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Maciej KRYZA[1*], Marek BŁAŚ[1], Anthony J. DORE[2] and Mieczysław SOBIK[1]

NATIONAL SCALE MODELLING OF THE CONCENTRATION AND DEPOSITION OF REDUCED NITROGEN AND ITS APPLICATION TO POLAND

MODELOWANIE STĘŻEŃ I DEPOZYCJI AZOTU ZREDUKOWANEGO I PRZYKŁADY ZASTOSOWAŃ DLA POLSKI

Abstract: The relative contribution of reduced nitrogen in acid and eutrophic deposition in Europe has been recently increasing as a result of SO_2 and NO_x emissions abatements. For Poland, the main source of spatial information on dry and wet deposition of NH_x is the EMEP model with a coarse 50 km x 50 km grid which may be insufficient for national scale studies, as the NH_x emissions, concentrations and depositions vary considerably over a short distance. The FRAME model is used to calculate the spatial patterns of annual average NH_x air concentrations and depositions with a 5 km x 5 km grid. The results correlate well with available measurement and with spatial patterns of concentrations and depositions of NH_x reported with the EMEP, but show higher spatial variability. The differences in deposition budgets calculated with FRAME and EMEP are less than 17% for wet and 6% for dry deposition. The differences between FRAME and the Polish Chief Inspectorate of Environmental Protection interpolation based wet deposition budget is 3%. Up to 93% of dry and 53% of wet deposition of NH_x comes from national activities. The western part of Poland and the mountains in the south are strongly influenced by the NH_x deposition from transboundary transport.

Keywords: ammonia, reduced nitrogen, deposition, FRAME, Poland

Introduction

Emissions of reduced nitrogen in Poland have fallen by 41% since 1985, compared with a 72% reduction of SO_2 and 46% reduction of NO_x emissions [1]. While SO_2 and NO_x emissions still show a downward trend, the NH_3 emission level has stabilized at about 320 Gg since the year 2000. The substantial reduction of SO_2 and NO_x emissions is a result of the successful application of abatement strategies in Poland [2]. The reduction of emissions resulted not only in decreased nitrogen and acid deposition but also the relative

[1] Department of Climatology and Atmosphere Protection, Institute of Geography and Regional Development, Wrocław University, ul. Kosiby 6/8, 51-670 Wrocław, tel. 71 348 54 41, fax 71 372 94 98
[2] Centre for Ecology and Hydrology, Edinburgh, Bush Estate, Penicuik, Midlothian, EH26 0QB UK, tel. +44 (0)131 4454343, fax +44 (0)131 4453943, email: todo@ceh.ac.uk
[*] Corresponding author: kryzam@meteo.uni.wroc.pl

contribution of chemical species in acid and eutrophic deposition has changed, with the NH_x deposition gaining in importance [3, 4].

Reduced nitrogen air concentration and deposition may have a number of environmental impacts, including soil acidification, eutrohpication of seminatural ecosystems, biodiversity decrease and formation of fine particulate matter in the atmosphere [5, 6]. Therefore detailed information on reduced nitrogen air concentration and deposition are necessary. Until recently, the main source of spatial information on atmospheric pollutants depositions and concentrations in Poland was the Unified EMEP model [7, 8], which works on a European scale with a 50 km x 50 km grid size. These data were used for the assessment of the critical loads exceedances and the environmental quality targets of the NEC Directive in Poland [2, 9]. The coarse resolution of the EMEP model is often insufficient for regional scale analysis, where detailed spatial information on atmospheric concentrations and deposition are necessary eg to calculate the critical levels and loads exceedances [10, 11]. This is especially important for reduced nitrogen, because of the very high spatial variation of its emission sources, concentrations and deposition [12, 13].

This study presents the results of applying a regional scale model, FRAME, to calculate spatial patterns of NH_3 and NH_4^+ air concentrations and deposition of reduced nitrogen for Poland. The FRAME model and input data are described and modelled results of annual average concentrations and annual depositions of reduced nitrogen are presented and compared with the available measurements. The FRAME deposition budgets for Poland are calculated and compared with EMEP data and CIEP (*Chief Inspectorate of Environmental Protection*) measurement-based estimates. The results of source-receptor analysis are also presented, to assess the fraction of reduced nitrogen deposition from national sources and the transboundary contribution.

Data and methods

Description of the FRAME model

A detailed description of the FRAME model can be found in [14-16]. The model was developed from an earlier European scale model, TERN (*Transport over Europe of Reduced Nitrogen* [17]).

The FRAME model is a statistical Lagrangian atmospheric transport model used to assess the annual mean air concentration and deposition of atmospheric pollutants. FRAME simulates an air column moving along straight-line trajectories. Trajectories are run at a 1° resolution for all grid squares at the edge of the model domain. The air column advection speed and frequency for a given wind direction is statistically derived from radio-sonde measurements [18]. The adoption of straight line trajectories was found to be successful in reproducing annual average measurements of gas and aerosol concentrations in air and wet deposition in the UK [15, 19-21].

The atmosphere is divided into 33 separate vertical layers extending from the ground to an altitude of 2500 m. Layer thicknesses vary from 1 m at the surface to 100 m at the top of the mixing layer. The high vertical resolution of the model makes it especially useful for reduced nitrogen assessment, as the anthropogenic emission sources of NH_3 are usually located near the ground surface. In fact, FRAME was originally developed as the *Fine*

Resolution AMmonia Exchange model and was used to simulate transport and deposition of reduced nitrogen over the UK [14].

Vertical diffusion in the air column is calculated using K-theory eddy diffusivity and solved with the Finite Volume Method. Point source emissions are treated individually with a plume rise model. Additional information on stack height, temperature and velocity of the outflow gases are used to calculate an effective emissions height [22]. The plume reaches its maximum height when temperature is equal to the surrounding environment and its momentum is dissipated. Buoyancy forces dominate the plume rise, which is parameterized separately for stable conditions and for neutral and unstable conditions according to the Pasquill-Gifford stability classes. The depth of the boundary layer in FRAME is calculated using a mixed boundary layer model with constant potential temperature capped by an inversion layer with a discontinuity in potential temperature. Solar irradiance is calculated as a function of latitude, time of the year and time of the day. Initial gas and aerosol concentrations at the edge of the model domain are calculated with FRAME-Europe, a European scale model working with a 50 km x 50 km resolution. Trajectories in FRAME-Europe were initialised with global background concentrations of gases and aerosol based on measurements from remote sites. The model was run over the EMEP 50 km x 50 km domain and the directionally dependent gas and aerosol concentrations output to datafiles which, after performing a GIS grid transformation from the EMEP to the Polish national grid, were used to initialise concentrations in a FRAME Poland simulation.

FRAME assumes constant NH_3 emissions over the year. Recent studies have shown however that the application of the seasonally varying emissions of ammonia is of certain importance, as a result of eg increased volatilisation due to high temperature in the summer, manure spreading etc [12, 23]. Therefore development of the seasonal version of the FRAME model is needed and will be undertaken in the future.

The chemical scheme used in FRAME is similar to the one employed in the EMEP Lagrangian model [24]. The model chemistry includes gas phase and aqueous phase reactions of oxidized sulphur and oxidized nitrogen and conversion of NH_3 to ammonium sulphate and ammonium nitrate aerosol. The prognostic chemical variables calculated in FRAME are: NH_3, NO, NO_2, HNO_3, PAN, SO_2, H_2SO_4, as well as NH_4^+, NO_3^- and SO_4^{2-} aerosol.

Dry deposition velocities of SO_2, NO_2 and NH_3 are ecosystem specific and are calculated individually to five different land cover categories (forest, grassland, moorland, urban and arable). For ammonia, the deposition velocity is generated from the sum of the aerodynamic resistance, the laminar boundary layer resistance and the surface resistance [14]. Information on wind speed needed for calculation of dry deposition is derived from climatological stations in Poland.

The model employs a constant drizzle approach using precipitation rates calculated from a climatological map of annual precipitation for Poland [25]. Wet deposition of chemical species is calculated using scavenging coefficients based on those applied in the EMEP model. An enhanced washout rate is assumed over mountainous areas due to the scavenging of cloud droplets by the seeder-feeder effect to calculate local scale orographic enhancement of precipitation and concentration [26]. The washout rate for the orographic component of rainfall is assumed to be twice that calculated for the non-orographic component. The factor of two for orographic enhancement is supported by the measurements performed in Poland by [26, 27]. These measurements showed elevated concentrations of dissolved ions in rain water in hill areas due to the scavenging of polluted hill cloud droplets by precipitation.

The model code is written in High Performance Fortran 90 and executed in parallel on a Linux Beowulf cluster comprising of 60 dual processors. The run time for a simulation employing 100 processors to calculate average annual concentration and deposition is 25 minutes.

Emission data

Total emission of NH_3 from Poland was estimated to be 325.0 Gg in the year 2002, which is the year of interest in this paper. The national total emissions are given here according to the national emission inventory [1] and the year 2002 is chosen as a reference because the detailed census data, necessary to estimate spatial patterns of emissions, are available for this year, therefore reducing uncertainties.

Agriculture contributed 96.5% (313.8 Gg of NH_3) of the total emission of NH_3 in Poland in 2002, the remainder came from waste treatment (2.5%) and production processes (1.0%). The national totals of NH_3 emissions from other sources, discussed by [28], including catalytic converters are not available. There are also large discrepancies in estimation of total agricultural emission of ammonia in Poland. This stems from different emissions factors that are in use. Recently [29] it was suggested that emission factors for Poland are generally lower than those that are currently in use for Western Europe, and which are applied by [1]. This is because of differences in agricultural practice. The total ammonia emission from agriculture, estimated by [29], is about 10% lower than calculated by [1] with emission factors suitable for Western Europe. The differences in the estimated emission from fertilizer application are even higher and exceed 30% which means that the spatial patterns of emissions from various agricultural sources can differ significantly if different emission factors are used. In this paper, the emission factors proposed by [1] are applied, as they are also used in official national reports, including EMEP reports. Because the FRAME model results are compared here with the EMEP estimates, the application of the emission factors used by [1] is justified.

Spatial patterns of NH_3 agricultural emission from animal breeding and fertilizer application for Poland were prepared with a 5 km x 5 km spatial resolution (Fig. 1) using the methodology similar to that proposed by [30]. Some modifications were necessary as the input data for spatial estimation of emissions were different from those used by [30]. Spatial data on the animal number and fertilizer consumption, provided by the [31], were combined with the emission factors [1] and Corine Land Cover data [32] to calculate the spatial patterns of yearly NH_3 emission from different agricultural sources for the area of Poland with 5 km x 5 km grid size. The data on animal numbers were available at commune level (average size of 126 km^2), while fertilizer consumption was only obtainable at province level. This should be considered as a drawback as the provinces are recognizable on the emission maps, but no other data on fertilizer application were available (Fig. 1). The emissions are gridded separately for cattle, pigs, poultry, sheep, horses and fertilizer consumption and mixed into the model lowest surface layers with a source-dependent emissions height. The rest of the model domain, ie countries bordering Poland, is covered with data from the EMEP expert emission inventory [33].

Spatial resolution of the NH_3 emission is set to 5 km x 5 km to be in accordance with the FRAME model grid size. The same spatial resolution is, for example, used in the UK for the regional scale modelling of the reduced nitrogen (and further for the critical levels and loads assessment [30, 34]. The recent studies with the FRAME model, performed by [35], show the need for higher spatial resolution of the model and emissions data and suggest 400 m x 400 m resolution for ammonia emissions inventory [12, 13, 23]. But this is certainly not at present a realistic proposition in the case of Poland because of lack on necessary input data.

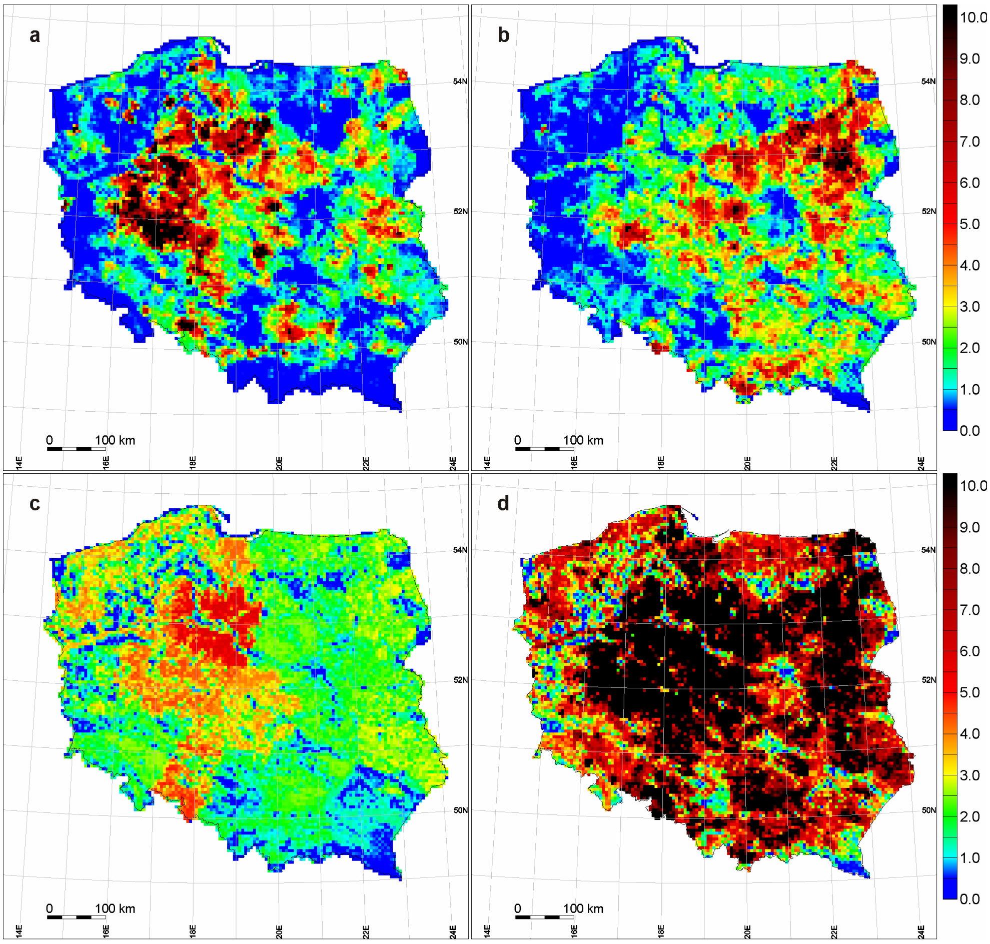

Fig. 1. Ammonia emission from three main sources in 2002 (units are kg(N-NH_3) ha^{-1} y^{-1}): a) pigs (95 Gg NH_3 in 2002), b) cattle (80 Gg), c) fertilizer application (84 Gg) and d) total agricultural emission of NH_3 (314 Gg of NH_3)

The uncertainty related with the aerial NH_3 emissions inventories is usually higher than for SO_2 or NO_x, as there is a large number of influencing factors [5, 12, 23, 34]. The emission factors may change according to the feeding and animal management practices.

Emission of ammonia depends also on meteorological conditions, that change both in space and time and, due to the input data limitations, were not considered in this paper. The other issue is related with the spatial allocation of the emission sources within a commune, which can be associated with considerable uncertainty in mapping emissions, particularly for intensively farmed livestock such as pigs and poultry. This issue has recently been discussed in detail by [34]. Recent studies have shown however that the application of the seasonally varying emissions of ammonia is of certain importance, as a result of eg increased volatilisation due to high temperature in the summer, manure speeding etc [12, 23]. These features are currently under development in FRAME and will be available in the forthcoming version of the model.

Point source emissions data for the European Union members were taken from the EPER database [36]. If available, additional information is provided to calculate the effective emission height. These include data on the stack height and diameter and temperature and exit velocity of the outflow gases. For the non-EU countries the EMEP expert emission data were used [33]. Exactly 43 large point sources from the area of the model domain were used for simulations, emitting over 4 Gg of NH_3. The point source emissions of NO_x and SO_2 were also taken from the EPER database. The NO_x and SO_2 emissions from area and line sources were spatially distributed using the method proposed by [37].

Meteorological data

Wind frequency and wind speed roses employed in FRAME use 6-hourly operational radiosonde data from the stations of Wroclaw, Legionowo, Leba, Greifswald, Lindenberg, Prague, Poprad and Kiev, spanning the whole 2002 year period (Fig. 2). The wind roses were calculated based on the methodology proposed by [18], which was previously successfully applied for the UK FRAME simulations. High resolution precipitation data for year 2002 were developed based on the methodology proposed by [25]. The data are based on the long-term precipitation measurements, spatially interpolated with the residual kriging. This is one of the multidimensional interpolation schemes, considering various atmospheric processes influencing spatial pattern of precipitation during the interpolation procedure.

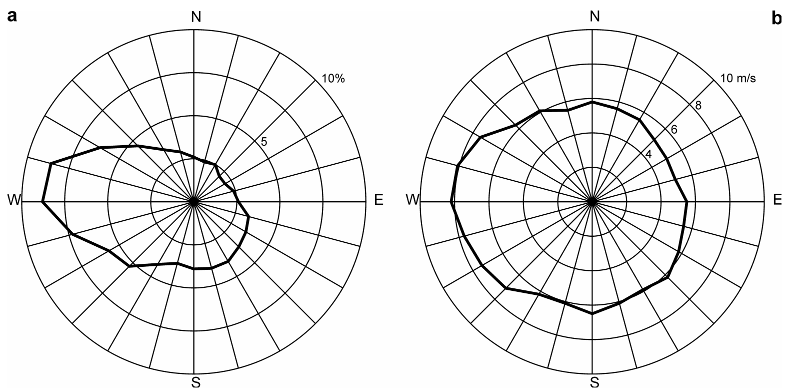

Fig. 2 Wind frequency rose (% per 15^o radial band) (a) and wind speed rose used for FRAME simulations for year 2002 (b)

Model evaluation

FRAME calculated spatial patterns of annual average air concentration and yearly deposition (dry, wet and total) for reduced nitrogen are compared with the EMEP model results. Wet, dry and total national deposition budgets are calculated for the FRAME model and compared with the estimates presented by EMEP and CIEP (wet deposition only [38]). It should be noticed that CIEP wet deposition budget estimates are based on 25 point measurements of ion concentrations in rainfall which are spatially interpolated to produce maps of wet deposition in Poland.

Ion concentrations of atmospheric pollutants in rainfall are measured at 25 stations in Poland and are used here to validate the FRAME modelled wet deposition of NH_4^+. Error statistics proposed by [39, 40] are calculated based on measured and modelled wet deposition data. The error metrics are: mean bias (MB), mean absolute gross error (MAGE), root mean square error (RMSE), mean normalized bias (MNB) and mean normalized absolute error (MNAE).

There are four stations, all operating in the EMEP network, which measure air concentrations of ammonia and ammonium (total mass of $NH_3 + NH_4^+$) with the filter pack samplers. A low number of sites that measure NH_3 and NH_4^+ concentrations in air is common also for other European countries, which makes model validation difficult. The EMEP measuring sites operating in Poland are Jarczew (51°49'N 21°59'E), Sniezka (50°44'N 15°44'E), Leba (54°45'N 17°32'E) and Diabla Gora (54°09'N 22°04'E) and these data are also used here as a complementary measure of model performance. A recently located fifth site has been running within the NitroEurope programme since the end of 2006 and therefore was not included.

It should be noticed that the FRAME model has been also extensively verified for the UK including comparison with an extensive monitoring network of over 90 sites for ammonia concentrations [15].

Source-receptor analysis

The aim of the source-receptor analysis is to asses the influence of national emissions and the transboundary contribution on deposition of reduced nitrogen in Poland in 2002. Two FRAME simulations were undertaken, one (base simulation) with the emissions data from Poland and surrounding countries, as well as boundary concentrations from FRAME-Europe (50 km x 50 km) included. For the second simulation (PL-only), only the Polish-based emission sources were taken into account and boundary concentrations were set to zero.

The transboundary contribution of reduced nitrogen deposition was calculated by subtracting the PL-only simulation results from the base simulation. The resultant maps, showing the differences between base and PL-only simulation as well as fraction of reduced nitrogen deposition from transboundary contribution are presented.

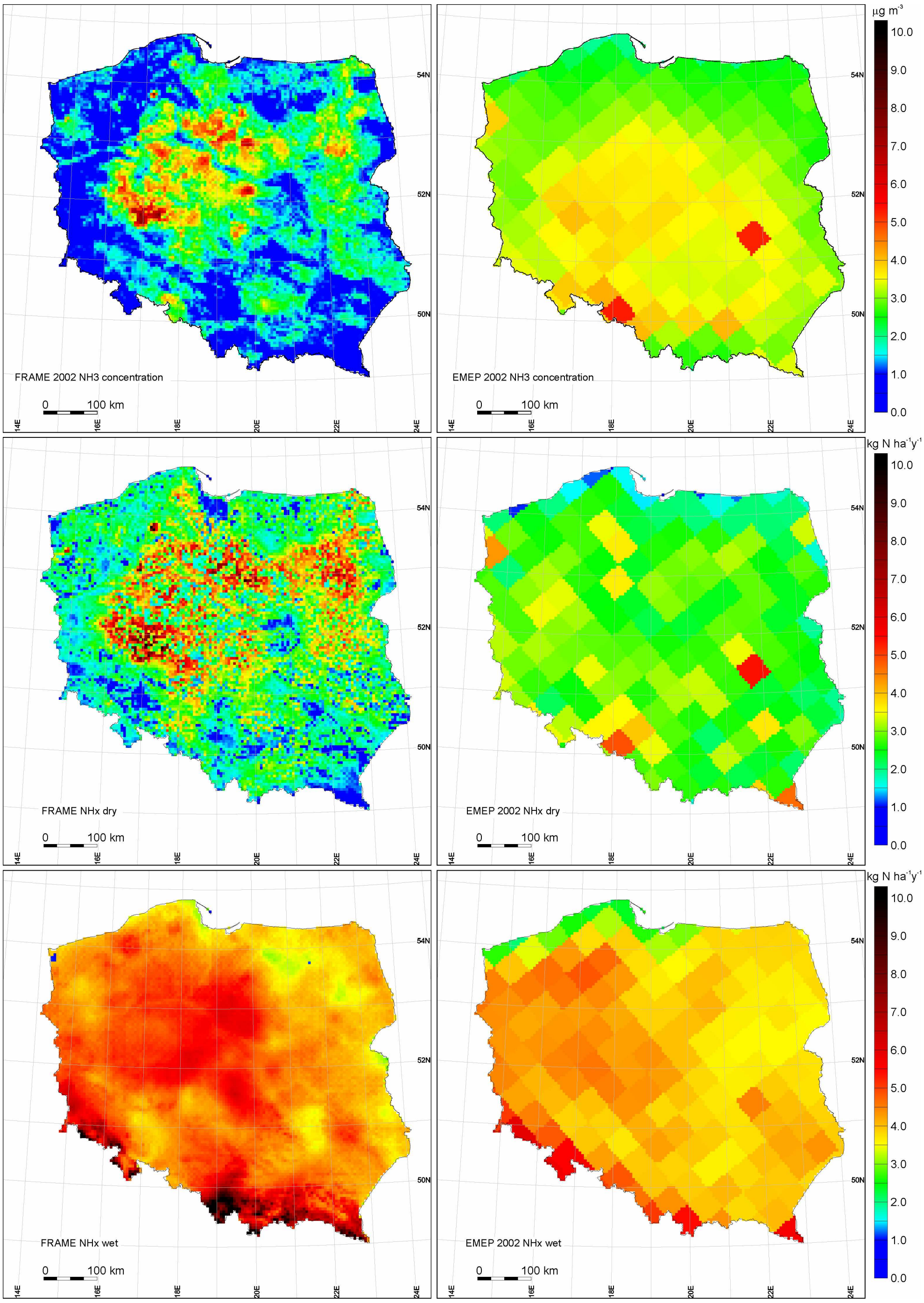

Fig. 3. FRAME (left column) and EMEP (right column) NH_3 concentrations (upper row) and NH_x dry (middle row) and wet (bottom row) depositions

Results and discussion

The highest NH_3 air concentrations are estimated by the FRAME model for the areas of intensive agriculture production in central Poland, ie in the vicinity of high emission from fertilizer application and animal breeding. Also the EMEP model predicts the high concentrations for that area although the FRAME calculated spatial pattern is more complex due to the higher spatial resolution of the FRAME model. The highest estimated annual average concentration of NH_3 exceeds 7 μg m^{-3} of NH_3 for the few 5 km x 5 km grid squares located in the area of high emission originating from pigs and cattle breeding (16°58'E, 51°41'N). This means that the long-term critical level, suggested by [41] of 3 μg m^{-3} of NH_3 might be exceeded there and cause shifts in species composition and also species extinction. The recent findings, presented by [35], show that in some cases 5 km x 5 km resolution of the FRAME model is still insufficient and there might be strong sub-grid variations in ammonia concentrations in the vicinity of emissions sources.

FRAME calculated spatial pattern of annual NH_3 air concentration is in general similar to that estimated by the EMEP model. The largest difference is located for the EMEP grid west from the 22° meridian and north from 51° parallel. In this area a large fertilizer production site is operating (Zakłady Azotowe "Pulawy"), with an annual NH_3 emission exceeding 600 Mg of NH_3 [34]. This emission source was included into the emission inventories used in FRAME (as a point source), but the EMEP emission inventory shows the high emission in this area from the agriculture (*Selected Nomenclature for Air Pollution* - SNAP sector 10) ie close to the surface, not production processes (SNAP sector 4) and this may be the cause of the observed discrepancies.

Monitoring of reduced nitrogen air concentrations in Poland occurs only at a small number of sites and provided as a total mass of NH_3+ NH_4^+ (gas + aerosol) nitrogen. One station (Leba) is located on the sea shore while Sniezka (1603 m a.s.l.) is on the mountain top, therefore these stations are not optimal for model validation. The FRAME modelled air concentrations of NH_3+ NH_4^+ are however in reasonable agreement with the measurements, with the MB of 0.32 and MAGE of 0.62 (error statistics for EMEP model are: 0.60 and 0.74, respectively).

Dry deposition, modelled with FRAME, is the highest in the source areas of central Poland, where large emission from animal breeding is combined with relatively high fertilizer consumption (Fig. 1). In contrary the EMEP model shows the highest dry deposition close to the fertilizer production site in south-east Poland. The possible explanation of these differences was previously discussed for concentrations. In general, FRAME modelled dry depositions are locally higher than those estimated by the EMEP model. This can be attributed to the finer grid resolution of the FRAME model, resulting in a larger spread in the range of modelled concentrations and depositions, particularly where high emissions are concentrated in small areas.

Remote mountainous areas in the south have dry deposition of reduced nitrogen well below 1 kg N $ha^{-1}y^{-1}$. In contrast, these areas suffer from high wet deposition (Fig. 3). This is due to higher precipitation and the influence of the seeder-feeder effect, which is represented in the FRAME model by an enhanced scavenging coefficient for orographic precipitation. Wet deposition of NH_4^+, calculated by the FRAME model can exceed 15 kg N $ha^{-1}y^{-1}$ where the EMEP model estimates do not exceed 8 kg N $ha^{-1}y^{-1}$.

In general, spatial patterns of wet deposition, calculated with the FRAME and EMEP model are similar. Both models estimate the highest wet depositions over the mountainous areas in the south, which is caused by high precipitation supported by the enhanced washout rate in FRAME where the seeder-feeder effect is concerned. Central Poland is the second region of above average wet deposition, as this is the source region of emission, causing large air concentrations of both NH_3 and NH_4^+.

The FRAME estimates of wet deposition are in good agreement with the measurements (Fig. 4), with a determination coefficient above 0.7. The FRAME model significantly overestimates the wet deposition for the Kasprowy station (Tatra Mts., 1987 m a.s.l.) for the year 2002. Simultaneously, for the other mountainous measuring site (Sniezka, 1607 m a.s.l.) the model calculates the wet deposition correctly. This might suggest that the seeder-feeder process parameterization is not accurate for the whole of Poland and this will be further investigated. In the more continental climate of the Tatra Mts., however, the air is on average less humid than in a more maritime climate so that we would expect the seeder-feeder effect to be less influential [27]. The problem might also be related to the specific meteorological conditions in 2002, for which a significant amount of yearly precipitation in the Tatra Mts. area (where Kasprowy station is located) was from summer convective rain. Ion concentrations in precipitation from convective clouds are usually low, therefore resulting in low wet deposition [26]. Here, for the seeder-feeder parameterization long-term climatological data were used.

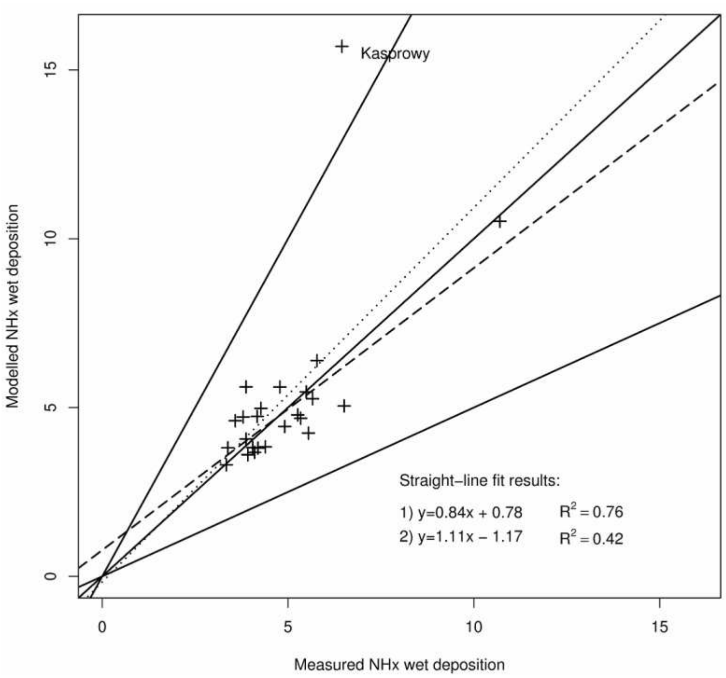

Fig. 4. FRAME vs measured wet deposition of reduced nitrogen [kg N $ha^{-1}y^{-1}$]. Linear regressions are for 1) all measurement stations except Kasprowy (dashed line) and 2) all stations (dotted line). The 1:1, 2:1 and 1:2 lines (solid) are shown for reference

Table 1

Error statistics for wet deposition of reduced nitrogen (Kasprowy station excluded)

Metrics	FRAME	EMEP
MB	–0.01	–0.77
MAGE	0.60	1.08
RMSE	0.73	1.46
MNB	0.01	–0.12
MNAE	0.13	0.21

Error statistics calculated for the wet deposition of the reduced nitrogen are summarized in Table 1 for the FRAME and EMEP models. In case of the FRAME model, all statistics suggest that the modelled wet deposition is accurate, with relatively small mean errors. MB is close to zero, therefore there is no general under or overestimation of the FRAME modelled wet deposition, which is important for the critical loads assessment in Poland. The error statistics, calculated for the FRAME and EMEP model suggest that the former performs better and the estimated wet deposition is more reliable. The EMEP model tends to underestimate wet deposition of the reduced nitrogen, with the average error being two times larger than calculated for the FRAME data.

Total mass of reduced nitrogen deposited in the year 2002 in Poland, as estimated by the FRAME model, is close to 227 Gg of N (Table 2) and this is about 40 Gg of N less than was emitted. The FRAME estimated dry, wet and total deposition budgets of reduced nitrogen are in close agreement with the EMEP data. The FRAME dry deposition budget is smaller than that estimated by the EMEP model, while wet deposition is larger. This is in accordance with the error statistics described above, which suggest the EMEP model tends to underestimate the wet deposition, while the MB calculated for the FRAME model is close to zero. According to the FRAME model estimates, NH_3 contributes to 86% of dry deposition within the domain. Simultaneously, FRAME wet deposition budget is close to the measurement-based estimates presented by CIEP [36] and the differences are smaller than 5%. The FRAME estimated wet deposition budget is 17% larger than estimated by the EMEP model. The wet deposition budget is dominated by NH_4^+, contributing 90%.

Table 2

Dry, wet and total deposition budget in Poland in 2002 (Gg of N)

	FRAME	EMEP	CIEP
Dry	80.4	85.9	Not available
Wet	146.5	125.1	151.3
Total	226.9	211.1	Not available

The differences between the FRAME and EMEP deposition budgets can be considered as small (approximately 6% for dry and 17% for wet deposition), taking into account different input data and model formulations. These include the emissions inventory and dissimilarities in models construction, particularly concerning the grid size and vertical resolution as well as chemical and physical parameterizations and the fundamental difference between a Eulerian model driven by a meteorological model and a statistical Lagrangian model employing simple representation of meteorological conditions. The effect of different grid size on modelled concentrations and depositions is summarized in Figure 5. While the mean concentrations and depositions, calculated for the area of Poland, are similar for FRAME and EMEP, there are large discrepancies in the maximum values. FRAME predicts higher maxima of concentrations and depositions, which are averaged over the coarse grids of the EMEP model. The other differences between the FRAME and EMEP model, which may be of special importance in NH_3 modelling, are related with high vertical resolution and assumed constant emission of NH_3 over the year for FRAME. The first vertical layer of the FRAME model has 1 m, while for EMEP it is 90 m. This may be important as the most NH_3 emissions come from sources located near the ground level.

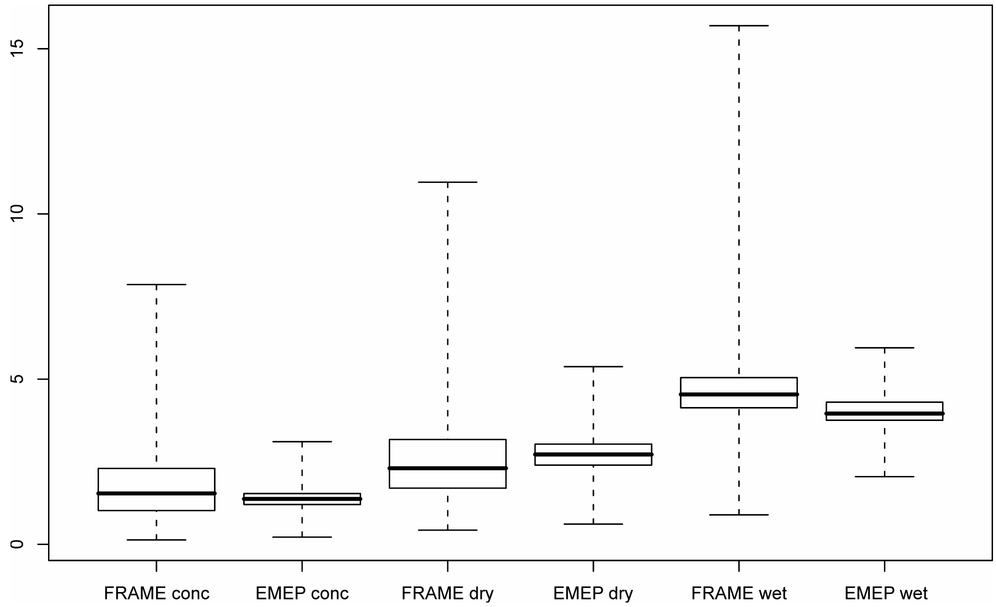

Fig. 5. Boxplots calculated from FRAME and EMEP estimates of NH_3 concentrations [$\mu g\ NH_3\ m^{-3}$] and dry and wet depositions of reduced nitrogen [$kg\ N\ ha^{-1} y^{-1}$] for the area of Poland. Statistics are: minimum, 1st quartile, mean, 3rd quartile and maximum

The source attribution analysis, performed with the FRAME model, shows that national emission of NH_3 is responsible for almost 64% of total deposition of reduced nitrogen in Poland, while the EMEP estimates suggest that the 58% of the total deposition is from national emissions sources [42]. According to FRAME estimates, up to 93% of dry deposition in Poland comes from national activities, while the transboundary contribution reaches 53% of wet deposition. This is in agreement with the deposition maps presented earlier, where dry deposition is high close to the emission sources and wet deposition being related, in general, to the areas of high precipitation and, to a lesser extend, with high emission (central Poland).

The western part of Poland and the mountainous areas in the south are strongly influenced by the deposition of reduced nitrogen from transboundary transport (Fig. 6). In the mountains over 80% (locally over 90%) of the total NH_x deposition comes from sources located outside of Poland. Very similar spatial patterns, calculated with the EMEP model, were presented by [42].

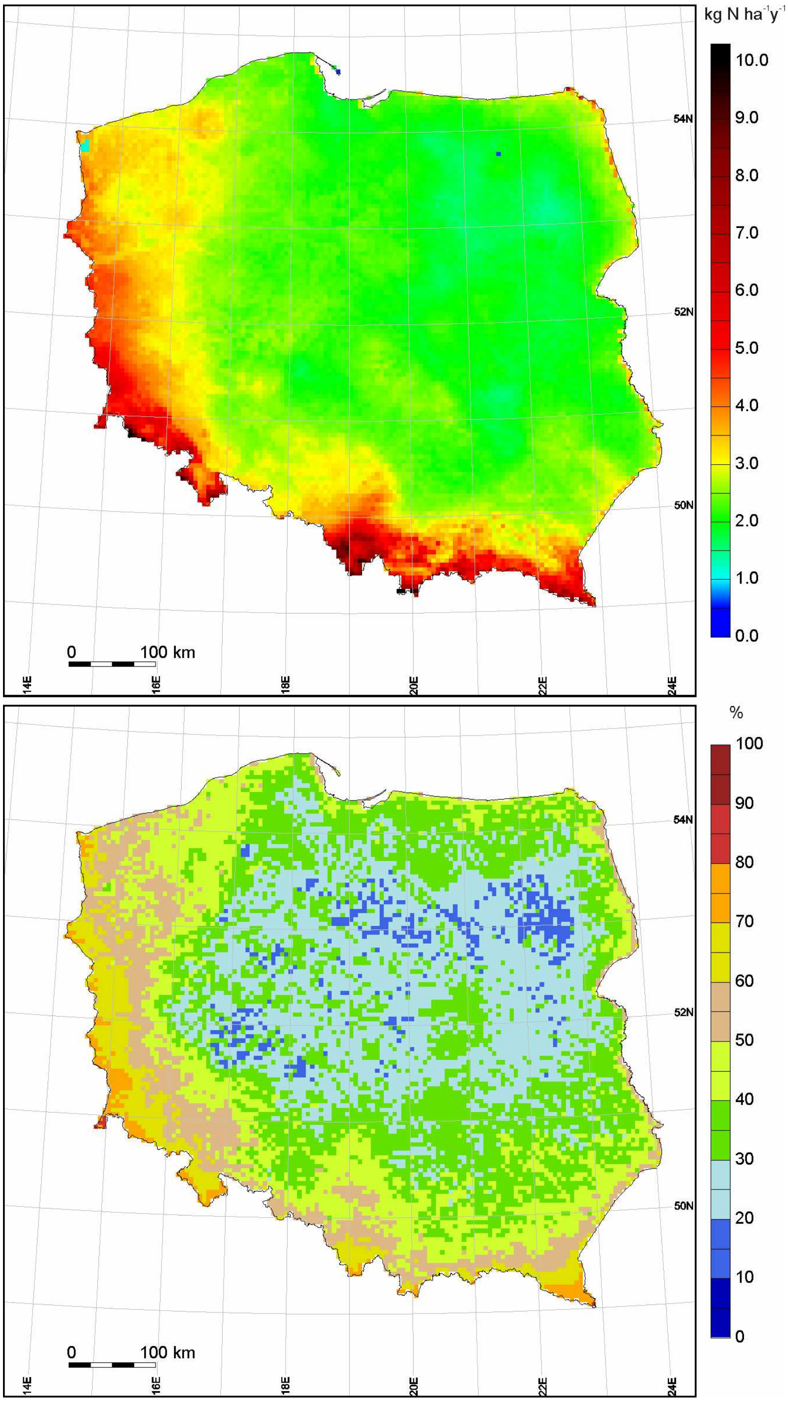

Fig. 6. Transboundary contribution to total reduced nitrogen deposition in Poland

Summary and conclusions

Spatial patterns of air concentrations of NH_3 and dry and wet deposition of reduced nitrogen were estimated at a 5 km x 5 km spatial resolution using the Fine Resolution Atmospheric Multi-pollutant Exchange model (FRAME). The model results show good agreement with the available measurements of wet deposition. For ammonia and ammonium concentrations only four measuring sites were available for comparison so a detailed verification of modelled concentrations could not be undertaken, although FRAME estimates were found to be in reasonable agreement with the measurements. As the reduced nitrogen is an issue of raising importance, the number of air concentration measuring sites should be increased.

Future improvements of the FRAME model should be focused on the seeder-feeder parameterization. The process is responsible for the prediction of high wet deposition over the mountainous regions in the south of Poland. This may lead to exceedance of critical loads for nitrogen deposition, therefore the reliable parameterization of the seeder-feeder process in the model is of importance. The other field of the future improvements is related with development of the seasonal version of the FRAME model. This may improve the modelling results, as suggested by [12, 23], as the ammonia emissions change within the year considerably.

The spatial patterns of concentrations and depositions, calculated with the FRAME model, are in general similar to those obtained by the Unified EMEP model. The high resolution of the FRAME model results however in locally higher concentrations and depositions estimates then in case of the EMEP model and leads to a better estimation of the wet deposition described by the error statistics. This might be crucial in critical levels and loads assessment as the higher concentrations and depositions calculated locally by FRAME can result in exceedances over the areas where the currently used EMEP deposition estimates are below the critical load threshold.

High spatial and vertical resolution, good correlations of results with measurements and with other data sources (EMEP, CIEP) are among the main advantages of the FRAME model. The short run time of the model, compared with complex Eulerian ones, is also a virtue especially if a large number of source-receptor simulations or emission scenarios have to be investigated. The Fine Resolution Atmospheric Multi-pollutant Exchange model, FRAME, can therefore be considered as a useful tool supporting government policy in assessing the effects of abatement of pollutant gas emissions, critical loads and levels exceedances and in Integrated Assessment Modelling in Poland.

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MODELOWANIE STĘŻEŃ I DEPOZYCJI AZOTU ZREDUKOWANEGO I PRZYKŁADY ZASTOSOWAŃ DLA POLSKI

Zakład Klimatologii i Ochrony Atmosfery, Instytut Geografii i Rozwoju Regionalnego, Uniwersytet Wrocławski
Centrum Ekologii i Hydrologii, Edynburg

Abstrakt: W wyniku skutecznej redukcji emisji SO_2 i NO_x wzrasta względny udział azotu zredukowanego w zakwaszaniu i eutrofizacji środowiska przyrodniczego w Polsce i całej Europie. Głównym źródłem przestrzennej informacji o suchej i mokrej depozycji NH_x w Polsce jest model EMEP (*European Monitoring and Evaluation Programme*), pracujący z przestrzenną rozdzielczością 50 km x 50 km. Ze względu na dużą zmienność przestrzenną w emisji, koncentracji i depozycji NH_x taka informacja może być niewystarczająca dla analiz w skali regionalnej. W pracy zastosowano model FRAME (*Fine Resolution Atmospheric Multi-pollutant Exchange*) do obliczenia średniorocznych stężeń azotu zredukowanego oraz jego suchej i mokrej depozycji w rozdzielczości przestrzennej 5 km x 5 km. Uzyskane wyniki wykazują dużą zgodność z dostępnymi danymi pomiarowymi oraz z przestrzenną informacją obliczoną za pomocą modelu EMEP, wykazując jednocześnie znacznie większą zmienność przestrzenną. Różnice w bilansie depozycji, obliczonym za pomocą szacunków FRAME i EMEP, nie przekraczają 17% dla depozycji mokrej i 6% dla suchej. Różnice między FRAME a szacunkami polskiego Głównego Inspektoratu Ochrony Środowiska są na poziomie 3% w przypadku mokrej depozycji. W pracy wykazano także, że 93% suchej i 53% mokrej depozycji azotu zredukowanego w Polsce pochodzi z emisji ze źródeł krajowych. Transgraniczny napływ zanieczyszczeń ma największe znaczenie na obszarze zachodniej Polski oraz w górach na południu kraju.

Słowa kluczowe: amoniak, azot zredukowany, depozycja, FRAME, Polska