

Special Article - Storm

On Nitrogen, Anthropogenic Aerosols, Farmland and Biodiversity Estimation

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Abstract

The complexity of the effects of greenhouse gases and especially nitrogen emission on atmospheric aerosols and nitrogen deposition has been puzzling for decades. Moreover, their effects on public health and biodiversity have increased the notion of urgency for drastic reforms and, as a result, have increased the complexity of the debate. This paper focusses on the interaction between agricultural sources of N-consumption and emissions and other anthropogenic activities as well as on the role of N-deposition on plant physiology and biodiversity impairment. Finally, the urgency of farmland reforms is situated in a socio-economic and historical context.

Keywords: Atmospheric aerosol formation; Anthropogenic effects; Agricultural N-emission and N-deposition; Biodiversity paradigm; Public Health; History of farmland reforms

Introduction

The negative impacts on the Earth's troposphere and biosphere of anthropogenic emissions of greenhouse gases [1-3], of anthropogenic aerosol formation [4,5] and the vertical distribution and degradation of stratospheric ozone [6-8] has been known and is well-documented already for several decades. Various modeling studies have addressed the role of cloud formation, rainfall and meteorological phenomena in the formation, vertical distribution and geographical spread of these chemical substances and of aerosols in particular [9-14]. Also the specific contributions of human traffic, industrial and agricultural activities on the emissions of S- and N-containing chemicals have been extensively estimated or measured at global and local scales, leaving little doubt as to the anthropogenic origin of these effects [15,16]. There is some consensus about the detrimental effects of smog, mineral dust and aerosols on public health [17].

The impact of nitrogen-deposition on (plant) toxicology and pollution also has been demonstrated long ago [18]. Moreover, nitrogen (N) has been designated as the limiting nutrient for many ecosystems, and, as a result, even a small increase in N-supply might in some cases lead to significant negative changes of ecosystem resilience (and biodiversity) [16]. In terrestrial ecosystems, ammonia-derived N may be take up directly through the plant leaves or indirectly through the root as N-rich soil water [16,18]. Indirectly, saturation of terrestrial ecosystems with N leads to a washout of soil nitrate (NO_3^-) and a slow acidification of the soil and loss of other soil-bound ions [16]. As a result, in European rural areas, agriculture-derived N-deposition is designated as the primary source for biodiversity loss [16].

Important questions remain about the role of biogenic emissions in marine environments [19], and the relative contribution of agricultural, industrial and traffic-born emissions on aerosol formation in relation to the weather and/or meteorological conditions.

Most puzzling, however, is the question why the political doctrine

of industrially scaled meat production and the use of N-rich fertilizers, feeding with N-rich livestock fodder and disposal of livestock manure on farmland has been continuously promoted for decades, in countries where the emission and N-deposition were known to be too high. In the words of the special mediator appointed for the nitrogen-crisis in the Netherlands (2022), Johan Remkes, former deputy prime minister of the Netherlands, a radical fifty % reduction of the national livestock, and a complete stop of peak pollutants nearby natural reserves will be inevitable, to prevent the country going into an economic lock-down. It means that either an economic lock, i.e. all other economic activities taking part into nitrogen-emission have to be frozen down to zero, or, a dramatic reduction of all activities related to animal farming is mandatory to 'unlock' the country. The urgency question of farmland reforms will be discussed later in this paper (see 7. The urgency of farmland reforms and political decision-making).

How could the situation come this far?

The deposition of N-rich mineral particles mainly is the result of the following reactions, the uptake and neutralization of gaseous ammonia (NH_3) with sulfuric acid (H_2SO_4) (basically the reaction occurs in two steps) or with nitric acid (HNO_3) [16]:



The reaction with nitric acid is reversible (in contrast to the reaction with sulfuric acid). The atmospheric concentration of nitrogen oxides (NO_x) has a dominant part originating from anthropogenic sources other than agriculture, namely traffic, industry, tourism, etc. Moreover, the formation of nitric oxide in the troposphere largely depends on the presence of ozone (O_3) and radical molecules that originate from O_3 breakdown, NO emission and NO formation. The fraction of NO in the total emission of nitrogen oxides (NO_x) plays a key-role in the oxidation processes that are in favor of the formation of O_3 and HO_x radicals (20) (see 3. Atmospheric aerosol formation

and anthropogenic activities). Water-borne radicals indeed form an very important link in the photo-oxidative destruction of ozone [19].

The deposition of N-rich aerosols thus is largely dependent on the meteorological conditions. An important distinction has to be made between dry and wet deposition [16]. Average deposition data form the usual instruments to calculate the relative contribution of various anthropogenic sources in relation to the total N-deposition [21]. Hereby, the notion of the half-life of aerosol particles and the average deposition radius from the emission source may become important key elements in the analysis of various contributions (see 4. Agricultural NH₃ emission and deposition).

In order to understand the impact of N-emission and deposition on ecosystem biodiversity, it is equally important to understand the mechanisms of ecosystem development and biodiversity resilience (see 5. The biodiversity paradigm). Recent meta-analyses of multi-decadal biodiversity trends in Europe have revealed an amalgam picture of the effect of changing land use and climate change upon the major biodiversity indexes in the European continent [22]. For instance, on the one hand a decreased abundance of terrestrial invertebrates is found, but an increased richness of birds and marine invertebrates. Also a decreased diversity in benthic algae, but increased diversity in birds and aquatic invertebrates, as well as an increased turnover in plants (following an influx of new species) were the result of the meta-analysis [22]. Previously, models to incorporate different taxonomic and heuristic levels of biodiversity analysis have been presented [23,24]. In the present paper, we will discuss the implications of the nitrogen issues (as summarized above) on biodiversity trends and will investigate the key determinants of the urgency paradigm of a potential ecosystem collapse.

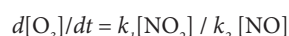
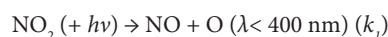
Atmospheric Aerosol Formation and Anthropogenic Activities

In the troposphere, a complex and highly variable interplay has been discovered between the processes causing the physical transport and chemical destruction of ozone (O₃) and the nitrogen oxides and other precursors of O₃-formation [20] (Figure 1). Whereas water (H₂O) and sunlight are important for the photolysis of O₃, resulting in the production of various free radical molecules (like hydroxyl [OH] and peroxy- [HO₂] radicals), these radicals together with nitrogen oxides (NO_x) are determining the production of O₃ too. The resulting production and decay cycles of tropospheric ozone form a multileveled dynamic equilibrium system, in which the ratio of NO₂ to NO concentrations is crucial for the photo-stationary state between NO, NO₂ and O₃ [20]. It appears that there is a sink for ozone (namely the O[¹D] reaction with H₂O) [25] and a source for ozone, resulting from the coupling between HO_x and NO_x pathways [20]. Moreover, radicals of the so-called 'odd hydrogen family' (defined as HO_x = [OH] + [HO₂]) play a key role in the oxidation of SO₂, NO₂, as well as in the breakdown of hydrogen peroxide (H₂O₂) and also in the formation of so-called peroxyacetyl nitrate (PAN). PAN is a secondary pollutant present in photochemical smog, originating from various carbonyl compounds present in polluted air, like acetaldehyde (CH₃CHO) and acetone (CH₃COCH₃) [20]. Interestingly, an important observation made by aircraft observations was the much higher [OH] radical concentrations above clouds compared to clear skies [26,27]. It remains to be investigated whether the importance

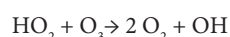
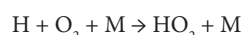
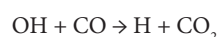
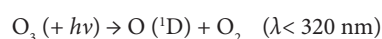
of these photochemical processes will augment in a changing climate with increasing average temperatures and increased water volume transport during precipitation [28].

Schematically, the key chemical reactions are summarized here:

1) regarding the photolysis of nitric oxide and the formation/ degradation of ozone, due to the coupling of the following reactions :

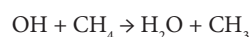


2) regarding the photolysis of ozone in the presence of water, carbon monoxide (CO) and radicals of the 'odd hydrogen family' [20]:



The latter reaction occurs at low NO_x concentrations, whereas in the presence of methane (CH₄) and higher concentrations of NO_x the removal of OH and HO₂ radicals may occur through the following chains of reactions [20]:

3) formation of methane radical and oxidation of this radical to formaldehyde and HO₂:



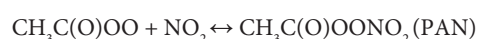
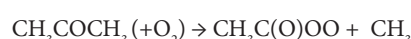
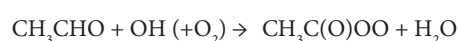
(It has been suggested that the high OH radical concentrations lead to the formation of high O₃ loads during the (wet) summer period, due to the OH-induced destruction of hydrocarbons and resulting reaction chains [16]).

4) formation of hydrogen peroxide and nitric acid:



Reactive nitrogen oxides are an important source for larger aerosol molecules, like peroxyacetyl nitrate (PAN), which can become an additional source for NO₂ through decomposition of PAN, or photolysis and oxidation of HNO₃ [20]:

5) formation of PAN from acetaldehyde (CH₃CHO) or acetone (CH₃COCH₃), through the reaction with oxygen and methane and/or hydroxyl radicals:



The latter equation describes the dynamic equilibrium for the

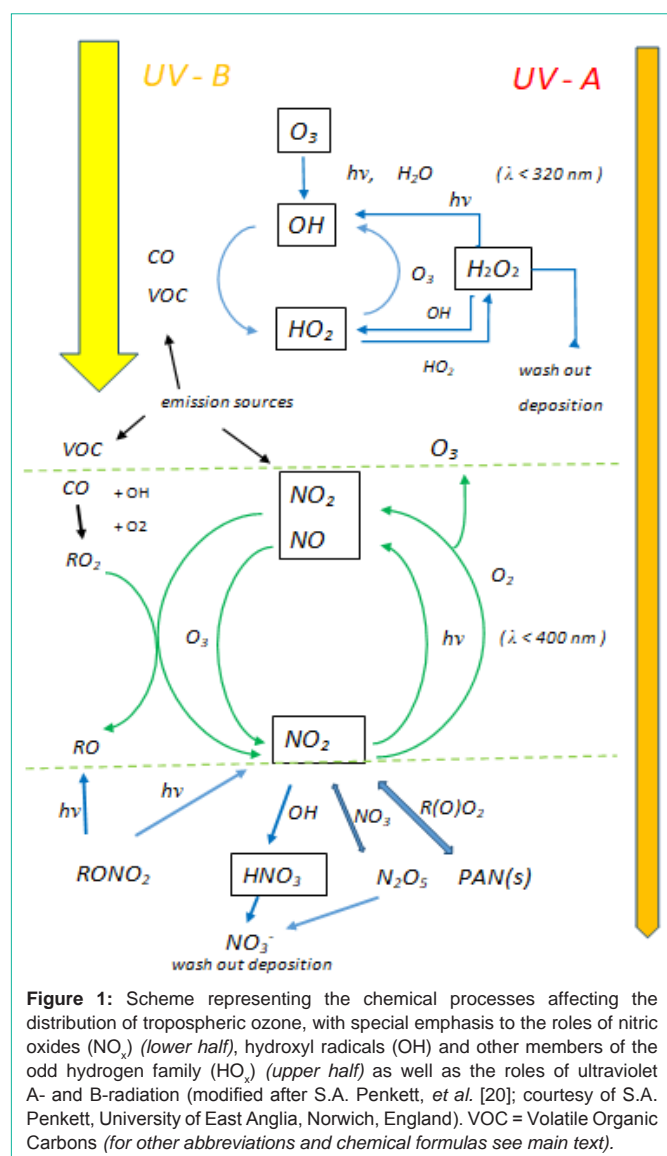


Figure 1: Scheme representing the chemical processes affecting the distribution of tropospheric ozone, with special emphasis to the roles of nitric oxides (NO_x) (lower half), hydroxyl radicals (OH) and other members of the odd hydrogen family (HO_x) (upper half) as well as the roles of ultraviolet A- and B-radiation (modified after S.A. Penkett, et al. [20]; courtesy of S.A. Penkett, University of East Anglia, Norwich, England). VOC = Volatile Organic Carbons (for other abbreviations and chemical formulas see main text).

formation of PAN. There is some similarity with the formation of ammonium nitrate (see 2. Introduction), although in the atmosphere, in the photo-oxidative chemical processes described above, ammonia (NH₃) doesn't take much part, if any at all [20]. In the presence of ozone, NH₃ is completely oxidized to nitrate, at least in wastewater [29]. Meteorological measurements of the vertical distributions of ammonia and related N-gases in aerosols revealed an upward NH₃ flux of 0.12 μgm⁻²s⁻¹ [30]. Moreover, a close correlation between the concentrations of NH₃ and HNO₃ gases was found, based on the concentration product [NH_{3(g)}] × [HNO_{3(g)}] (at temperatures above 0°C and relative humidity below 80 %). However, in other cases, higher NH₃ and/or HNO₃ concentrations in the gas phase were measured, deviating from the theoretically predicted values [30].

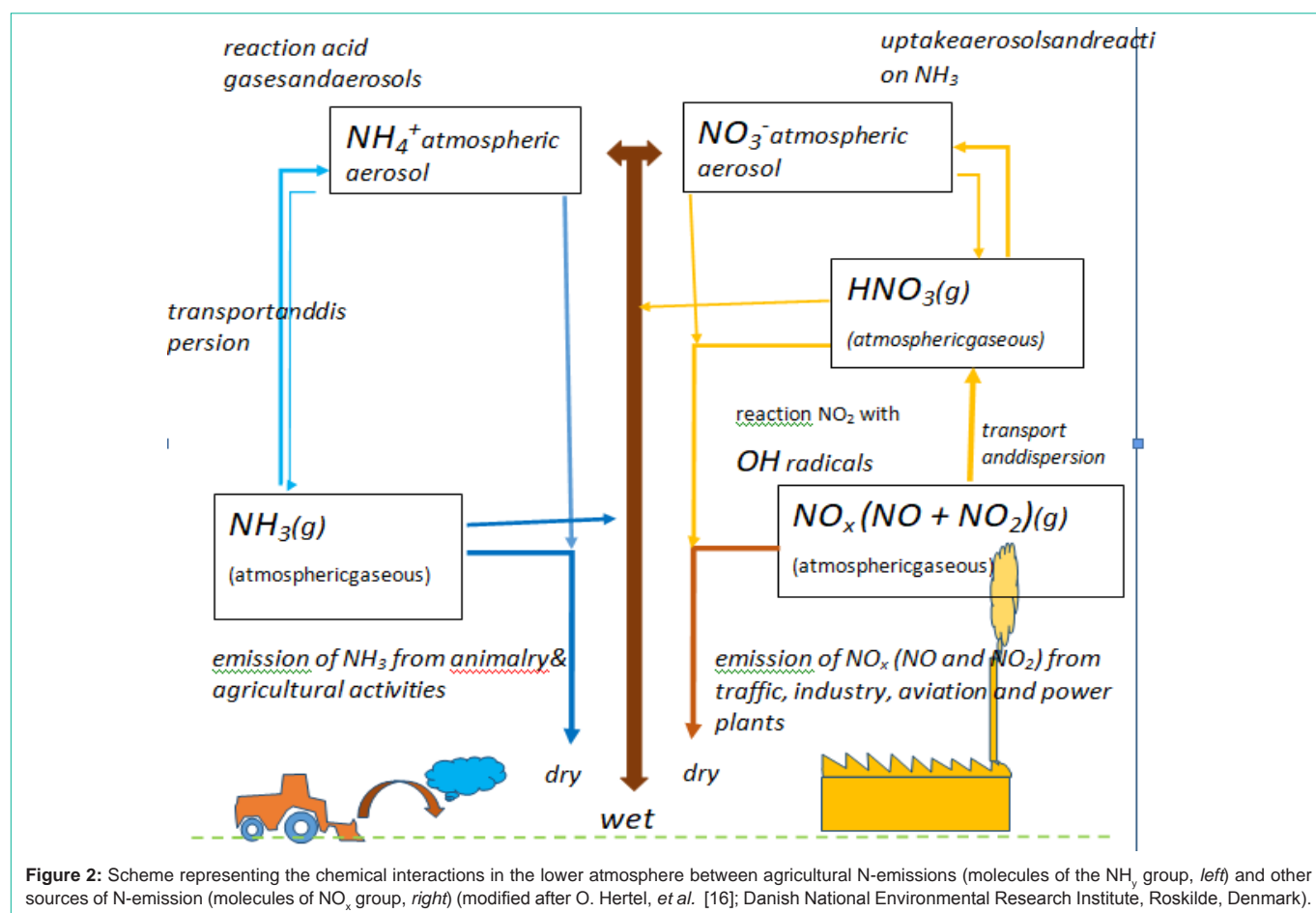
One of the most puzzling phenomena is that although HNO₃ has a much shorter half-life in the atmosphere compared to the half-life of NH₃ (which therefore could be transported over much longer distances), it nevertheless has been estimated that about 50% of the NH₃ emission is deposited within 50 km of the emission source [16].

The multitude of chemical reactions, both of anthropogenic or natural processes, are not the only cause of aerosol formation. The presence of enormous amounts of fine particles, of organic or inorganic source, from concentrated sea salt droplets to desert dust and plant pollen, they all create a diversified array of habitats to generate atmospheric aerosols [31]. Many of these processes are indirectly related to climate warming, to increased water evaporation or directly related to human activities (such as deforestation and desertification). Most of these effects have a negative impact on the climate, although a significant counteractive effect on radiative forcing (through greenhouse gases) has been demonstrated too [32]. As aerosol particles are strongly coupled with the gas-phase chemistry of the atmosphere including the clouds, in order to understand their properties and effects on climate and health, the gases, aerosol particles and clouds are regarded as one continuum, a single system, with scales ranging from molecules and nanometer-sized aerosol particles to frontal cloud systems spanning hundreds of squared kilometers [31]. These systems can react as sinks for reactive species and therefore may decrease or increase radiation-driven photochemical effects [31]. The influence of clouds on OH radical formation, as mentioned above [26,27], may extend as well to the much larger and diverse group of atmospheric chemicals (mentioned as well as others). For reasons of manageability and biological impact, we will hereafter focus on the lower troposphere and the impact of agricultural NH₃ emission and deposition (see 4. Agricultural NH₃ emission and deposition).

Agricultural NH₃ Emission and Deposition

It is important to note that most of the data in the present paragraph are derived from European studies and are best related to the European situation in agricultural and other forms of land use [18]. There is ample evidence that terrestrial NH₃-emission is predominantly derived from agricultural activities and these have (or had) a strong negative effect on plant biodiversity [16,18]. In Europe, there is also quite some seasonal variation in NH₃-emission which obviously follows the seasonal variation in agricultural activities. Field data in Denmark (referring to the period 1989-2003) have revealed a change of the peak emissions in spring and/or summer following the utilization of animal manure for crop fertilization, as a result of changes in the local legislation [16]. Peak NH₃-pollutions that are strongly correlated with animal production farms are reported in several European countries, although these emission data haven't been easily accessible to the public in previous decades.

However, a most important distinction has to be made regarding the dry and wet deposition of N-rich chemicals. Herein, it is obvious that the meteorological conditions play a decisive role too, in the proportion and source of agricultural versus other sources of N-deposition [16] (Figure 2). Whereas, nitrate-derived nitrogen is assimilated in plants, often in synergistic collaboration with fungal mycorrhiza [33], ammonia-derived nitrogen first needs to be nitrified (i.e. oxidized) through specific bacteria and other soil microbial organisms. NH₃ appears to stick to plant leaves and other surfaces. Adverse effects on vegetation occur when the rate of foliar uptake (through leaves) exceeds the capacity for detoxification of living plants [18]. In higher plants NH₃ uptake occurs mostly through the shoots, whereas NH₄⁺ uptake occurs through both shoots and roots



[18].

In the assimilation and detoxification of the couple $\text{NH}_3 \leftrightarrow \text{NH}_4^+$ (shortly denoted as NH_y), a significant role is played by the Glutamine Synthase (GS) – glutamate synthase (GOGAT) circular pathway [34]. Besides this, also glutamate dehydrogenase activity has an important contribution to the regulation of glutamate in higher plants [35]. The GS-COGAT pathway moreover is coupled to the Krebs cycle, through the reaction of 2-oxoglutaric (also known as α -ketoglutaric acid), formed from citric acid in the Krebs cycle, with glutamine resulting in the formation of glutamate (Figure 3). This reaction also depends on the presence of the Nicotinamide-Adenine-Dinucleotide-(Phosphate)-enzyme (NAD(P)H) – Ferredoxin (Fd) enzyme complex, which is present in the chloroplast and root cell plastids of higher plants [18]. As a result, NH_y uptake and assimilation results in enhanced amino acid and protein metabolism. In a number of studies [18], for instance in coniferous trees, increased concentrations of free N-rich amino acids (especially arginine) were found in pine needles [36]. In the latter study, in the coniferous trees studied, also a correlation of high N-availability with the occurrence of the parasitic fungus *Sphaeropsis sapinea* was detected [36]. Other distortions like an altered crop to weed ratio would result from too high NH_y assimilation [18]. When the relation with epiphytic lichens and the air pollution with SO_2 is considered, it appears that ‘nitrophytic’ lichens that are more sensitive to SO_2 , respond more rapidly than others to sudden decreases in SO_2 and consequently nitrophytic species may

colonize new sites more rapidly [37]. N-deposition therefore may interfere with the species balance in ecosystems and consequently may have an impact on biodiversity (see 5. The biodiversity paradigm).

Taking into account the adverse effects of NH_3 -assimilation, including the complex mechanisms of N-saturation through NH_4^+ and NO_3^- retention and/or NO_3^- washout from forest soils, studied in North-American [38,39] and European forests [40], a critical load for total N-deposition was repeatedly suggested [41]. Such critical N-deposition loads have been adopted at a *Convention on Long-Range Transboundary Air Pollution* [18,42]. It was suggested that a critical load of 5-10 $\text{kg ha}^{-1} \text{yr}^{-1}$ of total N-deposition (both dry and wet deposition combined of all atmospheric N substances) would be sufficient to protect the most vulnerable terrestrial ecosystems [18]. In forests, a somewhat higher critical load of 10-20 $\text{kg ha}^{-1} \text{yr}^{-1}$, depending on the soil conditions, was estimated (depending on the N-saturation and competition for N-availability in the tree species) [18].

Meanwhile in the Netherlands, the total N-deposition has decreased from above 2700 mol N/ha in 1990 to about 1500 mol N/ha in 2010 [43]. Between 2010 and 2020, however, the N-deposition showed no further decline, due to the lack of further lowering of the reduced (NH_y -derived) nitrogen from agricultural activities [43]. The oxidized (NO_x -derived) nitrogen on the other hand, showed a prolonged steady decline, also as a result of the use of better NO_x -

Effects on Human Health

In addition to the biodiversity impairment, the effects of N-deposition and anthropogenic aerosols on human well-being and health should not be under-estimated [52]. A lot of progress has been made in medical diagnosis and monitoring, since the first discoveries of serious health impairment due to polluted air inhalation in smog areas [53], already long before the Great Smog of London in 1952, when some 4000 deaths were counted in only one week [54]. In the past decades however, the problem of air pollution has travelled from the London city to the megacities and densely populated, industrialized and heavy traffic zones of the globe, especially in the developing countries and in China [55,56]. Air pollution was found to be the cause and established aggravating factor of a number of respiratory diseases like COPD, asthma [57,58] and lung cancer [59,60].

The list of detrimental health effects accompanying the industrialized meat production and meat consumptive habits has been extended recently, including the food contamination with dioxins and PCBs (polychlorinated biphenyls), micro-plastics, et cetera. However, one may ask whether such an orchestrated action against animal farming is really accountable to the agricultural activities and the contribution thereof to the environmental problems. On the other hand, the aggravating effects of a weakened respiratory system has proven to dramatically augment the morbidity in large groups of the population, during the recent corona pandemic(s) [61]. The pandemonium of health effects therefore emphasize the urgency of measures to counteract the nitrogen emission and aerosol pollution all the more.

The Urgency of Farmland Reforms and Political Decision-Making

It is well documented that the Netherlands occupy a frontrunner position in Europe, with respect to the consumption of nitrogen in agriculture, namely about $166 \text{ kg ha}^{-1} \text{ yr}^{-1}$, versus an average N-consumption of $48 \text{ kg ha}^{-1} \text{ yr}^{-1}$ in Europe (reference year 2019) [62]. These amounts of N-consumption are mainly due to the use of N-rich fodder and N-rich fertilization of farmland of which - again (!) - about 58 % is livestock-derived manure, the other part being derived from fertilizers [63]. From this yearly total N-load of about 669 million kg for the whole of the Netherlands (including some 18 million kg N-deposited from air)(reference year 2020), roughly 13,2% or 88 million kg nitrogen yr^{-1} is lost to the air, where as 32,7% is lost to the soil (corresponding to a national total of 307 million kg yr^{-1}) [63].

These problems aren't new, and the recent national farmer protests and upheaval of certain protest groups may seem strange to the fellow inhabitants of the world. But these reactions are not so strange, if they are put in a political and historical context. Not fortuitously, it was a Dutch minister of agriculture, Sicco Mansholt (1908-1995), who proposed the European Commission in 1952 an ambitious plan to re-organize European agriculture [64]. Mansholt became the first Commissioner for agriculture in the European Commission in 1958. The *Mansholt-plan* consisted of harmonizing the conditions of agricultural production, the settling of commercial prices (and fixing minimum prices to protect the farmer's income) and the regulation of import, and export of agricultural products [64].

The fact that the Netherlands became a very 'successful' export nation for both meat and crop products, was not in the least a by-product of the exploitation - since the early 1960s - of gas resources resulting in the availability of 'cheap' hydrogen. This hydrogen also became available for industrialized ammonia and ammonium-nitrate production for the fertilizer market. The discoveries of chemical techniques for ammonia and fertilizer production by the Germans Justus von Liebig (1803-1873), Fritz Haber (1868-1934) and Carl Bosch (1874-1940) (the so-called Haber-Bosch process for NH_3 production) in the early twentieth century, had enabled these chemical and economic breakthroughs in a previous era. However, the negative fame of these chemists, mainly due to their association with the chemical industries involved in toxic gas production, did not prevent the Nobel committee to award to Haber the Nobel prize for chemistry in 1918. According to some historical sources, both von Liebig and Mansholt afterwards deplored the impact of their chemical discoveries/agricultural reforms, respectively. Von Liebig warned against the large scale usage of chemical fertilizers already in 1861 [65]. Mansholt allegedly was also very disappointed, but the agricultural reforms would become irreversible, possibly due to the influential pressure of agricultural lobbyists.

It is obvious that the present conditions of 2022, both politically as regarding the 'cheap' exploitation of gas reserves in the Groningen area [66], have come to an immanent turning point. The largest gas reserves on the European continent, allegedly some 300 billion cubic meters [67], no longer are regarded as protecting the Dutch welfare state economy, since the gas exploitations are also causing billions of damages to housing, due to the numerous earthquakes in the Groningen area [66]. Within these socio-economic and environmental contexts, the recent political adaptations are more easily apprehended, but not at all justified.

Political adaptations require decisive choices to be made, but sometimes it is rather about avoiding the wrong choices. The choice of augmenting the conditions for food production in the postwar rural areas of Europe, e.g. given the limited availability of phosphorus in Dutch peaty soils [47], apparently is an illustrative example given the present N-crisis. Re-allotment of farmland was considered a necessary step in consolidating the up-scaling of agricultural activities, following the Mansholt reforms. But it also resulted in a dramatic loss of linear landscape elements and the impairment of biodiversity hot spots. The risks of forced collectivization of private farms, as experienced in other regions of the world before, were also mentioned by Mansholt in his Paris speech [64]. Replacing agricultural activities by other economic activities that could augment N-deposition, is understandable for the those who are eager to unlock the economy (see 2. Introduction). However, it can hardly be called a solution for the nitrogen crisis to replace one nitrogen emission source for another.

The urgency of the deterioration of the environmental conditions and the overall biodiversity decline (see also 5. The biodiversity paradigm) may represent a staggering contrast with the effective speed of reform measures in society. As shown above, the scientific studies that documented the negative effects of N-emission on atmospheric aerosol formation and N-deposition have been published several decades ago. Also conferences, discussions, declarations and manifestos have been put on paper already in the previous century, but still are awaiting the desired effect. In this respect it is interesting

to note a trade-off between the adaptation speed (S_{adapt}) and the costs of behavioral change ($P_{\Delta behavior}$) in the social groups concerned, resulting in the following formula for a human Resilience Strength Equation (huRSE):

$$huRSE = k(x, y) \cdot S_{adapt} \cdot P_{\Delta behavior}$$

Although the costs of behavioral changes are difficult to estimate, we may grasp its meaning by comparing the discourses of individuals or different groups in a socio-economic context. For a distant observer, a conversational phrase (while reading the morning newspaper) such as “Oh dear dear, .” (pouring himself another cup of coffee) could become the only noticeable difference. For another, it might be another political U-turn or the end of a career. Still, for the countless who see their investments and ambitions melting down or vanish in smoke, it might become all too serious. Farmers are known to be familiar with crop rotation (on a yearly basis), but real occupational transitions of a population of farmers are mostly characterized by the time span of a generation (i.e. several decades). It is tempting to investigate the role of GDP and population size, density and demographic structure on the latter equation constant ($k[x, y]$), but obviously this falls outside the scope of the present paper.

Conclusions

In scientific studies, the impending risks of greenhouse gas emissions were already reported more than half a century ago. This year 2022, the Rio de Janeiro conference on biological diversity ‘celebrated’ a thirty years anniversary. But there isn’t much enthusiasm for an anniversary party. In the first decades after the publication of the *Club of Rome’s* report [68], the main emphasis was laid on the environmental effects of greenhouse gases, on air pollution, acid rain as well as the effects on atmospheric ozone. In the recent decades, the effects of greenhouse gases on climate warming, as well as the alarming rates of biodiversity decline (especially due to global deforestation) came to the forefront. Although the role of nitrogen emission (both the oxidized forms and the ammonia-derived forms) in countries of the moderate climate zone were also known for thirty years (or more), so far effective measures to reduce agricultural nitrogen emission have been lacking in certain countries.

In this review paper, due attention is paid to both the complexity of nitrogen emission and interaction with atmospheric aerosols and the complexity of biodiversity estimation. It seems accurate to conclude that especially the nitrogen-enrichment of the rural environment, due to intensive agricultural activities aiming at meat production, poses an unacceptable load on the viability of plant communities and biodiversity nearby the emission sources. Biodiversity impairment in natural reserves, however, may not become reversible following the replacement of one nitrogen source for another. Moreover, the necessity of farmland reforms seems at conflict with the desired speed of behavioral changes in the population envisaged.

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