



Local-Scale Inversion of Agricultural Ammonia Emissions: A Case Study on Schiermonnikoog, the Netherlands

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Abstract. Ammonia (NH₃) is a major component of reactive nitrogen and a key alkaline gas in the atmosphere. With an increase in fertiliser application due to the intensification of agricultural livestock and industrial production, large amounts of NH₃ are emitted into the atmosphere at an increasing rate, posing a threat to both human and ecosystem health and contributing to climate change. Although many studies have examined the ammonia budget, large uncertainties remain in its emissions, distribution, and deposition.

Top-down, or inversion methods, using measurement data to estimate emissions, can help reduce these uncertainties. In this study, we have applied local-scale Bayesian inversions using ground-based measurements and the LOTOS-EUROS air quality model, with high-resolution emission inventories as prior input. To improve robustness, we estimated observational error by combining reported measurement uncertainties with residual errors and optimized Chi-Square statistics.

We applied this approach to the island of Schiermonnikoog in the Netherlands, where GVE (grazing livestock units) on Schiermonnikoog decreased from 639 to 541, with a particularly notable reduction in dairy cattle, corresponding to a 23% reduction in ammonia emissions between 2019 and 2022. Our inversion captured a similar trend, estimating a 51% decrease, with associated uncertainty derived from the posterior error covariance.

Moreover, we developed a method to assess the usefulness of individual observations and propose that adding a single high-quality continuous measurement in a strategically chosen location can significantly enhance the inversion performance. This strengthens the observational constraint and enhances the system's ability to resolve temporal variations in emissions.

1 Introduction

Ammonia (NH₃) is a crucial component of the global nitrogen cycle, playing a fundamental role in agriculture and atmospheric chemistry. As the most abundant alkaline gas in the atmosphere, it significantly influences air quality, ecosystem health, and climate. Since discovery of the the synthesis of ammonia from atmospheric dinitrogen in 1908 (Haber, 1920), the application in fertilisers has revolutionised global food production, sustaining over half of the world's population (Erisman et al., 2008; Smil, 2004, 2002). However, this agricultural success comes at an environmental cost. The widespread use of synthetic fertilisers and intensification of livestock farming has led to increasing atmospheric ammonia emissions, with profound consequences for air pollution, nitrogen deposition, and climate change (Erisman et al., 2008, 2013; Zhang et al., 2020; McCubbin et al., 2002).



25 Understanding ammonia's behavior in the atmosphere remains challenging due to its high spatial and temporal variability. Once emitted, ammonia has a relatively short atmospheric lifetime, often a few hours to a few days (Dammers, 2017; Dammers et al., 2019; Zhang et al., 2021), and can be rapidly deposited or converted into secondary particulate matter (Behera et al., 2013; Wyer et al., 2022). These processes create a dynamic system where emissions, transport, and deposition fluctuate significantly across different scales. While ammonia is a well-known contributor to nitrogen deposition and particulate matter
30 formation, large uncertainties persist in its atmospheric budget, particularly regarding emission estimates, spatial distribution, and deposition pathways.

1.1 Bridging the Gap: From Emission Inventories to Observational Constraints

Traditional ammonia emission inventories rely on bottom-up estimates, which aggregate data from agricultural activities, industrial processes, and other sources based on production statistics and emission factors (Eggleston et al., 2006; Kuenen et al.,
35 2022). However, these inventories suffer from inherent limitations. They often lack high temporal resolution, vary significantly between regions, and fail to capture the real-time dynamics of ammonia fluxes. Ammonia emissions are particularly sensitive to meteorological conditions, such as temperature and humidity, which can drive short-term fluctuations that are not well-represented in bottom-up models (Sutton et al., 2013; Ge et al., 2023).

To overcome these challenges, top-down approaches that integrate observational data into atmospheric models have gained
40 traction. These methods use measurements from satellites and ground-based networks to constrain and refine emission estimates, offering a more dynamic and data-driven perspective on ammonia fluxes. Several studies have successfully applied top-down techniques to improve ammonia emission estimates. To name a few, Paulot et al. (2014) used the adjoint GEOS-Chem model with ammonium wet deposition fluxes to infer emissions across the United States, Europe, and China. Similarly, Zhang et al. (2018) combined satellite TES (Tropospheric Emission Spectrometer) data with inverse modeling to enhance
45 ammonia emission inventories over China. More recently, van der Graaf et al. (2022) demonstrated the value of assimilating CrIS ammonia retrievals into the LOTOS-EUROS chemistry transport model, significantly improving the spatial and temporal representation of emissions. Additionally, Cao et al. (2022) implemented a 4D-Var inversion that accounted for bi-directional ammonia fluxes, leading to an accurate depiction of seasonal variability in ammonia exchange between the surface and the atmosphere.

50 Despite these successes, most top-down studies have focused on global or mesoscales, leaving a critical gap in understanding ammonia emissions at localized scales where emission sources, meteorology, and deposition processes interact in complex ways.

1.2 The Need for Local-Scale Ammonia Studies

Ammonia emissions and concentrations exhibit strong spatial and temporal variability due to their sensitivity to environmental
55 conditions and short atmospheric lifetime. This spatial variability is particularly pronounced within hundreds of meters to a few kilometers of emission sources (Schulte et al., 2022). NH_3 emission potential can increase by a factor of 9 with a temperature rise of twenty degrees Celsius (Ge et al., 2023; Sutton et al., 2013). The atmospheric lifetime of ammonia varies significantly

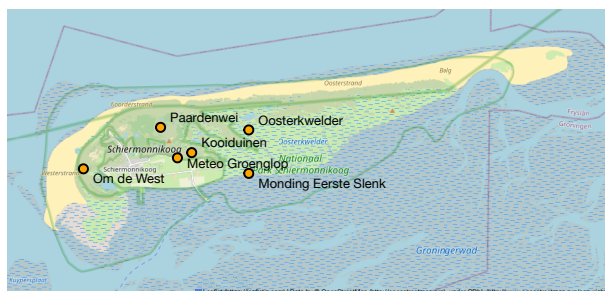


Figure 1. The map of Schiermonnikoog is provided by ©OpenStreetMap contributors (2017), in which the orange circles denote the MAN measurement sites.

depending on location and environmental factors. Furthermore, the average lifetime of ammonia in the atmosphere is between a few hours to a few days (Dammers, 2017; Dammers et al., 2019; Zhang et al., 2021; Norman and Leck, 2005). Given this variability, localized studies are essential to capturing the fine-scale processes driving ammonia dynamics and to designing targeted mitigation strategies.

A particularly relevant case study is the nitrogen crisis in the Netherlands, where high ammonia emissions from agricultural activities have led to excessive nitrogen deposition, biodiversity loss, and regulatory interventions (Erisman, 2019; Stokstad, 2019; Erisman et al., 2021). Within this context, Schiermonnikoog, a small island in the north of the Netherlands, serves as an ideal testbed for ammonia emission reduction at a fine spatial scale, shown in Fig. 1. The largest part of the island falls under the National Park Schiermonnikoog, with rich landscapes that include dunes, beaches, forests, mudflats, and polders. The National Park is one of the most important nature areas in the Netherlands, and its habitats are sensitive to nitrogen deposition (Sival and Strijkstra-Kalk, 1999). However, intensive dairy farming in the island's 275-hectare polder has historically contributed to ammonia loads exceeding critical thresholds (van Wijnen and Bakker, 1997).

In response, a feasibility study by Erisman and Hofstee (2016) proposed nature-inclusive agricultural strategies to mitigate ammonia emissions while supporting the economic viability of local farmers. Although the transition began in 2016, a more pronounced reduction occurred between 2019 and 2022, during which GVE (grazing livestock units) decreased from 639 to 541, with dairy cattle numbers dropping from 510 to 363, according to KringloopWijzer data (van Dijk et al., 2023). These changes contributed to an estimated 23% reduction in ammonia emissions, providing a valuable opportunity to evaluate whether current monitoring systems can effectively capture such changes and where improvements may be needed.

1.3 Observational Challenges and the Role of LOTOS-EUROS

Despite the availability of satellite observations, retrieving reliable ammonia concentrations over small islands like Schiermonnikoog remains inherently challenging. The limited land area of the island, combined with the coarse spatial resolution of current satellite instruments, means that the entire island and often parts of the surrounding sea are captured within a single satellite footprint. This spatial mismatch reduces the ability to resolve localized emission patterns. Moreover, the low ammonia column densities and weak thermal contrast between land and sea further degrade the retrieval quality and increase uncertain-



ties in satellite-based NH_3 measurements (Van Damme et al., 2014). As for the ground-based measurements, no high-temporal resolution ammonia monitoring stations exist in the region, leaving monthly observations from the Measuring Ammonia in Nature (MAN) network as the only continuous source of in situ data (Lolkema et al., 2015; Noordijk et al., 2020). These
85 ground-based measurements are crucial for validating and refining ammonia models at a local scale.

To bridge the gap between observations and models, we employ LOTOS-EUROS, a state-of-the-art regional chemistry transport model specifically designed for air quality applications in Europe. This model integrates atmospheric transport, deposition, and chemical transformations of ammonia, providing a high-resolution framework for emission quantification. One of its key advantages is its labelling functionality, which allows for precise source attribution, distinguishing between
90 agricultural, industrial, and natural contributions to ammonia concentrations. By combining MAN network data with LOTOS-EUROS simulations, we aim to refine spatial and temporal emission estimates, ultimately improving ammonia monitoring and mitigation strategies at the local scale.

1.4 Study Overview

In this study, we aim to refine local-scale ammonia emission estimates using a Bayesian inversion framework supported by
95 atmospheric modeling and ground-based observations. We begin by simulating baseline ammonia concentrations with the LOTOS-EUROS chemical transport model and comparing them to measurements from the MAN network. To support the inversion, we generate a set of controlled perturbation experiments to compute the Jacobian matrix and produce synthetic observations. These inputs allow us to quantify the sensitivity of the model and assess uncertainty through detailed error characterization. The inversion analysis proceeds in three stages:

- 100 – A comprehensive test incorporating MAN network uncertainties to evaluate model sensitivity.
- A refined inversion using real MAN network observations.
- A more optimized observation network design, outlining strategies to improve ammonia monitoring across different timescales.

These findings will provide valuable insights into the effectiveness of current monitoring networks and formulate future mea-
105 surement strategies to better quantify ammonia emissions at fine temporal scales.

2 Data and Methodology

To evaluate ammonia emissions at a local scale, we use a Bayesian inversion framework combining atmospheric modeling, synthetic data experiments, and observational constraints. This chapter begins with an overview of real-world ammonia mea-
110 surements from the MAN network, followed by a description of the synthetic data used for controlled tests. We then introduce the LOTOS-EUROS chemical transport model, along with the emission sources and prior estimates used in the simulations. Finally, we detail the inversion algorithm and the associated error characterisation.



2.1 Observations

2.1.1 In-situ measurements

The MAN (Measuring Ammonia in Nature) network is extensively used for monitoring atmospheric ammonia across the Netherlands. The spatial distribution of MAN sites is shown in Fig. 2. Unlike active optical techniques, the MAN network employs passive samplers, which measure monthly average ammonia concentrations via chemical absorption. This method is significantly cheaper than active optical techniques. Detailed descriptions of the measurement technique and associated uncertainties are provided in Lolkema et al. (2015); Noordijk et al. (2020). In this study, we utilize monthly data from 45 MAN sites—6 of which are located on Schiermonnikoog—for both annual and monthly inversion analyses.

In addition to the MAN network, the Netherlands also employs optical measurement techniques with higher temporal resolution. The Dutch National Air Quality Monitoring Network (Landelijk Meetnet Luchtkwaliteit, LML) operates miniDOAS (active differential optical absorption spectroscopy) instruments, providing hourly ammonia concentrations (Berkhout et al., 2017; van Zanten et al., 2017). However, the LML network has far fewer monitoring sites compared to MAN. Since our study area lacks LML measurements, we introduce synthetic LML-like observations in the final section to explore their potential impact on inversion performance.

2.1.2 Simulated observations

To better understand the inversion process, we conducted a series of controlled tests using simulated observations. These tests were based on high-resolution hourly data, which were subsequently averaged into monthly values to align with the temporal scale of the MAN measurements. This setup enabled us to quantify observational uncertainties and assess how synthetic measurements can enhance the spatial and temporal coverage of the current monitoring network.

Using LOTOS-EUROS output, we generated synthetic “observational data” that mimic the current monitoring network. A configuration of the simulated errors is provided in:

$$s_{\text{full error}} = \sqrt{0.90^2 + (0.28 \times c)^2} \times \mathcal{N}(0, \sigma^2), \quad (1)$$

in which c is the surface concentrations, designed to reflect the existing MAN measurement error structure (Lolkema et al., 2015; Noordijk et al., 2020).

Additionally, we integrated LOTOS-EUROS output into the current monitoring framework as additional measurements, exploring its potential to support monitoring network design by providing high-resolution, high-quality measurement proxies.

2.2 LOTOS-EUROS

We use the state-of-the-art air quality model LOTOS-EUROS (LOng-Term Ozone Simulation and European Operational Smog model), developed by TNO, as the forward model in our inversion framework. This study employs LOTOS-EUROS version 2.3.000, with its detailed configuration summarized in Table 1 (Manders et al., 2017; Manders-Groot and LOTOS-EUROS team, 2023).

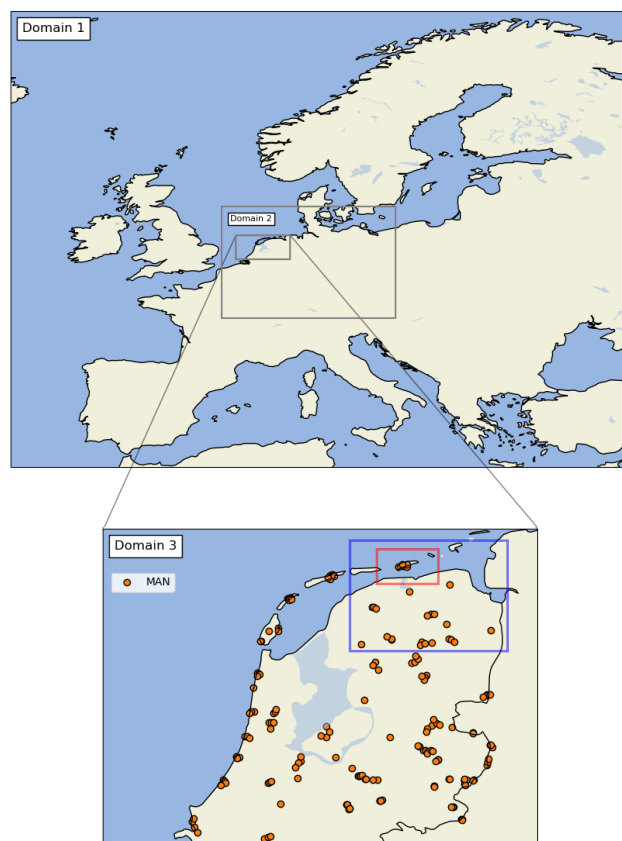


Figure 2. The nested domain configuration is shown. In Domain 3, the orange circles denote the MAN measurement sites. Schiermonnikoog is located in the red box. The measurement located within the blue box are used for inversion.

The model is driven by Integrated Forecast System (IFS) from the European Centre for Medium-Range Weather Forecasts (ECMWF Hersbach et al., 2020). Three-hourly meteorological parameters are interpolated to one-hour resolution for finer
145 temporal representation. To optimize computational efficiency and data storage, nested domains are used (Fig. 2): the coarsest domain covers from (35°N, 15°W) to (70°N, 35°E) with 0.5° and 0.25 ° resolution; the finest domain covers from (50.6°N, 3.15°E) to (53.7°N, 7.5°E) with 1.7 km × 2.15 km resolution.

Beyond standard air pollution modeling, LOTOS-EUROS includes a source apportionment function, which labels emissions based on their spatial and sectoral origins. In this study, we use this function to track ammonia contributions from key regions:

- 150
- Countries: Germany, Denmark.
 - Dutch regions: Schiermonnikoog, Groningen, Friesland, Drenthe, Gelderland, Overijssel (5 nearest provinces), and other locations.

**Table 1.** LOTOS-EUROS model configuration

Parameters	Setting	Notes/Citation
Time period	January 1 to December 31, 2019 and 2022	with a spin-up for 15 days
Spatial resolution	$0.025^\circ \times 0.0125^\circ$	$1.7 \text{ km} \times 2.15 \text{ km}$
Emission	CAMS-REG v5.1 REF2, GrETa and ER,	Kuenen et al. (2022), year 2019
Meteorology	ECMWF Reanalysis v5 (ERA5)	Hersbach et al. (2020)

For spatial resolution, the first and second column denote $(\Delta\text{lon} \times \Delta\text{lat})$ and $(\Delta x \times \Delta y)$, respectively.

Each emission source is further classified into two sectors: agricultural and non-agricultural emissions. This labeling framework enables precise source attribution and allows us to quantify the relative impact of various regions and sectors on ammonia concentrations over Schiermonnikoog. Detailed results are presented in Sect. 3.1.2.

2.3 Prior emission

The prior emissions used in the inversion, as well as the input for LOTOS-EUROS, are taken from the Copernicus Atmosphere Monitoring Service regional inventory (CAMS-REG v5.1 REF2, year 2019) for Domain 1 and GrETa and ER emission inventories for Domain 2 and 3. The temporal allocation follows the TNO-MACC (Monitoring Atmospheric Composition and Climate) inventory. This dataset includes emissions of major air pollutants, with detailed information available in Kuenen et al. (2021, 2022). The emissions are provided with a spatial resolution of $0.0167^\circ \times 0.0083^\circ$ in the finest domain.

In this study, we only optimize ammonia emissions from agricultural sources, as they are the dominant contributor to atmospheric NH_3 . Other emission sectors—traffic, residential, industrial, and transportation—are categorized as non-agricultural emissions. Further details on the agricultural sector are provided in the Supplement; more details can be referred to Kuenen et al. (2022, 2021).

2.4 Bayesian Inversion algorithm

To optimize ammonia emissions using observational constraints, we apply a Bayesian inversion framework, which efficiently integrates errors from prior emissions, observations, and the model itself. This approach is highly flexible, allowing for its application at local scales with different observational datasets.

Bayesian inversion aims to estimate the posterior probability density function (pdf) of the state vector $\mathbf{x}$ given observations $\mathbf{y}$. The revised version is expressed as (Rodgers, 2000; Turner and Jacob, 2015):

$$\begin{aligned}
 P(\mathbf{x}|\mathbf{y}) &= \frac{P(\mathbf{y}|\mathbf{x})P(\mathbf{x})}{P(\mathbf{y})} \\
 &\propto \exp \left\{ -(\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a) \right. \\
 &\quad \left. - [\mathbf{y} - \mathbf{F}(\hat{\mathbf{x}})]^T \mathbf{S}_o^{-1} [\mathbf{y} - \mathbf{F}(\hat{\mathbf{x}})] \right\},
 \end{aligned} \tag{2}$$



175 where:

- $\mathbf{x}$ is defined in logarithmic space such that $\mathbf{x} = \ln(\hat{\mathbf{x}})$, where $\hat{\mathbf{x}}$ represents the actual (linear) state vectors. This log transformation ensures positivity and accommodates multiplicative uncertainty in emissions;
- $\mathbf{y}$ is the vector with observations, which is monthly data from 45 MAN sites;
- $\mathbf{F}$ operates on $\hat{\mathbf{x}}$, to simulate corresponding concentrations;
- $\mathbf{x}_a$ is the prior state vector in logarithmic form;
- $\mathbf{S}_a$ and $\mathbf{S}_O$ are the prior and observational error covariance matrices, respectively (details in Sect. 2.4.1).

The optimal state $\mathbf{x}$ is found by maximizing Eq. (2), which corresponds to minimizing the cost function:

$$\mathcal{J}(\mathbf{x}) = (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a) + [\mathbf{y} - \mathbf{F}(\hat{\mathbf{x}})]^T \mathbf{S}_O^{-1} [\mathbf{y} - \mathbf{F}(\hat{\mathbf{x}})], \quad (3)$$

185 where $\mathbf{x}$ is of shape $(n \times 1)$; $\mathbf{y}$ is $(m \times 1)$; $\mathbf{S}_a$ is $(n \times n)$; $\mathbf{S}_O$ is $(m \times m)$. Since $\mathbf{F}(\hat{\mathbf{x}})$ is nonlinear due to ammonia chemistry, we approximate it linearly using the Jacobian matrix:

$$\mathbf{K} = \nabla_{\mathbf{x}} \mathbf{F} = \frac{\partial \mathbf{y}}{\partial \mathbf{x}} = \frac{\partial \mathbf{y}}{\partial (\ln \hat{\mathbf{x}})} = \hat{\mathbf{K}} \text{diag}(\hat{\mathbf{x}}), \quad (4)$$

where $\mathbf{K}$ is the Jacobian matrix in logarithmic space; $\hat{\mathbf{K}}$ is the Jacobian in linear space, defined as $\hat{K}_{ij} = \partial \mathbf{y}_{(i)} / \partial \hat{\mathbf{x}}_{(j)}$

To solve the Eq. (3), we apply the Levenberg–Marquardt approach (Rodgers, 2000; Chen et al., 2022, 2023), iteratively
190 updating the state vector:

$$\mathbf{x}_{N+1} = \mathbf{x}_N + \left[\mathbf{K}_N^T \mathbf{S}_O^{-1} \mathbf{K}_N + (1 + \kappa) \mathbf{S}_a^{-1} \right]^{-1} \cdot \left[\mathbf{K}_N^T \mathbf{S}_O^{-1} (\mathbf{y} - \hat{\mathbf{K}} \hat{\mathbf{x}}) - \mathbf{S}_a^{-1} (\mathbf{x}_N - \mathbf{x}_a) \right], \quad (5)$$

where $\mathbf{x}_N$ is the state vector at the Nth iteration; $\mathbf{K}_N$ is the Jacobian matrix at the Nth iteration; κ is the coefficient for determining the convergence rate and is set as 10 (Chen et al., 2022); $\hat{\mathbf{K}} \hat{\mathbf{x}}$ is equivalent to the forward model. The uncertainty in the
195 optimized emissions is given by the posterior error covariance matrix:

$$\hat{\mathbf{S}} = \left(\mathbf{K}_N^T \mathbf{S}_O^{-1} \mathbf{K}_N + \mathbf{S}_a^{-1} \right)^{-1}, \quad (6)$$

where $\mathbf{K}_N$ is the Jacobian at the final iteration. The degree of freedom for signal (DFS) is then calculated from:

$$\text{DFS} = \text{trace}(\mathbf{A}), \quad (7)$$

where $\mathbf{A}$ is the averaging kernel matrix:

$$200 \quad \mathbf{A} = \frac{\partial \hat{\mathbf{x}}}{\partial \mathbf{x}} = \mathbf{I} - \hat{\mathbf{S}} \mathbf{S}_a^{-1}. \quad (8)$$

The DFS quantifies how much information is gained from the observations. Higher DFS values indicate a stronger observational constraint on the emissions.



2.4.1 Error covariance matrices

The state vector in our inversion framework includes not only agricultural emissions from Schiermonnikoog but also contributions from external sources. These external influences are determined using the labeling functionality in the LOTOS-EUROS model, which identify regions that significantly impact ammonia concentrations on the island. If a labeled area has a strong contribution to Schiermonnikoog's ammonia levels, then uncertainties in emissions from that region are likely to propagate to the island's atmospheric NH_3 concentrations. The state vector is constructed with five elements, where the first represents the local emission source and the remaining four represent external influences for fixing the boundary condition:

1. agricultural emissions from Schiermonnikoog;
2. total contribution to concentration (agricultural and non-agricultural) from Groningen;
3. total contribution to concentration from Friesland;
4. total contribution to concentration from Germany;
5. a composite term ("Other") representing contributions to concentration from all remaining sources and sectors.

A major source of uncertainty in ammonia emissions are volatilization rates, which varies with temperature. The re-emission potential can increase by a factor of 9 for a twenty degrees Celsius temperature rise (Ge et al., 2023; Sutton et al., 2013), which shows that meteorological errors may lead to inaccurate estimates of ammonia. To account for this variability, and given the use of a logarithmic state vector, the diagonal elements of the prior error covariance matrix are defined as $(\ln \beta)^2$, where $\beta = 2$ represents the assumed annual emission variability factor, while external influences are assigned a factor of 1.5. For monthly emission inversion, the variability factors are set to $\beta = 4$ for emissions and 2 for external influences.

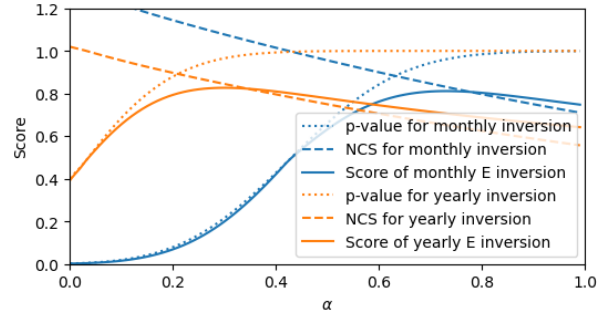
To estimate the observational error covariance matrix $\mathbf{S}_O$, we first follow the commonly used residual error method (Brasseur and Jacob, 2017):

$$\mathbf{S}_O = E[\varepsilon \varepsilon^T], \quad (9)$$

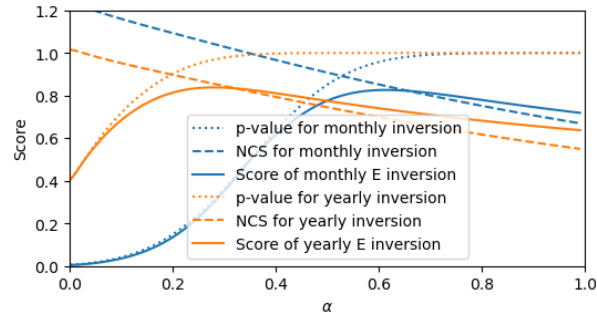
where:

- $\varepsilon = (z - \bar{z})$ is the residual error vector, assumed to represent random noise;
- $z = \mathbf{y} - \mathbf{F}(\hat{\mathbf{x}})$ denotes the misfit between observations and forward model output, assuming discrepancies arise primarily from emission uncertainties.

Initially, $\mathbf{S}_O$ is constructed using only diagonal elements with the residual error vector. However, this simplification introduces potential uncertainties. To improve representativeness, we adopt a hybrid approach that combines (1) residual error estimates and (2) established uncertainty descriptions from the MAN network (Noordijk et al., 2020; Lolkema et al., 2015). This integrated error representation enhances the robustness of the inversion system but might also overestimate the total uncertainty.



(a) 2019



(b) 2022

Figure 3. Evaluation of Chi-Square Test Results for the total observational error defined in Eq. (10). The model performance is scored by Eq. (11). A score is derived from the p-value and the Normalized Chi-square (NCS). An NCS value near 1 and a p-value close to 1 indicate that the error description is reasonable, reflecting a good fit and appropriate error covariance. Notably, the August and July 2019 data are eliminated because of the dune fire in 2019.

To assess the performance of this combined error formulation, we perform a series of Chi-Square tests and optimize the the integrated observational error to ensure an appropriate balance between model constraints and observational uncertainties (Rodgers, 2000). The total observational error is formulated as:

$$\epsilon = \sqrt{\epsilon_{\text{residual}}^2 + (\alpha \cdot \epsilon_{\text{MAN}})^2}, \quad (10)$$

where:

- $\epsilon_{\text{residual}} = (z - \bar{z})$;
- $\epsilon_{\text{MAN}} = \sqrt{0.90^2 + (0.28 \times c)^2}$, with c the monthly ammonia concentrations;
- α is a scaling factor optimized to best match model and observational uncertainty.



We define a performance score s_{χ^2} to simultaneously evaluate statistical consistency (via p-value) and goodness-of-fit (via normalized Chi-Square, NCS):

$$s_{\chi^2} = p \cdot e^{-|1-\text{NCS}|}, \quad (11)$$

where:

- p is the p-value from Chi-Square tests;
- $\text{NCS} = \frac{\mathbf{z}^T \mathbf{S}_0^{-1} \mathbf{z}}{\text{DOF}}$, with DOF as degrees of freedom.

This score reaches a maximum when both the model-observation agreement is close to ideal ($\text{NCS} \approx 1$) and the residuals are consistent with the assumed error distribution (high p-value). Note that here, DOF refers to the number of independent observational constraints used in the χ^2 calculation, and should not be confused with DFS, degrees of freedom for signal defined in Eq. (7), which measures how much information from observations is retained in the state vector after inversion. While both reflect aspects of information content, they apply to different parts of the inversion framework.

Based on the optimization (Fig. 3), we assign: for monthly emission inversions $\alpha = 0.5$; for annual emission inversions, $\alpha = 0.3$), reflecting the averaging over longer periods and the reduced influence of short-term fluctuations. For simulated observations, $\alpha = 0.3$ for monthly inversions and $\alpha = 0.15$ for annual inversions, reflecting the lower uncertainty in synthetic data compared to real-world observations. These adjustments ensure that the inversion framework maintains robustness while adequately capturing observational uncertainties across different timescales. Similar to Fig. 3, the optimization of Chi-Square statistics of simulated observations can be found in the supplement.

3 Results and Discussions

In this section, we first present the baseline results of the LOTOS-EUROS model, including the labelling function. Next, we validate the inversion framework using synthetic errors and simulated data. We then apply MAN measurements to perform the inversion on real-life data, analyze emission reductions, and investigate emission estimates across different timescales. Finally, we explore potential improvements to the inversion framework and propose an optimized observational design for enhanced emission monitoring.

3.1 Model performance and source apportionment

3.1.1 Model performance

Figure S1 illustrates the monthly variation of prior ammonia emissions and corresponding surface concentrations on Schiermonnikoog for 2019 and 2022. In both years, agricultural emissions peaked in March, following model time profile, with a secondary, smaller peak in August. Other local emissions, including non-agricultural sources, remain constant through the year. These emission patterns directly influence surface ammonia concentrations, which follow a characteristic bimodal seasonal cycle: a spring peak associated with manure spreading and a summer peak driven by higher temperatures and volatilization.



Table 2. The statistics of comparison between the model LOTOS-EUROS and the MAN measurements, including Pearson's correlation coefficient (r), root-mean-square error (RMSE), mean absolute error (MAE), mean absolute percentage Error (MAPE), normalized mean bias (NMB) and slope.

	MG	K	MES	O	OdW	P	All	Unit	
2019	r	0.48	0.57	0.41	0.49	0.55	0.34	0.41	unitless
	RMSE	6.39	3.27	3.15	1.89	1.80	2.22	3.49	$\mu\text{g}/\text{m}^3$
	MAE	5.05	2.30	2.42	1.52	1.41	1.78	2.41	$\mu\text{g}/\text{m}^3$
	MAPE	61	43	46	41	41	46	46	%
	NMB	-0.61	-0.42	-0.41	-0.18	-0.14	-0.22	-0.39	unitless
	slope	0.22	0.43	0.35	0.52	0.62	0.37	0.26	unitless
2022	r	0.64	0.75	0.87	0.78	0.78	0.84	0.67	unitless
	RMSE	3.24	1.57	1.37	1.27	1.34	1.25	1.81	$\mu\text{g}/\text{m}^3$
	MAE	2.96	1.36	1.20	0.90	0.94	0.89	1.38	$\mu\text{g}/\text{m}^3$
	MAPE	1.60	0.68	0.68	0.29	0.30	0.39	0.66	%
	NMB	-0.47	-0.22	-0.24	0.22	0.23	-0.03	-0.17	unitless
	slope	0.72	0.64	0.92	0.54	0.60	1.02	0.77	unitless

The abbreviations correspond to the site names: Schiermonnikoog-Meteo Groenglop, -Kooидуinen, -Monding Eerste Slenk, -Oosterkwelder, -Om de West, -Paardenwei.

Notably, although the same emission inventory is applied for both years, the emission rates vary greatly due to differences in the meteorological conditions under which the rates are calculated (Skjøth et al., 2011).

To evaluate the performance of the LOTOS-EUROS model in simulating ammonia concentrations, a statistical comparison was conducted against MAN network measurements. The assessment included Pearson's correlation coefficient (r), root-mean-square error (RMSE), mean absolute error (MAE), mean absolute percentage error (MAPE), normalized mean bias (NMB), and the slope of the regression between simulated and observed values. Table 2 presents the statistical results for six monitoring sites on Schiermonnikoog in both 2019 and 2022.

In 2019, the correlation coefficient (r) ranged from 0.34 to 0.55 across sites, reflecting a moderate agreement between model predictions and observations. By 2022, correlation values reached up to 0.87 at certain locations. Notable discrepancies occur, particularly at sites located near strong emission sources, such as Meteo Groenglo. This site consistently exhibited the highest RMSE and NMB values in both years, indicating that the spatial resolution of the model may not adequately capture local-scale variability in emissions and meteorological conditions. In 2019, the underestimation of peak ammonia concentrations is reflected in the strongly negative NMB values and correlation. This bias was reduced in 2022, especially at sites farther from major emission sources. Furthermore, the slope of the regression line between simulated and observed concentrations increased in 2022. The difference between two years may be due to the meteorological changes or an unaccounted-for occurrence of ammonia emission.

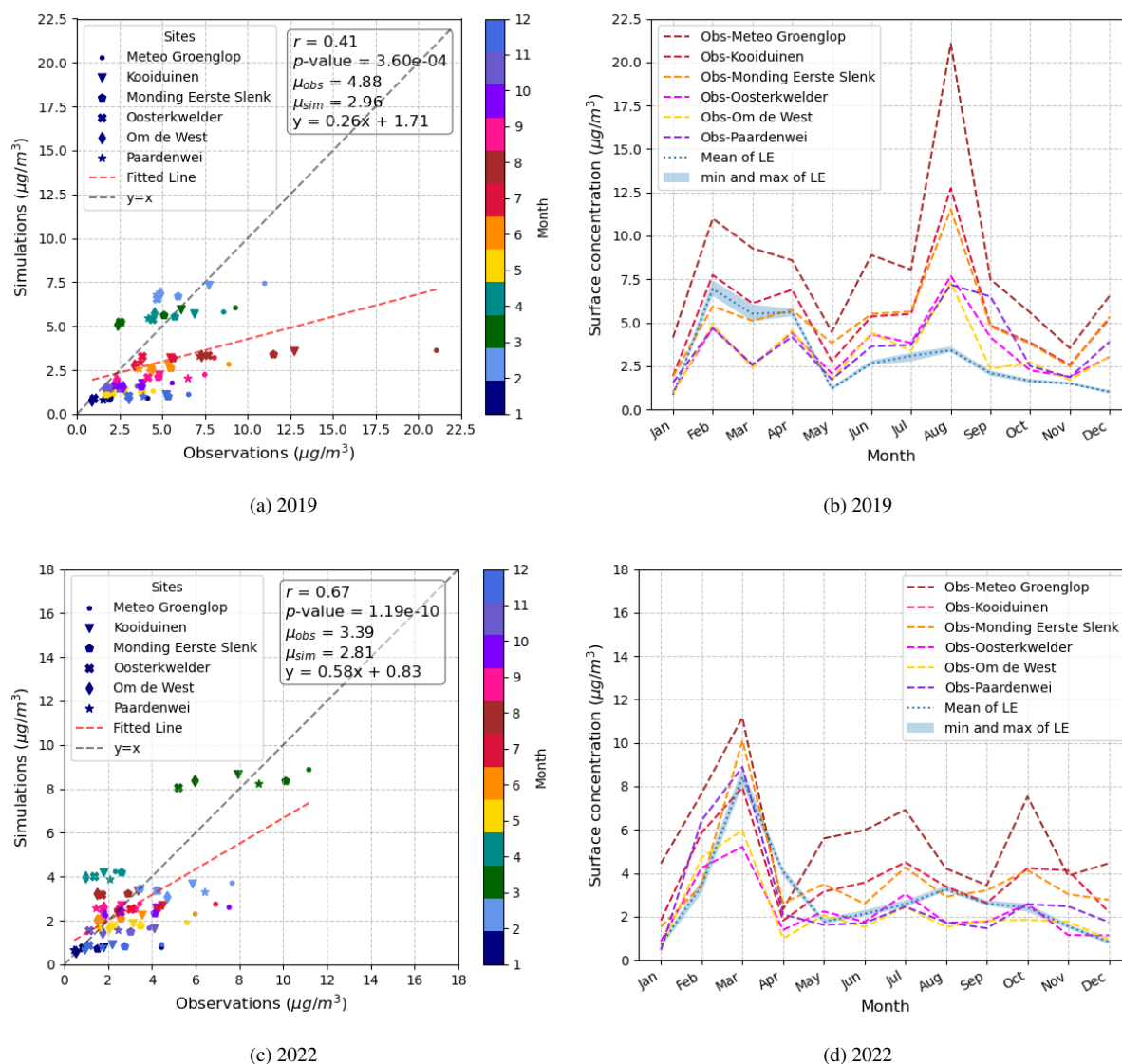
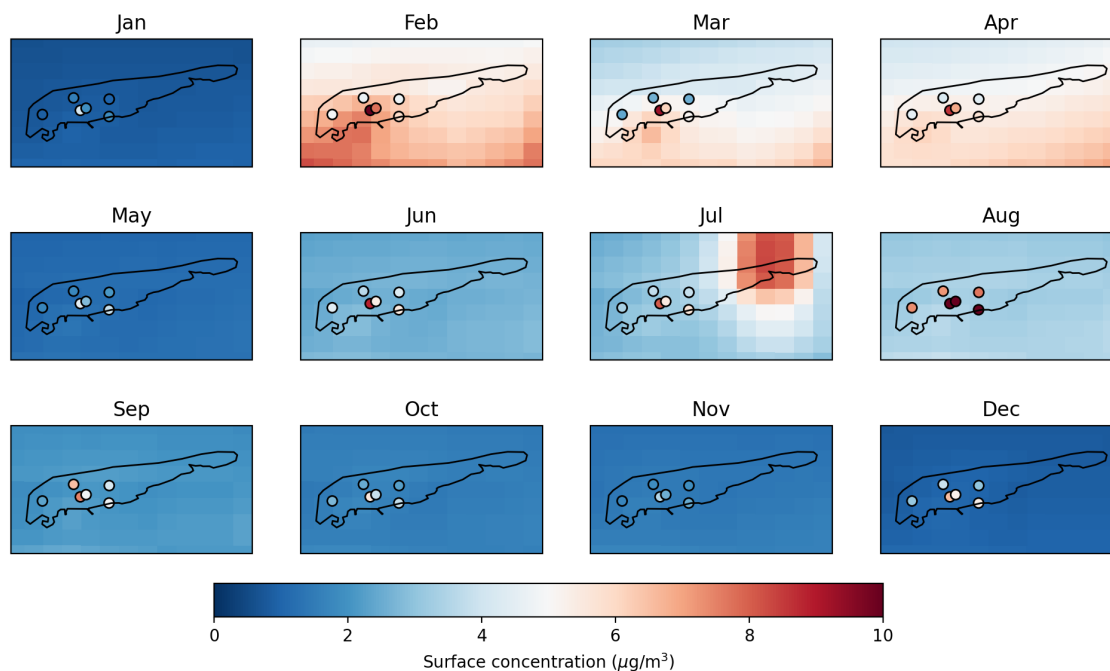
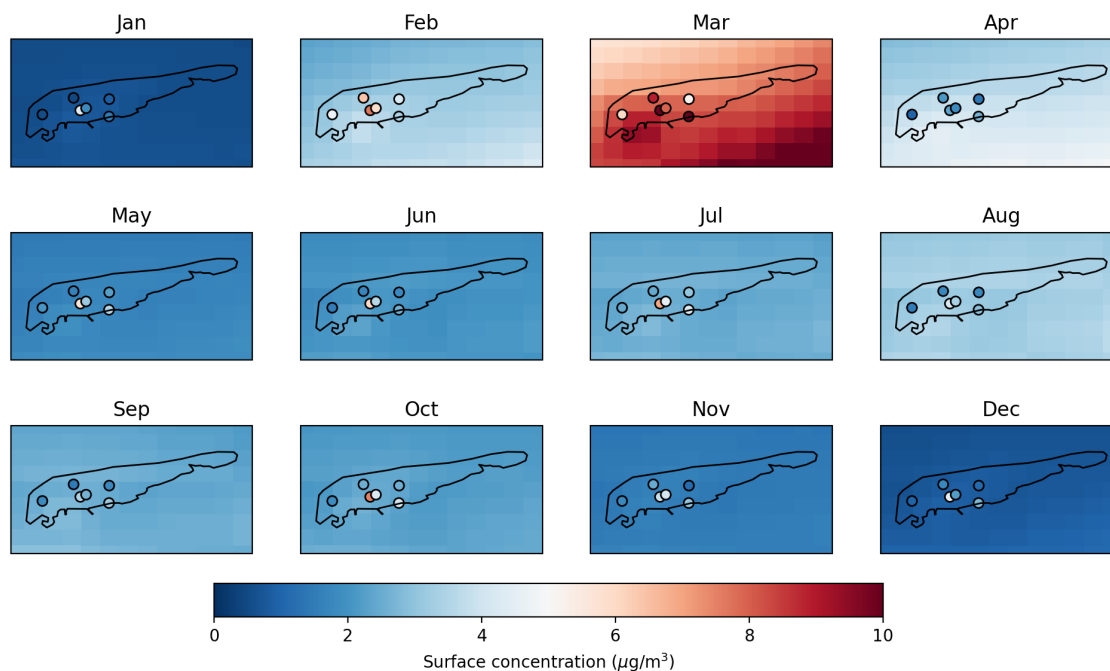


Figure 4. The comparison of model and observation in monthly average on Schiermonnikoog.

Digging deeper, a comparison of modeled and observed ammonia concentrations is illustrated in Fig. 4. The scatter plots for 2019 (a) and 2022 (c) demonstrate that the model underestimated ammonia concentrations in high-emission months, particularly during summer. The regression lines fitted in the scatter plots confirm that the model performed better in 2022, aligning more closely with the 1:1 reference line. Monthly variations, as shown in the right-hand panels of Fig. 4(b) and (d), reveal that while the model captured the seasonal cycle of ammonia, it underestimated peak concentrations in summer and overestimated lower values in winter. These discrepancies suggest that an improvement in the temporal variations of the emissions is needed.



(a) 2019



(b) 2022

Figure 5. Maps of surface concentrations on Schiermonnikoog for 2019 (a) and 2022 (b).



In addition to statistical performance, spatial variability was examined as shown in Fig. 5. These maps illustrate the modeled ammonia concentrations across Schiermonnikoog for each month in 2019 and 2022, overlaid with MAN measurement
295 locations. The maps highlight a clear seasonal pattern, with elevated concentrations during spring and summer, consistent with agricultural activity and temperature-driven volatilization. The spatial distribution of ammonia shows gradients, particularly in regions downwind of emission sources. The comparison between modeled fields and observations suggests that while LOTOS-EUROS effectively captures broad seasonal trends, local-scale concentration hotspots remain difficult to resolve due to the inadequate spatial resolution of the simulation, especially near emission sources. As shown in Fig. S2, the spatial resolution of the emission inventory does not always align with the true distribution of local ammonia sources, particularly on
300 Schiermonnikoog, where emissions from agricultural activities are concentrated in a small area. Since the model operates on a coarser grid, emissions may be spread over a larger area or displaced from their actual sources, leading to an underestimation of concentration peaks at specific measurement sites. This is particularly evident at the Schiermonnikoog-Meteo Groenglop site, where observed ammonia levels are consistently higher than simulated values, likely due to its proximity to actual emission
305 sources that are not well-represented in the model.

Moreover, external factors may have contributed to the high discrepancies observed in specific months. For instance, in July 2019 (see Fig. 5a), dune fires occurred in the eastern dunes of Schiermonnikoog, leading to increased ammonia concentration. Although fires were accounted for in the model with Global Fire Assimilation System (GFAS) emission inventory (Kaiser et al., 2012), the simulation remains highly uncertain due to the complexity of fire-induced emissions. Additionally, the timing
310 of passive sampler collection in the MAN network may not always align strictly with the first and last days of the month, potentially introducing inconsistencies between measured and simulated values.

3.1.2 Source Apportionment of Ammonia Concentrations

To better understand the origins of ammonia concentrations on Schiermonnikoog, a detailed source apportionment analysis was conducted, using the LOTOS-EUROS model's labeling function. Figure 6 presents both the monthly variations in source
315 contributions (a,b) and the annual summaries (c,d) for 2019 and 2022. These results reveal that local emissions play a relatively minor role in the island's ammonia budget, with the majority of the observed concentrations originating from external sources.

In 2019, emissions from Schiermonnikoog itself accounted for 6% of annual ammonia concentrations, with 4% from agriculture and 2% from other activities. Transported emissions from Friesland, Groningen, and Germany collectively contributed over 50%, with Friesland consistently showing the highest contribution, followed by Germany and Groningen. The distribution
320 was similar in 2022, though with a slight reduction in emissions from natural sources, primarily due to a lower frequency of wildfire events compared to 2019. A particularly significant contributor across both years was the "Other" label, representing over 10% of total concentrations. This category includes ammonia re-emission during deposition, as accounted for by the bi-directional flux scheme in LOTOS-EUROS. However, despite its modeled inclusion, this process remains highly uncertain.

The seasonal variability of ammonia sources is also evident. The monthly breakdown (Fig. 6a and b) indicates two distinct
325 peaks in ammonia concentration: one in spring, coinciding with manure application, and another in summer, driven by temperature-enhanced volatilization. These patterns are consistent with expected seasonal fluctuations in agricultural emis-

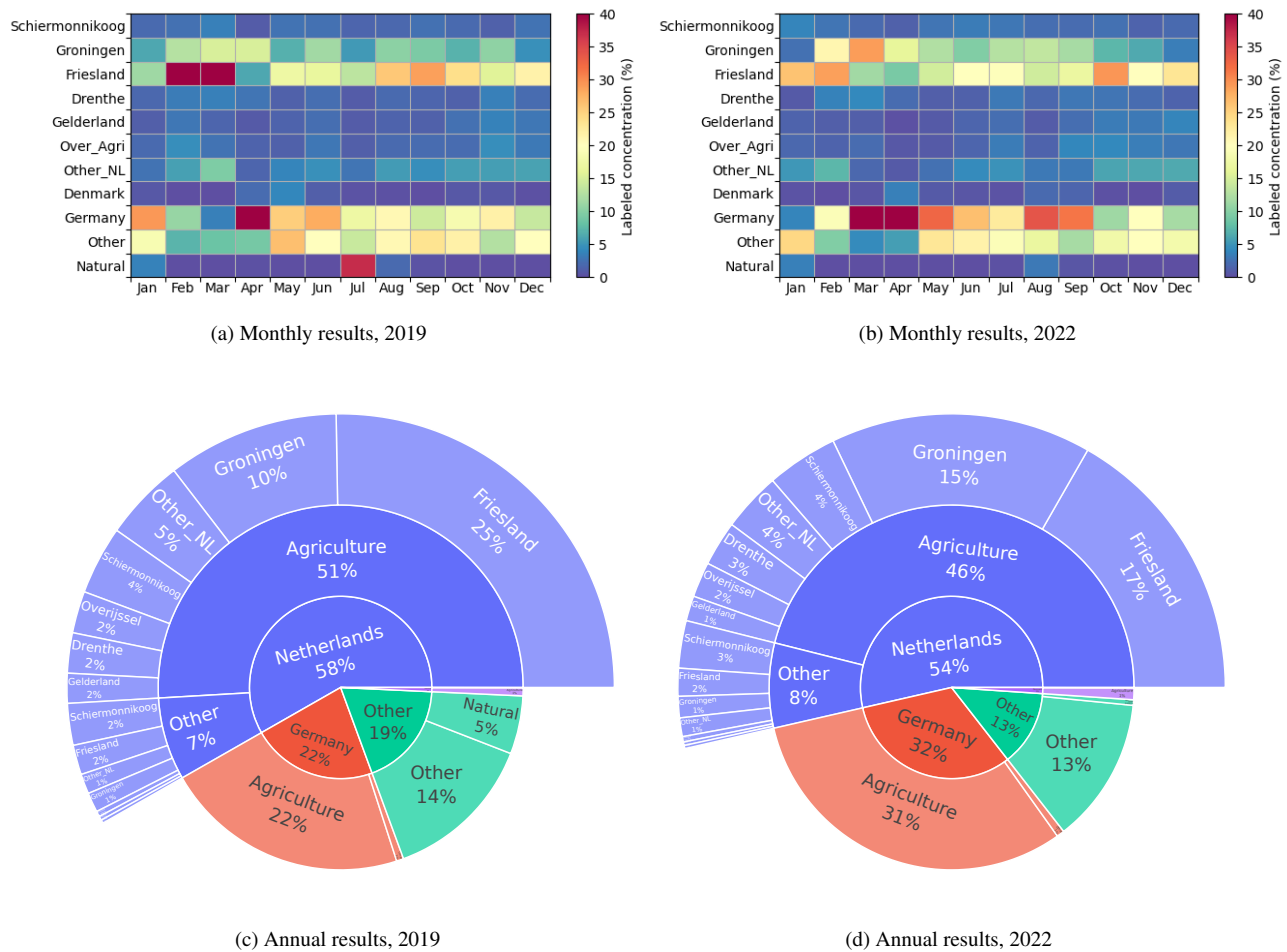
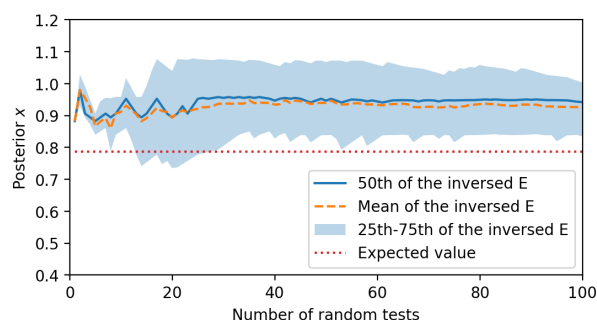


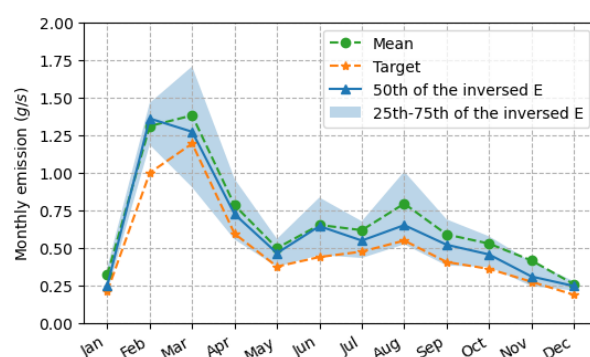
Figure 6. The relative contributions of different sources to the surface concentration of Schiermonnikoog in 2019 (a,c) and 2022 (b,d)

sions. Notably, the contribution from Germany is particularly high in spring, a period when Schiermonnikoog is frequently downwind of eastern air masses. Strong easterly winds during this time (see Fig. S3 for the seasonal wind field) enhance long-range transport of ammonia to the island, amplifying the external signal. Besides, 2019 exhibited an exceptionally high contribution from natural sources, especially in July, attributed to wildfires in the eastern dunes of Schiermonnikoog.

The annual source breakdown (Fig. 6c and d) further confirms the dominance of domestic agricultural emissions, which accounted for 51% of total ammonia emissions in 2019 and 46% in 2022. Other anthropogenic sources, including industrial, transportation, and residential emissions, remained relatively stable across both years, contributing approximately 7–8% to the total ammonia levels.



(a) Yearly emission inversion



(b) Monthly emission inversion

Figure 7. Results of yearly (a) and monthly (b) inversion of emission on Schiermonnikoog using synthetic observations. The results shown here correspond to the 2022 inversion.

3.2 Inversion results with synthetic observations

With synthetically constructed observations and an optimized observational error, the inversion system successfully reproduces known emission perturbations. As shown in Fig. 7(a), the yearly inversion for 2022 with synthetic observations demonstrates a decrease. Although the posterior state vectors fall between the prior value and the prescribed synthetic “true” emissions, the slight overestimation observed in the posterior emissions confirms that the inversion system does not overfit.

The results in Fig. 7(b) present the monthly inversion performance for 2022. While the ensemble average appears reasonable, individual monthly inversions suffer from high uncertainty, primarily due to the limitations of the simulated MAN error. The passive sampling approach and associated errors are not well-suited for resolving finer temporal variability. As such, these findings highlight the need for higher-quality, high-resolution measurements to improve inversion accuracy at monthly scales and also reduce the impact of seasonal mismatches between model and measurements.



Table 3. Inversion results of 2019 and 2022 with optimized observational error. The prior error covariances for the emission is 0.48 and for external influences are 0.16. Notably, the MAN data of July and August 2019 are eliminated because of the dune fires.

	Posterior $\bar{x}$	DFS	$\hat{S}$
2019, MAN	E	1.35	0.74
	Groningen	1.02	0.89
	Friesland	1.01	0.98
	Germany	0.92	0.9
	Other	1.08	0.98
2022, MAN	E	0.73	0.55
	Groningen	1.05	0.96
	Friesland	0.91	0.99
	Germany	0.61	0.82
	Other	0.72	0.97

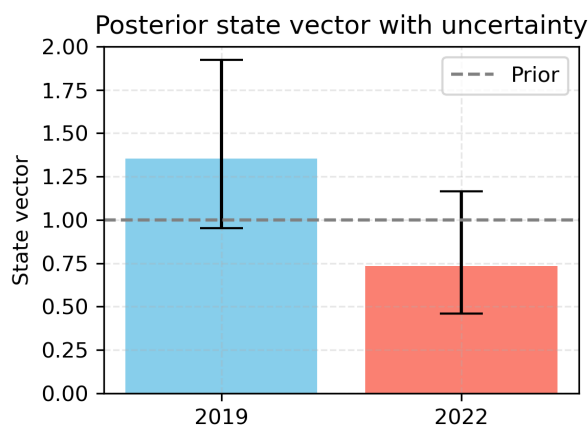
3.3 Inversion results of MAN

Table 3 presents the results of the ammonia emission inversion using MAN measurements for 2019 and 2022. The results show a notable reduction in total emission multipliers for Schiermonnikoog, from 1.35 (0.95-1.92) in 2019 to 0.73 (0.46-1.17) in 2022. Both inversions utilize the same 2019 emission inventory as prior. Although variations in meteorological conditions influence the emission rates between the two years, the inversion results consistently indicate a decrease in ammonia emissions.

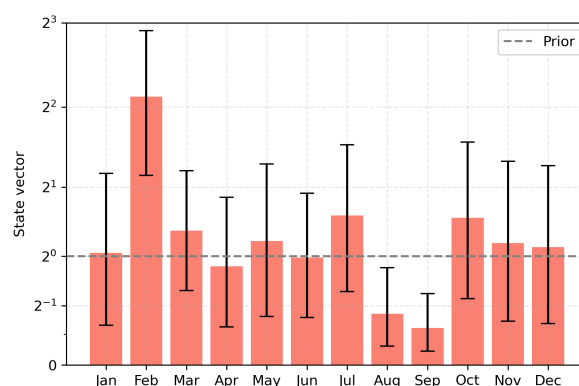
Without accounting for the potential systematic overestimation in 2022, the inversion results suggest a 51% reduction in ammonia emissions on Schiermonnikoog between 2019 and 2022. However, this figure may be an overestimate. To reflect the uncertainty in the posterior estimates, the credible intervals are derived using the transformation $\hat{x}_{\text{post}} \cdot \left[\exp\left(-\sqrt{\hat{s}}\right), \exp\left(\sqrt{\hat{s}}\right) \right]$, as shown in Fig. 8.

Despite the wide credible intervals—attributable to the sparse measurement network and high observational uncertainty—the inversion successfully captures both the direction and approximate magnitude of the emission reduction. These results indicate that the reduction in ammonia emissions on Schiermonnikoog is both detectable and significant.

We also attempted to use the MAN measurements for monthly emission inversion (see Fig. 8b). However, due to the high uncertainty in the data, which leads to large observational errors, the resulting inversions exhibited unrealistically high emission values in spring. Additionally, the degrees of freedom for signal (DFS) remained low, indicating that the system could not extract sufficient information from the observations to reliably constrain the emissions. In Sect. 3.4, we explore potential strategies to improve the performance of monthly emission inversions, including enhancements in measurement quality and network design.



(a) Yearly emission inversion



(b) Monthly emission inversion

Figure 8. Results of MAN-derived yearly ammonia emission (a) and monthly ammonia emission (b) with credible intervals, derived from the posterior error covariance matrix $\hat{\mathbf{S}}$. The results shown in Figure (b) correspond to the 2022 inversion.

3.4 Optimizing the measurement network for evaluation of emissions

Due to the limitations of the current MAN network on Schiermonnikoog, the inversion is only sufficient for quantifying annual emissions, albeit suboptimally. Furthermore, these measurements exhibit high uncertainty, particularly in low-emission regions, making localized emission changes difficult to detect due to measurement noise. To address these challenges, we propose potential enhancements to the existing monitoring network to improve emission tracking on Schiermonnikoog.

We apply a Monte Carlo approach to simulate Degrees of Freedom for Signal (DFS) under varying measurement network configurations. Specifically, we test the effects of increasing the number of monitoring sites across Schiermonnikoog and reducing observational errors to represent higher-quality measurements. Figure 9(a) shows the DFS results averaged over 20 randomized site configurations. The contour plot highlights how DFS evolves as a function of both site density and observa-

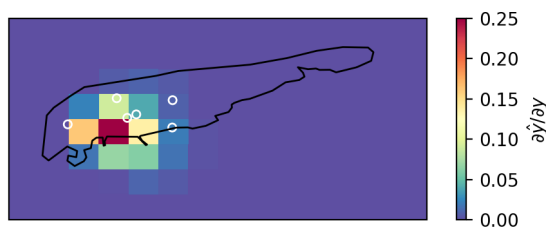
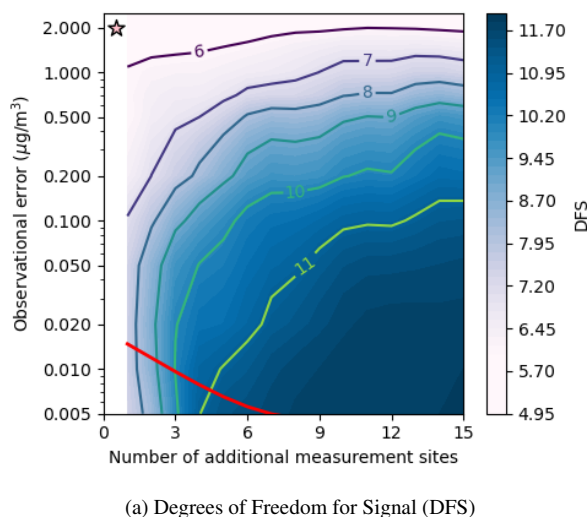


Figure 9. Figure (a) illustrates the DFS as a function of additional measurement sites and observational errors. The ridge line divides the plot into two regions: error-limited (above the ridge line) and site-limited (below the ridge line). Worth mentioning here, the measurement sites are added randomly; thus, Figure (a) shows the averaged results. The star in Fig. (a) represents the current monitoring network. Figure (b) highlights the spatial distribution of observational usefulness. Assuming high-quality measurements are available across the entire island, we calculate the contribution of each observation to the inversion result; see Eq. (12) for details. This diagnostic provides insight into which locations offer the greatest information gain, thus informing the strategic placement of future monitoring sites.

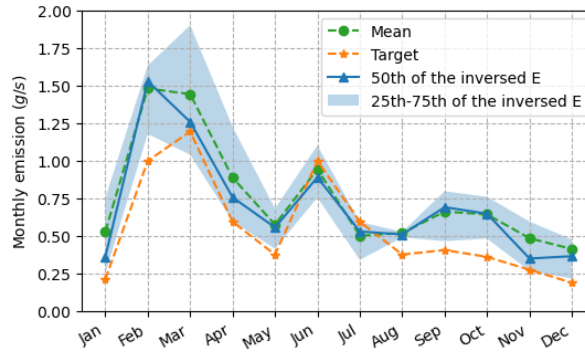
tional precision. The ridge line delineates the transition between two regimes: an error-limited regime (above the ridge), where measurement uncertainty dominates, and a site-limited regime (below the ridge), where sparse spatial coverage is the limiting factor. The results indicate that both increasing the number of sites and reducing observational errors can enhance DFS, but
375 their relative effectiveness depends on the regime:

- in site-limited conditions, adding more sites leads to substantial DFS improvement;
- in error-limited conditions, further reducing observational uncertainty becomes more impactful.

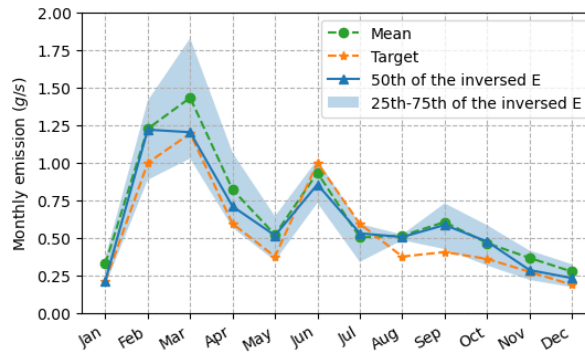
The star in Fig. 9(a) marks the current MAN network configuration, which is constrained by relatively high observational errors, resulting in low DFS. While increasing the number of high-quality measurements would greatly improve the system,



380 practical constraints such as cost and logistics make island-wide deployment of high-quality sensors difficult. This highlights the need for strategic placement of enhanced measurement sites to optimize inversion performance.



(a) Current MAN network



(b) Adding one LML-like measurement

Figure 10. The monthly inversion in 2022 with synthetic measurements and a synthetic summer peak. The observational error for the synthetic LML-like measurement is set at 3.5%. Figure (a) shows the posterior emission with the current MAN network. Figure (b) shows the posterior emission after adding one LML-measurement.

Figure 9(b) displays the spatial distribution of observational usefulness across Schiermonnikoog, averaged over 100 random test realizations. Assuming that high-quality measurements (with an observational error of $0.1 \mu\text{g}/\text{m}^3$) are available uniformly across the island, we quantify the relative contribution of each observation to the inversion outcome using the following
385 formulation (Rodgers, 2000):

$$\frac{\partial \hat{\mathbf{y}}}{\partial \mathbf{y}} = \frac{\partial \mathbf{K}_N \hat{\mathbf{x}}}{\partial \mathbf{y}} = \mathbf{K}_N \mathbf{G} \quad (12)$$

where $\mathbf{G} = \hat{\mathbf{S}} \mathbf{K}_N^T \mathbf{S}_0^{-1}$ is the gain matrix, $\hat{\mathbf{S}}$ is the posterior error covariance, $\mathbf{K}_N$ is the Jacobian matrix at the final iteration, and $\mathbf{S}_0$ is the observational error covariance matrix. To quantify the influence of each observation only on the inversion of Schiermonnikoog's emission, we take the first column and row of $\mathbf{K}_N$ and $\mathbf{G}$, respectively, corresponding to the local agricultural



emission component. The diagonal elements of this product reflect the influence of each observation on the inversion model outcome. These values are reprojected spatially in Fig. 9(b). This diagnostic provides an intuitive interpretation of where observations are most impactful for the inversion. Regions with higher values indicate greater potential for improving the accuracy of emission estimates, offering a useful basis for strategic sensor placement in future monitoring network designs.

We then propose adding an LML-like measurement site on Schiermonnikoog. Although Fig. 9(b) indicates that the most informative location would be directly at the island's main emission source, near the Schiermonnikoog-Meteo Groengloep site, this location is not ideal in practice. As discussed in Sect. 3.1.1, this site shows the least agreement between model and observation, likely due to unresolved spatial heterogeneity between reality and model. Moreover, measurement close to the source can lead to a large bias in regional misrepresentation of ammonia concentration (Schulte et al., 2022). The next model-suggested site is Schiermonnikoog-Om de West, at the western edge of the island. However, this site consistently records the lowest ammonia concentrations and is strongly influenced by sea winds, making it less suitable for detecting local agricultural emissions. Another candidate is the Schiermonnikoog-Paardenwei site, located in the north. While the model indicates it as a potentially useful location, this site is surrounded by dense vegetation, which in reality limits its sensitivity to nearby agricultural sources (see Sect. 3.1.1). Given these limitations, we propose placing the LML-like instrument near the Schiermonnikoog-Kooiduinen site. This location is downwind of the main emission source and shows good agreement with model predictions. Among the existing monitoring sites, Schiermonnikoog-Kooiduinen represents the best balance between observational consistency and proximity to emissions, making it the most suitable candidate for improved inversion performance.

Figure 10 illustrates the effect of incorporating a single high-quality, LML-like observation on monthly ammonia emission inversion for 2022. The synthetic observational error for the LML-like site is set at 3.5%, based on values reported by Dammers (2017); Blank (2001). Compared to the current MAN-only network, the addition of one strategically placed LML-like measurement significantly reduces uncertainty in most months. Posterior estimates not only align more closely with target values but also exhibit a narrower percentile spread across most months, indicating improved stability. While the inversion slightly overestimates emissions in March and April, the LML-enhanced setup still outperforms the MAN-only inversions.

Overall, this result demonstrates that even a single, well-placed, high-precision observation can substantially improve inversion performance, enhancing the system's ability to track temporal variability and increase DFS. With high-frequency, low-error measurements, it becomes feasible to detect near-real-time emission changes.

In addition, observational error can be reduced by averaging multiple measurements at the same site, effectively decreasing the random component of the total error, as suggested by the central limit theorem. According to Noordijk et al. (2020); Lolkema et al. (2015), the total monthly error in the MAN network comprises three components: random uncertainty, uncertainty from the calibration method, and systematic uncertainty from the calibration standard. The first two are random and can be substantially reduced by repeated sampling. By increasing the number of measurements at a given site and averaging them into a single "superobservation", the total error can be significantly lowered, approaching the quality of high-precision instruments. While enhancing a single MAN site alone does not achieve the same performance as adding a single LML-like site, substituting all six MAN sites on Schiermonnikoog with corresponding superobservations yields substantial improvements. In



fact, this approach performs even better than the LML-like configuration in March and April. More details are provided in the
425 Supplement.

4 Conclusions

4.1 Summary

In this study, we performed a local-scale Bayesian inversion of ammonia emissions using the LOTOS-EUROS chemical trans-
port model as the forward model, with MAN (Measuring Ammonia in Nature) observations serving as constraints and CAMS-
430 REG, GrETa, and ER inventories as prior estimates. To evaluate the influence of observational uncertainty, we performed
sensitivity analyses by integrating residual errors with reported MAN network uncertainties from Lolkema et al. (2015); No-
ordijk et al. (2020). By optimizing the Chi-Square statistics, we derived observational error covariance matrices for both annual
and monthly inversions, improving the robustness of the inversion framework.

Between 2019 and 2022, GVE (grazing livestock units) on Schiermonnikoog decreased from 639 to 541, with a particularly
435 notable reduction in dairy cattle, corresponding to an estimated 23% reduction in ammonia emissions based on activity data.
Our inversion successfully captured this trend. Using the current MAN network, we were able to invert annual emissions,
with the inversion indicating a 51% reduction between 2019 and 2022. However, this value may be overestimated. Using
the posterior error covariance, we derived a credible interval, describing the uncertainty of the inversion. In contrast, monthly
inversions remain challenging with the current observational network. High measurement uncertainty hinders the system's
440 ability to resolve short-term emission dynamics effectively.

To explore potential improvements, we conducted sensitivity analyses of the Degrees of Freedom for Signal (DFS) and
posterior error. Results showed that the existing network is limited by relatively high observational errors. Increasing the
number of high-quality measurements substantially improves the inversion performance. In particular, adding a single, high-
quality, LML-like observation at a strategically chosen downwind site on the island significantly enhanced the system's ability
445 to resolve temporal variability and increased DFS.

4.2 Discussion

Although the model captures overall seasonal trends in both years, significant discrepancies remain in specific months, partic-
ularly during episodic events such as the dune fires in July and August 2019. Such events substantially affect local ammonia
levels and underscore the need to include event-driven emissions in the modeling framework.

450 Furthermore, possible timing discrepancies in passive sampler collection, where measurements may not align precisely with
calendar months, introduce additional uncertainty when comparing observations to model outputs. These issues highlight the
need for refined emission timing, better spatial resolution, and improved observation methodologies to enhance simulation-
observation agreement.



Source apportionment analysis revealed that the "Other" category, primarily representing ammonia re-emission through bi-directional flux processes, also plays a key role. This includes volatilization from both soil and sea surfaces. While LOTOS-EUROS can capture sea-air exchange processes, these remain highly uncertain due to complex atmospheric interactions and limited validation data over marine environments.

4.3 Outlook

The findings from model evaluation and source attribution offer valuable insights for future ammonia inversion efforts. Persistent underestimation of summer ammonia concentrations suggests the need for improved temporal representation of agricultural practices, particularly manure spreading. Incorporating regional transport is also critical, as the island's ammonia concentrations are heavily influenced by upwind sources.

Refining prior emission inventories with higher spatial and temporal resolution is essential for improving inversion outcomes. Deploying high-resolution, continuous measurements, such as LML-like instruments at strategically chosen sites, would provide much-needed constraints and enable more accurate inversion at finer timescales.

In conclusion, while LOTOS-EUROS demonstrates strong capability in capturing broad seasonal ammonia trends, it struggles with peak events and local-scale gradients. The source apportionment confirms that Schiermonnikoog's ammonia levels are mainly driven by regional transport, reinforcing the need for regional-scale mitigation strategies in addition to local emission reductions. This highlights the importance of coupling localized monitoring efforts with national and transboundary air quality policies.

Future work will expand the inversion domain to include broader regions and higher-intensity emission sources. Furthermore, since the current framework assumes a linear relationship between emissions and concentrations, we will explore non-linear Jacobian formulations, which may better capture the dynamics over diverse and complex emission landscapes. For upcoming national-scale inversion studies, we plan to first characterize emission patterns before applying inversion, to enhance both accuracy and computational efficiency.

Code and data availability. The data of the farms and livestock can be found on <https://mijnkringloopwijzer.nl/>. The LOTOS-EUROS is an open-sourced model, and the newest version v2.3 can be found on the official TNO website (<https://airqualitymodeling.tno.nl/lotos-euros/open-source-version/>). The Measuring Ammonia in Nature (MAN) can be accessed through MAN website (<https://man.rivm.nl/>). LML measurement <https://www.luchtmeetnet.nl/>

Author contributions. SL: methodology, software, formal analysis, writing (original draft), visualization. ED: conceptualization, software, writing (review and editing), supervision, project administration, funding acquisition. JWE: conceptualization, validation, writing (review and editing), supervision, project administration, funding acquisition. AJS: methodology, writing (review and editing).



Competing interests. The authors declare that no competing interests are present.

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