A perspective on reactive nitrogen in a global, Asian and Indian context

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Reactive nitrogen (Nr) includes the inorganic (NH₃, NH₄, NO_x, HNO₃, N₂O, NO₃) and organic forms (urea, amines, proteins, nucleic acids) that readily participate in various reactions of the global N cycle. Over the last half a century, anthropogenic perturbations of the natural N cycle have led to the increasing accumulation of inorganic Nr in the soil, water and air, intentionally through agriculture and unintentionally through fossil-fuel consumption and other activities, adversely affecting human health, biodiversity, environment and climate change. One of the major emerging challenges of this century will be to ensure adequate availability of Nr inputs for agriculture and other activities, while preventing their unwanted accumulation. This article provides an overall perspective of the emerging issues related to Nr in the global, Asian and Indian contexts.

Keywords: Accumulation, anthropogenic perturbations, nitrogen cycle, reactive nitrogen.

NITROGEN (N) is a necessary nutrient for all forms of life. Its gaseous form (N₂) is one of the main constituents of air around us, but it is not usable by most living organisms due to its relatively unreactive nature. Barring a few N₂fixing microorganisms and legume crops that harbour them as symbionts, all other living organisms depend on reactive forms of N. Reactive nitrogen (Nr) refers to any form of nitrogen compound that is radiatively, chemically or biologically active in the biosphere and atmosphere of the earth. This includes inorganic, reduced forms of Nr like ammonia and ammonium (NH₃ and NH₄), inorganic oxidized forms like nitrogen oxides, nitric acid, nitrous oxide, and nitrate (NO_x, HNO₃, N₂O and NO₃), and organic compounds like urea, amines, proteins and nucleic acids. While plants and microorganisms are capable of utilizing inorganic forms of Nr, animals and human beings acquire them through the food chain in readily usable organic forms. The natural processes of producing Nr from atmospheric N₂ gas by lightning or by N₂-fixing microorganisms cannot meet the agricultural demands for food, feed and fibre production by the ever increasing human population, necessitating the anthropogenic production of Nr as fertilizers. However, anthropogenic sources of Nr in our soil, water and air also include by-products of other activities such as dairy effluents, domestic sewage, organic wastes and industrial and vehicular exhausts. When the total production of various forms of Nr exceeds the limits of their utilization and natural processes of their conversion by denitrification (back to inert N₂) in the biosphere, it leads to the accumulation of Nr in the earth's atmosphere, biomass, soils and water. During the last 150 years, the annual inputs of Nr to the earth's soils and fresh water bodies have more than doubled, primarily from anthropogenic sources^{1,2}. As a result, the global nitrogen cycle is being altered perhaps more than any other basic element cycle. There are reports of soil acidification, widespread pollution of groundwater and eutrophication of surface waters (including potable water sources) due to Nr in many places of the world, adversely impacting the health of both people and ecosystems³ (Table 1). While some of these concerns may be regional or national in nature, the greenhouse effects, directly due to N₂O and indirectly due to NO_x (through tropospheric O_3), as well as stratospheric ozone depletion due to N₂O emissions from the earth's surface, have global implications. N₂O gas is particularly significant in this regard, as it has 296 times higher global warming potential than carbon dioxide⁴ and the global consequences of their accumulation cannot be constrained by political boundaries or explained away by local policies. This article is an overview of the human influence on the nitrogen cycle in the global and Asian contexts, along with a brief description of the recent Indian efforts in this regard.

Human Nr creation/anthropogenic contribution of Nr

Production of food and energy has been the driver of human-induced alteration of the N cycle, with the former being the most important, on the average. As hunter-gatherers, humans had a limited impact on the nitrogen cycle due to their low populations, and because they were not creating new Nr, but rather converting existing Nr (e.g. by biomass burning). Thus the extent of the impact was limited relative to natural processes. Even with the advent of agriculture about 10,000 years ago, humans still did not add to

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the existing amount of Nr on the earth, but rather had local effects on the distribution of Nr (e.g. manure for fertilizer). It was not until the advent of legume cultivation over 5000 years ago that humans actually began to add to the amount of Nr on the earth. For thousands of years, this was the most important of the anthropogenic activities that created Nr.

As presented by Galloway et al.2,3, and restated recently⁵, the amount of cultivation-induced biological nitrogen fixation (C-BNF) grew with the global population such that by 1860, humans were adding about 15 Tg N yr⁻¹. This amount was large relative to the amount of Nr created due to fossil-fuel combustion (~0.3 Tg yr⁻¹), but still small relative to naturally occurring BNF in terrestrial systems (~120 Tg N yr⁻¹). By the early 1990s, the amount of C-BNF increased slowly (from ~15 to $\sim 33 \text{ Tg Tg N yr}^{-1}$), the amount of Nr created by fossilfuel combustion increased more rapidly (especially after 1950), and with the development of the Haber-Bosch process in the early 20th century, the amount of new Nr introduced into the environment started to increase rapidly, especially after 1960. By the early 1990s, the amount of Nr created by people (~160 Tg N yr⁻¹) was greater than that produced by natural terrestrial systems (~100 Tg N yr⁻¹; Figure 1). It is important to note that of the Nr created via human action, food production ac-

Table 1. Important negative effects of Nr (modified from UNESCO-SCOPE²⁴)

Direct and indirect effects on humans

Respiratory diseases due to high exposure to:

ozone

other photochemical oxidants

fine particulate aerosol

(on rare occasions) direct toxicity of NO₂

Nitrate contamination of drinking water

Increased allergenic pollen parasitic and infectious diseases

Blooms of toxic algae and decreased recreational use of water bodies

Direct effects on ecosystems

Ozone damage to crops, forests and natural ecosystems

Acidification effects on forests, soils, groundwaters and aquatic ecosystems

Eutrophication of freshwater and coastal ecosystems inducing hypoxia

Nitrogen saturation of forest soils

Biodiversity impacts on terrestrial and aquatic ecosystems Inducing damage by plagues and diseases

Other effects of societal importance

Odour problems associated with animal agriculture
Acidification effects on monuments and engineering materials
Regional hazes that decrease visibility at scenic vistas and airports
Accumulation of hazes in arctic regions of the globe
Depletion of stratospheric ozone by NO₂ from high-altitude aircraft
Global climate change induced by emissions of N₂O
Global climate induced by altered CO₂ and CH₄ exchange
Regional climate change induced by aerosol cooling

counted for \sim 77%, energy production for \sim 16%, and industrial uses for \sim 9%.

Global N budgets

To illustrate the impacts that human Nr creation has had on the global cycles of nitrogen, data pertaining to Nr creation by the geopolitical region for the early 1990s has been shown here² (Figure 2), based on a recent compilation of N cycles in 1860, early 1990s and projections to 2050

In 1860, the most important impacts of human action were from C-BNF, biomass burning and animal husbandry. Only the first resulted in the creation of Nr, the others mobilized existing Nr. Rates from all three of these activities were small relative to terrestrial BNF in unmanaged ecosystems. The one exception is that even in 1860, food production in general resulted in NH₃ emissions to the atmosphere (primarily from animal waste) that was about the same magnitude as those due to natural emissions from soils.

By the early 1990s, on a global basis anthropogenic Nr creation and the subsequent movement of Nr through environmental systems were either larger than natural processes or of the same magnitude. This was due to two reasons – first there were more people, and secondly, the per-capita rate of Nr creation almost tripled from 11 kg N yr⁻¹ in 1860 to 28 kg N yr⁻¹ in the early 1990s.

There are several important points to be noted from the comparison of the 1860 N budget to that of the early 1990s.

(i) Human activities create more Nr than do natural terrestrial activities. Anthropogenic Nr creation increased from ~15 to ~160 Tg N yr⁻¹, primarily due to increases in food and energy production; natural Nr

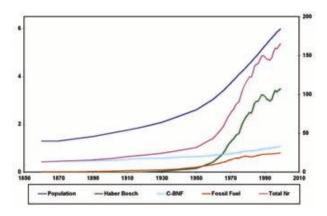


Figure 1. Global population trends from 1860 to 2000 in population (billions) and reactive nitrogen (Nr) creation (Tg N yr⁻¹). 'Haber–Bosch' represents Nr creation through the Haber–Bosch process, including production of ammonia for non-fertilizer purposes. 'C-BNF' represents cultivation-induced biological nitrogen fixation, and 'Fossil Fuel' represents fossil-fuel combustion (after Galloway *et al.*³).

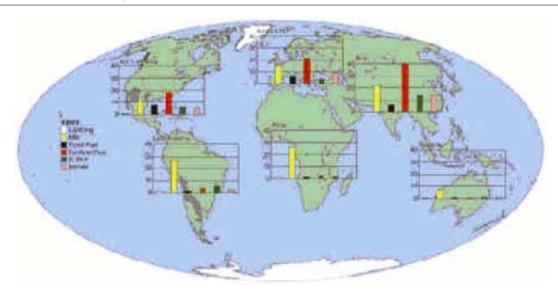


Figure 2. Nr creation by the geopolitical region for the early 1990s (Tg N yr⁻¹; after Galloway et al.²).

creation by terrestrial BNF decreased from ~ 120 to $\sim 107~{\rm Tg~N~yr^{-1}}$, primarily due to land-use changes.

- (ii) As more Nr is created, a larger proportion is emitted to the atmosphere. In 1860, 34 Tg N yr⁻¹ were emitted (24% of the total Nr created). By the early 1990s, 104 Tg N yr⁻¹ was emitted (~40% of total Nr created). This factor has significant implications for the atmospheric transfer of Nr from regions of high emissions (energy and food production areas) to downwind, unmanaged forests and grasslands.
- (iii) As Nr creation increased from 1860 to the early 1990s, the atmospheric emissions became increasingly more important relative to riverine flow. In 1860, the amount of Nr transferred from land to the coast was equal to the amount of Nr transferred from the land to the atmosphere (~28 Tg N yr⁻¹). However, by the early 1990s, twice as much Nr was emitted to the atmosphere (~100 Tg N yr⁻¹) compared to that transferred to coastal waters (48 Tg N yr⁻¹).
- (iv) Nr is accumulating in environmental systems. In the early 1990s it has been estimated that of the ~160 Tg N yr⁻¹ of anthropogenic Nr created, about 100 Tg N yr⁻¹ was denitrified, primarily in soil/water systems; most of the remainder, ~60 Tg N yr⁻¹ accumulated in terrestrial systems. While there is uncertainty in this value, it is unlikely that the accumulation rate is zero.

Thus between 1860 and the early 1990s on a global basis, human Nr creation surpassed natural terrestrial BNF, the atmosphere and commodity transport became increasingly important as distribution vectors for Nr, and there

was significant accumulation of anthropogenic Nr in the environment.

Regional N budgets

Analysis of the N budget on a global scale is useful for a general understanding of how humans have out-competed nature in both the creation and distribution of Nr. However, because Nr creation, use and distribution due to human activities are regionally dependent, the analysis has to also occur on a regional basis to fully understand the degree of alteration and the ultimate consequences.

There is substantial regional variability in the introduction of new Nr (Figure 2). Notable points based on the data include:

- (i) Natural Nr creation (BNF) dominates in Africa and Latin America.
- (ii) Anthropogenic Nr creation with fertilizer production as the largest Nr input dominates in Asia, Europe and North America.
- (iii) Commodity transport of Nr is important. In the early 1990s, inter-regional transport of N-containing commodities, primarily N fertilizer and grain, was substantial. For most regions it was the most rapidly growing source of new Nr.

The regional differences in Nr creation and distribution illustrate that some regions have an abundance of Nr and add as sources for other regions (either inadvertently through atmospheric transport or purposefully via commodity export). Other regions (notably Africa) are deficient in Nr. Both types of regions suffer the consequences of N-cycle alterations, as discussed in the next section.

Consequences of N-cycle alteration

Increased food production is the most important consequence of N-cycle alteration by human activities. It has been estimated⁶ that currently, ~40% of the world's population is sustained by Nr created with the Haber–Bosch process. Unfortunately, with the benefits also come problems. During the incorporation of Nr into grains⁷ and meat⁸, of the Nr created for food production, less than ~20% is consumed by humans. The rest is lost to the environment via the atmosphere, hydrosphere or is stored in soils. In addition, energy production (via fossil-fuel combustion) also creates Nr, and it is emitted directly to the atmosphere. Roughly calculated, of the ~160 Tg N yr⁻¹ of Nr created in the early 1990s, over 90% was released directly into the environment. These releases have negative impacts on the health of both people and ecosystems².

The combination of the multitude of effects coupled with the dynamic nature of the nitrogen biogeochemical cycle, has led to the concept of the 'nitrogen cascade'³, which simply stated is that once a molecule of Nr is created by energy or food production, it will move through the environment contributing to numerous effects, in sequence. The nature of the effect, and its magnitude, depends on the sensitivity of the environmental system to increases in Nr concentrations and the residence time of the Nr. It is noteworthy that at the Third International Nitrogen Conference, the N cascade was adapted to an economic analysis of the impact of human alteration of the N cycle⁹ on the Chesapeake Bay, USA.

The importance of Asia

Eleven thousand years ago, humans began an experiment that continues to this day on the environmental conesquences of resource use. The development of agriculture in the Fertile Crescent region and northern China, and its spread to other regions of the world in the following millennia resulted in landscape changes that increased chemical fluxes to the environment, especially to the atmosphere. The environmental changes were at first local, but by 8000 years ago, it had begun to manifest at the regional and global scale.

The changes occurred due to both direct and indirect drivers. As delineated by the recently completed Millennium Ecosystem Assessment, direct drivers were habitat change, climate change, invasive species, over exploitation, and chemical release. Indirect drivers included population change, economic change, technological change, and sociopolitical and cultural factors.

At the beginning of the experiment, Asia played a 'founding' role. Populations were centred there, and agriculture and associated land-use change also began there. Over time, Asia has continued to be a major factor in the experiment. Over half of the world's population is cen-

tred there, and thus agricultural drivers of change are also centred there. In the 20th century, other regions of the world became the dominant force in other global-scale changes (e.g. fossil-fuel combustion), but their dominance was temporary – Asia is projected over the next few decades to be once again the dominant force in the alteration of the global environment.

It is thus fitting that this study focuses on the human alteration of the N cycle of Asia on current impacts there, how these might change in the future and how N-related changes in Asia influence changes elsewhere.

Assessment of the nitrogen cycle of Asia has received substantial attention over the past decade^{2,10–12}. The common findings are that:

- Human activities are the major source of new reactive
 N in Asia
- 2. Asia is the largest consumer of fertilizer N on a global basis
- 3. There are major ecosystem and human health impacts due to increased Nr in the Asian environment.
- Asia is predicted to become an even larger creator of Nr over the next few decades as both population and per-capita resource use continue to grow.

The most recent N budget for Asia contrasts the major inputs (Nr creation by lightning), biological nitrogen fixation (natural and anthropogenic-induced), fertilizer production, imports of N fertilizer, grain and meat and fossil-fuel combustion into Asia and the outputs (riverine flow, atmospheric advection (by difference)) from Asia for 1995 (Figure 3). Nr inputs total 96 Tg N yr⁻¹ and Nr outputs total 32 Tg N yr⁻¹. In other words, we can account for a third of the Nr that enters Asia, but are missing the other two-thirds! There are estimates for a small (but important) flux – N₂O. Mosier and Zhu¹³ estimate the flux to be 2.1 Tg N yr⁻¹ in the 1994 from agricultural systems, with most of it derived from Nr created by anthropogenic activity. While this flux is small relative to

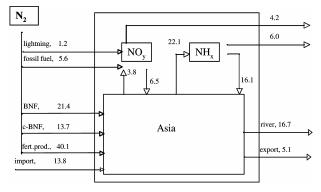


Figure 3. Nr creation and distribution in Asia for the early 1990s (Tg N yr $^{-1}$; Galloway *et al.* 2).

other fluxes, its importance is due to the fact that it is a greenhouse gas in the troposphere and in the stratosphere, it contributes to ozone destruction.

Even though the N₂O flux is important, adding it to the known fates of the Nr introduced into Asia does not help much with tracking the 'missing Nr'. There are two additional fates that need to be addressed, both probably large, and both unfortunately uncertain – Nr storage and Nr converted to N₂ via denitrification. On a global basis, Galloway *et al.*² estimate that there is a 1:2 mix of Nr storage vs Nr denitrification. However, they stress the large uncertainty of both fluxes and conclude that both are certainly important. The same has to be true for Asia. Also true is that it is vitally important that the balance between storage and denitrification be determined, for the former retains Nr in the system for potential impacts in the future, while the latter represents a true and permanent loss of Nr from Asia.

Predictions of future trends of Nr creation in Asia are fraught with challenges. But there is one aspect where the prediction will be certain - Nr creation in Asia will continue to increase in the future, and while population growth will level-off by the mid-21st century, per-capita Nr creation rates will continue to increase as energy use increases and dietary preferences continue to evolve. In 1995, total Nr creation in Asia (fertilizer, fossil-fuel combustion, biological nitrogen fixation (cultivationinduced)) was ~60 Tg N yr⁻¹ and per-capita Nr creation in Asia was 16 kg N capita⁻¹ yr⁻¹. By 2050, if the per-capita Nr creation rate does not change, but population does (~50% increase relative to 1995), Asia will create ~120 Tg N yr⁻¹. This is probably a minimal projection for, given the economic growth rates of Asia, the increased reliance on fossil-fuels and the changing diets (i.e. more meat), the per-capita Nr creation rate is likely to increase over its 1995 levels. How high will it go? While we do not know the answer, we can do the simple calculation showing that if, in 2050, the Asian per-capita Nr creation rate is the same as in North America in 1995 (100 kg N capita⁻¹ vr⁻¹), then the total Asian Nr creation rate would be ~570 Tg N yr⁻¹, or about a ten-fold increase relative to 1995. Is this possible by 2050? Probably not, but is it possible at some time post-2050? We do not know, but unless society does a better job at managing nitrogen, and making its use in agriculture more efficient, it will be more likely to occur and if it does, it is difficult to imagine the state of ecosystems not only in Asia, but also in the world.

The Indian scenario

The need for an integrative approach to research and policy regarding Nr in Indian agriculture, industry and environment was realized in 2004, when the Society for Conservation of Nature (SCON), a voluntary body of sci-

entists, brought together some concerned Indian experts from diverse backgrounds to discuss the issue. This was followed by a series of nationwide consultations in association with the National Academy of Agricultural Sciences (NAAS) in 2005 and with the Union Government's Department of Biotechnology and Indian National Science Academy (INSA) in 2006, with active support from other agencies such as the Ministry of Environment and Forests (MOEF) and Council of Scientific and Industrial Research. The discussions at NAAS on Nr and N use efficiency in Indian agriculture led to the adoption of a policy paper¹⁴. A network of nitrogen researchers and experts called 'Indian Nitrogen Group' (ING), has also been formalized as an outcome of the INSA workshop in 2006. A compilation of position papers and reviews from the nationwide consultations on agriculture, health and environment in the Indian context has also been published recently as a book15, highlighting the need for an integrative understanding of Nr¹⁶. ING has begun to identify gap areas and catalyse research into them, for example, through a recent initiative with the National Fund for basic and strategic research under the Indian Council of Agricultural Research. Based on the policy initiatives of SCON/ING, the Planning Commission is currently considering long-term policy support and funding mechanisms for coordinated research on Nr during the 11th fiveyear plan. ING also came into contact with the International Nitrogen Initiative (INI) (http://initrogen.org), formed in 2003 to address similar concerns and bring about international coordination. Other recent regional initiatives elsewhere, such as an organization called 'Nitrogen in Europe' and the 'NitroEuropeIP', a project for integrated European research into the N cycle, highlight the growing concerns related to nitrogen at the national, regional or international level. ING will coordinate with such bodies to harmonize the national, regional and international concerns and priorities on Nr. As a part of this effort, ING-SCON will host the Fifth International Nitrogen Conference in November-December 2010 at New Delhi.

This special issue is the first ever attempt in a professional journal to distil the essence of the ongoing consultations in the ING with regard to various aspects of Nr. It encompasses most aspects of the nitrogen cycle with special reference to the Indian context, and summarizes the current knowledge as well as identifies the gaps in it for informed decisions on further research and policy. Apart from being a unique reference source at the national level, we hope that this collection of articles will be of interest to all those who wish to understand country-specific issues in the wider international context. Some of the salient issues of Indian concern are summarized below.

In India, as elsewhere, agriculture and fossil-fuel consumption constitute the two most important sources of Nr. However, gross inequalities in development meant that the anthropogenic Nr contribution in India is not uni-

formly distributed within and between states, or within and between sectors. In agriculture, for example, Fertiliser Association of India has estimated that the country consumed about 23 million tonnes of urea alone in 2005– 06. India is currently the third largest producer and consumer of fertilizers (after China and USA), and fertilizer usage is bound to increase with further intensification of agriculture. However, un-uniform expansion of fertilizer use has meant that while chronic N-deficiency continues to be a problem in majority of the Indian soils, the fertilizer N use may be as high as 300 kg ha⁻¹ yr⁻¹ in the intensively cropped regions of northwestern India. This has led to a peculiar situation in which genuine demands for expansion of fertilizer N use in some areas coexist with the concerns over the environmental hazards of excessive and inefficient N fertilizer use in other areas, such as pollution of groundwater and surface water bodies. Loss of N from the soil-plant system results from gaseous plant emission, nitrification-denitrification, surface run-off, volatilization and leaching beyond rooting zones of crops. Considering that the average NUE for cereal production is as low as 33%, the remaining 67% represents an annual waste of up to Rs 72,000 crores (10 billion Euro or 17 billion US\$) in Indian agriculture¹⁴. Therefore, losses/ leakages of reactive N from the agricultural systems are a cause of serious concern for both economic and environmental reasons. Improvement of N-use efficiency through appropriate mix of cultivar choice, fertilizer formulation and agronomic and management practices are important areas of concern in this regard 17-20

The transport of surface reactive N from unused fertilizers, animal wastes and other domestic and industrial sources into streams and rivers and eventually into estuaries and coastal waters is becoming a matter of great concern, as O₂-deficiency in Indian coastal waters is being reported due to enhanced nitrogen loading from land. The contribution of gaseous emissions of Nr from fossil-fuelconsuming industries (especially the power sector) and automobile exhausts is also becoming an increasingly significant factor in the rapidly growing Indian economy²¹. Human health problems due to Nr from both dietary intake as well as pulmonary intake by breathing are being increasingly documented²². There is a paucity of reliable nationwide data on many aspects of Nr, though localized surveys and estimates are available from many regions that enable rudimentary constructions of the Indian N cycle. For example, the contribution of pulses, biofertilizers, natural geodeposits, animal husbandry and dairy industry to Nr in the Indian environment is not well documented. The Indian aspects of the emerging picture of the biology of the N cycle are also not well understood²³. We need a precise understanding of the scale of the nitrogen use/ misuse/release through various agricultural, industrial, vehicular and other activities and their contribution to the pollution of waters and air, with special reference to various point and non-point sources and the biogeochemical N cycle. In this respect, one of the major challenges before the scientific community is to provide policy makers with reliable estimates of Nr transfers to different ecosystems and to describe balanced, cost-effective and feasible strategies and policies to reduce the amount of reactive nitrogen where it is not wanted.

Mitigation options

Considering that N is an essential part of our developmental paradigm, options to minimize the accumulation of Nr in our environment will have to be addressed at many different levels, such as establishing/updating national N information systems, improvements in fertilizer/ biofertilizer formulations, enhancement of the N-use efficiency of our crops and farming systems/practices, reduced dependence on non-renewable energy sources, improvements in fossil-fuel quality, fuel-use efficiency and reduction of fossil-fuel use/abuse, reduction in NOx emissions from farming, industrial and vehicular sources, minimizing anthrophogenic (including agri-industrial) reactive N load in naturally overloaded areas (e.g. geodeposits) and fragile ecosystems, better management of wetland ecosystems that return Nr to the atmosphere as N₂, etc. The sheer diversity of research areas, expertise and approaches it encompasses and the various levels at which the problems need to be identified/tackled, calls for an integrated network approach to harness our intellectual, financial and infrastructural resources effectively. Above all, the scale of the challenge presented by the changing N cycle remains under-appreciated in both policy and scientific circles, and it is hoped that that this special issue will create the necessary awareness to stimulate further research and policy, especially in the Indian con-

Conclusion and future perspective

The impact of food and energy production on the nitrogen cycle has global and regional aspects^{24,25}, and the latter is becoming increasingly obvious at the Asian and Indian levels as well. As the ING has demonstrated in this volume and elsewhere¹⁵, not only is the N cycle seriously altered, but the consequences on both ecosystems and people are significant. In this regard, there are two contrasting but important points to be made. First, the alteration of the Indian N cycle will increase significantly in the future as population, economy and per-capita resource use increase. With this enhanced alteration will come the increased potential for negative impacts on both ecosystems and people. Secondly, the early attention by the ING to the development of an integrated nitrogen management approach will increase the likelihood that the benefits of nitrogen will be maximized, and the problems of excess nitrogen will be minimized.

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