

Nitrogen as a threat to the European greenhouse balance

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Executive summary

Nature of the problem

- Reactive nitrogen (N_r) is of fundamental importance in biological and chemical processes in the atmosphere–biosphere system, altering the Earth's climate balance in many ways. These include the direct and indirect emissions of nitrous oxide (N_2O), atmospheric N_r deposition and tropospheric ozone formation (O_3), both of which alter the biospheric CO_2 sink, N_r supply effects on CH_4 emissions, and the formation of secondary atmospheric aerosols resulting from the emissions of nitrogen oxides (NO_x) and ammonia (NH_3).
- Human production and release of N_r into the environment is thus expected to have been an important driver of European greenhouse balance. Until now, no assessment has been made of how much of an effect European N_r emissions are having on net warming or cooling.

Approaches

- This chapter summarizes current knowledge of the role of N_r for global warming. Particular attention is given to the consequences of atmospheric N_r emissions. The chapter draws on inventory data and review of the literature to assess the contribution of anthropogenic atmospheric N_r emissions to the overall change in radiative forcing (between 1750 and 2005) that can be attributed to activities in Europe.
- The use of N_r fertilizers has major additional effects on climate balance by allowing increased crop and feed production and larger populations of livestock and humans, but these indirect effects are not assessed here.

Key findings/state of knowledge

- Due to its multiple, complex effects on biospheric and atmospheric processes, the importance of N_r for the European greenhouse gas balance has so far received insufficient attention.
- The main warming effects of European anthropogenic N_r emissions are estimated to be from N_2O (17 (15–19) mW/m^2) and from the reduction in the biospheric CO_2 sink by tropospheric O_3 (4.4 (2.3–6.6) mW/m^2). The main cooling effects are estimated to be from increasing the biospheric CO_2 sink by atmospheric N_r deposition at -19 (-30 to -8) mW/m^2 and by light scattering effects of N_r containing aerosol (-16.5 (-27.5 to -5.5) mW/m^2), in both cases resulting from emissions of NO_x and NH_3 .
- The production of O_3 from European emissions of NO_x is estimated to have a modest warming effect (2.9 (0.3–5.5) mW/m^2), which is largely offset by the cooling effect of O_3 in reducing the atmospheric lifetime of CH_4 (-4.6 (-6.7 to -2.4) mW/m^2), giving an uncertain net warming of $+1.7$ (-6.4 to $+3.1$) mW/m^2 .
- Overall, including all of these terms, European N_r emissions are estimated to have a net cooling effect, with the uncertainty bounds ranging from a substantial cooling effect to a small warming effect (-15.7 (-46.7 to $+15.4$) mW/m^2).

Major uncertainties/challenges

- The largest uncertainties concern the aerosol and N_r fertilization effects, and the estimation of the European contributions within the global context.
- Published estimates suggest that the default N_2O emission factor of 1% used by IPCC for indirect emissions from soils following N_r deposition is too low by at least a factor of two.
- The wider effects of fertilizer N_r in allowing increased biospheric C cycling, food and feed production and populations of livestock and humans are a major uncertainty. Industrial production of N_r can be considered as having permitted increased overall consumption (of food, feed and fuel) with major net warming effects. These interactions remain to be investigated.

Recommendations

- The contribution of anthropogenic alteration of the nitrogen cycle to the radiative balance needs to be specifically accounted for in any greenhouse gas reporting (e.g., UNFCCC).
- Although individual components of N_r emissions have cooling effects, there are many opportunities for ‘smart management’ in linking the N and C cycles. These can help mitigate greenhouse gas emissions, while reducing the other N_r -related environmental threats such as eutrophication, acidification, air quality and human health.

19.1 Introduction and objectives

This chapter aims to characterize how inputs of reactive nitrogen compounds (N_r) to the biosphere have affected the concentration of atmospheric trace substances and particles that are important for the radiative balance of the earth system. Based on our current understanding, and with a specific focus on Europe, this chapter furthermore evaluates how historic, present day and future changes in biospheric N_r inputs have and will feedback on the European greenhouse gas (GHG) balance. By including the additional cooling effects of aerosol, we extend the GHG estimates to assess the overall effect on radiative balance.

The pathways of N_r input to the biosphere and how they are influencing atmospheric composition, and thus the radiative balance, are complex. They involve microbiological, plant-physiological, animal-physiological and physico-chemical processes, as well as manure management, industrial processes or atmospheric chemistry (Figure 19.1).

Sources of anthropogenic N_r additions to the global biosphere are primarily related to fertilizer production, combustion processes, including the transport sector, or cultivation of leguminous plants (Galloway *et al.*, 2004).

Once N_r has entered the biosphere, it can directly or indirectly affect the radiative balance of the earth by various processes. Direct effects are generally related to the formation of N_2O , a greenhouse gas which is approximately 296 times as powerful as CO_2 on a 100-yr timescale and per unit of weight (IPPC, 2007). The dominant source of both, natural or anthropogenic emissions is microbial production by nitrification and denitrification (see Butterbach-Bahl *et al.*, 2011, Chapter 6, this volume).

Indirect effects of N_r additions on the radiative balance involve a multitude of mainly biological processes on the ecosystem scale, but also physicochemical processes in the atmosphere (Figure 19.1), with the most prominent ones being as follows.

- Changes in ecosystem C fluxes and C sequestration, affecting CO_2 exchange.
- Changes in ruminant and ecosystem CH_4 production and consumption.
- Changes in N_2O production and emission.
- Changes in atmospheric chemistry and specifically nitrogen oxides (NO_x), ammonia (NH_3) increasing aerosol formation and associated changes in the oxidative capacity of the troposphere with relevant feedbacks on biospheric processes, e.g., tropospheric ozone (O_3) and plant growth.

The primary effects of N_r inputs (which are easy to understand, but not to quantify) are increased emissions of N trace gases

(N_2O , NH_3 , NO_x) to the atmosphere. The processes driving the biosphere-atmosphere exchange of these compounds, such as nitrification and denitrification (N_2O and NO) or volatilization (NH_3) depend significantly on the availability of N_r in the plant–soil system. Thus, increased N_r inputs to agricultural systems (with livestock farming systems having the highest N_r use intensity in Europe) lead to increased losses of N trace gases (NH_3 , NO_x , N_2O) at the site of N_r input. However, following the cascade of nitrogen downwind or downstream into other ecosystems, N_2O emissions affect a broader regional scale (Davidson, 2009; Oenema *et al.*, 2009). Thus, N_r trace gas emissions from natural and semi-natural terrestrial ecosystems, as well as emissions from water bodies, such as lakes, rivers or coastal waters, need to be considered (Figure 19.1).

As a macro-nutrient, N_r positively affects photosynthesis and thus, the assimilation of atmospheric CO_2 in plant biomass (Figure 19.2) (Liu and Greaver, 2009). Furthermore, N_r can stimulate the growth of the soil microbial community and in particular stimulate low affinity CH_4 oxidation in rice paddies or inhibit high affinity CH_4 oxidation in upland soils (Bodelier and Laanbroek, 2004; Figure 19.2), i.e., the availability of N_r affects the tendency or strength of binding of the enzyme CH_4 -monooxygenase for catalyzing the oxidation of CH_4 to methanol. Even the process of methane production in anaerobic sediments (but also in the enteric fermentation in ruminants) is affected by the addition of N_r , as the stimulation of plant growth is a positive feedback upon rhizodeposition of C compounds and plant litter production, both of which serve as substrates for methanogenesis. Since N_r is highly mobile within the biosphere, it also affects aquatic ecosystems, e.g., with regard to eutrophication and the biosphere–atmosphere exchange of CO_2 and CH_4 or by changing the source strength of coastal waters for N_2O .

Besides these biological processes driving biosphere–atmosphere exchange of CO_2 , CH_4 and N_2O (NO_x), the importance of NH_3 volatilization and industrial processes, as well as soil NO_x emissions for particle formation and tropospheric O_3 concentrations needs to be considered. Similarly, feedback loops need to be addressed for the quantification of the net effect of N_r on the GHG balance. In the case of O_3 , not only is it important as a greenhouse gas, but it also has a detrimental effect on plant productivity and, thus on atmospheric CO_2 removal by terrestrial ecosystems, which needs to be accounted for.

Therefore, to characterize and quantify effects of N_r on the radiative balance at continental to global scales, it is necessary to evaluate the effects of N_r on N_2O -, CO_2 -, CH_4 -exchange as well as on exchange of NO_x - and NH_3 and their consequences for aerosol and O_3 formation.

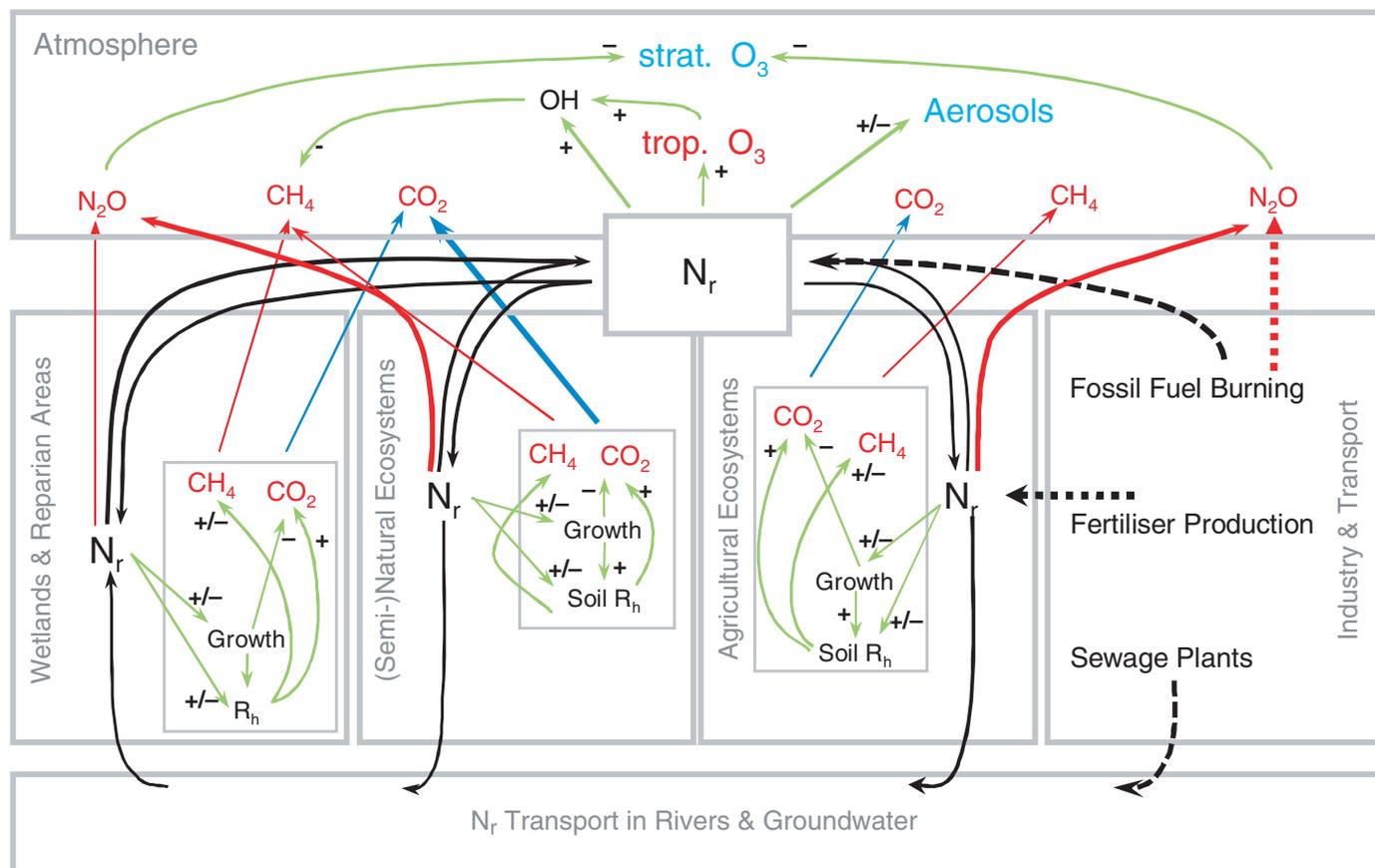


Figure 19.1 Effects of reactive nitrogen (N_r) on various biospheric processes in terrestrial and aquatic ecosystems and on atmospheric chemistry. Feedbacks on the production and consumption of atmospheric compounds directly or indirectly affecting the global radiative balance are indicated by arrows, where the thickness of the arrow gives an indication about the relative importance of a particular process. (Black arrows: N_r fluxes; green arrows: effects (in terms of positive or negative feedbacks); Radiatively active compounds red or fluxes (arrows) of them are marked red (blue) if they tend to increase (decrease) the radiative forcing. Dashed arrows: Direct effects from anthropogenic additions of N_r ; Black arrows: N_r fluxes; Green arrows: Effects (in terms of positive or negative feedback); Red arrows: compounds that increase the radiative forcing (warming); Blue arrows: compounds that decrease the radiative forcing (cooling).)

This chapter estimates, for the first time, the effect of European nitrogen usage on the climate system, taking into account: (i) direct emissions of the long-lived GHG nitrous oxide (N_2O), (ii) the effect of N_r on the biospheric control of other GHGs, and (iii) the effect of N_r emissions on long-lived (e.g., methane, CH_4) and short-lived radiative forcing agents (e.g., O_3 , particles).

Several different metrics are used to quantify the climate effect of a change, e.g., in atmospheric composition or land cover. The most commonly used are the radiative forcing (RF) and the global warming potential (GWP). The RF is the global, annual mean radiative imbalance to the Earth's climate system caused by human activities. The GWP of a trace gas is defined as the instantaneous mass emission of carbon dioxide that gives the same time-integrated radiative forcing as the instantaneous emission of unit mass of another trace gas (such as CH_4 or N_2O), when considered over a given time horizon. Thus, this metric is particularly useful in quantifying and comparing the future climate impact that is due to current emissions of long-lived GHGs. However, GWP is less suited to quantifying the impact of short-lived agents. These radiative forcing metrics do not account for the climate sensitivity to the forcing. For example, climate sensitivity of radiative forcing due to changes

in O_3 may not be the same as for RF due to CO_2 and the sensitivity might differ geographically (Hansen *et al.*, 1997; Joshi *et al.*, 2003).

Both GWP and RF are used in this chapter: the GWP is applied to assess the impact of N_r on current GHG emissions in Europe, and the RF concept to assess the indirect effects on air chemistry and to quantify the integrated effect that European N_r emissions have had on the global climate system.

19.2 Effects of reactive nitrogen on net N_2O exchange

The major driver for changes in atmospheric N_2O concentrations is the increased use of N_r fertilizer, which on the one hand allowed humans to dramatically increase global agricultural production and, thus, to feed the current global world population (Erisman *et al.*, 2008; Jensen *et al.*, 2011, Chapter 3, this volume), but on the other hand increased N_r availability and thus microbial N_2O production. Owing to cascading of applied N_r onto landscape, regional and even global scales following the volatilization of NH_3 and NO_x , leaching of nitrate to water bodies or erosion processes, fertilizer N_r has also affected the source strength of non-agricultural terrestrial and aquatic

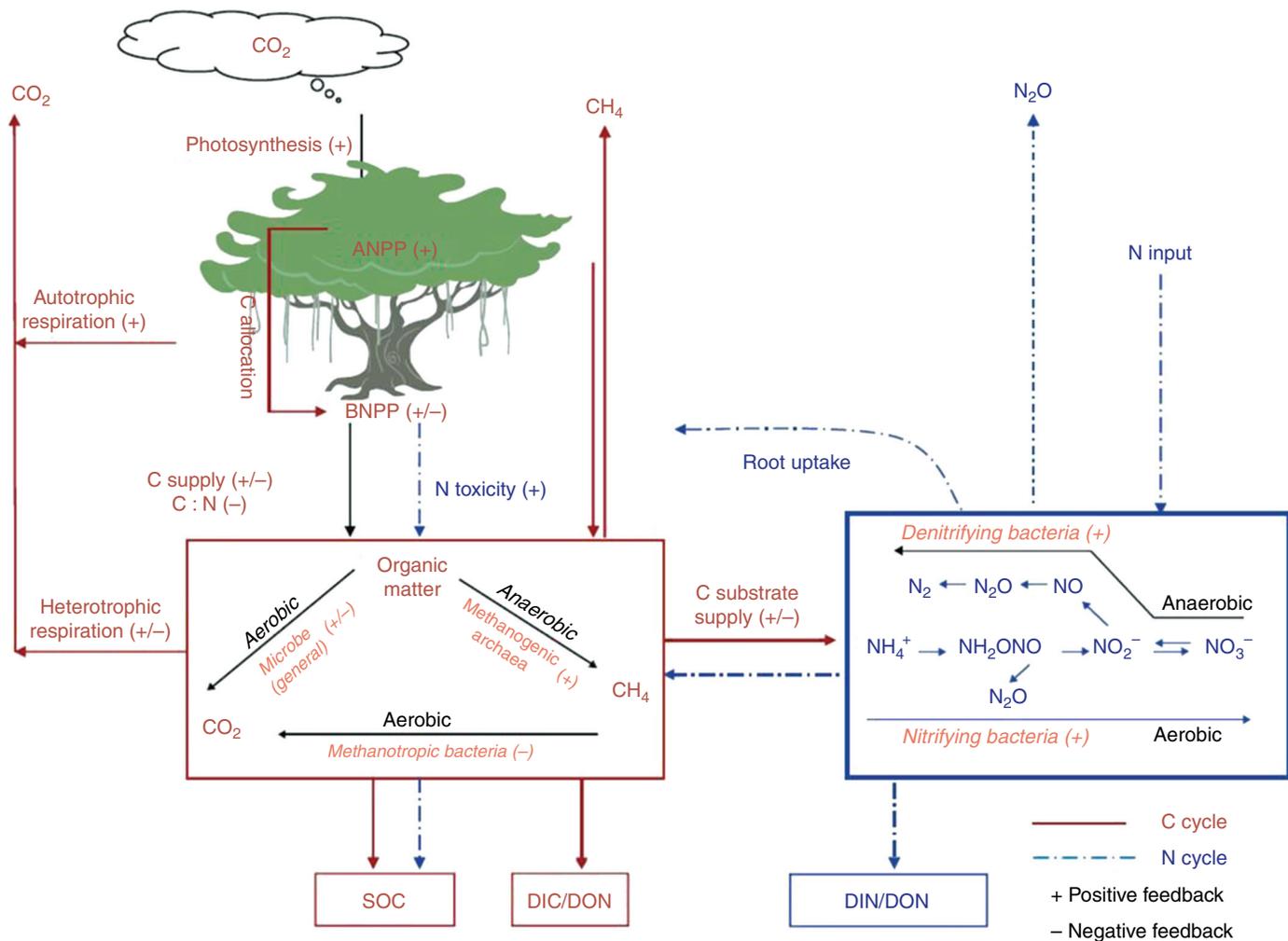


Figure 19.2 Potential mechanisms regulating the terrestrial ecosystem responses of CO_2 , CH_4 and N_2O production and consumption to increased availability of N, (figure adapted from Liu and Greaver, 2009). (ANPP, aboveground net primary productivity; BNPP, belowground net primary productivity; SOC, soil organic carbon; DOC, dissolved organic carbon; DIN, dissolved inorganic nitrogen; DON, dissolved organic nitrogen.)

systems for N_2O (Galloway *et al.*, 2004; Butterbach-Bahl *et al.*, 2011, Chapter 6, this volume). Also the emission of N_r from industry and combustion (including transport) has directly and indirectly contributed to changes in the global atmospheric N_2O source strength. In addition, emissions of NO_x from all combustion processes has resulted in huge increases in the atmospheric loading of N_p , with consequences for N_r deposition to terrestrial and aquatic ecosystems and thus also for N_r availability for microbial processes and finally N_2O production (Sutton *et al.*, 2007; Simpson *et al.*, 2011, Chapter 14, this volume). This section evaluates separately each source category for N_2O and how the source strength may have changed with time.

19.2.1 Direct N_2O emissions from agricultural activities

N_2O emissions from the agricultural sector are mainly related to direct N_2O emissions from soils following the application of N_r – either in the form of synthetic fertilizer or in the form of manure – or from N_2O emissions related to livestock production,

specifically during manure storage, livestock grazing or from paddocks. The IPCC (2006) guidelines specifically list all of these sources and provide emission factors for estimating N_2O emissions from them. Other top-down or bottom-up approaches have investigated only some of these sources or have amalgamated several sources together. Therefore, we provide a general overview of results for different approaches in Table 19.1.

N_2O emissions from soils are mostly estimated by using emission factor approaches (EFs), expressing proportionality between N_2O efflux and fertilizer N input rate. However, it needs to be noted that these factors have a wide range of uncertainty (Eggleston *et al.*, 2006), and their use can underestimate the cumulative effect fertilizer N production may have on worldwide N_2O formation. While using the IPCC default factor (1% of N_r applied being directly emitted as N_2O plus indirect N_2O emissions following N_r cascading downwind/downstream of ecosystems due to volatilization/deposition, leaching/run-off or sewage emissions, see below) may still reflect the average situation at the plot scale, its overall effect on the global emission situation may be underestimated as the increase in atmospheric concentrations is observed to be more

in the order of 2%–2.5% of manure and fertilizer N_f (Davidson, 2009) or 3%–5% of newly produced N_f (Crutzen *et al.*, 2008; see further below).

In many regions of Europe, N_f added to soils exceeds the removal via crop and animal products. Velthof *et al.* (2009) published the N_f losses to the environment for EU-27, using a deterministic and static N cycling model that calculates N_f emissions from agricultural activities on an annual basis (MITERRA-EUROPE). Based on their evaluation, 171 Mha of agricultural land in EU-27 receives on average 143 kg N_f ha⁻¹ yr⁻¹, most of which is in the form of mineral fertilizer (46%) and manure (43%). On average, N_2O emissions have been estimated at 2 kg N ha⁻¹ yr⁻¹ agricultural land (EU-27), which amounts to approx. 377 GgN in the year 2000 for the agricultural sector (Oenema *et al.*, 2009; see also Leip *et al.*, 2011, Chapter 16, this volume). Total denitrification losses including both N_2O and N_2 as estimated by a N emission factor and mass balance approach were however much larger: 44 kg N ha⁻¹ yr⁻¹ agricultural land on average (range: 8–183 N ha⁻¹ yr⁻¹ agricultural land) or 7486 Gg N for EU-27 (Velthof *et al.*, 2009; Oenema *et al.*, 2009), indicating that emissions of N_2 are dominating. N_2O emissions from the agricultural sector of EU-27 as reported by the EU member states in the framework of the Kyoto protocol are in the same range as reported by Oenema *et al.* (2009), i.e., approx. 400 Gg N in the year 2000 for direct soil N_2O emissions and emissions of N_2O from pastures and manure management (EEA, 2009).

The figures presented above are mainly based on IPCC emission factor (EF) approaches. However, Crutzen *et al.* (2008) compared bottom-up and top-down approaches and suggested that the EF factors as recommended by IPCC may miss part of the effects over the lifetime of an N_f molecule. As a consequence, the overall emissions from releasing N_f to the environment as a fertilizer are not fully characterized by the 1.4% estimated according to IPCC (1% direct + 0.4% indirect; IPCC, 2006; note that 1% direct emissions is a global average which is based on field measurements, see e.g., database by Stehfest and Bouwman, 2006), but 3%–5% for synthetic fertilizer N_f or otherwise newly created N_f (Crutzen *et al.*, 2008). In a recent re-evaluation of available data on temporal trends in global fertilizer and manure use and changes of atmospheric N_2O concentrations over the past 150 years, Davidson (2009) suggested that during manure or mineral fertilizer use, 2% or 2.5% of the nitrogen is converted to N_2O , respectively, and that double accounting of newly fixed N may be appropriate if the same N atom is first applied as newly fixed synthetic mineral fertilizer to produce animal feed and later gets recycled onto the soil as manure. It should be noted that the EF of Davidson and Crutzen *et al.* include both direct and indirect emissions. When using the Davidson (2009) approach and figures for EU-27 for total manure application (11 302 Gg N) and fertilizer use (10 678 Gg N) (Oenema *et al.*, 2009), this would result in total N_2O emissions from the agricultural sector of 493 Gg N. If an overall conversion factor is applied of 3%–5% from newly fixed N (synthetic fertilizer N plus biological N fixation) to N_2O -N, including indirect sources from cascading N_f (Crutzen *et al.*, 2008), then N_2O emissions from EU-27 due to agricultural activities would range from

364–607 Gg N for the year 2000 (Table 19.1) (see also De Vries *et al.*, 2011, Chapter 15, this volume).

Drainage of peatlands for agricultural use or for improving forest growth is widely practiced in Northern Europe. The resulting emissions of greenhouse gases from such organic soils may constitute a significant contribution to national GHG emissions. For example, Kasimir-Klemetsson *et al.* (1997) estimated that CO_2 and N_2O emissions from farmed organic soils in the Netherlands, Sweden and Finland, though representing a minor fraction of arable soil, may contribute 3%–10% of total national greenhouse gas emissions (see also Freibauer, 2003). However, since land use conversion to arable land cannot directly be attributed to N_f use this source has not been considered in this study.

19.2.2 Direct N_2O emissions from livestock farming and feedlots

Livestock production systems and management of livestock manure exert various influences on the environment and make up a relatively large share of the total emissions of nitrous oxide (N_2O). The influences on the environment greatly depend on the livestock production system itself, the management and the environmental conditions. Much of the influence of livestock systems on the environment occurs via its effects (direct and indirect) on land use (changes) and nutrient element cycling. These effects have increased greatly over the last decades, particularly in response to the current trends in livestock production: up-scaling, intensification, specialization and regional conglomeration (Tamminga, 2003; Foley *et al.*, 2005; Naylor *et al.*, 2005; Steinfeld *et al.*, 2006). These trends are facilitated by the availability of cheap energy, transport infrastructure and cheap N_f fertilizers for boosting the production of animal feeds.

Though livestock consumes less than 3% of the global net primary production (Smil, 2002), its contribution to the global burden of NH_3 , CH_4 and N_2O in the atmosphere ranges between 10% and 40% (Bouwman *et al.*, 1997; Oenema and Tamminga, 2005). Globally, livestock excrete about 100 (70–140) Tg N_f per year, but only 20%–40% of this amount is recovered and applied to crops (Sheldrick *et al.*, 2003; Oenema and Tamminga, 2005).

The total amount of N_f excreted by livestock in EU-27 was about 7–8 Tg N_f in the early 1960s and increased to 11 Tg in the late 1980s. Thereafter it tended to decrease again. These amounts are in the same order of magnitude as the N_f fertilizer use. Fertilizer N_f use was 4 Tg in 1960, peaked at 12 Tg per year in the late 1980s, and was about 10.5 Tg in 2002 (FAOSTAT, 2006).

Losses of N via N_2O emissions from manure management are presented in Table 19.2. The table shows that soil-based N_2O emissions (application and grazing) were higher than N_2O emissions from housing and storage.

19.2.3 Direct N_2O losses from sewage treatment and waste disposal

Both heterotrophic denitrification and nitrification (more specifically nitrifier denitrification, see Butterbach-Bahl *et al.*, 2011,

Table 19.2 Total excretion of N, by livestock and emission of N₂O (kton) from animal manure management systems in EU-27 in 2000 (Oenema *et al.*, 2009)

Livestock category	N _r excreted	Housing & Storage	Land Application	Grazing	Total
		N ₂ O ^a	N ₂ O	N ₂ O	N ₂ O
Dairy cattle	2670	18	18	12	48
Other cattle	3210	18	14	27	59
Pigs	1687	9	17		26
Poultry	1750	7	9		16
Other	1055	2	3	24	29
Total	10 372	54	61	62	177

^aN₂O emissions given here are the sum of emissions from housing systems and storage and do not include losses from mineral fertilizer applications to soil.

Chapter 6 this volume) are responsible for N₂O emission during the process of wastewater treatment (Kampschreur *et al.*, 2009). Under either totally anoxic or fully oxic conditions, N₂O production is rather limited; however, it increases significantly under low oxygen partial pressures (typically in the range 0.3% to 1.5% O₂ saturation) (Tallec *et al.*, 2006, 2008). Under actual operating conditions of wastewater aerobic activated sludge treatment N₂O emission can represent 0.1% to 0.4% of the NH₄⁺-N load oxidized (Tallec *et al.*, 2006). Similarly, nitrate removal through denitrification at zero oxygen could result in N₂O emissions representing 0.4±0.3% of the NO₃⁻-N eliminated (Tallec *et al.*, 2008). The latter figure is in the range of 0–1% mentioned by Hanaki *et al.* (1992) for a bench-scale study of sewage plants with sludge ages of more than 2.5 days and influent chemical oxygen demand/NO₃⁻-N ratios higher than 2.5.

One basic difficulty is that it is not always easy to distinguish clearly wastewater treatment from other waste management, e.g. the burning of sewage sludge, both industrial and domestic. EDGAR (2009) estimates annual emissions from waste water handling at about 25.7 GgN representing ~5% of total European N₂O emissions. For this emission source, the size of the population in the European countries is a good measure for the amounts emitted.

Solid waste disposal includes the incineration of solid wastes (mandatory for future years according to the *Waste Incineration Directive*, 2000/76/EC; OJ L332, P91–111), as well as its combustion for power and heat generation, and finally its disposal in landfills. In total, solid waste disposal sources contributed about 4.98 Gg N₂O-N in the year 2000, representing 0.8% of European emissions (EDGAR, 2009).

19.2.4 Direct N₂O emission fluxes from the energy sector, industry, transport, etc.

Like NO_x, N₂O may be formed as a side product during combustion. Temperatures favouring N₂O formation are somewhat lower than those of NO_x, so that N₂O emission factors in medium-temperature installations (500–600 °C) are clearly higher than at higher temperatures. Fluidized bed boilers, which employ lower temperatures (partly to abate NO_x) are thus an N₂O source. N₂O may also be formed during reduction of NO_x, during selective non-catalytic reduction, as well as in

three-way catalysts used for vehicles. In conclusion, NO_x abatement is often a cause of N₂O formation.

Referring to figures on combustion emissions from EU-27, reveals that combustion is nevertheless only a minor source for N₂O emissions. For 2007, EEA (2009) reports 7.6 Tg CO₂-eq from power plants, and 13.4 Tg CO₂-eq from transport (diesel and gasoline fuel), which is small fraction of the agricultural emissions.

The only major point sources of N₂O are large industrial facilities. The production of nitric acid via the oxidation of ammonia (a process that is also employed to produce caprolactam) and especially the application of concentrated nitric acid as an oxidizer for production of a handful of special chemicals (mainly adipic acid but also glyoxylic acid/glyoxal) are associated with high N₂O concentrations downstream and in consequence have high emission factors. Nitric acid plants exist typically in each medium sized European country, with several plants in large countries. By contrast, installations to produce the specialized chemicals are limited to very few sites in Europe: adipic acid is produced in three installations in Germany and one each in the UK and France. Two small facilities also exist in the Ukraine with one in Italy. Glyoxal, another compound along the nitric acid pathway, is produced in France only, while caprolactam is mainly produced in Belgium and the Netherlands.

The very limited number of installations and the high concentrations in the exhaust gas make it possible to monitor the emissions, so that the reports of EEA (2009) can be considered reliable. Furthermore, N₂O mitigation measures have become available at very low costs or could even be integrated into the overall process as product recovery. In consequence, already before 2000, all large adipic acid plants were equipped with mitigation devices, such that in EU-27 N₂O emissions dropped from 60 Tg CO₂-eq in 1990 to 9 Tg CO₂-eq, changing from a major to a minor source of N₂O emissions. Within the EU, only the Italian plant followed relatively late, after 2005. At the same time, the N₂O emissions from the other small industrial products mentioned decreased markedly (from 4.7 to 2 Tg CO₂-eq).

The N₂O emissions from nitric acid production are still relevant. For 2007, EEA (2009) reports emissions of 40 Gg CO₂-eq, about one quarter of the direct soil emissions.

Abatement is technically possible and has been successfully applied in at least one nitric acid plant, but as plume concentrations of N_2O are lower it is more expensive than in other processes. Nevertheless, the perspective of emission trading seems to make it financially attractive for European nitric acid producers to apply abatement measures as needed. According to industry reports, the phase-out of substantial N_2O emissions is under way.

The estimate for the UNFCCC reporting is 261 Gg N for the industry and transport sectors (EEA, 2009) is significantly lower than estimates by EDGARv4 with 461 Gg N (Reis *et al.*, 2009). Given that emissions from the chemical industry, as well as from the energy and transport sectors are well understood, differences between UNFCCC and EDGAR data are most likely due to different emission factors (or activity rates) in both approaches (Reis *et al.*, 2009).

19.2.5 Indirect N_2O emissions from natural/semi-natural terrestrial ecosystems

Nitrogen deposited in natural and semi-natural ecosystems via atmospheric input mainly comes from two sources: agricultural activities and associated NH_3 volatilization and NO_x emissions through the burning of fossil fuels, from the industrialized continents, shipping and aircraft or biomass burning, lightning and production from soil microbes (Simpson *et al.*, 2006). The N demand of a growing forest is approx. 5–10 kg N_f /ha/yr (Scarascia-Mugnozza *et al.*, 2000). Thus, N_f deposition > approx. 10 kg N will lead to an enrichment of the ecosystem with N_f , i.e., increased N_f availability in the soil-plant system. Indications for N_f enrichment are a reduction of the C:N ratio of the litter, forest floor or mineral soil, increased concentrations of nitrate and ammonium in the soil solution (Kristensen *et al.*, 2004; Mannig *et al.*, 2008), as well as increased emissions of N_2 and N_f trace gases from the soil. Several studies show that N_f deposition and N_2O as well NO emissions from forest soils are positively correlated (Brumme and Beese, 1992; Brumme *et al.*, 1999; Papen and Butterbach-Bahl, 1999; Van Dijk and Duyzer, 1999; Butterbach-Bahl *et al.*, 1997; Pilegaard *et al.*, 2006; Skiba *et al.*, 2006). The observed stimulation of fluxes is mainly attributed to the increased availability of N_f (as NH_4^+ and NO_3^-) for the microbial processes of nitrification and denitrification (Rennenberg *et al.*, 1998; Corré *et al.*, 1999), i.e., the key microbial processes responsible for N trace gas production in soils.

A possible further explanation for increased N_2O emissions due to ecosystem N_f enrichment was recently provided by Conen and Neftel (2007). They speculated that increased N_f availability may have reduced N_2O reduction in soils via denitrification, i.e., that the ratio of $N_2O:N_2$ increases with increasing N_f availability. Since increased N_f deposition also affects nitrate leaching and runoff (Dise *et al.*, 1998; Borken and Matzner, 2004), indirect N_2O emissions from water bodies due to N_f deposition to natural systems also need to be considered.

However, a thorough evaluation and quantification of N_f deposition effects on soil N trace gas emissions remains difficult, since environmental conditions, such as meteorology or

soil and plant properties, significantly affect the magnitude, temporal course and composition of the emitted N gases.

Having in mind these difficulties, there have been several attempts to estimate the stimulating effect of N_f deposition on N_2O emissions from forest soils. Skiba *et al.* (2006) used a gradient approach, with measuring sites located in a mixed forest at increasing distances from a poultry farm, i.e., a strong NH_3 source. They estimated that >3% of the N deposited to the woodland sites was released as N_2O . Butterbach-Bahl *et al.* (1998) used a regression type approach, time series of nitrogen deposition throughfall data and continuous N_2O and NO emission measurements at the long-term monitoring site at Höglwald Forest for estimating N_f -deposition driven N_2O losses. Their estimate is comparable to that in the study by Skiba *et al.* (2006), i.e., 1.4% for coniferous forests and 5.4% for deciduous forest. Also, a literature review by Denier van der Gon and Bleeker (2005) showed that N_f deposition to forests stimulates N_2O emissions within the same range; they concluded that the stimulating effect was higher for deciduous forests (5.7% of deposited N is lost as N_2O) than for coniferous forests (3.7%).

In a scenario study at the EU scale, Kesik *et al.* (2005) estimated N_f deposition effects on forest soil N_2O emissions by running the biogeochemical model Forest-DNDC either with best estimates of atmospheric N_f deposition or by assuming that N_f deposition was zero. The results indicated that, across Europe, 1.8% of atmospheric N_f deposition was returned to the atmosphere as N_2O . All published estimates, therefore, show that the default N_2O emission factor of 1% used by IPCC for indirect emissions from soils following N deposition (Mosier *et al.*, 1998; IPCC, 2006) is most likely too low by at least a factor of two.

19.2.6 Indirect N_2O emissions from riparian areas, rivers and coastal zones

Although direct emission from agricultural soils is the dominant process responsible for N_2O emission by the agricultural sector, indirect emissions linked to the cascading of agricultural N_f 'downstream' from the fields might also play a significant role.

Reactive nitrogen inputs to rivers and coastal waters include both natural and anthropogenic components; the latter is dominated by applied fertilizer N_f lost through leaching and runoff, followed by sewage and atmospheric sources (Seitzinger and Kroeze, 1998). Most of the N_f may already be denitrified in riparian areas in the direct vicinity of the sites of N_f application (e.g., arable fields). From a review of available data on N_2O emissions from riparian wetlands, Groffman *et al.* (2000) concluded that although current data are inadequate to propose a quantitative emission factor for N_f entering riparian areas, these emissions are likely to be significant in many regions. A nitrogen budget of the Seine hydrographical network (Billen *et al.*, 2001, 2007, 2009), reveals that up to 25%–30% of the N_f input to surface water from agricultural soils is denitrified to N_2O and N_2 in riparian zones, compared to only 5%–10% in-stream. If the percentage of N_2O loss with respect to nitrate denitrified in riparian zones is the same as in the drainage network, N_2O

emissions from riparian zones would represent about 10% of the estimated total direct N_2O emissions from the agricultural soils of the watershed (Garnier *et al.*, 2009).

Also with regard to N_2O losses from coastal areas, estimates are highly uncertain. The total flux of N_r and the fraction of fertilizer N reaching coastal waters is both variable and difficult to estimate. Early studies of individual rivers and/or estuaries, targeted both measured and estimated inputs from various land use activities such as N_r in sewage and atmospheric deposition (Billen *et al.*, 1985; Larsson *et al.*, 1985; Jaworski *et al.*, 1992; Boynton *et al.*, 1995; Nixon *et al.*, 1995; Howarth *et al.*, 1996). Other work examined dissolved N_r export in relation to specific watershed characteristics, such as human population and energy use (Cole *et al.*, 1993), and point and non-point sources (Cole and Caraco, 1998). Another study modelled river and estuarine N_2O production globally, using functions of nitrification and denitrification that were related to external N loading rates derived by adapting local/regional models of watershed environmental parameters to global databases (Seitzinger and Kroeze, 1998). The results indicated (i) that ~ 8% of the N_r input to terrestrial ecosystems is exported as dissolved inorganic nitrogen (DIN) in rivers; (ii) the DIN export to estuaries globally (year 1990) is ~20.8 Tg N_r per yr; (iii) about 1% of the N_r input from fertilizers, atmospheric deposition, and sewage to watersheds is lost as N_2O in rivers and estuaries; hence rivers and estuaries might account for 20% of current global anthropogenic N_2O emissions and are thus similar in magnitude to previously identified sources such as direct anthropogenic N_2O emissions from soils (Seitzinger and Kroeze, 1998).

Such model-derived estimates should, however, be approached with caution. First, for denitrification the heterogeneity of microbial ecosystem structure (Rich and Myrold, 2004), oxygen status (Helder and De Vries, 1983) and physicochemical aspects such as sediment porosity and grain size (Garcia-Ruiz *et al.*, 1998), ambient temperatures, pH and water content (Berounsky and Nixon, 1990), and levels of suspended particulate matter (Owens, 1986) all co-vary to constrain N_2O production. Second, the simple linear functions relating DIN loading to N_2O in the global-scale models (Seitzinger and Kroeze, 1998; Seitzinger *et al.*, 2000) are not well supported by individual studies, which reveal a wide range. For example, previous work in the Humber estuary (UK) implied that ~25% of the terrestrial DIN input converts to N_2O via sediment denitrification (Barnes and Owens, 1999), a far larger conversion than the mean of about 0.15% employed in the global-scale models (Seitzinger and Kroeze, 1998; Seitzinger *et al.*, 2000). By contrast, a dynamic model of estuarine DIN cycling in the Tyne estuary (UK) showed only 3.9% of the DIN load to be nitrified, in comparison to a value of 60% assumed in the global scale models, with only 0.009% of the DIN load converted to N_2O (Rodrigues *et al.*, 2007). A corresponding value for the Scheldt estuary was 0.17% of nitrified N_r being converted to N_2O , much closer to the global scale average (Rodrigues *et al.*, 2007). Nevertheless, this study concluded that the amount of atmospheric N_2O derived from agricultural sources in general, including estuarine transformations of N, might need to be

revised downward, consistent with constraints set by atmospheric N_2O growth.

Although the global models (Seitzinger and Kroeze, 1998; Seitzinger *et al.*, 2000) included some major European river/estuaries in their development, direct comparisons of DIN load to N_2O production were not readily available. Given the variability among the few direct comparisons that have been made and ranges of more than an order of magnitude in both DIN inputs and nitrification rates in European estuaries (Rodrigues *et al.*, 2007), a meaningful representative mean value for the ratio for DIN input to N_2O production for European estuaries appears to remain not well constrained.

Seitzinger and Kroeze (1998) presented the first comprehensive estimate of N_2O emissions from terrestrial and aquatic sources at the scale of Europe (more specifically the watersheds of North Eastern Atlantic, the Baltic, the Black Sea and the Mediterranean). They calculated N_2O emissions from rivers, estuaries and shelf areas as a percentage (0.3 or 3%) of nitrification and denitrification rates in rivers and estuaries which were, in turn, calculated as a function of external inputs of dissolved inorganic nitrogen (DIN). Although the method is rather rough and questionable, it provides a first order of magnitude of the fluxes at European scale: 0.23 Tg N_2O -N per yr from rivers and 0.11 Tg N_2O -N per yr from estuaries and shelves. Based on a compilation of available data of N_2O concentration in estuarine and coastal marine waters, Bange (2006) arrived at overall higher figures, based on component estimates of 0.13–0.16 and 0.20–0.41 Tg N_2O -N per year for European shelf and estuarine waters, respectively. However the major merit of the approach used by Seitzinger and Kroeze (1998) is to explicitly link N_2O emissions from aquatic systems to the nitrogen load reaching surface waters from agricultural sources, as is suggested by the site specific data presented above. Even if one assumes that 50% of emissions are natural, the remaining magnitude of N_2O emissions from coastal zones in Europe would still be at least in the same magnitude as indirect emissions from leaching and N_r deposition (Table 19.1).

19.3 Effects of N_r on net CH_4 exchange

Reactive nitrogen availability has been reported to affect both the capacity of upland soils to serve as a sink for atmospheric CH_4 and as a source of CH_4 emissions from wetlands (Figure 19.2). Furthermore, indirect effects of N_r on animal feed quality, in this case, changes in indigestible carbohydrates and crude protein contents, can also affect ruminant CH_4 emissions. These three aspects are briefly discussed here with their potential feedback on the EU radiative balance. A summary of all effects is presented in Table 19.3.

19.3.1 Net CH_4 oxidation by upland soils

Globally, biological methane oxidation is estimated at 17 ± 9 Tg CH_4 -C per yr (Dutaur and Verchot, 2007). By comparison, Boeckx and Van Cleemput (2001) estimated the total EU15 CH_4 oxidation (grassland and agricultural land) to be 0.2 Tg C per yr. In general, higher uptake rates than for grasslands and agricultural land are reported for other upland ecosystem types

Table 19.3 Changes in CH₄ fluxes in EU-24/EU-27 and C sequestration by terrestrial ecosystems due to the use of N_i and its dispersal in the environment (positive values are emissions and negative values are uptake)

	Forest	Grassland	Arable	Other semi natural land	Wetlands	Ruminants	Terrestrial C sequestration
Total area (km ²) ^a	1 030 635	820 219	1 174 325	264 932	46 915		
CH ₄ exchange (kg CH ₄ -C ha ⁻¹ -yr ⁻¹) ^b	-2.7 (-2.2 - 6.3)	-1.6 (-0.5 - 3.1)	-1.0 (-0.7 - 2.2)	-2.7 (-2.2 - 6.3)	3.92 ^e	4.98 ^g	
Total CH ₄ exchange EU24 (Tg CH ₄ -C yr ⁻¹)	-0.27 (-0.23 - 0.65)	-0.12 (-0.04 - 0.25)	-0.11 (-0.08 - 0.26)	-0.07 (0.06 - 0.17)			
Effect of N _i on CH ₄ exchange (Tg CH ₄ -C yr ⁻¹)	0.02 ± 0.0006 ^c	0.09 ^d (0.09 - 0.27)	0.2 ^d (0.18 - 0.48)	<0.01 ^c	<0.01 ^f	? ^h	
Ecosystem CO ₂ fluxes (Tg C yr ⁻¹)							-125 ⁱ
N _i effect on ecosystem CO ₂ fluxes (Tg C yr ⁻¹)							-36.6 ± 12.9 ^j
O ₃ effects on ecosystem carbon sequestration (Tg C yr ⁻¹)							5.0 - 14.4 ^k
N _i effect on EU-27 radiative balance							
							-9.3 to -44.5
							-34.1 to -163

^a European Environment Agency (2006).

^b Boeckx and Van Cleemput (2001).

^c Based on the linear equation by Liu and Greaver (2009) and an average N deposition to EU forests and semi-natural land of 13 kg N_i yr⁻¹ or 10 kg N_i yr⁻¹, respectively.

^d Assuming that rates would equal those of upland forest soils if N_i use would be abandoned.

^e Saarnio *et al.* (2009).

^f Assuming a mean atmospheric N_i deposition of 10 kg N ha⁻¹ yr⁻¹ and using the N_i response factor as provided by Liu and Greaver (2009) together with the total area of wetlands in EU-27.

^g EEA (2009); includes only the values for enteric fermentation (year 2000).

^h The effects of N_i supply on forage digestibility and CH₄ emissions per animal, and the effect of N_i in allowing increased animal stocking rates and overall animal numbers are not quantified here.

ⁱ Schulze *et al.* (2009), all terrestrial ecosystems (forest only = -1.09 Tg C yr⁻¹)

^j De Vries *et al.* (2007) and section 19.4 (this volume).

^k See Section 19.5.2 (this volume).

such as forests and heathlands in the temperate and boreal zones (Smith *et al.*, 2001; Butterbach-Bahl, 2002; Dutaur and Verchot, 2007). Uptake of CH₄ by upland soils in EU25 has been approximately calculated as 4 Tg CH₄-C per yr (Schulze *et al.*, 2009). This sink strength, however, has been and still is affected by N_r (Figure 19.2) and land use change.

Increased atmospheric N_r deposition increases [NH₄⁺] in the soil and usually decreases CH₄ uptake by well-drained soils (Stuedler *et al.*, 1989; Gullledge and Schimel, 1998). Three mechanisms have been postulated for the partial inhibition (slowing down) of CH₄ uptake by well-drained soils in response to increased N_r input: (a) competitive inhibition of the methane mono-oxygenase by ammonia, (b) inhibition of methane consumption by toxic intermediates and end products of methanotrophic ammonia oxidation such as hydroxylamine and nitrite, or (c) osmotic stress due to high concentrations of nitrate and/or ammonium (Schnell and King, 1996; Bradford *et al.*, 2001; Bodelier and Laanbroek, 2004; Reay and Nedwell, 2004).

Changes in land use affect soil methane oxidation, for example by the demand for increased production for food, fibre and biofuels. Generally CH₄ oxidation in upland soils sharply decreases with intensity of landuse, i.e. CH₄ oxidation will be highest in forests and natural grasslands, somewhat lower in managed grasslands and negligible in arable soils.

On the basis of a meta-data analysis of published studies on N_r effects on CH₄ uptake by upland soils, Liu and Greaver (2009) estimated the detrimental effects of N_r addition to soils on CH₄ uptake as 0.012 ± 0.006 kg CH₄-C ha⁻¹ yr⁻¹ for upland agricultural fields and as 0.016 ± 0.004 kg CH₄-C ha⁻¹ yr⁻¹ for non-agricultural ecosystems, per 1 kg N_r ha⁻¹ yr⁻¹ added to the ecosystem. Comparable values for the inhibitory effect of N_r addition to forest ecosystems in Europe via N_r deposition on CH₄ uptake are reported by Butterbach-Bahl *et al.* (1997) and De Vries *et al.* (2008). They reported a factor of 0.031 kg CH₄-C ha⁻¹ yr⁻¹ per 1 kg N ha⁻¹ yr⁻¹ in the form of NH₄⁺. If one assumes that the total molar ratio of reduced to oxidized atmospheric N_r deposition in Europe is approx. 1:1 (which is in accordance with Simpson *et al.*, 2006) this would result in precisely the same number as in the Liu and Greaver study, assuming oxidized N does not affect CH₄ oxidation.

These findings can be used to approximate the effect of N_r additions via atmospheric deposition or fertilizer application on CH₄ uptake for natural/semi-natural ecosystems in Europe. For grassland and agricultural land, an approach which considers N_r application and uses the regression line provided by Liu and Greaver (2009) yields somewhat unrealistic values if the N_r use intensity in Europe is taken into account. However, for agricultural land one can assume that CH₄ uptake activity may recover to a maximum of values observed in natural upland systems (if no N_r is applied and assuming that no management alteration). On the basis of an extensive literature review on existing measurements of methane uptake in Europe and elsewhere, Boeckx and van Cleemput (2001) summarized ranges for CH₄ uptake for forest soils of (2.2–6.3 kg CH₄-C ha⁻¹ yr⁻¹, mean: 2.7 CH₄-C ha⁻¹ yr⁻¹), for grasslands (0.5–3.1 kg CH₄-C ha⁻¹ yr⁻¹, mean: 1.6 CH₄-C ha⁻¹ yr⁻¹) and for arable soils (0.7–2.2 kg CH₄-C ha⁻¹ yr⁻¹, mean: 1.0 CH₄-C ha⁻¹ yr⁻¹). These ranges

are in general agreement with a more recent global analysis of CH₄ uptake on upland soils by Dutaur and Verchot (2007).

If one assumes that CH₄ uptake would, at maximum, equal the rates of uptake in forest soils, then agricultural activity and use of N_r has decreased CH₄ uptake by on average 0.1 Tg CH₄-C per yr for grassland soils and by 0.2 Tg CH₄-C per yr for arable soils, respectively (at the scale of EU-24), see Table 19.3. For forest the formula as provided by Liu and Greaver (2009) was used. Average atmospheric N_r deposition in Europe was assumed to be on average 13 kg N_r ha⁻¹ yr⁻¹ for forests (Simpson *et al.*, 2006) or 10 kg N_r ha⁻¹ yr⁻¹ for other semi-natural upland systems (e.g., heathland, macchia), respectively (Table 19.3). With this approach we do not consider that large parts of forests in Northern Europe receive <13 kg N ha⁻¹ yr⁻¹, whereas forests in Central Europe are exposed to values of atmospheric N deposition >13 kg N ha⁻¹ yr⁻¹, i.e. the spatial variability of N_r deposition was disregarded for this calculation. However, the effect of N_r atmospheric deposition on rates of atmospheric CH₄ uptake by forest soils and soils of other semi-natural land uses at the scale of EU-27 can be seen to be small at <0.05 Tg CH₄-C yr⁻¹. In summary, accounting for all land uses, the effect of all N_r in suppressing CH₄ uptake by soils in Europe was estimated at 0.27–0.75 Tg CH₄ yr⁻¹ (Table 19.3).

19.3.2 Net CH₄ emissions from wetlands

The effect of N_r additions to wetland ecosystems will largely depend on the N status of the respective ecosystems. For highly managed wetland ecosystems, such as rice paddies, that receive high loads of N_r fertilizers, inorganic N_r additions have been shown to result in reductions of CH₄ emissions. This effect is most likely due to a stimulation of CH₄ oxidizing bacteria following increased N_r availability (Bodelier and Laanbroek, 2004) or due to a slower decrease in redox potential and thus delayed onset of methanogenesis (if nitrate fertilizers or ammonium sulphate fertilizers are used). On the other hand, N_r additions will increase plant productivity and thus also rhizodeposition of C substrates, which can fuel methanogenesis and can lead to a net increase in CH₄ emissions, as has been shown for rice paddies and natural wetlands (Fumoto *et al.*, 2008). Furthermore, significant rates of N_r addition to wetlands (>approx. 10–20 kg N_r ha⁻¹ yr⁻¹) promotes the growth of vascular plants (to the detriment of moss numbers) and thus potentially increases evapotranspiration – which may lower the water table – and stimulates CH₄ oxidation in the rhizosphere (Berendse *et al.*, 2001; Bouchard *et al.*, 2007).

However, reports on the effect of atmospheric N_r deposition on CH₄ emissions from wetlands are scarce (Dise and Verry, 2001), so that the possible consequences of N_r additions to wetlands and on CH₄ fluxes are difficult to assess. In their meta-analysis of data sets, Liu and Greaver (2009) therefore included studies on N_r addition effects on CH₄ emissions from rice paddies, because of scarcity of relevant datasets and because, in their evaluation, the response to N_r addition was not different between the two ecosystems. The calculated response factor suggests that, per kg N_r, CH₄ emissions are stimulated by 0.008 ± 0.004 kg CH₄-C ha⁻¹ yr⁻¹. Compared to the overall CH₄

emissions from wetlands and water bodies in Europe, which were estimated at 3.92 Tg CH₄-C yr⁻¹ (Saarnio *et al.*, 2009), the N_r deposition effect (assuming a mean value of 10 kg N ha⁻¹ yr⁻¹) on total emissions is negligible and <0.01 Tg CH₄-C yr⁻¹.

19.3.3 CH₄ emissions from ruminants and their relationship to nitrogen

Ruminants are an important source of CH₄, which is a product of microbial degradation of feed constituents. In the long term, some feeding practices on intensively managed farms have improved considerably and can be expected to continue to do so, mainly because of the gradual improvement of ruminant genotypes which requires a permanent adaptation of nutritional strategies. In the case of extensive feeding systems, changes over time in diet composition were much smaller.

There are several interactions between N_r management and ruminants CH₄ emissions. These include the use of N_r fertilizer to supply increased amounts of feed, allowing larger livestock populations, the effects of forage and feed quality on CH₄ emissions, and management interactions concerning the fate of livestock excreta.

Possibly the largest effect of N_r supply on ruminant CH₄ emissions in Europe is by providing fertilizer to grow crops used as forage and feed for ruminants. Without substantial N_r inputs – with a significant share coming from feed imports – also Europe would not be able to support so many livestock, especially in the dairy, pig and poultry sectors. By comparison, beef cattle and especially sheep are typically grazed on more extensive systems, often with much lower N_r fertilizer inputs. In broad terms, Erismann *et al.* (2008) estimated that around half of the world population depends on N_r fertilizers and this percentage is expected to be higher for Europe. In this case, a minimum of 50% of European ruminant CH₄ emissions (i.e. ~2.5 Tg CH₄-C per year) could therefore be attributed to N_r supply. However, this figure remains very uncertain due to the different mixtures of land use, forage and feed (including feed imports) used to support the European ruminant population. In the following paragraphs, we therefore focus on the direct interactions of N_r with CH₄ emissions from ruminants.

The key for a high N use efficiency of the animal and thus a low N emission potential of the manure lies in the optimization of the rumen microbial protein synthesis. These microbes require both degradable protein and carbohydrates (starch, sugars). The most prominent change in the last decades in European feeding practices concerns changes in the carbohydrate sources. This is through the gradual exchange of part of the forage (rich in fibre) by concentrate (rich in starch and often also protein). Under certain conditions (e.g., sufficient ruminal availability of the concentrate's carbohydrates, adjusted amino acid supply) this improves N use efficiency by up to 50% and mitigates potential N emissions accordingly. By contrast, forages from extensive grasslands are often low in digestibility (few ruminally degradable carbohydrates) and those from intensively managed grasslands are excessive in protein, thus being inferior to mixed forage-silage maize-concentrate diets in that respect.

The consequences for methane emission of these changes are variable; however, a proportionate decline in methane formation per unit of digestible carbohydrates, and therefore especially per unit of livestock-source food, can still be expected from the use of mixed diets (Beauchemin *et al.*, 2008). Mitigating effects of concentrate on proportionate methane emission range between 0 and 70%. Hindrichsen *et al.* (2006) noted a reduction of methane emission from 36 to 19 and 14 g CH₄ per kg of milk in cows yielding 10, 20 and 30 kg milk day⁻¹, respectively, showing that the efficiency gain is much lower when starting from a higher level of yield. Additionally, any reduction with this diet change may be at least partially compensated for by a concomitantly increased methane emission from the manure (Hindrichsen *et al.*, 2006).

The second most important measure to improve N use efficiency in livestock is given by a reduction of excessively high (crude) protein contents of the diet. This is very effective in reducing potential N emission (Külling *et al.*, 2001), but effects on methanogenesis are small and inconclusive (Külling *et al.*, 2001, 2003). The only exception arises when a dietary protein reduction below a critical value adversely affects fibre degrading rumen microbial species. These species are also those providing most of the hydrogen required by the methanogens. This situation rarely occurs in Europe, except in very extensive systems. In developing countries, the use of either urea-molasses licking blocks, or ammonia/urea treated straw is a major option for dealing with the limitation in degradable N. The reduced methane emission resulting from this strategy is associated with a massive increase in performance (Moss *et al.*, 1994; Islam and Begum, 1997).

The source of ruminally degradable protein (grass, oil meals, urea, etc.) has no direct influence on the ruminal N use efficiency and therefore does not affect either N or methane emissions in a substantial way. However, the metabolic N use efficiency of the animal for milk and meat production can be improved by using ruminally-undegradable protein sources when dietary protein is concomitantly reduced (Kröber *et al.*, 2000). Such protein sources include naturally and artificially rumen-protected protein or even single protected amino acids. As protein is not a major substrate for the methanogens, again this set of feeding measures would only affect N_r emissions and not methanogenesis.

New attempts in the mitigation of CH₄ and associated noxious gases are based on feeding with low dietary concentrations of effective secondary plant metabolites (Beauchemin *et al.*, 2008). There is a huge variability in compounds and plant species available, and some of them have already been processed to marketable products, others might be grown as forages by European farmers. Effective plants are often rich in tannins, saponins, essential oils and sulphur-containing compounds. As these ingredients may partially inhibit ruminal protein degradation to ammonia and at the same time the methanogens, these measures are particularly promising. However, they are rarely cost-effective unless mitigation of noxious gases also improves animal growth or milk yield or is included in a payment scheme.

Methane emissions per unit of food have substantially declined with increasing feed quality over the past decades, but also due to improvements in the genetic characteristics of the herd and the improvements in management. However, this change was not accompanied by a corresponding reduction in the number of animals needed to produce our food, as eating habits simultaneously increased towards higher consumption of these now cheaper foods. This means that global methane emission from ruminant husbandry has continued to increase during the last decades, as fueled by substantial N_r fertilizer inputs, and thus contributes to the ongoing raise in atmospheric CH_4 concentration.

19.4 Effects of reactive nitrogen on ecosystem net CO_2 exchange

Reactive nitrogen is a key nutrient for both vegetation and soil biota and because of the limited (natural) supply it is a limiting factor for plant growth and soil organic matter decomposition in many terrestrial ecosystems (Vitousek and Howarth, 1991; LeBauer and Treseder, 2008). Field studies have demonstrated a positive effect of low to medium level N_r additions on plant growth and carbon accumulation (Vitousek and Howarth, 1991; Aber *et al.*, 1993; Bergh *et al.*, 1999; Franklin *et al.*, 2003). N_r additions affect vegetation growth by increasing tissue N content and leaf-level photosynthesis, as well as decreasing the (relative) investment into below-ground carbon allocation (Poorter and Nagel, 2000; Magill *et al.*, 2004). Both mechanisms generally increase above-ground productivity and lead to a higher accumulation of above-ground woody biomass. However, the effectiveness of these mechanisms is limited when other processes such as water limitation, micronutrient availability, and competition for light become more limiting than N_r availability. For example, a 15-year-long N_r amendment study in Harvard Forest, USA, showed that the large increases in carbon accumulation predicted using a linear relationship between leaf N_r content and photosynthesis, failed to materialize in the field at least for red pine (Bauer *et al.*, 2004). As a consequence, the vegetation response is strongest in young fast growing forest ecosystems (Oren *et al.*, 2001) and boreal forest ecosystems (Bergh *et al.*, 1999; Jarvis and Linder, 2000), in which N_r is the primary constraint of growth. Furthermore, the N_r addition effect is expected to saturate or even decline in ecosystems with high N_r input ecosystems (Aber *et al.*, 1998; Brumme and Khanna, 2008).

Soil respiration rates, both from autotrophic and heterotrophic sources, have been shown to be generally reduced under elevated N_r (Fog, 1988; Agren *et al.*, 2001; Hagedorn *et al.*, 2003; Knorr *et al.*, 2005; Olsson *et al.*, 2005). The reason for the decline is likely to be an alteration of the microbial decomposition of organic matter by uncoupling the degradation of polysaccharides and polyphenols (Sinsabaugh *et al.*, 2002, 2005). Increased N_r availability stimulates cellulolysis, which tends to accelerate the decomposition of labile litter, and inhibits the expression of oxidative enzymes required for the breakdown of lignin and other secondary compounds. In consequence labile organic matter stocks may turnover more

rapidly, thus shrink in abundance, while humified fractions accumulate (Sinsabaugh *et al.*, 2005). The net effect on C accumulation in soil depends on whether changes in the decomposition rate or increased C inputs from increased biomass and litter production dominate (Schulze *et al.*, 2000).

Rained, ombrotrophic bogs are a special case in the response of soil organic matter to N_r addition. Low N_r inputs promote the growth of the peat building Sphagnum plants; however, higher N_r availability generally favors the growth of vascular plants (Berendse *et al.*, 2001; Bubier *et al.*, 2007). Because of (co-)limitation with phosphorus (P) and potassium, N_r input does not necessarily increase growth (Limpens *et al.*, 2004). However, even where growth is increased, the net effect on C accumulation may still be zero or even negative because of the higher degradability of vascular plant litter (Gunnarsson *et al.*, 2008). N_r additions generally increase the decay of dead material in peatlands (i) due to reduced microbial N_r limitation and (ii) indirectly due to improved litter quality (Bragazza *et al.*, 2006). Enhanced decomposition in peatlands due to N_r may however, be limited, as several studies with high N_r inputs show signs of P limitation (Limpens *et al.*, 2004; Bragazza *et al.*, 2006).

At the whole ecosystem scale, the rate of C accumulation in response to N_r addition is determined by the fate of the N_p and the stoichiometry of vegetation and soil organic matter (Nadelhoffer *et al.*, 1999, 2004). For example, in forests, because of the high C:N ratio of woody biomass, N stored in wood will involve a much stronger C accumulation than in N storage in soil organic matter. Depending on the fate of the added N_p , the C accumulation per unit N_r could vary between zero (in ecosystems with no or little N retention) and several hundred (where most N accumulates in woody tissue). A number of recent studies have aimed at quantifying the response of C accumulation to additions based on fertilizer trials (Hyvonen *et al.*, 2007), application of ^{15}N tracer (Nadelhoffer *et al.*, 1999, 2004), observations at long-term monitoring plots (De Vries *et al.*, 2006; Solberg *et al.*, 2009; Laubhann *et al.*, 2009), as well as interpretation of net ecosystem production data from eddy-covariance CO_2 measurements (Magnani *et al.*, 2007; Sutton *et al.*, 2008). The published estimates of the response of carbon sequestration to N addition in above-ground biomass and in soil organic matter for forests and heathlands have recently been summarized by de Vries *et al.* (2009) (Table 19.4). The results of the various studies are in close agreement and show that above-ground accumulation of carbon in forests is generally within the range 15–40 g C per g N_r . In heathlands, a range of 5–15 g C per g N_r has been observed based on low-dose N_r fertilizer experiments. The uncertainty in C sequestration per kg N_r addition in soils is larger than that for above-ground biomass and varies on average between 5–35 g C per g N_r for both forests and heathlands. All together these data indicate a total carbon sequestration range of 5–75 g C per g N_r deposition for forest and heathlands, with a most common range of 20–40 g C per g N_r . Such low values are to be expected as ^{15}N tracer studies suggest that most added N_r becomes stored in soil organic matter (Nadelhoffer *et al.*, 1991; Tietema *et al.*, 1998).

The results are in line with a meta-analysis of studies on CO_2 fluxes from N_r additions in multiple terrestrial and

Table 19.4 Estimated ranges in carbon sequestration per kg nitrogen addition in above-and-below ground biomass in forest at various scales (after de Vries *et al.*, 2009)

Approach	Carbon sequestration (kg C per kg N _r)		Scale of application		Authors
	Above ground	Below ground	Total		
Forests					
Empirical field data					
Correlation between NEP and total N deposition	—	—	68–177 ^a	Chronosequences (5) in boreal and temperate forests of Eurasia and North America	Magnani <i>et al.</i> (2007) as re-evaluated by Sutton <i>et al.</i> (2008)
Correlation between the average growth increase of nearly 400 Intensive Monitoring plots and N deposition in a multivariate analysis	15–38	—	—	Nearly 400 Intensive forest Monitoring plots	Solberg <i>et al.</i> (2009), Laubhann <i>et al.</i> (2009)
¹⁵ N experimental data					
Extrapolation of ¹⁵ N experimental data with average C/N ratios of forest ecosystem compartments	30–70	11–18	41–88	One forest site in Sweden	Melin <i>et al.</i> (1983)
Extrapolation of ¹⁵ N experimental data with average C/N ratios of forest ecosystem compartments	25	21	46	Generic average	Nadelhoffer <i>et al.</i> (1999)
Extrapolation of ¹⁵ N experimental data with site specific data at 6000 plots in Europe	33	15	48	European average	De Vries <i>et al.</i> (2006)
Results of fertilizer experiments					
Average results from 30 year low dose (34 kg N _r /ha/yr) fertilizer experiments	25	—	—	Forest plot in Sweden	Högberg <i>et al.</i> (2006)
Average results from 14–30 year fertilizer experiments	25	11	36	Two forest plots in Sweden and Finland	Hyvönen <i>et al.</i> (2008)
Average results from 10 year chronic N addition (30 kg N _r /ha/yr) experiments	17	23	40	Four forest plots in the USA	Pregitzer <i>et al.</i> (2008)
Results of model simulations					
Range in results of three process-based models (high N _r supply)	—	—	10–30	One forest site in Sweden	Levy <i>et al.</i> (2004)
Range in results of three process-based models (medium N _r supply, 9–25 kg N _r /ha/yr)	—	—	43–75	One forest site in Sweden	Sutton <i>et al.</i> (2008)
Range in results of five process-based models	15–25	—	—	Two forest plots in UK	Rehfuess <i>et al.</i> (1999)
Average result of the process-based model EFM	—	—	41–54	22 forest plots in Europe	Milne and van Oijen (2005); Sutton <i>et al.</i> (2008)
Range in results of the process based model SUMO	20–30	—	—	Dutch forests	Wamelink <i>et al.</i> (2009a)
Range in average results per latitude of the process based model chain SMART2-SUMO2	3–12	5–11	7–24	166 forest plots in Europe	Wamelink <i>et al.</i> (2009b)
Range in results based on the process-based model O-CN	—	—	37 (2–79)	87 forest plots in Europe and Northern America	Zaehle and Friend (2010)

Table 19.4 (cont.)

Approach	Carbon sequestration (kg C per kg N _r)		Scale of application		Authors
	Above ground	Below ground	Total		
Heathlands					
Results from 5–11 year N fertilizer experiments at 20–120 kg N _r /ha/yr	5–15	20–34	25–49	2 heathland plots	Evans <i>et al.</i> (2006); Evans, pers. comm.
Model simulations for the N fertilizer experiment sites	-	21–32	-	3 heathland plots	Evans <i>et al.</i> (2006); Evans, pers. comm.

^a The high value assumes no covariation between N_r and climate drivers; the low value attributes variation first to climate (growing degree days).

wetland ecosystem types by Liu and Graever (2009). The analysis included 68 publications that contained 208 observations across North and South America, Europe and Asia. The overall results showed that the effect on net ecosystem CO₂ exchange for non-forest ecosystems (grassland, wetland and tundras) was not statistically significant (very large differences), while the effect on ecosystem net carbon storage for forest ecosystems showed on average a statistically significant 6% increase, with annual N_r additions ranging from 25 to 200 kg N_r ha⁻¹ yr⁻¹. On average, forest ecosystems sequestered 24.5 ± 8.7 kg CO₂-C ha⁻¹ yr⁻¹ per kg N_r ha⁻¹ yr⁻¹ (-89.8 ± 32.0 kg CO₂ equivalents ha⁻¹ yr⁻¹) added to the ecosystem. Note that these results cannot be extrapolated to systems with very high N_r inputs, nor to other ecosystems such as peatlands, where the impact of N_r is much more variable, and may range from C sequestration to C losses.

It should be noted that although a study of forest chronosequences (Magnani *et al.*, 2007) might appear to give a very high response of ~400 g CO₂-C per kg N_r (Hogberg, 2007), part of this high value can be related to the need to account for dry N_r deposition, in which case the results show a *prima facie* response of ~177 g CO₂-C per g N_r depositon (de Vries *et al.*, 2008; Sutton *et al.*, 2008). While this remains a high value, it may be explained by spatial covariation with climatic differences between sites, accounting for which gave a smaller estimate of 68 g CO₂-C per g N_r deposition (Sutton *et al.*, 2008). Nevertheless, even that lower estimate is high compared with the other forest studies shown in Table 19.4. Overall, the mean forest C responses in Table 19.4 for above- and below-ground are 25 (20–30) and 15 (14–17) g CO₂-C per g N_r input, respectively, amounting to an overall response of 41 (35–47) g CO₂-C per g N_r input. (This would equate to 47 (33–61) g CO₂-C per g N_r if the total responses shown were used, including the higher dry-deposition-corrected estimate of Magnani *et al.*, 2007, and Sutton *et al.*, 2008.)

Taking the spatial distribution of forests N_r deposition into account, the total (wet+dry, reduced+oxidized) N_r deposition over European forests has increased from 1860 to 2000 by 1.5 Tg N_r yr⁻¹ over a forest area of 188 Mha (Dentener *et al.*, 2006, Zaehle *et al.*, 2010). This forest area is somewhat larger than reported in the forest statistics for EU-27 as this estimate

accounts for all forested and woody land cover types, including those not reported in forest statistics. The increase in N_r deposition implies a mean increase of ~7.9 kg N_r ha⁻¹ yr⁻¹. Note that this estimate refers to above-canopy deposition velocities, not N_r catch by an ecosystem. Based on this N_r deposition rate, and the above reported NEP response (24.5 ± 8.7 g C g⁻¹ N_r from Liu and Graever, 2009), C sequestration due to N_r deposition would average 36.6 ± 13.0 Tg C yr⁻¹ for Europe. This figure is in good agreement with recent simulations by Zaehle *et al.* (unpublished) using the OCN model (Zaehle *et al.*, 2010) and same rates of N_r deposition and forest cover changes. Such a calculation resulted in a net forest uptake rate due to N_r deposition of 23.5 ± 8.5 Tg C yr⁻¹ (mean and standard deviation for the years 1996–2005), compared to an estimated net C uptake of 140 Tg C yr⁻¹ resulting from the historical changes in atmospheric [CO₂], climate, N_r addition (deposition and fertilizer application) and land-cover change. In addition, the N_r effect on unmanaged grasslands accounts for a further sink of 2.8 Tg C yr⁻¹. This mechanistic model implies an average C storage of 37 g CO₂-C g⁻¹ N_r across the sites considered by Magnani *et al.* (2007), with a range of 2–72 g CO₂-C g⁻¹ N_r, and a comparably low average response for European forests of 14.5 CO₂-C g⁻¹ N_r, due to the interaction with other growth limitations in this model (e.g., from water shortage in Southern Europe). The simulations with OCN suggest that N_r deposition has played only a minor role in terrestrial C cycling prior to the 1950s, after which the effect increased to the mid 1980s. The effect has thereafter remained relatively constant with some inter-annual variations related mainly to the interactions of N_r availability with climatic variability.

It is obvious that there remains significant uncertainty in the overall response of ecosystem carbon sequestration to N_r inputs. While the very high forest C response to N_r observed by Magnani *et al.* (2007) is an outlier, there remain significant differences between the estimate of Liu and Graever (2009) at 24.5 ± 8.7 g C g⁻¹ N_r and the mean of the values summarized in Table 19.4, at 41 (35–47) g CO₂-C per g N_r input, indicating the need for further research. Key uncertainties for forests concern the sensitivity of the C response to the amount, form and manner of N_r input (i.e., non-linear response with dose, dosing frequency, NO_y vs NH_x, above- vs below-canopy addition, etc.) and the

successional status of the forest, including the biophysical and biochemical legacies of prior land-use history.

Nitrogen additions also alters carbon cycling in agricultural systems due to positive effects on biomass yields, crop residues inputs to soil or stimulation of humus formation at high N_r availability (Christopher and Lal, 2007). On the other hand, fertilizer N has been shown to promote the decomposition of crop residues and soil organic matter (Khan *et al.*, 2007). In their metadata-analysis Liu and Greaver (2009) found that N_r additions to agricultural soils may slightly increase soil organic carbon stocks by an average of 2%. However, this estimate is based on a limited number of studies and, thus, cannot consider the variability of the chemical composition of the SOC pool across sites and regions, so that the estimate remains highly uncertain. Only if N is added to the soil in the form of manure soil C sequestration may significantly increase (Christopher and Lal, 2007). In view of the rather uncertain mineral N effects on soil C stocks we therefore assumed that they are neutral.

19.5 Effects of reactive nitrogen through atmospheric chemistry

Emissions of N_r compounds, in particular of NO_x from combustion sources and fertilized soils, and ammonia (NH_3) from livestock wastes and fertilizers, exercise multiple indirect effects on the climate system, through their participation in atmospheric chemistry. Several recent studies have attempted to attribute climate forcing components to emissions of pollutants, including NO_x and NH_3 . The IPCC (2007) report does this in a simplified manner, without taking into account chemical interactions, while the more recent study of Shindell *et al.* (2009) provides an improved allocation to emissions.

19.5.1 NO_x and changes in atmospheric oxidation capacity

Nitrogen oxides affect the oxidation capacity of the atmosphere: NO_2 undergoes photolysis which results in the formation of ozone (O_3). Tropospheric O_3 represents one of the most important greenhouse gases. The radiative forcing (RF) due to long-lived greenhouse gases (CO_2 , CH_4 , N_2O) does not vary significantly with location of its emission, so that the RF can be estimated from the emissions alone. By contrast, the lifetime of O_3 is much shorter and highly variable (from days to many weeks). Furthermore, the O_3 production per molecule of NO_x emitted depends non-linearly on other emissions, primarily of CO and non-methane hydrocarbons (NMHCs). To complicate matters further, the RF of O_3 also depends on its vertical distribution. As a consequence, the efficiency of NO_x in producing RF through O_3 production depends greatly on where this NO_x is emitted.

Increases in O_3 (associated with NO_x emissions) also lead to increases in OH. Similarly, NO reacts with HO_2 to form OH directly. The concentration of OH governs the lifetime of CH_4 , and thus the increase in OH has had a negative RF effect by reducing the CH_4 burden. Similarly, the increase in the CH_4 burden since pre-industrial times has increased the photochemical

production of O_3 , an effect which the IPCC attributes to CH_4 (rather than O_3) and which accounts for about 20% of the RF of CH_4 .

The net RF of NO_x is therefore the result of competing effects and will differ depending on where and when the NO_x is emitted, which makes it much harder to estimate the contribution of continents to the RF. For example, aircraft emissions appear to be particularly efficient in generating ozone, while spiking experiments have found the net RF from NO_x spikes is negative for January and July, and positive for April and October (Berntsen *et al.*, 2005).

A few modelling studies have tried to quantify the response of greenhouse gases to changes in NO_x emissions on different continents. Some of these were perturbation experiments where the aim was to find out the response to relatively modest changes in present-day emissions, to estimate the efficacy of abatement strategies. The extrapolation to zero anthropogenic emissions is somewhat uncertain due to non-linearities in the chemical system.

Derwent *et al.* (2008) estimated that a January emission pulse of 2 Tg NO_2 in Europe results in global time-integrated emission responses of -0.6 and $+0.013$ ppb years for CH_4 and O_3 , respectively. Thus, a sustained cut of all anthropogenic NO_x emissions in Europe (5.0 Tg N_r yr^{-1}) would increase the global average concentration of CH_4 by approximately $+6.5$ ppb years and decrease that of O_3 by -0.14 ppb years. Applying a relationship between RF and concentration of 0.37 $mW\ m^{-2}ppb^{-1}$ for CH_4 (Schimel *et al.*, 1996) results in an estimate of the RF of current European NO_x emissions of -2.4 $mW\ m^{-2}$ through the effect on reducing global CH_4 and a net effect of $+1.4$ $mW\ m^{-2}$ ($+2.0$ $mW\ m^{-2}$ short-term and -0.6 $mW\ m^{-2}$ long-term) through the effect on O_3 . Model results indicate that the sensitivity of CH_4 and O_3 to NO_x increases with increasing NO_x concentrations and thus the extrapolation of a 2 Tg NO_2 pulse to a -5 Tg yr^{-1} would tend to underestimate the overall effect. Also, emission spikes in one single month (January) may not be representative for the average effect over the year (Berntsen *et al.*, 2005).

It should be noticed that industrial NO_x emissions originate from the same sources as emissions of CO and NMHC. We here estimate the effect of the NO_x emissions alone. However, without these emissions, emissions of CO and NMHC would also be lower and the O_3 RF forcing responds very sensitively to the combined change.

Table 19.5 summarizes the different estimates of the global average RF due to European anthropogenic emissions of NO_x through their effect on tropospheric O_3 and CH_4 lifetime. Here, the O_3 RF is the relatively small difference between the short-term cooling effect (through reducing O_3 in the short term) and the long-term warming effect (through reducing the oxidative capacity in the longer term; see Derwent *et al.*, 2008).

19.5.2 NO_x and tropospheric O_3 feedbacks on plant growth

Elevated tropospheric ozone, resulting largely from NO_x and VOC emissions, is well-known to cause reductions in the growth

Table 19.5 Summary of estimates of the global RF due to the effect of European anthropogenic NO_x emissions on CH₄ and O₃ (mW m⁻²)

Model	Study	CH ₄ RF	O ₃ RF	Total	Comment
LMDzINCA	Berntsen <i>et al.</i> (2005)	-5.0	+4.3	-0.7	Calculated as -5× estimated response to 1 Tg pulse
Oslo-CTM2	Berntsen <i>et al.</i> (2005)	-4.2	+5.5	+1.3	Calculated as -5× estimated response to 1 Tg pulse
MOZART-2	Naik <i>et al.</i> (2005)	-6.7	+0.3	-6.4	Derived as 10× the estimated response to a 10% cut
STOCHEM	Derwent <i>et al.</i> (2008)	-2.4	+1.4	-1.0	Derived here (see text)
Mean		-4.6	+2.9	-1.7	

of crops and tree species, and changes in species composition in experimental grasslands (cf. Karnosky *et al.*, 2007; Fuhrer, 2009). By inhibiting wood production, ozone directly leads to reduced aboveground C storage in forests (Wittig *et al.*, 2009) and through its effects on plant C assimilation and C allocation belowground, ozone could affect soil carbon sequestration, and thus soil C storage. Ozone may change decomposition processes in the soil through effects of ozone on plant residue mass and on the concentration of nutrients, secondary metabolites, lignification and/or the C/N ratio of above- and below-ground plant parts, in combination with indirect effects on soil microbial communities (Kim *et al.*, 1998; Kanerva *et al.* 2008; Chen *et al.*, 2009). Results from short-term experiments are, however, not consistent, and it is thus difficult to draw a consistent picture of the impact of ozone on belowground C cycle processes. While reduced relative C allocation to roots leading to lower root litter production is generally observed, consequences of ozone stress for long-term C stabilization involving microbial processes are less clear. In aspen (*Populus tremuloides*) and in mixed aspen-birch (*Betula papyrifera*) stands, Loya *et al.* (2003) observed that ozone strongly inhibited extra stable soil C formation from elevated CO₂. Under wheat and soybean, elevated ozone caused a change in soil C quality towards high molecular weight and more aromatic components (Islam *et al.*, 1999). While biogeochemical model simulations consistently project reduced soil C sequestration and C stocks (Ren *et al.*, 2007; Sitch *et al.*, 2007), long-term field studies of soil C under elevated ozone are lacking.

Sitch *et al.* (2007) estimated that global gross primary production is projected to decrease in 2100 as compared to 1901 by 8%–23% owing to plant ozone damage. However, there are still large uncertainties since counteracting effects of changes in atmospheric O₃ and CO₂ concentration cannot easily be delineated. As a first approach we assumed that by the year 2000 ecosystem sequestration in Europe was already reduced by 4%–11.5% due to increased atmospheric O₃ concentration, thereby neglecting regional differences in C sequestration and atmospheric O₃. We applied this assumption to ecosystem CO₂ fluxes in EU 25, which were approximately -125 Tg C per yr in the period 2000–2005 (Schulze *et al.*, 2009). Thus, contemporary O₃ concentrations may already have reduced total C sequestration in Europe by 5–14 Tg C per yr as an upper bound (Table 19.3).

19.5.3 Nitrous oxide and stratospheric ozone

Forster *et al.* (2007) assess the total radiative forcing from observed changes in stratospheric ozone to be -0.05+/-0.1 W m⁻² (medium confidence), subject to the spatial and temporal distribution of the changes, with mean decreases relative to pre-1980s values of 6% and 3%, in the Southern and Northern Hemisphere, respectively. Only a rather small fraction of this decline could potentially be attributed to the observed increases in atmospheric N₂O concentrations, given the model experiments by Nevison and Holland (1997), however, such an assertion is fraught with uncertainty due to the complexities of stratospheric ozone chemistry. Given that the EU-27 currently emits around 10.6% of the global N₂O (Table 19.7) the approximate effect of anthropogenic N_r increase from the EU-27 is uncertain, but likely to be comparatively small.

19.5.4 Nitrogen containing aerosols

Aerosols are known to have a cooling effect on the climate through direct scattering of sunlight (the direct effect). Emissions of NH₃ and NO_x are associated with the formation of aerosol sulphates and nitrates. Ammonia neutralizes sulphuric acid (H₂SO₄) to produce ammonium bisulphate and ammonium sulphate aerosols, which are stable salts (although they may revolatilize through cloud processing (Bower *et al.*, 1997). Oxidation of NO_x results in the production of HNO₃ which interacts with NH₃ to form ammonium nitrate (NH₄NO₃), which establishes a dynamic equilibrium with the gas precursors: the aerosol phase favoured in cold, humid conditions and high gas concentrations. Any NH₄NO₃ may therefore revolatilize into NH₃ and HNO₃ if temperatures rise, relative humidity drops or if the gas concentrations decrease. Sulphates and NH₄NO₃ form in the accumulation mode (0.1–1 μm). This aerosol mode undergoes the slowest removal from the atmosphere and therefore survives longest, providing the largest surface area for the condensational processes to occur. This size range is highly efficient in scattering light and therefore contributes to the direct forcing of aerosols. Nitric acid also interacts with coarse (sub-micron) crustal and seasalt aerosol to form calcium and sodium nitrate (Ca(NO₃)₂, NaNO₃), respectively. Thus there are several processes by which the emission of N_r compounds impacts on the direct RF of aerosol (Table 19.6).

Table 19.6 Summary of different estimates of the anthropogenic direct aerosol forcing global and averaged over Europe. These values implicitly include neutralization by ammonia

Model	Study	Global (W m ⁻²)		European average (W m ⁻²)		European contribution to global (mW m ⁻²)	
		Sulphate	Nitrate	Sulphate	Nitrate	Sulphate	Nitrate
GCM II-prime	Adams <i>et al.</i> (1999)	-0.96	-0.19				
GATORG	Jacobson (2001)	-0.32	-0.05				
GISS	Liao and Seinfeld (2005)	-0.57					
GEOS-CHEM v5.03	Martin <i>et al.</i> (2004)	-0.36	-0.069	-1.36			
Oslo CTM-2	Myhre <i>et al.</i> (2004, 2005)	-0.37	-0.02				
GISS	Bauer <i>et al.</i> (2007) ^a	-0.34	-0.11	-0.81	-0.34	-26.5	-11.1

^a European values: S. Bauer, personal communication.

Modulation of H₂SO₄ production from SO₂

As discussed above, NO_x emissions affect OH concentrations and therefore the main source of H₂SO₄ in the atmosphere, production through SO₂ oxidation. Shindell *et al.* (2009) attribute a global RF of -0.13 W m⁻² to the effect of NO_x emissions on SO₄²⁻. Regional contributions of this effect do not appear to have been derived. A first estimate of the European effect is here derived by multiplication of this RF estimate with the ratio of the total European/global SO₄²⁻ RF.

In large parts of North America and China the aerosol is often observed to be acidic (i.e. sulphate exists as bisulphate or even sulphuric acid), and the same holds for European areas affected by higher SO₂ emissions or subject to warm temperatures (e.g., Eastern and Southern Europe or Northern Scandinavia; Nemitz *et al.*, 2010). By contrast, in Central and Western Europe sulphate now tends to be fully neutralized by NH₃ and in these areas, sulphuric acid competes with nitric acid for the ammonia. As a consequence a reduction in H₂SO₄ would increase the amount of NH₃ available for NH₄NO₃ formation, which would at least partially offset the reduction in (NH₄)₂SO₄. Thus the total aerosol RF over Europe is likely to be less sensitive to H₂SO₄ production than at the global average.

Neutralization of sulphuric acid

Sulphuric acid condenses to form aerosol even in the absence of NH₃. However, any NH₃ present is readily incorporated into this aerosol neutralizing sulphuric acid to bisulphate and eventually to sulphate. These three forms of sulphate have different optical properties and can hold different amounts of water, leading to different effective aerosol sizes. Jacobson (2001) attributed a global tropopause forcing of +0.06 W m⁻² to anthropogenic ammonium, by reducing the water holding capacity of the aerosol compared with H₂SO₄ at a given RH. Martin *et al.* (2004) modelled the optical properties of the NH₄⁺-SO₄²⁻-NO₃⁻ system based on the two different branches of the deliquescence/efflorescence curve. Because the atmospheric lifetime of sulphate is quite long (days to weeks; Garland, 2001), regional sulphate fields cannot easily be linked to regional emissions. As a

first estimate, we derive the contribution of European anthropogenic emissions by multiplying the global value by the ratio of European to global anthropogenic SO₂ emissions (Smith *et al.*, 2001), which does not consider the fact that the deposition and oxidation of SO₂ will vary in different parts of the world.

Coarse nitrate formation

Adding only a thin layer to the existing aerosol, the reaction of HNO₃ with coarse aerosol does not significantly change the radiative properties of this size mode, which is already less important for the climate system.

Ammonium nitrate formation

Formation of NH₄NO₃ exercises an RF contribution that can be clearly attributed to N_r emissions. Fewer model studies have attempted to estimate the direct forcing of nitrates, compared with sulphates, and the general picture is that, globally, the nitrate effect is about ¼ of the sulphate effect. NH₄NO₃ forms where large emissions of NH₃ and NO_x (from agricultural and combustion sources, respectively) co-exist at relatively low temperatures, and it volatilizes in remote areas with an atmospheric life time of hours to days. Thus, fine nitrate RF is regionally variable, with Europe representing one of the major global hotspots, together with China and parts of the USA. Figure 19.3 demonstrates that the nitrate field over Europe can be clearly attributed to Europe itself and concentrations decrease towards the edge with little intercontinental transport. This allows the regional nitrate RF to be linked to the European emissions. Thus the regional top-of-the-atmosphere forcing of nitrate averaged over Europe, scaled to the globe, provides a reasonable estimate of the contribution of European NH₃ and NO_x to the global RF, although there is likely to be some net export of NO₃⁻ to the east, which is not captured by this approach. See also Figure 19.4.

Ammonium and nitrate indirect effects

In addition, to the direct effect, aerosols exercise a number of indirect effects, e.g., by modulating the number of cloud condensation nuclei, with associated increases in cloud albedo

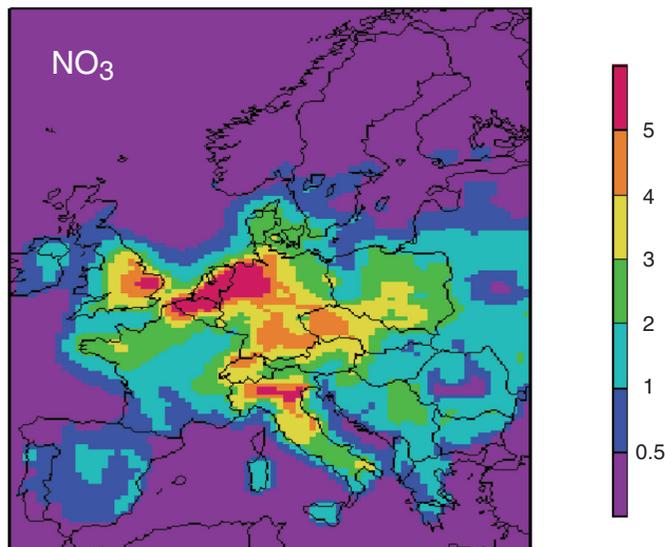


Figure 19.3 Modelled fine nitrate (NO_3^-) concentration over Europe (in $\mu\text{g m}^{-3}$), from Schaap *et al.* (2004). Estimates of the European forcing from sulphates and nitrates are summarized in Table 19.6. While several studies have quantified the effect at the global scale, only the authors of the study of Bauer *et al.* (2001) provided European averages on request, the fields of which are shown in Figure 19.4. The comparison with Figure 19.3 illustrates that the global coarse resolution models at present have limited skill in reproducing the European nitrate fields and the European forcings therefore have to be treated with some caution.

(1st indirect effect), the cloud lifetime/thickness (2nd indirect effect) or changing the albedo of snow (mostly important for black carbon). These effects are poorly quantified for sulphate aerosol and no global quantification appears to exist for the contribution of nitrate aerosol. Li *et al.* (2009) recently simulated the effect of nitrate indirect forcing for China and estimated a RF of -1.63 W m^{-2} in January and -2.65 W m^{-2} in July.

In addition to the effect on the Earth's radiation balance, aerosols are thought to stimulate plant growth and thus carbon storage. Through scattering of light, fine particulates tend to increase diffuse radiation. This may result in an increase in ecosystem production, since photosynthesis seems to be more efficient under diffuse light conditions (Mercado *et al.*, 2009). Furthermore, increased availability of N, also seems to have a positive impact on the reflectivity (albedo) of vegetation. This would imply an additional cooling effect, although the mechanism is yet not clear (Ollinger *et al.*, 2008). These two latter effects were not considered here.

19.6 Integration: comparing present trends with the past

This section attempts to answer the question: 'What is the overall climate effect of European nitrogen emissions?' Although several approaches could be taken to answer this question, here the following constraints were considered: (a) since the overall effect includes short-lived agents, the quantification should be based on the RF metric; (b) since the overall effect includes long-lived GHGs, the estimate needs to consider the effect on the global rather than European regional RF; and (c) the

estimate should illustrate how European N management could change the European contribution to climate change. In extension to the IPCC approach, the question is therefore refined to: 'What has been the contribution of European anthropogenic nitrogen emissions to the atmosphere to the overall change in global radiative forcing (between 1750 and 2005) that can be attributed to European activity?'

To answer this question poses two additional challenges: (i) the European contribution of the IPCC RF bar graph needs to be isolated, which does not appear to have been done before, and (ii) the present-day GWPs and RFs derived above need to be turned into RFs integrated over the 1750–2005 time window.

19.6.1 European contribution to the change in global RF 1750–2005

The RF of European GHG emissions is estimated by applying the total global RF components in relation to the present-day contribution of European to total global emissions (Table 19.7). This does not take into account that this ratio has evolved over the time-scale that affects present-day concentrations of GHGs. Approximately 23% of the global jet fuel is currently sold in Europe (source: USEIA, 2009) and we have therefore assumed that the same proportion of contrails can be attributed to European activity. The European effect of black carbon (BC) on snow was similarly estimated from the present day ratio of European to global BC emissions (Bond *et al.*, 2004).

The change in surface albedo due to landuse change has been highly variable geographically, and largest albedo increases have been derived for North East US, Europe (with a possible decrease in the Iberian Peninsula), East Asia and Brazil (Mhyre *et al.*, 2005). However, once the geographical variability in available solar radiation to calculate the effect of RF is included, the effect is weighted towards the low latitudes. There is substantial disagreement in the spatial patterns of the RF due to surface albedo changes (cf. Hansen *et al.*, 1998; Mhyre *et al.*, 2005; Betts *et al.*, 2006), but qualitative agreement that the albedo increase in Europe has been larger than the global average. Thus, we derive a first estimate of the RF due to European landuse change as twice the value expected by ratioing European to global landmass.

19.6.2 Conversion of nitrogen-related present day RF and GWP to change in RF for 1750–2005

Estimating the radiative forcing (RF) due to N_r deposition effects on terrestrial C storage cannot be done directly from present-day net exchanges, because of the long life-time of CO_2 in the atmosphere. By contrast, the spatial patterns of net exchanges are of lesser importance. This implies that the combined impact of the historical net terrestrial, marine and fossil C exchanges with the atmosphere on atmospheric $[\text{CO}_2]$ need to be taken into account. Calculation based on the historical emission estimates from fossil fuel statistics (Boden *et al.*,

Table 19.7 Contribution of Europe (OECD states) to total global anthropogenic emissions of CO₂, CH₄ and N₂O and importance of N₂O for GHG emissions in Europe. Global and European (OECD states) data were taken from the EDGAR (2009) emissions database

	CO ₂	CH ₄	N ₂ O		
	Tg CO ₂	Tg CH ₄	Tg CO ₂ -equiv.	Gg N ₂ O	Tg CO ₂ -equiv.
Global	29 913	321	7 382	12 208	3 613
EU (OECD countries)	3 861	16.4	376.4	1 299	384.6
%-share EU	12.9	5.1	5.1	10.6	10.6
N ₂ O effect on GHG emissions (Table 19.3)	-34 to -163	0.4-1.0	8.3-23	910-2260	269-669
%-share in relation to total-EU emissions	0.01-0.05		2.2-6.1	100	100

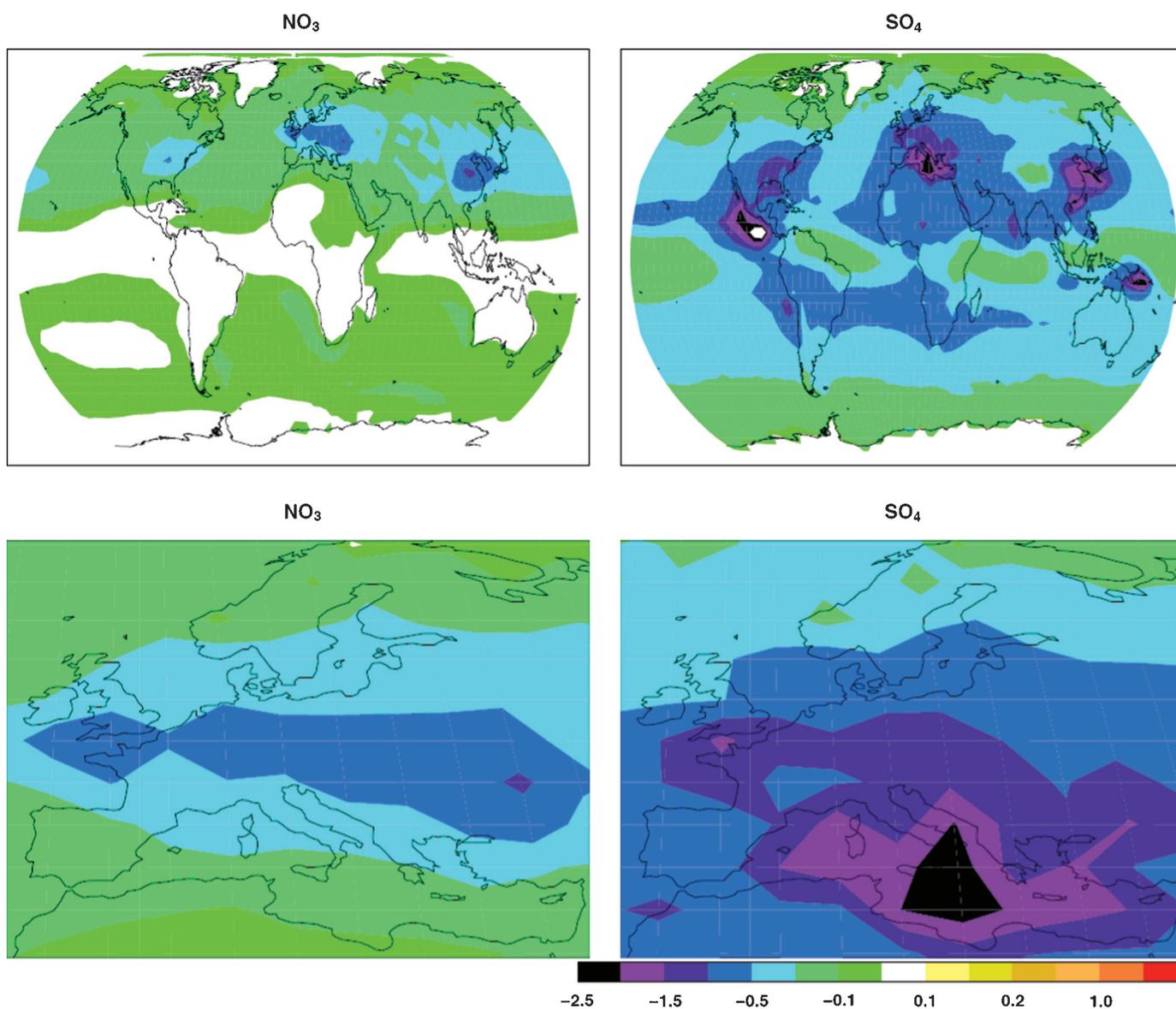


Figure 19.4 Example model result of NO₃⁻ and SO₄²⁻ top-of-the-troposphere forcing in W/m² at the global and zoomed-in European scale (based on the data by Bauer *et al.* (2007 kindly replotted by the authors)).

Table 19.8 Summary of best estimates of N_r global RF attributed to European anthropogenic emissions, and their uncertainty ranges (in mW m⁻²)

Effect	Best estimate	Min	Max
Biosphere interactions			
Increase in terrestrial C sequestration due to atmospheric N _r deposition ^a	-19	-30	-8
Decrease in terrestrial C sequestration due to tropospheric O ₃ from NO _x	4.4	2.3	6.5
Decrease in CH ₄ soil uptake due to atmospheric N _r deposition	+0.13	+0.03	+0.24
N ₂ O			
Increase in atmospheric [N ₂ O]	+17	14.8	19.1
Gas phase chemistry			
Reduction in CH ₄ lifetime	-4.6	-6.7	-2.4
Tropospheric O ₃ production – radiative effect	+2.9	+0.3	+5.5
Aerosol direct effects			
Total sulphate effect	-5.4	-9.4	-1.4
Increase in H ₂ SO ₄ production from SO ₂	-10.1	N/A	N/A
Neutralization of H ₂ SO ₄	+4.7	N/A	N/A
Coarse nitrate production	negligible		
NH ₄ NO ₃ direct effect	-11.1	-18.1	-4.1
Aerosol indirect effects			
Effect on cloud albedo	No estimate		
Effect on precipitation / cloud lifetime	No estimate		
Stimulation of plant growth through diffuse radiation	No estimate		

^a This estimates accounts for the atmospheric N_r deposition to forests and unmanaged grasslands; it does not include the effect of agricultural fertilization with N_r.

2009) and marine (Le Quere *et al.*, 2009) and terrestrial forest (Zaehle *et al.*, 2010) modelling, suggests a net radiative cooling of 19 mW m⁻² due to atmospheric N_r deposition effects on forests and natural grasslands from EU-27, compared with a total cooling effect of 74 mW m⁻² from EU-27 terrestrial carbon storage, which is roughly an order of magnitude less than the radiative forcing from EU-27 fossil fuel emissions (Zaehle, unpublished results) (Table 19.8). The predicted C storage due to N_r deposition is estimated to be 23.48 Tg C yr⁻¹ by forests and 2.79 Tg C yr⁻¹ by unmanaged grasslands, which is lower than the estimate of 34–163 Tg C yr⁻¹ derived for forests on the basis of Section 19.4 (cf. Table 19.7). This reflects the fact that the N_r-sensitivity in this mechanistic model lies at the lower end of the values found in the literature (Section 19.4). Consequently, the radiative effect predicted here must therefore be considered conservative. It should also be noted that the estimate of the carbon response of managed (agricultural) ecosystems is not included.

A similar modelling approach was not available to estimate the effect of N_r related increases in ozone concentrations on plant growth over the 1750–2005 time period. A first estimate of the effect of +4.4 mW m⁻² was derived by comparison of the reduced C storage of -5 to 14 Tg C per yr estimated in Section 19.5.2 to the increased C storage due to N_r deposition predicted by the OCN model of +36.6 Tg C per yr (Section 19.4). This assumes that the time-lines in N_r and associated O₃ damage have been similar.

Again in the absence of a time-integrating model calculation, a first estimate of the radiative forcing associated with reduced CH₄ uptake by soils was derived from the ratio of the estimated reduction in CH₄ uptake to the anthropogenic European emissions (Table 19.7), which led to a small warming of 0.13 mW m⁻².

The current European RF of N_r-related aerosol is taken as an approximation for the 1750–2005 change. Clearly, some N_r-related aerosol effects would have been in place in 1750 due to NO_x and NH₃ emissions from the natural biosphere and lightning.

The resulting estimates of the European contribution to the change in global RF and the effects of N_r are summarized in Figure 19.5 and Table 19.8. The European contribution to the global RF is estimated to be about 410 mW m⁻² equating to about 27% of the global anthropogenic change in RF. European N_r emissions to the atmosphere cause warming and cooling effects, which add to a net cooling effect of -15.7 mW m⁻² which equates to a reduction of the European RF of about 4%. Because the indirect aerosol effects (e.g., cloud albedo effect) are not contained in this figure, the best estimate of the net cooling effect of N_r would be larger. However, since the errors associated with the individual components are significant, the range of possible values is estimated to be -46.7 to +15.4 mW m⁻², i.e. -11% to +4% of the total contribution.

The estimates of the effects of N_r emissions to the atmosphere on (i) carbon sequestration, (ii) aerosols (sum of nitrate and sulphate) and (iii) N₂O emissions are each estimated to be

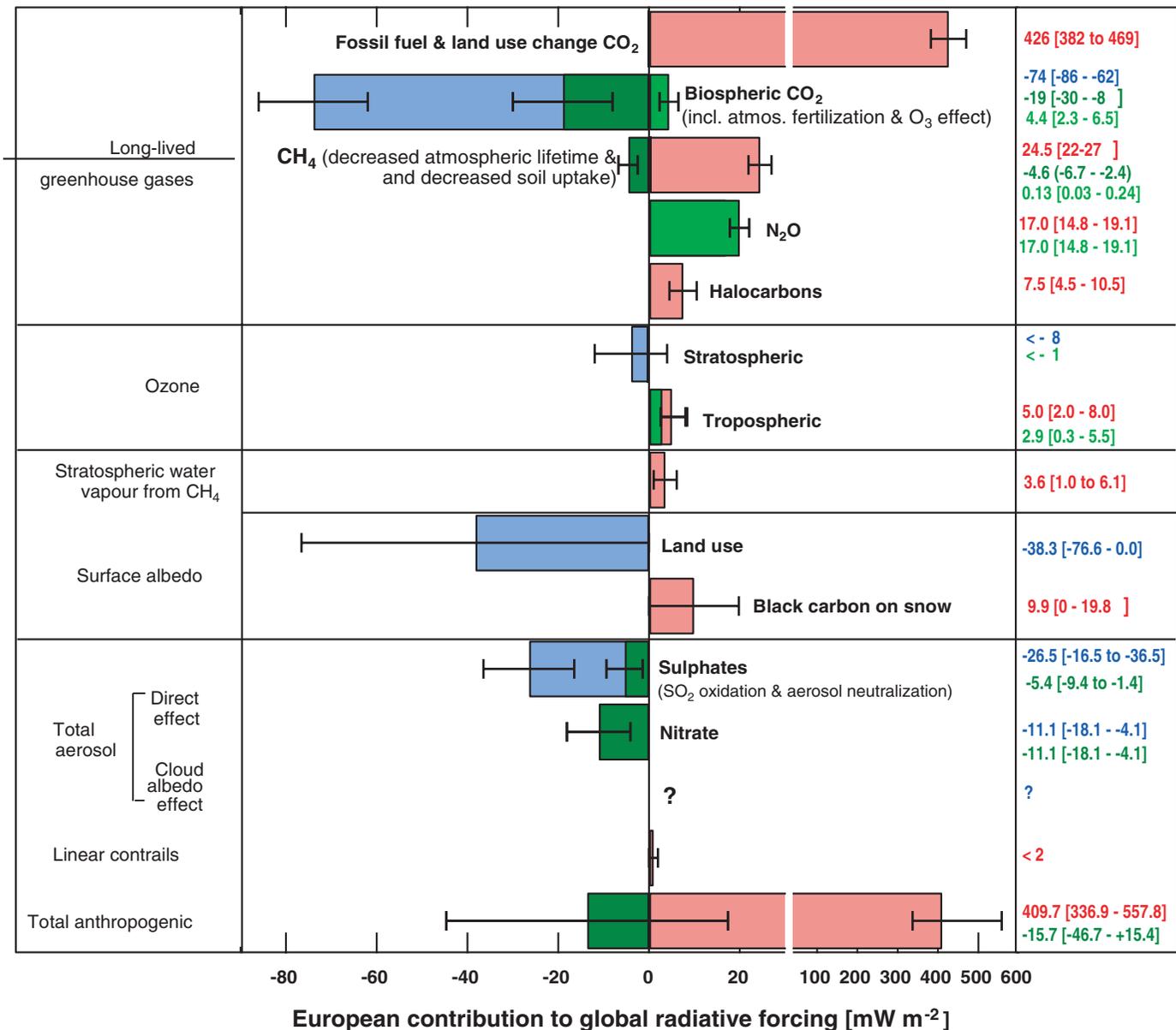


Figure 19.5 A first estimate of the change in global radiative forcing (RF) due to European emissions and the effect on European anthropogenic N_i emissions to the atmosphere. The RF components due to European anthropogenic activity have been derived as described in the text. The N_i effect is taken from Table 19.8. Red bars: positive radiative forcing; light green bars: positive radiative forcing due to direct/indirect effects of N_i; blue bars: negative radiative forcing; dark green bars: negative radiative forcing due to direct / indirect effects of N_i. For biospheric CO₂, the dark green bar represents the additional CO₂ sequestered by forests and grasslands due to N_i deposition, while the light green bar represents the decrease in productivity due to effects of enhanced O₃ caused by NO_x emissions. For CH₄ the positive (not visible) and negative contributions represent the effects of N_i in reducing CH₄ uptakes by soil and the decreased atmospheric lifetime, respectively.

about $\pm 18 \text{ mW m}^{-2}$ and are therefore of similar magnitude. While the emission of reactive nitrogen (NO_x and NH₃) has a net cooling effect, the effect of N₂O emissions is obviously warming. However, it should be noted that some of this N₂O emission is due to previous deposition of N_r from NO_x and NH₃ emissions.

For comparison, IPCC (2007, Figure 2.21 and Table 2.13) represented a first estimate of the global RF attributed to global emissions (and their changes) of individual anthropogenic precursors). This includes the effects of N₂O and NO_x emissions

on direct GHG concentrations and atmospheric chemistry, which are quantified as $+0.14$ and -0.21 W m^{-2} respectively, but does not include the effect of NH₃ emissions. A more detailed study (Shindell *et al.*, 2009) attributed a total cooling of -0.29 and -0.09 W m^{-2} to global emissions of NO_x and NH₃, respectively, the sum of which roughly balances the warming effect of N₂O emission at the global level. However, unlike the estimate presented here, neither of these two global studies considered biosphere feedbacks through the promoting CO₂ sequestration and ozone damage.

Table 19.9 Impacts of legislation on nitrogen-triggered atmospheric radiation effects and expectations beyond current implementation

Legislation/policy	Primary effect on	Relevance for radiative budget	Extension beyond current legislation
Nitrate Directive	Water quality	High (limits N inputs to soils, thus also N ₂ O emissions)	See water framework directive
Water Framework Directive	Water quality		Waters directive as such is challenging to implement
Emission Ceilings Directive/Gothenborg Protocol	Air quality – NO _x	Medium (ozone precursor)	Further abatement is technically feasible
	Air quality – NH ₃	Medium (precursor of cooling aerosol, but opportunities in coupling of N _r and C measures, e.g. improving N use efficiency)	Further abatement is technically feasible
	Air quality – PM (under discussion – not implemented yet)	Low and highly uncertain (may be either increasing or decreasing radiative effects)	Extends beyond current legislation
Common Agricultural Policy	Market regulation instrument	Medium (decreased agricultural overproduction affects N _r compounds, C and net radiative forcing)	Decreasing agricultural production subsidies may give more weight to environmental initiatives
Kyoto Protocol/UNFCCC	Greenhouse gas emissions	High (but focus currently on CO ₂ ; potential for including N _r management options)	More direct focus on agriculture as a low-cost mitigation option seems realistic

19.7 Future trends and mitigation opportunities

Reactive nitrogen compounds will continue to affect the formation and burden of radiatively active compounds in the atmosphere of the future. Scenarios of these effects deal with the trends leading to the release of precursor compounds and with measures aimed at limiting environmental effects (see Winiwarter *et al.*, 2011, Chapter 24 this volume).

The climate-relevant effects of N_r compounds have been outlined in the current chapter. They comprise not only the effects of atmospheric N₂O, but also the indirect effects of N_r on atmospheric CH₄, CO₂, O₃ and particles. Effects of any policy mitigating N_r emissions will therefore have legacy effects also for these other climatically relevant compounds. Conversely, because of the multiple interactions between these compounds, future emission limitation for any of these substances also relates to the respective contribution of N_r. The same is the case for future mitigation beyond current national or international agreements. In Table 19.9, we summarize the current international policy instruments, and their possible efficacy in relation to radiative effects and foreseeable future developments beyond current agreements.

Overall, it should be noted that while reduction of N_r emissions as N₂O is of direct benefit for European climate forcing, the situation is more complex for NO_x and NH₃ emissions. In the case of NO_x, a number of cooling effects (e.g., N fertilization and aerosol effects) and warming effects (tropospheric ozone, indirect N₂O emissions) apply, with Figure 19.5 suggesting an overall net cooling effect. For NH₃ emissions, the main effects are cooling (N fertilization and aerosol effects), which will be larger than the warming effect (indirect N₂O emissions).

In principle, therefore, control of NO_x and NH₃ emissions for other environmental reasons, such as threats to biodiversity and air quality, needs to be considered against a ‘climate penalty’, as is already well established for the control of SO₂ emissions. In this situation it is important to quantify the trade-offs between the different environmental effects of the N_r forms (see Brink *et al.*, 2011, Chapter 22 this volume).

Nevertheless, it must be emphasized that, despite the overall cooling effect of European NO_x and NH₃ emissions, it is expected that there are opportunities for reducing emissions of these pollutants that can lead to net climate benefits. This may be achieved where the measures concerned provide simultaneous reductions for several pollutants. In the case of NO_x emissions, the challenge is to identify measures that reduce both CO₂ and NO_x emissions, especially reducing overall fuel consumption. In the case of agriculture, reducing NH₃ and nitrate losses is central to improving nitrogen use efficiency and to reduce soil N₂O and NO emissions, with the potential to reduce fertilizer N_r consumption and the CO₂ emissions associated with its manufacture. Similarly, measures to trap biogas from stored manures have co-benefits for both NH₃ and CH₄ emissions. These interactions highlight the need for integrated approaches to manage to N_r, climate and other issues, as discussed by Oenema *et al.* (2011, Chapter 23 this volume).

It should be noted that, a complete balance on the effect of European activities on radiation budgets should ideally also cover the consequences of European demand on the release of N_r compounds outside Europe and their conversion to substances affecting the radiation budget. Considering this ‘footprint’ would allow us to monitor leakages, i.e., export of emission intensive activities to other countries outside Europe while importing the products for less problematic further action. In

many respects, the future development is easier to understand for the whole globe than for a sub-region (see Erisman *et al.*, 2008, who have projected the global future of the application and release of nitrogen). This is beyond the possible discussion here, especially as abatement measures beyond Europe cannot be covered.

19.8 Research needs

This chapter demonstrates that N_r emissions significantly affect biospheric and atmospheric processes that are of importance for the Earth's radiative balance in a number of ways. The effects of N_r additions to our biosphere on radiative balance are either direct – as in the case of N_2O – or (in most cases) indirect. That is to say that, in addition to the stimulation of N_2O emissions from N_r use (e.g., from soils, water bodies, or during sewage and waste treatment), other effects of N_r for ecosystem productivity and, hence, biosphere–atmosphere exchange of CO_2 , biosphere–atmosphere CH_4 exchange and atmospheric chemistry (aerosol production, O_3 chemistry, atmospheric lifetime of CH_4) are likely even more important in terms of their effect on the European radiative forcing balance.

The estimates of the individual components presented here have a large uncertainty. Taking the importance and contribution of individual processes to the European radiative balance into account, the uncertainty may be ranked as follows: aerosol N_r effects > N_2O emissions from soils and water bodies > N_r effects for biospheric CO_2 sequestration > N_r effects for O_3 chemistry > N_r effects on CH_4 exchange and atmospheric lifetime. These large uncertainties can only be reduced through targeted research, thereby overcoming classical sectorial research, e.g., by linking research communities working on atmospheric chemistry, terrestrial processes or aquatic nutrient cycling. In particular, although we have focused on the role of atmospheric emissions, N_r has even wider effects on society. Anthropogenic N_r affects world food and feed supplies, livestock numbers and human population, all of which would need to be considered in addressing the overall effect of N_r on climate balance.

It is essential that future research projects should explicitly address the importance of N_r for the radiative balance. This implicitly includes that a new view and perception of the importance of nitrogen for the radiative balance needs to be established, e.g., by specifically account for N_r effects in any greenhouse gas reporting (e.g., Kyoto protocol). Until now, the focus has been predominantly on carbon. Yet, it is obvious that nitrogen is a key threat for the European and global climate and, therefore, N_r research should become more fully integrated into the scientific assessment, societal debate and environmental policies.

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