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The application of the FRAME model for modelling concentrations and deposition of selected atmospheric pollutants and mapping the critical levels and loads exceedance for Poland

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Abstract A critical level is the concentration of a pollutant in the atmosphere above which direct adverse effects on receptors, such as human beings, plants, ecosystems or materials, may occur according to present knowledge (UNECE 1996). In this paper the Fine Resolution Atmospheric Multi-pollutant Exchange model (FRAME) is used to asses the spatial patterns of yearly averaged air concentrations of SO_2 and NH_3 for the area of Poland. FRAME is a Lagrangian model with high spatial (5x5 km) and horizontal resolution (33 layers) and was originally developed for the area of the United Kingdom. The model was recently tested for the United Kingdom, showing close agreement with measurements. It has also been used as a tool supporting decision making processes by DEFRA (Department for the Environment, Food and Rural Affairs) and to estimate the critical loads and critical levels exceedance in the UK. The modelled yearly averaged air concentration maps are used to determine the areas where the critical loads are exceeded.

Key words: *atmospheric pollution, pollutant deposition, critical levels, critical loads, FRAME, Poland, numerical modelling*

1. Introduction

The critical load and level concept has been developed since the 1980s under the UNECE Convention on Long-range Transboundary Air Pollution, providing a sustainable reference point against which pollution levels can be compared. Critical level and loads can be used for calculating emission ceilings for individual countries with respect to acceptable air pollution levels (UBA 2004).

In this paper, a high-resolution long-range transport model – FRAME (Fine Resolution Atmospheric Multi-pollutant Exchange; Singles et al. 1998, Fournier et al. 2004) is applied to calculate yearly averaged concentrations of SO_2, NO_2 and NH_3 and the total deposition of nitrogen for the area of Poland. These data are also used to calculate the exceedances of the critical level and loads for the semi-natural ecosystems. A simple approach was chosen, based on empirical critical loads/levels proposed by UBA (2004), since the main aim of this work is to present an example of possible applications of the FRAME model. A more detailed study on the critical load exceedance will be continued utilizing critical load functions of acidity and modelled critical loads of nutrient nitrogen for Polish terrestrial ecosystems, developed at the Institute of Environmental Protection for the ongoing review of the Gothenburg Protocol (Mill et al. 2007).

2. Fine Resolution Multi-pollutant Exchange (FRAME) model

FRAME has been used here to calculate yearly averaged concentrations of SO_2, NO_2 and NH_3 and the total deposition of atmospheric nitrogen (NH_x and NO_y). The model was developed in the Centre for Ecology and Hydrology Edinburgh and has been successfully used for modelling long-range transport and deposition of atmospheric pollutants in the United Kingdom. Recently FRAME has been adopted for the area of Poland.

FRAME is a regional Lagrangian model with 5 x 5 km resolution. An air column is divided into 33 layers moving along straight-line trajectories with a 1° angular resolution. The air column advection speed and frequency for a given wind direction is statistically derived from radio-sonde measurements (Dore et al. 2006). The layer thickness varies from 1 m at the surface to 100 m at the top of the mixing layer. Vertical diffusion in the air column is calculated using K-theory eddy diffusivity and solved with the Finite Volume Method. Wet deposition is calculated using a diurnally varying scavenging coefficient depending on a mixing layer depth and a 'constant drizzle' approximation. An enhanced washout rate is assumed over mountainous areas due to the scavenging of clouds droplets by the seeder-feeder effect to calculate the local scale orographic enhancement of precipitation and concentration (Dore 1992, 1999). The washout rate for the orographic component of rainfall is assumed to be twice that calculated for the non-orographic component. The dry deposition of NH_3 is ecosystem specific and includes five land classes: forest, moorland, grassland, arable, urban and water. A canopy resistance parameterisation is employed including an optional canopy compensation point module for representation of the bi-directional exchange of NH_3. The model chemistry includes gas phase and aqueous phase reactions of oxidised sulphur and oxidised nitrogen and the conversion of NH_3 to ammonium sulphate and ammonium nitrate aerosol.

3. Emission inventory

A detailed inventory of annual emissions from individual point sources (568, 742 and 43 sources for SO_2, NO_x and NH_3 respectively) for the year 2004 was provided by the EPER (European Pollutant Emission Register, 2006). The point sources emissions of SO_2, NO_2 and NH_3 wee rescaled to give correct totals for the year 2002, according to the National Inventory Report (Olendrzyński, 2003) estimates. The low level emissions of SO_2 and NO_2 from non-industrial (residential) combustion for the area of Poland were spatially estimated based on the National Inventory Report and data provided by the National Statistical Office (2006) at the commune level. NO_2 emissions from road transport were estimated using the detailed information on the traffic intensity provided by the General Directorate for National Roads and Motorways. Maps of the ammonia emission (mainly from agriculture) were developed using the methodology proposed by Dragosits at al. (1998) and information on the animal numbers and fertilizer consumption, provided by the National Statistical Office (2006). The low-level emission of NH_x was calculated separately for different sources: for cattle, pigs, poultry and sheep, and fertiliser (Kryza et al. 2007) and mixed into the lowest surface layers with a source-dependent emission height.

This emission inventory is in agreement with the National Inventory Report for 2002 (Olendrzyński 2003) and it was used to calculate the yearly averaged concentrations of SO_2, NO_x, NH_3 and total deposition of atmospheric nitrogen (both reduced and oxidized) with the FRAME model. The FRAME concentration and deposition maps have been used to calculate the critical loads/levels exceedances.

4. Exceedance calculations

The exceedance $Ex(X_{dep/conc})$ of the critical load/level $CL(X_{dep/conc})$ is given as (UBA 2004):

$$Ex(X_{dep/conc})=X_{dep/conc} - CL(X_{dep/conc})$$

where $X_{dep/conc}$ is the deposition (dep) or concentration (conc) of a pollutant X. If the $X_{dep/conc}>CL(X_{dep/conc})$, the critical load or level is exceeded, and the amount of the exceedance is marked on the presented maps.

The critical levels for yearly average concentrations of SO_2, NO_2 were set, after UBA (2004), to 20 and 30 $\mu g/m^3$ respectively. The critical level of NH_3 was set to 3 $\mu g/m^3$ according to Sutton, Sheppard & Fowler (2006).

This work is based on the empirical critical loads for nitrogen deposition to natural and semi-natural ecosystems, proposed by UBA (2004). The critical loads for many ecosystems are currently estimated within the range 10-15 kg N $ha^{-1}y^{-1}$ (Krupa 2002, UBA 2004). Here, the threshold of 10 kg N $ha^{-1}y^{-1}$ was chosen. The forest and semi-natural areas were selected based on the CORINE Land Cover database.

5. Results

The spatial patterns of yearly average ground level air concentrations of SO_2, NO_x and NH_3 show the highest values close to the emission sources for all chemical species (Fig. 1). The maps show general agreement with the EMEP estimates (EMEP plots are not presented), which were previously used to calculate the exceedances of the critical load and levels. The FRAME modelled concentrations are locally significantly higher than those reported by the EMEP model. This is because of the high spatial resolution of the FRAME model.

The modelled SO2 and NOx air concentrations, calculated with the FRAME model, are in close agreement with the measurements available for the year 2002 with the R2 for both SO2 and NOx close to 0.7 (Fig. 2). It should be noted that only the background stations were used for comparison. The urban and traffic stations are strongly underestimated by the model, which is clearly visible for the NOx concentrations (Fig. 2). This is because of the large subgrid scale variations in concentration. NH3 concentration is not routinely measured at the monitoring stations in Poland, therefore it is not possible to validate the model estimates.

Fig. 3 shows spatial patterns of the total deposition of oxidized, reduced and total atmospheric nitrogen. The mountainous areas in the south of the country are exposed to large deposition of both oxidized and reduced nitrogen. This for the two reasons:

1. the precipitation over the mountainous area is substantially higher than that in the lowlands and this increases the wet deposition;
2. the FRAME model incorporates the seeder-feeder effect by doubling the washout coefficient over the mountains;
3. high levels of pollutants are advected from the other European sources in prevailing westerly winds.

The FRAME modelled wet, dry and total deposition budget of NO_y and NH_x was checked against the results obtained with the EMEP model and interpolation-based estimates of wet deposition calculated by GIOS/IMGW

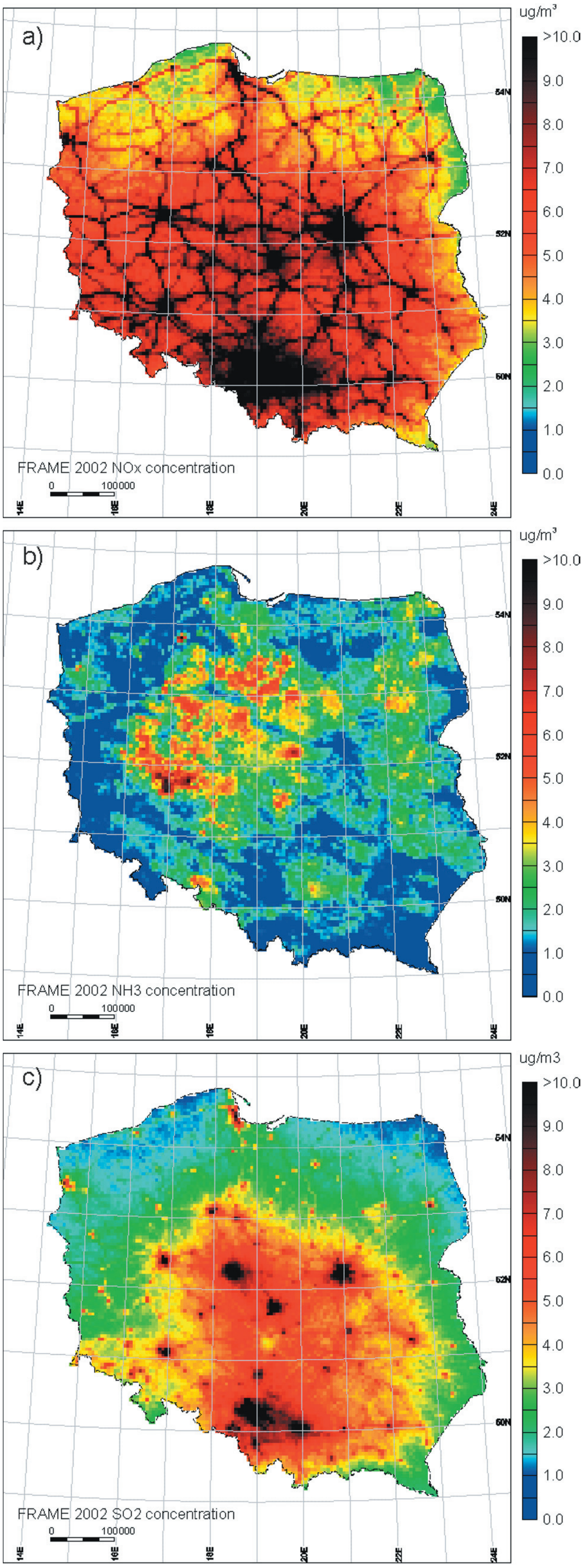

Fig. 1 Yearly average air concentration of a) NO_x b) NH_3 and c) SO_2

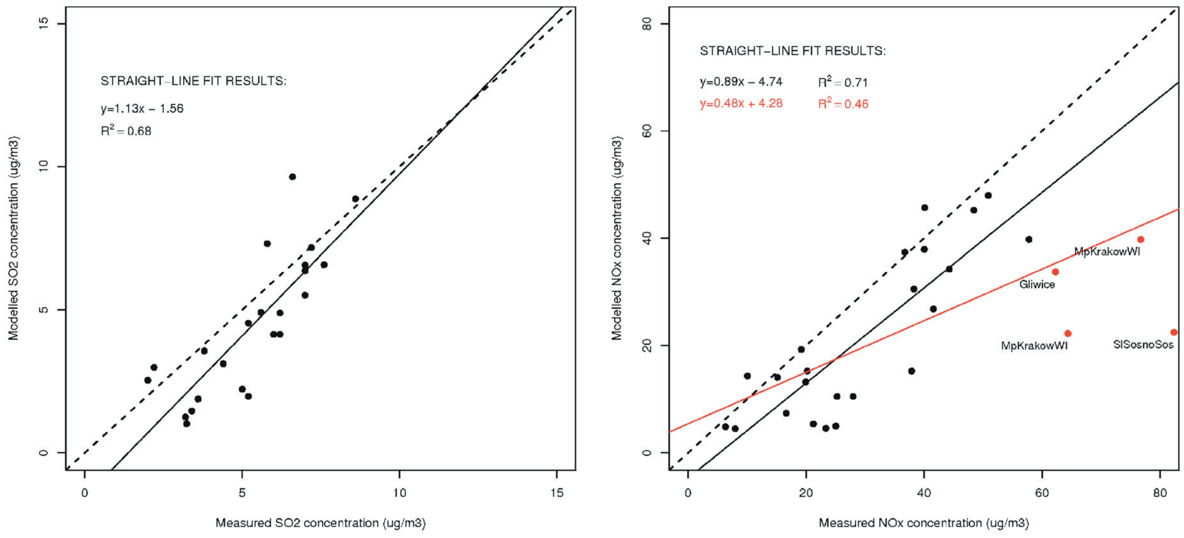

Fig. 2 Correlation between modelled and measured air concentrations of SO_2 and NO_x

(Chief Inspectorate of the Environmental Protection 2006) for the year 2002 (Table 1). The dry deposition budget for FRAME is generally lower than that reported by EMEP. The FRAME modelled wet deposition budget is in close agreement with GIOS/IMGW estimates.

Table 1 Deposition budget for NO_x and NH_x (Gg N/y)

	FRAME	EMEP	GIOS/IMGW
NOx dry	57.7	72.2	-
NOx wet	93.0	98.2	94.4
NOx tot	150.7	170.4	-
NHx dry	79.6	85.9	-
NHx wet	146.7	125.1	151.3
NHx tot	226.3	211.1	-

According to the FRAME model estimates, the critical level of SO_2, suggested by UBA (2004), is not exceeded for the seminatural areas. There are small areas where the NO_x critical load is exceeded (Fig. 4, Table 2) and they are mainly located in the close vicinity of large cities and main roads. Although spatially not extensive, the exceedances of the NO_x critical loads are in some regions significant. The SO_2 and NO_x concentrations calculated with the EMEP model do not exceed the critical levels.

The total extent of the seminatural areas where the critical level for the ammonia is exceeded according to the FRAME model surpasses 6000 km² (Table 2). For the majority of these areas, the exceedance is lower than 2.5μg. The critical level is exceeded mainly in central Poland. This is the region with the intensive agriculture and thereby the source region of a high NH_3 emission. The exceedance of the NH_3 critical level locally exceeds 5.0 μg.

For the EMEP model, the total extent of the area with the exceedance of the NH_3 critical level is substantially smaller than estimated with FRAME and the exceedances are not greater that 2.5 μg (Table 2). Within the 0-2.5 span most of the grids have the exceedance is not larger than 0.5 μg.

For almost 90% of the seminatural areas, the critical load for atmospheric nitrogen deposition is exceeded (Fig. 7, Table 2). The highest exceedances are noticed in the mountainous areas of south Poland. This is because of the high wet deposition of both oxidized and reduced nitrogen caused by the high precipitation. The wet deposition in the mountainous areas is additionally increased because of the seeder-feeder effect implemented in the FRAME model.

It should be mentioned that in the mountainous terrain cloud/fog water also have a substantial proportion in total nitrogen deposition due to direct interception by earth surface. The efficiency of cloud/fog droplets deposition is not incorporated into the model because of significant differentiation over short distances due to various height, structure and size of trees, as well as frequency of gaps in the forest canopy (Błaś and Sobik, 2003). An extreme pollutant deposition rate via cloud/fog is observed at convex landforms on a windward side of a larger massif covered by forest. In the Izera Mts. (Western Sudety Mts.) at the same site (forest edge at the Stóg Izerski Mt.) the total pollutant deposition was composed of 30% of wet deposition, 19% dry

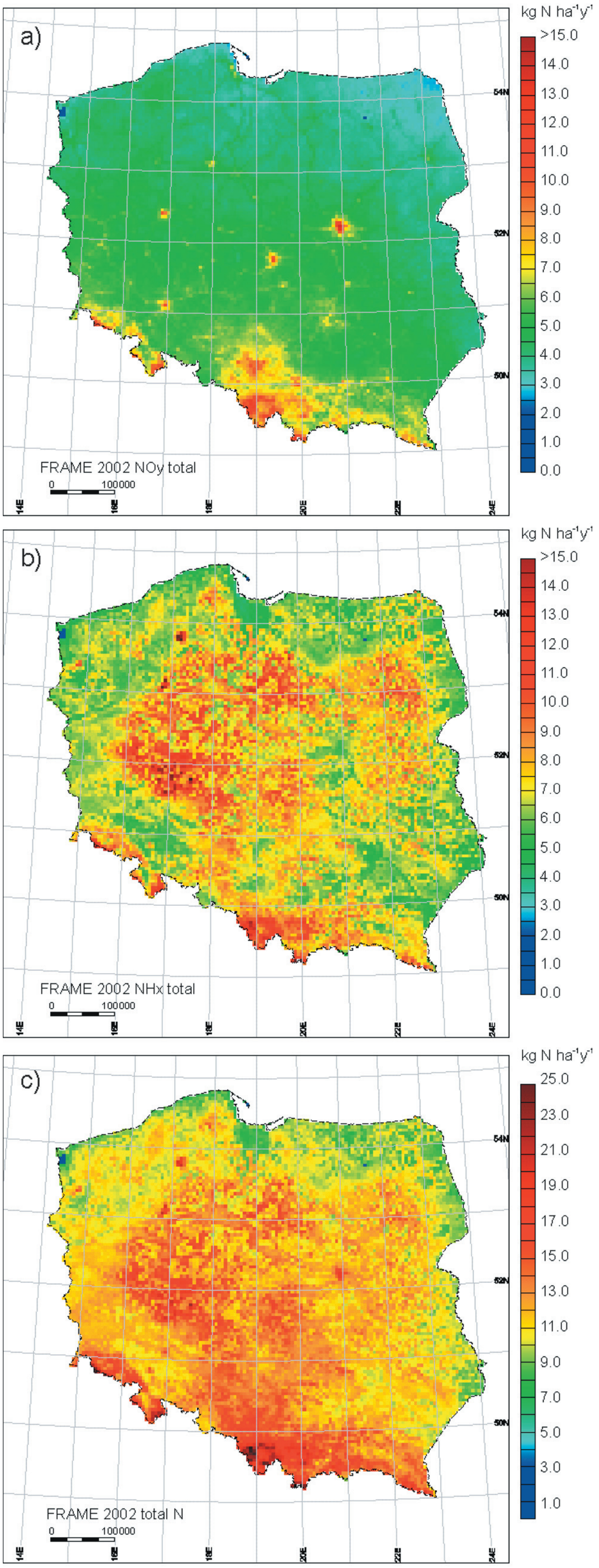

Fig. 3 Annual total deposition of a) oxidized nitrogen, b) reduced nitrogen, and c) total atmospheric nitrogen

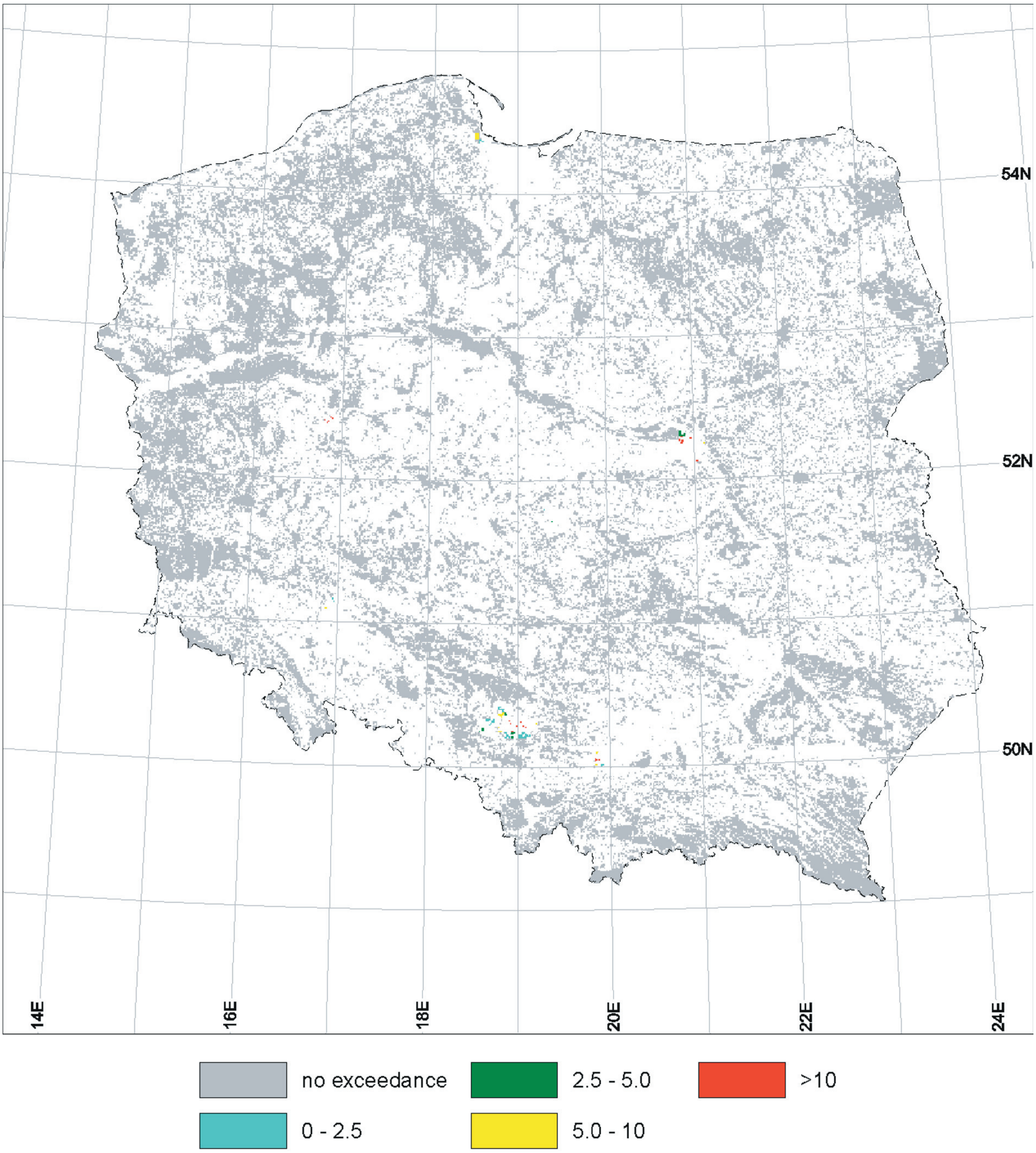

Fig. 4 The exceedance of the critical level of NO_2 for the FRAME model [μg]

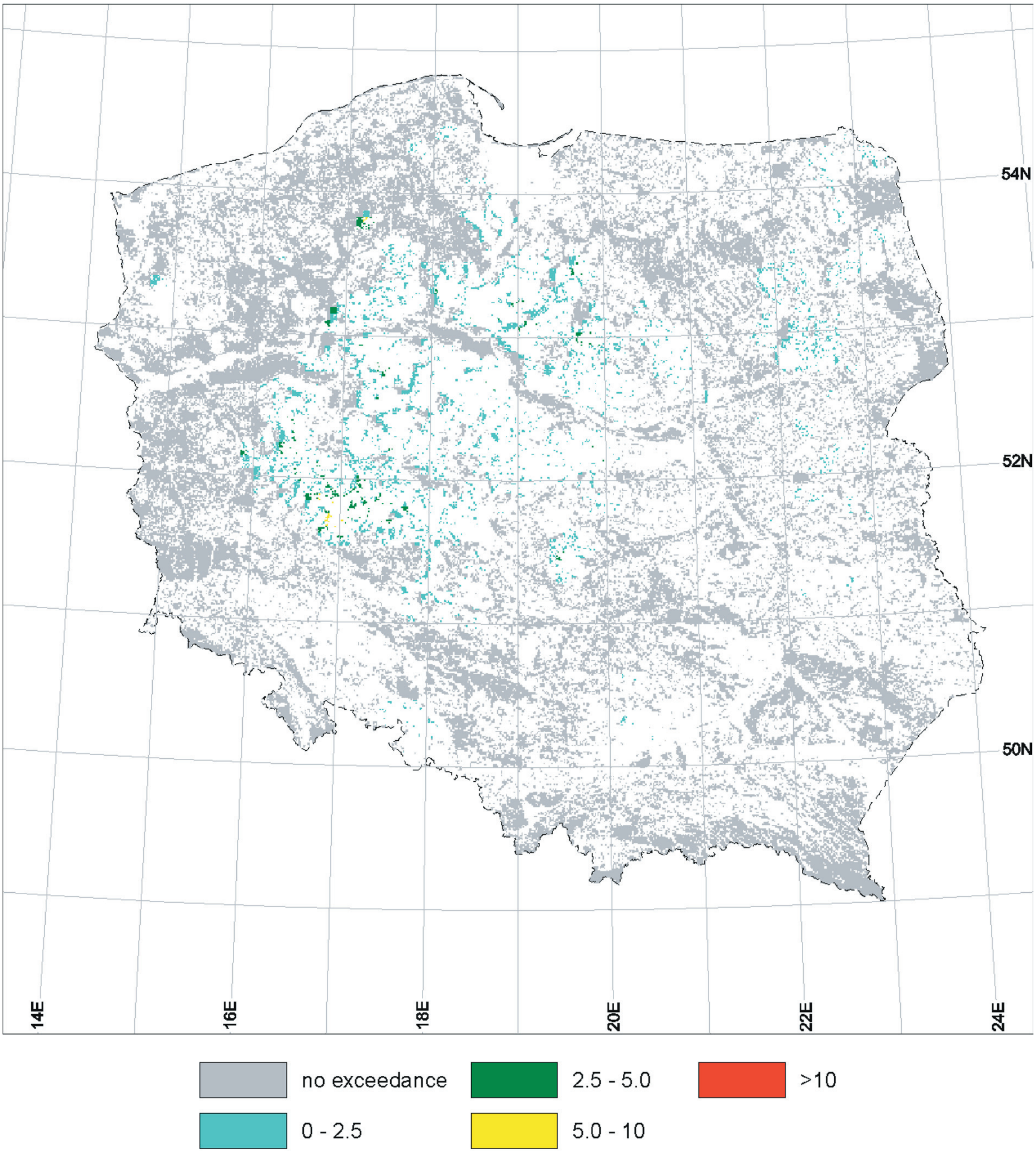

Fig. 5 The exceedance of the critical level of NH_3 based on the FRAME model [μg]

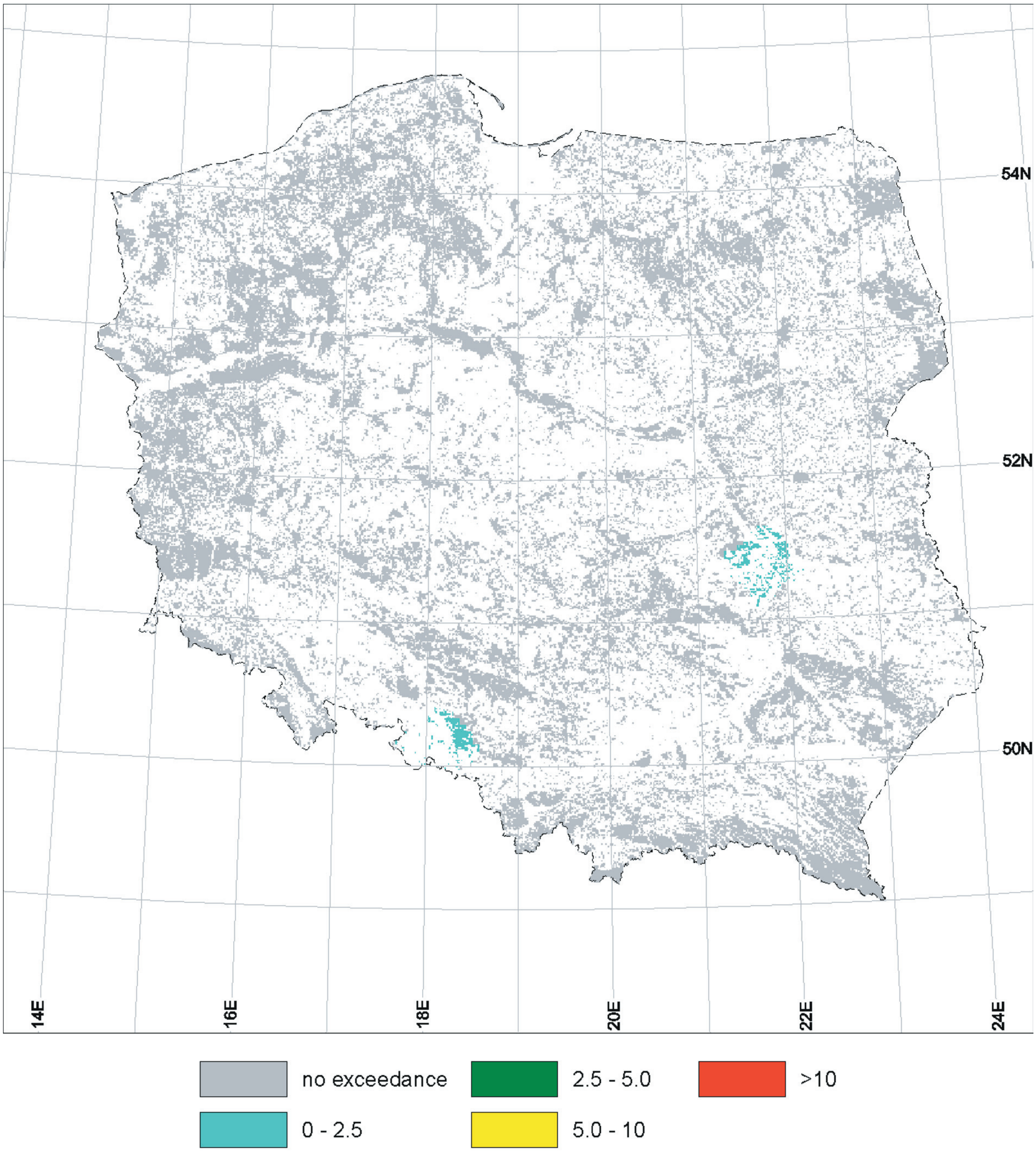

Fig. 6 The exceedance of the critical level of NH_3 based on the EMEP model [μg]

deposition and 51% direct fog/cloud deposition, while the associated proportion at a nearby lowland site was 58%, 40% and 2%, respectively. The total deposition, measured as a chemical input by throughfall at the edge of a spruce forest site during 5 months of the 2004 growing season equals 1.98 Moles m-2 with nitrogen (from both nitrate and ammonia) being the main component (0.91 Moles m-2). This value is equivalent to 305.8 kg of N ha-1 yr-1, which can be compared to 20.2 kg of N ha-1 yr-1 under a spruce canopy at a nearby low land site (Błaś et al., 2005). It shows that the local exceedances of the critical load for atmospheric nitrogen can be much larger than showed on the Fig. 7.

Significant exceedances of the critical levels for atmospheric nitrogen can be also noticed in the seminatural areas of central Poland. The reason for this is the high emission of the NH_3 due to the intensive agriculture and large dry deposition of NH_x close to the emission source.

In general, the EMEP modelled deposition of N gives similar spatial pattern. The exceedances are lower than calculated with the FRAME deposition data, with the exception to the western parts of Poland. This can be probably explained by the advection of the polluted air from the other European sources in the prevailing westerly winds.

Table 2 Total areas with the critical level (NO_x, NH_3) and loads (total N) exceeded

	NO_x		**NH_3**		**Total N**	
	FRAME	EMEP	FRAME	EMEP	FRAME	EMEP
No exceedance	94884	95062	88729	94091	11508	4283
0-2.5	79	0	5872	971	47819	42965
2.5-5.0	30	0	429	0	26316	40500
5.0-10.0	37	0	32	0	8030	7314
>10.0	32	0	0	0	1389	0
Total:	95062	95062	95062	95062	95062	95062

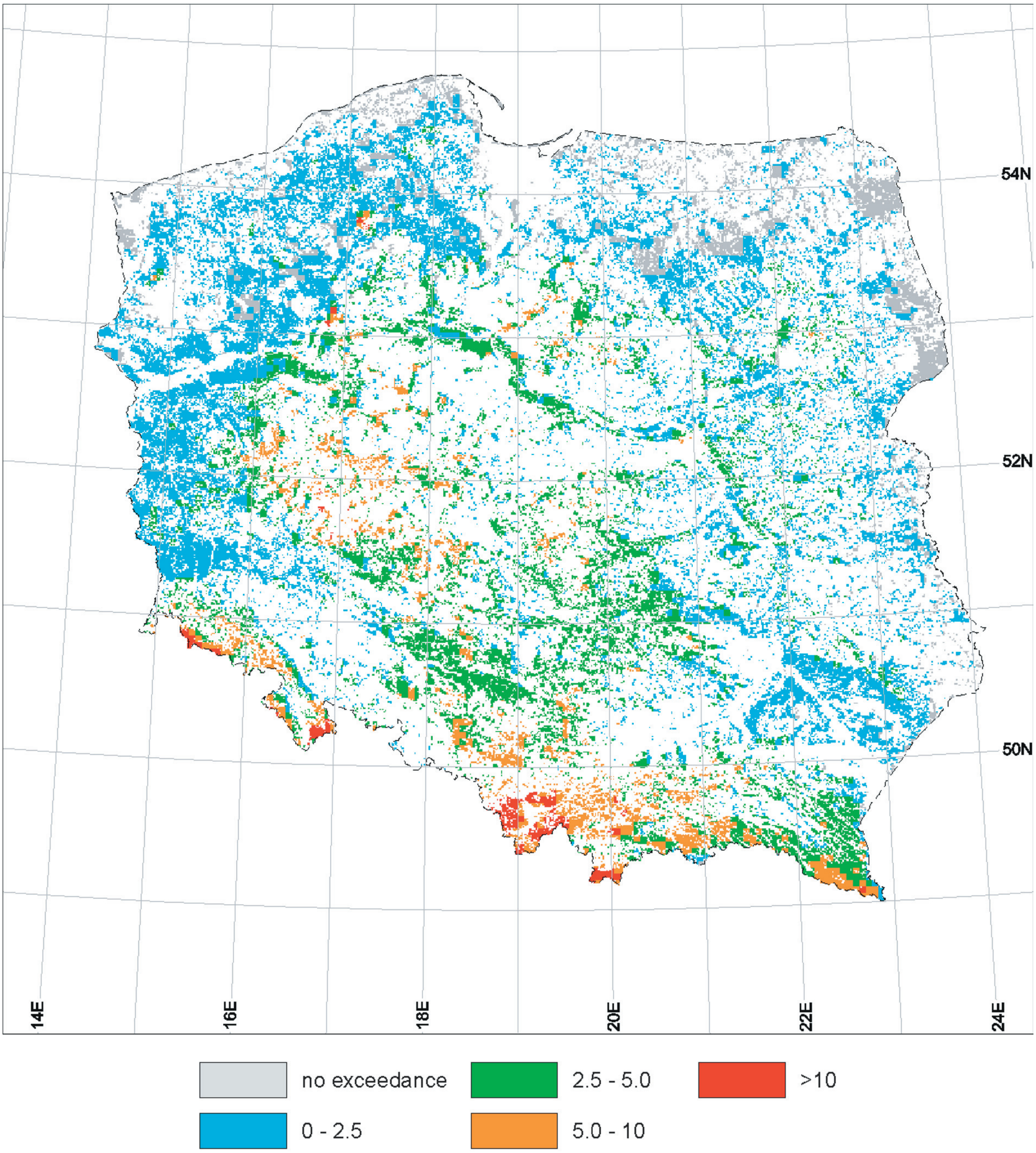

Fig. 7 The exceedance of the critical load for the total atmospheric nitrogen based on the FRAME model

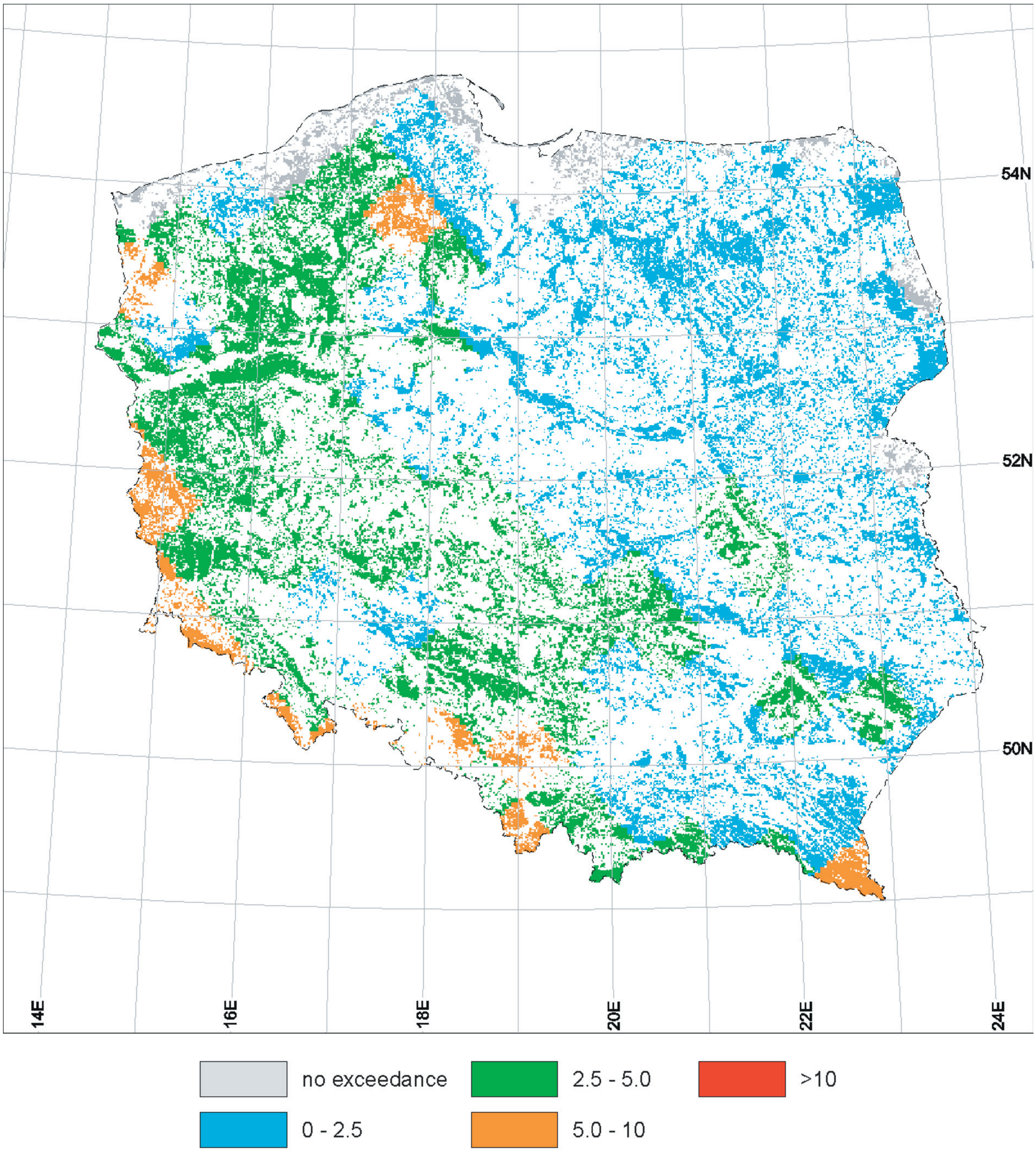

Fig. 8 The exceedance of the critical load for the total atmospheric nitrogen based on the EMEP model

6. Summary and conclusions

Our study shows a possible application of the Fine Resolution Atmospheric Multi-pollutant Exchange model to delineate the areas with the critical level/loads exceeded. The FRAME model has been used to calculate the spatial patterns of yearly averaged concentrations of SO_2, NO_x and NH_3 and the total deposition of oxidized and reduced nitrogen. The correlation of the SO_2 and NO_x modelled concentrations with the measurements is found to be high, with the determination coefficient close to 0.7.

FRAME deposition budgets, calculated separately for wet and dry deposition of the reduced and oxidized nitrogen, are close to the EMEP and GIOS/IMGW reports, which we consider encouraging.

The exceedances of the critical level have been noticed only for oxidized and reduced nitrogen concentrations (the latest only for the FRAME model concentrations). No seminatural areas with the exceedance of the critical level for SO_2 are found for both FRAME and EMEP. For the oxidized nitrogen, the areas with the critical level exceeded are quite small and placed in the vicinity of large cities and main roads.

There are some differences in the extension of the area with the NH_3 critical level exceeded for the FRAME and EMEP model. The total area with the critical level exceeded is larger if calculated with the FRAME data.

The FRAME-based exceedances are also greater then obtained with the EMEP data. This suggest that the difference in the grid size (5 km for the FRAME and 50 km for the EMEP model) can lead to this discrepancies.

There are large areas where the deposition of the total atmospheric nitrogen exceeds the level of 10kg N $ha^{-1}y^{-1}$. However, this calculation is based on a simplified approach, as no spatial information on the critical load is available. The cause of the critical load exceedance may differ from region to region. In the mountainous areas the exceedance is due to the increased wet deposition of both oxidized and reduced nitrogen caused by the higher precipitation, as well as to the seeder-feeder effect leads to the exceedance. Seminatural areas in central Poland are subjected to high dry deposition of the NH_x, which takes place close to the emission sources. Therefore the EMEP model gives higher exceedances in western Poland.

The extension of the area with the N critical load exceedance is larger if calculated with the EMEP deposition data. Simultaneously, the exceedances are larger if calculated with the FRAME data. These discrepancies can also be explained, at least partly, by the different grid sizes. The next reason for this is different import of the aerosols from the other European countries in the prevailing westerly winds.

The FRAME model can be used as a useful tool for assessing different environmental problems in Poland. The modelled results show close agreement with the measurements, and can be considered as reliable. The exceedance of the critical loads and levels is one of the many fields where the FRAME model can be used. Because of the simplifications assumed in this work, the critical levels and loads should be further investigated.

Acknowledgements

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