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Improved modelling of atmospheric ammonia over Denmark using the coupled modelling system DAMOS

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Abstract

A local-scale Gaussian dispersion-deposition model (OML-DEP) has been coupled to a regional chemistry-transport model (DEHM) in the Danish Ammonia Modelling System, DAMOS. Thereby it has been possible to model the distribution of ammonia concentrations and depositions on a spatial resolution down to 400 m × 400 m for selected areas in Denmark. DAMOS has been validated against measured concentrations from the dense measuring network covering Denmark. Here measured data from 21 sites are included and the validation period covers 2–5 yr within the period 2005–2009. A standard time-series analysis (using statistic parameters like correlation and bias) show that the coupled model system captures the measured time-series better than the regional scale model alone. However, our study also shows that about 50 % of the modelled concentration level at a given location originates from non-local emission sources. The local-scale model covers a domain of 16 km × 16 km and of the locally released ammonia (NH₃) within this domain, our simulations at five sites, show that 14–27 % of the locally emitted NH₃ also deposit locally. These results underline the importance of including both high-resolution locale-scale modelling of NH₃ as well as the regional scale component described by the regional model. The DAMOS system can be used as a tool in environmental management in relation to assessments of total nitrogen load of sensitive nature areas in intense agricultural regions. However, high spatio-temporal resolution in input parameters like NH₃ emissions and land-use data are required.

1 Introduction

The EEAs third European environmental assessment published in 2003 concluded that despite considerable improvements over the 1990ties, critical loads for nitrogen (N) were exceeded for almost 60 % of the European ecosystems (EEA, 2003). In recent years, the agricultural nutrient balance has improved for many of the EU member

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states, but according to the most recent estimates more than 40 % of sensitive terrestrial and freshwater ecosystem areas in these countries are still subject to atmospheric N depositions above critical loads (EEA, 2010). In addition, it is well documented that also for many coastal marine ecosystems; the atmospheric depositions play a significant role for the overall N loads (Spokes et al., 2006; Spokes and Jickells, 2005). Exceedances of critical loads may on the long term lead to loss of biodiversity (Bobbink et al., 2010; Stevens et al., 2004, 2010), where valuable rare species are in danger of being lost (Krupa, 2003). Critical load exceedances have received increasing scientific and political attention in Europe over the last couple of decades. This awareness calls for new strategies (Sutton et al., 2011) that in a balanced way deal with all major nutrient sources (Conley et al., 2009), and includes high quality assessment tools that deals with these sources for the management and policy making, both regarding protection of coastal areas (Bartnicki et al., 2011; Geels et al., 2011; Hertel et al., 2003; Langner et al., 2009) and of sensitive ecosystems (Sutton et al., 2007).

In the close vicinity of intense agricultural activities, deposition of locally emitted ammonia (NH_3) may totally dominate the N loads to the terrestrial ecosystems (Hertel et al., 2006). In such an area the contribution from a single farm to the annual atmospheric deposition of N within the first few hundred meters down-wind in the dominating wind direction may be of the order of 50 or even up to 100–200 $\text{kg N ha}^{-1} \text{yr}^{-1}$, depending on the local dispersion conditions, the distance to the source, and the size of the emission (Hertel et al., 2006). Such high loads are far beyond critical loads for sensitive terrestrial ecosystems, which for many systems are in the range of 10 to 20 $\text{kg N ha}^{-1} \text{yr}^{-1}$ (UNECE, 2004). The footprint from local NH_3 emissions may vary significantly and has been investigated by a range of methods. Indirect estimates using mass balance, ^{15}N labelling, SF_6 to NH_3 ratio methods, as well as modelling studies, have thus been applied to determine the fraction of emitted NH_3 which is recaptured within the first 2 km downwind from the source. The results vary widely showing recapture fractions within the first 2 km downwind from the source between 2 % and up to 60 %, but in most cases in the range between 10 % and 40 % (Asman, 1998; Dragosits

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et al., 2002; Loubet et al., 2001, 2009; Sommer and Jensen, 1991). The wide range in recapture fractions reflect the variability in deposition velocities to different surface types, the variability in meteorological conditions, but also the uncertainties in the determination of various parameters and governing processes. One important issue that has attracted much attention in the recent years is the bi-directional flux of NH_3 that can occur in regions, where high atmospheric NH_3 levels have saturated the surface (Nemitz et al., 2004; Sutton et al., 2007). Progress has been made towards a better understanding and description of this process, but so far the deposition schemes commonly applied for NH_3 do not take this bi-directional flux into account (Massad et al., 2010).

It has been known for some time that coarse resolution in deposition mapping combined with critical load limits leads to underestimation of the exceedances of critical loads (Spranger et al., 2001). A variety of studies have shown that higher spatial resolution in NH_3 emission data lead to significant changes in modelled N depositions (Duyzer et al., 2001; Pul et al., 2004), and our studies have shown that in addition to the high spatial resolution also high temporal resolution may be crucial for the performance of chemistry-transport models (CTMs) (Skjøth et al., 2004, 2011). This is also supported by a review with focus on air quality forecasting, stating that dynamical calculations of NH_3 emissions are needed for a better prediction of high particulate matter episodes (Menut and Bessagnet, 2010). The emission pattern of NH_3 is generally characterised by a large number of point sources (animal housings and storages) surrounded by area sources (fields to which manure is applied) in the rural areas. The scavenging of NH_3 from the atmosphere is a competition between two relatively fast processes; the reactions with acid gases and aerosols leading to aerosol phase ammonium (NH_4^+), and the dry deposition (V_d in the order of 0.03 to 5.0 cm s^{-1} depending on surface and meteorological conditions) (Hertel et al., 2006). The rate of chemical conversion has been decreasing in recent years (especially when compared to the situation in the 1980ties and 1990ties) due to reductions in mainly sulphur emissions (leading to reductions in ambient air levels of sulphuric acid; H_2SO_4) (Van Jaarsveld,

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2004). In combination with the heterogeneity of the NH_3 emissions, this leads to large gradients in NH_3 concentration and deposition pattern; gradients that complicate interpretation of observed NH_3 levels as these generally cannot be considered spatially representative (Hertel et al., 2007). For a proper assessment of the impact of NH_3 this heterogeneity requires a very dense monitoring network (Erisman et al., 2005) and since this is not feasible (especially on the long run), CTMs are typically applied e.g. in combination with observations making use of integrated monitoring and assessment (Hertel et al., 2007).

Current regional CTMs (typically Eulerian) with a resolution of 50 km to 10 km tend to over- or under predict observed NH_3 levels across Europe (Simpson et al., 2011). A few national-scale models (with resolutions from 5 km down to 1 km) have shown better agreement with observations (Dragosits et al., 2002; Nemitz and Sutton, 2004; Pul et al., 2009; Theobald et al., 2004). Eulerian models are well suited for describing long-range to regional-scale atmospheric transport, but for the local-scale dispersion on resolution of 1 km or even higher, the Gaussian approach is useful and commonly applied – an example is the OPS model used in the Netherlands (Van Jaarsveld, 2004). In order to obtain improved model performance from applying high resolution local-scale models, it is required that high quality emission inventories with sufficiently high temporal and spatial resolution are available (Kryza et al., 2011). Denmark is a highly intensive agricultural country and many sensible ecosystems are closely surrounded by areas used for agricultural purposes. Many natural and semi-natural ecosystems are therefore located as a patchwork within the agricultural landscape. It is our hypothesis that use of high resolution emission inventories in combination with local scale and regional scale CTMs in a coupled system, improve the model performance significantly with respect to assessing ambient NH_3 levels and resulting depositions to this patchwork of ecosystems. We demonstrate this through a presentation of the Danish Ammonia Modelling System (DAMOS) – a combination of the regional scale Danish Eulerian Hemispheric Model (DEHM) (Christensen, 1997) and the local scale Gaussian dispersion-deposition model OML-DEP (Sommer et al., 2009). The DAMOS

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results are compared with observations from Danish rural monitoring stations, where we within the period 2005–2009 have a unique data set with measurements from up to 21 different sites. In the model validation, the differences between regional scale model alone and the coupled model system are discussed. Moreover, the impact of local versus non-local emissions at different locations in the country are discussed in relation to implications for environmental management and assessment of emission reduction strategies.

2 Methodology

In the subsequent sections first the monitoring methods and sites are described. Then the national emission inventory and the applied model setup are explained.

2.1 Monitoring methods and sites

Within the framework of the Danish Background Air Quality Monitoring Program, concentrations and depositions of eutrophying and acidifying air pollutants have been monitored in Denmark since 1989. Currently the concentrations of several nitrogen gases are measured at five of the stations in the Danish monitoring network using the filter pack method (Fig. 1). The stations are located in background areas in order to represent the background levels for the particular areas rather than the direct impact of local sources. The stations Anholt and Keldsnor are characterized as coastal sites in areas with only a few local sources for ammonia. Ulborg, Lindet and Tange are all located within agricultural areas in Jutland/Denmark and are characterized as forests sites (Ulborg and Lindet) and an agricultural site (Tange). At the five stations, diurnal sampling is carried out with the Danish filter pack sampler (a slightly modified version of the EMEP filter pack sampler). This has in the past been shown to give a reasonable separation between gaseous ammonia and particulate ammonium (NH_4^+) under Danish conditions (Andersen and Hovmand, 1994). In the present paper the yearly mean

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concentration of ammonia determined by the filter pack method at the five locations are presented. Additionally long term denuder measurements (weekly to half monthly) of ammonia and ammonium are carried out at Ulborg, Anholt, Keldsnor and Lindet. The denuder is a slightly modified version of the denuder described by (1979), with oxalic acid coating and a flow rate of 1 l min^{-1} .

During the years 2006–2009 ammonia has been measured at several locations in Denmark (Fig. 1) with passive diffusion tube samplers ALPHA (Adapted Low-cost Passive High Absorption). Ammonia is absorbed on a filter impregnated with citric acid. The ALPHA samples have been tested against the denuder method at Danish stations with different ammonia levels (for further details see Andersen et al., 2009; Ellermann et al., 2009; Tang and Sutton, 2003). The measurements cover different regional levels of background concentrations of ammonia, different types of nitrogen sensitive ecosystems and different years. The measurements are run with a half monthly to monthly temporal resolution and comparisons to model results are included in the present paper.

Table 1 gives an overview of locations, measuring method and time resolution for the concentrations used in the comparisons of measured and modelled values. The uncertainty of the determination of the concentration of ammonia is assumed to be on the order of up to 20 % (Andersen et al., 2009; Andersen and Hovmand, 1994).

2.2 Ammonia emissions for Denmark

A dynamical ammonia emission model based on a detailed national inventory and describing the temporal variation throughout the year has been developed for Denmark (Gyldenkærne et al., 2005; Skjøth et al., 2004). The inventory is based on information from national data bases: the Central Husbandry Register and The General Agricultural Register, which gives the geographical location of each agricultural building including stables, storages and fields throughout the country. A standardised ammonia emission for each live stock farm and associated fields are then estimated based on the information on animal type/number and type of crops. The final inventory includes

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the total annual ammonia emission coming from:

1. point sources (farms, storages etc.) at the exact location of these in Denmark,
2. area sources i.e. emissions from fields due to grown crops as well as emission due to application of manure and mineral fertilizer.

5 The area sources are distributed within a 100 m × 100 m grid covering Denmark. Examples of the distribution of NH₃ emissions across Denmark and within the area around the Tange site are seen in Figs. 1b and 2b. The temporal variability of the NH₃ emissions is described by 15 additive functions reflecting different agricultural activities and a 16th function describing the contribution from traffic. The functions describe the temporal variations of the emission by specific parameterisations characterising each activity (see Table 2 and for details see Gyldenkærne et al., 2005). An example of the seasonal variations of the different functions and hence the hourly NH₃ emission is shown for 2007 at the Tange site (Fig. 3). The parameterisations are based on knowledge on emissions factors and the sensitivity of the emission to temperature, wind and ventilation; a simple crop growth model; information on current agricultural practise etc., and the derived temporal variation in the emissions have been shown to agree with measured ammonia levels in Denmark (Skjøth et al. 2004) as well as reflecting the changes in Danish regulations (Skjøth et al., 2008). Recently the inventory has been extended to cover Northern Europe, which resulted in improved NH₃ calculations at sites in Germany and even lead to additional improvements at the Danish sites (Skjøth et al., 2011).

The inventory is updated every year with:

1. new data on actual farm size, taking into account the structural development in the agricultural sector,
2. updated nitrogen excretion rates per animal, as these change between years due to increased efficacy (Poulsen et al., 2001; Poulsen, 2011),

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3. information on fields allocated to each farm,
4. updated amount of manure available at farm level (farm production minus export and plus export),
5. meteorological data for the specific year, which is used for estimating the temporal variations of the emissions.

The most recent inventory available for this study is for 2007. For the total national emission the uncertainty is assumed to be in the range of 5–10 %, while the uncertainty on farm/field level is in the range of 25–35 %.

2.3 The Danish Ammonia Modelling System

The Danish Ammonia Modelling System (DAMOS) is a coupled system consisting of the regional air pollution model DEHM and the local-scale dispersion and deposition model OML-DEP. The overall structure of DAMOS, the necessary input data to the models and the flow of information between the two models is described in the following.

2.3.1 The DEHM model

The Danish Eulerian Hemispheric Model (DEHM) is a 3-D Eulerian transport model covering the Northern Hemisphere. The model exist today in several versions and has through the years been applied for numerous studies, for example studies of transport of air pollution to the Arctic (Christensen, 1997; Christensen et al., 2004; Genualdi et al., 2011; Hansen et al., 2008), for analyses of atmospheric CO₂ over Europe (Geels et al., 2004, 2007), for health assessments (Brandt et al., 2011) and for studying the future air pollution levels in a changed climate (Hedegaard et al., 2008, 2011; Langner et al., 2012). Furthermore, the model is used in a model system, THOR, integrating the regional scale with an urban background model and an urban street canyon model (Brandt et al., 2001). This system is applied for describing the air pollution in cities,

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taking into account higher resolution emissions as well as different physical local scale characteristics (as e.g. dispersion) similar to the DAMOS system. In relation to nitrogen the model has been used for several studies related to e.g. impact studies in the Arctic (Forsius et al., 2010) and projections of the N load to the Baltic Sea (Geels et al., 2011). The model is on a yearly basis applied in the Danish Background Air Quality Monitoring Program for calculations of nitrogen depositions to the Danish land and sea areas (Ellermann et al., 2009).

In the current study the chemistry version of DEHM is applied, including 67 chemical compounds and 122 chemical reactions (Brandt et al., 2012; Christensen, 1997; Frohn et al., 2002). The horizontal resolution is 150 km × 150 km in the mother domain and the setup includes three nested domains with the highest resolution of 5.56 km × 5.56 km over Denmark. Vertically the model is divided into 29 unevenly distributed σ -levels; with the highest resolution towards the ground (lowest level is approximately 25 m thick). The necessary meteorological input is provided by the MM5v3 model (Grell et al., 1995), with a setup of grid definition and model domains that is identical to the DEMH model.

The emissions of the primary pollutants (NH_3 , NO_x , SO_2 , CO , CH_4 , VOC, $\text{PM}_{2.5}$, PM_{10} and TSP) are based on a combination of the EMEP (European Monitoring and Evaluation Programme, Mareckova et al., 2008) and RCP (Representative Concentration Pathways, Lamarque et al., 2010) emission inventories. Natural emissions of NH_3 and NO_x are taken from the GEIA database (Global Emission Inventory Activity, Graedel et al., 1993), while emissions from wildfires are based on Schults et al. (2008). For the Danish area the dynamical ammonia emission model described in Sect. 2.2 is applied as well as detailed emissions from ships (Olesen et al., 2009). For Denmark emissions from road traffic (1 km × 1 km) and other emissions sources (16.67 km × 16.67 km) are based on results from the HYSCENE project (Jensen et al., 2010).

Dry deposition velocities for the gaseous compounds are based on the methodology applied in the EMEP Unified model (Simpson et al., 2003). The dry deposition

is calculated for 16 land use categories, where the surface resistance is divided into a stomatal and a nonstomatal component. For NH_3 the nonstomatal component includes, among other things, an acidity ratio between SO_2 and NH_3 (Emberson et al., 2000; Simpson et al., 2003), while a stomatal compensation point is not included. Wet deposition processes in DEHM includes in-cloud and below-cloud scavenging of particles and gases.

2.3.2 OML-DEP and the coupling to DEHM

OML-DEP is an extension of the OML model, which is a Gaussian plume model used for modelling of atmospheric dispersion on the local scale of up to ca. 20 km from point and area sources (Olesen et al., 1992). It is the recommended model to be used for environmental impact assessments when new industrial sources are planned in Denmark. OML-DEP has been developed in order to include dry deposition of ammonia on local scale applying high resolution land use data for Denmark (Nielsen et al., 2000) and the previously described dynamical ammonia emission model. This version of the model has e.g. been validated in connection with a measuring campaign around a poultry farm in Denmark, where it was shown that the model calculations clearly reflected the measured NH_3 concentration and N deposition around the farm (Sommer et al., 2009).

In DAMOS the DEHM and OML-DEP models use the same input data and parameterisations, to the extent it is possible, in order to ensure consistency. Hence, the dry deposition of NH_3 is in OML-DEP parameterised in the same way as in the regional model. Similarly the applied meteorology is the same in OML-DEP and the DEHM model in the form of hourly time series of relevant meteorological parameters extracted from the MM5v3 fields with a resolution of $5.56 \text{ km} \times 5.56 \text{ km}$ over Denmark. In OML-DEP the chemical transformation of NH_3 to NH_4^+ is calculated by a first-order reaction velocity (Asman et al., 1989), that on average reduces the NH_3 concentration with about 10% within transport distances of 8 km. The OML-DEP model uses a quadratic grid with 40×40 receptor points. In DAMOS, OML-DEP is applied for

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a 16 km × 16 km domain centred at the monitoring site or nature area in focus. This gives a grid resolution of 400 m × 400 m in the resulting concentrations and depositions, which is considered to give a reasonable coverage of the local-scale sources and the transport and transformation of NH₃ near these sources. In order to account for the medium to long-range transport of ammonia from sources outside the OML-DEP domain, time series of NH₃ concentrations are provided by the DEHM model. The hourly time series are interpolated from the DEHM grid at the upwind border of the OML-DEP domain and averaged over the boundary layer. Likewise, hourly time series of surface concentrations of SO₂ are provided by DEHM for the acidity ratio in the dry deposition formulation. Due to the low SO₂ concentrations in Denmark the ratio rarely has any impact on the depositions.

3 Results

3.1 Spatial patterns

Examples of the model results are displayed in Figs. 1 and 2 for the year 2007. The spatial patterns of NH₃ concentrations (lowest model layer) and the total N deposition across the Danish land areas estimated by the regional model are shown in Fig. 1c,d. In both maps a gradient across Denmark is seen with highest depositions in the south-western part of the country. This gradient is caused partly by atmospheric transport of mainly NO_x (and its reaction products) from Northern Europe and partly by the local ammonia emissions (Fig. 1b) that are highest in this agricultural part of Denmark. In 2007 the N load was more than 20 kg N ha⁻¹ in the southern part of Jutland, close to the German border.

The geographical distribution of the NH₃ concentration and dry deposition can be estimated in more detail with the OML-DEP model. An example is seen in Fig. 2c,d, which show the annual mean concentration and dry deposition in the 16 km × 16 km area surrounding the measuring site Tange. The spatial distribution of the NH₃ concentrations

and depositions follow the distribution of the emissions closely (Fig. 2b). The highest concentrations and depositions of more than $5 \mu\text{g NH}_3\text{-N m}^{-3}$ and up to ca. $40 \text{ kg NH}_3\text{-N ha}^{-1}$ are seen around the point sources (animal houses and manure storages), with a steep decrease in the concentration/deposition with distance from the source. Spatial variability in the deposition field is also seen due to variations in the surface characteristics (a few land-use characteristics are shown in Fig. 2a) with low depositions e.g. over the lake central in the model domain and relative higher depositions in forested areas east of the lake.

3.2 Temporal patterns

3.2.1 Inter-annual variations

In Fig. 4 the measured and modelled annual mean concentration of ammonia at the five main Danish monitoring stations are shown for the period 2005–2009. The model results from DAMOS are shown as the yearly value at the receptor point located at the monitoring station, while the results from the DEHM model are interpolated from the model grid to the location of the stations.

Lowest levels in the range $0.14\text{--}0.19 \mu\text{g NH}_3\text{-N m}^{-3}$ are measured at the island Anholt, while the highest levels of up to $1.63 \mu\text{g NH}_3\text{-N m}^{-3}$ are measured at Lindet in the southern part of Jutland, where the levels in general are high (see Fig. 1). Generally the DAMOS system (including both DEHM and OML-DEP) captures the measured concentrations better than DEHM alone. This is especially the case at the Lindet site where the yearly level is captured within 5 %, when averaged over the full period. At Anholt the levels are on average underestimated by 12 %, while the NH_3 levels are overestimated by about 19 % at the Keldsnor and Tange sites. At the Ulborg site the measured levels are overestimated on average by about 53 %. The DEHM model overestimates by 30–50 % at four of the sites and by up to 114 % at the Ulborg site. In the

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individual years the DAMOS and DEHM numbers can be in better/worse agreement with the observations. An example is 2007, where the DAMOS model captures the measured levels at Anholt, Keldsnor, Lindet and Tange within $\pm 15\%$.

DAMOS use reported NH_3 emission from 2007 to cover the calculations for the years 2007, 2008 and 2009. The differences in the model results between 2007, 2008 and 2009 are therefore driven by meteorological variations alone. At the Lindet and Ulborg sites these variations leads to approximately 7–17% higher concentrations in 2008–2009 in the models. At the other sites the variability is between –8% and 8%.

3.2.2 Intra-annual variations

The DAMOS system is also evaluated on the seasonal time-scale by using the mainly half monthly to monthly data from all the Danish sites (Fig. 5). Statistical data for the correspondence between both models and the measured time series are given in Table 3 and time series of the NH_3 concentration at four sites are shown in Fig. 6 as an example.

From this evaluation it can be seen that the overall measured pattern with a main peak in spring, a few less pronounced peaks throughout late summer and fall and low concentrations during the winter months are captured by both models. Correlation coefficients of 0.67–0.94 are obtained for the DAMOS system, while the regional model obtains correlation coefficients in the range of 0.55–0.94. As seen in Table 3 improved correlations are obtained at some stations in the DAMOS simulations compared to DEHM alone, but at other stations the opposite can be the case. A clearer picture is seen for the bias (model mean-measured mean), which for the majority of the stations is smallest with the simulations from DAMOS. From the time series in Fig. 6 it is seen that both models tends to overestimate the overall level during the spring and summer months, while the measured level is better captured by the models (especially DAMOS) during winter.

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3.3 Direct impact of local emissions

In spite of the high-resolution NH_3 emission inventory (Fig. 1b) applied for Denmark in the DEHM model, some of the heterogeneity in the emissions near a given site will be lost in the $5.56 \text{ km} \times 5.56 \text{ km}$ model simulations. This is likely part of the reason why the regional model in general overestimates the observed NH_3 level more than the high resolution DAMOS simulations. However, in order to confirm this, the distribution of the local emissions has to be viewed in detail at each site.

In order to illustrate the density of the local NH_3 emissions around the five main sites, the emission within circle areas around each site has been calculated. Based on the NH_3 inventory used in DAMOS, the emission per ha for five circle areas with radius from 500 m to about 8000 m and centred at the measuring site are displayed in Fig. 7 (the total emission is also given in Table 4). The resolution of the DAMOS model is $400 \text{ m} \times 400 \text{ m}$, so the inner circle closest to the centre illustrates the emission that directly influences the concentration in the centre. The regional model has a resolution of $5.56 \text{ km} \times 5.56 \text{ km}$ and the concentration at the measuring site is found by interpolation between the nearest grid points. The emissions directly influencing this concentration will therefore be represented by the circles with a radius between ca. 2000 m to 5000 m.

Based on the results in Fig. 7 and the results described above, the following can be seen:

- The local emissions pr ha is small within the first 1000 m from the main sites, with only a small contribution from area sources. This is in accordance with the fact that the locations of the sites have been carefully selected in order to minimize the impact from larger local sources like farms.
- The local emissions in the Anholt area are seen to be very close to zero. At the other sites we see a steep increase in the emissions pr ha from around 1000 m to 2000 m from the sites. These emissions are in DEHM instantly distributed in a grid cell of $5.56 \text{ km} \times 5.56 \text{ km}$ and a layer thickness of 25 m and will directly influence

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the concentration calculated by the DEHM model. This will lead to higher NH_3 concentrations than obtained with the DAMOS model.

- The local scale model has a more realistic dispersion both horizontally and vertically close to the sources. This is for example seen in the time series (Fig. 6) where DEHM at the Lindet and Ulborg sites especially overestimates the peaks during spring to autumn, implying a too high impact of local emissions that are highest during this time of year.

It should be noted that this analysis does not take the combined effect of location of source and wind direction into account. For example will a farm located upwind of the measuring site have more impact on the measured ammonia levels than a farm located downwind from the site.

3.4 Local emissions versus non-local emissions

In order to answer the question “why not only use a local scale model” we have utilized the DAMOS system with and without local emissions. Thereby we can identify the impact of local emissions versus non-local emissions. The local emissions are here defined as the NH_3 emissions included in a given DAMOS domain ($16 \text{ km} \times 16 \text{ km}$), while the non-local-emissions refers to all NH_3 emissions upstream from this domain that will have an impact on the NH_3 background concentration provided by the DEHM model at the boundary of the local scale model. In Fig. 8 the resulting yearly NH_3 concentrations and depositions at the five main sites are shown for the two different model runs.

As expected from the analysis in Sect. 3.3 the Anholt site is controlled by inflow from non-local sources. At the four other sites the background contributes with about 50 % of the overall NH_3 concentration/deposition. Small differences in the impacts are, however, seen as the local signal dominates at the Lindet and Tange sites, while the background signal slightly dominates at the Keldsnor and Ulborg sites. When comparing the absolute values in Fig. 8, the background level as well as the local contribution

is seen to be highest at the Lindet and Tange sites, which is in agreement with the regional scale emissions (Fig. 1) and the local emissions (Fig. 7).

These model results can also be used to estimate the fraction of locally emitted NH_3 that is dry deposited locally within the model domain. In Table 4 the yearly dry deposition is calculated for each site as a sum over all the grid cells in a given $16 \text{ km} \times 16 \text{ km}$ domain. Also the total annual emissions within the domains are given. From these two set of numbers we can calculate that about 14 % to 27 % of the locally (that is within a radius of about 8 km) emitted NH_3 is also deposited locally at the five sites in focus here.

4 Discussion

Comparing the observed and calculated concentrations of ammonia at a number of sites in Denmark shows that our overall initial hypothesis holds: the annual to monthly ammonia concentrations calculated with the DAMOS system, which couples regional and local scale models, generally matched the observed levels better than values based on the regional model alone. However, both models tend to overestimate the observed levels. The improved performance of the DAMOS system was most pronounced at sites located in agricultural regions. Here the regional model tends to overestimate the NH_3 levels as the direct impact of local emissions becomes too high when applying a $5.56 \text{ km} \times 5.56 \text{ km}$ grid resolution in the model. However, by coupling the regional model to the local-scale model, the emissions pattern was resolved in greater detail and the resulting concentrations matched the levels better (the bias is smaller at a majority of the sites). Similarly, Fournier et al. (2002) concluded that a $5 \text{ km} \times 5 \text{ km}$ resolution model for the British Isles did not fully describe the local variability observed. Dragosits et al. (2002) extended the study and applied a local scale atmospheric dispersion model for NH_3 at a higher spatial resolution ($50 \text{ m} \times 50 \text{ m}$). They concluded again that the local variability is high and that the national assessments at the 5 km grid in the UK will give an incomplete picture of the occurrence of critical load exceedances.

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In relation to dry deposition and exceedances it is important to resolve the surface roughness. The coupled system therefore also improved the result through its higher resolution of the surface roughness (land-cover) as seen in the example of the estimated dry deposition of ammonia in the OML-DEP domain centred at Tange (Fig. 2). Here, the high deposition areas close to the sources are clearly seen, but higher depositions can also be seen in the regions with forests (with high roughness).

At sites like Keldsnor, Anholt and Raabjerg Mose located in areas with fewer local sources the improvements by using the DAMOS system are less obvious. These sites are mainly influenced by regional to long-range transport of ammonia. In this case, the regional model captured the observed level and the background level from DEHM dominated the signal from the coupled model. So, both the local-scale and regional-scale model are important for the results, while the relative importance of the local-scale versus the long-range contribution will vary depending on the emission density of a given area as well as on meteorological conditions (e.g. dominating wind speed/direction).

The pronounced intra-annual cycle of the ammonia concentration across Denmark is generally captured by the DAMOS system. Previous sensitivity studies with the DEHM model, has shown that this variability is captured due to the detailed description of the spatial and temporal variation in the NH₃ emissions (Skjøth et al., 2011). So far the necessary high resolution emission data and detailed information on agricultural praxis are still only available in few European countries; effort is currently put into extending the data coverage (see e.g., Reis et al., 2012, with focus on the UK). Modelling of the seasonal variability has importance as the dry deposition is calculated as a product of the concentration and a parameterized deposition velocity, which during Danish conditions has a seasonal variability due to a strong influence from humidity. Additional, a future implementation of compensation points in the models gives a strong need for as good concentration estimations as possible. In relation to effects of nitrogen load/level to sensitive nature areas this variability might be important to capture by models, as the plants sensitivity to the nitrogen load/level might differ throughout the year (Sheppard et al., 2009).

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In relation to regulation of ammonia emissions the range of influence of emitted NH_3 is highly relevant and has been studied e.g. by the use of various types of models. As previously mentioned, a review of a number of European models, state that the fraction of NH_3 that is recaptured locally is in the range of 2–60 % (Loubet et al., 2009). With the DAMOS model we estimate that the fraction of locally emitted NH_3 depositing locally is 14–27 % at five Danish locations. In a recent study for the US, Dennis et al. (2010), uses the CMAQ model to estimate that about 8–15 % of the NH_3 emitted within a 12 km grid cell is also deposited in the same cell. Model results by Kryza et al. (2011) covering Poland with at 5 km × 5 km and the UK with 5 km and 1 km (Hallsworth et al., 2010) suggest that both large areas as well as smaller hot-spots can have exceedances of critical loads due to intense agricultural production. These studies suggest that near strong emission sources a higher spatial resolution than 5 km is needed for a meaningful assessment of model based critical loads to sensitive ecosystems. The studies by Kryza et al. (2011), however, also suggest that high spatial resolution modelling with transport models only makes sense, if the this modelling is accompanied with equally high resolution in the emission inventories.

These results hence all confirm that NH_3 emissions should not only be viewed and treated as a local problem, but also as an issue on the regional scale. As previously mentioned part of the emitted NH_3 will in the atmosphere be transformed into aerosols containing ammonium (NH_4^+). As the main removal path of these aerosols is wet deposition they can be transported over long distances (>1000 km) before deposited (Hertel et al., 2006), which also supports that NH_3 should be viewed in regional perspective. However, some of the problems in relation to excess nitrogen from agriculture can only be meaningfully solved by taken local variations into account. The studies presented here shows that high resolution calculations that takes both regional and local scale variations into account are technically possible without to large computational cost and that such calculations makes scientifically sense, when high quality emission data are available.

It has long been acknowledged that the flux of ammonia can be bi-directional in agricultural areas (Farquhar et al., 1980). Like many other regional models (Massad et al., 2010) the parameterisation of dry deposition in DAMOS is based on the resistance method and the stomatal compensation point for NH_3 . Hence, a possible a bi-directional flux is not directly accounted for. However, the effect of bi-directional fluxes is indirectly accounted for in the calculations of the Danish ammonia emission inventory as it takes into account the emission from grown crops due to bi-directional fluxes (Gyldenkærne et al., 2005).

Bi-directional flux can also take place over marine areas, where a previous preliminary study points towards a redistribution of the deposition in the coastal area and this redistribution tends to reduce the gradients in NH_3 deposition (Sorensen et al., 2003). Recent studies with the regional CTM model LOTOS-EUROS includes a new flux parameterisation accounting for bi-directional flux in both terrestrial and marine areas confirm these findings (Wichink Kruit et al., 2010; Van Zanten et al., 2010). The DAMOS system is currently not taking this into account for marine areas.

In general the descriptions of bi-directional flux have mainly been used at field scale and input parameters (e.g. stomatal compensation points for various canopy types) needed for more general use at for example the regional scale have not been available (Massad et al., 2010; Zhang et al., 2010). Recently more generalised parameterisations have been proposed (e.g., Wichink Kruit et al., 2010) and we are currently working on an updated version of the DAMOS system, where parts of the parameterisation developed by Massad et al. (2010) will be included to more explicitly account for the bi-directional flux of NH_3 over terrestrial ecosystems.

In this study we have only validated the model system against concentrations of ammonia. So far only very few data on deposition rates appropriate for model validation are available, as measuring dry deposition of ammonia is very complex and uncertain (Hertel et al., 2011). On way to test if the models included in DAMOS covers the important process for ammonia deposition is to compare the model results with the result of other models. The OML-DEP has been a part of an inter-comparison exercise with

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other local-scale models (ADMS 4, AERMOD, LADD and OPS-st) against measured concentrations around a pig farm and performed the best (Theobald et al., 2009).

The reason for the general overestimation by the models is not yet fully explored, but may be related to the following possible issues:

- an overestimation of the national ammonia emissions,
- the resolution of the local scale model is too coarse leading to a direct impact of emissions in the 400 m × 400 m grid cell including the measurement site,
- the parameterisation of the emission does not reflect the true diurnal cycle, but e.g. overestimates the night-time emission, which combined with typical limited night-time atmospheric mixing leads to an up-concentration of ammonia near the surface,
- an underestimation of the chemical conversion from ammonia to ammonium, which is directly linked to the content of acid components in the ambient air or the parameterisation of the exchange between gas phase and aerosol component,
- an underestimation of the vertical dispersion leading to overestimation of the concentrations close to the source,
- the background concentration used as input on the boundaries of the local scale model is an average for the full boundary layer in DEHM. This means that in agricultural/emission areas the concentration near the surface is underestimated close to the borders of the local scale model compared to real concentrations. Thereby the deposition of NH₃ also will be underestimated near the border and too much NH₃ is transported across the domain contributing to an overestimation of the concentration at the measurement sites,

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- the models does not take an increased deposition at the edges of woods into account. This might lead to an underestimation of the deposition and hence an overestimation of the concentration.

An underestimation of the chemical conversion is likely one of the main issues. When looking at the other nitrogen components it is for example seen that the DEHM results show a tendency for underestimating the ammonium concentrations at the Danish sites, while the sum of NH_3 and NH_4 (NH_x) is well captured. This could be linked to a general underestimation of sulphate in the model.

We are currently working on a modified version of the local-scale model OML-DEP, where the background concentration from DEHM is included as a profile instead of an average for the full boundary layer in DEHM. This newer version of the model will hopefully also be able to run with a higher resolution (100 m × 100 m) and the same domain size of 16 km × 16 km. Thereby the impact of emissions within the grid cell including the measuring site can be handled more realistically and it will generally lead to a somewhat smaller impact of local emissions.

5 Applications of the system

The DAMOS system is applied on a yearly basis at a number of selected nature areas as part of the Danish Background Air Quality Monitoring Program. The system has also been included in assessments carried out for a few Environment Centres in Denmark as the basis for their management of the Danish NATURA 2000 locations. As part of the Danish implementation of the Habitat and Bird Directives, a management plan for each of the ca. 250 2000 sites has been made (Dinesen and Bjerregaard, 2011). For such assessments the system is used to map the N deposition to selected nature areas, estimate impact of local vs. non-local emissions and to compare the local N load with critical load values (Frohn et al., 2008, 2010; Geels et al., 2008; Hertel et al., 2012). The Centres have access to maps similar to the maps in Fig. 2c,d. in order

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to make priorities for future regulations and actions in order to protect specific nature areas. The total N load is estimated as the sum of NH_3 deposition from DAMOS and wet deposition (of NH_3 , NH_4 and NO_3) and dry deposition of NH_4 and NO_3 as extracted from the regional model. In Fig. 9 the simulated total N load at 26 specific nature areas in the Aarhus region in Denmark are displayed together with the critical loads related to the general nature types within each nature area. At 13 areas the upper limit of the critical load is seen to be exceeded and for 9 of these sites the DAMOS results based on different scenarios showed that it is not possible to get below this upper limit even if the local sources are reduced. At none of the sites the estimated N deposition was below the critical load. These assessments have been described in more detail in Hertel et al. (2012).

The described assessments are based on model simulations for a specific year. However, the current analysis of the inter-annual variations (Sect. 3.2.1) show that year-to-year variation in the meteorology alone can lead to variations in the annual nitrogen load on the order of approximately 10–20%. Such variations should be taken into account when the estimated nitrogen loads are used for management. Alternatively, the estimated nitrogen load should be based on model simulations covering several years.

The model results are extracted for the land-use category corresponding to the specific nature areas, which gives a more precise estimate of the total N load. For example at a specific coniferous forest (see Table 5) the total N load would in the DAMOS system be estimated about 22 kg N ha^{-1} . The regional model estimated a load of ca. 29 kg N ha^{-1} to the forest, while a load of ca. 17 kg N ha^{-1} is estimated for an average over all the nature types within the $5.56 \text{ km} \times 5.56 \text{ km}$ grid cell including the specific coniferous forest. Wet deposition of NH_x and NO_y (Table 5) are of courses the same in the three estimates, as this is not dependent on the land-use type. However, differences are seen for the calculated dry deposition. When extracting dry deposition to the forest, the regional model alone gives a very high estimate for NH_3 . This is mainly because the heterogeneity in the emissions is not resolved in the regional

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model. However, when averaging over all the land-use types in the grid cell, lower values of dry deposited NH_x as well as NO_y are obtained. This example emphasize the importance of using a model system that covers both local to regional scale as well as a system that includes deposition to different land-use types of both NH_3 and the other nitrogen components. Even in an agricultural country like Denmark, the other nitrogen components contribute significantly to the total N load at a given site.

6 Summary and conclusions

The main goal of this research has been to develop a coupled model system in order to improve the modelling of atmospheric ammonia in a landscape with a mix of agricultural and natural/semi-natural areas. The model system has been validated against measured NH_3 concentrations from 21 sites in Denmark. Our findings that are summarized in the following can be used to bring environmental management a step forward:

- The results show that the regional model DEHM overestimate observed local ammonia concentrations across Denmark due to the fact that local agricultural emissions are distributed over grid cells of $5.56 \text{ km} \times 5.56 \text{ km}$.
- Coupling the regional model DEHM with the local-scale model OML-DEP, as it is done in DAMOS, improves the results.
- A detailed spatial and temporal description of NH_3 emissions is crucial for the improved modelling of ammonia also at the intra-annual timescales.
- Detailed land-use information is also important for precise estimates of the deposition to specific nature areas.
- The fraction of locally emitted NH_3 depositing locally (here in a $16 \text{ km} \times 16 \text{ km}$ domain) is on the order of 15 % to 30 % and close to intense agricultural sources these may alone exceed critical loads of local ecosystems.

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– For Danish areas the NH_3 contribution from non-local NH_3 sources are on the order of 50%. However, when including wet deposition and dry deposition of other N components, the non-local sources dominate the local signal and in total exceed the critical loads of many sensitive ecosystems. The local contributions are hence often moderate.

– A background concentration reflecting the non-local emissions within the region of a specific area is hence important when making assessments of the total nitrogen load. This is obtained with a model system like DAMOS where models covering both the regional to local scale are applied in combination with detailed surface information (emissions and land-cover).

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Table 1. Details about the monitoring sites applied in the current study: location, land-use, measuring period and time resolution and measuring method. The geographical positions are shown in Fig. 1a.

Station	Land-use	Measuring period*	Time resolution	Measuring method
Anholt	Grass, coastel	2005–2009	Daily	Filter pack
		2007–2009	Half monthly	Denuder
Keldsnor	Grass, coastel	2005–2009	Daily	Filter pack
		2005–2009	Half monthly	Denuder
Lindet	Forest	2005–2009	Daily	Filter pack
		2005–2009	Weekly	Denuder
Tange	Grass	2005–2009	Daily	Filter pack
Ulborg	Forest	2005–2009	Daily	Filter pack
		2005–2009	Half monthly	Denuder
Hjelm Hede	Heath land	2005–2006	Half monthly	Denuder
Idom Hede	Heath land	2005–2009	Half monthly	Denuder/ALPHA
Husby	Heath land, coast	2006–2007	Half monthly and winter time monthly	ALPHA
Lønborg Hede	Heath land	2006–2007	Half monthly and winter time monthly	ALPHA
Ovstrup Hede	Heath land	2006–2007	Half monthly and winter time monthly	ALPHA
Bisgyde	Dry grassland	2007–2009	Monthly	ALPHA
Diesbjerg	Heath land	2007–2009	Monthly	ALPHA
Hammer Bakker	Heath land	2007–2009	Monthly	ALPHA
Helm Polde	Heath land	2007–2008	Monthly	ALPHA
Holmkær	Meadow	2007–2008	Monthly	ALPHA
Nybo Mose	Raised bog	2007–2009	Monthly	ALPHA
Randbøl Hede	Heath land	2007–2009	Monthly	ALPHA
Råbjerg Mose	Raised bog	2007–2009	Monthly	ALPHA
Storelung	Raised bog	2007–2009	Monthly	ALPHA
Ulvholm	Rich fen	2007–2009	Monthly	ALPHA
Ulvshale	Heath land	2007–2008	Monthly	ALPHA

* The measuring period used in this study. At some of the sites the measuring period is longer (see <http://www.dmu.dk/en/Air/Monitoring/Programmes/>).

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Table 2. A short description of the 16 different functions describing the temporal variation in NH_3 emissions from various activities (like Table 1 in Skjøth et al., 2011).

Function	Description
Fkt 1	Animal houses with forced ventilation
Fkt 2	Open animal houses (non-forced ventilation)
Fkt 3	Manure storages
Fkt 4	Winter crops (no emission simulated in this study)
Fkt 5	Spring crops (no emission simulated in this study)
Fkt 6	Late spring crops (no emission simulated in this study)
Fkt 7	Grass
Fkt 8	Spring application of manure on bare soil
Fkt 9	Application of manure on crops
Fkt 10	Summer application of manure
Fkt 11	Autumn application of manure
Fkt 12	Spring application of fertilizer (90 % of all fertilizer)
Fkt 13	Summer application of fertilizer (10 % of all fertilizer)
Fkt 14	Emission related to grassing cattle
Fkt 15	Emissions related to ammonia treated straw
Fkt 16	Emissions related to personal vehicles with catalytic converters

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Table 4. The modelled annual NH_3 deposition within the OML-DEP domain related to the local emissions alone. Given as both $\text{kg NH}_3\text{-N ha}^{-1} \text{ yr}^{-1}$ and as % of the NH_3 emitted from local sources.

Station	Deposition due to local emissions	Local emissions	Local emissions dep. in domain
Unit	$\text{kg NH}_3\text{-N ha}^{-1} \text{ yr}^{-1}$	$\text{kg NH}_3\text{-N ha}^{-1} \text{ yr}^{-1}$	%
Anholt	6.4×10^{-4}	2.4×10^{-3}	27
Keldsnor	4.0×10^{-1}	2.1	19
Lindet	2.5	17.4	14
Tange	2.3	15.1	15
Ulborg	1.6	11.4	14

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Table 5. An example of the modelled annual deposition (dry and wet) of NH_x and NO_y to a coniferous forest in Denmark. The first row presents the deposition estimated by the DAMOS system. The next two rows show the estimates from the DEHM model alone, when extracted for a coniferous forest or as mean over all land-use types in the specific grid cell. Note that the differences between the numbers will depend on e.g. the relative occurrence of different land-use types in the specific grid cell.

	Dry NH_x ($\text{kg N ha}^{-1} \text{ yr}^{-1}$)	Dry NO_y ($\text{kg N ha}^{-1} \text{ yr}^{-1}$)	Wet NH_x ($\text{kg N ha}^{-1} \text{ yr}^{-1}$)	Wet NO_y ($\text{kg N ha}^{-1} \text{ yr}^{-1}$)	Total N dep ($\text{kg N ha}^{-1} \text{ yr}^{-1}$)
DAMOS to con. forest	6.3	7.3	4.2	3.8	21.5
DEHM to con. forest	13.8	7.3	4.2	3.8	29.0
DEHM to mean grid	6.0	3.0	4.2	3.8	17.0

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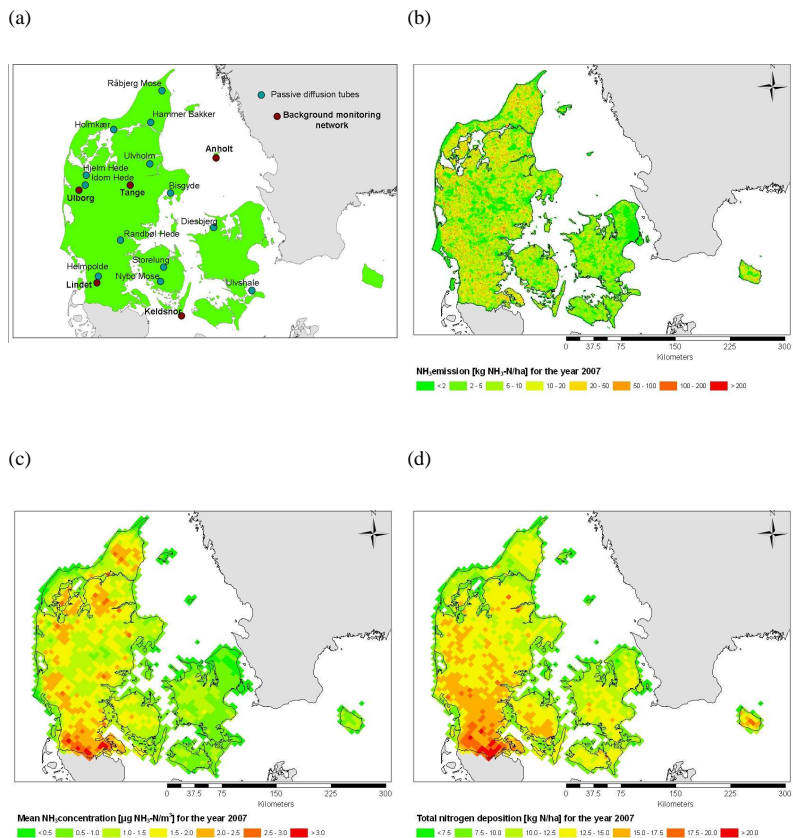


Fig. 1. (a) The location of the measuring sites included in this study. (b) An example of the distribution of NH₃ emissions across Denmark on a 1 km × 1 km grid for the year 2007. (c) The yearly averaged NH₃ concentration over Denmark as calculated by the DEHM model for the year 2007. (d) The yearly total nitrogen load (kg N ha⁻¹) over Denmark as calculated by the DEHM model for the year 2007.

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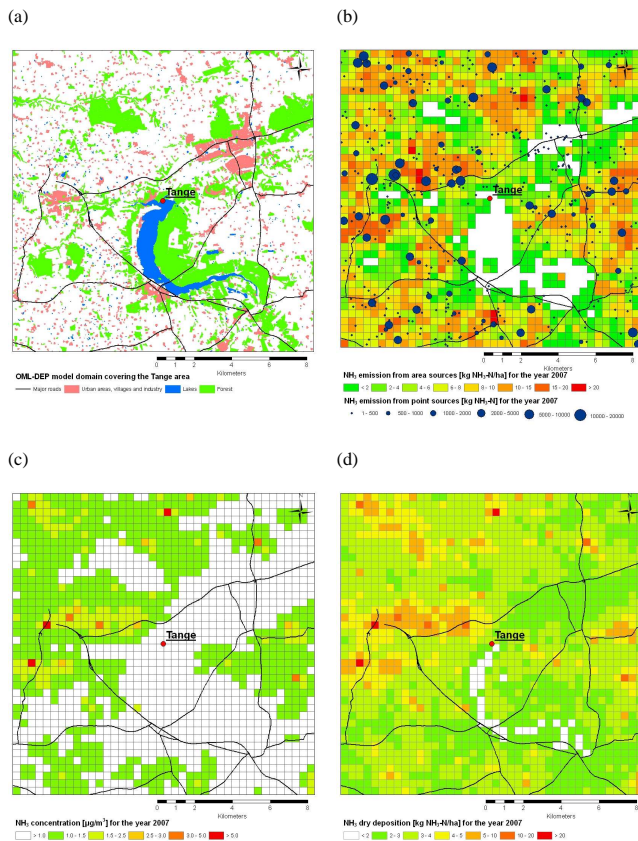


Fig. 2. (a) The OML-DEP 16 km \times 16 km model domain centered around the Tange site. The resolution in the local-scale model is 400 m \times 400 m. (b) The applied NH₃ emissions divided in to area and point sources for the year 2007. (c) The yearly averaged NH₃ concentration as calculated by the DAMOS system for the year 2007. (d) The yearly total dry deposition of NH₃ as calculated by the DAMOS for the year 2007.

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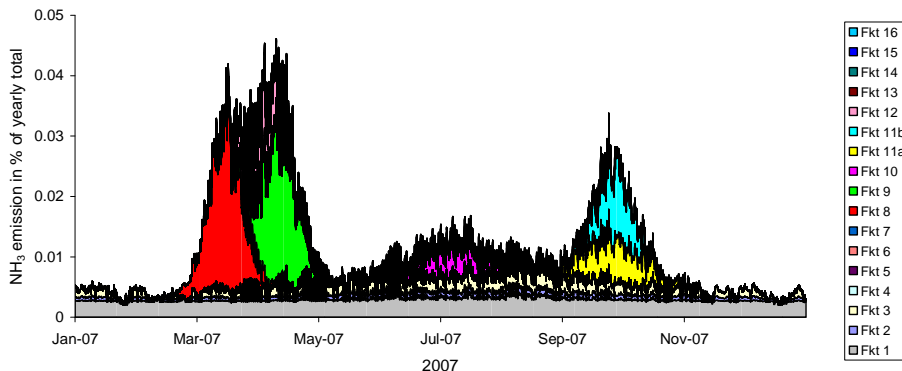


Fig. 3. Seasonal variations of the hourly NH_3 emissions separated in to the various agricultural emission categories (described in Table 2) in the applied emission parameterisation. The emissions are shown as percentage of the yearly sum of all the 16 categories for the Tange site in the year 2007.

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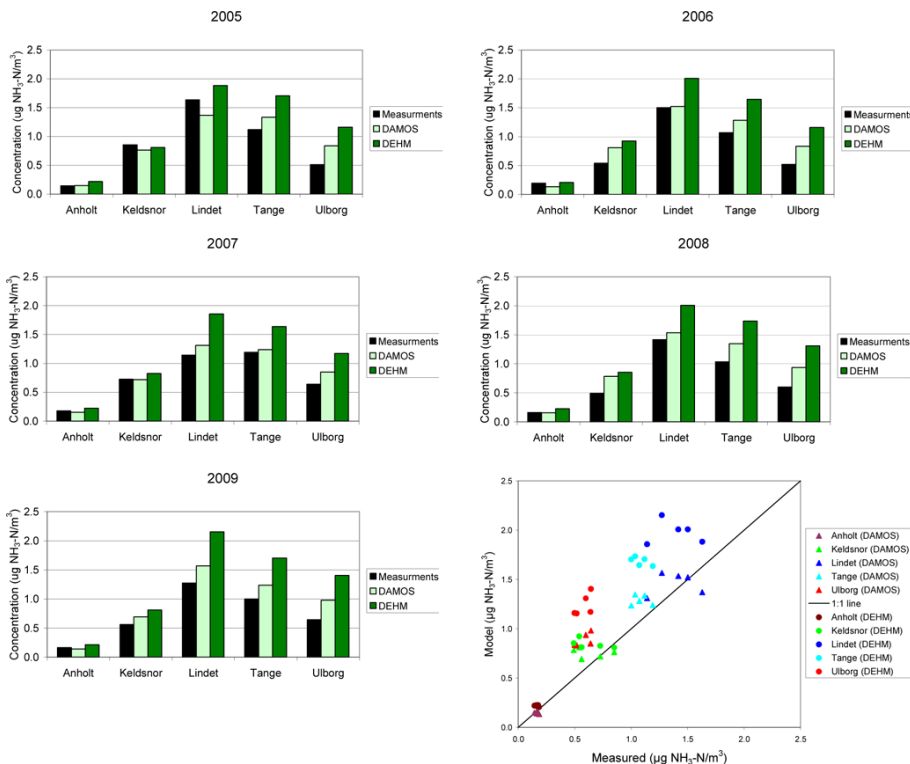


Fig. 4. Measured and modelled yearly NH_3 concentrations at the five main monitoring sites as a column diagram for each of the years 2005–2009 and collected in a scatter diagram.

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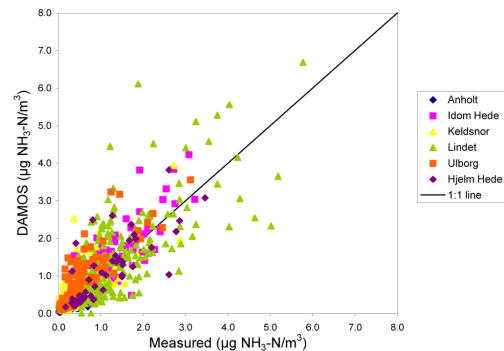
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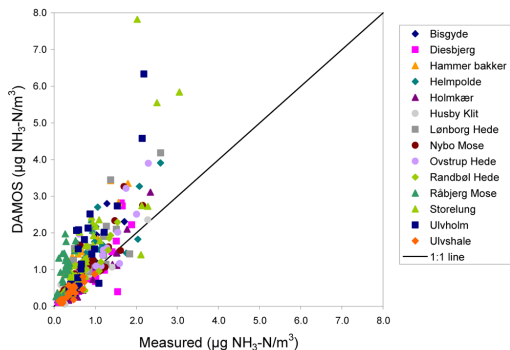
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(a)



(b)

Fig. 5. Measured and modelled monthly or 1/2 monthly concentrations at the Danish sites **(a)** where the denuder method has been applied and **(b)** where the passive diffusion tube samplers ALPHA have been applied (see also Table 1 for more details about each site).

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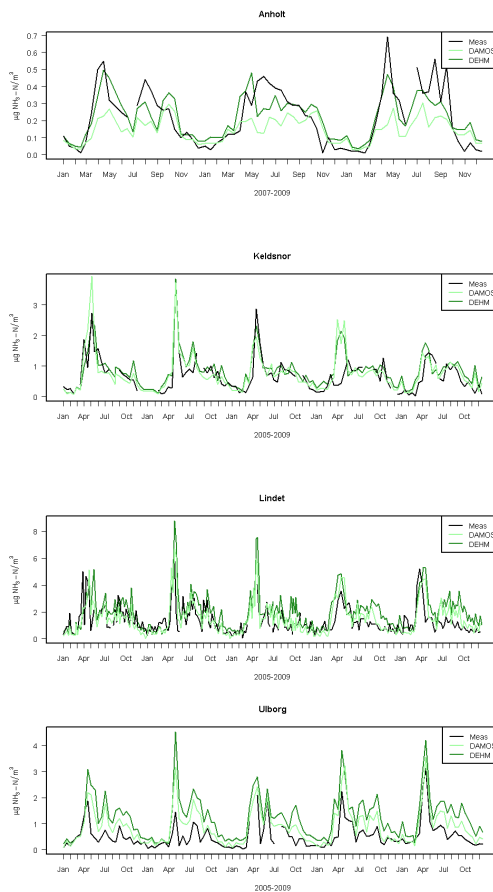


Fig. 6. Time series of measured and modelled NH_3 concentrations at four sites in Denmark covering the period 2005–2009 or 2007–2009. At the Lindet site mean weekly samples are shown, while at Anholt, Keldsnor and Ulborg 1/2 monthly samples are displayed.

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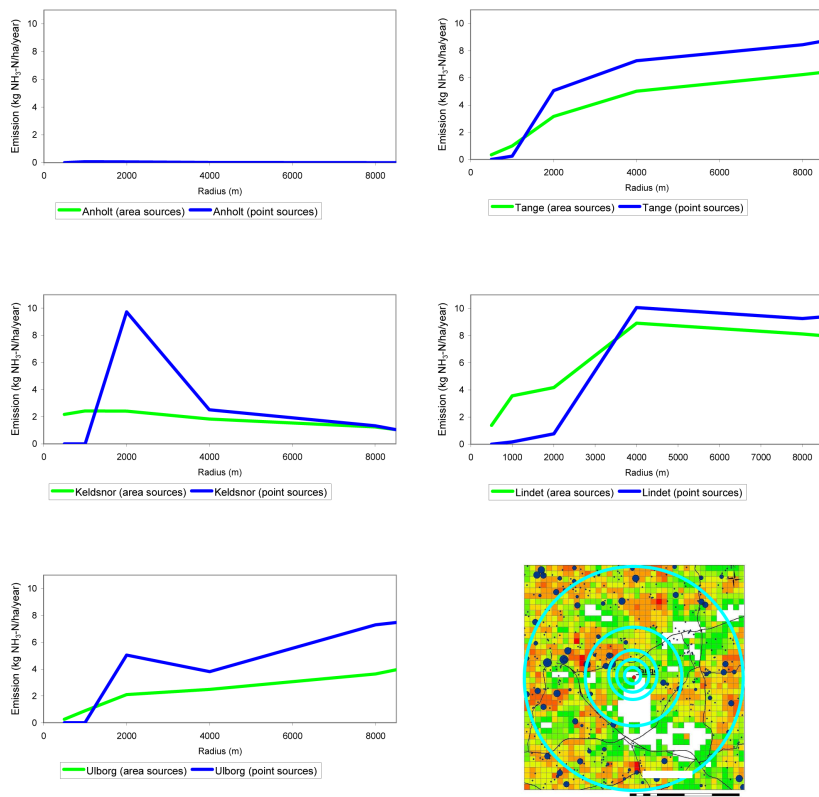


Fig. 7. The average emission of NH_3 from area and point sources within different circle areas around each of the five main monitoring sites. Based on the emissions used in DAMOS for the year 2007. The radius of the circle areas (centred around the sites) are given on the x-axis and the emissions are normalized by the circle area. Note that the emissions give at 8500 m are the sum over the full domain. The location of the circle areas are shown together with the NH_3 emissions for the domain at Tange as an illustration (lower right plot).

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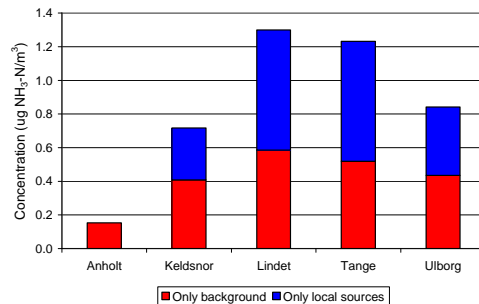
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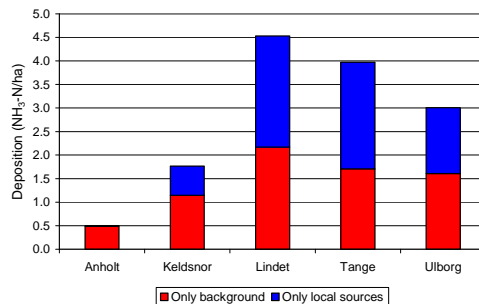
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(a)



(b)

Fig. 8. The modelled NH₃ concentration **(a)** and deposition **(b)** at the five main sites separated into “only background” and “only local sources” for the year 2007. The “background” refers to the part coming from non-local sources i.e. due to NH₃ transported into the OML-DEP domain from upwind sources in the DEHM model domain. The “local sources” refers to the part coming from the local NH₃ emissions within the OML-DEP domain. The size of the local emissions can be seen in Fig. 7.

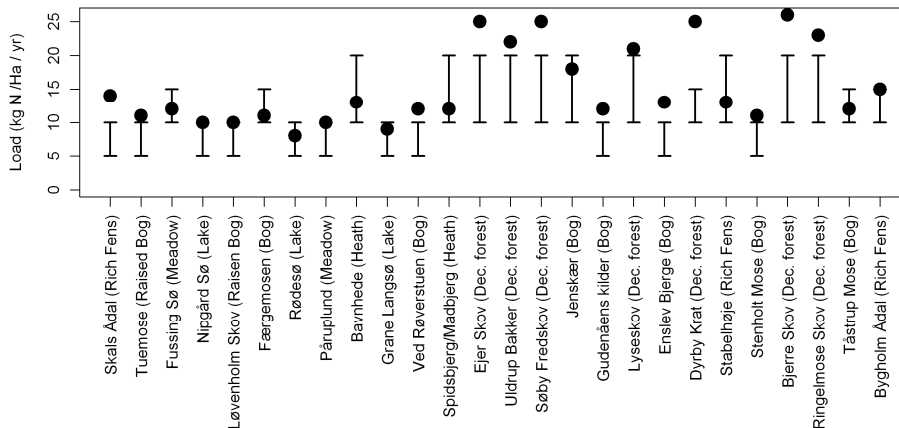


Fig. 9. The estimated N load (dots) to specific nature areas in the Aarhus region in Denmark, displayed in relation to the critical load levels (bars) corresponding to the dominating nature types. This figure is also shown in Hertel et al. (2012).

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