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Manure Acidification and Air Cleaners for Ammonia Abatement: A Holistic Assessment of the Costs and Effects on Terrestrial, Freshwater and Marine Ecosystems

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Abstract: Manure acidification has been introduced as an abatement to reduce ammonia (NH_3) emissions to improve air quality and protect terrestrial and aquatic environments from nitrogen deposition. A successful regulation of NH_3 emissions using manure acidification might, however, result in increased nitrogen leaching from fertilized fields with adverse effects on freshwater and marine ecosystems, if the overall fertilizer application rate in the fields is not adjusted according to the increased fertilizer value of the manure. We apply a holistic model framework encapsulating all important environmental compartments to assess the ecological and economic consequences of a specific agricultural practice or a combination of these. The results show that manure acidification combined with air cleaners reduces NH_3 emission and atmospheric nitrogen deposition with substantial positive effects on the terrestrial environment. Although manure acidification results in a slight increase in total nitrogen input into freshwater and marine ecosystems, the subsequent increase in chlorophyll *a* concentration and decrease in water transparency is insignificant. Hence, according to the model results, manure acidification will improve terrestrial nature quality, with no significant adverse effects on the aquatic environments.

Keywords: agricultural practice; manure; mitigation methods; pollution swapping; atmospheric deposition; environmental effects; eutrophication; cost efficiency

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1. Introduction

Since the 1950s, anthropogenic activities have altered the nitrogen (N) cycle significantly. The global anthropogenic production and use of reactive nitrogen (Nr) is now approximately three-fold higher than the production caused by natural terrestrial processes [1]. The increased anthropogenic use of Nr is related to negative impacts on the environment, including natural terrestrial habitats and aquatic ecosystems, and human health (as reviewed by [2]). Reactive nitrogen is typically emitted as the chemical forms of reduced nitrogen (ammonia (NH_3) and ammonium (NH_4^+)) and oxidized nitrogen (nitrogen oxides (NO_x), nitrate (NO_3^-) and others). The many forms can interreact and transform and have a cascade of effects through different compartments of the environment [3,4], where it is continuously accumulating [5]. In the atmospheric compartment, Nr contributes to the formation of particulate matter and adds thereby to negative impacts on human health [6–8]. After removal from the atmosphere (by dry or wet deposition), Nr is a major driver of biodiversity loss and can lead to reduced resilience of terrestrial ecosystems [9,10]. Freshwater and marine environments also receive nitrogen from the atmosphere, but mainly through run-off from land, causing large problems with eutrophication leading to algae blooms, decreased water transparency and oxygen deficiency [11–14] (. The costs of reducing the N loads to terrestrial and aquatic environments have increased with more extensive load reduction requirements [15], but assessments indicate that the benefits exceed the costs [16,17].

The agricultural sector is a major contributor to the emission of Nr into the atmosphere. At the European Union scale (EU-28), the total NH_3 emission in 2018 was 3853 kt NH_3 [18], of which 93% (3578 kt) had its origin in the agricultural sector. Only moderate reductions have been obtained for NH_3 within EU-28 in the last decades, compared to other important air pollutants like SO_x and NO_x. Reduction targets for NH_3 have therefore been set for many European countries [19], which urge the agricultural sector to apply new technologies that minimize the emissions related, e.g., to manure handling. The total NH_3 from animal manure from EU-28 can be estimated from the individual countries' submissions to the LRTAP Convention [20] to have been about 2809 kt NH_3 in 2018. Of this, only 183 kt NH_3 was due to urine and manure deposited directly during grazing. The remaining 2625 kt NH_3 came from emissions from animal manure in livestock buildings, which thus constitutes about 68% of total EU-28 emissions. This share can be technically manipulated in a number of different ways. This includes technologies that can be both simple, such as coverage of manure stores and fast incorporation of field-applied manure, to very advanced air-scrubbers or liquid manure injection into the soil.

Agriculture is an important sector in Denmark and more than 60% of the land is used for farming [21]. Through the years, a number of policy action plans for protecting the terrestrial, marine and freshwater environments have been implemented [21]. To support that, research related to effects, abatement technologies, emission factors and the transformation/transport in the atmosphere and hydrosphere has been carried out for many years and has supported the use of cost-effective measures [22,23].

Acidification of manure slurry has been recognized as an effective NH_3 emission abatement technology and has been used by farmers in Denmark for a number of years [24–26]. This method reduces the formation and evaporation of NH_3, and Nr is kept in the manure as NH_4^+. Sulfuric acid (H_2SO_4) is normally used to acidify the manure and approximately 6–12 kg of concentrated H_2SO_4 per m^3 slurry is required to reach a pH of max 6 in the manure [25,27]. Based on a meta-analysis including abatement methods related to slurry treatment, storage and application, acidification was found to be the most effective [28]. Manure acidification can take place in different ways: (1) in the barn (barn acidification), (2) in the storage tank (storage acidification) and (3) at the slurry tanker during field application (field acidification). All three technologies are offered today by different companies and approximately 10–12% of the slurry spread on fields in Denmark in 2017 was acidified [29].

Although manure acidification is known to be an efficient abatement measure for reducing NH_3 emission into the atmosphere, potentially leading to a reduced N deposition, the acidification technology could also result in increased leaching of Nr from fertilized fields, with adverse environmental effects on marine and freshwater systems. Hence, successful management and regulation of NH_3 emissions might lead to pollution swapping [30] and result in an increase in solute Nr forms polluting the aquatic environment. The risk of unintended pollution swapping can be investigated by applying integrated and holistic management approaches. By following the fate of Nr in detail, a holistic approach covering the flow between the main environmental compartments can ensure that an improvement in one compartment is not counteracted by deterioration in another compartment.

We therefore applied a holistic methodology of a coupled model system to test the following hypothesis.

Manure acidification in the barn, combined with air cleaners, reduces NH_3 emissions into the atmosphere, resulting in reduced Nr deposition with positive effects on terrestrial ecosystems. This could, however, potentially lead to increased Nr leaching from fertilized soils, resulting in increased riverine Nr loading with subsequent negative effects on marine and freshwater systems.

The hypothesis will be tested by applying the DEEMON (**D**ynamic **E**cological and **E**conomic **Mo**dels of **N**utrients) system for a selected area in Denmark.

2. Material and Methods

The DEEMON model system is based on existing state-of-the-art models of Nr dynamics in relevant environmental compartments, combined and customized to provide insights into the ecological and economic consequences of different management strategies related to Nr regulation of the agricultural sector. An overview of the system and the flows between the compartments are shown in Figure 1. In the following sections, the study area and the emission scenarios are described. Then the setup of the different models (DEHM, SWAT, GOTM-FABM-PCLake and PLS), as well as the methods for quantifying the resulting effects, are described.

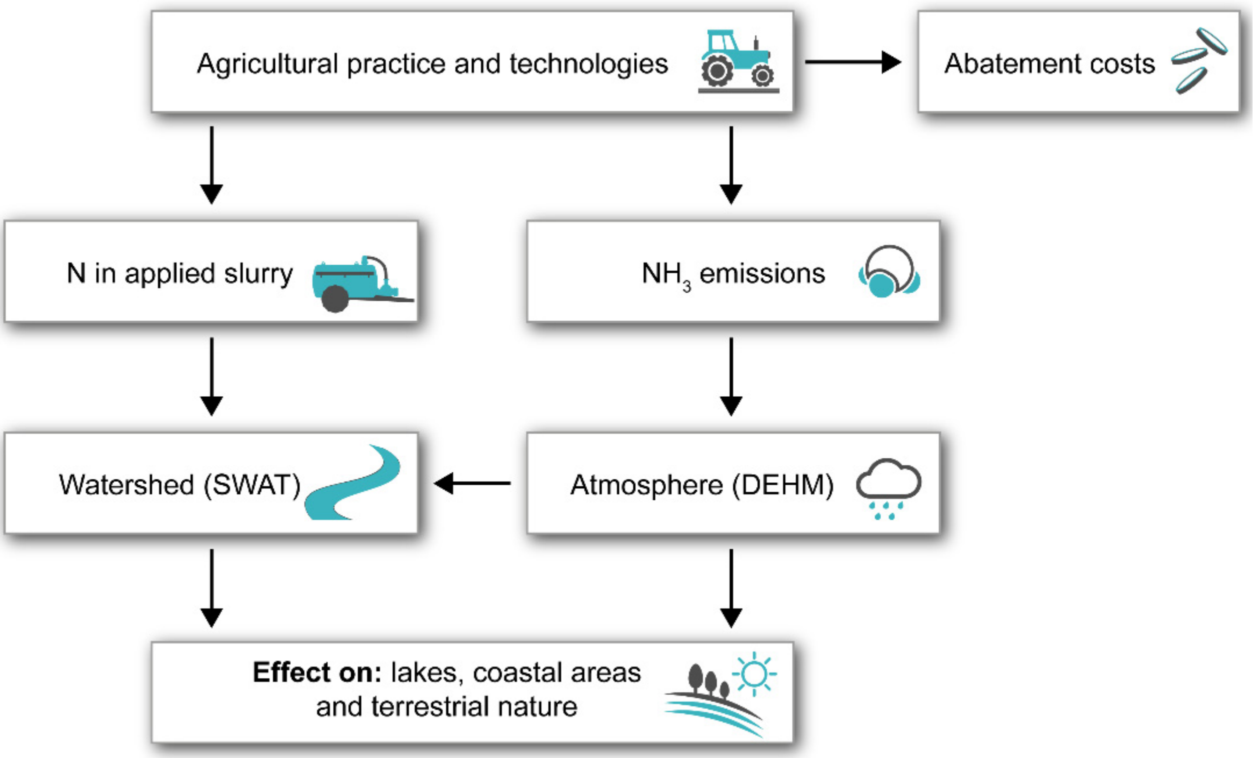


Figure 1. A schematic overview of the DEEMON model system linking the main compartments of the environment. The atmosphere compartment is described by the DEHM model; the watershed is described by the SWAT model, while the aquatic areas are described by the GOTM-FABM-PCLake and PLS models. Quantification of exceedances of critical loads were used to study the effects on terrestrial nature, and the economic consequences of solutions are estimated as changes in abatement costs for Nr reductions into air and the aquatic environment.

2.1. Study Area

The model system has been set up for an area in the northern part of Denmark. The area covers the Limfjord—a 180 km long branched sound connecting the North Sea and Kattegat—and the associated catchment areas. The study area has been chosen as it covers an intensive and important agricultural region of Denmark, as well as Limfjorden, the largest estuary in Denmark, important for several bird species, marine mammals and for the Danish shellfish industry (Figure 2).

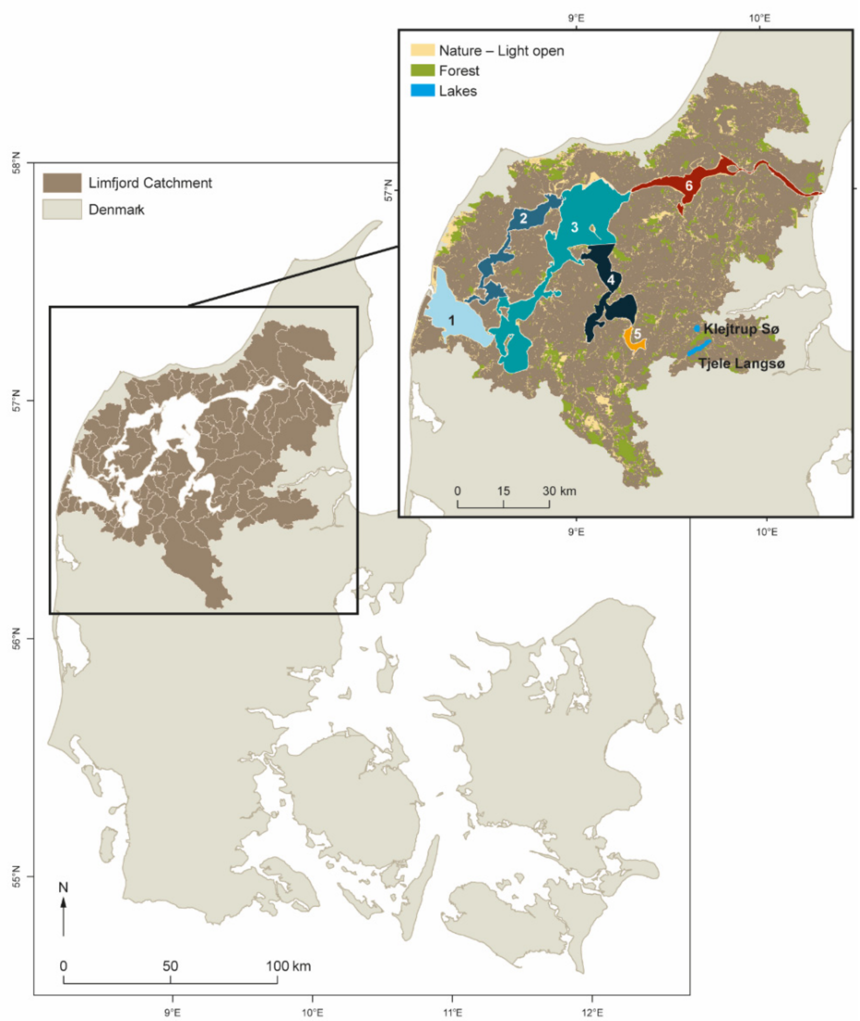


Figure 2. A map of Denmark and the location of the Limfjord with its catchment area divided into 118 sub-basins constituting the study area. The catchment area contains the two lakes Klejstrup and Tjele Langsø, as well as large terrestrial nature areas. The zoom to the right shows the six marine areas of the Limfjord in different colors: 1. Nissum Bredning, 2. Thisted Bredning, 3. Løgstør Bredning, 4. Skive Fjord–Lovns bredning–Riisgårde Bredning, 5. Hjarbæk Fjord and 6. Nibe Bredning.

Limfjorden is a complex shallow sound (mean depth of about 5 m) with numerous branches, broads, islands and sills and a total area of 1500 km^2. Limfjorden is eutrophic because of the input of nutrients from the catchment area, as well as from the North Sea and the atmosphere [31]. The phytoplankton concentration is high and seasonal eutrophication-induced oxygen depletion events occur every year, especially in the middle part of the fjord (Figure 2, basin 2 and 4). The catchment area is about 7.600 km^2, of which more than 65% is intensively farmed with a livestock density of 1.3 livestock/ha, which is higher than the average for Denmark [32,33]. Mean annual (1990–2019) precipitation (corrected for gauge under catch) is approximately 883 mm y^{-1}, with no pronounced seasonality [34]. The catchment has a mean annual (1990–2019) runoff of about 344 mm y^{-1} and the mean annual nitrogen load to the Limfjord for the years 2010–2019 is estimated to be about 12.500 ton-N y^{-1} [35]. Although the Limfjorden catchment area is intensively farmed, about 624 km^2 of valuable and protected terrestrial nature and about 246 km^2 of lakes are located within the catchment area. The catchment has 180 km² salt meadows, 94 km² meadows, 196 km² semi-natural dry grasslands, 136 km² heathland and 29 km² forest protected under the Habitat Directive. These areas have a high biodiversity of both flora and fauna, including a variety of migrating and stationary bird species. Several streams and small lakes are located in the catchment area, including lakes Klejtrup and Tjele Langsø, two typical Danish lakes with a mean depth of 2.7 m and 17 m, respectively. Both lakes receive nutrients from the catchment, as well as from the atmosphere, and drain into the Limfjord. Like most other Danish lakes, Klejtrup and Tjele Langsø are eutrophic with high phytoplankton concentrations, occasional cyanobacteria blooms and low water transparency; especially during the growth season. During summer, the deep Tjele Langsø also experiences an annually reoccurring period of oxygen depletion where the lake is thermally stratified.

2.2. Baseline and Emission Scenario

A baseline and an emission scenario have been set up and details of the data and methods applied are given below. The main characteristics of the scenarios are:

- The baseline scenario reflects the actual agricultural situation and practice in Denmark in 2012. This includes type, size and location of animal houses and storage facilities, field location, crop types and manure application, as well as the actual NH_3 emission and N content in manure.
- The emission scenario assumes the same conditions as in the baseline scenario, but with:
    1. Barn acidification of all cattle and pig manure slurry at all Danish farms regardless of size of the farm. It is assumed that the barn acidification will have a full effect on the subsequent NH_3 emission from storage and from manure application.
    2. Air cleaning at all poultry farms in Denmark.

2.2.1. Manure Production and NH_3 Emission from Animal Houses and from Field Application (Baseline Scenario)

NH_3 Emission from Animal Houses and Storage Facilities

Data on manure production (rate, type and location), nitrogen excretion and NH_3 emission used in the baseline scenario are based on the farmers' nitrogen accounting for 2012, which includes 97–99% of the total amount of nitrogen in animal manure in Denmark. The farmers' reporting on nitrogen excretion rates is generally based on the normative Danish nitrogen accounting system [36] and corrections by the individual farmers to their own feeding and production efficacy rates. In the current setup, the estimated emission from housing and storage was 34.4 kt NH_3-N for 2012.

NH_3 Emission from Field Application

NH_3 emissions from field applications are based on the emission model described in [37]. This model includes knowledge of each field: crop and location and the farmer's nitrogen accounting for the amount of nitrogen in animal manure available on that specific farm (production minus export and plus import). The NH_3 emission from each field differs depending on the crop. The model encompasses six different crop types and five application times. The emission factors are estimated with ALFAM 1.0 [38] combined with new measurements included in the ALFAM2 model [39] corrected for actual temperatures during manure application combined with expert judgement. The emission factors (EFs) for acidified manure have been adjusted to a general value of 50% for untreated manure. The currently used emission factors for slurry application range between 0.85 and 17.8% of total N depending on crop type and time of manure application [40]. The total NH_3 emission from manure application in Denmark amounts to 12.7 kt NH_3-N in the current study for 2012.

2.2.2. Effects of Acidification and Air Cleaning on NH_3 Emission (Emission Scenario)

The emission scenario is constructed from the baseline scenario by applying documented effects (relative reduction factors) of acidification and air cleaners on NH_3 emission rates from barns and fields, as well as effects (relative increase) of acidification on nitrogen content in the manure.

Effects of Barn Measures on NH_3 Emission

It is assumed that barn acidification reduces NH_3 emissions by 65% from the barn and storage facility and during field application (Table 1). The acidification is assumed to take place inside the animal houses. Thereby, the ammonia emissions from the animal houses and storage facilities and from manure applied in the field are lowered.

Table 1. Relative reduction factors in NH_3 emission that are due to barn acidification and air cleaners compared to untreated barns in the baseline scenario. Source: [8,41].

Source of Emission	Measure	Animal Houses
Cattle	Barn acidification	65%
Pigs	Barn acidification	65%
Poultry (full-time housed)	Air cleaners	75%
Poultry (free range)	Air cleaners	37.5%

A number of biological air purification tests have been conducted on different biological air cleaners in Denmark [42,43] showing results of reduction efficiency on ammonia emission from 94% to 77%. In our scenario, air cleaners are installed in all poultry houses with an assumed efficiency of 75% compared to barns with no air cleaners (Table 1).

Effects of Acidification on Nitrogen Content in Manure

Manure acidification not only results in a reduced ammonia emissions but also in an increased content of nitrogen within manure available for field application. As the input data for the current situation (baseline scenario) are actual farm data describing the amount of nitrogen in the manure before field application, the amount of nitrogen in manure in the scenario has been increased with the avoided ammonia emission. This increases the fertilizer value of the manure (Table 2). As solid manure cannot be acidified, changes in the ammonia emission from solid manure and deep litter are not included in the scenario.

Table 2. Change in nitrogen content in animal manure that are due to acidification relative to untreated manure slurry in the baseline scenario.

Manure Source	Change in Nitrogen Content
Cattle	110%
Pig	114%
Mixture cattle and pig	112%

2.2.3. Summarizing Input Data Used in Baseline and Emission Scenario

The data characterizing the baseline and emission scenario, including data on type and amount of manure, ammonia emission from barns and manure storage facilities and nitrogen in manure applied to soil, as well as the effects of manure acidification and air cleaners, are summarized in Table 3. In the table, the input values for both scenarios are aggregated to national scale; however, the emissions cover all of Denmark with a special resolution of 1 km^2 in the further calculations.

Table 3. Total yearly nitrogen from all animals in Denmark in manure and ammonia emission used in the baseline and emission scenario. Based on 2012 data.

	Baseline * Scenario [kt N]	Emission * Scenario [kt N]
National manure production incl. deposited during grazing	261.2	261.2
NH_3 emission, houses and storage [as NH_3-N]	34.4	13.7
N applied to soil, excl. grazing	205.0	226.4
NH_3 emission, manure application [as NH_3-N]	12.7	9.4
N available for plants	192.3	216.9

* Values for the baseline scenario are based on data from 2012 and values for the emission scenario are calculated from the baseline values using experimental determined changes in emission factors that are due to manure acidification and air cleaners, as described above.

2.3. Atmospheric Transport and Deposition

The Danish Eulerian hemispheric model (DEHM) is a chemistry-transport model describing the fate of air pollution in the atmosphere [44–46]. The model covers the Northern Hemisphere and includes several nests with higher resolution over specific regions. In the current setup, the area of Denmark is covered by a horizontal grid with a resolution of 5.56 km × 5.56 km. The model describes the chemical transformation, transport and removal of 67 different chemical species (gases and aerosols). Details on the driving meteorology, emissions and evaluation of the model are given in the supplementary. The anthropogenic emissions for Denmark are based on a detailed inventory on a 1 km × 1 km resolution grid based on the SPREAD (spatial distribution of emissions to air) model [47,48] and the temporal variation of the NH_3 emissions from the agricultural area is based on a parameterization describing the impact of meteorology, regulation, production methods, etc. on the hourly emission [49,50].

In the current study two one-year simulations have been carried out:

- The baseline with actual emissions for 2012. Here, the total yearly agricultural NH_3 emission amounts to 59 kt NH_3-N for Denmark (this includes the emissions from houses, storage and manure application as given in Table 3, as well as emissions from crops and fertilizer use).
- The scenario with acidification and air cleaners. Here the total yearly agricultural amounts to NH_3 35 kt NH_3-N for Denmark.

The meteorological conditions have a significant impact on the overall deposition of N and also for the distribution between wet and dry deposition. Here we apply meteorological data for the year 2009, which in Denmark can be considered close to an "average" meteorological year in terms of wind direction and precipitation when compared to the previous 30-year period.

2.4. SWAT Calculations of Riverine Nitrogen Loads

The SWAT model (Soil and Water Assessment Tool, version 2009, [51]) is set up for the North Jutland region and includes 182 sub-basins of which 118 drain into the Limfjord. Details on the input data (e.g., soil and climate data) and calibrations are given in the supplementary. The agricultural practice is included as 14 five-year crop rotations representing different forms of agriculture (pig farms, dairy farms, and arable farms at different fertilization intensities). The representation of agriculture in SWAT matches average regional statistics on crop use (area covered and relationship to farm type) and fertilizer application (chemical and/or manure applied to what crop at what time in accordance to farm type) for the year 2005 [52,53].

The baseline and the emission scenario are simulated in the SWAT model by changing two different model inputs. The changes in atmospheric deposition, as obtained from the DEHM model, are incorporated by the NO_3 concentration in rainfall. The changes in nitrogen content in slurry (organic fertilizer) are incorporated by adjusting the nitrogen concentrations in the applied organic fertilizers (Table 3).

2.5. Quantifying Effects on Freshwater Ecosystems

Results of the SWAT model were used as input into the next model in the overall system, where the effects on the freshwater ecosystems in two typical Danish lakes are quantified. The GOTM-FABM-PCLake ecosystem model [54,55] was set up to represent a shallow and a deep Danish lake, represented by the lakes Klejtrup and Tjele Langsø (Figure 2). The model simulates a wide range of ecosystem state variables, which can help indicate the level of integrity of a freshwater ecosystem. We used total chlorophyll *a* and coverage of submerged macrophytes as indicators when estimating potential impacts of manure acidification on freshwater aquatic ecosystems. The baseline calculation for the two lakes included river and nutrient inflow boundary conditions based on a national model complex (DK-QNP) by [56] and local meteorological forcing (see the supplementary for detail). The changes in nitrogen runoff into waterways as simulated by the SWAT model were used to change the river boundary conditions for the lake ecosystem models, relative to the baseline calculation. Thereby could the changes in the two key state variables be quantified.

2.6. Quantifying Effects on Terrestrial Ecosystems

Critical loads for terrestrial ecosystems are defined as a quantitative estimate of an exposure to a pollutant, below which significant harmful effects on specified sensitive elements of the environment do not occur [57]. Critical loads have been a highly successful tool in, especially, European air pollution policies addressing the widespread and severe problems of acidification and eutrophication caused by elevated sulfur and nitrogen depositions in the 1980s and 1990s [58]. In Denmark, biodiversity-based critical loads for nitrogen have been established and used in the assessment of air pollution effects on biodiversity based on a method developed by [59]. A coupled soil geochemical/plant occurrence model, VSD+/MOVE [60], was used to analyze scenarios for air pollution effects on biodiversity in Danish nature areas and to isolate the effect of air pollution from the effects of other pressures. A suitable indicator (no net loss of biodiversity compared to 1992 as reference year [61]) and criteria were established for biodiversity effects, and based on this, critical loads for a number of the Habitat Directives Annex 1 nature types have been calculated for different biodiversity targets.

Exceedance of critical loads for five nature types in the Limfjord catchment were evaluated in the baseline scenario and in the emission scenario using maps of the nitrogen deposition from the DEHM model. For light-open nature not protected under the Habitat Directive but protected under the Danish Nature Protection Act (§ 3), median values of empirical-based critical loads have been used [62].

2.7. Quantifying Effects on Marine Ecosystems

The effects on marine ecosystems caused by the changed input of nutrients in the scenario were quantified by the use of empirical models. These models were based on the time series of marine data and nutrient loadings from the national marine monitoring program (NOVANA), as well as on meteorological observations provided by the Danish Meteorological Institute. The empirical models were partial least squares (PLS) regression models, with summer chlorophyll *a* concentration and light attenuation as the dependent variables and eight independent variables consisting of N-loading (ton year^{-1}), P-loading (ton year^{-1}), freshwater inflow (m^3 month^{-1}), wind power (m^3 s^{-3}), surface irradiance (µmol photons m^{-2} s^{-1}), salinity (psu), water column stability (s^{-1}) and sea surface temperature (°C). An iterative process using cross-validated multiple linear regression (MLR), in combination with stepwise exclusion of the explanatory variable that gave the lowest model error in the form of "Root Mean Square Error of Cross Validation", was used for selecting independent variables for each model (see the supplementary and [63] for details). The selected variables were used to produce a PLS regression and for each variable the root mean squared error of both the calibration data and the validation data was quantified and parameters that increased the error in the prediction of the validation data were left out. The PLS regression analysis was done in MATLAB® using a PLS program package from Eigenvector®.

2.8. Quantifying the Abatement Costs Related to Nr Reductions from Acidification of Manure

The abatement costs associated with barn acidification on all farms and air cleaning of all poultry farms are estimated based on data for investment and operating costs, including labor costs, from [64], adjusted to the 2020 price level. The costs are estimated for farm size classes of between 75 and 950 livestock units for both cattle and pigs, as relatively high investment costs result in decreasing costs per animal for increasing herd sizes. The costs at farm level are aggregated to the national level using data from the General Husbandry Register for the distribution of types and numbers of animals per farm. The dataset is from 2012 and is the same as the dataset used for the assessment of emissions in Section 2.2.

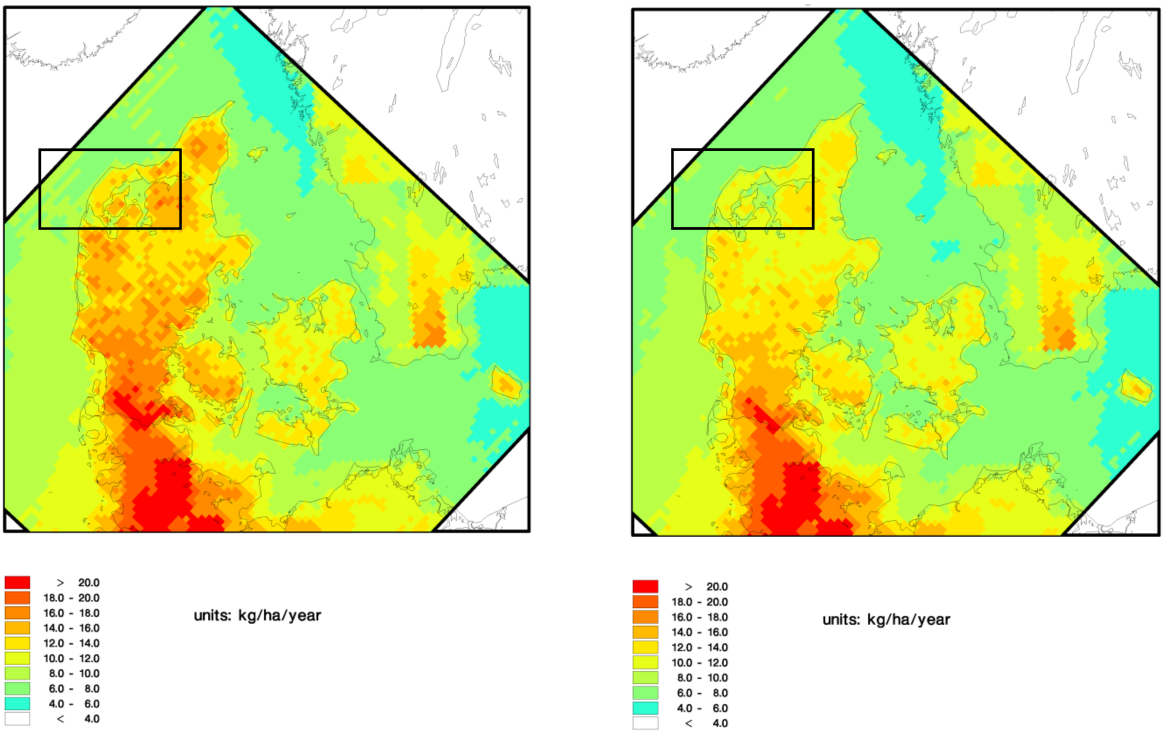
The capital costs for acidification contain investments in equipment including the mixing tank and installation. The investment costs are annualized using an interest rate of 4%, and the lifetime of the investments related to acidification in the barns is set to 12 years. The lifetime might be longer (15 years), so this is a conservative assumption. The operating costs for acidification also include an increase in the use of liming and labor, as well as energy consumption. The costs of air purification mainly constitute investment costs but also include operating costs for labor, change of filters, water and electricity, as well as maintenance. The assumed lifetime for the air purification is 10 years. For both cattle and pig production, the nitrogen efficiency of the manure will increase, and this increase can be measured as saved costs for fertilizers. The acid used enriches the manure and substitutes acid in fertilizers, and therefore reduced costs for both nitrogen fertilizers and sulfur are accounted for, using the price for sulfur and nitrogen fertilizers.

The differentiation of the investment and operating costs on herd sizes as done in this assessment is supported by [65], who concluded that there are economics of scale, based on a review of studies.

3. Results

3.1. Changes in Nitrogen Deposition and Effects on Terrestrial Ecosystems

The resulting atmospheric deposition of total Nr as simulated by the DEHM model is shown in Figure 3 for the baseline and scenario.



(**a**) (**b**)

Figure 3. The modeled total deposition of Nr in the baseline (**a**) and the scenario (**b**) runs. From the 5.56 km × 5.56 km inner nest of the DEHM model. The Limfjord area is indicated by the black box.

The only difference between the simulations is that the Danish ammonia emissions are about 40% lower in the scenario. This reduces the total N deposition by about 11% on average for the Danish land areas, with some regional differences.

The largest reduction is seen in the central and northern part of Jutland, where the agricultural activities in general are most intensive. In the Limfjord area, the N deposition is reduced by ca. 17% and the largest reductions are obtained in the deposition linked to dry deposition of NH_x (Table 4). The overall deposition is unchanged in the southern part of Jutland, where the southwesterly winds lead to a large impact from sources in Germany and the rest of Europe.

Table 4. The modeled annual depositions of NOx and NHx (wet and dry) into the Limfjord catchment area and to the marine areas of the Limfjord in the two simulations. The total deposition and the change between the baseline and the scenario are also given.

Deposition (kg ha^{-1} yr^{-1})	Baseline	Scenario	Change (%)
NOx wet	3.2	3.2	0
NOx dry	1.9	2.0	6
NHx wet	4.0	3.3	−17
NHx dry	4.7	2.9	−39
Total N dep	13.7	11.4	−17

The modeled deposition with separate values for forest and light-open nature has been used to calculate the effects on terrestrial ecosystems, expressed as areas with an exceedance of critical loads. Habitat Directive Annex 1 nature types and light-open nature protected under the Danish Nature Conservation Act cover, in total, 62,400 ha in the Limfjord catchment area (including a 2900 ha Annex 1 forest). In the baseline, the critical loads are exceeded in about 61% of the nature areas; this is reduced to about 54% of the area in the acidification scenario. The average accumulated exceedance is ca. 4.5 kg N ha^{-1}y^{-1} in the baseline and is reduced to 2.8 kg N $ha^{-1}y^{-1}$ in the scenario. The exceedances at the main ecosystem types are given in Table 5. The largest changes are seen for the nature types dune, heath, dry grassland, and Annex 1 forest, where present depositions overlap the critical loads. The more sensitive nature types such as sensitive mires, bogs and fens have a critical load interval of 5–10 kg N $ha^{-1}y^{-1}$, and for these ecosystems the critical load is exceeded in nearly 100% of the areas in both the baseline and in the scenario.

Table 5. Percentage areas on which critical loads are exceeded in the baseline and scenario.

Area Type	Baseline	Scenario
Annex 1 forest	90.0	42.1
Dune	86.0	68.2
Heath	32.5	18.9
Dry grassland	25.7	12.3
Sensitive mire, bog and fen	98.4	98.4

3.2. Changes in N Input to Limfjorden and Effects on Marine Ecosystem

The changes in nitrogen load to the marine areas as estimated by the SWAT model are given in Table 6.

Table 6. Percentage change between baseline and scenario in nitrogen load to the six marine areas (shown in Figure 2).

Marine Area	NO_3	NH_4^+	NO_2	Tot N
1	0.8	0.0	0	0.7
2	0.6	1.0	0	0.5
3	1.3	−0.1	0	1.3
4	1.3	0.0	0	1.2
5	1.5	0.1	0	1.5
6	1.6	0.0	0	1.5

The changes are given as the differences between the baseline and the emission scenario for the six marine areas defined in Figure 2. The largest changes are seen for NO_3. The differences between the inputs from the different catchments are mainly driven by differences in agricultural land use. In catchments with a high share of agricultural land use, the scenario with increased N concentration in slurry leads to higher N loads than in the other catchments.

Although primary production in Limfjorden is believed to be N, limited especially during summer, the minor increase (<2%) in nitrogen loading only had minor effects (<1.5%) on chlorophyll *a* concentrations and negligible effects (<0.5%) on light attenuation (Table 7).

Table 7. Percentage changes in summer chlorophyll *a* concentrations and light attenuation in the different areas of Limfjorden that are due to changes in nutrient input in the scenario. No data indicate that nitrogen load was not selected as a controlling variable.

Marine Area	Chlorophyll *a*	Light Attenuation
1	0.75	
2		
3	0.6	0.24
4	1.4	0.27
5		
6	1.3	

The results indicate that chlorophyll *a* responds more strongly on changes in nutrient loadings compared to light attenuation, which is not surprising since light attenuation is often influenced by a variety of parameters, whereas chlorophyll *a* is more directly linked to eutrophication. There were minor differences between the sub-basins, reflecting differences in both loadings and sensitivity.

3.3. Changes in N Input into Lake Kjelstrup and Tjele and Effects on Freshwater Ecosystems

The nitrogen nutrient loads to both lakes were adjusted according to changes simulated with the SWAT model (Table 6). As expected, the effects of minor changes in nitrogen load on the two Danish freshwater lakes, in which the primary producers for the majority of a year are considered phosphorus-limited, are negligible. Changing the annual external nitrogen load by 1.6% resulted in changes in annual average chlorophyll and submerged macrophytes by <1‰.

3.4. Costs of Abating Nr with Acidification

The total cost of the emission scenario is estimated to be EUR 231 million, including both barn acidification in cattle and pig production and air purification in poultry production. As mentioned in Section 2, acidification will increase the nitrogen (N) efficiency of the manure, which is used as fertilizer, and this increased efficiency is measured as a saving in the costs of commercial fertilizer. As mentioned, the reduced costs include both N fertilizers and sulfur (S). Therefore, the fertilizer value of the slurry increases when acid is added, as a larger share of the N stays in the manure when acidified. In addition, the added S increases the fertilizer value of the manure. This value of the added S depends on crop choice and is therefore assumed to differ between pig and cattle livestock farms because the crop composition differs on those farm types. Accounting for value added by the increased content of N in manure, as well as the added amount of S, the actual operating costs are reduced, even if there are costs for purchasing the sulfur for the acidification. This can be seen from the first and second rows in Table 8. The adjusted total cost is EUR 181 million, corresponding to a unit price of 8.8 EUR/kg^{-1} N, where the lowest costs per kg N are found for air purification in poultry (4.2 EUR/kg^{-1} N) and the highest costs per kg N are found for cattle and pigs (11.2 EUR/kg^{-1} N), i.e., an equal cost-effectiveness ratio for these productions.

In comparison, [66] estimated a cost-effectiveness ratio between 3–7 EUR/kg^{-1} NH_3-N for ammonia abatement by acidification in Denmark, and barn acidification was included. The assumptions related to these estimates are not quite clear, but differences in the assumption of the distribution of farm sizes might be an important difference, as the costs are high for the smaller farms.

Table 8. The abatement costs for the scenario: total costs and costs distributed across the different animal types.

	Total Costs	Poultry (Air Cleaning)	Cattle (Acidification)	Pigs (Acidification)
Cost (EUR, Million)	231	76	94	129
Cost incl. the value of N and S (EUR)	181	56	79	96
Cost-effectiveness ratio (EUR/kg N)	8.8	4.2	11.2	11.2

4. Discussion

4.1. Reducing Ammonia Emissions

Regulation has led to significant reductions in emissions of many key air pollutants, while only small reductions or even increases have been seen for NH_3 globally and in

Europe [18,67]. On top of that, the direct emission of NH_3 from agricultural activities has been projected to increase in a warming climate [68], underlining the importance of implementing additional measures to also obtain reduced emissions in the longer time scales. As most terrestrial ecosystems are highly vulnerable to Nr deposition [9], there is an urgent need to reduce NH_3 emissions as close to the source as possible, and in this respect, manure acidification and especially air cleaners are efficient mitigation measures.

In addition to the impacts on ecosystems, emissions of NH_3 are also related to impacts on human health. Exposure to NH_3 and NH_4^+ has, e.g., been shown to increase the risk of developing asthma in childhood [69]. More importantly, NH_3 adds to the formation of secondary fine particulate matter within the atmosphere, which is linked with both morbidity and mortality [70,71]. It has been estimated that about 220 premature deaths each year in Denmark can be associated to air pollution related to the national agricultural sector [72].

As manure acidification as an NH_3 abatement technique potentially affects several environmental matrices (soil, air, fresh water and marine systems), proper assessment requires a holistic assessment system encompassing the relevant matrices. For this study, we have created the DEEMON modeling chain, allowing for running emission scenarios and tracing the effects through atmospheric deposition and land run-off to effects in terrestrial, freshwater and marine recipients. Our scientific hypothesis was that this approach can lead to "pollution swapping", where abatement measures leading to, e.g., reduced NH_3 emissions into the atmosphere can potentially lead to increased N loadings in other parts of the environment. The model scenario results revealed that manure acidification and air cleaners reduced NH_3 emissions in Denmark by 40% compared to the actual agricultural practice (with no NH_3 abatement) in 2012. This led to a reduction of approximately 17% in the total N deposition in the Limfjord catchment area and had significant positive effects on sensitive terrestrial ecosystems. For forests, dunes and heaths, the modeled N deposition got below the critical level, so that significant reductions were seen in a number of areas where the critical loads are exceeded. However, for the most sensitive nature types (mires, bogs and fens), the critical loads were still exceeded. When moving on to the aquatic environment, our analysis showed that the scenario only led to a small (<2%) increase in the nitrogen loading in lakes and marine environments in the Limfjorden catchment, with either negligible negative environmental effects (lakes) or minor negative effects (coastal areas, Table 7). Hence, for this test case, manure acidification did not induce any significant nitrogen pollution swapping and our hypothesis proved wrong. This is supported by a recent study by [73], showing that application of acidified slurry did not increase nitrate leaching compared to slurry injection under Mediterranean conditions.

4.2. Other Side Effects

There might, however, be other potential side effects of using manure acidification as an ammonia emission abatement measure. As current manure acidification technologies involve acidification with H_2SO_4, this results in reduced manure and soil pH (short term for soil) and a potential increase in sulfur. Reduction in manure pH has been shown to affect rates and pathways of microbial organic matter degradation [74,75]. As most enzymes and microorganisms are pH sensitive, the enzymatic and microbially controlled degradation of slurry and soil organic matter will be affected as a result of lower pH. This will often result in a higher content of large, dissolved organic compounds and lower content of non-dissolved and small, dissolved organic compounds. Changes in degradation patterns are, however, likely to be relevant only for long-term storage tank acidification and less relevant for acidification in field application [75]. Although manure acidification affects degradation patterns and, thus, likely the microbial community, there are no clear evidence of changes in the microbial composition in either acidified slurry [75] or in soil after application of acidified slurry [76] compared to non-acidified slurry.

The addition of substantial amounts of H_2SO_4 to the soil will likely increase the SO_4-S soil pore water concentration as documented by [76]. The environmental implications are so far not well-documented. A few studies have indicated that sulfuric acid increases the fertilizer value of manure [77] as plants cannot efficiently use nitrogen in the absence of sulfur [78,79], but it also increases the risk of odor from sulfur compounds [76]. In addition, excess sulfur is prone to leach from agricultural soils, even from soils with good adsorption capacity [76], with adverse effects in eutrophic lakes. In these lakes, sulfate will be reduced to hydrogen sulfide in the lake sediments, eventually leading to the dissolution of ironbound phosphorus and thereby increasing eutrophication [80].

4.3. Assumptions and Uncertainties

The individual models have been applied in other studies, e.g., as part of the monitoring of the environment in Denmark [81–83], but only a modeling chain such as the DEEMON system allows for a consistent evaluation from management regulation to environmental effects. In the following, we discuss the main underlying assumptions and uncertainties related to the model chain and the scenarios. The model system is based on (semi-) steady state assumptions, where potential time lags between implementation of measures and environmental effects in lakes and coastal waters are largely ignored. Significant time lags can, however, be expected as changes in agricultural systems are not immediately reflected in the nutrient loadings. In addition, responses in the environmental conditions of lakes and coastal waters following changes in nutrient loading may also be prolonged. The length of the time lag from the agricultural system to changes in nutrient loadings depend on the size and type of the catchment, including the age of the groundwater, and transport time lags have been found to mask trends in N loads following changes in agricultural systems in large river basins [84,85]. However, for smaller sandy and loamy Danish catchments, [86] found by particle tracking in a 3D groundwater model that the median age of oxidized groundwater input to stream water was less than one year. They also showed that the model could not recreate the long groundwater travel times in the chalk catchments found in the southeastern part of the Limfjord catchments. Although marine and coastal ecosystems show signs of recovery after decreasing trends in nutrient loadings [87], significant time lags (or hysteresis) can be expected [88] resulting from long-term accumulation of organic matter and reduced compounds [89], as well as from deterioration of habitats and food webs.

All parts of the applied modeling chain are associated with significant uncertainties, and in the current setup, we focus on a specific case study area and scenario results based on a single meteorological year. As described above, we apply state-of-the-art models that are continuously evaluated using long-term (up to 30 years) monitoring data, reducing the risk of model bias. The scenario used to assess the impact of manure acidification is based on the meteorological year 2009 and the impact of the applied meteorological year for the atmospheric deposition was assessed by comparing it to other years. Here we find that the modeled atmospheric deposition (wet and dry) for the Limfjord area is within 4–6% of the average dry and wet deposition for the period 2005–2014. As we mainly focus on the differences between two scenarios, we believe that it is reasonable to assume that the results are robust and can be generalized to other years. In the Limfjorden catchment, atmospheric N deposition only constitutes a minor (<5%) part of the total nitrogen input into lakes and coastal waters. Hence, impacts of decreased atmospheric N deposition that are due to manure acidification are negligible and not expected to counteract the increase in riverine N input. For surface waters less affected by riverine N input, atmospheric N deposition might be important, and here, reduced N deposition is expected to have a positive environmental effect despite a minor increase in riverine N input.

The expected environmental effects of manure acidification are, to a large extent, determined by the sensitivity of atmospheric N deposition relative to the sensitivity of riverine N inputs. Hence, the scenario results from the Limfjorden catchment can be considered a "worse case" example of the impacts of pollution swapping caused by manure acidification. In less-eutrophied surface waters mainly affected by atmospheric N deposition, manure acidification will not only improve terrestrial environments but might also improve the environmental condition of lakes and coastal waters.

The cost-effectiveness ratio for acidification and air cleaning is between EUR 4–11 per kg N. As these technologies are not yet mature and widespread, the costs can be expected to decrease as the technology is more widely taken up. The cost-effectiveness ratio is somewhat higher than former estimated costs and this can be explained by both differences in the approach, where attention is paid to the economics of scale here, and also by differences in the effects on ammonia emissions, which are modeled in a detailed manner here. Only 150 barn acidification tanks were established in Denmark in 2020 [90], which makes our cost estimates somewhat uncertain. Air cleaning is, however, becoming more widely used, and in the Netherlands 46% of pig farms had this technology installed in 2015. Moreover, this technology will be required on large pig farms in Schleswig-Holstein [65,66,91]. The costs of air cleaning are increasing with cleaning efficiency, so that 100% cleaning is much more expensive than lower cleaning efficiency [90].

The emission scenario is based on existing and well-tested acidification technologies addressing barn acidification, as well as on existing air cleaning systems for poultry farms. Hence, the scenario covers realistic reductions in NH_3 emissions targeting the main types of animal houses in Denmark. Further ammonia emission reductions could be achieved by, e.g., combining air cleaning in pig houses with acidification in slurry tanks. Such a scenario is, however, seen as more unrealistically costly and therefore not included here.

5. Conclusions

By combining existing models, we have set up a holistic system for evaluating the potential positive and negative effects of abatement measures in the agricultural sector. As a test case, the system was set up for the Limfjord and the surrounding catchment in Denmark, where the combination of intensive agriculture and sensitive marine and terrestrial ecosystems represents a region where the cascading effects of reactive nitrogen need to be fully described. Environmental management in such regions is typically divided into the various forms and compartments of Nr. Our scientific hypothesis was that this approach can lead to "pollution swapping", where abatement measures leading to, e.g., reduced NH_3 emissions into the atmosphere can potentially lead to increased N loadings in other parts of the environment.

We tested this by setting up an emission scenario assuming manure acidification and air cleaners in the agricultural sector in Denmark and estimating the effects on terrestrial and aquatic ecosystems. The emission scenario was evaluated in comparison to the actual management practice in 2012. The scenario resulted in reduced NH_3 emissions (40% lower than the annual national total for 2012) and, hence, to significant reductions in nitrogen deposition across Denmark. For the Limfjord region, this led to significant reductions in the areas where critical loads for terrestrial ecosystems are exceeded. Increased N content in the applied slurry in the scenario run led, however, only to very small changes (<2%) in the N load within the catchment and, hence, to very small changes in the environmental parameters in the aquatic ecosystems. The increased content in the applied slurry could, though, be compensated for through a decrease in the application of chemical fertilizers. In the marine systems in the Limfjord, the chlorophyll *a* concentrations and light attenuation were estimated to change by less than 1.5%. In the two freshwater lakes within the catchment, the effect of the increased N load was also estimated to be insignificant. We can thereby conclude that for this test case our hypothesis proved wrong. The suggested abatement measures resulted in reduced impacts on terrestrial ecosystems, while the increased N load in the aquatic systems was so small that the potential negative impact on the aquatic ecosystems was very limited. By assessing the costs related to investment and the operational costs for acidification and air cleaning, a cost-effectiveness ratio on the order of EUR 4–11 per kg N was found. The DEEMON modeling chain nevertheless proved operational and can now be used to test other scenarios and relevant abatement measures in the future.

The current model scenario also illustrates that the combination of manure acidification and air cleaners is a highly effective mitigation measure that, if implemented at the national scale, could significantly reduce the emission of ammonia into the atmosphere. Even if this is shown to reduce the related negative impact on the terrestrial ecosystems in the test region, the critical loads for the most sensitive ecosystems are still exceeded.

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Reference

1. Galloway, J.N.; Winiwarter, W.; Leip, A.; Leach, A.; Bleeker, A.; Erisman, J.W. Nitrogen footprints: Past, present and future. *Environ. Res. Lett.* **2014**, *9*, 115003. https://doi.org/10.1088/1748-9326/9/11/115003.
2. Sutton, M.A.; Howard, C.M.; Erisman, J.W.; Billen, G.; Bleeker, A.; Greenfelt, P.; Grinsven, H.V.; Grizzetti, B. *The European Nitrogen Assessment—Sources, Effects and Policy Perspectives*; Cambridge CB2 8RU; Cambridge University Press: Edinburgh, UK, 2011; p. 612.
3. Galloway, J.N.; Aber, J.D.; Erisman, J.W.; Seitzinger, S.P.; Howarth, R.W.; Cowling, E.B.; Cosby, B.J. The Nitrogen Cascade. *BioScience* **2003**, *53*, 341–356. https://doi.org/10.1641/0006-3568(2003)053[0341:TNC]2.0.CO.;2.
4. Battye, W.; Aneja, V.P.; Schlesinger, W.H. Is nitrogen the next carbon? *Earth's Futur.* **2017**, *5*, 894–904. https://doi.org/10.1002/2017ef000592.
5. Galloway, J.N.; Leach, A.; Erisman, J.W.; Bleeker, A. Nitrogen: The historical progression from ignorance to knowledge, with a view to future solutions. *Soil Res.* **2017**, *55*, 417. https://doi.org/10.1071/sr16334.
6. Brandt, J.; Silver, J.D.; Christensen, J.H.; Andersen, M.S.; Bønløkke, J.H.; Sigsgaard, T.; Geels, C.; Gross, A.; Hansen, A.B.; Hansen, K.M.; et al. Contribution from the ten major emission sectors in Europe and Denmark to the health-cost externalities of air pollution using the EVA model system–an integrated modelling approach. *Atmos. Meas. Tech.* **2013**, *13*, 7725–7746. https://doi.org/10.5194/acp-13-7725-2013.
7. Geels, C.; Ellermann, T.; Hertel, O.; Brandt, J.; Skjøth, C.A.; Winiwarter, W.; Im, U.; Hansen, K.M.; Christensen, J.H. Changing Agricultural NH3 Emissions Since 1979: The Impact on N Deposition and Health Effects Across Europe and the Potential for Further Reductions in the Future. In *Air Pollution Modeling and its Application XXV*; Mensink, C., Kallos, G., Eds.; ITM 2016; Springer Proceedings in Complexity; Springer: Berlin/Heidelberg, Germany, 2017; pp. 477–482. https://doi.org/10.1007/978-3-319-57645-9_75.
8. Malley, C.S.; Hicks, W.K.; Kulyenstierna, J.C.I.; Michalopoulou, E.; Molotoks, A.; Slater, J.; Heaps, C.G.; Ulloa, S.; Veysey, J.; Shindell, D.T.; et al. Integrated assessment of global climate, air pollution, and dietary, malnutrition and obesity health impacts of food production and consumption between 2014 and 2018. *Environ. Res. Commun.* **2021**, *3*, 075001. https://doi.org/10.1088/2515-7620/ac0af9.
9. Stevens, C.J.; Duprè, C.; Dorland, E.; Gaudnik, C.; Gowing, D.J.; Bleeker, A.; Diekmann, M.; Alard, D.; Bobbink, R.; Fowler, D.; et al. Nitrogen deposition threatens species richness of grasslands across Europe. *Environ. Pollut.* **2010**, *158*, 2940–2945. https://doi.org/10.1016/j.envpol.2010.06.006.
10. Phoenix, G.K.; Emmett, B.A.; Britton, A.J.; Caporn, S.J.M.; Dise, N.B.; Helliwell, R.; Jones, L.; Leake, J.R.; Leith, I.D.; Sheppard, L.J.; et al. Impacts of atmospheric nitrogen deposition: Responses of multiple plant and soil parameters across contrasting ecosystems in long-term field experiments. *Glob. Chang. Biol.* **2011**, *18*, 1197–1215. https://doi.org/10.1111/j.1365-2486.2011.02590.x.
11. Cloern, J. Our evolving conceptual model of the coastal eutrophication problem. *Mar. Ecol. Prog. Ser.* **2001**, *210*, 223–253. https://doi.org/10.3354/meps210223.
12. Anderson, D.M.; Glibert, P.M.; Burkholder, J.M. Harmful algal blooms and eutrophication: Nutrient sources, composi-tion, and consequences. *Estuaries* **2002**, *25*, 704–726.

13. Howarth, R.W.; Marino, R. Nitrogen as the limiting nutrient for eutrophication in coastal marine ecosystems: Evolving views over three decades. *Limnol. Oceanogr.* **2006**, *51*, 364–376. https://doi.org/10.4319/lo.2006.51.1_part_2.0364.
14. Diaz, R.J.; Rosenberg, R. Spreading Dead Zones and Consequences for Marine Ecosystems. *Science* **2008**, *321*, 926–929. https://doi.org/10.1126/science.1156401.
15. Hasler, B.; Hansen, L.B.; Andersen, H.E.; Termansen, M. Cost-effective abatement of non-point source nitrogen emissions—The effects of uncertainty in retention. *J. Environ. Manag.* **2019**, *246*, 909–919. https://doi.org/10.1016/j.jenvman.2019.05.140.
16. Hyytiäinen, K.; Blyh, K.; Hasler, B.; Ahlvik, L.; Ahtiainen, H.; Artell, J.; Ericsdotter, S. *Environmental Economic Research as a tool in the Protection of the Baltic Sea: Costs and Benefits of Reducing Eutrophication*; Nordic Council of Ministers: Copenhagen, Denmark, 2014; Volume 504. https://doi.org/10.6027/TN2014-504.
17. Jensen, C.L.; Jacobsen, B.H.; Olsen, S.B.; Dubgaard, A.; Hasler, B. A practical CBA-based screening procedure for identification of river basins where the costs of fulfilling the WFD requirements may be disproportionate—Applied to the case of Denmark. *J. Environ. Econ. Policy* **2013**, *2*, 164–200. https://doi.org/10.1080/21606544.2013.785676.
18. EEA, 2020. Available online: https://www.eea.europa.eu/data-and-maps/dashboards/necd-directive-data-viewer-3 (accessed on 1 September 2022).
19. EU. Directive (EU) 2016/2284 of the European Parliament and of the Council of 14 December 2016 on the Reduction of National Emissions of Certain Atmospheric Pollutants, Amending Directive 2003/35/EC and Repealing Directive 2001/81/EC. Available online: https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=uriserv%3AOJ.L_.2016.344.01.0001.01.ENG (accessed on 1 September 2022).
20. EMEP. Available online: https://www.emep.int/ (accessed on 1 September 2022).
21. Dalgaard, T.; Hansen, B.; Hasler, B.; Hertel, O.; Hutchings, N.J.; Jacobsen, B.H.; Jensen, L.S.; Kronvang, B.; Olesen, J.E.; Schjorring, J.K.; et al. Policies for agricultural nitrogen management-trends, challenges and prospects for improved efficiency in Denmark. *Environ. Res. Lett.* **2014**, *9*, 115002. https://doi.org/10.1088/1748-9326/9/11/115002.
22. Hellsten, S.; Dalgaard, T.; Rankinen, K.; Tørseth, K.; Bakken, L.; Bechmann, M.; Kulmala, A.; Moldan, F.; Olofsson, S.; Piil, K.; et al. Abating N in Nordic agriculture—Policy, measures and way forward. *J. Environ. Manag.* **2019**, *236*, 674–686. https://doi.org/10.1016/j.jenvman.2018.11.143.
23. Wulff, F.; Humborg, C.; Andersen, H.E.; Blicher-Mathiesen, G.; Czajkowski, M.; Elofsson, K.; Fonnesbech-Wulff, A.; Hasler, B.; Hong, B.; Jansons, V.; et al. Reduction of Baltic Sea Nutrient Inputs and Allocation of Abatement Costs Within the Baltic Sea Catchment. *AMBIO* **2014**, *43*, 11–25. https://doi.org/10.1007/s13280-013-0484-5.
24. Husted, S.; Jensen, L.S.; Jørgensen, S.S. Reducing ammonia loss from cattle slurry by the use of acidifying additives: The role of the buffer system. *J. Sci. Food Agric.* **1991**, *57*, 335–349. https://doi.org/10.1002/jsfa.2740570305.
25. Kai, P.; Pedersen, P.; Jensen, J.E.; Hansen, M.N.; Sommer, S.G. A whole-farm assessment of the efficacy of slurry acidification in reducing ammonia emissions. *Eur. J. Agron.* **2008**, *28*, 148–154. https://doi.org/10.1016/j.eja.2007.06.004.
26. Petersen, S.O.; Andersen, A.J.; Eriksen, J. Effects of Cattle Slurry Acidification on Ammonia and Methane Evolution during Storage. *J. Environ. Qual.* **2012**, *41*, 88–94. https://doi.org/10.2134/jeq2011.0184.
27. Pexas, G.; Mackenzie, S.G.; Wallace, M.; Kyriazakis, I. Environmental impacts of housing conditions and manure management in European pig production systems through a life cycle perspective: A case study in Denmark. *J. Clean. Prod.* **2020**, *253*, 120005. https://doi.org/10.1016/j.jclepro.2020.120005.
28. Emmerling, C.; Krein, A.; Junk, J. Meta-Analysis of Strategies to Reduce NH_3 Emissions from Slurries in European Agriculture and Consequences for Greenhouse Gas Emissions. *Agronomy* **2020**, *10*, 1633. https://doi.org/10.3390/agronomy10111633.
29. Mikkelsen, M.H.; og Nyord, T. *Opdaterede Aktivitetsdata for Udbringning af Husdyrgødning og Konsekvenserne for Nationale Emissionsopgørelser. Rådgivningsnotat, 15.02.2019*; DCA—Nationalt Center for Fødevarer og Jordbrug, Aarhus Universitet: Aarhus, Denmark, 2019. https://pure.au.dk/portal/files/145164819/Levering_Gylleudbringsningsteknologianvendelse.pdf. (In Danish), (accessed on 1 September 2022).
30. Reis, S.; Bekunda, M.; Howard, C.M.; Karanja, N.; Winiwarter, W.; Yan, X.; Bleeker, A.; Sutton, M.A. Synthesis and review: Tackling the nitrogen management challenge: From global to local scales. *Environ. Res. Lett.* **2016**, *11*, 120205. https://doi.org/10.1088/1748-9326/11/12/120205.
31. Riisgård, H.U.; Andersen, P.; Hoffmann, E. From Fish to Jellyfish in the Eutrophicated Limfjorden (Denmark). *Estuaries and Coasts*. **2012**, *35*, 701–713. https://doi.org/10.1007/s12237-012-9480-4.
32. Mst.dk. Available online: https://mst.dk/natur-vand/vandmiljoe/vandomraadeplaner/vandplaner-2009-2015/hoeringer/hoering/basisanalyser/1-2-limfjorden/ (accessed on 1 September 2022).
33. Kristensen, I.T.; Rasmussen, B.M.; Staunstrup, J.K.; Mouritsen, A.K.M.; Bøcher, P.K.; Hansen, J.F. *Geografisk Variation i Nøgleparametre for Dansk Landbrug*; Danmarks Jordbrugs Forskning: Tjele, Denmark, 2005.
34. Allerup, P.; Madsen, H.; Vejen, F. Correction of precipitation based on off-site weather information. *Atmos. Res.* **2000**, *53*, 231–250. https://doi.org/10.1016/s0169-8095(99)00051-4.
35. Thodsen, H.; Tornbjerg, H.; Bøgestrand, J.; Larsen, S.E.; Ovesen, N.B.; Blicher-Mathiesen, G.; Rolighed, J.; Holm, H.; Kjeldgaard, A. *Vandløb 2019—Kemisk Vandkvalitet og Stoftransport. NOVANA*; Aarhus Universitet, DCE—Nationalt Center for Miljø og Energi: Aarhus, Denmark, 2021. http://dce2.au.dk/pub/SR452.pdf. (accessed on 1 September 2022).
36. Poulsen, H.D. Normtal for Husdyrgødning–2016, 35 sider. 2017. Available online: http://anis.au.dk/fileadmin/DJF/Anis/dokumenter_anis/Forskning/Normtal/Normtal_2017.pdf (accessed on 1 September 2022).

37. Gyldenkærne, S.; Skjøth, C.A.; Hertel, O.; Ellermann, T. A dynamical ammonia emission parameterization for use in air pollution models. *J. Geophys. Res. Atmos.* **2005**, *110*, D07108. https://doi.org/10.1029/2004jd005459.
38. Søgaard, H.; Sommer, S.; Hutchings, N.; Huijsmans, J.; Bussink, D.; Nicholson, F. Ammonia volatilization from field-applied animal slurry—The ALFAM model. *Atmos. Environ.* **2002**, *36*, 3309–3319. https://doi.org/10.1016/s1352-2310(02)00300-x.
39. Hafner, S.D.; Pacholski, A.; Bittman, S.; Burchill, W.; Bussink, W.; Chantigny, M.; Carozzi, M.; Génermont, S.; Häni, C.; Hansen, M.N.; et al. The ALFAM2 database on ammonia emission from field-applied manure: Description and illustrative analysis. *Agric. For. Meteorol.* **2018**, *258*, 66–79. https://doi.org/10.1016/j.agrformet.2017.11.027.
40. Nielsen, O.-K.; Plejdrup, M.S.; Winther, M.; Nielsen, M.; Gyldenkærne, S.; Mikkelsen, M.H.; Albrektsen, R.; Thomsen, M.; Hjelgaard, K.; Fauser, P.; et al. *Denmark's National Inventory Report 2020. Emission Inventories 1990-2018—Submitted under the United Nations Framework Convention on Climate Change and the Kyoto Protocol*; Scientific Report No. 372; Aarhus University, DCE—Danish Centre for Environment and Energy: Aarhus, Germany, 2020; p. 900. http://dce2.au.dk/pub/SR372.pdf. (accessed on 1 September 2022).
41. Nyord, T.; Liu, D.; Eriksen, J.; Adamsen, A.P.S. Effect of acidification and soil injection of animal slurry on ammonia and odour emission. In Proceedings of the The Ramiran Conference, Versailles, Paris, 3–5 June 2013.
42. Sørensen, K. *Test of Biological Air-Cleaner Dorset Milieutechniek*; Videncenter for Svineproduktion, Den Rullende Afprøvning: Copenhagen, Denmark, 2011.
43. Riis, A.L. Test of *Farm Airclean 3-step Bio Flex from SKOV A/S in a Fattening Pig Barn*; Videncenter for Svineproduktion Den Rullende Afprøvning: Copenhagen, Denmark, 2012.
44. Christensen, J.H.; 1997. The Danish Eulerian hemispheric model—A three-dimensional air pollution model used for the Arctic. *Atmos. Environ.* **1997**, *31*, 4169–4191.
45. Brandt, J.; Silver, J.; Frohn, L.; Geels, C.; Gross, A.; Hansen, A.; Hansen, K.; Hedegaard, G.; Skjøth, C.; Villadsen, H.; et al. An integrated model study for Europe and North America using the Danish Eulerian Hemispheric Model with focus on intercontinental transport of air pollution. *Atmos. Environ.* **2012**, *53*, 156–176. https://doi.org/10.1016/j.atmosenv.2012.01.011.
46. Geels, C.; Hansen, K.M.; Christensen, J.H.; Skjøth, C.A.; Ellermann, T.; Hedegaard, G.B.; Hertel, O.; Frohn, L.M.; Gross, A.; Brandt, J. Projected change in atmospheric nitrogen deposition to the Baltic Sea towards 2020. *Atmos. Meas. Tech.* **2012**, *12*, 2615–2629. https://doi.org/10.5194/acp-12-2615-2012.
47. Plejdrup, M.S.; Gyldenkærne, S. *Spatial Distribution of Emissions to Air—The SPREAD Model*; NERI Technical Report no. FR823; National Environmental Research Institute, Aarhus University: Aarhus, Denmark, 2011; p. 72. http://www.dmu.dk/Pub/FR823.pdf. (accessed on 1 September 2022).
48. Plejdrup, M.S.; Nielsen, O.-K.; Brandt, J. Spatial emission modelling for residential wood combustion in Denmark. *Atmos. Environ.* **2016**, *144*, 389–396. https://doi.org/10.1016/j.atmosenv.2016.09.013.
49. Skjøth, C.A.; Geels, C.; Berge, H.; Gyldenkærne, S.; Fagerli, H.; Ellermann, T.; Frohn, L.M.; Christensen, J.; Hansen, K.M.; Hertel, O. Spatial and temporal variations in ammonia emissions—A freely accessible model code for Europe. *Atmos. Meas. Tech.* **2011**, *11*, 5221–5236. https://doi.org/10.5194/acp-11-5221-2011.
50. Geels, C.; Andersen, H.V.; Skjøth, C.A.; Christensen, J.H.; Ellermann, T.; Løfstrøm, P.; Gyldenkærne, S.; Brandt, J.; Hansen, K.M.; Frohn, L.M.; et al. Improved modelling of atmospheric ammonia over Denmark using the coupled modelling system DAMOS. *Biogeosciences* **2012**, *9*, 2625–2647. https://doi.org/10.5194/bg-9-2625-2012.
51. Arnold, J.G.; Srinivasan, R. A Continuous Catchment-Scale Erosion Model. *Model. Soil Eros. Water* **1998**, *55*, 413–427. https://doi.org/10.1007/978-3-642-58913-3_31.
52. Thodsen, H.; Andersen, H.E.; Blicher-Mathiesen, G.; Trolle, D. The combined effects of fertilizer reduction on high risk areas and increased fertilization on low risk areas, investigated using the SWAT model for a Danish catchment. *Acta Agric. Scand. Sect. B-Soil Plant Sci.* **2015**, *65*, 217–227. https://doi.org/10.1080/09064710.2015.1010564.
53. Trolle, D.; Nielsen, A.; Rolighed, J.; Thodsen, H.; Andersen, H.; Karlsson, I.; Refsgaard, J.; Olesen, J.; Bolding, K.; Kronvang, B.; et al. Projecting the future ecological state of lakes in Denmark in a 6 degree warming scenario. *Clim. Res.* **2015**, *64*, 55–72. https://doi.org/10.3354/cr01278.
54. Hu, F.; Bolding, K.; Bruggeman, J.; Jeppesen, E.; Flindt, M.R.; van Gerven, L.; Janse, J.H.; Janssen, A.B.G.; Kuiper, J.J.; Mooij, W.M.; et al. FABM-PCLake—Linking aquatic ecology with hydrodynamics. *Geosci. Model Dev.* **2016**, *9*, 2271–2278. https://doi.org/10.5194/gmd-9-2271-2016.
55. Andersen, T.K.; Nielsen, A.; Jeppesen, E.; Hu, F.; Bolding, K.; Liu, Z.; Søndergaard, M.; Johansson, L.S.; Trolle, D. Predicting ecosystem state changes in shallow lakes using an aquatic ecosystem model: Lake Hinge, Denmark, an example. *Ecol. Appl.* **2020**, *30*, 2160. https://doi.org/10.1002/eap.2160.
56. Windolf, J.; Thodsen, H.; Troldborg, L.; Larsen, S.E.; Bøgestrand, J.; Ovesen, N.B.; Kronvang, B. A distributed modelling system for simulation of monthly runoff and nitrogen sources, loads and sinks for ungauged catchments in Denmark. *J. Environ. Monit.* **2011**, *13*, 2645–2658. https://doi.org/10.1039/c1em10139k.
57. Nilsson, J.; Grennfelt, P. Critical Loads for Sulphur and Nitrogen. Report from Skokloster Workshop. Skokloster, Sweden. 1998. Available online: https://doi.org/10.1007/978-94-009-4003-1_11 (accessed on 1 September 2022).
58. Sliggers, J.; Kakebeeke, W. *Clearing the Air—25 Years of the Convention on Long-Range Transboundary Air Pollution, Economic Commission for Europe*; United Nations: Geneva, Switzerland, 2004, ISBN 92-1-116910-02005.
59. Bak, J.L. Critical Loads for Nitrogen Based on Criteria for Biodiversity Conservation. *Water Air Soil Pollut.* **2014**, *225*, 2180. https://doi.org/10.1007/s11270-014-2180-x.

60. De Vries, W.; Wamelink, G.W.W.; van Dobben, H.; Kros, J.; Reinds, G.J.; Mol-Dijkstra, J.P.; Smart, S.M.; Evans, C.D.; Rowe, E.C.; Belyazid, S.; et al. Use of dynamic soil–vegetation models to assess impacts of nitrogen deposition on plant species composition: An overview. *Ecol. Appl.* **2010**, *20*, 60–79. https://doi.org/10.1890/08-1019.1.
61. Rowe, E.; Jones, L.; Dise, N.; Evans, C.; Mills, G.; Hall, J.; Stevens, C.; Mitchell, R.; Field, C.; Caporn, S.; et al. Metrics for evaluating the ecological benefits of decreased nitrogen deposition. *Biol. Conserv.* **2017**, *212*, 454–463. https://doi.org/10.1016/j.biocon.2016.11.022.
62. Bobbink, R.; Hicks, K.; Galloway, J.; Spranger, T.; Alkemade, R.; Ashmore, M.; Bustamante, M.; Cinderby, S.; Davidson, E.; Dentener, F.; et al. Global assessment of nitrogen deposition effects on terrestrial plant diversity: A synthesis. *Ecol. Appl.* **2010**, *20*, 30–59. https://doi.org/10.1890/08-1140.1.
63. Erichsen, A.C.; Timmermann, K.; Christensen, J.P.A.; Kaas, H.; Markager, S.; Møhlenberg, F. *Development of Models and Methods to Support the Danish River Basin Management Plans*; Scientific documentation; Aarhus University Department of Bioscience and DHI: Aarhus, Denmark, 2017; p. 191.
64. Hansen, M.J.; Nyord, T.; Hansen, L.B.; Martinsen, L.; Hasler, B.; Jensen, P.K.; Melander, B.; Thomsen, A.G.; Poulsen, H.D.; Lund, P.; et al. *Miljøteknologier i det Primære Jordbrug: Driftsøkonomi og Miljøeffektivitet*; DCA Rapport, nr. 29; DCA—Nationalt Center for Fødevarer og Jordbrug. *Aarhus,* Denmark, 2013; Volume 29.
65. Latacz-Lohmann, U. *Economic Analysis of Ammonia Regulation in Germany (SchleswigHolstein) in Relation to the Hab-itat Directive*; Final report, 21 November 2017; Department of Agricultural Economics, University of Kiel: Kiel, Germany, 2017.
66. Jacobsen, B.H. Why is acidification of slurry a success only in Denmark? Transfer of environmental technology across borders. In Proceedings of the IFMA Congress, Edinburgh, UK, 2–7 July 2017, ISBN:978-92-990062-5-2.
67. Amann, M.; Kiesewetter, G.; Schöpp, W.; Klimont, Z.; Winiwarter, W.; Cofala, J.; Rafaj, P.; Höglund-Isaksson, L.; Gomez-Sabriana, A.; Heyes, C.; et al. Reducing global air pollution: The scope for further policy interventions. *Philos. Trans. R. Soc. A Math. Phys. Eng. Sci.* **2020**, *378*, 20190331. https://doi.org/10.1098/rsta.2019.0331.
68. Skjøth, C.A.; Geels, C. The effect of climate and climate change on ammonia emissions in Europe. *Atmos. Meas. Tech.* **2013**, *13*, 117–128. https://doi.org/10.5194/acp-13-117-2013.
69. Holst, G.; Thygesen, M.; Pedersen, C.B.; Peel, R.G.; Brandt, J.; Christensen, J.H.; Bønløkke, J.H.; Hertel, O.; Sigsgaard, T. Ammonia, ammonium, and the risk of asthma: A register-based case–control study in Danish children. *Environ. Epidemiol.* **2018**, *2*, e019. https://doi.org/10.1097/ee9.0000000000000019.
70. Héroux, M.-E.; Anderson, H.R.; Atkinson, R.; Brunekreef, B.; Cohen, A.; Forastiere, F.; Hurley, F.; Katsouyanni, K.; Krewski, D.; Krzyzanowski, M.; et al. Quantifying the health impacts of ambient air pollutants: Recommendations of a WHO/Europe project. *Int. J. Public Health* **2015**, *60*, 619–627. https://doi.org/10.1007/s00038-015-0690-y.
71. Raaschou-Nielsen, O.; Thorsteinson, E.; Antonsen, S.; Holst, G.J.; Sigsgaard, T.; Geels, C.; Frohn, L.M.; Christensen, J.H.; Brandt, J.; Pedersen, C.B.; et al. Long-term exposure to air pollution and mortality in the Danish population a nationwide study. *Eclinicalmedicine* **2020**, *28*, 100605. https://doi.org/10.1016/j.eclinm.2020.100605.
72. Ellermann, T.; Nordstrøm, C.; Brandt, J.; Christensen, J.; Ketzel, M.; Massling, A.; Bossi, R.; Frohn, L.M.; Geels, C.; Solvang Jensen, S.; et al. Luftkvalitet 2019. In *Status for den Nationale Luftkvalitetsovervågning*; Aarhus Universitet, DCE—Nationalt Center for Miljø og Energi: Aarhus, Denmark, 2020.
73. Cameira, M.D.R.; Valente, F.; Li, R.; Surgy, S.; Abreu, F.G.; Coutinho, J.; Fangueiro, D. Band application of acidified slurry as an alternative to slurry injection in Mediterranean winter conditions: Impact on nitrate leaching. *Soil Tillage Res.* **2018**, *187*, 172–181. https://doi.org/10.1016/j.still.2018.12.001.
74. Ottosen, L.D.; Poulsen, H.V.; Nielsen, D.A.; Finster, K.; Nielsen, L.P.; Revsbech, N.P. Observations on microbial activity in acidified pig slurry. *Biosyst. Eng.* **2009**, *102*, 291–297. https://doi.org/10.1016/j.biosystemseng.2008.12.003.
75. Fangueiro, D.; Hjorth, M.; Gioelli, F. Acidification of animal slurry—A review. *J. Environ. Manag.* **2015**, *149*, 46–56. https://doi.org/10.1016/j.jenvman.2014.10.001.
76. Edesi, L.; Talve, T.; Akk, E.; Võsa, T.; Saue, T.; Loide, V.; Vettik, R.; Plakk, T.; Tamm, K. Effects of acidified pig slurry application on soil chemical and microbiological properties under field trial conditions. *Soil Tillage Res.* **2020**, *202*, 104650. https://doi.org/10.1016/j.still.2020.104650.
77. Eriksen, J.; Sørensen, P.; Elsgaard, L. The Fate of Sulfate in Acidified Pig Slurry during Storage and Following Application to Cropped Soil. *J. Environ. Qual.* **2008**, *37*, 280–286. https://doi.org/10.2134/jeq2007.0317.
78. Fazili, I.S.; Jamal, A.; Ahmad, S.; Masoodi, M.; Khan, J.S.; Abdin, M.Z. Interactive Effect of Sulfur and Nitrogen on Nitrogen Accumulation and Harvest in Oilseed Crops Differing in Nitrogen Assimilation Potential. *J. Plant Nutr.* **2008**, *31*, 1203–1220. https://doi.org/10.1080/01904160802134905.
79. Mathot, M.; Mertens, J.; Verlinden, G.; Lambert, R. Positive effects of sulphur fertilisation on grasslands yields and quality in Belgium. *Eur. J. Agron.* **2008**, *28*, 655–658. https://doi.org/10.1016/j.eja.2007.12.004.
80. Roden, E.E.; Edmonds, J.W. Phosphate mobilization in iron-rich anaerobic sediments: Microbial Fe(III) oxide reduc-tion versus iron-sulfide formation. *Arch. Für Hydrobiol.* **1997**, *139*, 347–378.
81. Ellermann, T.; Nygaard, J.; Christensen, J.H.; Løfstrøm, P.; Geels, C.; Nielsen, I.E.; Poulsen, M.B.; Monies, C.; Gyldenkærne, S.; Brandt, J.; et al. Nitrogen Deposition on Danish Nature. *Atmosphere* **2018**, *9*, 447. https://doi.org/10.3390/atmos9110447.
82. Molina-Navarro, E.; Andersen, H.E.; Nielsen, A.; Thodsen, H.; Trolle, D. Quantifying the combined effects of land use and climate changes on stream flow and nutrient loads: A modelling approach in the Odense Fjord catchment (Denmark). *Sci. Total. Environ.* **2018**, *621*, 253–264. https://doi.org/10.1016/j.scitotenv.2017.11.251.

83. Andersen, R.C. *Undersøgelse af DMI´s Nedbørsdata til Anvendelse for Hydrologiske Formål*; DMI-rapport 21-40; Undersøgelser af DMI's nedbørsdata til anvendelse for hydrologiske formal; Danmarks Meteorologiske Institut: Copenhagen, Denmark, 2021; p. 33.
84. Grimvall, A.; Stålnacke, P.; Tonderski, A. Time scales of nutrient losses from land to sea—A European perspective. *Ecol. Eng.* **2000**, *14*, 363–371. https://doi.org/10.1016/s0925-8574(99)00061-0.
85. Bouraoui, F.; Grizzetti, B. Long term change of nutrient concentrations of rivers discharging in European seas. *Sci. Total. Environ.* **2011**, *409*, 4899–4916. https://doi.org/10.1016/j.scitotenv.2011.08.015.
86. Højberg, A.L.; Windolf, J.; Børgesen, C.D.; Troldborg, L.; Tornbjerg, H.; Blicher-Mathiesen, G.; Kronvang, B.; Thodsen, H.; Ernstsen, V. *National Kvælstofmodel. Oplandsmodel til Belastning og Virkemidler*; Geological Survey of Denmark and Greenland: Copenhagen, Denmark, 2015. (In Danish).
87. Riemann, B.; Carstensen, J.; Dahl, K.; Fossing, H.; Hansen, J.W.; Jakobsen, H.; Josefson, A.B.; Krause-Jensen, D.; Markager, S.; Stæhr, P.A.; et al. Recovery of Danish Coastal Ecosystems After Reductions in Nutrient Loading: A Holistic Ecosystem Approach. *Estuaries Coasts* **2015**, *39*, 82–97. https://doi.org/10.1007/s12237-015-9980-0.
88. Jones, H.P.; Schmitz, O.J. Rapid Recovery of Damaged Ecosystems. *PLoS ONE* **2009**, *4*, e5653. https://doi.org/10.1371/journal.pone.0005653.
89. Middelburg, J.J.; Levin, L.A. Coastal hypoxia and sediment biogeochemistry. *Biogeosciences* **2009**, *6*, 1273–1293. https://doi.org/10.5194/bg-6-1273-2009.
90. Foged, H. *Staldforsuring som Instrument til Implementering af Dansk Klimapolitik*; Organe Institute: Skødstrup, Denmark, 2020.
91. Jacobsen, B.H. *Omkostninger Ved Virkemidler der Kan Bidrage til Reduktion af Ammoniakemission i 2020 og 2030, 35 s*; IFRO Udredning, nr. 2019/05; Institut for Fødevare- og Ressource Økonomi, Københavns Universitet: Copenhagen, Denmark, 2019.