Chapter

2 The European nitrogen problem in a global perspective

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Executive summary

Nature of the problem

- Reactive nitrogen has both positive and negative effects on ecosystems and human health. Reactive nitrogen is formed through the use of fossil fuels releasing large amounts of nitrogen oxides into the atmosphere and through the production of ammonia by the Haber–Bosch process and using it in agriculture to increase our food, feed and fuel production. While the use of nitrogen as a fertilizer and chemical product has brought enormous benefits, losses of fertilizer nitrogen and combustion nitrogen to the environment lead to many side effects on human health, ecosystem health, biodiversity and climate.

Approaches

- The European nitrogen problem is placed in a global perspective, showing the European nitrogen fixation, transport and environmental impacts compared with different regions of the globe.

Key findings/state of knowledge

- Humans, largely through agriculture, but also through burning of fossil fuels, have had a huge impact on the nitrogen budget of the Earth. Europe is one of the leading producers of reactive nitrogen, but it is also the first region in the world where the issue was recognized and in some parts of Europe the reactive nitrogen losses to the environment started to decrease. Europe is a nitrogen hotspot in the world with high nitrogen export through rivers to the coast, NO_x and particulate matter concentrations and 10% of the global N_2O emissions.
- The consequences of nitrogen losses in Europe are visible and are on the average more pronounced than in the rest of the world. Nitrogen contributes to all environmental effects to some extent.
- There is a clear policy on reducing nitrogen oxide emissions that led to reductions by implementation of end of pipe technology. Europe is ahead compared to the rest of the world with NO_x policies.
- Fertilizer production and use decreased in Europe in the early 1990s, in particular, due to the economic recession in the Eastern part of Europe. Currently, the fertilizer use in Europe is about 12 Mton, which is 4 Mton lower than in the 1980s, but increasing again. The nitrogen use efficiency of nitrogen in the EU, defined as the net output of N in products divided by the net input is about 36%. This is lower than the world average (50%) as fertilization rates in Europe are much higher.

Major uncertainties/challenges

- More quantification of the effects is needed to establish cause–effect relationships. Most is known about the exceedances of critical limits, but more quantitative results are needed on impacts, including biodiversity loss, ground water pollution and eutrophication of ecosystems; eutrophication of open waters and coastal areas resulting in algal blooms and fish kills; increased levels of NO_x and aerosols in the atmosphere resulting in human health impacts and climate change; and the increased emissions of the greenhouse gas nitrous oxide resulting in climate change. The effects of nitrogen affecting the other biogeochemical cycles such as carbon and phosphorus need to be quantified on different scales.
- The complexity of multi-pollutant–multiple-effect interactions is a major hurdle to improving public awareness.

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2.1 Introduction

Nature and its biodiversity could only exist because of the availability, even if limited, of reactive nitrogen (N_r) in the system, which is defined as all nitrogen compounds except for N_2. This reactive nitrogen was provided by limited natural sources such as lightning, biomass burning and biological nitrogen fixation. Because of the limited availability, nature became very effective in conserving and re-using reactive nitrogen compounds. Nitrogen, together with other nutrients and water, is the limiting factor for the production of food. Mankind has sought for different ways to increase the crop production necessary for food to sustain a growing population. This has led to the development of synthetic fertilizer production based on the Haber–Bosch process (Smil, 2001; Erisman *et al.*, 2008). This additional availability of reactive nitrogen has led to increased crop production and to the intensification of agriculture. The large increase in population is due to intensification and extension of agricultural land, but also due to the availability of fertilizers. A recent estimate of the current human population supported by synthetic fertilizer is 48%, 100 years after the invention of the synthesis of ammonia from its elements (Erisman *et al.*, 2008; Figure 2.1).

To maximize crop production, the availability of cheap fertilizer in the industrialized world led to excessive use of nitrogen, resulting in a large nitrogen surplus and increased nitrogen losses. As the use of fossil fuel in the industrial revolution expanded, fertilizer production increased similarly (see Figure 2.1). The industrial revolution was accelerated by the combustion of fossil fuels producing heat and power, but also polluting gases, such as carbon dioxide, sulphur dioxide and nitrogen oxides. The use of fossil fuels led at the same time to an increase in the production of fertilizer through the Haber–Bosch process, and to a replacement of manpower by machines increasing the productivity and yield per hectare, further accelerating excess nitrogen. Furthermore, the availability of fossil fuels made globalization possible, transporting food, feed, goods and products all over the world, and depleting nutrients in one area and concentrating nutrients in another area, e.g. in intensive livestock production (Galloway *et al.*, 2008). These leakages from agriculture, industry and transport, in their turn, have led to a cascade of N through the global environment causing a number of different environmental effects: loss of biodiversity, eutrophication of waters and soils, drinking water pollution, acidification, greenhouse gas emissions, human health risks through exposure to oxidized nitrogen (NO_x), ozone (O_3) and particulates, and destruction of the ozone layer.

Europe has benefited to a large extent from the increase in nitrogen, both economically as well as socially (see Jensen *et al.*, 2011, Chapter 3 this volume). Agriculture has contributed to a large extent to GDP development and, apart from some poorer areas in Europe, hunger is no longer a key issue. The situation has, however, developed into overuse of nitrogen in agriculture as a straightforward 'cheap' insurance against low yields with all the concomitant negative side effects. Therefore the focus is to deal with the unwanted downside: optimizing use while minimizing adverse effects.

This chapter of the European Nitrogen Assessment (ENA) provides an overview of the European nitrogen problem in a global perspective. The chapter reviews existing knowledge, bringing different studies together to assess the European nitrogen situation relative to different priorities in other areas in the world. The specific processes and effects are addressed in more detail in the following chapters in this book. This chapter starts with an introduction on reactive nitrogen formation in nature, agriculture and through fossil fuel combustion. Then the nitrogen fluxes are described, including the losses to air and water, followed by a section describing the negative effects of nitrogen in Europe in a global perspective.

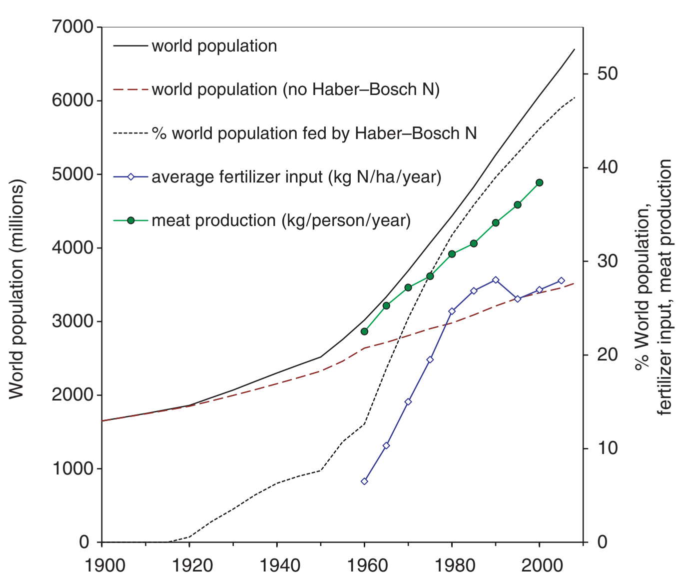

Figure 2.1 Trends in human population and nitrogen use throughout the twentieth century (Erisman *et al.*, 2008). Of the total world population (solid line), an estimate is made of the number of people that could be sustained without reactive nitrogen from the Haber–Bosch process (dashed line), also expressed as a percentage of the global population (short dashed line). The recorded increase in average fertilizer use per hectare of agricultural land (blue symbols) and the increase in per capita meat production (green symbols) are also shown.

2.2 Reactive nitrogen

Reactive nitrogen, N_r, is defined here as all other nitrogen forms in our system apart from N_2. This includes oxidized nitrogen, mainly NO, NO_2, NO_3; reduced forms of nitrogen: NH_4^+, NH_3 and organic nitrogen: proteins, amines, etc., with different states of oxidation (Table 2.1)

Natural sources of the formation of N_r include volcanoes, biological nitrogen fixation in natural soils and lightning (Figure 2.2, Box 2.1) (Smil, 2001; Reid *et al.*, 2005; Schlesinger, 2009; Sutton *et al.*, 2008 and Hertel *et al.*, 2011 (Chapter 9 this volume), Simpson *et al.*, 2011 (Chapter 14 this volume), where details are provided) and weathering of rocks (Holloway and Dahlgren, 2002). Senescence of plants, wildlife and forest fires are natural processes that result in the re-distribution of N_r in the biosphere. Most of the abiotic natural sources of N_r are in oxidized forms, although wildlife and volcanos also emit reduced forms (Galloway *et al.*, 2003; Schlesinger, 2009). In Europe natural sources of N_r are estimated to create annually 2–3 Mton N_r (van Egmond *et al.*, 2002; Galloway *et al.*, 2004). Anthropogenic activities enhancing the formation of N_r include cultivated Biological Nitrogen Fixation (BNF) in agriculture, N_2 fixation through the Haber–Bosch process, the burning of fossil fuels and forest fires (Figure 2.2).

Table 2.1 Examples of nitrogen components and their oxidation state

Oxidation state	Example	Component name
Reduced forms		
−3	NH_3	Ammonia
−2	NH_2NH_2	Hydrazine
−1	HNNH	Diimide
0, non-reactive	N_2	Di-nitrogen
Oxidized forms		
+1		
+2	NO	Nitrogen oxide
+3	HNO_2	Nitrous acid
+4	NO_2	Nitrogen dioxide
+5	HNO_3	Nitric acid

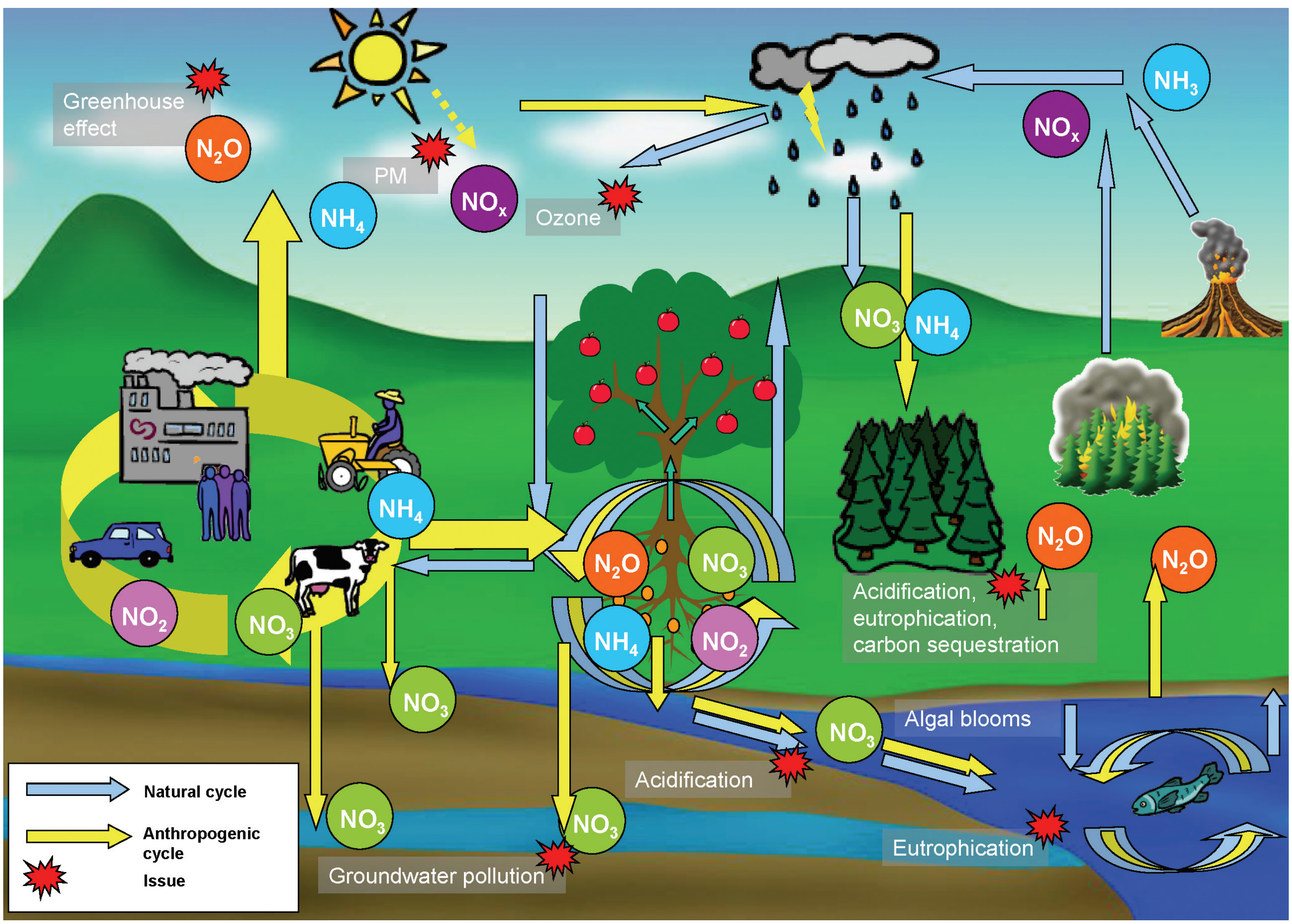

Figure 2.2 The N_r cycle and the main fluxes (picture by Anne-Christine LeGall).

Of the major nutrients needed for biomass production, nitrogen is most commonly the limiting one, at least in terrestrial systems (De Vries *et al*., 2011, Chapter 15 this volume). Phosphorus and potassium are the other major limiting nutrients. Higher plants can mostly only use nitrogen after it has been converted to reactive forms such as nitrate (NO_3^-) or ammonium (NH_4^+). There are basically three ways of accomplishing this:

- decay of organic matter by microbes in soils (dead organisms, leaves, manure, etc.) and eventually release biologically available forms of N;
- Biological Nitrogen Fixation (BNF): nitrogen-fixing organisms (e.g. bacteria) 'fix' atmospheric N_2 into biologically available forms of reactive nitrogen;
- production and application of reactive nitrogen as inorganic fertilizer.

2.2 Human intervention in the nitrogen cycle

The nitrogen cycle with an explanation of the most important processes is given in Box 2.1. Humans increase the creation of N_r by three processes. The first process is the combustion of fossil fuels for energy production, which generates nitrogen oxides from the oxidation of N_2 or fossil organic N in the fuel. Second, the production of fertilizers and chemicals (e.g. nylon, explosives), mainly through the Haber–Bosch process, which creates NH_3 by the reaction of N_2 and H_2. And third, the planting of nitrogen-fixing crops (e.g. legumes) which convert N_2 to NH_3 incorporated in the organic matter.

The primary emission of oxidized nitrogen is through the formation of nitrogen oxides (NO and NO_2) and nitrous oxide (N_2O). Through reactions in the atmosphere other oxidized nitrogen compounds are formed. Oxidized nitrogen has several sources, but is formed mainly by combustion processes where fuel N is oxidized or atmospheric N_2 is oxidized at high temperatures. These processes occur in industries, fuel combustion for transportation and energy production. Among the other sources of oxidized nitrogen, soils are most important (Skiba *et al*., 1994; 1997).

The Haber–Bosch process has facilitated the production of agricultural fertilizers on an industrial scale, dramatically increasing global agricultural productivity in most regions of the world. The number of humans supported per hectare of arable land has increased from 1.9 to 4.3 persons between 1908 and 2008 (Erisman *et al*., 2008). This increase was only possible because of nitrogen from the Haber–Bosch process.

An additional use of fertilizer is the production of crops and biomass for bioenergy and biofuels. Currently, bioenergy contributes 10% to the global energy use, while biofuels contribute 1.5% and the influence on global fertilizer use is still marginal. However, present climate and energy policies tend to stimulate biofuel production so that the influence of Haber–Bosch nitrogen will tend to grow, depending on which soils and crops are used and in how far N-efficiencies in food production can be increased (Erisman *et al*., 2009). The environmental impacts of inefficient use of fertilizer and livestock breeding systems result in groundwater pollution, airborne emission of ammonia and nitrous oxides, contributing to excess nitrogen cascading through the environment with negative impacts on human health and ecosystem services.

Sources of reduced nitrogen include emissions from the fertilizer industry and/or other industry applying ammonia as a

Box 2.1 The global nitrogen cycle and the main processes

The nitrogen cycle consists of the following main processes.

- Biological Nitrogen Fixation. $2N_2 + 3H_2 \rightarrow 2NH_3$. This is a natural process performed by a number of diazotrophs, such as *Anabaena* (a cyanobacterium), and *Rhizobium* (the symbiotic bacterium found in legume root nodules).
- Nitrogen fixation. Humans influence the nitrogen cycle through industrial N fixation. Conversion of N_2 into NH_3 by the Haber–Bosch process.
- Nitrification. This is the oxidation of ammonia to oxyanions. The initial oxidation to nitrite, $2NH_3 + e^- + 3O_2 \rightarrow 2NO_2^- + 2H_2O + 2H^+$, is performed by bacteria such as *Nitrosomonas* and the next step oxidation to nitrate, $2NO_2^- + O_2 \rightarrow 2NO_3^-$ is performed by *Nitrobacter*.
- Denitrification. Some bacteria, such as *Pseudomonas*, are able to use nitrate as a terminal electron acceptor in respiration: $2NO_3^- + 12H^+ + 10e^- \rightarrow N_2 + 6H_2O$.
- Assimilation. Plants assimilate nitrogen in the form of nitrate and ammonium. The nitrate assimilated is first reduced to ammonium, and then combined into organic forms, generally via glutamate. Animals generally assimilate nitrogen by first breaking protein down into amino acids.
- Decay (ammonification) and excretion. When plants and animals decay, putrefying bacteria produce ammonia from the proteins they contain. Animals also produce breakdown products such as ammonia, urea, allantoin and uric acid from excess dietary nitrogen. These compounds are also targets of ammonification by bacteria.
- Annamox reaction. This is conversion of nitrite and ammonium to pure nitrogen gas (N_2) in seas and oceans, hot springs, hydrothermal vents, and many freshwater wetland ecosystems, which than escapes to the atmosphere. The reaction mechanism is triggered by a newly discovered bacterium, called *Brocadia anammoxidans*.
- Volatilization. Turns fertilizers and manures on the soil surface into gases like NH_3, N_2O and N_2 that also join the atmospheric pool.
- Weathering of rocks. The process where stored nitrogen in rocks is released by wind, rain and erosion.
- Runoff. Carries the nitrogen in fertilizers and manure and the nitrogen in the soil into rivers and streams causing a concern for water quality.
- Leaching. Carries nitrates deep into the soil so that plants can no longer use them, producing a dual concern; for lost fertility and for water quality, as nitrates enter the groundwater and wells that provide drinking water.

chemical, from fertilizer application, and plant senescence, from livestock manure (in the field, housing systems and application of manure), cars equipped with three way catalysts to reduce NO_x, industrial emissions applying urea or ammonia Selective Catalytic Reduction (SCR) to reduce emissions, households using ammonia as a cleaning agents and excretion by cats and dogs.

Total denitrification of nitrate by microbes in soils, groundwater and surface water is considerable. Particularly when nitrate concentrations are comparatively high, denitrification is incomplete and small quantities of N_2O are released in the process. N_2O is also released during nitrification. N_2O is also a by-product in the Haber–Bosch process of synthetic fertilizer production.

2.3 Nitrogen use in agriculture

Europe is one of the world's largest and most productive suppliers of food and fibre. In 2004 Europe produced 21% of global meat production and 20% of global cereal production. About 80% occurred in Europe, defined here as the 25 European countries, EU25 (IPCC, 2007). The productivity of European agriculture is generally high, in particular in Western Europe: average cereal yields in the EU are more than 60% higher than the global average (EFMA, 2010). The major sources of N_r in agriculture are Biological Nitrogen Fixation and the use of inorganic and organic fertilizers. This section will outline the major sources for European conditions comparing this with global context. Important for the environmental aspect of agricultural nitrogen is the nitrogen use efficiency. Finally, the influence of bioenergy on the agricultural nitrogen is described.

2.3.1 Biological Nitrogen Fixation (BNF)

Biological nitrogen fixation (BNF) is a vital biological process which allows atmospheric molecular di-nitrogen (N_2) to be converted into mineral nitrogen (NH_3) that can then be assimilated by living organisms. This process is carried out by specific N-fixing bacteria that are either free-living in soil or water or associated with the root nodules of legume plants.

Global terrestrial BNF is estimated at 120 Tg N/yr, of which a little less than half is fixed in oceans, the rest on land (Smil, 2001). BNF in Europe is estimated at 14.8 Tg N/yr in natural soils (Galloway *et al*., 2004). Crop BNF, the human induced BNF, was estimated by Galloway *et al.* (2004) to be 30 Tg N on the global level and for Europe 3.9 Tg N/yr, while Velthof *et al.* (2009) estimated for Europe a value of 5 Tg N/yr. The distinction between natural and crop BNF is difficult to make when fertilizers are added or atmospheric deposition substantially contributes to nitrogen inputs as this can suppress BNF (van Kessel and Hartley, 2000).

2.3.2 Nitrogen in mineral and organic fertilizer

Mineral fertilizer

Nitrogen is an essential element for plant growth, being a component of chlorophyll, amino acids, proteins and enzymes and increased nitrogen application leads to higher crop production (see e.g. Olson and Kurtz, 1982). Sufficient supply of nitrogen is required for plant metabolism, and addition of N will essentially increase the efficiency of photosynthesis to produce carbohydrates. Higher inputs of nitrogen have increased yields as shown in Figure 2.3 where the changes in yield and fertilizer intensity are plotted for 1961–1990. In the USA and Europe the yield has decreased during recent years showing that the efficiency of the added nitrogen has become less.

Nitrogen fertilizers are manufactured by combining atmospheric N_2 with hydrogen from methane or gasified coal to produce ammonium nitrate, ammonium sulphate, or urea. Nitrogen fertilizer data throughout the world shows that the annual use rate is increasing (FAO, 2010c; Davidson 2009). For Europe there is no inventory of fertilizer data before 1960. Therefore, we used application rates in kilograms of fertilizer per hectare for the years 1910, 1920, 1935 and 1950 as reported by Moïssey Postan and Rich (1952) to construct the European time series. Figure 2.4 illustrates the trends in amounts of applied fertilizer in Europe compared to the whole world. There is an overall increase in fertilizer application on a global scale. There is some anti-correlation with the gas prices explaining the reductions in application. Throughout the 1930s, European consumption of nitrogenous fertilizers remained above half of the world's total while the continent's arable land accounted for only 12% of all cropland. The applications were heavily concentrated in Germany, Benelux, England and France and applications outside the Northwestern part of the continent remained marginal. After a decrease of production during the Second World War their synthesis and applications began to grow substantially only during the early 1950s. The big drop in 1990–1992 has two main reasons: (1) the collapse of the Eastern European countries economy, which had a dramatic effect; and (2) the impact of the McSharry reform (new CAP in 1990). This was the introduction of the mandatory set aside, and the farming community has 'heavily overreacted' to this new policy. Then we have seen a slight recovery in the following years (until 1996), shown in Figure 2.4. Europe accounts currently for about 10% of the global use of N fertilizers. After the political changes in Eastern Europe around 1989, the economic breakdown resulted in a strong decrease in fertilizer application, which is reflected in Figure 2.4. The mean national application rate varies very much in Europe from the lowest level of 42 kg/ha of agricultural land in Portugal to 243 kg/ha on grassland in the Netherlands.

Manure and livestock

The process of livestock production does not directly lead to the creation of new N_r, but to a concentration of N_r and a redistribution of it over different spatial scales. This occurs through the intake of feed, international and local transport of feed and the emissions to air and soil/water. Therefore, livestock production is very important for N_r as an external

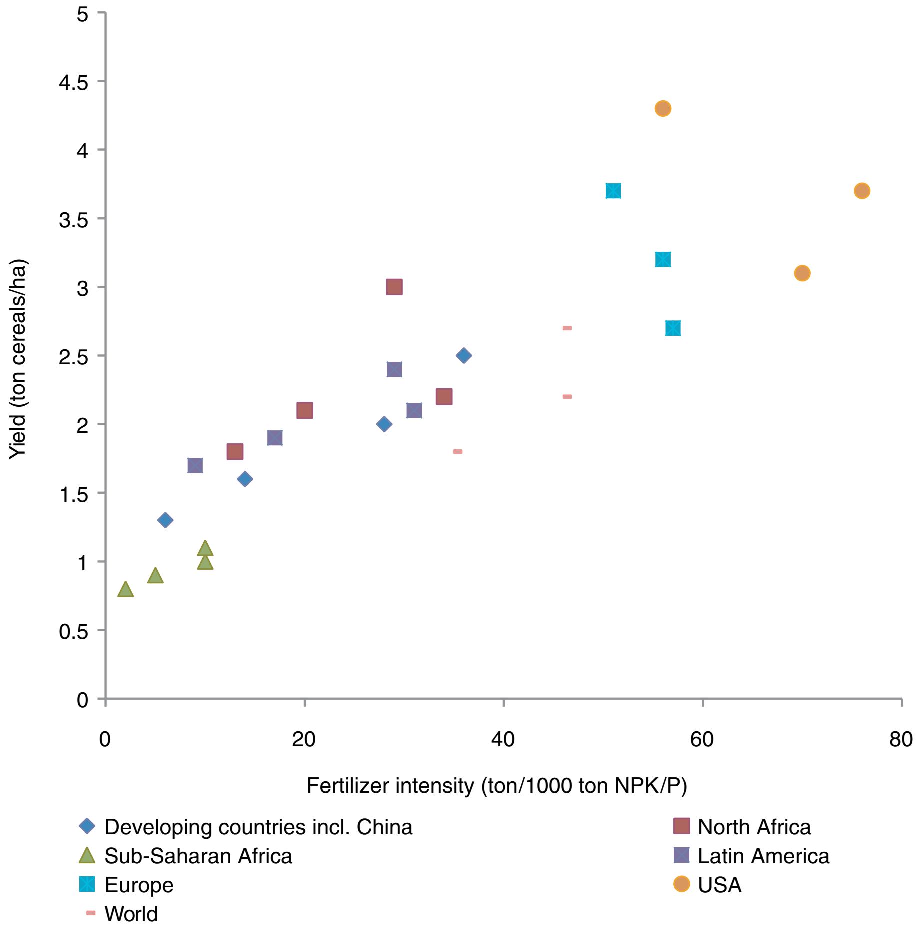

Figure 2.3 Overall yield (Y) and fertilizer intensities (FI) for the developing regions, and other countries and regions between 1960 and 1990. The fertilizer intensity is the fertilizer input (expressed as the amount of N + P_2O_5 + K_2O) as a fraction of total biomass production (FAO, 2010a).

input in different regions or an internal cycling of nitrogen with appreciable losses to the environment. Europe has a large share of animals and animal breeding farms in the world (Table 2.2; Figure 2.5). Meat production in Europe in 2004 was about 20% of the world production, milk production about 30% (source FAOstat: FAO, 2010c). The number of animals is shown in Table 2.2. The regions of most intensive livestock production in Europe include Denmark, the Netherlands, Belgium, Brittany, Spain, Poland, the UK and the Po Valley (Italy). The density of pigs per unit area is among the highest in the world together with some regions of China and the USA as can be seen in Figure 2.5. Cattle are more equally distributed over the world and are linked to the grassland areas (land bound). Nitrogen cycling within European livestock production is very important with the total N excretion from animals almost equalling the use of mineral fertilizer in Europe (Oenema *et al.*, 2007). Although livestock production does not directly lead to the production of new N_r, there are relevant indirect interactions. The nitrogen supply for animal feeds is dependent on new N_r supply both through fertilizers and BNF. Thus increasing livestock production, results in additional demands for nitrogen fixation.

Livestock manure production in the world was plotted by Davidson (2009) and is shown in Figure 2.4. European data are not available for the period before 1960. Data were obtained from Buijsman (1986) and OECD to compile a similar trend for Europe, plotted in Figure 2.4, with the global and European fertilizer use for the same period. The data are highly uncertain. There is a difference between the ratio of manure to fertilizer use globally and that in Europe. Currently, about equal amounts of nitrogen are applied in manure and fertilizers in Europe. Globally, however, the amount of nitrogen applied in manure was an order of magnitude higher than that applied in fertilizer, currently decreased to a factor of two. Apparently much manure is produced globally on unfertilized (grass)lands, whereas in Europe most agricultural land is fertilized. The inputs in agriculture in Europe are further specified and presented in De Vries *et al.*, 2011 (Chapter 15 this volume) and Leip *et al.*, 2011 (Chapter 16 this volume).

2.3.3 Nitrogen use efficiency

During the last decade the EU Common Agricultural Policy (CAP) has been reformed to reduce overproduction, reduce environmental impacts and improve rural development. This is not expected to greatly affect agricultural production in the short term (OECD, 2001). Excessive application of nitrogen leads to an imbalance as not all applied nitrogen can be taken

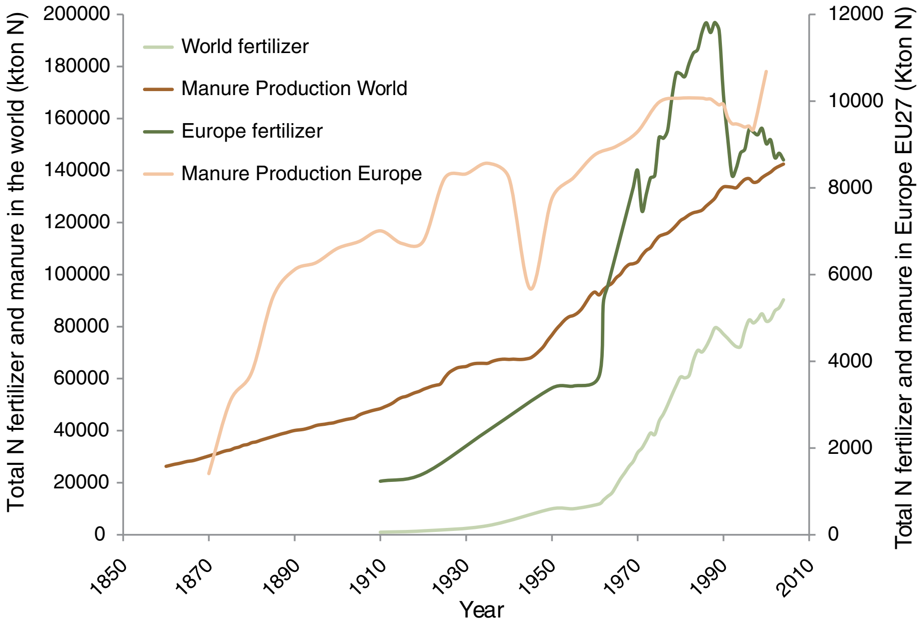

Figure 2.4 Global and European livestock manure and fertilizer nitrogen consumption (Kton N). Global data are obtained from Davidson (2009). European data are constructed from animal numbers and excretion factors by Buijsman (1986) and the INTEGRATOR model (de Vries *et al.*, 2009).

Table 2.2 Livestock numbers in Europe and the world in 2005

Livestock (1000 head)	Europe	World
Poultry	1 329 162	15 146 608
Pig	164 794	917 635
Cattle	100 508	1 310 611
Small ruminant	142 476	1 722 175

Source: From Steinfeld *et al.*, 2006

up by the crops. The nitrogen surplus or nitrogen balance is an indicator for the agricultural pressure on the environment. The gross nutrient balance is calculated by subtracting the sum of the total nitrogen output in harvested crops and forage from the total nitrogen input calculated as the sum of total fertilizer N (inorganic fertilizers, organic fertilizers: organic inputs from non-agricultural sources: urban compost and sewage sludge spread on agricultural land), livestock manure production, manure stocks (stock levels, imports and exports of livestock manure), biological nitrogen fixation and atmospheric deposition of nitrogen compounds (EEA, 2005b; OECD, 2001; Campling *et al.*, 2005).

Nitrogen use efficiency (NUE) can be defined in different ways. Usually in agriculture it is the nitrogen in the product leaving the farm divided by the nitrogen input to the farm. The nitrogen use efficiency (%), generally increases with a decrease in N input and N surplus as calculated by the nitrogen balance. The efficiency is less than 50% in countries with an N surplus above 80 kg/ha/yr (the Netherlands, Belgium, Denmark and UK), between 50% and 70% in countries with an N surplus between 50–80 kg/ha/yr and more than 70% in countries with an N surplus below 50 kg/ha/yr, except for Portugal and Spain (OECD, 2006).

The worldwide database compiled by FAO shows that the NUE has decreased exponentially in all countries except Western Europe and the United States (Figure 2.6). The driver for these trends is the increasing amount of N fertilizer applied in all world regions except Europe. In the analysis, the grain production was computed as the sum of all cereal crops and maize production. Grain production has increased linearly since 1960 in the United States and Western Europe (Hatfield and Prueger, 2004). These changes in grain production have caused a slight increase in NUE in the past decade. However, these trends may hide the effect of manure, which is applied in large amounts. In addition, the NUE of fertilizer in the United States and Western Europe is low because of over-application (see also Figure 2.3).

Another approach for defining the NUE is to consider the consumed amounts of calories and of protein as effectively used by humans. Ultimately, nitrogen for food production aims to provide the necessary proteins. Animal protein is much more inefficient in terms of NUE than plant proteins. The more efficiently protein is obtained by humans, the higher the NUE (van Grinsven *et al.*, 2003). Estimates for different regions of the world (Figure 2.7, van Grinsven *et al.*, 2003) show major regional differences in protein consumption per capita. The differences mainly result from variation in the fraction of protein in diet provided by animal products and in the type of animal product. In the developed countries more animal protein is consumed than in developing countries, where, especially in the low protein countries almost all proteins are consumed through vegetable food products.

Rough estimates can be made of the amount of nitrogen that was used in agriculture to produce the consumed amount of protein. This yields a nitrogen consumption efficiency for the different regions in the world (Figure 2.7). In western societies about 60% of the harvested crop, or whole animal is converted

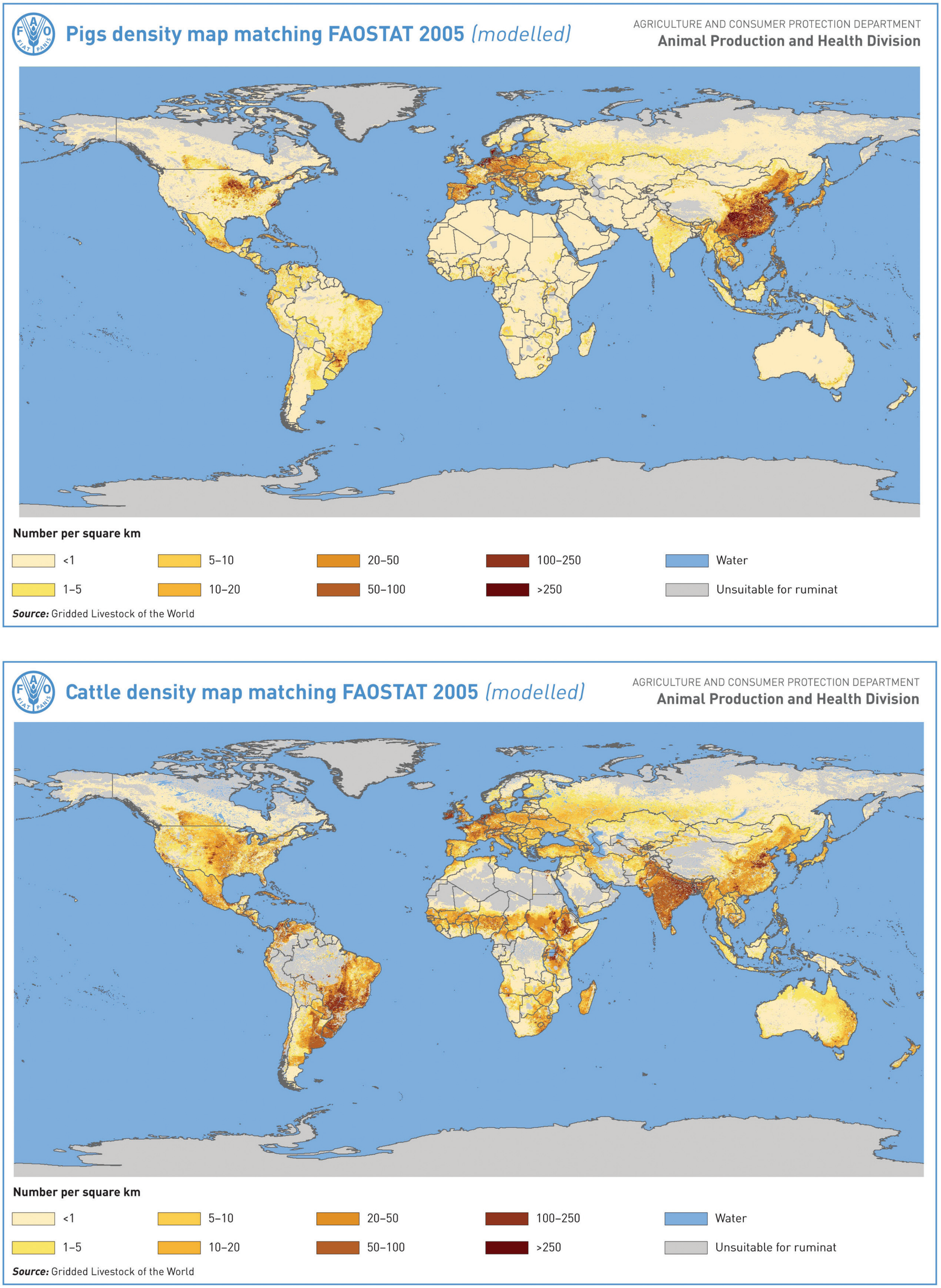

Figure 2.5 Global pig (top) and cattle (below) density in 2005 (FAO, 2010b).

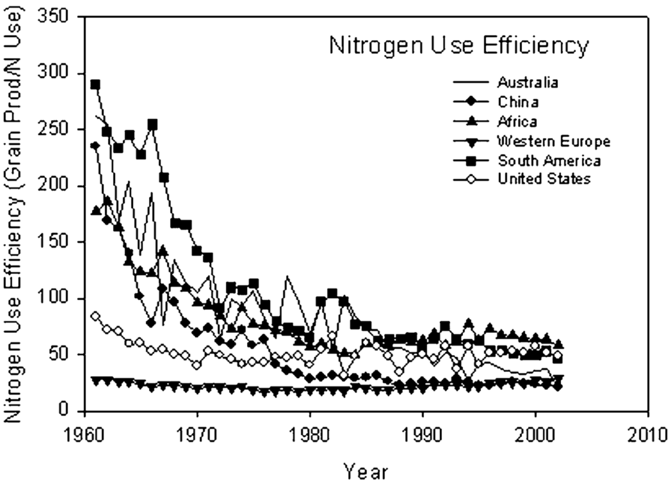

Figure 2.6 Nitrogen use efficiency for grain production relative to N fertilizer use for 1961–2002 for selected regions in the world (Hatfield and Prueger, 2004). Data source is http://faostat.fao.org/faostat.

to food products. Without correcting for over consumption of proteins as compared to the recommended intake of protein (60 g per day capital or 3.5 kg N per year capita; recommended by the World Health Organisation (WHO, 2007), the nitrogen efficiency of consumption varies from 10% in the USA to 28% in China (van Grinsven *et al.*, 2003). Present Chinese consumption is far less N-efficient compared to the data in Figure 2.7, because animal protein consumption has increased substantially since 1995.

2.3.4 Biomass and food production and future fertilizer consumption in Europe

Biomass is the oldest resource of energy used by mankind and has been the main source of energy until a century ago (Smil, 2004). Because of the inherently low efficiency of the photosynthetic process, no form of energy supply has such low power densities, and hence such high land demands (and fertilizer), as does the production of phytomass (Smil, 2004). In principle, there is globally enough annual growth of new biomass to cover up to four times the human annual energy use (Dornburg *et al.*, 2007). However, in order to grow, collect and use biomass in a sustainable way to satisfy the human energy requirements, a well regulated and optimized process is needed. The European Fertilizer Manufacturer Association (EFMA) reports that in Europe the most ambitious Action Plans for biofuels production are still those of France and Germany. However, the UK, Sweden, Italy and Greece now also have ambitious objectives for biofuels production, closely followed by Austria and Denmark, the original 'pioneer' countries in this domain (EFMA, 2010). There are sufficient domestic resources to meet the EU targets set for the year 2010 but if more stringent goals are set for bioenergy in the future, it will be challenging to find sufficient resources in Europe and biomass imports from outside the EU (Fagernäs *et al.*, 2006; Londo and Deurwaarder, 2007). There is a major challenge to reach the targets in a sustainable way and there is much discussion on the availability of different biomass sources for bioenergy application, especially in relation to the additional use of fertilizer and the effect on greenhouse gas emissions (N_2O).

These developments are already affecting the food area as we see that energy crops are grown on former cropland and grassland, even without using the energy crop premium. According to EFMA (2010), in the coming decade, the production of biofuels will contribute to the 4.7% increase of nitrogen consumption in the EU-27 between 2009 and 2019. These prospects do not take into account possible new generations of bioenergy which might present an additional potential. For the longer term, recent scenario's predict a much higher increase of fertilizer application as the result of increased food demand, biofuels production and limited land availability (Erisman *et al.*, 2009).

2.4 Energy, transport and industry

The major link of energy, transport and industry to the nitrogen issue is the direct emission into the atmosphere of nitrogen oxides (NO_x) from combustion of fossil fuels. The gases disperse, react and are eventually lost through deposition to the earth surface as gas or aerosol. Nitrogen oxides contribute to a variety of adverse effects, such as the formation of tropospheric ozone, the deposition of acidifying and eutrophying substances and the formation of secondary aerosols (mainly ammonium nitrates). While aerosols having an impact in Europe are largely formed from European emissions, background ozone levels in particular are significantly affected by NO_x emissions throughout the Northern Hemisphere (with substantial contributions from emissions in Asia and North America).

There are other links with energy, transport and industry to the nitrogen issue, which are much less well quantified. Energy (coal or natural gas) is needed to produce nitrogen fertilizers and thus related to energy. Furthermore, through the use of fossil fuels the labour by man and draught animals has been replaced by machines and agriculture could be expanded and intensified leading to higher production in total and per ha. Furthermore, increased transportation of fertilizer, feed, food, fuel and other products has led to a redistribution of N_r over the world, while emitting NO_x on the way.

2.4.1 NO_x formation processes

NO_x is mainly formed by two processes: thermal NO_x, when nitrogen and oxygen in the combustion air combine with one another at the high temperatures in a flame, and fuel NO_x by the reaction of nitrogen bound in the fuel with oxygen in the combustion of air. A third and generally less important source of NO_x formation is *prompt* NO_x that forms from the rapid reaction of atmospheric nitrogen with hydrocarbon radicals (Dean and Bozzelli, 1999). Large combustion plants in power generation contribute to NO_x emissions from high stacks,

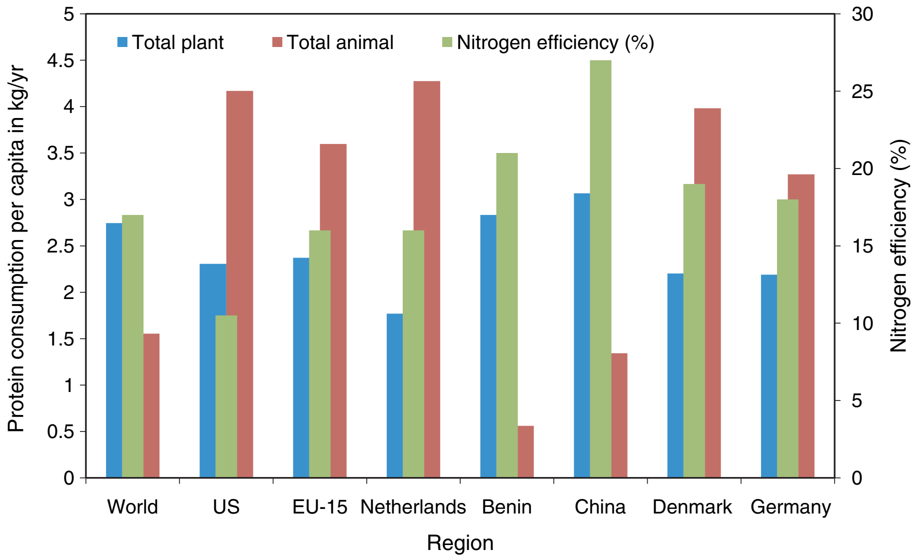

Figure 2.7 Protein consumption per capita as direct nitrogen intake in kg/yr in 1995 for different regions of the world and the nitrogen efficiency of protein consumption relative to N inputs (%) (van Grinsven *et al.*, 2003).

while road transport sources are mainly line sources. Urban traffic, residential and commercial combustion as well as off-road sources can be classified as area sources. NO_x emissions in Europe have fallen markedly in the last decades, mainly due to stringent emission controls applied to large combustion plants (EC Large Combustion Plants Directive, EEA, 2005a) and the EURO standards for road transport vehicles. At the same time, overall emission control due to effect-based regulations (EC National Emissions Ceiling (NEC) Directive and Gothenburg Protocol) have led to reductions in other sectors which have contributed to this decline (see Hertel *et al.*, 2011, Chapter 9 this volume).

Total non-transport NO_x emissions in Europe are currently about 2000 kton/yr (EEA, 2005a). Emissions of NO_x from public electricity and heat production in the EU fell by 45% over the period 1990 to 2004. If the structure of power production had remained unchanged from 1990 then by 2004 emissions of NO_x would have increased by 33% above their 1990 levels, in line with the additional amount of electricity and heat produced. This decoupling of NO_x emissions and electricity and heat production over the period 1990 to 2004 has been due to the following (EEA, 2005a).

- The introduction of low-NO_x combustion technology and flue gas treatment, which led to a 49% reduction.
- Efficiency improvements, which resulted in a 14% reduction.
- The switch in the fuel mix, away from coal and fuel oil towards natural gas, which led to an 8% reduction.
- The lower share of nuclear and non-thermal renewable energy (i.e. excluding biomass) in 2004 compared to 1990, which actually increased emissions by 3%.

The overall effect was a 45% reduction in NO_x emissions in 2004 compared to 1990 levels.

The total transport emission of NO_x from Europe is currently about 8000 kton/yr. The specific emissions of air pollutants from passenger and freight transport decreased for most modes of transport, more so for passenger transport than for freight transport (EEA, 2007). The highest reduction of specific emissions can be found in the road sector, following the increasingly stricter emission standards. Rail only slightly improved its performance over the past decade. Inland waterway freight transport stabilized its emissions per tonne-kilometre, while maritime passenger and freight transport increased their specific emissions over the past decade.

2.4.2 Additional NO_x from bioenergy use

Biofuels and bioenergy are forms of energy (heat, power, transport fuels or chemicals) based on different forms of biomass. Recently, the EU adopted new targets for sustainable energy and greenhouse gas (GHG) emission reductions: 20% GHG reductions and 20% contribution of sustainable energy sources, including a target of 10% share of biofuels in the transportation sector in 2020 (EU, 2009). It is clear that biomass as transport fuel (biofuels), electricity and heat production (bioenergy) and Substitute Natural Gas (SNG or Green gas) will be a major component necessary to reach the targets. By 2050, it is estimated that biomass and waste utilization could rise from 9.0 to 13.5 EJ/a (215–320 Mtoe) (EU Biomass Action Plan, 2007).

Increased biomass production potentially requires more fertilizer inputs, which will accelerate the nitrogen cycle (see Section 2.7). Additional fertilizer use will also cause additional N_r losses. Furthermore, bioenergy emits NO_x into the atmosphere when combusted without de-NO_x installations such as SCR. Additional emissions of NO_x might be expected because the fuel-N is higher compared to that in fossil fuels and/or no de-NO_x installations will be used for small scale applications and because more energy (combustion) is needed to produce one unit of electricity or transport. The

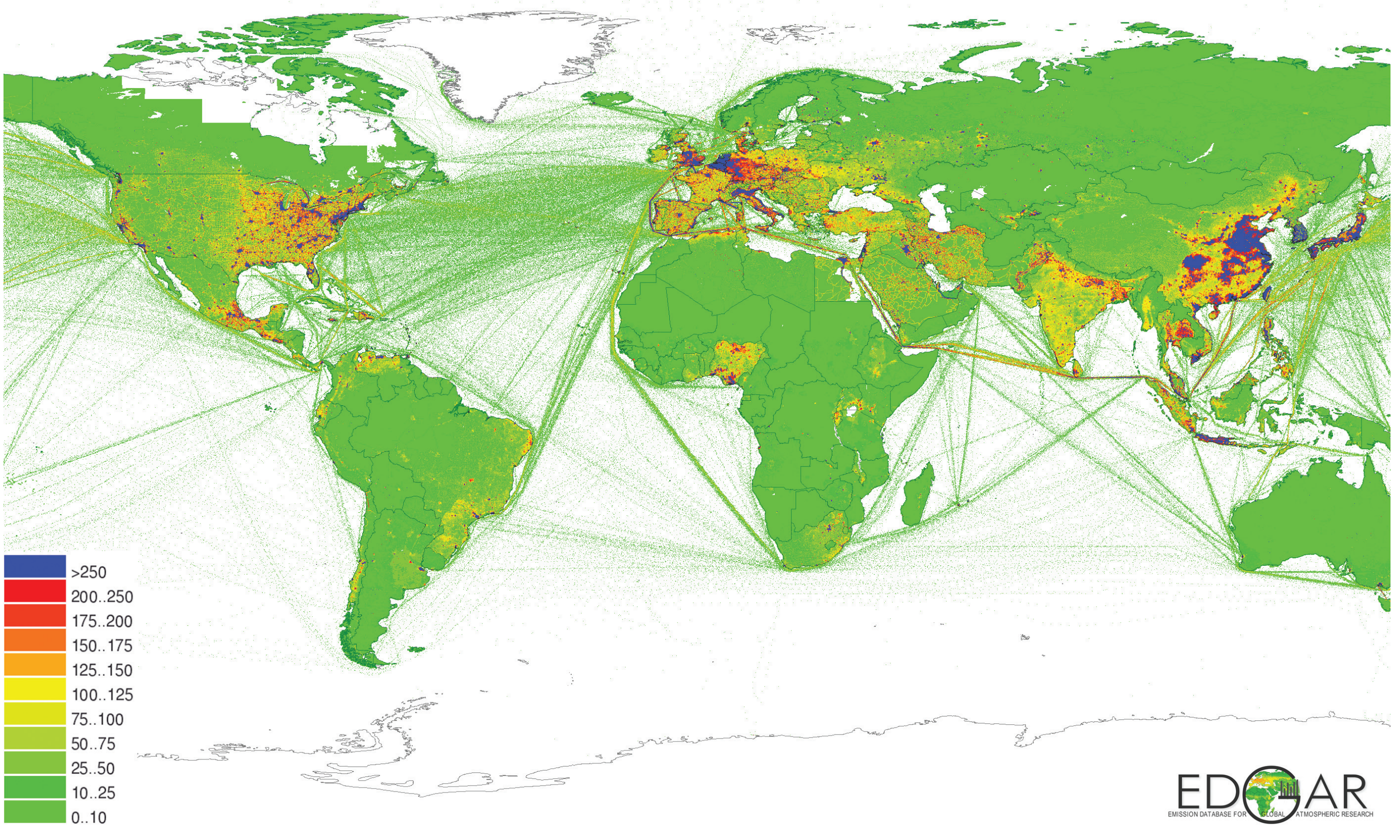

Figure 2.8 Global emissions of nitrogen oxides (NO_x) (EDGAR, 2010).

direct nitrogen emissions from different options to produce heat and power were compared by Pehnt (2006). Power generating systems excluding biomass are considerably better than the 'reference mix' which is based on fossil fuels, but biomass systems are well above the reference mix. An exception to this is systems with co-combustion of forest wood. This is due, in particular, to the fact that the NO_x emissions of small combustion plants tend to be higher due to the lower temperature and efficiency. A special case is the biogas system. The nitrogen emissions of this system are more than the reference mix owing to the ammonia emissions resulting from the animal manure of the agricultural system prior to combustion (Pehnt, 2006).

2.4.3 European NO_x emissions in a global perspective

Figure 2.8 shows global NO_x emissions for the year 2000 (EDGAR, 2010). Europe contributed about 14% of global NO_x emissions in the year 2000, which was lower than that by North America (17%) and Asia (12% for S/SE Asia and 14% for East Asia, respectively). By far the largest sectoral contribution (Figure 2.9) to European NO_x emissions stems from mobile combustion sources, contributing an estimated 62% (road transport 30.7%, other mobile sources 16.2% and international shipping 15%), followed by stationary combustion (15.7% from large combustion plants and 4.5% residential and commercial combustion). Current inventories, such as the EDGAR inventory used here, often do not include data on natural and biogenic sources of emissions (or if so only partially), as can be seen in Figure 2.9 showing a very low contribution from biogenic and natural sources.

Figure 2.10 indicates the estimated atmospheric transport distance of NO_x emissions of Europe and North America (Sanderson *et al.*, 2008) showing that Europe is substantially impacting parts of Asia and North America, and vice versa Europe is mostly influenced by emissions from North America. A few percent of the NO_x emissions from North America are reaching Europe. The Taskforce on Hemispheric Transport of Air Pollution states that on average 75% of the NO_x emissions in Europe is deposited within Europe, with small fractions falling on North America (1%) (Sanderson *et al.*, 2008); South Asia (2%); East Asia (2.5%), and the remainder deposited in the oceans, and Russia.

2.5 Global and European nitrogen budget

Globally, it is estimated that about 57% of anthropogenic nitrogen fixation results from the manufacture of nitrogen-containing fertilizers, 29% from cultivation of nitrogen-fixing crops, and 14% from burning fossil fuels (see Table 2.3, Erisman *et al.*, 2005). Fixation occurs in marine systems

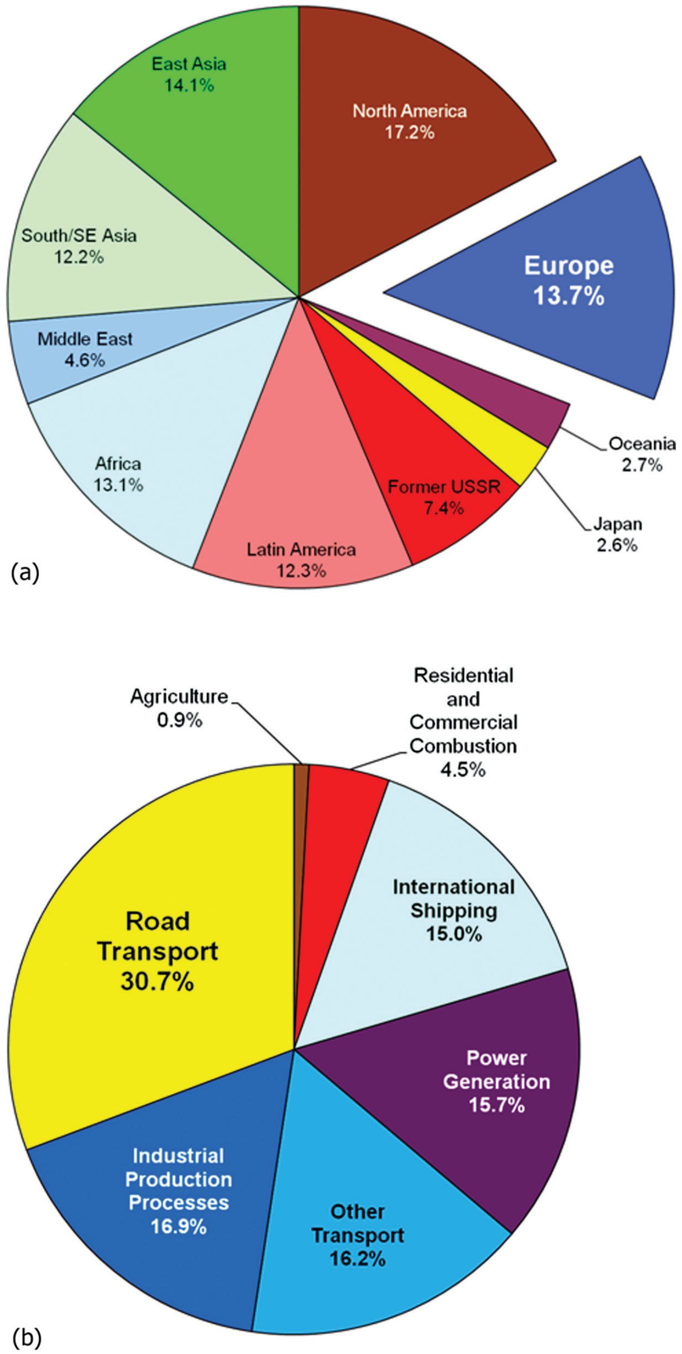

Figure 2.9 NO_x contribution by different regions of the world (a) and different sources (b), based on EDGAR data for the year 2000 (EDGAR, 2010).

as well, but those rates are highly uncertain. Van Egmond *et al.* (2002) presented the estimated input and output flows for Europe. Updated values for Europe are presented by Leip *et al.*, 2011 (Chapter 16 this volume). The export is lower than the import of N_r. The remaining part is stock increase in vegetation, soils and water, but the largest part is denitrified to the atmosphere.

The atmospheric emissions consist of oxidized and reduced forms of N. The total NO_x emission in EU27 is currently 11 Mton NO_x and its distribution is shown in Simpson *et al.*, 2011 (Chapter 14 this volume). The total emissions of reduced nitrogen to the atmosphere amount to 4 Mton NH_3 (EU27) (Hertel *et al.*, 2011, Chapter 9 this volume). The total emission of oxidized and reduced nitrogen in Europe is not much different, but the spatial and temporal variation is different and the chemical behaviour in the atmosphere is different. Therefore they have a different footprint, with oxidized nitrogen being transported over much larger distances. As a result, NO_x emissions are much more of a global problem (also linked to O_3 background) than the NH_3 emissions.

Total deposition of oxidized nitrogen in Europe (EU27) in 2006 was 1.7 Mton N and for reduced nitrogen 2.3 Mton, the total nitrogen deposition being 4 Mton N per year and the distribution is given in Hertel *et al.*, 2011 (Chapter 9, this volume). The highest nitrogen deposition occurs in central Europe. Deposition of oxidized N is significant in the UK, the Netherlands, Germany and the Po valley (Italy). In addition, these areas present the highest deposition of reduced N, as a result of the intensive livestock production. In the regions of Europe with intensive agriculture spatial variability of nitrogen emissions and the deposition climate is high.

More than half of the fertilizer that is produced in Europe is exported as fertilizers or agricultural products, mainly to the USA and Asian countries. Most of the nitrogen emitted into the air is deposited again on the land surface even though about one third is exported outside Europe, mainly as NO_x and particles. Riverine transport to outside Europe is somewhat higher than atmospheric transport. The difference in the nitrogen balance in different regions can be derived from Figure 2.11, where the nitrogen budgets for the continents are given (Galloway *et al.*, 2004). There are large differences, with BNF being the largest source in Latin America, Oceania and Africa and in all other regions it is fertilizer input. Also the major outputs differ: in Africa, Asia and Latin America most output is through riverine transport, in Oceania most of the output occurs via the atmosphere and in Europe and Northern America most of the N_r is exported through fertilizer and products.

Global and European N_r production in 2000 and the different fluxes are presented in Table 2.3. Based on the numbers in Table 2.3 it can be calculated that the nitrogen efficiency, defined as the product output divided by the total inputs, in agriculture in Europe in the year 2000 was 36% compared to a global average of 50%. The nitrogen consumption by humans in Europe was 5.5 kg N per person compared to 4.3 kg N per person globally. The gross input necessary for consumption in Europe was 75 kg N per person compared to 45 kg N per person globally. The agricultural system and consumption in Europe therefore uses and wastes much more nitrogen than the global system.

Apart from the creation of new N_r, there is a fair amount that is transported over the globe and within Europe, concentrating N_r in certain regions where its use is not always efficient. In 2005, ~45 Tg N of the ~190 Tg N of N_r created was traded internationally (Figure 2.12). Over the preceding decade, global trade of N-commodities increased twofold faster than the rate of N_r fixation (Galloway *et al.*, 2008).

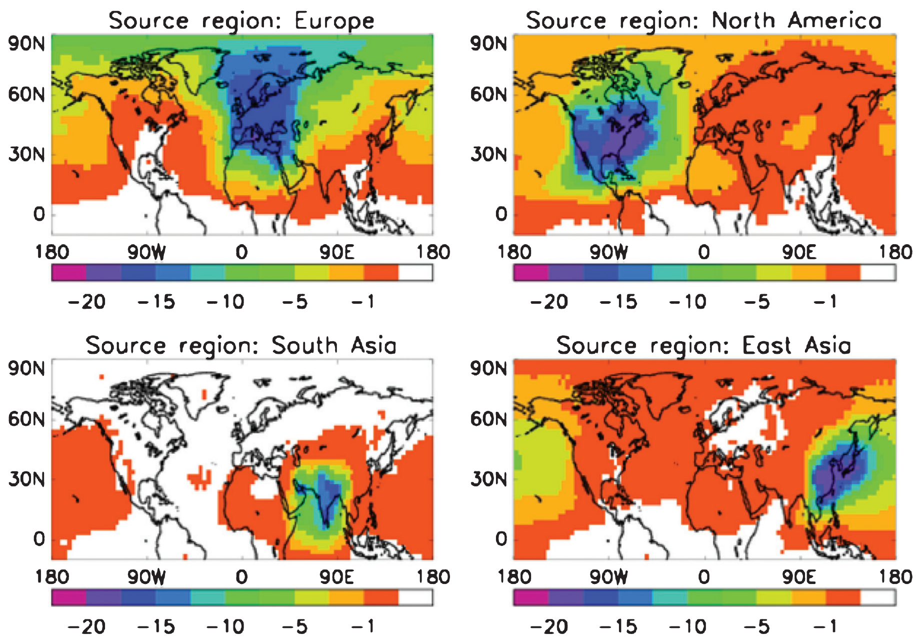

Figure 2.10 The transport distance of NO_x emissions of Europe and North America over the globe. Shown is the percentage change in deposition of NO_y in each NO_x emission perturbation experiment relative to the control run, using multi-model annual mean deposition fluxes (Sanderson *et al.*, 2008).

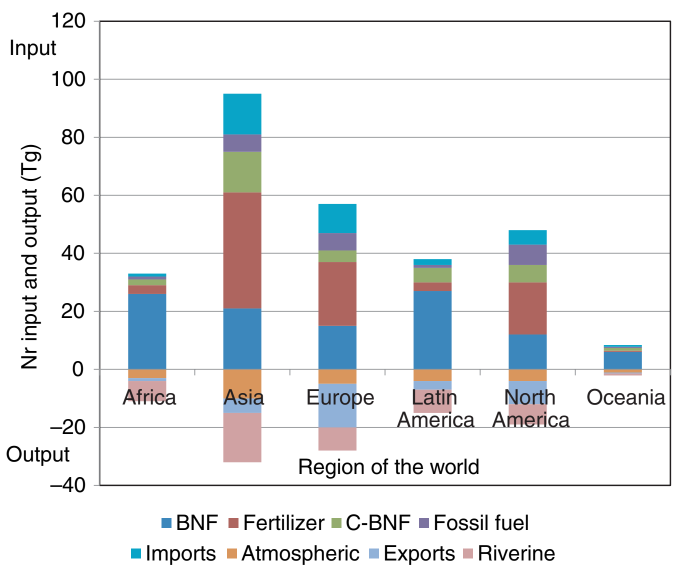

Figure 2.11 Nitrogen input and output (Tg) for different regions of the world (data from Galloway *et al.*, 2004).

Unlike aquatic or atmospheric transport, where N_r is diluted to varying degrees, commerce typically results in injection of N_r to ecosystems in more concentrated doses. Regions that consume N-containing products, such as meat and milk, are often far removed from regions that produce the commodity and thus do not have to bear the environmental cost of the production.

Table 2.3 Global and European current inputs of N to the biosphere and per person. In brackets: the percentage of the total budget (Erisman *et al.*, 2005)

	Global	Europe	Global	Europe
	Tg N (%)	Tg N (%)	Tg N per person	Tg N per person
Biological N fixation	90 (24)	14.8 (28)	15.9	55.0
Lightning	5 (1)	0.1 (0)	0.9	0.4
Total	*95 (25)*	*14.9 (28)*	*16.8*	***55.4***
Haber–Bosch N fertilizer & industry	85 (23)	21.6 (41)	15.0	80.3
Biological N fixation in agriculture	33 (9)	3.9 (7)	5.8	14.5
Animal feed imports	—	7.6 (14)		28.3
Combustion in industry and transportation	21 (6)	6.1 (11)	3.7	22.7
Total	*140 (37)*	*39.2 (74)*	*24.5*	***145.7***
Natural N fixation in oceans	140 (37)	—	24.7	
Total	*375*	*53.2*	***66.1***	***201.1***

2.6 Consequences of the nitrogen cascade

There are many benefits of nitrogen, especially through the Haber–Bosch production. These are discussed by Jensen *et al.* 2011 (in Chapter 3, this volume). Here the focus is on the adverse effects in Europe in the global context. Nitrogen in its various chemical forms plays a major role in a great number of environmental issues (see Box 2.2). It contributes to acidification and eutrophication of soil, groundwater and surface waters, decreasing ecosystem vitality and biodiversity and causing groundwater pollution through nitrate and aluminium leaching. Nitrogen compounds play an important role in carbon sequestration, global change, and formation of ozone, oxidants and aerosols, potentially posing a threat to human health and affecting visibility. Each of the emissions takes part in the cycling of N causing a number of different effects with its consequent linkages. For example, reactive N emitted to the atmosphere from fossil fuel combustion, in sequence can cause tropospheric ozone levels to increase, visibility to decrease and atmospheric acidity to increase. Once deposited from the atmosphere, reactive N can acidify soils and waters, over-fertilize forests, grassland and coastal ecosystems, and can then be re-emitted to the atmosphere as nitrous oxide contributing to global warming and stratospheric ozone depletion. The environmental changes will continue as long as N_r remains in circulation, for reactive N once created, and then lost to the environment, can be transported to any part of the Earth system, no matter where it was introduced. This sequence of effects has been termed the nitrogen cascade. In principle every pollutant can cause a cascade of effects, however nitrogen stands out because it can occur in many very mobile compounds that can cause a wide range of effects.

Box 2.2 Most important adverse effects of reactive nitrogen (modified from Cowling *et al.*, 1998)

Direct effects on humans

Respiratory disease in people caused by exposure to high concentrations of:

- ozone
- other photochemical oxidants
- fine particulate aerosol
- (on rare occasions) direct toxicity of NO_2

Nitrate contamination of drinking water

Increase allergenic pollen production, and several parasitic and infectious human diseases

Blooms of toxic algae and decreased swimability of water bodies

Direct effects on ecosystems

Ozone damage to crops, forests, and natural ecosystems

Acidification effects on forests, soils, ground waters, and aquatic ecosystems

Eutrophication of freshwater lakes and coastal ecosystems inducing hypoxia

Nitrogen saturation of forest soils

Biodiversity impacts on terrestrial and aquatic ecosystems

Inducing damage by plagues and diseases

Effects on other societal values

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 NO_2 from high-altitude aircraft

Global climate change induced by emissions of N_2O

Regional climate change induced by aerosol cooling

Enhanced deterioration of archaeological artefacts

2.7 Effects of nitrogen on the European environment

While some environmental problems are strictly local, like soil and groundwater pollution or exposure to high concentrations, N-related problems include the regional to global scales. The emissions of N_2O readily spread across the atmosphere and have a global contribution. NO_x has a continental character and can be transported over long distances between continents; NH_3 is also continental but less than NO_x and has smaller intercontinental exchange. The scale of N problems in estuaries and coastal seas depends on the extent of the river basin feeding them. The scales are important for the abatement strategy.

2.7.1 Nitrogen leaching in soil and groundwater

Water quality is a major concern throughout Europe and other regions of the globe. Nitrate pollution of groundwater poses a recognized risk for its use as drinking water, while eutrophication of surface water due to excessive nutrient loads can lead to algal growth, oxygen deficiencies, and fish kills. Agriculture puts the largest pressure on groundwater and also on surface water pollution (EEA, 2005a). During the 1990s the nitrate concentrations slightly decreased in some European rivers, while they have remained constant in groundwater and high in some regions. Although some improvements have been carried out to reduce the nutrient input from wastewater discharge, diffuse pollution of agricultural origin remains a major threat for waters in the EU (EEA, 2005a). In the period 2000–2003, in EU15 nearly 40% of the groundwater monitoring stations (average values) exceed 25 mg NO_3/l, and almost 50% of the surface water monitoring stations presents values greater than 10 mg NO_3/l (EC, 2007a). These values are based on the information reported by EU Member States, and they are affected by the inhomogeneous distribution of sampling stations. The European Community Nitrates Directive (Council Directive 91/676/EEC) aims to control N losses and requires Member States of the European Union to identify areas contributing to N pollution of groundwater and surface water (EC, 2007a). In these areas agriculture may also be restricted. For example, the application of fertilizers should balance the needs of the crops, and the application of manure should not exceed 170 kg N/ha. Nitrate concentrations in drinking water should not exceed 50 mg/l (EC Drinking Water Directive, EC, 2007a).

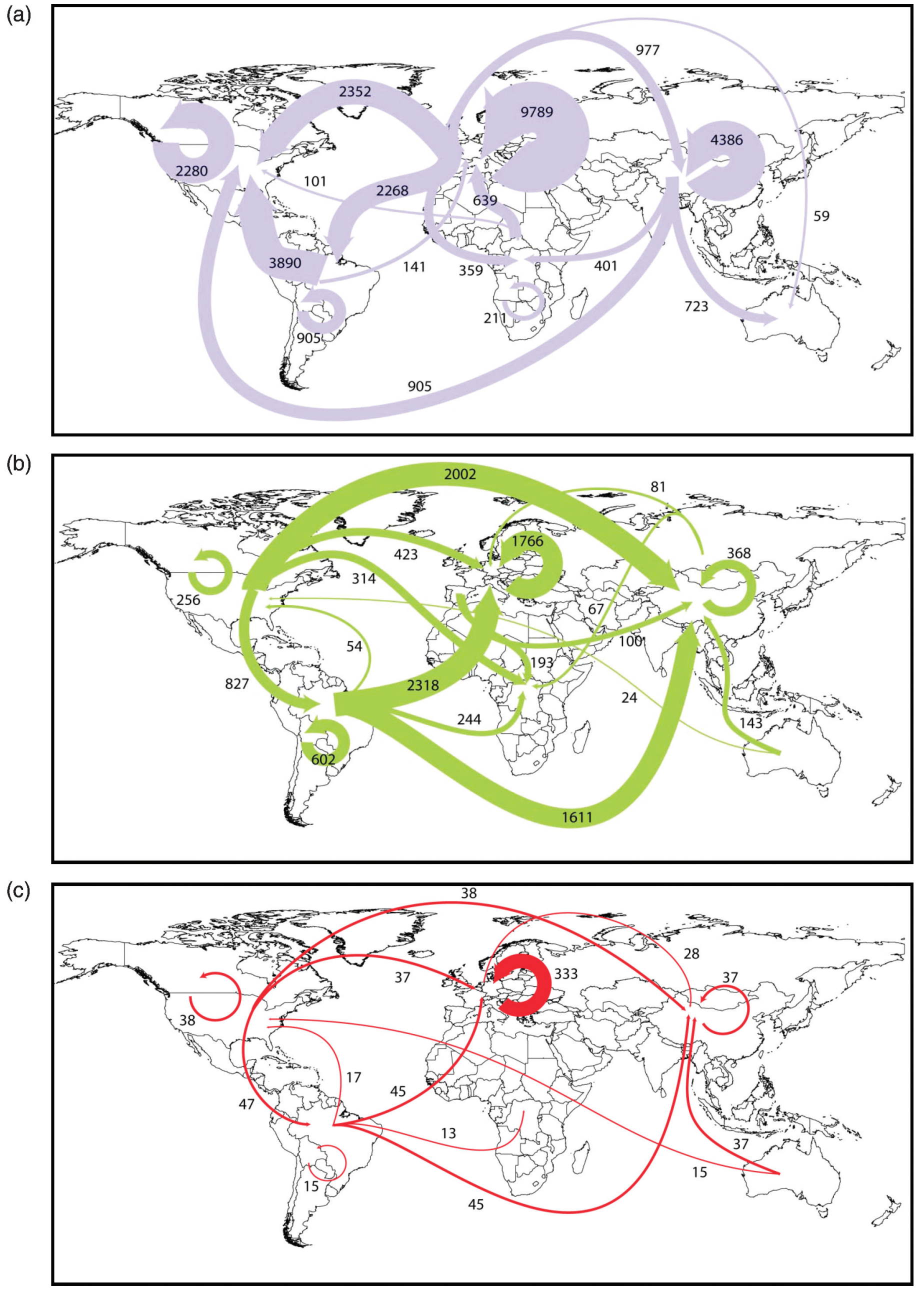

Figure 2.12 Amounts of N contained in internationally traded products: (A) fertilizer (31 Tg N), (B) grain (12 Tg N), and (C) meat (0.8 Tg N). Data are for 2004 and are in units of thousand of tons. Minimum requirements for drawing a line are 50 000 tons N, 20 000 tons N and 10 000 tons N for fertilizer, grain and meat respectively (UNEP, 2007; Galloway *et al*., 2008).

Exceedances of the nitrate standards are a common problem across Europe, particularly from shallow wells. It is often a problem in rural water supplies. For example, in Belgium 29% of 5000 wells examined had concentrations in excess of the limit value (OECD, 1997) and in Bulgaria it was estimated that, in the early 1990s, up to 80% of the population was exposed to nitrate concentrations that exceeded the limit value (OECD, 1995). In about a third of the groundwater bodies for which information was available nitrate concentrations exceeded the recommended limit. In general, there has been no substantial improvement in the nitrate situation in European groundwater and hence nitrate pollution remains a significant problem (EEA, 2003). The same is true for other parts of the globe, where nitrogen leaching to groundwater and subsequent riverine and watershed increase in nitrates are recognized as an increasing issue (UNEP, 2007).

2.7.2 Wastewater discharge to surface water

In surface waters, the overall trend is that N concentrations have remained relatively stable throughout the 1990s and are highest in those Western European countries where agriculture is most intensive. Also in Europe's seas the nitrate (nitrogen) concentrations have generally remained stable. A few stations in the Baltic, Black and North Seas, though, have demonstrated a slight decrease in nitrate concentrations (EEA, 2003).

Although the most important, agriculture is not the only contributor of nitrogen in European streams. Other inputs of nitrogen come from the atmospheric deposition, household scattered dwellings, and from the direct discharges from sewerage, wastewater treatment plants and industries. The nitrogen input from direct discharges from sewerage, wastewater treatment plants and industries constitute a threat for surface waters. According to what is reported by Member States, in the year 2000 in EU15, about 80% of wastewaters received adequate treatment before reaching the water bodies and the number of 'big cities' (agglomeration with waste water discharges greater than 150 000 population equivalents) without sufficient treatments has declined from 27 in 1999 to 17 in 2003 (EC, 2007b). However, the percentage of population connected to wastewaters treatment in Southern and Eastern Europe and in the accession countries is relatively low (EEA, 2005a) and information is often missing or not easily accessible (Mulligan *et al.*, 2006). The load can be estimated based on the map of population density, emissions factor per population equivalent, and national statistics of population connected to sewerage system and level of wastewater treatment (Grizzetti and Bouraoui, 2006) and is given in Billen *et al.*, 2011 (Chapter 13 this volume) and Grizzetti *et al.*, 2011 (Chapter 17 this volume).

According to this estimate, the regions affected by higher nitrogen losses to surface waters include Belgium, the Netherlands, the Po valley (Italy), the Brittany region (France). Most of these areas are already totally or partially designated as Nitrates Vulnerable Zones to meet the EU Nitrate Directive.

The EU makes progress in controlling point sources of pollution from industry and households through wastewater treatment. The Urban Waste Water Treatment Directive aims at 75% removal of the N load to the treatment plants in sensitive areas. However, by the end of 1998, still some 37 out of 527 cities had no treatment at all, including Brussels, Milan and Porto, while 57 others, including Aberdeen, Athens, Barcelona, Dublin, Florence, Liège and Marseille, were discharging a large part of their effluents untreated. The situation is generally improving and some of these cities made the necessary investments (EEA, 2005a).

The GEMSTAT database of UNEP (www.gemstat.org) contains currently over 600 000 stations measuring nutrients in ground, surface and estuary waters. The data is used in different assessments to determine the watershed nutrient loads. In order to compare the European situation with the rest of the world the Nitrogen loading indicator is used (Figure 2.13). This indicator provides a measure of potential water pollution by explicitly mapping out the extent of both natural and anthropogenic nitrogen loading to the land and aquatic systems (Green *et al.*, 2004). Global, continental, regional, and coastline-specific estimates of nitrogen loadings onto the continental land mass are derived by applying a mass balance assessment of nitrogen loads to the landscape providing an accounting of nitrogen sources, uptake, transport and leakages to terrestrial and riverine systems. In Europe the water pollution from nitrogen is mainly the result of fertilizer and lifestock production with the latter being dominant (Figure 2.13). Only in India and the southern parts of Latin America lifestock production is the dominant contributor. In Northern America fertilizers is dominant and in the rest of the world fixation dominates.

2.7.3 Eutrophication and acidification of terrestrial ecosystems

The deposition of N_r is far above levels that the ecosystems are able to absorb and handle without adverse consequences for its vitality. Many ecosystems have changed from N limited systems to N saturated systems where N is not limiting any longer. Heathlands, e.g. in the Netherlands and Denmark, have turned into grasslands and forests once dominated by blueberries and lingon-berries have now a large occurrence of grasses (Bobbink *et al.*, 2010). Long-term high N deposition loads to ecosystems will also lead to N leaching into groundwater and surface water runoffs. A substantial fraction, not uncommonly in the order of 30% of the deposition may in this way be leached and transported to marine areas and contribute significantly to the marine eutrophication (EEA, 2005a). Direct deposition of nitrogen to sea surfaces is also of significant importance for the overall N input to marine ecosystems (see Voß *et al.*, 2011, Chapter 8 this volume). About one third of the overall N input to the Baltic Sea, which is suffering from severe algae blooms every summer, is caused by N deposition (Billen *et al.*, 2011, Chapter 13 this volume).

The issues of acidification and eutrophication have been effectively, but not sufficiently, tackled by policy measures in the EU since the 1980s (EEA, 2005a). Several international agreements under the Convention on Long-range Transboundary Air Pollution (LRTAP) have been reached to reduce emissions. With respect to air pollution control, the EU has adopted emission and fuel quality standards for its Member States. In addition, many European countries have adopted national standards and other types of regulation reflecting the seriousness of pollution and national environmental quality priorities (Oenema *et al.*, 2011, Chapter 4 this volume).

An impact indicator that has been extensively used in Europe to assess the policy responses is the proportion of ecosystems where 'critical loads' of acidity and eutrophication are exceeded. The critical loads and critical levels refer to thresholds, which can serve as a tool to assess the occurrence of effects in natural ecosystems due to acid deposition. A critical load is a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects do not occur according to current knowledge. Critical loads are ecosystem specific and show a large variation over Europe. Current European policies are anticipated to substantially improve the environmental conditions for Europe's nature. The area affected by acidification is expected to decrease from 25% in 1990 to less than 5% in 2010. The eutrophication indicator shows the percentage of unprotected ecosystems improving from 55% to 41% (Hettelingh *et al.*, 2008). This underlines that eutrophication is a far larger problem in Europe than acidification and needs further abatement/attention. Critical loads are developed for other areas in the world, such as Asia and the USA.

2.7.4 Eutrophication of marine ecosystems

Pollution of coastal seas occurs by the influx of nitrates and DON (dissolved organic N) through – often transboundary – rivers and by atmospheric deposition. Spatially explicit,

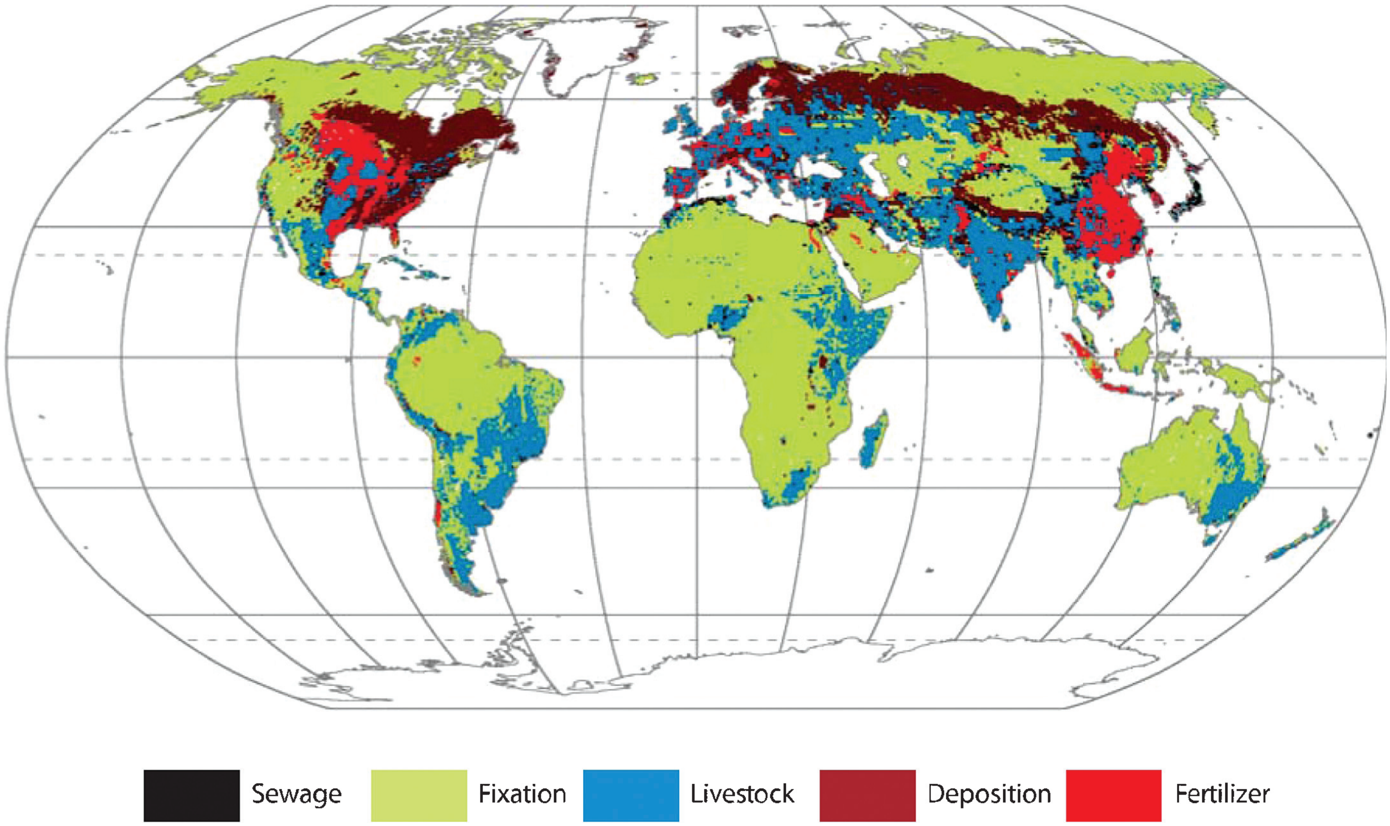

Figure 2.13 Nitrogen loading onto the land mass and aquatic systems as a source for delivery to the coastal zone; a measure of potential water pollution. Total and inorganic nitrogen loads as deposition, fixation, fertilizer, livestock loads, human loads and total distributed nitrogen to the land and aquatic system. Map prepared by Water Systems Analysis Group, University of New Hampshire.

quantitative assessments of N inputs to coastal waters and marine ecosystems are not developed in most large-scale assessment reports. However, there are published studies of N inputs for individual estuaries in some regions as well as spatially explicit regional and global river N export models that provide considerable information. One of the first global syntheses of measurements of river nitrogen export, by nitrogen form, was by Meybeck (1982). Since then, several databases have been created documenting measured nitrogen export from rivers for specific regions and globally (Peierls *et al*., 1991; Meybeck and Ragu, 1995; Smith *et al*., 2003; LOICZ; UNEP/GPA, 2006). The creation of these databases has highlighted the large variation among rivers, both in terms of nitrogen flux density (kg N/km watershed/yr) and nitrogen load (kg N/watershed/yr), and made it possible to develop a more refined understanding of patterns of nitrogen export at local, regional and global scales. There is considerable spatial variation at local, regional and global scales in the magnitude of nitrogen loading (amount per watershed) as well as nitrogen yield (amount per unit area of watershed) from watersheds to coastal systems (Figure 2.14), with many hotspots around the world.

It is clear from these maps that Europe forms a hot spot in the world with about the highest increases in nitrogen transport to the river mouth.

These hot spots are the result of the growing nitrogen surplus, especially in agriculture. The source contribution varies very much among the different river deltas. Also the environmental influence on transboundary outputs is variable.

The amount of nutrients entering the oceans tend to vary significantly over time and from region to region (see Figure 2.15; UNEP/GPA, 2006), as do the actions to control the problem. Nutrient enrichment between 1960 and 1980 in the developed regions of Europe, North America, Asia and Oceania resulted in major changes in coastal ecosystems. Estuaries and bays are most affected, but eutrophication is also apparent over large areas of semi-enclosed seas, including the Baltic, North Adriatic and Black Seas in Europe, the Gulf of Mexico and the Seto Inland Sea in Japan (UNEP/GPA, 2006).

2.7.5 Global warming: N_2O emissions and other effects of nitrogen

Although the absolute quantities are small, the increasing N_2O production plays an important role in the global warming issue since N_2O is a powerful greenhouse gas. Europe's emission is estimated at 0.8 Mton N_2O-N – 65% of which is due to ecosystem denitrification (EDGAR, 2010). The greenhouse gas targets for Europe defined in the Kyoto Protocol is a reduction of 8% compared to 1990 (EDGAR, 2010). These targets have to be met during the period 2008–2012. Europe contributes 10.8% of the global N_2O emissions. Nitrous oxide has emerged as such a major GHG issue from agriculture and there has been some

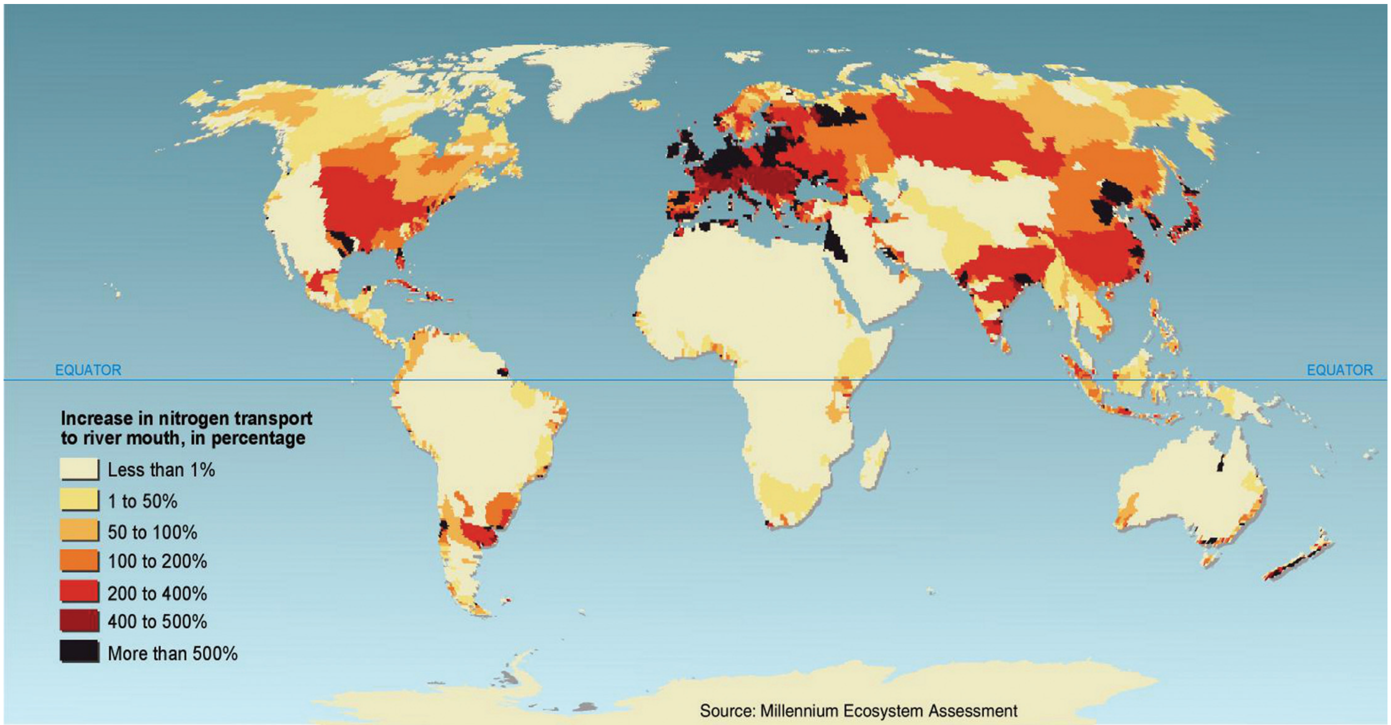

Figure 2.14 Increase in nitrogen transport to river mouth between 1980 and 2000 (Reid *et al.*, 2005).

debate about the validity of the emission factors used within IPCC (see e.g. Crutzen *et al.*, 2008). More work is needed to provide consistent factors and use them for abatement strategies (Davidson, 2009).

Apart from N_2O there are indications that other chemical forms of nitrogen are emitted that could have a major impact on the global warming potential (GWP). Nitrogen trifluoride is about 17 000 times more potent than carbon dioxide. Its estimated worldwide release into the atmosphere this year is equivalent to the total global-warming emissions from Austria (Wen-Tien Tsai, 2008).

Other impacts of nitrogen on the GHG emissions and the net GWP include the effect on carbon sequestration in waters, soils and plants; the effect on aerosol formation causing a direct and indirect cooling effect (through clouds) on the radiation balance and the effect on the emissions of other GHG, such as methane. De Vries *et al.* (2008) for example estimated that the effect of nitrogen deposition on the net GHG emissions for European forests yielded a net reduction in GWP through the additional sequestration of CO_2. Recent debate has focused on the response of forests to this effect. The reported amounts of carbon stored per kg N added show a large range from 40 to 400 kg C per kg N deposition (Högberg, 2007; Magnani *et al.*, 2007; De Vries *et al.*, 2008; Reay *et al.*, 2008). Meanwhile, further efforts are being directed to understand the overall effect of N_r on greenhouse gas balance, including the interactions with nitrous oxide, methane, ozone and aerosols (see Butterbach-Bahl *et al.*, 2011, Chapter 19 this volume).

The nitrogen cycle links with several other cycles, the most important being phosphorus and carbon, acidity and sulphur. For some issues, the complex role of the nitrogen cycle is well appreciated and discussed in Sutton *et al.*, 2011 (Chapter 5 this volume). Climate change is one example where these multifaceted interactions are understood, as the roles of N_2O and tropospheric O_3 (enhanced due to increased NO_x emissions) are well understood as a contributing factor in greenhouse gas emissions. However, for other issues, there is a poor understanding of the role of the nitrogen cycle, including its place in the process of carbon sequestration and the interactions among the nitrogen, carbon and phosphorus cycles (Gruber and Galloway, 2008).

2.7.6 Effects of nitrogen on human health

Excess nitrogen inputs to land, air and water can influence human health and welfare in both direct and indirect ways. Some such connections are well known. For example, exposure to high levels of NO_x in urban areas or along roads cause human health problems, N-driven increases in tropospheric O_3 pose direct health threats to humans (Levy *et al.*, 2005) and cause substantial losses in agricultural productivity (Reilly *et al.*, 2007); the combination of these effects likely has a multi-billion dollar cost. N_r in the air also contributes to the formation of fine particulates, which are in turn a substantial health threat in polluted regions such as urban areas (Wolfe and Patz, 2002). Excess nitrate in drinking water may also pose risks for some types of cancer and reproductive problems, though epidemiological data on these links remains too sparse to draw firm conclusions and there is considerable debate and a lack of consensus on the interpretation of medical evidence (van Grinsven *et al.*, 2006; Ward *et al.*, 2005). Nitrate intake through drinking water is only part of the total dietary intake; with the main dietary intake of nitrate for many people being from vegetables and meats.

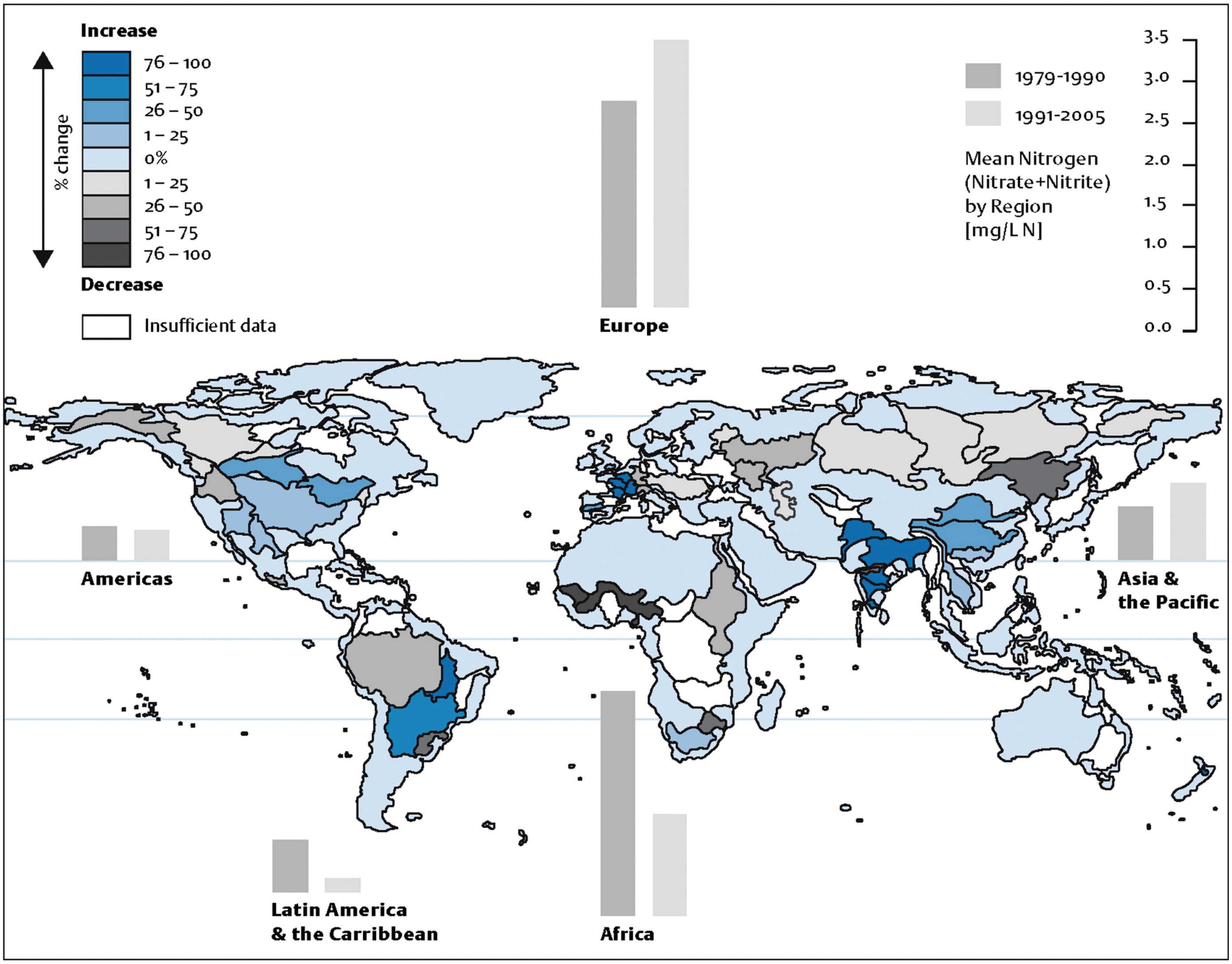

Figure 2.15 Changes in nitrogen concentrations for significant global watersheds (percentage) and by region (concentration): 1979–1990 and 1991–2005 (UNEP GEMS, 2006).

Jakszyn and González (2006) concluded that: '*The available evidence supports a positive association between nitrite and nitrosamine intake and gastric cancer, between meat and processed meat intake and gastric and oesophagal cancer and between preserved fish, vegetable and smoked food intake and gastric cancer, but is not conclusive*'. Van Grinsven *et al*. (2006) concluded that there are both experimental and epidemiologic studies that indicate possible chronic health effects associated with consumption of elevated levels of drinking water nitrate, although there is no consistency across all studies. Therefore, the uncertainties associated with risk estimates are considerable, and hamper the design of cost-effective specific preventive measures for sensitive subpopulations or regions. Moreover, the enhanced risk of nitroso compounds (NOC)-induced toxicity as a result of high drinking water nitrate in combination with other individual risk factors, such as inflammatory diseases, emphasizes the importance of changing the limit values only when such risks have been carefully evaluated. At this moment this is not the case. Likewise, uncertainties do not allow an estimate of the health losses related to methemoglobinemia due to drinking water nitrate. Evidence is emerging for possible benefits of nitrate/nitrite as a potential pharmacological tool for cardiovascular health (Wink and Paolocci, 2008).

Although it is not yet possible to estimate net health loss due to nitrate, it is possible to make estimates of potential exposure. Based on data reported to the European Commission about the implementation of the Drinking Water Directive and data on the present nitrate levels in groundwater at drinking water extraction depths, the population in ten west European countries potentially exposed to drinking water exceeding the 50 mg/l nitrate standard, or the 3 mg/l nitrite standard, was estimated at over 9 million (2.7%).

Other feedbacks remain poorly known but are potentially important and costly, including the possible effects of excess nutrients on human infectious and parasitic diseases (Townsend *et al*., 2003). Diseases that show signs of change following N (and/or P) caused eutrophication include malaria, West Nile virus, cholera and schistosomiasis (Townsend and McKenzie, 2007). These effects are more relevant for other parts in the world. In Europe and in parts of Asia and the USA the exposure of humans to NO_x and PM and the intake of NO_3 is the main threat. Nonetheless, the facts that tropical regions will experience marked increases in nutrient loading and also contain the greatest diversity of human parasitic and infectious diseases highlights the need to understand these connections (Townsend and McKenzie, 2007). Finally, it is important to note that a healthy immune system requires adequate nutrition, thus one of the most critical links between fixed nitrogen and many tropical diseases may be via its greater supply in fertilizer to undernourished regions (Sanchez and Swaminathan, 2005).

2.7.7 Conclusions

The nitrogen cascade effect is expected to be relevant in Europe. Through long-range atmospheric transport, river transport or groundwater transport the effects extend from regional to continental (acidification, eutrophication, carbon sequestration, aerosols) and even global dimensions (N_2O). The cascade depends on the nitrogen status of a region: this is defined as the amount of excess nitrogen in the system (or region) causing effects at different levels in the cascade of N causing a number of different effects. If the nitrogen excess increases, the number of effects in the cascade likely will increase (the cascade length increases). At the same time the area that is affected by nitrogen pollution increases (higher contribution to long-range transport or N_2O emissions). While the linkages in the cascade effect still require to be quantified at the different scales, the available information already highlights its importance. Only at the beginning of the cascade the form of N_r is of importance. In the next stages of the cascade, it will be transformed either in the oxidized or reduced form and the origin is of little importance, whether it comes through the atmosphere or directly from manure or through mineralization or nitrification in the soil.

Within the global context, Europe can be regarded as an excess nitrogen area, in contrast to developing regions such as Africa where nitrogen is limited in food production. Europe was one of the first regions where nitrogen became an environmental issue, with hotspots in the Netherlands, Denmark, France and Italy. Other areas in the world currently experience similar issues, such as parts of the USA, China, India and Latin America. It is expected that the nitrogen situation will become worse. Knowledge on the European nitrogen fixation rates, the transport through environment and cascading effects as described in this European Nitrogen Assessment might serve as input for these other regions. Of enormous significance is that excess nitrogen is linked to many of the major global and regional challenges that policymakers face today, such as globalization, strong development of growing economies, increase in human population, political stress, environmental aspects, etc. A prerequisite to reducing these problems is the development of a sound scientific base to help identify policy options. Furthermore, these issues need be recognized at scientific and political levels. The focus on food production in developed and developing countries should take environmental impacts of nitrogen into consideration. For the future it is envisaged that the focus on the production of biofuels and the increased use of fertilizer will yield similar issues. The basis for a successful approach was laid down in the Nanjing Declaration on nitrogen management (Erisman, 2004). A comprehensive overview of N-related policies in Europe is given in (Oenema *et al*., 2011, Chapter 4 this volume).

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References

Billen, G., Silvestre, M., Grizzetti, B. *et al*. (2011). Nitrogen flows from European watersheds to coastal marine waters. In: *The European Nitrogen Assessment*, ed. M. A. Sutton, C. M. Howard, J. W. Erisman *et al.* Cambridge University Press.

Bobbink, R., Hicks, K., Galloway, J. *et al*. (2010). Global assessment of nitrogen deposition effects on terrestrial plant diversity: a synthesis, *Ecological Applications*, **20**, 30–59.

Buijsman, E. (1986). *Historical Trend in the Ammonia Emission in Europe (1870–1980)*, Report R-86-9, Institute for Meteorology and Oceanography, University of Utrecht, The Netherlands.

Butterbach-Bahl, K., Nemitz, E., Zaehle, S. *et al*. (2011). Nitrogen as a threat to the European greenhouse balance. In: *The European Nitrogen Assessment*, ed. M. A. Sutton, C. M. Howard, J. W. Erisman *et al*. Cambridge University Press.

Campling P., Terres J. M., Walle S. V., Orshoven J. V. and Crouzet, P. (2005). Estimation of nitrogen balances from agriculture for EU-15: spatialisation of estimates to river basins using the CORINE Land Cover. *Physics and Chemistry of the Earth*, **30**, 25–34.

Cowling, E., Erisman, J. W., Smeulders, S. M., Holman, S. C. and Nicholson, B. M. (1998). Optimizing air quality management in Europe and North America: justification for integrated management of both oxidised and reduced forms of nitrogen. *Environmental Pollution,* **102**, 599–608.

Crutzen, P., Mosier, A. R., Smith, K. A. and Winiwater, W. (2008). N_2O release from agro-biofuel production negates global warming reduction by replacing fossil fuels. *Atmospheric Chemistry Physics*, **8**, 389–395.

Davidson, E. (2009). The contribution of manure and fertilizer nitrogen to atmospheric nitrous oxide since 1860. *Nature Geoscience,* **2**, 659–662.

De Vries, W., Solberg, S., Dobbertin, M. *et al*. (2008). Ecologically implausible carbon response? *Nature,* **451**, E1–E3.

De Vries, W., Kros, J., Reinds, G. J. *et al*. (2009). INTEGRATOR: a modelling tool for European-wide assessments of nitrogen and greenhouse gas fluxes in response to changes in land cover, land management and climate. Calculation procedures, application methodology and examples of scenario results, Alterra Report (in press), Alterra Wageningen UR: Wageningen, The Netherlands.

De Vries, W., Leip, A., Reinds, G. J. *et al*. (2011). Geographic variation in terrestrial nitrogen budgets across Europe. In: *The European Nitrogen Assessment*, ed. M. A. Sutton, C. M. Howard, J. W. Erisman *et al.* Cambridge University Press.

Dean, A. M. and Bozzelli, J. W. (1999). Combustion chemistry of nitrogen. In: *Gas-Phase Combustion Chemistry*, ed. W. C. Gardiner, Jr., Springer, New York, pp. 125–341.

Dornburg, V., Faaij, A., Verweij, P. *et al*. (2007). *Biomass Assessment: Global Biomass Potentials and Their Links to Food, Water, Biodiversity, Energy Demand and Economy.* Climate Change Scientific Assessment and Policy Analysis, PBL, The Netherlands.

EDGAR (2010). *Integrated Project of Climate Change and Impact Research: The Mediterranean Environment,* Project No. 036961–CIRCE. http://www.mnp.nl/edgar/model/v32ft2000edgar/

EFMA (2010). *Forecast of Food, Farming and Fertilizer Use in the European Union 2009–2019*. EFMA, Brussels, Belgium (www.fertilizerseurope.com).

Erisman, J. W. (2004). The Nanjing declaration on management of reactive nitrogen. *BioScience*, **54**, 286–287.

Erisman, J. W., Domburg, N., de Vries, W. *et al*. (2005). The Dutch N-cascade in the European perspective. *Science in China*, *Series C, Life Sciences*, **48**, 827–842.

Erisman, J. W., Galloway, J. N., Sutton, M. S., Klimont, Z. and Winiwater, W. (2008). How a century of ammonia synthesis changed the world. *Nature Geoscience*, **1**, 636–639.

Erisman, J. W., van Grinsven, H., Leip, A., Mosier, A. and Bleeker, A. (2009). Nitrogen and biofuels; an overview of the current state of knowledge. *Nutrient Cycling in Agroecosystems*, **86**, 211–223.

European Commission (2007a). COM(2007) 120 final. Report from the Commission to the Council and the European Parliament on implementation of the Council Directive 91/676/EEC concerning the protection of waters against pollution caused by nitrates from agricultural sources for the period 2000–2003. 11 pp.

European Commission (2007b). SEC(2007) 363. Annex to the Communication from the Commission to the European Parliament and the Council 'Towards Sustainable Water Management in the European Union' First stage in the implementation of the Water Framework Directive 2000/60/EC. 4th Commission Report (Executive Summary) on the Implementation of the Urban Waste Water Treatment Directive.

European Environment Agency (EEA) (2003). *Europe's Environment: The Third Assessment.* Environmental Assessment report no. 10. Copenhagen.

European Environment Agency (EEA) (2005a). *The European Environment: State and Outlook 2005*. Copenhagen.

European Environment Agency (EEA) (2005b). *Agriculture and Environment in EU-15: the IRENA Indicator Report*. EEA Report no. 6/2005. Copenhagen.

European Environment Agency (EEA) (2007). *NEC Directive Status report 2006.* Technical report no.15/2007. Copenhagen.

EU (2007). *Biomass Action Plan.* Brussels.

EU (2009). Directive 2009/28/EC of the European Parliament and of the Council of 23 April 2009 on the promotion of the use of energy from renewable sources and amending and subsequently repealing Directives 2001/77/EC and 2003/30/EC.

Fagernäs, L., Johansson, A., Wilén, C. *et al*. (2006). *Bioenergy in Europe: Opportunities and Barriers*. Espoo VTT Tiedotteita, Finland.

FAO (2010a). (http://www.fao.org/docrep/w5146e/w5146e00.htm).

FAO (2010b). (http://www.fao.org/agriculture/lead/tools/densities/en/).

FAO (2010c). (http://faostat.fao.org/faostat).

Galloway, J. N., Aber, J. D., Erisman, J. W. *et al*. (2003). The nitrogen cascade. *BioScience*, **53**, 341–356.

Galloway, J. N., Dentener, F. J., Capone, D. G. *et al*. (2004). Nitrogen cycles: past, present and future. *Biogeochemistry*, **70**, 153–226.

Galloway, J. N., Townsend, A. R., Erisman, J. W. *et al.* (2008). Transformation of the nitrogen cycle: Recent trends, questions, and potential solutions. *Science*, **320**, 889–892.

GEMS Water Triennial Reports (2006). United Nations Environment Programme GEMS/WATER (http://www.cciw.ca/gems/intro.html).

Green, P. A., Vörösmarty, C. J., Meybeck, M. *et al*. (2004). Pre-industrial and contemporary fluxes of nitrogen trough rivers: a global assessment based on topology. *Biogeochemistry*, **68**, 71–105.

Grizzetti, B. and Bouraoui, F. (2006). *Assessment of Nitrogen and Phosphorus Environmental Pressure at European Scale*. Report EUR 22526 EN.

Grizzetti, B., Bouraoui, F., Billen, G. *et al*. (2011). Nitrogen as a threat to European water quality. In: *The European Nitrogen Assessment,* ed. M. A. Sutton, C. M. Howard, J. W. Erisman *et al*. Cambridge University Press.

Gruber, N. and Galloway, J. N. (2008). An earth-system perspective of the global nitrogen cycle. *Nature*, **451**, 293–296.

Hatfield, J. L. and Prueger, J. H. (2004). Nitrogen over-use, under-use, and efficiency. In: *Proceedings of the 4th International Crop Science Congress*, 26 Sept–1 Oct 2004, Brisbane, Australia, CD-ROM, www.cropscience.org.au.

Hertel, O., Reis, S. and Ambelas Skjøth, C. (2011). Nitrogen processes in the atmosphere. In: *The European Nitrogen Assessment*, ed. M. A. Sutton, C. M. Howard, J. W. Erisman *et al.* Cambridge University Press.

Hettelingh, J.-P., Posch, M. and Slootweg, J. (2008). Status of the critical load database and impact assessment. In: *Critical Load, Dynamic Modelling and Impact Assessment in Europe*, ed. J.-P. Hettelingh, M. Posch and J. Slootweg, CCE Status Report 2008, www.pbl.nl/cce.

Högberg, P. (2007). Nitrogen impacts on forest carbon. *Nature*, **474**, 781–782.

Holloway, J. M. and Dahlgren, R. A. (2002). Nitrogen in rock: occurrences and biogeochemical implications. *Global Biogeochemical Cycles*, **16**, 1118.

IPCC (2007). *Fourth Assessment Report on Climate Change: Impacts, Adaptation and Vulnerability for Researchers, Students, Policymakers*. Cambridge University Press.

Jakszyn, P. and González, C. A. (2006). Nitrosamine and related food intake and gastric and oesophagal cancer risk: a systematic review of the epidemiological evidence. *World Journal of Gastroenterology*, **12**, 4296–4303.

Jensen, L. S., Schjoerring, J. K., van der Hoek, K. *et al*. (2011). Benefits of nitrogen for food fibre and industrial production. In: *The European Nitrogen Assessment*, ed. M. A. Sutton, C. M. Howard, J. W. Erisman *et al.* Cambridge University Press.

Leip, A., Achermann, B., Billen, G. *et al*. (2011). Integrating nitrogen fluxes at the European scale. In: *The European Nitrogen Assessment*, ed. M. A. Sutton, C. M. Howard, J. W. Erisman *et al.* Cambridge University Press.

Levy, J. I., Chemerynski, S. M. and Sarnat, J. A. (2005). Ozone exposure and mortality: an empiric bayes metaregression analysis. *Epidemiology*, **16**, 458–468.

Londo, M. and Deurwaarder, E. (2007). Developments in EU biofuels policy related to sustainability: overview and outlook. *Biofuels, Bioproducts and Biorefining*, **1**, 292–302.

Magnani, F., Mencuccini, M., Borghetti, M. *et al.* (2007). The human footprint in the carbon cycle of temperate and boreal forests. *Nature*, **447**, 848–850.

Meybeck, M. (1982). Carbon, nitrogen, and phosphorus transport by world rivers. *American Journal of Science*, **282**, 401–450.

Meybeck, M. and Ragu, A. (1995). *River Discharges to the Ocean: an Assessment of Suspended Solids, Major Ions, and Nutrients*. United Nations Environment Program, Nairobi.

Moïssey Postan, M. and Rich, E. E. (1952). *The Cambridge Economic History of Europe: Trade and Industry in the Middle Ages*, Cambridge University Press.

Mulligan, D., Bouraoui, F., Grizzetti, B., Aloe, A. and Dusart, J. (2006). *An Atlas of Pan-European Data for Investigating the Fate of Agrochemicals in Terrestrial Ecosystems*. Report EUR 22334 EN.

OECD (Organisation for Economic Co-operation and Development) (1995).

OECD (Organisation for Economic Co-operation and Development) (1997).

OECD (Organisation for Economic Co-operation and Development) (2001). *Environmental Indicators for Agriculture Methods and Results*, Volume 3. OECD Publishing, Paris.

OECD (2006). *Key Environmental Indicators*. OECD Environment Directorate, Paris, France. http://www.oecd.org/dataoecd/32/20/31558547.pdf

Oenema, O., Oudendag, D. and Velthof, G. L. (2007). Nutrient losses from manure management in the European Union. *Livestock Science*, **112**, 261–272.

Oenema, O., Bleeker, A., Braathen, N. A. *et al.* (2011). Nitrogen in current European policies. In: *The European Nitrogen Assessment*, ed. M. A. Sutton, C. M. Howard, J. W. Erisman *et al.* Cambridge University Press.

Olson, R. A. and Kurtz, L. T. (1982). Crop nitrogen requirements, utilization and fertilization. In: *Nitrogen in Agricultural Soils*, ed. F. J. Stevenson, pp. 567–599. American Society of Agronomy, Ann Arbor, MI.

Pehnt, M. (2006). Dynamic life cycle assessment (LCA) of renewable energy technologies. *Renewable Energy*, **31**, 55–71.

Peierls, B. L., Caraco, N. F., Pace, M. L. and Cole, J. J. (1991). Human influence on river nitrogen. *Nature*, **350**, 386–387.

Reay, D. S., Dentener, F., Smith, P., Grace, J. and Feely, R. (2008). Global nitrogen deposition and carbon sinks. *Nature Geoscience*, **1**, 430–437.

Reid, W. V., Mooney, H. A., Cropper, A. *et al.* (2005). *Millennium Ecosystem Assessment: Ecosystems and Human Well-Being: Synthesis*. Island Press, Washington, DC.

Reilly, S., Paltsev, B., Felzer, X. *et al.* (2007). Global economic effects of changes in crops, pasture, and forests due to changing climate, carbon dioxide, and ozone. *Energy Policy*, **35**, 5370–5383.

Sanderson, M. G., Dentener, F. J., Fiore, A. M. *et al.* (2008). A multi-model study of the hemispheric transport and deposition of oxidised nitrogen. *Geophysics Research Letters*, 35, L17815.

Sanchez, P. A. and Swaminathan, M. S. (2005). Public health. Cutting world hunger in half. *Science*, **307**, 357–359.

Schlesinger, W. H. (2009). On the fate of anthropogenic nitrogen. *Proceedings of the National Academy of Sciences of the USA*, **104**, 203–208.

Simpson, D., Aas, W., Bartnicki, J. *et al.* (2011). Atmospheric transport and deposition of nitrogen in Europe. In: *The European Nitrogen Assessment*, ed. M. A. Sutton, C. M. Howard, J. W. Erisman *et al.* Cambridge University Press.

Skiba, U., Fowler, D. and Smith, K. A. (1994). Emissions of NO and N_2O from soils. In: *Non-CO_2 Greenhouse Gases*, ed. J. van Ham, L. J. Janssen and R. J. Swart. Kluwwer, Dordrecht, pp. 153–158.

Skiba, U., Fowler, D. and Smith, K. A. (1997). Nitric oxide emissions from agricultural soils in temperate and tropical climates: sources, controls and mitigation options. *Nutrient Cycling in Agroecosystems*, **48**, 139–153.

Smil, V. (2001). *Cycles of Life: Civilization and the Biosphere*. Scientific American Library, New York.

Smil, V. (2004). World history and energy. In: *Encyclopedia of Energy*, ed. C. Clevaland, Volume 6. Elsevier, Amsterdam, pp. 549–561.

Smith, S. V., Swaney, D., Talaue-McManus, L. *et al.* (2003). Humans, hydrology, and the distribution of inorganic nutrient loading to the ocean. *BioScience*, **53**, 235–245.

Steinfeld, H., Gerber, P., Wassenaar, T. *et al.* (2006). *Livestock's Long Shadow: Environmental Issues and Options*, LEAD/FAO, Rome.

Sutton, M. A., Erisman, J. W., Dentener, F. and Möller, D. (2008). Ammonia in the environment: from ancient times to the present. *Environmental Pollution*, **156**, 583–604.

Sutton, M. A., Howard, C. M., Erisman, J. W. *et al.* (2011). The need to integrate nitrogen science and policies. In: *The European Nitrogen Assessment*, ed. M. A. Sutton, C. M. Howard, J. W. Erisman *et al.* Cambridge University Press.

Townsend, A. R., Howarth, R. W., Bazzaz, F. A. *et al.* (2003). Human health effects of a changing global nitrogen cycle. *Frontiers Ecology and Environment*, **1**, 240–246.

Townsend, A. R. and McKenzie, V. J. (2007). Parasitic and infectious disease responses to changing global nutrient cycles. *Ecohealth*, **4**, 384–396.

UNEP/GPA (2006). *The State of the Marine Environment: Trends and processes*. UNEP/GPA, The Hague.

UNEP (2007). *Global Environmental Outlook – 4* . UNEP, Nairobi, Kenya.

van Egmond, K., Bresser, T. and Bouwman, L. (2002). The European nitrogen case. *Ambio,* **31**, 72–78.

van Grinsven, J. J. M., van Schijndel, M. W., Schotten, C. G. J. and van Zeijts, H. (2003). Integrale analyse van stikstofstromen en stikstofbeleid in Nederland: Een nadere verkenning. Integrated analysis of nitrogen flows and nitrogen policy in the Netherlands further explored in Dutch – 81 pp. – 2003 Onderzoeksrapport – RIVM rapport 500003001.

van Grinsven, H. J., Ward, M. H., Benjamin, N. and de Kok, T. M. (2006). Does the evidence about health risks associated with nitrate ingestion warrant an increase of the nitrate standard for drinking water? *Environmental Health: A Global Access Science Source 2006*, **5**, 26.

van Kessel, C. and Hartley, C. (2000). Agricultural management of grain legumes: has it led to an increase in nitrogen fixation? *Field Crops Research*, **65**, 165–181.

Velthof, G., Oudendag, D., Witzke, H. P. *et al.* (2009). Integrated Assessment of Nitrogen Losses from Agriculture in EU-27 using MITERRA-EUROPE. *Journal Environmetal Quality*, **38**, 402–417.

Voss, M., Baker, A. and Bange, H. W. (2011). Nitrogen processes in coastal and marine systems. In: *The European Nitrogen Assessment*, ed. M. A. Sutton, C. M. Howard, J. W. Erisman *et al.* Cambridge University Press.

Ward, M. H., de Kok, T. M., Levallois, P. *et al.* (2005). Workgroup report: drinking-water nitrate and health: recent findings and research needs. *Environmental Health Perspective*, **113**, 1607–1614.

Wen-Tien Tsai (2008). Environmental and health risk analysis of nitrogen trifluoride (NF_3), a toxic and potent greenhouse gas. *Journal of Hazardous Materials*, **159**, 30, 257–263.

WHO (2007). *Protein and Amino Acid Requirements in Human Nutrition : Report of a Joint FAO/WHO/UNU Expert Consultation*, WHO Technical Report Series no. 935. World Health Organization, Geneva.

Wink, D. A. and Paolocci, N. (2008). Mother was right: eat your vegetables and do not spit ! When oral nitrate helps with high blood pressure. *Hypertension,* **51**, 1–3.

Wolfe, A. H. and Patz, J. A. (2002). Reactive nitrogen and human health: acute and long-term implications. *Ambio*, **31**, 120–125.