Modelling the Transport and Deposition of Sulphur and Reduced and Oxidised Nitrogen in the UK.

Status Report to DEFRA, as a contribution to
Long Range Transport of Pollutants in the UK

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Summary

1. The **FRAME** (**F**ine **R**esolution **A**tmospheric **M**ulti-pollutant **E**xchange) model has been applied to simulate the wet and dry deposition of sulphur and reduced and oxidised nitrogen over the United Kingdom for the years 1990, 1996 and 2010.

2. Total deposition for the different scenarios is found to be strongly correlated to the emissions. For the 2010 future emissions scenario, deposition becomes shifted towards an increased proportion of gaseous phase dry deposition and a lesser contribution from wet deposition.

3. The results of FRAME have been compared with the NEGTAP estimates of deposition budgets for the United Kingdom, to site measurements of wet deposition and gas and aerosol concentrations and to deposition maps generated by the **CEH DMP** (**D**eposition **M**apping **P**rocedure), which is based upon interpolation of measured gas concentrations and wet deposition

4. Good agreement is found between the NEGTAP and FRAME deposition budgets with some under-estimation of NO_Y dry deposition and over-estimation of NO_Y wet deposition by FRAME. Good correlation was found with measurements of wet deposition and ammonium aerosol concentrations.

5. Considerable scatter was evident in the correlation with ammonia gas concentration measurements due to the high sub-grid spatial variability for this species. For SO_2 concentrations, FRAME was successful in estimating concentrations at remote sites but less accurate for concentrations near strong sources, which again may be linked to sub-grid variability.

6. A general tendency of the FRAME model was to generate lower estimates of deposition than the DMP for the remote areas of Scotland with higher values near source areas, particularly in northern England.

7. The exceedance of critical loads for the United Kingdom was calculated for the different emissions scenarios. The present and planned abatement of pollutant gas emissions resulted in a fall in the percentage of habitats with exceedance of critical loads for acidic deposition from 81% to 36% during the period 1990 to 2010.

8. For nutrient nitrogen deposition, the percentage of habitats with exceedance of the critical load fell from 65% to 46% over the same time period of 1990-2010.

1. Description of the Model

1.1 History

The **FRAME** (**F**ine **R**esolution **A**tmospheric **M**ulti-pollutant **E**xchange) model is a Lagrangian atmospheric transport model used to assess the long-term annual mean deposition of reduced and oxidised nitrogen and sulphur over the United Kingdom. A detailed description of the FRAME model is contained in Singles *et al.* (1998). Fournier *et al.* (2002) describe the development of a parallelised version of the model with an extended domain that includes Northern Ireland and the Republic of Ireland. The model was developed from an earlier European scale model, **TERN** (**T**ransport over **E**urope of **R**educed **N**itrogen, ApSimon *et al.* 1994). FRAME was developed initially to focus in particular on transport and deposition of reduced nitrogen and was named the **F**ine **R**esolution **AM**monia **E**xchange model. Recent developments in the treatment of sulphur and oxidised nitrogen (Fournier *et al.*, 2003b) mean that it may now be considered as a robust multi-chemical species tool. The new name reflects these changes whilst preserving the familiar acronym. The current version of the model in use is FRAME 4.17.

The system used to generate UK maps of dry and wet deposition from site measurements of gas concentrations and wet deposition was previously referred to as the ‘CEH deposition model’. This has now been renamed the CEH **D**eposition **M**apping **P**rocedure (**DMP**) to distinguish it from FRAME.

1.2 Model Domain

The domain of the model covers the British Isles with a grid resolution of 5 km and grid dimensions of 172 x 244. Input gas and aerosol concentrations at the edge of the model domain are calculated using **FRAME-EUROPE**, a larger scale European simulation which was developed from TERN to run a statistical model over the entirety of Europe with a 150 km scale resolution. FRAME is a Lagrangian model that simulates an air column moving along straight-line trajectories. The atmosphere is divided into 33 separate 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 domain. Separate trajectories are run at a 1^o resolution for all grid edge points. A year-specific wind rose is used to give the appropriate weighting to directional deposition and concentration for calculation of total deposition and average concentration.

1.3 Emissions

Emissions of ammonia are estimated for each 5 km grid square using national data of farm animal numbers (cattle, poultry, pigs and sheep) as well as fertiliser application, crops and non-agricultural emissions (including traffic and contributions from human sources, wild animals etc). The ammonia emissions inventory is described in Dragosits *et al*. (1998). NH_3 is emitted into the lowest layer. Emissions of SO_2 and NO_X are from the National Atmospheric Emissions Inventory for the United Kingdom (Salway *et al.*, 1999). For SO_2 approximately 80% of 1996 emissions from the UK are associated with a small number of strong point source emissions. For NO_X, point source emissions account for approximately 25% of the total.

1.4 Plume Rise

Point source emissions of SO_2 and NO_X are treated individually with a plume rise model which uses stack height, temperature and exit velocity to calculate an 'effective emissions height' (Vieno, 2003). The plume reaches its maximum height when its temperature is equal to that of the surrounding environment and its momentum is dissipated. Buoyancy forces dominate the plume rise, which is parameterised separately for stable conditions and for neutral and unstable conditions according to the Pasquill-Gifford stability classes.

1.5 Diffusion

Diffusion of gaseous and particulate species in the vertical is calculated using K-theory eddy diffusivity and solved with a Finite Volume Method. The vertical diffusivity K_Z has a linearly increasing value up to a specified height H_Z and then remains constant (K_{max}) to the top of the boundary layer. During day time, when diffusivity depends on a combination of mechanical and convective mixing, H_Z is taken as 200 m and K_{max} is a function of the boundary layer depth and the geostrophic wind speed. At night-time these values depend on the Pasquill stability class.

1.6 Chemistry

The chemical scheme in FRAME is similar to that employed in the EMEP Lagrangian model (Barrett and Seland, 1995). 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^{--}aerosol. For oxidised nitrogen, a suite of gas phase reactions are considered. These include photolytic dissociation of NO_2, oxidation of NO by ozone, formation of PAN (peroxyacetyl nitrate) and the creation of nitric acid by reaction with the OH· free radical. NH_4NO_3 aerosol is formed by the equilibrium reaction between HNO_3 and NH_3. A second category of large nitrate aerosol is present and simulates the deposition of nitric acid on to soil dust or marine aerosol. The formation of H_2SO_4 by gas phase oxidation of SO_2 is represented by a predefined oxidation rate. H_2SO_4 then reacts with NH_3 to form ammonium sulphate aerosol. The aqueous phase reactions considered in the model include the oxidation of S(IV) by O_3, H_2O_2 and the metal catalysed reaction with O_2.

1.7 Wet Deposition

The model employs a constant drizzle approach using precipitation rates calculated from a climatological map of average annual precipitation for the British Isles. Wet deposition of chemical species are calculated using scavenging coefficients based on those used in the EMEP model. An enhanced washout rate is assumed over hill areas due to the scavenging of cloud droplets by the seeder-feeder effect. The washout rate for the orographic component of rainfall is assumed to be twice that calculated for the non-orographic component (Dore *et al.*, 1992). The model incorporates the directional dependence of orographic rainfall by considering two components of rainfall: non-orographic precipitation which has no directional dependence and orographic precipitation which is directionally dependent and stronger for wind directions associated with humid air masses. The directional orographic rainfall model is described in detail in Fournier *et. al*, 2001 and 2003(a).

1.8 Dry Deposition

Dry deposition of SO_2, NO_2 and NH_3 is calculated individually to five different land categories (arable, forest, moor-land, grassland and urban). For ammonia, deposition is calculated individually at each grid square using a canopy resistance model (Singles *et al.,* 1998). The deposition velocity is generated from the sums of the aerodynamic resistance, the laminar boundary layer resistance and the surface resistance. Dry deposition of SO_2 and NO_2 is calculated using maps of deposition velocity derived by the CEH 'big leaf' model (Smith *et al.* 2000), which takes account of surface properties as well as the geographical and altitudinal variation of wind-speed. Other species are assigned constant values of deposition velocity.

1.9 Diurnal Cycle

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. At night-time, a single fixed value is used for the boundary layer depth according to Pasquill stability class and surface windspeed.

1.10 Wind Rose

The wind rose employed in FRAME uses 6-hourly operational radiosonde data from the stations of Stornoway, Hillsborough, Camborne and Valentia spanning a ten year period (1991-2000) to establish the frequency and harmonic mean wind speed as a function of direction for the British Isles. This is illustrated in figures 1(a) and 1(b) for data averaged over the ten year period.

1.11 Computational Performance

The model code is written in High Performance Fortran 90 and executed in parallel on a Linux Beowulf cluster comprising of 60 dual processors. Run time for a simulation employing 50 processors is approximately 15 minutes.

2. Results of FRAME Simulations

This status assessment of FRAME describes the use of emissions inventories corresponding to the years 1990, 1996 and 2010. For the latter scenario, emissions were scaled to meet the requirements of the National Emissions Ceiling Directive. The total UK emissions are illustrated in Table 1.

Figures 1(a)-(f) illustrate maps of wet deposition and grid-averaged dry deposition for reduced nitrogen, oxidised nitrogen and oxidised sulphur calculated by FRAME using a 1996 emissions inventory. Wet deposition occurs primarily due to the washout of aerosol particles from the atmosphere by precipitation. As aerosol particles are associated with long range transport, the areas of high wet deposition are not related to areas of high emissions of primary pollutants but more closely correlated to areas of high rainfall. In particular the mountain areas of the Pennines and north Wales are notable for high deposition which exceeds 10 kg N Ha^{-1} for NH_X and NO_Y and 15 kg S Ha^{-1} for SO_X. Figure 2(d) illustrates the grid-averaged dry deposition of NH_X, which occurs mostly due to NH_3 gas. Due to the low-level emissions of NH_3 and its reactive nature, much ammonia is re-deposited in to the grid square of emissions, so that the deposition pattern is closely correlated to the spatial distribution of emissions. Dry deposition of NO_Y occurs mostly due to deposition of gaseous NO_X. As illustrated in figure 2(e), NO_Y deposition is highest in the greater London region where emissions from vehicle exhausts are greatest. Dry deposition of SO_X is associated with SO_2 gas. For the year 1996, approximately 80% of SO_2 emissions are associated with large power plants. Many of these are located in northern England, which is seen to be the area of highest SO_X dry deposition.

	1990	1996	2010
NH_3 (kT) N	306	285	245
NO_X (kT) N	840	615	355
SO_2 (kT) S	1860	1014	293

Table 1. Total UK emissions of NH_3, NO_X and SO_2 for the model simulation years

The budgets for total deposition of pollutants to the United Kingdom for the different model simulation years are illustrated in Table 2(a). These are compared with the values calculated by the **N**ational **E**xpert **G**roup on **T**ransboundary **A**ir **P**ollution (NEGTAP, 2001). NEGTAP budgets are based on weekly measurements of wet deposition at 32 stations as well as gas concentrations at national monitoring stations across the U.K. The 3-yearly annual average measurement values are interpolated across the country with the application of appropriate deposition velocities to gas concentrations using the **DMP** (**D**eposition **M**apping **P**rocedure**,** Smith *et al.*, 2000) to generate maps of wet and dry deposition. A very close agreement is found between the NEGTAP and FRAME budgets for both wet and dry deposition of NH_X. This suggests that FRAME is able to accurately represent the rates of vertical mixing and dry deposition of ammonia as well as the chemical transformation rates to ammonium aerosol and washout rates of gas phase and particle phase reduced nitrogen. For NO_Y, FRAME gives a lower value for dry deposition and a higher value for wet deposition than NEGTAP. This suggests that the gas phase species of oxidised nitrogen in FRAME (which dominate the dry deposition component) may be converted too rapidly to particulate form (which is the predominant component of wet deposition. In particular, FRAME has been found to predict much lower values of nitric acid concentrations than those measured. The NEGTAP contribution of nitric acid deposition (65 kT) was found to be the major component of NO_Y dry deposition.

Comparing the change in deposition for FRAME between 1990 and 2010, the total deposition of NH_X to the United Kingdom is seen to decrease by 21% from 214 kT to 168 kT N, a similar change to the 20% decrease in emissions. For NO_Y, the total deposition decreases by 58% from 239 to 100 kT N over the same period. Again the deposition decrease is the same as the 58% decrease in UK emissions. For SO_X, the total UK deposition decreases by 82% from 625 to 111 kT, which is similar to the 84% decrease in emissions. A strong linearity in the relationship between emissions and deposition is thus evident for all three chemical components. A temporal change is apparent however in the relative contributions of wet and dry deposition. For 1990, wet deposition contributes more to total deposition of reduced nitrogen than dry deposition. For the year 2010, this balance is reversed. For 1990, wet deposition is the greater component in the total depositions of oxidised nitrogen and sulphur. For 2010, the balance is shifted further towards the dominance of wet deposition. This trend can be explained in terms of the reduction in gaseous emissions generally slowing down the rates of gas to particle conversion so that pollutants are preferentially dry deposited in their primary gaseous phase state. This has implications for long range transport of pollutants. Gases tend to be locally dry deposited whilst particles are deposited more slowly through washout by precipitation and can travel greater distances. The shift towards the gaseous phase thus corresponds to a reduction in transport distance of pollutants.

(kT N or S/year)	NEGTAP 95-97	FRAME 1990	FRAME 95-97	FRAME 2010
NH_X Dry Dep	99	100	97	88
NH_X Wet Dep	110	114	110	80
NH_X Total Dep	209	214	207	168
NO_Y Dry Dep	87	95	63	36
NO_Y Wet Dep	91	144	132	64
NO_Y Total Dep	178	239	195	100
SO_X Dry Dep	125	311	130	39
SO_X Wet Dep	171	314	240	72
SO_X Total Dep	296	625	370	111

Table 2(a). 1990, 1995-1997 & 2010 UK annual deposition budgets for the model and for NEGTAP (National Expert Group on Trans-boundary Air Pollution) for 1995-1997.

UK budget (Gg N or S/year)	NEGTAP			FRAME		
	NH_X	NO_Y	SO_X	NH_X	NO_Y	SO_X
Import	30	60	90	58	155	135
Emissions	282	615	1014	285	615	1014
Dry Dep’	99	87	125	97	63	130
Wet Dep’	110	91	171	110	132	240
Export	103	497	808	136	575	779

Table 2(b) UK annual budgets for import, export, emissions and wet and dry deposition for the model and for NEGTAP for 1995-1997.

At the start of each trajectory in FRAME, atmospheric aerosol and gas loadings are initialised with the concentrations generated by FRAME-Europe which is run on the EMEP European grid. Table 2(b) illustrates the components of import and export calculated by FRAME for the United Kingdom, as compared to NEGTAP. FRAME Generally estimates greater import than NEGTAP. The relative amount of import compared to the sum of national emissions and import are 17%, 20% and 12% for NH_X, NO_Y and SO_X respectively. It is evident that the United Kingdom is a major net exporter of reduced and oxidised nitrogen and sulphur.

Correlation plots of the model compared to measurements are illustrated in Figure 3. For wet deposition, figures 3(a)-3(c), good correlation coefficients of R^2 are obtained with values of 0.78, 0.70 and 0.63 for NH_X, NO_Y and SO_X respectively. The slope of the graphs is good for NH_X (0.90) and SO_X (0.89) though there is a tendency for FRAME to overestimate the wet deposition of NO_Y (slope 1.22). Figure 3(d) shows the correlation between modelled and measured ammonia concentrations. There is a tendency for the model to overestimate the concentrations and considerable scatter is evident. This can be attributed in part to the fine scale variability in ammonia concentrations, which occurs on a sub-5km scale. As the model estimates grid-square average concentrations, measurements will be sensitive to the presence or absence of local ammonia sources. A better correlation of $R^2=0.79$ is obtained for ammonium aerosol concentrations, which have less fine scale spatial resolution, as shown in figure 3(e). The tendency of FRAME to overestimate NH_3 concentrations and underestimate NH_4^+ concentrations may suggest that gas to aerosol conversion is proceeding too slowly in the model. Figures 3(f) and 3(g) show the correlations of modelled to measured NO_2 and SO_2 concentrations. The slopes of both curves are 1.0 with R^2 values of 0.57 and 0.47 respectively. Outlying points are evident particularly in areas of high emissions where the simple meteorology employed in FRAME is not able to accurately represent the three dimensional motion and dispersion of a plume emitted from a stack.

Figures 4(a)-4(g) illustrate an alternative approach to comparing the model with measurements. The ratio plots show for each 5km grid square in the UK the ratio of the FRAME modelled value of deposition compared to that generated by the measurement-interpolation procedure employed in the DMP. Ratios of 1.0 correspond to an exact agreement between the two systems. In general the target ratio may be considered to lie between the values 0.5 and 1.5, which corresponds to grid squares where FRAME deposition values are within +/- 50% of those generated by DMP. Figure 4(a) shows that a very good spatial correlation is obtained for wet deposition of NH_X across the U.K. FRAME tends to estimate higher values in south-east England and lower values in northern Scotland than DMP. Similarly figures 4(b) and 4(c) show higher wet deposition of nitrate and sulphate in southeast England and lower values in northern Scotland with FRAME than for the measurement system. This effect may be explained by the straight line trajectories employed in FRAME-Europe which predict high import of pollutants from Europe for south-easterly trajectories and low concentrations for westerly trajectories. In practice, curved trajectories may cause re-circulation of European air masses, particularly when high-pressure systems are present over northern Europe. Figure 4(d) shows that FRAME gives higher deposition values of reduced nitrogen than DMP in England and lower values in the remote areas of Scotland. For dry deposition of oxidised nitrogen, figure 4(e), FRAME values tend to be significantly higher than those for DMP, as the dry deposition of nitric acid was not incorporated in to this data set for the DMP model. Dry sulphate deposition tends to be higher for FRAME in the source regions of northern England and lower in the remoter regions of south-western England and Scotland, suggesting that SO_2 may be too rapidly dry deposited by FRAME.

3. Calculation of Critical Load Exceedance for Acidification and Eutrophication

The deposition data generated by FRAME may be used to calculate the exceedance of critical loads of acidity and nitrogen to different habitats in the UK. The deposition data were ‘calibrated’ to the measurement-based estimates of the DMP. This technique assumes that the best available estimate of annual wet and dry deposition for the period 1995-1997 is obtained from the measurement-based data for this period. Having established such a baseline data set, the model is run with a 1996 emissions scenario as well as for the 1990 and 2010 emissions scenarios. Estimates of deposition for future and past scenarios are made by applying the modelled relative change in wet and dry deposition for each grid square in the UK domain to the measurement-based data set for 1995-1997. This calibration approach ensures consistency between the official 1995-1997 deposition data and modelled past and future deposition data sets. In this study, however, we specifically consider the influence of calibration on calculations of exceedance of critical loads .

To assess the impacts of different deposition scenarios on critical loads exceedance for nutrient nitrogen and acidic deposition, the National Focal Centre (NFC) for critical loads at CEH Monks Wood has designed a suite of programs in ARC/INFO macro language (AML), linked via a C program (Hall *et al*., 2001). The program suite is referred to as EXCEED and was originally created in November 1998, but has since undergone a series of modifications to continually address the needs of Defra. EXCEED imports the appropriate deposition fields, the 1 km habitat areas and 1 km habitat-specific critical loads. It calculates acidity exceedances via the Critical Loads Function and nutrient nitrogen exceedances separately. The exceedances are calculated at 1 km resolution for each habitat. Therefore deposition data are also treated as 1 km data by assuming that the deposition values remain constant across the larger 5km FRAME grid squares. This exceedance information is stored and used by the program to calculate:

- The area of each ecosystem exceeded in England, Wales, Scotland, NI, GB and UK.
- The Accumulated Exceedance (AE) values for each ecosystem in England, Wales, Scotland, NI, GB and UK.
  AE = exceeded area * exceedance value (keq/year = ha * keq/ha/year)

In addition, this information is summed for each 5 km grid square, to enable maps to be produced, which show:

- The total area of ecosystems exceeded in each 5 km square of the UK.
- The total Accumulated Exceedance (AE) values for all ecosystems in each 5 km square.

DMP and FRAME deposition data were applied to the critical loads habitat categories using the following convention:

- Moorland deposition was used for all low-growing vegetation (acid grassland, calcareous grassland, dwarf shrub heath, bog, montane and supralittoral sediments (coastal dune grasslands)).
- Woodland deposition was used for all woodland habitats (managed coniferous woodland, managed broadleaved woodland, unmanaged woodland and Atlantic Oaks).

- Average deposition was used for freshwaters, which have catchment areas of mixed habitat types.

Work is currently being undertaken to generate a new data set of land classification for use in DMP and FRAME with an expanded number of land types. This will include a distinction between intensively managed, semi-improved and natural grassland and between broad-leafed and deciduous woodland. In the future this should allow a closer match between deposition land classes and those used for critical loads calculations.

The national habitat critical loads used for this exercise are the data for February 2003 (Hall *et al*., 2003a). The habitats for which critical loads and exceedances are calculated are also described in detail in Hall *et al*., (2003a). In the tables 3(a)-3(d) illustrated below, the coniferous and broadleaved woodland categories refer to managed woodland only. The unmanaged woods consist of both coniferous and broadleaved woodland. For nutrient nitrogen, the critical loads for this habitat are set to protect the woodland ground flora. Nutrient nitrogen critical loads for Atlantic oak woods are set to protect epiphytic lichens in this woodland habitat. Freshwater exceedance statistics refer only to the catchment areas of 1610 sites sampled throughout the UK, not all UK freshwaters. Further information on these habitat critical loads and exceedances can be found in Hall *et al*., 2003 a and b.

Table 3(a) shows the percentage habitat areas in the UK for which the critical loads for acid deposition are exceeded for the DMP data for 1995-1997 and calibrated FRAME data for 1990 and 2010. The trend towards reduced exceedances over the period of two decades is noticeable, with the percentage area exceeded for coniferous woodland being halved (from 95% to 45%). This is mostly attributed to the large reductions of SO_2 and NO_X emissions, which have already occurred and are further envisaged by 2010. Nonetheless, significant areas of habitats remain with exceedances forecast for 2010.

Broad habitat	FRAME calibrated 1990	DMP 1995-1997	FRAME calibrated 2010
Acid grassland	92	86	63
Calcareous grassland	11	0	0
Dwarf shrub heath	75	56	17
Bog	84	72	34
Montane	95	87	35
Coniferous woodland	95	73	45
Broadleaved woodland	86	74	53
Unmanaged woods	85	61	33
Freshwaters	26	23	11
All habitats	81	67	36

Table 3(a) Statistics for percentage areas of habitats in the UK with exceedance of acidity critical loads for 1990 calibrated FRAME data, DMP data for 1995-1997, and 2010 calibrated FRAME data.

Table 3(a) illustrates the areas of ecosystems exceeded but does not indicate the magnitude of exceedance. The accumulated exceedance is a better measure of the amount by which acidic deposition exceeds critical loads. The data is presented in this format in Table 3(b) for the purposes of comparison of the magnitude of exceedance generated by FRAME and by DMP for 1995-97. It can be seen that the FRAME data generates higher values of accumulated exceedance for many of the ecosystems (excepting managed woodland), notably acid grassland, dwarf shrub heath and bog. Despite the smaller areas of ecosystem exceeded with the FRAME data (57%), than with the DMP data (67%), FRAME generally gives larger accumulated exceedances. This illustrates that in FRAME there is a tendency for material to be deposited closer to source, in polluted areas. With the DMP data there is a smoother distribution, producing lower deposition in polluted regions and greater deposition in cleaner regions than in FRAME. For the year 2010, it can be seen that applying the calibration procedure results in a small increase in the accumulated exceedance of critical loads for acidification for some habitats and a decrease for others.

Broad habitat	DMP 1995-1997	FRAME 1995-1997	FRAME 2010 calibrated	FRAME 2010 uncalibrated
Acid grassland	1700	2000	540	620
Calcareous grassland	0	10	0	0
Dwarf shrub heath	950	1200	150	220
Bog	350	450	100	130
Montane	190	180	30	26
Coniferous woodland	830	640	300	160
Broadleaved woodland	920	910	350	310
Unmanaged woods	310	320	92	85
Freshwaters	68	92	19	21
All habitats	5300	5800	1600	1600

Table 3(b) Statistics for accumulated exceedance of acidic deposition (M eq year^{-1}) to ecosystems in the United Kingdom for DMP data and FRAME for 1995-1997 and for calibrated and uncalibrated FRAME data for 2010.

Table 3(c) illustrates the percentage of habitat areas with exceedance of critical loads for nutrient nitrogen to various habitats in the UK for the official data sets. There is again a temporal trend towards lower values of exceedance with the percentage area of all habitats exceeded falling from 65% to 46% between 1990 and 2010. The decrease however is smaller than for acidification because the decreases in ammonia and oxidised nitrogen emissions are less than those for sulphur dioxide.

Broad habitat	FRAME calibrated 1990	DMP model 1995-1997	FRAME calibrated 2010
Acid grassland	70	67	42
Calcareous grassland	84	77	29
Dwarf shrub heath	37	35	19
Bog	47	46	39
Montane	86	82	49
Coniferous woodland	94	93	87
Broadleaved woodland	98	98	96
Unmanaged woods	96	96	93
Atlantic oak	90	84	68
Supralittoral sediment	59	52	22
All habitats	65	63	46

Table 3(c) Statistics for percentage area of habitats in the UKwith exceedance of nutrient nitrogen critical loads for 1990 calibrated FRAME data, DMP data for 1995-1997 and 2010 calibrated FRAME data.

Accumulated exceedances of nutrient nitrogen deposition are shown for FRAME and DMP data for 1995-97 in Table 3(d). For this data set, FRAME generates larger accumulated exceedances for certain habitats (acid grassland, calcareous grassland, dwarf shrub heath, bog and supralittoral sediment) but lower accumulated exceedances for woodland habitats. For most habitats the difference between the FRAME and DMP accumulated exceedances are relatively small. FRAME however generally generates smaller percentage area exceedances (57% for all habitats) compared to DMP (63%). It is important to note that the figures for the UK mask the spatial differences between FRAME and DMP. There is a tendency for FRAME to estimate greater values of exceedance of nutrient nitrogen critical loads, than DMP for England and Northern Ireland and lower values for Scotland and Wales. This is evident from figure 4(d), which shows a map of the dry deposition ratio for reduced nitrogen, which makes a major contribution to nutrient nitrogen deposition, particularly for moorland and forest ecosystems. The exceedance of supralittoral sediment is notable in having a significantly higher value for the area exceeded with the FRAME data. This habitat occurs specifically along the coastline where FRAME deposition is generally higher than DMP. Also for the year 2010, it can be seen that the calibration procedure has a relatively small influence on the accumulated exceedance for nutrient nitrogen deposition, representing an increase from 2000 to 2300 Meq year^{-1}.

Broad habitat	DMP 1995-1997	FRAME 1995-1997	FRAME 2010 calibrated	FRAME 2010 uncalibrated
Acid grassland	550	650	190	250
Calcareous grassland	96	230	23	91
Dwarf shrub heath	410	500	140	180
Bog	190	220	100	110
Montane	90	80	33	20
Coniferous woodland	870	530	560	260
Broadleaved woodland	1200	1100	880	730
Unmanaged woods	490	470	350	300
Atlantic Oak	74	48	48	28
Supralittoral sediment	31	80	8	39
All habitats	4000	3900	2300	2000

Table 3(d) Statistics for accumulated exceedance of nutrient nitrogen deposition (M eq year^{-1}) to ecosystems in the United Kingdom, for DMP and FRAME data for 1995-1997 and calibrated and uncalibrated FRAME data for 2010.

Figures 5(a) to 5(c) illustrate maps of exceedance of critical loads for acidic deposition and nutrient nitrogen deposition for FRAME calibrated data for 1990 and 2010 and for DMP data for 1995-1997. For acidic deposition, it is evident that large ecosystem areas in Scotland, Wales and northern England are subjected to critical loads exceedance. For many 5 km grid squares, the accumulated exceedance for 1990 is greater than 2000 keq year^{-1}. The situation is improved for the 1995-1997 data set, with further reductions in exceedance by 2010. It is notable for 2010 that large areas in northern Scotland and southern England are no longer exceeded although areas with exceedance of critical loads remain in northern England and Wales. A major reduction in the magnitude of accumulated exceedance can be observed with few grid squares having exceedance greater than 500 keq year^{-1}. For nutrient nitrogen, significant areas in Scotland, northern England and Wales can be seen to have exceedance of critical loads in the range 1000-2000 keq year^{-1}. For the year 2010, major improvements are seen in Scotland where the area of habitats with exceedance of nutrient nitrogen critical loads is greatly reduced. Furthermore the magnitude of accumulated exceedance is reduced such that, for most grid squares, this is below 500 keq year^{-1}. A relatively small number of 5 km grid squares remain with accumulated exceedances above 1000 keq year^{-1}.

Finally, it should be noted that the national habitat critical load maps are based on empirical or steady-state mass balance methods, used to define critical loads for systems at steady-state. Therefore exceedance of these critical loads is an indication of the potential for harmful effects to systems at steady-state, and a habitat that is currently exceeding its critical load is not necessarily already showing the signs of damage. In addition, reducing deposition to below the critical load does not mean the ecosystems immediately recover. There are time lags before chemical recovery takes place, and further delays before biological recovery. The timescales for both chemical and biological recovery could be very long, particularly for the most sensitive habitats.

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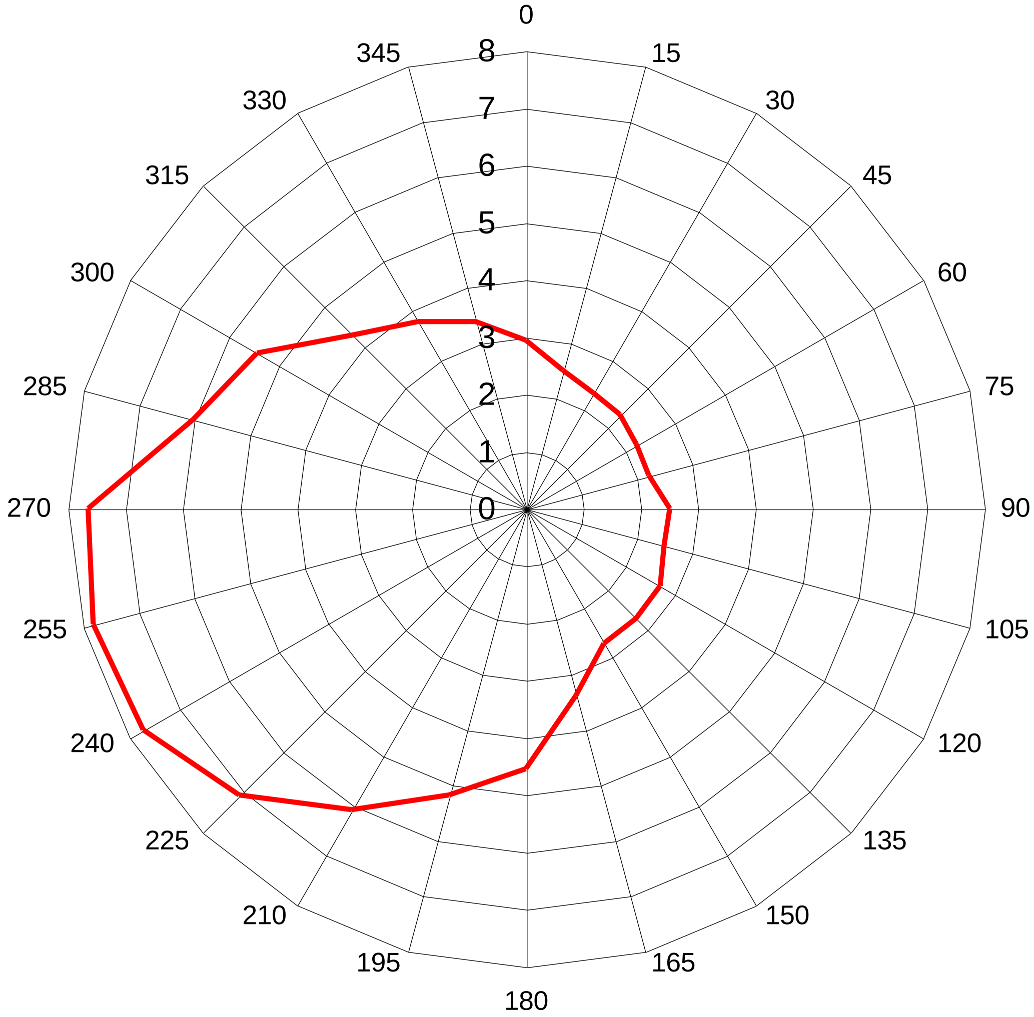


Figure 1(a). Frequency of wind (%) as a function of wind direction calculated from radiosonde data for the period 1991-2000.

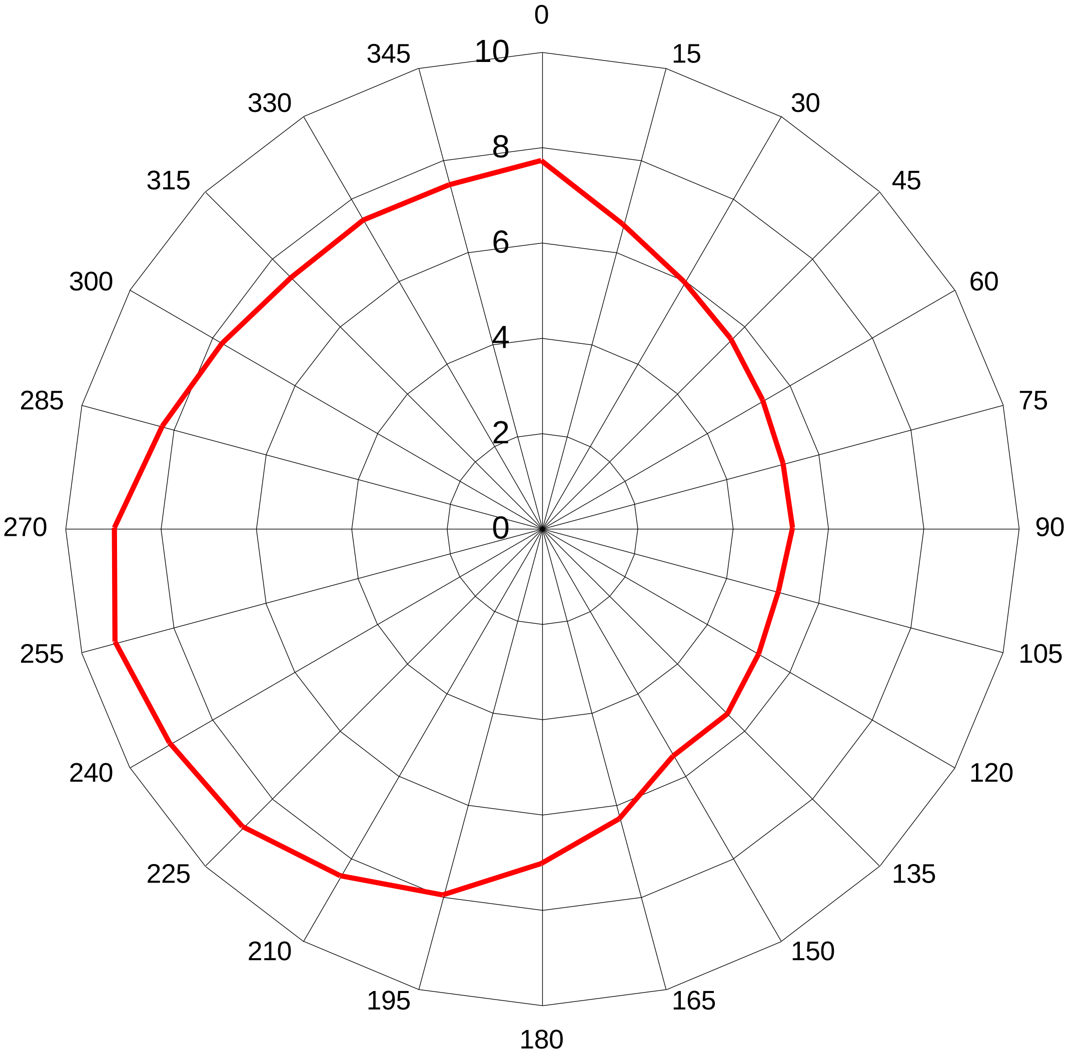


Figure 1(b). Wind speed (m s^{-1}) as a function of wind direction calculated from radiosonde data for the period 1991-2000.

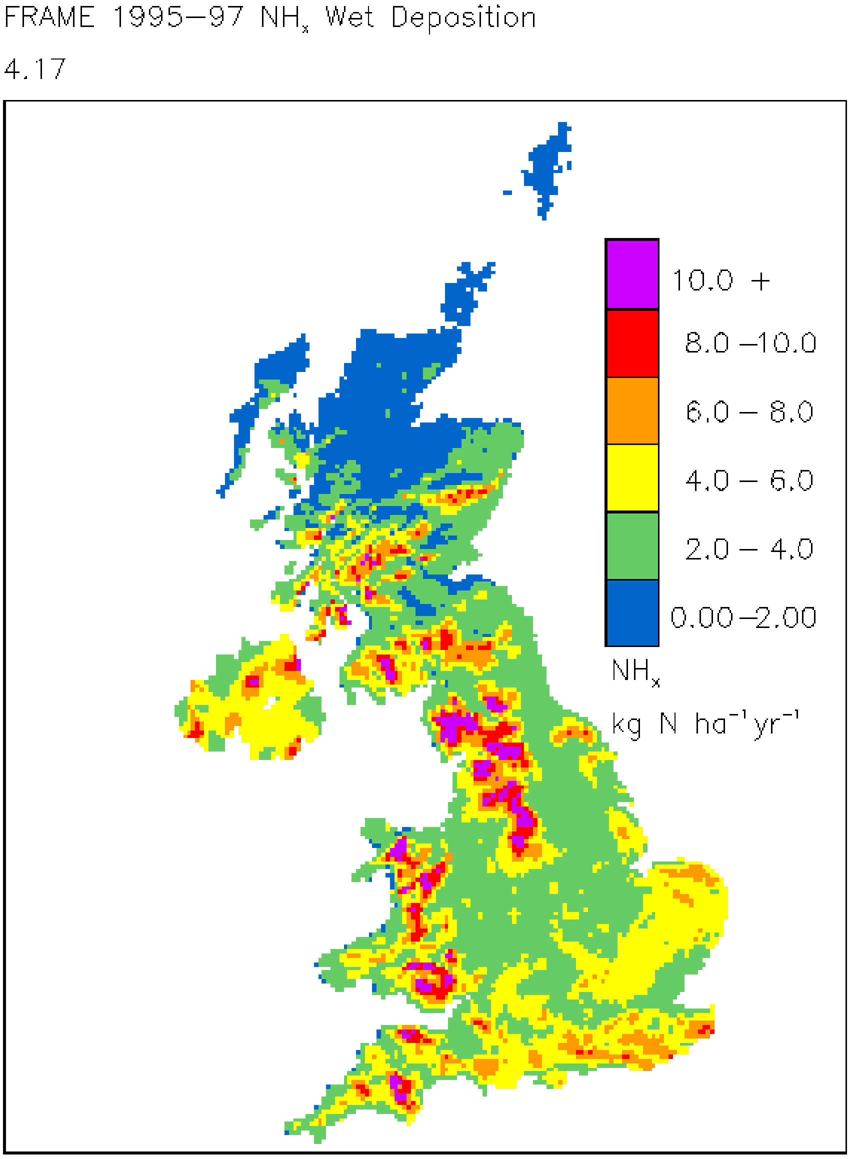


Figure 2(a) FRAME NH$_X$ wet deposition

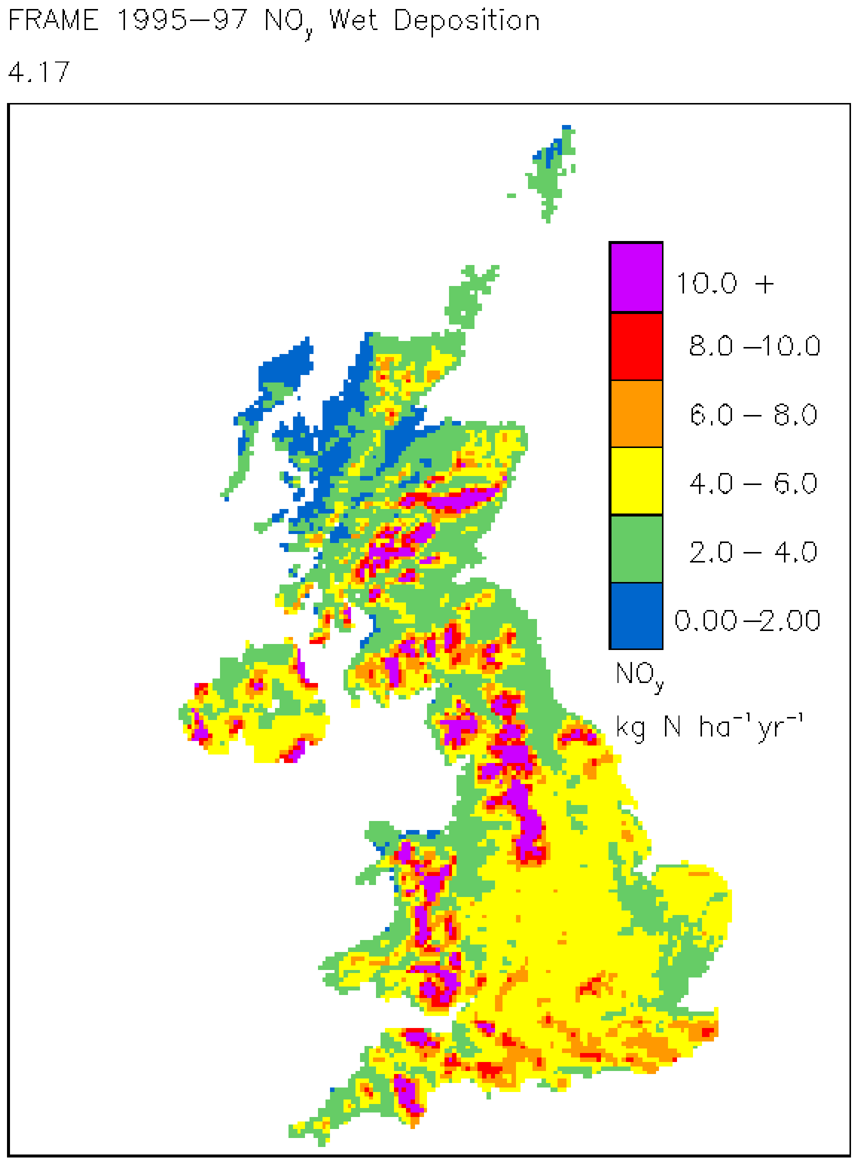


Figure 2(b) FRAME NO$_Y$ wet deposition

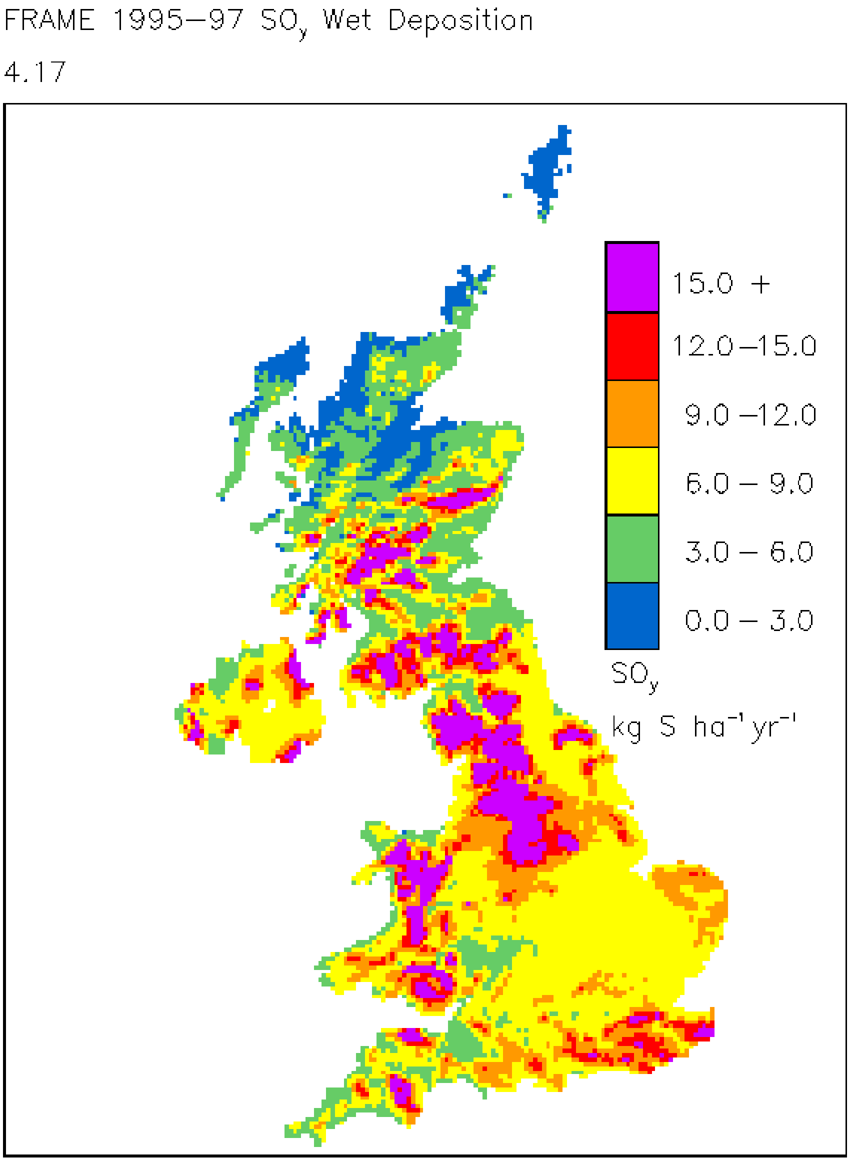


Figure 2(c) FRAME SO_X wet deposition

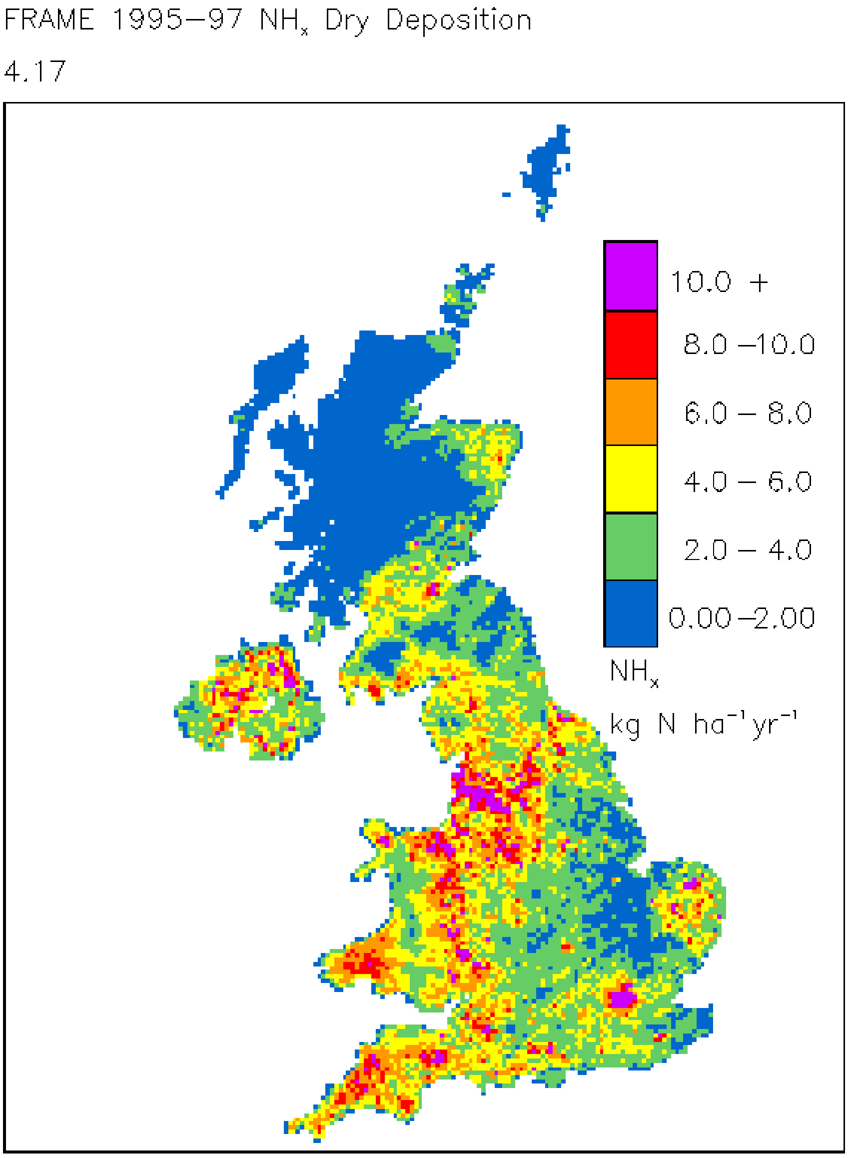


Figure 2(d) FRAME grid-average NH$_X$ dry deposition

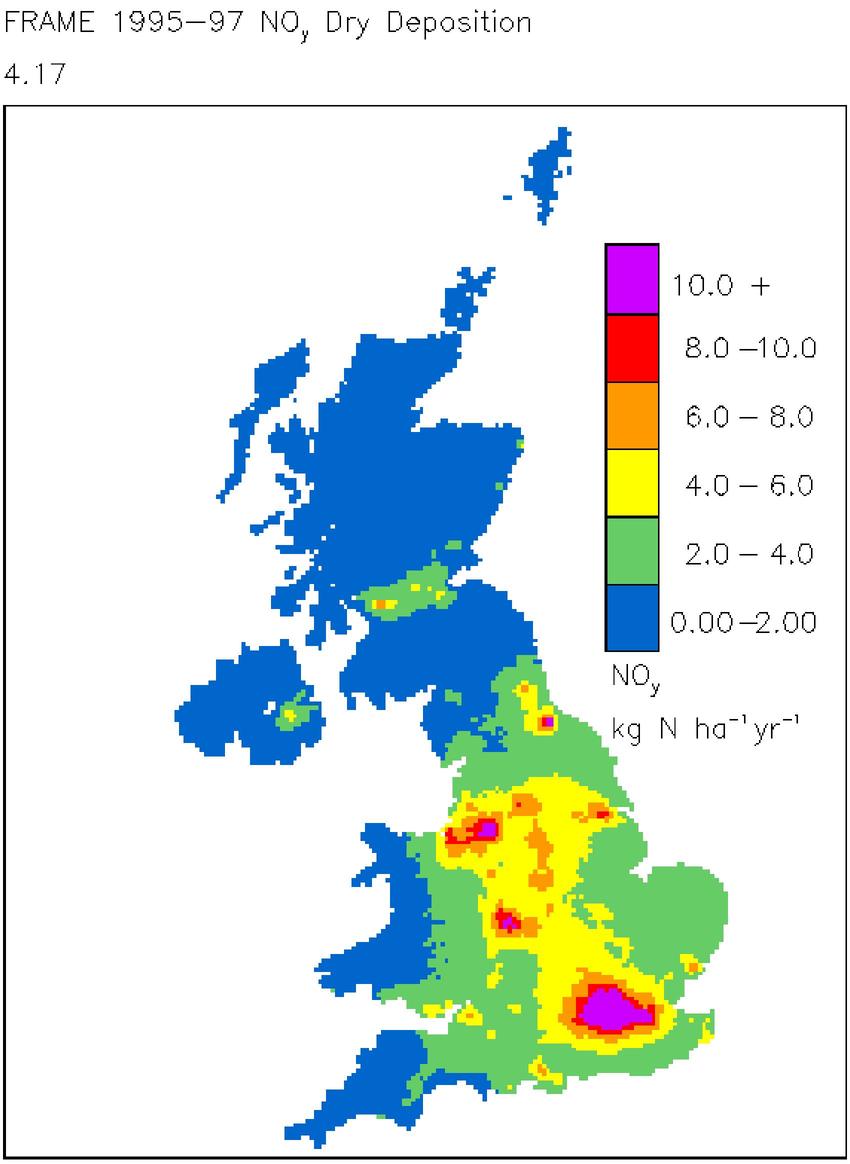


Figure 2(e) FRAME NO$_Y$ grid-average dry deposition

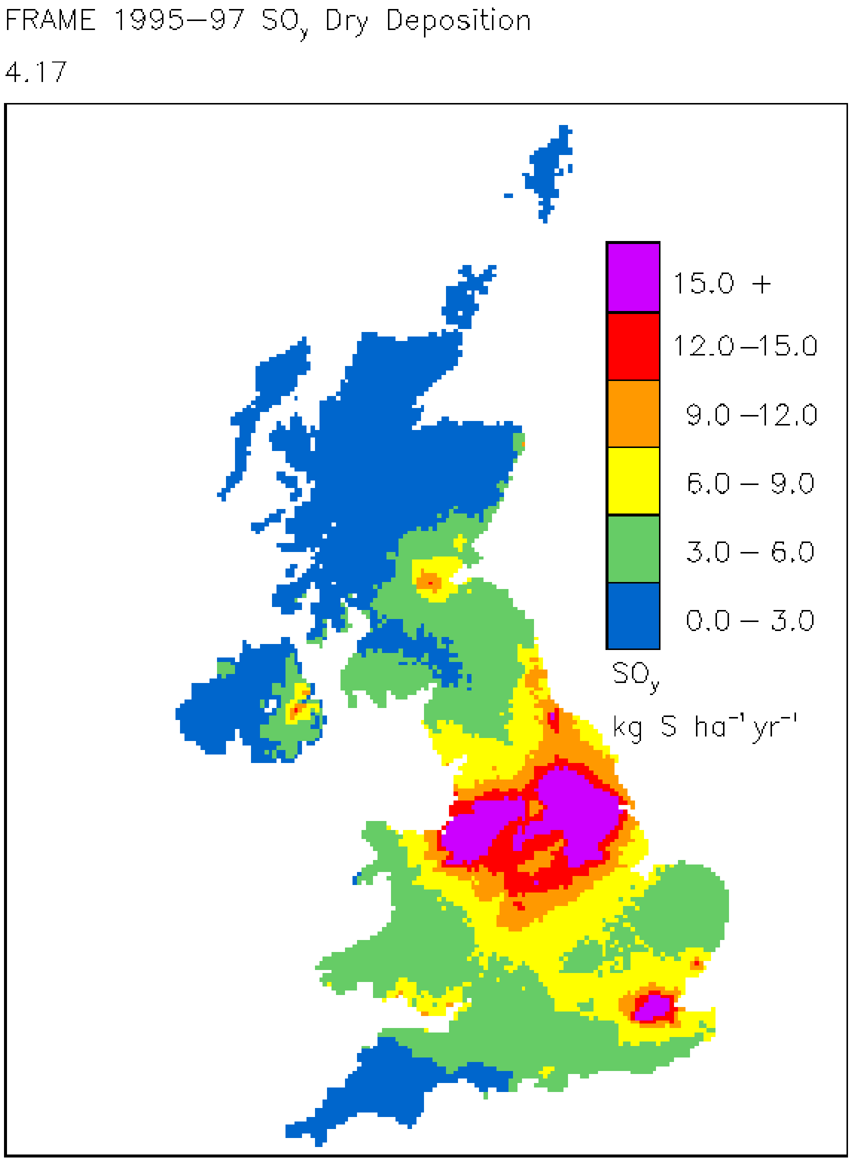


Figure 2(f) FRAME SO$_X$ grid-average dry deposition

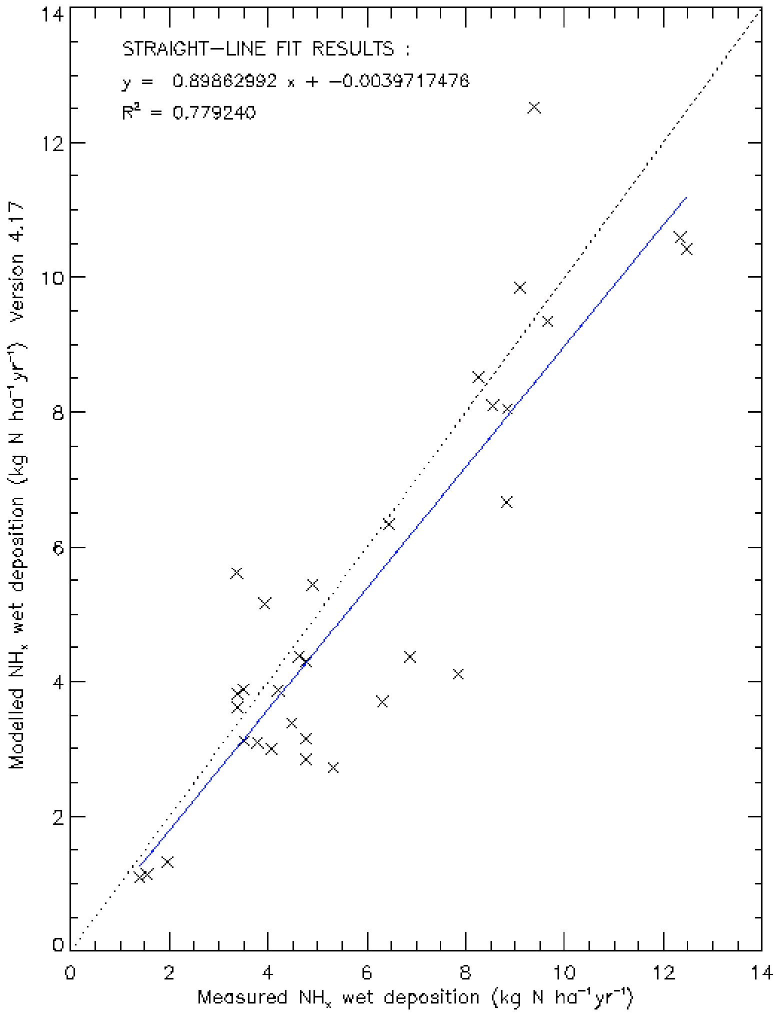


Figure 3(a) NH_X wet deposition correlation with measurements

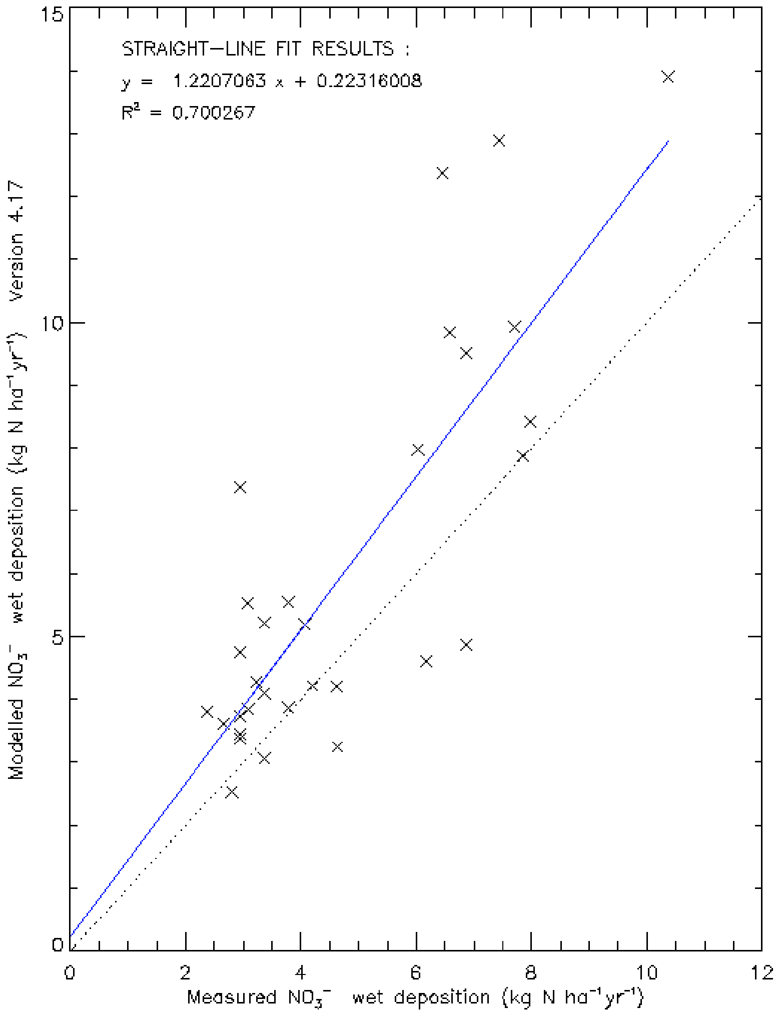


Figure 3(b) NO_Y wet deposition correlation with measurements

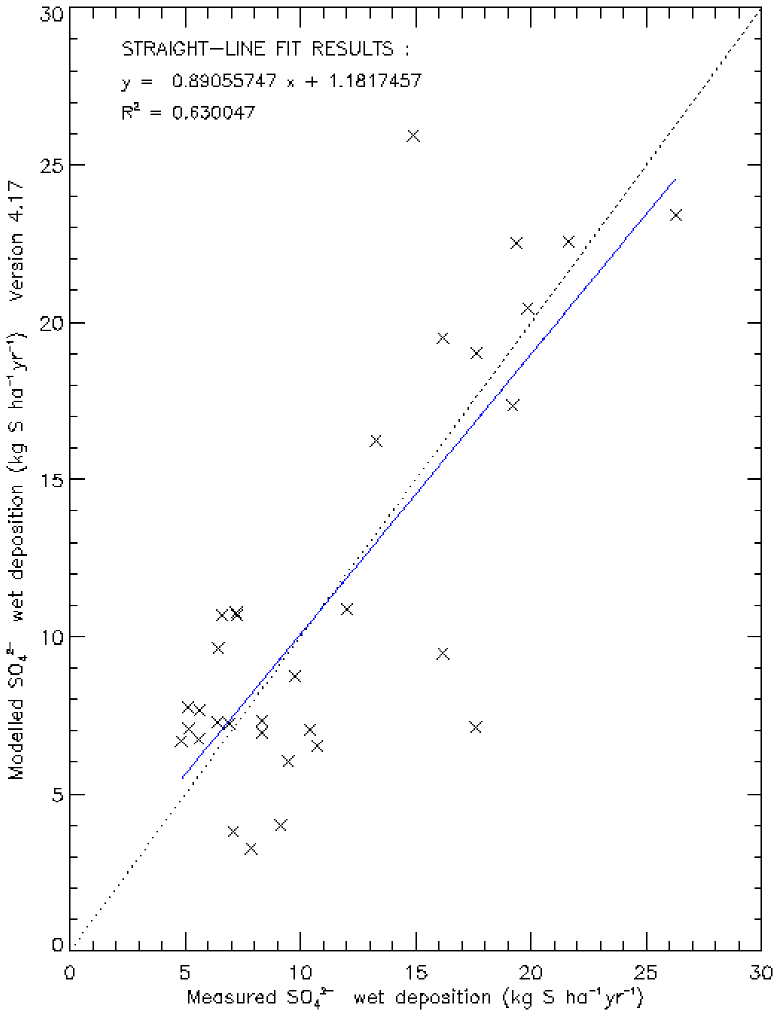


Figure 3(c) SO_x wet deposition correlation with measurements

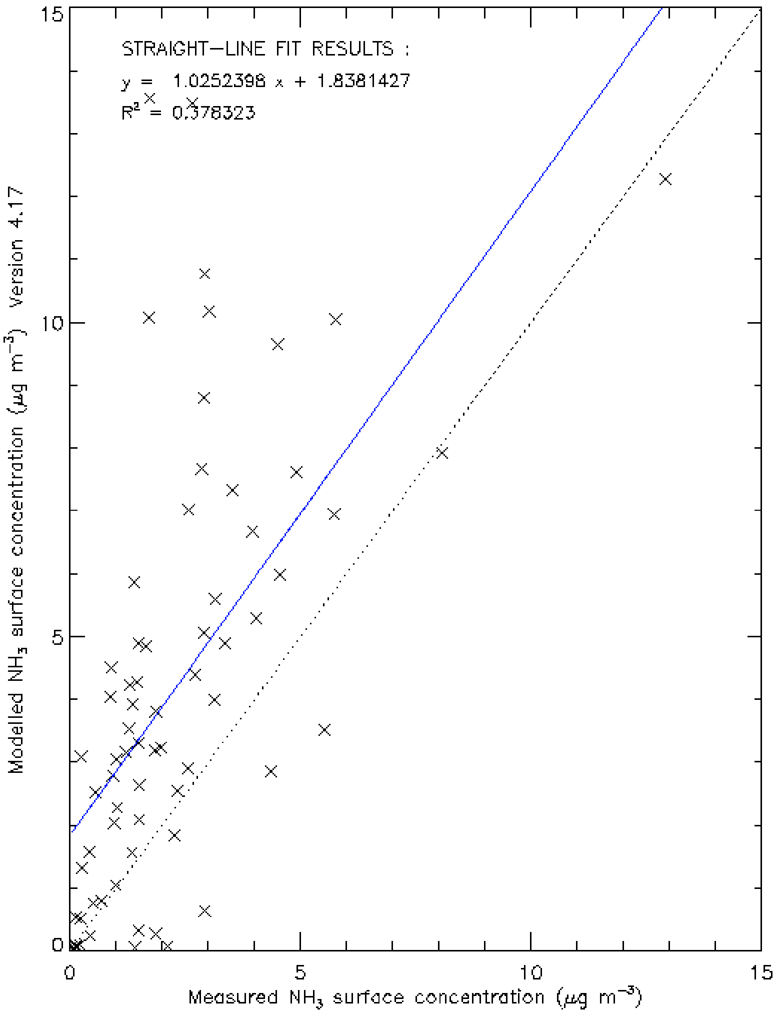


Figure 3(d) NH_3 concentration correlation with measurements

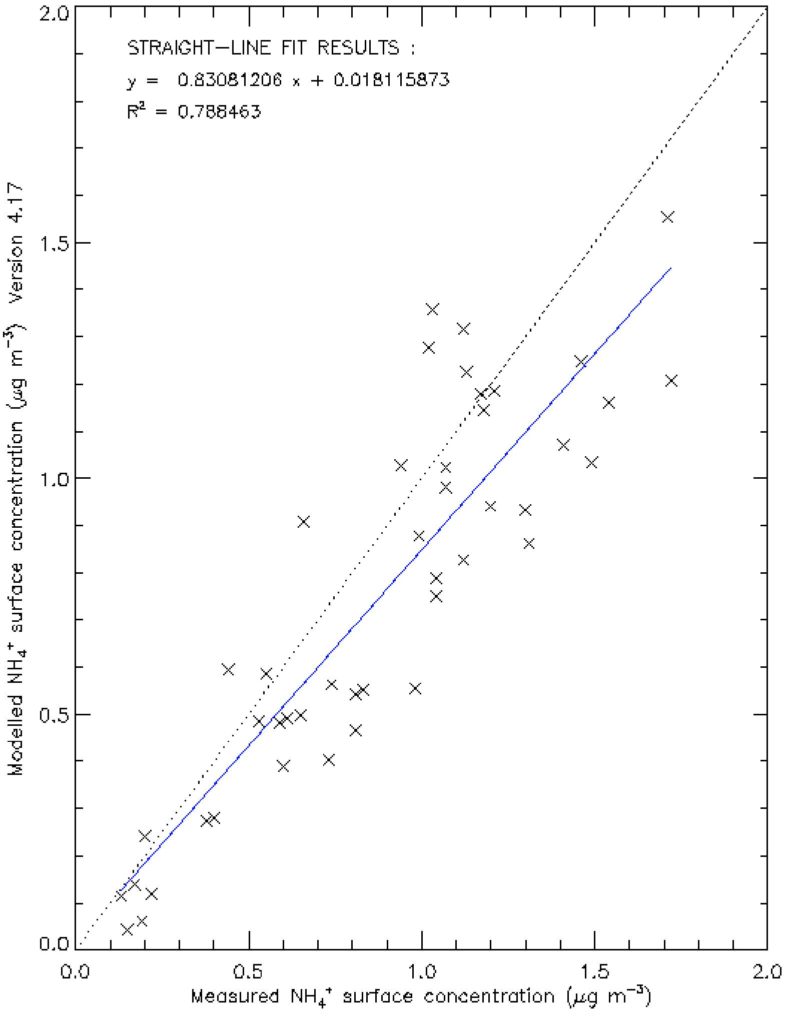


Figure 3(e) NH_4^+ aerosol concentration correlation with measurements

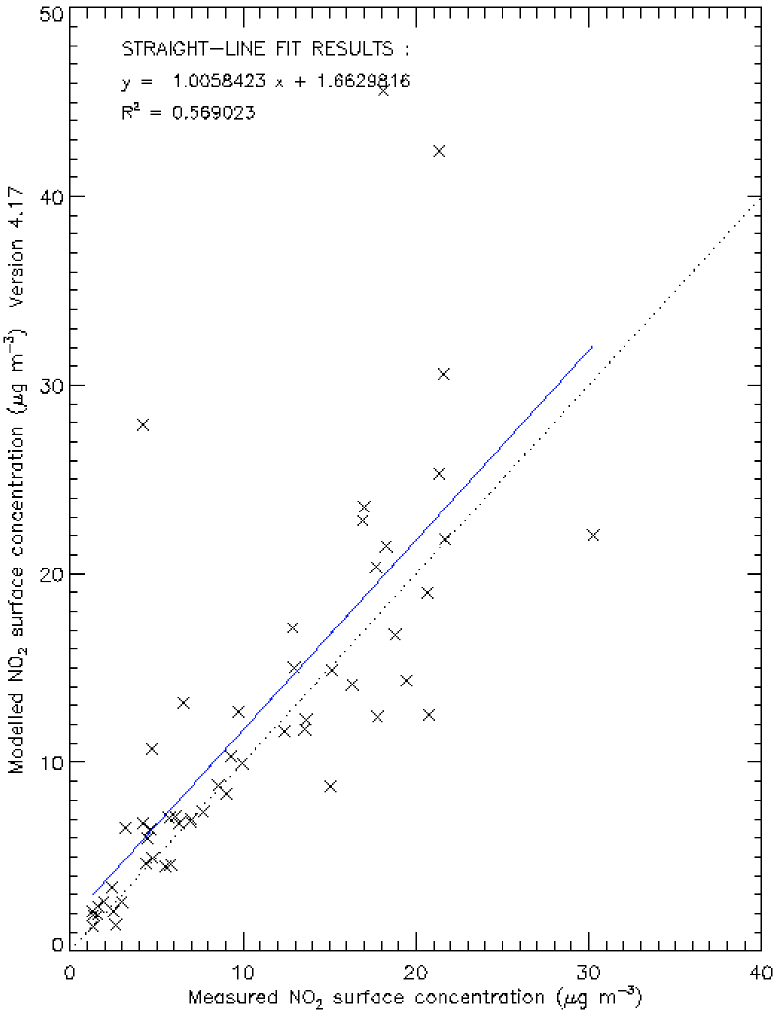


Figure 3(f) NO_2 concentration correlation with measurements

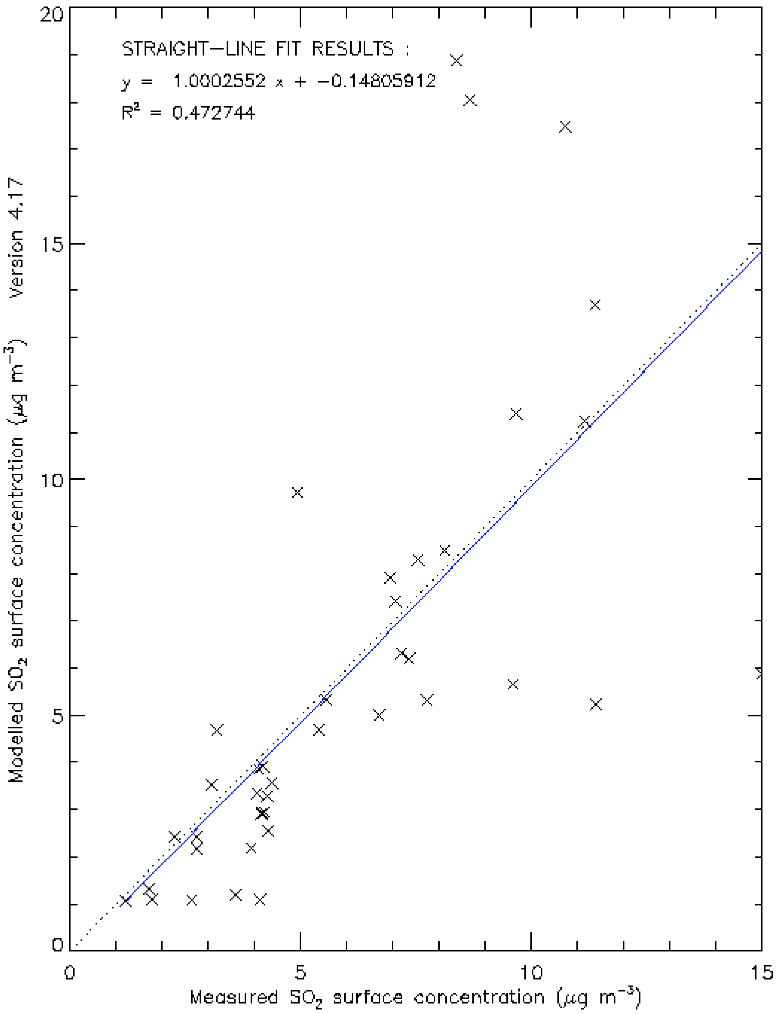


Figure 3(g) SO_2 concentration correlation with measurements

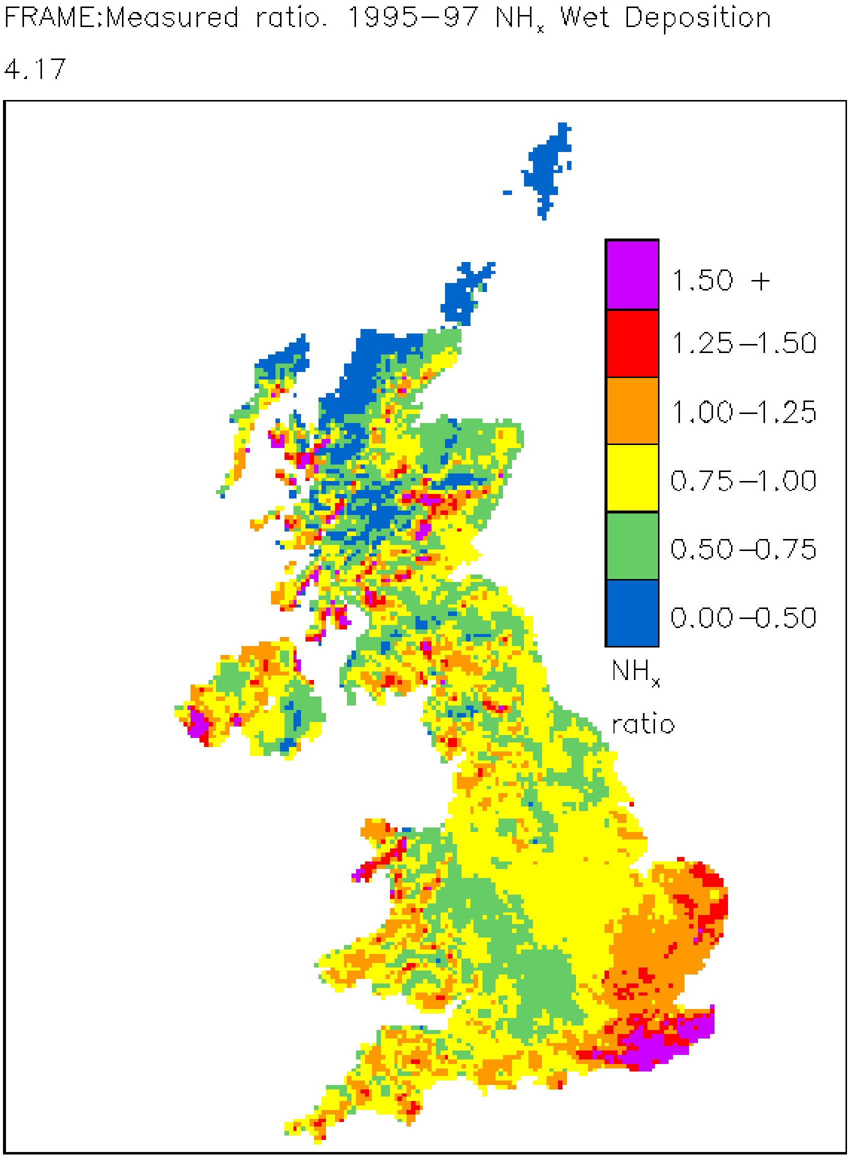


Figure 4(a) Map of ratio of modelled: measured-interpolated NH$_X$ wet deposition

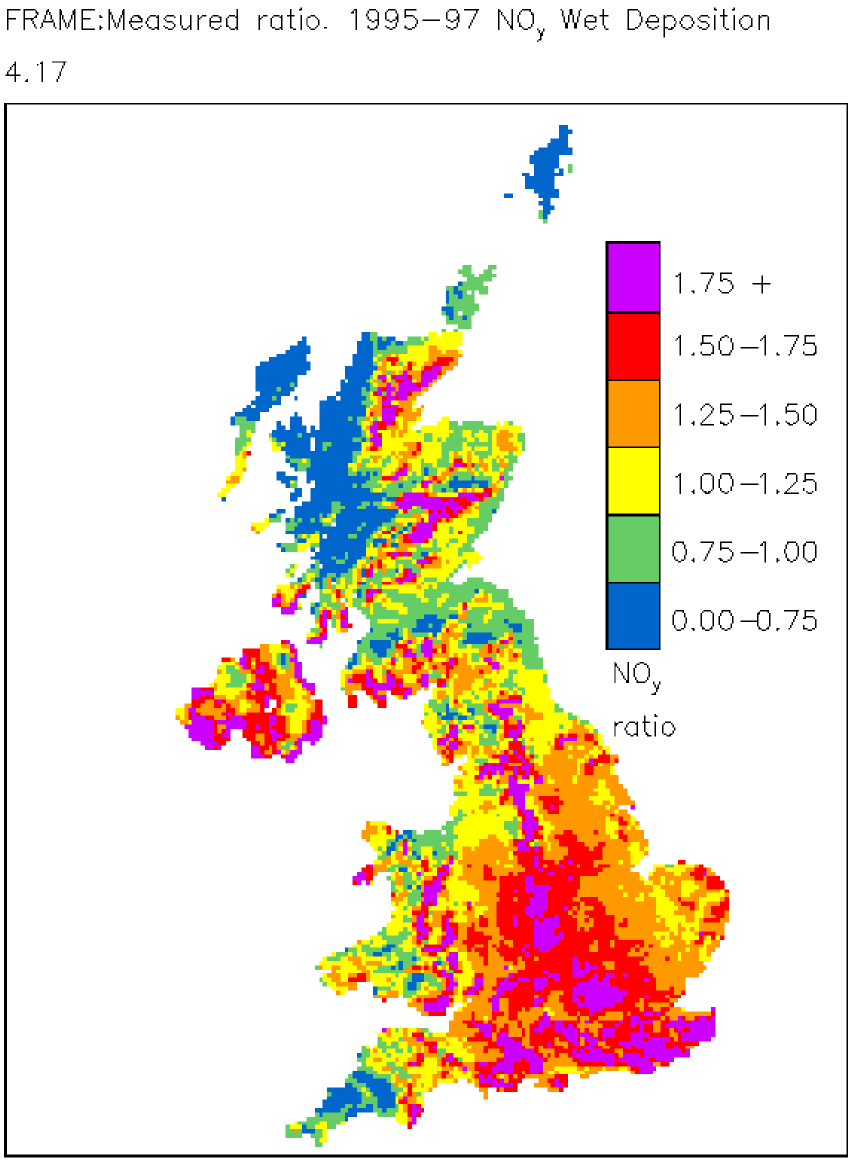


Figure 4(b) Map of ratio of modelled: measured-interpolated NO$_Y$ wet deposition

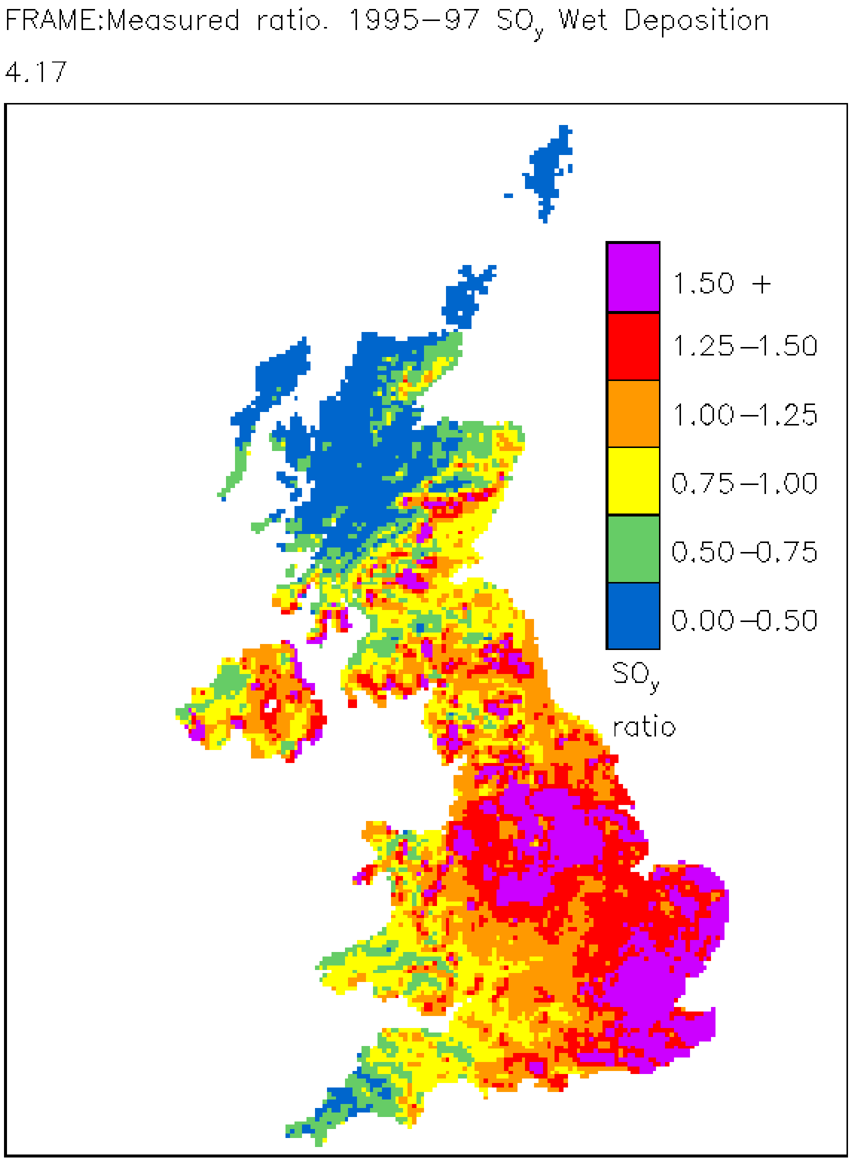


Figure 4(c) Map of ratio of modelled: measured-interpolated SO_X wet deposition

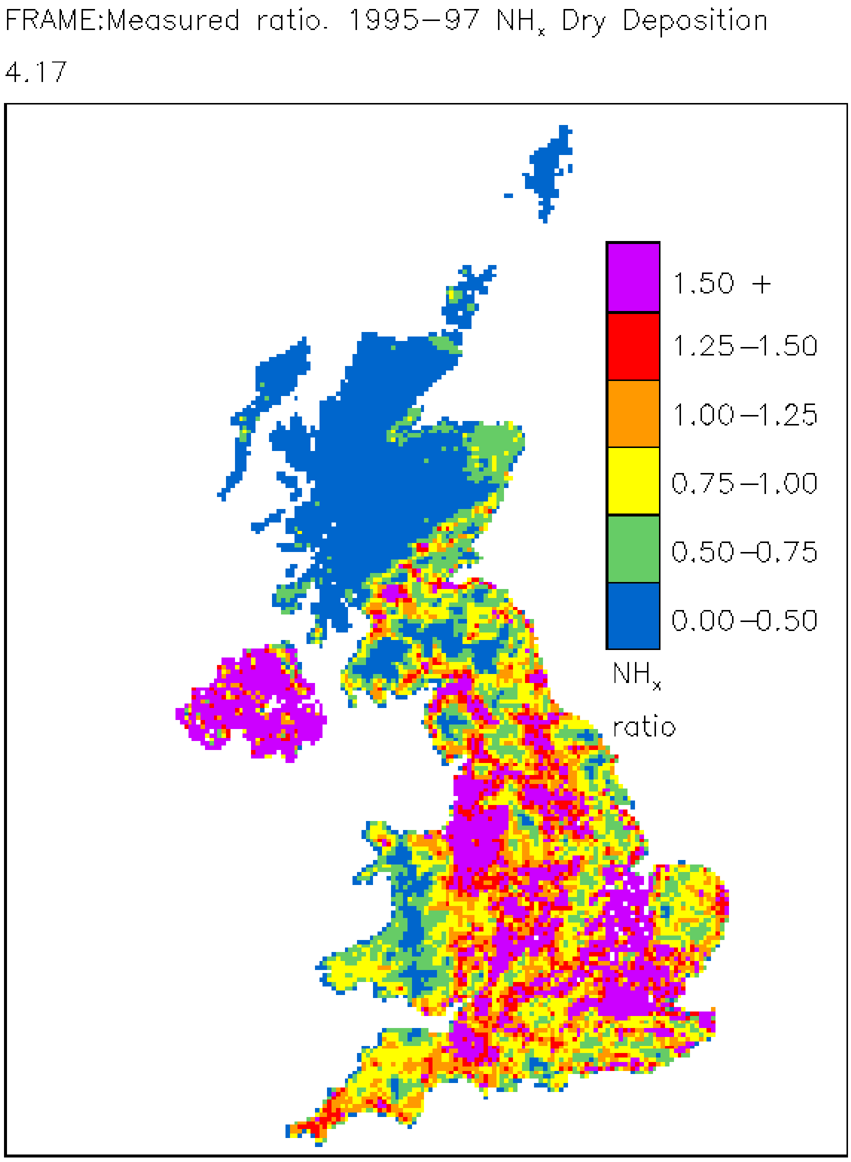


Figure 4(d) Map of ratio of modelled: measured-interpolated NH$_X$ dry deposition

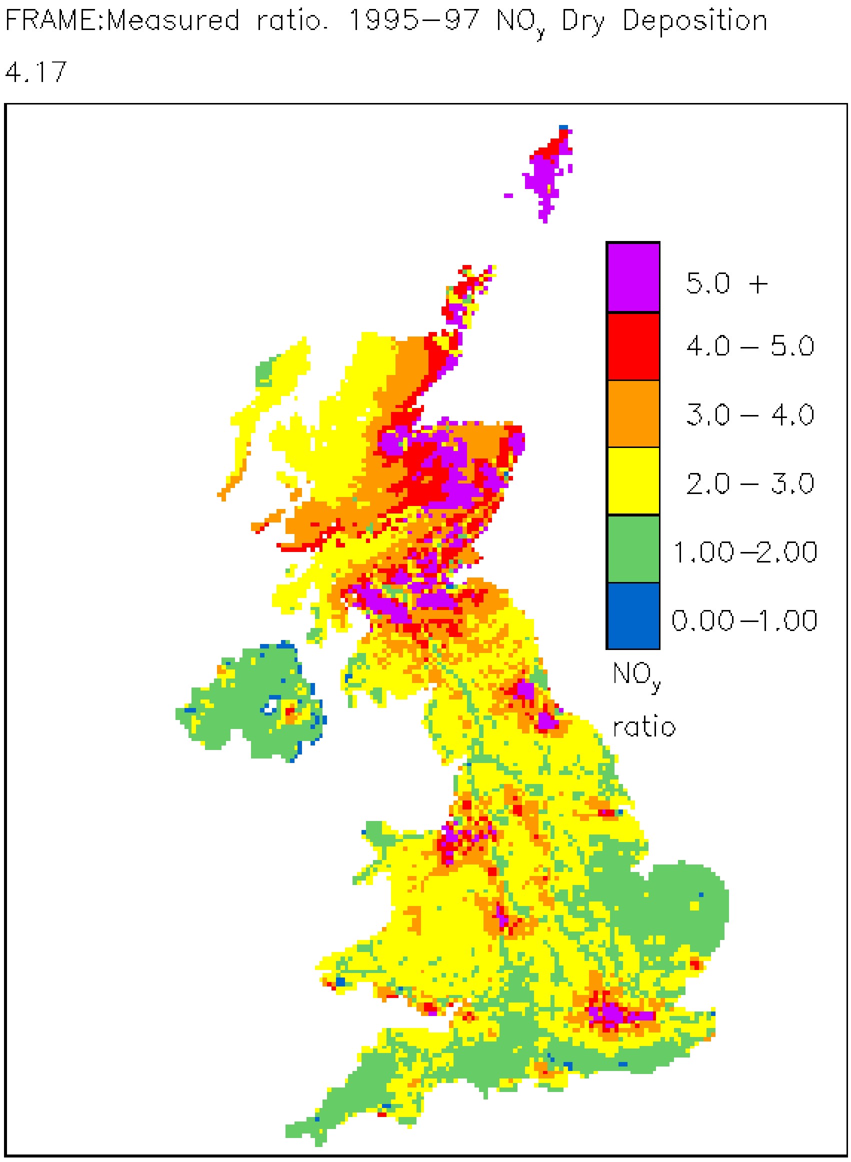


Figure 4(e) Map of ratio of modelled: measured-interpolated NO$_Y$ dry deposition

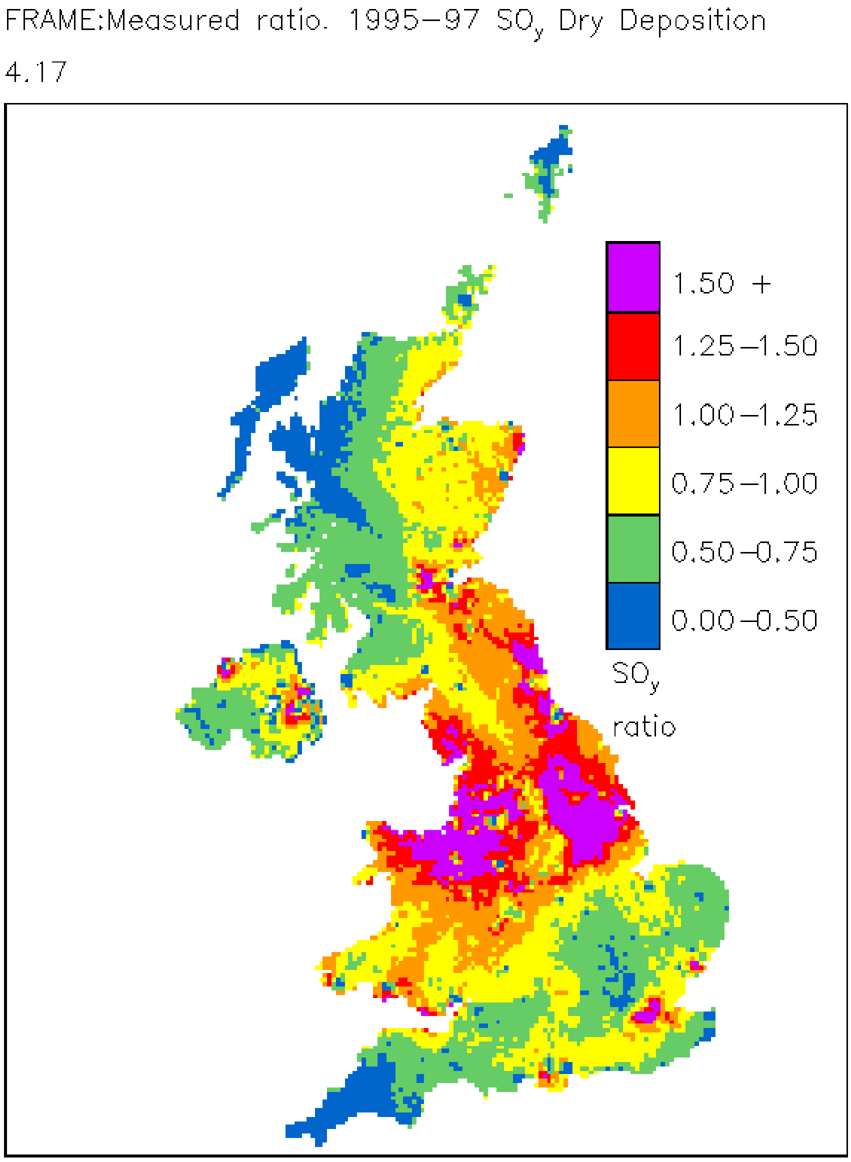
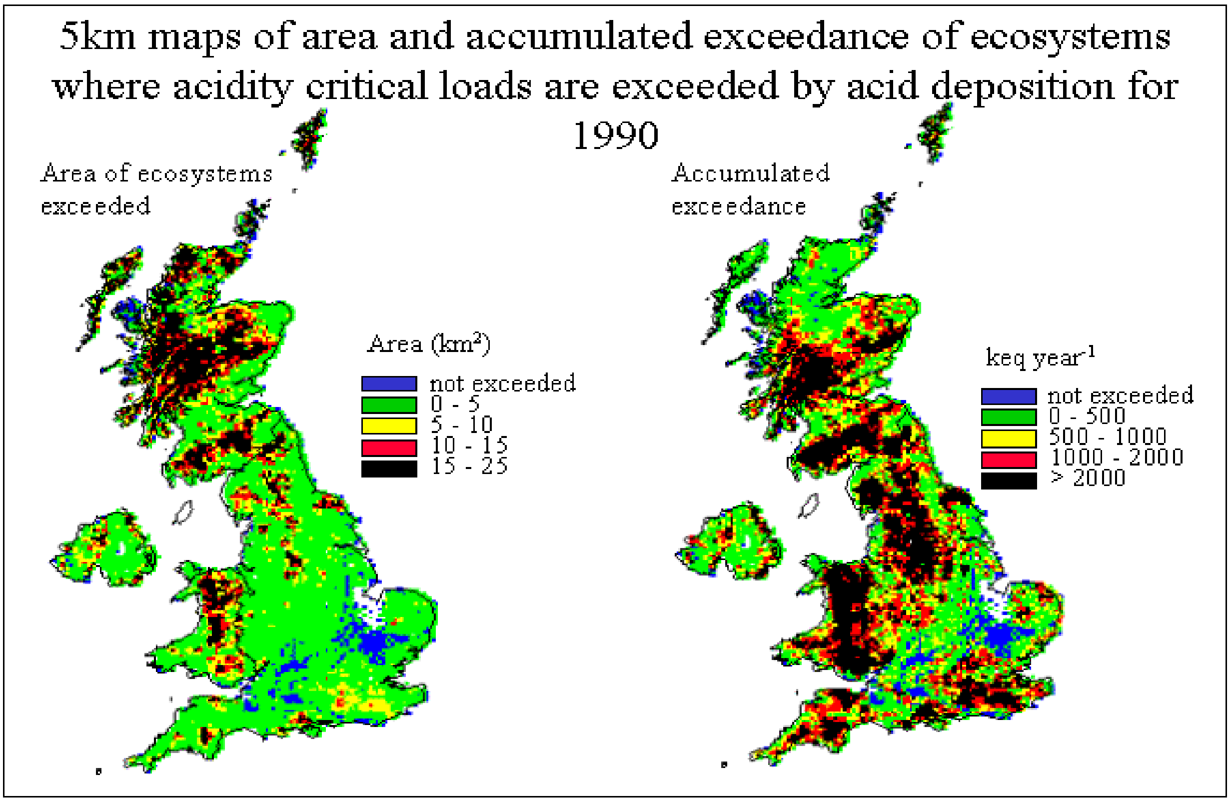


Figure 4(f) Map of ratio of modelled: measured-interpolated SO_Y dry deposition



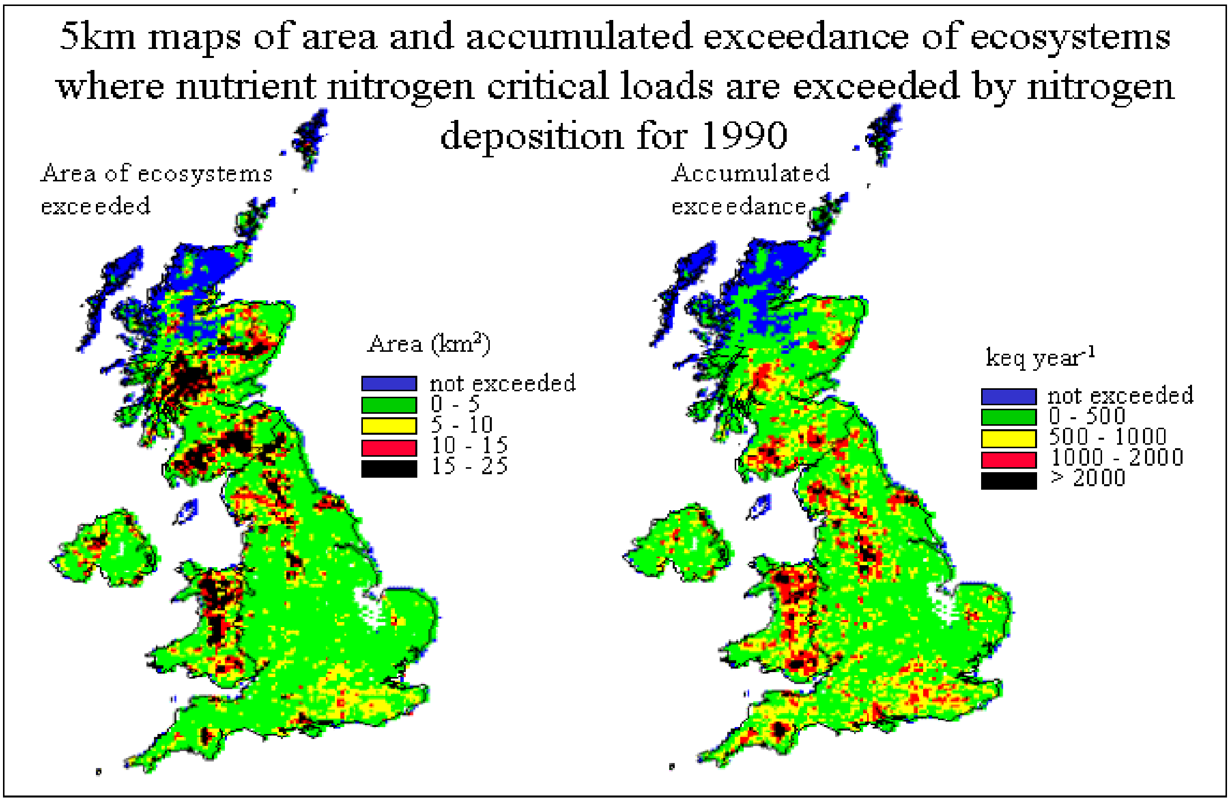
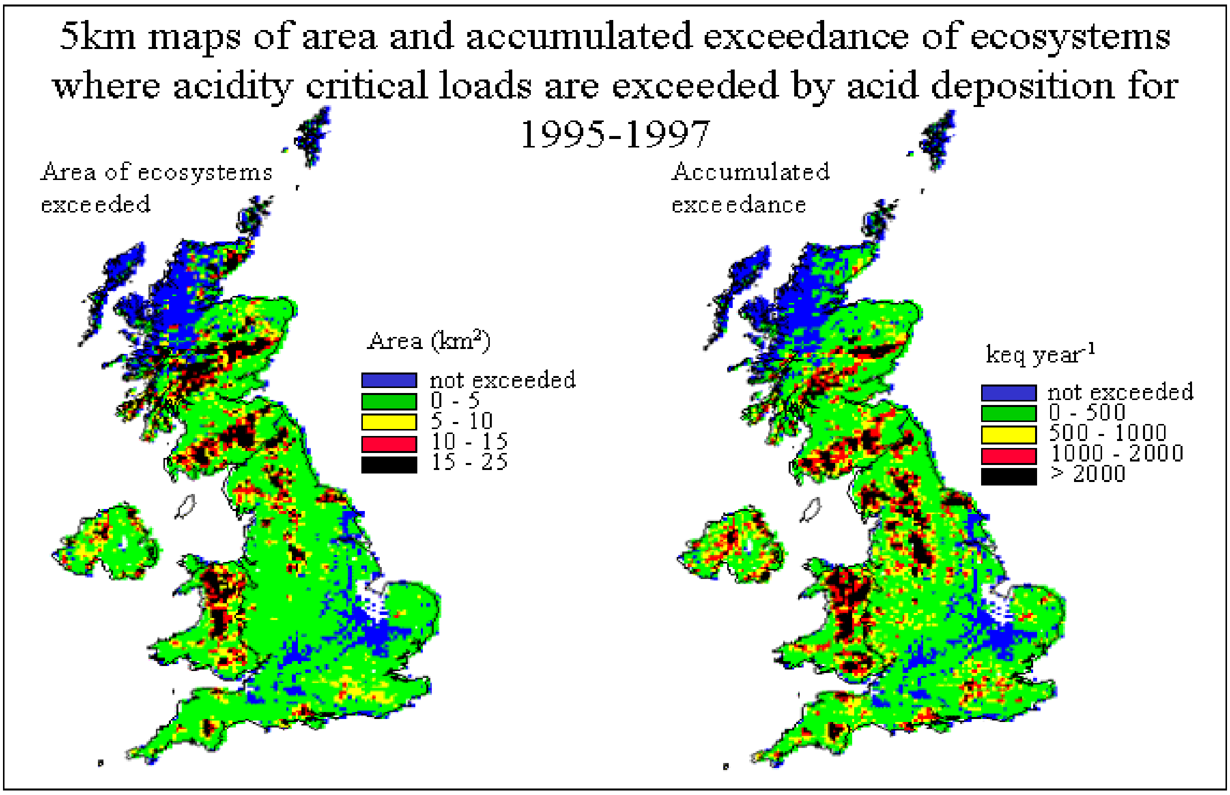


Figure 5(a). Exceedances of critical loads by FRAME calibrated deposition for 1990



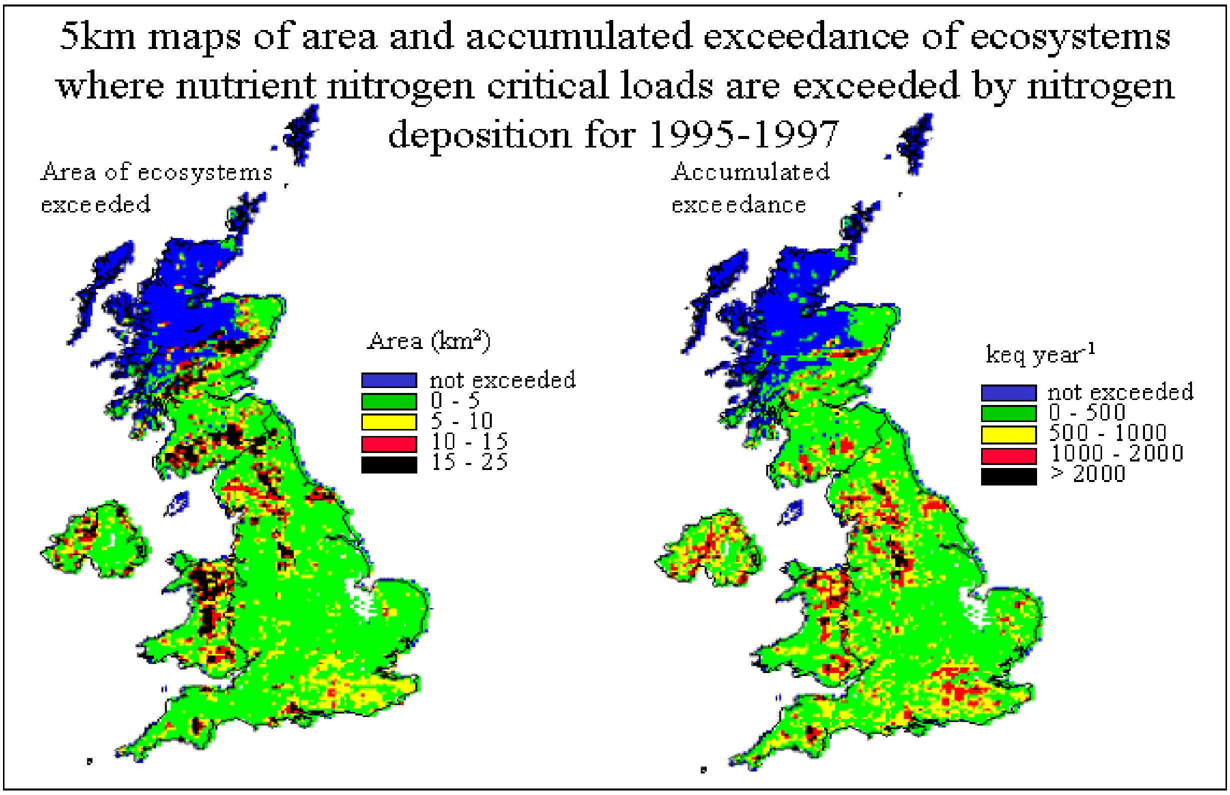
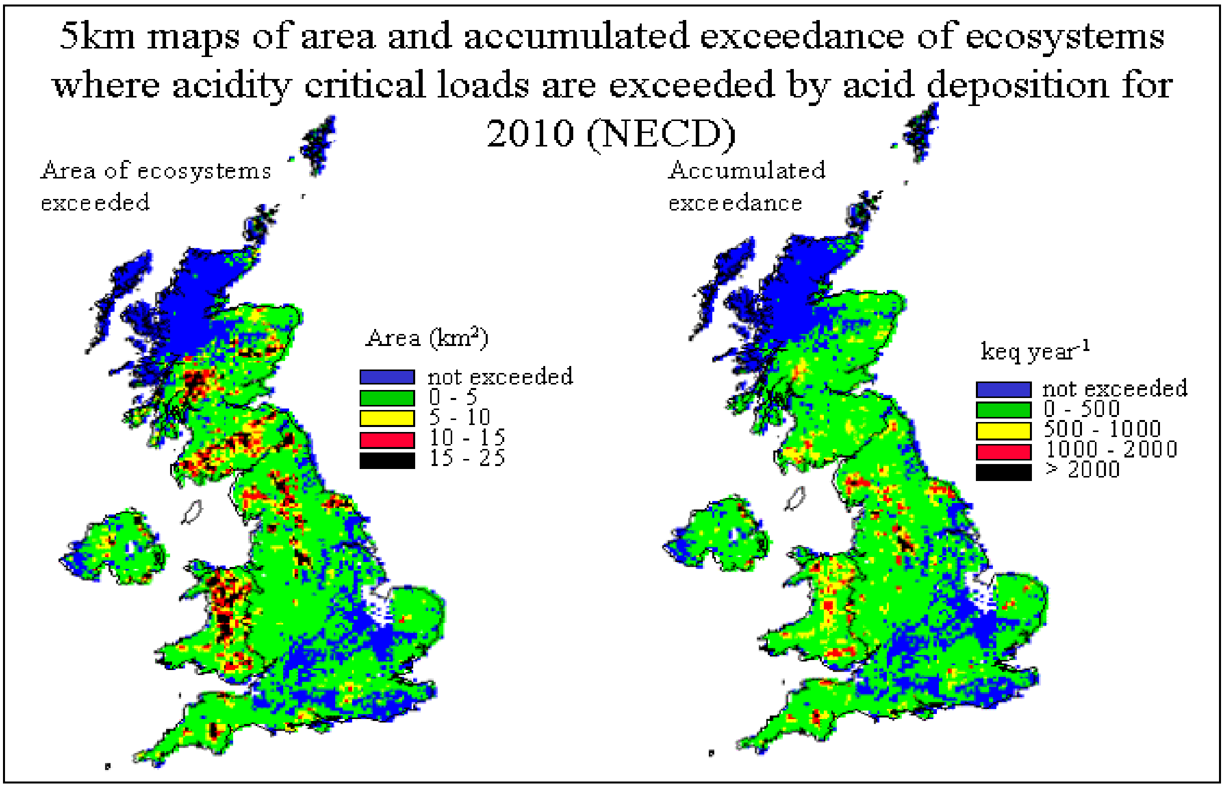


Figure 5(b). Exceedances of critical loads by DMP deposition for 1995-1997



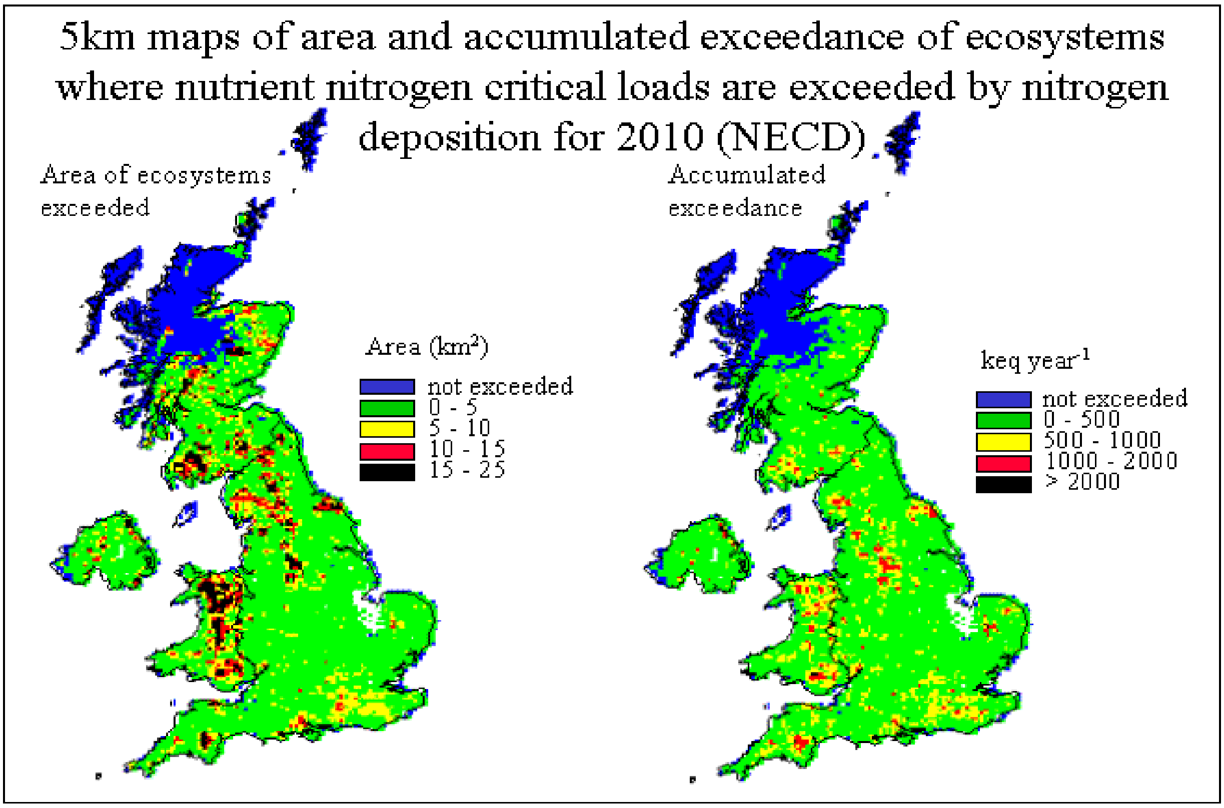


Figure 5(c). Exceedances of critical loads by FRAME calibrated deposition for 2010