



National Institute for Public Health  
and the Environment  
*Ministry of Health, Welfare and Sport*

**Implementation of a data fusion approach  
to assess the concentration and dry  
deposition of ammonia in the Netherlands**

RIVM letter report 2020-0076  
R. Wichink Kruit et al.





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## Colophon

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DOI 10.21945/RIVM-2020-0076

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Published by:

**National Institute for Public Health  
and the Environment, RIVM**

P.O. Box 1 | 3720 BA Bilthoven  
The Netherlands

[www.rivm.nl/en](http://www.rivm.nl/en)

## Synopsis

### **Implementation of a data fusion approach to assess the concentration and dry deposition of ammonia in the Netherlands**

#### **Method for determining the concentration and dry deposition of ammonia further refined**

RIVM has further refined the method to assess the concentration and dry deposition of ammonia in the Netherlands. Concentration is the amount of ammonia that is in the air. Dry deposition is the amount of ammonia that gets down from the air to the surface in dry weather. With the refined method, the measured and calculated ammonia concentrations correspond better.

#### **The method**

The new method is a post-processing on the calculated concentration and dry deposition of ammonia. The ratio between calculated and measured ammonia concentrations may differ locally. The new method takes into account these local differences better than before and adapts the calculated values to the local situation. The earlier method used a fixed, national factor for this. The local adjustments are now possible, because many measurement points have been added to the Measuring Ammonia in Nature network (MAN) in recent years.

#### **Better insight**

With the new method, differences between the measured and calculated ammonia concentrations are smaller. The calculated concentration and dry deposition are higher in the (south)west of the Netherlands with the new method. In the east of the Netherlands they are lower. The average concentration and dry deposition of ammonia in the Netherlands remain virtually the same.

Keywords: ammonia, nitrogen, deposition, Natura 2000 areas, MAN, OPS



## Publiekssamenvatting

### **Implementatie van een datafusie methode om de concentratie en droge depositie van ammoniak in Nederland te bepalen**

#### **Methode om de concentratie en droge depositie van ammoniak vast te stellen is verder verfijnd**

Het RIVM heeft de methode om de concentratie en 'droge depositie' van ammoniak in Nederland vast te stellen verder verfijnd. De concentratie is de hoeveelheid ammoniak die in de lucht zit. De droge depositie is de hoeveelheid ammoniak die bij droog weer vanuit de lucht op de bodem terecht komt. Met de verfijnde methode komen de gemeten en berekende ammoniakconcentraties nog beter overeen.

#### **De methode**

De nieuwe methode is een nabewerking op de berekende concentratie en droge depositie van ammoniak. Lokaal kan de verhouding tussen berekende en gemeten ammoniakconcentraties verschillen. De nieuwe methode houdt hier beter rekening mee dan voorheen en past de berekende waarden aan de lokale situatie aan. De eerdere methode gebruikte hiervoor een vaste, landelijke factor. De lokale aanpassing is nu mogelijk, omdat er in de afgelopen jaren veel meetpunten zijn bijgekomen in het Meetnet ammoniak in Natuurgebieden (MAN).

#### **Beter inzicht**

Met de nieuwe methode zijn lokale verschillen tussen de gemeten en berekende ammoniakconcentraties kleiner. De berekende concentratie en droge depositie met de nieuwe methode zijn hoger in het (zuid)westen van Nederland. In het oosten van Nederland zijn die juist lager. De gemiddelde concentratie en droge depositie van ammoniak over heel Nederland blijft vrijwel hetzelfde.

Kernwoorden: ammoniak, stikstof, depositie, Natura 2000-gebieden, MAN, OPS



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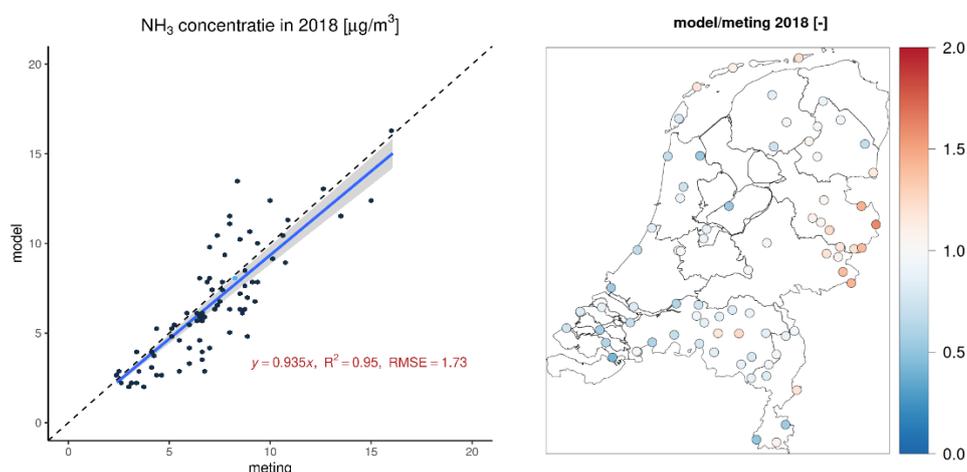
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## Samenvatting

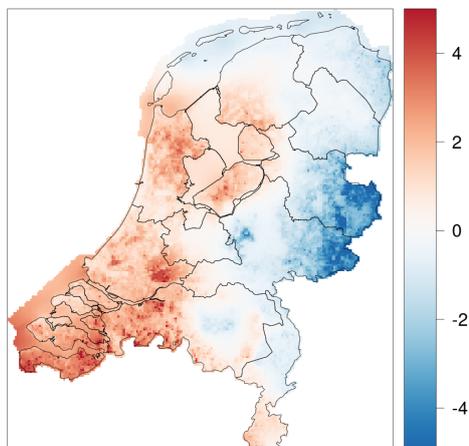
Ammoniak heeft veel betrekkelijk kleine lokale bronnen. Het is een reactieve stof waardoor er veel interactie is met andere stoffen en de bodem. Om een landsdekkend beeld te verkrijgen worden ammoniakconcentraties in de lucht berekend met een model. In zulke berekeningen zijn vergelijkingen met metingen essentieel voor een betrouwbaar resultaat. Van oudsher waren hiervoor 8 meetlocaties in het Landelijk Meetnet Luchtkwaliteit beschikbaar. De laatste jaren is het aantal meetlocaties sterk uitgebreid door metingen met samplers in het Meetnet Ammoniak in Natuur (MAN, zie <http://man.rivm.nl>). Als onderdeel van de Grootschalige Concentraties Nederland (GCN) wordt de jaargemiddelde ammoniakconcentratie en -depositie op 1x1 km berekend. Om de berekende concentraties in overeenstemming te brengen met de metingen worden ze gekalibreerd aan de jaargemiddelde concentratiemetingen. Dit gebeurt met een constante factor die over heel Nederland hetzelfde is, zie figuur 1a. Met het sterk toegenomen aantal meetpunten is te zien dat er rond dit gemiddelde ook een bepaalde spreiding is. Nadere analyse heeft geleerd dat de spreiding niet willekeurig over Nederland verdeeld is maar patronen vertoont (zie figuur 1b). Bijvoorbeeld in Zeeland kan de berekende concentratie tot 50% lager zijn dan de metingen en in oosten van Overijssel en Gelderland tot 50% hoger.



Figuur 1. De relatie tussen de gemeten ammoniakconcentraties en de berekeningen. Het linker plaatje, 1a, toont de huidige kalibratie met 1 constante nationale kalibratiefactor. Het rechterplaatje, 1b, toont de ruimtelijke verdeling van de verhouding tussen de berekeningen en de metingen.

Het gevonden patroon biedt de mogelijkheid om een meer verfijnde kalibratie op de berekeningen uit te voeren. Via de ruimtelijke interpolatie methode 'kriging' is een passend kalibratieveld uitgerekend en toegepast. Deze analyse blijkt over de laatste 5 jaar een zeer stabiel resultaat op te leveren hetgeen vertrouwen geeft in de robuustheid van de methode. De verandering in de berekende ammoniakconcentratie in 2018 is te zien in figuur 2.

verschil in  $\text{NH}_3$  concentratie nieuwe - huidige methode 2018 [ $\mu\text{g}/\text{m}^3$ ]



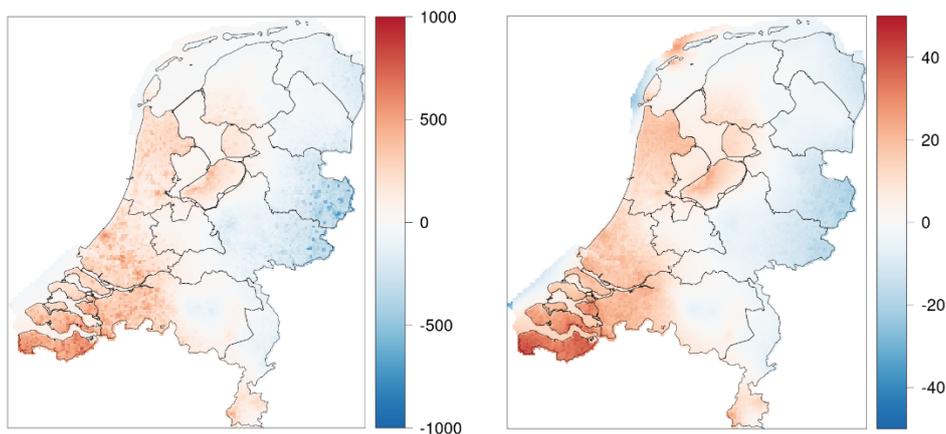
*Figuur 2. Verschilkaart van de  $\text{NH}_3$  concentratie berekend met de nieuwe ruimtelijke kalibratie methode en de huidige constante kalibratie methode.*

### Impact

De verandering van de berekende ammoniakconcentraties leidt ook tot een verandering van de berekende droge depositie van ammoniak. Dit heeft weer een verandering in de berekende totale stikstofdepositie tot gevolg (Figuur 3). 80% van de veranderingen ligt tussen de -9% en +18%. Lokaal in Zeeland en in het oosten van Gelderland en Overijssel zijn de verschillen groter.

verschil in totale N depositie nieuwe - huidige methode 2018 [mol/ha]

verschil in totale N depositie nieuwe - huidige methode 2018 [%]

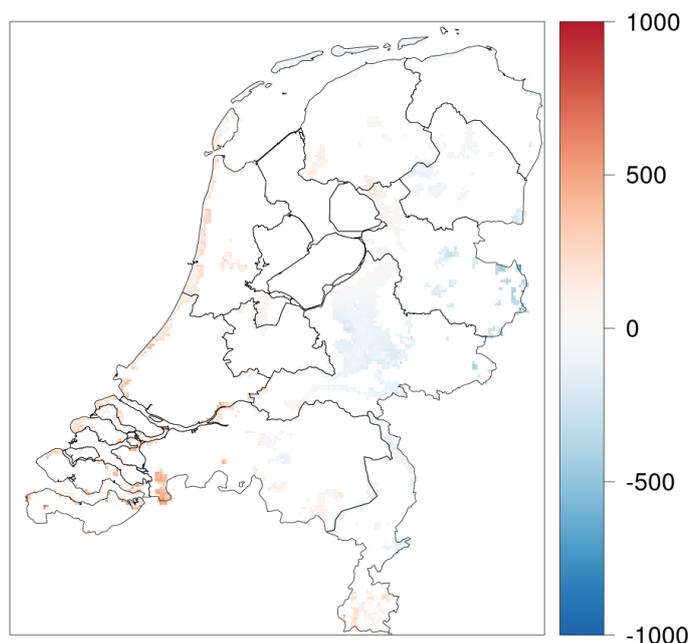


*Figuur 3. Absolute (links) en relatieve (rechts) verschilkaart van de totale stikstofdepositie berekend met de nieuwe ruimtelijke kalibratie methode en de huidige constante kalibratie methode.*

Voor de monitoring van de stikstofdepositie is het verloop van de stikstofdepositie in de tijd belangrijk. Het ruimtelijk geïnterpoleerde kalibratieveld over Nederland is echter stabiel over de jaren en daarom is het effect op de trend verwaarloosbaar en niet verder onderzocht. De depositiecijfers worden echter ook gebruikt om overschrijdingen van de Kritische Depositie Waarde (KDW) te berekenen. Figuur 4 laat het effect zien van de nieuwe methode op de overschrijding van de KDW in de

Natura 2000-gebieden. Vooral in het (zuid)westen van het Nederland gaat de berekende overschrijding omhoog, maar in het oosten van Nederland gaat de berekende overschrijding juist naar beneden. Dit betekent overigens niet dat er ecologisch iets verandert. De veranderingen in de overschrijdingen zijn namelijk enkel het gevolg van een andere berekeningswijze. De effecten van de nieuwe berekeningswijze zullen in AERIUS Monitor gerapporteerd worden.

verschil in KDW overschrijding nieuwe - huidige methode 2018 [mol/ha]



*Figuur 4. Verschilkaart van de overschrijding van de kritische depositiewaarden in Natura 2000-gebieden met stikstofgevoelige habitattypen berekend met de nieuwe ruimtelijke kalibratie methode en de huidige constante kalibratie methode.*

### **Onzekerheden**

De onzekerheid in de berekeningen van de stikstofdepositie zijn betrekkelijk groot ten opzichte van (andere) concentratie berekeningen. Op het ruimtelijke schaal niveau van 1x1 km wordt de totale onzekerheid geschat op 35% (1 sigma; Hoogerbrugge et al., 2020). Een belangrijke component van de onzekerheid komt voort uit het de hierboven beschreven verschil tussen de gemeten en berekende ammoniak concentraties. Bij toepassing van de nieuwe kalibratiemethode neemt de standaarddeviatie af van 1,67 naar 1,18  $\mu\text{g}/\text{m}^3$ . Bij een gemiddelde concentratie in natuurgebieden van 5  $\mu\text{g}/\text{m}^3$  daalt de onzekerheid in de berekende ammoniak concentraties door de nieuwe kalibratie methode van 33% naar 24% (1 sigma). De nieuwe kalibratiemethode is daarmee een effectieve manier om te corrigeren voor het verschil tussen de gemeten en gemodelleerde concentraties. Het blijft echter belangrijk om de andere belangrijke factoren, zoals mogelijk ontbrekende of onzekere emissies, op te lossen en modelverbeteringen door te voeren.

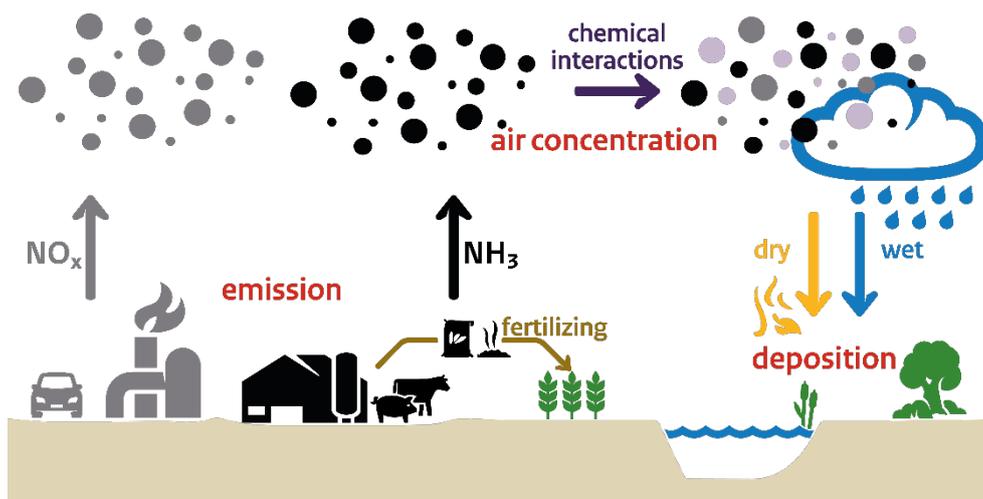


## 1 Introduction

Nitrogen ( $N_2$ ) is a colorless and odorless gas that is all around us. About 78% of all air consists of nitrogen. Nitrogen is not harmful to humans or the environment. But there are also compounds of nitrogen in the air that can be harmful to humans and the environment. These are nitrogen oxides ( $NO_x$ , a compound of nitrogen and oxygen) and ammonia ( $NH_3$ , a compound of nitrogen and hydrogen). The amount of nitrogen oxides and ammonia in the air is called the concentration.

To properly understand the so-called "nitrogen problem", three terms are important:

- Emission (how much gets into the air)
- Concentration (how much is in the air)
- Deposition (how much gets on the surface)



### 1.1 Concentration

The concentration in the air is the amount of a pollution in the air. The units we use are  $\mu g/m^3$ . Too much nitrogen oxides in the air, or: a high nitrogen oxide concentration, is harmful to health. Especially people with lung complaints and asthma suffer from it. Too much ammonia in the air is also harmful to humans. But in ambient air the ammonia concentrations are rarely that high. However, high ammonia concentrations are not harmless, they have an indirect negative effect on the environment (see section 1.3 about deposition).

Ammonia concentrations are measured on 6 locations of the National Air Quality Monitoring Network (LML). Since 2005 ammonia is also measured in Natura 2000-areas using passive samplers (MAN, [man.rivm.nl](http://man.rivm.nl)). Currently, the MAN-network consists of more than 80 Natura 2000-areas with in total more than 300 individual measurement locations.

## 1.2 Emission

To determine the concentration, it is important to know the amount of nitrogen oxides and ammonia released into the air. The release of these substances is called emission. The units we use are million kg nitrogen per year (or M kg N).

- Nitrogen oxides are mainly released when burning (fossil) fuels. The main sources of nitrogen oxides are traffic, power plants and industry.
- Ammonia is mainly released from agriculture, but traffic and people also produce ammonia. Farmers use fertilizer and manure because it ensures better plant growth and higher crop yields. Nitrogen evaporates from the manure as ammonia and enters the air. Another part of the nitrogen can leach into the groundwater; this is called (nitrate) leaching.

Under the direction of the National Institute for Public Health and the Environment (RIVM), various organizations work together to collect and establish data on the emission of pollutants into air, water and soil. These are reported on the website of the Pollutant Release and Transfer Register (PRTR).

The figure below shows the emissions of nitrogen (ammonia plus nitrogen oxides) from various sectors to the air in the period between 1990 and 2017:

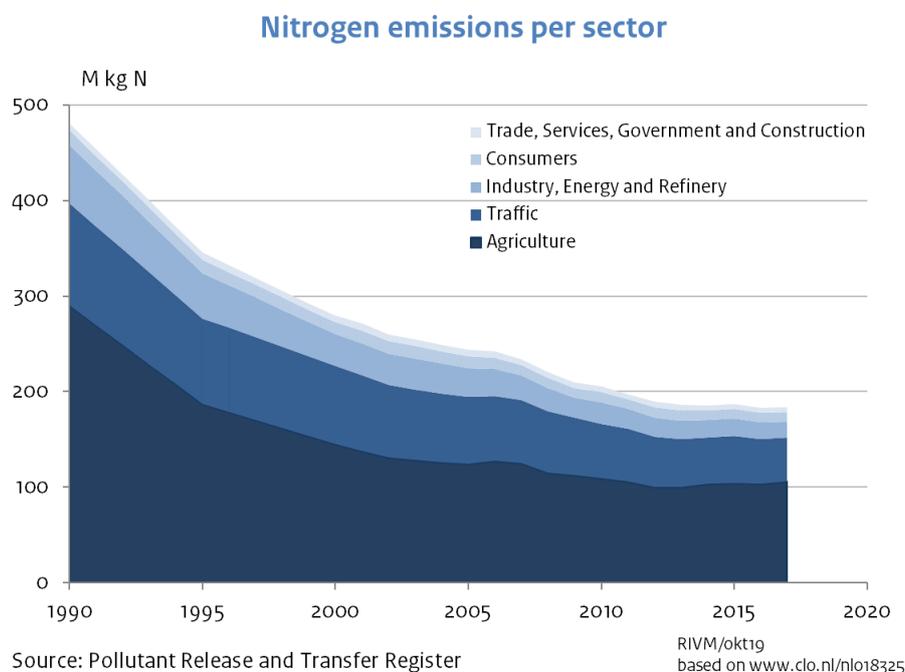


Figure 1. Nitrogen emissions per sector from 1990 – 2017 (From the Dutch Pollutant Release and Transfer Register).

## 1.3 Deposition

The nitrogen oxides and ammonia in the air eventually get back to the earth's surface again. This is called nitrogen deposition. The units we use here are mol nitrogen per hectare (mol/ha). The deposition of

nitrogen oxides and ammonia enriches the soil in nutrients. This can become a problem especially in nature reserves, as rare plant species that favor a nutrient-poor soil disappear. In general, eutrophication of the soil leads to a decline in biodiversity (the variety of plant and animal life). If vegetation and soil absorb nitrogen directly from the air, this is called dry deposition. If the substances can come along with precipitation on the surface, this is called wet deposition.

RIVM measures and calculates deposition. The dry deposition of ammonia is measured at 3 locations in Natura 2000 areas with different ecosystems: in Bargerveen (moorland), in Oostelijke Vechtplassen (bogland) and Hoge Veluwe (heathland); see <https://www.rivm.nl/stikstof/meten/drogedepositieNH3>. We are extending this monitoring network with 2 new locations. Besides, RIVM collects rain samples at 8 LML locations every 2 weeks using wet-only samplers (which put a lid on the collector when it is not raining to avoid evaporation of rainwater and collection of dry deposition). The amount of ammonium and nitrate in the rain samples is then analyzed in a laboratory.

#### **1.4 Assessment of the deposition in the Netherlands**

It is not possible to equip the entire country with measuring instruments. That would be too expensive and practically impossible. In order to provide a nationwide picture of the nitrogen concentration and deposition, and to be able to make future predictions, RIVM uses model calculations. The model that is used to do the model calculations is the OPS-model (Sauter et al., 2018). This model is the basis of the GDN/GCN maps and is also used in AERIUS, which is the calculation tool for the living environment in the Netherlands.

All sorts of data from different sources are used in modeling. Not only from RIVM but also from other institutes like the Royal Netherlands Meteorological Institute (KNMI Royal Meteorological Institute), Central Bureau of Statistics (CBS Central Bureau of Statistics), Wageningen University (WUR Wageningen University & Research), the Netherlands Environmental Assessment Agency (PBL Netherlands Environmental Assessment Agency) and TNO. This includes data on weather conditions, the emission of substances per source, data on the road distribution and traffic intensities in the Netherlands and economic expectations (for future predictions).

The OPS-model uses information on emissions, land use and meteorology to calculate the concentrations in the air and the deposition from the air to the surface. In these calculations, the OPS-model accounts for all relevant atmospheric processes like dispersion, transport, chemical reactions in the atmosphere, removal by rain and removal by the surface (vegetation or water).

For the GCN/GDN maps, OPS calculates the annual average concentrations and deposition values on a 1x1 kilometer grid. In the AERIUS applications the calculations from individual sources are carried out at a higher spatial resolution of approximately 100 x 100 meters.

The OPS-model calculates each of the components of the deposition, e.g. dry  $\text{NH}_x$ , wet  $\text{NH}_x$ , dry  $\text{NO}_y$  and wet  $\text{NO}_y$ . To assess the total nitrogen deposition in the Netherlands the model calculations are combined with measurements. The two wet components can be validated and corrected with direct observations of the wet deposition at 8 measuring stations across the Netherlands (Hoogerbrugge et al., 2020). Due to the limited number of stations but the spatial homogeneous character of the wet deposition, this correction is based on the average difference between the modelled and measured wet deposition. For the dry deposition of  $\text{NO}_y$  a fixed correction is applied which was derived by Buijsman et al. (2008).

Ideally, the modelled annual dry deposition is calibrated with the measured annual dry deposition. However, the number of locations with dry deposition measurements is too low to do this accurately at the moment. The use of the dry deposition measurements in the validation and calibration process is still under investigation and is beyond the scope of this report. The current method to correct the dry deposition of ammonia is based on the property that the dry deposition of  $\text{NH}_3$  is approximately proportional to the  $\text{NH}_3$  concentration (see Appendix A. Proportionality of dry deposition and concentration of ammonia). For the dry deposition of  $\text{NH}_x$  a constant national calibration factor is derived from a comparison of the modelled and measured  $\text{NH}_3$  concentrations. This constant national calibration factor is used to correct both the modelled  $\text{NH}_3$  concentrations and the dry  $\text{NH}_x$  deposition. The calibration of the  $\text{NH}_3$  concentration and dry deposition of  $\text{NH}_x$  is the main topic of this report.

Every year, RIVM reports the development of the national average annual total nitrogen deposition on the Environmental Data Compendium (Figure 2). The national average total nitrogen deposition is based on the combination of model results and measurements as described above. The figure shows the development of the dry and wet  $\text{NH}_x$  and  $\text{NO}_y$  deposition in time. It shows that the dry  $\text{NH}_x$  deposition is an important contributor to the total nitrogen deposition.

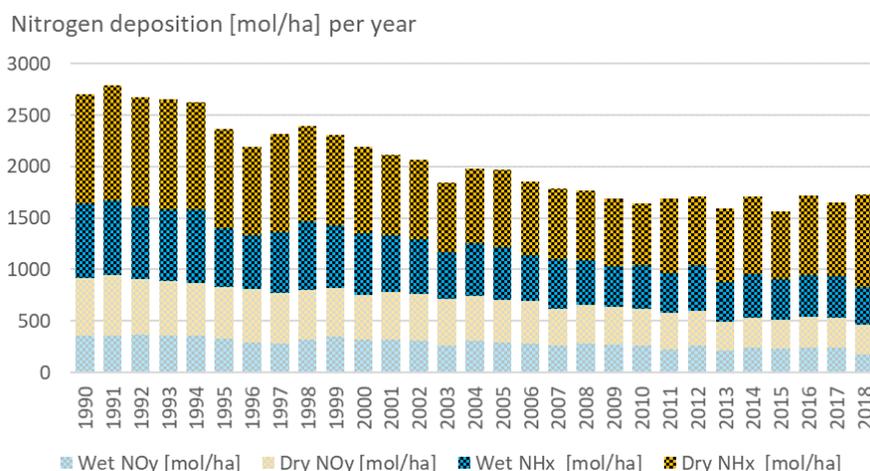


Figure 2. Annual average total nitrogen deposition in the Netherlands between 1990 and 2018 (based on [www.clo.nl/nl0189](http://www.clo.nl/nl0189)).

In this report, we focus on the validation of the  $\text{NH}_3$  concentration and dry deposition of  $\text{NH}_x$ . We describe the current method to correct the  $\text{NH}_3$  concentration and dry deposition of  $\text{NH}_x$  in Chapter 2. In Chapter 3 we explore the possibilities of a new data fusion approach. In Chapter 4 we do a cross validation check to see how robust the current method and the new data fusion approach are. In Chapter 5, we describe the consequences for the assessment of the total nitrogen deposition and the exceedance of critical loads in the Netherlands. In Chapter 6, we briefly discuss our findings.



## 2 Current method to correct the NH<sub>3</sub> concentration and dry deposition of NH<sub>x</sub>

In the current method to correct the NH<sub>3</sub> concentration and dry NH<sub>x</sub> deposition, the individual MAN-observations are linearly averaged to one area averaged (per MAN-area) value. To determine the under- or overestimation of the model a regression analysis between the annual averaged measured and modelled NH<sub>3</sub> concentration is carried out. The constant national calibration factor is obtained from the linear fit through the origin. Figure 3a shows the comparison of measured versus modelled annual area average ammonia concentrations in 71 Natura 2000 areas plus 6 LML measuring stations (of which two with passive samplers). The constant national calibration factor, i.e., 0.935 slightly deviates from the reported correction factor of 0.932 (=1/1.073) in the GCN report (Hoogerbrugge et al., 2019). This is due to an incorrect coordinate in one of the 77 locations, which has been corrected now. When the constant national calibration factor is applied to the modelled concentrations, the corrected concentrations are in line with the measured concentrations (Figure 3b).

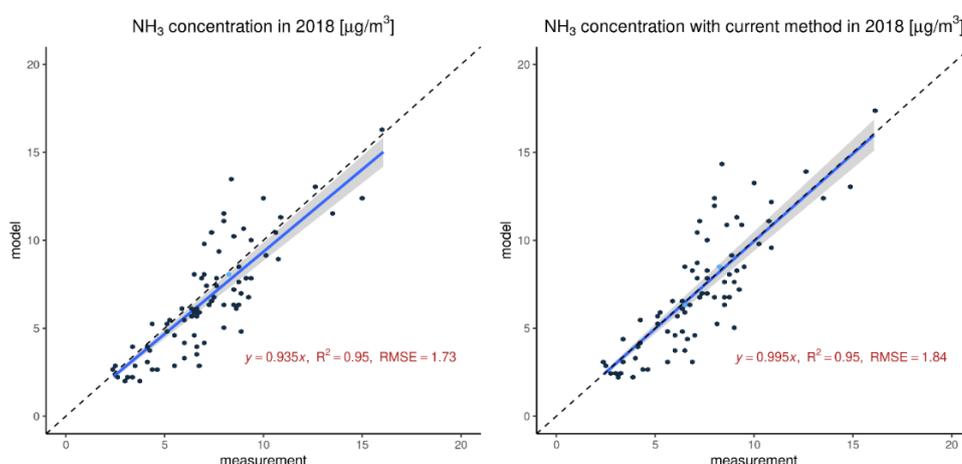
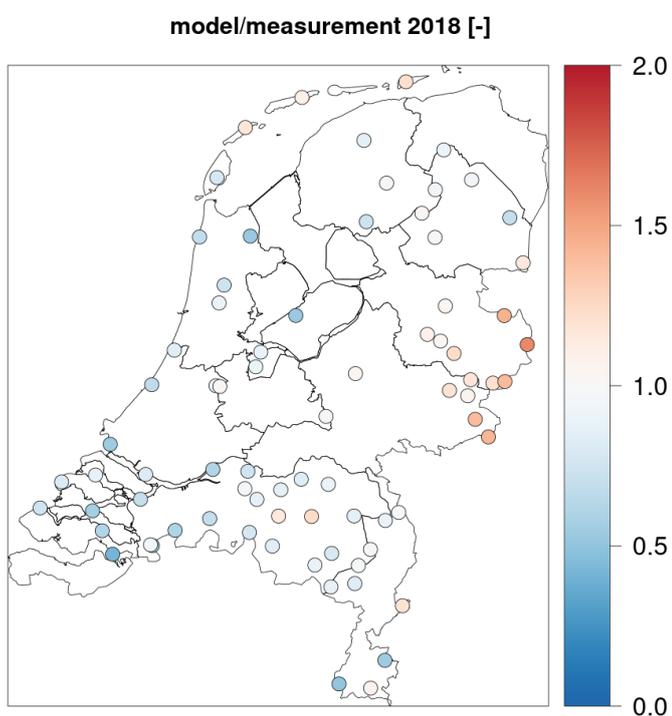


Figure 3. Scatterplot of measured versus modelled annual average ammonia concentrations in 2018 before (left) and after correction with the current method (right). Coloured points are overlapping points.

The scatterplot of the measured versus the modelled annual area averaged ammonia concentrations (Figure 3) shows that at some locations the concentrations are overestimated by the model, while at other locations the concentrations are underestimated by the model.

Figure 4 shows the spatial distribution of the ratio between the modelled and measured annual area averaged concentrations throughout the Netherlands. A ratio  $> 1$  indicates that the model overestimates the concentrations compared to the observations, while a ratio  $< 1$  indicates that the model underestimates the concentrations compared to the observations. The figure shows that ratios  $> 1$  are clustered in the eastern part of the Netherlands, while the ratios  $< 1$  are mainly located in the (south)western part of the country. Figure 4 indicates that the correction of the  $\text{NH}_3$  concentration and dry deposition of  $\text{NH}_x$  can be improved by accounting for these regional differences.



*Figure 4. Spatial distribution of the ratio between modelled and observed concentrations.*

### 3 The data fusion approach to spatial correct the NH<sub>3</sub> concentration and dry deposition

A method for spatial interpolation that is commonly used in geostatistics is called kriging (Oliver and Webster, 2014). The purpose of kriging is to predict the unknown value of a variable (in this case the ratio between modelled and measured NH<sub>3</sub> concentration) at a given location from the known values of the variable given at a number of fixed locations. Kriging is known to provide the best linear unbiased predictions. Kriging uses a covariance function as a way to describe how much samples correlate depending on the distance between those samples. This covariance function can be estimated from the so-called empirical semi-variogram, which depicts the semi-variance between all pairs of samples as a function of the distance between them. Samples taken far apart will generally vary more than samples close to each other. The experimental variogram shows the mean semi-variance between values as a function of distance. The variogram model is the fit through these aggregated semi-variances, weighted by the number of pairs. Here, we use the variogram model based on Matern, M. Stein's parameterization using the R-package 'autoKrige'.

Figure 5 shows the experimental variogram and the fitted variogram model based on 5 years of model calculations and measurements of ammonia concentrations in the Netherlands. The use of 5 years of model calculations and measurements is more robust than the use of individual years (see Figure 19 in Appendix B). The numbers in the plot reflect the number of data pairs within the distance class.

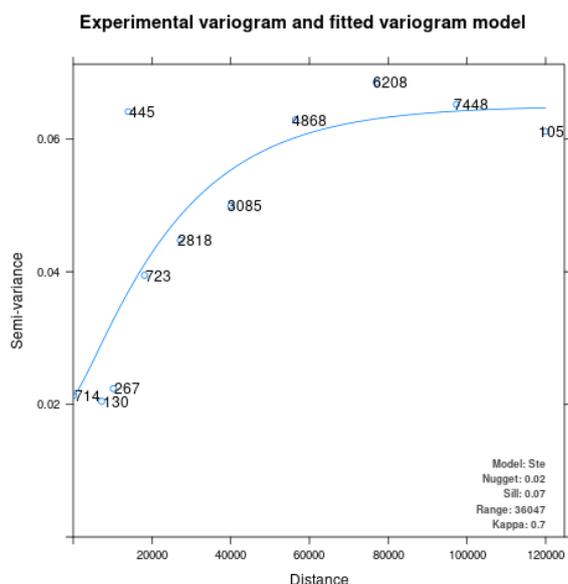
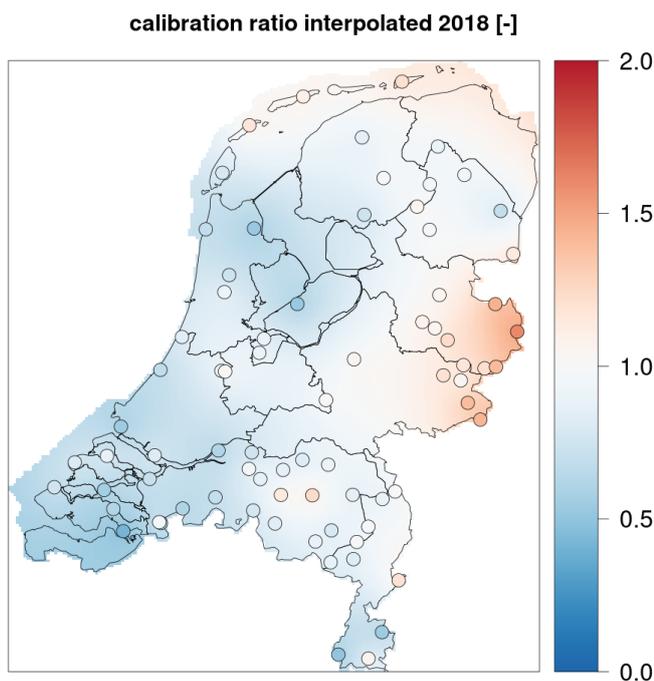


Figure 5. Experimental variogram (blue dots) and fitted variogram model (blue line) based on area averaged<sup>1</sup> NH<sub>3</sub> concentrations (5 years of data). The numbers indicate the number of pairs of samples

<sup>1</sup> When there is more than one measurement location within a Natura 2000-area, the values of all measurement locations within this Natura 2000-area are linearly averaged to a single 'area averaged' value.

The fitted variogram model from Figure 5 is subsequently used to interpolate the ratios<sup>2</sup> between modelled and measured  $\text{NH}_3$  concentrations, which then results in an interpolated calibration ratio field (Figure 6). Note that to correct the modelled nationwide  $\text{NH}_3$  concentrations, we need to divide the concentrations by the interpolated calibration field.



*Figure 6. Spatially interpolated ratio between modelled and measured  $\text{NH}_3$  concentrations in the Netherlands in 2018, data fusion approach.*

When the spatially interpolated calibration ratio is applied to the modelled concentrations, the scatter between the corrected concentrations and the measured concentrations reduces considerably (Figure 7). This results in a higher correlation coefficient (0.99) and a smaller Root Mean Square Error (RMSE = 0.64) then with the current method ( $R^2=0.95$ , and RMSE = 1.84; see also Figure 3).

<sup>2</sup> Technically the log of the ratios are used to construct the calibration field. After back transformation the field is ready for use.

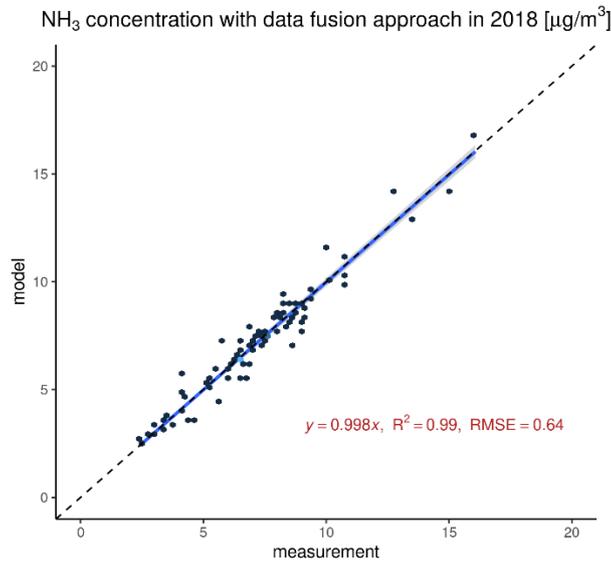


Figure 7. Scatterplot of observed versus modelled ammonia concentrations after correction with the data fusion approach in 2018. Coloured points are overlapping points.

In the current method to correct the NH<sub>3</sub> concentration, the individual observations within a Natura 2000-area were averaged to one area averaged (per Natura 2000-area) value. In the data fusion approach, information about the spatial correlation is lost when area averages are calculated. Therefore, we used all the individual measurement locations in the Natura 2000-areas to calculate the experimental variogram and the fitted variogram model (Figure 8).

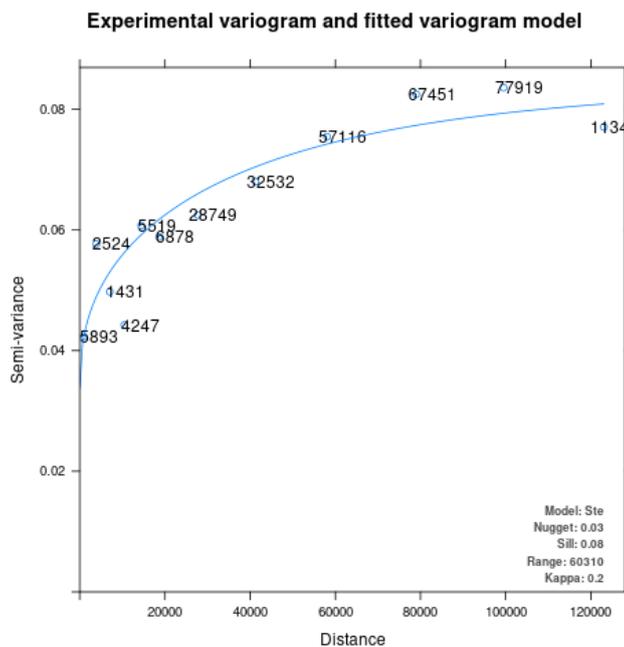


Figure 8. Experimental variogram (blue dots) and fitted variogram model (blue line) based on NH<sub>3</sub> concentrations at the individual measurement locations (5 years of data). The numbers indicate the number of pairs of samples.

Because all individual measurement locations are involved, the number of data pairs increases considerably. Also the differences between the experimental semi-variogram and the variogram model reduces presumably due to the larger number of data and subsequent noise reduction.

The resulting spatially interpolated calibration ratio is shown in Figure 9 (right panel). For comparison, the spatially interpolated calibration ratio obtained with the area averaged values (Figure 6) is shown in the left panel. The figure shows that the new interpolation technique is not very sensitive to outliers. The spatially interpolated calibration ratios roughly show the same pattern as the spatially interpolated calibration ratios that were obtained with the area averaged values.

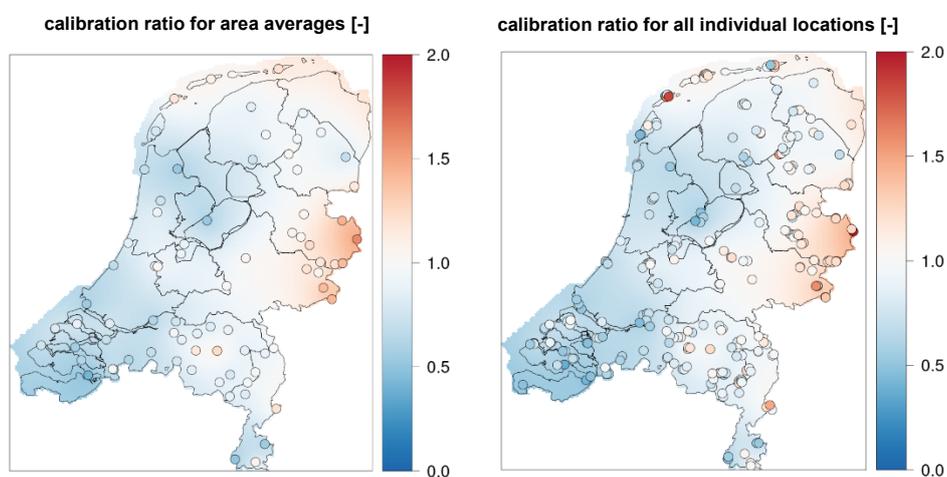


Figure 9. Spatially interpolated ratio between modelled and measured  $\text{NH}_3$  concentrations (i.e. calibration ratio) in the Netherlands in 2018. Left: based on area averaged values; Right: based on individual locations.

In Figure 3 and Figure 7 we showed the scatter plots of the *area averaged* measured versus modelled  $\text{NH}_3$  concentrations with the uncorrected model results, the corrected model results with the current method and the corrected model results with the data fusion approach, respectively. In Figure 11, we show the same scatter plots of the measured versus modelled concentrations, but now for *all individual measurement locations*.

When the spatially interpolated calibration ratio is applied to the modelled concentrations for all measurement locations, the agreement between the corrected concentrations and the measured concentrations is much improved compared to the current method (Figure 10 lower right panel compared to the lower left panel,  $R^2$  increases from 0.93 to 0.98 and RMSE from 2.04 to 0.97).

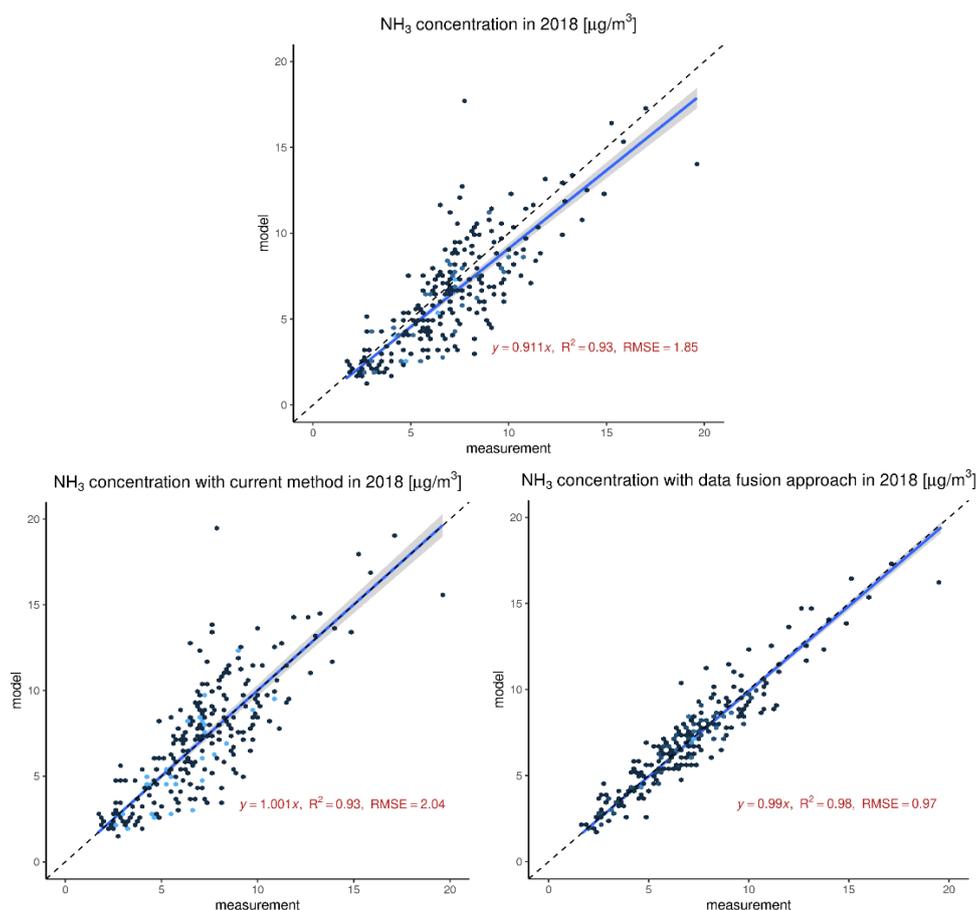


Figure 10. Scatterplot of measured versus modelled ammonia concentrations for all measurement locations before (upper) and after correction with the current method (lower left) and the data fusion approach (lower right) in 2018. Coloured points are overlapping points.

The modelled NH<sub>3</sub> concentrations as calculated by the OPS model (uncorrected) are shown in the upper left panel of Figure 11. The measurements at all measurement locations are plotted with coloured dots in the same figure (using the same colour scale). All modelled and measured NH<sub>3</sub> concentrations in the map correspond to a point in the upper scatter plot of Figure 10. The upper right panel in Figure 11 shows the modelled concentrations after correction with the current method (corresponding to the lower left scatter plot of Figure 10). The lower left panel shows the modelled concentrations after correction with the data fusion approach (corresponding to the lower right scatter plot of Figure 10). The lower right panel of Figure 11 shows the absolute difference in the NH<sub>3</sub> concentration between the two correction methods.

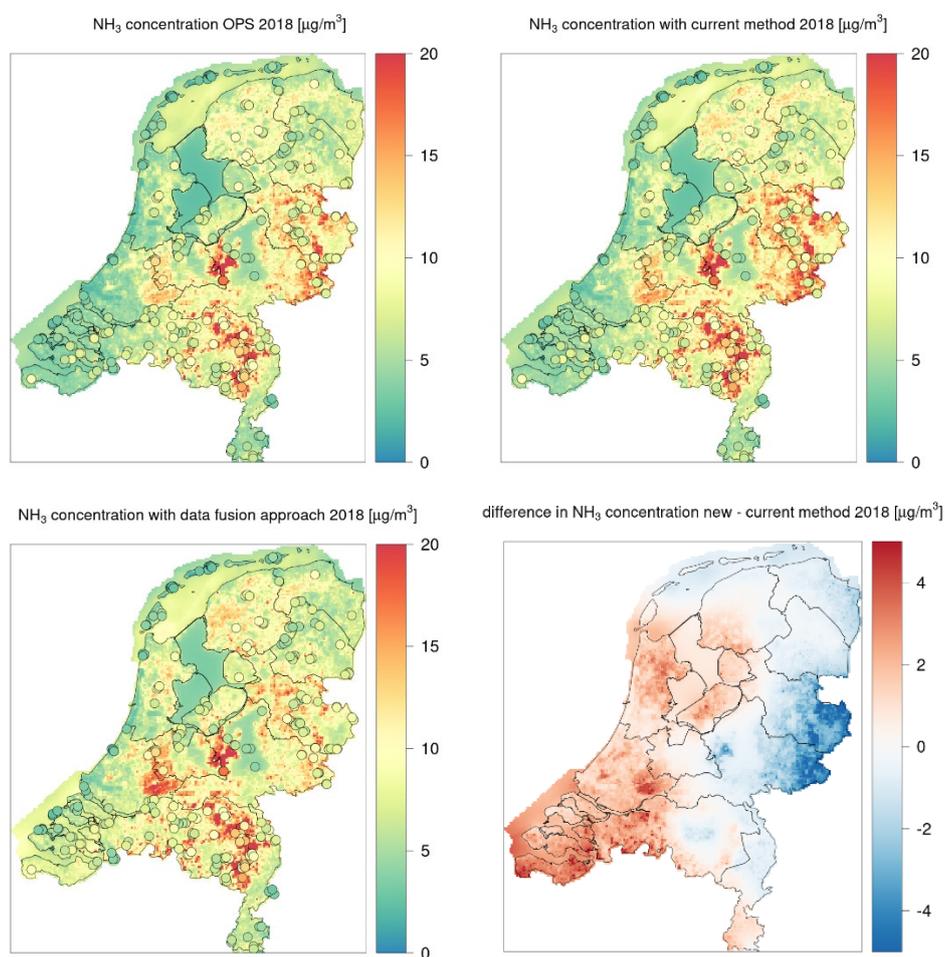


Figure 11. Annual average  $\text{NH}_3$  concentration as calculated by the OPS model (upper left), after correction with the constant national calibration factor (upper right), after correction with the data fusion approach (lower left) and the difference between the  $\text{NH}_3$  concentration after correction with the data fusion approach and the  $\text{NH}_3$  concentration after correction with the constant national calibration factor (lower right). The points in the first 3 figures are the measured concentrations at the measurement locations in the same colour scale as the modelled  $\text{NH}_3$  concentrations.

## 4 Comparison of new and current method with cross validation

### 4.1 The cross validation method

To check if the new data fusion approach performs better than the current method, a cross validation is carried out for both methods using all individual measurement locations. Cross validation is a model validation technique for assessing how the results of a statistical analysis will generalize to an independent data set. In the cross validation that we carried out, 80% of the data is used to correct the remaining 20% of the data. For the remaining 20% of the data, we evaluate the Root Mean Square Error (RMSE) and the coefficient of determination ( $R^2$ ) of the corrected model data versus the observations. This procedure is repeated 100 times to get an impression of the robustness of both methods. This method is also known as bootstrap method.

### 4.2 Results of the cross validation

Figure 12 shows the RMSE for the current method (left) and the new data fusion approach (right). Ideally, the RMSE is close to zero. The figure shows that the RMSE is much smaller for the correction with the new data fusion approach than with the current method. Also for the RMSE we see that the variation within one year is smaller for the new method.

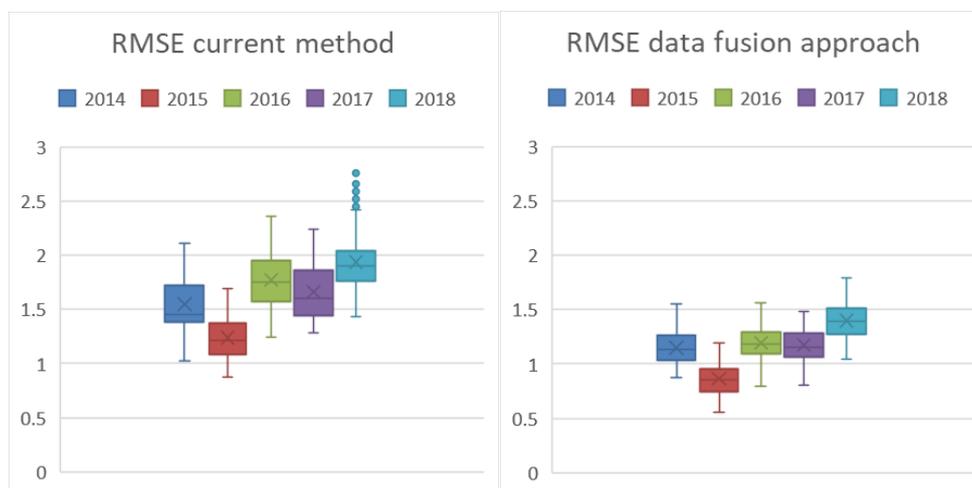


Figure 12. RMSE of the 100 linear regression lines through the scatter of measured ammonia concentrations versus modelled ammonia concentrations after correction with the current method (left) and the new data fusion approach (right).

Figure 13 shows the coefficient of determination ( $R^2$ ) for the current method (left) and the new data fusion approach (right). In the case that  $R^2$  is 1, the model results perfectly match the observations. The figure shows that the  $R^2$  is larger for the correction with the new data fusion approach than current method. As found with the RMSE again, the spread in the obtained  $R^2$  is smaller for the new data fusion approach.

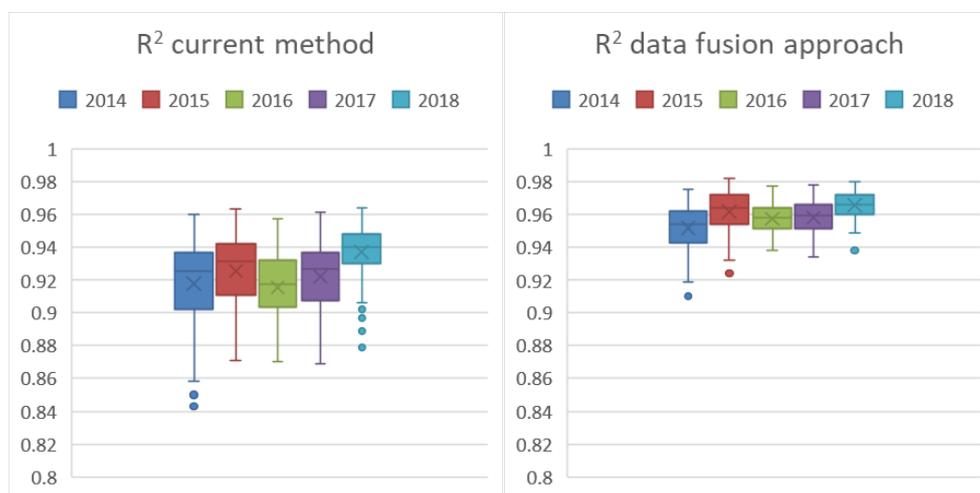


Figure 13.  $R^2$  of the 100 linear regression lines through the scatter of measured ammonia concentrations versus modelled ammonia concentrations after correction with the current method (left) and the new data fusion approach (right).

### 4.3 Using the cross validation in uncertainty analysis

The cross validation results are used as an unbiased estimator of the uncertainty of the calibrated concentration maps. When we combine the validation results for all 5 years the RMSE of the old method is  $1.67 \mu\text{g}/\text{m}^3$ . For the new method the RMSE reduces to  $1.18 \mu\text{g}/\text{m}^3$ . For Dutch nature 2000 area's the average concentration is approximately  $5 \mu\text{g}/\text{m}^3$ . Then the relative standard deviation will be respectively 33 and 24%. Assuming a normal distribution, a RSME of  $1.18 \mu\text{g}/\text{m}^3$  implies that a calculated model value has a 95% confidence interval of  $2.36 \mu\text{g}/\text{m}^3$  around the model value. This is much better than the original confidence interval of  $3.34 \mu\text{g}/\text{m}^3$ .

Using the cross validation results might introduce a small overestimation of the uncertainty since each cross validation data set is 20% smaller than the calibration set in the operational setting. In principle the application of sample sites from the some Natura 2000 area might introduce couples of extremely correlated data which can cause an underestimation of the variance found in the cross validation. Figure 8 shows that the semi variance at very small distances fit quite well into the overall semi variance curve. This implies that close couples are not extremely correlated and therefore probably don't disturb the cross validation process. Both the potential under and overestimation of the uncertainty are presumably small and therefore ignored.

## 5 Effect of data fusion approach on the dry $\text{NH}_x$ deposition

### 5.1 Change in the dry $\text{NH}_x$ deposition pattern

The current calibration method and new data fusion approach lead to different concentration and dry deposition maps. Figure 14 shows the effect of both calibration methods on the calculated dry deposition field. The upper left panel shows the original (uncorrected) calculation. The upper right panel shows the corrected dry  $\text{NH}_x$  deposition with the constant national calibration factor obtained by the linear regression method that is currently applied. The lower left panel shows the dry  $\text{NH}_x$  deposition obtained with the new spatially interpolated calibration ratio, i.e. the data fusion approach. The lower right panel shows the difference between the two methods. The figure shows that the new method results in a shift in the dry deposition pattern over the Netherlands. The dry deposition is reduced in the eastern part of the Netherlands, while in the (south)western part of the country the dry deposition is increased.

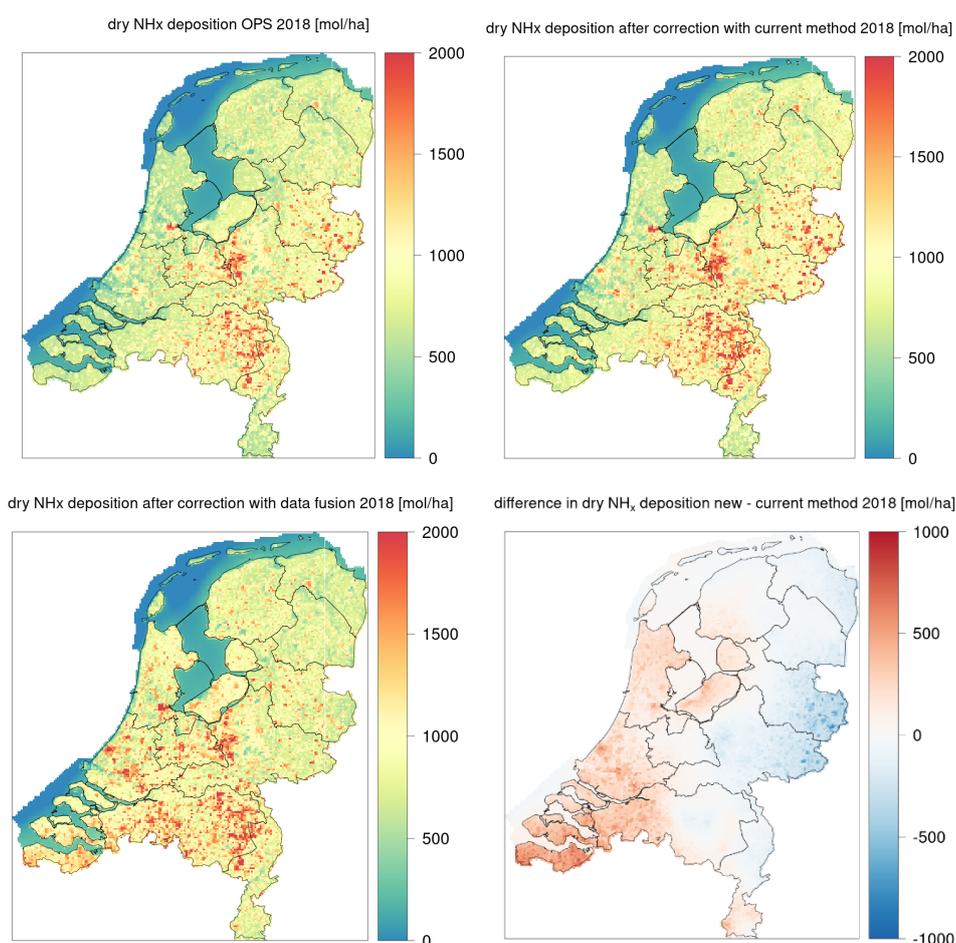


Figure 14. Annual dry  $\text{NH}_x$  deposition as calculated by the OPS model (upper left), after correction with the constant national calibration factor (upper right), after correction with the data fusion approach (lower left) and the difference between the two methods (lower right).

## 5.2 Effect on the total nitrogen deposition

The different correction methods lead to different maps of the dry  $\text{NH}_x$  deposition and consequently to different maps of the total N deposition. Figure 15 shows the total nitrogen deposition obtained with the current method and the new data fusion approach to correct the dry  $\text{NH}_x$  deposition (upper left and upper right panel respectively). Note that the wet deposition of  $\text{NH}_x$  and  $\text{NO}_y$  and the dry deposition of  $\text{NO}_y$  are kept the same in both cases. How these components are calculated and corrected is described in Hoogerbrugge et al. (2020). The absolute difference between the two methods (lower left panel) is therefore exactly the same as the lower right panel in Figure 14 as we only changed the correction method of the dry  $\text{NH}_x$  deposition. The lower right panel shows the relative difference in the total N deposition between the two methods. The two lower panels show that the total nitrogen deposition increases in the (south)western part of the Netherlands, while it decreases in the eastern part of the country.

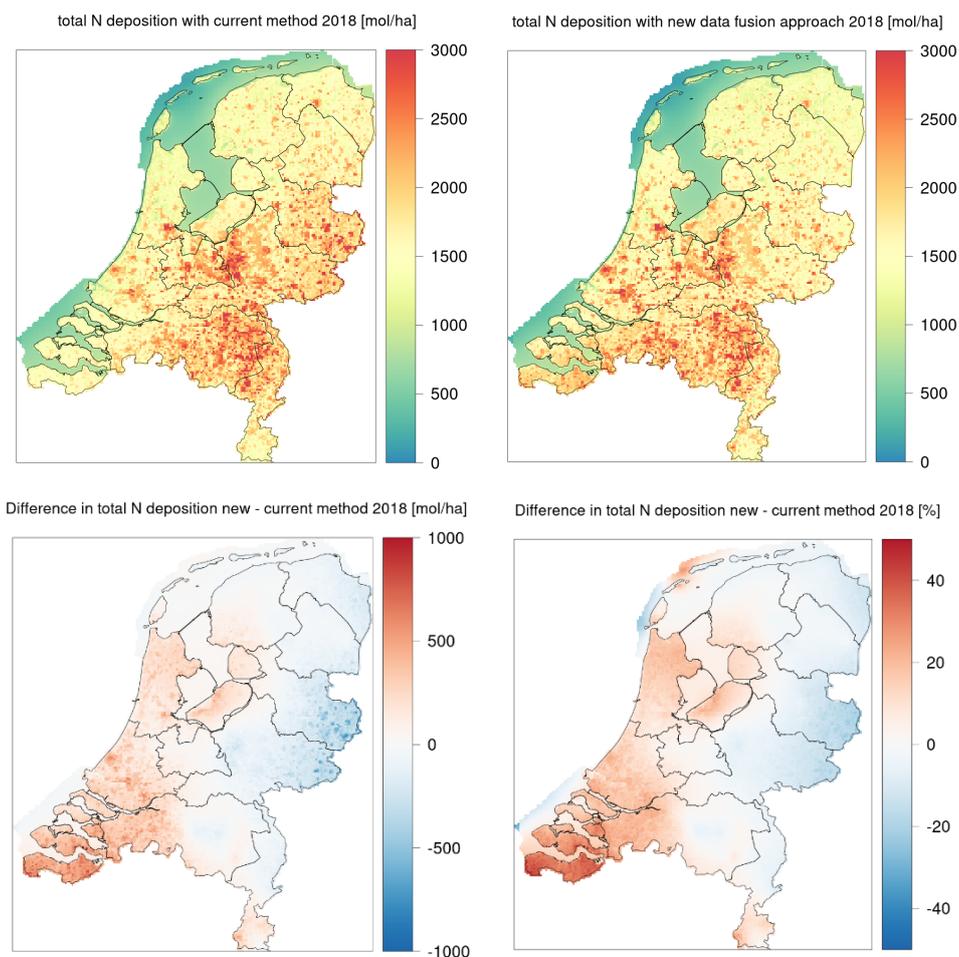


Figure 15. Annual total nitrogen deposition as obtained with the current method (upper left) and the new data fusion approach (upper right). The absolute and relative difference between the annual total nitrogen deposition as obtained with the two methods is shown in the bottom left and bottom right panel respectively.

The absolute and relative differences in the total N deposition between the two methods are summarized in Table 1. The table shows the values of the absolute and relative differences in the total nitrogen deposition for the different percentiles over all grid cells in the Netherlands. The median of the difference (both absolute and relative) is close to 0, which is expected as we only add a spatial component to the correction. Locally, the absolute difference can amount up to 1139 mol/ha at a local hotspot for which the effects are largest. The relative difference in the total nitrogen deposition can amount up to almost 50%. However, 80% of the differences are within -9% and +18%. These values are well within the uncertainty of 35% (1 sigma) that is estimated for the uncertainty in total nitrogen deposition on a 1x1 km<sup>2</sup> grid cell in Hoogerbrugge et al. (2020).

*Table 1. Distribution of the absolute and relative (to the current method) differences in the total nitrogen deposition over the Netherlands.*

<b>percentile</b>	<b>Absolute difference [mol/ha]</b>	<b>Relative difference [%]</b>
minimum	-854	-35
1%	-381	-18
5%	-218	-12
10%	-144	-9
25%	-52	-4
50% (median)	13	1
75%	122	10
90%	263	18
95%	356	22
99%	529	34
maximum	1139	48

### 5.3 Effect on the exceedance of critical loads

An important indicator for biodiversity loss is the exceedance of critical nitrogen loads for nitrogen sensitive habitats. The critical nitrogen load is defined as a quantitative estimate of the exposure to nitrogen deposition below which significant harmful effects on a specified nitrogen sensitive habitat do not occur according to present knowledge. Figure 16 shows the critical nitrogen loads for each 1x1 grid cell containing nitrogen sensitive habitats within the different Natura 2000-areas in the Netherlands. The figure shows the critical loads for the most sensitive habitat type within each grid cell. It should be noted that the calculations in AERIUS use more detailed information on emissions, land use and habitats. Therefore, the figures presented here only give an indication of the effects of the new data fusion approach. Figure 16 shows that nitrogen sensitive habitats are present all over the country. In general, the critical loads are well below 2000 mol/ha, while the total nitrogen deposition is larger in large parts of the country (Figure 15, the country average annual nitrogen deposition being around 1730 mol/ha in 2018). This means that critical loads are exceeded in most of the Natura 2000-areas.

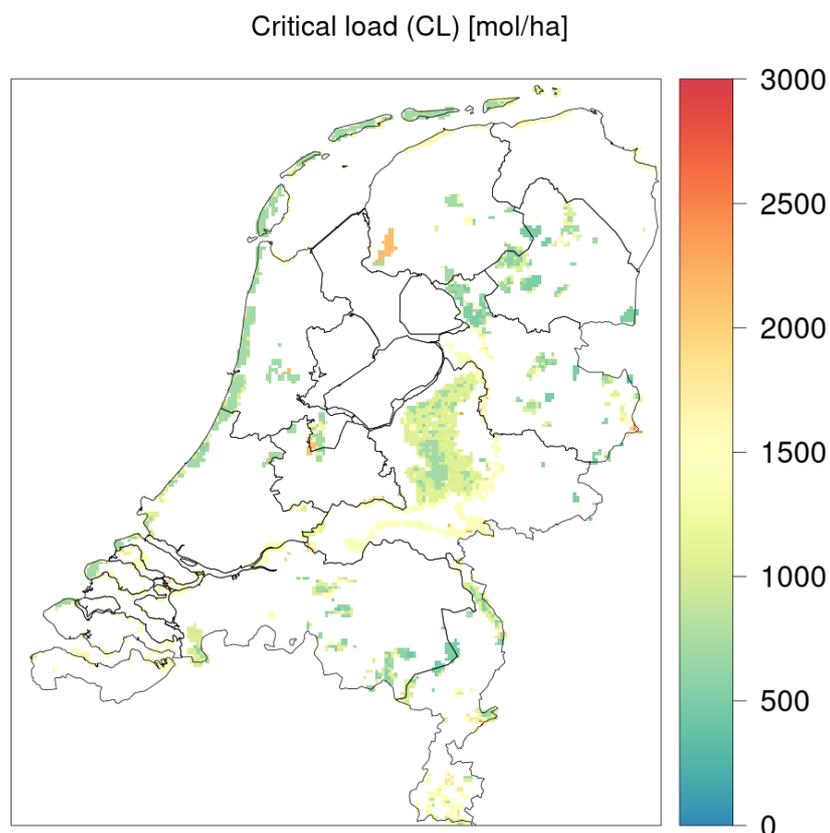


Figure 16. Critical nitrogen load for each 1x1 grid cell containing nitrogen sensitive habitats within the different Natura 2000-areas in the Netherlands.

The exceedance of the critical loads (CL) is calculated as the total nitrogen deposition minus the critical load value in each 1x1 km grid cell. The CL exceedances as calculated with the current method and with the data fusion approach are shown in Figure 17 (upper left and right panel, respectively). On first sight the maps look rather similar, but the shift in the total nitrogen deposition pattern also leads to changes in the critical load exceedance. This is illustrated in the map with the absolute difference in the CL exceedance between the two methods (bottom left panel). The most obvious changes appear in the (south)western part of the country, where the critical load exceedance increases, but there are also some areas in the eastern part of the country where the critical load exceedance decreases. Table 2 summarizes the distribution of the absolute differences in the CL exceedance between the two methods. 80% of the changes are within  $-130$  to  $190$  mol/ha. However, locally the changes amount up to  $\pm 750$  mol/ha. The map in the bottom right panel shows the relative difference in the CL exceedance between the two methods relative to the CL values. Table 2 also gives an overview of the distribution of these relative differences. The table shows that most (80%) of the changes are within  $-14\%$  to  $19\%$  of the CL values. But also here, local differences can be very large. In an extreme case, a reduction in the exceedance is found of  $155\%$  of the CL value.

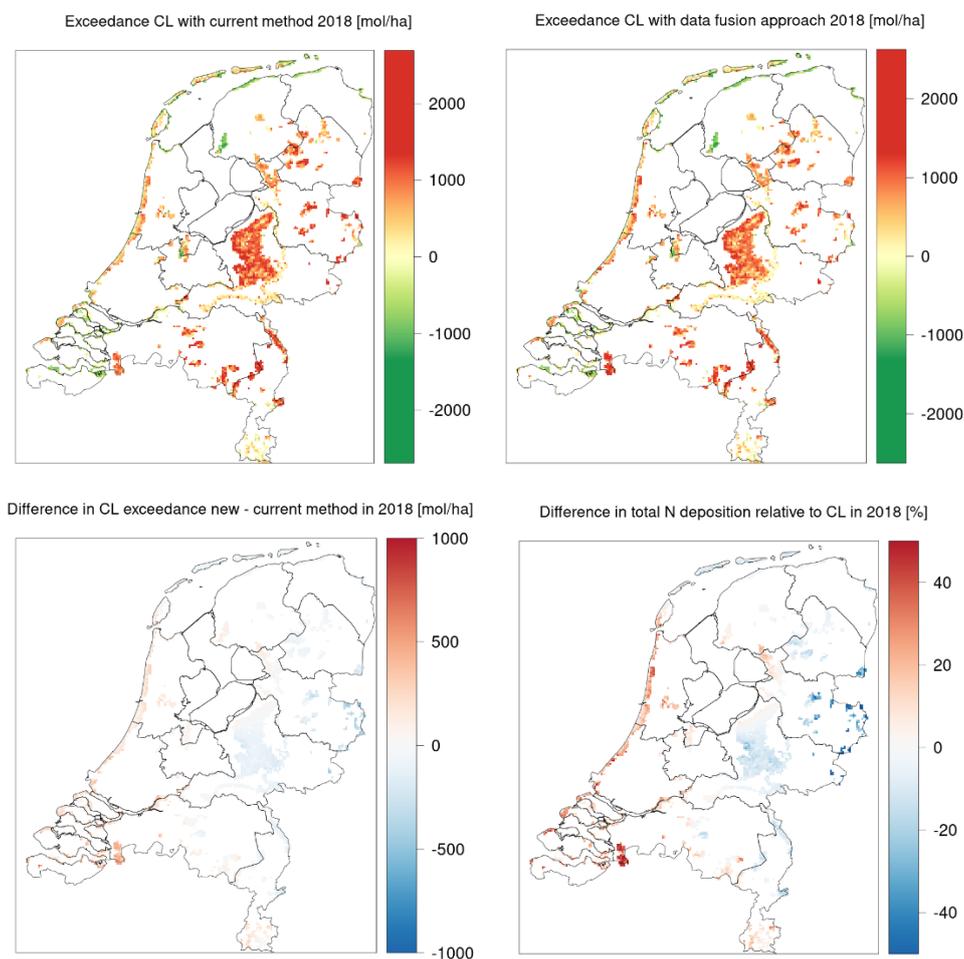


Figure 17. Critical load exceedance as calculated with the current method to correct the dry  $\text{NH}_x$  deposition (upper left) and with the data fusion approach (upper right). The absolute difference in the CL exceedance between the two methods (bottom left) and the relative difference in the CL exceedance between the two methods relative to the CL values (bottom right).

Table 2. Distribution of the absolute differences in the CL exceedance between the two methods and the relative differences in the CL exceedance between the two methods relative to the CL values.

percentile	Absolute difference [mol/ha]	Relative difference [%]
minimum	-741	-155
1%	-373	-53
5%	-179	-21
10%	-130	-14
25%	-78	-8
50% (median)	-11	-1
75%	84	8
90%	190	19
95%	277	27
99%	447	43
maximum	757	83



## 6 Discussion

In this report, a new data fusion approach for correcting the  $\text{NH}_3$  concentration and the dry  $\text{NH}_x$  deposition is described, using  $\text{NH}_3$  concentration measurements across the Netherlands. The current and new correction methods are both based on the assumption that the dry deposition of  $\text{NH}_x$  is proportional to the  $\text{NH}_3$  concentration level. A check of this assumption shows that this is indeed the case for different ranges of roughness lengths. This illustrates why the dry deposition can be corrected using the ratio between modelled and measured  $\text{NH}_3$  concentrations. There are some studies, e.g. Cape et al. (2008), that indicate that really close to a source (<100m) the deposition is not linearly dependent on the ammonia concentration. As most of the measurement locations are located in Natura 2000-areas, not in the proximity of sources, the possible non-linear behaviour close to sources is not likely to affect our correction methods.

Due to the large number of observations and the rather homogeneous distribution of the measurement locations over the country, kriging was used to correct the dry deposition of  $\text{NH}_x$ . The spatial pattern in the correction factor, i.e., ratios between modelled and measured  $\text{NH}_3$  concentrations different from 1, should ideally not be present and is subject for further research. At this stage, there is no clear reason for the spatial pattern. It might be caused by imperfections in the emissions or in the model descriptions. The spatial pattern of the difference in  $\text{NH}_3$  deposition between the new and the current method is found to be rather consistent in time (see appendix C). The spatial interpolation method might be further improved by including measurements and model calculations for locations outside of the Netherlands.

The uncertainty in the nitrogen deposition calculations is relatively large compared to (other) concentration calculations. At the spatial scale level of 1x1 km, the uncertainty in the total nitrogen deposition is estimated to be 35% (1 sigma; Hoogerbrugge et al., 2020). Main reason for this relatively large uncertainty is due to uncertainties in the dry deposition velocity. This current uncertainty estimate is however a somewhat worst-case value. At this moment the uncertainty is being re-assessed. The uncertainty in the deposition also has an effect on the comparison with the critical loads. The critical loads can be exceeded less or more due to the new calculation method. It even can lead to a change whether a critical load of a nature area is being exceeded or not. This illustrates that great caution is needed when critical loads and their exceedances are used for policy support. The numerical change in the exceedance due to a new calculation technique does not mean a change in exposure in the field. So for instance, when a nature area is now exceeded due to the new calculation method, this does not automatically mean that the ecological situation has changed. In general, it can be stated that using the deposition numbers and the exceedance numbers in a relative way in policy support is far more robust than using the absolute values of deposition and exceedance. The use of absolute deposition or exceedance data should preferably only be used as

indicative since they can change over time due to methodological adjustments.

An important component of the uncertainty arises from the difference between the measured and calculated ammonia concentrations. For a pure determination of the uncertainty, a bootstrap method is used. When applying the new data fusion approach, the standard deviation of the uncertainty decreases from 1.7 to 1.2  $\mu\text{g}/\text{m}^3$ . With an average concentration in nature areas of about 5  $\mu\text{g}/\text{m}^3$ , the relative standard deviation becomes 33% and 24%. The new data fusion approach is an effective way to correct for the difference between the measured and modelled concentrations. It remains important to resolve the underlying causes, such as missing or uncertain emissions and model imperfections.

## 7 References

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Sauter F., Van Zanten, M., Van der Swaluw, E., Aben, J., De Leeuw, F., Van Jaarsveld, H., 2018. The OPS-model. Description of OPS 4.5.2. RIVM report. Bilthoven, the Netherlands. <https://www.rivm.nl/media/ops/v4.5.2/OPS-model-v4.5.2.pdf>



## Appendix A. Proportionality of dry deposition and concentration of ammonia

The current method to correct the dry deposition of ammonia is based on the assumption that the dry deposition of  $\text{NH}_3$  is proportional to the  $\text{NH}_3$  concentration. To validate this assumption, Figure 18 shows the modelled concentration versus the modelled dry deposition of ammonia of all grid cells ( $1 \times 1 \text{ km}^2$ ) covering the land surface in the Netherlands (roughly 40000 point). It is known that dry deposition strongly depends on the roughness of the surface; a rough surface generates more turbulence and therefore more deposition. Different colours are used to mark the roughness lengths at the measurement locations. The figure illustrates that more deposition is calculated for the grid cells with a larger roughness lengths. The figure also shows that the assumption that the  $\text{NH}_3$  deposition is proportional to the  $\text{NH}_3$  concentration is approximately valid within certain ranges of roughness lengths. In practice, when the model overestimates the concentrations by 10%, the dry deposition is also overestimated by 10%.

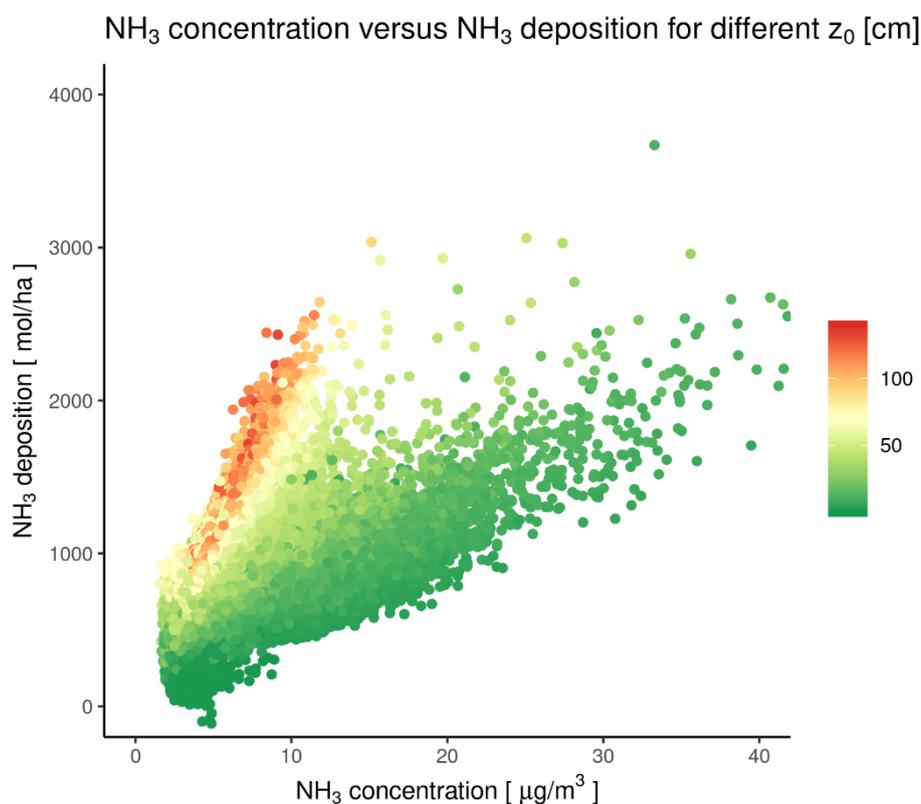


Figure 18. Modelled  $\text{NH}_3$  concentration versus  $\text{NH}_3$  deposition for different ranges of the roughness length ( $z_0$ ).

## Appendix B. Experimental variograms in 2014-2018

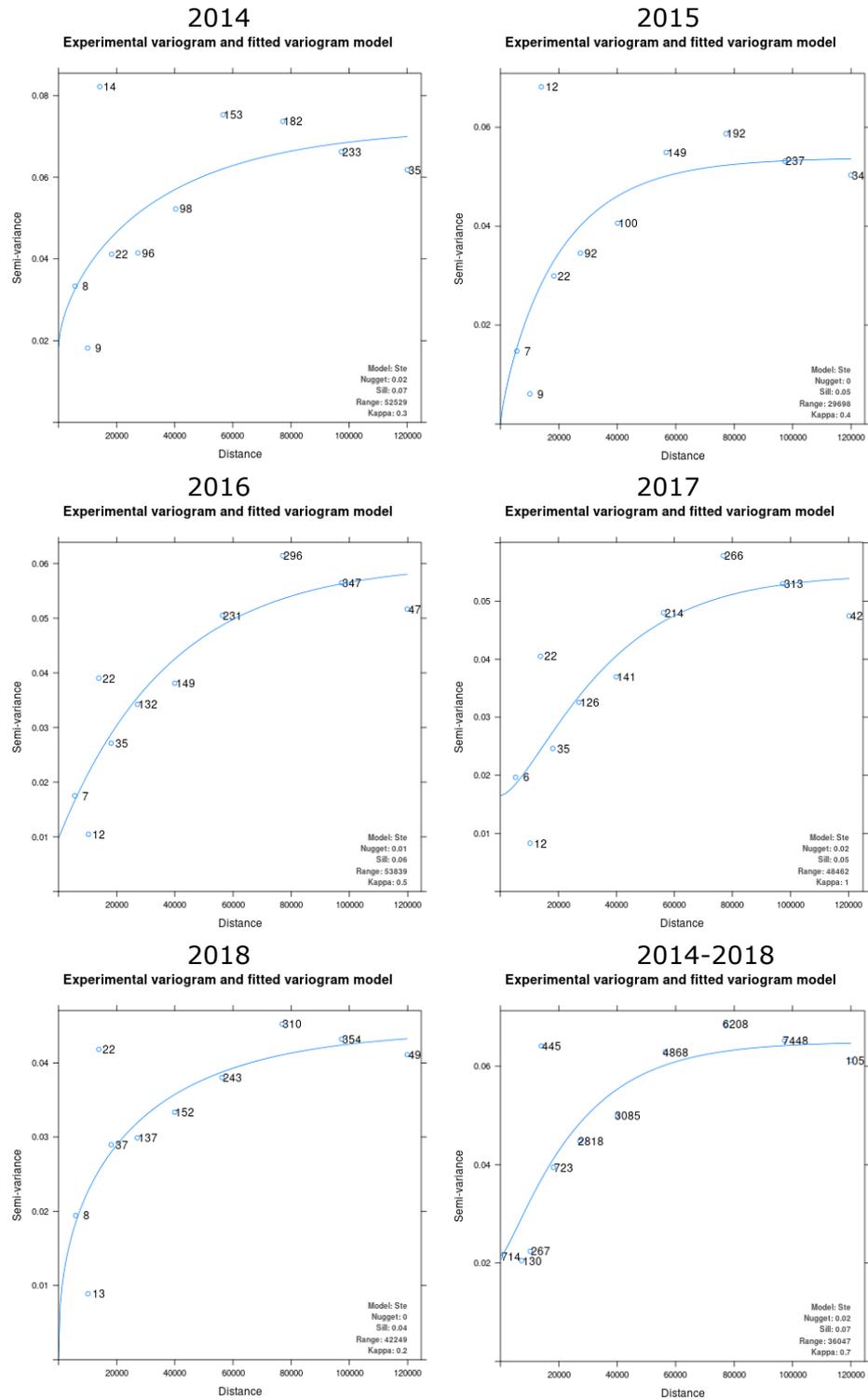


Figure 19. Experimental variogram (blue dots) and fitted variogram model (blue line) based on measured and modelled area averaged  $\text{NH}_3$  concentrations using individual years (2014-2018) and all 5 years.

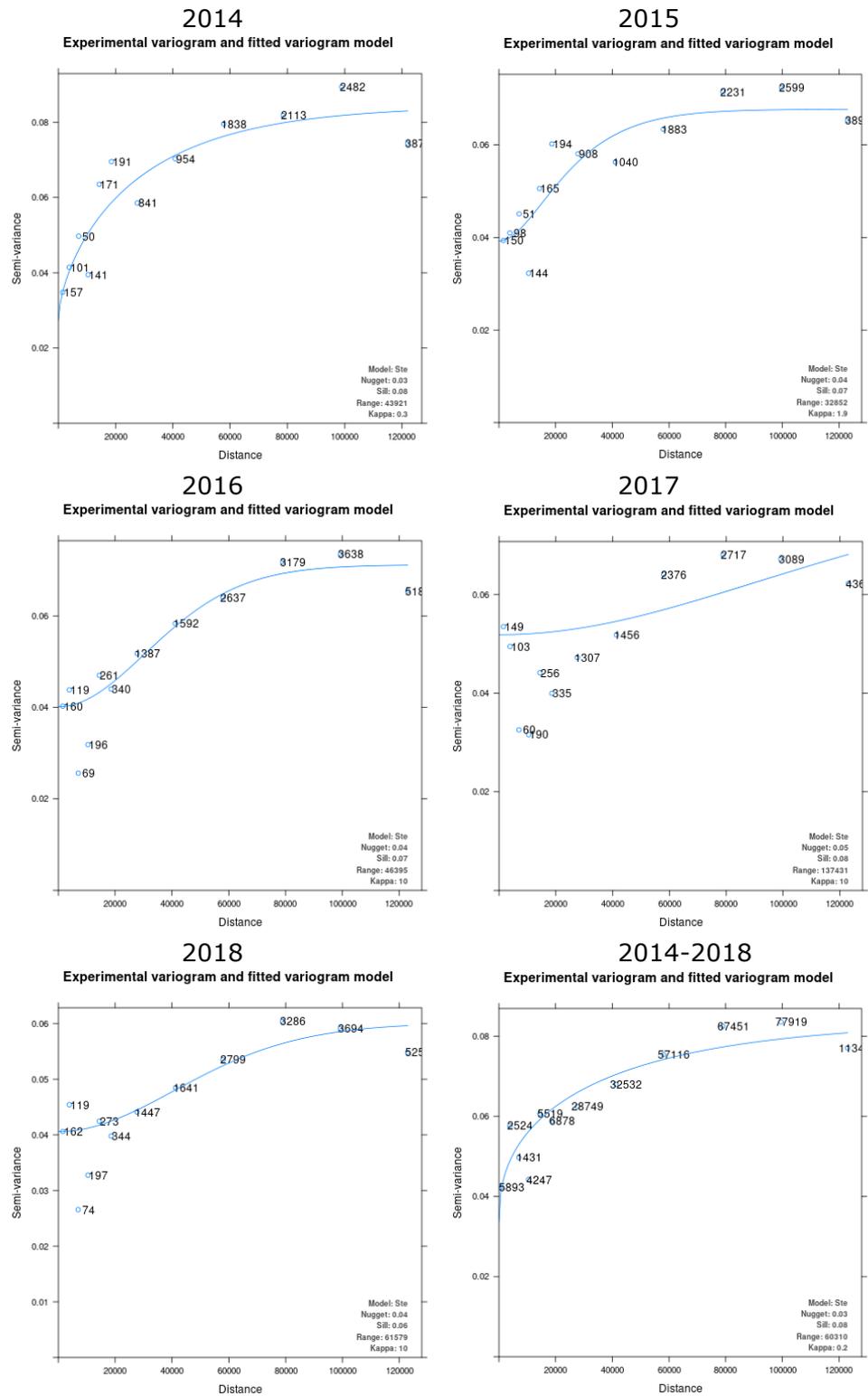
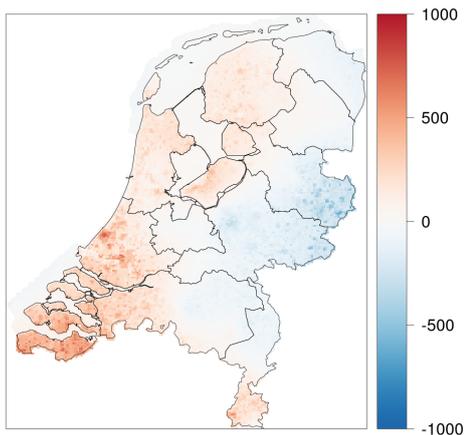


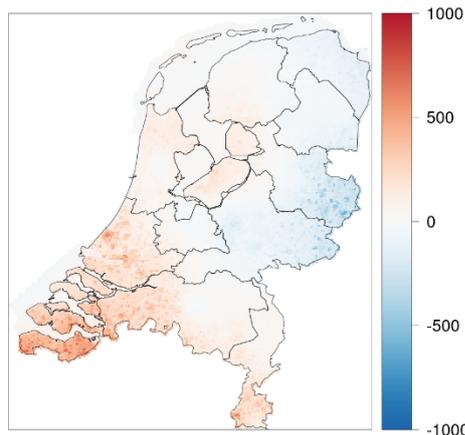
Figure 20. Experimental variogram (blue dots) and fitted variogram model (blue line) based on measured and modelled NH<sub>3</sub> concentrations at all the individual measurement locations in the Natura 2000-areas using individual years (2014-2018) and all 5 years.

## Appendix C. Difference in total N deposition due to new correction method in 2014-2018

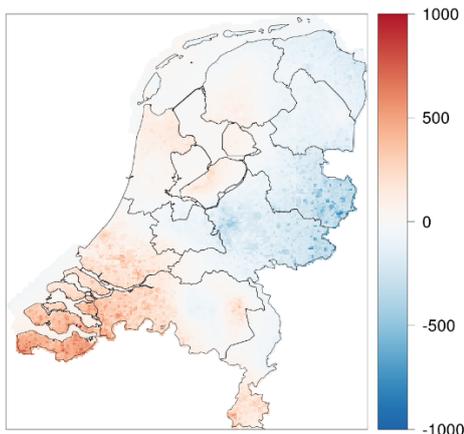
difference in total N deposition new - current method 2014



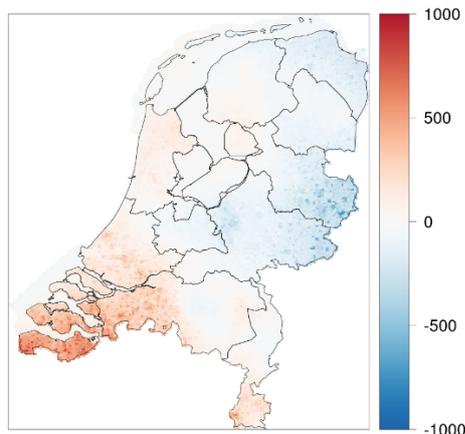
difference in total N deposition new - current method 2015



difference in total N deposition new - current method 2016



difference in total N deposition new - current method 2017



difference in total N deposition new - current method 2018

