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UNCERTAINTY ESTIMATES FOR NITROGEN DEPOSITION CALCULATIONS IN THE NETHERLANDS

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Abstract: Reliable estimates of the nitrogen deposition loads on sensitive ecosystems are important for developing policy to protect these ecosystems. In The Netherlands such estimates are published annually, for both past years and future years, by combining model outcomes of RIVM's OPS-LT model with observations. So far, a framework for assessing the quality of these estimates was lacking. Here we present a method to assess the uncertainty in the nitrogen deposition estimates, by comparing estimates of both deposition fluxes and concentrations with observations in The Netherlands over the period 2005-2021. The fluxes that are considered in this study are the dry deposition fluxes of ammonia, nitrogen oxides and of the secondary inorganic aerosols, and the wet deposition fluxes of both oxidized and reduced nitrogen. After deriving the uncertainties in these separate fluxes, they are combined to obtain the uncertainty in the total nitrogen deposition flux. We found that the uncertainty in the estimated local total nitrogen deposition on N2000 habitats is 60-70% (2σ). Uncertainties in the dry deposition velocities contribute most to the uncertainty in the total nitrogen deposition. These results give policy makers insights in the uncertainties in the reported values. In addition, our insights help to direct research efforts into model improvements and measurement strategies. Finally, the presented methods could potentially be applied to other models to compare the performance between models.

Key words: *Uncertainty, Nitrogen deposition, OPS model***.**

INTRODUCTION

Nitrogen, in its reactive forms, is an important nutrient for plant growth. However, too much nitrogen can be detrimental for ecosystems. It causes nitrogen tolerant species to outcompete more sensitive plants and fungi, and it contributes to acidification of soils. The atmosphere is the dominant source of nitrogen in natural ecosystems. Reactive nitrogen is emitted to the air by combustion processes and by livestock, and is transported by the atmosphere to deposit on plants, water and soil (Figure 1).

In The Netherlands, maps of nitrogen deposition loads are published annually by the National Institute for Public Health and the Environment (RIVM), at a resolution of 1x1km (GDN maps; Hoogerbrugge et al., 2023) and at a higher resolution of 1 ha for sensitive Natura 2000 (N2000) habitats (Marra et al., 2023). Nitrogen deposition maps are based on computations with the Operational Priority Substances (OPS-LT) model (Sauter et al., 2023). The model results are bias corrected using an extensive network of concentration measurements and wet deposition measurements. These maps, together with critical deposition loads that have been stablished for all N2000 habitats in The Netherlands, are the basis for Dutch policy that is being developed to reduce nitrogen emissions and protect sensitive ecosystems.

Given the central role of the nitrogen deposition maps in policy making it is important to provide uncertainty estimates of the reported values. Besides informing policy makers on the quality of the maps, such uncertainty estimates can yield valuable insights that help to direct research efforts into model improvements and measurement strategies. Finally, uncertainty estimates can be used to compare performance between models.

Assessing the uncertainty of models can be done in roughly two ways. The first approach is to estimate the uncertainty of the model input and its parameters, and study how these uncertainties propagate through the model to obtain an estimate of the uncertainty in model output. Such approach requires uncertainty estimates of all relevant model input and parameters, which are not always available. The second approach is simply comparing the model output with observations. This approach is only feasible when sufficient observations are available, and requires that these observations are not used for model calibration.

In this study we demonstrate how we derived uncertainty estimates for the Dutch nitrogen deposition maps using a combination of the two approaches described above. Our method makes use of the extensive concentration measurement networks of The Netherlands to obtain uncertainty estimates of the concentration calculations, and relies on uncertainty propagation to arrive at uncertainties in the total nitrogen deposition flux.

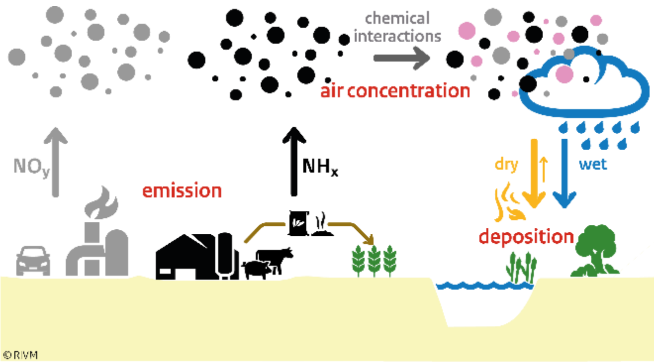

Figure 1. Pathways for nitrogen deposition.

METHODS

Uncertainty in the sum of fluxes

The OPS-LT model considers six pathways for nitrogen deposition (Sauter et al., 2023; Figure 1).

1. dry deposition of the primary reduced compounds, F_d NH_3
2. dry deposition of the primary oxidized compounds, F_d NO_x
3. dry deposition of the secondary reduced compounds, F_d NH_4^+
4. dry deposition of the secondary oxidized compounds, F_d $HNO_3 + NO_3^-$
5. wet deposition of the reduced compounds, F_w NH_x
6. wet deposition of the oxidized compounds, F_w NO_y

The primary oxidized compounds are composed of the gases NO, NO_2 and HNO_2, whereas the secondary compounds are mostly in the form of particulate matter.

To obtain an uncertainty estimate for the sum of these fluxes s_{tot}, we need to compute uncertainties in each of the fluxes and propagate them. The variance in the sum of dependent random variables is calculated with the sum of (co-)variances:

$$s_{\mathrm{tot}}^2 = \sum_{i=1}^{6} s_i^2 + 2 \sum_{j=1}^{5} \sum_{i=j+1}^{6} s_i s_j \rho_{ij} \tag{1}$$

where the first term takes the sum of uncertainty of the fluxes s_i, and represents the uncertainty if the random variables were independent. In our case this means that the uncertainties in the deposition fluxes are independent. This is certainly not the case, given that the deposition fluxes are governed mostly by the same processes, regardless of the compound that is being deposited. Therefore we also need to consider the sum of covariances between uncertainties of flux pairs, represented by the second term in equation (1). The covariance between uncertainties of flux pairs is the product of their uncertainties, $s_i s_j$, which we estimate at all available measurement sites, and the correlation between model residuals, ρ_{ij}, which we can only estimate at sites where both fluxes are measured. Hence, to obtain the uncertainty in total nitrogen deposition we need uncertainties in each flux, and the correlation of uncertainties between each flux pair.

Uncertainty estimation for measured fluxes

We define the uncertainty in a quantity X_{calc} as:

$$s_{X,\text{calc}}^2 = \frac{\sum_{i=1}^{n}(X_{i,\text{obs}} - X_{i,\text{calc}})^2}{n} - s_{X,\text{obs}}^2 \quad (2)$$

The first term is the average of the squared differences between the calculated and the observed value of X. With $X_{i,\text{obs}}$ and $X_{i,\text{calc}}$ being the observed and the calculated value of X at site i, and n being the number of observations. The second term subtracts the observation uncertainty, $s_{X,\text{obs}}$, which explains part of the difference between observation and calculation. Observation uncertainties were obtained from literature (Siteur et al., 2023), and are assumed to be independent of the uncertainties in the calculations.

The published nitrogen deposition maps are bias-corrected using the same observations as used in this study. To make a fair comparison between bias-corrected model output and observations, a leave-one-out cross-validation was performed. This means that $X_{i,\text{calc}}$ represents model output that is bias corrected using all observations except for $X_{i,\text{obs}}$.

Uncertainty estimation for unmeasured fluxes

We can apply equation (2) to the wet deposition fluxes of NH_x and NO_y (both 135 annual averages from six sites) and the dry deposition of NH_3 (18 annual averages from three sites). However, no measurements of dry deposition fluxes of the other compounds are available. To estimate the uncertainty in these unmeasured fluxes, we make use of the relationship

$$F_d = V_d \cdot C \quad (3)$$

where V_d is the dry deposition velocity and C is the concentration. Hence, the uncertainty in the dry deposition flux is composed of uncertainties in V_d and C.

By estimating the uncertainty in C and V_d, and assuming these uncertainties are independent, we can propagate them to obtain an uncertainty in F_d using (Goodman, 1960):

$$s_{F_d}^2 = F_d^2\left[\frac{s_{V_d}^2}{V_d^2} + \frac{s_C^2}{C^2} + \frac{s_{V_d}^2}{V_d^2}\frac{s_C^2}{C^2}\right] \quad (4)$$

The uncertainties in the concentrations are obtained from concentration measurements (NH_4^+ and NO_3^- both 70 annual averages from four sites, NO_2: 793 annual averages from 92 sites). Uncertainties in V_d of the unmeasured dry deposition fluxes are unknown. However, we can derive a relative uncertainty in V_d of NH_3, and use that as an approximation for the uncertainties in the dry deposition velocity of the unmeasured fluxes. This can be done by rewriting equation (4) to obtain:

$$\frac{s_{V_{d,\text{eff}}}^2}{V_{d,\text{eff}}^2} = \left[\frac{s_{F_d}^2}{F_d^2} - \frac{s_C^2}{C^2}\right]\left[1 + \frac{s_C^2}{C^2}\right]^{-1} \quad (5)$$

Here the relative uncertainty in s_C is obtained directly from comparison with NH_3 measurements (3123 annual averages at 278 sites) and $V_{d,\text{eff}}$ is the effective deposition velocity for NH_3. The effective deposition velocity is the long term average deposition velocity, resulting from deposition and reemission of NH_3 from soils and vegetation. We apply the obtained relative uncertainty for the effective deposition velocity of NH_3 to the unmeasured fluxes using equation (4), after subtracting the uncertainties due to reemission (see Siteur et al., 2023). We do this because reemission is only significant for NH_3, not for the other compounds.

Correlations between uncertainties in flux calculations

Correlation between uncertainties of two fluxes can be estimated directly from residuals, provided that both fluxes are measured at the same locations during the same period. Unfortunately, in The Netherlands this is only the case for the wet deposition fluxes. For dry deposition we therefore make use of the many concentration measurements that are available with overlapping locations and period. Assuming independence between concentrations and deposition velocities, the correlations in concentration uncertainties can be translated to correlations in deposition uncertainties using (Bohrnstedt and Goldberger, 1969):

$$\rho_{F_{d,i}F_{d,j}} = \frac{F_{d,i}}{s_{F_{d,i}}}\frac{F_{d,j}}{s_{F_{d,j}}}\left(\rho_{C_iC_j}\frac{s_{C_i}}{C_i}\frac{s_{C_j}}{C_j} + \rho_{V_{d,i}V_{d,j}}\frac{s_{V_{d,i}}}{V_{d,i}}\frac{s_{V_{d,j}}}{V_{d,j}} + \rho_{C_iC_j}\frac{s_{C_i}s_{C_j}}{C_iC_j}\rho_{V_{d,i}V_{d,j}}\frac{s_{V_{d,i}}s_{V_{d,j}}}{V_{d,i}V_{d,j}}\right) \quad (6)$$

Here values of $\rho_{C_iC_j}$ are be computed with:

$$\rho_{C_iC_j} = \frac{1}{s_{C_i}s_{C_j}} \frac{\sum_{k=1}^{m}(C_{i,\text{obs}} - C_{i,\text{calc}})(C_{j,\text{obs}} - C_{j,\text{calc}})}{m} \tag{7}$$

Here m is the number of observations of C_i and C_j that overlap in place and time. In this equation s_C is only estimated using these overlapping observations. In absence of measurements, we estimate the correlation between the uncertainties of deposition velocities by computing the correlation between the modelled deposition velocities in N2000 areas.

$$\rho_{V_{d,i}V_{d,j}} = \frac{1}{s_{V_{d,i}}s_{V_{d,j}}} \frac{\sum_{k=1}^{l}(\langle V_{d,i,\text{calc}}\rangle - V_{d,i,\text{calc}})(\langle V_{d,j,\text{calc}}\rangle - V_{d,j,\text{calc}})}{l} \tag{8}$$

Where l is the number of 1x1 km grid cells that contain N2000 habitats, and $\langle \quad \rangle$ takes the average over these grid cells. In this equation, s_{V_d} represents the standard deviation in modelled V_d values, instead of the uncertainty in V_d.

RESULTS

Table 1 shows the relative uncertainties, obtained by combining comparison with observations and uncertainty propagation. The highest relative uncertainties were found for the dry deposition fluxes. This is mainly due to uncertainties in the dry deposition velocity. On average, the dry deposition of NH_3 contributes most to deposition in N2000 areas. Therefore, the absolute uncertainties of this flux will in general be higher than for the other dry deposition fluxes.

Table 1. Average contribution of each flux to the total nitrogen deposition in sensitive N2000 habitats according to the calculated deposition maps (2005-2021) and uncertainty in calculations of concentrations (*C*), dry deposition velocities (V_d) and deposition fluxes (*F*), expressed as 1 sigma relative uncertainty. Uncertainties are estimated directly from observations (Equation 2; green), through propagation of uncertainties (Equation 3 and 4; blue) and approximated using the uncertainty of the NH_3 dry deposition velocity and removing uncertainties related to reemission (purple).

Flux	**Average contribution**		**Relative uncertainty**		
	mol/ha/y	*%*	***C***	V_d	***F***
F_d NH_3	575	37	21%	55%	60%
F_d NO_x	227	15	8%	44%	45%
F_d NH_4^+	29	2	35%	44%	58%
F_d $HNO_3 + NO_3^-$	120	8	21%	44%	50%
F_w NH_x	415	27	-	-	16%
F_w NO_y	196	13	-	-	13%

We can use the relative uncertainties in Table 1, and multiply these with the fluxes as computed at a given location. This gives the absolute uncertainties in the fluxes at that location. These absolute uncertainties and the correlation between uncertainties of the fluxes (Table 2), can then be entered in Equation 1 to obtain the estimated uncertainty of the total nitrogen deposition. Doing this for all computed 1x1 km resolution deposition calculations for The Netherlands yields the uncertainty map of Figure 2A.

Table 2. Correlation between uncertainties in deposition fluxes

ρ	F_d NH_3	F_d NO_x	F_d NH_4^+	F_d $HNO_3 + NO_3^-$	F_w NH_x	F_w NO_y
F_d NH_3	1.00	0.74	0.58	0.67	-0.03	-0.09
F_d NO_x		1.00	0.55	0.57	-0.01	-0.05
F_d NH_4^+			1.00	0.76	0.34	0.24
F_d $HNO_3 + NO_3^-$				1.00	0.30	0.22
F_w NH_x					1.00	0.67
F_w NO_y						1.00

To get a sense of the uncertainty in deposition on N2000 habitats, we computed the average uncertainty of the uncertainty map of Figure 2A, weighted with the cover of relevant N2000 habitats (Figure 2B). This gives an average uncertainty of 491 mol/ha/y, or 31% of the total nitrogen deposition on N2000 habitats. However, locally the uncertainties can be higher or lower, depending on the composition of the nitrogen that is deposited (see Figure 2A). The relative uncertainty is between 30-35% for most N2000 habitats. Expressed in two standard deviations this gives uncertainties in the range of 60-70%.

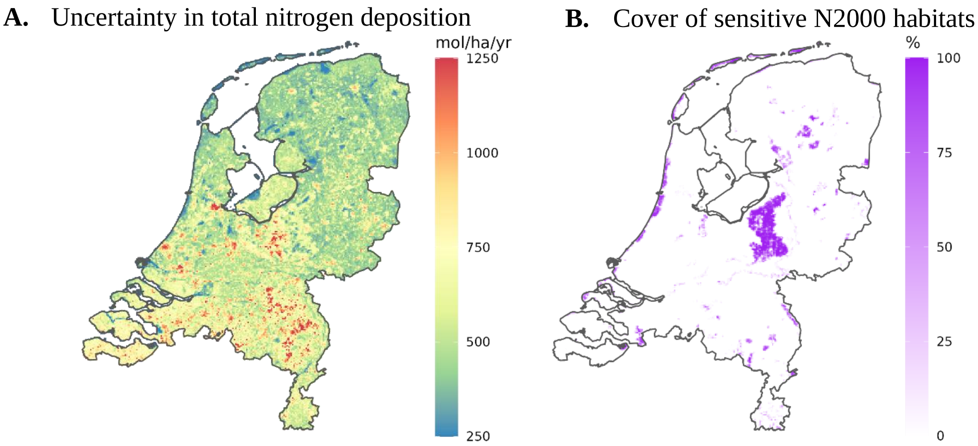

Figure 2. A: 1x1 km resolution maps of uncertainty in total nitrogen deposition. B: the percentage of relevant N2000 habitats per grid cell. N2000 are considered sensitive if the critical load is less than 2400 mol/ha/y.

CONCLUSION AND DISCUSSION

We found that the uncertainty in the calculated total nitrogen deposition on 1x1 km grid cells that contain N2000 habitats is 60-70% (2σ). This uncertainty estimate gives context to the deposition maps that are published by the RIVM, and informs policy in trying to protect sensitive ecosystems. The obtained uncertainty estimates can also be used to direct research efforts. For the OPS model used in the Netherlands, it is clear that efforts need to be made in reducing uncertainties in the deposition velocity. The presented method gives an approximation of the uncertainty in the total nitrogen deposition, even in cases when not all contributing nitrogen deposition fluxes are measured. This allows applying it to a broad range of countries and models. Evaluation of our method would require long term measurements of dry deposition fluxes of NO_x and particulate matter, which are currently not available for The Netherlands.

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