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Atmospheric Ammonia in Sweden: Regional Modeling and Assessment of National Emission Reduction Benefits

ORIGINAL RESEARCH PAPER

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ABSTRACT

Ammonia (NH_3) is a central component of atmospheric reactive nitrogen, and a precursor gas that forms airborne particulate matter including nitrate and sulfate aerosol. Reducing ammonia emissions, primarily from agriculture, is a major challenge for air pollution mitigation and environmental protection. This work assesses the effects of ammonia emission reductions in Sweden, which is one of the EU Member States that are currently not meeting their emission reduction commitments. We apply the regional chemical transport model MATCH with improved NH_3 treatment, and benchmark the updated model setup against surface observations of atmospheric NH_3 and other nitrogen species for a large-scale model domain over Europe. We perform high-resolution regional simulations over Sweden, and compare the effects of reducing the emissions of (1) agricultural NH_3, and (2) nitrogen oxides (NO_x) from road transport, which is the second major nitrogen source in Sweden. The results show the potential of national NH_3 emission reductions to mitigate NH_3 levels, fine particulate matter ($PM_{2.5}$) and nitrogen deposition within Sweden. While cutting NO_x emissions is important for air pollution abatement through NO_x mitigation, in Sweden the reductions have only minor or negligible effects on $PM_{2.5}$ and nitrogen deposition. This demonstrates the benefits of NH_3 reduction for both air quality and environment, and the impact of national abatement actions.

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1 INTRODUCTION

Atmospheric ammonia (NH_3) has significant environmental impacts through various effects on air quality, ecosystems and climate (Behera *et al*., 2013; Nair and Yu, 2020). Ammonia contributes to the formation of airborne particulate matter (PM), which has significant health and climate impacts. Deposition of nitrogen species also has detrimental effects on water, soil and plants. The dominating ammonia emission source is agricultural activities, including fertilizer use and animal husbandry (Anderson, Strader and Davidson, 2003; Beaudor *et al*., 2023; Wyer *et al*., 2022). While anthropogenic emissions of other nitrogen species, primarily nitrogen oxides (NO_x), are generally decreasing in Europe, reducing ammonia emissions has proved difficult (Van Damme *et al*., 2021). In the European Union, ammonia remains the most significant challenge for the national emission reduction commitments, with several Member States yet not reaching their targets (EEA, 2024a).

Ammonia is a central component of secondary aerosol formation: gaseous NH_3 forms particulate ammonium nitrate and ammonium sulfate, which make a substantial contribution to total PM (Wyer *et al*., 2022). PM, characterized by the $PM_{2.5}$ and PM_{10} metrics corresponding to the total mass of particles smaller than 2.5 and 10 µm, respectively, is among key air pollutants, causing respiratory and cardiovascular diseases and premature mortality (Apte *et al*., 2018; Lelieveld *et al*., 2015). The majority of the urban population in Europe continues to be exposed to unsafe PM concentrations, with exceedances of EU and WHO limit values also in the Nordic countries that are among regions with lowest pollution levels (EEA, 2024c).

Atmospheric particles are also climate forcers with a significant role in the global radiation budget, with nitrate aerosols causing a net cooling effect (Xu and Penner, 2012). As sulfate aerosols are reduced due to declining sulfur emissions, nitrate aerosols are expected to become more important (Bauer *et al*., 2007; Pye *et al*., 2009). Consequently, agricultural ammonia emissions are a central question for the future climate forcing attributed to anthropogenic aerosols (Hauglustaine, Balkanski and Schulz, 2014). In addition to secondary aerosol formation onto existing particles, ammonia contributes to nucleation of new particles from gases through the sulfuric acid–ammonia nucleation pathway (Dunne *et al*., 2016; Smith *et al*., 2021). New-particle formation has substantial effects on particle numbers, which are critical for cloud formation and indirect aerosol effects through aerosol–cloud–climate interactions (Kerminen *et al*., 2012). The nucleation pathway can often be limited by the availability of ammonia, thus making ammonia relevant for both particle mass and number.

Deposition of atmospheric nitrogen species to soil and water is a major environmental threat, causing eutrophication, nutrient imbalances and loss of biodiversity (Dise *et al*., 2011; Guthrie *et al*., 2018). Despite the declining trend in nitrogen deposition due to emission reductions, current deposition levels still exceed those of the preindustrial era (Engardt *et al*., 2017; Kanakidou *et al*., 2016). Northern Europe covers both ecosystems under significant stress from eutrophication, including the brackish Baltic Sea, as well as relatively pristine areas with comparatively low historical nitrogen deposition, such as natural areas in Lapland and the Scandinavian Mountain Range (Andersen *et al*., 2017; Manninen *et al*., 2024; Nordin *et al*., 2005). Continued decline in nitrogen deposition is a prerequisite for ecosystem recovery and preventing disturbances in potentially sensitive ecosystems.

In Sweden, the main atmospheric anthropogenic nitrogen sources are road transport, industry and agriculture, including emissions of NO_x, NH_3 and to a minor extent N_2O (Moldan *et al*., 2022; SMHI, 2025c). The transport sector is the largest emitter of NO_x, however with a continuous declining trend in the emissions. NH_3 emissions from the agricultural sector, on the other hand, have not decreased significantly during the last decades (SMHI, 2025c). As for now, Sweden is not meeting the emission reduction commitments for 2030 for NH_3 or NO_x, and further reductions are needed for both species (EEA, 2024a). In addition to Swedish emissions, long-range atmospheric transport makes a major contribution to the Swedish nitrogen budget (SMHI, 2025b). For example, most nitrogen deposition in Sweden has been attributed to emissions elsewhere (Moldan *et al*., 2022). In the vicinity of Sweden, major emitters of nitrogen and NH_3 include e.g. the Netherlands, Germany and also Denmark with a relatively large agricultural sector (Dalgaard *et al*., 2014; EEA, 2024b). Nitrogen and other main air pollutants in Sweden are monitored in the national monitoring program for air, including measurements and regional model assessments (Alpfjord Wylde, Leung and Andersson, 2023; SMHI, 2025b; Tørseth, 2016). The main monitored pollutants include ozone (O_3), nitrogen dioxide (NO_2), $PM_{2.5}$ and PM_{10}, and deposition of nitrogen and sulfur. While regional modeling includes NH_3 emissions, ammonia is not assessed or evaluated separately.

This work presents regional model assessments of ammonia and related nitrogen species in Europe, with focus on Sweden. The purpose is to:

1. Benchmark the model performance for the relevant species in order to establish a model setup for assessments of NH_3 effects, also including possibility to integrate NH_3 to other national air quality modeling.
2. Assess the possibilities to mitigate ammonia and nitrate species in Sweden by reducing national agricultural emissions.

For this, we apply the chemical transport model MATCH (Multi-scale Atmospheric Transport and Chemistry model; Robertson, Langner and Engardt, 1999) over Europe, and over Sweden at a higher spatial resolution. Simulations over the European domain are used for model evaluation against measurement data from various European field stations, and the nested Swedish domain is applied to assess the national emission reduction effects. We use anthropogenic emission data from the national emission database, and introduce model updates for the treatment of surface–atmosphere NH_3 fluxes.

Emission reduction tests are performed by scaling down Swedish emissions by 20%. This is an ambitious but realistic reduction target considering the Swedish emission reduction commitments within the European Green Deal framework (EEA, 2024a; European Commission, 2019). We address specifically the following questions:

- How much can Sweden affect the national levels of NH_3, fine particulate matter $PM_{2.5}$ and deposited nitrogen by decreasing its own NH_3 emissions?
- How efficient is reducing agricultural emissions of NH_3 as a measure to abate atmospheric nitrogen, as compared to reducing NO_x from road transport by the same relative amount?

The reference test case with NO_x is motivated by NH_3 and NO_x emissions being the major nitrogen sources, with each of them involving a single largest emission sector: agriculture for NH_3, and road transports for NO_x (excluding international shipping). The total annual emissions from these two sectors are currently of the same order in Sweden (SMHI, 2025c). Both species contribute to the nitrogen budget and to the formation of inorganic $PM_{2.5}$ together with sulfur species.

2 METHODS

2.1 REGIONAL CHEMICAL TRANSPORT MODELING

The MATCH model (Robertson, Langner and Engardt, 1999) is an established regional-scale chemical transport model (CTM) with various gas and particle chemistry and physics schemes, including photochemistry, oxidation and other gas-phase reactions, secondary aerosol formation from gases, and dry and wet deposition of gases and aerosols. MATCH is used widely in research and operational work, including national air quality modeling in Sweden (e.g. Alpfjord Wylde, Leung and Andersson, 2023; Andersson, Langner and Bergström, 2007; CAMS, 2025; SMHI, 2025a).

Detailed descriptions of the model transport schemes and boundary layer parameterization can be found in previous works (Robertson, Langner and Engardt, 1999; Colette *et al.*, 2025). The gas-phase chemistry follows the EMEP MSC-W EmChem09 scheme (Simpson *et al.*, 2012) with modified isoprene chemistry (Carter, 1996), and reaction rates updated according to EmChem19 (Bergström *et al.*, 2022). Details regarding the gas-phase chemistry mechanism are given in Supplementary Information Section S3.4. Gas-to-particle conversions include the formation of secondary inorganic aerosol (SIA; consisting of ammonium sulfate and nitrate) and secondary organic aerosol (SOA). Particles are treated as bulk aerosol in two size classes, corresponding to fine and coarse PM with diameters smaller and larger than 2.5 µm, respectively. Ammonium nitrate (NH_4NO_3) equilibrium is calculated according to temperature- and relative humidity-dependent equilibrium coefficient (Mozurkewich, 1993), and coarse nitrate formation is described by transfer of gaseous nitric acid (HNO_3) to aerosol-phase NO_3 (Strand and Hov, 1994).

Primary organic aerosol emissions are treated as non-volatile in the model setup used in this study. Secondary organic aerosol formation from oxidation of volatile organic compounds (VOC) is treated using a volatility basis set (VBS) scheme (Bergström *et al.*, 2012). This includes anthropogenic SOA (ASOA) mainly from aromatic hydrocarbons, and biogenic SOA (BSOA) from isoprene, monoterpenes and sesquiterpenes. A simple scheme that does not include atmospheric aging of the SOA species is used (Langner *et al.*, 2020), which is a slightly modified version of the VBS-NPNA scheme (Bergström *et al.*, 2012).

MATCH has a number of different options for the treatment of wet and dry deposition of gases and particles. Here, relatively simple scavenging coefficients are used for gases and particles that are subject to wet deposition, as described in Supplementary Information Section S3.5. Dry deposition of gases by the resistance model, aerosol dry deposition, and stomatal resistance are described following the EMEP MSC-W schemes (Simpson *et al.*, 2012). In this work, the dry deposition description of NH_3 is updated to a bidirectional flux parameterization (Wichink Kruit *et al.*, 2017 and references therein) within the model development project Nordic Nature & Nitrogen to improve simulation of nitrogen in the Nordic countries (Frohn *et al.*, 2025). While the standard resistance model assumes only one-directional flux of NH_3 from the atmosphere to the surface, the bidirectional flux approach considers also the opposite flux to the atmosphere. The opposite flux is based on compensation points, corresponding to concentrations already present in the media, that determine the ability to take up more gas. Here, the main focus is uptake on vegetated surfaces, as described in Supplementary Information Section S3.1.

MATCH is applied for the European domain at a spatial resolution of 0.2° × 0.2° and for a nested domain over Sweden at a resolution of 5 km × 5 km (see e.g. Figure 1 and Figure 2 for the two domains). The European domain

is used for model evaluation by large sets of available quality-controlled observation data (Sections 2.2 and 2.3). The high-resolution domain is designed to be used in national air quality modeling for Sweden, and to study the national emission reduction effects. European-scale modeling is performed for selected full years, including 2017 and 2019, which are considered representative for recent trends. This excludes years corresponding to the COVID-19 pandemic and 2018, which was an unusually warm year in Sweden with exceptional heatwaves and forest fires. We focus on 2019, and perform additional model evaluation for 2017 to ensure the robustness of the model results. The Swedish emission reduction assessments are conducted for 2019.

The MATCH simulations use meteorological data from the European Centre for Medium-Range Weather Forecasts Integrated Forecast System (IFS) (ECMWF, 2025) with 50 hybrid vertical levels, reduced to 25 levels in the MATCH model corresponding to a vertical extent of approximately 6–8 km. In the present study, land-use data are based on the CCE CLC/SEI database (LRTAP, 2025); 16 different land-cover classes are included (as in Simpson *et al.*, 2012). For the European-scale simulations, anthropogenic emissions are obtained from the CAMS-REG emission inventory, using version V5.1c and with emission year 2018 applied for all years 2017–2019 (Denier van der Gon, Gauss and Granier, 2023, pp. 8–18; Kuenen *et al.*, 2022). Swedish anthropogenic emissions from the SMED database (SMED, 2025) are used for the Swedish-domain simulations. Emissions are distributed temporally according to the CAMS-REG-TEMPO-v3.1 profiles (Denier van der Gon, Gauss and Granier, 2023, pp. 32–47; Guevara *et al.*, 2021). The emission-sector-dependent split of total volatile organic compounds into the different VOC model species (Table S2) is based on data from CAMS (Kuenen *et al.*, 2022; following a similar methodology as Bergström *et al.*, 2022). Details and further information about the emission setup are given in Supplementary Information Section S3.2.

Biogenic emissions of isoprene and monoterpenes are calculated in the model (using the methodology of Simpson *et al.*, 2012); sesquiterpene emissions are also added based on plant chamber experiments with no observable biotic stress (5% of the monoterpene emissions; following Bergström *et al.*, 2014). Emission of sea salt particulate matter is modelled based on a parameterisation considering wind speed, water salinity and water temperature (Sofiev *et al.*, 2011). Natural aeolian dust emissions are modelled based on the DEAD model (Zender, Bian and Newman, 2003). Gaseous and particulate emissions from biomass fires are taken from the GFAS fire emission database (Kaiser *et al.*, 2012) (Supplementary Information Section S3.3). Emissions of sulfur species from seas and volcanoes (here Etna, Stromboli, Vulcano) are included in a simplified manner; oceanic dimethyl sulfide is treated as oxidized to SO_2 and sulfate before being introduced in the model.

Boundary data for the European-scale domain is obtained from the IFS global forecasts for the following species: NO, NO_2, HNO_3, PAN, O_3, SO_2, CO, CH_4, C_2H_6, C_2H_4, C_5H_8, HCHO, sulfate and desert dust similar to the operational CAMS system (Colette *et al.*, 2025), but in the present study the global boundary data are read with 6-hour intervals. For other model species, the model uses seasonal climatological boundary concentrations. Results of the European-scale simulations are used as boundary data for the nested Swedish-scale simulations for all advected model components; these boundary data are read with 3-hour intervals and interpolated in the model.

2.2 STUDIED SPECIES

To assess the impacts of emission reductions, we examine selected air quality and environmental indices that are expected to be notably affected by ammonia:

- Gas-phase NH_3
- $PM_{2.5}$
- Nitrogen deposition (Ndep)

For model evaluation, we also apply available observation data for related nitrogen species:

- Total ammonia and ammonium NH_x = NH_3(g) + NH_4^+(pm)
- Total nitric acid and particulate nitrate ΣNO_3 = HNO_3(g) + NO_3^-(pm)

Finally, in addition to the species directly related to ammonia, we benchmark the model setup against observation data for other common pollutants, namely:

- O_3
- NO_2
- PM_{10}

We study and evaluate the atmospheric concentration data at hourly or daily time resolution, depending on the component: O_3 and NO_2 components exhibit distinct diurnal cycles that are typically well described by models, and thus hourly resolution is applied. For other gas and aerosol components, daily mean values are used due to both availability of observation data (see Section 2.3) and diurnal variations being less important for model evaluation. For deposition, seasonal accumulated values are studied.

2.3 OBSERVATION DATA

Model results are evaluated against surface observations from atmospheric monitoring sites at hourly or daily temporal resolution, retrieved from the EBAS atmospheric database (EBAS, 2025). For NH_3, we use also additional observations collected from national databases: data collected by Aarhus University for Denmark (Ellermann

et al., 2021; stations DK-01, DK-09, DK-10), and the RIVM database for the Netherlands (RIVM, 2025; stations NL10131, NL10444, NL10538, NL10633, NL10738, NL10929).

For some species, here NH_3, $PM_{2.5}$ and PM_{10}, observation data are available at both hourly and daily resolutions, typically for different stations. For these species, hourly data are converted to daily means for consistency and for improved spatial data coverage. For calculating evaluation statistics for given periods (e.g. monthly or seasonal), at least 67% (2/3) observation data coverage is required.

For NH_x, data are combined by complementing available NH_x data with additional reported NH_3 and NH_4 data aggregated into the sum component NH_x. This allows including four Danish stations that only report NH_3 and NH_4 separately but are relevant for evaluating model performance in Northern Europe.

2.4 EMISSION REDUCTION ASSESSMENTS

The effects of Swedish national emission reductions are tested by scaling down the emissions from the given sectors by 20%. The scaling is applied temporally and spatially uniformly to (1) NH_3 emissions from the agricultural sector, and (2) NO_x (as NO_2) emissions from the road transport sector. These tests are performed using the high-resolution domain over Sweden and surrounding areas.

The magnitude of the test reductions is in line with the Swedish commitments for 2030, for which NH_3 and NO_x emissions need to be reduced by 7% and 37%, respectively (EEA, 2024a). For simplicity, the same reduction is applied for both components. For NH_3 the scaling factor is more ambitious, but such goals are relevant for assessing the potential effects and benefits of lower NH_3 emissions, considering the urgent need to reduce the emissions both in Sweden and elsewhere.

3 RESULTS AND DISCUSSION

The results are organized as follows: First, model results are presented and evaluated focusing on the larger European domain, with additional evaluation for the nested high-resolution domain (Sections 3.1 and 3.2). Second, assessments of national emission reduction effects are presented for the Swedish domain (Section 3.3). As NH_3 emissions have a strong seasonal dependence, we focus on season-wise analysis. The main findings and implications are summarized and further discussed in Section 3.4. Results are shown for 2019; the model evaluation for 2017 gives similar results as summarized in the text.

3.1 MODELED REGIONAL CONCENTRATIONS

Figures 1 and 2 present seasonal means of gas-phase NH_3 simulated by the MATCH model for the European and Swedish domains, respectively. The highest concentrations in Europe, appearing at spring and summer time, are

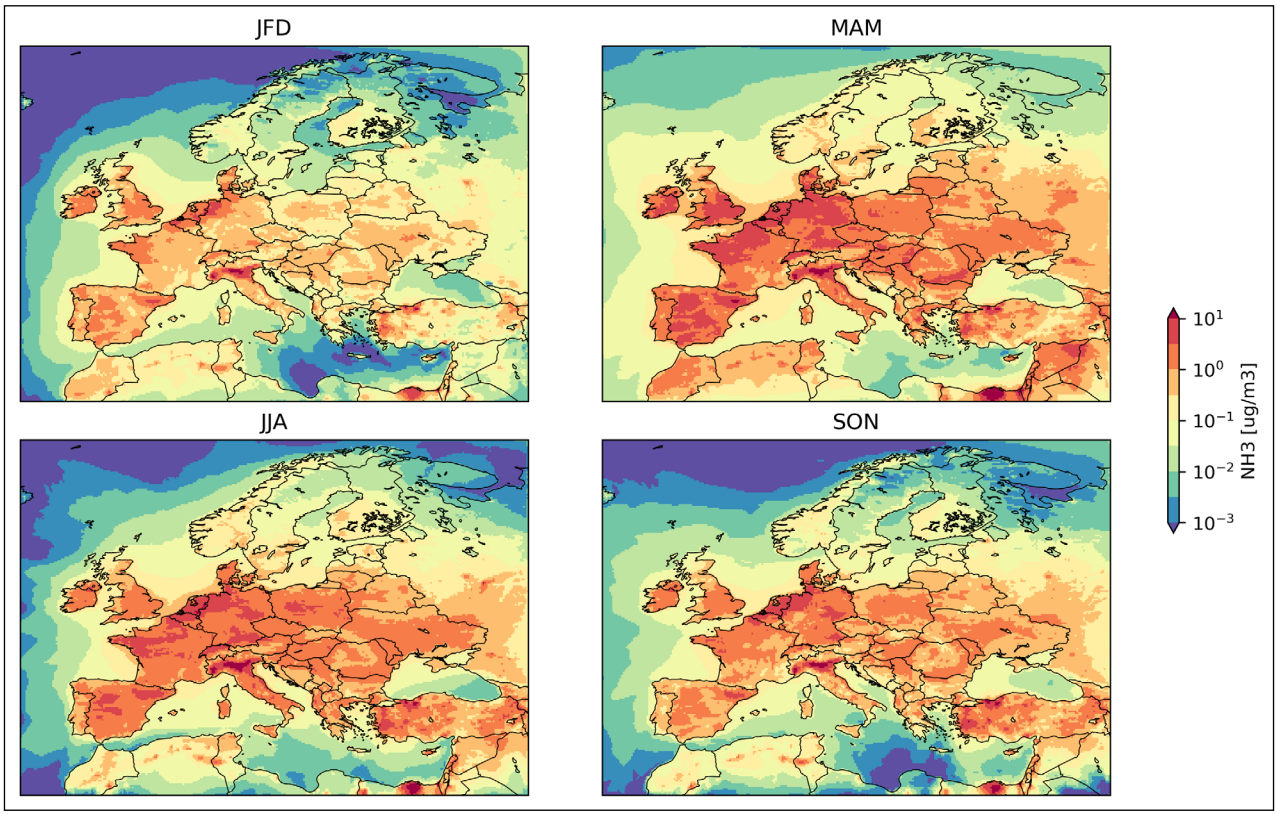

Figure 1 Seasonal (3-month) mean values of modeled NH_3 concentrations for the European model domain for 2019. The seasons are as follows: JFD: January, February, December; MAM: March, April, May; JJA: June, July, August; SON: September, October, November.

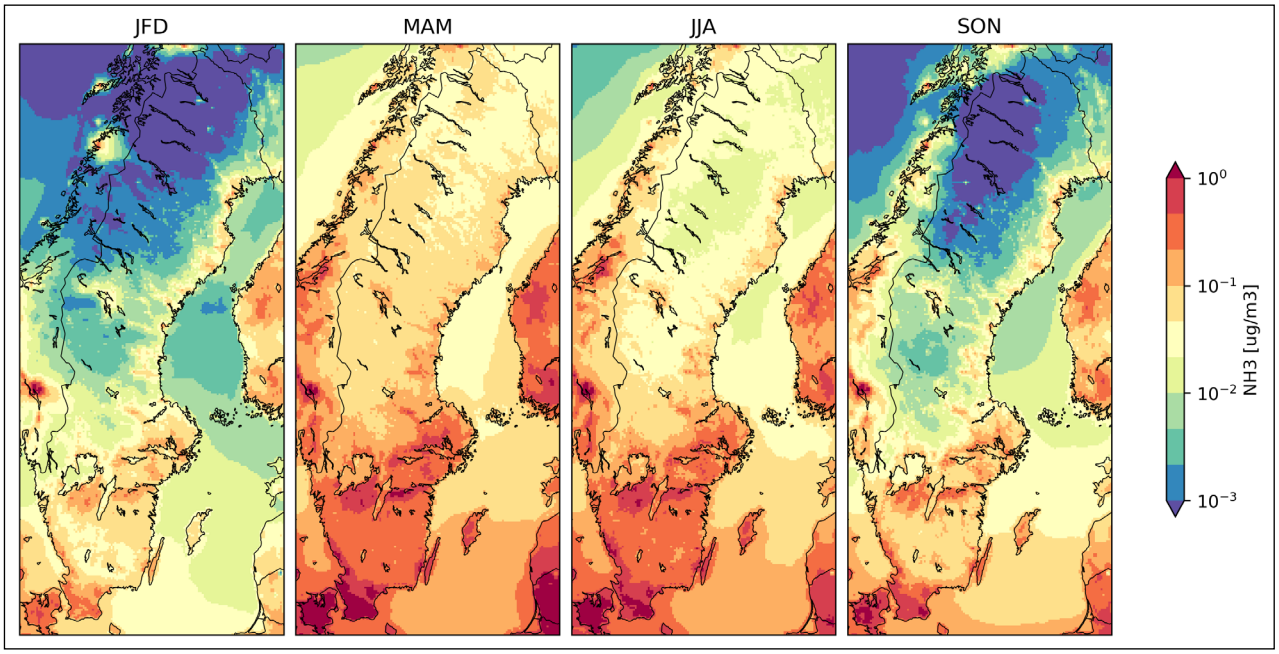

Figure 2 Seasonal mean values of modeled NH_3 concentrations for the Swedish model domain for 2019. Note the different y-axis scale compared with Figure 1.

located around major agricultural sources (Figure S18). In the vicinity of Sweden, these include sources in Denmark, the Netherlands and Northern Germany (Figure 1). As expected, total reduced nitrogen species NH_x exhibit similar patterns as NH_3 (to some extent, the modeled total oxidized nitrogen also shows similar distributions; Figure S1). NH_3 concentrations are generally lower in Northern Europe except for Denmark, with the highest Nordic concentrations (excluding Denmark) located in southern parts of Sweden and Finland, and Southern and mid-Norway. In Sweden, elevated NH_3 levels are centered upon the southernmost areas (Figure 2).

3.2 MODEL–MEASUREMENT COMPARISONS

Figure 3 shows monthly modeled and observed NH_3 concentrations, normalized mean bias (NMB) and correlation over all stations for the two model domains. Seasonal station-wise NMB and correlation for the European domain are shown in Figure 4; the station-wise statistics are similar for the Swedish-domain simulation. The global monthly trends, here characterized by the median and the 25th and 75th percentiles, are qualitatively well represented by the model (Figure 3, panels (a) and (d)). The modeled and observed distributions show a spring peak due to application of fertilizers, followed by elevated levels in summer time and decline towards winter.

The available observations primarily cover Fennoscandia and Netherlands, with a few individual stations in United Kingdom and Eastern Europe (Figure 4). In general, modeled concentrations tend to be lower than measured values, with some overestimation in spring and autumn seasons mainly at a few stations in Denmark and Southern Sweden (Figure 4a). Correlation is generally higher in spring and summer with elevated NH_3 (Figure 4b). The global monthly NMB for all available stations in the European domain varies between −76% and −19% (Figure 3b), with a global annual NMB of −44% (Table S1). For global correlation, the monthly and annual values are 0.59...0.86 (Figure 3c) and 0.65 (Table S1), respectively. The NMB and correlation values can be considered satisfactory compared to previous NH_3 model studies applying a regional CTM, reporting absolute NMB of 48% and correlation of 0.59 for weekly or monthly surface observations in Netherlands and Germany (Ge *et al.*, 2020).

The overall model results are improved with the updated NH_3 scheme, which decreases the positive bias in Denmark and Southern Sweden (Section S2.2; Figure S13). As shown in Figure S2, the new scheme predominantly reduces the predicted NH_3 in the European domain, although some increases occur depending on location and season. The changes are due to the net effects of the updated deposition flux and the introduction of the compensation points (Supplementary Information Section S3.1). The compensation points, that determine the upward flux, peak in spring and summer, as summarized in Supplementary Information Section S1.1. Within Sweden, modeled NH_3 is decreased by up to approximately a factor of 2. The relative changes are similar over the main NH_3 emission areas in Southern and Central Sweden, except for the most intensive national agricultural hotspots that are less affected likely due to elevated compensation points. It can be noted that the modeled NH_3 may involve more uncertainties in clean remote regions with no observations. Compensation points in such environments are less well understood, and ground emissions may occur especially under warm and dry conditions (Walker *et al.*, 2023; Wu *et al.*, 2023).

Evaluation statistics for the other nitrogen species NH_x and ΣNO_3, summarized in Figures S14, S15 and S17, are comparable to those of NH_3, or in some cases better. For NH_x, the global monthly NMB varies between −13% and 72%, but values higher than 25% (40%...72%) only occur in March–May. For ΣNO_3, the bias is within −28%...24%. Global monthly correlation for both species is within 0.60...0.81 (Supplementary Information Section S2.2). Model evaluations for the standard regulated pollutants O_3, NO_2, $PM_{2.5}$ and PM_{10} are presented in Figures S9–S12 and S16, showing performance similar to previous MATCH and other model studies (e.g. Frohn *et al.*, 2022). It can be noted that while $PM_{2.5}$ and PM_{10} involve more prominent systematic bias compared to the other species, this is attributed to PM components other than ammonium and nitrate that are the focus of this work (Supplementary Information Section S2.1). The European-domain simulation for 2017 shows generally similar evaluation results for the studied species in terms of NMB and correlation (Table S1), and their temporal and spatial trends. The main difference to 2019 is that NH_3 doesn't exhibit a sharp global peak in April (as in panel (a) in Figure 3). Instead, observations and model results for 2017 show only moderately elevated levels or a low maximum, respectively. This can be partly due to a few more observation stations in central Europe being available and included in the statistics. ΣNO_3 involves less global month-wise variation with a rather flat median curve (compared with panel (a) in Figure S15) for both observations and model. Overall, the simulation years show similar model performance for the nitrogen species, supporting the application of the model setup for the ammonia reduction assessments. While multi-year simulations are outside the scope of the present study, long-term trends in PM simulated by the MATCH model have been evaluated in previous work (Tsyro *et al.*, 2022). The previous European-scale simulations for 2000–2010 show decreases in PM following the European emission reductions, in line with observations and with consistent model bias without significant year-to-year variability.

3.3 AIR POLLUTION MITIGATION POTENTIAL IN SWEDEN BY NH_3 REDUCTION

Modeled changes in nitrogen-containing species for the Swedish national emission reduction tests are presented in Figures 5–7. Decreases in gaseous NH_3 upon 20% reduction of agricultural NH_3 emissions are highest in regions with elevated NH_3 levels and hotspots in

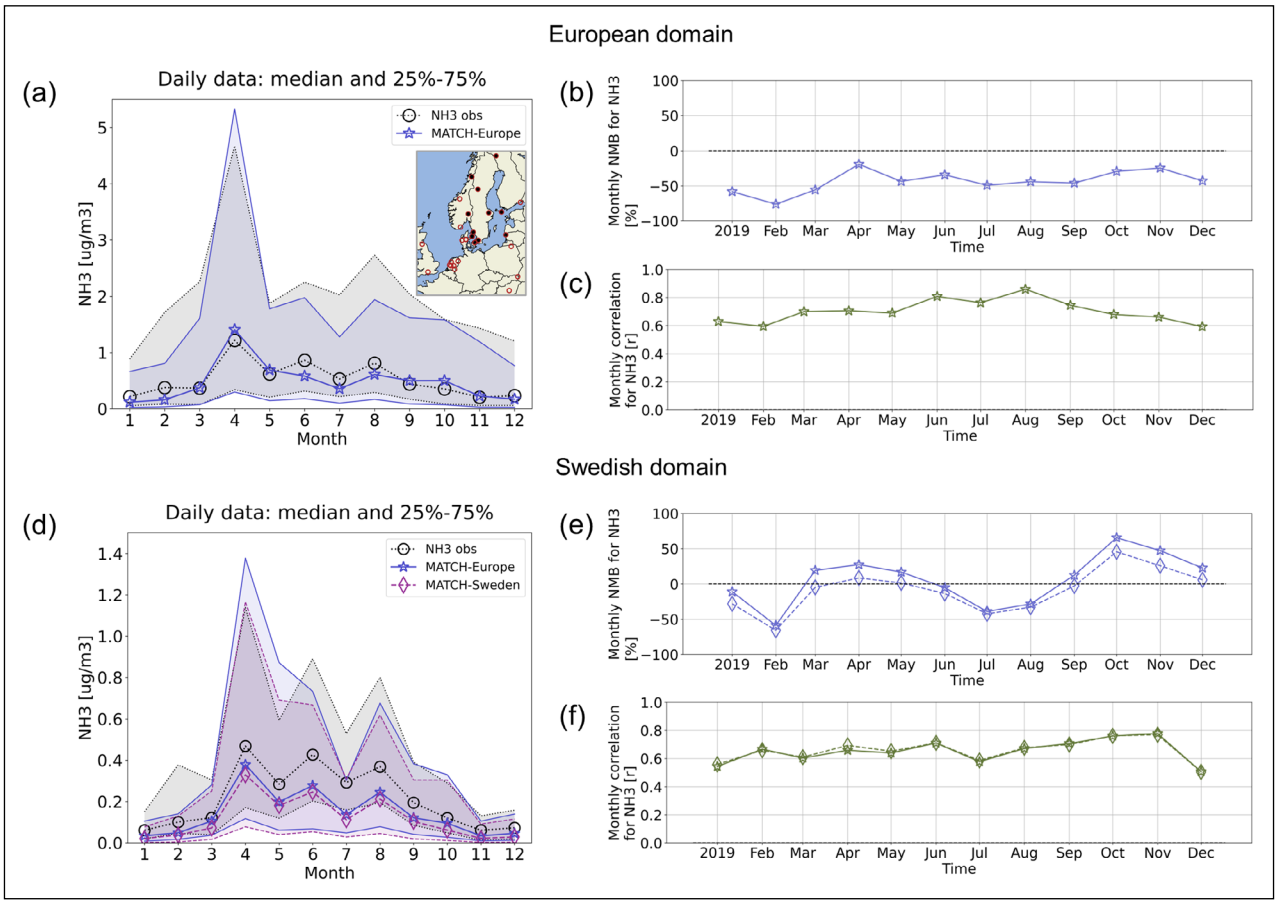

Figure 3 Monthly NH_3 concentrations and evaluation statistics over all measurement stations for the European (panels **(a)–(c)**) and Swedish (panels **(d)–(f)**) model domains. Panels (a) and (d): Annual distribution of modeled and observed daily mean NH_3 concentrations. Panels (b) and (e): Monthly global normalized mean bias (NMB). Panels (c) and (f): Monthly global Pearson correlation coefficient. Panels for the Swedish model domain ((d)–(f)) include only the stations in the Sweden-centered domain and show results from both the Swedish-scale and the coarser-resolution European-scale simulations. All stations are marked with red circles on the inset map in panel (a), with the Swedish-domain stations filled with black.

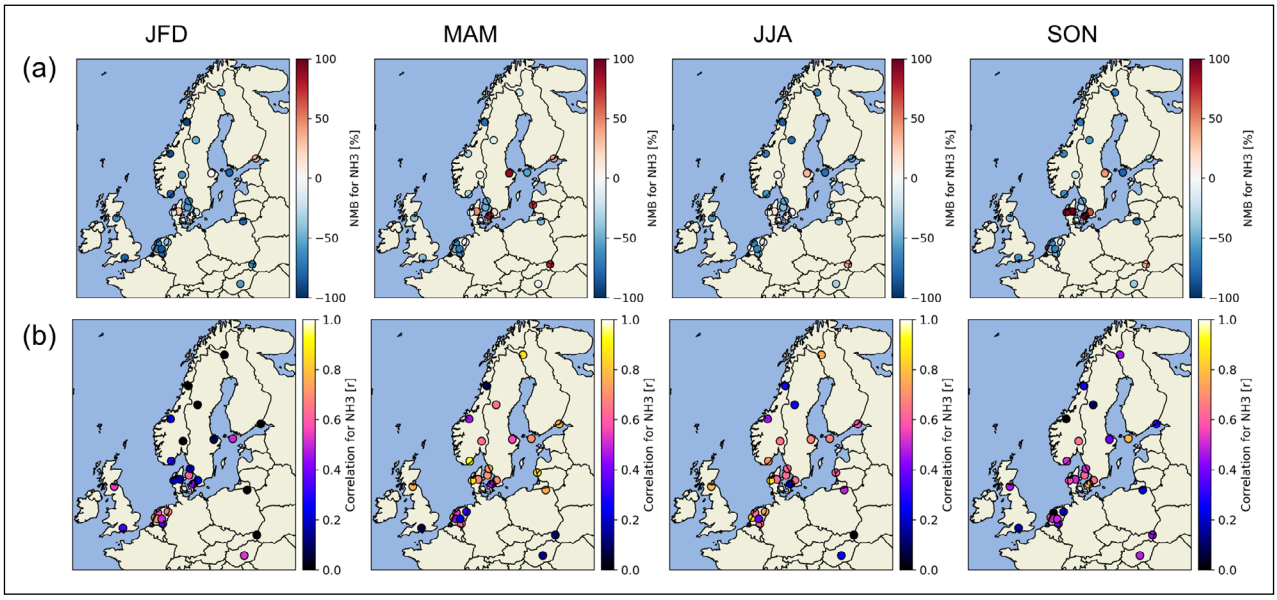

Figure 4 Seasonal evaluation statistics for modeled daily mean NH_3 concentrations for the European model domain. Panel **(a)**: Normalized mean bias (NMB). Panel **(b)**: Pearson correlation coefficient.

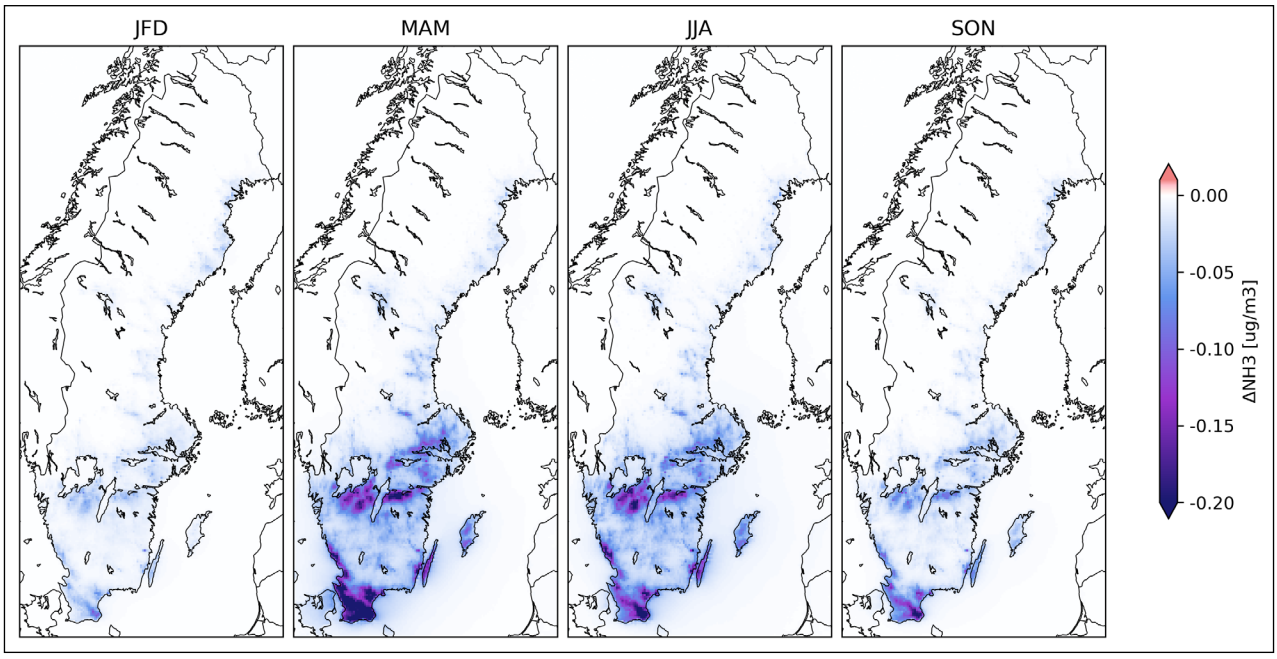

Figure 5 Predicted changes in seasonal mean NH_3 concentrations in model scenario with reduced Swedish emissions for agricultural NH_3.

Southern Sweden (Figure 5; cf. Figure 2). In these areas, the corresponding relative changes are within ca. −10... −20%, which suggests that such emission reductions can effectively decrease NH_3 levels.

Concentrations of fine particulate matter $PM_{2.5}$ and predicted effects of 20% national emission reductions in (1) agricultural NH_3 and (2) road-transport NO_x (as NO_2) are shown in Figure 6. The decreases in seasonal $PM_{2.5}$ upon reduction of NH_3 (panel (b)) show similar spatial patterns as decreases in NH_3 (Figure 5), as NH_3 can directly contribute to inorganic PM by nitrate or sulfate formation with NO_x and SO_2 oxidation products, respectively. The largest decreases in $PM_{2.5}$ occur during the winter and autumn seasons, when NH_3 concentrations are lower. The impact is generally smaller during spring and summer with elevated NH_3 levels, as PM formation is less limited by ammonia under these seasons.

By contrast, effects of NO_x reduction on $PM_{2.5}$ are close to negligible (panel (c)), even if NO_2 levels are decreased especially in Stockholm region (Figure S3). NO_x needs to undergo oxidation to form nitric acid, which finally can contribute to nitrate PM in the presence of NH_3. That is, NO_x reduction effects on secondary inorganic PM are limited by the availability of both oxidants and NH_3, and the contribution of sulfate PM. These results agree well with previous European-scale assessments of season-dependent NH_3 contributions to $PM_{2.5}$ (Backes *et al.*, 2016; Clappier *et al.*, 2021), and with European and global estimates of the benefits of NH_3 abatement over NO_x (Clappier *et al.*, 2021; Gu *et al.*, 2021). In Figure 6c,

the minor decreases in $PM_{2.5}$ upon NO_x reduction apply mainly to areas affected by agricultural NH_3, but their magnitude is not sensitive to the NH_3 levels (Figure 2). NO_x reduction can also increase $PM_{2.5}$ through increases in O_3 (and other oxidants) and hence in the oxidative potential of the atmosphere (Gaubert *et al.*, 2021; Jhun *et al.*, 2015). Here, very small increases in $PM_{2.5}$ are seen in Northern Sweden due to minor increases in O_3 (Figure S4), mainly affecting secondary organic PM. The O_3 effect may also make inorganic PM reduction less efficient due to increased NO_2 and SO_2 oxidation (Clappier *et al.*, 2021).

Figure 7 shows the impacts of NH_3 and NO_x reductions on total deposited nitrogen mass (Ndep), including dry and wet deposition. Similarly to $PM_{2.5}$ (Figure 6), reducing NH_3 decreases Ndep following the NH_3 reduction patterns (Figure 5), while NO_x shows no or negligible effects. Here,

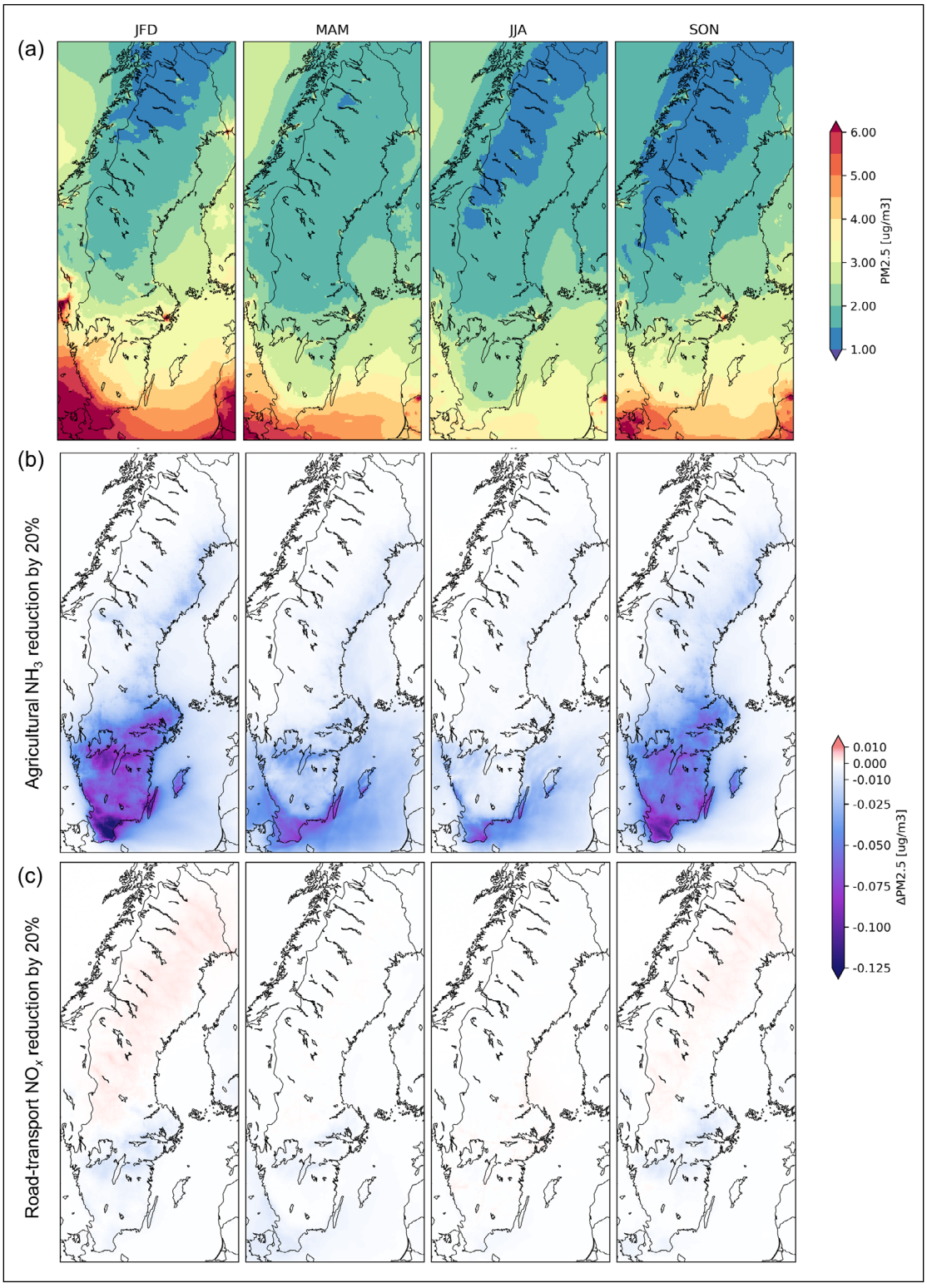

Figure 6 Panel **(a)**: Modeled seasonal mean concentrations of $PM_{2.5}$. Panels **(b)** and **(c)**: Predicted changes in model scenarios with reduced Swedish emissions for agricultural NH_3 (panel (b)), and NO_x from road transport (panel (c)).

also the season-dependent magnitude of the Ndep reduction follows that of the NH_3 reduction, i.e. it shows largest reductions in spring and summer time, as the atmospheric concentration is directly linked to deposition. Here, NO_x effects are limited by the relatively smaller contribution of nitrogen mass to the total emissions compared with NH_3 (as a large fraction of emitted NO_x mass is oxygen). A substantial contribution to the decreases in Ndep upon NH_3 reduction (Figure 7b) comes from reduced dry deposition of gaseous NH_3. Dry deposition, for which NH_3 gas is a major component, constitutes a notable fraction of total nitrogen deposition as shown in Figures S5 and S6. Here, changes in dry deposition of NH_3 contribute significantly to the total Ndep changes (panels (b) in Figure S5 and Figure 7, respectively). Reduced wet deposition has large impacts, especially in the areas with the largest total Ndep (and NH_3) decreases in Southern Sweden in the summer period (Figure 7b).

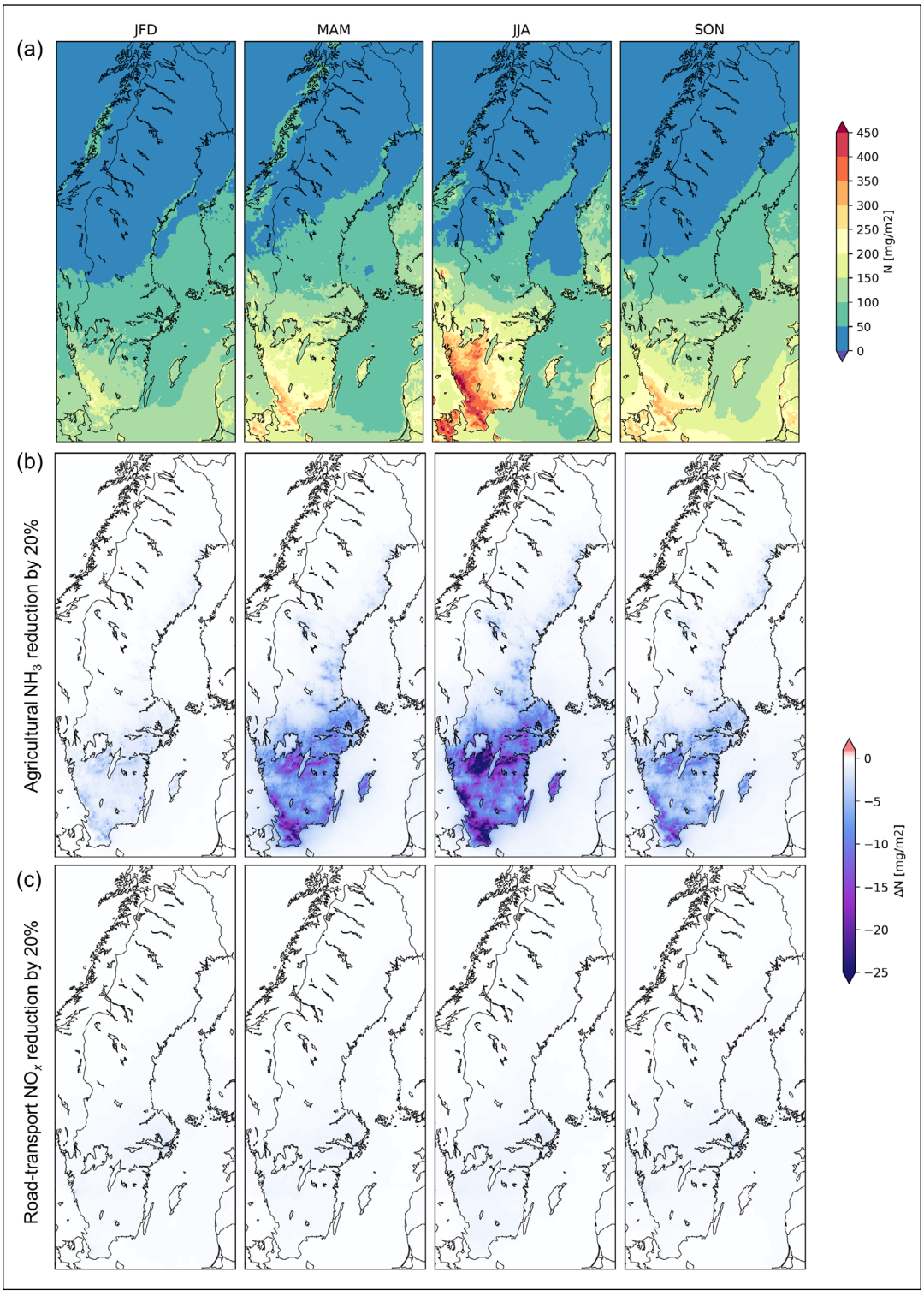

Figure 7 Panel **(a)**: Modeled seasonal accumulated deposition of total nitrogen. Panels **(b)** and **(c)**: Predicted changes in model scenarios with reduced Swedish emissions for agricultural NH_3 (panel (b)), and NO_x from road transport (panel (c)).

3.4 IMPLICATIONS

Overall, the model performance for the studied reduced and oxidized nitrogen components NH_3, NH_x = NH_3(g) + NH_4^+(pm) and ΣNO_3 = HNO_3(g) + NO_3^-(pm) is satisfying, especially in capturing the qualitative seasonal variations. The observed temporal trends in monthly medians and 25th–75th percentiles are qualitatively reproduced by the model, and the NMB and correlation statistics are comparable to previous regional model studies (Section 3.2).

Results obtained by the model setup for NH_3 and NO_x emission reduction impacts in Sweden highlight the following implications:

- Reducing agricultural NH_3 emissions is potentially beneficial for both human health and the environment through decreases in airborne fine particulate matter ($PM_{2.5}$) and nitrogen input to ecosystems by atmospheric deposition (Ndep), respectively. Although the nitrogen budget of Sweden is affected by emissions in other countries through long-range transport, reducing national NH_3 emissions has distinct impacts on air quality and environmental indicators in Sweden, especially in regions around the emission sources. It can be noted that here the reduced nitrogen input through direct deposition primarily applies to land ecosystems over areas affected by the agricultural sources, with only minor reductions for the Baltic Sea and coastal areas.
- Sweden's most urgent reduction targets currently include both NH_3 and NO_x. NO_x reductions contribute to improved air quality by decreased NO_x concentrations, but the present assessments suggest very minor or no effects on $PM_{2.5}$ and Ndep. The impacts naturally depend on the applied emission scaling factors, here 20% for both species, corresponding to approximately equal reductions in total emitted mass (9.5 ktons NO_x and 9.3 ktons NH_3, respectively). The potential to reduce Ndep depends on the reductions in emitted nitrogen (N) mass, which is here approximately 2.6 times higher for NH_3 (i.e. the relative reduction in NO_x emissions would need to be substantially higher than 20% to achieve similar theoretical potential as NH_3 to reduce the nitrogen inputs). On the other hand, large NO_x reductions will also lead to changes in concentrations of O_3 and other oxidants (OH and nitrate radicals), which may enhance secondary PM formation—and thus lead to increasing effects on $PM_{2.5}$—in some regions and seasons. This makes the health and environmental impacts of changes in NO_x emissions more complex.
- The NH_3 reduction impacts on $PM_{2.5}$ exhibit a significant seasonal dependence due to the seasonally varying NH_3 concentrations in relation to acidic PM precursors. NH_3 reductions are most beneficial in ammonia-limited conditions, that is, in the winter half-year with low NH_3 concentrations. However, it must be noted that NH_3 reduction is always beneficial for decreasing Ndep, and especially at high NH_3 concentrations. Here, a notable contribution to decreases in total nitrogen deposition is due to reduced direct dry deposition of gaseous NH_3. The minor effects of NO_x reduction on $PM_{2.5}$ suggest that $PM_{2.5}$ formation in Sweden is primarily not limited by NO_x. In addition to NH_3 reductions, secondary inorganic PM could potentially be mitigated by sulfate reduction through SO_x emissions, especially in the summer season when neither NO_x nor NH_3 reduction show significant effects. On the other hand, Swedish SO_x emissions are already relatively low and the potential for further sulfate PM reductions may be limited.

4 CONCLUSIONS

The present model assessments of agricultural ammonia emission reduction effects in Sweden indicate potential to mitigate both fine particulate matter and nitrogen input to ecosystems within Sweden. The applicability of the model setup for modeling of ammonia and related nitrogen species, including improved treatment of NH_3 through bidirectional surface–atmosphere fluxes, is verified against surface observations. In terms of $PM_{2.5}$ abatement, NH_3 reduction is most efficient in winter and autumn when NH_3 emissions are lower and PM formation is NH_3-limited. However, reducing the higher NH_3 emissions in spring and summer is required to efficiently reduce the atmospheric nitrogen burden and deposition to ecosystems. While Sweden also needs to reduce NO_x emissions, the present assessments suggest no favorable co-effects on $PM_{2.5}$ or Ndep mitigation, as neither of these is sensitive to NO_x. Overall, the results show direct benefits of national NH_3 reduction to air quality and environmental protection in Sweden.

Ammonia is currently the most challenging component for air pollutant emission reductions, and Sweden is among the EU Member States that are not yet reaching their present ammonia reduction goals. This work demonstrates the possibility to integrate the modeling of ammonia to national air pollution surveillance, including model assessments of emission reduction impacts. The national emission scaling tests are used to study the effects of different pollutants and emission sectors on air quality and environmental indicators, here applied to the two largest nitrogen-emitting sectors. This supports assessing the benefits and the potential of national abatement actions to mitigate pollution and harmful environmental impacts, and planning of optimal mitigation strategies.

DATA ACCESSIBILITY STATEMENT

Observation data are available in the EBAS atmospheric database (ebas.nilu.no). The additional NH_3 observation data for Denmark are available from Aarhus University (Lise Marie Frohn Rasmussen and Zhuyun Ye) and for the Netherlands at data.rivm.nl/data/luchtmeetnet. MATCH simulation data are available from the authors upon request.

ADDITIONAL FILE

The additional file for this article can be found as follows:

- **Supplementary Information.** Atmospheric Ammonia in Sweden: Regional Modeling and Assessment of National Emission Reduction Benefits. DOI: https://doi.org/10.16993/tellusb.1881.s1

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COMPETING INTERESTS

The authors have no competing interests to declare.

AUTHOR CONTRIBUTIONS

Conceptualization: RB, TO; Data curation: RB, TO; Formal analysis: TO; Investigation: RB, TO; Methodology: RB (model development and modeling), TO (model evaluation); Software: RB, TO; Validation: RB, TO; Visualization: TO; Writing – original draft: TO; Writing – review & editing: RB, TO.

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