Consequences of human modification of the global nitrogen cycle

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Consequences of human modification of the global nitrogen cycle

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The demand for more food is increasing fertilizer and land use, and the demand for more energy is increasing fossil fuel combustion, leading to enhanced losses of reactive nitrogen (Nr) to the environment. Many thresholds for human and ecosystem health have been exceeded owing to Nr pollution, including those for drinking water (nitrates), air quality (smog, particulate matter, ground-level ozone), freshwater eutrophication, biodiversity loss, stratospheric ozone depletion, climate change and coastal ecosystems (dead zones). Each of these environmental effects can be magnified by the 'nitrogen cascade': a single atom of Nr can trigger a cascade of negative environmental impacts in sequence. Here, we provide an overview of the impact of Nr on the environment and human health, including an assessment of the magnitude of different environmental problems, and the relative importance of Nr as a contributor to each problem. In some cases, Nr loss to the environment is the key driver of effects (e.g. terrestrial and coastal eutrophication, nitrous oxide emissions), whereas in some other situations nitrogen represents a key contributor exacerbating a wider problem (e.g. freshwater pollution, biodiversity loss). In this way, the central role of nitrogen can remain hidden, even though it actually underpins many trans-boundary pollution problems.

1. Introduction Q2

Reactive nitrogen (Nr) is created from N_2 naturally by biological nitrogen fixation, biomass burning and lightning. Nitrogen is an essential nutrient for the growth and functioning of plants, animals and humans, and an essential element for food security [1,2]. However, limited amounts of natural nitrogen fixation have led to the world's ecosystems becoming adapted to low rates of Nr supply, with limited productivity but high biodiversity. Because of their limited availability, some essential Nr molecules are efficiently conserved and re-used in most natural environments. Nitrogen is commonly a limiting factor for the production of food. Humankind has sought different ways to increase crop production to provide food to sustain a growing population. This has led to the development of synthetic fertilizer production based on the Haber–Bosch process [3,4]. Nitrogen currently provides many benefits to society, in particular to agriculture and industry [5].

While the industrial fixation of N_2 is essential for food production, it is not without costs for the environment and human health. The amount of Nr used to produce food is on average about 10-fold higher than its consumption, owing to inefficiencies in the food production–processing–consumption chain [6–8].

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Table 1. Overview of Nr emission-related health impacts and a summary of limit values for concentrations in air, as set by EU policies [9,10]. AOT40, accumulated exposure over a threshold ozone concentration of 40 ppb; MAC, maximum allowable concentration.

emission	routes	health impacts	indicators	limit values/targets
NO_x	inhalation: — direct impacts of NO_2 — impacts via O_3 — impacts via PM visibility (PM)	asthma, respiratory disorder, inflammation of air ways, reduced lung functions, bronchitis, cancers	NO_x	40 $\mu g\ m^{-3}$ (annual mean) 200 $\mu g\ m^{-3}$ (hourly mean) 400 $\mu g\ m^{-3}$ (threshold, 3 hrs) 30 $\mu g\ m^{-3}$ (annual mean; plant damage)
			O_3	180–240 $\mu g\ m^{-3}$ (hourly mean) AOT40 120 $\mu g\ m^{-3}$ (hourly mean)
NH_3	inhalation: — direct impacts (negligible) — impacts via PM	see NO_x		18 ppm (MAC value) 1 $\mu g\ m^{-3}$ (annual mean, plants)
	odour	modest odour contribution		—
N_2O	health impact owing to global warming, often enhanced by eutrophication health impact owing to loss of stratospheric ozone depletion	enhancement of vectors for infectious diseases (e.g. malaria) and frequency of infestations (e.g. algae blooms, insects)		—

Agricultural sources of Nr produce atmospheric emissions of ammonia (NH_3), nitrogen oxides (NO_x) and nitrous oxide (N_2O) from agriculture to the air, and nitrate (NO_3) to groundwater [8]. At the same time, combustion processes in energy production, transport and industry have led to the formation of new Nr through the emission of NO_x as an unintentional waste product.

Nr is highly mobile. Most of it dissipates into the environment and cascades through air, waters and terrestrial ecosystems where it contributes to a multitude of effects, including adverse impacts on human health, ecosystem services, biodiversity and climate change [4,7,8]. The endpoint of the cascade is ultimately the conversion back to unreactive N_2 gas, although Nr is being produced more rapidly than it is being converted back to N_2, so in many regions Nr is accumulating in the environment.

Overall, as human fixation of nitrogen continues to rise, the direct public health benefits through food production will probably continue to rise. However, the negative health consequences on ecosystems and people may become more diverse, and might in total increase more rapidly than the benefits.

This paper presents an overview of the consequences of Nr in the global environment. In the following sections, we elaborate in more detail on the impacts of Nr on (i) air quality and human health, (ii) aquatic and terrestrial ecosystems, and (iii) climate change, including global overviews whenever available.

2. Impacts on air, water quality and human health

(a) Impacts on human health: air quality

When released into the lower atmosphere, NO_x can increase tropospheric ozone (O_3) formation, smog, particulate matter (PM) and aerosols. Particulate nitrate can be formed following the oxidation of NO_2 to nitric acid (HNO_3), which can then further react with NH_3 to form ammonium nitrate, $NH_4NO_3 \cdot NO_3$ and NH_4 are two of the major inorganic components in urban aerosol particles. In the atmosphere, NH_3 reacts not only with HNO_3 but also with aerosols and other acid gases such as H_2SO_4 and HCl to form ammonium-containing particles (e.g. $(NH_4)_2SO_4$; NH_4NO_3; NH_4Cl). NO_x can also contribute to formation of the secondary organic aerosol particles in photochemical smog. **Q3**

Table 1 provides an overview of N-related health impacts. The direct impacts of NH_3 are mainly of importance within or close to Nr sources such as animal housing units or manure storage tanks. Direct impacts of NO_x exposure and indirect impacts through PM and O_3 exposure are most important for health. NO_2 is an irritant gas and can cause severe damage to the lungs if inhaled. High indoor NO_2 levels can also induce a variety of respiratory illnesses. Concentrations above 60–150 ppm can cause coughing and a burning sensation deep inside the lungs. Damage to the lungs can be visible after 2 to 24 h. These concentrations are, however, an order of magnitude higher than ambient levels and occur in special conditions. Continuous exposure to low concentrations of NO_2 can cause a cough, headache, loss of appetite and stomach problems. Environmental studies have shown that children exposed to chronically elevated NO_2 in their environment are more likely to develop respiratory diseases and reduced breathing efficiency [9].

Concentrations of NO_2 are often strongly correlated with those of other toxic pollutants with similar sources in industry and transport, and, being relatively simple to measure, NO_2 is often used as a surrogate for the pollutant mixture as a whole [10]. Achieving guideline concentrations for individual pollutants such as NO_2 may, therefore, reduce the level of many other pollutants that have the same source, bringing public health benefits that exceed those anticipated on the basis of estimates of a single pollutant's toxicity.

O_3 is an important pollutant affecting human health, almost exclusively through inhalation [9,11]. Adverse health impacts that can be initiated and exacerbated by O_3 exposure include coughs and asthma, short-term reductions in lung function and chronic respiratory disease [9,11]. A recent overview of health risks of ozone by the World Health Organization (WHO) indicates a clear increase in mortality and respiratory morbidity rates with increasing levels of ozone in the environment [12]. An estimated 21 000 premature deaths in the EU member states are associated with ozone levels exceeding a maximum daily 8-h average of 35 ppb. Ozone is also associated with 14 000 respiratory hospital admissions annually in the EU member states [12]. A statistically significant increase in mortality risk has been observed at O_3 concentrations above 70 $\mu g\ m^{-3}$ (35 ppb). Outdoor air pollution contributes to 5 per cent of all cardiopulmonary deaths worldwide [13]. In many countries approximately 20–40 deaths per 100 000 population are reported to be due to cardiopulmonary illness. In 2008, urban outdoor air pollution was responsible for an estimated 1.3 million annual deaths, representing 2.4 per cent of the total deaths in the world, mainly in the Eur-Asia region and in urban areas. Worldwide, urban air pollution is estimated to cause about 9 per cent of lung cancer deaths, 5 per cent of cardiopulmonary deaths and about 1 per cent of respiratory infection deaths [13].

PM is the most significant contributor to adverse health effects from air pollution [13]. It is an environmental health problem that affects people worldwide, but middle-income countries disproportionately experience this burden. According to a recent study on behalf of the European Environment Agency, pollution of fine particles is associated with more than 455 000 premature deaths every year in the EU27 member states [14]. The scattering and absorbing of light owing to particles also affects visibility in cities and scenic areas.

Apart from direct effects on humans, ozone damages crops and forests, and leads to reduced agricultural yields [15–17]. O_3 is absorbed into plants via stomatal pores on the leaf that open during the day to allow CO_2 absorption for photosynthesis and evaporation of water. O_3 damages cell walls and membranes leading to cell death and reduction in photosynthesis rates [17]. This negatively affects crop and horticultural plant yields and CO_2 uptake. Global relative yield losses owing to ozone exposure are estimated to range from 7 to 12 per cent for wheat, 3 to 4 per cent for rice, 3 to 5 per cent for maize and 6 to 16 per cent for soybeans. In Europe, the regionally aggregated yield losses for these crops are estimated to be 5 per cent, 4 per cent, 5 per cent and 27 per cent, respectively [15].

(b) Impacts on human health: nitrogen enrichment of drinking water and food

It is important to note that a healthy immune system requires adequate nutrition, thus one of the most important links between fixed nitrogen and many tropical diseases may be that better access to nutrients in undernourished regions increases the overall health and disease resistance of the population [18]. However, when in excess, different forms of Nr can cause human health problems.

Nitrate pollution of groundwater poses a recognized risk to human health. The WHO standard for drinking water is 50 mg $NO_3^-\ l^{-1}$ (as NO_3^-) for short-term exposure, and 3 mg $NO_3^-\ l^{-1}$ for chronic effects [13]. Agriculture puts the largest pressure on both groundwater and surface water pollution owing to reactive N [19,20]. Although nitrate concentrations have slightly decreased over the past decades in some European rivers, levels have remained high in others and, overall, nitrate levels in groundwater have remained constant. Although some improvements have been made in reducing nutrient inputs from wastewater discharge, diffuse pollution of agricultural origin remains a major threat for waters in the EU [19]. From 2000 to 2003, nearly 40 per cent of the groundwater monitoring stations in the EU exceeded average values of 25 mg $NO_3^-\ l^{-1}$, and almost 50 per cent of the surface water monitoring stations exceeded average values of 10 mg $NO_3^-\ l^{-1}$ [19]. Similar high levels occur in other parts of the world where high levels of fertilizer are used [20].

There are other impacts related to the intake of Nr through our food system. Diets in developed countries generally contain more protein than required for human health [8,21,22]. The WHO reports that current knowledge of the relationship between protein intake and health is insufficient to enable clear recommendations about either optimal intakes for long-term health or to define a safe upper limit [22]. High protein uptake can also lead to high urea production and elevated blood pH, leading to an overreaction of the immune system. Furthermore, the kidneys can be overloaded causing possible kidney failure. A high blood pH can lead to loss of bone mass. Gout has also been associated with high purine foods such as meat [22].

People normally consume more nitrates from vegetables than from cured meat products. Spinach, beets, radishes, celery and cabbages are among the vegetables that generally contain very high concentrations of nitrates [23]. Nitrates can be reduced to nitrites by certain micro-organisms present in foods and in the gastrointestinal tract. This has resulted in nitrite toxicity in infants fed vegetables with a high nitrate level. No evidence currently exists implicating nitrite itself or nitrate as a carcinogen [24]. There are both experimental and epidemiologic studies that indicate possible chronic health effects associated with consumption of elevated levels of nitrate in drinking water, although results are inconsistent. Likewise, there are no good estimates of damage to health related to methaemoglobinemia owing to drinking water nitrate. Evidence is emerging for possible benefits of nitrate or nitrite as a potential pharmacological tool for cardiovascular health [18].

The available evidence supports a positive association between nitrite and nitrosamine intake and gastric cancer, between meat and processed meat intake and gastric and oesophageal cancer, and between preserved fish, vegetable and smoked food intake and gastric cancer, but is not conclusive **Q4** [25]. A diet high in red meat is associated with the formation of nitrosamines through the additives (sodium nitrite) that increase the red colour of the meat. The natural breakdown products of proteins can combine with nitrites to form compounds such as nitrosamines. There are many different types of nitrosamines, most of which are known carcinogens in test animals. It is unknown at what levels, if any, nitrosamines are formed in humans after they eat cured meat products, or what constitutes a dangerous level in meat or in humans.

3. Impacts of Nr on natural ecosystems

Nr can both acidify and eutrophy ecosystems. The impact of Nr on a species or ecosystem depends on several factors, including the duration of exposure, total amount and form of nitrogen;

the sensitivity of the species; and intrinsic ecosystem properties such as fertility and acid neutralizing capacity [26]. High concentrations of Nr (especially reduced N) can be toxic to organisms that adsorb elements directly from the environment, such as sensitive algae, lichens or bryophytes [27]. More commonly, Nr acts indirectly on organisms through factors such as nutrient enrichment, oxygen depletion (in aquatic ecosystems), soil or water acidification, altering nutrient ratios, or intensifying the impact of other stressors such as pathogens or climate change. In this section, major impacts of Nr on aquatic and terrestrial ecosystems are presented.

(a) Aquatic ecosystems

(i) Acidification

Aquatic ecosystems with a low acid neutralizing capacity (primarily freshwater) can be acidified by atmospheric deposition of reactive N and S. With a sharp decline in sulfur emissions beginning in the mid-1980s, Nr has become the major component of acidic deposition in many areas of Europe and North America, and a growing problem in many developing countries. With persistent acidification, species composition at the base of the food chain is shifted, and often simplified, to favour acid-tolerant macrophytes and phytoplankton. Early life stages of fish and aquatic invertebrates can be especially sensitive to acidification, but direct and indirect impacts have been reported at all higher trophic levels, including zooplankton, benthic invertebrates, amphibians and birds [28,29].

(ii) Eutrophication

Nutrient enrichment of freshwater and coastal ecosystems usually originates from surface sources such as fertilizer runoff, erosion of nutrient-rich sediments or sewage discharge. In oligotrophic ecosystems, biomass or diversity may increase with increasing nutrient load [30]. However, as levels of Nr and P increase, phytoplankton capable of efficiently assimilating these nutrients are increasingly favoured over species more limited by other factors (e.g. diatoms, requiring silica, or benthic primary producers, requiring light). Low-diversity algal or cyanobacterial blooms can result, leading to surface water hypoxia and the release of toxic compounds. This in turn impacts sensitive higher trophic level organisms, such as invertebrates and fish [27,31]. Sedimentation and decomposition of biomass from phytoplankton blooms can deplete oxygen in bottom waters and surface sediments, especially in ecosystems with low rates of water turnover [31]. This further shifts the benthic community towards fewer tolerant species. Changes in the benthic community alter nutrient cycling in the sediments and overlying water, feeding back to further alter the rest of the aquatic ecosystem ([32], and references therein).

Coastal eutrophication has recently emerged as a global issue of serious concern, with a steady growth in the extent and persistence of eutrophic, hypoxic and anoxic coastal waters [31,33], and related incidences of toxic algal blooms such as red tides [34]. Coral reefs, sea grass beds, wild or farmed fish and shellfish can be particularly sensitive to eutrophication and oxygen depletion. Similar to acidification, impacts on lower trophic levels can move up the food chain to seabirds, mammals and other marine animals.

A recent estimate identifies 415 eutrophic and hypoxic coastal systems worldwide [33], with 169 documented hypoxic areas, 233 areas of concern and only 13 systems in recovery. These numbers are very likely underestimates owing to low data availability in many areas, particularly Asia, Africa, Latin America and the Caribbean. The most underrepresented region is likely Asia, with relatively few documented eutrophic and hypoxic areas despite large increases in intensive farming methods, industrial development and population growth over the past 20 years.

(b) Terrestrial ecosystems

In high concentrations, Nr can cause direct foliar damage, primarily to lower plants. NH_3, NO_x and NH_4^+ are especially phytotoxic [35]. This is a particular problem downwind of direct sources such as intensive livestock production. Whereas direct foliar damage is usually due to high local concentrations of Nr, broader ecosystem-scale changes to soil and vegetation often arise from chronically elevated regional Nr deposition. Nr is the limiting nutrient for plant growth in many natural and semi-natural terrestrial ecosystems. Over time, species composition changes, and diversity often declines, as characteristic species of oligotrophic, mesotrophic or circumneutral habitats are out-competed by more nitrophilic or acid-resistant plants. Forbs, bryophytes, lichens and nutrient-poor shrubs are the most impacted functional types; graminoids adapted to higher nutrient levels are the main beneficiaries of elevated Nr deposition.

Chronically elevated Nr deposition can also enhance susceptibility to stress, such as frost damage, herbivory or disease ([36] and references therein). Northern temperate, boreal, arctic, alpine, grassland, savannah and Mediterranean biomes are particularly sensitive to Nr deposition [37]. As with aquatic ecosystems, effects of Nr have been identified at all trophic levels, including indirect impacts on above-ground fauna such as insects and birds.

Within the soil, Nr fertilization can reduce the allocation of organic carbon from the vegetation to mycorrhizal fungi, because the increasing supply of Nr from above reduces the plant's dependence on mycorrhizae for scavenging Nr from the soil [38]. Free-living fungi and N-fixing bacteria are also sensitive to Nr. Changes in the microbial community in turn impact soil processes such as organic matter mineralization and nutrient cycling. The soil fauna—protozoa, worms, insect larvae, etc.—primarily react to Nr indirectly, through changes in the microbial community, microbial-driven processes or vegetation growth and composition [39,40]. Changes in macrobiota in turn influence the physical properties of soil, such as soil aggregation, water infiltration and organic matter turnover [41,42].

Exceedance of critical loads for nutrient nitrogen is linked to reduced plant species richness in a broad range of ecosystems and 5–10 kg N ha^{-1} yr^{-1} has been used as a general threshold value, although effects may occur over the long-term at lower levels ([26,36] and references therein). Combining global modelled N deposition with the spatial distribution of protected areas (PAs) under the convention on biological diversity, Bleeker *et al*. [43] showed that 40 per cent of all PAs (11% by area) are projected to receive N deposition higher than 10 kg N ha^{-1} yr^{-1} by 2030 (figure 1). These cover almost all of southern Asia and the eastern US, as well as parts of Africa and South America. **Q5**

(i) Ozone exposure

As with impacts on food crops (described above), O_3 also affects natural ecosystems. The most prominent effect is that

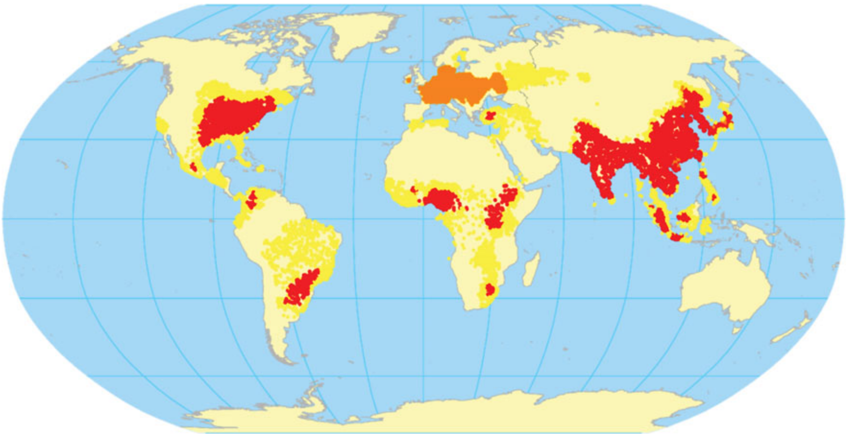

Figure 1. Distribution of Nr deposition classes and exceedance of deposition levels in the period 2000–2030 on Protected Areas (PAs) under the Convention on Biological Diversity [41]. Red PAs show an exceedance of 10 kg N ha^{-1} yr^{-1} and deposition 2030 higher than 2000, Orange PAs show a current exceedance, but deposition in 2030 lower than 2000. Yellow PAs might be under threat in the near future since Nr deposition exceeds 5 kg N ha^{-1} yr^{-1}, but is increasing over the period 2000–2030.

it reduces forest productivity and thereby carbon sequestration. In a meta-analysis, Wittig *et al*. [44] estimated the magnitude of the impacts of current and future O_3 concentrations on the biomass, growth, physiology and biochemistry of trees representative of northern hemisphere forests. They found that current ambient O_3 concentrations (40 ppb on average) significantly reduced the total biomass of trees by 7 per cent compared with trees grown in charcoal-filtered controls, which approximates pre-industrial O_3 concentrations. Their results are in line with estimates of forest yield losses of 6 per cent for Norway spruce in Europe owing to current exceedances of O_3 critical levels [16,45].

(c) Interactions

Nr also acts with other human-influenced impacts on natural ecosystems, such as land-use change, climate change, sulfur deposition, ground-level O_3 enrichment and exotic species invasion. Research on the impacts of multiple stressors is increasing, but much more understanding is needed; an in-depth review of the existing studies is beyond the scope of this paper. Effects may be additive, synergistic or antagonistic. For instance, a recent evaluation suggests that enhanced exposure of ground-level O_3 to acid grasslands in Europe may impact different plants, and in different ways, than Nr deposition, thus making Nr an additive stress [46].

4. Reactive nitrogen and climate change

Nr has many direct and indirect links to climate (summarized by Erisman *et al*. [47]). The most important warming effects of Nr on climate are:

— N_2O formation during industrial fertilizer production, incomplete combustion or microbial denitrification and nitrification—notably after fertilizer and manure application to soils. Excess Nr can also lead to hypoxia and anoxia in the ocean and surface waters, enhancing rates of denitrification and N_2O release;

— ground-level O_3 formation from NO_x. O_3 is an important greenhouse gas. It is also formed in the troposphere as a result of NO_x and VOC emissions. O_3 reduces plant productivity, and therefore reduces CO_2 uptake from the atmosphere; and

— changes in ecosystem CH_4 production and consumption. Nr deposition to wetlands may increase vascular plant production, thus increasing root exudation of low-molecular weight carbon compounds such as acetate, a major substrate source for some groups of methanogenic Archaea. A shift towards vascular plants such as sedges also increases the rate of release of CH_4 to the atmosphere through stems, bypassing CH_4 oxidation in the soil. Nr may also increase rates of CH_4 consumption by methanotrophic bacteria in wetlands; however, the opposite may be the case in upland soils, with the balance depending on the background levels of both CH_4 and nutrients.

The most important cooling effects of Nr on climate include:

— enhancement of the biospheric CO_2 sink owing to increased supply of Nr. Because N (often together with P) is commonly a growth-limiting element, increased Nr increases primary productivity, and thus CO_2 uptake from the atmosphere, in many terrestrial ecosystems, rivers, estuaries and areas of the open ocean. Nr may reduce productivity in very high N deposition areas; however, these are fairly rare. Increasing Nr may increase or reduce the rate of organic matter breakdown, dependent upon the background level of Nr in the environment and the type of organic matter. Estimates of the quantitative importance of N on carbon sequestration vary widely;

— N-containing aerosols. This occurs both directly via absorbing terrestrial radiation and scattering solar radiation, and indirectly, e.g. by influencing cloud formation;

— changes in CH_4 production and emission from ruminants. Increased Nr supply can be associated with more digestible diets, potentially reducing CH_4 emission from these animals. This effect is, however, small; and

— effects of O_3 on CH_4. Elevated tropospheric O_3 increases the formation of the hydroxyl radical (·OH), which is a major sink for atmospheric CH_4. However, O_3 can also reduce the emission of CH_4 from wetland plants, possibly by impacting photosynthesis and reducing root exudation of carbon [42].

Q6

Table 2. Societal costs of nitrogen emissions in ranges based on the references [49–51]. Units are euro per kg Nr.

Nr flux	health	ecosystem/ coastal systems	crop decline O_3	climate	total
NO_x–N to air	10–30	2–10	1–3		13–43
NH_3–N to air	1,1–20	2–10			3,1–30
Nr to water	0–4	5–50			5–54
N_2O–N to air	1–3			1–15	2–18

Estimating the net effect of these major interactions between Nr on climate at the global scale, Erisman *et al*. [47] calculated an overall small net cooling effect of $-0.24\ W\ m^{-2}$, but with a large uncertainty range of -0.5 to $+0.2\ W\ m^{-2}$. This cooling effect should not be taken as indicating that Nr is not an issue for climate policies. We should conclude from this that, whatever measures are taken that affect Nr emissions, potential climate effects should be evaluated to insure that the role of Nr does not change to make it more of a contributor to climate warming. Furthermore, it is very relevant to include the climate effect when addressing Nr reduction for environmental reasons in order to prevent trade-offs or pollutant (issue) swapping. The reason for this is that environmental policies will affect different sources and sectors, which all contribute to only a part of the N-cycle.

Climate change is, however, of central importance to the Nr budget. At a direct level, enriching the atmosphere in CO_2 can enhance net primary production rates and thus accelerate nitrogen cycling. In addition, when the climate changes, many factors such as temperature, precipitation, run-off, sea level, ocean chemistry and wind may change: these factors can strongly influence nitrogen/nutrient dynamics. For example, nitrification rates in the ocean appear to be reduced by ocean acidification resulting from increased CO_2 dissolution [48]. The consequence of this is not only reduced availability of nitrogen for phytoplankton and other micro-organisms, but also a reduction in N_2O emissions.

(a) Cost–benefit analysis for Nr

Comparing the societal costs of different effects of Nr provides a means of evaluating these different effects on the same scale. Recently cost–benefit analyses of Nr have been attempted for the Chesapeake Bay in the US [49], for Europe [50] and as a broad overview for the US [51]. Table 2 shows the ranges of estimated societal costs per Nr component loss and impact, based on the 'willingness to pay' method [50]. Based on these costs, the most important component of the Nr cycle is the emission of NO_x, owing to the health impacts of both particulates and ozone. Ammonia is also important, but the health effects are less certain. There is a large uncertainty for the cost of Nr enhancement of surface- and groundwater. Brink *et al*. [50] estimated that the agricultural benefits of Nr in Europe are €25 and €130 billion per year, whereas the total environmental costs based on the numbers in table 2 adds up to €13 and €65 billion per year and are appreciable compared with the benefits.

(b) Synthesis and importance of the Nr effects

Figure 2 is a first attempt by expert judgement to describe the major consequences of human induced Nr losses to the environment as synthesized in this paper. This figure shows two parameters: (i) the exceedance of the effects levels of Nr for ecosystems or human population and (ii) the contribution of Nr to the total effect, relative to other components or causes (e.g. natural) of the problem. For each problem an attempt is made to define the level above which effects are expected and its exceedance. The figure extends from the local scale, through the regional scale, to the global/stratospheric scale and hence, represents the cascade of Nr through the environment. Overall the figure provides direct insight in where Nr is an issue and if it needs attention based on the exceedance and relative to the other stresses needs to be addressed. Note that the two parameters are not necessarily related! The following problems are included and explained:

- *Nitrate or nitrite intake by humans*. The figure describes the estimated fraction of the global population with a nitrate or nitrite intake above-recommended levels. The intake comes from drinking water with excess nitrate, air pollution inhalation of nitrate particles and nitrate in food. Food and drinking water are by far the major sources for nitrate, and cured meat is the major source for nitrite [22]. We estimate that about 70 per cent of the global population has a higher intake than recommended. The human induced Nr share of the total intake of health-impacting substances through food and drinking water is large at 80 per cent; 20 per cent being of natural origin.
- *Air pollution* (*human health*). This is expressed as the fraction of the world's the population exposed to levels above health thresholds, such as described by WHO [9,10] (table 1). According to WHO 60 per cent of the global population in urban areas is exposed to PM, NO_2, and other toxic (N) substances (such as nitrosamines) at levels above the thresholds, and a substantial fraction of rural dwellers are exposed to O_3 and PM levels above the thresholds. Nitrogen constitutes a major source of O_3 precursor emissions: 60 per cent of the O_3 increase since 1900 is due to an increase in NO_x, with the remaining owing to an increase in emissions of CO, CH_4 and NMVOC [10]. Nr globally contributes about 20 per Q6 cent to the formation of fine particles [13]. All health impact assessments (e.g. by WHO) show that particle pollution dominates total health impacts (approx. 95%).
- *Air pollution* (*crop loss*). Crop loss owing to air pollution is mainly caused by increased levels of surface O_3 [15,17]. The range of crop losses given in the literature is 6–11% [45]; with a NO_x contribution of 70 per cent, we set this to 4 per cent. We estimate that the Nr (as air pollution) contribution to air pollution-based crop loss, not including other stresses such as water stress, is above 50 per cent.
- *Freshwater pollution*. Fresh water eutrophication is defined as areas where the concentration of nitrate exceeds 1 mg NO_3–N l^{-1} [20,27,52]. The Millennium Ecosystem

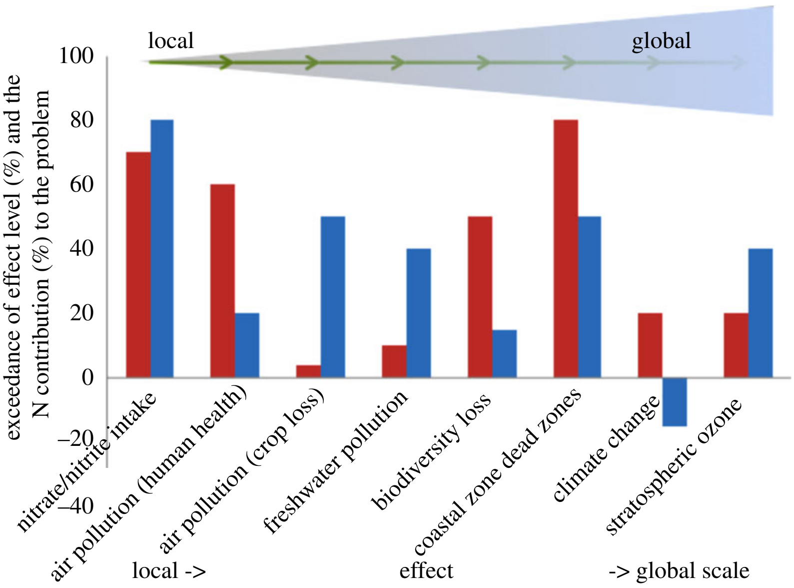

Figure 2. The exceedance (red bar) of the effects levels of Nr for ecosystems or human population, and the contribution of Nr (blue bar) to the total effect, relative to other components or causes (e.g. natural) of the problem. The figure extends from the local scale to the global/stratospheric scale and thus represents the Nr cascade (green arrow). The exceedance and contribution can be summarized as follows: nitrate or nitrite intake: exceedance, 70% of global population exposed to above-recommended levels of either NO_3^- or NO_2^- in air, water or food; contribution of Nr, 80% (20% of the exposure to above-recommended Nr is due to natural sources). Air pollution (human health): exceedance, 60% of global population exposed to air quality above-recommended safe levels; contribution of Nr, 20% of the formation of fine particles is due to human-caused Nr. Air pollution (crop loss): exceedance, 4% of global crop loss owing to air pollution; contribution of Nr, 50% of crop loss is due to human-caused Nr, primarily through tropospheric ozone enrichment. Freshwater pollution: exceedance, 10% of freshwater 'systems' area where NO_3-N exceeds 1 mg l^{-1}; contribution of Nr, 40% relative to other freshwater pollution and natural causes. Biodiversity loss: exceedance, 50% of the total area of biodiversity hot spots in which N deposition exceeds 5 kg N ha^{-1} yr^{-1}; contribution of Nr, 15% of global biodiversity loss estimated to be due to Nr. Coastal zone dead zones: exceedance, 80% of large marine ecosystems (64 in total) 'with a Nr problem'; contribution of Nr, 50% of global coastal zone pollution estimated to be due to Nr. Climate change: exceedance, 20% of the pre-industrial N_2O concentration; contribution of Nr, net cooling of 15% owing to all Nr impacts on drivers of radiative forcing. Stratospheric ozone: exceedance, 20% of the pre-industrial N_2O concentration; contribution of Nr, 40% of all stratospheric ozone depletion is estimated due to Nr.

Assessment shows that in most of the continents, apart from North and South America, this level is exceeded (60% of freshwater systems). Based on the Global Environmental Outlook (GEO-4, [20]) we estimate that globally about 10 per cent of the freshwater area exceeds the 1 mg limit. The contribution of Nr relative to other freshwater pollution is 40 per cent; most of the other pollution resulting from industrial leaching of toxic substances and run-off of fertilizers, and faecal and organic pollution where apart from Nr also P and other pollutants are of concern.

— *Biodiversity loss*. Biodiversity loss owing to Nr deposition has been linked to the critical load for Nr, which, for sensitive terrestrial ecosystems, is between approximately 5 and 10 kg N ha^{-1} yr^{-1} [26]. If we take the global deposition estimates by Dentener *et al*. [53] and the distribution of biodiversity hot spots or eco-regions, the global exceedance of 5 kg N ha^{-1} yr^{-1} is 50 per cent [43]. Overall, biodiversity loss is primarily caused by land-use change, probably followed by climate change, with Nr deposition estimated to account for about 5–15% of current global biodiversity loss [36,37]. Food production and its associated Nr use drives land-use change, although on the other hand land change is avoided by intensified production through Nr use. These land-use change effects on biodiversity are not taken into account here.

— *Coastal dead zones*. The reported number of coastal dead zones has increased from nine in the 1960s to 460 currently [33]. There are currently 64 Large Marine Ecosystems, defined as relatively large areas of ocean space of approximately 200 000 km^2 or greater. These LMEs are **Q6** located in coastal waters adjacent to continents; primary productivity is generally higher than in open ocean areas [54]. The LMEs produce about 80 per cent of the annual world's marine fisheries catch. Globally they are centres of coastal ocean pollution, nutrient over-enrichment, habitat degradation (e.g. sea grasses, corals, mangroves), overfishing, biodiversity loss and climate change effects. According to the National Oceanic and Atmospheric Administration of the United States, most LMEs are subjected to significant eutrophication in coastal waters [54]. There is a Nr problem in about 80 per cent of the LMEs [54]. Compared with other pollution issues of coastal zones, such as phosphorus, the contribution of Nr is estimates to be about 50 per cent.

— *Climate change*. Radiative forcing owing to Nr is expressed as the current N_2O concentration level (325 ppb) above the pre-industrial level (approx. 270 ppb; [55]), which is regarded as the 'safe' level because there are currently no thresholds defined [56,57]. The exceedance of the pre-industrial level is 20 per cent. The contribution of all Nr emissions to the total radiative forcing is a net cooling of 15 per cent as determined by Erisman *et al*. [47] and explained in this paper. This assumes that the short-lived cooling effect lasts for a hundred years if no emission reduction measures are taken. Additionally, the long-term N_2O effects will outweigh the other long-term effect, the additional carbon stored per kg N, which is also much more uncertain.

- *Stratospheric ozone depletion*. Contribution of N_2O to stratospheric O_3 depletion can be expressed similarly to the contribution to climate change: 20 per cent exceedance of the pre-industrial concentration. N_2O emission is currently the single most important ozone-depleting agent, and is expected to remain the largest throughout the twenty-first century [58]. The contribution of Nr has increased because of the reduction of the other stratospheric O_3-depleting substances, and it is now the dominating factor (40%).

Figure 2 shows that, for those issues where both the exceedance and the contribution of Nr are high, there is a clear need for focus in Nr policies. This holds especially for nitrate or nitrite intake, air pollution, coastal dead zones and stratospheric ozone. There is a tendency for the Nr contribution to the effect to decrease as the scale increases from local to global, suggesting that local-scale intervention will be especially effective for reducing Nr impacts. At larger scales, Nr abatement also becomes more difficult. Finally, because of the cascade of Nr, focusing on local-scale issues has a clear benefit for the larger scale.

5. Concluding remarks

Much evidence exists for Nr effects on eutrophication of coastal zones, increased concentrations of ozone and PM in the atmosphere, ozone depletion in the stratosphere and biodiversity loss in terrestrial and aquatic ecosystems. Less is known about the relationship with human health (air and water) and climate. Furthermore, although there is strong evidence for the Nr cascade of effects, better data are needed to quantify the components of the cascade to best support policy options. This review presents as far as possible quantified impacts on the global scale. On smaller scales there are still many uncertainties owing to spatial and temporal variability, and insufficient knowledge.

Current assessments, such as the IPCC AR5, Global Environmental Assessment and regional assessments need better quantitative relationships between nitrogen levels and effects, and we also need to improve our knowledge of the impact of a shortage of nitrogen for many societies. Overall there is large spatial and temporal variability in nitrogen shortages, excesses, fluxes, sources and effects. This is made even more complex through the cascade of nitrogen through the environment and related linked effects. Coupling of the different scales is, therefore, very important, although we still lack effective tools to do so. Although local sources (air emissions or run-off of Nr) contribute primarily to local effects, they also contribute to effects on regional, national, continental and sometimes global scales. Focusing effort on reducing local Nr sources and impacts, therefore, can reap significant benefit at the larger scale.

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