$

$$N_Ern = N_Ers + N_Erb + N_Erm + N_Ere \quad (A12)$$

$$N_Ers_k = [(N_Dm_{k-1} + N_Om_k + N_Sf_k) \times (1 - f_Rl_k) + N_Bnfl_k] \times (f_Nh3sfu + f_Noxsfu) + [(N_Dm_{k-1} + N_Om_k + N_Sf_k) \times f_Rl_k + N_Bnfr_k] \times f_Nh3sfp \quad (\text{when } k = 1, N_Dm_{k-1} \approx N_Dm_k) \quad (A13)$$

$$N_Erb = (f_Nh3 + f_Nox + f_Rcn) \times \sum_{j=1}^{13} (N_Rs_j \times f_Rb_j) \quad (A14)$$

$$N_Erm = f_Enn \times N_Wah \quad [\text{in which } N_Wah = \sum_{i=1}^7 (P_Ah_i \times f_Wah_i)] \quad (A15)$$

$$N_Ere = N_Erep + C_Ct \times f_Ct + C_Gt \times f_Gt \quad (A16)$$

$$N_Erep = C_Pt \times (f_Ppi \times f_Pi + f_Pap \times f_Pa + f_Ptf \times f_Pt + f_Prf \times f_Pr + f_Pof \times f_Po) \quad (A17)$$

$$N_Sg = N_In - N_Ot \quad (A18)$$

$$N_In_k = N_Sf_k + N_Om_k + N_Bnfr_k + N_Bnfl_k + N_Dm_k + N_Sg_{k-1} \times f_Mn \quad (\text{when } k = 1, N_Sg_{k-1} \approx 0) \quad (A19)$$

$$N_Bnfl = (1 + f_N2os + f_Nns / 2 + f_Lc) \times N_Lc \quad (A20)$$

$$N_Ot = N_Lr + N_Ers + N_En2os + N_En2s + N_Up \quad (A21)$$

$$N_Lr = [N_In \times (1 - A_Ra / \sum_{j=1}^{13} A_Cr_j) - N_Bnfl] \times f_Uf + A_Ra \times f_Rf + N_Bnfl \times f_Lc \quad (A22)$$

$$N_En2os = N_In \times f_n2os \quad (A23)$$

$$N_En2s_k = [(N_Dm_{k-1} + N_Om_k + N_Sf_k) \times (1 - f_Rl_k) + N_Bnfl_k] \times f_N2up + [(N_Dm_{k-1} + N_Om_k + N_Sf_k) \times f_Rl_k + N_Bnfr_k] \times f_N2pr \quad (A24)$$

$$N_Gnr = N_Ern + N_N2o \quad (A25)$$

$$N_N2o = N_En2os + N_En2olr + N_N2om + N_En2od + N_N2ob \quad (A26)$$

$$N_En2olr = N_Lr \times f_n2olr \quad (A27)$$

$$N_En2od = N_Ern \times r_Dl \times (1 - A_Al / A_Land) \times f_n2os + N_Ern(1 - r_Dl) \times f_n2olr \quad (A28)$$

$$N_En2odc = [f_Pna \times \sum_{j=1}^{13} (N_Rs_j \times f_Rb_j) + N_Ern \times r_Dl \times (A_Al / A_Land)] \times f_n2os \quad (A29)$$

$$N_N2om = N_En2oan + N_En2olq + N_En2osd + N_En2opp + N_En2ods + N_En2of + N_En2oos \quad (A30)$$

$$N_En2oan = f_N2oan \times \sum_{i=1}^7 (P_Ah_i \times f_Wah_i \times f_Wahan_i) \quad (A31)$$

$$N_En2olq = f_N2olq \times \sum_{i=1}^7 (P_Ah_i \times f_Wah_i \times f_Wahlq_i) \quad (A32)$$

$$N_En2osd = f_N2osd \times \sum_{i=1}^7 (P_Ah_i \times f_Wah_i \times f_Wahsd_i) \quad (A33)$$

$$N_En2opp = f_N2opp \times \sum_{i=1}^7 (P_Ah_i \times f_Wah_i \times f_Wahpp_i) \quad (A34)$$

$$N_En2ods = f_N2ods \times \sum_{i=1}^7 (P_Ah_i \times f_Wah_i \times f_Wahds_i) \quad (A35)$$

$$N_En2of = f_N2of \times \sum_{i=1}^7 (P_Ah_i \times f_Wah_i \times f_Wahf_i) \quad (A36)$$

$$N_En2oos = f_N2oos \times \sum_{i=1}^7 (P_Ah_i \times f_Wah_i \times f_Wahlq_i) \quad (A37)$$

$$N_N2ob = f_N2ob \times [\sum_{j=1}^{13} (N_Rs_j \times f_Rb_j)] \quad (A38)$$

$$N_Nrc = N_Sf + N_Bnfr + N_Bnfl + N_Ere \quad (A39)$$

$$N_Nrs = N_En2s + N_En2lr + N_En2m + N_En2b \quad (A40)$$

$$N_En2lr = N_Lr \times f_En2lr \quad (A41)$$

$$N_En2m = f_N2pr \times [\sum_{i=1}^7 (P_Ah_i \times f_Wah_i \times (f_Wahan_i + f_Wahlq_i))] \quad (A42)$$

$$N_En2b = f_N2 \times \sum_{j=1}^{13} (N_Rs_j \times f_Rb_j) \quad (A43)$$

Note: For definitions of the variables and parameters involved refer to Appendix B, and for the values of the parameters refer to Appendix C.

APPENDIX B

Definitions of the variables for the equations in Appendix A

Variable	Definition, value, and/or data source
i	Animal type ID (refer to Appendix IV for the ID code of each type)
j	Crop type ID (refer to Appendix V for the ID code of each type)
k	Number of year (defined by users)
A_Al	Area of arable land of a certain region, available as statistical data

A_Land	Land area of a certain region, available as statistical data
A_Ra	Paddy rice harvest area of a certain region, available as statistical data
A_Cr	Harvest area of individual crops, available as statistical data
C_Ct	Consumption amount of coal, available as statistical data
C_Gt	Consumption amount of dry natural gas, available as statistical data
C_Pt	Consumption amount of petroleum, available as statistical data
N_Bnfl	Biological N fixation by all legume crops
N_Bnfr	Biological N fixation by rice cultivation
N_Cr	Required commercial N to sustain annual crop production
N_Dm	Deposited N from the atmosphere, which is induced by reactive N emissions from agricultural systems and fossil fuel energy consumption
N_En2lr	N ₂ released from Nr removed out of the cultivated soil layer by leaching and runoff
N_N2o	Total N ₂ O emissions due to agricultural activity
N_En2m	N ₂ released from manure management systems
N_En2oan	Annual total N ₂ O-N emissions from anaerobic manure management systems
N_N2ob	Annual total N ₂ O-N emissions from crop residue burning
N_En2od	N ₂ O emissions from deposited N out of croplands, which originated from N volatilization from agriculture and energy consumption
N_En2odc	N ₂ O emissions from the N deposited to croplands
N_En2ods	Annual total N ₂ O-N emissions from daily spread of manure
N_En2of	N ₂ O emissions from manure during burning of fuel
N_En2olq	Annual total N ₂ O-N emissions from liquid manure management systems
N_En2olr	N ₂ O emissions from the N removed to deep sediments and water bodies by leaching and runoff
N_En2oos	Annual total N ₂ O-N emissions from other systems
N_En2opp	Annual total N ₂ O-N emissions from pasture range and paddock systems
N_En2os	Annual total N ₂ O-N emissions from cropland soils
N_En2osd	Annual total N ₂ O-N emissions from solid storage and drylot systems
N_En2s	N ₂ -N emission from cultivated soils by denitrification
N_Erb	Reactive N emissions from crop residue burning
N_En2b	N ₂ released to the atmosphere from residue burning
N_Ere	Emitted reactive N from fossil energy consumption
N_Erep	NO _x -N emissions from petroleum consumption
N_Erm	Emitted reactive N from human and animal manure
N_Ern	Annual total reactive N released from agricultural systems to the atmosphere
N_Ers	Emitted reactive N from agricultural soils
N_Gnr	Gaseous Nr released from agricultural activity to the atmosphere
N_N2ob	N ₂ O emissions from crop residue burning
N_N2om	Annual total N ₂ O-N emissions from manure management systems
N_Nrs	Nr sink rate
N_Nrc	Nr source rate
N_Om	Applied organic manure and returned crop residues and roots
N_Sf	Annually consumed synthetic N fertilizer, available as statistical data
N_Sg	Soil N gain
N_Sp	Annual surplus of commercial synthetic N fertilizer
N_In	Annual total N introduced into cropland soils
N_Ot	Annual total N out of cropland soils
N_Up	Annual total crop uptake of N of a country
N_Wah	Annual total amount of excreted N by animal and rural human population
N_Mah	Annually applied animal and human manure N
N_Hp	N in harvested crop products
N_Rr	N in crop roots
N_Rs	N in crop straw and stubble

N_Lr	Leaching and runoff Nr from croplands
N_Lc	Annual legume crop uptake of N of a country
P_Ah	Annual animal or rural human population, available as statistical data
P_Hp	Harvest production, available as statistical data

APPENDIX C

Definitions, values and references for the parameters involved in Appendix A

Parameter	Definition	Value	Reference(s)
f_bnfr	Rate of biological N fixation in rice paddy fields ($\text{kg N hm}^{-2} \text{ yr}^{-1}$)	32 (14–50)	Kundu and Ladha (1995)
f_Ct	NO_x -N emission rate from coal combustion ($10^{-12} \text{ kg N J}^{-1}$)	73.68	IPCC (1997)
f_Enn	Fraction of NH_3 & NO_x emissions from animal or human excreta (no dimension)	0.20	IPCC (1997)
f_Gt	NO_x -N emission rate from dry natural gas combustion ($10^{-12} \text{ kg N J}^{-1}$)	78.95	IPCC (1997)
f_Lc	Leaching factor of N from upland fields growing legume crops (no dimension)	0.024	Xing and Zhu (2000)
f_n2olr	Emission factor of N lost by leaching and runoff (no dimension)	0.025	IPCC (1997)
f_En2lr	N_2 emission factor of N flowing through rivers, estuaries and shelves (no dimension)	0.95	Galloway et al. (2003)
f_N2pr	N_2 emission factor of N available in submerged paddy rice soils (no dimension)	0.309	Zhu and Wen (1992);
f_N2up	N_2 emission factor of N available in dry cultivated soils (no dimension)	0.071	Ryden et al. (1979);
f_N2	N_2 emission factor from biomass burning (no dimension)	0.467	Appendix F
f_Om	Utilization efficiency of N in organic manure (0–1 factor, no dimension)	0.27 ± 0.11	Zhu and Wen (1992)
f_Mn	Utilization efficiency of mineral N (0–1 factor, no dimension)	0.36 ± 0.15	Zhu and Wen (1992)
f_N2oan	N_2O -emission factor of manure N of anaerobic systems (no dimension)	0.001	IPCC (1997)
f_N2ods	N_2O emission factor of daily spread	0.000	IPCC (1997)
f_N2ob	Partitioning factor of N_2O -N in burned residue N (no dimension)	0.007	Appendix F
f_N2olq	N_2O -N emission factor of manure N stored in anaerobic systems (no dimension)	0.001	IPCC (1997)
f_N2oos	N_2O -N emission factor of manure N in other management systems (no dimension)	0.005	IPCC (1997)
f_N2opp	N_2O -N emission factor of manure N in pasture range and paddock systems and daily spread (no dimension)	0.02	IPCC (1997)
f_N2of	N_2O -N emission factor of manure N burned as fuel (no dimension)	0.007	Appendix F

f_n2os	National mean direct emission factor of N ₂ O-N of cultivated soils (no dimension)	0.0135±0.0122	Zheng et al. (2004)
f_N2osd	N ₂ O-N emission factor of manure N stored in solid storage and drylot systems (no dimension)	0.02	IPCC (1997)
f_N2pr	Emission factor of N ₂ -N from rice paddy fields by denitrification (no dimension)	0.3088	Zhu and Wen (1992)
f_N2up	Emission factor of N ₂ -N in upland fields by denitrification (no dimension)	0.0708	Ryden et al. (1979); Zhu and Wen (1992)
f_Nh3fp	Emission factor of NH ₃ -N from N fertilizer applied in rice paddies (no dimension)	0.18	Xing and Zhu (2000)
f_Nh3sfu	Emission factor of NH ₃ -N from N fertilizer applied in upland fields (no dimension)	0.09	Xing and Zhu (2000)
f_Nh3	NH ₃ emission factor of residue burning	0.034	Appendix F
f_No _x	NO _x emission factor of residue burning	0.121	Appendix F
f_Rcn	RCN emission factor of residue burning	0.033	Appendix F
f_Nns	Emission factor of NH ₃ and NO _x from fertilizer applied to agricultural soils (no dimension)	0.10	IPCC (1997)
f_No _x sfu	Emission factor of NO _x from synthetic N fertilizer applied to upland fields (no dimension)	0.020	Zheng et al. (2003)
f_Pa	NO _x -N emission factor from petroleum consumption in aviation (10 ⁻¹² kg N J ⁻¹)	110.53	IPCC (1997)
f_Pap	Partitioning fraction of petroleum consumed in aviation (no dimension)	0.038	China data in 1998 (EIA)
f_Pi	NO _x -N emission factor from petroleum consumption in industries (10 ⁻¹² kg N J ⁻¹)	73.68	IPCC (1997)
f_Pna	Portioning fraction of PNA from burned residue N (no dimension)	0.338	Appendix F
f_Po	NO _x -N emission factor of other petroleum consumption (10 ⁻¹² kg N J ⁻¹)	211.11	IPCC (1997)
f_Pof	Partitioning fraction of other petroleum consumption (no dimension)	0.337	China data in 1998 (EIA)
f_Ppi	Partitioning fraction of petroleum consumption in industries	0.248	China data in 1998 (EIA)
f_Pr	NO _x -N emission factor from petroleum residential consumption (10 ⁻¹² kg N J ⁻¹)	36.84	IPCC (1997)
f_Pr _f	Partitioning fraction of residual consumption (no dimension)	0.195	China data in 1998 (EIA)
f_Pt	NO _x -N emission factor from petroleum consumed in road transportation (10 ⁻¹² kg N J ⁻¹)	221.05	IPCC (1997)
f_Ptf	Partitioning fraction of petroleum consumption in road transportation (no dimension)	0.183	China data in 1998 (EIA)
f_Rb	Fraction of burned residue N (no dimension)	0.7	Cao and Zhuang (1996); MOA/DOE PET (1997)
f_Rf	Leaching and runoff factors of N from rice paddy fields (kg N hm ⁻² yr ⁻¹)	26.9	Xing and Zhu (2000); Ma et al. (1997); Lu et al. (1991)
f_Ri	Fraction of residue (including stubble) incorporation or return (no dimension)	0.14±0.03	Survey by the authors
f_Rl	Ratio rice harvest area to total crop harvest area		Quantified with FAO database

f_Uf	Leaching and runoff factors of N from upland fields growing non-legume crops (no dimension)	0.142	Xing and Zhu (2000)
f_Wah	Excretion rate of manure N by per capita (kg N yr ⁻¹)	Appendix D	
f_Wahan	Partitioning factor of excreta to anaerobic manure management systems (no dimension)	Appendix G	
f_Wahlq	Partitioning factor of excreta to liquid manure management systems (no dimension)	Appendix G	
f_Wahos	Partitioning factor of excreta to other systems (no dimension)	Appendix G	
f_Wahds	Partitioning factor of excreta for daily spread (no dimension)	Appendix G	
f_Wahpp	Partitioning factor of excreta to pasture range and paddock systems (no dimension)	Appendix G	
f_Wahsd	Partitioning factor of excreta to solid storage and drylot systems (no dimension)	Appendix G	
f_Wahf	Partitioning factor of excreta for use as fuel (no dimension)	Appendix G	
f_Dr	Fraction of dry matter (no dimension)	Appendix E	
f_Np	N fraction in harvest production (no dimension)	Appendix E	
f_Nr	N fraction in root biomass (no dimension)	Appendix E	
I_Hi	Crop harvest index (no dimension)	Appendix E	
R_Rs	Shoot-to-root ratio of individual crops (no dimension)	Appendix E	
r_Dl	Fraction of reactive N released from Asia and re-deposited to Asian land (no dimension)	0.819	Galloway et al. (2004)

APPENDIX D

Nitrogen excretion rates from livestock and rural human populations

ID code (<i>i</i>)	Population type (animal or human)	Excretion rate of per capita (kg N yr ⁻¹)
1	Swine	3.22 ^b
2	Sheep	0.7 ^b
3	Non-dairy cattle and buffaloes	11.35 ^b
4	Dairy cattle	60.0 ^a
5	Poultry	0.6 ^a
6	Horse, ass, mule, camel, goat	9.7 ^b
6	Horse, ass, mule, camel, goat	9.7 ^b
7	Rural people	0.69 ^b

^aDefault value recommended by IPCC (1997) and Moiser et al. (1998). ^bAsia-specific N excretion rates provided by Zhu et al. (1997) and Xing and Zhu (2000).

APPENDIX E

Parameters of crops related to N uptake estimation

Code (<i>j</i>)	Crop	I_Hi _j	R_Rs _j	f_Dr _j	f_Np _j	f_Nr _j
1	Maize	0.441 ^a	0.14–0.2 ^g	0.4 ^e	0.017 ^l	0.0058 ⁿ

2	Wheat	0.368 ^a	0.1–0.3 ^f	0.83 ^e	0.014 ^f	0.00516 ⁿ
3	Rice	0.43 ^a	0.1–0.15 ^g	0.83 ^e	0.01 ^f	0.00753 ⁿ
4	Sorghum	0.386 ^a	0.12–0.25 ^g	0.4 ^k	0.017 ^l	0.0073 ⁿ
5	Barley	0.39 ^a	0.19–0.25 ^h	0.83 ^e	0.014 ^f	0.00516 ⁿ
6	Rapeseed	0.256 ^a	0.1–0.2 ^g	0.83 ^e	0.00548 ^m	0.00548 ^e
7	Soybean	0.436 ^a	0.1–0.3 ^g	0.83 ^e	0.06 ⁿ	0.02284 ^e
8	Pulses excluding soybean	0.436 ^b	0.1–0.3 ^g	0.83 ^e	0.05 ⁿ	0.02284 ^e
9	Seed cotton	0.383 ^a	0.1–0.3 ^g	0.83 ^e	0.00548 ^m	0.00548 ^c
10	Root and tubers	0.714 ^c	0.05 ⁱ	0.45 ^e	0.00507 ^c	0.00507 ^c
11	Sugar cane	0.83 ^d	0.26 ^j	0.4 ^e	0.0058 ^l	0.0058 ^o
12	Sugar beet	0.83 ^e	0.05 ⁱ	0.4 ^e	0.00507 ^c	0.00507 ^c
13	Vegetables and melons	0.83 ^d	0.2–0.3 ^g	0.15 ^e	0.008 ⁿ	0.008 ⁿ

Notes: j , number of crop; LHi_j , harvest index; $R.Rs_j$, root-to-shoot ratio; $f.Dr_j$, dry matter fraction; $f.Np_j$, N content in harvest products; $f.Nr_j$, N content in residue. ^aRefer to Zhang and Zhu (1990); ^btake the value of soybean; ^ctake IPCC (1997) value for potatoes; ^dtake IPCC (1997) value for sugar beet; ^eIPCC (1997) value; ^frefer to Meng et al. (2001); ^grefer to JNHA (1991); ^hrefer to Tang et al. (2000); ⁱset by experience of the authors; ^jrefer to Chen et al (2001); ^ktake the IPCC (1997) value for maize; ^lrefer to Bao (2000); ^mtake the IPCC (1997) value for residue of rapeseed; ⁿrefer to Xing and Zhu (2000); ^otake value for residue.

APPENDIX F

Fraction of each N component from burned biomass N

Species (X)	Emission factor ratio ^a (X/CO ₂ , mol mol ⁻¹)	Emission factor ratio ^b (X/NO _x , mol mol ⁻¹)	Partitioning fraction of burned biomass N (kg kg ⁻¹)
NH ₃	1.3×10^{-3}	0.277	0.034 ^d
N ₂	11×10^{-3}	3.860	0.467 ^d
N ₂ O	0.1×10^{-3}	0.055	0.007 ^d
NO _x	2.1×10^{-3}	1.000	0.121 ^c
RCN ^g	0.5×10^{-3}	0.269	0.033 ^d
PNA ^h			0.338 ^f

^aEmission factor (X to CO₂ ratio by molecule) of biomass burning, given by Sanhueza and Crutzen (1998) and references therein.

^bEmission factor ratio (X to NO_x ratio by molecule), calculated with X/CO₂. ^cIPCC (1997) default value of NO_x emission factor from burned biomass N. ^dPartitioning fraction calculated with X_i/NO_x . ^eValue from Cao and Zhuang (1996). ^fPartitioning fraction calculated by subtracting the fractions of all gaseous species and PNS from 1. ^gRCN=(CN)₂+HCN+CH₃CN+C₂H₅CN+C₂H₄CN (Sanhueza and Crutzen, 1998). ^hParticle N released to atmosphere from biomass burning.

APPENDIX G

Partitioning factor of manure among management systems (IPCC, 1997)

ID code (i)	$f.Wahan_i$	$f.Wahlq_i$	$f.Wahos_i$	$f.Wahds_i$	$f.Wahpp_i$	$f.Wahsd_i$	$f.Wahf_i$
1	0.01	0.38	0	0.01	0	0.53	0.07
2	0	0	0.17	0	0.83	0	0
3	0	0	0	0.16	0.29	0.14	0.40
4	0.06	0.04	0	0.21	0.24	0	0.46
5	0.01	0.02	0.52	0	0.44	0	0.01
6	0	0	0.05	0	0.95	0	0
7	0	1	0	0	0	0	0

Notes: $f.Wahan_i$, partitioning factor of excreta to anaerobic manure management systems (no dimension); $f.Wahlq_i$, partitioning factor of excreta to liquid manure management systems; $f.Wahds_i$, partitioning factor of excreta for daily spread; $f.Wahpp_i$, partitioning factor of excreta to pasture range and paddock systems; $f.Wahsd_i$, partitioning factor of excreta to solid storage and drylot systems; $f.Wahf_i$, partitioning factor of excreta for use as fuel; $f.Wahos_i$, partitioning factor of excreta to other systems.

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