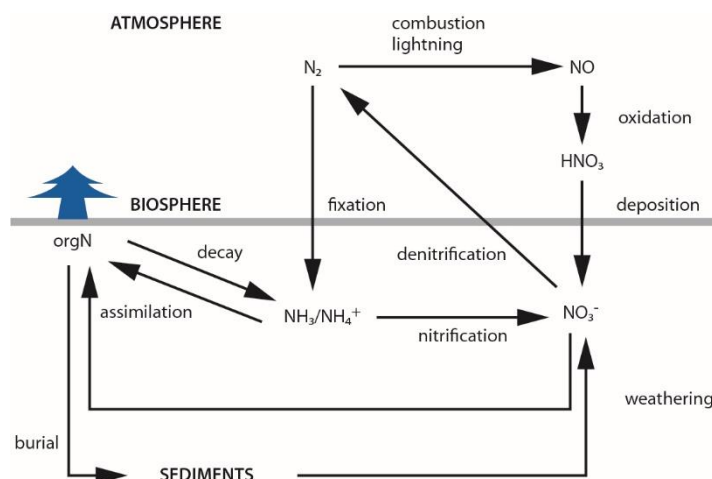


## Annex A. Basic facts on nitrogen

### A.1 The nitrogen cycle

Conversion of the highly stable (“inert”) dinitrogen ( $N_2$ ) molecule to biologically available (“reactive”) nitrogen, a process called “fixation”, is difficult. Fixation is achieved in soil and water by specialised bacteria which can reduce atmospheric dinitrogen to ammonia ( $NH_3$ ) or ammonium ( $NH_4^+$ ) (Figure A.1).<sup>1</sup> Microorganisms early on in the Earth's history developed the ability to use enzymes to produce or “fix”  $NH_4^+$  from dinitrogen, possibly because the availability of nitrogen through abiotic routes were biologically limiting (McRose et al., 2017). Nitrogen-fixing prokaryotes (bacteria and archaea) live in water (e.g. cyanobacteria), in soil (e.g. *Azotobacter*), in association with plants (e.g. *Azospirillum*), or in symbiosis with leguminous plants such as peas, clover and soyabeans (e.g. *Rhizobium*). In the latter case, the prokaryote shares the nitrogen with the plant; in exchange, the plant supplies the prokaryote with a source of carbon and energy for growth.

Figure A.1. The nitrogen cycle: major processes



*Note:* Ammonia ( $NH_3$ ) is highly soluble in water. When dissolved in water, a portion of  $NH_3$  reacts to form ammonium ( $NH_4^+$ ) as a function of the acidity of the water, i.e. its pH (“potential of hydrogen”). pH refers to the concentration of hydrogen ions ( $H^+$ ) in water. A decrease in pH results in an increase in  $NH_4^+$  and a decrease in  $NH_3$ .

*Source:* Jacob (1999).

The  $NH_3/NH_4^+$  is “assimilated” as organic nitrogen by the bacteria or by their host plants, which may in turn be consumed by animals. Eventually these organisms excrete the nitrogen or die; the organic nitrogen is eaten by bacteria and mineralised to  $NH_3/NH_4^+$ ,<sup>2</sup> which may then be assimilated by other organisms.

Bacteria may also use  $\text{NH}_3/\text{NH}_4^+$  as a source of energy by oxidising it to nitrite ( $\text{NO}_2^-$ ) and on to nitrate ( $\text{NO}_3^-$ ). This process (“nitrification”) requires oxygen (aerobic conditions).<sup>3</sup>  $\text{NO}_3^-$  is highly mobile in soil and is readily assimilated by plant and bacteria, providing another route for formation of organic nitrogen.

When oxygen is depleted in water or soil (anaerobic conditions), bacteria may use  $\text{NO}_3^-$  as an alternate oxidant to convert organic carbon to carbon dioxide ( $\text{CO}_2$ ). This process (“denitrification”) converts  $\text{NO}_3^-$  to dinitrogen ( $\text{N}_2$ ) and thus returns nitrogen from the biosphere to the atmosphere.<sup>4</sup> Denitrification may generate nitrous oxide ( $\text{N}_2\text{O}$ ).

An additional pathway for fixing dinitrogen is by high-temperature oxidation of dinitrogen to nitric oxide ( $\text{NO}$ ) in the atmosphere during combustion (e.g. forest fire) or lightning, followed by atmospheric oxidation of  $\text{NO}$  to nitric acid ( $\text{HNO}_3$ ) which is water-soluble and scavenged by rain.

Nitrogen is transferred to the lithosphere by burial of dead organisms (including their nitrogen) in the bottom of the ocean. These dead organisms are then incorporated into sedimentary rock. Eventually the sedimentary rock is brought up to the surface of the continents and eroded, liberating the nitrogen and allowing its return to the biosphere. Rock (geologic) nitrogen comprises a potentially large pool of nitrogen (Holloway and Dahlgren, 2002). Rock nitrogen concentrations range from trace levels (<200 mg N per kg) in granites to more than 1 000 mg N per kg in sedimentary rocks. Nitrate deposits accumulated in arid and semi-arid regions are also a large potential pool.

## A.2 The nitrogen problem in brief

According to anthropogenic and natural nitrogen flux estimates compiled by Battye et al., 2017, human nitrogen production is estimated to have increased fivefold in the last half-century (since 1960). According to Bleeker et al., 2013, human emissions of nitrogen oxides ( $\text{NO}_x$ )<sup>5</sup> and  $\text{NH}_3$  to the atmosphere have increased about fivefold since pre-industrial times. Atmospheric nitrogen deposition in large regions of the world exceeds natural rates by an order of magnitude. Much is deposited in nitrogen-limited ecosystems, leading to unintentional fertilisation and loss of biodiversity.

Still according to Bleeker et al., 2013, the transfer of nitrogen from terrestrial to coastal systems has doubled since pre-industrial times. As with terrestrial ecosystems, many of the coastal ecosystems are nitrogen-limited, such that abundance in nitrogen leads to algal blooms and a decline in the quality of aquatic ecosystems.

In addition, nitrogen has direct and indirect effects on climate change, being itself a greenhouse gas (GHG) and influencing emissions and the uptake of other GHGs such as  $\text{CO}_2$ . It also enhances ground-level ozone (GLO) formation, depletes stratospheric ozone, increases soil acidification and stimulates the formation of particles in the atmosphere, all of which have negative effects on people and the environment.

The social cost of nitrogen impacts appears to be largely underestimated.<sup>6</sup> Considering only the health impact of air pollution by nitrogen, the social cost is already in the hundreds of billions of USD. This is because nitrogen represents a

significant part of urban pollution with fine particles (PM<sub>2.5</sub>)<sup>7</sup> whose health cost (premature deaths) is estimated at some USD 1.8 trillion in OECD countries and USD 3.0 trillion in BRIICS countries (Roy and Braathen, 2017). When adding GLO pollution, of which nitrogen is also a precursor, health costs amount to some USD 1.9 trillion in OECD countries and USD 3.2 trillion in BRIICS countries (ibid).

To this must be added the health cost of water pollution by nitrogen as well as the cost of nitrogen pollution on ecosystems and climate. Keeler et al., 2016 made estimates for the state of Minnesota in the United States (see Chapter 2. ).

Global warming could make nitrogen pollution worse (Conniff, 2017). Nitrogen availability changes in response to climate change, generally increasing with warmer temperatures and increased precipitation. For example, Sinha et al., 2017 projects that climate change-induced precipitation changes alone will increase runoff nitrogen in U.S. waterways by 19% on average over the remainder of the century under a business as usual climate scenario. Toxic blue-green algae (or cyanobacteria) blooms, fuelled by nitrogen pollution, are being exacerbated by warmer temperatures and increased rainfall associated with climate change (Paerl et al., 2016).

However, there is not yet a thorough understanding of the complex interactions between climate change and the nitrogen cycle (e.g. in the case of seasonally inundated environments) and what the net effect might be under different scenarios of future anthropogenic activity. In particular, more research is needed on the joint impact of climate change and nitrogen on plant diversity. For example, an experiment in arid habitat in southern California has shown that nitrogen combined with changing precipitation can result in a community of native shrubs to shift to non-native grasses (Rao and Allen, 2010).

### A.3 Supplementary information on nitrogen impacts

Chapter 1 provides an overview of the main externalities associated with excess nitrogen in the environment. The following sections provide additional factual information. Section A.3.1 focuses on the troposphere.<sup>8</sup>

#### A.3.1 Air quality

Nitrogen dioxide (NO<sub>2</sub>) and, to a lesser extent, ammonia (NH<sub>3</sub>), are directly harmful to human health. Nitric oxide (NO) is not considered to be hazardous to health at typical ambient concentrations, but at high concentrations NO<sub>2</sub> is toxic, which is why it is classified by the World Health Organisation (WHO) as a hazardous atmospheric pollutant.

Parts of the secondary particulate matter (PM) are formed in the atmosphere from nitrogen oxides (NO<sub>x</sub>) and NH<sub>3</sub> precursors. NO<sub>x</sub> is also an essential precursor to the formation of ground-level ozone (GLO) (in the presence of sunlight).

#### *Nitrogen dioxide (NO<sub>2</sub>)*

There is strong evidence of respiratory effects (asthma exacerbation) following short-term NO<sub>2</sub> exposures, typically minutes to hours (in part, following USEPA, 2016a and 2017). In addition to the effects of short-term exposures, there is likely

to be a causal relationship between long-term NO<sub>2</sub> exposures and respiratory effects, based on the evidence for asthma development in children. People with asthma, as well as children and the elderly are generally at greater risk for the health effects of NO<sub>2</sub>.

#### *Ammonia (NH<sub>3</sub>)*

Short-term inhalation exposure to high levels of NH<sub>3</sub> in humans can cause irritation and serious burns in the mouth, lungs, and eyes (in part, following USEPA, 2016b). Chronic exposure to airborne NH<sub>3</sub> can increase the risk of respiratory irritation, cough, wheezing, tightness in the chest, and impaired lung function. In animals, breathing NH<sub>3</sub> at sufficiently high concentrations can similarly result in effects on the respiratory system. Animal studies also suggest that exposure to high levels of ammonia in air may adversely affect other organs, such as the liver, kidney, and spleen.

#### *Particulate matter (PM)*

PM pollution stands for a mixture of solid particles and liquid droplets found in the air (in part, following USEPA, 2013a). These particles come in many sizes and shapes and can be made up of hundreds of different chemicals. Some (primary PM) are emitted directly from a source, such as construction sites, unpaved roads, fields, chimneys or fires. Most form in the atmosphere as a result of chemical reactions; these are secondary PMs such as ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>).

PMs can be inhaled and cause serious health problems. They can get deep into lungs, and some may even get into bloodstream. Exposure to PM increases the risk of premature death in people with heart or lung disease, nonfatal heart attacks, irregular heartbeat, aggravated asthma, decreased lung function, and increased respiratory symptoms, such as irritation of the airways, coughing or difficulty breathing. People with heart or lung diseases, children, and older adults are the most likely to be affected by particle pollution exposure. The health effects are associated with long- and short-term exposures. The risk of mortality is higher for long-term exposure. The risk of morbidity for short-term exposure is higher for cardiovascular than respiratory diseases.

#### *Ground-level ozone (GLO)*

Breathing GLO (or "bad" ozone) can trigger reduced lung function, increased respiratory symptoms and pulmonary inflammation, particularly for children, the elderly, and people with asthma or other lung diseases, and other at-risk populations (in part, following USEPA, 2015). Health effects are associated with long- and short-term exposures. Research also indicates that GLO exposure can increase the risk of premature death from heart disease, although more research is needed to understand how GLO may affect the heart and cardiovascular system. GLO is most likely to reach unhealthy levels on hot sunny days in urban environments, but can still reach high levels during colder months.

GLO can also have harmful effects on sensitive vegetation and ecosystems, including crops, in particular during the growing season. When sufficient GLO enters the leaves of a sensitive plant, it can reduce photosynthesis, slow the plant's growth and increase the plants' risk of disease. The effects of GLO on individual

plants can then have negative impacts on ecosystems, including loss of species diversity, changes to species composition, changes to habitat quality and changes to water and nutrient cycles.

### *A.3.2 Greenhouse balance*

Three forms of nitrogen have a direct impact on the greenhouse effect (Erisman et al., 2011). First is nitrous oxide (N<sub>2</sub>O), which has a strong global warming potential over a 100-year time scale (GWP<sub>100</sub> close to 265).<sup>9</sup> GWP<sub>100</sub> conversion factors have changed in successive reports of the Intergovernmental Panel on Climate Change. There are various reasons for this, including temperature of the atmosphere and the increasing concentration of the greenhouse gases (GHG) themselves. These conversion factors do not include the indirect effects that N<sub>2</sub>O has on carbon-cycle feedbacks, such as the impact the warming N<sub>2</sub>O causes will have on how much carbon the world's oceans and forests will be able to store in the future. These effects are thought to be large – their inclusion would increase the 2013 conversion factor for N<sub>2</sub>O from 265 to 298 (IPCC, 2013).

Second is nitrogen oxides (NO<sub>x</sub>) emissions which contribute to (i) formation of ground-level ozone (GLO); (ii) a decrease of methane (CH<sub>4</sub>); and, (iii) formation of nitrate aerosols. CH<sub>4</sub> and GLO are, respectively, the 2<sup>nd</sup> and 3<sup>rd</sup> most important GHGs after carbon dioxide (CO<sub>2</sub>).<sup>10</sup> Nitrogen-containing aerosols have a cooling effect, both direct and indirect (through cloud formation).<sup>11</sup> CH<sub>4</sub> has both a warming effect (as a GHG) and a cooling effect (through formation of aerosol in the troposphere). The net effect of all three NO<sub>x</sub>-related contributions is cooling. Third is ammonia (NH<sub>3</sub>) emissions which contribute to aerosol formation and a cooling effect.

Nitrogen also impacts indirectly on climate change (Erisman et al., 2011). First, microbial ammonification of dissolved organic nitrogen (DON) in soils (litter decomposition) contributes to soil respiration and hence CO<sub>2</sub> emissions. Second, nitrogen increases plant productivity and hence CO<sub>2</sub> uptake in terrestrial ecosystems, except in situations where it accelerates organic matter breakdown, thereby increasing release of CO<sub>2</sub>. Third, nitrogen increases marine productivity and hence CO<sub>2</sub> uptake in oceans, except in situations where it causes ocean acidification. Fourth, GLO reduces plant productivity, and hence CO<sub>2</sub> uptake by plants.

If considered in all its forms, and not only N<sub>2</sub>O, and in all its direct and indirect effects, Erisman et al, 2011 estimated that there was no significant net effect of nitrogen on overall radiative balance. However, this estimate does not take into account the fact that the effects of NO<sub>x</sub> and NH<sub>3</sub> have a relatively short life span whereas N<sub>2</sub>O persists in the atmosphere for about a century. Measures that reduce short-lived aerosols but do not address long-lived N<sub>2</sub>O would, for example, increase the net warming effects of nitrogen.

### *A.3.3 Water quality*

Anthropogenic increase of nitrogen in water poses direct threats to human and aquatic ecosystems (fresh and marine waters). High nitrate/nitrite (NO<sub>3</sub><sup>-</sup>/NO<sub>2</sub><sup>-</sup>) levels in drinking water may cause a potentially fatal blood disorder in infants under six months of age called methemoglobinemia or "blue-baby" syndrome. With this disorder there is a reduction in the oxygen carrying capacity of blood,

which can cause shortness of breath and a blueness of the skin of infants or even lead to the infant's death.

The US Environmental Protection Agency (USEPA) has not mandated a Maximum Contaminant Level (MCL) for ammonia (NH<sub>3</sub>). However, it has been known, since early in this century, that NH<sub>3</sub> is toxic to fish and that the toxicity increases with increasing pH and temperature of the water (USEPA, 2013b). When NH<sub>3</sub> is present in water at high enough levels, it is difficult for aquatic organisms to sufficiently excrete the toxicant, leading to toxic build-up in internal tissues and blood, and potentially death. In 2013, USEPA has issued a Final Ammonia Criteria for the toxic effect of NH<sub>3</sub> for aquatic life.

In aquatic ecosystems, nutrient enrichment (eutrophication) is responsible for algal blooms (including toxic algal blooms<sup>12</sup>) on the surface. This can lead to the reduction or even the disappearance of oxygen and thus of fish in the deep waters that become “dead zones”.

Research continues to reveal nitrogen-related impacts on health and aquatic ecosystems. For example, Zhang et al., 2015 found a link between the proliferation of toxic blue-green algae and the risk of death from liver disease. Another example relates to coral. Corals are adapted to thrive in the sun-lit, nutrient-poor waters of tropical oceans<sup>13</sup> thanks to their intimate relationship with microscopic algae and nitrogen-fixing microbes (called diazotrophs). Photosynthesis of algae provides the coral animal with a source of carbon and energy, while diazotrophs provide nitrogen for metabolism and growth. In this relationship, corals regulate the algal growth by limiting their access to nitrogen (Pogoreutz et al., 2017). However, sugar-enriched discharges in coastal waters (e.g. from wastewater) feed the diazotrophic activity, which means they fix more nitrogen. This excess nitrogen available for algae causes the breakdown of coral-algae symbiosis and triggers bleaching (ibid).<sup>14</sup>

#### *A.3.4 Ecosystems and biodiversity*

Species and communities most sensitive to chronically elevated nitrogen deposition are those that are adapted to low nutrient levels, or are poorly buffered against acidification.<sup>15</sup> In the United Kingdom, nitrogen affects many threatened vascular plant, bryophyte and lichen species as well as certain fungal groups (Plantlife and Plant Link UK, 2017). Early evidence also suggests that habitat changes resulting from nitrogen deposition may also affect other taxonomic groups such as insects and birds, although further research is required (ibid). In addition, after an initial increase, the productivity of forests and grasslands is reduced beyond a certain threshold of nitrogen loading.

Hernández et al., 2016 showed that 78 of the 1 400 species of invertebrates, vertebrates and plants listed under the US Endangered Species Act (approximately 6%) were affected by the direct toxicity of nitrogen; eutrophication of their habitat or the spread of non-native plant species. Simkin et al., 2016 found that nitrogen deposition exceeded critical loads for loss of plant species richness in 24% of the 15 136 sites they examined in the United States. Those sites included woodland and grassland sites across the United States. They found that the effects of nitrogen deposition on plant species richness was more pronounced in acidic versus neutral or basic soil and in dry versus wet climates.

SRU, 2015 reveals that in 2009, 48% of Germany's natural and semi-natural terrestrial ecosystems were affected by eutrophication and 8% by acidification.

Jones et al., 2014 estimated the impact of declines in nitrogen deposition on the value of six ecosystem services in the United Kingdom: two provisioning (timber and grassland production), two regulating (CO<sub>2</sub> sequestration and reduction of N<sub>2</sub>O emissions) and two cultural services (recreational fishing and appreciation of biodiversity). They found a net benefit with reduced emissions of N<sub>2</sub>O as a GHG and enhanced cultural services outweighing costs (loss of value) of reduced CO<sub>2</sub> sequestration and provisioning services.

## Notes

- <sup>1</sup> Converting dinitrogen into  $\text{NH}_3/\text{NH}_4^+$  is the role of “nitrogen-fixing bacteria”.
- <sup>2</sup> Decomposition of organic matter is the role of decomposers (“ammonifying bacteria”).
- <sup>3</sup> Converting  $\text{NH}_3/\text{NH}_4^+$  into  $\text{NO}_2^-$  and  $\text{NO}_3^-$  is the role of “nitrifying bacteria”.
- <sup>4</sup> Returning  $\text{NO}_3^-$  back into dinitrogen, thereby closing the nitrogen cycle, is the role of “denitrifying bacteria”. Denitrifying bacteria comprise more than 150 known species and probably hundreds of unknown species.
- <sup>5</sup> Nitrogen oxides ( $\text{NO}_x$ ) include nitric oxide (NO) and nitrogen dioxide ( $\text{NO}_2$ ).
- <sup>6</sup> It was estimated within a range of EUR 75–485 billion per year in the EU-27 (Van Grinsven et al., 2013) and USD 81–441 billion per year in the United States (Sobota et al., 2015).
- <sup>7</sup> For example, Huang et al., 2017 estimated that nitrate and ammonium aerosols accounted for up to a third of  $\text{PM}_{2.5}$  emissions measured in Beijing, Tianjin and Shijiazhuang from June 2014 to April 2015. The  $\text{PM}_{2.5}$  emissions consisted mainly of organic matter (16.0%–25.0%), sulphate aerosol (14.4%–20.5%), nitrate aerosol (15.1%–19.5%), ammonium aerosol (11.6%–13.1%) and mineral dust (14.7%–20.8%). The nitrogenous aerosol formation pathways are detailed in Chapter 1.
- <sup>8</sup> The troposphere starts at Earth's surface and goes up to a height of 7 to 20 km above sea level, depending on latitude and season (and whether it is day or night). Atmospheric layers (i.e. troposphere, stratosphere) have been defined to reflect significant variations in temperature and pressure with altitude.
- <sup>9</sup> This means that one tonne of  $\text{N}_2\text{O}$  emitted this year causes 265 times as much heating over the next hundred years as a tonne of  $\text{CO}_2$ .
- <sup>10</sup> Because of its local and short-lived nature, ground-level ozone (GLO) does not have in itself strong global warming effects, but may have a radiative forcing (warming) effect on regional scales; there are regions of the world where GLO has a radiative forcing up to 150% of  $\text{CO}_2$ . The radiative forcing effect from GLO is strongly height- and latitude-dependent through coupling of GLO change with temperature, water vapour and clouds (Bowman et al., 2013).
- <sup>11</sup> Aerosols in the atmosphere scatter and absorb visible radiation, limiting visibility. They affect the Earth's climate both directly (by scattering and absorbing radiation) and indirectly (by serving as nuclei for cloud formation).
- <sup>12</sup> Zhang et al., 2015 found evidence of blue-green (toxic) algal blooms in 62% of the 3 100 U.S. counties surveyed.
- <sup>13</sup> Much of the world's marine waters lack biologically available nitrogen, which limits photosynthesis in the oceans.
- <sup>14</sup> Bleaching occurs when algae that live inside corals and give them their colour are expelled (e.g. due to increased sea temperatures). Bleached corals continue to live but as the algae provide the coral with up to 90% of its energy, after expelling the algae the coral begins to starve and is subject to mortality.
- <sup>15</sup> Grassland, heathland, peatland, forest, and arctic/montane ecosystems are recognised as vulnerable habitats in Europe; other habitats may be vulnerable but are still poorly studied (Sutton et al., 2011).



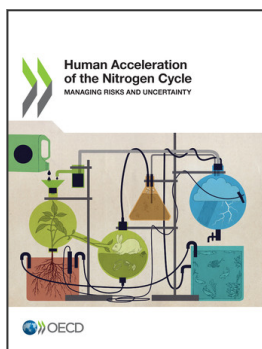
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