Nitrogen deposition around dairy farms: spatial and temporal patterns

Executive summary

Picture by Bram Ebben

Authors:

Albert Tietema, Henrik Barmentlo, Emiel van Loon, Roland Bol, Bram Ebben, Tamar Tulp, Marijn Tromp, Claudia Schwennen, Lea Maas and Julia Averkamp

Ecosystem and Landscape Dynamics, Institute of Biodiversity and Ecosystem Dynamics, University of Amsterdam, Science Park 904, 1098 XH Amsterdam

Universiteit van Amsterdam
Instituut voor Biodiversiteit en Ecosysteem Dynamica

Mesdag
Stichting Mesdag-Zuivelfonds NLTO

Nitrogen deposition around dairy farms: spatial and temporal patterns

Summary (EN)

This study investigates the spatiotemporal patterns of N deposition around dairy stables. In our comprehensive investigation, we measured wet- only and bulk N deposition, atmospheric ammonia (NH_3), compared the OPS model to field measurements, used automated flux chambers to assess the bi-directional exchange of NH_3 between the plant-soil system and the atmosphere, and explored the potential of low-tech bio-monitors to quantify the impact of deposition sources.

Atmospheric NH_3 concentration and bulk N deposition displayed the same temporal and spatial patterns around the stable. Normal practices at the farm like fertilization with manure, artificial fertilizer and mowing, caused an increase in atmospheric NH_3 concentration and bulk N deposition. The NH_3 concentration and bulk N deposition originating from the stable decreased exponentially with the distance to the stable. N deposition was particularly high in the first 100 m from the stable.

Dry deposition was determined as the difference between bulk deposition and wet-only deposition. This definition does not include interactions with surface areas, vegetation and soil, such as gaseous NH_3 that is directly taken up by the plants through stomata. These interactions are very difficult to measure. Our approach for estimating these interactions was to use the outcome of the OPS model, which was validated on measurements from our two farms. This combination of measurements and model results was chosen because in this way a complex system such as deposition with processes that are difficult to measure can be understood.

An application of the OPS model, that was validated with measurements from our two farms, showed that around 91% of total emission (stable and manure) is dispersed into the higher atmosphere and transported over distances beyond 500 m. Complementary, 9% of the total emission from the stable and manure on farm fields ended as deposition in a 500 m radius circle around the stable. At (and beyond) 500 m from the stable there is still an impact from the farm. However, the contribution of one specific farm outside the 500 m circle is low compared to the background concentration of NH_3. By accounting for background values and effects of manure application, the study was able to identify the proportion of measured atmospheric NH_3 originating from the stable only. This proportion decreased with distance to the stable: at 25 m from the stable 75% of the NH_3 concentration originated from the stable, this was 25% at 500 m from the stable.

With only adding a realistic representation of the emitting stable, the spatial distribution of the NH_3 concentration modeled by a local version of the OPS model closely matched the measured data (with a R^2 of 0.9). The model only underestimated the measured concentration very close (< 15 m) to the stable. When the validated model was applied on our two farms, the outcome revealed that total dry deposition (including plant- and soil uptake) on the circle with radius 500 m was approximately 3,6 times higher than dry deposition without uptake, measured with the bulk-samplers.

Automated flux chambers were used to measure the balance between NH_3 emission and deposition (net NH_3 exchange rate) in the grassland surrounding the farm. The results showed that after fertilization with manure peaks of net NH_3 emission occurred. The accumulated net annual emission after all agricultural practices amounted to 9 kg ha^{-1}. In between these practices and in winter, the field was a small source of NH_3 of 3 kg $ha^{-1}y^{-1}$. As a result, in this year, this field acted as a small NH_3 source, mainly due to application of manure.

In this study low-tech, alternative methods to measure N deposition were evaluated. Several types of bio-monitors were included. Criteria were the comparison with (1) measured patterns of atmospheric NH_3 concentration and N deposition around the farm and with (2) measured values of total deposition from the literature. Two out of three experiments with ryegrass bio-monitors around the stable, showed a comparable pattern as atmospheric NH_3 concentration and bulk deposition. However, the estimates of total N deposition on the various sites were far too high if compared to reported values in the literature. These high values might be explained by unrealistic upscaling assumptions. The ratio of N stable isotopes ^{15}N and ^{14}N in biomass formed around the farms (by moss, ryegrass and periphyton) was clearly linked to the importance of NH_3 in emissions. This indicated a potential successful application as a bio-monitor of the source of deposition.

Stikstof depositie rond melkveebedrijven: ruimtelijke en temporele patronen

Samenvatting (NL)

In deze studie hebben we in de periode van 2000 tot en met 2002 de ruimtelijke en temporele patronen van N-depositie rondom melkveestallen onderzocht. In ons uitvoerige onderzoek hebben we de natte en totale N-depositie gemeten, evenals atmosferisch ammoniak (NH_3). We hebben het OPS-model vergeleken met veldmetingen, gebruikgemaakt van geautomatiseerde fluxkamers om de uitwisseling van NH_3 tussen het plant-bodemsysteem en de atmosfeer te evalueren, en de mogelijkheden van bio-monitors onderzocht om depositiebronnen te kwantificeren.

Atmosferische NH_3 concentratie en bulk N depositie vertoonden dezelfde temporele en ruimtelijke patronen rond de stal. Normale activiteiten op het bedrijf, zoals bemesting met drijfmest, kunstmest en maaien, zorgden voor een toename van de atmosferische NH_3 concentratie en bulk N depositie. De NH_3 concentratie en bulk N depositie afkomstig van de stal namen exponentieel af met de afstand tot de stal. Vooral de eerste 100 m vanaf de stal was de N depositie hoog.

Droge depositie werd gemeten als het verschil tussen bulkdepositie en natte N depositie. Interacties met oppervlaktes, vegetatie en bodem vallen niet onder deze definitie, zoals ammoniakgas (NH_3) dat

rechtstreeks door de planten wordt opgenomen via de huidmondjes. Deze interacties zijn erg moeilijk te meten. Onze aanpak voor het schatten van deze interacties was om de uitkomst van het OPS-model te gebruiken, dat werd gevalideerd op metingen van onze twee boerderijen. Er is gekozen voor deze combinatie van metingen en modelresultaten omdat op deze manier een complex systeem als N depositie met moeilijk meetbare processen kan worden begrepen.

Na een toepassing van het OPS-model, gevalideerd op metingen van onze twee boerderijen, bleek dat ongeveer 91 % van de totale emissie (stal en mest) wordt verspreid in de hogere atmosfeer en over afstanden van meer dan 500 m wordt getransporteerd. Dit betekent dat 9% van de totale emissie van de stal en mest op het veld valt als N depositie in een straal van 500 m rond de stal. Op 500 m (en verder) van de stal heeft de boerderij nog een effect van de boerderij, maar de bijdrage van één specifiek bedrijf buiten de 500 m-cirkel is echter laag ten opzichte van de achtergrondconcentratie van NH_3. Door rekening te houden met achtergrondwaarden en effecten van bemesting, kon in het onderzoek worden bepaald welk deel van de gemeten atmosferische NH_3 afkomstig was uit de stal. Dit aandeel nam af met de afstand tot de stal: op 25 m van de stal was 75% van de NH_3-concentratie afkomstig van de stal, terwijl op 500 m nog 25% van de NH_3 concentratie afkomstig was van de stal.

Door alleen een realistische weergave van de emitterende stal aan het model toe te voegen, kwam de ruimtelijke verdeling van de NH_3 concentratie gemodelleerd door een lokale versie van het OPS model goed overeen met de gemeten gegevens (R^2 = 0,9). Het model heeft wel de gemeten concentratie zeer dicht (< 15 m) bij de stal onderschat. Bij het toepassen van het gevalideerde model op onze twee boerderijen, bleek dat de gemodelleerde totale droge depositie (dus inclusief plant- en bodem opname) in de cirkel met straal 500 m, in totaal 3,6 keer hoger was dan de droge depositie gemeten met de bulk-sampler zonder opname.

Automatische fluxkamers zijn gebruikt om de balans tussen NH_3 emissie en depositie (netto NH_3 uitstoot) te meten in het grasland rondom het bedrijf. Uit de resultaten bleek dat na bemesting met drijfmest pieken in de netto NH_3 uitstoot werden gemeten. De geaccumuleerde netto NH_3 uitstoot na alle landbouw activiteiten bedroeg 9 kg N ha^{-1} j^{-1}. Tussen deze activiteiten door en in de winter was het veld een bron van NH_3 van 3 kg N ha^{-1} j^{-1}, dus een totale emissie vanuit het grasland zelf van 12 kg ha^{-1} j^{-} Hierdoor fungeerde het grasland over het jaar als een NH_3-bron voornamelijk als gevolg van bemesting, die overigens zeer beperkt is vergeleken met de emissie vanuit de stal.

In deze studie werden low-tech, alternatieve methoden om N depositie te meten geëvalueerd. Er werden verschillende soorten bio-monitors meegenomen in deze studie. Criteria voor succes waren de vergelijking met (1) gemeten patronen van atmosferische NH_3 concentratie en N depositie rondom het bedrijf en met (2) gemeten waarden van totale depositie uit de literatuur. Twee van de drie experimenten met raaigras bio-monitoren rond de stal lieten een vergelijkbaar patroon zien als atmosferische NH_3 concentratie en bulkdepositie. De schattingen van de totale N depositie op de verschillende locaties waren echter veel te hoog vergeleken met gemeten en gemodelleerde fluxen en gerapporteerde waardes in de literatuur. Deze hoge waarden in deze studie kunnen worden verklaard door onrealistische aannames wat betreft het opschalen van de resultaten. De verhouding van de stabiele N isotopen ^{15}N en ^{14}N in de rond de boerderijen gevormde biomassa (door mos, raaigras en periphyton) hing duidelijk samen met de dominantie van NH_3 in emissies, en dus landbouw en niet verkeer als belangrijkste bron op deze plekken. Dit duidde op een mogelijk succesvolle toepassing als bio-monitor van de bron van de depositie.

Nitrogen deposition around dairy farms: spatial and temporal patterns

Executive summary

Contents

Nitrogen deposition around dairy farms: spatial and temporal patterns

Executive summary

Preface

Nitrogen (N) deposition is currently a very topical subject in the Netherlands. Many of the Dutch nature reserves are under threat by excessive nitrogen deposition, which is why many N producing sectors are currently on hold due to problems with financing, legislation and/or permits. Dairy farms are an important source of N emissions. In 2018, RIVM reported that Dutch agriculture was responsible for 46% of Dutch N emissions, of which dairy farmers contributed a major part. At present, the policy of limiting deposition by dairy farming in order to protect nature by reducing emissions, is dominated by reducing cattle and farmers. The "Stichting Mesdag-Zuivelfonds NLTO", a fund representing the interests of dairy farmers, has approached us, the Institute for Biodiversity and Ecosystem Dynamics (IBED) of the University of Amsterdam, to conduct research in this area. This report is the final report of this study.

The research aimed to answer the following questions.

(1) What are the temporal and spatial patterns of atmospheric ammonia (NH_3) concentration and N deposition around two farms?
(2) Is a fertilized grassland a source or sink of NH_3?
(3) Can we simulate the measured local patterns measured at (1) with the OPS model?
(4) Which part of the emitted NH_3 is deposited within the circle with a radius of 100 meters, as well as within the area with radius between 100 to 500 meters around the farm?
(5) Can we measure N deposition around a farm and in nature areas with bio-monitors as a low-tech method?
(6) Can we use the natural abundance of N isotopes in biomass for source determination?
(7) Can we use periphyton and plankton in ditches as bio-indicators of N deposition?

After a short introduction to the problem, this final report consists of extended summaries of seven scientific papers, each addressing one of the above mentioned questions. The papers themselves are confidential until published in open-access, peer-reviewed, scientific journals.

1. Introduction

1.1 The nitrogen problem

Our atmosphere consists of 80% of nitrogen (N) in the form of N_2 (Söderlund & Svensson, 1976). In this form, N is not reactive due to the triple covalent bond that needs a high amount of energy to break. Anthropogenic activities result in an increase in the environmental abundance of reactive N molecules (N_r) that are biologically available (Galloway et al., 2003). The global amount of N_r in the environment has more than doubled in the last century. This increase of environmental N_r results in environmental effects like soil acidification, bio-diversity loss and global warming as a result of N_2O emissions (Erisman et al., 2011).

1.2 The nitrogen cycle

Reactive N can disperse through a variety of pathways, and many factors influence its fluxes, rendering the system complex, diverse and dynamic (De Vries et al. 2003). N_r occurs in several forms, the most important being: (i) in oxidized form as NO_x, which is predominantly emitted by transport, construction and industry; and (ii) in reduced form as NH_y, which is mainly emitted by agriculture. Industrial fixation of N for fertilizer production is another major source of N_r compounds. Natural N_r is often the product of wildfires, lightning and biological fixation of N_2. Denitrification by aerobic bacteria can in turn transform N_r, via N_2O, back to N_2.

Reactive N compounds can either become aerosols or dissolve in water. Aerosol N_r can subsequently be transported through wind and brought back to the surface by N deposition. Reactive N can be deposited through wet- or dry deposition, meaning respectively through either dissolving in precipitation, fog or snow or via airflow and then gaseous/aerosol exchange between atmosphere and organisms or soil/water (Díaz-Álvarez et al. 2018). Besides by atmospheric deposition and fixating organisms, soils and surface waters are enriched with N_r through a variety of pathways. Examples are surface runoff, eutrophication of ground water and water currents that cross ecosystem borders (Oenema et al. 2005).

Deposited N_r can be taken up by ecosystems and removed by cropping or grazing, or it can leach into surface- or groundwaters. The N_r that cannot be taken up by ecosystems is called the surplus, which can accumulate, disperse past ecosystem borders and entail adverse effects if abundantly present. According to Jacobsen et al. (2019), a large part of N_r that ends up in the environment originates from agricultural surpluses. Thus, in order to optimize the protection of earth's geo-ecosystems, an adequate understanding and quantification of the biogeochemical fluxes of N_r in agricultural plots is essential.

1.3 Nitrogen in the Netherlands

A majority of the 75 habitat types present in the Netherlands can be harmed or drastically altered as a result of excessive deposited N. To quantify this impact, the Critical N load values for Dutch Natura 2000

habitat types have been calculated (Hettelingh, 2017). Critical N load is defined as the threshold above which there is a risk that the habitat is significantly altered or damaged. In 2018, the critical N loads were exceeded in circa 70% of the Netherlands (CBS et al., 2022). Many biomes already suffer from eutrophication and acidification as a result of continuous excessive N deposition. RIVM reported in 2021 that around 46% of the amount of deposited N_r in Natura 2000 habitats originated from agricultural activities (RIVM, 2021). Partly, this can be explained by the fact that Natura 2000 habitats in the Netherlands are relatively small and often neighboring intensively used agricultural land, inducing an increased edge effect (Wuyts et al. 2009).

1.4 Measuring N deposition and emission

In this explanatory study, the focus was on monitoring N deposition and emission close to a dairy farm. Deposition consists of several components that are difficult to measure directly and separately. This section 1.4 deals with the methods we used in this study and is not aimed at giving an overview of existing methods measuring N deposition and emission.

To address the spatial- and temporal dynamics of environmental N_r, several parameters close to two dairy farms were measured. Atmospheric NH_3, which is a precursor of the NH_3 and NH_4^+ flux to the geo-ecosphere interface, was measured using the passive sampler ALPHA (Tang et al. 2001). In addition, bulk deposition of NO_3^- and NH_4^+ was measured using a funnel on top of a bottle. Bulk N deposition consists of wet deposition (particles captured by the funnel and bottle during precipitation events) and part of dry deposition (the gaseous part that settles on the surface of the funnel and is leached into the bottle during the next precipitation event). These parts were separated by an Eigenbroth wet-only sampler (NSA 181/D HD-PE). The funnel of this sampler is normally closed by a lid on top (so nothing is captured when it is dry). This lid automatically opens if it rains, allowing only wet deposition to enter the bottle. The difference between the two measurements is dry deposition, but not all dry deposition. The other part of dry deposition is gaseous NH_3 that interacts with the geo-ecosphere, e.g. surface area interactions, uptake by vegetation through the stomata), microbes and soils. Measuring this latter part is still a challenge. In our research, bulk deposition, wet deposition directly and part of dry deposition in bulk deposition was measured, both directly and indirectly. Bio-monitors (see section 1.5 for an explanation) were used to estimate total deposition. By difference, this would give an estimate of dry deposition.

There are two important sources of NH_3 emission at a dairy farm: manure and farming practices. In the present study, all cows are located year-round in a stable in the central part of the farm. They produce manure and urine that is stored close to the stable. As a result, the stable is a year-round point-source of NH_3 volatilizing from the mixture of solid manure and urine. Secondly, a large part of the manure/urine mixture is applied as fertilizer to the production grassland around the farm about three to four times a year. In addition, the farmer adds artificial fertilizer and the grass is mowed several times a year. All these normal agricultural practices are expected to influence NH_3 emission during that moment of application. In contrast to the stable emission, this is an occasional and diffuse source and not an almost constant, point source.

Not all NH_3 emission fluxes were measured. The emission originating from the stable was not measured because the farmer added a new stable halfway our measuring period. This would have frustrated the measurements. Instead, we used data from “de kringloopwijzer”. This is an administrative tool from the government (https://mijnkringloopwijzer.nl/) that dairy farmers have to fill in every year. This datasheet with detailed information on the farm is analysed by a model with a scientific basis. Calculated annual NH_3 emission of those particular farms for the specific measuring year was used as emission from the stable. The sink/source strength (the net flux of NH_3 on the interface of soil/vegetation/atmosphere) of the surrounding grassland after applying manure and artificial fertilizer, and mowing was measured year-round with an automated flux chamber designed for NH_3.

1.5 A model validated by measurements

It is impossible to carry out nation-wide environmental monitoring to capture and evaluate ambient conditions and effects of mitigating regulations (Schrader et al. 2018). In order to achieve this, models of the behaviour of atmospheric pollutants are needed. These models can both interpolate and extrapolate the behaviour of pollutants and are able to capture nation-wide spatial and temporal variation. A prerequisite is that these models are validated with measurements, preferably on the scale the models are used. In the Netherlands, the Operational Priority Substances model (OPS), developed by RIVM, is publicly available and widely used. OPS accounts for the emission, dispersion, transport, chemical conversion and deposition processes of atmospheric substances, including NH_3, NO_x, and their secondary products. OPS is a pivotal tool used in AERIUS, which is the instrument used for N deposition calculations and monitoring of Natura2000 habitats in the Netherlands. The necessity that measurements and modelling should go hand by hand is widely accepted. In this study we validated both our measurements and the OPS model by applying the model at the scale of our measurements at the farms.

1.6 Bio-monitors

Physical and chemical monitoring of N deposition is precise but impossible to perform on a large spatial scale due to the high costs. Because of this, there is an urgent need for relatively simple and cheap ways to measure landscape impacts (Sommer, 1988). Bio-monitors and bio-indicators appear to be promising (supplementary) methods to achieve this, due to their integration of complex processes and cost-effectiveness. The terms ‘bio-monitor’ and ‘bio-indicator’ are defined as an organism that serves as a (semi-)quantitative or qualitative proxy, respectively, for environmental chemical change. Opposed to physio-chemical analyses, bio-monitors also highlight indirect ecosystem responses. Site specific research needs to be conducted on the most suitable bio-monitors for each specific area. Several types of bio-monitors and bio-indicators were added to our experimental setup to provide an absolute and a relative estimate of total deposition (Boltersdorf et al. 2014).

2. Results and discussion

This chapter titled “Results and discussion” contains extended abstracts of seven scientific papers representing our work. The full scientific papers are already or will be submitted to open-access, peer-reviewed scientific journals and thus will be fully open after their acceptance.

In the first part of this chapter (section 2.1) titled “Measuring and modelling N deposition around two farms”, the focus is on spatial-temporal patterns of N cycling in the vicinity of two dairy farms in the Netherlands. These patterns are evaluated in relation to normally occurring practices at a Dutch dairy farm. In section 2.1.1, conventional methods for measuring biweekly atmospheric ammonia concentration, bulk deposition and wet-only deposition are measured. In section 2.1.2, an automated flux-chamber to measure net NH_3 fluxes during and after agricultural practices and in the period in between is used. Then, in section 2.1.3 the outcome of the N dispersion model (OPS) applied on the two farms is evaluated. Measurements of atmospheric NH_3 concentration and bulk N deposition are used for evaluation. Finally, in section 2.1.4, all measured and modelled N fluxes at both farms are integrated and evaluated.

In the second part of this chapter (section 2.2) titled “Bio-monitors and bio-indicators measuring N deposition”, alternative methods to measure N deposition are used, the so called bio-monitors and bio-indicators. Our aim is to test if these low-tech methods lead to comparable results as the conventional methods, and if, in the future, these methods can be used to measure or give a relative measure of such complex process like N deposition. At first, in section 2.2.1, we use pots with ryegrass, raised under N poor conditions. Then, in section 2.2.2, the possibility of using the isotopic composition of biomass-N produced in mosses, ryegrass in the bio-monitors and periphyton in ditches to determine the source of N deposition is tested. Finally, in section 2.2.3, we examine the use of periphyton and phytoplankton in ditches close to the farm as bio-indicator of N deposition and availability.

2.1 Measuring and modelling nitrogen deposition around dairy farms

2.1.1 Spatiotemporal patterns of reactive nitrogen compounds in bulk deposition and the atmosphere at a dairy farm in the Netherlands

The spatiotemporal trends of local nitrogen (N) deposition related to ammonia (NH_3) emission from dairy farms remain under-explored. In order to fill this gap, the present study with measurements in 2021 and 2022 was aimed at: (i) examining year-round spatiotemporal patterns of atmospheric NH_3 concentration and bulk ammonium (NH_4^+) deposition at a dairy farm located in the northern part of the Netherlands; (ii) identifying the factors determining these patterns; (iii) evaluating the relative contributions of wet and dry deposition to the bulk deposition, and (iv) identifying the proportion of

measured atmospheric NH_3 and bulk NH_4^+ originating from the stable, by accounting for background values and effects of manure application.

Atmospheric NH_3 concentration and bulk N deposition were measured fortnightly according to the methods described in section 1.4, in four directions around the farm and at six distances varying of 15 to 500 meters to the farm. For finding factors that drive the measured variables, linear mixed effect models were used (Pinheiro & Bates, 2000).

Atmospheric NH_3 concentration decreased exponentially with distance to the stable, with average concentration of 34.0 µg NH_3-N m^{-3} at 15 meters and 5.8 µg NH_3-N m^{-3} at 500 meters (Figure 1). Our analysis revealed that atmospheric NH_3 concentrations were positively associated with temperature, farming practices and wind projections, and negatively with precipitation and distance to the stable. Temperature plays the most prominent role, likely because of the thermodynamic properties of gases, as can be seen by looking at the higher values in Figure 2 (left).

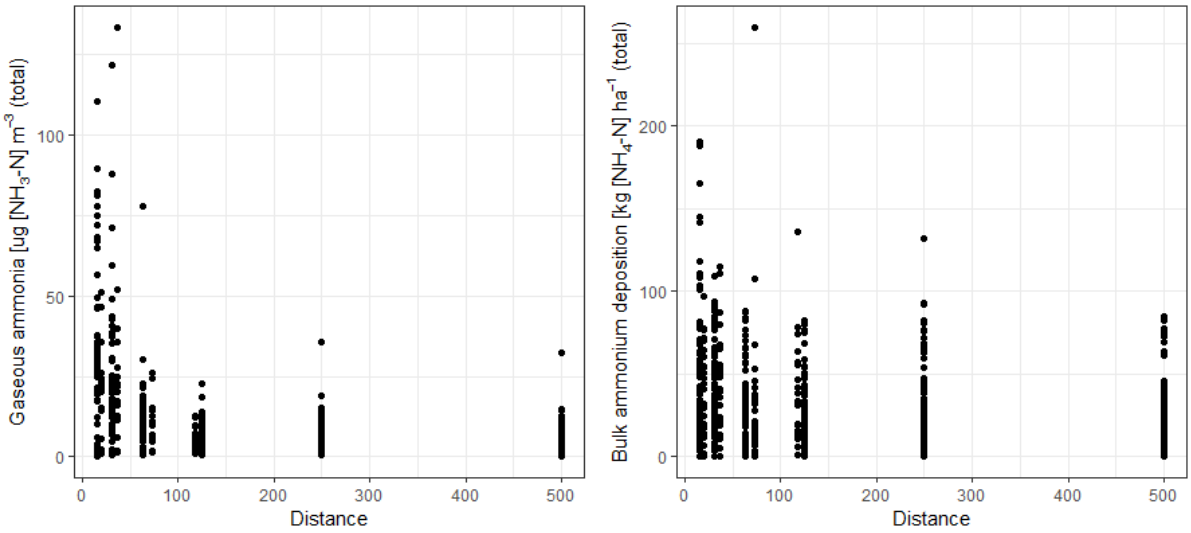

Figure 1. Atmospheric NH_3 concentration (left panel), and bulk deposition (right panel), plotted as a function of distance to the stable as point source of NH_3.

Bulk NH_4^+ deposition also displayed a sharp decrease with distance to the farm, with average values decreasing from 48.6 kg NH_4^+-N ha^{-1} y^{-1} at 15 meters to 25.9 NH_4^+-N ha^{-1} y^{-1} at 500 meters (Figure 1). The spatiotemporal dynamics of bulk NH_4^+ deposition were driven by temperature fluctuations, precipitation, farming practices, the stable's proximity, wind conditions, and seasonal variations, with the positive interaction between rainfall and farming practices emerging as the most dominant factors, likely due to flushing of atmospheric NH_3, that can be seen in Figure 2 (right).

The study found a temporal variation in the proportion of bulk deposition originating from wet and dry deposition, which was closely associated with rainfall patterns. On average, dry deposition contributed to 65% of the bulk deposition, although it should be noted that this dry deposition does not encompass all processes, as interactions with vegetation, soil and microbes are not accounted for.

The portion of measured atmospheric NH_3 concentration traced back to the dairy stable diminished from 75% near the stable to 25% at a distance of 500 meters (Figure 3). Similarly, the bulk NH_4^+-N deposition traced back to the stable decreased from 50% near the stable to 20% at 500 meters. On average, the 'background' deposition was 5.6 kg [NH_4^+-N] ha^{-1} y^{-1}, although it should be noted that this value includes the effect of farming practices.

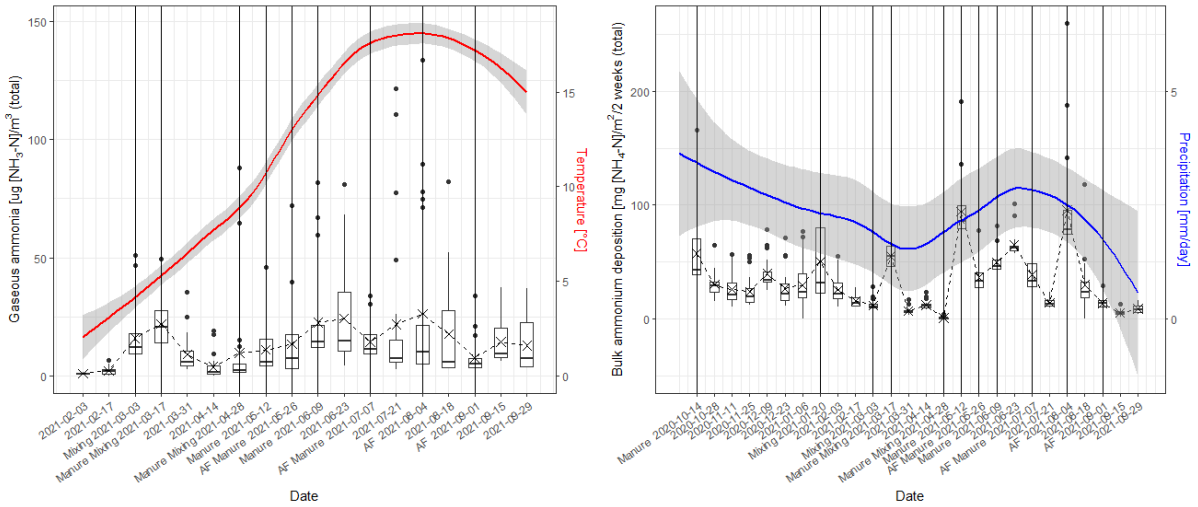

Figure 2. Atmospheric NH_3 concentration (left panel) and bulk deposition (right panel), plotted for each sampling moment. Sampling moments that had farming practices are highlighted by the vertical lines, the practices can be seen at the labels on the x-axes (AF = Artificial Fertilizer; AF Manure = Application of manure). Temperature is plotted in red (left plot, right y-axis), and precipitation is plotted in blue (right plot, right y-axis). In the boxplots, mean values are indicated by the crosses, and median values are indicated by the lines.

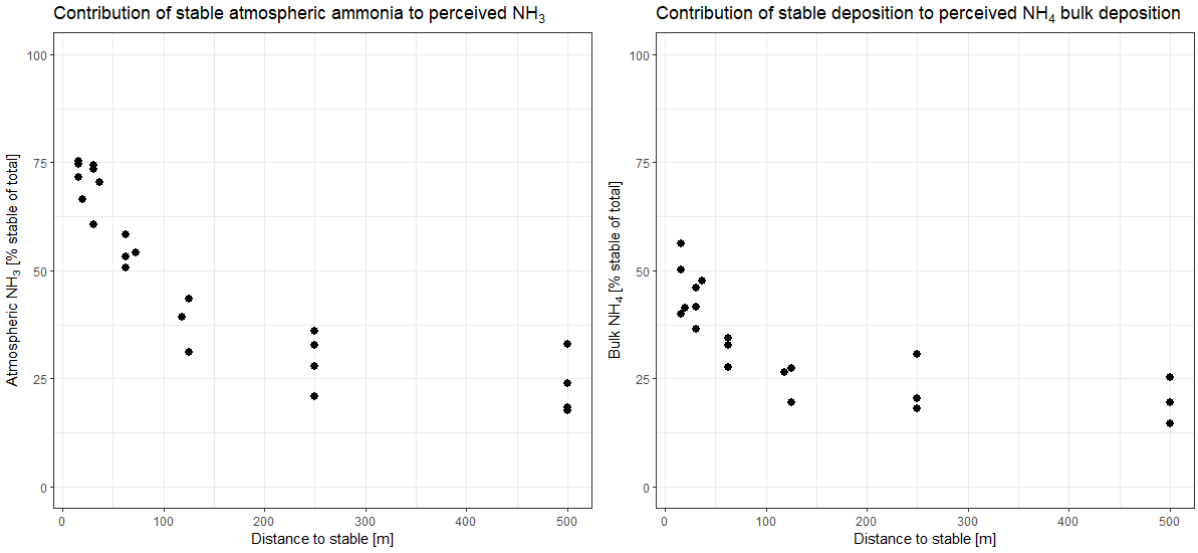

Figure 3. Average contributions of the stable to atmospheric NH_3 concentration (left panel) and bulk NH_4^+-N deposition (right panel) over distance, each point represents a measuring location.

By identifying spatiotemporal dynamics of NH_3 around a dairy farm, quantifying the impacts of farming practices, and emphasizing the importance of meteorology, the study reveals the necessity for localized investigations to tailor farm management strategies and environmental policy for the mitigation of N_r sources.

2.1.2 Assessing the net ammonia flux of a production grassland surrounding a dairy farm in the Netherlands using automated and manual closed chambers

Picture by Claudia Schwennen

Figure 4. Set up of the three automatic flux-chambers with the manual control boards (orange cases) attached to the side of each chamber. The equipment housing (green cart) containing the sensor and other peripherals was positioned behind the chambers (i.e. not in line with the stable and the chambers). The air inlet for ambient concentration measurements was attached to the equipment housing. On a regular basis fertilizer was applied to the grassland. At the same time the practices within and around the chambers were closely simulated by hand with the same amount and composition of manure and fertilizer.

The objective of this study was to quantify the spatiotemporal patterns in net NH_3 fluxes at a production grassland surrounding a dairy farm in the north of the Netherlands. For a full year, net NH_3 fluxes on the atmosphere/soil/plant interface were measured with a high frequency (Figure 4). At the dairy farm normal agricultural practices like fertilization with manure, artificial fertilizer and regular mowing took place. The NH_3 sink/source strength of the grassland was evaluated based on the timing and the extent of these practices and the time in between.

To determine the temporal variability, three automated flux chambers were employed 30 m in northeastern direction from the NH_3-emitting stable. The chambers were developed to measure NH_3 which is considered a difficult gas to work with due to its sticky behavior to many surfaces and to its high solubility in water (McGinn & Janzen, 1998; Görres et al. 2015). Each of the three chambers was measured with a frequency of once every 90 minutes. To determine spatial variation, manual flux chambers with the same dimensions were used during the second half of the growing and harvesting season at various locations.

The results showed that most of the NH_3 volatilization occurred during the growing and harvesting season, largely due to substantial emission rates directly after application of manure. Maximum fluxes of NH_3 just after application of manure in three of the four cases were 10 - 35 µg N-NH_3 m^{-2} s^{-1} (Figure 5). The maximum flux after the last application was much lower (0,5 – 1,0 µg N-NH_3 m^{-2} s^{-1}), possibly due to the fact that during that particular application atmospheric NH_3 concentration approached saturation, affecting emission. Very low net NH_3 flux rates rates occurred during fall and winter. In this period a temporal pattern of alternating net NH_3 emissions and deposition was found. In total, this led to a net NH_3-N emission of 9 kg ha^{-1} yr^{-1} at the growing and fertilizing season and 3 kg ha^{-1} yr^{-1} at the dormant season, adding up to 12 kg ha^{-1} annually. Though spatial variation had been observed, the

spatial pattern seemed to vary over time. Since background fluxes represented only a small fraction relative to fluxes measured after manure application (spread over the whole field), a more or less uniform annual net NH_3 emission of 12 NH_3-N kg ha^{-1} across the whole production grassland can be assumed.

This study suggests that this grassland, in this year, acted as a net NH_3 source. The peaks in net NH_3 emission after application of manure determined the sink/source strength. As a result, the amount, type, composition, way of applying, are all factors that can have an influence on the balance between sink and source. As net NH_3 fluxes in between the periods of practices and in the dormant season are very low and alternating between sink and source, gross emission and dry deposition rates are not expected to change substantially, affecting net NH_3 fluxes.

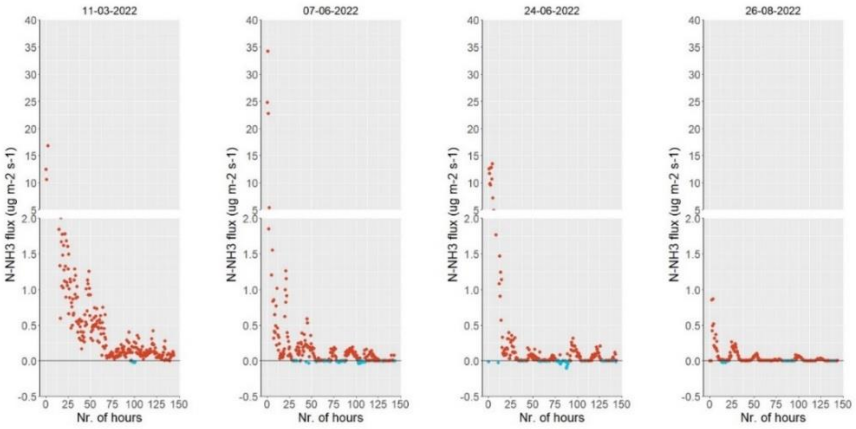

Figure 5. Course of NH_3-N fluxes in µg m^{-2} s^{-1} following each manure application during the 150 hours post-application. The red dots indicate net NH_3 emissions, the blue dots net NH_3 deposition.

2.1.3 Validating a farm-scale application of the atmospheric dispersion model OPS with local measurements of atmospheric NH_3 concentration and N bulk deposition

As part of this research we have applied the Operational Priority Substances (OPS) model to the research sites to investigate if the fine-scale atmospheric NH_3 concentration and deposition measurements can be described accurately with this model, while not calibrating this model but using it with the default parameter values and existing parameterization. OPS is an atmospheric dispersion model that is used to model dispersion, transport, chemical conversion and deposition processes of atmospheric substances for given emissions such as ammonia, nitrogen oxides, and their secondary products (Van Jaarsveld & De Leeuw, 1993; Sauter et al., 2023; https://github.com/rivm-syso/OPS). In this study, local NH_3 dispersion up to a distance of 500 m around a farm was modelled, using a realistic representation of the emitting stable in every direction. To test the sensitivity of the model, multiple scenarios mimicking these emission characteristics were implemented and the outcome of these scenarios was

compared with measurements of NH_3 atmospheric concentrations and N bulk deposition at different distances from the dairy farms.

Modelled N deposition, using an emission value of the farm reported in the "kringloopwijzer" indicated that approximately 9% of the farm's NH_3 emission (this includes emission from the stable and manure application on the field) did return as deposition within a circle with a 500 meter radius around the farm. The remaining NH_3 emission was deposited further away (with exponentially declining N deposition rates – in the 500 to 1000 meter ring +/- 15% of the emission is deposited). The measured NH_3 concentrations and those calculated by the OPS-model correlated well when studied on a fine spatio-temporal scale, although the model underestimated high concentration measurements close (15 m) to the stable. We think this difference could be explained by lower turbulence and lower wind-speeds in reality than assumed by the OPS-model. The total deposition values estimated by bulk samplers were considerably lower than when predicted by the OPS-model while the spatial patterns matched well. This comparison must be handled with care since the bulk sampler observations give a measure that is only representing a part of what is defined in the model as dry deposition. Hence, given this context, the results are consistent with what is expected theoretically (total deposition from bulk deposition observations lower than modelled total deposition). In addition, the deposition measurement with bulk samplers was associated with some uncertainties (related to material properties, handling in the field and the lab etc.). Furthermore, we can attribute part of the discrepancy between model outputs and deposition observations to limitations in the model input data: we did not incorporate spatiotemporally specific manure application in the input data files (but rather homogeneous emissions from manure in time and over the study domain).

In conclusion, with regard to NH_3 concentration in the air, the OPS-model predicts this well at fine spatio-temporal resolutions – provided that the emission is specified appropriately at a high level of detail. The current observations provide a good validation case study for this. With regard to total NH_3 deposition we think the model output is consistent with the observations, but we can't claim a good match nor any form of validation because the observed entities (bulk deposition measurements) are different from those represented in the OPS-model.

2.1.4 Integration of patterns of nitrogen fluxes at two dairy farms: comparison of measurements and model outcome

We would like to compare and integrate the measurements (section 2.1.1) with the model results (section 2.1.3) for two reasons:

1. To get a picture as complete and accurate as possible about the redistribution of N around the stable along the different pathways.
2. As an internal check on consistency and (whenever possible) also the validity of the measurements as well as model.

We will discuss both aspects here and use a schematic figure to clarify the pathways we were able to distinguish (see Figure 6).

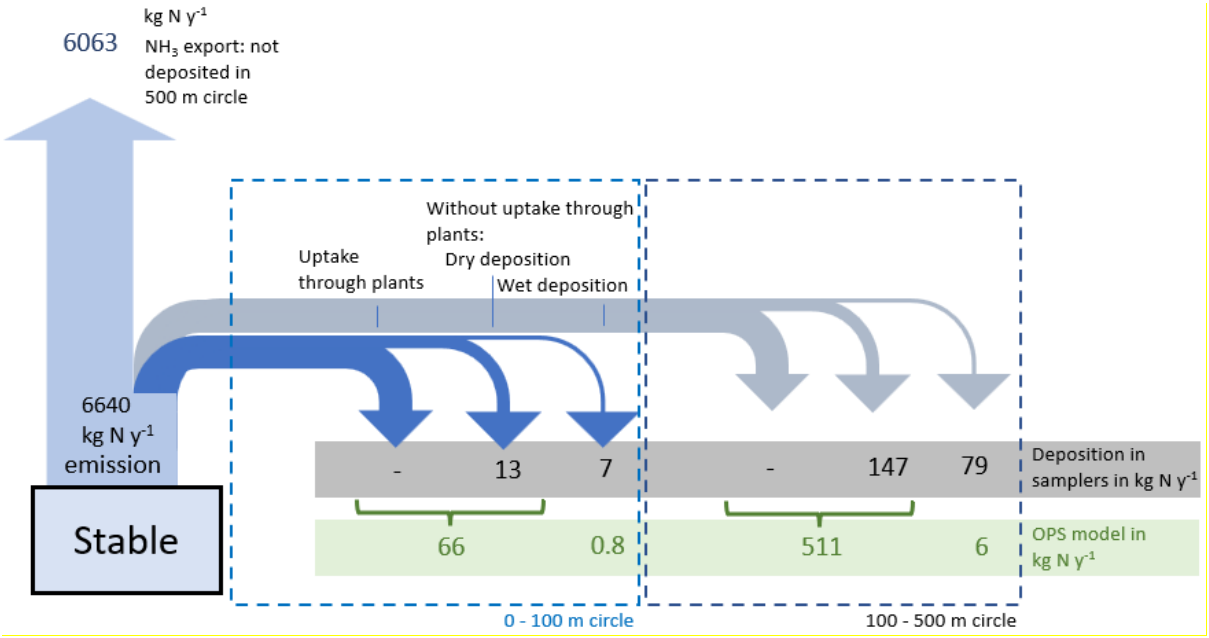

Figure 6. Schematic illustration of the different N (via NH_3) pathways from the stable to the surrounding area. The vertical upward arrow represents all the N not deposited in a 500 m radius. The two sets of horizontal arrows represent (from left to right) N that is taken-up by plants, N that is captured as dry deposition (in the bulk samplers) and N that is captured by the wet-only sampler or modeled by OPS as wet deposition. The dash means that this process, uptake by plants, could not be measured with the bulk-samplers. The blue arrows represent the part that is deposited on the land in the 100 m radius around the stable, the grey arrows represent the part that is deposited in the 100-500 m ring. The two horizontal bars (in grey and green) represent the different types of measurement and model (see the labels at the right, we will call these 'data sources'). In the two bars, the N-deposition values for the respective spatial domain are given.

The first aspect that stands-out in Figure 6 is the discretization of space: the 100 m radius, 500 m radius and everything beyond. This was an intentional simplification to make the discussion a bit easier but also to enable an aggregation of values within these spatial units, which reduces some of the variability that can be expected from individual measurements (like individual bulk samplers). Secondly, the split of dry deposition between a part that is taken-up by plants and the part that is deposited as small particles is somewhat unusual. This distinction was made to clarify conceptual differences between outcomes from the bulk samplers (which only measures dry deposition that appears as small particles) and the OPS model (which only describes the combination of the two dry deposition fluxes).

For the two different data sources (deposition samplers and OPS model – represented by the two horizontal bars in Figure 6) it is clear that from top to bottom the N deposition values should become larger: the samplers don't include the uptake by plants, while OPS does. This pattern (N dry deposition values higher in OPS model) appeared to be the case for the data collected in this study.

The relatively high values observed in the wet-only sampler (see values 7 and 79 in the grey bar in Figure 6, compared to what we see in the green bar for the OPS model, but also compared to other experiments reported in the scientific literature) can in the first place be explained by the nature of this observation. In this research there was only one wet-only sampler per farm (located very close to the stable), while

for the bulk-samplers there was a spatial array (along 4 transects up to 500 m from the stable). This implied that the background concentration could not be adequately removed from the wet-only sampler. In contrast, the wet-only values in OPS were based on the emission from the stable only. By not fully removing the background values, the N in wet-only samplers should thus be expected to be considerably higher that the values described by OPS. Another aspect which further contributed to upward biased values by the wet-only sampler is the simple way by which a spatial distribution was generated for this data: the spatial pattern seen in the bulk-only samplers was used to extrapolate the wet-only observations, using a constant ratio between dry- and wet-only as observed at the single location. It is likely that the share of wet-only N-deposition near the stable is higher than further away. In conclusion, also the discrepancy in the wet-only values deserves further study. It will need the creation of a more sophisticated extrapolation method, which would not use a constant ratio between wet- and dry-deposition but make this distribution dependent on the NH_3 concentration and possibly also rainfall properties.

We conclude that for NH_3 deposition the different data sources are consistent (smallest and largest values occur where they are expected), but that this comparison does not allow for an internal validation – some data values are too far apart. To achieve internal validation we have to explain the discrepancies among the different data sources quantitatively (possibly by new experiments and additional modelling). Currently we have explained the discrepancies qualitatively and formulated hypothesis about the reasons for the observed discrepancies.

2.2 Bio-monitors measuring N deposition

2.2.1 The challenges and opportunities of using ryegrass as bio-monitors for determining N deposition in the proximity of a dairy farm and in nature areas

Because N deposition is such a complex process, monitoring is a major challenge. Bio-monitoring is a low-tech and relatively simple method. Nevertheless, it requires a lot of man-power and chemical analyses. In this study bio-monitors for N deposition were used close to a dairy farm and in three nature areas. These bio-monitors can be a valuable method because theoretically they measure all processes involved in deposition.

The bio-monitors consisted of pots with ryegrass (Figure 7) and were placed in four directions at six different distances around a dairy farm. This way it was possible to determine a spatial pattern of total N deposition around the farm. In addition, the same bio-monitors setup was used in three nature

reserves. All bio-monitors were incubated in the field for three consecutive exposure periods of four weeks during spring/summer (farm) and autumn (nature) in 2022. Afterwards, total N deposition was calculated by determining the N balance of the bio-monitor by measuring the change in N storage in each compartment (water, soil and plant) of the bio-monitor after field incubation.

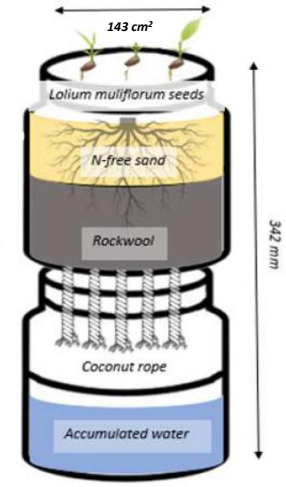

Figure 7. The bio-monitor set-up (capture area is 143 cm^2). The bio-monitor consist of two HDPE pots (3,6 L) joined together. The top half was filled with N free sand as a seedbed for 0.16 g ryegrass seeds. 5 holes were drilled into the joining section were coconut rope was pulled through as a means for excess water to drain to the bottom-half and for the plants to re-absorb it if needed. A layer of rockwool was placed between sand and the ends of the rope to increase the capillary function.

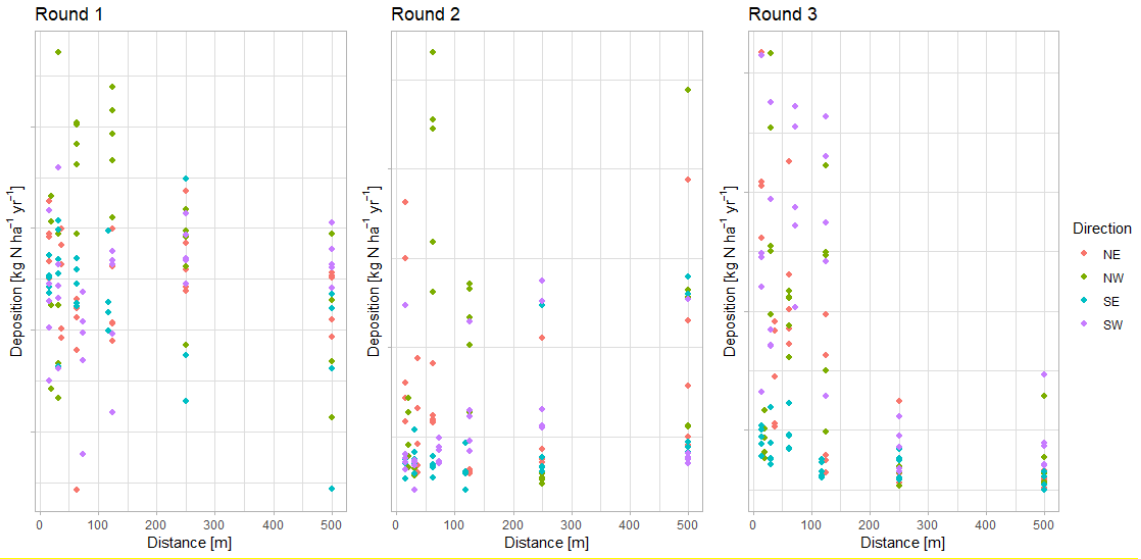

Figure 8. The estimated N deposition based on the ryegrass bio-monitors plotted against its proximity to the N source. In total 120 bio-monitors were placed at positions at various distances (± 16, 31, 62, 125, 250, 500 m) from a dairy farm in four directions (NE; northeast: NW; northwest: SE; southeast: SW; southwest). After a four-week growth period at a greenhouse under N limiting circumstances, the bio-monitors were incubated in three time periods (Round 1, 2 and 3) of four weeks (see text). After incubation, the change in N-balance was determined and the total N deposition was calculated in kg N ha^{-1} yr^{-1}. The x-axis shows the distance of the bio-monitors in relation to the farm in meters. The y-axis shows total deposition of each bio-monitor in kg N ha^{-1} yr^{-1}. The colors indicate the directions of the bio-monitors.

Linear mixed modelling showed that rainfall, temperature and bulk deposition were most significant for positively predicting total N deposition. The atmospheric NH_3 concentration was not significant in predicting deposition levels, indicating that the ryegrass had other available N sources besides atmospheric NH_3 and did not use NH_3 as a N-source (which was one of the assumptions). Only during period 1 and 3 both distance to the farm and atmospheric NH_3 concentrations were found to have a positive effect on the bio-monitor results, indicating that only during these periods grasses used the available NH_3 to grow. That the plants used the NH_3 emitted from the farm during this period was confirmed by the lowered $\delta^{15}N$ in biomass in the monitors close to the farm, and by increased biomass and N% of the ryegrass close to the farm.

The results of the bio-monitor on the farms were evaluated using two criteria; the patterns of deposition around the farms and the absolute numbers of deposition. The results of exposure period round 1 and 3 showed similar patterns as the measured atmospheric deposition with distance to the N-source, the stable. In round 2, no relation was found between total N deposition and distance to the farm. Another way of evaluating the bio-monitor method is determine if the method gives realistic values. If the absolute numbers of total deposition were compared to the other measured and modeled fluxes, it was concluded that the bio-monitor method resulted in far too high numbers.

There are a few problems with the current design of the bio-monitor setup may have influenced the absolute numbers of the total N deposition estimates. One of the problems is temperature, a limiting factor in a field incubation experiment that can't be controlled. If temperature is the limiting factor (i.e. in winter), more N availability as NH_3 or NH_4^+ might not lead to more N plant uptake. As a result, seasonality, more particular, temporal upscaling might be important affecting the absolute results, with a growing season of 7 months instead of 12, causing a decrease in total deposition . The second problem is a spatial upscaling problem. A bio-monitor can be visualized as a bulk deposition sampler with three separate compartments, water at the bottom, in between a poor sandy soil and on top a plant (ryegrass). Both water and soil have a fixed "capture area" of 143 cm^2. It is unlikely that the capture area for the plant uptake of gaseous NH_3 is the same as for the other compartments in the bio-monitor. Diffusion of NH_3 from the atmosphere to the plant is driven by a NH_3 concentration gradient that might result in a larger capture area, eventually leading to a lower total deposition estimate. Changing the gaseous NH_3 capture area into a circle around the bio-monitor with a diameter of 19,1 cm (instead of 13,4 cm), would mean a doubled capture area of 286 cm^2 (instead of 143 cm^2) and a decrease of the total deposition estimate with a factor 2. Especially this last factor is still a guess as no information in the literature was found about the size of this capture area.

However, if these two upscaling problems are solved in a new design and associated data processing, the bio-monitor method has a promising future for measuring total N deposition in a low-tech way.

2.2.2 Spatial patterns of N deposition around two dairy farms: using natural abundance of stable N isotopes in biomass as bio-indicators of the source of deposition

The possible environmental threats of N deposition clearly highlight the necessity of monitoring N deposition as a prerequisite for developing policies and management actions. However, existing methods like automated air quality monitoring networks, in combination with models are expensive to maintain and result often in uncertainties and in debates on the results. This study aimed at testing an alternative approach. The composition of stable isotopes of N (^{14}N and ^{15}N) was measured in the biomass of several bio-indicators sampled at various distances from the farm. It was hypothesized that the isotopic signature of the biomass would reveal the relative importance of NH_3 and NO_x as source for N deposition (Choi et al, 2017; Díaz-Álvarez et al. 2018). The isotopic composition of manure and of inorganic N (NH_4^+ and NO_3^-) in bulk deposition was also measured, both being important on-site sources of N in deposition. As possible on-site bio-indicators we evaluated biomass of periphyton (see also section 2.2.3), biomass of ryegrass (*Lolium multiflorum*) from the bio-monitors (see also 2.2.1) as well as bryophytes (*Brachythecium rutabulum*) (Díaz-Álvarez et al. 2019).

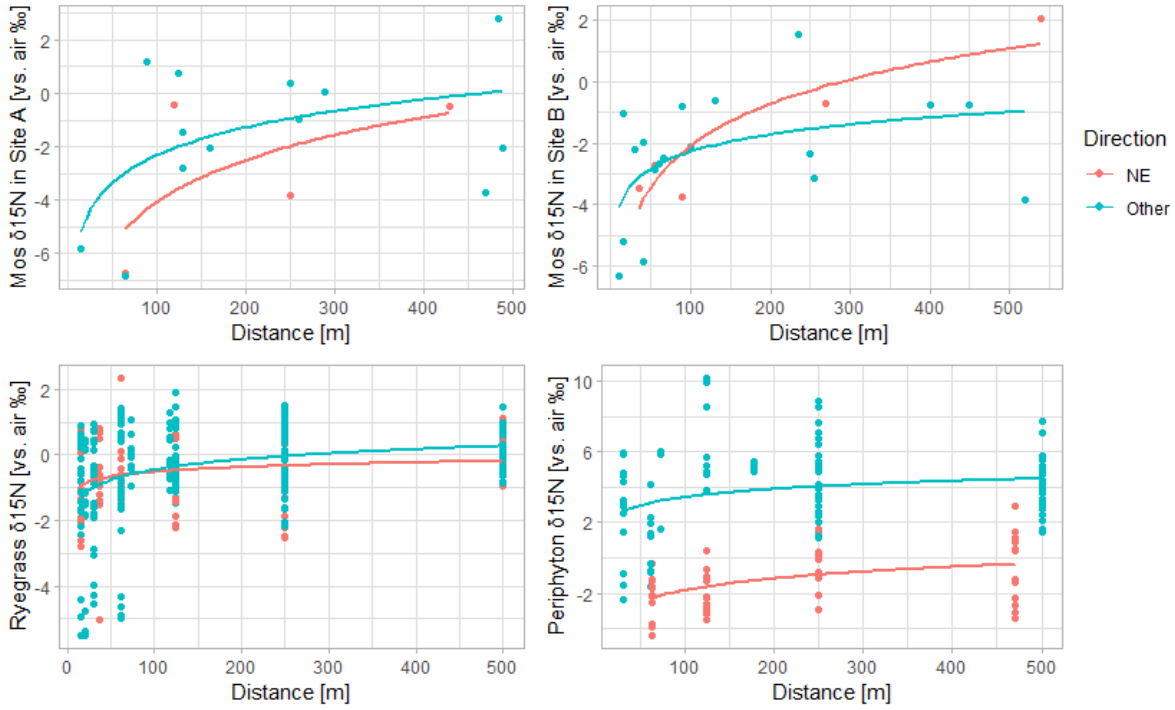

Figure 9. Natural abundance of stable N isotopes (‰ $\delta^{15}N$) in biomass as a function of distance to the farm, separately for the northeast direction (NE) and the other directions to the stable (southeast, southwest and northwest together). Site A is the site in Friesland, with only agricultural activities in the surrounding, site B is the site in Flevoland neighbouring a city and a motorway. Ryegrass and periphyton data are only sampled at site A. The lines represent a generalized additive model (GAM) smoothed curve, fitted through the individual datapoints.

We found a clear spatial gradient in all bio-indicators of lower $\delta^{15}N$ values (more ^{14}N and less ^{15}N compared to air-N_2 as a standard) at a closer distance to the farm (Figure 9). The measured pattern of $\delta^{15}N$ in biomass followed the relationship between distance to the farm and atmospheric NH_3 concentration and inorganic N concentration in bulk-deposition. The negative $\delta^{15}N$ signature in all biomass samples close to the farm was likely due to the influence of NH_3, emitted as a result of agricultural activity. Positive $\partial^{15}N$ values in manure (7,5‰) indicated that fractionation favoring $^{14}NH_3$ occurred during volatilization. This would lead to depleted ^{15}N in atmospheric NH_3. Indeed, in a review by Díaz-Álvarez et al. 2019, the authors reported that the isotopic values of atmospheric NH_3 originating from volatilization of NH_3 from manure can be as negative as −40‰, whereas the $\delta^{15}N$ in NH_4^+ and NO_3^- in bulk deposition can range from −15‰ to 15‰. We found in bulk deposition measured over a full year low negative $\partial^{15}N$ values in NO_3^- (-2,2‰) (compared to the low positive $\partial^{15}N$ values (+2.9‰) in NH_4^+), with no significant difference between the two sites. Plant available N is a mixture of soil NH_4^+ and NO_3^- in bulk deposition, soil with a $\partial^{15}N$ around zero and atmospheric NH_3 with a large negative $\partial^{15}N$ (Felix et al. 2014). We measured a higher NH_3 concentration close to the farm, as a result the relative (negative) contribution of NH_3 in plant uptake (the source), and the resulting effect on biomass, will be larger (a more negative sink) close to the farm.

Mosses, as perennials, showed a larger difference between $\delta^{15}N$ in biomass sampled close to both farms and at 500 meters distance, compared to the other studied biomass (Díaz-Álvarez et al. 2019). This was likely due to their longer exposure to NH_3 deposition and the subsequently cumulative effect (Díaz-Álvarez et al. 2019). Nevertheless, a comparable pattern was found in ryegrass (incubated 4 weeks at the farm) and even periphyton (incubated 2 weeks at the farm). Mosses were sampled at both farms. We expected a different isotopic signatures in a NH_3 dominated site in Friesland with only agricultural activity in the surrounding vs a NO_x dominated site close to a large city and motorway in Flevoland. However, this difference between the sites in natural abundance of inorganic N in bulk-deposition, as well as a different pattern of $\delta^{15}N$ in mosses was not found. Nevertheless, isotopic analysis of biomass produced at various distances to the farms was successfully applied to determine patterns of N deposition around these farms and can be used as a reliable additional or even an alternative method to models and monitoring networks.

2.2.3 Bio-monitoring of dairy farm emitted NH_3 in surface waters using phyto-plankton and periphyton

Although being quantified as an important pathway of N_r, the local spatiotemporal patterns of NH_3 emitted by dairy farming and its resulting effect on surface waters has not been quantified to a great extent (Ge et al., 2023). In order to reduce this knowledge gap, this study: (i) analysed spatiotemporal patterns in atmospheric NH_3 and aqueous NH_4^+ at a dairy farm in the Netherlands; (ii) determined meteorological variables which influenced these patterns; and (iii) assessed the suitability of periphyton and phytoplankton as bio-monitors for atmospheric NH_3 spreading and the spatial impacts of the stable.

Measurements of biomass production of periphyton and phytoplankton were conducted fortnightly between 4th August 2021 and 13th October 2021.

Similar to atmospheric NH_3 concentration (see 2.1.1), surface water concentration of NH_4^+ were significantly regulated by the stable. Concentrations of NH_4^+ were positively associated with atmospheric NH_3 concentration, and displayed a sharp decrease over distance. Interestingly, while atmospheric NH_3 concentrations in the different directions were within range of each other at 500 meters from the stable, surface water concentration at this distance in the northeast direction were roughly a factor of 6 higher compared to the other directions (3.7 mg vs 0.6 mg [NH_4^+ -N]/L), which is likely explained by the dominant wind direction (from the southwest), legacy effects and aquatic streams. For reference, eutrophication typically occurs with TN values above the 0.5-1.0 mg/L range (Camargo & Alonso, 2006). Looking at the composition of TN values in surface waters, N from NH_4^+ was dominant (median of 55%). Perceived dynamics of aforementioned ammonia nitrogen compounds corresponded with all analyzed bio-monitors. Periphyton biomass, total N and isotopic ratios all significantly reflected the spatiotemporal dynamics of atmospheric NH_3 and aqueous NH_4^+, as did the total N concentration in phytoplankton. Total N values in the bio-monitors decreased with distance to the stable, and natural abundance of ^{15}N of periphyton increased with distance (Figure 10).

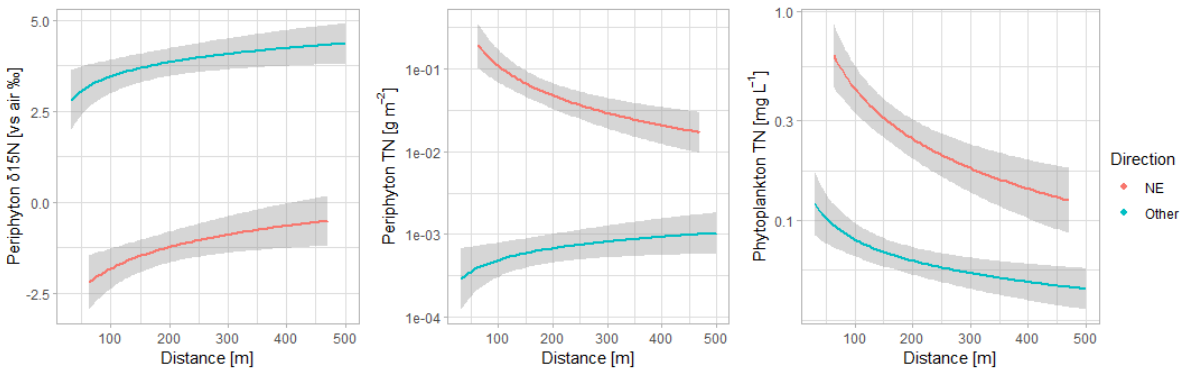

Figure 10. Bio-monitor parameters (i.e. $\delta^{15}N$ (left), periphyton TN (middle) and phytoplankton TN (right) over distance to the stable, per direction. NE = northeast and Other = southeast, southwest and northwest together. The lines represent a 'loess' curve, i.e. a non-parametric smoothing technique used to capture the underlying trend in the data without assuming a specific form. The grey area surrounding the lines represents the standard error.

In conclusion, this study found that both aquatic bio-monitors were influenced by and reflected dairy farm emitted NH_3 and that this followed a sharp dilution with distance. Our study thus strongly underlines the effect of dairy farming on local water quality via atmospheric transport of ammonia. This study shows that aquatic bio-monitoring with periphyton and phytoplankton is a promising supplementary tool to enable optimized farm management, because they can reflect (long-term) enhanced N_r deposition on small scales, and in an integrative and responsive manner.

3.0 References

Boltersdorf, S. H., Pesch, R., Werner, W. (2014) Comparative use of lichens, mosses and tree bark to evaluate nitrogen deposition in Germany. Environmental Pollution 189, 43-53

Camargo, J. A., & Alonso, Á. (2006). Ecological and toxicological effects of inorganic nitrogen pollution in aquatic ecosystems: A global assessment. *Environment International*, *32*, 831–849

CBS, PBL, RIVM, WUR (2022). Milieudruk door stikstofdepositie op landnatuur, 2020 (indicator 1592, versie 04 , 2 juni 2022). www.clo.nl. Centraal Bureau voor de Statistiek (CBS), Den Haag; PBL Planbureau voor de Leefomgeving, Den Haag; RIVM Rijksinstituut voor Volksgezondheid en Milieu, Bilthoven; en Wageningen University and Research, Wageningen

Choi, W-J., Kwak, J-H., Lim, S-S, Park, H-J., Chang, S.X., Lee, S-M., Arshad, M. A., Yun, S-I., Kim, H-Y. (2017) Synthetic fertilizer and live-stock manure differently affect ∂15N in the agricultural landscape: A review. Agriculture, Ecosystems and Environment 237, 1-15

Dämmgen, U., Erisman, J. W., Cape, J. N., Grünhage, L., & Fowler, D. (2005) Practical considerations for addressing uncertainties in monitoring bulk deposition. Environmental Pollution, 134, 535–548

David, M., Loubet, B., Cellier, P., Mattsson, M., Schjoerring, J. K., Nemitz, E., ... & Sutton, M. A. (2009). Ammonia sources and sinks in an intensively managed grassland canopy. *Biogeosciences, 6*(9), 1903-1915

De Vries, W., Kros, J., Oenema, O., & De Klein, J. (2003). Uncertainties in the fate of nitrogen II: A quantitative assessment of the uncertainties in major nitrogen fluxes in the Netherlands. Nutrient Cycling in Agroecosystems, 66, 71-102

Díaz-Álvarez, E. A., Lindig-Cisneros, R., & de la Barrera, E. (2018). Biomonitors of atmospheric nitrogen deposition: potential uses and limitations. *Conservation Physiology*, *6*(1), coy011

Díaz-Álvarez, E. A., de la Barrera, E., Arciga-Pedraza, A., Arróniz-Crespo, M. (2019) Bryophyte enzymatic responses to atmospheric nitrogen deposition: A field validation for potential biomonitors. The Bryologist, 122, 369-403

Erisman, J. W., Galloway, J., Seitzinger, S., Bleeker, A., & Butterbach-Bahl, K. (2011). Reactive nitrogen in the environment and its effect on climate change. Current Opinion in Environmental Sustainability, 3, 281-290

Felix, J. D., Elliott, E. M. , Gish, T., Maghirang, R., Cambal, L., Clougherty, J. (2014) Examining the transport of ammonia emissions across landscapes using nitrogen isotopes ratios. Atmospheric Environment 95, 563-570

Galloway, J. N., Aber, J. D., Erisman, J. W., Seitzinger, S. P., Howarth, R. W., Cowling, E. B., & Cosby, B.J. (2003) The nitrogen cascade. Bioscience, 53, 341-356

Ge, X., Schaap, M., & De Vries, W. (2023) Improving spatial and temporal variation of ammonia emissions for the Netherlands using livestock housing information and a Sentinel-2-derived crop map. *Atmospheric Environment: X*, *17*(100207)

Görres, C. M., Kammann, C., & Ceulemans, R. (2015) Automation of soil flux chamber measurements: potentials and pitfalls. *Biogeosciences Discussions, 12*(17).

Hettelingh J-P, Posch M & Slootweg J (eds.) (2017) European critical loads: database, biodiversity and ecosystems at risk, CCE Final Report 2017, Coordination Centre for Effects, RIVM Report 2017-0155, Bilthoven, Netherlands. doi: 10.21945/RIVM-2017-0155 https://www.rivm.nl/publicaties/european-critical-loads-database-biodiversity-and-ecosystems-at-risk-cce-final-report

Jacobsen, B. H., Latacz-Lohmann, U., Leusink, H., Michels, R., Stahl, L. (2019) Costs of regulating ammonia emissions from livestock farms near Natura 2000 areas - analyses of case farms from Germany, Netherlands and Denmark. Journal of Environmental Management 246, 897-908

McGinn, S. M. & Janzen, H. H. (1998) Ammonia Sources in agriculture and their measurement. Canadian Journal of Soil Science, 78, 139-148

Oenema, O., van Liere, L., & Schoumans, O. (2005) Effects of lowering nitrogen and phosphorus surpluses in agriculture on the quality of groundwater and surface water in the Netherlands. Journal of Hydrology, 304, 289-301

Pinheiro, J. C., & Bates, D. M. (2000) Mixed-Effects Models in S and S-PLUS. *Statistics and Computing*. Springer-Verlag, New York, NY

RIVM. (2021). Stikstof. From https://www.rivm.nl/stikstof. Retrieved June 2023

Sauter, F., Sterk, M., Van der Swaluw, E., Wichink Kruit, R., De Vries, W., Van Pul., A (2023) The OPS-model, Description of OPS 5.1.1.0, ops_documentation_5.1.1.0_20230705.docx, 2023-07-05, RIVM

Schrader, F., Schaap, M., Zöll, U., Kranenburg, R., Brümmer, C. (2018) The hidden cost of using low-resolution concentration data in the estimation of NH_3 dry deposition fluxes. Scientific Reports 8, 969

Sommer, S. G. (1988) A simple bio-monitor for measuring ammonia deposition in rural areas. Biology and Fertility of Soils 6, 61-64

Söderlund, R., & Svensson, B. H. (1976) The global nitrogen cycle. Ecological Bulletins, 23-73

Stokstad, E. (2019) Nitrogen crisis threatens Dutch environment—and economy. *Science, 366*, 1180-1181

Tang, Y.S., Cape, J.N., Sutton, M.A. (2001) Development and types of passive samplers for monitoring atmospheric NO_2 and NH_3. Scientific World 1, 513–529

Van Jaarsveld, J. A., & De Leeuw, F. A. (1993) OPS: An operational atmospheric transport model for priority substances. *Environmental Software, 18*(2), 91-100

Wuyts, K., De Schrijver, A., Vermeiren, A., Verheyen, K. (2009) Gradual forest edges can mitigate edge effects on throughfall deposition if their size and shape are well considered. Forest Ecology and Management 257, 679-687

Zhang, Q., Yanan, L.,Wang, M., Wang, K., Meng, F., Liu, L., Zhao, Y., Ma, L., Zhu, Q., Xu, W., Zhang, F. (2021) Atmospheric nitrogen deposition: A review of quantification methods and its spatial pattern derived from the global monitoring networks *Ecotoxicology and Environmental Safety* 2021, 112180

Acknowledgements

This study was made possible by financial support of the “Stichting Mesdag-Zuivelfonds NLTO” (MZF). We thank the board of MZF for their trust in us as UvA research team. Agricultural entrepreneurs and individual farmers who made a donation to make this research possible are greatly acknowledged. In addition, we are grateful to the farmers who were so kind to allow us on their land. We were guided by a “klankbordgroep” consisting of scientists involved in comparable research. Thanks for your always positive criticism.

This research would never have been possible without a whole team of students and interns at the University of Amsterdam. Some of them were co-authors, some helped with lab- and fieldwork or just accompanied the fieldworkers on the long trip to Friesland. Also, we had a huge amount of help of the staff of IBED, the research institute where this research was carried out.

On behalf of the initial research team Emiel, Roland, Henrik, Bram and me, thanks to everybody for their help!

Albert Tietema