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Trends and geographic variation in adverse impacts of nitrogen use in Europe on human health, climate, and ecosystems: a review

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Abstract

This paper presents a review of the trends and geographic variation of impacts of reactive nitrogen (N) inputs on in Europe through impacts on air, soil and water quality. It illustrates those impacts, by assessing temporal and spatial variation in air, soil and water quality indicators and their exceedances of critical thresholds in view of impacts on human health, terrestrial and aquatic ecosystems, during 1990-2019. Trends are derived from regular inventory and monitoring data and from simulated trends in air quality using the EMEP model.

Well quantified adverse impacts of increased N use are: (i) the effects on human health due to increased tropospheric concentrations of NO_x and ozone, and N-induced increases in fine particulate matter, (ii) the contribution of N_2O to climate change and stratospheric ozone depletion, (iii) the loss of plant and faunal diversity both in terrestrial and aquatic ecosystems via direct and soil mediated impacts, (iv) the acidification of forest soils, with impacts on tree forest nutrition, tree growth and tree vitality, and (v) the eutrophication of marine ecosystems, and associated biodiversity loss and occurrence of harmful algae blooms.

Over the period 1990-2019, N inputs to agriculture stayed relatively constant, but the emissions of ammonia (NH3) decreased by 27%, while emissions of nitrogen oxides (NOx) decreased by 57%. In response to those reductions, concentrations of NO_x, and of N in particulate matter also declined, although less than 50%. In contrast, the reduction in NO_x-induced ozone concentrations and ozone related indicators (AOT40, SOMO35 and POD) was much less (ca 15-20%). Exceedances of critical ozone concentrations for human health and of critical AOT40 and POD vales for vegetation reduced in a similar order of magnitude. Despite decreasing NH_3 emissions, NH_3 concentrations showed a slight and steady increase from 1995 onwards, due to the large reduction in SO_x emissions. Nitrogen deposition and exceedances of critical N loads for terrestrial ecosystems decreased by ca 60%, but the area exceeding critical N loads only reduced by ca 10%. Unlike N, the area exceeding critical acid loads has declined by more than 90% due to high reduction in SO_x and NOx emissions. Trends in nitrate (NO_3) concentrations in groundwater varied across Europe, but showed overall limited changes over the last two decades. . However, N concentrations in surface water and the area exceeding critical levels in view of aquatic biodiversity has decreased and the same holds for N concentrations in coastal regions. Nevertheless, the eutrophication condition of coastal waters has overall not improved due to adverse impacts of elevated phosphorus inputs. Finally the negative impacts of N induced N_2O emissions on climate are estimated to be outweighed by the positive effects of N induced CO_2 sequestration, mainly in forests, and this holds for the whole period 1990-2019. Nitrogen hotspots, being areas with high exceedances in critical levels and loads of N compounds in air

and water, are concentrated in intensive agricultural areas with high livestock densities and in urban region with strong industrial and traffic activities.

Cost-benefit analysis shows that environmental costs of reactive N release to the environment are substantial and tend to exceed the direct economic benefits for agriculture. Given the relevance of N for safeguarding food production it is key to develop integrated and targeted plant nutrition strategies following a food system approach and practices that minimize trade-offs between productivity and the environment. In addition, targeted strategies to further reduce NO_x emissions are needed to reduce air quality related health and biodiversity impacts.

Key words: air pollution, aquatic ecosystems, biodiversity, forest vitality, human health, nitrogen, terrestrial ecosystems, soil and water quality

1 Introduction

Human activities have significantly contributed to the conversion of atmospheric nitrogen gas (N_2) into various reactive nitrogen (N_r) compounds including all nitrogen forms that are biologically, radiatively or photochemically active (Galloway et al., 1995; De Vries et al., 2017; Gu et al., 2023). These reactive compounds include gaseous forms such as nitrogen oxides (NO_x) and ammonia (NH_3) that play a role in chemical processes in the troposphere. Additionally, dissolved nitrogen forms such as nitrate (NO_3) and ammonium (NH_4) are the predominant forms of N taken up by organisms while nitrous oxide (N_2O) is a notable greenhouse gas. Compared to pre-industrial times, the global cycling of reactive N due to human activities has undergone a substantial increase, estimated to have more than doubled. The anthropogenic fixation of N_2 to reactive forms of nitrogen (around 210 Tg N yr^{-1} globally) exceeds the combined contributions of all terrestrial natural processes (Fowler et al., 2013).

Within the European Union (EU) the input of N_r has even tripled throughout the 20^{th} century, primarily due to the use of inorganic fertilisers in agriculture (Sutton et al., 2011a,b). The use of fertilizers doubled the calorie production from land and quadrupled protein consumption

worldwide (Lassaletta et al., 2016; Mueller & Lassaletta, 2020) facilitating the global rise in meat consumption (e.g., Erisman et al., 2008). From all N inputs in the European agriculture less than 25% ultimately becomes human food, 40% is denitrified to N_2 and the remainder is lost to the environment, leading to adverse environmental impacts (Sutton et al., 2011a) as well on human health (UNEP, 2003). These undesirable "cascading effects" (Galloway et al., 2003, 2004) do not stop until the reactive N is converted back to N_2. Both globally and within the EU, fertilizer production emerges as the largest anthropogenic Nr source, contributing to 60-70% of the total N_r production. On a global scale, the second-largest source is biological N fixation (Figure 1) whereas in Europe, high-temperature combustion industrial processes are the second-largest source. In 2010, the N input by fertilizer production is 11.5 Tg N yr^{-1}, being three to nine times higher than industry (3.7 Tg N yr^{-1}) and biological N_2 fixation (1.3 Tg N yr^{-1}), respectively, all showing a substantial increase over the past century (Leip et al., 2011; Sutton et al., 2011a).

In both natural and agricultural ecosystems, additional N is released via mineralization of litterfall, crop residue or organic matter, which converts organic matter into NH_4 (ammonification). Nitrification then occurs, converting NH_4 to NO, NO_3 and N_2O, predominantly under aerobic conditions. Conversely, denitrification can take place in (partially) reduced environments, converting NO_3 to NO, NO_2, N_2O and ultimately back to N_2. Emissions of NO_x increases the ozone production in the troposphere, while emissions of N_2O contribute to stratospheric ozone depletion. Furthermore, emissions of both NO_x and NH_3 result in the formation of ammonium nitrate and ammonium sulphate aerosols, leading to the formation of aerosols or particulate matter (PM). These aerosols have implications for human health, vegetation, and climate dynamics, as they scatter light and act as cloud condensation nuclei, ultimately exerting a net cooling effect. Elevated concentrations of N compounds in terrestrial and aquatic ecosystems can lead to eutrophication and acidification with adverse impacts on flora and fauna. This N cascade is therefore intricately linked to major global and regional environmental challenges that policymakers currently face (De Vries, 2021). These N related

challenges encompass human health impacts, climate change, terrestrial and aquatic biodiversity loss, as well as pollution of surface and groundwater. Due to all those interaction, reactive nitrogen affects most UN sustainable development goals' with agriculture being a key sector (Winiwarter et al., 2021).

The fundamental importance of the N cycle in human health issues, ecosystem functioning and climate change is illustrated in Figure 1. Apart from the beneficial role of N fertilization in view of food and feed production and of N deposition on forest growth, the fixation of reactive N has also many adverse impacts on the environment. These impacts include: (i) enhanced losses of NO_x and NH_3, which affects ozone and particulate matter production in the atmosphere, and causes elevated nitrate concentrations in drinking water, affecting humane health, (ii) increased NH_3 and NO_x deposition and nitrate runoff affecting terrestrial and aquatic biodiversity and (iii) enhanced the emission of the greenhouse gas nitrous oxide, affecting climate. Since the contribution of excess N_r to these issues varies across regions due to the strong dependency on regional and local agro-ecosystem properties, it is crucial to understand the principles of the 'Nitrogen Cascade' in view of critical impacts on the environment.

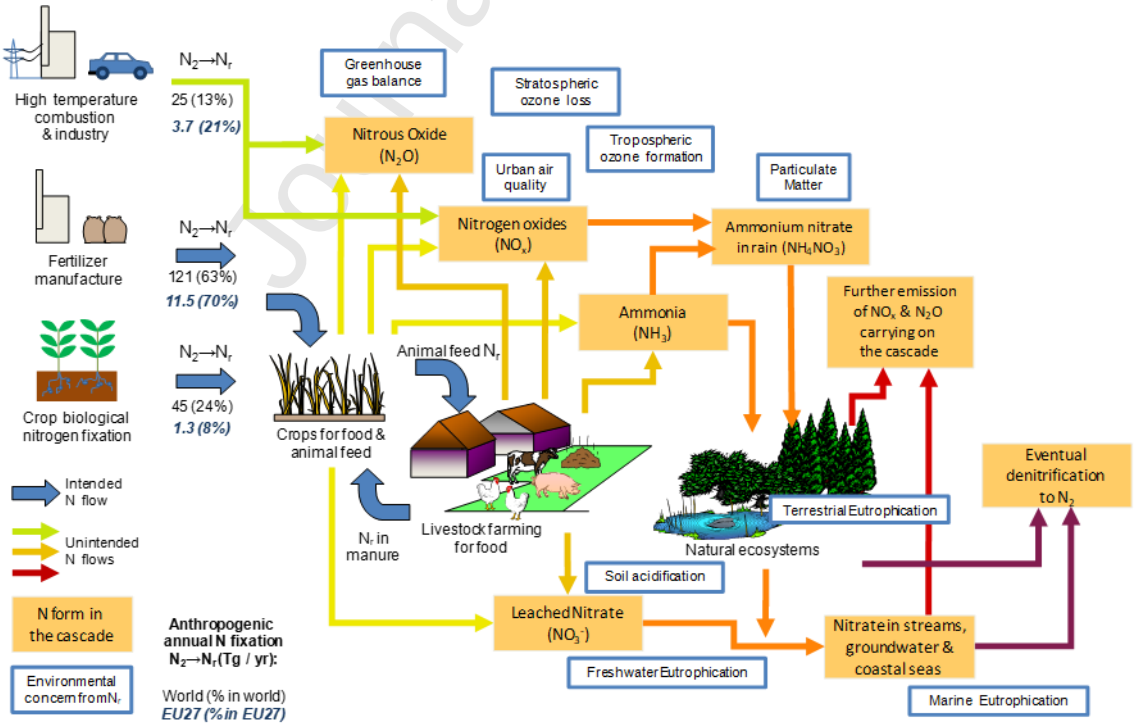


Figure 1. Simplified view of the 'Nitrogen Cascade'. At both European and global scales, the largest anthropogenic source of reactive nitrogen (N_r) enters agriculture via chemically fixed atmospheric N_2 and leaves agriculture in harvest products and via N losses to air (NH_3, N_2O and NO_x), groundwater and surface waters (NO_3, NH_4, Dissolved Organic Nitrogen). Additional sources are biological N fixation and the inadvertent fixation of N through high-temperature combustion processes releasing NO_x in the industrial and transport sectors. NO_x emissions lead to ozone formation, and both NO_x and NH_3 contribute to particulate matter formation. These losses and their transformations create a sequence of ecological and human health effects, allowing one atom of fixed N to exert several different effects, with nine main threats illustrated here (based on Sutton et al., 2011a).

In 2011, the European Nitrogen Assessment (Sutton et al., 2011b) provided a comprehensive overview of the current N budgets for Europe, specifically focusing on the year 2000 (De Vries et al., 2011a; Leip et al., 2011). This study also explored the impacts of N on air quality (Moldanová et al., 2011), greenhouse gas balance (Butterbach-Bahl et al., 2011), soil quality (Velthof et al., 2011), terrestrial biodiversity (Dise et al., 2011), freshwater (Grizzetti et al., 2011), and marine systems (Voss et al., 2011). However, limited attention was paid to impacts on human health and forest ecosystems, the integration of flows and impacts across all nitrogen induced impacts, and the spatial and temporal variation of drivers and impacts across Europe, in particular in view of their exceedances for existing critical levels and loads.

In Europe N emissions and corresponding deposition decreased since the 1990s (Engardt et al., 2017) due to a combination of emission abatement policies and economic transformation (Erisman et al., 2015). Since 1990, national monitoring systems related to air quality, ground and surface water quality (e.g. EEA, 2023a) and to impacts on forest ecosystems in terms of soil quality, forest nutrition, tree growth and biodiversity (ICP on forest; De Vries et al., 2003; Schmitz et al., 2019; 2023) were implemented. In addition, results of model applications related to air quality are available since 1990 at a high spatial resolution (5 arcmin x 5 arcmin). However, until now, a systematic review on trends and spatial variation in adverse impacts since 1990, evaluating the impacts of N emission reductions across the various compartments (air, soil and water) and their receptors (ecosystems, health and climate) has not been carried out.

In this paper, we present a comprehensive review of the adverse impacts of reactive N on human health, climate, and ecosystems at the European scale and examine trends in N compounds and N-related (or N-induced) threat indicators in air and water over the period 1990 to 2019. The trends are based on an analysis of data available in regular inventories and long term monitoring systems for air, ground and surface water quality and forest ecosystems as indicated above. In addition, we used results of the EMEP air quality model (see Simpson et al., 2012, 2023 and references therein for details on this model and Fagerli et al., 2023 for the calculation setup) on trends in concentrations of NO_x, NH_3, O_3 and N-$PM_{2.5}$ and deposition of N at a spatial resolution of 50 km × 50 km (https://www.emep.int/mscw/mscw_moddata.html). Trends in ecological and human health risks are assessed by the exceedance of critical levels and critical loads of N. Critical levels refer to the direct effects of elevated pollutant concentrations and are defined as "*pollutant concentrations above which adverse effects on receptors, including human beings, plants, ecosystems, or materials, may occur based on current knowledge*" (UBA, 1996, 2004). Critical loads refer to the indirect (soil-mediated) effects of elevated N deposition on terrestrial ecosystems and are defined as "*quantitative estimates of exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur based on current knowledge*" (Nilsson & Grennfelt, 1988). We assess what the time delays are between emission reductions and reductions in air, soil and water quality in terms of exceedances in critical levels and loads of N compounds and associated pollutants. More specifically, trends in observed and calculated concentrations of N_r compounds and N induced pollutants (O_3 and PM) and deposition levels or N loadings are compared with critical levels in view of their impact on human health, climate, and terrestrial and aquatic ecosystems as given in Table 1.

Table 1. Nitrogen compounds, and pollutants and GHGs affected by them, which are used as indicators of air, soil and water quality with related effects on human health, climate and the functioning (services) of terrestrial and aquatic ecosystems. PM is particulate matter including NH_4, NO_3 and SO_4. NO_x is a mixture of NO and NO_2. Other N compounds not shown here include nitric acid, nitrous acid, amines and a plethora or oxidized and reduced organic nitrogen compounds.

Impacts	Air quality			Soil quality	Water quality	
	Primary	Secondary	GHGs	Soil	Ground	Surface

	pollutants		pollutants				solution	water	water
	NH_3	NO_x	O_3	PM	N_2O	CO_2, CH_4	NO_3, NH_4, pH, Al	NO_3, Al	N total, NH_4, pH, Al
Human health	x	x	x	x				x	(x)
Climate			x	x	x	x			
Terrestrial ecosystems	x	x	x	(x)[1]			x		
Aquatic ecosystems									x

[1] N-induced PM, such as $(NH_4)_2SO_4$ may also affect terrestrial ecosystems by affecting photosynthesis and plant physiology, but this effect is included in impacts of gaseous NH_3 on plants and PM is not used as an indicator for these effects.

In more detail, we discuss impacts on (see Table 1):

- Trends in N use and N surpluses in agriculture and related N losses by emissions to air (NH_3 and NO_x) and leaching and runoff to water (NO_3, total N) in combination NH_3 and NO_x emissions by trade and industry (Section 2).
- Impacts on human health due to: (i) direct adverse impacts of elevated NO_2 concentrations, (ii) NO_x induced O_3 formation, (iii) NH_3 and NO_x induced PM formation, and (iv) pollution of freshwaters due to NO_3-leaching and acidification induced Al leaching (Section 3).
- Impacts on greenhouse gas exchange due to: (i) direct and indirect emissions of nitrous oxide, (ii) NH_3- and NO_x-deposition induced increased forest productivity and impacts of N on CO_2 exchange and on and methane exchange, (iii) NO_x-induced formation of tropospheric ozone and on CO_2 exchange by reduced productivity, and (iv) NH_3- and NO_x-induced increased formation of particulate matter (Section 4).
- Impacts on terrestrial ecosystems including: (i) plant damage through NO_x-induced ozone formation, (ii) N enrichment and acidification of soils with related impacts on forest nutrition and plant (tree) growth and increased susceptibility to secondary stresses, (iii) and biodiversity decreases due to eutrophication and acidification (Section 5).

- Impacts on aquatic fresh water and marine ecosystems including eutrophication and acidification, leading to biodiversity loss and toxic algal blooms (Section 6).

Considering the trends in N use and associated environmental impacts, we also include a short analysis of costs and benefits of measures reducing or mitigating the impacts of N (Section 7). We finalize this review with conclusions with respect to: (i) the evidence for adverse N effects across the various compartments (air, soil and water) and related receptors (ecosystems, health and climate), (ii) trends and geographic variation in those adverse impacts since 1990 and (iii) recommendations for future research (Section 8).

2 Trends in nitrogen use and nitrogen losses

The spatial variation and temporal trends in N impacts are closely linked to the spatial patterns of N use and N losses to air and water. In this section, we present an overview of these trends for the period spanning 1990 to 2020 while focussing on N fertilizer consumption and N manure production, NH_3 and NO_x emissions to air and the nitrogen surplus, being the difference in N input and N uptake (crop N removal), which strongly affects NO_3 leaching and thereby NO_3 concentrations in groundwater and surface water.

Trends in total N inputs on croplands and fertilized grassland (given by Einarsson et al., 2021 for the period 1961-2019) slightly increased up to 1996, followed by a decrease up to 2008 and an increase again up to 2018, varying around an average number near 22 Tg N yr^{-1} (Figure 2, top left) Synthetic fertilizers were the main N source, accounting for about 50% of total N inputs, followed by manure, accounting for approximately 35% of the N input . Total fertilizer consumption rates declined slightly from 11 Tg N yr^{-1} in 1990 down to 10 Tg N yr^{-1} in 2008 and gradually increased up to 11 Tg N yr^{-1} in 2015, while the manure input stayed relatively constant near 8.5 Tg N yr^{-1} (Figure 2 top left; see also Eurostat, 2023a,b).

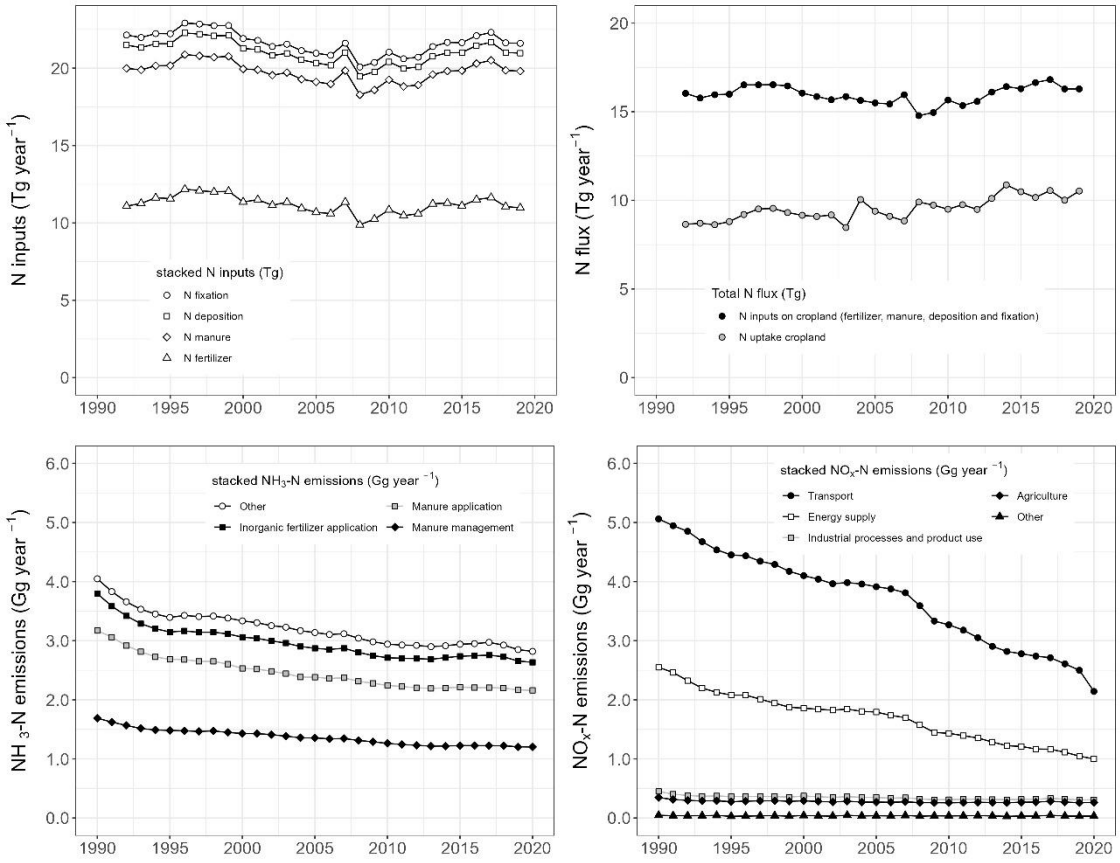


Figure 2. Stacked nitrogen inputs to EU from 1990-2019 on cropland and fertilized grassland (top left), cropland N harvest (uptake) and N inputs on cropland (top right) in Tg N yr^{-1} and stacked N emissions from both agriculture and other sources to air as NH_3 (bottom left) or NO_x (bottom right) in Gg N yr^{-1}. Data of N inputs and uptake are from Einarsson et al. (2021), the NH_3 and NO_x emissions to air from (EEA,2023b). Note that the difference between total N inputs and N uptake in top left figure is the N surplus.

Total N inputs on croplands are relatively stable over the years 1990 to 2019 increasing from 16.0 -16.3 Tg N yr^{-1} while the N uptake increased from 8.6 -10.5 Tg N yr^{-1} , implying a decrease in N surplus, defined as total N input minus crop N removal, of 7.4 -5.8 N yr^{-1} (Figure 2 top right). However, in that period, the area of agricultural cropland also decreased from 124 to 109 M ha. Consequently, the average N application rates on cropland, derived by dividing the total N input by the cropland area, increased from 130 to 149 kg N ha^{-1} yr^{-1} between 1990 and 2019. The averaged N fertilizer input rates on crop land in 2019 was 89 kg N ha^{-1} whereas the manure N input was around 4 kg N ha^{-1}, whereas the remaining N input from deposition and fixation was around 16 kg N ha^{-1}. The N surplus on cropland slightly decreased from ca 60-50 kg N ha^{-1} between 1990 and 2011 due to changes in agricultural management (e.g., fertilizer application techniques), after which the N surplus remained near the same value. The N inputs in European

agriculture since 1990, with related losses of NH_3 and NO_x to air and nitrate to water, combined with NH_3 and NO_x emissions from other sources (see Figure 2 bottom), is associated with negative impacts on biodiversity and ecosystems, as discussed later.

Agriculture is the main source of NH_3 emissions, accounting for 93% of total emissions (Figure 2 bottom left). The major sources of NH_3 are manure management (approximately 43% during the whole time period), manure application (decreasing from 37% in 1990 to 34% in 2020), and fertilizer use (increasing from 15% in 1990 to around 16% in 2020). Over the period of 1990 to 2008, NH_3 emissions show a decline of 27% (from 4.1 to 3.0 Tg N), mainly due to a changes in the handling and management of organic manures, and a reduced use of N fertilizers since 1990 (Erisman et al., 2015). Since 2008, emissions stay rather constant (varying between 2.8 and 3.0 Tg N) and a recent decadal satellite record even suggest an increase over the period 2008–2018 (Van Damme et al., 2021)

Compared to NH_3, the contribution of agriculture to NO_x emissions is minor (Figure 2 bottom right) increasing from 5.9% in 1990 to 10.3% in 2020. Main sources of NO_x emissions include the combustion of fossil fuels in transportation (increasing from 50% in 1990 to 54% in 2020), followed by energy supply (decreasing from 42% in 1990 to 32% in 2020). Through the implementation of the EU's National Emissions Ceilings Directive and the adoption of cleaner technologies, NO_x emissions declined with 57% between 1990 and 2020, reaching a level of 2.2 Tg N in 2020. The nitrous oxide (N_2O) emissions also decreased from 330 to 231 Mg N (-30%) over the same period due to mandatory measures implemented under the Nitrate Directive (EC, 2000), the Common Agricultural Policy, and the Landfill Waste Directive (EC, 1999).

3 Impacts on human health

Changes in the N cycle have both positive and negative implications for human health. While N fertilizers play a crucial role to maintain and increase food production in view of required human food consumption (Erisman et al., 2011; de Vries et al., 2021), this section focuses on the adverse health effects associated with reactive N compounds. Specifically, we examine the

negative impacts on human health caused by inhaling N-containing or N-induced compounds (such as NO_x, O_3, and PM) and ingesting N compounds in water (primarily NO_3), including direct and indirect effects. We delve into the health effects, critical levels established for these compounds, and trends in exceeding those levels, particularly focusing on the impacts of NO_x, O_3 and PM in the atmosphere, and NO_3 in drinking water. Additionally, we briefly summarize other potential hypothesized health effects associated to elevated N inputs.

3.1 Air quality: nitrogen oxides, ozone and particulate matter

3.1.1 Relationships between reactive N and human health

Air pollution is widely recognized as a significant public health concern (Landrigan et al., 2018). The contribution of N compounds to air pollution is considerable, as they can elevate local concentrations of NO_x, which, in turn, contribute to the production of tropospheric ozone (Chameides et al., 1994) and fine particulate matter. These pollutants have detrimental effects on human health, as established by several studies (Wolfe & Patz 2002; WHO, 2006a, b; WHO, 2008). The rate of O_3 production depends on the ratio of NO_x to volatile organic compounds (VOCs), including methane, and this relationship is nonlinear. A change in NO_x has a relative greater impact on O_3 (up to five times greater) than a change in non-methane VOCs (Wang & Jacob, 1998; Simpson, 1995), thereby neglecting the effect of changes in CO and CH_4. About 60% of the increase in O_3 since 1900 can be attributed to the rise in NO_x (Wang & Jacob, 1998).

Nitrogen emissions also increase the presence of fine particulate matter (PM) due to the interaction of NO_x and NH_3 in the atmosphere (de Leeuw & Horálek, 2009). In the atmosphere secondary particles such as ammonium sulphate and ammonium nitrate can be formed. These particles can travel long distances and contribute to regional exposure of humans to both PM_{10} and $PM_{2.5}$, where the numbers 10 and 2.5 refer to the size of the particles (less than 10 and 2.5 μm, respectively). The N compounds act as atmospheric oxidants and favour conditions for oxidation (e.g. increased pH), neutralise sulphuric acid thereby producing ammonium sulphate. The contribution of N_r to fine particulate matter ($PM_{2.5}$) varies significantly across regions, being two times higher in urban than in rural areas (Malm et al., 2000; Putaud et al., 2004). In

densely populated regions of Europe, NH_3 contributes 10-20% to the formation of small secondary inorganic aerosols (SIA) and this percentage can be higher in areas with intensive livestock farming (Hendriks et al., 2013).

In urban areas, particulates containing NH_4 and NO_3 make up a significant portion (~70%) of the SIA. These SIA particles, in turn, contribute about 40-50% to $PM_{2.5}$. Measures reducing NH_3 can therefore lead to substantial reductions in $PM_{2.5}$. According to Pozzer et al. (2017), a reduction of 50 or 100% in NH_3 emissions would decrease the annual $PM_{2.5}$ concentrations over Europe by 11% and 34%, respectively. Reducing the NH_3 emission by 50% also leads to a 19% reduction in air pollution-related mortality in Europe.

3.1.2 Health impacts and critical levels

Human health impacts are often expressed as mortality impacts (premature deaths or years of life lost, YLL) or impacts on the years lived with disabilities (YLD) or disability-adjusted life years (DALY). Mortality refers to the number of deaths that have occurred because of a specific disease, and is expressed either as premature deaths (deaths that occur before a person reaches an expected age) or YLL (the years of potential life lost because of premature death). Morbidity is the state of having a disease, often being expressed as years lived with disability (YLD), meaning years of healthy life lost to disability. A Disability-Adjusted Life Year (DALY) is one lost year of a 'healthy' life.

Extensive research has established a clear association between air pollution and premature mortality as well as various diseases (Lelieveld et al., 2015). Exposure to air pollution increases the risk of mortality from respiratory conditions such as chronic obstructive pulmonary disease, pneumonia, and asthma (Garshick, 2014; Chen et al., 2017; Pirozzi et al., 2018; Liu et al., 2019). Additionally, air pollution has been linked to an increased number of hospital visits for allergic rhinitis, asthma, and serum Immunoglobulin E (IgE)-mediated allergies (Villeneuve et al., 2007; Hu et al., 2020; Hou et al., 2021). The detrimental effects of air pollution extend to young children, who face a higher risk of hospitalization for respiratory diseases due to

exposure (Huang et al., 2022). Furthermore, this exposure during childhood contributes to increased airway obstruction in adolescence (Milanzi et al., 2018).

In 2020, air pollution in EU-27 as determined by pollutant concentrations above guideline levels of the World Health organization (WHO), being of 5 μg m-3 for PM2.5, 15 μg m^{-3} for NO_2 , 60 μg m^{-3} for O_3 and 5 μg m^{-3} for $PM_{2.5}$, respectively (see also Table 2), led to 49,000 premature deaths due to NO_2 exposure, 24,000 premature deaths due to O_3 exposure and 238,000 premature deaths due to $PM_{2.5}$ exposure (WHO, 2021; EEA, 2023b). The total number of years of life lost (YLL) in the EU was 249,000 due to exposure to O_3, 484,000 due to exposure to NO_2, and increased up to 2,427,000 due to exposure to $PM_{2.5}$.

Nitrogen oxides impacts

Increased emissions of NO_x contribute to the presence of high atmospheric NO_x levels. Urban areas tend to experience the highest concentrations of NO_x, which can exacerbate and prolong common viral infections, posing substantial risks to individuals with asthma (Spannhake et al., 2002). Among nitrogen oxides, NO and NO_2 are free radicals and show a variety of biological effects. NO_2 is a strongly oxidizing toxicant, although NO, not oxidizing as NO_2, is toxic in that it interacts with haemoglobin to form nitrosyl- and methaemoglobin, which can be life-threatening. Animal studies have demonstrated that short-term exposure to NO rarely causes permanent effects. However, prolonged and continuous exposure to NO concentrations up to 1880 μg m^{-3} over several weeks to months can have severe consequences, particularly for the lungs, as well as other organs such as the liver and blood (WHO, 2003). It is important to note that ambient concentrations of NO are typically two to four orders of magnitude lower than the aforementioned exposure levels of 1880 μg m^{-3}. Nitrogen dioxide can cause harm to the lungs when inhaled. Numerous studies, published since 2004 and reviewed by the World Health Organization (WHO, 2013a, 2013b, 2021), have demonstrated associations between both short-term and long-term exposure to NO_2 and adverse health outcomes, including mortality, hospital admissions, and respiratory symptoms. These effects have been observed at concentrations equal to or below the current EU limit value of 40 μg m^{-3}, whereas the most recent WHO Air

Quality Guidelines decreased this limit via intermediate targets to a final limit of 10 μg m^{-3} (WHO, 2021). Despite the fact that these critical concentrations are exceeded in various parts of Europe, some health impact assessments (e.g., Holland et al., 2005a, 2005b) have not directly considered the effects of NO_2. Instead, they evaluated the health damage caused by NO_2 by applying the dose-response functions associated with the impacts of particulate matter with respect to the nitrates that are formed in the atmosphere from NO_x.

Ozone impacts

Ozone is an air pollutant that primarily affects human health through inhalation (von Mutius, 2000; WHO, 2003). Exposure to O_3 can lead to various adverse health impacts, including coughs, asthma (reactive airways diseases, RAD), short-term reductions in lung function, and chronic respiratory disease. Children who engage in physical activity in high O_3 environments have a 40% higher likelihood of developing asthma (McConnell et al., 2002), a disease that continues to spread in many parts of the world despite advancements in treatment (Beasley, 1998). Health risks of ozone include a clear increase in mortality and respiratory morbidity rates due its short-term impact on lung function, inflammation, permeability, and respiratory systems (WHO, 2008). A study by Turner et al. (2016) suggests that long-term exposure to ambient O_3 contributes to the risk of respiratory and circulatory mortality. In the EU member states, it is estimated that O_3 exceeding 35 ppb is associated with ~13,600 premature deaths (EEA, 2017) and 14,000 respiratory hospital admissions (WHO, 2008) per year. An increase in mortality risk has been observed at O_3 concentrations above 60 μg m^{-3} (WHO, 2021).

Fine particulate matter impacts

The physicochemical properties of particulate matter, such as their size, shape, and composition, determines their fate in the environment as well their impact on human health. Especially particle size (in μm) is important, being the reason to distinguish between PM_{10} and $PM_{2.5}$, with the lower size aerosols having a larger effects at the same concentration. Numerous studies have demonstrated positive associations between elevated levels of fine PM and various health

issues, including cardiovascular diseases, respiratory diseases, asthma, reduced lung function, and increased overall mortality (e.g. Pope et al., 2002).

Secondary inorganic aerosols, predominantly composed of nitrates and sulphates, constitute a significant portion of $PM_{2.5}$ (see Rohr & Wyzga, 2012). There has been some uncertainty on the health effects of N in these secondary aerosols (Kelly et al., 2004) because laboratory studies fail to determine a biological effect to exposure levels higher than those found in ambient air. In contrast, epidemiological studies have frequently found adverse health effects of secondary inorganic aerosols (Kelly & Fussell, 2012). In the ambient air, nitrates and sulphates can alter the hygroscopicity of PM, potentially increasing exposure to soluble metals and other toxic components. In their review Kelly & Fussell (2012) showed that N compounds have similar health impacts than other PM constituents (Fowler et al., 2013; WHO, 2013b; Brunekreef et al., 2015).

Limit values and critical limits

A summary of regulatory limit values for NO_2, PM_{10} and $PM_{2.5}$, and O_3 as outlined in the European Air Quality Directive (EC, 2008) and effect-based air quality standards defined by WHO (WHO, 2021) is given in Table 2. Both Air Quality Standards and limit values pertain to both acute effects resulting from short-term exposures and chronic effects associated with long-term exposures (annual means). Note that the EU limit values are less stringent than the WHO air quality guidelines (Table 2), since the latter represent scientifically determined values (consistent with critical levels) while the EU values incorporate compromises as a result of policy negotiation.

Table 2. Limit values of NO_2 and particulate matter (PM) and limit values of O_3 as defined in the European Air Quality Directive 2008/50/EC (EC, 2008) for the protection of human health, compared with the air quality standards as recommended by the WHO (WHO, 2021) All values are given in μg m^{-3}.

Period	Limit/target issued by	NO_2[1,2]	O_3[1]	PM_{10}[1]	$PM_{2.5}$[1]
Calendar year	EU	40	-	40	25
	WHO	10	60	15	5
Short term[2]	EU	200	120	50	-
	WHO	25	100	45	15

[1] The EU limit values for NO_2 and PM_{10} and the limit values for ozone since 1 January 2010; the $PM_{2.5}$ value is set as limit value since 2010 and has become legally binding by 2015

[2] Limit values for health are given for NO_2 specifically and not for NO_x, unlike impacts on biodiversity (see Table 6).

[3] The WHO values refer to an hourly average for NO_2, the highest 8-hour average during a day for O_3 and a daily average for PM_{10} and $PM_{2.5}$. Exceedances of the EU limit values are allowed for 18 hours (NO_2), 25 days (O_3) and 35 days (PM_{10}).

On the other hand, exceedance of the WHO air quality guidelines does not necessarily imply the need for action, whereas non-compliance of an EU limit value implies that measures must be implemented to bring the concentrations below the specified limit value. In the event of exceeding an O_3 limit value, Member States are also required to take all necessary actions to reduce concentrations; however, these measures should not incur disproportionate costs in meeting the limit value. The rationale for the relatively less stringent approach to O_3 compared to NO_2 and PM is the significant contribution of O_3 producing sources located outside of Europe.

The EU has not defined limit values for NO in terms of human health, as toxic values are far above ambient levels. For NO_2 the limit value of 40 μg m^{-3} for the annual average concentration appears to be the most stringent, and its attainment generally implies achievement of the hourly limit value, being 200 μg m^{-3}. For O_3 there is only a limit value for short-term exposures given the limited epidemiological evidence for long-term impacts (Jerrett et al., 2009), being slightly less stringent than the WHO air quality guidelines for critical limits. Health effects of PM_{10} or $PM_{2.5}$ are to be expected by exposures to annual critical limits above 15 and 5 μg m^{-3},

respectively, as set by the guidelines of WHO, where short-term critical limits are set at 45 and (PM_{10}) and 15 ($PM_{2.5}$) μg m^{-3}. It is widely accepted, however, that there is no critical threshold below which no effects do not occur, implying that even exposure to below these low levels of PM_{10} or $PM_{2.5}$ can have some detrimental health effects (WHO, 2004, 2021).

3.1.3 Geographic variation and trends in air pollutant concentrations

Nitrogen oxides

Concentrations of NOx (sum of NO and NO_2, expressed in NO_2), over Europe, based on EMEP calculations, reveals a decreasing trend since 1990, particularly in the early 90s for Central Europe (Figure 3, left,). The trends are presented for the overall mean across Europe and for three sub-regions, with each region covering approximately 2.3-2.9 Mkm^2: North (>55ºN), South (<46ºN) and Central. These findings align with a slight downward trend in measured NO_2 concentrations in rural and urban areas (Figure 3, right).

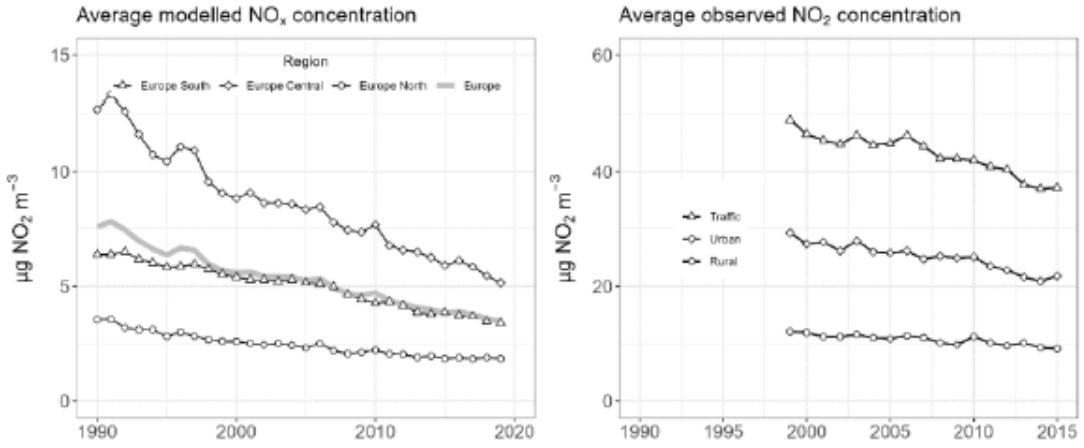


Figure 3. The 1990–2019 temporal development of modelled average ground-level NO_2 concentration in three European regions and overall European average (thick grey line; left) and observed NO_2 concentrations at rural background, (sub)urban stations and at stations close to traffic (source: EEA), averaged over all stations operational in the period 1999-2015 (right).

Highest concentrations are detected near traffic locations. Critical limits and limit values are not exceeded except for locations in proximity to urban or traffic locations. Observed

concentrations and trends in NO_x concentrations, expressed in NO_2, in rural areas, varying from 12-9 μg m^{-3} are in fair agreement with the model results (see also: https://www.emep.int/mscw/index.html). Note, however, that the resolution of the EMEP model is insufficient to capture the higher NO_2 concentrations around traffic or urban location.

A more detailed analysis of NO_2 observations at individual sampling stations confirms the observed decline since 1990 (Figure 4, left). NO_2 concentrations usually decline with 0-4% a year, being concordant with the observed decline in NO_x emissions across Europe since 1999 (Mol et al., 2010), partly related to the increased use of catalysts in combustion engines. Only a limited number of stations show an increasing trend ($P > 0.05$), in particular in Austria. On the Iberian Peninsula, the situation is more diverse with a mix of decreasing and increasing trends. The stabilization or occasional increase near traffic locations can be attributed to the growing number of diesel cars (Carslaw, 2005).

Though the EU limit value for the annual average concentration of NO_2 of 40 μg m^{-3} was expected to be achieved by 2010, NO_2 concentrations in 2015 frequently exceed this limit (Figure 4, right). These exceedances occur in 22 of the EU28 countries, but also in Norway, Switzerland, and Serbia. Between 2000 and 2015, the proportion of people in urban areas exposed to NO_2 concentrations above 40 μg m^{-3} decreased from 26% to 8.7% (EEA, 2018).

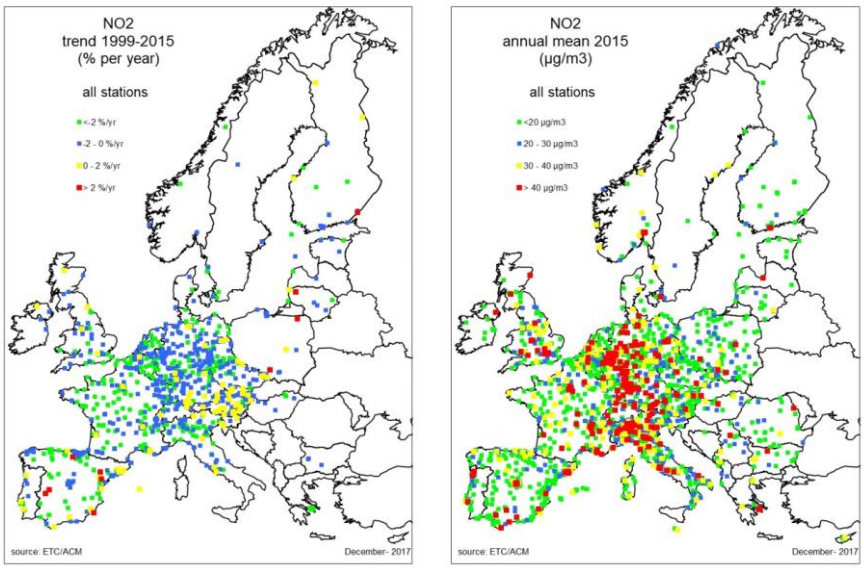


Figure 4. Observed trends in measured annual average NO_2 concentrations over the period 1999-2015 (relative changes in concentration in % per year for all station types; left) and the

annual mean concentrations in 2015, where a red dot indicates stations exceeding the EU limit value of 40 μg m^{-3} (right). Source concentration data: ETC/ACM (2013); EEA (2017).

Ozone

Concentrations of ozone show a slightly decreasing trend over Europe, based on EMEP calculations at a spatial resolution of 50 km × 50 km, as illustrated for both the maximum 8-hour average O_3 concentrations and the sum of ozone means exceeding the 35 ppb (SOMO35) for the period 1990 to 2019 (Figure 5). Significant inter-annual variability exists, particularly around the year 2003, likely due to unusually high summer temperatures. Over the past 25 years, both ozone indicators show a decreasing trend in the central and southern regions, while the reduction in the northern region is relatively small. The simulated values in maximum 8-hour average O_3 concentrations generally fall within the range of 30 to 60 ppb (Figure 5, left), consistent with previous studies indicating that daytime O_3 concentrations in the temperate latitudes of the northern hemisphere typically range from 25 to 65 ppb (Akimoto, 2003; Oltmans et al., 2006), with an average of approximately 40 ppb (Ehhalt et al., 2001). Note that these levels are about twice as high as the estimated pre-industrial O_3 concentrations, which were around 10 to 15 ppb (Volz & Kley 1988; Akimoto, 2003).

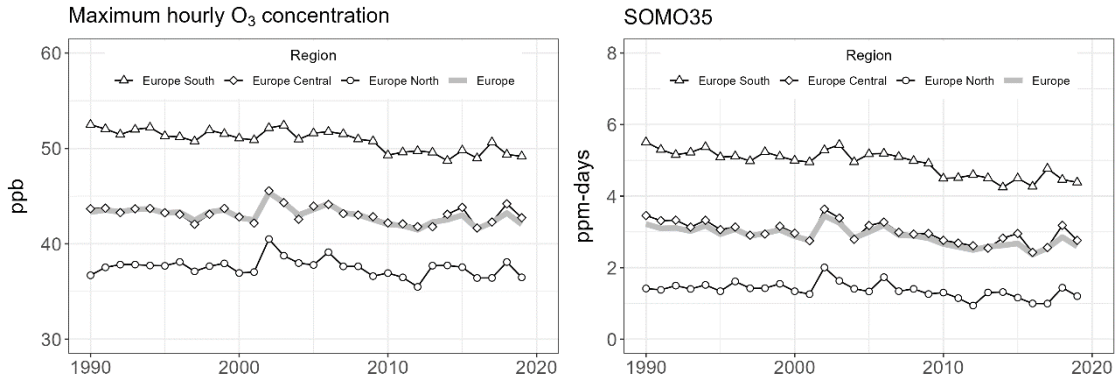


Figure 5. The 1990-2019 trends in modelled ground-level annual average maximum hourly O_3 concentration per day (left) and sum of 8-hourly average O_3 concentrations above 35 ppb (SOMO35;right) in three European regions and the overall European average (thick grey line).

The observed annual mean and the 98th percentile of hourly O_3 concentrations across Europe increased for locations nearby traffic (n = 90) and in urban areas (n = 537) and decreased in

rural areas (n = 537), confirming the simulated trends by EMEP. In rural areas the mean annual O_3 concentrations declined with 0.2% per year. The changes in hourly concentrations are slightly more pronounced than the changes in mean annual concentrations. The increase in O_3 concentrations around traffic locations, and to some extent also in urban areas, partly follows the decline in NO_x as less O_3 is consumed in the titration reaction with NO.

The declining trend in rural areas is further demonstrated in Figure 6, which showcases the geographic variation in maximum O_3 concentrations (left) and SOMO35 (right) as calculated by the EMEP model for the years 1990 and 2019. eed urban ones. The majority of ozone exceedances occur in Central and Southern Europe.

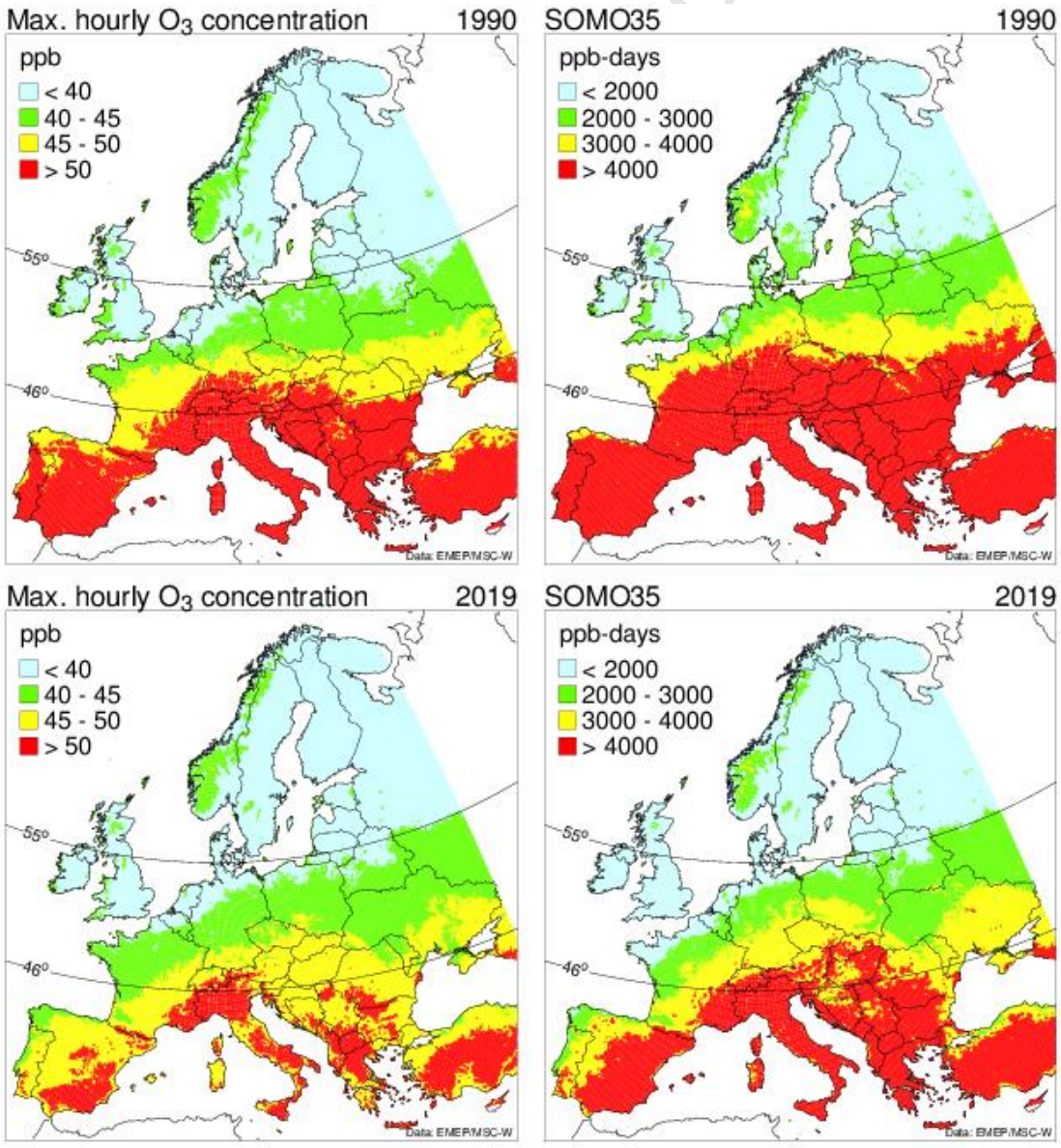


Figure 6. Mean of modelled ground-level daily maximum O_3 concentration) (left) and sum of 8-hourly average O_3 concentrations above 35 ppb (SOMO35; right) in 1990 (top) and 2019 (bottom). The 46ºN and 55ºN parallels separate three regions 'North', 'Central' and 'South'.

Ozon concentrations exceeding the short term (25 days) EU limit value of 120 µg m^{-3} (Table 2), being equal to 60 ppb, are likely to occur for more than 25 days per year in various regions, particularly in the Mediterranean, where the maximum O_3 concentration exceeds 50 ppb. These findings align with actual measurements. Between 2013 and 2015, it was estimated that 7-30% of the urban population in Europe experienced ozone concentrations exceeding the EU limit value for more than 25 days per year, compared to 55% in 2003 (EEA, 2018). However, the rural population tends to face higher exposure levels as rural concentrations generally exc

Note that short-term peak concentrations across Western Europe seems to decline whereas the mean annual O_3 concentrations seems to increase. The decline in peak concentrations is largely influenced by the reduced emissions of volatile organic compounds (VOC) and NO_x and partly counteract the increase in the hemispheric background concentrations that tends to increase due to rising methane concentrations, increased ship emissions, augmented hemispheric transport of O_3 precursors from other countries in the Northern Hemisphere, and reduced O_3 deposition under hot and dry conditions. This implies a reduction in the effects of acute O_3 exposure, which the EU limit values address, but an increase in low-level chronic exposure, which is not accounted for by the current limit values.

Particulate matter

Concentrations of nitrogen-containing fine PM (N-$PM_{2.5}$), being the sum of fine PM nitrate and ammonium, strongly declined over the years 1990 to 2020 (Figure 7).

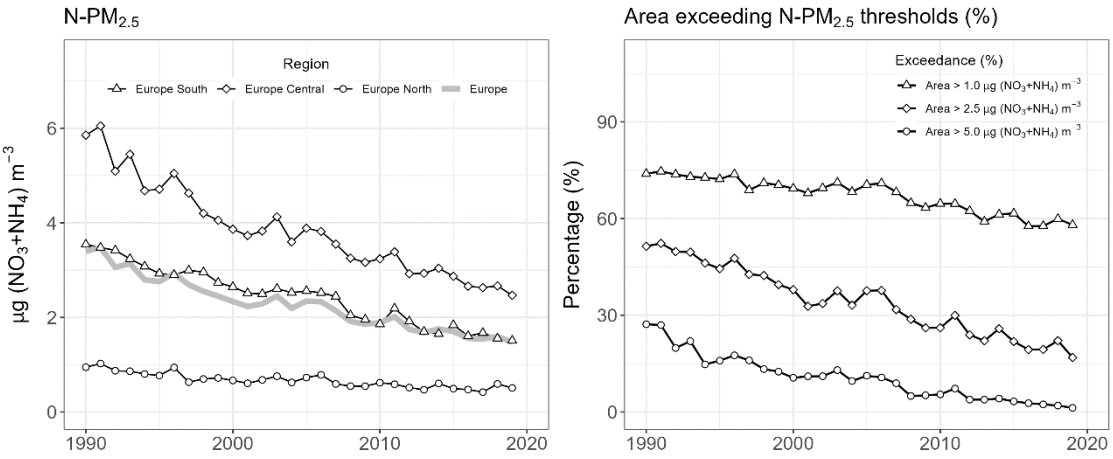


Figure 7. The 1990–2019 temporal development of modelled average sum of fine particulate matter nitrate and ammonium (N-PM2.5) in three European regions and the overall European average (thick grey line; left) and the percentage of European land area (west of 40ºE) with modelled N-PM2.5 above 1, 2.5 and 5 μg (NO_3 + NH_4) m^{-3} (right)..

The N-$PM_{2.5}$ concentrations declined from 6 to 2.5 μg m^{-3} in Central Europe, from 3.5 to 1.5 μg m^{-3} in the South of Europe and slightly reduced from 1 to 0.5 μg m^{-3} in the North of Europe (Figure 7 left). The contribution of those N compounds to $PM_{2.5}$ varies between 10-50%, implying that total $PM_{2.5}$ can be 2-10 times larger. This implies that for concentrations of N-$PM_{2.5}$ above 5 μg m^{-3} the WHO critical limit of 5 μg m^{-3} for $PM_{2.5}$ is absolutely exceeded, while the EU limit of 25 μg m^{-3} for $PM_{2.5}$ (Table 2) is likely exceeded. The area exceeding this value in Europe, is however predicted to decrease from ca 30% in 1990 to only 1% in 2019 (Figure 7 right).

Estimated trends in $PM_{2.5}$ observations can be based on measurements in monitoring stations that are officially reported to the European Environment Agency (EEA) by its member countries, distinguishing traffic stations, industrial stations and background stations in rural areas (EEA2023b). During the period from 2000 to 2015, 96% of those measurements demonstrate a significant downward trend (EEA, 2023b). On average, concentrations of PM_{10} decrease by 2.8% per year. Despite this overall downward trend, there are still several regions across Europe where the concentration of $PM_{2.5}$ exceeds its limit value as illustrated for the year 2015 (Figure 8). Moreover, PM_{10} concentrations continue to surpass the EU limit values in a significant portion of Europe, exceeding the daily limit value of 50 μg m^{-3}.

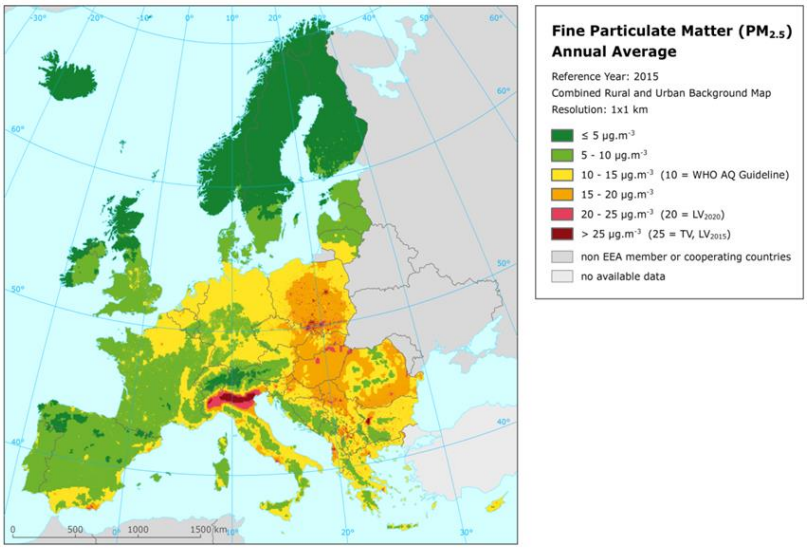


Figure 8. Map of the annual average measured concentrations of PM2.5 for the year 2015. The critical levels for PM2.5 are 5 µg m^{-3} (WHO, 2021) and limit levels of 25 µg m^{-3} (EC, 2008). Source: Horálek et al. (2018). The mapping is based on 'residual kriging' of air quality measurements, combined with chemical transport model results and other supplementary data (such as altitude and meteorology) to interpolate the results. The methodology does not allow for formal compliance checking with limit or target values.

3.2 Water quality: nitrate in groundwaters

Groundwater nitrate contamination occurs widespread across the EU (Oenema et al., 1998), particularly in areas with intensive livestock production (Mallin, 2000). As nitrate leaching negatively impacts the groundwater suitability for drinking water, diverse policies are at place to avoid contamination of groundwater with nitrate (Ward et al., 2018). About 9 million consumers are potentially exposed to drinking water exceeding the critical limit of 50 mg NO_3 l^{-1} (Erisman et al., 2011).

3.2.1 Health impacts and critical levels

The potential health effects associated with elevated nitrate levels are diverse and include reproductive problems (Kramer et al., 1996), methemoglobinemia (commonly known as "blue-baby" syndrome), and digestive tract and colorectal cancer (Van Grinsven et al., 2006). Infants are particularly vulnerable to methemoglobinemia, but the mortality rate is extremely low in Europe, and in other regions symptoms are often caused by pathogens present in drinking water rather than nitrate itself (Powlson et al., 2008). Excess nitrate in drinking water has also been linked to potential risks for certain types of cancer, specifically colorectal cancers (Van

Grinsven et al., 2010; Espejo-Herrera et al., 2016; Schullehner et al., 2018). While there is a growing body of epidemiological data supporting these associations, a lack of consensus remain due to inconsistencies between epidemiological and clinical findings (Ward et al., 1996, 2005; Van Grinsven et al., 2006; Gupta et al., 2017). Though clear cause-and-effect relationships between nitrate and health impacts could not be established, there are numerous indications that high nitrate levels might have negative impacts on human health, even for nitrate levels occurring below regulatory levels (Ward et al., 2005).

The WHO adopted a 10 mg l^{-1} NO_3-N limit for safe drinking water, being close to the EU limit value of 11.3 mg l^{-1} NO_3-N (50 mg l^{-1} of NO_3), limit values that are set to protect against methemoglobinemia and other human health risks (Gupta et al., 2000). Several studies show that even at lower limit values (5 to 6 mg l^{-1} NO_3-N) health risks might increase due to the formation of nitrosamines (Van Maanen et al., 1996) and associated risks for colon cancer (De Roos et al., 2003), bladder and ovarian cancer (Weyer et al., 2001), non-Hodgkin's lymphoma (Ward et al., 1996) and neural tube defects (Ward et al., 2005). Conversely, Powlson et al. (2008) concludes that evidence for methemoglobinemia and cancer at levels below 10 mg NO_3-N l^{-1} is scarce, whereas others (Avery, 1999; Addiscott & Benjamin, 2004) suggest that it is safe to raise the lower limit up to 65 – 100 mg l^{-1} of NO_3(14.7-22.6 mg NO_3-N l^{-1}). Though debated (Van Grinsven et al., 2006) and remaining high uncertainties, the strong carcinogenic effects of N-nitroso compounds in humans and associated health risks of nitrates in drinking water emphasize that adapting the lower limits requires a careful evaluation of these risks. At this moment this is not the case.

3.2.2 Geographic variation and trends in nitrate concentrations

Nitrate concentrations have been reported in groundwater between 2000 and 2015 in four year reporting periods, i.e. 2000-2003 (EC 2007), 2004-2007 (EC 2010), 2008-2011 (EC 2013), in more than 16,000 monitoring stations. This number more than doubled to ~35,000 for the 2016-2019 reporting period (EC 2021). Across 461 groundwater bodies, the nitrate concentration was relatively constant, varying between 20 and 21 NO_3 l^{-1} though earlier EEA reports showed an

increase from 21 NO_3 l^{-1} in 1992 to 23 mg NO_3 l^{-1} in 2015 (EEA, 2023a, Figure 9). This trend is consistent with monitored nitrate concentrations in wells, where 30% showed a decline and 36% showed an increase between 1996-1999 and 2000-2003 (EC, 2007), with no decline observed in the subsequent 10 years (EC, 2010) as well the years thereafter (EEA, 2023a).

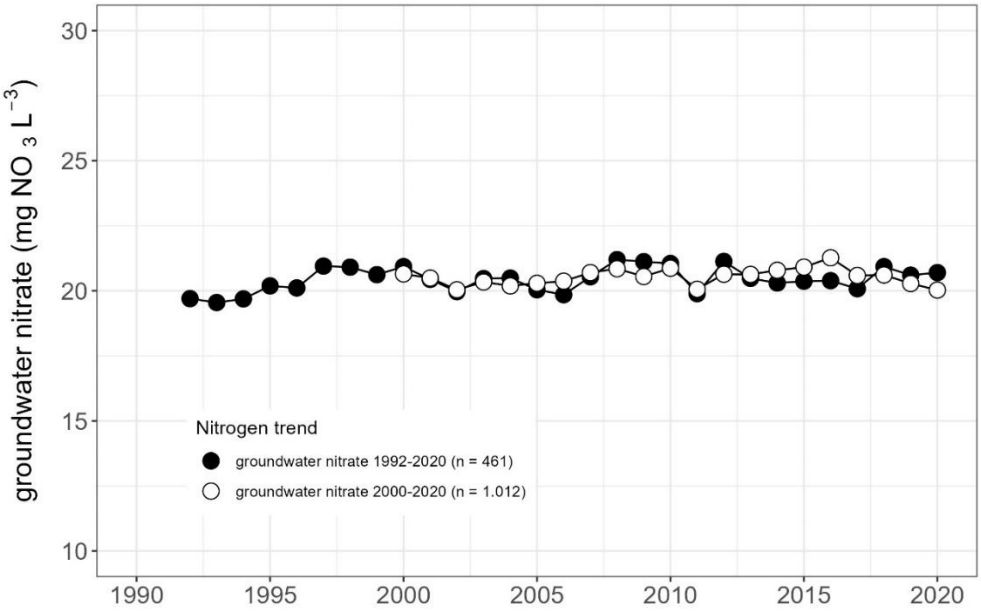


Figure 9. Trends in nitrate in European groundwater as reported by the European Environment Agency (EEA, 2023a). The geographical coverage is the 38 EEA member countries, but only complete time series are included. The shown time series are aggregated to European level by averaging across all sites for each year.

Nationally averaged groundwater nitrate concentrations are all below the critical level, although it occurs in some regions in Germany, Poland and Spain (Table 10, left) but national and even regional aggregation masks considerable variation at the scale of individual monitoring stations, with ~13% of groundwater monitoring stations across Europe exceeding the critical vale of 50 mg NO_3 l^{-1} (Figure 10). Between 1992 and 2004 this pattern has remained relatively stable (see Figure 9), lying around 10% (not shown in Figure 10). In the period 2005-2009, ca 15% of the stations exceeded the critical limit followed by a slight decrease in the period 2009-2015 (See results in Table 10 right for EU28, for the periods 2004-2007, 2008-2011 and 2012-2015. On average the water quality remained the same in 74% of the stations. In the years after 2015, the

number of groundwater monitoring stations with nitrate concentrations exceeding the 50 mg NO_3 l^{-1} has not been reduced (EEA, 2023a).

Note that large differences occur within countries, depending on soil geo-hydrology, land use, N fertilizer practices as well the depth of monitoring. The highest proportion of exceedance in 2009 was observed in Belgium (30%), Denmark (26%), Spain (22%) and Cyprus (19%) but high levels were also found in Austria, Germany, Bulgaria, Montenegro and Czech Republic (11-15%). The spatial variation in NO_3 concentrations coincides with the calculated spatial variation in nitrate leaching in 2010, which varied from <2.5 kg N ha^{-1} yr^{-1} in Southern Europe to >20 kg N ha^{-1} yr^{-1} in Western and Eastern Europe (De Vries et al., 2021).

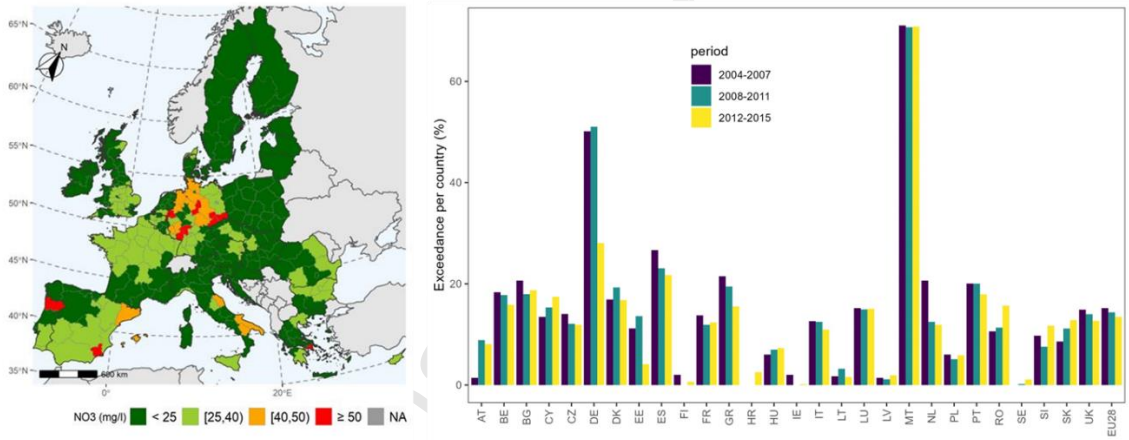


Figure 10. Spatial variation in annual average nitrate concentrations in groundwater for the reporting period 2016-2019 (left; source EEA, 2023), and percentage of sampling points exceeding the 50 mg NO_3 l^{-1} per country over periods 2004-2007, 2008-2011 and 2012-2015 (right; source EC, 2021).

In most cases, the percentage of declining trends is higher for rivers (see section 6.2) than for groundwater. This is probably reflecting, in part, the lag times associated with the transport of nitrate from soil layers to deeper groundwater (10 – 40 years). Declining trends in groundwater nitrate can typically be attributed to improvements in the management of agricultural land.

3.3 Other human related impacts

Other nitrogen emission impacts on human health

An indirect health effect of nitrous oxide emission is the destruction of the stratospheric ozone layer, thereby increasing UV radiation causing increased occurrence of skin cancers.

Ravishankara *et al.* (2009) showed that N_2O emissions are currently the most important O_3 depleting since the "Montreal Protocol on Substances that Deplete the Ozone Layer" reduced the use of halocarbon refrigerants, whereas Slaper et al. (1996) predicted that future skin cancer incidence at global level would double to quadruple. Recent calculations for Europe indicate that excess skin cancer incidences caused by ozone depletion might well rise to around 10-12% of the normal incidence around 2065 in large parts of the most populated areas in Europe (Van Dijk et al., 2008). Given the impact of N_2O on ozone measures taken to reduce N_2O (UNEP, 2013) might additionally help to reduce the risk of skin cancer.

Nitrogen also impacts human health via N induced pollen pollution and various diseases as summarized by Townsend *et al.* (2003). We describe here only the main impacts without analysing spatial and temporal variation across Europe. Human allergic response to pollen is a pervasive environmental health issue. Millions suffer from hay fever, allergenic rhinitis, and allergenic asthma following exposure and sensitisation to pollen (NIH, 1993). Pollen counts are rising not only due to climatic change, disturbance-driven changes in species composition, increased atmospheric CO_2 (Wayne et al., 2002), but also by increased N availability (Lau et al., 1995). The effects of N on pollen are likely to vary with species, but it is noteworthy that N additions to ragweed, a widespread producer of allergenic pollen, caused dramatic increases in pollen production (Townsend et al., 2003).

In addition, many infectious diseases are controlled by vector hosts. Some evidence suggests that the abundance and distribution of these hosts, including the mosquito hosts of malaria and West Nile virus, may be affected by N availability (Townsend et al., 2003). Other diseases potentially affected by eutrophication are cholera and schistosomiasis (Epstein, 1993; Colwell & Huq, 2001; Cottingham et al., 2003; McKenzie & Townsend, 2007), often occurring in the tropics (McKenzie & Townsend, 2007). Note that ecological interactions between vector hosts, insects and diseases are complex, driven not only by the organism's direct response, but also by its food sources and the parasitic and predatory species that affect its abundance (Comiskey et al., 1999). Experimental data on disease responses are rare and thus a comprehensive

assessment of nutrient-disease connections requires further research. This also holds for the influence of N:P ratios on human health, suggesting a potential role in the the proliferation of infectious diseases, including Zika and malaria, but also in non-infectious diseases, such as cancer (Penuelas and Sardans 2023)

Nitrogen emission impacts on visibility and materials

Impacts of air pollution caused by N emission not yet discussed include reduced visibility and damage to materials. Nitrogen containing aerosols contribute to visibility problems (Diederen et al., 1985; Kean et al., 2000). Ten Brink et al. (1996) found that aerosol light-scattering, in particular its humidity dependence, increases with the degree of air pollution. Around 1980 the median visibility in Western Europe was around 9 km (Diederen et al., 1985) in areas with high population density, high air pollution levels and a high relative humidity. Analysis of visibility data for eight Meteorological Office network sites in the UK indicated that around 1980 the visibility at those sites ranged between about 17 km at an urban (airport) site and 37 km at a rural site (Doyle & Dorling, 2002). Over the period from 1950 to 1997 the visibility improved due to changes in personal behaviour, fuel use and vehicle fleet efficiency. On the basis of daily-averaged visibility data during the period 1994-1998, visibility reduction appears to be significantly higher in Europe than in the United States (Husar et al., 2000). Currently, most of the European continent (93%) Is below the critical threshold for visibility reduction, i.e. an extinction of 5% (RIVM, 2001).

Nitrogen compounds can affect our cultural heritage, i.e. historical buildings and monuments, in particular in urban areas with high NO_x emissions and associated acidifying effects. Ammonia can be corrosive at very high concentrations and ammonium salts can be nitrified by bacteria forming nitric acid. This nitric acid can ‘dissolve’ the chalk of statues and buildings as demonstrated by Keuken et al. (1990) for an old church building in the Netherlands. Ammonia also corrodes some metals and composites and NO_x affects the deterioration of calcareous stone (Massey, 1999). The extent of corrosion can be higher by the presence of water (Massey, 1999). Copper, tin, zinc, and composites of these metals corrode in the presence of ammonia. NH_3 can

also have an effect on non-metal materials: it softens wood by interaction with the cellulose vessels, expand natural rubber and has impacts on concrete. Lastly, nearly all kinds of plastics are sensitive to ammonia (Erisman, 2000; Erisman et al., 2014).

4 Impacts on greenhouse gas emissions and radiative forcing

4.1 Relationships between reactive nitrogen production and radiative forcing

The human-induced disruption of the global N cycle by the release of reactive N has significant implications for the Earth's climate, as it impacts the ecosystem nitrogen and carbon (C) cycles, thereby influencing the emissions of greenhouse gases (GHGs) such as nitrous oxide (N_2O), carbon dioxide (CO_2), and methane (CH_4) from agricultural and natural systems (Butterbach-Bahl et al., 2011; Erisman et al., 2011). The main pathways how N_r inputs affect GHG emissions (Figure 11) include:

1. Direct and indirect emissions of N_2O from fertilizers and manure use in agriculture.
2. Changes in net ecosystem production (NEP) due to N inputs and associated carbon sequestration in biomass, soils and marine environments.
3. Increases in ozone (O_3) formation due to NO_x, which has direct warming effects (as O_3 is a GHG) and indirect warming effects by reducing NEP, impacting C sequestration, and affecting the atmospheric lifetime of CH_4.
4. Changes in the exchange of CH_4 between the biosphere and the atmosphere as response to N inputs to soils, freshwater ecosystems, and oceans.
5. Enhanced aerosol formation due to emissions of NO_x and NH_3, thereby influencing the radiation balance.

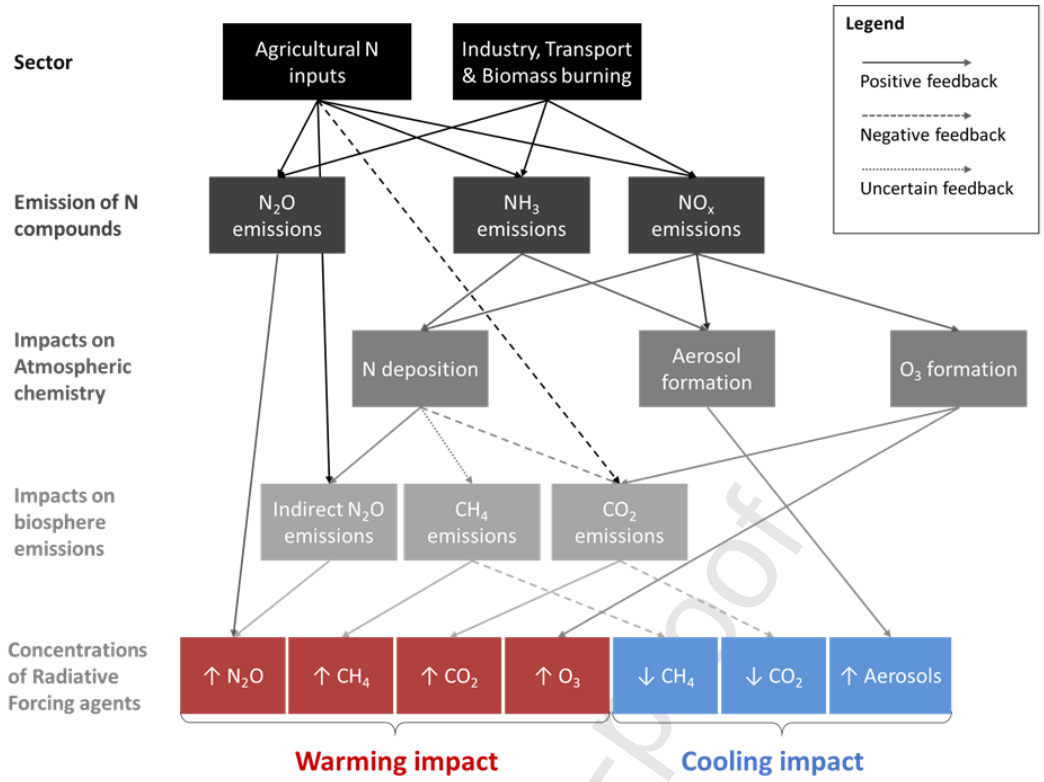


Figure 11. Overview of impacts of intended and unintended reactive nitrogen production on radiative forcing (De Vries et al., 2023).

First, the increase in N_r in the biosphere has led to a rise in N_2O emissions from various sources such as soils, sediments, and water bodies. This increase can be attributed to direct and indirect N_2O emissions, where the relative importance of both pathways remains uncertain (Voss et al., 2011). Direct N_2O emissions originate from the application of inorganic fertilizers, manure, and the cultivation of N fixating crops (Velthof et al., 2009, De Vries et al., 2010). Indirect N_2O emissions occur in terrestrial and aquatic systems (e.g. Hasegawa et al., 2000; Beaulieu et al., 2011; Yao et al., 2020) when these systems are enriched with N as a result of volatilization or leaching of N from surrounding agricultural areas (Mosier et al., 1998; Durand et al., 2011; Voss et al., 2013) where N_2O is produced during microbial conversion of ammonium to nitrate and finally to N_2 (Butterbach-Bahl et al., 2013). The microbial mediated N_2O release increases with substrate availability, high moisture content and temperatures, and soil pH, and consequently by management measures such as fertilization, liming and tillage (Li et al., 2005). Other anthropogenic sources of N_2O include emissions from manure storage, biomass burning, fossil fuel combustion, sewage, and industrial processes. The main natural N_2O sources include

forests (e.g. Kesik et al., 2005), coastal waters and oceans (Bange, 2006; Werner et al., 2007; Syakila & Kroeze, 2011), where O_2 is the main driver regulating N_2O (Bange, 2006). Globally, most of the N_2O emitted from aquatic systems is associated with agriculture (Yao et al., 2020).

Second, increased availability of N_r impacts CO_2 uptake in various ecosystems. In many terrestrial ecosystems, the biomass growth is limited by nitrogen availability (Vitousek & Howarth, 1991; LeBauer & Treseder, 2008). Increased N deposition enhances net primary production and stimulates carbon sequestration in biomass growth (Nadelhoffer et al., 1999; De Vries et al., 2006, 2008, 2009, 2014b; Solberg et al., 2009; Thomas et al., 2010) and soils (Conant et al., 2001; Vitousek & Howarth, 1991; LeBauer & Treseder, 2008) due to increased litter production and reduced decomposition (Janssens et al., 2010). An increase in N input might also accelerate SOM decomposition, whereas the actual sequestration in agricultural soils strongly depends on site history and soil, crop, and manure management (Schipper et al., 2010). Though forests account for only 60% of the global terrestrial NPP (Field et al., 1998), they account for more than 90% of the terrestrial C sink (Pan et al., 2011; Harris et al., 2021). At continuous high N inputs, however, forests can become N-saturated (Aber et al., 1989; Aber et al., 1998), resulting in tree growth decline and even death (e.g. Magill et al., 1997, 2004). Nitrogen limitation is also widespread in marine ecosystems. The increased availability of N in marine environments can enhance the growth of primary producers (Duce et al., 2008; Suntharalingam et al., 2012), such as phytoplankton, leading to greater carbon uptake and storage within marine ecosystems (Krishnamurty et al., 2007).

Thirdly, the increased emissions of NO_x have notable impacts on radiative forcing through two mechanisms. The first mechanism includes a positive impact on O_3 production (warming effect) and the second mechanism includes the long-term negative impact of NO_x on the lifetime of CH_4 (being oxidised by the positive impact of NO_x on the hydroxyl radical OH, resulting in a cooling effect), subsequently affecting the background O_3 concentration (Fiore et al., 2008; Isaksen & Dalsøren, 2011). The rising concentrations of tropospheric O_3 caused by NO_x emissions are a cause for concern. Ozone not only acts as a greenhouse gas, ranking as the third

most potent in terms of radiative forcing (Forster et al., 2007; Myhre et al., 2013), but also negatively affects global terrestrial primary production (e.g. Sitch et al., 2007), leading to decreases in crop yields (e.g. Van Dingenen et al., 2009), forest growth (e.g. Cailleret et al., 2018) and grassland productivity (e.g. Xia et al., 2021), followed by lower C sequestration in soils. Overall, Xia et al. (2021) estimated that elevated O_3 concentrations may have decreased global crop production by about 9%. These impacts are particularly important in forests due their long-term sequestration. Increased O_3 levels can limit the C sink of forest ecosystems (IPCC, 2007; Sitch et al., 2007; Wittig et al., 2009; Pretzsch et al., 2010), due to reduced net photosynthesis, accelerated leaf senesence and increased dark respiration (Matyssek et al., 2010a,b). The impact of NO_x-induced O_3 on biomass production and CO_2 sequestration in oceans is considered negligible.

Fourth, increased N_r inputs to both agricultural and forest soils have an impact on the uptake and release of CH_4 by well-drained soils as well of that of ruminants (Beauchemin et al., 2008). Several mechanisms have been proposed to explain this phenomenon (Schnell & King, 1994; Bradford et al., 2001; Bodelier & Laanbroek, 2004; Reay & Nedwell, 2004), and studies have shown a decrease in CH_4 consumption rates in response to N fertilization (Steudler et al., 1989; Van den Pol-van Dasselaar et al., 1999), commonly practiced in commercial forests to enhance productivity (Castro et al., 1994; Chan et al., 2005). However, there have been reports of insignificant or even opposing impacts. In wetland paddy soils N additions might reduce CH_4 emission due to CH_4 oxidizing bacteria (Bodelier & Laanbroek, 2004) but also increase CH_4 emission by increased rhizodeposition of C substrates fuelling methanogenesis (Fumoto et al., 2008). In addition, Whalen & Reeburgh (2000) and Borken et al. (2002) concluded that increased N deposition on boreal forest soils did not affect CH_4 uptake where Xia et al. (2020) found reduced CH_4 uptake only in subtropical and tropical forests at N addition levels that resemble N deposition rates. Aquatic ecosystems contribute to 53% of the total global CH_4 emissions with freshwater wetlands and lakes being the dominant source (Rosentreter et al., 2021) where the CH_4 emission increases by eutrophication (Beaulieu et al., 2019), in particular

when N limitation occurs (Maberly et al., 2020; Paerl et al., 2020; Janse et al., 2022). Overall, the relationship between N inputs and CH_4 soil release is complex and can vary depending on soil type, ecosystem type, and the magnitude of N addition.

Lastly, NH_3 and NO_x, together with SO_2, are precursors of secondary aerosols. These aerosols have a cooling impact in the atmosphere through the scattering of sunlight. Moreover, aerosols have various indirect effects, such as modifying the number of cloud condensation nuclei, which can lead to increases in cloud albedo and changes in cloud lifetime or thickness. Additionally, aerosols can alter the albedo of snow (Butterbach-Bahl et al., 2011) and the increase in diverse radiation might increase ecosystem production since photosynthesis seems more efficient under diffuse light conditions (Mercado et al., 2009).

4.2 Nitrogen-induced impacts on the GHG exchange and ozone formation

Impacts on GHG exchange

Impacts of N inputs on GHG exchange have been assessed by empirical models (e.g. De Vries et al., 2011b, 2017) and process-based models (e.g. (Tian et al., 2011) . Empirical models quantify the impact by multiplying linear response ratios, estimating the exchange (emission or uptake) of CO_2-C, N_2O-N and CH_4-C per unit N, with the N input, while differentiating between N sources (e.g. fertilizer N input, N deposition, N runoff) and ecosystems (e.g. agricultural soils, forest ecosystems, aquatic ecosystems). De Vries et al. (2011b) thus estimated CO_2-C, N_2O-N and CH_4-C exchange factors per kg N input, for agricultural and non-agricultural terrestrial systems and for aquatic and marine ecosystems as presented in Table 3. The factors terrestrial ecosystems are based on a summary review by De Vries et al. (2009) for CO_2-C exchange, on a literature review by Lesschen et al. (2011) for N_2O-N exchange and on a meta-data analysis by Liu and Graever (2009) for CH_4-C exchange. The factors for aquatic systems were assessed by De Vries et al. (2011) for CO_2-C exchange and derived from Eggleston et al. (2006) for N_2O-N exchange and for marine systems they were derived from Duce et al (2008) for both CO_2-C and N_2O-N exchange.

Table 3 N induced exchange (emission or uptake) factors for CO_2-C, N_2O-N and CH_4-C, based on data presented in De Vries et al. (2011b).

System	F_{CO2-C} (kg CO_2-C/kgN)	F_{N2O-N} (kg N_2O-N/kgN)	F_{CH4-C} (kg CH_4-C/kgN)
Arable land	-0.53	0.25-0.0225[1]	0.012
Grass land	0	0.0017-0.0040[1]	0.016
Semi-natural	-25 (0 - -50)	0.018	0.016
Forest	-50 (-25 - -75)	0.018	0.016
Aquatic	-1.07	0.0075	-
Marine	-4.0	0.03	-

[1] Variation as a function of N source, land use and soil as given in De Vries et al. (2011b).

Using these ratios, the direct and indirect impacts of N_r on CO_2, N_2O and CH_4 exchange at European scale for the year 2000 has been quantified by multiplying them with the relevant N input fluxes (De Vries et al., 2011b). The total impact was estimated on -2.5 Tg CO_2-eq yr^{-1} with a net GHG production of 160 Tg CO_2-eq yr^{-1} via N_2O and a net GHG consumption of -172 Tg CO_2-eq yr^{-1} via CO_2 sequestration, predominantly occurring in forests (Table 4). This implies that the negative impacts of N_2O emissions are outweighed by the positive effects of C sequestration in forests, and to a lesser extent by other ecosystems.

Table 1 Estimated impacts of N use in agriculture at EU 27 level on the emissions of CO_2, N_2O and CH_4 in Tg CO_2 equivalents in 2000, as derived after De Vries et al. (2011b).

System	Area (million km^2)	N inputs (Tg N yr^{-1})	CO_2 (Tg CO_2 eq yr^{-1})	N_2O (Tg CO_2 eq yr^{-1})	CH_4 Tg CO_2 eq yr^{-1})	GHG (Tg CO_2 eq yr^{-1})
Arable land	1.40	13.7[1]	-26.7	96.4	4.87	74.4
Grass land	0.56	8.9[1]	0.00	38.5	4.21	42.9
Semi-natural	0.43	0.15[2]	-13.9	1.28	0.07	-12.5
Forest	1.45	0.61[2]	-112	5.10	0.30	-106
Aquatic	0.05	1.50[3]	-5.90	5.24	-	-0.66
Marine	3.28	0.96[2]	-14.1	13.4	-	-0.70
Total	7.16	25.6	-172	160	9.5	-2.5

[1] Includes N inputs by fertilizer, manure, and NH_3-N deposition.
[2] NH_3-N deposition only. The marine area is limited to the continental shelf.
[3] N runoff from agriculture (estimated at 1.46 Tg N yr^{-1}) and NH_3-N deposition on aquatic ecosystems (estimated at 0.036 Tg N yr^{-1}).

Similar findings were presented by De Vries et al. (2023) for the global scale while quantifying the N induced impacts on GHG exchange in 2010 using an empirical approach and the Dynamic Land Ecosystem Model (Tian et al., 2011). The N induced impacts on CH_4 exchange in

terrestrial ecosystems are very limited in both approaches. The N-induced CO_2 sequestration dominates in croplands and forests, while the contribution of pastoral grassland (or pasture and range land) is limited. Most of the N_2O emission originated from cropland (>60%), followed by grassland (~30%). Globally, the N-induced effect on GHG exchange in inland & coastal waters is a net warming, with N-induced CH_4 emissions being much larger than in terrestrial ecosystems (De Vries et al., 2023).

Impacts on exchange rates of CO_2 through the formation of ozone

Negative impacts of N inputs on C sequestration in forests (trees) and soils via ozone exposure (e.g. Ashmore, 2005; Paoletti et al., 2007) has been assessed via empirical models by Wittig et al. (2009) and Xia et al. (2021). Ozon exposure and uptake via leaf stomata would decline the photosynthesis of living plants and trees and subsequently the carbon sequestration (Sitch et al., 2007; Goldstein et al., 2004). Observations from Wittig et al. (2009) suggest that the forest growth would remain unaffected as long as the O_3 concentration is below 10 ppb after which the growth declined with 1% per 4 ppb O_3 increase up to 40 ppb, and a 1% growth reduction per 6 ppb O_3 increase above 40 ppb. Results from Xia et al. (2021) showed that elevated O_3 exposure significantly decreased plant photosynthesis (26%), crop yield (26%), net primary productivity (22−29%) and soil C pools (5.8%) from global terrestrial ecosystems. These results are in line with estimates of forest yield losses in Europe of around 10% (Broadmeadow, 1998). Similar findings were found for empirical forest models using exceedances of hourly ozone concentrations over 40 ppb and the accumulated phytotoxic ozone dose (Büker et al., 2015) as well for the Dynamic Land Ecosystem Model that uses an empirical ozone response coefficient to analyse the O_3 effects on photosynthesis (Tian et al., 2016). On a global scale the N induced O_3 effects reduced the C sequestration by 359 to 419 Tg CO_2-eq yr^{-1} for biomass and by 192 to 342 Tg CO_2-eq yr^{-1} for soil. Hence, the O_3 effects on reduced C sequestration are twice as high for biomass (limited to forests only) than for soils. Assuming that present elevated O_3 concentration is for about 70% due to the increase in NO_x emissions (see Section 2.1), this implies that NO_x emissions are responsible for about 5-7% of the forest yield decline in Europe.

Impacts on ozone forcing, methane lifetime reduction and particulate matter formation

Anthropogenic N induced increases and O_3 induced reductions in terrestrial carbon sink, NOx emissions also affect the radiative forcing through their effects on short-lived climate forcers. This includes a warming effect by ozone formation and a cooling effect by reducing the CH_4 lifetime and by aerosol formation. IPCC estimates (Table 5) show that the cooling effect of reduced CH_4 lifetime and of N induced aerosol formation is significantly larger than the warming effect of NO_x on short-lived O_3 (Myhre et al., 2013; Szopa et al., 2021). Even when accounting for the NO_x induced impact of O_3 on the terrestrial carbon sink, the net impact of NO_x emissions is therefore resulting in a cooling effect (Aamaas et al., 2017).

Table 5. Direct radiative forcing (RF, in W m^{-2}) for the period 1750-2011 excluding aerosol-cloud interactions[1] by emitted components as presented in IPCC AR5 and IPCC AR6 (values in brackets; both from Figure 8.17 in Myhre et al., 2013).

N Component emitted	Reaction products				Total
	Ozone formation	Methane (including background ozone)	Nitrate aerosols	Sulphate aerosols	
NO_x	0.14 (0.13)	-0.25 (-0.38)	-0.04 (-0.02)	-	-0.15 (-0.27)
NH_3			-0.07 (-0.03)	0.01 (-)	-0.06 (-0.03)
(NO_x+ NH_3)	0.14 (0.13)	-0.25 (-0.38)	-0.11 (-0.05)	0.01 (-)	-0.21 (-0.31)

[1] *Short-lived climate forcers (SLCFs) such as aerosols do not only directly change the radiative forcing, but also cause a number of secondary radiative responses on a short time scale ('rapid adjustments'), for instance aerosols modifying cloud lifetime and cloud brightness. Since IPCC's AR5, the concept of 'effective radiative forcing' (ERF) has been used which includes both the instantaneous radiative forcing (IRF) and these so-called rapid adjustments.*

4.3 Geographic variation and trends in nitrogen induced greenhouse gas emissions

Figure 12 shows the geographic variation in net GHG exchange due to agricultural N-induced GHG emissions for the years 1990 and 2015, using the approach by De Vries et al. (2011) with N emissions being calculated for ca 40.000 calculation units accounting for spatial variability in soil type, land use, climate, and fertilizer practices. The CH_4 and CO_2 emissions or sinks in response to N inputs were estimated using the response ratios given in Table 3. The results illustrate that not only Scandinavia is a net GHG sink because of its large area of forests, but also substantial parts of Central and Southern Europe even though the forested area is limited.

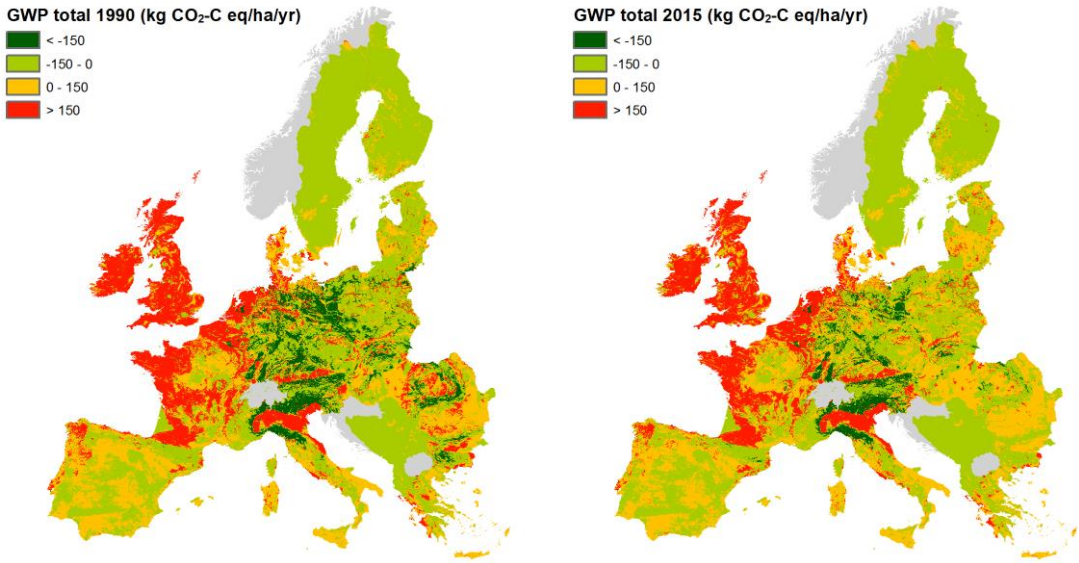


Figure 12. Geographic variation in the average agricultural N-induced global warming potential due to N_2O-N emission, CH_4–C uptake and CO_2-C sequestration in 1990 (left) and 2015 (right). Results are an update of the assessment for the year 2000 given in De Vries et al. (2011a).

This is because of the larger atmospheric NH_3-N input (up to 25 kg NH_3-N) and thus higher C sequestration by these forests (De Vries et al., 2011a). The change in N inputs in agriculture between 1990 and 2015 has been limited and thus results are quite comparable.

5 Impacts on terrestrial ecosystems

5.1 Relationships between elevated nitrogen inputs and ecosystem impacts

Nitrogen deposition is a main driver of plant biodiversity loss in terrestrial ecosystems (Emmett, 2007; Bobbink et al., 2010; Dise et al., 2011; Sala et al., 2000; Vellend et al., 2013) via direct and indirect effects. Direct effects occur due to high concentrations of NO_x and NH_3 or ozone (section 5.2) whereas indirect effects occur via eutrophication and acidification (section 5.3). The actual impacts depend on the N accumulation rate and exposure duration (Figure 13).

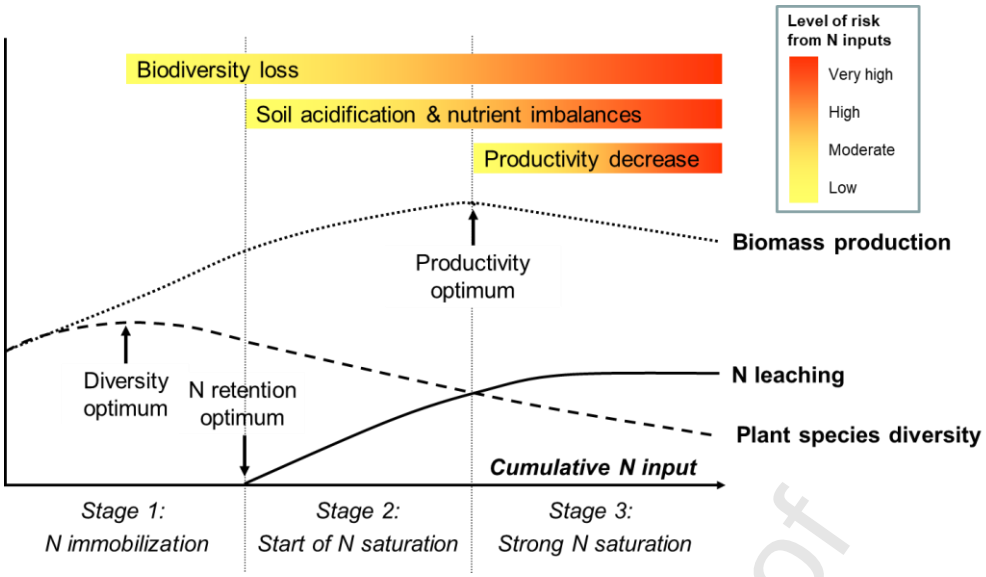


Figure 13. Hypothetical relationship between cumulative nitrogen input and biomass production (dotted line), plant species diversity (dashed line) and nitrogen leaching (solid line), indicating three stages of progressive nitrogen saturation. Figure from De Vries and Schulte-Uebbing (2018), being an update of the original figure by Aber et al. (1989), further updated by Aber et al. (1998), Gundersen et al. (2006) and Bobbink et al. (2010).

At very low N deposition levels and low cumulative N inputs (stage 1), plant species diversity may not decrease (or even slightly increase) with increasing N inputs up to a threshold. After passing the threshold, plant communities shift towards more nitrophilic species (Ellenberg, 1985; Bobbink et al., 1998; Bobbink & Hettelingh, 2011) due to asymmetric N utilization and growth rates among species (Figure 13, dashed line). Plant species diversity of N-limited ecosystems usually decline at deposition levels above 5 kg N ha^{-1} yr^{-1} (Stevens et al., 2010a), although critical deposition levels up to 10 kg N ha^{-1} yr^{-1} have been found (Roth et al., 2013). At this stage, most of the N inputs is retained in biomass or soil, and N leaching is generally negligible (Aber et al., 1998). With increasing N inputs (stage 2), the ecosystem starts to saturate and N leaching increases (Figure 13, solid line). Long-term observations from hundreds of European forest plots show increasing N leaching rates when N deposition exceeds the 10 kg N ha^{-1} yr^{-1} (Dise et al., 2009). At this stage, plant species diversity further declines due to competition exclusion or increased susceptibility to secondary stressors, such as pathogens, herbivory, frost and drought. The increased nitrate (and associated base cation) leaching decreases the acid neutralising capacity and the soil starts to acidify. Biomass productivity, on the other hand, still increases whereas adverse impacts on plant species diversity and soil quality

already occur (Figure 13, dotted line). When the N inputs increase further (stage 3) soil acidification and associated release of aluminium (with toxic effects on fine roots and mycorrhiza) and nutrient imbalances (e.g. base cation deficiencies and excess of N) may reduce biomass production and further reduce plant species diversity. Further N accumulation can even deplete seed banks (Basto et al., 2015), potentially limiting the recovery of plant species diversity following a decrease in N deposition. Results from many long-term N addition and forest monitoring studies in boreal and temperate regions indicate that these adverse impacts occur when deposition exceeds the 15–35 kg N ha^{-1} yr^{-1} (De Vries et al., 2014b, c).

5.2 Direct impacts of exposure to ammonia, nitrogen oxides and ozone

5.2.1 Vegetation impacts and critical levels

Adverse effects of NH_3 on vegetation occur when the rate of foliar uptake is greater than the rate and capacity for in vivo detoxification. The sensitivity of plant species to NH_3 increases from lichens to native vegetation, to forests up to agricultural crops. Adverse effects on higher plants includes damage of the epicuticular wax layer, increased susceptibility to drought stress due to prolonged stomatal opening and higher transpiration rates, and an enhanced risk for fungal infections and pest attacks (see Krupa (2003) and Cape et al. (2009) for an overview). Critical concentrations to avoid adverse impacts by *long-term exposure* to NH_3 have been set at annual average concentrations of 1 to 3 μg NH_3 m^{-3}, with the lower level for lichens and bryophytes and the upper level for higher plants, including trees (Cape et al., 2009). Critical levels for *short-term* (daily or monthly) exposures to NO_x are set at 75 μg NO_x m^{-3} and at 23 μg NH_3 m^{-3} for higher plants (Table 6). Critical levels are not available for intensively managed agricultural grasslands and arable crops, which are often sources rather than sinks of ammonia.

Adverse effects of elevated NO_x concentrations also reduce plant growth for both natural vegetation and agricultural crops (Van der Eerden & Duym, 1988). Forests have been considered less sensitive to NO_x than other (semi)natural vegetation. The mean annual critical level for NO_2 is set at 30 μg m^{-3} (CLRTAP, 2004; WHO, 2001), based on the impacts of both NO and NO_2 but expressed as NO_2.

Table 6 Critical levels (μg m^{-3}) in view of vegetation impacts used for short-term and long-term exposures to NO_x (expressed as NO_2) and NH_3[1].

Exposure	Vegetation	NO_x	NH_3
Long term (Annual mean)	Lichens and bryophytes[1]	-	1[2]
	Higher plants[3]	30[4]	3[2]
Short term[5]	Higher plants[3]	75[4]	23

[1] Includes ecosystems where lichens and bryophytes are key parts of the ecosystem.

[2] Based upon Cape *et al.* (2009). Fir higher plants, a range is suggested of 2-4 μg m^{-3}. See also Sutton et al. (2009)

[3] Includes heathland, grassland and forest ground flora in case of NH_3. For NO_x it also includes pastures and arable crops.

[4]. Basis for both values is WHO (2000) and used under the UNECE Convention on Long-range Transboundary Air Pollution, CLRTAP. Sutton (2019), however, indicates a lower uncertainty bound for the most sensitive vegetation of 15 μg m^{-3} NO_x expressed as NO_2.

[5] 24 hour mean for NO_x and monthly mean for NH_3.

The knowledge to establish separate critical levels for the two gases is still lacking, even though there is evidence that NO is more phytotoxic than NO_2 (Morgan et al., 1992; UNECE, 2009). In 2000, the WHO set a lower uncertainty bound of 15 μg m^{-3} NO_2 to protect the most sensitive vegetation (Sutton, 2019).

Adverse impacts of O_3 includes growth changes, yield losses, visible injuries, and reduced seed production. Most severe impacts are caused by the ozone that is taken up through the stomata into the leaf interior (Reich, 1987; Ashmore et al., 2004; Manning 2005), leading to stomatal closure and reducing the uptake of CO_2 and associated photosynthesis (Karnosky et al., 2003). Critical levels for forests trees have therefore been set for the stomatal uptake (flux) of ozone rather than for the O_3 concentration (e.g. Karlsson et al. 2004; Matyssek et al., 2007). The adverse impacts are not only shown in controlled experimental studies (Matyssek et al., 1995; Sandermann et al., 1997; Novak et al. 2005) but also in field and modelling studies in Europe, where ozone have reached concentrations high enough to produce phytotoxic effects (Taylor et al. 1994; Skärby et al. 1998; Matyssek & Innes 1999; Wittig et al., 2009). Observed O_3 impacts have even led to considerable economical losses (Ashmore 2005, King et al. 2005, Morgan et al. 2006; Matyssek et al. 2010a, 2010b; Pretzsch et al. 2010). The estimated yield losses for the EU25 range between 4% and 5% for wheat, between 21% and 27% for soybean, between 3% and 5% for rice and maize, and is around 10% for forests (Van Dingenen et al., 2009; Hollaway et al., 2012; Broadmeadow, 1998).

Unlike critical levels for NO_x and NH_3, which are described in terms of mean annual, monthly or daily concentrations (Table 6), critical levels for O_3 are defined as cumulative concentration exposures or fluxes through plant stomata. A commonly used indicator for cumulative concentration exposures is AOT40, being the accumulated hourly ozone concentration over a concentration threshold of 40 ppb for specific time intervals during the day and for a given period over the year, expressed in ppm hours. There are various definitions for AOT40, depending on assumed height of the ozone concentrations (top of crop or 2-3m), whether daytime is defined by fixed hour periods or by global radiation, and length of accumulation period (Mills et al., 2018). Here we use the CLRTAP definition, with AOT being related to daylight hours and using a period of 3 months (May-July) for crops and a latitude-dependent period of 6 months or forests. Critical AOT40 levels thus derived, corresponding to a 2-10% reduction in yield or biomass, are set at 2 - 6 ppm hours, depending on the vegetation (UBA, 1996, Table 7).

Table 7 Critical levels used for concentration-based exposures (AOT40) and flux-based exposures (POD_1, POD_3 or POD_6) to O_3.

Vegetation	Effect considered	O_3 exposure term	
		AOT40[1] (ppm h)	POD[2] (mmol m^{-2})
Crops			
- *Wheat*	5% yield reduction	3	POD_6 of 1
- *Potato*	5% yield reduction	-	POD_6 of 5[3]
- *Tomato*	5% yield reduction	6	POD_6 of 2
- *Generic crop*	5% yield reduction	-	POD_3 of 3[4]
Forests			
- *Coniferous trees (Norway spruce)*	2% growth reduction	5[4]	POD_1 of 8[5]
- *Deciduous trees (birch and beech)*	4% growth reduction	5[4]	POD_1 of 4[5]
Semi-natural vegetation			
- *grasslands*	10% biomass reduction	5	POD_1 of 2[6]

[1] Based on Mapping Manual (UNECE, 2009)
[2] Based on Harmens et al. (2010).
[3] A correction of the value of 4, as given Harmens et al. (2010), corrected in UNECE (2010).
[4] Based on a 5% yield reduction, using results presented in Harmens et al. (2010).
[5] Using 5% (10%) growth reduction as the effect considered leads to a POD_1 of approximately 20 (40) for Norway spruce and 5 (10) for beech/birch.
[6] This refers to both productive grasslands and grassland areas of high conservation value for which Trifolium spp (clover species) are used as an indicator species

The Phytotoxic Ozone Dose (POD), being the accumulated hourly stomatal O_3 flux over a flux threshold expressed in mmol m^{-2} (UNECE, 2009; Harmens et al., 2010; ICP Vegetation, 2010), is a more accurate indicator, accounting for the impact of climatic conditions on the stomatal opening, moisture availability and growth stage (e.g. Ashmore et al., 2004). Critical POD

values, calculated with a flux a threshold of 1 nmol m^{-2} s^{-1} for (semi-)natural vegetation and forests (POD_1) and a flux threshold of 6 nmol m^{-2} s^{-1} for agricultural and horticultural crops (POD_6), vary from 1 to 8 mmol m^{-2} (Table 7).

5.2.2 Geographic variation and trends in pollutant exceedances of critical level

Trends in average NO_x concentrations, denoted in NO_2, in three European regions and the overall average over Europe varied between 1-8 μg NO_2 m^{-3} in the period 1990-2019 (Figure 3). The geographic variation indicates that the critical level of 30 μg NO_2 m^{-3} for higher plants was exceeded in a few areas of Europe in 1990 (Figure 14), mostly around large urban centres and intensive agricultural and industrial areas, as in the Netherlands, the German Ruhr area, and the Po valley in Italy. In 2019 these exceedances hardly existed any more.

Despite decreasing NH_3 emissions over Europe in the period 1990-2019 (Figure 2) the NH_3 concentrations show a slight steady increase from 1995 onwards, especially in the Southern and Central parts of Europe (Figure 15 left) in line with observations (see https://www.emep.int/mscw/index.html).

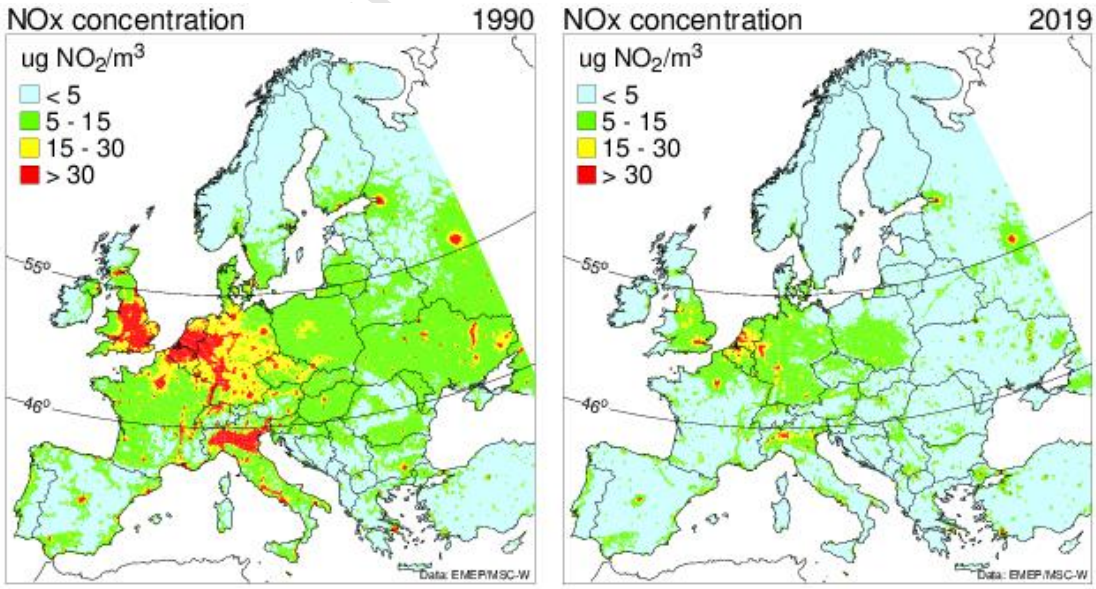


Figure 14. Geographic variation in modelled NO_x concentrations in 1990 and 2019. The 46ºN and 55ºN parallels separate three regions 'North', 'Central' and 'South'.

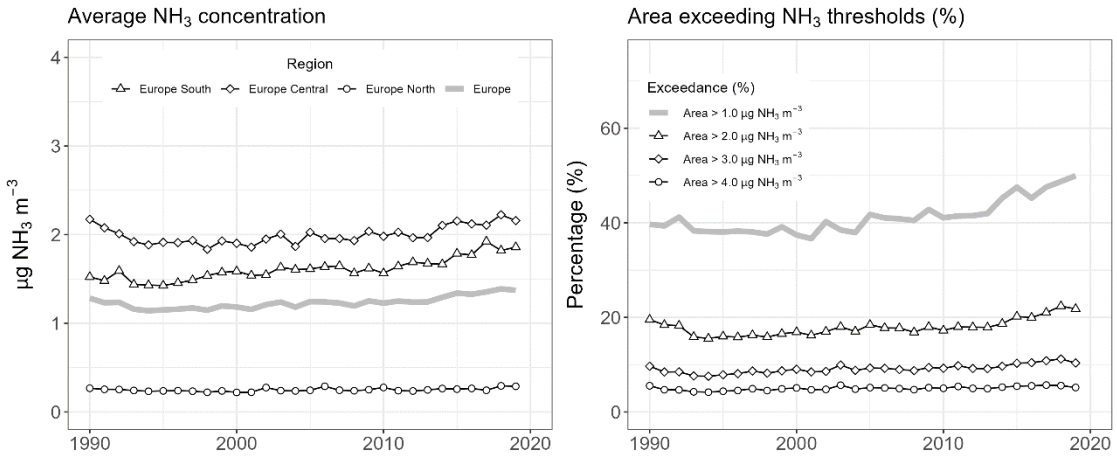


Figure 15. The 1990–2019 temporal development of modelled average NH_3 concentrations in the three European regions and overall European average (thick grey line; left) and the percentage of European land area (west of 40ºE) with NH_3 concentration above critical limits of 1 µg NH_3 m^{-3} for lichens and bryophytes and of 3 (uncertainty range 2-4) µg NH_3 m^{-3} for higher plants (right).

The estimated areas with NH_3 concentration exceeding the critical limit of 1 µg NH_3 m^{-3} for lichens thus increased from ca 40% to 50% (Figure 15 right). This increase is caused by the large reduction in SO_x emissions and associated ammonium sulphate aerosols, leading to elevated concentrations of ammonium nitrate aeresols (e.g. Fagerli & Aas, 2008). Unlike ammonium sulphate, NH_4NO_3 is in equilibrium with gaseous NH_3 and HNO_3, being only a partial sink of NH_3. In addition, decreasing levels of SO_4 have led to less acidic surfaces on vegetation, reducing the deposition sink of NH_3 (Fowler et al., 2009). The area exceeding the critical limit for herbaceous plants of 3 µg NH_3 m^{-3} stayed almost constant near 10% (Figure 15). The limit of 1 µg NH_3 m^{-3} for lichens and bryophytes is violated throughout most European countries, both in 2015 and 2019, except for Fennoscandia (Figure 16). The total area exceeding this limit also increased (Figure 15). The limit for herbaceous plants of 3 µg NH_3 m^{-3} is also exceeded exceed throughout the whole of Europe, both in 1990 and 2019, but specifically in in high livestock regions in Western Europe, eastern Ireland and Northern Italy (Figure 16).

The simulated averaged AOT40 and POD values, using a latitude-dependent growing period (Simpson et al., 2012), showed a steady decline from 1995 onwards, especially in Southern and Central Europe (Figure 17 left). The percentage of the European land area exceeding the critical

AOT40 value of 5 ppm-hours for forests and grasslands (Table 8) slightly decreased from 85 to 75% (Figure 17 right).

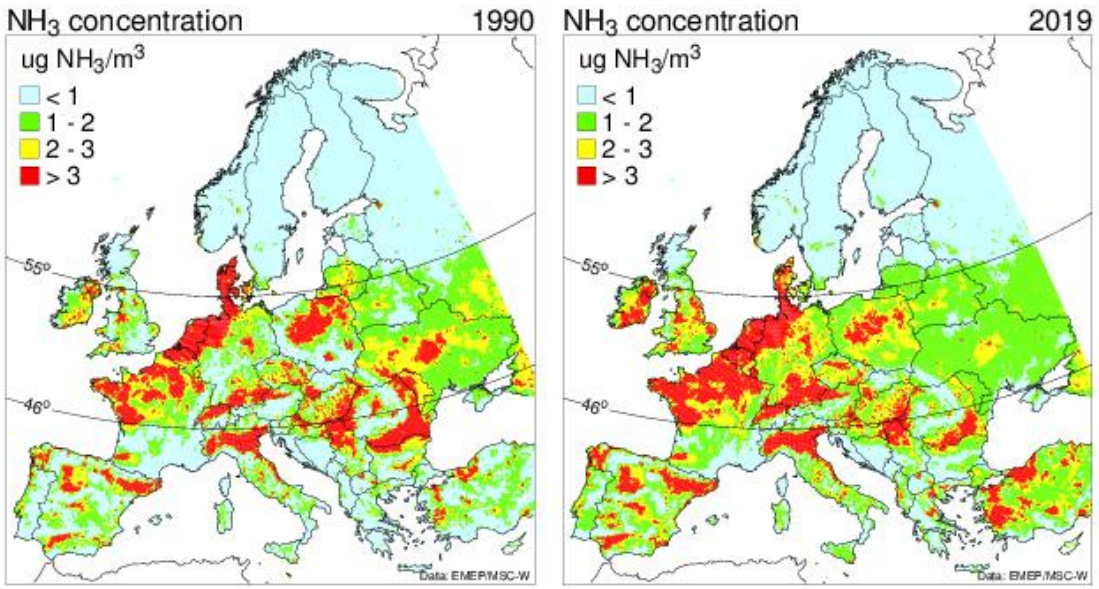


Figure 16. Geographic variation in modelled NH_3 concentrations (in µg m^{-3}) in 1990 and 2019. The 46ºN and 55ºN parallels separate three regions 'North', 'Central' and 'South'.

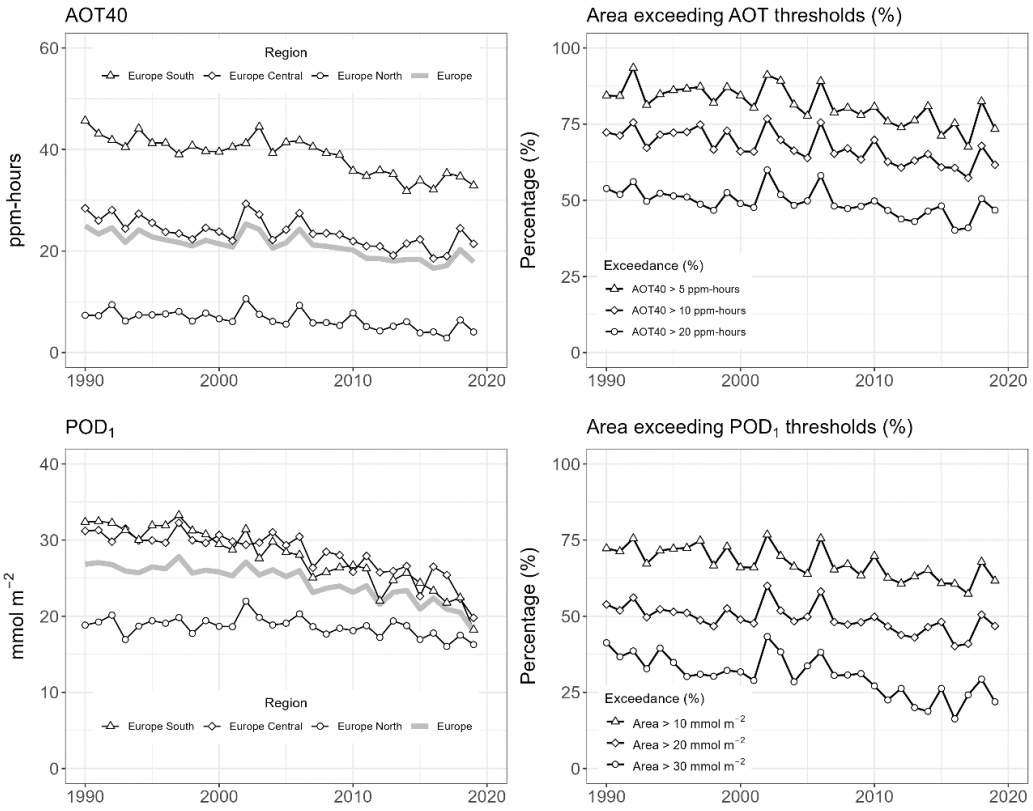


Figure 17. The 1990–2019 temporal development of modelled average AOT40 values (top left) and POD_1 (bottom left) in three European regions and overall average (thick grey line) and the percentage of European land area (west of 40ºE) with AOT40 above 5, 10 and 20 ppm-hours (top right) and POD_1 above 10, 20 and 30 mmol m^{-2} (bottom right).

Similar results were found for the critical value of 10 ppm-hours (from 73% to 65%) but the total area exceeding the 20 ppm-hours stayed relatively constant, fluctuating around 50% (Figure 17, top right).

Unlike AOT40, the change in areas exceeding a POD_1 of 10 and 20 mmol m^{-2}, related to ca 10-20% growth reduction, changed less than the areas exceeding a POD_1 of 30 mmol m^{-2} (Figure 17, bottom right). The spatial variability in AOT40 and POD_1 showed a clear gradient from Northern to Southern Europe, but this trend was much more clear for AOT40 than for POD, with highest values in Italy and in coastal areas (Figure 18).

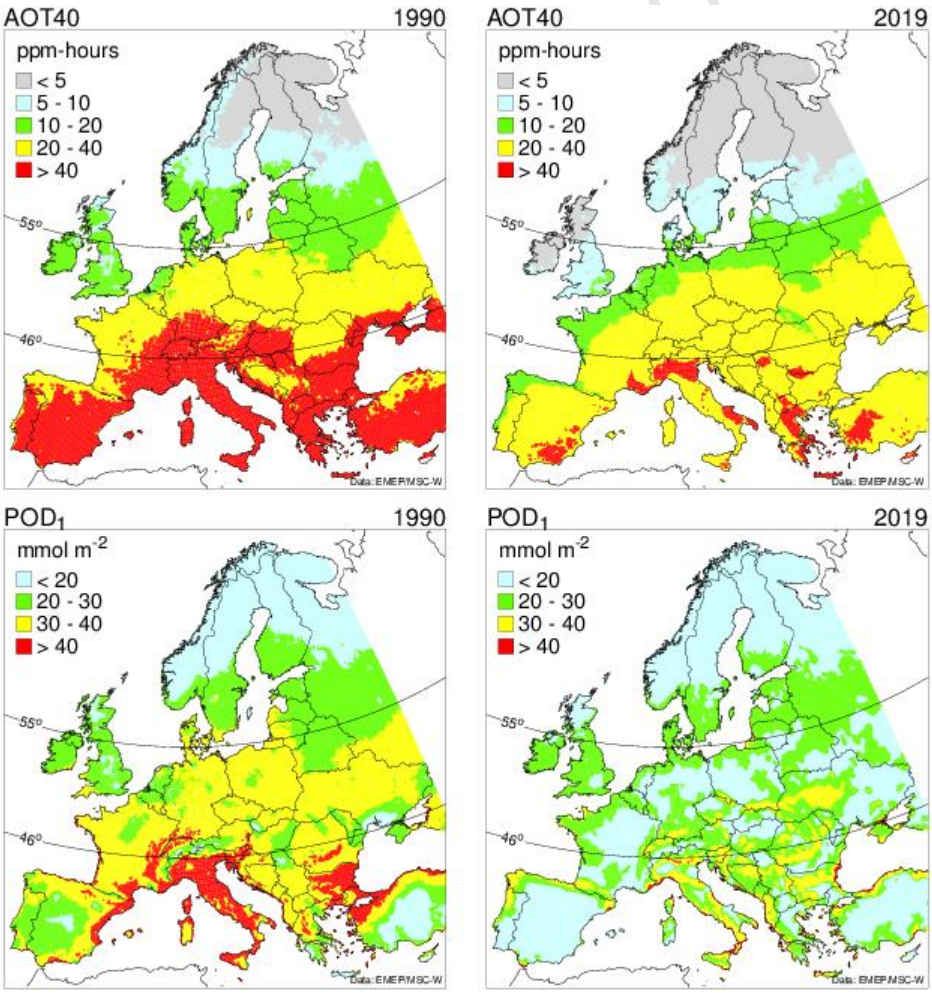


Figure 18. Geographic variation in modelled AOT40 values (in ppm h^{-1}) (top) and phytotoxic ozone dose (in mmol m^{-2}) above 1 nmol $m^{-2}s^{-1}$ (POD_1) for trees (bottom) in 1990 and 2019.The 46ºN and 55ºN parallels separate three regions 'North', 'Central' and 'South'.

The high values near the coast are driven by the fact that O_3 deposition on sea is almost zero (Klingberg et al., 2012) implying that O_3 can build up over sea areas from continental plumes containing NO_x and flow inlands from the sea at a later moment.

5.3 Soil-mediated impacts in forests and related critical loads

External N inputs cause soil acidification as a result of direct H^+ inputs and indirect H^+ inputs originating from nitrification and NO_3^- leaching, where the latter causes a decline in base cations and an increase of toxic Al (Bowman et al., 2008; De Vries et al., 2015). Enriching the ecosystem with N enhances the capacity of soils to supply N and supports the abundance of nitrophilic species. The fate of deposited N and its impacts in ecosystems have extensively been studied using ^{15}N tracer experiments, long-term monitoring networks, N addition experiments and regional studies across N deposition gradients (Du and De Vries, 2023a).

5.3.1 Impacts on forest ecosystems, their services and critical loads

In Europe's forests, N deposition causes a variety of changes (Table 8), including impacts on soil (solution) chemistry, and increased leaching of N from forest soils to surface and ground waters (Dise et al., 2009; Gundersen et al., 2006), soil tree productivity (Perkins et al., 2000; De Vries et al., 2006, 2017b; Erisman & De Vries, 2000), tree nutrition reflected in foliar nutrient concentrations (Jonard et al., 2015; Sardans et al., 2016b; Waldner et al., 2015), sensitivity of trees to biotic and abiotic stress (Bobbink & Hettelingh, 2011), the composition of understory vegetation (Dirnböck et al., 2014; Van Dobben & De Vries, 2017; Perring et al., 2023) and ectomycorrhizal fungal and microbial communities (Van der Linde et al., 2018; Forstner et al., 2019; Lilleskov et al, 2023). A recent overview of those effects is summarized in Du and De Vries (2023c). Those impacts of atmospheric N deposition affects the forest ecosystem services such as wood production (provision service), carbon sequestration (climate regulation service), buffer capacity (water quality regulation service) and pest/disease regulation (see Table 8).

Table 8. Possible effects of increased atmospheric N loading and exposure to NO_x and NH_3 on forest ecosystems (after Erisman & de Vries 2000).

Forest ecosystem compartment	Effects	
	Chemistry	Ecosystem
Soil (solution)	- increased N concentrations in soil (solution)	- increase in nitrophilous species/ decrease in biodiversity - increase in NO_3 leaching
	- increased ratios of NH_4 and Al to base cations	- inhibition of uptake (nutrient imbalances) - root damage and mycorrhiza decline
Trees (foliage)	- increased N concentrations in foliage	- nutrient deficiency absolute or relative (to N), imbalanced nutrition /discoloration - increased biomass production/ water demand - increased ratio of foliage to roots (risk of drought and nutrient deficiency - increased frost sensitivity - increased parasite injury (insects, fungi, virus)
	- increased arginine concentrations in foliage	- growth reduction

Nitrate leaching and soil acidification

A notable indication of adverse impacts of N inputs in forest ecosystems is increased leaching of N, causing increased NO_3 concentrations in soil solution (Dise & Wright, 1995). Site properties controlling the nitrate leaching are C:N ratio of the forest floor (Gundersen et al., 1998a), clay content, soil pH, groundwater depth and a variety of stand characteristics controlling the ecosystem N cycling (Lovett & Goodale, 2011 Driscoll et al., 2001; Wessel et al., 2021).

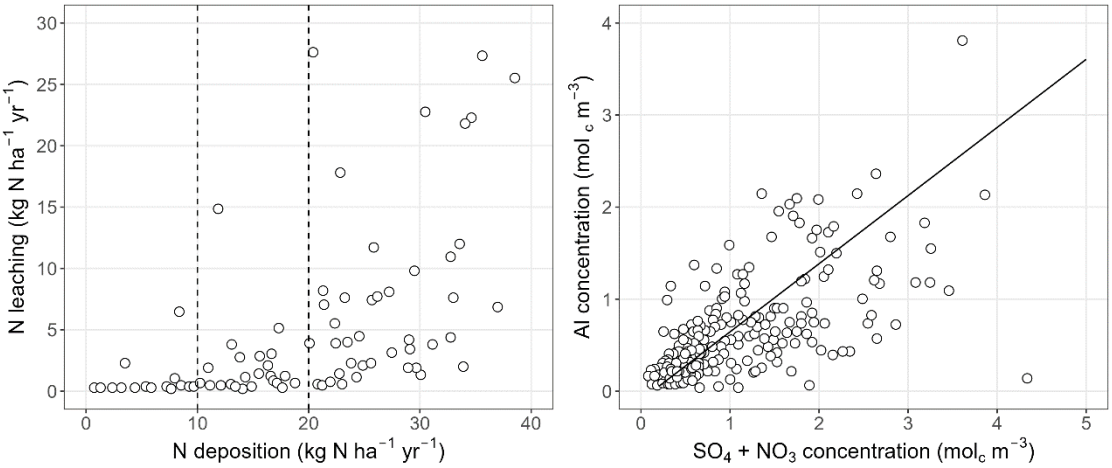
When forest soils start to saturate nitrate concentrations often exceed the 1 mg N l^{-1} during all months over the year (Stoddard, 1994). Median concentrations in more than 500 sites across Europe ranged from 0.35 to 6.7 mg N l^{-1} where the highest concentrations were found in Danish and Dutch forests, sites subject to high N deposition. The European nitrate threshold for drinking water (11.3 mg NO_3-N l^{-1}) was exceeded at 9 to 30% of the sites (Gundersen et al., 2006). The fraction of forest sites per region exceeding the threshold of 1 mg N l^{-1} in seepage water varied between 11 and 93%. The leaching of N is negligible when N deposition is lower than 10 kg N ha^{-1} yr^{-1} (Figure 19, left) being confirmed by monitoring data from Dise et al. (1998a, 1998b) and Gundersen *et al.* (1998a, 1998b). When the N deposition exceeds the 20 kg N ha^{-1}, N leaching increases (Figure 19). The nitrate leaching is often higher in deciduous than in coniferous forests (De Vries et al., 2007). The quality of groundwater and surface water is threatened when the N inputs exceeds the 10 to 15 kg N ha^{-1} yr^{-1} (Table 9).

Table 9. Effects of N deposition on (forest) ecosystem services and related critical N loads (after De Vries et al.,2015).

Ecosystem services	**Examples of nitrogen effects**	**Causal link with nitrogen deposition**	**Crit. N load (**kg N ha^{-1} yr^{-1})
Provisioning services			
Wild plants and animal products	Decline in biodiversity	N-induced eutrophication and soil acidification affect plant and faunal species diversity and thereby the provision of biodiversity-based products.	5–10
Timber/ wood fuel	Increase in wood production	In N-limited forests, N increases forest growth and wood production but in N-saturated forests, N can reduce growth.	20–35
Regulating services			
Climate regulation	Increased carbon (CO_2) sequestration (cooling effect)	In N-limited systems, N deposition increases forest growth and related carbon sequestration, though it can enhance mortality in some species. N deposition can also increase litterfall and reduce decomposition, leading to soil carbon sequestration.	20–35
	N_2O and NO_x emissions (warming effect)	N deposition enhances N_2O and NO_x emissions. NO_x emissions in turn induce O_3 formation. N_2O and O_3 are greenhouse gases, and O_3 is toxic for plants and reduces forest growth and thus carbon sequestration.	5–10
Water quality regulation (water purification)	Decline in N retention and buffer capacity affecting groundwater (drinking water) and surface water quality	N eutrophication and N-induced soil acidification cause a decrease in C:N ratio and base cations/pH, leading to: o Increasing NO_3, Cd and Al concentrations in groundwater and surface water, may exceed drinking water criteria in view of human health o Increasing Al concentrations in acid-sensitive surface waters resulting in reduction or loss of fish (salmonid) populations and reduction of aquatic diversity at several trophic levels (acidification). o Increasing NO_3 concentrations in surface waters causing fish dieback by algal blooms and anoxic zones (eutrophication). Eutrophication is also affected by silica and phosphorus in estuaries.	10–15
Pest/disease regulation	Increase in forest diseases and pests	Elevated N input weakens the resilience of forests and increases infestation rates, such as beech bark disease, in response to e.g. increased foliar N concentrations.	15–20

Soil acidification, induced by nitrate leaching, accelerates the leaching of base cations and the mobilization of toxic Al, compromising tree health (Driscoll et al., 2006; Boudot et al., 1994;

De Vries et al., 2014a; De Wit et al., 2010a, 2010b; Cecchini et al., 2019). Adverse impacts of acidification particularly occur when the pH level declines below 4.5 due to Al dissolution. Higher nitrate and sulphate concentrations in soil solution are strongly correlated to Al



concentrations in acidic forest soils (Figure 19, right), showing that an increase in N deposition enhances the Al release.

Figure 19. Scatter plots of N leaching against total N deposition (left) and of total Al against total SO_4+NO_3 (right) in the subsoil of more than 100 intensive monitoring plots. The solid line in the left plot represents a regression line for sites with a pH < 4.5, reading Al = -0.095 + 0.74 (SO_4+ NO_3) (R^2 = 0.86). Source: De Vries et al. (2003b).

Tree nutrition

Foliar element concentrations and their ratios reflect the nutritional status of trees (Flückiger & Braun, 2003). Unbalanced N:P ratios in foliar tissues are frequently associated with defoliation (Kazda, 1990; Bontemps et al., 2011; Ferretti et al., 2015; Veresoglou et al., 2014; Waldner et al., 2015) and an increasing risk of attacks by parasites (Flückiger & Braun, 1998) and herbivores (Pöyry et al., 2016) as well as decreasing plant capacity to respond to abiotic stressors such as drought, heat, and frost (Fangmeier et al., 1994; Sardans & Peñuelas, 2012). In addition, changes in N:P ratios might alter community composition, species richness and understory vegetation (Peñuelas et al., 2013) and resistance to biotic stressors (Sardans et al., 2016a). High foliar N concentrations might cause nutritional imbalances leading to deficiencies for macronutrients and micronutrients like B, Mn and Mo. Observations from 100 monitoring

plots across Europe (de Vries et al., 2000, 2001, 2003a, 2003b) showed that P deficiencies occurred in 50% of the sites, followed by Mg deficiencies. The relative shortage of P, K, Mg and Ca compared to an increased N content can be aggravated by the acidifying impact of both N and S compounds because the release of toxic Al^{+3} might reduce the availability of base cations, reduce root growth and root uptake (e.g. Sverdrup & Warfvinge, 1993). Considering all base cations, the median N deposition was 10 kg N ha^{-1} yr^{-1} at the plots with a balanced nutrition and 21 kg N ha^{-1} yr^{-1} at the plots with an unbalanced nutrition, suggesting that unbalanced nutrition hardly occur at N deposition levels below 10 kg N ha^{-1} yr^{-1}.

Tree productivity

There are various broad-scale and regional studies investigating the effect of N deposition on tree growth, while accounting for the impacts of other drivers, such as changes in temperature and precipitation (Braun et al., 2017; Kint et al., 2012; Kolář et al., 2015; Solberg et al., 2009). Most forests in N limiting environments show a positive impact of N inputs on wood production and associated carbon sequestration (cooling effect, Townsend et al., 1996) when the N inputs increase up to 20-35 kg N ha^{-1} yr^{-1} (Table 9). Note that N deposition also enhances the N induced growth reduction via ozone (warming effect) when N inputs exceed 5-10 kg N ha^{-1} yr^{-1}. At sustained high levels of N addition, soil N retention decreases due to increased leaching (Aber et al., 1989; Forstner et al., 2019; Templer et al., 2012), evidenced by a decreasing response to N with increasing levels of N application in fertilization experiments (Högberg et al., 2006, 2014; Liu & Du, 2021; Schulte-Uebbing & De Vries, 2018; Tian et al., 2016). The increase in forest carbon per unit N deposited declined with soil N levels (Schulte-Uebbing et al., 2021).

Using stoichiometric scaling, Vries *et al.* (2014b) estimated the averaged forest C–N response for biomass and soil combined at 40 kg C per kg N in boreal forests and 31 kg C per kg N in temperate forests (with 35-45% sequestered in the soil), being the major forest types in Europe. Long-term N addition experiments show biomass C–N responses of 15–25 kg C per kg N (De Vries et al., 2009) and soil C-N responses of 10 kg C per kg N for boreal forests (Maaroufi et

al., 2015) and 36 kg C per kg N for temperate forests (Janssens et al., 2010), where the N-induced increase in C sequestration was lower at higher N deposition rates (Schuelte-Uebing & de Vries, 2018). Lastly, nitrogen deposition gradient studies in Europe showed a C-N response varying between 50-75 kg C per kg N for biomass and soil (Sutton et al., 2008) and a C-N response of 19-26 kg C per kg N for aboveground biomass N (Laubhann et al., 2009, Solberg et al., 2009). A summary of the estimated responses of C sequestration to N deposition (kg C per kg N) in global forest biomes, based on meta-analysis of N addition experiments, stoichiometric scaling and model simulations indicates a range of 15-60 C per kg N in boreal forests and 10-35 kg C per kg N in temperate forests (Du and de Vries, 2023b).

The positive biomass and carbon response to N deposition likely declines with the total N availability (Schulte-Uebbing et al., 2021) where the tree growth declines when the total N inputs exceed the 15 to 20 kg N ha^{-1} yr^{-1} due to the antagonistic effects to tree vitality via soil acidification, nutrient imbalances and increased susceptibility to biotic and abiotic stress (Aber et al., 1998; De Vries et al., 2014a, c; Dobbertin, 2005). Under specific situations these thresholds can be much higher (Schmitz et al., 2018). Productivity in European forests has been observed to decrease at N deposition rates above around 25 kg N ha^{-1} yr^{-1} (Perkins et al., 2000; Flechard et al., 2020). The decline in biomass production at higher N deposition levels also suggests that particularly polluted forest stands (mostly located in Central and Western Europe) might benefit from declining N deposition, as decreases have been strongest in the formerly most polluted regions.

Tree vitality

Tree vitality can be defined as the plant's capacity to grow and reproduce at a specific site (Körner, 2018), as well as its capacity to restore the physiological efficiency after a disturbance (Lloret et al., 2011; Bussotti & Pollastrini, 2021). Defoliation is the most important diagnostic parameter to assess crown condition and tree vitality in forest monitoring programmes (Sanders et al., 2016, 2017a, 2017b). Several fertilizer experiments and observational studies highlighted the relevance of N deposition as a factor potentially modifying the overall physiological status

of trees and their responses to environmental pressures and disturbances over time. Increasing N deposition might enhance the photosynthetic activity and the synthesis of defence compounds, such as non-structural carbohydrates (Huang et al., 2021; Fusaro et al., 2017; Fleischer et al., 2013) but can also increase the susceptibility to insect attacks, pathogens, frost, and storm damages (Bobbink & Hettelingh, 2011; Jönsson et al., 2004; Li et al., 2016; Walter et al., 2021). In addition, N deposition evidently causes changes in mycorrhiza (Arnolds, 1991; Braun et al., 2010; De Witte et al., 2017; Duquesnay et al., 2000; Jaenike, 1991; Kjøller et al., 2012; Van der Linde et al., 2018), changes in the rooting system and aluminum toxicity to roots (Dziedek et al., 2017; Godbold & Kettner, 1991; Haynes, 1982; Jonard et al., 2012; Ostonen et al., 2007), depletes base cations from soil (Jonard et al., 2012; Prietzel et al., 1997), increases the vulnerability to tropospheric ozone (Marzuoli et al., 2018) and drought (Dziedek et al., 2016) as well as deficient nutrient supply other than nitrogen (Jonard et al., 2015; Mellert & Ewald, 2014; Neirynck et al., 1998; Ochoa-Hueso et al., 2013; Peñuelas et al., 2013, 2020; Sardans et al., 2015; Sardans and Peñuelas, 2012; Thelin et al., 1998). The high complexity and interactions among all these mechanisms makes it difficult to underpin the exact effect of (changes in) N deposition on tree vitality. Critical values for N concentrations in foliage related to tree vitality in view of sensitivity to pests and diseases range between 18 and 20 g N kg^{-1} dry weight (Roelofs et al., 1985, 1988; Van Dijk et al. 1992, Flückiger & Braun, 1998) a range often being considered optimal for growth (McNulty et al., 1991). Increasing root biomass and root vitality was reported when trees were protected from N deposition for Scots pine (Boxman et al., 1995), Douglas fir (Murach & Parth, 1999) and Norway spruce (Persson & Ahlström, 2002). Reports on the N effects on frost sensitivity are contradictory, ranging from positive (DeHayes et al., 1989; Klein et al., 1989; L'Hirondelle et al., 1992; L'Hirondelle & Mehl, 1992) over neutral (Hellergren, 1981; Thomas & Ahlers, 1999; Jönsson et al., 2004; Fløistad, 2002) to negative effects (Aronsson, 1980; Jönsson, 2000). Note that site factors including stand age, drought, and frost conditions strongly contribute to the tree vitality independent of the N input (e.g. Eickenscheidt et al., 2016; Klap et al., 2000). For that reason no clear critical loads for N inputs in view of tree vitality could be set, though De Vries et al. (2015) derived preliminary

critical loads varying between 15 and 20 kg N ha^{-1} yr^{-1} to avoid weakening of the resilience against pests and diseases, being a simple proxy for tree vitality (Table 9).

Understory vegetation

Forests provide habitat for understory vegetation, bryophytes, lichens as well as microbial and animal communities. Elevated N inputs often promote nitrophilic species, but can also modify interactions between trees and fungi, the invasibility of exotic species and thereby the understory species composition (Gilliam, 2006; Gilliam et al., 2019). Not only N deposition, but also litter quality, light availability, density of large herbivores, and differences in forest management control the understory plant communities (Bernhardt-Römermann et al., 2015; Perring et al., 2017; Verheyen et al., 2012). Nevertheless, elevated N deposition has clearly contributed to diversity loss in epiphytic lichens in European forests (Bobbink & Hettelingh, 2011; Giordani et al., 2014; Hauck et al., 2013; Mayer et al., 2013). Similarly, major impacts in the community composition and diversity of mycorrhiza were identified at the European level (Suz et al., 2014, 2021; Van der Linde et al., 2018) and in various regional studies (Bobbink & Hettelingh, 2011). Studies detailing the relation between N deposition and animal diversity in Europe's forests are scarce, partly due to the complex dynamics of animal populations and corresponding food-webs (Nijssen et al., 2017). Critical loads in view of biodiversity decline in forest vary mostly from 5 to 15 kg N ha^{-1} yr^{-1} depending on the type of forest (Table 9; Table A1).

5.3.2 Geographical variation and trends of N induced impacts

Spatial patterns of soil solution nitrate under forests are highly variable but partly reflect spatial patterns in N deposition, with higher levels in the Netherlands, Belgium, parts of Germany, Switzerland, Northern Italy and Denmark and lower levels in parts of France, Norway, Northern Sweden, and Finland (Balestrini et al., 2019; Boxman et al., 2008; Braun et al., 2020; De Vries et al., 2007; Evans et al., 2001; Gundersen et al., 1998a; Jonard et al., 2012; Mellert et al., 2004, 2008; Moffat et al., 2002; Pannatier et al., 2010; Pihl Karlsson et al., 2011; Rothe et al., 2002;

Ukonmaanaho et al., 2014; Van der Heijden et al., 2011; Verstraeten et al., 2012). There are relatively fewer reports of elevated nitrate in soil solution in Southern and Eastern Europe, and N deposition is mostly lower in these regions (Waldner et al., 2015).

At the European scale, trends in soil solution N below forests are mostly weak or non-significant. For example, Johnson et al. (2018) found a slight reduction in nitrate concentrations at 40-80 cm depth for 162 plots across Europe between 1995 and 2012. In addition, inorganic N in runoff water declined in 17 plots across Europe between 1990 and 2017 (Forsius et al., 2021). National and regional studies show variable results with obvious reductions in nitrate concentrations in response to decreasing N deposition in Netherlands, Flanders, Northern Italy, and Switzerland (Balestrini et al., 2019; Boxman et al., 2008; Braun et al., 2020; Verstraeten et al., 2012) whereas other sites in Germany and Czech Republic showed no effect of lower N deposition on nitrate concentrations in soil solution (Meesenburg et al., 2016; Oulehle et al., 2011; Švik et al., 2020). Though the leaching of nitrate is generally accompanied by a decrease in base cations and total Al concentrations, there were no clear trends in soil solution pH and acid neutralizing capacity (ANC) found in recent decades (Iost et al., 2012; Johnson et al., 2013, 2018). Despite considerable heterogeneity, Schmitz et al. (2019) reviewed all findings from observations across Europe and concluded that a decreasing trend in soil solution NO_3^- concentrations was found at the European scale. Experimental studies tend to report a faster and more pronounced reaction of soil solution NO_3^- concentrations compared to the findings from large-scale observational studies.

Results from observational studies across Europe for responses in soil, ground vegetation, and trees (nutrition, growth, and vitality) to decreasing N deposition indicate considerable spatial variability in the trends published for these parameters (Schmitz et al., 2019). Several observational studies in European forest showed an increase in nitrophilic forest understory plant species due to elevated N deposition, but those changes were not accompanied by a broad scale decline in plant diversity (Verheyen et al., 2012; Dirnböck et al., 2014; Van Dobben & De Vries, 2017). Similar results were also found for bryophytes in central European forest (Dittrich

et al., 2014). In contrast, elevated N deposition has clearly contributed to the dramatic diversity loss in epiphytic lichens in European forests (Mayer et al. 2009, 2013; Hauck et al. 2013). The degree to which decreasing trends in N deposition contributed to the trends in foliage composition and tree vitality was not clear due to strong variation in confounding site factors also affecting these trends. However, results from observational and experimental studies corroborate the concept of a unimodal response of tree growth to N deposition, with thresholds above which N deposition negatively affects tree growth ranging from 15 - 30 kg N ha^{-1} yr^{-1}. This suggests that forest stands experiencing high N deposition, mostly located in Central and Western Europe, might have benefitted from declining N deposition, as decreases have been strongest in the formerly most polluted regions. Few trends in N deposition have been observed in less polluted areas like Northern Scandinavia, suggesting that a growth decline due to decreased N deposition in these areas is less likely.

5.4 Soil-mediated impacts in terrestrial ecosystems and related critical loads

5.4.1 Impacts on plant species diversity

Impacts of N deposition on plant diversity have been assessed through experimental N addition studies (De Schrijver et al., 2011; Fu & Shen, 2016; Humbert et al., 2016; Soons et al., 2017) and observational studies that investigate plant species diversity across N deposition gradients, either in time series analysis (e.g., Stevens et al., 2010a, 2010b) or over a spatial gradient (e.g., Duprè et al., 2010; Jones et al., 2004). Advantages and disadvantages of both approaches are summarized in Table 10, and discussed in more detail by Du and De Vries (2023a).

Table 10. Advantages and disadvantages of N manipulation experiments, surveys along N-deposition gradients and re-surveys in time.

Approach	Advantages	Disadvantages
Manipulation experiments	Evaluate uniquely the impact of N, can provide cause-effect relationships, can identify thresholds	Assess relatively short-term responses (few exceed 20 years), potential for artefacts (e.g. high N concentrations), systems may already be impacted by N
Spatial field surveys	Can provide insights into longer-term responses, can cover a wide range of N-deposition, avoid experimental artefacts	Cannot prove causality, other drivers on diversity need to be accounted for
Re-surveys over time	Type of evidence that can directly identify changes occurring over long periods of time, without experimental manipulation	Confounding influence of other factors (e.g. land use change, climate, etc), methodology changes, incomplete data records.

The advantage of experimental N addition studies is that effecs can directly be attributed to added nitrogen. However, experimental studies typically assess relatively short-term (several years) responses only, which may cause an underestimate of N deposition-induced biodiversity loss at N addition levels that are in the range of current atmospheric N deposition.

Hettelingh et al.(2015) used N addition experiments to derive dose-response relationships between plant species richness ratio and total N inputs and found significant negative relationships between both (as illustrated in Figure 20 for grassland soils). Results might, however, be influenced by experimental design and local environmental conditions, which limits regional and global extrapolation. The methodological issues can be addressed by setting up globally distributed experiments such as the Nutrient Network (Borer et al., 2014), or by synthesizing multiple N-addition experiments with meta-analysis, allowing to derive a more general quantitative response of plant species diversity to N enrichment (Midolo et al., 2019).

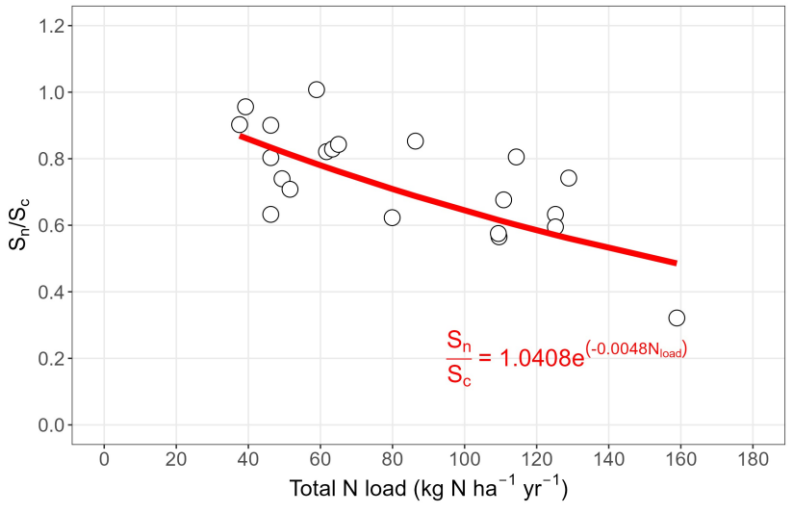


Figure 20. Relationships between the species richness ratio (y=Sn/Sc, where Sn = species number in treatment and Sc=species number in control) and total N load in grassland habitats

(six countries; n = 22; p < 0.001) derived from N addition experiments. Source: Hettelingh et al. (2015).

Observational gradient studies provide insight into longer-term responses, cover a realistic range of N depositions, and avoid experimental artefacts, but need to correct for confounding site factors and cannot prove causality (Dise et al., 2011). In acidic grasslands in Great Britain, Stevens et al. (2004) identified the potential pollutant impacts on plant species richness and found that the diversity declined linearly as a function of the N deposition rate. This study was expanded in 2010 to encompass acid grassland habitats across western Europe (Stevens et al., 2010a, b), and showed the same pattern of species richness decline with increasing N deposition (Figure 21). For the mean N deposition of central Europe (17 kg N ha^{-1} yr^{-1}) this implies a 23% species reduction compared with grasslands receiving the lowest levels of N deposition (5 kg N ha^{-1} yr^{-1}).

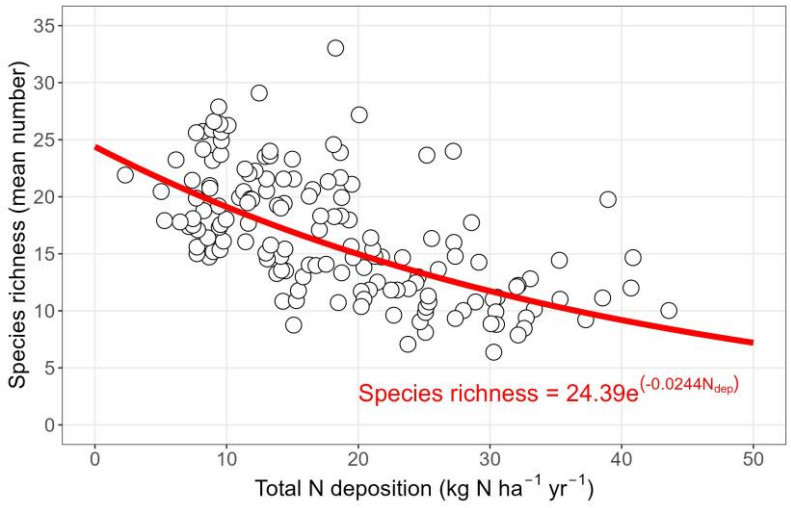


Figure 21. Relationship between species richness of acid grasslands and total inorganic N deposition across western Europe. Adapted from Stevens et al. (2010a).

Several long-term ecological surveillance studies in Europe report a decline in species characteristic for low-nutrient conditions and an increase in nitrophilic plant species over recent decades, including botanical inventories in the UK, Spain and Portugal (Preston et al., 2002) and Great Britain, The Netherlands and Germany (Duprè et al., 2010). The cumulative N deposition was strongly related to a decline in the proportion of vascular plant species (Figure

22). The consistency of results among the various approaches supports the conclusion that high N inputs reduce biodiversity in terrestrial ecosystems (Dise et al., 2011).

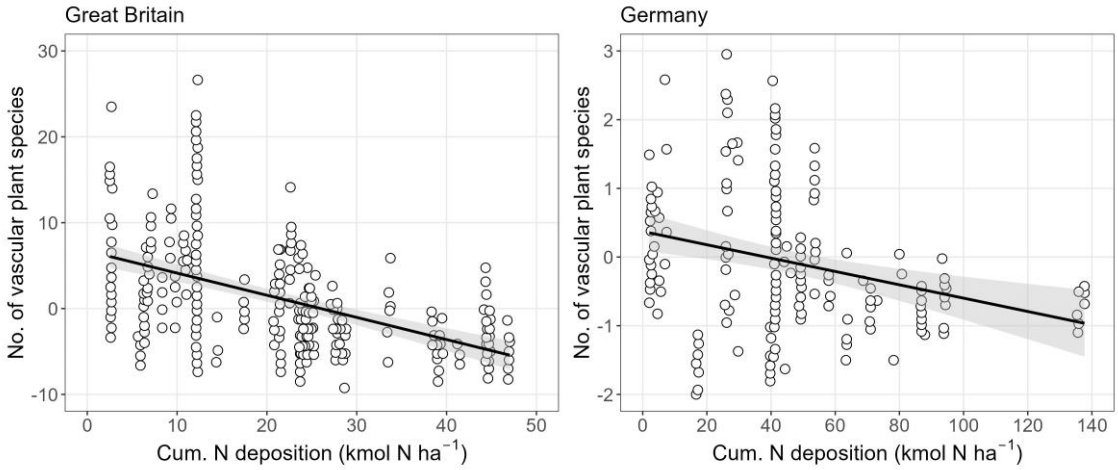


Figure 22. Relationships between vascular plant species richness and cumulative N deposition (in kmol N ha^{-1}) from analyses of plot data from the 1940s onward in Great Britain and Germany. Source: Duprè et al. (2010).

Based on a global synthesis of experimental observed N deposition effects on terrestrial plant diversity, Bobbink et al. (2010) showed that across different ecosystems, N mainly alters species composition by increasing N availability, thereby altering competitive interactions and/or making conditions unfavourable for some species. Other effects, such as ammonium toxicity, soil acidification, and secondary stress are more ecosystem- and site-specific. They also concluded that N deposition is one of the major risks to plant species diversity loss in temperate regions in northern Europe, while it can also be a serious risk in boreal forests, and Mediterranean systems. Similar findings have been reported by others (Tilman, 1987; Aerts & Berendse, 1988; Sala et al., 2000). A recent meta-analysis of N addition experiments also showed that plant species diversity declined with annual N addition rate and experimental duration, in particular at warmers sites and for soils with a lower acid buffer capacity (Midolo et al., 2019).

An increase of N inputs in oligotrophic N-deficient systems enhances the competition abilities of nitrophilic species, thereby reducing the unproductive species adapted to N deficiency. The

associated plant species diversity loss is thus mainly due to indirect soil-mediated effects by N enrichment, accompanied by eutrophication, acidification and increased susceptibility to secondary stress and disturbance factors, operating at different spatial and temporal scales (Bobbink et al., 2010; Dise et al., 2011). The relative impact of these processes depends on the rate and duration of N deposition, the predominant N species, the soil acid neutralizing capacity buffering the N inputs, the soil nutrient availability, and climatic conditions (Dise et al., 2011; Bobbink & Hicks, 2014), as illustrated in Figure 23. Additionally, direct toxic effects of oxidized and reduced N gases and aerosols may also play a role besides the land use history where the latter mainly controls the composition and functioning of plant communities (Bernhardt-Römermann et al., 2015).

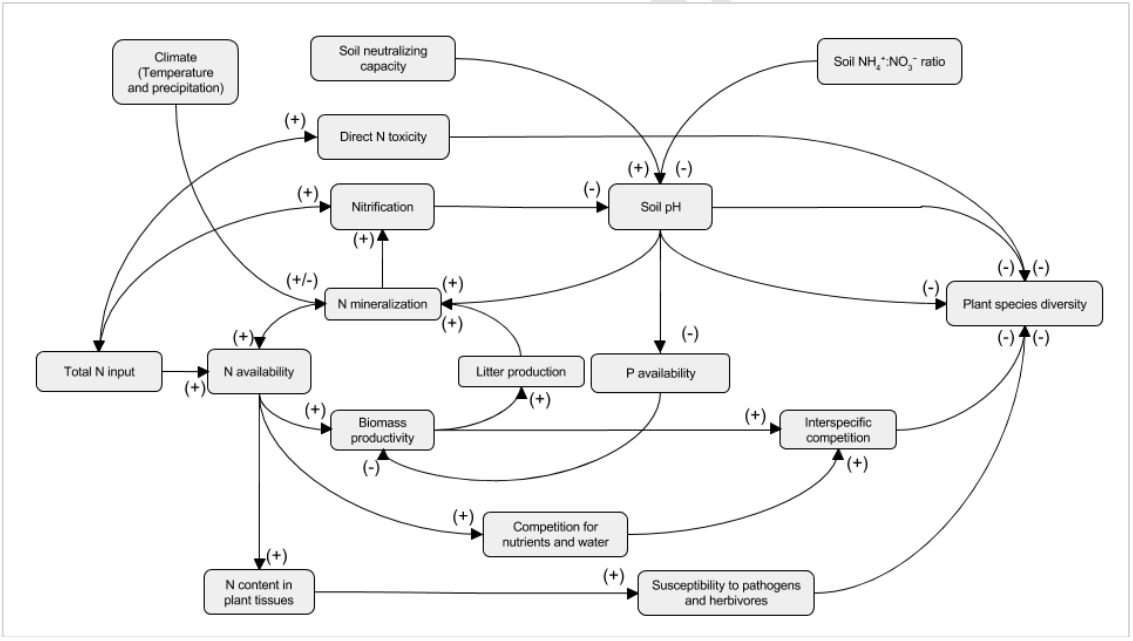


Figure 23. Interactions between main influencing factors and processes that affect plant diversity in response to increased N input (based on Dise et al., 2011).

Data on impacts are mostly limited to Europe (Achermann & Bobbink, 2003; Bobbink et al., 2003; Bobbink & Hettelingh, 2011) and the US (Pardo et al., 2011). Effects of N deposition are now recognised in nearly all oligotrophic nutrient-poor natural ecosystems across Europe; these include grasslands (including tundra and Mediterranean grasslands), heathlands, coastal habitats, oligotrophic wetlands (mire, bog and fen), forests, aquatic habitats and marine habitats. Species and communities most sensitive to chronically elevated N deposition are those that are adapted to low nutrient levels or are poorly buffered against acidification. Grassland, heathland,

peatland, and forest ecosystems are recognised as vulnerable habitats in Europe. Many semi-natural grassland communities are dominated by species with low nutrient requirements, are sensitive to acidification, eutrophication or both, and occur in areas with elevated N deposition. Ecosystems of cold climates, including montane, boreal, tundra, subarctic and arctic habitats, are also vulnerable to N deposition (Bobbink et al., 2010; Dise et al., 2011). Table 11 summarises the effects on plant diversity in these environments and gives examples of key studies that provide evidence of effects. In cases for which there is limited European evidence (i.e. Mediterranean ecosystems), additional evidence from systems in North America is given.

Table 11. Effects of nitrogen deposition on plant biodiversity reported across the major bio-climatic zones in Europe (after Dise et al., 2011)

Habitat	Observed effects on plant biodiversity	Overall weight of evidence in Europe
Grassland	-*Acid grassland*: Reduced species richness, particularly of forbs. -*Calcareous grassland:* Change in species composition. -*Temperate*: Invasion of nitrophilic species; loss of epiphytic lichen species.	Strong for species-rich acid and calcareous grasslands of temperate regions; limited for others.
Forest	-*Boreal*: Decreased cover of ericaceous shrubs; decline of characteristic bryophytes.	Strong for boreal and temperate forests; limited for other forests.
Peatland	-Decline of characteristic bryophyte species. -Loss of sundew.	Strong, with a range of studies.
Heathland	-Loss of characteristic lichen species. -Invasion of nitrophilic acid grassland species. -Reduced species richness, particularly of bryophytes.	Strong for temperate dry heaths, limited for others.
Coastal dune	-Reduction in species diversity. -Increased grass growth. -Loss of lichen species.	Limited to a small number of studies.
Mediterranean ecosystems	-*Forest*: loss of sensitive lichen species. -*Grassland and shrub*: loss of native forb species. -Reduced cover of lichens.	Very limited.
Tundra	-Increased cover of vascular plants. -Changes in bryophyte species composition.	Limited to a small number of studies.
Arctic and alpine	-*Grasslands*: Increased cover of sedges, reduced cover of grasses and forbs. -*Heaths*: Loss of characteristic lichen species.	Limited to a small number of studies.

Major impacts of N deposition on the community composition and diversity of mycorrhiza were found at European level (Suz et al., 2014; Van der Linde et al., 2018) and in various regional studies (Bobbink & Hettelingh, 2011) with potential cascading effects from soil microbes to plants (Farrer & Suding, 2016). More details on trends in plant species diversity in response to changes in N deposition are given in Schmitz et al. (2019). In addition, there are indications that increased N availability in response to N deposition can lead to a decrease in the growth of peat mosses, such as *Sphagnum*, due to P limitation and even toxicity at very high N levels (Limpens & Berendse 2003; Granath et al., 2012). A Europe-wide study indicates that N deposition

between 1950 and 2000 depressed production between 0 and 25% relative to 1900, particularly in temperate regions (Granath et al., 2014).

5.4.2 Impacts on faunal species diversity

Diversity effects of N deposition on plants can also cascade from plants to animal species, including insects, butterflies, birds and mammals (Nijssen et al., 2017). Research on fauna is complicated, as different species use the landscape at different spatial scales and animal species outnumber plant species by about 25 to 1. Consequently, research on the effects of N deposition on fauna is largely lacking. There is, however, a clear impact by affecting food and environmental conditions. Increased N deposition foremost causes changes in plant growth, the nutrient content and also plant species composition and thereby the micro-climate (temperature and moisture regimes) experienced by animals. As an example, a change in the occurrence and quality of host plants by N deposition affects the population size of herbivorous animals like caterpillars (Bink, 1992; Soontiëns & Bink 1997; Kerslake et al., 1998). Changes in nutrient composition of soil organic matter affects the fate of detritivores, as e.g. Vos et al. (2000, 2002) showed for aquatic invertebrates. In addition to the direct impact on food quality and micro-climate there is also an indirect impact of N inputs on landscape configuration and heterogeneity. Increasing the N inputs reduce the variation in biomass, species and associated agricultural management across the landscape, leading to a reduction in animal diversity by at least three mechanisms. First, species may depend on specific conditions, which are only present in transitions between different biotopes. Second, many animal species require different parts (biotopes) of the landscape for reproduction, resting, foraging, etc. Third, heterogeneity creates the possibility of risk spreading, leading to a higher persistence of species (Verberk et al., 2002; Verberk & Esselink, 2004).

For example, Mulder *et al.* (2005) showed that butterfly abundance decreased steadily over the period 1994-1999 but they could not relate that to a negative impact of acidification of rainwater or climate change. Nevertheless, the butterfly decline was identified as a secondary effect of heavy metal stress on local plants, which in turn can be related to acidification, since pH largely

affects the bioavailability of heavy metals (e.g. Lofts et al., 2004). An interpretation that ascribes the biodiversity and density of the butterfly community to secondary effects of N related pollutants on plants is further supported by the well-known negative correlation between pollen density and the concentration values of NO_2, SO_2 or O_3 (e.g. Bessonova, 1992; Black et al., 2000). Similarly, Nijssen et al. (2001) could explain differences in ground beetle populations by the N-induced changes in microclimate via encroachment with the grasses *Calamagrostis epigejos* and, to a lesser extent, *Ammophila arenaria*, for 15 coastal dune grasslands in the Netherlands. In addition, the observed decline of the red-backed shrike (*Lanius collurio*) illustrates how the effects of increased N deposition have repercussions across the entire food web (Beusink et al., 2003). This bird species strongly declined from 1900 onwards throughout Western Europe. To ensure a constant and sufficient energy supply, red-backed shrikes require a high diversity of large prey species, which in turn depends on landscape heterogeneity (Esselink et al., 1994). In Dutch coastal dunes, increased N deposition led to the encroachment by tall grasses and bushes, a decrease of open sandy areas and a loss of succession stages rich in species. The decline in landscape heterogeneity seriously affected the prey availability for red-backed shrikes (Beusink et al., 2003).

5.4.3 Critical loads of nitrogen for plant and faunal diversity

In order to set standards and targets for emission reduction policies the concept of 'critical load' has been developed since the mid-1980s. A widely accepted definition of a critical load is 'a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge' (Nilsson & Grennfelt, 1988). Critical loads have been defined for specific combinations of pollutants, effects and receptors (Nilsson & Grennfelt, 1988; Bobbink et al., 1996, Achermann & Bobbink, 2003). Since 1994, critical loads of N and acidity have played an important role in European air pollution abatement (Hettelingh et al. 2001, 2007, 2017; Spranger et al., 2008; De Vries et al., 2015).

Eutrophication

Critical loads of N in view of eutrophication and associated biodiversity effects are estimated empirically or by modelling. The empirical approach uses mainly *manipulation experiments* in which N is added in experimental fields or 'mesocosms', while results of *spatial field surveys* or *re-surveys over time* are used as additional evidence. An overview of empirical N critical loads related to impacts on plant species diversity, including impacts on mosses, lichens and mycorrhizae, in (semi-) natural terrestrial ecosystems (coastal habitats, wetlands, grasslands, and heathlands/ tundra) is presented in Table 12, using the classes of the EUropean Nature Information System (EUNIS). Results for marine habitats, inland surface waters, and forests are given in Table A1

Table 12. Selection of empirical critical loads of N (kg N ha^{-1} yr^{-1}) for natural and semi-natural terrestrial ecosystems as revised in 2010 plus a description of effects that can occur when critical loads are exceeded (after Bobbink & Hettelingh, 2011).The reliability is qualitatively indicated by ## reliable; # quite reliable and (#) expert judgement. Changes with respect to values derived in 2003 (Bobbink et al., 2003) are indicated in bold.

Ecosystem type	Critical load kg N ha^{-1} yr^{-1}	Reliability	Indication of exceedance
Coastal habitats (B)			
Shifting coastal dunes	10-20	(#)	Biomass increase, increased N leaching
Coastal stable dune grasslands (grey dunes)	**8-15**	#	Increase in tall graminoids, decrease in prostrate plants, increased N leaching, soil acidification, loss of typical lichen species
Coastal dune heaths	10-20	(#)	Increase in plant production, increased N leaching, accelerated succession
Moist to wet dune slacks	**10-20**	(#)	Increased biomass tall graminoids
Mires, bogs and fens (D)			
Raised and blanket bogs	**5-10**	##	Increase in vascular plants, altered growth and species composition of bryophytes, increased N in peat and peat water
Valley mires, poor fens and transition mires	**10-15**	#	Increase in sedges and vascular plants, negative effects on bryophytes
Rich fens	**15-30**	(#)	Increase in tall graminoids, decrease in bryophytes
Montane rich fens	**15-25**	(#)	Increase in vascular plants, decrease in bryophytes
Grasslands and lands dominated by forbs, mosses or lichens (E)			
Sub-Atlantic semi-dry calcareous grasslands	15-25	##	Increase in tall grasses, decline in diversity, increased mineralisation, N leaching, surface acidification
Mediterranean xeric grasslands	**15-25**	(#)	Increased production, dominance by graminoids
Non-Mediterranean acidic and neutral grasslands	**10-15**	##	Increase in graminoids, decline in typical species, decrease in total species richness
Inland dune pioneer grasslands	**8-15**	(#)	Decrease in lichens, increase in biomass
Inland dune siliceous grasslands	**8-15**	(#)	Decrease in lichens, increase in biomass, increased succession
Low- and medium-altitude hay meadows	20-30	(#)	Increase in tall grasses, decrease in diversity
Mountain hay meadows	10-20	(#)	Increase in nitrophilous graminoids, changes in diversity
Moist and wet oligotrophic grasslands			
• *Molinia caerulea* meadows	15-25	(#)	Increase in tall graminoids, decreased diversity, decrease in bryophytes

• Heath (*Juncus*) meadows and humid (*Nardus stricta*) swards	10-20	#	Increase in tall graminoids, decreased diversity, decrease in bryophytes
Moss- and lichen-dominated mountain summits	5-10	#	Effects on bryophytes or lichens
Alpine and subalpine acidic grasslands	**5-10**	#	Changes in species composition; increase in plant production
Alpine and subalpine calcareous grasslands	**5-10**	#	Changes in species composition; increase in plant production
Heathland, scrub and tundra (F)			
Tundra	**3-5**	#	Changes in biomass and in species composition in bryophyte layer, decrease in lichens
Arctic, alpine and subalpine scrub habitats	5-15	#	Decline in lichens, bryophytes and evergreen shrubs
Northern wet heath			
• 'Calluna-dominated wet heath (Upland moorland)	10-20	#	Decreased heather dominance, decline in lichens and mosses, increased N leaching
• Erica-dominated wet heath (Lowland)	**10-20**	(#)	Transition from heather to grass dominance
Dry heaths	10-20	##	Transition from heather to grass, decline in lichens, changes in plant biochemistry, increased sensitivity to abiotic stress
Mediterranean scrub	**20-30**	(#)	Change in plant species richness and community composition

Field addition experiments and mesocosm studies with independent N treatments and realistic N loads and durations (below 100 kg N ha^{-1} yr^{-1} for more than one year) were used for updating and refining critical load values in 2011 (Bobbink & Hettelingh, 2011). Only in cases where no appropriate N-addition studies were available, gradient and retrospective studies were given a higher weight. The ranges of critical loads can be made more specific in relation to a number of modifying factors, but it is very difficult to quantify such modifying factors on regional scales, and consequently it is recommended to use the minimum value of the ranges of empirical critical loads to assess exceedances of critical loads of N (Bobbink & Hettelingh, 2011). Note that critical N loads near 10–20 kg N ha^{-1} yr^{-1} in view of impacts on ectomycorrhizal fungi in forests have been updated to a value around 5–6 kg N ha^{-1} yr^{-1}, based on a substantial ectomycorrhizal shift at higher N inputs, as derived in a large-scale N-deposition gradient study over European forests (Van der Linde et al., 2018).

Note that the use of empirically derived critical loads can be criticized based on data selection, impacts of site conditions and methodological issues like experimental duration and background-N inputs, and the lack of fixed criteria for which a critical load is derived. In view of these disadvantages, model-based critical N loads for eutrophication are also derived, based on a mass balance, which balances the N deposition to an ecosystem with its long-term capacity

to buffer this input without harmful effects (Hettelingh et al., 2001, Spranger et al., 2008, Reis et al., 2012). The harmful effects are defined in terms of critical limits above which a negative effect is assumed to occur. An overview of those limits is given in De Vries et al. (2007). Details on the calculation of critical N loads are given in Posch *et al.* (2015).

Acidification

The (excess) input of N (and sulphur) to ecosystems also contributes to the acidification of soils and surface waters. There are no common empirical critical loads for acidity. Instead, a model-based approach is used, which balance the acid load to an ecosystem with its long-term capacity to buffer this input without harmful effects (Hettelingh et al., 2001; Spranger et al., 2008). Critical loads of acidifying N and S have always been modelled using steady-state charge and mass balances and a chemical criterion that reflects what is to be protected by the deposition at (or below) the critical load (e.g. fine roots in forest soils, fish in surface waters, etc.). While for eutrophication a single critical load number is computed ($CL_{eut}N$), for acidification a critical load function (CLF) is obtained that describes all pairs of N_{dep} and S_{dep}, for which the chemical criterion is not violated. A comprehensive description of the methods to model critical loads of eutrophication and acidity can be found in Posch et al. (2015).

5.5 Geographic variation and trends in exceedances of nitrogen critical loads

The average deposition of inorganic N across all land-use types in Europe decreased from 10.3 kg N ha^{-1} yr^{-1} in 1990 to 6.6 kg N ha^{-1} yr^{-1} in 2018 (after Engardt et al., 2017). The trends are distributed heterogeneously in space. While many forests in areas with higher absolute levels of N deposition (e.g. in Central and Western Europe) experienced a decrease in N inputs, less clear trends have been reported for Northern Scandinavia and parts of Southern Europe.

If a deposition is greater than a given critical load (CL), which is a time-independent ecosystem-specific quantity, that CL is said to be exceeded. For a CL that is given by a single number, such as the CL for eutrophication, this is straight-forward; but for two pollutants, such as N and S in the case of acidity, the exceedance is characterised by the (shortest) distance of the point (N_{dep}, S_{dep}) from the critical load function (in the N_{dep}-S_{dep} plane) as described by Posch et al. (2001).

In order to provide a single exceedance number for a grid cell or country/region, the so-called accumulated average exceedance (AAE) is computed, which is the area-weighted average of all exceedances within that region (Posch et al., 2015). Under the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP), critical loads for eutrophication and acidification have been compiled for Europe since 1990 and used in the negotiations of effects-oriented emission reduction agreements in Europe (see, e.g. Sliggers & Kakebeeke 2004; Reis et al. 2012; Hettelingh et al., 2013). Here we use the European critical loads database from Hettelingh et al. (2017) to evaluate their exceedances over time.

Trends in average accumulated exceedance over Europe of critical loads of eutrophication due to N deposition for the period 1990-2019 are declining from 1990 onwards, especially in North and Central Europe (Figure 24, top).

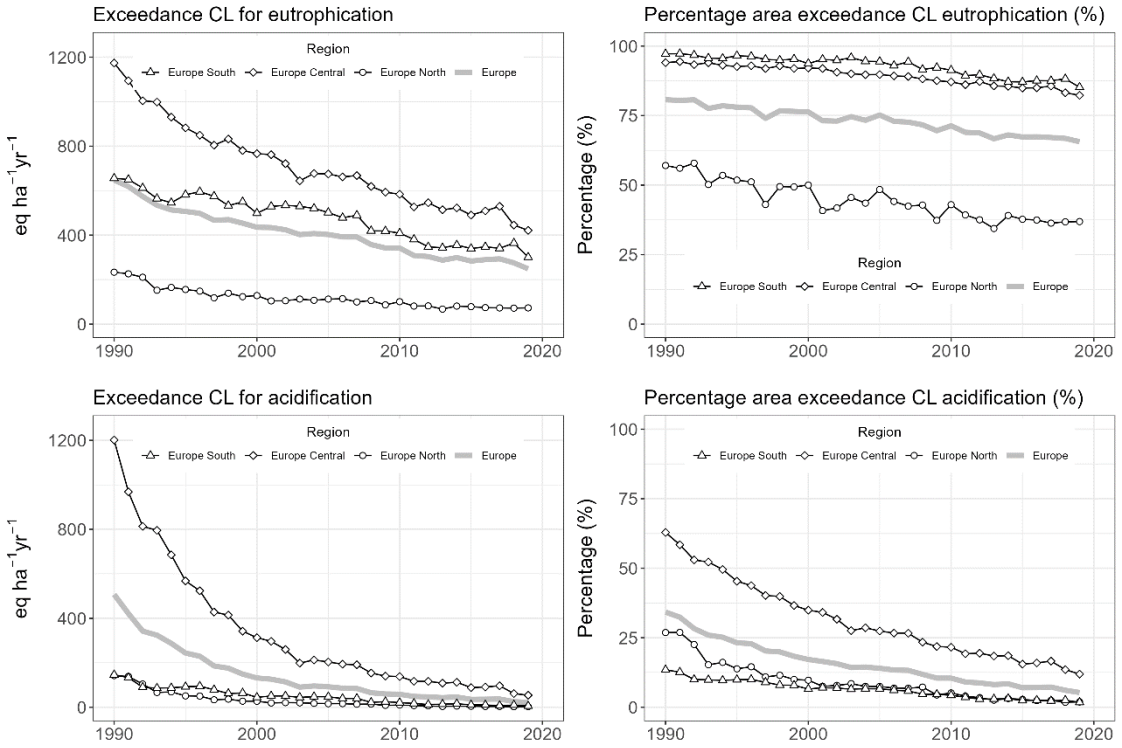


Figure 24. The 1990–2019 temporal development of the modelled average accumulated exceedance (left) and the percentage of terrestrial ecosystem area exceeded (right) of the critical loads of eutrophication ($CL_{eut}N$, top) and acidification (bottom) in the three regions of Europe west of 40°E and overall European averages (thick grey lines).

Overall the exceedance declined on average by ca 60% from ca 650 to 300 eq ha^{-1} yr^{-1} (Figure 24 top left) while the area exceeding critical loads reduced only ca 10%, from ca 80 in 1990 to 65% in 2019 (Figure 24 top right). The results are in line with Slootweg et al. (2015), who showed that despite N deposition reductions, 62% of the European ecosystem area exceeded the critical N loads for eutrophication in 2015. This decline mainly originates from the large reduction in NO_x emissions. Much stronger reductions were found for the accumulated average exceedance for acidification due to N and S deposition, being higher than 80% (Figure 24, bottom). This decline mainly originates from the large reduction in SO_2 emissions, in addition to NO_x emissions.

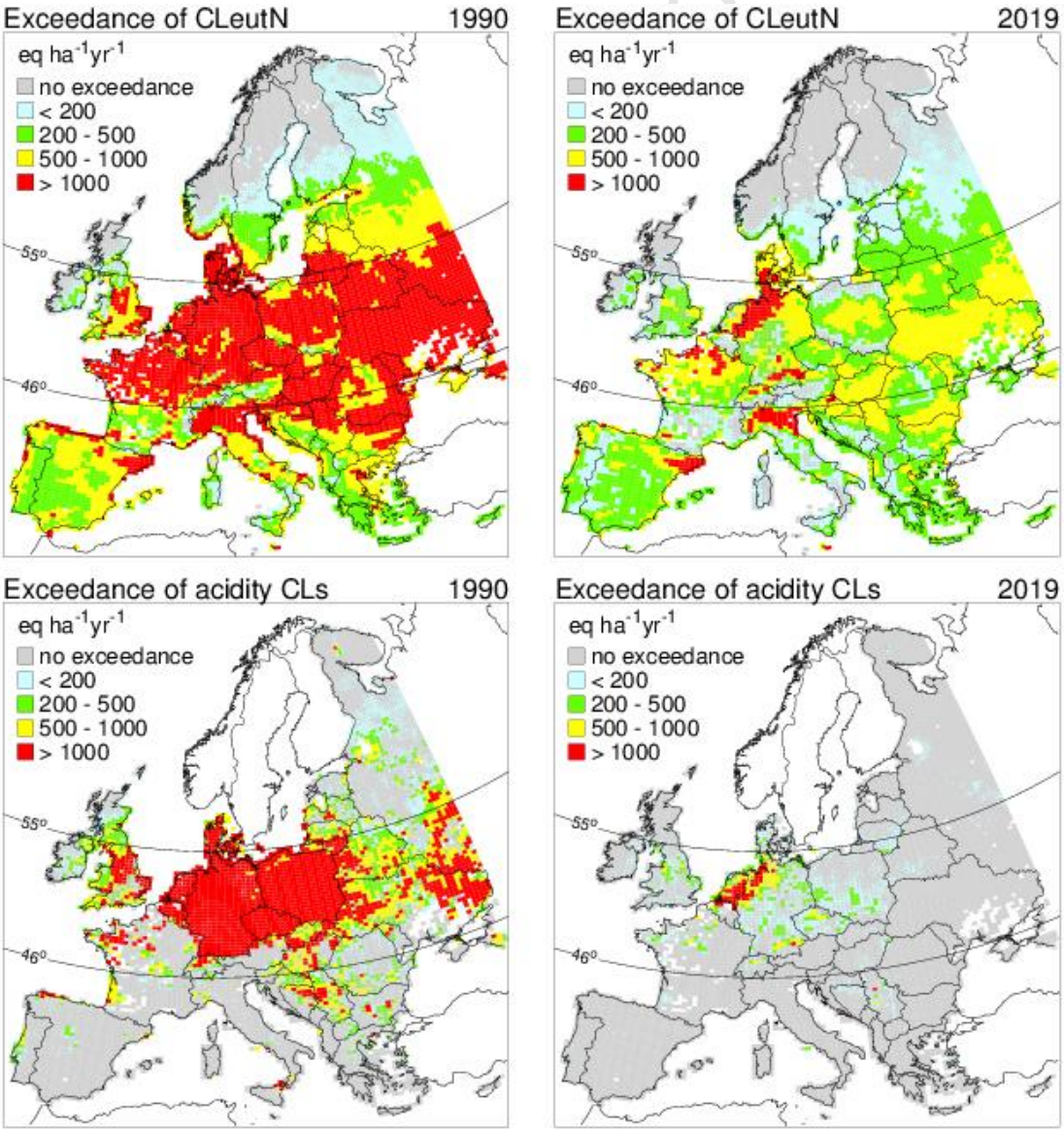


Figure 25. Geographic variation in modelled average accumulated exceedance of the critical loads of eutrophication ($CL_{eut}N$, top) and acidification (CLs, bottom) for terrestrial ecosystems in 1990 and 2019. The two parallels separate the three regions 'North', 'Central' and 'South'.

The geographic variation in the AAE of critical loads of N in view of eutrophication for 1990 and 2019 show that exceedances still occur almost everywhere in Europe (Figure 25), but that they have declined considerably in magnitude, leaving only a few areas with high exceedances, such as the Po-valley in Italy and the Dutch-German-Danish border regions. For acidity, there are hardly any exceedances left except for the Netherlands (Figure 25).

6 Impacts on aquatic ecosystems

6.1 Relationships between elevated nitrogen inputs and ecosystem impacts

European water policies aim to attain good ecological status (defined as no more than a slight deviation from near-natural conditions) in all rivers, lakes, coastal and transitional waters by 2027 the latest (EC, 2000). Currently 57% of rivers, 44% of lakes, 40% of coastal waters and 66% of transitional waters fail to achieve this (Poikane et al., 2019), caused by various factors from which nutrient enrichment remains the main reason (EEA, 2018; Grizzetti et al., 2017). Aside from eutrophication, chemicals with endocrine-disrupting properties, pesticides and metals have also adverse impacts on aquatic life, threatening the aquatic biodiversity in freshwater and marine ecosystems. The sources of freshwater pollution are extremely diverse and vary considerably with geographical location given the fact that the extent, speed, and pathways by which pollutants are transported from agricultural land to freshwater bodies are diverse (Goulding et al., 2000).

Eutrophication

The enrichment of nitrogen in freshwaters is due to: (i) runoff from diffuse sources within the catchment, such as deposition, fertiliser, and manure application, (ii) direct input from point sources, such as sewage treatment works, and (iii) atmospheric deposition onto the water body. Elevated N in surface waters and coastal/marine waters contribute to eutrophication and affects

phytoplankton and benthic algae, zooplankton, water plant communities (macrophytes) and fish of freshwater ecosystems and coastal/ marine ecosystems. Specifically, in marine ecosystems, where N is considered to be the most important element limiting phytoplankton growth, effects can be considerable and include many negative effects (see Carpenter et al., 1998; Smith, 2003; Smith & Schindler, 2009; Pinay et al., 2017), such as: i) changes in species abundance, composition and diversity, ii) loss of subaquatic vegetation as excessive phytoplankton and algal growth reduce light penetration, iii) decline in coral reef health and associated communities, as increased nutrient levels favour algae growth over coral larvae, iv) harmful algal blooms which produce toxic effects on aquatic organisms, and v) formation of hypoxic or oxygen-depleted waters potentially leading to ecosystem collapse.

The severity of eutrophication of freshwaters by N largely depends on the characteristics of the aquatic system, including climate, morphology, water residence time, nutrient ratios, trophic web status, and ecosystem resilience (Durand et al., 2011; Grizzetti et al., 2011). Therefore, similar nutrient loads may produce different effects among catchments. Similarly, the impacts are related not only to N loads, but rather to its specific synergies with the availability of other elements, such as phosphorus, carbon, and silica (Billen & Garnier, 2007). Scientists use the concept of "limiting element", which is the element that limits the potential rate of primary production (Howarth, 1988). Nevertheless, N (and P) concentrations in surface waters, being a major driving force of the problems, can be used as proxy to evaluate the risk for water eutrophication (Phillips et al., 2008).

Acidification

Acid deposition modifies the transport fluxes of cations and anions from soil to water, changes the biogeochemical cycles of elements in the water system, and even kills living species susceptible to low pH. The intensity of aquatic acidification is controlled by (i) the acid deposition on the catchment, with regard to the exposure time, and (ii) the natural susceptibility and vulnerability of the catchment to acidification given geological, geographical, and climatic conditions. For example, if most of the soils in a catchment are acidic, the waters acidify more

quickly due to leaching of hydrogen and aluminium than in catchments with soils having a higher buffer capacity.

The effects of acidification on freshwater ecosystems have been thoroughly studied and documented, in particular since the 1970s (Hultberg & Stenson, 1970; Beamish & Harvey 1972; Jensen & Snekvik, 1972; Beamish, 1974; Leivestad & Muniz, 1976; Austnes et al., 2018). Evidence for large scale die-back of fish populations in the Nordic countries became evident in the 1980s (Hesthagen et al., 1999). Severe impacts, on fish as well as other aquatic organisms were reported for Sweden, Finland, the UK, Canada, and the USA (Tammi et al., 2003; Monteith et al., 2005; Gray et al., 2012). Water acidity is often found to exert a tight control on the health and species composition of biological communities ranging from primary producers, e.g. algae (e.g. Battarbee and Charles, 1986), to top predators, including fish (Baker & Schofield, 1982), and there is widespread evidence that acidification has led to major losses in diversity and shifts in species composition (e.g. Stoner et al., 1984; Hesthagen & Hansen, 1991; Hermann et al., 1993; Herrmann & Svensson, 1995; Watt et al., 1983, 2000; Fjellheim & Raddum, 2001; Lacoul et al., 2011). Several physiological effects are related to toxic effects of H and Al (e.g. Rosseland & Staurnes, 1994; Gensemer & Playle, 1999). Both low pH (Schofield, 1976) and increased Al levels (Baker & Schofield, 1982) are directly toxic to fish, causing mortality of adult fish and recruitment failure (Hesthagen et al., 2011), and negative effects on the gills that regulate the ion exchange and respiration rates (Exley et al., 1991). In addition, low pH and increased Al cause chronic stress that may lead to a lower body weight and smaller size, either as a chronic effect on the fish or as indirect effects of changes in food and habitat (Lien et al., 1996). The toxic effects are reduced at higher calcium concentration and in the presence of complexing agents such as dissolved organic matter (Howells et al., 1983; Neville, 1985). There is evidence that sensitive populations increase in size in chemically recovering waters (Monteith et al., 2005), though the pre-acidification state is difficult to regain (Battarbee et al., 2014).

6.2 Impacts on freshwater ecosystems

Ecological impacts from freshwater eutrophication are initialized by the increase in primary productivity resulting from enhanced nutrient uptake by autotrophs, thereby prompting the increase in water turbidity, odour, and, subsequently, the decomposition of organic matter, water temperatures, and the depletion of dissolved oxygen. The latter is particularly detrimental to heterotrophic species. The depletion of sunlight caused by increased water turbidity also enhances competition for light by photosynthesizing organisms, which, in some cases, may lead to the synthesis of toxic substances (allelochemicals) by competing phytoplankton (Carpenter et al., 1998; Leflaive & ten Hage, 2007). Ultimately, they may prompt losses in biodiversity, e.g., decline in genera richness (Struijs et al., 2011).

The common paradigm is that phosphorus levels in freshwaters instigate primary production more than those of nitrogen (Schindler et al., 2008; Schindler, 2012). However, the accumulation of "legacy" P in lakes over decades of nutrient enrichment have moved lakes towards N limitation (Paerl et al. 2020; Maberly et al., 2020). N limitation is also common in headwater streams draining natural and semi-natural landscapes (e.g. Burrows et al., 2015; Wurtsbaugh et al., 2019). Furthermore, controlling only P inputs to freshwaters and ignoring the large anthropogenic inputs of N can reduce algal uptake of N and thus allow more N to be transported downstream where it can exacerbate eutrophication problems in estuarine and coastal marine ecosystems (Conley et al., 2002, 2009). It is thus in most cases essential, to implement a dual-nutrient-reduction strategy when developing measures to control eutrophication in freshwater ecosystems

6.2.1 Eutrophication and critical limits

Nitrogen impacts on eutrophication

N and P exports can have profound effects upon the quality of receiving waters (Carpenter et al., 1998; Correll, 1998). The degradation of water resources by eutrophication can result in losses of their component species, as well as losses of the amenities or services that these systems provide (Carpenter et al., 1998, Postel & Carpenter, 1997). These effects of nitrogen were only

considered recently (Stoddard, 1994) since the dominant view since the 1970s was that the primary productivity in aquatic ecosystems was controlled by phosphorus (Schindler 1971, 1977, Schindler & Fee, 1974; Hecky & Kilham, 1988; Vitousek et al., 1997) and controlled by numerous physical, chemical, and biotic factors such as light availability, hydraulic flow regime and herbivore grazing (Smith et al., 1999). Though this dominant view was questioned in the 1990s (Kratzer & Brezonik, 1981; Elser et al., 1990) and challenged in some reviews (Elser et al., 2007; Lewis & Wurtsbaugh, 2008; Sterner, 2008), it was finally updated after various stream enrichment studies confirmed the role of N in eutrophication of freshwaters (Smith et al., 1999).

Impacts of N on the biology are particularly visible in nutrient-poor aquatic ecosystems, often found in in arctic, boreal, and temperate regions (see De Wit & Lindholm, 2010). Effects of N on freshwater systems were documented by various types of studies: historical records of lake sediments (paleolimnology), lake surveys and experimental studies focusing on algae, water plants (macrophytes) and invertebrates (insects, snails). First, the paleolimnological studies cover a wide range in geographical and climate conditions and showed that N enrichment was able to change the phytoplankton community structure in oligotrophic lakes (Baron et al., 2000; Wolfe et al., 2001) or interactions of climate and N deposition (Saros et al., 2003; Wolfe et al., 2006; Holmgren et al., 2009; Pla et al., 2009; Hobbs et al., 2010). There are also shifts in paleolimnological records that are attributed to climate only (Birks et al., 2004; Ruhland et al. 2008). The evidence presented supports the hypothesis that N deposition increases algal productivity and can change algal community structure at N deposition rates of 1 to 3 kg N ha^{-1} yr^{-1}, but especially in lakes receiving more than 10 kg N ha^{-1} yr^{-1}. Second, lake surveys and experimental studies resulted in conflicting outcomes in view of N limitations (Schindler, 1977; Fee, 1979; Schindler et al., 2008; Scott & McCarthy, 2010) though the productivity of both phytoplankton and sediment-dwelling algae increased after addition of N (Axler & Reuter, 1996; Jansson et al., 2001). Lake surveys along various gradients (deposition, climate, vegetation cover) are also used to statistically relate water chemistry to changes in algal growth.

Regional surveys in boreal lakes show higher chlorophyll concentrations per unit P in areas with higher N deposition, indicative of higher primary production (Bergström et al., 2005, 2008; Bergström & Jansson 2006). Experimental nutrient additions in lakes via mesocosm studies and bioassays support this finding by showing that co-limitation (N and P) or N limitation of algal growth is common, especially when N availability is low (Morris & Lewis, 1988; Nydick et al., 2003, 2004; Lafrancois et al., 2004; Saros et al., 2005; Andersson & Brunberg, 2006). It has even been suggested that N deposition may have shifted primary productivity in oligotrophic lakes from N limitation to P limitation (Bergström & Jansson, 2006; Elser et al., 2009). Critical N loads in view of impacts on surface water vary mostly from 5 to 20 kg N ha^{-1} yr^{-1} depending on the type of surface water (Table A1).

Critical limits for nitrogen concentration in surface water

In Europe, the Water Framework Directive (WFD; EC, 2000) was adopted to protect and enhance the status of aquatic ecosystems. Under the WFD, ecological status is assessed through biological quality elements (phytoplankton, benthic flora, benthic invertebrate and fish fauna) together with supporting hydromorphology and physicochemical parameters, including nutrient conditions. The WFD stipulates that, at good ecological status, nutrient concentrations must "not exceed the levels established so as to ensure the functioning of the ecosystem and the achievement of values specified (for good status) for the biological quality elements". Thus, the WFD does not provide nutrient concentration targets but requires the derivation of nutrient criteria ensuring good ecological status. While a huge effort has been devoted to the development of the assessment methods using biological quality elements, much less attention has been paid to setting nutrient criteria (Poikane et al., 2019).

The type-specific nutrient criteria also imply that no general standards for N concentrations exist, since the effects of N depends on P availability and other catchment properties. Limited P availability in European lakes is mainly found in northern latitudes, low population density areas, or mountainous regions, while in Western and Central Europe most lakes are relatively rich in phosphorus, and an intermediate level is found in most lakes in the Baltic States, Poland,

and Spain (EEA, 1999). Furthermore, if P loads increase, N can become growth-limiting. Nitrogen often limits phytoplankton growth in lakes in which water plants are abundant and the species diversity of water plants is also largely determined by the availability of N.

Camargo & Alonso (2006) performed an extensive study on the ecological and toxicological effects of inorganic N in aquatic ecosystems. They showed that total N levels lower than 0.5-1.0 mg N l^{-1} are needed to prevent aquatic ecosystems from developing eutrophication and/or acidification. The proposed limit however does not apply for ecosystems naturally rich in N or clearly limited by P availability. For the Netherlands, Van Liere & Jonkers (2002) derived critical nutrient concentrations for small streams, ditches, lakes, rivers, and coastal waters, and showed that they varied from 0.3 to 2.0 mg N l^{-1}. Poikane et al. (2019) reviewed all critical limits for N and P used by the EU Member States to support good ecological status. They found median critical values for total N ranging from 0.48 to 1.6 mg N l^{-1} for lakes in correspondence to literature linking N concentrations to the desired aquatic ecology (Phillips et al., 2018; Poikane et al., 2019). For river types, the critical median N values that should not be exceeded without adverse impacts on aquatic biodiversity ranged between 1.5 and 2.8 mg N l^{-1} where literature data varied from 1.1 to 3.5 mg N l^{-1} (Phillips et al., 2018). For coastal and transitional waters the critical values ranged between 1 and 1.2 mg N l^{-1} (HELCOM, 2015, 2017; Salas Herrero et al., 2019). In general, critical N thresholds seem to range between 0.5 and 2.8 mg N l^{-1}.

6.2.2 Acidification and critical limits

Historically, acidification of surface waters was one of the major environmental impacts of air pollution leading up to the signing of the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP) in 1979. The resulting reductions in N and particularly S deposition resulted in a gradual decline in degree and extent of surface water acidification. However, there is a time lag in chemical and, particularly, biological recovery. Acidification occurs when the buffering capacity of the catchments is insufficient to balance the level of acid deposition. Effects of acidification are therefore usually confined to lakes and streams in areas with acidic

bedrock (Brönmark & Hansson, 2002) and to moorland pools on sandy soils (Roelofs et al., 1996). However, also in lakes with more calcareous sediments, such as southern Swedish waters, vegetation changes can occur due to an increase in bicarbonate levels, supporting the development of submerged Sphagnum and Juncus (Lükewille et al., 1997). Acidification and eutrophication are interacting because N deposition enhances acidification, but the acidity state also affects the N transformations. Consequently, not only the increased bicarbonate levels but also the increased N levels, specifically for NH_4, might trigger vegetation changes, as illustrated by a pH and N species induced plant community shift from *isoetes*, *lobelia* and *littorella* to a dominant community of *sphagnum* and *juncus* (Smolders et al., 2000; Schuurkes et al., 1986).

Critical limits for surface waters are generally derived for the protection of aquatic organisms, mostly fish species, but also invertebrates and aquatic plant species are being used as indicators. The link between water chemistry and fauna has mostly been evaluated from survey data, used to establish empirical relationships between water chemistry parameters and abundancy of organisms. In general, low pH and high Al concentrations cause a decline in species abundance. When focusing on pH, critical values range between 5 and 6. For example, Jeffries & Lam (1993) established a critical pH of 6 to protect fish and other aquatic organisms in Canadian lakes whereas Rask et al. (1995) derived critical pH values ranging between 5 and 5.8 for Finnish lakes. When focusing on aluminium^{+}, Rask et al. (1995) derived critical concentrations ranging from 20 mg m^{-3} for roach up to 90 mg m^{-3} for perch. In addition, Hultberg (1988) gave a value of 50 mg m^{-3} for perch.

The most common indicator to assess whether a water body is acidified, is the Acid Neutralising Capacity (ANC), with a commonly used critical limit of 20 meq m^{-3} to avoid adverse impacts on the fish and invertebrate populations (Lien et al., 1996). Of course, pH, Al and ANC are interrelated but their relationship depends, in addition to the partial pressure of CO_2, mostly on the amount of dissociated organic anions and complexed Al. The advantage of ANC is that it is a robust variable, e.g., it is not affected by the partial pressure of CO_2. This limit has been used for surface waters in northern Russia (Moiseenko, 1994), Nordic countries (Posch et al., 1997),

southern central Alps (Boggero et al., 1998) and China (Duan et al., 2000). Other limits were set at zero in the United Kingdom (CLAG, 1995) and at 40 meq m^{-3} in Canada (Henriksen et al., 2002). The critical limit of 20 meq m^{-3} is frequently used in critical load calculations.

6.2.3 Geographic variation and trends in water quality

Changes in nitrogen concentrations in view of eutrophication

To monitor the surface water quality within Europe a total of ~22,000 monitoring stations were operational in 2000-2003, compared to ~14,000 in 1996-1999 (EC, 2007). In 2007 the number of groundwater stations grew to ca 27,000 (EC, 2010). Unlike ground water, the average nitrate concentrations across 1258 rivers declined from 2.3-1.9 mg NO_3 l^{-1} (Figure 26). Histograms of the percentage of sampling points with NO_3 concentrations in surface water in 2004-2007 and the changes to 2000-2003 in different classes, based on reported data in EC (2010) are given in SI, Figure A1.

The average river nitrate concentration decreased steadily over the period 1992 to 2009 and levelled off since then (EEA, 2018). Rivers that drain land with intense agriculture or a high population density generally have the highest nitrate concentrations. There has been a decrease in river nitrate concentrations at 47% of the monitoring sites since 1992, and an increase at 16% of the sites. Czechia, Denmark, Germany, and Slovakia had the highest proportion of significantly decreasing trends (63-100%). France, Ireland, Spain, and Switzerland had similar proportions of significantly increasing and decreasing trends, while Estonia had the highest proportion of significantly increasing trends (44%).

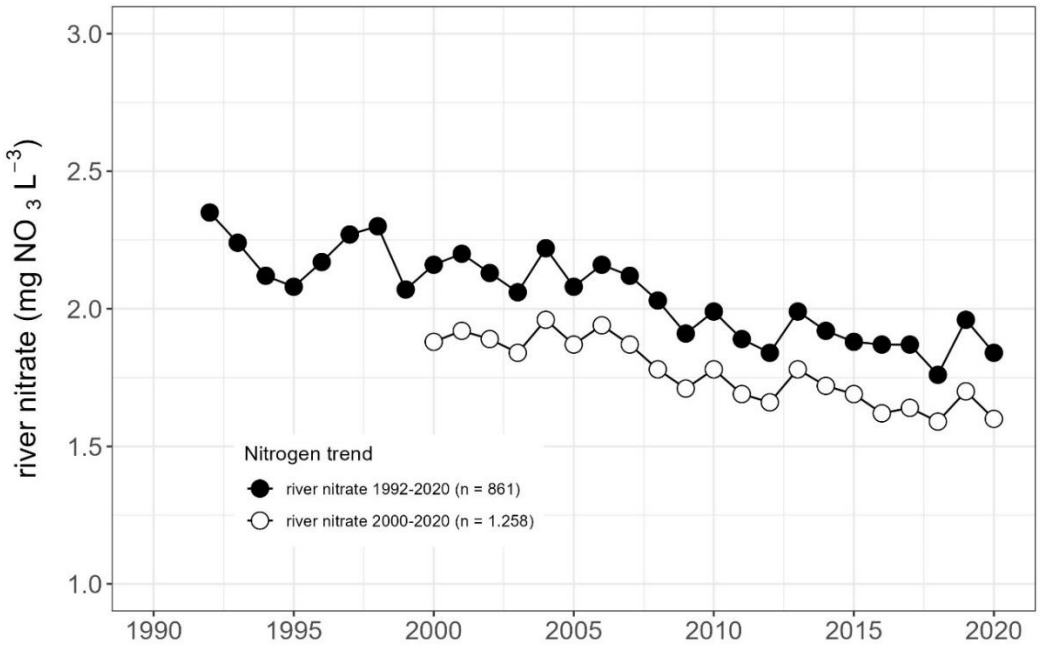


Figure 26. Trends in nitrate in European rivers as reported by the European Environment Agency (EEA, 2023a). The geographical coverage is the 38 EEA member countries, but only complete time series are included. The shown time series are aggregated to European level by averaging across all sites for each year.

An overall decline in total N concentrations, although slowing in recent years, is observed for Austria, Belgium, Czechia, Denmark, Germany, Serbia, and Sweden (Figure 27), contributing to the pattern seen in the European time series. The trends in nitrate and nitrogen concentrations shows that in a large majority of surface waters (Vigiak et al., 2023), the N concentrations are decreasing (55%) or stable (31%).

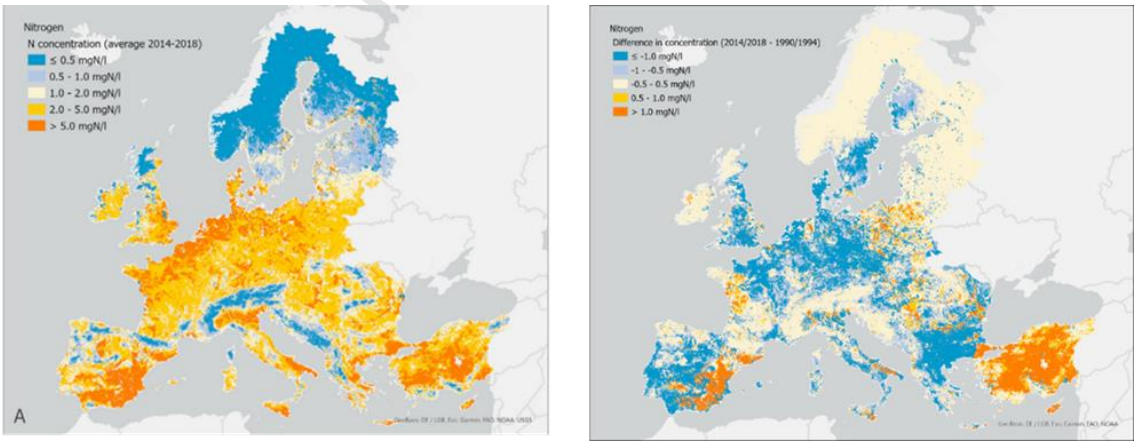


Figure 27. Geographic variation in modelled average total N concentration of freshwaters for the years 2014-2018 (left) and the change in total N concentration from 1990-1994 up to 2014-2018 (right). Source: Vigiak et al. (2023).

Over the whole of Europe, the total N load to surface water declined from 31.5 Tg year^{-1} in the early 1990s to 29.7 Tg yr^{-1} in 2018, i.e. a reduction of 6%, which occurred mainly in the first

half of the period (Vigiak et al., 2023; Bouraoui & Grizetti, 2011), due to a reduction in fertiliser use and a substantial decrease in emissions from scattered dwellings. There were notable regional differences (Figure 27) with the largest reduction occurring in the Greater North Sea (-14%) and even an increase occurring in the Black Sea region (+8%). Large parts of the surface waters across Europe had mean annual concentrations above 5 mg N l^{-1}, particularly along the Greater North Sea coastline, the UK and Ireland, and the Mediterranean regions. Areas of very high nutrient concentration shrank considerably compared to the start of the period (Figure 27), and from 1990 to 2018, mean annual nutrient values shifted consistently towards lower concentrations. The share of stream network length with concentrations compatible with good ecological status (< 2 mg N l^{-1}; < 60 μg P l^{-1}) increased from 47 to 55% for N, and from 30 to 43% for P. The improvement for N occurred mostly in upstream reaches, probably related to the reduction of atmospheric N deposition (Vigiak et al., 2023).

Changes in nitrate and sulphate concentrations in view of acidification

Trends in surface water chemistry have been reported for the period 1990-2008 using observations from long-term (ICP Waters) monitoring programs in more than 20 countries (Skjelkvåle & De Wit, 2011; Garmo et al., 2014). These studies show a consistent pattern of decreasing SO_4 concentrations in response to the widespread decline in S deposition. Despite the reduction in N deposition, NO_3 concentrations exhibit positive, negative, and neutral trends (Figures 28 and 29), being consistent with an earlier analysis by Stoddard *et al.* (1999).

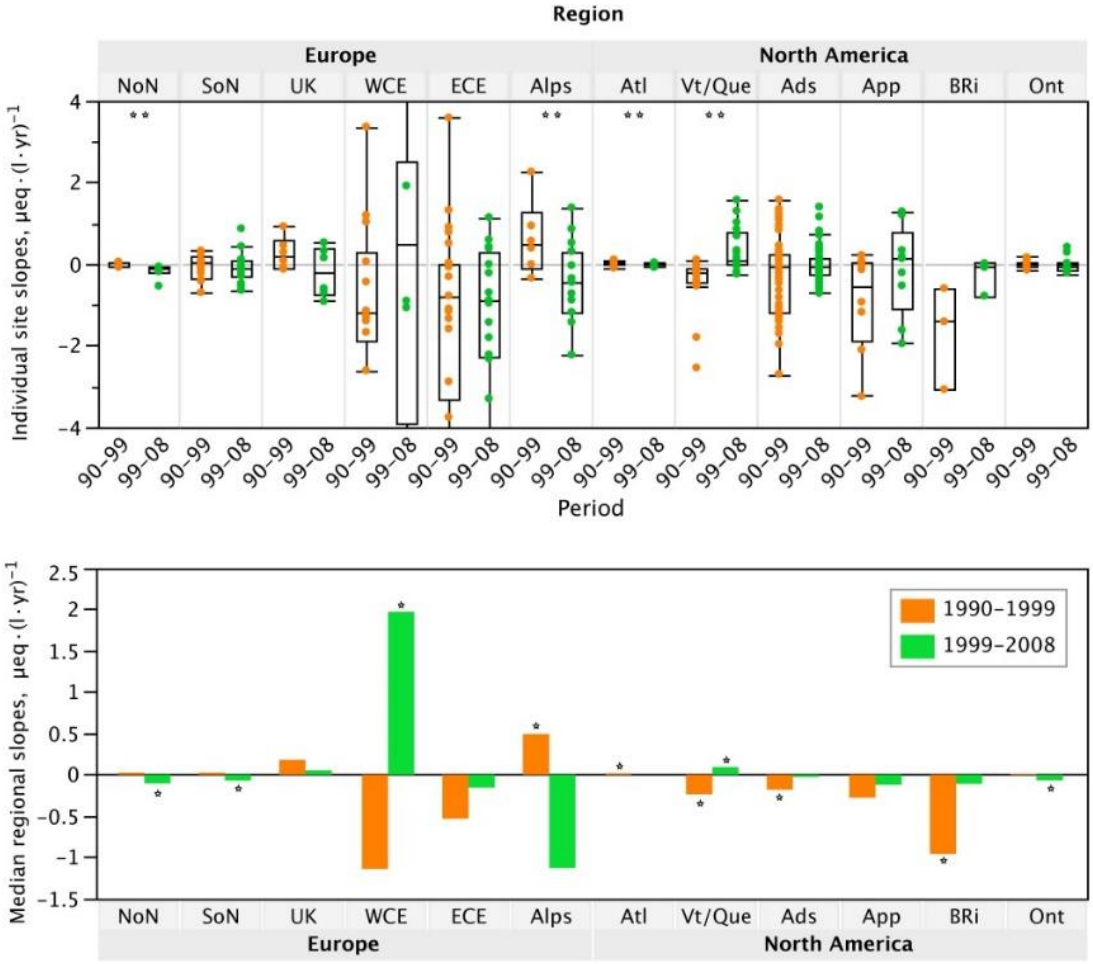


*Figure 28. Slopes of regional trends in concentrations of NO_3^- for twelve regions in Europe and North America (North Nordic: NoN, South Nordic: SoN, United Kingdom: UK, West Central Europe: WCE, East Central Europe: ECE, Alps: Alps, Maine and Atlantic Canada: Atl, Vermont and Quebec: Vt/Que, Adirondacks: Ads, Appalachians: App, Blue Ridge Mountains: Bri and Ontario: Ont) for the time spans 1990-1999 and 1999-2008. The first figures indicate the individual site slopes with the boxes and whiskers covering the 25th to 75th and 10th to 90th percentiles of slopes, while the median is indicated in the second graph. Asterisks indicate the significance level (** p<0.05, * p<0.10) of a non-parametric test for difference between the distributions of slopes for the two time periods. Source: Skjelkvåle and de Wit (2011).*

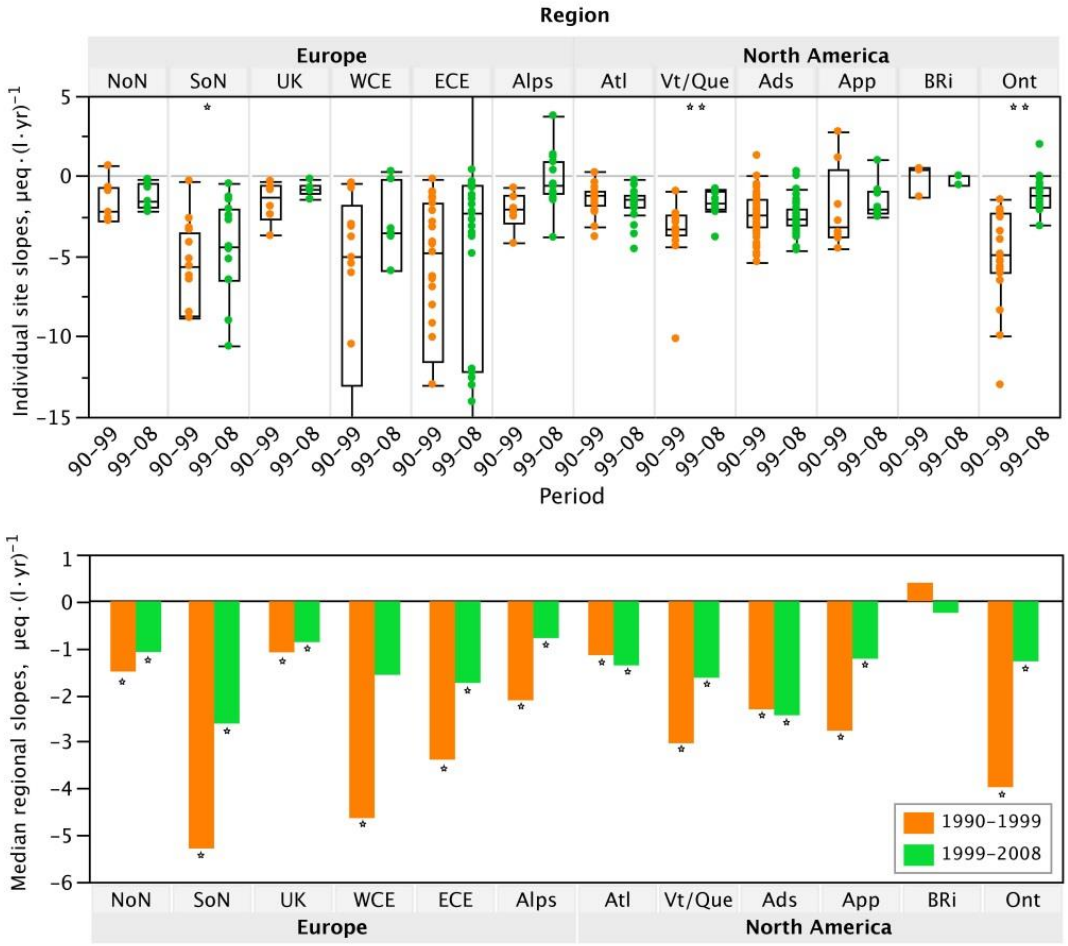


Figure 29. Slopes of regional trends in concentrations of non-marine SO_4^{2-} for twelve regions of Europe and North America for the time spans 1990-1999 and 1999-2008. For region description, interpretation of boxplots and significance levels, see Figure 28. Source: Skjelkvåle and de Wit (2011).

While a NO_3 decline seems to be far more common than a NO_3 increase, the majority of catchments show no significant trend, opposite to the trends observed for total N (Vigiak et al., 2023). The long-term monitoring data indicate no major change in N saturation during the 1990s, indicating that progression to increased N saturation is a slow process with a time scale of decades. Compared with S, N is much more influenced by biological processes within ecosystems. Hence changes in N deposition not always directly correlates with changes in inorganic N leaching and runoff. In addition, nitrogen losses are highly dynamic given changes in climate and hydrology (Helliwell et al., 2014), thus affecting acidification and eutrophication.

Time trend data show not only evidence of chemical recovery from surface water acidification, but sometimes also evidence of biological recovery. One well documented long-term dataset is from Lake Saudlandsvatn in southernmost Norway, where the abundance of brown trout (*Salmo*

trutta), caddisfly (*Hydropsyche siltalai)* and zooplankton (*Daphnia longispina)* has been monitored together with water chemistry (Hesthagen et al., 2011). The lake was highly acidified in the 1970s and 1980s, with episodic pH values below 5.0, and the critical load was exceeded fivefold. Chemical recovery following reduced S deposition is observed since the late 1990s and the S deposition dropped below the critical load around 2000.

The brown trout population was stable until the early 1980s, then started to decline and nearly went extinct in the 1990s before starting to recover ever since. Parallel with the trends in water chemistry it was also observed that the caddisfly disappeared from the tributaries in the 1980s, but reappeared in 1996 and increased in abundance from 2000 onwards. Recovery in all three organism groups (brown trout, caddisfly, and zooplankton) coincided with an ANC of > 20 meq m^{-3} and a toxic inorganic Al concentration of < 30 mg m^{-3} (Hesthagen et al., 2011).

Surface waters (and in particular lakes) have been the earliest ecosystems for which critical loads (CLs) have been established on a broader scale, albeit with respect to acidification (see, e.g. Henriksen et al., 1990); critical loads for acidification for rivers, streams and ditches have hardly been considered. The CLs of acidity are mostly derived with the First-order Acidity Balance (FAB) model that builds on the earlier 'Henriksen model'. This approach is the aquatic equivalent of a simple mass balance model for soils, and relates the ANC-limit to the N and S deposition onto the catchment while accounting for N and S retention by in-lake processes (Henriksen & Posch 2001, Posch et al. 2012, 2015). In addition to the Nordic countries also the United Kingdom and Ireland have mapped CLs for surface waters on a national scale, and Figure 30 shows the temporal development (1990-2019) of the exceedance of the acidity CLs for 7749 lakes in Finland, Norway, Sweden, Ireland and the United Kingdom due to the modelled N and S deposition (see Curtis et al., 2015). While the average exceedance declines by about an order of magnitude, the number of lakes exceeded is going down about two thirds (from 37% to about 12%).

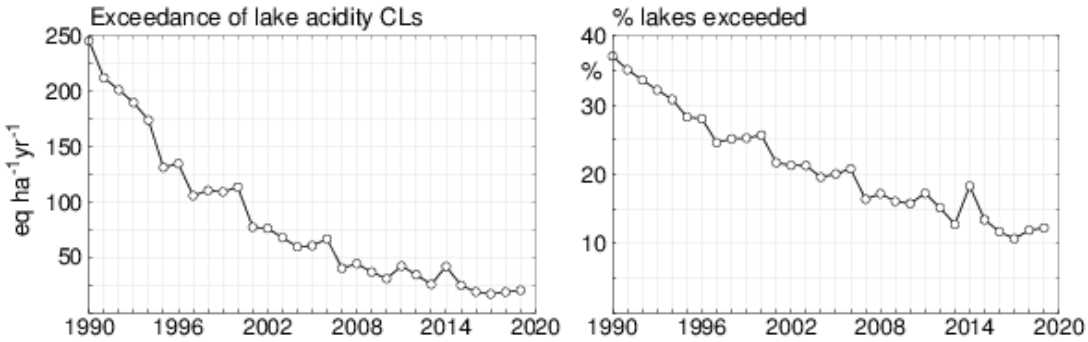


Figure 30. The 1990–2019 temporal development of the modelled average exceedance (in eq $ha^{-1}a^{-1}$; left) of critical loads of acidity and of the percent of lakes exceeded (right) in Finland, Norway, Sweden, Ireland, and the United Kingdom (7749 lakes). Source of the critical loads is Curtis et al (2015) and of the depositions are EMEP model results.

6.3 Impacts on coastal and marine ecosystems

6.3.1 Eutrophication and critical levels

A clear and widespread effect of an accelerated N cycle is the discharge of N to the marine ecosystem causing coastal eutrophication (Tyrell, 1999; NRC, 2000). There often is a gradient from freshwater to estuarine and coastal systems with regard to the role N plays as a limiting nutrient, with P limitation and N and P co-limitation prevailing in upstream freshwater systems to almost exclusive N limitation in more saline downstream estuarine and coastal systems (Elser et al., 2007; Paerl and Piehler 2008). Marine primary plankton has over millennia established a constant relationship of nutrients referred to as Redfield ratio; 16 moles of nitrate and 1 mole of phosphate is the mean required nutrient ratio for marine phytoplankton (Redfield, 1958). Since most of the nutrients lost from land are delivered as nitrate, and much less as phosphate, the N:P ratio steadily increases (Tilman et al., 2002). This increase in stoichiometric N:P ratios is observed in many rivers and lakes in catchments dominated by agriculture (Elser et al., 2009), and increased in almost all European catchments over the last decade (Vigiak et al., 2023). From 1990 to 2018 the N:P ratio increased by 0.7 units in the Mediterranean Sea and up to 4 units in the Atlantic Ocean and the Black Sea. For the whole of Europe, the N:P ratios increased from 11.5 to 14.0, with six of the nine catchments exceeding the Redfield ratio (Vigiak et al., 2023).

This corresponds to global oceanic studies where the atmospheric deposition is now estimated ~20 times the Redfield ratio for N : P (Jickells, 2006; Peñuelas et al., 2011, 2013).

In estuaries, high nutrient inputs with N:P ratios exceeding the Redfield ratio, microbial processes are favoured that recycle and retain the N-compounds in river water (Lunau et al., 2012). The surplus of N will be released by the phytoplankton as dissolved organic nitrogen (DON), fuelling the microbial loop with associated N_2O losses (Lunau et al., 2012) and retaining the remaining N in the aquatic environment. Consequently, the total N inputs in the coastal and marine environment increases, as illustrated by observations from the Elbe River (Dähnke, 2008).

Excess N and P runoff from farm fertilisers, sewage, and industrial pollutants together with atmospheric deposition may cause blooms of microscopic algae known as phytoplankton. Atmospheric N deposition is perhaps even more consequential than coastal eutrophication due to the vast expanses of open ocean affected (Jickells et al., 2017). When algae and other organisms die, they sink to the bottom, and are decomposed by bacteria, using the available dissolved oxygen. Salinity and temperature differences between surface and subsurface waters lead to stratification, limiting oxygen replenishment from surface waters that can lead to the formation of oxygen-depleted, so called hypoxic or "dead" zones. Because very few organisms can tolerate the lack of oxygen, anoxic conditions can destroy the habitat for fish, shellfish, and most other forms of life. Excess N concentrations may thus cause oxygen-starved (hypoxia) areas of the world's oceans that are devoid of higher life forms like mussels, polychaetes or fish (Boesch et al., 2001; Boesch, 2002; Rabalais et al., 2014).

A threshold value for oxygen concentrations related to hypoxia often used for estuaries and shallow coastal waters is 2 mg l^{-1} (Gray et al., 2012), with the threshold being related to impaired recruitment. Since 1950, more than 500 sites in coastal waters have reported oxygen concentrations $\leq$2 mg l^{-1} at global scale (Diaz & Rosenberg, 2008; Breitburg et al., 2018). However, he tolerance to hypoxia varies greatly among marine taxa; with mobile fish and crustaceans being generally most sensitive, requiring oxygen concentrations higher than 2 mg

l^{-1} to avoid unimpaired recruitment and growth, while others are adapted for life in low oxygen conditions (Vaquer-Sunye & Duarte, 2008; Isensee et al., 2016). Despite the variability in sensitivity and thereby the uncertainty in directly linking oxygen concentrations ≤2 mg l^{-1} to oxygen-starved areas in coastal waters, the trends in those areas increased from about 10 documented cases in 1960 (UNEP, 2003; Diaz & Rosenberg, 1995, Diaz et al., 2004) to at least 169 in 2007 at global scale (Selman et al., 2008). Notable dead zones in Europe are found in the Baltic Sea, Black Sea, Adriatic Sea (part of the Mediterranean Sea) and North Sea (Anonymous, 1993).

Nutrient imbalances affect the potential for eutrophication (Salas Herrero et al., 2019; Garnier et al., 2021; Sardans et al., 2021a, 2021b) and favour Harmful Algal Blooms (HABs) (Glibert & Burfort, 2017), however, it is hard to predict ecosystem responses. While nutrient pollution is the primary driver of eutrophication, the relationship between nutrient pollution and HABs is more complex than previously thought (Glibert et al., 2005; Heisler et al., 2008). The occurrence of HABs lies at the intersection of the physiological adaptations of the harmful algal species, the environmental conditions, and interactions with co-occurring organisms that alter abiotic conditions and/or aggregate or disperse cells (or can alter abiotic conditions in a favourable or unfavourable manner), in turn promoting or inhibiting their growth (Glibert et al., 2018). Some toxic cyanobacteria are more common under elevated N:P ratios or when total N concentrations are high, and several cross comparative studies show that cyanobacterial biomass can be predicted from increasing N concentrations and from changes in N:P ratios (Smith, 1983; Downing, 1997, Downing et al., 2001; Kosten et al., 2012; Dolman et al 2012). Therefore, species composition is important to consider (Dolman et al., 2012) as, in fact, some of the most toxic cyanobacteria are not N_2-fixing species. Glibert et al. (2014) argue that increasing N:P ratios in waters favour HABs when the N form is disproportionately in reduced form (i.e. urea, NH_4) relative to oxidized form (i.e., NO_3), and underpins this by physiological evidence and long-erm trends in HABs and fertilizer use in China.

6.3.2 Acidification and critical loads

Anthropogenic N and S deposition to the oceans alters surface seawater chemistry, leading to acidification and reduced total alkalinity. However, ocean acidification is generally more associated with increased CO_2 levels causing the increased dissociation of protons from carbonic acid. While the oceans are strongly buffered with a pH above 7, even a slight decrease in pH stresses saltwater organisms and increases mortality rates. Lower pH and carbonate concentrations are of particular concern for a range of benthic and pelagic organisms that form calcareous shells such as corals, coralline algae, foraminifera, pteropods, and coccolithophores (Orr et al., 2005; Kleypas et al., 2006; Chan & Connolly, 2013; Fabricius et al., 2011). Acidification impacts on single species and ecosystems by reducing species diversity and thereby altering ecosystem functioning (Kroeker et al., 2013; Przeslawski et al., 2015; Sunday et al., 2016). The acidification effects, although not as large globally as those of anthropogenic CO_2 uptake (Feely et al., 2004; Andersson & Brunberg, 2006), are particularly significant in coastal regions because these regions are already vulnerable to other human impacts, including eutrophication, pollution, overfishing, and climate change (Doney et al., 2007). Ocean acidification is therefore a significant threat to ecosystems, including coral reefs and coastal benthic and planktonic food webs dominated by calcifying organisms (Hutchins & Capone, 2022).

6.3.3 Geographic variation and trends

Eutrophication and acidification in marine, coastal and estuarine ecosystems is a consequence of nutrient over-enrichment, with N and P coming from land-based sources, marine activities, and atmospheric N deposition, as well as fluxes from neighbouring water bodies. Between 1980 and 2019, inorganic N levels declined overall in the Greater North, Baltic and Celtic Seas (Figure 31) as a consequence of EU policy implementation. Time series data were not sufficient to identify trends in other regional seas. The occurrence of reduced oxygen concentrations in bottom waters of coastal and marine waters has increased both in spatial extent and duration in the last decades, owing mainly to a combination of natural causes and anthropogenic pressures

such as eutrophication (Figure 31). Results indicate that conditions have deteriorated in 9% of the cases, improved in 3%, while for the majority (88%) no trend could be established. The Baltic and Black Seas suffer the most from oxygen depletion, caused by stratification, but which has intensified due to eutrophication and ocean warming.

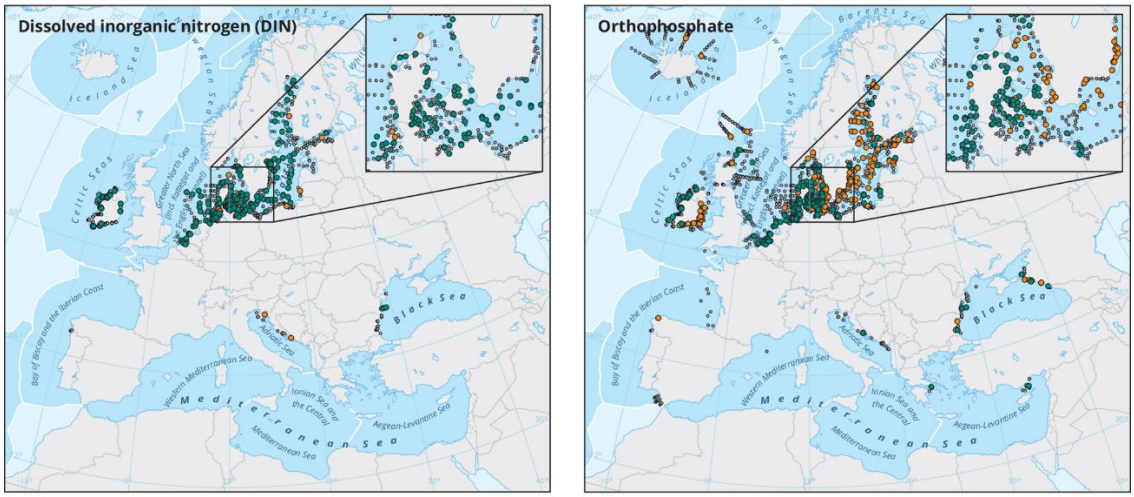


Figure 31. Trends in dissolved inorganic N and P concentrations in transitional, coastal, and marine waters in Europe. Blue dots are decreasing, orange dots increasing and grey dots did not change over the period 1980 to 2019 (EEA, 2023a).

The Baltic Sea is a prime example of human impacted eutrophication and expansion of dead-zones (Carstensen et al., 2014a, 2014b) in both the central basin and in coastal zones (Conley et al., 2011). The Baltic Sea has intensive phytoplankton blooms in spring and especially in summer, driven by the nutrients from land and from atmospheric deposition and the presence of N fixing cyanobacteria (Vahtera et al., 2007). Release of P under anoxic conditions and high denitrification rates at the anoxic-oxic interface leads to P accumulation and a decline in N:P ratios. Phosphate can only be removed on long time scales through burial of organic matter in the sediments. This internal nutrient cycle implies that the Baltic Sea will suffer from oxygen deficiencies for decades (Steckbauer et al., 2011) even when the external nutrient inputs substantially decline. This might explain why the occurrence of anoxic regions remain even with declining nutrient inputs from land (Andersen et al., 2017). The trend in nutrient reductions is illustrated in Figure A2, which shows that the eutrophication ratio (ER) of various coastal and open waters of the Baltic Sea declined or stayed relatively constant during the period 1980-2010.

Inputs of nutrients to the North Sea also decreased during the last decades as a result of emission reductions (OSPAR, 2000, 2017). Specifically, the P load has been reduced (Behrendt et al., 2000) leading to an increase in N:P ratios. Skogen *et al.* (2004) showed that the production regime in the southern North Sea is therefore P limited, while N remains the limiting nutrient in the northern North Sea. In addition, the oxygen levels decline since 2000 due to warming of the North Sea water (Queste et al., 2013). Coastal and inshore waters still experience problems from eutrophication (OSPAR, 2017) and substantial nutrient reductions in the agricultural catchments is needed to achieve long-term recovery of the marine environment.

The Mediterranean Sea, isolated from Atlantic Ocean, has specific properties. The arid climate and high evaporation lead to high salinity while fresh water input through rivers play a minor role. Therefore, the Mediterranean Sea is an oligotrophic ocean with strong P depletion and high N:P ratios of 28:1 (Krom et al., 2010). The role of groundwater discharge in nutrient delivery to the sea is at least as high as the nutrient delivery by riverine discharge, which is due to the karstic soil causing high leaching to groundwater (Rodellas et al., 2015). Only the Northern Adriatic Sea, receiving the discharge of the largest Italian River Po, is affected by eutrophication and hypoxic bottom waters (Justić, 1991; Gooday et al., 2009). Historical records suggest that extensive blooms have always occurred in the region, but have recurred more often since the 1980s until today (Kraus & Supić, 2015).

7 Costs and benefits of reducing nitrogen inputs for Europe

This review showed that since the last decade of 20th century, losses of N_r to the environment in the EU27 have stabilized or decreased, which is a result of mitigation measures related to EU and national policies and a decreasing use of N-fertilizer. Important pieces of EU legislation for N were the Nitrates Directive, Water Framework Directive, Groundwater Directive, Ambient Air Quality Directive, National Emissions Ceilings Directive, Urban Waste Water Treatment Directive, Marine Strategy Framework Directive, Integrated Pollution Prevention and Control (IPPC) and the Habitats Directive. An important question with respect to policies related to the

reduction of N emissions and its impacts is whether the reductions can be achieved in such a way that the benefits, in terms of human welfare improvement, exceed (or at least are in balance with) the associated costs. In this context, Brink et al. (2011) and Van Grinsven et al. (2013) explored the potential of cost-benefit assessment to set priorities for an integrated improved management of the N-cycle in the EU27. Here we applied a slightly updated approach to assess trends in N-associated costs of impacts over the period 1990-2010, based on trends in N losses to air and water.

Unit costs per kg emitted nitrogen and total costs for EU27

The concepts and data for costs and benefits of nitrogen in the EU27 environment are given in Table A.2. The economic value of N-damage was based on 'Willingness to Pay' (WTP) of citizens to prevent premature death and loss of health, to prevent degradation of ecosystems or restore ecosystems, or to reduce greenhouse gas emissions (based on the carbon price in emission trading system). The monetary value per kilogram of N_r emission or N_r loss was determined by dividing the economic value of the N share of an impact (euro yr^{-1}) by the associated N_r flux (kg yr^{-1}). The economic value of benefits of N in agriculture was based on yield response of wheat, oilseed rape and milk to N and current world market prices of crops and synthetic fertilizer, and the response of wood yields to N_r deposition. For crops we used long-term yield response to N. For details we refer to Brink et al. (2011) and Van Grinsven et al. (2013).

Unit damage costs for N_r per unit of emissions to the environment (Table 13) show a wide range between different N_r compounds and are often very uncertain.

Table 13. Estimated cost of different N_r -threats in Europe per unit N_r emitted based on WTP data in the period 1995-2005 (After Brink et al. 2011).

Emission	Health €/kg N_r	Ecosystem €/kg N_r	Climate €/kg N_r	Crops, wood €/kg N_r	Total €/kg N_r
NO_x -N to air	10-30[1]	2-10[2]	-9-2[3]	1-2[4]	3-42
NH_3-N to air	10-20[5]	2-10[2]	-3-0[3]		9-30
N_2O-N to air	1-3[6]		5-15		6-18
N_r to water	0-4[7]	5-20[8]			5-24
N_r fertilizer			0.03-0.3	-2 - -5[3]	-2 - -5
N deposition		5-20[8]		-3[3]	2-17

[1] Human health effects by particulate matter, NO_2 and O_3

[2] Includes 2 – 10 €/kg N_r for impacts on terrestrial ecosystems (eutrophication, biodiversity) and in case of NO_x an additional.
[3] Negative values indicate benefits; especially increase in crop yield in response to N fertilizer and increase in forest growth in response to N deposition. Furthermore cooling effects of NO_x-N and NH_3–N emissions due to aerosol formation
[4] 1 – 2 euro/kg N_r for crop damage by ozone
[5] Human health effects by particulate matter.
[6] Human health effects due to increased ultraviolet radiation from ozone depletion.
[7] Human health effects due to NO_3 in drinking water (via leaching to groundwater).
[8] Impacts on aquatic ecosystems (eutrophication, biodiversity).

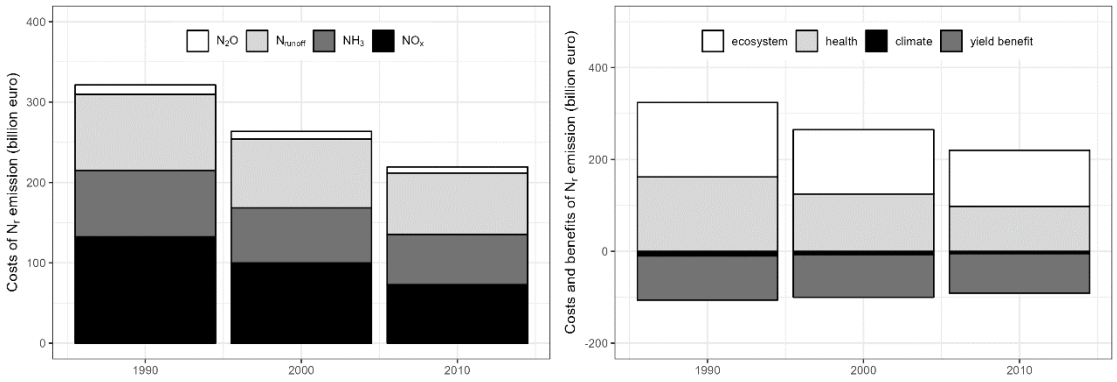
Highest values are associated with air pollution effects of NO_x on human health, followed by the effects of N_r loss to aquatic ecosystems and the effects of NH_3 on particulate matter. The smallest values are estimated for the effects of nitrates in drinking water on human health and the effect of N_2O on human health by depletion of stratospheric ozone.

To calculate the national N costs for 1990, 2000 and 2010 in the EU27, national emission and deposition fluxes from EMEP, UNFCCC and the IMAGE model (Beusen et al., 2016) were combined with the marginal N costs and benefits (Table 13). Missing data were estimated from countries having comparable agricultural and socioeconomic conditions. Because temporal changes in unit damage costs were unavailable, calculated differences among the years were mainly due to differences in N_r emissions. Improvements compared to the cost benefit assessment for the EU27 for the year 2008 (Van Grinsven et al., 2013) include (1) an increase of the lower bound for health impacts by ammonia aerosols in view of more consensus on occurrence of these impacts, (2) accounting for ecosystem impacts of N_r deposition on sea areas and (3) for benefits of N_r deposition for wood production.

Trends in total costs of nitrogen for EU27

The direct N inputs from fertilizer, manure, and biological N fixation, and of N_r emissions to air of NO_x and NH_3, decreased from 35 Tg in 1990 to 26 Tg in 2010. The resulting sum of environmental losses to land and seas, as atmospheric deposition, and to rivers, from non-point and point sources, decreased from 15 Tg in 1990 to 11 Tg in 2010. The total N removal by crops and wood, denitrification and storage in soil and sediment decreased from 20 Tg in 1990 to 15 Tg in 2010. The total environmental costs of N_r in 2010 was estimated at €220 billion per year (uncertainty range €85-€415) of which 65% is related to emissions to air and 35% to

emissions to water. The cost share of total impacts to human health is 46%, to ecosystems 57%, while the net impact on global warming is not significant (-2%); warming impacts of N_2O are cancelled out by cooling impacts of N containing aerosols and N driven carbon sequestration



(Figure 32).

Figure 32. External cost of N pollution for various N components (left) and for various impacts including benefits for crop and wood production (right) for 1990, 2000 and 2010, using time-constant unit damage costs.

In line with Van Grinsven et al. (2013), about half of the total costs is related to agricultural N_r emissions. The total mean costs decreased from €320 billion in 1990, to €265 billion in 2000, to €220 billion in 2010 (Figure 32). Net N_r benefits in this period for crops and wood remained constant at €95 billion; the yield decrease by reduced N fertilization was compensated by the decrease of ozone damage.

The total damage costs in 2000, taking into consideration that 'Willingness to Pay' (WTP) data for unit costs apply to the period 1995-2005, was €550 per person, ranging from €210 up to €1050. This equates to 1-5% of the average GDP of €16,300 per capita in 2000 and therefore constituted a substantial welfare effect. Total damage in 2010 was substantially lower at €440 per person, ranging between €170 and €840, and accounting for 0.5-3% of the average GDP in 2010 of €30,250. This estimate of GDP loss is an underestimate as WTP in the EU27 increases proportionally with GDP. Taking this effect into account, the range of GDP loss would increase up to 1-6%. The per capita cost is about twice as high as the present 'Willingness to Pay' to

control global warming by carbon emissions trading, which is 100-300 €/capita (taking a CO_2-eq emission of 11 ton/capita (2004) and CO_2 emission trading price of 10-30 €/ton).

Benefit-cost considerations for N-mitigation

The estimates of potential welfare loss due to N_r in the EU27 allow an estimation of the level of mitigation costs up to which there is a net gain of welfare. Combining the marginal mitigation cost curves from the GAINS-model and the marginal damage cost values (Table 13) allows calculation of a marginal Benefit-Cost ratio's (BC-ratio). For improvement of N-management in agriculture it is most meaningful to express N-damage costs and benefits per unit of N applied to agricultural land. This is possible by combining unit damage costs for N-compounds in Table 13 with emission factors for N-compounds per unit of N application (e.g. Velthof et al. 2009). This leads to a maximum range of damage costs of 0.4-6.8 € kg^{-1} of added N, assuming the use of calcium ammonium nitrate, which is the most used N fertilizer in Europe. In comparison, (short-term) benefits of N-fertilizer for producing bulk commodities such as cereals, potatoes, sugar beet and milk range between 0.3-3.3 € kg^{-1} fertilizer-N. The results for costs and benefits of nitrogen for the EU27 thus show the high likelihood that environmental costs of N_r losses to the environment are very substantial and at present tend to exceed the direct economic benefits for agriculture.

Results of the calculation show that robust (using the lower bound of the estimates) marginal welfare increases for NO_x are to be expected up to an emission reduction of 0.25 Tg (7% of total EU27 emissions), for NH_3 up to 0.6 Tg (17%) and for N_2O up to 0.3 Tg (38%) (Brink et al. 2011). The rather small proportion of NO_x and NH_3 reduction implies that without technological innovation, there is a modest scope for N_r mitigation with robust welfare increase. Based on a meta-analysis of 1,521 worldwide field observations, Gu et al. (2023) identified 11 key measures to reduce the nitrogen losses from croplands to air and water by 30-70% while increasing crop yield by 10-30%. The costs of these mitigation measures are 25 times lower than the benefit from reduced pollution and increased crop yield. This implies that numerous cost-efficient measures exist to increase N_r efficiencies and reduce N_r losses, particularly for

manure, without any effect on farm income or total food production. Results also indicate that spatial relocation of N polluting agricultural activities within the EU can be a measure to reduce external costs, which was demonstrated for pig production (Van Grinsven et al., 2018). Similarly, Liu et al. (2023) showed that that strategies for cost-effective $PM_{2.5}$ mitigation in Europe shift in favour of NH_3 controls, as NH_3 controls up to 50% remain 5 to 11 times more cost-effective than NO_x per unit $PM_{2.5}$ reduction, emphasizing the priority of NH_3 control policies for Europe. For NO_x emission from energy and transportation there is also scope for improvement, but the emission reduction range with robust net social N_r benefits is rather narrow (7%) in view of the steep marginal mitigation cost curves. So here there is clear need for technological innovation and for integrated measures that focus on climate benefits.

8 Conclusions

Nitrogen related impacts and the evidence for adverse effects

An overview of N related impacts as well the scientific evidence for these impacts, the level of scientific understanding and the possibility to quantify these impacts on a European scale is given in Table 14.

When the evidence for adverse effect is acknowledged to be very high, it implies that enhanced N emissions definitely result in adverse effects. The level of scientific understanding indicates the extent of insight into the mechanisms (cause-effect relation) behind a particular effect.

The most certain and widely accepted adverse impacts include:

- impacts on human health due to elevated atmospheric NO_x, ozone and particulate matter concentrations;
- eutrophication and acidification (together with sulphur) of soils and surface waters;
- impacts on flora diversity in terrestrial and aquatic systems;

- impacts on climate warming due to increased N_2O emissions, counteracted by N induced enhanced CO_2 sequestration, mainly due to elevated productivity of forests, causing global cooling

Table 14 Overview of N-related impacts with their evidence for effect, level of understanding and possibility (reliability) for European quantification.

Impacts	Evidence for effect[1]	Level of scientific understanding[2]	Possibility for European quantification[2]
Air and water quality and human health impacts			
– Air pollution			
NO_x and ozone	++	+	+
Fine particulate matter	+	+	+
Pollen pollution	?	+/-	-
– Nitrate in drinking water	+	+/-	+/-
Air quality and climate impacts			
– N_2O emissions	++	+	+
– CO_2 emissions	++	+/-	+/-
– CH_4 emissions	+	+/-	+/-
– Ozone and aerosols	+	+/-	+
Air and soil quality and species diversity of terrestrial ecosystems			
– Plant species diversity			
Nature	++	+	+
Forests	+	+/-	+/-
– Faunal species diversity	+	+/-	-
Soil quality and forest nutrition			
– Nutritional imbalance	++	+	+/-
– Soil acidification	++	+	+
– Increased sensitivity to frost, drought, and diseases	+	+/-	-
Water quality and species diversity of aquatic ecosystems			
– Surface water ecosystems	++	+	+
– Coastal /marine ecosystems	++	+/-	+/-
Other impacts			
Visibility	+	+	+
Materials	?	+/-	+/-

[1] The evidence refers to the role of N in the effects: ++ = has certainly an effect; + = has probably an effect; - = has probably no effect; - - = has certainly no effect; ? = not well known.

[2] Scientific understanding and possibilities for European wide quantification: +: high; +/-: intermediate; - low.

Finally, evidence for adverse effects on faunal species diversity and marine systems exists, but the level of scientific understanding is less. Highest uncertainty about adverse effects exists for forest diversity, pollen pollution and materials, probably because the N effect is rather small and often confounded by other causes. The same is true for health implications arising via complex ecological feedbacks, such as the increase in infectious and parasitic diseases. Unravelling the exact role of nitrogen in these complex cycles requires substantial additional research.

Trends and geographic variation in adverse impacts of nitrogen use i

Over the period 1990-2019, N inputs to agriculture stayed relatively constant, but the emissions of ammonia (NH_3) decreased by ca 25%, specifically between 1990 and 2010, while emissions of nitrogen oxides (NO_x) decreased more than 50%. In response to those emission reductions, concentrations of NO_x, and of N in particulate matter also declined almost 50%, but the

reduction in NO_x-induced ozone (O_3) concentrations and O_3 related indicators (AOT40, SOMO35 and POD) was much less (ca 15-20%). Exceedances of critical O_3 concentrations for human health and of critical AOT40 and POD values for vegetation reduced in a similar order of magnitude.

Despite decreasing NH_3 emissions, however, NH_3 concentrations show a slight and steady increase from 1995 onwards, due to the large reduction in SO_x emissions, increasing the estimated areas exceeding critical levels for sensitive lichens by 10%. Unlike NH_3 concentrations, however, exceedances of critical N loads for terrestrial ecosystems have decreased by ca 60%, although the area exceeding critical N loads reduced by ca 10% only. Unlike N, the area exceeding critical acid loads has declined by more than 90% with only a few areas left with exceedances, due to high reduction in SO_x and NO_x emissions.

Trends in nitrate (NO_3) concentrations in groundwater showed a limited change averaged over Europe, but overall, there has been an increase in the area exceeding critical concentration NO_3 levels, in view of impact on human health, by ca 5%. However, N concentrations in surface water and the area exceeding critical levels in view of aquatic biodiversity has decreased and the same holds for N concentrations in coastal regions. Nevertheless, the eutrophication condition of coastal waters has overall not improved due to adverse impacts of elevated phosphorus inputs.

Finally the negative impacts of N induced N_2O emissions on climate are estimated to be outweighed by the positive effects of N induced CO_2 sequestration, mainly in forests, and this holds for the whole period 1990-2019.

Nitrogen hotspots, being areas with high exceedances in critical levels and loads of N compounds in air and water, are concentrated in intensive agricultural areas with high livestock densities and in urban region with strong industrial and traffic activities.

This overview shows that air quality responds relatively fast to emission reductions, although atmospheric chemistry can affect air pollutant concentrations. An example is the strong (ca

90%) reduction in SO_2 emissions, implying a comparably strong reduction in the area exceeding critical acid loads, but enhancing NH_3 concentrations despite NH_3 emission reductions. Unlike air quality, however, the response of soil and water quality and related impacts on terrestrial and aquatic biodiversity is much slower. This is mainly due to the buffer processes in the soil, including N immobilisation versus N mineralization, sulphate adsorption versus desorption, and cation exchange, implying a damage delay time after an increase in N inputs, but also a recovery delay time after a decrease in N inputs (see e.g. Schmitz et al., 2019 and Gilliam et al., 2019 for an overview of delayed impacts of N deposition reductions on forest soils and forest ecosystems). Similarly the environmental effects of improved N management in agriculture are often delayed in time. Reduction of external nutrient loads, for example, does not lead to an immediate shift in the eutrophic condition of the system (see Rabalais (2002) for an overview). In marine systems, overfishing and nutrient pollution of coastal zones also often coincide and contribute synergistically to degradation. This requires a multi-step approach to restore coastal ecosystems (Boesch et al., 2001).

Need for an integrated research and policy approach to nitrogen

In the last decades, there has been concentrated research effort on the impacts of elevated N use in different compartments, including air quality, soil quality and water quality, with its related impacts on humans and ecosystems. This has caused a lot of insight in each of those areas, but the co-benefits and trade-offs of measures in view of different impacts but has gained less attention and this holds specifically for related N policies.

Despite its relevance to most UN Sustainable Development Goals (SDGs), N pollution still lacks coordinated global governance (Sutton et al., 2021). There is an urgent need to develop a holistic ‘integrated’ approach to N that enhances the multiple benefits of N use for food and energy production while simultaneously reducing the multiple threats of N pollution for water quality, air quality, soil quality and greenhouse gas emissions, with its consequences for health, ecosystems and biodiversity environment and climate This includes a sustainable circular

economy for nitrogen to maximize synergies and avoid trade-offs. In this context, there is a clear need for a focus on sustainable food production, apart from the need to further reduce NO_x emissions, alongside with CO_2 emission reductions from industry and transport.

Even though the environmental impacts of reactive N compounds are serious, a world without the use of N fertilizers is not an option given the (growing) demand for food (Cassman et al., 2003). World agricultural output has grown at an average annual rate of about 2.2% during the past 60 years to feed the growing world population, and the increasing access to mineral fertilizers has played an important role in this increase. Current estimates suggest that at global scale only around 20% of applied nitrogen end up in food or feed products, with up to 80% lost to the environment in different forms. It is therefore critical to develop integrated and targeted plant nutrition strategies and practices that minimize trade-offs between productivity and the environment. Following a food system approach, responsible plant nutrition aims to (Dobermann et al., 2022):

- improve income, productivity, nutrient efficiency and resilience of farmers and businesses supporting them;
- increase nutrient recovery and recycling from waste and other under-utilized resources;
- lift and sustain soil health;
- enhance human nutrition and health through nutrition-sensitive agriculture;
- minimize greenhouse gas emissions, nutrient pollution, and biodiversity loss.

The outcome of this transformation will be a new societal plant nutrition optimum rather than a purely economic optimum. The new nutrient economy will become an integral component of a low carbon emission, environment-friendly and circular economy, supporting the food and nutrition requirements of a rising global population and improving the income and livelihood of farmers worldwide. This urgent need is well expressed by Dobermann et al. (2022): "*So far we have failed to achieve the goals stated above, despite many scientific and technical solutions that have existed for decades. Achieving it now, within one generation, will require a far more concerted effort by everyone involved, from the fertilizer industry to farmers and consumers of*

food and other agricultural products. Fast action - grounded in long-term sustainability thinking - is need to facilitate the transition towards a new paradigm for plant nutrition." Considering the adverse impacts of nitrogen, further efforts are thus needed to reduce the N losses during the whole food supply chain, as currently aimed for in the 'Farm to Fork Strategy' with a goal to reduce nutrient losses by 50% in 2030.

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List of abbreviations used in this paper

Organizations

CLRTAP UNECE Convention on Long-range Transboundary Air Pollution
EEA European Environmental Agency
EMEP European Monitoring and Evaluation Programme
IPCC Intergovernmental Panel on Climate Change
ICP International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects
UNEP United Nations Environment Programme:
WHO World health organization

Elements and exposures

VOCs Volatile organic compounds
PM Particulate matter
GHGs Greenhouse gases
AOT40 Accumulated hourly ozone concentration over a concentration threshold of 40 ppb
POD Phytotoxic Ozone Dose (),
SOMO35 Sum of ozone means exceeding 35 ppb

Impacts

YLL Years of life lost
YLD Years lived with disabilities
DALY Disability-adjusted life years
AAE Accumulated average exceedance
CL Critical load

CLF Critical load functio

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Highlights

- Adverse impacts of reactive nitrogen are most evident for human health and for aquatic and terrestrial ecosystems
- Effects of nitrous oxide emissions on climate are largely counteracted by N induced carbon sequestration
- Ecosystem restoration in response to declined N inputs in Europe since 1995 shows a time lag
- There is large spatial variation across Europe in N impacts, requiring regional policies
- Environmental costs of reactive nitrogen varied from 0.4 to 6.8 euro per kg N.