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The role of annual circulation and precipitation on national scale deposition of atmospheric sulphur and nitrogen compounds

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Abstract:

Atmospheric circulation and rainfall are important factors controlling the deposition of atmospheric pollutants. This paper aims to quantify the role of these factors in the deposition of sulphur and nitrogen compounds, using case studies in the United Kingdom and Poland. The FRAME model has been applied to calculate deposition for the base year (2005), dry and wet years (2003 and 2000 for the UK and 2003 and 1974 for Poland, respectively), and for years with contrasting annual wind patterns (1986 and 1996 for the UK, and 1998 and 1996 for Poland).

Variation in annual wind and rainfall resulted in statistically significant changes in spatial patterns of deposition and the national deposition budget of sulphur and nitrogen compounds in both countries. The deposition budgets of S and N are 5% lower than for the reference year if the dry year is considered in both countries. For the wet year, there is an increase in country total deposition by up to 17%. Years with an increased frequency of eastern winds are associated with an increase in deposition of up to 14% in Poland and 8% in the UK. The national deposition budget is below the average for the years with high frequencies of W winds, especially for the UK (up to 13%). Wet deposition varies due to meteorological factors to a larger extent than dry deposition. In Poland, the changes in national deposition budget due to meteorological factors exceed the changes resulting from emission abatements in years 2000 – 2009 for nitrogen compounds. In the UK, emission abatements influence the

national deposition budget to a larger extent than meteorological changes (except for NH_x). The findings are important in relation to future climate changes, especially considering the potential increase in annual precipitation. This may lead to an increase in deposition over mountainous areas with sensitive ecosystems, where annual rainfall brings significant load of S and N. Changes in annual wind speed and frequency can modify the spatial pattern of deposition. An increased frequency of W winds will benefit both countries through reduced S and N deposition. NW areas of Poland and the UK will suffer from above- average deposition during years with enhanced easterly flow, and this may result in critical loads for acid and nitrogen deposition being exceeded over the areas that are at present sufficiently protected from acidification and eutrophication, despite the ongoing emission abatements.

Keywords: sulphur deposition, nitrogen deposition, national deposition budget, FRAME

1. Introduction

Poland and the United Kingdom are among the European countries with the highest SO_2 and NO_x emissions. These chemical species, together with reduced nitrogen (NH_x) emitted mainly from agricultural activities, lead to acidification and eutrophication of ecosystems and to loss of biodiversity. Since the 1990's, a significant decrease in emissions of SO_2 (67% in Poland and 81% in the UK in 1990-2005) and NO_x (ca. 50% in Poland and the UK) has been observed in both countries, as a result of a successful emission abatement policies. In Poland, the downward trend in NO_x emission after the year 2002 is less pronounced (or even reversed) due to the rising number of cars and increase of the transport sector share in the total emission of NO_x. A large decrease (46%) of NH_3 emissions in Poland took place at the beginning of the 1990's as a result of economic changes (Mill, 2006) and, over recent years, the annual values of NH_3 emission oscillated around 300Gg of NH_3. The decrease in emission of NH_3 in the UK has been relatively modest compared to those of SO_2 and NO_x. Importantly, both countries differ also in geographical, climatological and environmental conditions. The UK, an island with a maritime climate, is relatively remote from pollutant sources in neighbouring countries, which are downwind during prevailing south-westerly winds. In contrast, Poland, one of the largest countries in central Europe, has a more continental climate and significant transboundary exchange of pollutants with neighbouring countries.

The long-term trend in emission is reflected in changes in measured and modelled air concentrations and deposition of atmospheric pollutants, but it can be significantly modified by year to year changes in meteorological factors and thus confound attempts to measure decreases in pollutant deposition due to emissions abatement (Jonson et al., 2006; Andersson et al., 2007; Hess and Mahowald, 2009; Matejko et al., 2009). Andersson et al. (2007) report that the changes in deposition of sulphur and nitrogen due to meteorological factors in Europe may reach 9% for dry and 20% for wet deposition, and they emphasise the significant importance of different meteorological parameters for depositional trends. Matejko et al. (2009) suggest that the variations in wet deposition over the UK are strongly affected by joint changes of precipitation and annual synoptic patterns, resulting in a non-linear relation in the period 1990 – 2005. These non-linearities are also linked by some authors (Fowler et al., 2007; Fagerli and Aas, 2008) with the shift in equilibrium between nitric acid and ammonium nitrate towards particulate phase, caused by the reductions in the SO_2 emissions, on deposition of sulphur and nitrogen compounds. All factors that influence the inter-annual variability or long term trends in deposition of sulphur and nitrogen should be taken into consideration in emission control strategies and for Integrated Assessment Modelling (Oxley et al., 2003). The changes in deposition resulting from year to year variations in meteorology are also of significant importance when considering future climate changes and the smaller decrease in emission over the recent years if compared to the beginning of the 1990's.

The main focus of this paper is the quantification of the influence of the inter-annual changes in meteorological conditions on the spatial patterns of total, dry and wet depositions of sulphur and nitrogen compounds, and on national deposition budget of Poland and the UK. A number of years with specific meteorological conditions have been selected for both countries and used for modelling with the Fine Resolution Atmospheric Multi-pollutant Exchange (FRAME) model. The impact of year to year changes in wind (speed and direction) and in precipitation on sulphur and nitrogen deposition is quantified separately. To distinguish the role of meteorological factors from the changes in emissions, the latter is kept constant for all model runs. The results for specific meteorological conditions are compared with the FRAME run with the meteorological year 2005, used as a reference year. To give a more general picture of the deposition changes due to variation in meteorological conditions, two additional model runs have been performed, with meteorological data for year 2005 and emission data for year 2000 and 2009. This was done to compare the results of anthropogenic emission abatements due to national and international regulations and changes caused by factors that are not human – dependent.

2. Data and Methods

2.1. FRAME model description

The atmospheric transport model FRAME provides information on the annual mean oxidised sulphur and oxidised and reduced nitrogen atmospheric concentrations and deposition. A detailed description of the FRAME model is given in Singles et al. (1998), Fournier et al. (2004), Dore et al. (2006) and Vieno et al. (2010). Details on the model configuration for Poland can be found in Kryza et al. (2010). FRAME is a Lagrangian model which describes the main atmospheric processes in a column of air moving along straight-line trajectories following specified wind directions. The model consists of 33 vertical layers of varying thickness, ranging from 1 m at the surface, and increasing to 100 m at the top of the domain. Trajectories are advected with different starting angles at a 1° resolution using directionally dependent wind speed and frequency roses. Wind speed and wind frequency roses are calculated using radiosonde and calendar classification data and are described in section 2.4 below. Vertical diffusion of gaseous and particulate species is described with K-theory eddy diffusivity, and solved with the Finite Volume Method. The vertical diffusivity (K_z) has a linearly increasing value up to specified height (H_z) and then remains constant (K_{max}) to the top of the boundary layer. During daytime, H_z is taken as 200 m and K_{max} is a function of the boundary layer depth and the geostrophic wind speed. For night-time, these values depend on the Pasquil stability classes. The FRAME model chemistry scheme is similar to the one used in the EMEP Lagrangian model (Barret and Seland, 1995).

Dry deposition is calculated by determining vegetation dependent velocities (V_d) to each chemical species derived from the dry deposition model (Smith et al., 2000). The model derives maps of deposition velocity taking into account surface properties and geographical and altitudinal variation of wind speed. Wet deposition is calculated with scavenging coefficients and a constant drizzle approach, using precipitation rates calculated from a map of average annual precipitation. The wet deposition flux to the surface is the sum of wet removal from all volume elements aloft, assuming that the scavenged material comes down as precipitation. There is no difference between in-cloud and below-cloud processes and an averaged value of scavenging ratio (Δ_i) is applied in the FRAME model. To produce the scavenging coefficient λ_i, Δ_i is combined with the precipitation rate and the depth of the mixing layer ΔH_{mix}. An increased washout rate is assumed over hill areas due to the seeder-feeder effect. It is assumed that the washout rate for the orographic component of rainfall due to the seeder-feeder effect is twice that used for the non-orographic components (Dore et al., 1992).

FRAME has a grid resolution of 5 km x 5 km and grid dimensions of 172 x 244 cells for the UK and 160 x 160 cells for Poland. Aerosol concentrations at the boundary of the model domain are calculated with the FRAME-Europe model for both countries. FRAME-Europe is a model similar to FRAME, but runs for the entire Europe on the EMEP grid at 50 km x 50 km resolution.

2.1.1. Evaluation of the FRAME model results

Assessment of the accuracy of FRAME in estimating concentrations and deposition has been previously undertaken by Dore et al. (2007), Matejko et al. (2009) and Kryza et al. (2010, 2011), and only the main issues are presented here for clarity. Both for the UK and Poland, the model results were compared with national monitoring networks that measure air concentrations and wet deposition of atmospheric pollutants. Long term dry deposition is measured directly only at a very few sites, therefore direct model-measurement comparison of dry deposition is not feasible. The FRAME results for the UK were compared with Concentration Based Estimated Deposition (CBED) data estimates for national wet deposition budgets (Smith et al. 2000). For Poland the estimates of wet deposition budget are provided by the Polish Chief Inspectorate of Environmental Protection (CIEP), and were used for evaluation of FRAME.

FRAME modelled concentrations and wet deposition of sulphur and nitrogen for the year 2005, which is used as the base year in this study, are in good agreement with the measurements, with the correlation coefficients close to or higher than 0.8, both for the UK and Poland. FRAME modelled wet deposition budgets are in close agreement with measurement-based estimates of CBED and CIEP. For the UK, FRAME has a tendency to constantly give higher values for SO_x wet deposition, and lower for NO_y, in comparison to CBED estimations. For the wet deposition in Poland, the FRAME estimates are below the values reported by CIEP, with the differences less than 15%. In general, the model was found to satisfy the criteria of being ‘fit for purpose’ that over 50% of modelled data points should be within 0.5 times and 2 times the measured value. The good agreement with the measurements shows that the model works correctly for both the UK and Poland and can be applied to assess the influence of extreme atmospheric circulation and precipitation on pollutant concentration and deposition.

2.2. Assessment of the role of meteorological conditions in deposition of atmospheric pollutants in Poland and the UK

To quantify the impact of meteorological conditions on dry, wet and total deposition of sulphur and nitrogen, the following procedure has been applied in this study:

1. FRAME was run with the emission inventory and meteorological conditions (wind speed, frequency and precipitation) for year 2005. The results from this simulation form the baseline deposition information (base simulation, BS, see Table 1 for a summary of the model simulations).
2. To assess the role of wind speed and direction frequency in annual dry, wet and total deposition, FRAME was run with emission and precipitation data as for BS, but with changed wind speed and frequency. Two simulations were performed for each country to determine spatial patterns of deposition during the extreme years in terms of general circulation. The years selected for analysis are described in section 2.4. The difference between the BS and results from a model run for a specific wind speed and frequency was then calculated.
3. To assess the role of precipitation in annual dry, wet and total deposition, FRAME was run with emission and wind data as for BS, but with a changed map of annual precipitation. The procedure used was similar to that described above for wind conditions.

Similarly, the FRAME model was run to quantify the changes in deposition due to changes in emission for the years 2000 and 2009. The meteorological conditions in these simulations were kept constant and equal to the base simulation (Table 1).

The differences between a given FRAME simulation and a base run scenario are presented spatially on maps and tested for statistical significance using a non parametric Wilcoxon test for mean deposition value and an Ansari-Bradley test for variance. The Wilcoxon test was used to compare both paired (grid to grid) and unpaired (country total) deposition values. The paired test is based on differences in deposition calculated in two different model runs for the corresponding grid cells (two grids form a pair). This accounts for both: differences in country total deposition value and spatial location in deposition. The unpaired Wilcoxon test does not account for the grid to grid difference, only for the overall difference in median value. The Ansari-Bradley test is used to check if the variance of deposition values in a FRAME run for a given year and a base run scenario differ significantly. The tests quantify if the mean deposition for a given scenario is significantly different from the base run (unpaired

Wilcoxon test), the variance in deposition differs (Ansari-Bradley test) and if there is a significant difference in spatial distribution of deposition (paired Wilcoxon test).

Table 1 Annual emission, wind and precipitation data used for FRAME model simulations

Simulation name	Emission	Wind	Precipitation
UKBS	2005	2005	2005
PLBS	2005	2005	2005
UKW	2005	1986	2005
UKE	2005	1996	2005
PLW	2005	1998	2005
PLE	2005	1996	2005
UKdry	2005	2005	2003
UKwet	2005	2005	2000
PLdry	2005	2005	2003
PLwet	2005	2005	1974
UK2000	2000	2005	2005
UK2009	2009	2005	2005
PL2000	2000	2005	2005
PL2009	2009	2005	2005

2.3. Emission data

Emissions for year 2005 were used in the FRAME model base runs both for the UK and Poland. The total mass of SO_2, NO_x and NH_3 emitted in year 2005 is summarized in Table 2, together with emissions for 2000 and 2009 that were used here to drive FRAME for simulations UK2000, PL2000 and UK2009, PL2009.

Table 2 Sulphur and nitrogen emissions from Poland and the UK used in modelling [Gg of SO_2, NO_2 and NH_3]

	SO_2	NO_x	NH_3
Poland 2005	1222	811	326
Poland 2000	1511	838	322
Poland 2009	861	820	273
UK 2005	687	1682	305
UK 2000	1226	1877	330
UK 2009	398	1086	288

Emissions of SO_2 and NO_x for the UK were taken directly from the National Atmospheric

Emissions Inventory (NAEI, www.naei.org.uk). Ammonia emissions are estimated for each grid square using the AENEID model (Atmospheric Emissions for National Environmental Impacts Determination) that combines data on farm animal numbers with land cover information, as well as fertiliser application, crops and non-agricultural emissions (Dragosits et al., 1998).

For Poland, point sources emissions with chimney parameters (stack height, diameter, temperature and velocity of the outflow gases) were provided by the Institute of Environmental Protection KASHUE/KOBIZE. For the remaining emission sources, the national emissions inventory for the year 2005, organized by SNAP sectors, including area, line and point sources, was taken from Dębski et al. (2009) and, in a spatial form suitable for modelling, from Kryza et al. (2010, 2011).

Emissions data for years 2000 and 2009 for the UK and Poland were derived from the 2005 emission maps by applying emission sector-dependent scaling factors (SF). SFs were provided for each SNAP sector and were calculated from the official emissions reported by NAEI for the UK and by KASHUE for Poland. This method was applied to assure homogeneous spatial patterns of emission, and therefore to eliminate the influence of spatial changes in the location of emission sources (Matejko et al., 2009).

2.4. Meteorological data

2.4.1. Precipitation data

FRAME requires annual average meteorological information on wind speed, direction and precipitation. Precipitation data for the UK was generated by interpolation of measurements from the tipping bucket rain gauges gathered at the Meteorological Office national network at approximately 5000 stations. Precipitation data for Poland was developed using measurements from about 200 weather stations and spatially interpolated with the residual kriging procedure supported by a high resolution map of the long-term precipitation (Kryza, 2008).

To select the extreme years for precipitation, the period 1986-2006 was analysed for the UK and 1951-2006 for Poland. The periods were selected based on the data availability. For the UK, the national mean annual precipitation of the period was 1124 mm, with standard deviation of 119 mm. The wettest years were 2000 (1331 mm, 118 % of the average) and 2002 (1281 mm, 114 % of the average), whereas the driest were 2003 (881 mm, 78 % of the average) and 1996 (920 mm, 82 %). For Poland, the average annual precipitation for the period 1951 – 2006 was 653 mm, with standard deviation of 78 mm. The unusually dry years in Poland were 1982 (483 mm, 74 % of the average), 1953 (517 mm, 79 %) and 2003 (525 mm, 80 %). The extremely wet years were 1966 (808 mm, 124 % of the average) and 1974 (803 mm, 123 %). Finally, the years 2000 (wet) and 2003 (dry) for the UK were selected and 1974 (wet) and 2003 (dry) for Poland.

The differences between the year 2005 and selected years with extreme rainfall can also be compared more quantitatively, by calculating the grid to grid correlation coefficient and mean difference. The first measure quantifies the spatial shifts in precipitation between the year 2005 and a given year, while the second describes the average difference for all 5 km x 5 km grids covering the UK or Poland. The correlation between the year 2005 precipitation and dry and wet years for both the UK and Poland is above 0.8. This suggests that the spatial pattern of the annual precipitation does not change significantly from average to dry or wet years. However, the mean differences between the year 2005 and dry and wet years in both countries are high and exceed ±200 mm, with the exception of dry year 2003 for Poland, where the mean difference is 91 mm.

2.4.2. Wind conditions

Airflow data were based on the Lamb-Jenkinson weather types classification for the UK (Lamb, 1972; Hulme and Barrow, 1997), and the Niedźwiedź circulation type classification for Poland (Niedźwiedź, 2009), together with radiosonde information from both countries. Analysis of circulation conditions for the UK was conducted for the same period as for rainfall (1986-2006) and for the years 1951-2009 for Poland.

For the UK, the average circulation pattern for the period selected illustrates the predominant wind directions from the SW-W, and low frequency of the NE-SE sector. To select extreme wind roses for the period, the contribution of two sectors, from 120 to 225^{o} and from 225 to 320^{o} are analysed. The first sector is responsible for the transport of relatively polluted air from continental Europe. The second brings relatively clean air from the Atlantic Ocean. On average, the relation of frequency of airflow from the first sector to the second amounts to 1.8. For the oceanic year (frequent advections of relatively clean air from the Atlantic Ocean; 1986), this factor decreases to 1.2 and for the continental year (frequent advections of polluted air masses from Eastern Europe; 1996) increases to 2.4. The correlation coefficients between the wind frequencies from a given direction in year 2005 and 1986 for the UK are 0.88 and 0.75 if years 2005 and 1996 are compared.

Fig. 1 Wind frequency roses used in FRAME model runs for the UK (a) and Poland (b)

For Poland, the average westerly direction frequencies were approximately twice those of easterlies and the frequency of airflow for the broader sector (SW+W+NW) was 1.6 times higher than for NE+E+SE sector. The largest differences were noted in 1990 (extremely oceanic circulation) and 1963 (extremely continental) when the previously mentioned factors were: 4.76, 3.24 (broad SW-NW sector) and 0.88, 0.79, respectively. Almost the same extreme circulation conditions as for the years 1990 and 1963 appeared for two other years: 1998 (predominant W winds) and 1996 (high frequency of E winds), and these were selected for the analysis because of the availability of radiosonde measurements (Fig. 1). For the year 1998, the western direction appeared 5 times more frequently than the eastern, and the W sector (SW+W+NW) was 2.2 times often than the E sector (NE+E+SE). In the case of 1996, a slight predominance of the eastern sector is observed (the corresponding factors are 0.93 for W to E direction and 0.90 for the W and E sectors). The examples of years 1996 and 1998 show that in recent years large circulation contrasts are still present being neither suppressed nor amplified by the warmer climatic phase of the last two decades. The correlation coefficients between the wind frequencies from a given direction in year 2005 and 1998 for Poland are 0.93 and 0.21 for 2005 – 1996. This suggests a larger year to year variability of annual wind patterns in Poland than in the UK, which is also supported by the long term climatological data.

The wind speed roses for the FRAME model runs were calculated for the selected years using radiosonde data for the level 500-1000 m above sea level, according to the methodology proposed by Dore et al. (2006). For the UK, data was taken from seven different geographical locations and the station selection criteria were data completeness and geographical representation of the northern, southern, western and eastern extent of the British Isles. The selected stations were: Aberporth, Camborne, Herstmonceaux West End, Larkhill, Lerwick, Nottingham Watnall and Shoeburyness Landwick. An average wind speed for the period was 7.0 m s^{-1}. The highest wind speeds are for the SW-W and N directions (oceanic circulation) and the lowest are characteristic of the easterly winds. For the FRAME runs for Poland, radiosonde data from stations Wrocław, Łeba, Warszawa (all located in Poland), Greifswald, Lindenberg (Germany), Prague (Czech Republic), Poprad (Slovakia), and Kiev (Ukraine) were used to calculate wind speed roses. For the year 1998, an average wind speed at a higher boundary layer was 7.1 m s^{-1}, whereas for 1996, it was 5.9 m s^{-1}.

3. Results

The results are organized as follows: first, the UK and PL base runs for sulphur and nitrogen deposition are presented and, afterwards, the emission scenarios runs (simulations UK2000, UK2009, PL2000 and PL2009) are compared with the FRAME base runs for year 2005. The results for various wind roses (simulations UKW, UKE and PLW, PLE) and for dry and wet years (UKdry, UKwet and PLdry, PLwet) are then presented. Each group of the FRAME runs (emission, circulation and precipitation) is presented in a separate subsection which includes the spatial patterns of the changes presented on maps, the national deposition budget calculated for each simulation and the information whether the differences between the results of a given simulation and the base run are statistically significant.

3.1. Deposition of sulphur and nitrogen compounds in Poland and the UK in the year 2005

Total deposition of oxidised sulphur and nitrogen compounds for the UK and Poland for year 2005 is shown in Fig. 2. In both countries, emission source areas have high total deposition values. Increased depositions of sulphur and nitrogen are also calculated for remote mountainous regions. This can be attributed to increased precipitation and the influence of the seeder-feeder effect. National deposition budget for the reference year 2005 is presented in Fig. 3 and 4 for the United Kingdom and Poland, respectively. The main difference between these two countries is in deposition of oxidised sulphur, which is significantly higher for Poland due to higher domestic emission and transboundary transport. In both countries, and especially in the UK, wet deposition is responsible for the majority of the deposited mass of S and N (Fig. 3-4).

Fig. 2 Total deposition of oxidized sulphur (left), oxidised nitrogen (middle) and reduced nitrogen (right) compounds in the UK and Poland for a base model run

Fig. 3 The UK national total deposition budget of oxidised sulphur, oxidised nitrogen and reduced nitrogen (dark colour – dry deposition, pale – wet) and its change relative to the reference year 2005 (in percentage)

Fig. 4 Poland national total deposition budget of oxidised sulphur (left), oxidised nitrogen (middle) and reduced nitrogen (right) (dark colour – dry deposition, pale – wet) and its change relative to the reference year 2005 (in percentage)

3.2. Changes in total deposition of sulphur and nitrogen compounds due to emissions abatement during 2000 – 2009

For the UK, emissions of SO_2, NO_x, and NH_3 in the year 2000 were at 179 %, 115 % and 107 % of the 2005 emissions. The respective values for the year 2009 were 57 %, 70 % and 93 %. Changes in emissions are reflected in the national deposition budget, but the percentage change in deposition is smaller than in emission (Fig. 3-4). This can be attributed to both: nonlinearities due to atmospheric chemistry and change in pollution export. Within this study, it is not possible to quantify these effects separately. For the UK, the changes in national deposition budget over the entire period of 2000-2009 are especially large for oxidised sulphur, and smaller for nitrogen compounds, especially for NH_x.

In Poland, emissions in the year 2000 were at 124 %, 103 % and 99 % of the 2005 values for SO_2, NO_x and NH_3 emissions, respectively. For the year 2009, the respective numbers were 71 %, 101 % and 84 %. Similarly to the UK, the total mass of oxidised sulphur deposited in Poland was decreased in the period 2000-2009, as a result of national and international emission abatements. However, the changes in nitrogen deposition are different from those calculated for the UK. Deposition of oxidised nitrogen showed a small increase when the years 2000, 2005 and 2009 are compared. For reduced nitrogen, the highest deposition was calculated for year 2005.

The changes of dry deposition budget, resulting from the emission abatements, are smaller in Poland than in the UK. This can be attributed to the differences in the source of the emissions, especially to the large share of residential combustion in sulphur and nitrogen emission in Poland. This emission sector provides 10% of NO_x and 28% of SO_2 emission in Poland (due to more common use of coal as a domestic fuel), compared to 6% and 10% in the United Kingdom. The relatively low level emissions from residential combustion result in high deposition in the vicinity of the emission sources, regardless of the annual average meteorological conditions (Kryza et al. 2010). The changes in deposition for the model runs with 2000 and 2009 emissions are statistically significant for both the UK and Poland in terms of variance (Ansari-Bradley test) and mean value (Wilcoxon test), if compared to the base runs.

3.3. The impact of annual circulation pattern on deposition of

sulphur and nitrogen compounds

There is a significant change in total deposition of sulphur and nitrogen compounds due to changes in annual circulation pattern both in the UK and Poland (Fig. 5-6). The increased frequency of westerly winds results in an overall decreased of total deposition in both countries. In contrast, the high frequency of winds from the east results in an increased total deposition budget. The spatial pattern of changes is also similar – high frequency of winds from the west results in a decreased deposition over NW and W areas of the countries. This can be attributed to the fact that the air masses from the W and NW bring relatively clean air from the ocean, especially in case of the UK. The NW and W areas of Poland and the UK suffer from higher than average deposition during the years of increased frequency of the eastern winds. The main industrial areas with high emission, both in the UK and Poland, are located in central, S and SE regions of the countries. Winds from the east transport the domestic pollutants to the N and NW areas of both Poland and the UK, which results in higher than normal deposition. For Poland, lower wind speeds are associated with easterly winds, leading to longer residence time of the domestic pollutants within the country borders and increased deposition. The differences between baseline model runs for Poland and the UK and PLW, PLE and UKW, UKE simulations are statistically significant for all chemical species considered in terms of deposition mean value (Wilcoxon test) and variance (Ansari-Bradley test).

Fig. 5 Changes in total deposition of oxidised sulphur due to changes in annual wind pattern. Left column – UKW and PLW, right – UKE and PLE (as % of total deposition in year 2005)

Fig. 6 Changes in total deposition of oxidised nitrogen due to changes in annual wind pattern. Left column – UKW and PLW, right – UKE and PLE (as % of total deposition in year 2005)

Increased frequency of westerly winds decreases the national deposition budget for both the UK and Poland if compared to the baseline model run (Fig. 3-4). Increased frequency of easterly winds changes the national deposition budget to a smaller extent, especially for the UK (up to 108 % of the year 2005 deposition budget for oxidised nitrogen). In Poland, increased frequency of eastern winds increases the deposition budget of sulphur by over 14 %, if compared with the year 2005, and the changes for nitrogen compounds are significantly smaller (Fig. 4). The largest changes in both countries are calculated for the wet deposition budget. The changes in dry deposition are small, but consistent and show an increase in dry deposition for years with increased frequency of eastern winds.

3.4. The impact of rainfall on deposition of sulphur and nitrogen compounds

Changes in deposition due to precipitation are more pronounced than changes caused by annual circulation pattern for both countries. The exception is the UK NO_y deposition, for which changes in annual circulation can influence the national deposition budget to a larger extent than precipitation. For the dry year, the total mass of deposited sulphur and nitrogen compounds is smaller than for the base run, and the decrease is at a similar level (5%) for the UK and Poland (Fig. 3-4). Decrease in total deposition during the dry year is mainly due to the decrease in wet deposition.

Dry deposition is higher for the PLdry and UKdry model runs if compared with the base run. This can be attributed to the higher concentrations of atmospheric pollutants calculated by the FRAME model in the dry year, especially for oxidised sulphur and nitrogen. The increased air concentrations can be attributed to the decreased rainfall and wet deposition, leaving more sulphur and nitrogen available for dry deposition.

Considering the wet year in Poland, the country average precipitation is 123% of the 2005 value. This results in an increase in the deposition budget to 116% for all chemical species. For the UK, the changes in deposition budget in the wet year (118% of the 2005 precipitation) vary from 115% for SO_x to 111% for nitrogen compounds (Fig. 3-4).

The spatial pattern of the changes in deposition due to precipitation is not as homogenous spatially as that calculated for the PLW, PLE and UKW, UKE model runs or the emission scenarios, but is similar for all chemical species (see SO_x presented as an example in Fig. 7). This reflects rather heterogeneous spatial changes in precipitation. During the dry year, the contribution of local individual precipitation episodes to the annual sum of rainfall (usually of convective nature, especially in Poland) was found to increase, resulting in high diurnal sums, with large differences over a short distance.

Fig. 7 Changes in total deposition of oxidised sulphur due to changes in annual precipitation pattern. Left column – UKdry and PLdry, right – UKwet and PLwet (as % of total deposition in year 2005)

4. Summary and discussion

In this study, the FRAME model with 5 km x 5 km spatial resolution has been used to quantify the role of individual meteorological parameters (precipitation, wind speed and direction) on deposition of sulphur and nitrogen compounds in Poland and the United Kingdom. The results have been compared with the changes in deposition due to national emission strategies employed over the years 2000 – 2009. The results obtained allow assessment of the importance of two important meteorological factors, precipitation and annual circulation, in shaping both the spatial pattern and national deposition budget in the United Kingdom and Poland.

In the UK, the variations in deposition due to meteorological factors are found to be relatively small if compared with the changes attributed to the emission abatements that took place over the last decade. This is the case for oxidised sulphur and nitrogen deposition. In Poland the changes in deposition due to emissions exceed the changes due to meteorological factors only for sulphur. For the nitrogen compounds in Poland, the meteorological factors, especially precipitation, modify the spatial pattern and national deposition budget to a greater extent than the emission abatements during the years 2000-2009. This is also the case for reduced nitrogen deposition in the UK. The national deposition budget of NH_x and the spatial pattern of deposition vary mainly due to changes in meteorology in both countries. Long lasting changes in atmospheric circulation, especially an increased frequency of eastern winds may result in increased deposition of S and N in terms of national deposition budget and statistically significant changes in spatial allocation of deposition. This is potentially important for environmental management in terms of ecosystem protection, as the changes may result in critical loads being exceeded over areas that are at present sufficiently protected from acidification and eutrophication, despite the ongoing emission abatements. The W and NW areas of the UK and Poland are especially at risk, due to spatial differences in the relative contribution of national and foreign emission sources to total deposition.

In the UK, the variation in annual precipitation changes the national deposition budget to a similar extent as the variation in annual circulation. In Poland, the changes in annual precipitation are much more important for the national deposition budget than the changes in annual circulation. These differences between the UK and Poland can be attributed to the "emission neighbourhood", which is more homogeneous for Poland, surrounded by countries with large emissions (except for the northern border). In the UK, the dominant direction of transboundary transport of pollutants is from SE. The oceanic air masses from N and NW are relatively clean. Therefore even small changes in the annual wind rose may result in statistically significant changes in deposition, as the chemical composition of the air coming from N and NW differs significantly when compared to the air coming from the European mainland. Despite the importance for the national deposition budget, both meteorological factors considered can result in statistically significant changes in spatial pattern of sulphur and nitrogen deposition in both countries. The changes in national deposition budget of sulphur and nitrogen in the UK and, especially, in Poland are mainly due to the changes in wet deposition. Changes in dry deposition flux are smaller when compared to wet deposition for the scenarios analysed.

The FRAME model results support earlier findings presented by Andersson et al. (2007) for Europe, that meteorological factors can change the sulphur and nitrogen deposition by c.a. 20%. It has been shown here in our study that the change of a single meteorological factor may influence both the spatial pattern of deposition and national deposition budget to a similar or higher extent than long-term international emission abatements. The findings presented here are of importance considering the climate predictions for the next years, provided by the Intergovernmental Panel on Climate Change (IPCC). According to the IPCC report, annual precipitation is very likely to increase in northern Europe (Solomon et al. 2007). The predictions for central Europe are less certain, but annual rainfall is also expected to increase especially during winter, i.e. the season of increased emissions of sulphur and nitrogen caused by residential combustion and power generation. Considering the findings reported in this paper and the IPCC predictions, it might be expected that sulphur and nitrogen deposition will increase as a result of increased precipitation, if the emission stays at the current level. Moreover, the largest wet deposition is observed over the mountainous areas that contain natural or semi-natural ecosystems sensitive to acid and eutrophying deposition (Mill, 2006). Considering the possibilities of increased precipitation and wet deposition, these areas might be affected by acidification and eutrophication, or the ecosystems recovery might be slower. Further studies are ncessary to investigate these issues, also in the context of the prediction of annual circulation changes, which are currently less certain (Solomon et al., 2007).

Emission source regions and the mountains are generally areas of high deposition of S and N in both the UK and Poland. Deposition over the mountains is especially important because of the presence of sensitive ecosystems in upland regions. In Poland, the majority of ecosystems have not yet fully recovered from the ecological disaster of the ‘80ies (when sulphur emissions were at their highest), and for over 90% of ecosystems the nutrient critical load is exceeded. The mountains and emission source areas have above-average deposition in the reference year 2005 and in all years with specific meteorological conditions (wet/dry and W/E dominated winds). This means that adequate protection of ecosystems can only be achieved by national and international emission abatements, which should also take into consideration persistent changes of meteorological conditions (e.g. an increase in annual precipitation) as these are expected to be favorable for increased deposition of atmospheric pollutants (Solomon et al., 2007). The emission scenarios for the years 2020 and 2030 suggest ongoing abatements of oxidized sulphur and nitrogen, but not for reduced nitrogen (Amann et al., 2011). In Poland and neighboring countries (e.g. Ukraine and Belarus) it is expected that NH_3 emissions will go up in the next 20 years. Considering both climate change (increased precipitation) and emission scenarios, the current state of widespread eutrophication in Poland may not improve. More effort to reduce ammonia emission is needed, primarily at national level in Poland, as domestic emissions contribute over 64% of the national NH_x deposition budget (Kryza et al., 2010).

5. Conclusions

Non-linearities in the relationship between national scale pollutant emissions and deposition occur due to the long range trans-boundary transport of pollutants, complex atmospheric chemical reactions and the influence of variable inter-annual meteorology. Understanding these processes is important for policy makers to inform decisions on control of emissions of pollutants and predict their expected impact on the natural environment. The results of this study demonstrate that sulphur and nitrogen deposition can be highly sensitive to changes in annual general circulation and precipitation. Such changes in annual meteorology can mask attempts to assess reductions in sulphur and nitrogen deposition using measurements of wet deposition from national monitoring networks. Atmospheric transport models have an advantage that they can be applied either with varying annual meteorology or with constant meteorology allowing the influence of emissions abatement and of variable meteorology to be calculated separately. The message to the environmental managers and policymakers is that the changes in meteorology should be considered in future emission control policies, as the meteorological factors are responsible for significant changes in spatial distribution of deposition, which is also supported by other studies (Giorgi and Meleux 2007, RoTAP 2009).
National scale simulations of S and N deposition in the two European countries have been undertaken with independent modification of annual pollutant emissions and meteorology.

The results show that inter-annual variability in both general circulation and total precipitation can cause major changes to atmospheric inputs to natural ecosystems. This demonstrates the need for both the application of chemical transport models and the monitoring of air pollutants over multi-year periods. Long term analysis is a necessity in order to detect trends in sulphur and nitrogen deposition caused by policy-driven emissions reductions within the natural year to year variation due to meteorology. This study also demonstrates the importance of precipitation and atmospheric circulation on deposition of S and N compounds in the UK and Poland. The persistent increase of precipitation and shift of prevailing cyclone tracks polewards resulting in an increased frequency of stagnation, may be favorable for increased S and N. The importance of these two factors was earlier shown by Jacob and Winner (2009) for ozone concentrations. This may slow down chemical and biological recovery from the effects of acid deposition and lead to increased eutrophication. However, the trends in regional climate for both countries are uncertain, especially for precipitation. Further studies on regional climate change, preferably at high spatial resolution, and climate change – long range transport of atmospheric pollutants are recommended to provide solid scientific background for policy makers and environmental managers in terms of future ecosystem protection and sulphur and nitrogen emission abatements policy.

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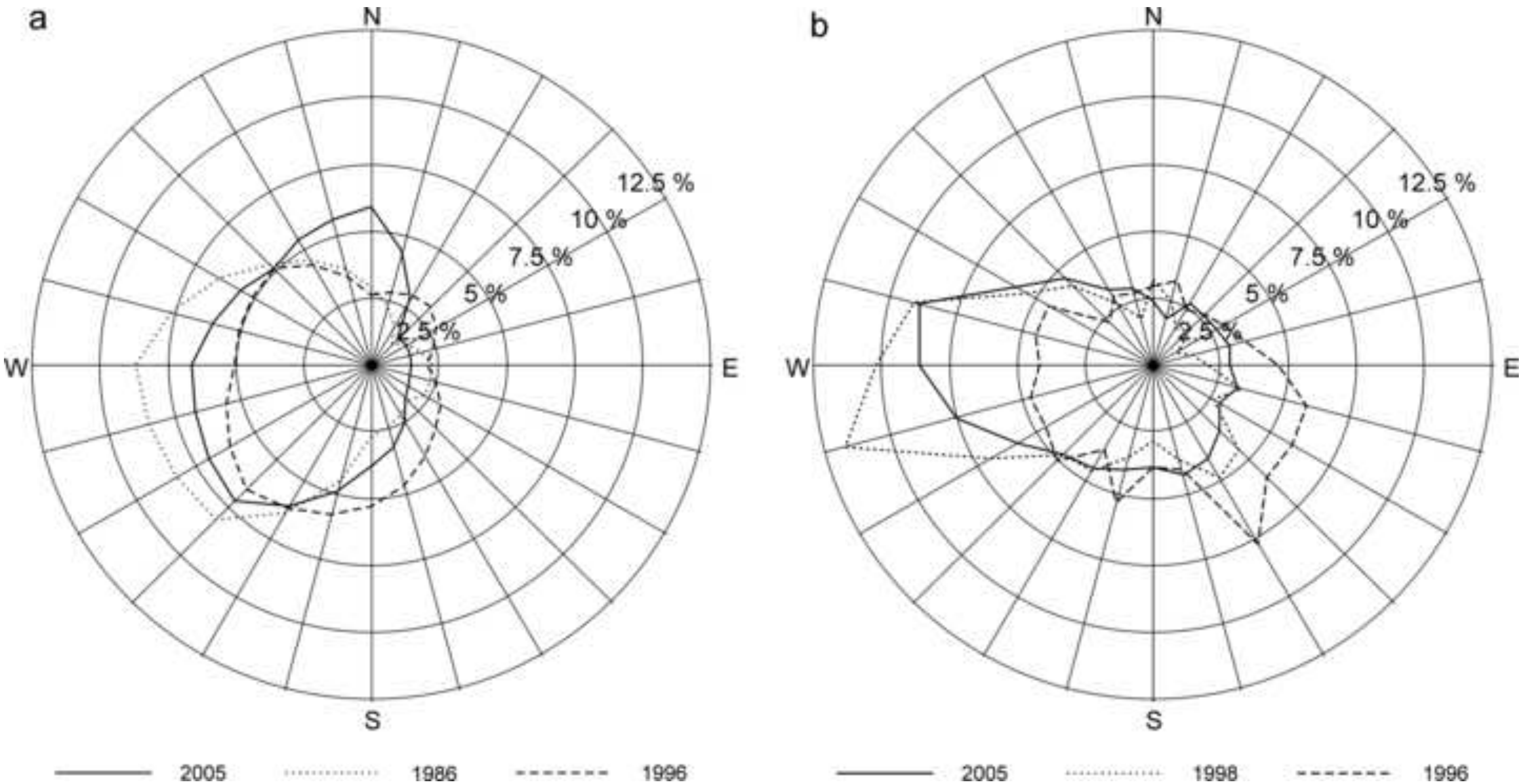

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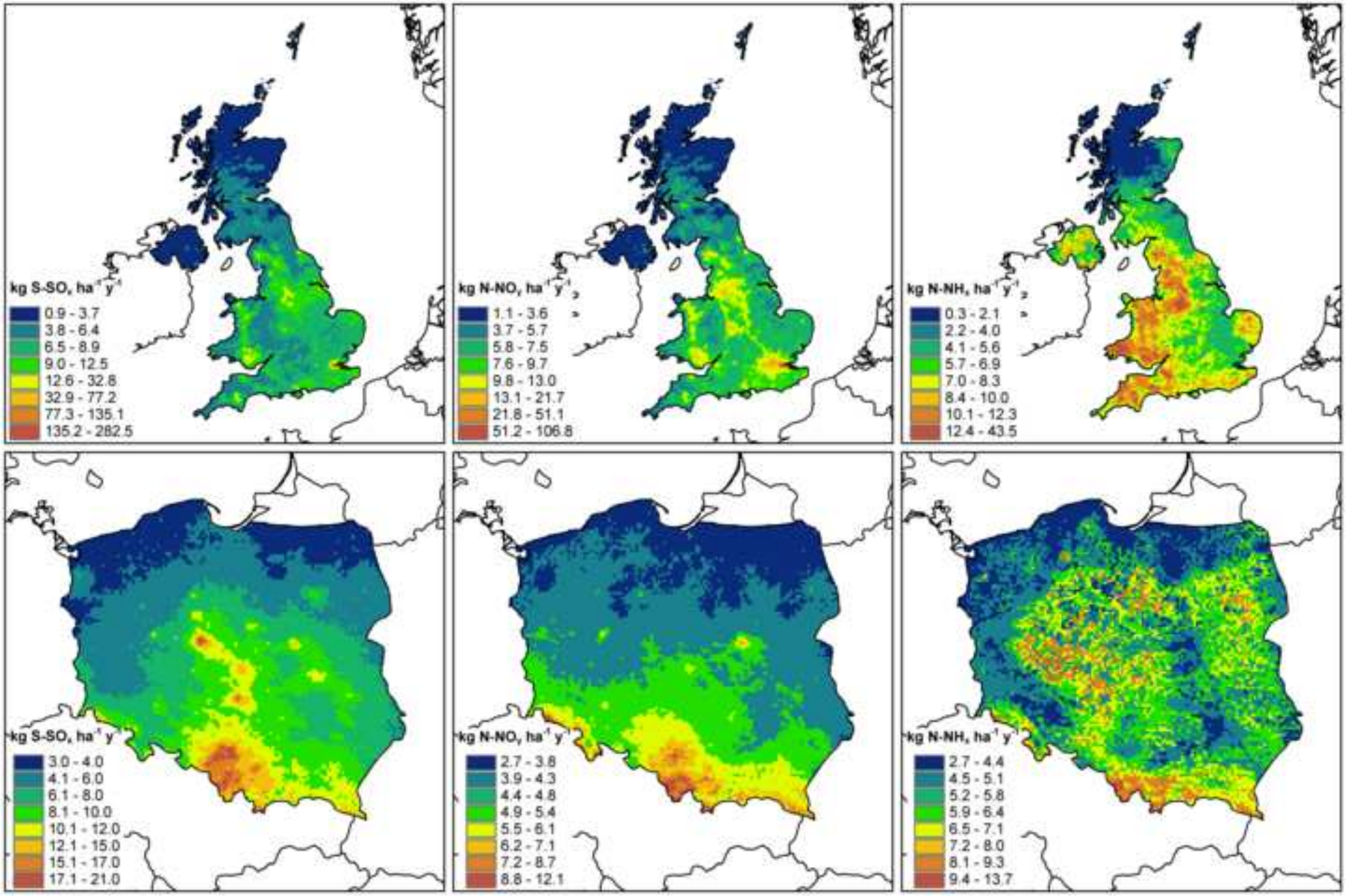

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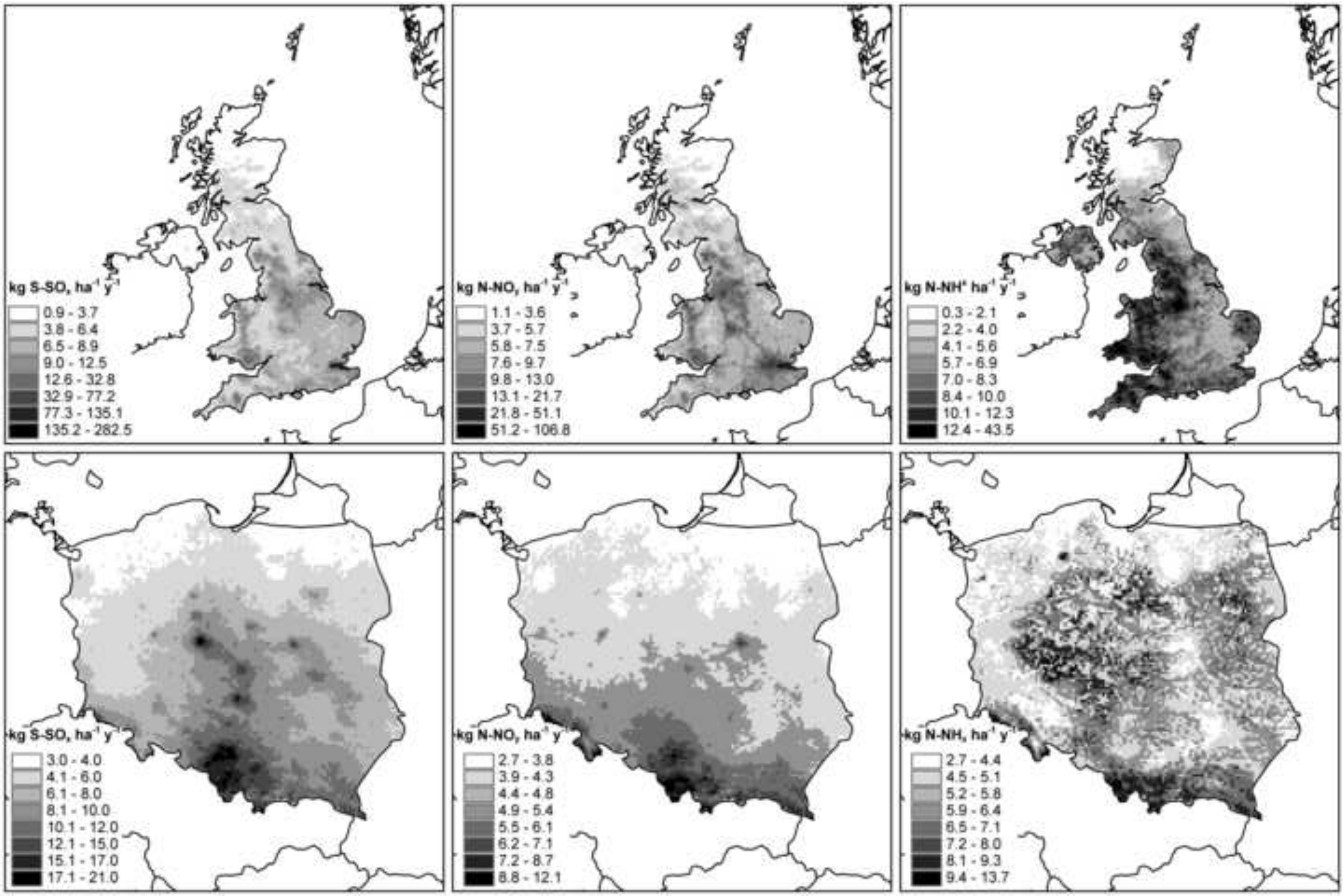

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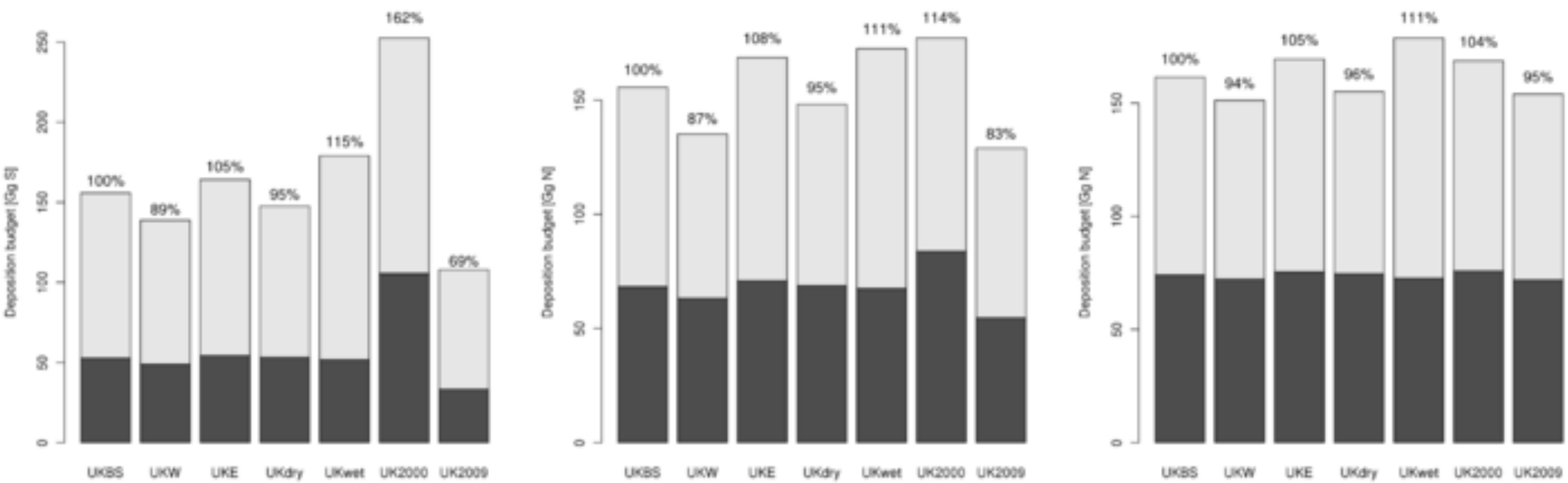

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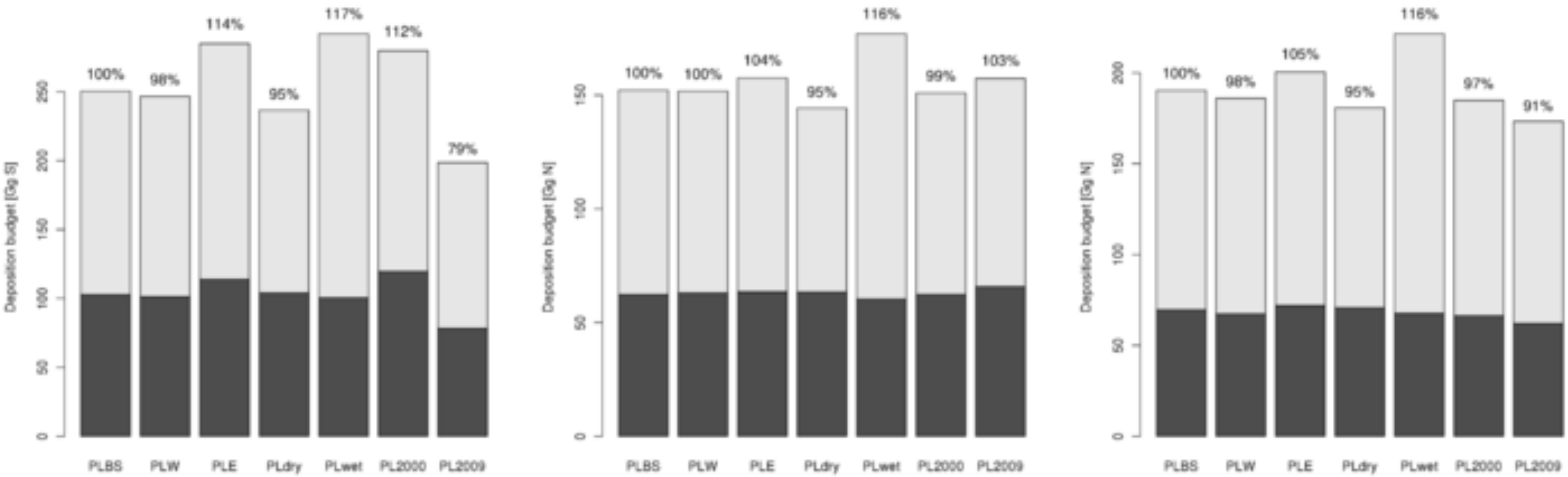

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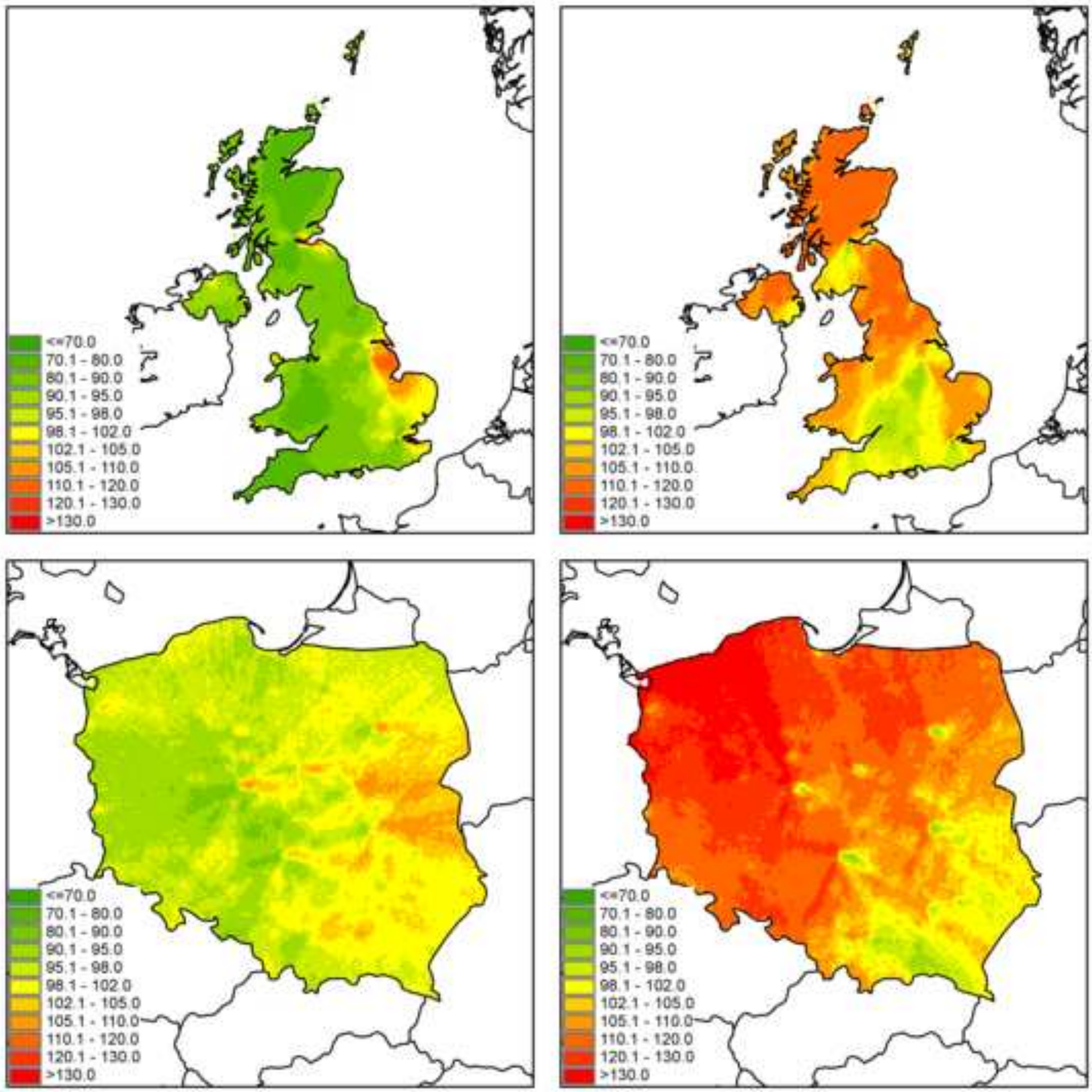

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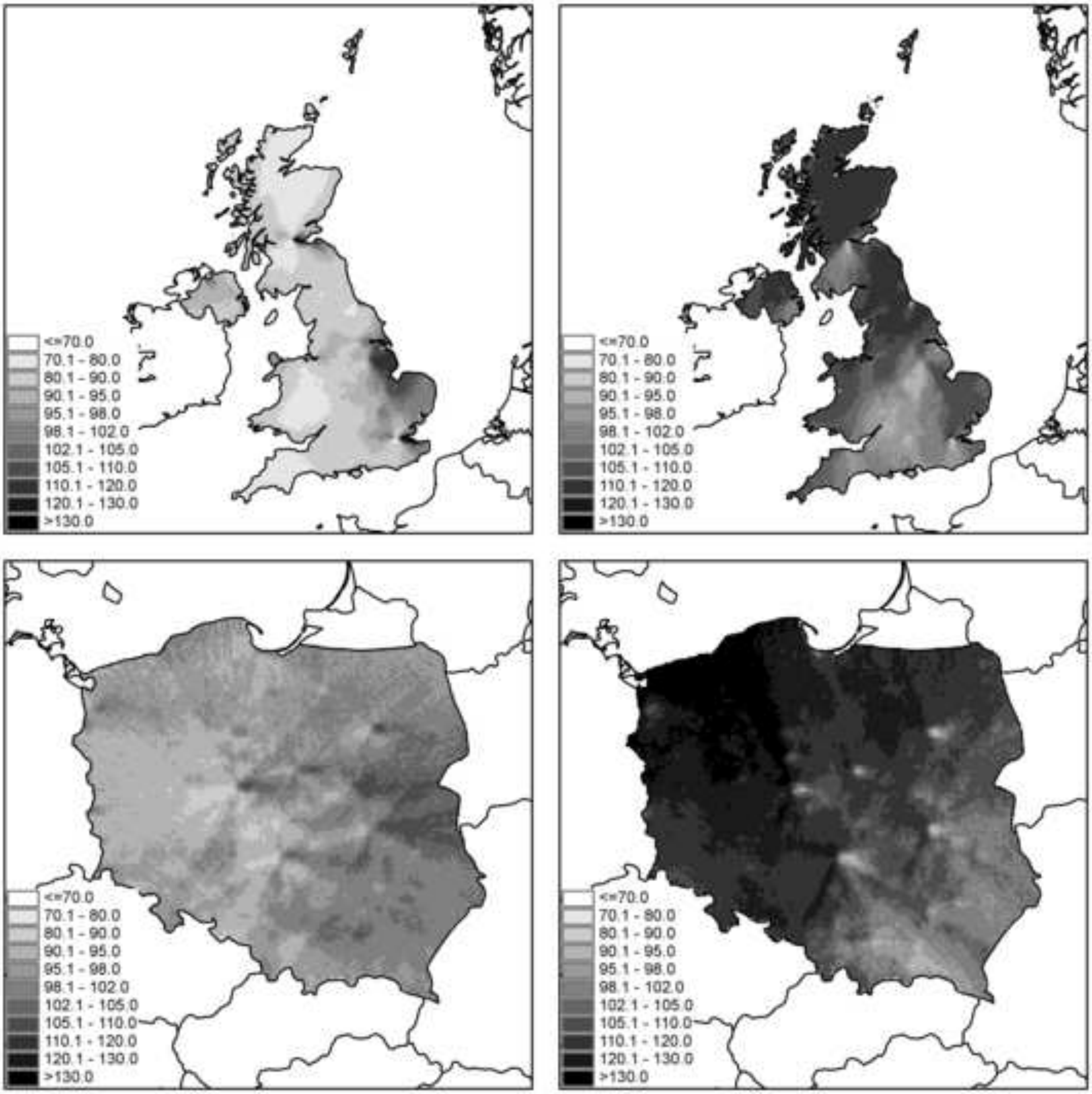

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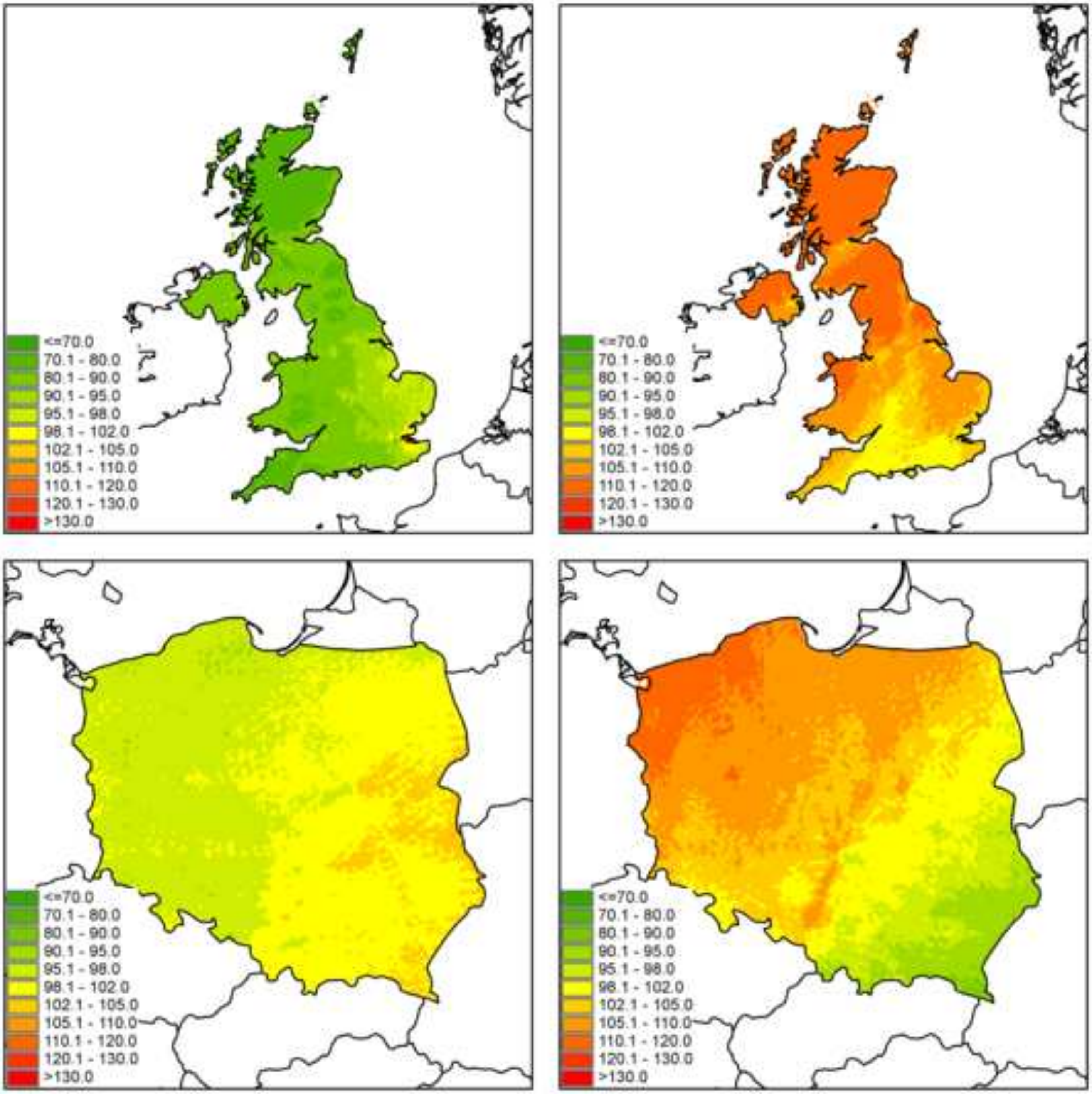

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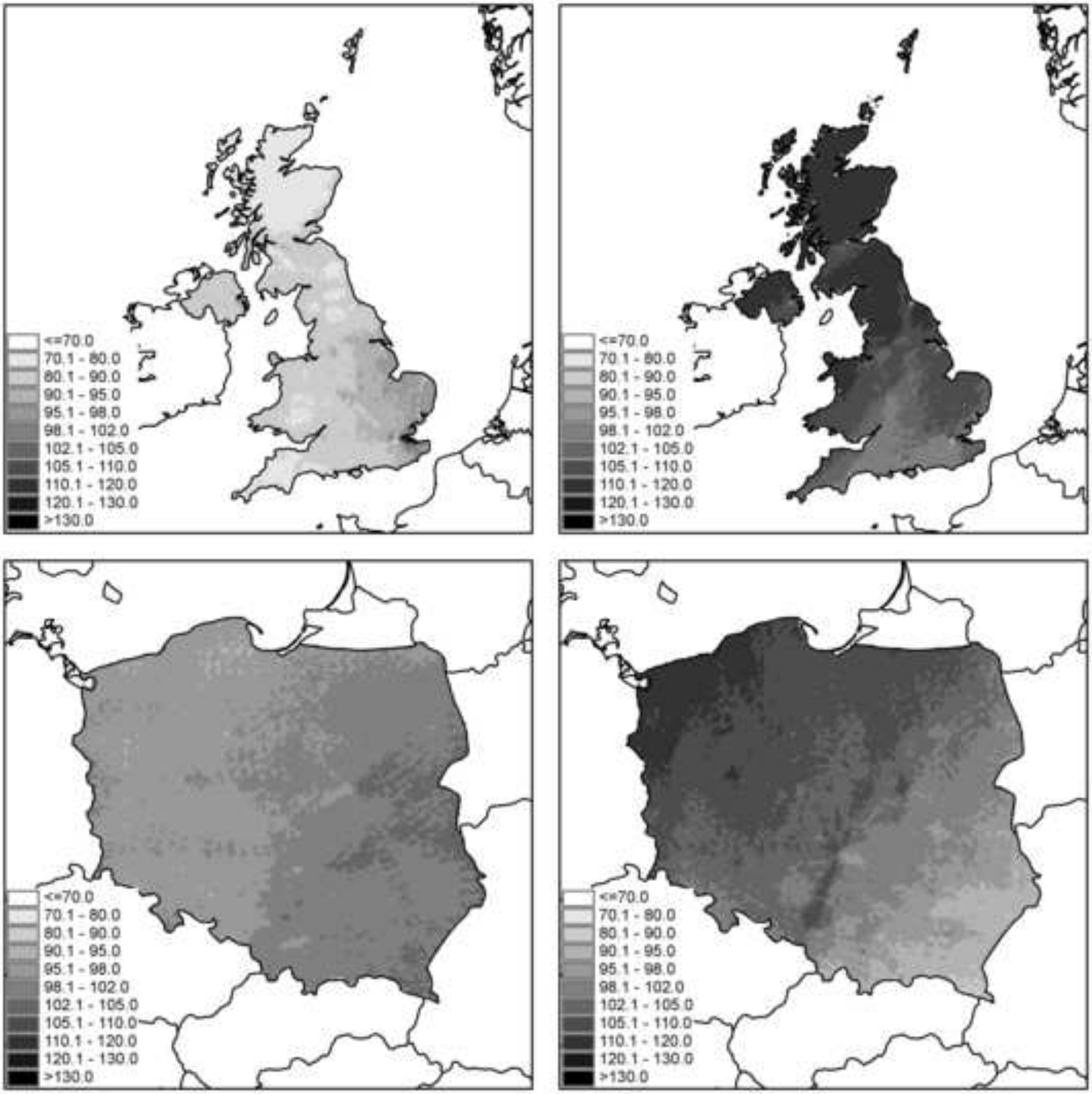

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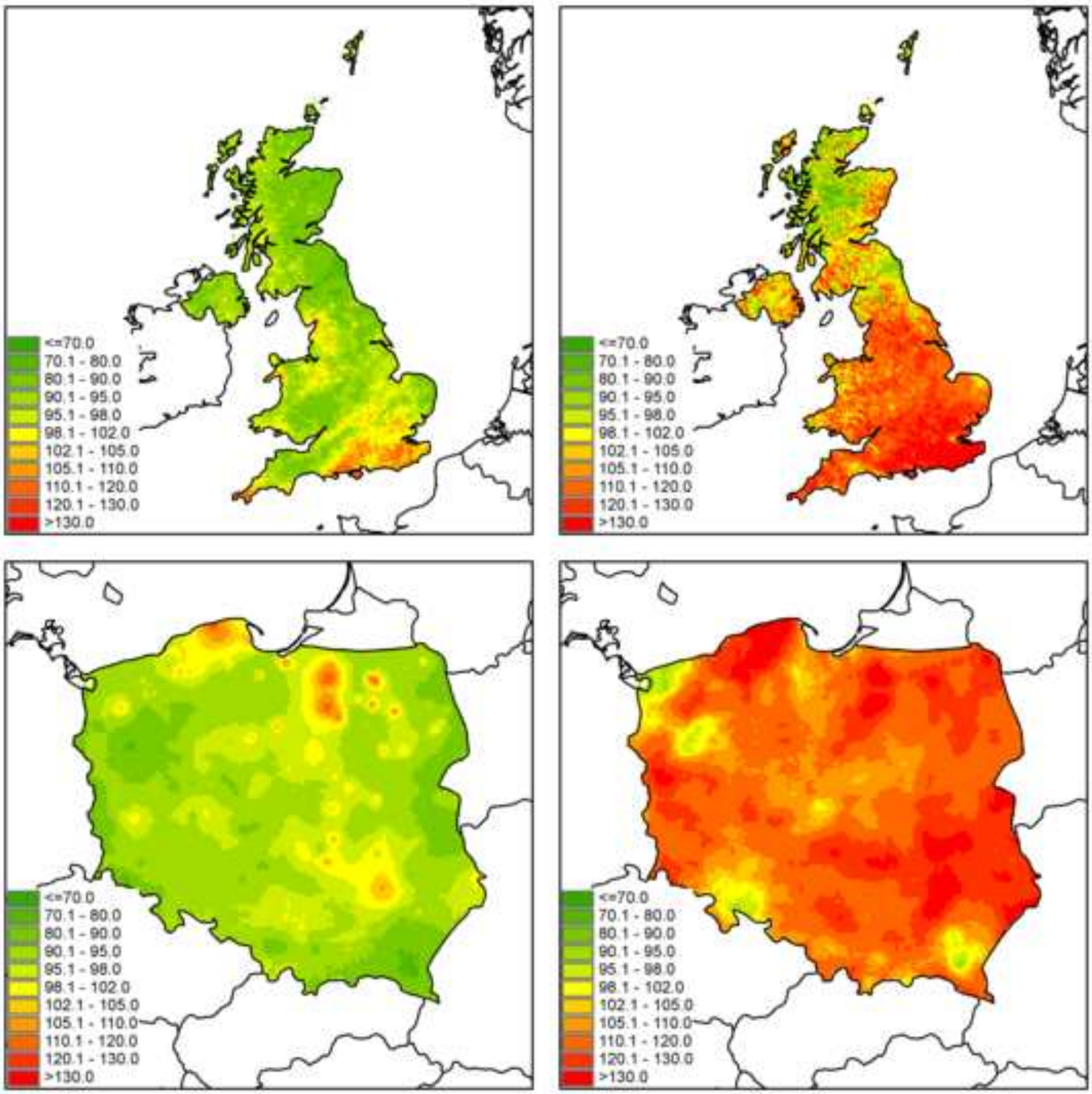

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