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Reactive nitrogen in the environment and its effect on climate change

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Reactive nitrogen in the environment and its effect on climate change

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Humans have doubled levels of reactive nitrogen in circulation, largely as a result of fertilizer application and fossil fuel burning. This massive alteration of the nitrogen cycle affects climate, food security, energy security, human health and ecosystem services. Our estimates show that nitrogen currently leads to a net-cooling effect on climate with very high uncertainty. The many complex warming and cooling interactions between nitrogen and climate need to be better assessed, taking also into account the other effects of nitrogen on human health, environment and ecosystem services. Through improved nitrogen management substantial reductions in atmospheric greenhouse gas concentrations could be generated, also allowing for other co-benefits, including improving human health and improved provision of ecosystem services, for example clean air and water, and biodiversity.

Addresses

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Introduction

Several drivers, or forcing agents, can affect the Earth's biological, chemical and physical processes at a planetary scale. Humans are now the prime driver of global change, which includes climate change, reduced water quality and availability, biodiversity loss and degraded ecosystem services — the services provided by nature for free — for example, fertile soils, clean air, pollination and water

purification [e.g. [1–4,5*]]. These also affect human health.

The human drivers include a growing population and changing per capita consumption patterns both of which affect food, feed, livestock and fiber production, energy (fossil fuel and biofuel) use, land-use changes as well as social equity. Directly or indirectly, these lead to planetary-scale changes to biogeochemical cycles of nitrogen, carbon, phosphorous, sulfur, etc., which result in changes in our biological and ecological systems pushing the earth system to its planetary boundaries or even beyond [5*]. The anthropogenic induced changes of the biogeochemical cycles of carbon, C, as well as of nitrogen, N, have already resulted in changes of the atmospheric composition, with detectable consequences for the climate system. Obviously, due to the close interaction of the biogeochemical cycles human intervention creates many feedbacks such as between increased atmospheric CO₂ concentrations and increased plant C sequestration, which will have direct and indirect consequences for the rate of climate change.

Of increasing importance to these climate feedbacks are the short-lived atmospheric species (e.g. ozone, O₃; nitrogen oxides, NO_x; ammonia, NH₃; aerosols) [e.g. [6*,7–9]]. Most previous studies have addressed the O₃-aerosol impacts, including black carbon, on the radiative balance. While attention is being given to these short-lived species, to date environmental assessments, including the IPCC assessments, have largely ignored the effects of reactive nitrogen (N_r) on climate and climate feedbacks, with the exception of nitrous oxide, N₂O. This is understandable since most reactive nitrogen species such as NO_x and NH₃ are short-lived and therewith irrelevant to long-term climate forcings. Furthermore, there have been high uncertainty levels regarding the links between increased reactive nitrogen levels and the carbon cycle. But, knowledge of the role of reactive nitrogen in climate is expanding rapidly.

The element nitrogen is essential for all life forms on Earth. All plants, animals and humans, need N to live. It is essential for producing protein (N is a part of it), carbohydrates (plants need proteins to assemble carbohydrates so protein availability affects growth rate as well as rates of CO₂ fixation) and fibers [e.g. [10*,11,12]]. Nature and its biodiversity can only exist because of the availability, even if limited, of N_r in the system. N_r is defined as all

nitrogen compounds except for N_2 and includes, for example, NH_3 , nitrate (NO_3), NO_x , N_2O , amines, and organic forms of nitrogen. Naturally available N_r is produced mainly by lightning, wildfires and biological N_2 fixation (BNF). Humankind has developed technology to produce synthetic N_r fertilizer, which is used to increase crop and thereby also meat production [50]. Food for about half of the current human population can only be produced due to the availability of synthetic fertilizers [11]. Additionally, farmers increasingly introduced leguminous crops in their crop rotations, thereby increasing nitrogen input to agricultural systems via biological nitrogen fixation. Moreover, mankind also creates N_r accidentally. Fossil-fuel combustion by industry, the transportation sector and the energy sector not only emits gases such as CO_2 and sulfur dioxide, but also NO_x .

Once a N_r molecule has been created, it may remain in the environment for a considerable time. N_r is highly mobile and over time, one molecule of N_r can contribute to several environmental effects as it cascades through the environment [13^{••}]. During the cascade it drives different environmental impacts such as eutrophication and acidification in space and time, some of which affect radiative forcing (e.g. increased C-sequestration by forest ecosystems or O_3 formation due to NO_x emissions) and therefore climate. The endpoint of the cascade is ultimately

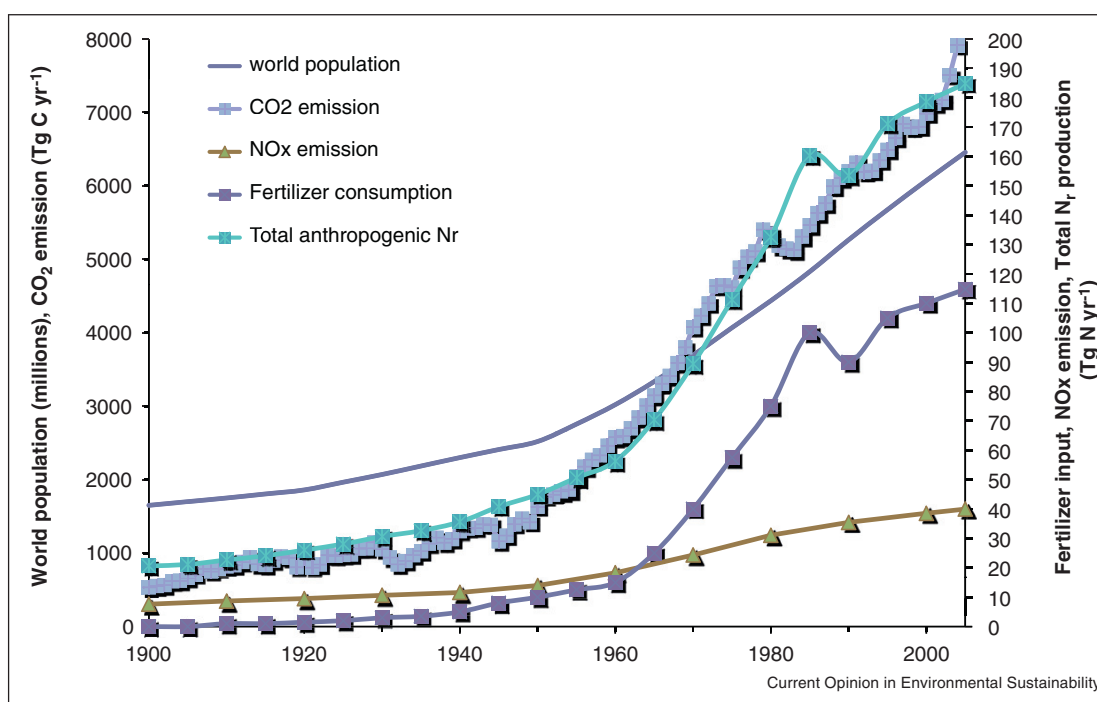
the emission of N_2 or N_2O ; the latter contributes to climate change and stratospheric ozone depletion [2,14]. During the cascade N_r can influence greenhouse gas exchange with the atmosphere, aerosol production, tropospheric O_3 , or can increase biological productivity, which requires carbon. All of these have climate impacts.

This paper provides a discussion on the status of reactive nitrogen in the environment and its effect on climate change. It first addresses the relationship between N_r production and CO_2 emissions, and then discusses why the N_r cycles are relevant for the on-going climate change debate. Then the climate impacts of N_r are quantified in terms of its effect on the radiative balance and finally the policy implications of the above are discussed.

Nitrogen creation and CO_2 emissions

In the past 50–100 years, human activities have caused the global anthropogenic rate of N_2 fixation to increase so that it is now 1.5–2 times the natural rate of BNF in terrestrial ecosystems (Figure 1) [13^{••},50]. Globally, the level of terrestrial and agricultural BNF is reasonably well known, but regionally still large uncertainties exist with regard to natural terrestrial BNF. On land the natural BNF is estimated to be about 110 Tg [15] while anthropogenic sources add 100–125 Tg N [10[•],15,50]. Estimates of N_r inputs to the open ocean suggest a natural oceanic

Figure 1



Global trends in human population, CO_2 emissions in Tg C [16] and total anthropogenic reactive nitrogen production (in Tg N) throughout the 20th century [based on [10[•],17]]. The graph also includes average fertilizer production and the increase in NO_x emissions from fossil fuel burning. The natural terrestrial BNF is about 110 Tg $N\ yr^{-1}$ and the natural oceanic BNF is about 140 Tg $N\ yr^{-1}$ [15].

BNF rate of approximately 140 Tg N yr^{-1} , while 90 Tg N yr^{-1} are added from various human derived N sources, such as atmospheric deposition and riverine inputs [15]. The proportion of anthropogenic nitrogen therefore differs between the terrestrial and the marine environment; on land the anthropogenic N_r input is higher than the natural input, but this is not yet the case in the ocean.

Figure 1 shows the global trends in N_r production. The graph depicts ammonia production for fertilizer and industry, total N_2 fixation, NO_x emissions from fossil fuel burning, together with global fossil fuel related CO_2 emissions and the world population. There is a high correlation between CO_2 emissions and N_r production because of the similarity between the drivers (food, feed and energy) and, to some extent, the sources. This shows the strong correlation between nitrogen enrichment and climate change (through CO_2 emissions) at a source level, as nitrogen is the key parameter allowing sustaining a growing world population with its increasing demand for energy from fossil fuels.

Since the 80s of the last century the human N_r production exceeds that of the natural terrestrial BNF. Furthermore, since then the graph shows a strong linear correlation between both CO_2 and N_r with the increase in world population. For each increase of the world population with one billion people another approximately 25 Tg N and $1 \text{ Pg } CO_2\text{-C}$ is added annually to the environment, which makes prediction easy if no measures are taken.

The relevance of short-lived N_r to climate

A characteristic of climate change is the long atmospheric residence time of non-reactive greenhouse gases (GHG), such as N_2O and CO_2 . The climate warming effects of the GHG are expressed in the global warming potential (GWP). CO_2 is the reference chemical for determining GWP, so it has a value of 1. N_2O has a value of 298 (i.e. a 298 times stronger GHG in terms of GWP) and CH_4 of 26 over a 100-year time horizon [2]. The change in radiative forcing due to GHG and aerosols is computed based on 'unperturbed' values, defined by the Intergovernmental Panel on Climate Change (IPCC) as the measured difference relative to the year 1750. A positive forcing (more incoming energy stays in the system) tends to warm the earth system, while a negative forcing (more outgoing energy) tends to cool it. Here we assume that the forcing through short-lived constituents can be calculated as done in [2] using their continuous supply over the last 100 years. Current policies will only take effect in the long term because measures to mitigate climate change will not reduce atmospheric concentrations of GHG. At best, concentrations will stabilize over the coming decades. Historical, present day and future anthropogenic changes of the N-cycle and effects on the biosphere-atmosphere exchange of N_r compounds are characterized mainly as

short-lived, that is, shorter atmospheric residence times compared to the lifetime of GHGs, except for N_2O .

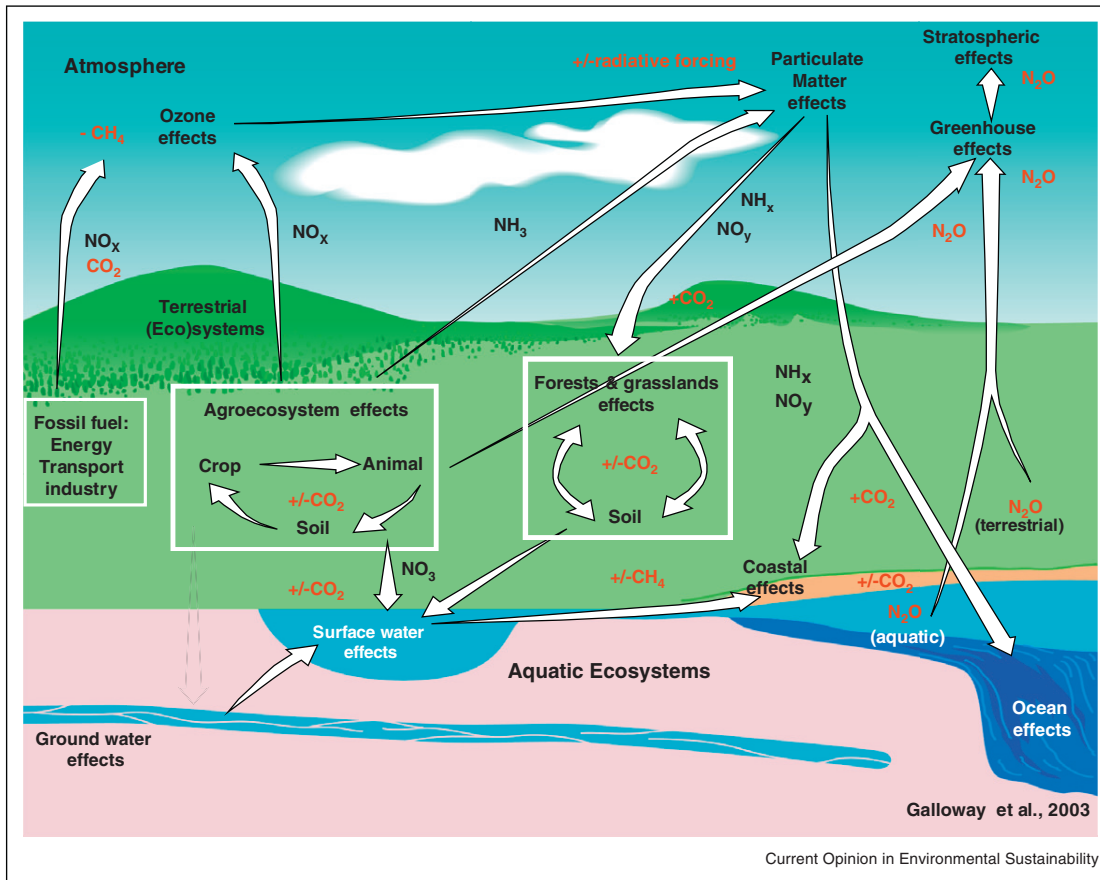
Figure 2 shows major areas where nitrogen and climate interact, visualized by the flows in the N-cycle and the greenhouse gas and radiative forcing links. The long-term effects represent the effect on carbon storage in the soil and the effects on greenhouse gas emissions, such as N_2O . N_2O can be formed and emitted in several places along the N cascade. As shown in Figure 2, N_r is involved in numerous processes forming short-lived atmospheric substances having a significant impact on the Earth's radiative balance in the short term. A good example is particulate matter or aerosols and the contribution of nitrogen to aerosol formation processes. A significant share of aerosols is, for example, ammonium sulfate or ammonium nitrate-aerosols, which are affecting the Earth's radiative balance directly (they reflect incoming solar radiation) and indirectly by affecting cloud formation. However, their lifetime in the atmosphere is in the order of hours to weeks [e.g. [2,9,18,19]].

Aerosols and other short-lived pollutants do not have the cumulative and sustained effects on climate that greenhouse gases do. Decreasing aerosol formation, therefore, has a short-term effect on climate and therefore these short-lived species are not considered relevant for the climate-change policies. The same arguments have been used for limited attention to the other changes in the N-cycle. However, there are arguments against this. Firstly, in developed countries it has been proven difficult to reduce nitrogen and other air pollutants from the sources, such as fertilizers, industry and energy. In other parts of the world these sources will increase for several decades until measures are taken. So N_r emissions in the form of NH_3 or NO_x will continue to result in an increased supply of aerosols in the atmosphere for a long time [e.g. [11]]. Secondly, the atmospheric cycle of N in aerosols might be relatively fast, but the N_r reservoirs created in other parts of the biosphere have a much longer turnover time and so a longer potential effect on GHG emissions (e.g. nitrogen accumulation in soils and respective changes in soil C:N ratios may be effective for decades) [18]. Thirdly, N_r affects the GHG concentrations in many ways as we will see from the next section and therefore, better nitrogen management, largely through improved agricultural management, will have many advantages for GHG management [17,20]

Direct and indirect links between nitrogen and climate

At different points of the cascade one nitrogen molecule can have a direct or indirect effect on GHG sources and sinks and on climate (Figure 2). Here we list the most important of these links and provide a preliminary quantification. More scientific background on these links is provided in [17,40**] and is applied for Europe in [35**].

Figure 2



Major nitrogen and climate coupling (modified from [13**]) visualized by the flows in the N-cycle and the greenhouse gas and radiative forcing links (in red).

The most important *direct links* between N_r and climate include:

- a. N_2O formation during industrial processes (e.g. fertilizer production), combustion, or microbial conversion of substrate containing nitrogen — notably after fertilizer and manure application to soils. N_2O is a strong greenhouse gas [e.g. [23*]].
- b. Ground level O_3 formation from NO_x . O_3 is the third most important greenhouse gas.
- c. Aerosol formation affecting radiative forcing, where N-containing aerosols have a direct cooling effect (which is in addition to an indirect cooling effect through cloud formation).

The most important *indirect links* between N_r and climate include:

- d. Alteration of the biospheric CO_2 sink due to increased supply of N_r . About half of the carbon that is emitted to the atmosphere is taken up by the biosphere; N_r affects net CO_2 uptake from the atmosphere in

- terrestrial systems, rivers, estuaries and the open ocean in a positive direction (by increasing productivity or reducing the rate of organic matter breakdown) and negative direction (in situations where it accelerates organic matter breakdown). CO_2 uptake in the ocean causes ocean acidification, which reduces CO_2 uptake.
- e. Excess N_r deposition either increasing or reducing ecosystem productivity and so C-sequestration. Therefore, the level of N_r production and deposition is important. For example, excess N_r leads to hypoxia and anoxia in the ocean as well as in surface waters [21]. Hypoxic zones are mostly formed along the coastlines and via stimulated denitrification more N_2O may be produced and emitted.
- f. Changes in ecosystem CH_4 production and consumption. N_r deposition to wetlands may fuel vascular plant production, thus increasing C substrate supply to the system and fuel CH_4 production. It may also change overall CH_4 consumption by bacteria and leading to an overall change in CH_4 emissions from wetlands.
- g. Changes in CH_4 production and emission from ruminants. Increased N_r supply can be associated

with more digestible diets, potentially reducing CH₄ emission from these animals.

- h. O₃ formed in the troposphere as a result of NO_x and VOC emissions reduces plant productivity, and therefore reduces CO₂ uptake from the atmosphere.
- i. O₃ effects on atmospheric OH radical concentrations and thus atmospheric lifetime of atmospheric CH₄.

Quantification of global N_r effects on climate N₂O (a)

The most direct effect of N_r on climate is through the formation of N₂O, which is responsible for 11% of the net anthropogenic radiative forcing [22]. The primary source of increasing N₂O is agriculture, largely due to the production of fertilizers and the expanding use of N fertilizers and animal manures for maintaining highly productive crop and pasture lands (e.g. [23[•],24–26]). N₂O emissions are strongly affected by the availability of N in soil, estuaries and shallow sediments [23[•],27]. There is some uncertainty in the emissions of N₂O. In addition to top-down estimates by Crutzen *et al.* [25], Davidson [23[•]] recently updated the global N₂O emissions to 19.8 Tg N₂O-N per year, which is higher than the 17.7 Tg N₂O-N in AR4 [2]. Between 1750 and 2005, the change in radiative forcing at the top of the atmosphere due to increases in global atmospheric concentrations of N₂O (a) was +0.16 W m⁻² [22].

Atmospheric influence of reactive N_r emissions (b, c, i)

For the atmospheric influence of reactive nitrogen emissions to aerosol, O₃ and CH₄ forcing we use the emission based estimates by Shindell *et al.* [19], who calculated atmospheric composition changes, historical radiative forcing, and forcing per unit of emission due to aerosol and tropospheric ozone precursor emissions in a coupled composition–climate model. They calculated the radiative forcing (from 1750 to 2000) for NH₃ to be -0.09 W m⁻² as a contribution to aerosol cooling and for NO_x a net cooling comprised 0.02 via O₃ and -0.31 W m⁻² via reduced lifetime of tropospheric CH₄ and NO₃ and SO₄ aerosols effects (net -0.29 W m⁻² for NO_x).

Carbon and nitrogen interactions in terrestrial systems (d + h)

Of the human emission of the greenhouse gas CO₂, 45% remains in the atmosphere, the remaining 55% is taken up by terrestrial ecosystems (30%) and oceans (25%) [28]. The terrestrial sink is influenced by increased nitrogen availability [29,30]. The C and the N cycles are linked through the ecosystem C:N ratio [31,32]. Increasing CO₂ uptake can be due to an increase in ecosystem C:N ratio, or due to a shift of N from soils (low C:N) to vegetation (high C:N) [32]. Uptake of atmospheric CO₂ is, therefore, limited by nitrogen availability [31,33[•]]. Several review papers [4,34,35^{••}] provide ranges in C sequestration per kg N addition in above-ground biomass and in soil organic

matter for forests and heathlands, based on: (i) empirical relations between spatial patterns of carbon uptake and influencing environmental factors including nitrogen deposition (forests only), (ii) ¹⁵N field experiments, (iii) long-term low dose N fertilizer experiments and (iv) results from ecosystem models. A summary of the results is given in Figure 3. Most studies range between 35 and 65 kg C/kg N, except for the empirical field data, which are higher but showing a large range. Furthermore, there is great uncertainty whether ecosystems will continue to retain exogenous N following N saturation [13^{••}]. The global change in radiative forcing is estimated -0.1 to -0.3 W m⁻². This does not take the CO₂ fertilization effect into account.

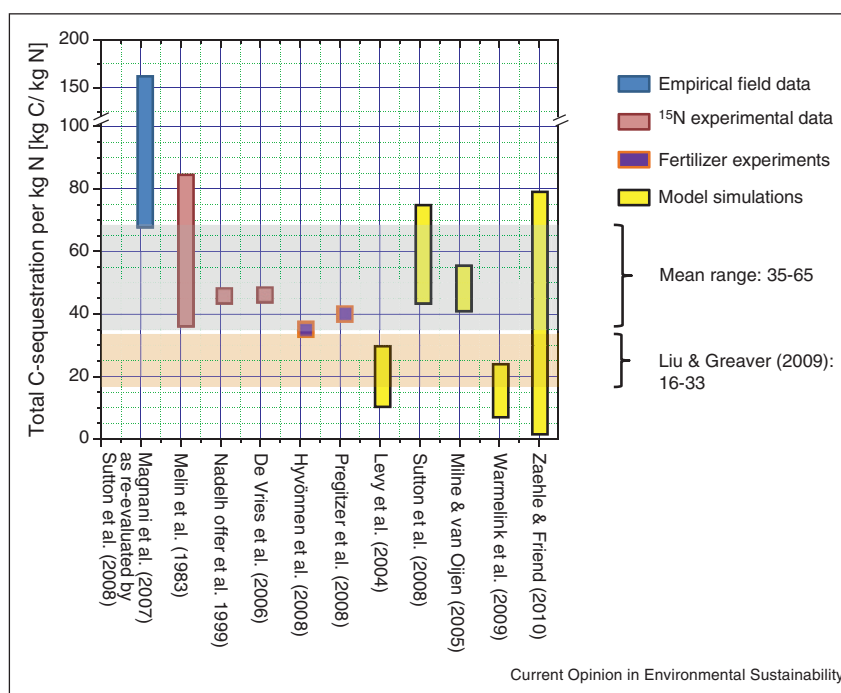
The acceleration of the natural N-cycle as a consequence of global warming is another mechanism to increase N availability to the vegetation. This mechanism works via faster litter decomposition and associated N mineralization rates [36]. Global integrated responses of net land C exchange to variation in temperature and precipitation are significantly damped by C–N cycle coupling [33[•]]. Warming will enhance soil–nitrogen mineralization, making more nitrogen available to sustain enhanced plant growth.

Several authors have evaluated the effect of including C–N coupling in carbon and/or climate models [36–41]. Anthropogenic N deposition is predicted to have increased net primary productivity due to increases in foliage area and foliage N, contributing 0.2–0.5 Pg C yr⁻¹ to the 1990s global net C uptake [39^{••}]. Simulations with coupled C–N cycles show a strong reduction of the future CO₂ fertilization effect (-0.4 to -0.8 W m⁻²) in comparison to models that do not account for terrestrial N dynamics [42^{••}]. The temperature effect due to the GHG concentration change is not taken into account.

Biophysical vegetation changes due to enhanced N availability are also likely to feedback on climate. Known mechanisms are adaptations in ecosystem composition, biodiversity and stature [36], changes in vegetation albedo at the leaf level [43] and vegetation shading of snow in high latitude forests [44]. These may change the surface energy balance, particularly when there are shifts from low vegetation to shrubs or forests. In drought regions, increased vegetation stature may lead to more sensitivity to soil moisture. In terrestrial systems close to agricultural and industrial areas the N deposition might be so high that the biodiversity is strongly reduced and even ecosystem damage occurs [10[•],11].

NO_x in the atmosphere lead to ozone production, which is damaging to crops and forests and reduces yields when critical levels are exceeded [45,46]. Loss of forest production due to excess O₃ concentrations was estimated as

Figure 3



Ranges in C sequestration per kg N addition as number of observations in above-ground biomass and in soil organic matter for forests and heathlands, based on [4,34,35**].

6% of the annual yield [45] and this was assumed to be representative for the reduced C sequestration. Arneeth *et al.* [42**] estimate that the detrimental effects of O₃ on C uptake could increase global radiative forcing by 0.08–0.17 W m⁻².

Carbon and nitrogen interactions in oceans (e)

The naturally occurring oxygen minimum zones (OMZ) in the open ocean and in coastal upwelling areas may increase in size under the continuous addition of N_r. Atmospheric deposition is a major source of nitrogen in remote areas also on the ocean [47]. Assuming today's transport pathways it is suggested that a significant increase in the magnitude or size of low oxygen areas may occur especially in the tropical and subtropical part of the ocean, which is oligotrophic today. If the areas grow in size and N-removal processes like denitrification increase, then the ratios of NO₃⁻: PO₄³⁻ may decrease. Since these waters will be circulated and upwelled to the surface ocean the low N:P ratio may affect the productivity of the ocean [47]. Moreover N₂O is generated during denitrification and the OMZ may produce large quantities of N₂O. Increasing quantities of atmospheric anthropogenic fixed N entering the open ocean could account for up to about a third of the ocean's external (non-recycled) N_r supply and up to ~3% of the annual new marine biological production, ~0.3 Pg of C per year.

The change in radiative forcing is estimated between -0.1 and -0.3 W m⁻² [47].

One of the points of interest is the coupling between terrestrial/atmospheric nitrogen inputs and ocean pH, in particular in the coastal zone. Next to stimulating primary production, the increased N_r inputs will increase denitrification in the coastal zone. Besides the effect of N₂O, denitrification will create alkalinity, which in turn raises pH (thus counteracting ocean acidification) and would permit CO₂ uptake from the atmosphere [e.g. [48]]. The alkalinity increases will, however, be on the coastal margins, and will be small relative to the overall acidification of the entire ocean.

CH₄ (f and g)

There is evidence that increased N_r deposition reduced methane oxidation in forest soils, and thus the sink strength of soils for atmospheric CH₄ [e.g. [49]]. However, this effect of increased N_r availability seems only to work in soils, which are functioning as net sinks for atmospheric CH₄. In ecosystems, which are net emitters of CH₄ (i.e. peatlands, where this really matters), increased N_r availability may stimulate low affinity CH₄-oxidation, which may result in decreased CH₄ emissions [49]. On the other hand N_r deposition to wetlands may increase plant production and, thus, substrate supply

Table 1

The best estimate influence of nitrogen on the radiative forcing at the global scale (W m^{-2})

Process	Forcing (W m^{-2})	Remark
N_2O emission	0.16	[2]
NO_x emission atmosphere	-0.09	[19]
NH_3 emission atmosphere	-0.29	[19]
N and C in terrestrial systems and oceans	-0.2	Terrestrial systems [39**,42**]
	-0.2	Oceans [47]
	0.25	Mineralization [42**]
O_3 phytotoxicity	0.13	[42**]
CH_4	0	Negligible. Effect on the atmospheric lifetime is included in NO_x emissions
Total	-0.24	

for methanogenic bacteria, thereby increasing CH_4 production and emission. On the basis of a detailed literature review Liu and Greaver [34] calculated an emission factor of $0.008 \pm 0.004 \text{ kg CH}_4\text{-C/kg N}$, which is an order of magnitude smaller than the emission factors in Figure 3 and can be neglected.

We do not have estimates of changes in CH_4 production and emission from ruminants as the result of more feed through more fertilizer N availability.

Table 1 summarizes the overall effect of nitrogen on the radiative forcing based on the estimations given above. We estimate an overall small net effect of -0.24 W m^{-2} , with a range of $+0.2$ to -0.5 W m^{-2} , based on the uncertainty ranges estimated for Europe [35**]. The uncertainty in the different components is very high, especially in the atmospheric components due to the short lifetime and in the deposition effect due to the uncertainty in deposition estimates and the nutrient cycling in different systems [35**]. Furthermore, most of the data are obtained from the developed parts of the world and data from developing areas are scarce.

There are many more (in)direct interlinkages, which are unquantified, small or negligible, including NH_3 as a greenhouse gas; industrial formation and release of nitrogen trifluoride (NF_3), which is a very strong greenhouse gas; the role of organic N in the N-cycle and its relation to the carbon cycle; reduction in soil microbes, CO_2 and water uptake in areas where excess N deposition leads to nutrient imbalances in these ecosystems; manure storage processing producing biogas and CH_4 leakages resulting from these facilities; interaction between N-fertilizer, agricultural productivity and land use change; the NH_3 -based chemical industry affecting climate during the processes and from the wastes; etc. The overall impact of these linkages is unknown.

IPCC estimated the current net radiative forcing to be about 1.5 W m^{-2} and N has already been taken into account for N_2O and aerosols [2]. Our estimate of 0.24 W m^{-2} is significant, but due to the high uncertainty

and regional variability it needs to be better investigated. It shows, however, that since there is a balance between positive and negative influences of different sources on different impacts on the N-cycle, it is important to focus policies both from the environmental side as well as from the climate side in such a way that the maximum positive results are obtained.

Conclusions and policy implications

Initial quantifications of the effect of N_r on climate show that there are many positive and negative influences on the GHG balance and the radiative forcing, which compensate each other leading to a small net cooling effect of -0.24 W m^{-2} , with a range of $+0.2$ to -0.5 W m^{-2} . The uncertainty in the individual processes is very large and given this uncertainty it is safe to say that there is a balance between cooling and warming effect of N which is slightly more on the cooling side, especially in the Northern Hemisphere. Ammonia emissions contribute most to the cooling effect mainly through the aerosol formation and to the N-deposition, which stimulates C sequestration. NO_x has more interactions and lead to a small net cooling effect. The warming effect of N_r is mainly through N_2O emissions. For the long-term, if emission scenarios to reduce N_r would be successful the effect could be significant. However, the radiative forcing of N_r depends on the location and source type of the N_r losses and, thus, a simple upscaling approach of effects of reductions of N_r on global radiative balance remain difficult.

The effects of increased N_r in the environment have led to policies limiting emissions of N_r to the air and losses to soil and waters. Current policies on reducing N_r emissions mainly concentrate on NO_x emissions to the atmosphere. Policies on agricultural losses of N_r are necessary to reduce the warming impacts mainly through N_2O and to reduce the other impacts on human health and the environment. However, N management options need to be assessed to make sure that they lead to a net benefit for climate and at the same time to improved environmental quality and ecosystem services. The effects of increasing concentrations of GHGs have led to policies reducing its

emissions. The drivers of nitrogen and climate are essentially the same and there is merit in finding the synergies between mitigation and adaptation policies. These are important reasons why the N cycle should be taken into account in discussions on climate and to take advantage of positive synergetic effects to address global change issues. This, however, requires an integral and multi-disciplinary approach including: multi-source/actor, multi-pollutant, multi-problem, multi-benefits, multi-receptor, multi-effect, multi-scales, etc.

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- of outstanding interest

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