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Atmospheric Supply of Nitrogen (Emissions and Deposition) to the OSPAR Convention Waters

Summary Report for UBA

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Summary

This report presents the results of the EMEP calculations of atmospheric transport and deposition of nitrogen to the OSPAR Convention Waters, as well as estimates of atmospheric emissions of nitrogen for the years 1990 – 2001. It is an update of the previous EMEP report (Bartnicki and Fagerli, 2003) with the results of the source-receptor computations for the year 2000 as a major new addition.

The EMEP Unified Eulerian model system has been used for all nitrogen computations presented here. This system has undergone a major overhaul the last two years, where the previous EMEP models (Lagrangian as well as Eulerian) have been merged and re-written in order to produce the EMEP Unified Eulerian model. It has been verified against measurement data at EMEP stations for nine different years (1980, 1985, 1990, 1995-2000) in EMEP Status Report 1/2003, Part II.. A comparison of computed and measured nitrogen depositions at the OSPAR stations for 1990, 1995, 1996, 1997, 1998, 1999, and 2000 is described in the present report.

For most of the OSPAR Contracting Parties a reduction of nitrogen oxides emissions (NO_2) has been reported for the period 1990 – 2001. Out of 18 countries relevant for OSPAR, annual emissions are declining in 14 countries. United Kingdom, Germany and France are the largest emitters of nitrogen oxides (NO_2) among the OSPAR Parties. In these three countries, emissions are significantly lower in 2001 than in 1990 by 39%, 42% and 26%, respectively. In the three largest contributors to nitrogen deposition in the OSPAR Convention Waters from outside OSPAR Parties, Poland, Italy and the part of the Russian Federation included in the EMEP area, emissions in 2000 are reported to be lower than in 1990 by 37%, 29% and 35%, respectively. Thus, with some guardedness of the uncertainty associated with the reporting of emissions, there seem to be evidence for NO_2 emissions going down in the OSPAR area.

International ship traffic on the OSPAR Convention Waters is the largest single source of NO_2 emissions in the area of interest. Unfortunately, the latest official data on these emissions are available for 1990. It is very important to update ship emissions for further calculations of the depositions with the EMEP model.

Annual emissions of ammonia (NH_3) are lower in 2001 than in 1990 for most of the OSPAR Contracting Parties and all selected additional three countries outside OSPAR. Out of 18 countries relevant for OSPAR, annual emissions in 15 countries are lower in 2001 than in 1990, sometimes significantly e.g. 45%, 39% and 36% emission reduction in the Russian Federation, Poland and in the Netherlands, respectively.

Annual 2000 emissions of nitrogen oxides (NO_2) as a sum of emissions from all OSPAR Parties and selected additional three countries outside OSPAR are 13% higher than the corresponding annual 2001 emissions of ammonia (NH_3), in terms of nitrogen emitted.

For oxidized nitrogen, annual deposition to the main OSPAR regions is lower in 2000 than in 1990 in all main OSPAR regions, by 21%, 9%, 20%, 17% and 20% for Arctic Waters, Greater North Sea, Bay of Biscay and Wider Atlantic, respectively. Thus, the declining pattern of nitrogen oxides (NO_2) emissions is followed by the deposition pattern for the same period.

For both oxidized and reduced nitrogen, a clear gradient of the deposition towards the open sea can be noticed with maxima of the deposition in the OSPAR Region 2, Greater North Sea.

Deposition of oxidized nitrogen (maximum 630 mg N m^{-2} in 2000) is higher than the deposition of reduced nitrogen (maximum 537 mg N m^{-2} in 2000). **This means that the nitrogen emitted from combustion sources contributes more to the deposition than the nitrogen emitted mainly from sources related to agriculture.**

Annual deposition of reduced nitrogen in 2000 is higher than in 1990 in the main OSPAR Region 2, Greater North Sea (22%) and in the main OSPAR Region 3, Celtic Sea (6%). In the three remaining main OSPAR Regions it is lower by 23%, 15% and 30%, for Arctic Waters, Bay of Biscay and Wider Atlantic, respectively.

There is a large inter-annual variability in nitrogen depositions, caused by the different meteorological conditions in the different years.

Annual deposition of oxidized nitrogen to all sub-regions (of the main OSPAR Region 2) except sub-regions 5 and 13 is lower in 2000 than in 1990 by 8% to 16%. Depositions of oxidized nitrogen to sub-regions 5 and 13 remain on the same level in 2000 as in 1990.

Except for sub-region 2 (4%decrease), annual deposition of reduced nitrogen in 2000 is higher than in 1990 in all sub-regions of the Greater North Sea. For some of sub-regions deposition increase is significant e.g. 50% and 35% for sub-regions 13 and 5, respectively.

Measured and computed wet annual depositions of oxidized, reduced and total nitrogen were compared for the years 1990, 1995, 1996, 1997, 1998, 1999 and 2000 for the stations located in the main OSPAR regions and in the sub-regions of the OSPAR Region 2 (Greater North Sea). **For the majority of the stations, the modelled and measured**

concentrations agree well. In general, modelled wet depositions for stations situated in the sub-regions of the Greater North Sea follow observations better than modelled wet depositions for stations located in the main regions. This may be caused by uncertainties in the model boundary conditions as well as local influence on the observations. **It is reassuring that although the model underestimates wet depositions somewhat, it reproduces very well the observed inter-annual variability.**

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1. Introduction

This report has been prepared for the German Federal Environmental Agency (UBA) and for the OSPAR Commission (for the Protection of the Marine Environment of the North-East Atlantic). It is based on model estimates and monitoring results presented to the twenty-seventh session of the Steering Body of EMEP (Co-operative Programme for Monitoring and Evaluation of Long-Range Transmission of Air Pollutants in Europe). The present report is a follow up of the first report published in 2003 (Bartnicki and Fagerli, 2003). Compared to the content of the last year report, the present version includes emissions and computed nitrogen depositions to the main regions and sub-regions of the Greater North Sea, for one additional year 2001. In addition, the results of the source-receptor computations are also included in this report, which give the contribution of nitrogen emissions in 15 OSPAR Contracting Parties and three additional selected countries to nitrogen deposition in or regions and sub-regions.

The role of atmospheric deposition of nitrogen in the total fluxes to the North Sea and North Atlantic, as well as discussion of related chemical and biological processes has been a subject of many scientific publications (e.g. Rendell *et al.,* 1993; Peierls and Paerl, 1977; Shultz *et al.,* 1999 and Klein, L. M., 2002). However, most of the studies related to atmospheric supply of nitrogen to the OSPAR Convention Waters were based on measurements and limited to rather small regions, not covering the entire area of interest. In the present report we have used model calculations for the estimation of annual nitrogen depositions to all five main regions of the OSPAR Convention Waters. In addition, we have also calculated annual depositions to 13 sub-regions of the OSPAR Main Region 2 – Greater North Sea. The model calculations cover seven years, 1990, 1995, 1996, 1997, 1998, 1999 and 2000. All specific results (maps, tables, ascii files with data) of this study are available on internet on the EMEP web site www.emep.int/index_facts.html.

The EMEP Unified Eulerian model system has been used for all nitrogen computations presented here. This system has undergone a major overhaul the last two years, where the previous EMEP models (Lagrangian as well as Eulerian) have been merged and re-written in order to produce the Unified EMEP Eulerian model. The model has been documented in detail in the EMEP Status Report 1/2003, Part I (Simpson *et. al.,* 2003),. It has been verified against measurement data at EMEP stations for nine different years (1980, 1985, 1990, 1995, 1996, 1997, 1998, 1999 and 2000) in the EMEP Status Report 1/2003, Part II (Fagerli *et al*., 2003). A short description of the main EMEP model features is given in the next chapter.

Similar computations as for the OSPAR Convention Waters have already been performed for the Baltic Sea in the frame of co-operation between the Helsinki Commission (HELCOM) and EMEP (Bartnicki *et al.,* 2002). As agreed with UBA, the results of the

computations for the OSPAR Convention Waters are presented here in the similar way as it has been done for HELCOM.

The following topics are included and discussed in the present report:

- Annual emissions of nitrogen oxides (NO_2) and ammonia (NH_3) from the OSPAR Contracting Parties and three largest outside contributors to nitrogen deposition in the OSPAR Convention Waters in the period 1990 – 2000.
- Modelled annual depositions to the five main OSPAR regions and 13 sub-regions of the Greater North Sea for the years 1990, 1995, 1996, 1997, 1998, 1999 and 2000.
- Comparison of model results and measurements currently available from the OSPAR stations, for wet depositions of nitrogen in the years 1990, 1995, 1996, 1997, 1998, 1999 and 2000.

2. Short description of the Unified EMEP model

The Unified EMEP model is an Eulerian model that has been developed at EMEP/MSC-W (Meteorological Synthesizing Centre – West of EMEP) for simulating atmospheric transport and deposition of acidifying and euthrophying compounds as well as photo-oxidants in Europe. The latest model version has been documented in EMEP Status Report I, Part I (Simpson *et. al.,* 2003), and here we only give a short description of the basic features of the model. Model details and its applications can be also found on the EMEP web site www.emep.int.

The model domain covers Europe and the Atlantic Ocean (Figure 1). The model grid (of the size 170×133) has a horizontal resolution of 50 km at 60^{o}N, which is consistent with the resolution of emission data reported to CLRTAP (Vestreng, 2003). In the vertical, the model has 20 sigma layers reaching up to 100 hPa. Approximately 10 of these layers are placed below 2 km to obtain high resolution of the boundary layer which is of special importance to the long range transport of air pollution.

The Unified Model uses 3-hourly resolution meteorological data from the PARLAM-PS model, a dedicated version of the HIRLAM (High Resolution Limited Area Model) Numerical Weather Prediction model.

The emissions consist of gridded annual national emissions of sulphur dioxide, nitrogen oxides, ammonia, non-methane volatile organic compounds and carbon monoxide. They are available in each of the 50×50 km^2 model grid. These emissions are distributed temporally according to monthly and daily factors derived from data provided by the University of Stuttgart (IER).

Concentrations of 71 species are computed in the latest version of the Unified EMEP model (56 are advected, 15 are short-lived and not advected). The sulphur and nitrogen chemistry is coupled to the photo-chemistry, which allows a more sophisticated description of e.g. the oxidation of sulphur dioxide to sulphate.

Dry deposition is calculated using the resistance analogy and is a function of the pollutant type, meteorological conditions and surface properties. Parametrization of wet deposition processes includes both in-cloud and sub-cloud scavenging of gases and particles using scavenging coefficients.

3. Definitions of the OSPAR regions and sub-regions in the EMEP grid system

Annual deposition of nitrogen was calculated for the five main regions of the OSPAR Convention Waters and for 13 sub-regions of the selected main region.

OSPAR Convention waters are divided into five main regions:

Region 1: Arctic Waters
Region 2: Greater North Sea
Region 3: Celtic Sea
Region 4: Bay of Biscay
Region 5: Wider Atlantic

Definitions of the OSPAR border lines for five main regions are available in the OSPAR WEB site as descriptive geographical co-ordinates. Definitions of 13 sub-regions of the OSPAR Region 2 – Greater North Sea, were provided to EMEP by UBA, also in the form of geographical co-ordinates in accordance with the sub-regions used by OSPAR within the OSPAR Quality Status Reports.

The sub-regions of the Greater North Sea are related to the ICES Boxes in the following way:

Sub-region 1: ICES Box 1
Sub-region 2: ICES Box 2a
Sub-region 3: ICES Box 2b
Sub-region 4: ICES Box 3a
Sub-region 5: ICES Box 3b
Sub-region 6: ICES Box 4
Sub-region 7: ICES Box 5a
Sub-region 8: ICES Box 5b
Sub-region 9: ICES Box 6
Sub-region 10: ICES Box 7a
Sub-region 11: ICES Box 7b
Sub-region 12: ICES Box 8
Sub-region 13: ICES Box 9

In order to calculate nitrogen deposition to OSPAR regions, the borders of the OSPAR main regions and sub-regions had to be converted into the EMEP grid system, which is shown in Figure 1. In the first stage, descriptive geographical coordinates were transformed into the sets of discrete geographical coordinates describing each main region or sub-region. A major problem in this stage was the lack of a data set with geographical coordinates defining the western border of Region 3 – Wider Atlantic (see Figure 2). These coordinates were finally read manually from the map.

In the second more difficult and time consuming stage, borders of the regions in the EMEP grid system were used to calculate, what percentage of the considered OSPAR region or sub-region was included in each EMEP grid square. In this way, a separate percentage file was created for each region and sub-region.

A map with the borders of the main OSPAR regions in the EMEP grid system is shown in Figure 2, and a map with the borders of the sub-regions of the OSPAR Region 2, The Greater North Sea, is shown in Figure 3.

The border of the main OSPAR Region 5, Wider Atlantic, goes slightly outside the EMEP model grid system resulting in the underestimation of computed nitrogen deposition in this region. The underestimation is small, estimated to be less than 1%, because of the small number of missing grids and small values of the depositions in the missing grids, which are located far away from the significant sources of nitrogen emission.

An additional reason for the underestimation of nitrogen deposition in the Wider Atlantic Region is the formulation of boundary conditions in the EMEP model. According to this formulation, the values of the deposition in the lowest row in the EMEP grid system are always equal to zero. Also in this case underestimation is practically insignificant, below 1% of the total deposition value in this region.

These two problems can be solved by the extension of the EMEP model domain to the south. Such a solution is not possible at present, but the extension of the model domain is included in the future plans of EMEP.

4. Atmospheric emissions of nitrogen

In the deposition calculations, official figures on national emissions reported to EMEP were used (Vestreng, 2003). Emission inventories from the EMEP Parties, which include OSPAR Contracting Parties and the three selected countries (Italy, Russian Federation and Poland) with the largest deposition of nitrogen to the OSPAR Convention Waters, are presented in this report for the period 1990 – 2000. There are two kinds of nitrogen emissions used as input files to the EMEP model calculations: nitrogen oxides emissions as NO_2 and ammonia emissions as NH_3.

The three largest contributors to nitrogen deposition in the North Atlantic and North Sea from outside OSPAR Parties are Poland, Italy and Russian Federation. We assume that those countries are also the largest contributors to nitrogen deposition in the OSPAR Convention Waters from outside the OSPAR Parties.

An additional and very important, for the OSPAR Convention Waters, source of atmospheric NO_2 emissions is the international ship traffic. Official data for 1990 give 1266 ktonnes NO_2 annual emission from the international ship traffic on the North-East Atlantic and 648 ktonnes NO_2 from the international ship traffic on the North Sea. This is the largest source of NO_2 emissions in the OSPAR area of interest. Unfortunately, information about this source rather old and exists only for 1990. It is important to update the NO_2 emission data for more recent years as soon as possible.

A map with time series of nitrogen oxides (NO_2) emissions from the OSPAR Contracting Parties, and additionally Italy, Russian Federation and Poland, is shown in Figure 4. A similar map for ammonia (NH_3) emissions is shown in Figure 5. The same data are available as numbers in Tables 1 and 2, for nitrogen oxides (NO_2) and ammonia (NH_3) emissions, respectively.

For most of the OSPAR Contracting parties a reduction of nitrogen oxides emissions (NO_2) has been reported for the period 1990 – 2001 (Figure 4 and Table 1). Out of 18 countries relevant for OSPAR, only in four countries (Portugal, Spain, Iceland and Ireland) annual emissions of nitrogen oxides (NO_2) are reported to be higher in 2001 than in 1990 by 35%, 8%, 8% and 6% respectively. Moreover, in two of these countries (Iceland and Ireland) emissions in both years are low (9 and 38 kt N a^{-1}, respectively) compared to the rest of the countries. United Kingdom, Germany and France are the largest emitters of nitrogen oxides (NO_2) among the OSPAR Parties. In these three countries, emissions are significantly lower in 2001 than in 1990 by 39%, 42% and 26%, respectively. The three largest contributors to nitrogen deposition in the OSPAR Convention Waters from outside OSPAR Parties are Poland, Italy and the part of the Russian Federation included in the EMEP area. In all these countries, Poland, Russian Federation and Italy, emissions in 2001 are reported to be lower than in 1990 by 37%, 35% and 29%, respectively.

Annual emissions of nitrogen oxides, as a sum of emissions from all OSPAR Parties and selected additional three countries outside OSPAR are 29% lower in 2001 than the annual emissions in the year 1990. Thus, with some guardedness of the uncertainty associated with the reporting of emissions, there seem to be evidence for NO_2 emissions going down in the OSPAR area.

Ammonia (NH_3) emissions are shown in Figure 5 and Table 2. Also annual emissions of ammonia (NH_3) are lower in 2001 than in 1990 for most of the OSPAR Parties and all selected additional three countries outside OSPAR. Out of 18 countries relevant for OSPAR, annual emissions in 15 countries are lower in 2001 than in 1990, sometimes significantly e.g. 45%, 39% and 36% emission reduction in the Russian Federation, Poland and in the Netherlands, respectively.

In the Russian Federation, Poland and Germany ammonia (NH_3) emissions decreased in the beginning of the period investigated (approximately until 1994). From 1994 to 2001 emissions remained on the same level in almost all countries except Spain, where emissions are slightly rising from 1994 to 2001.

The rate of ammonia (NH_3) emission reductions is slightly lower than the rate of nitrogen oxides (NO_2) emission reduction in the period 1990 – 2001. Annual emissions of ammonia, as a sum of emissions from all OSPAR Parties and selected additional three countries outside OSPAR are 20% lower in 2001 than the annual emissions in the year 1990.

Annual emissions of nitrogen oxides (NO_2), as a sum of emissions from all OSPAR Parties and selected additional three countries outside OSPAR are 13% higher than the corresponding annual emissions of ammonia (NH_3) in the year 2001, in terms of nitrogen emitted.

5. Atmospheric supply of nitrogen to the OSPAR Convention Waters

Atmospheric depositions of oxidized nitrogen and reduced nitrogen were calculated for seven years; 1990, 1995, 1996, 1997, 1998, 1999 and 2000. Oxidized nitrogen deposition consists of the sum of PAN, NO_2, HNO_3 and aerosol nitrate (ammonium nitrate + coarse nitrate) depositions. Deposition of reduced nitrogen includes depositions of NH_3 and aerosol ammonium (ammonium sulphate + ammonium nitrate).

Maps of modelled annual deposition of oxidized nitrogen to the five main OSPAR regions in 1990 and 2000 are shown in Figures 6 and 7, respectively. Maps of annual deposition of reduced nitrogen in 1990 and 2000 are shown in Figures 8 and 9, respectively. Numerical values of the deposition to five main OSPAR regions for all seven years are included in Tables 3 and 4 for oxidized and reduced nitrogen, respectively.

For oxidized nitrogen, annual deposition to the main OSPAR regions is lower in 2001 than in 1990 in all main OSPAR regions, by 24.8%, 14.8%, 16.8%, 11.8% and 5.3% for Arctic Waters, Greater North Sea, Bay of Biscay and Wider Atlantic, respectively. Thus, the declining pattern of nitrogen oxides (NO_2) emissions is followed by the deposition pattern for the same period.

In case of reduced nitrogen, annual deposition in 2001 is higher than in 1990 in the main OSPAR Region 2, Greater North Sea (0.7%) and in the main OSPAR Region 3, Celtic Sea (5.6%). In the three remaining main OSPAR Regions it is lower by 29.9%, 10.6% and 15.5%, for Arctic Waters, Bay of Biscay and Wider Atlantic, respectively. It is clear from the figures that there is a large inter-annual variability of the depositions, probably caused by the different meteorological conditions in the different years. The year-to-year variations are large and have the same magnitude as the change in depositions in the ten year period from 1990 to 2001. This illustrates that in order to detect trends caused by changes in the emissions, many years of observational data are necessary. Moreover, model calculations can be a helpful tool to assess whether observed changes are related to changes in the meteorological conditions or to emission changes. For instance in the Wider Atlantic, depositions of nitrogen peaked in 1996/1997, whilst the relevant reported emissions exhibit no such peak. Both observations and model results at the Portuguese Atlantic coast point towards high wet depositions of nitrogen these years. Thus, the peaks of nitrogen depositions at the Wider Atlantic in the mid-1990's were likely caused by the specific meteorological conditions.

For both oxidized and reduced nitrogen, a clear gradient of the deposition towards the open sea can be noticed with maxima of the deposition in the OSPAR Region 2, Greater North Sea. Deposition of oxidized nitrogen (maximum 630 mg N m^{-2} in 2000) is higher than the deposition of reduced nitrogen (maximum 537 mg N m^{-2} in 2000). This means

that the nitrogen emitted from combustion sources contributes more to the deposition than the nitrogen emitted mainly from sources related to agriculture.

Annual total nitrogen depositions to the main OSPAR regions are shown in Figures 10 and 11, for the years 1990 and 2000, respectively. Numerical values of total (oxidized + reduced) nitrogen depositions for all seven years are given in Table 5. Also in this case a clear gradient of the deposition towards the open sea can be noticed with maximum of the deposition in the OSPAR Region 2, Greater North Sea. Compared to 1990, annual depositions of total nitrogen in 2000 were higher only in the main OSPAR Region 2 – Greater North Sea.

Annual nitrogen depositions to the entire area of the OSPAR Convention Waters are shown in Figure 11 for the years 1990, 1995,1996, 1997, 1998, 1999, 2000, 2001. Depositions of oxidized and total (oxidized + reduced) nitrogen are going down from 1990 to 1995, whereas depositions of reduced nitrogen are slightly higher in 1995 than in 1990. There is a clear maximum for all three types of nitrogen deposition in the year 1996 and then decreasing until 1999.. From 1999 to 2001, depositions of oxidized and total nitrogen are going slightly up, but depositions of reduced nitrogen are going slightly down.

Maps of modelled annual deposition of oxidized nitrogen to the sub-regions of the OSPAR Region 2 (Greater North Sea) in 1990 and 2000 are shown in Figures 12 and 13, respectively. Maps of annual deposition of reduced nitrogen in 1990 and 2000 are shown in Figures 14 and 15, respectively. Numerical values of the deposition to 13 sub-regions of the Greater North Sea for all seven years are given in Tables 6 and 7 for oxidized and reduced nitrogen, respectively.

For oxidized nitrogen, annual deposition to all sub-regions except sub-regions 2 and 4 is lower in 2001 than in 1990 by 2% to 30%. Depositions of oxidized nitrogen to sub-regions 6 and 13 remain on the same level in 2001 than in 1990. Maxima (over 600 mg N m^{-2}) of the deposition can be noticed close to the Norwegian and Swedish costs in sub-region 12 and minimum (below 150 mg N m^{-2}) in sub-region 2.

Annual deposition of reduced nitrogen in 2001 is higher than in 1990 in 8 out of 13 sub-regions of the Greater North Sea. For some of sub-regions deposition increase is significant e.g. 30% and 22% for sub-regions 2 and 9, respectively. Maxima (over 1000 mg N m^{-2}) of the deposition can be observed close to German, Belgian and French cost and minimum (below 150 mg N m^{-2}) again in sub-region 2. Lower deposition in 2001 than in 1990 can be especialy noticed in sub-regions 9 and 12 with 22% and 20% reduction, respectively.

Annual total nitrogen (oxidized + reduced nitrogen) depositions to the sub-regions of the Greater North Sea are shown in Figures 16 and 17, for the years 1990 and 2001, respectively. Numerical values of total (oxidized + reduced) nitrogen depositions for all seven years are given in Table 8. Compared to 1990, annual depositions of total nitrogen in 2001 are higher in 5 out of 13 sub-regions and especially in the east-south border of the Greater North Sea with maximum over 1000 mg N m^{-2}.

Maps with time series of oxidized, reduced and total nitrogen deposition to the five main OSPAR regions are shown for the years 1990, 1995, 1996, 1997, 1998, 1999 and 2000 in Figures 18, 19 and 20, respectively. For all types of the deposition and for all components maximum in the four main OSPAR regions (Arctic Waters, Celtic Sea, Bay of Biscay, Wider Atlantic) occurs in 1996. In the Greater North Sea region maximum of the deposition for all components occurs in 2000.

Maps with time series of oxidized, reduced and total nitrogen deposition to the 13 sub-regions of the main OSPAR Region 2, Greater North Sea are shown for the years 1990, 1995, 1996, 1997, 1998, 1999 and 2000 in Figures 21, 22 and 23, respectively. Deposition values of oxidized nitrogen in 2000 are lower than 1990 depositions in all sub-regions. Deposition of reduced nitrogen is higher in 2000 then in 1990 in many sub-regions and no clear trend is visible in the deposition pattern. The same applies to the deposition of total nitrogen.

6. Comparison of computed vs. measured depositions at

OSPAR stations

Maps with modelled vs. measured annual deposition of oxidized, reduced and total (oxidized and reduced) nitrogen for the years 1990, 1995, 1996, 1997, 1998, 1999 and 2000 at the stations located in the main OSPAR regions are shown in Figures 24, 25 and 26, respectively. Similar maps with measured versus calculated depositions of oxidized, reduced and total nitrogen, for the stations located in the OSPAR Region 2 (Greater North Sea), are shown in Figures 27, 28 and 29, respectively.

The agreement between model results and observations depend not only on the "model performance" and the adequacy of emissions employed, but also on the quality and representativeness of the measurement sites. It is worth mentioning here that for some stations, double sets of measurement data (e.g. obtained with different methods) have been reported. In some cases, these data sets differ by as much as 20-30 %. This indicates that there is a substantial uncertainty in the measurements. Thus, the following discussion on model *underestimation* and *overestimation* simply implies that the calculated values are lower or higher than the observations and does not refer to model deficiency only.

For the majority of the stations, the modelled and measured concentrations match well. In general, modelled wet deposition for stations situated in the sub-regions are in better agreement with observations than stations located in the main regions. For instance, wet deposition of both oxidized and reduced nitrogen for the Portugal station PT0010R, situated at the Azores, is substantially underestimated by the model. The observed values are relatively high for a background station, presumably due to the observations being influenced by local sources.

Both this station and the Norwegian station at Spitzbergen, NO0059R, are situated close to the boundary of the model domain. Therefore, the model results are sensitive to the choice of boundary conditions in the model. Consequently, the results for these stations are expected to be worse than for other stations.

Observed wet deposition for sites situated in the sub-regions agree very well with modelled wet deposition. It is especially encouraging that the model manages to reproduce the monitored year-to-year variations.

In general, wet deposition of ammonium and nitrate is somewhat underestimated. It is well known that dry deposition of NH_3 to open bulk collectors can account for a substantial part of the measured wet deposition. Thus, the apparent under prediction of wet depositions may partly be caused by the bias in measured wet deposition.

7. Source-receptor matrices for the main OSPAR regions

8. Source-receptor matrices for sub-regions of the Greater

North Sea

9. Remarks about uncertainties

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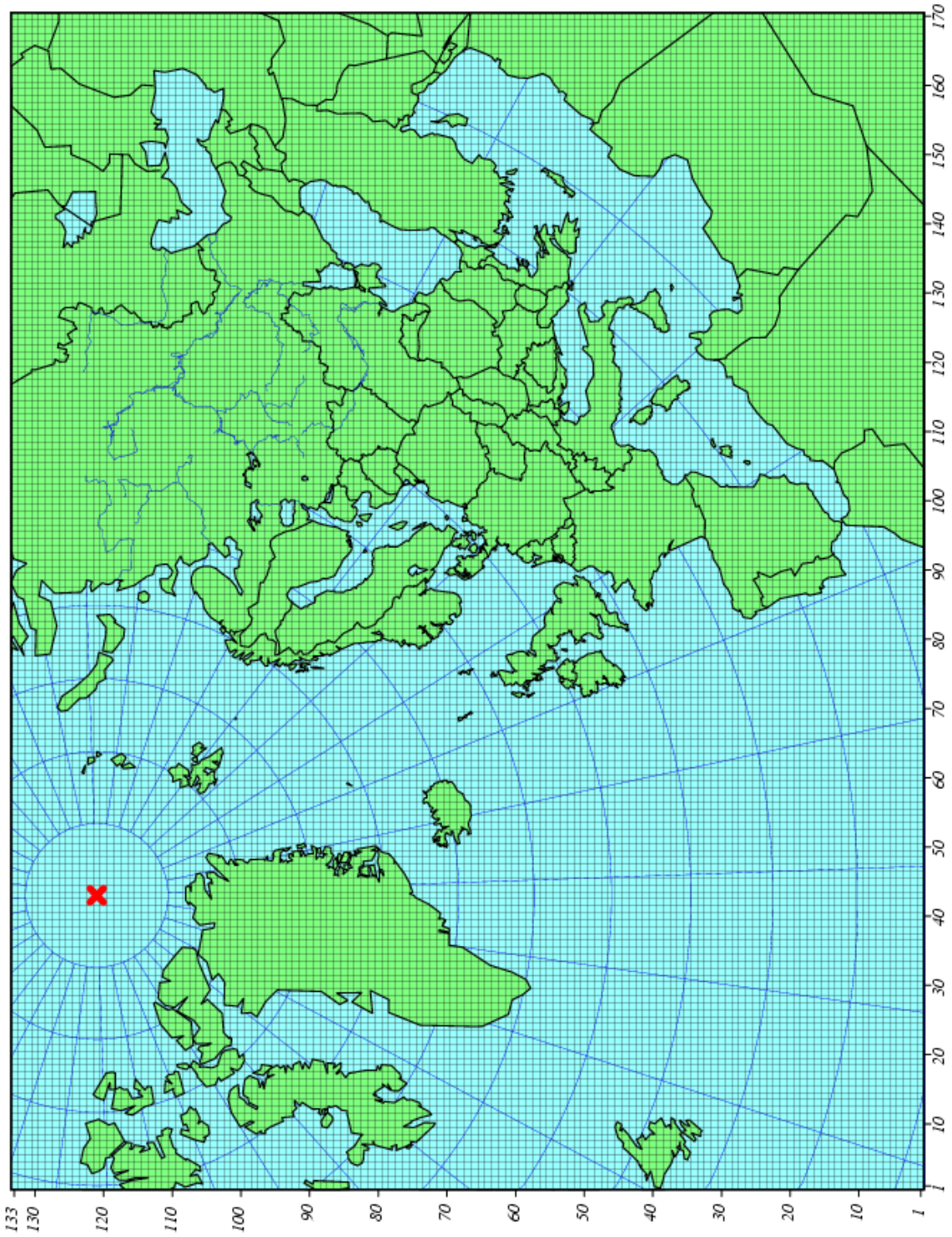

Figure 1. The EMEP grid system of the size 170×133 in the Polar Stereographic projection. The grid resolution is 50 km at 60°N.

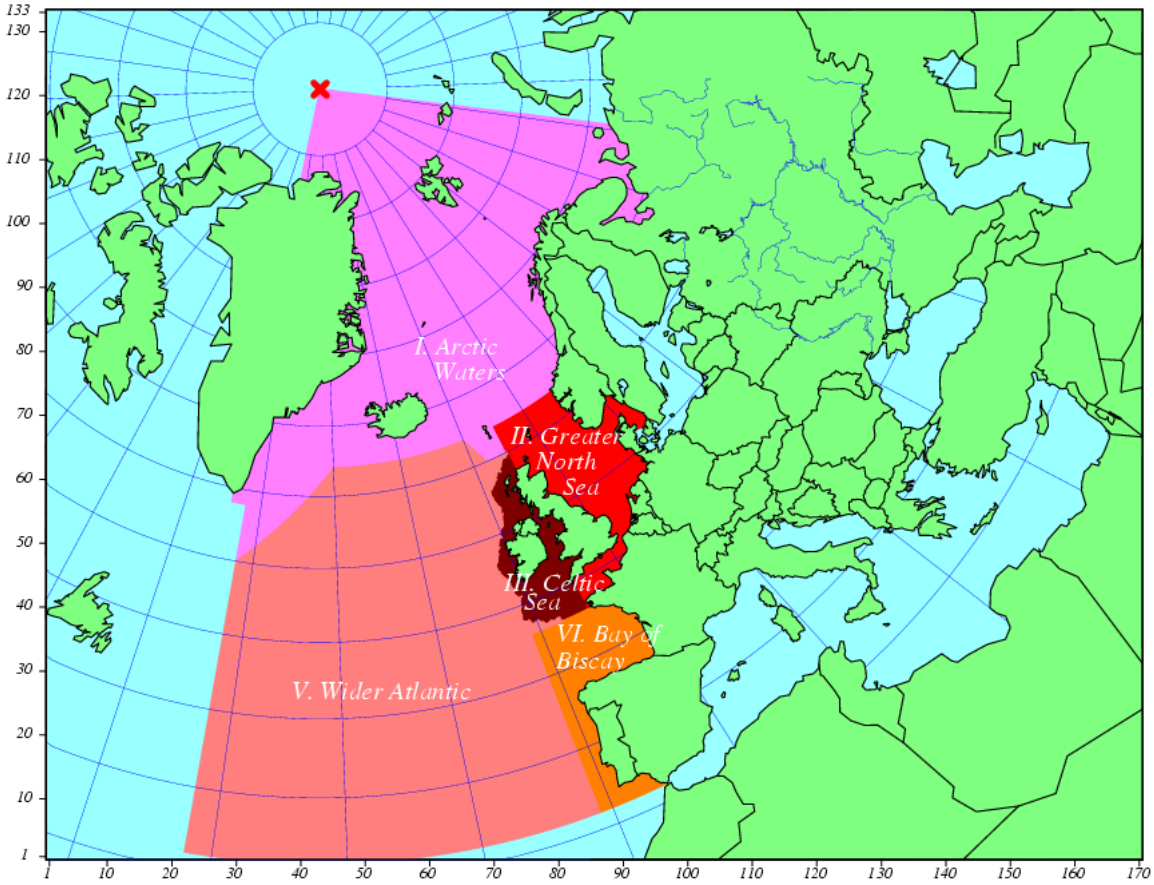

Figure 2. Borders of the five main OSPAR regions in the EMEP grid system.

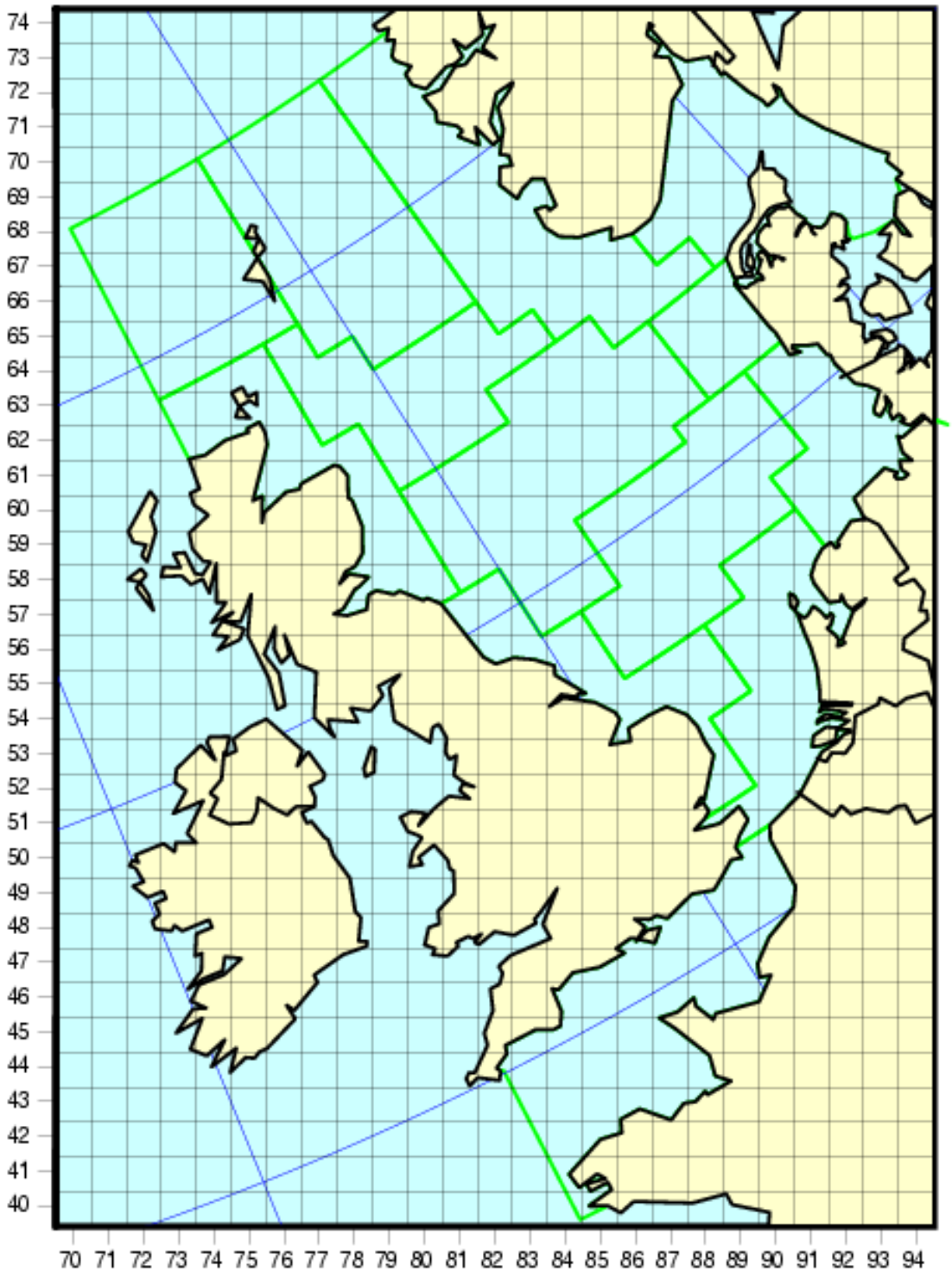

Figure 3. Borders of the 13 sub-regions of OSPAR Region 2 (Greater North Sea) in the EMEP grid system.

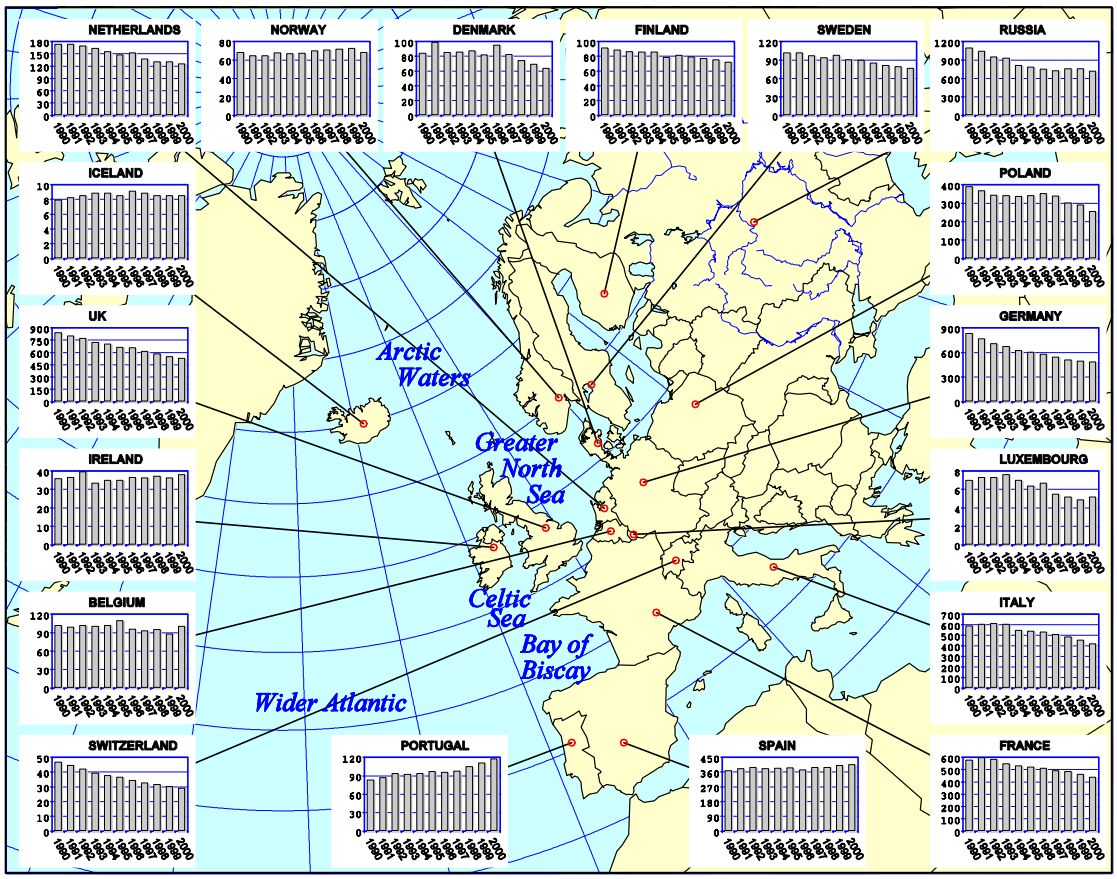

Figure 4. Time series of nitrogen oxides (NO_2) emissions from the OSPAR Contracting Parties, and selected countries (Poland, Italy and part of the Russian Federation within the EMEP domain) with the largest contribution to deposition on the OSPAR maritime area. Unit: kt N a^{-1}.

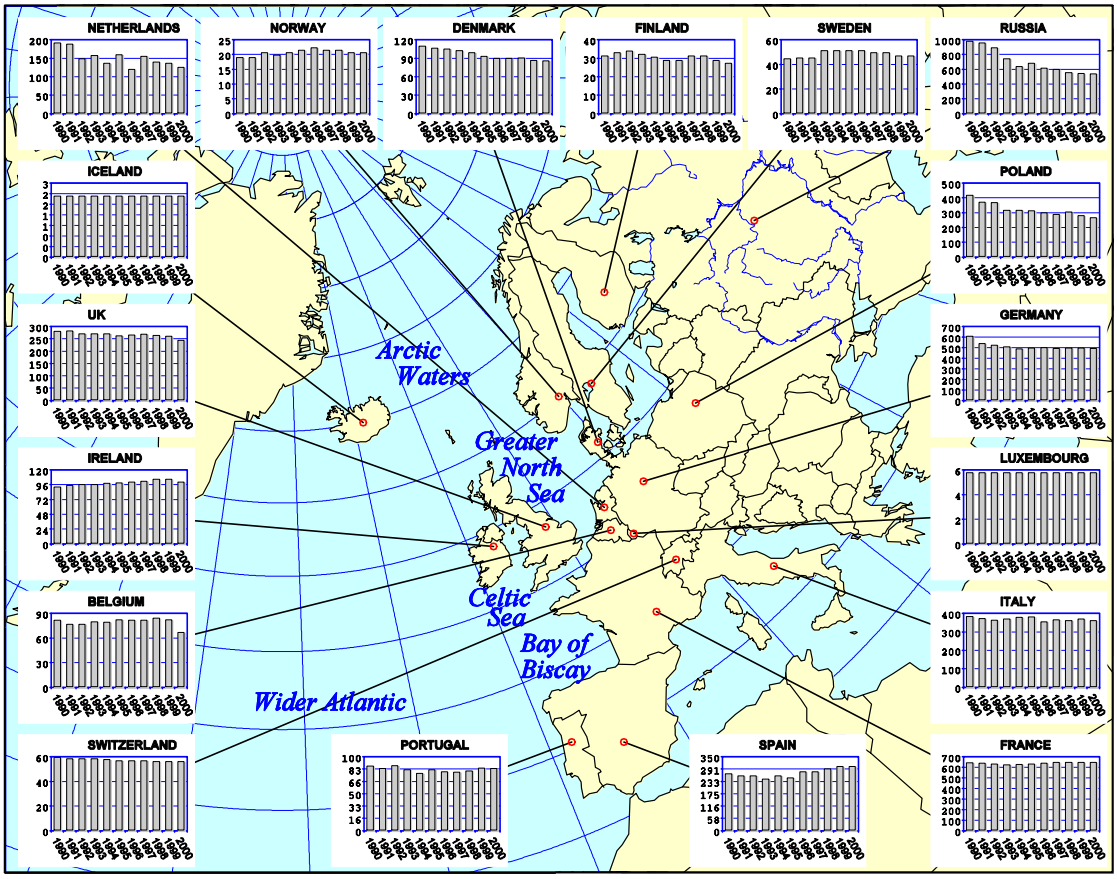

Figure 5. Time series of ammonia (NH_3) emissions from the OSPAR Parties, and selected countries (Poland, Italy and part of the Russian Federation within the EMEP domain) with the largest contribution to deposition on the OSPAR maritime area. Unit: kt N a^{-1}.

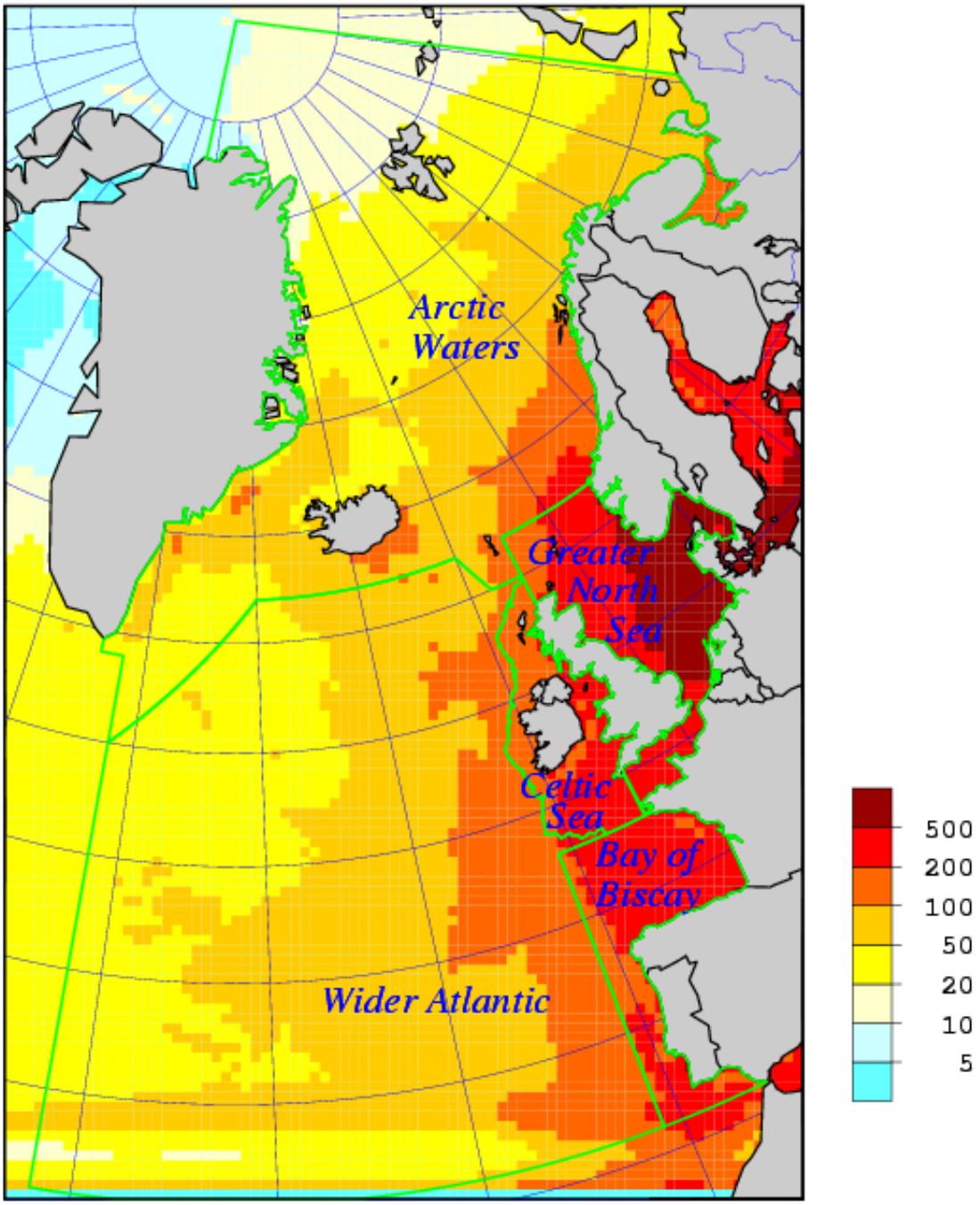

Figure 6. Map of modelled annual oxidized nitrogen deposition to the five main OSPAR regions in 1990. Unit: mg N m^{-2}.

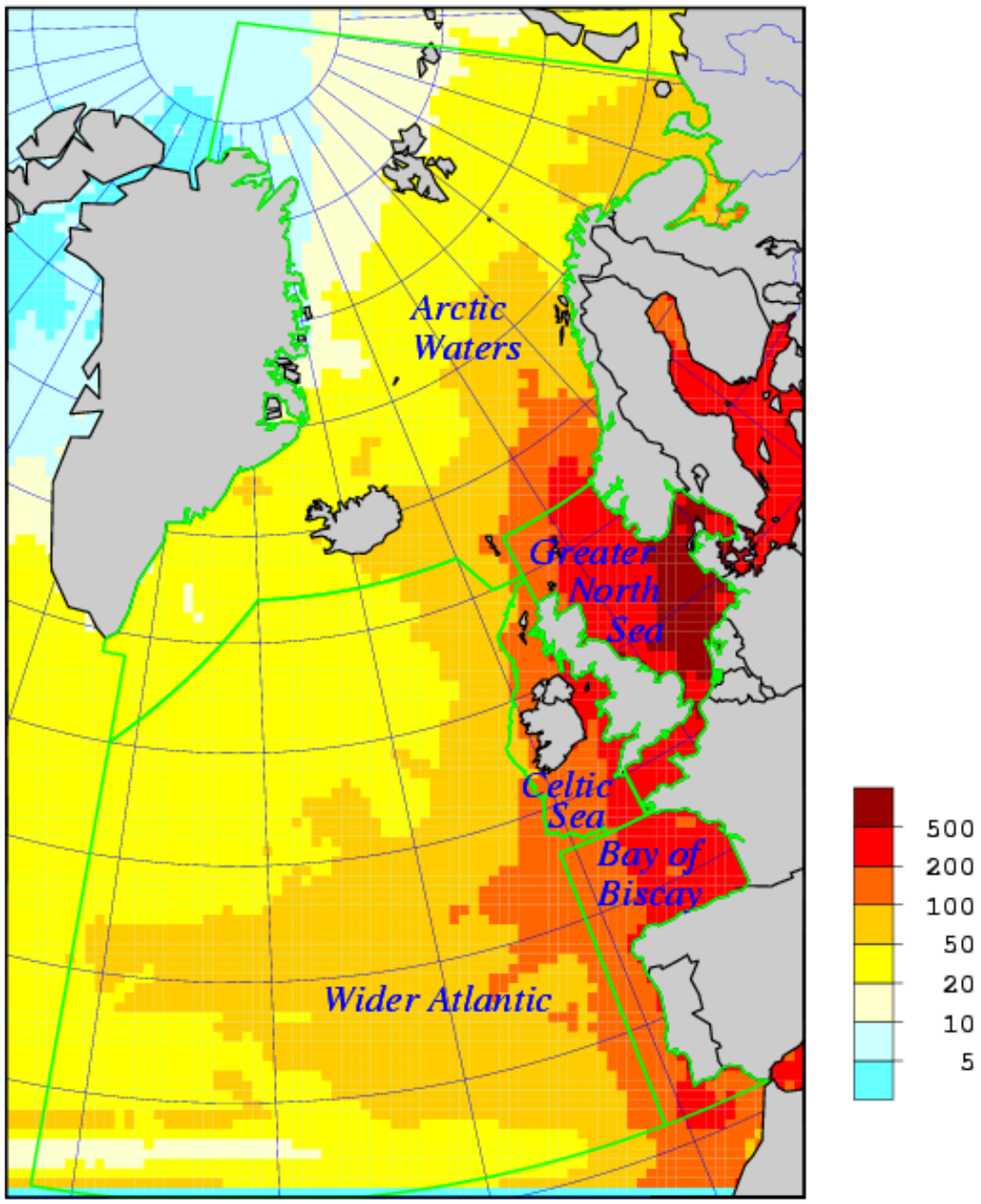

Figure 7. Map of modelled annual oxidized nitrogen deposition to the five main OSPAR regions in 2000. Unit: mg N m^{-2}.

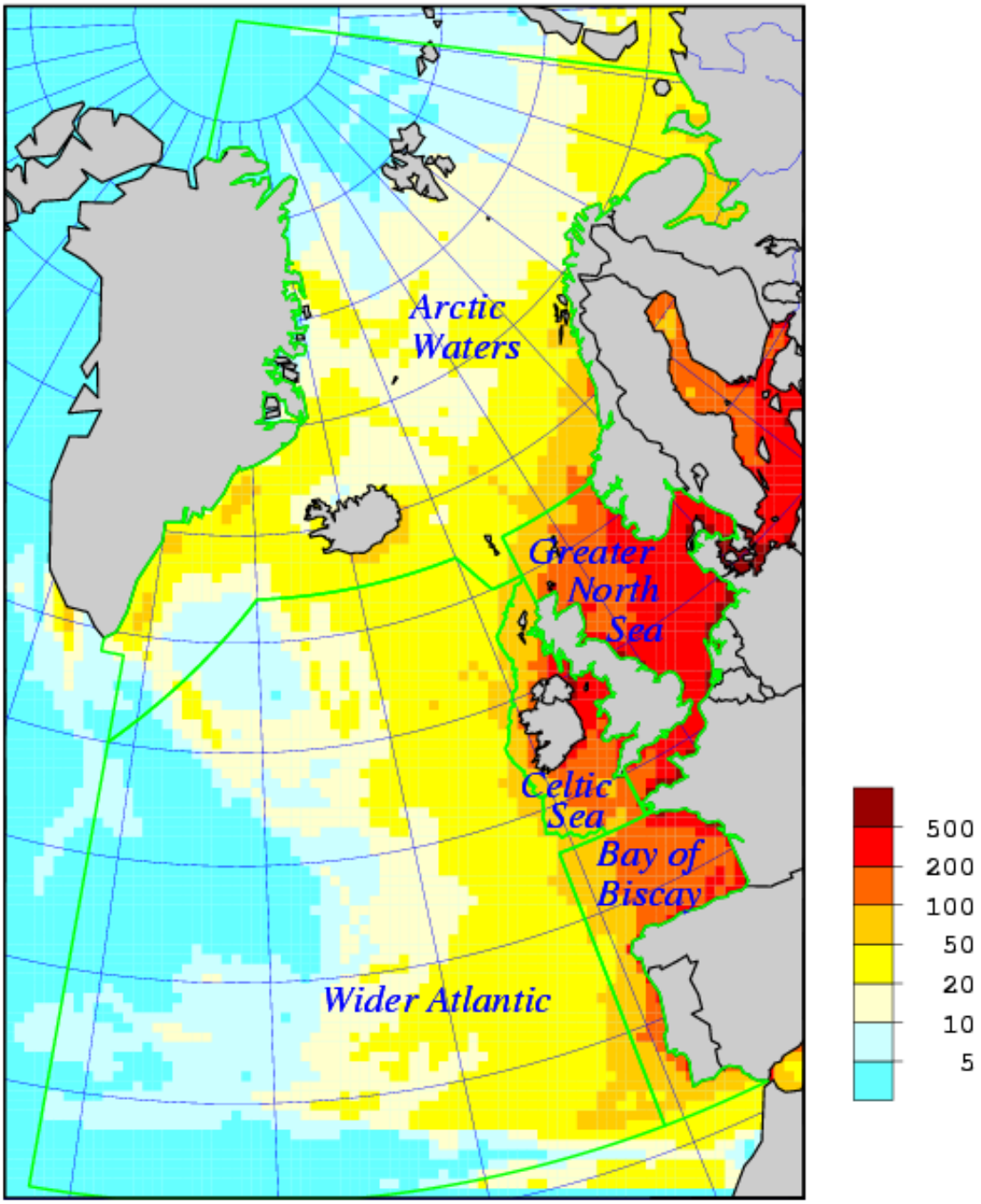

Figure 8. Map of modelled annual reduced nitrogen deposition to the five main OSPAR regions in 1990. Unit: mg N m^{-2}.

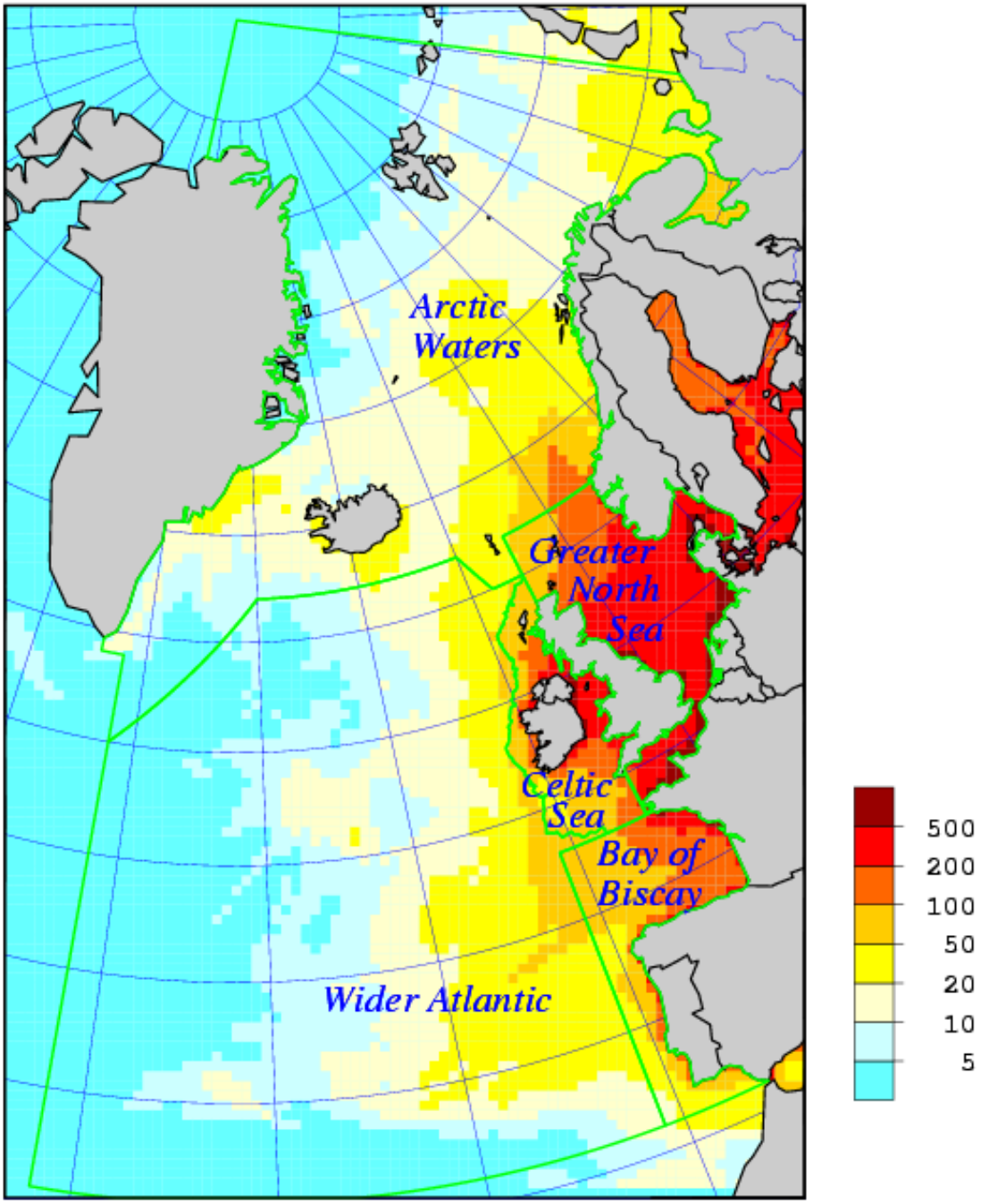

Figure 9. Map of modelled annual reduced nitrogen deposition to the five main OSPAR regions in 2000. Unit: mg N m^{-2}.

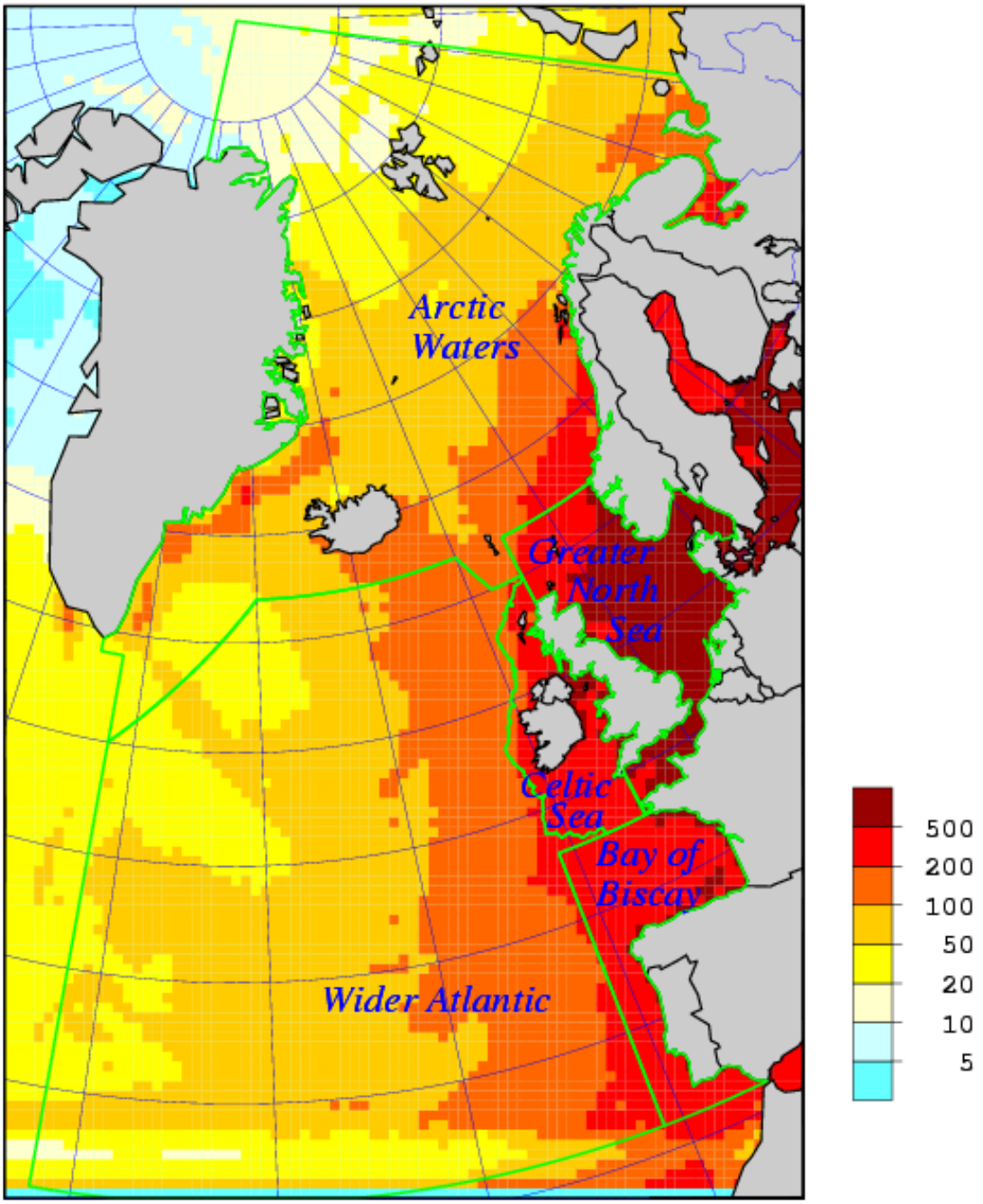

Figure 10. Map of modelled annual total nitrogen deposition to the five main OSPAR regions in 1990. Unit: mg N m^{-2}.

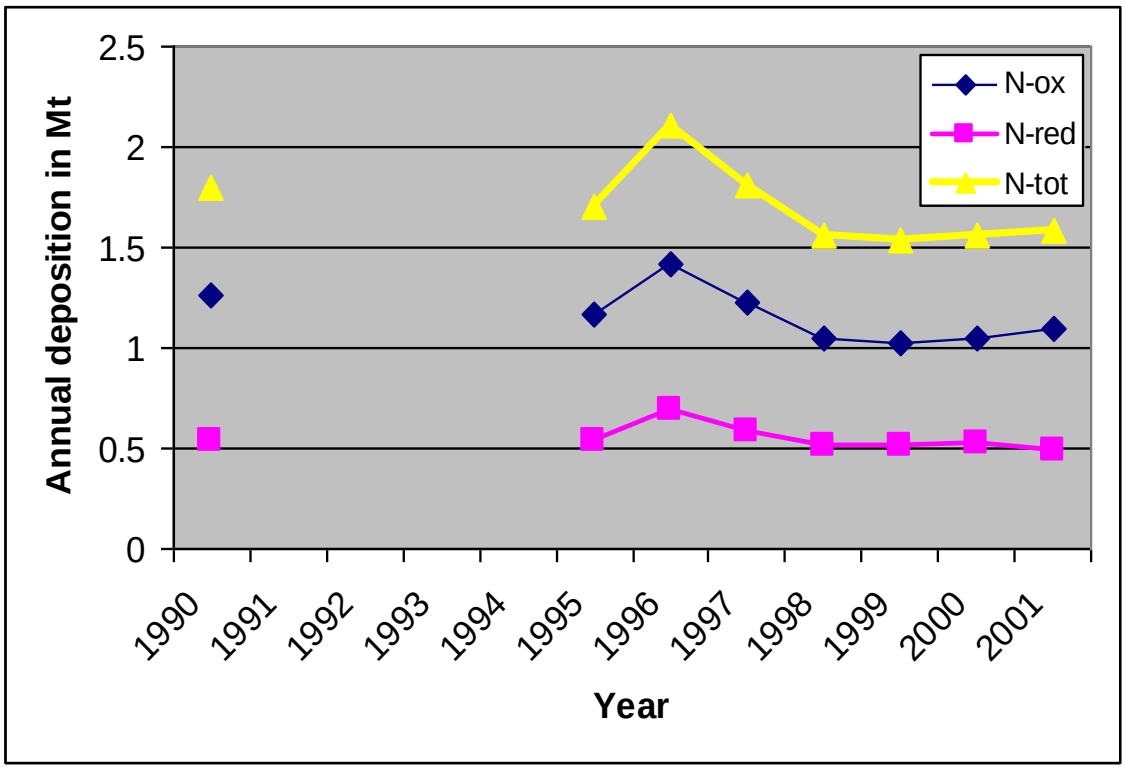

Figure 11. Annual nitrogen depositions to the entire area of the OSPAR Convention Waters in the period 1990 - 2001. Unit: Mt N a^{-1}.

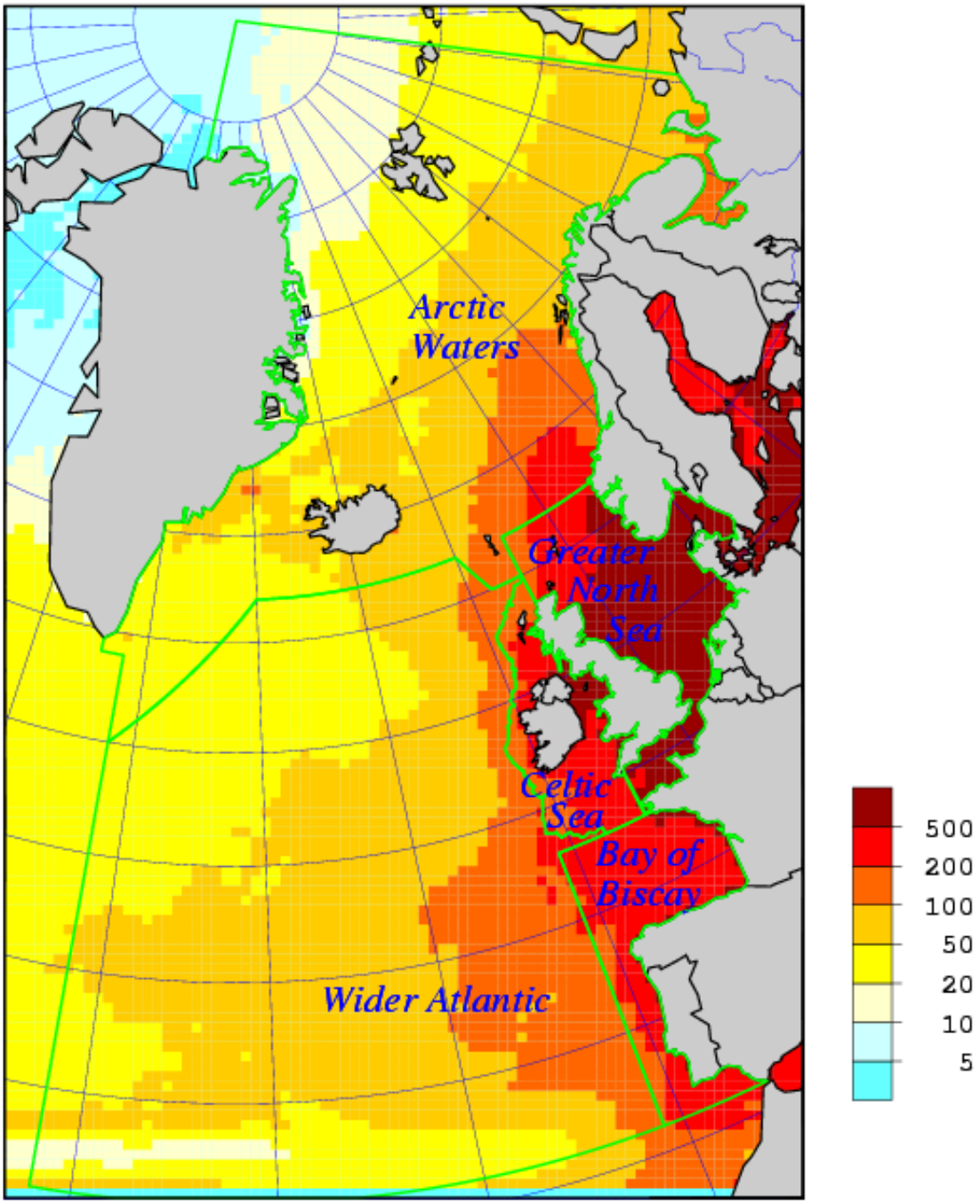

Figure 12. Map of modelled annual total nitrogen (oxidized and reduced) deposition to the five main OSPAR regions in 2000. Unit: mg N m^{-2}.

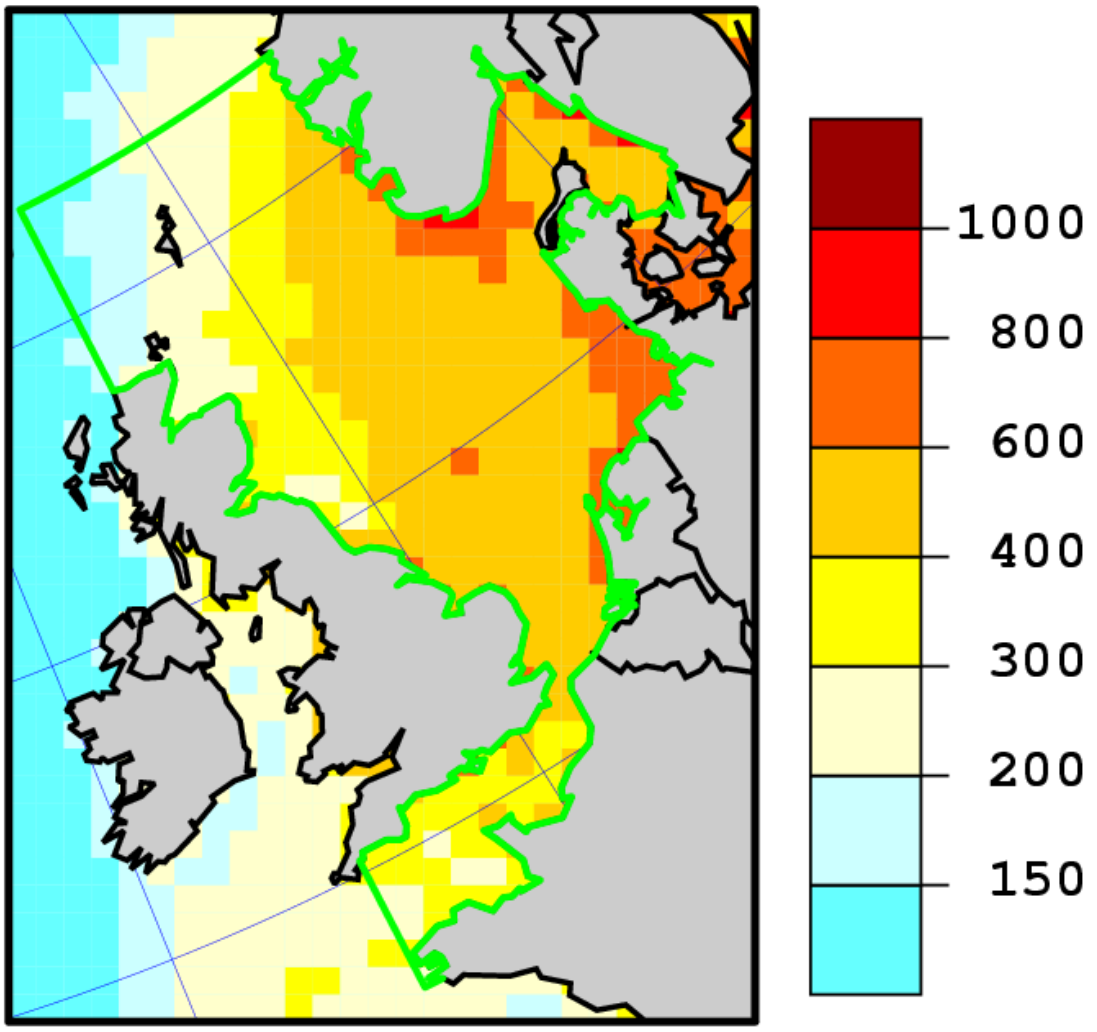

Figure 13. Map of modelled annual oxidized nitrogen deposition to 13 sub-regions of the OSPAR Region 2 (Greater North Sea) in 1990. Unit: mg N m^{-2}.

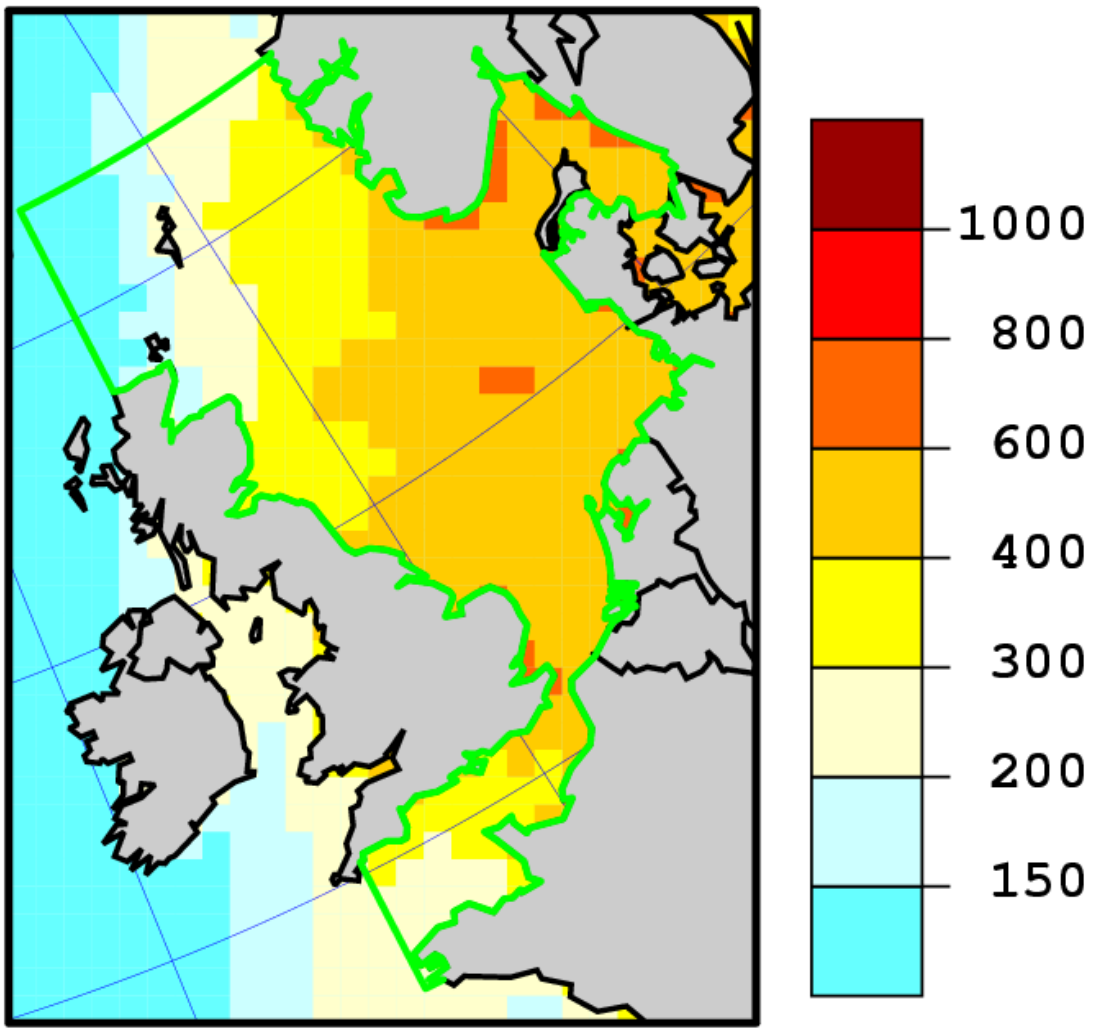

Figure 14. Map of modelled annual oxidized nitrogen deposition to 13 sub-regions of the OSPAR Region 2 (Greater North Sea) in 2000. Unit: mg N m^{-2}.

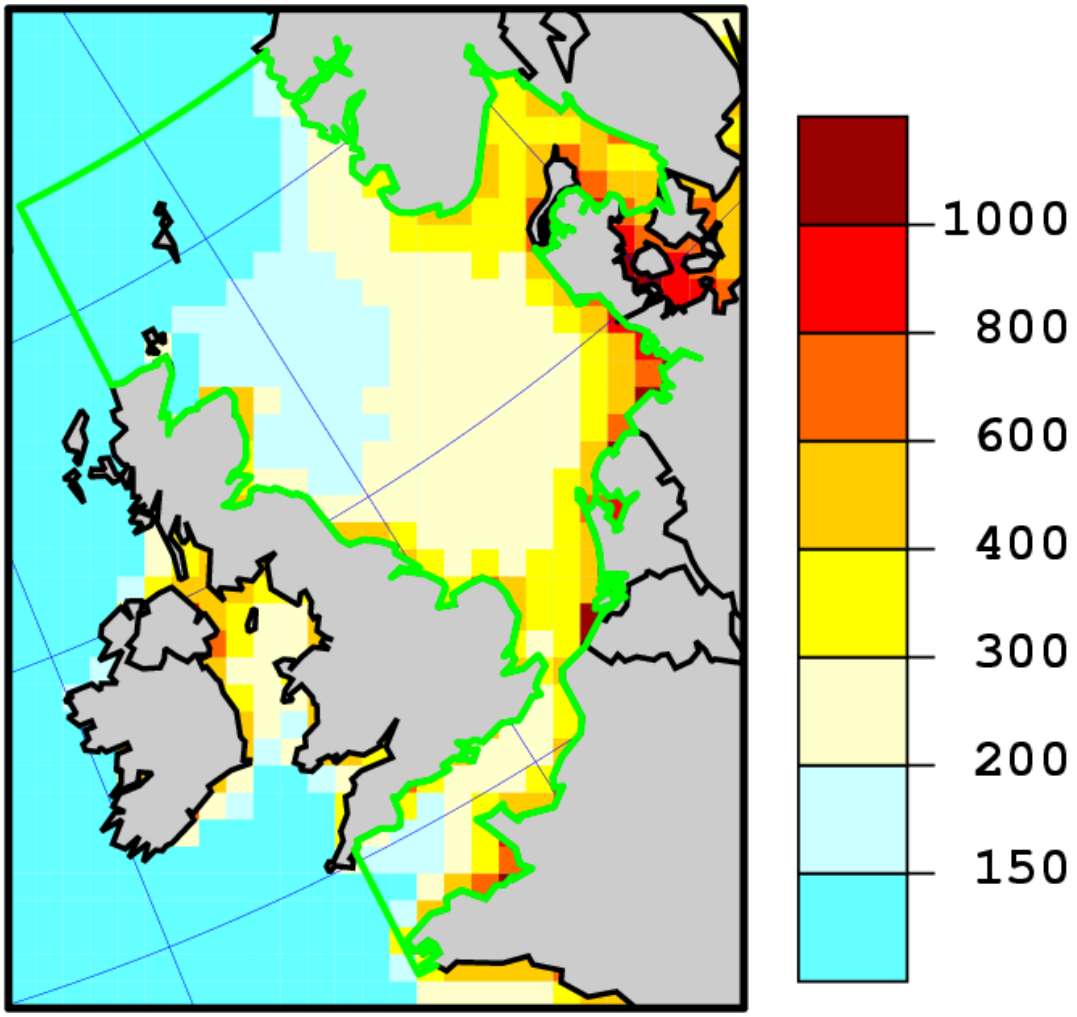

Figure 15. Map of modelled annual reduced nitrogen deposition to 13 sub-regions of the OSPAR Region 2 (Greater North Sea) in 1990. Unit: mg N m^{-2}.

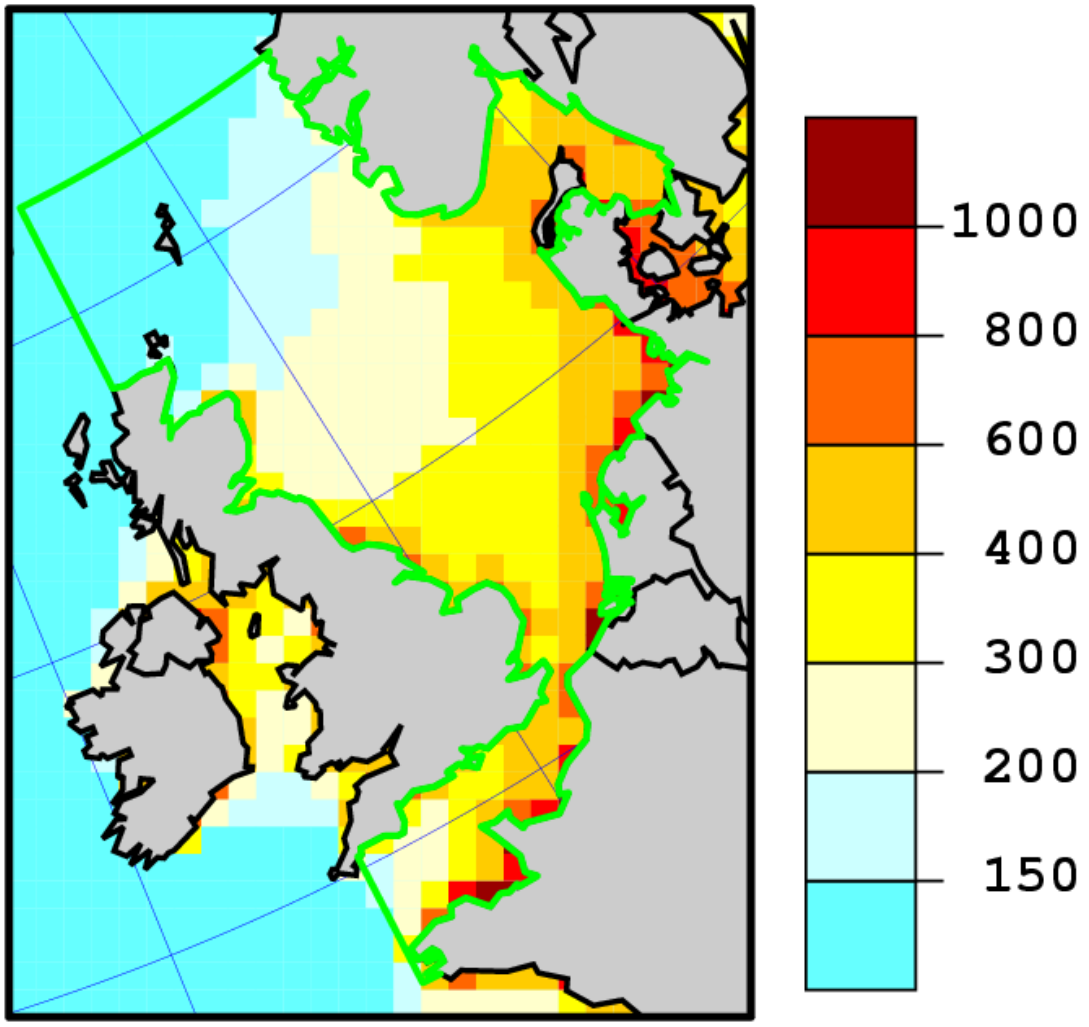

Figure 16. Map of modelled annual reduced nitrogen deposition to 13 sub-regions of the OSPAR Region 2 (Greater North Sea) in 2000. Unit: mg N m^{-2}.

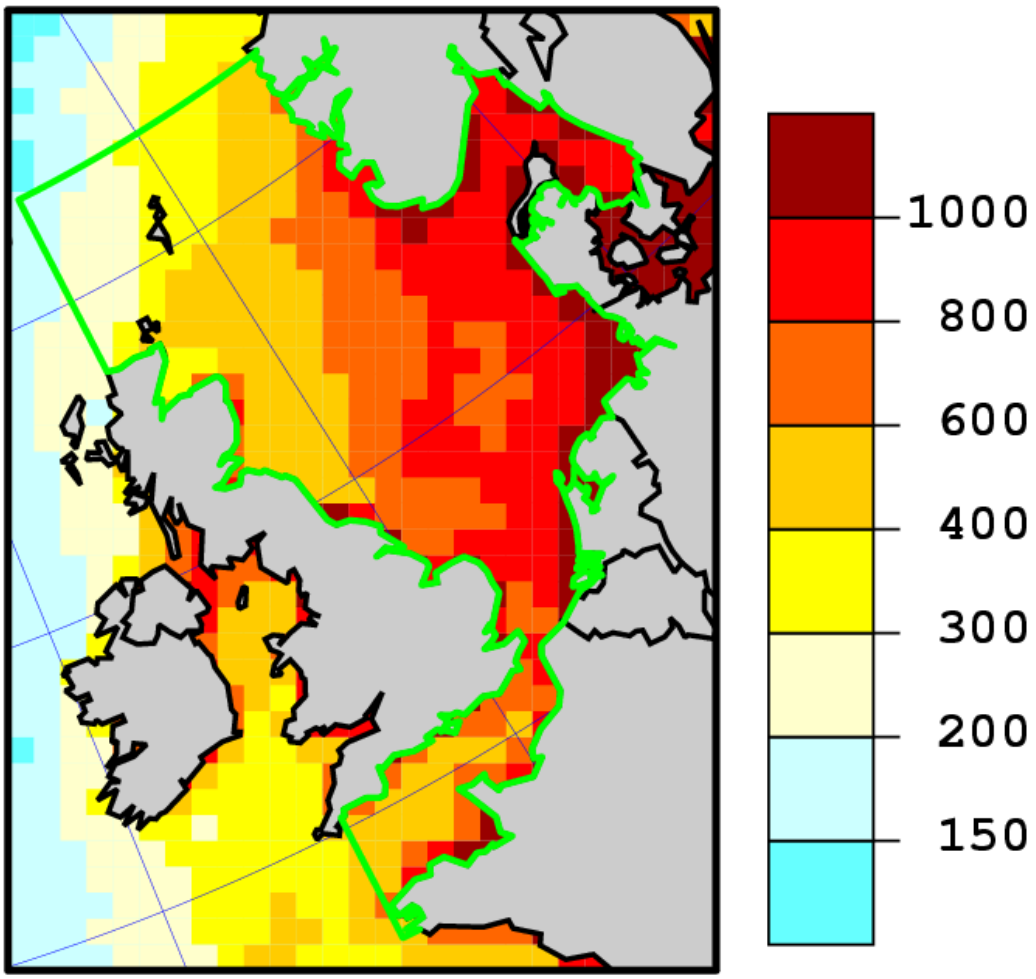

Figure 17. Map of modelled annual total nitrogen (oxidized and reduced) deposition to 13 sub-regions of the OSPAR Region 2 (Greater North Sea) in 1990. Unit: mg N m^{-2}.

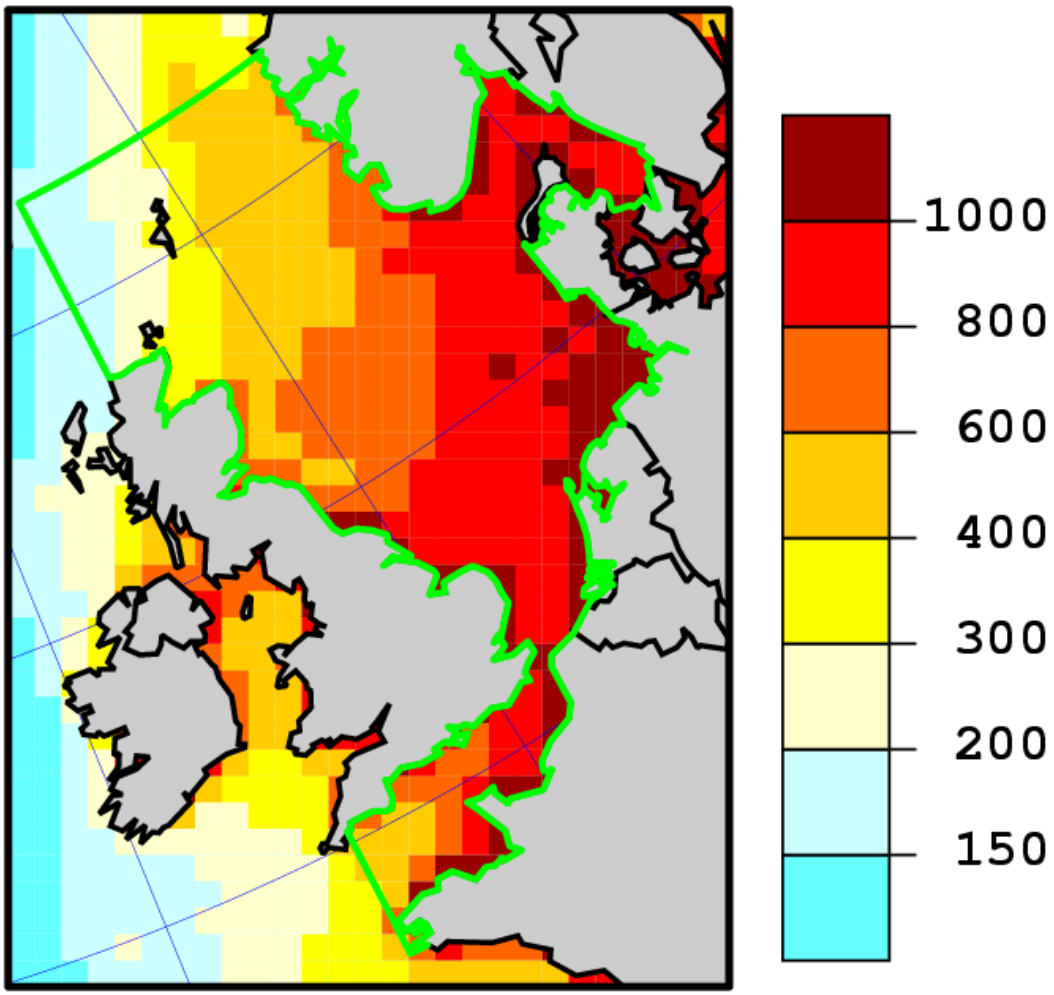

Figure 18. Map of modelled annual total nitrogen (oxidized and reduced) deposition to 13 sub-regions of the OSPAR Region 2 (Greater North Sea) in 2000. Unit: mg N m^{-2}.

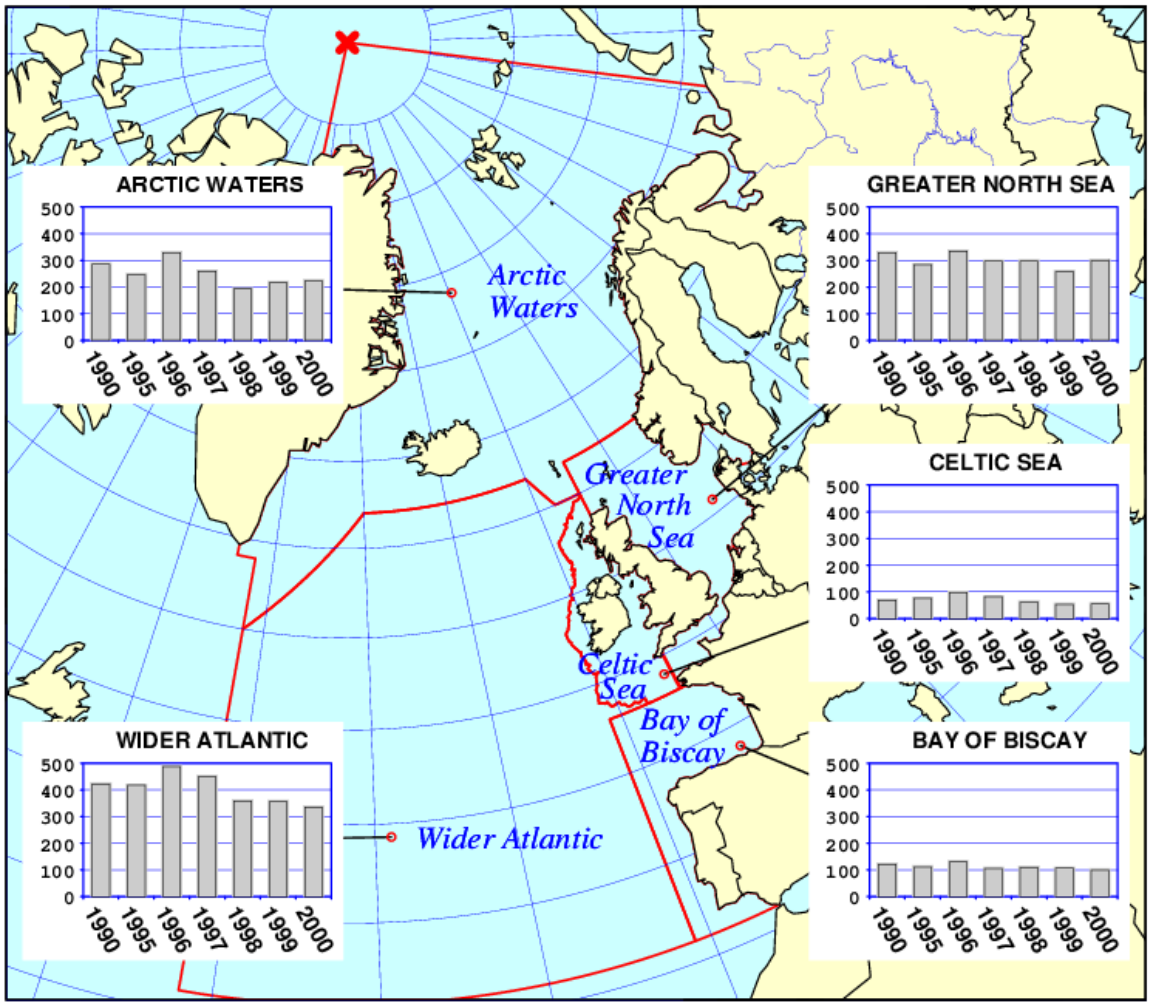

Figure 19. Map with time series of modelled annual oxidized nitrogen depositions to the five main OSPAR regions. Unit: kt N a^{-1}.

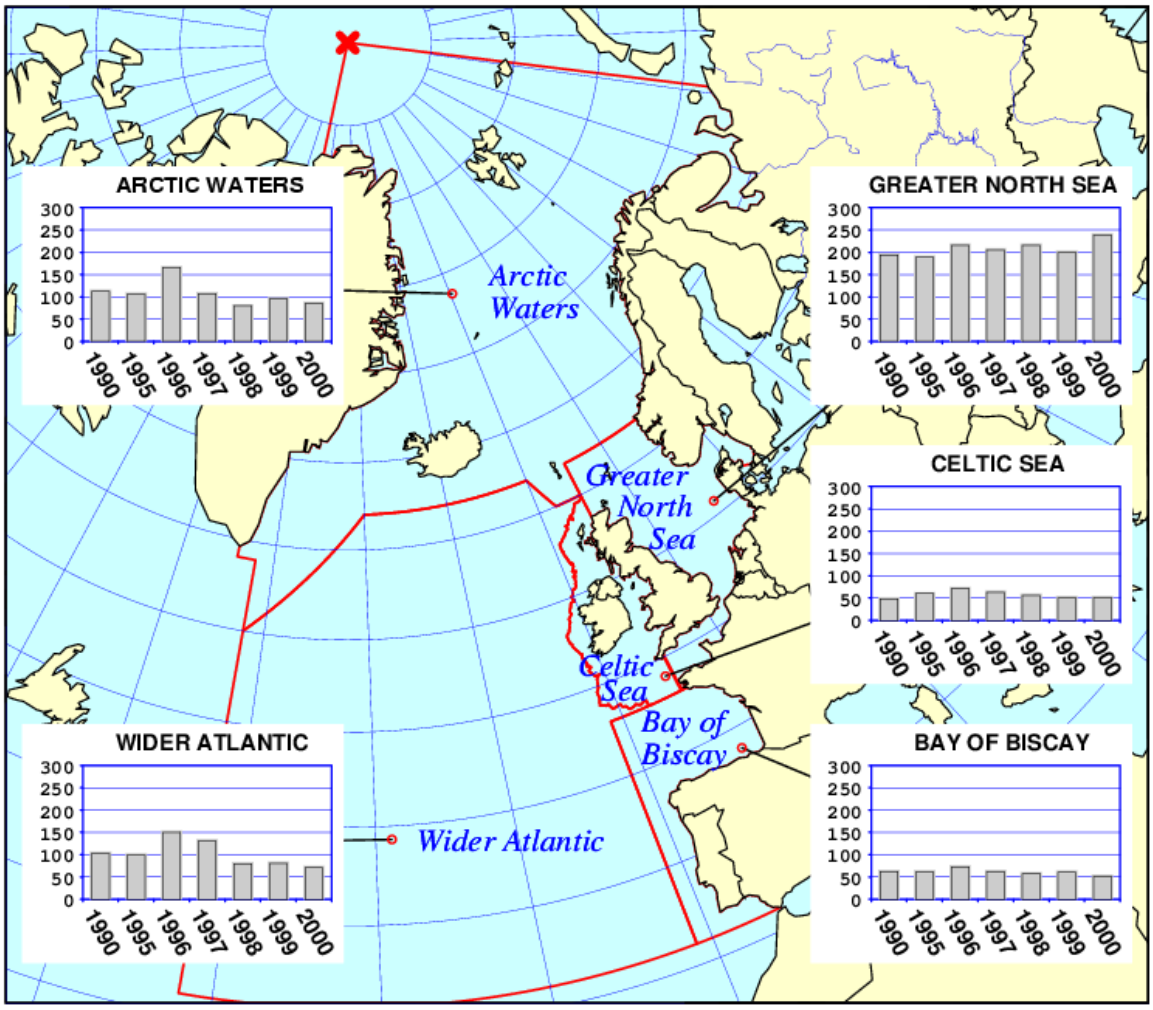

Figure 20. Map with time series of modelled annual reduced nitrogen depositions to the five main OSPAR regions. Unit: kt N a^{-1}.

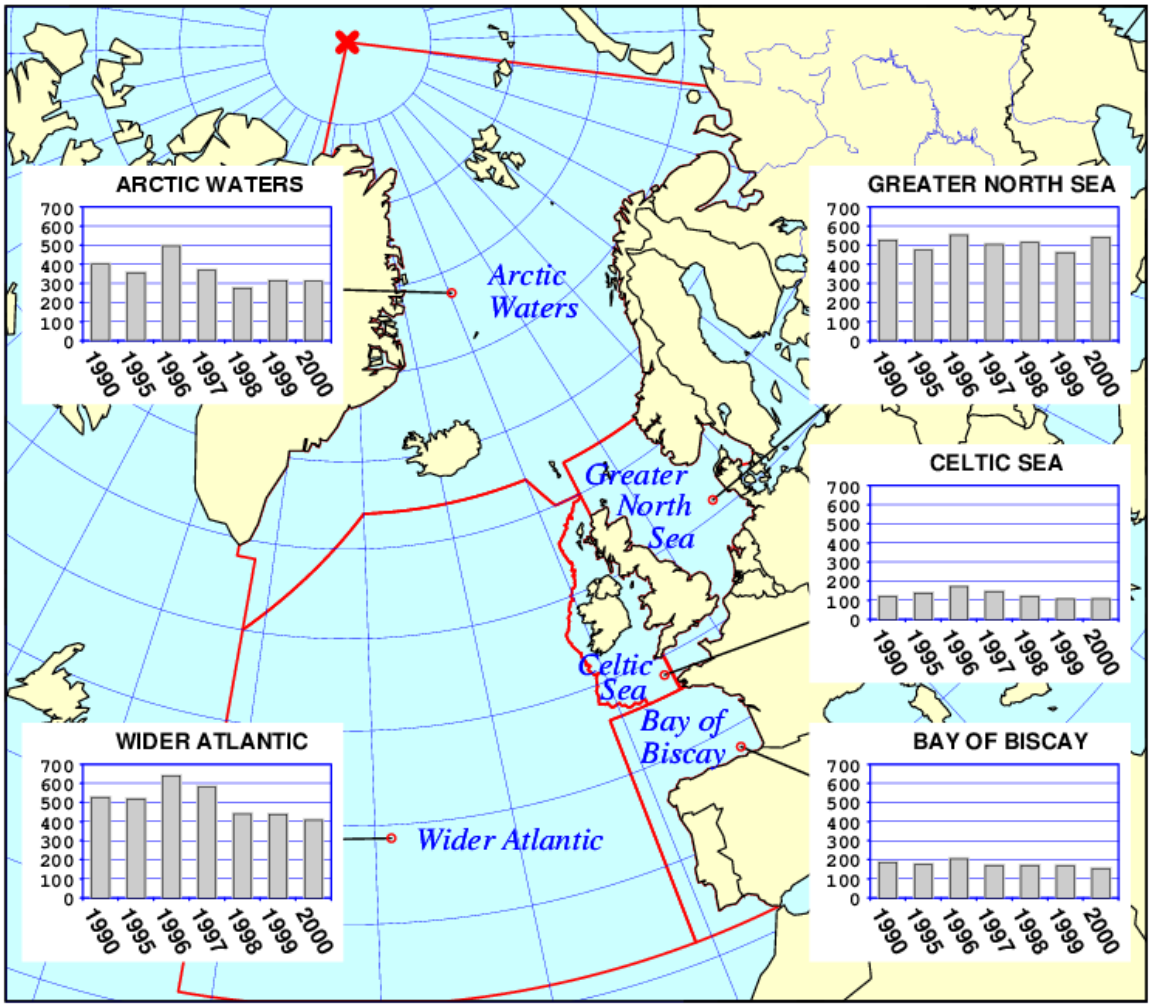

Figure 21. Map with time series of modelled annual total nitrogen (oxidized + reduced) depositions to the five main OSPAR regions. Unit: kt N a^{-1}.

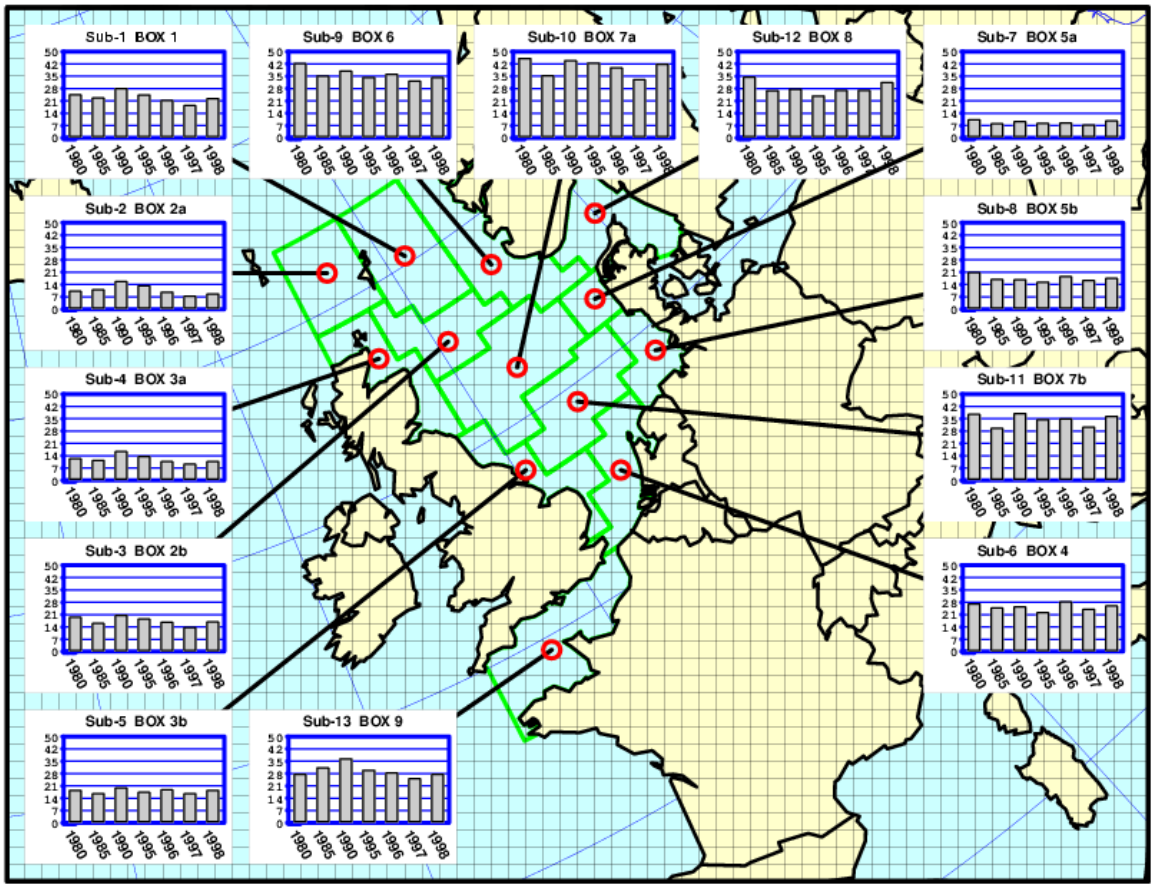

Figure 22. Map with time series of modelled annual oxidized nitrogen depositions to 13 sub-regions of the OSPAR Region 2 (Greater North Sea). Unit: kt N a^{-1}.

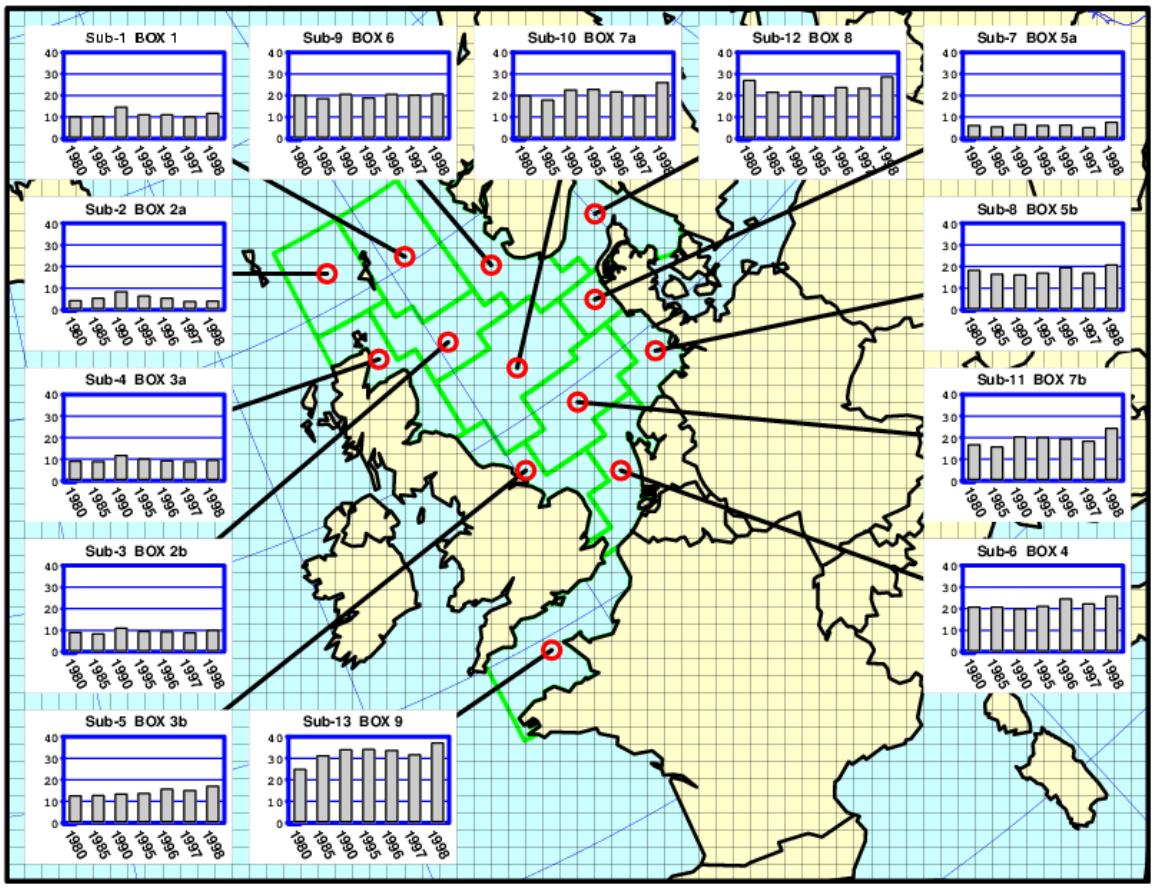

Figure 23. Map with time series of modelled annual reduced nitrogen depositions to 13 sub-regions of the OSPAR Region 2 (Greater North Sea). Unit: kt N a^{-1}.

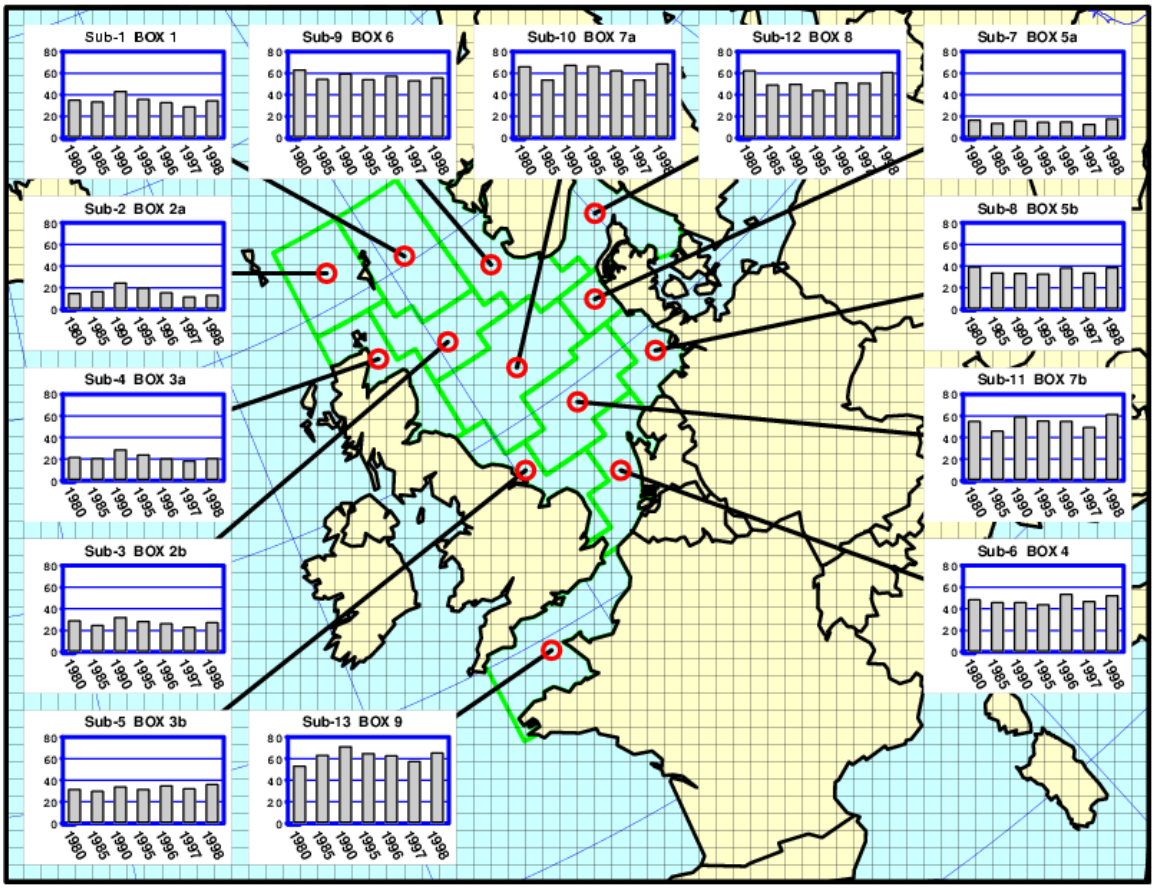

Figure 24. Map with time series of modelled annual total nitrogen (oxidized + reduced) depositions to 13 sub-regions of the OSPAR Region 2 (Greater North Sea). Unit: kt N a^{-1}

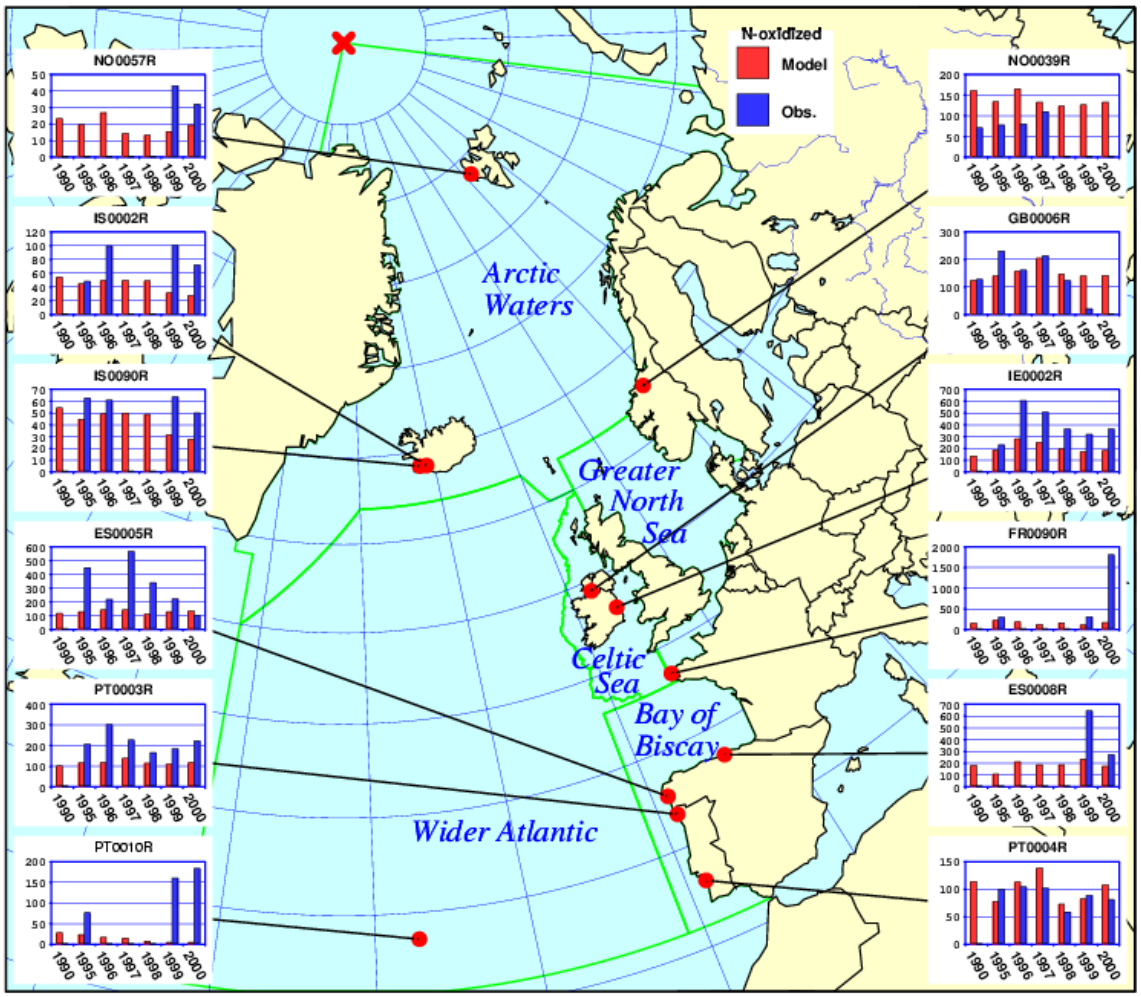

Figure 25. Map with time series of modelled versus measured annual oxidized nitrogen depositions at the stations in the five main OSPAR regions. Unit: mg N m^{-2}.

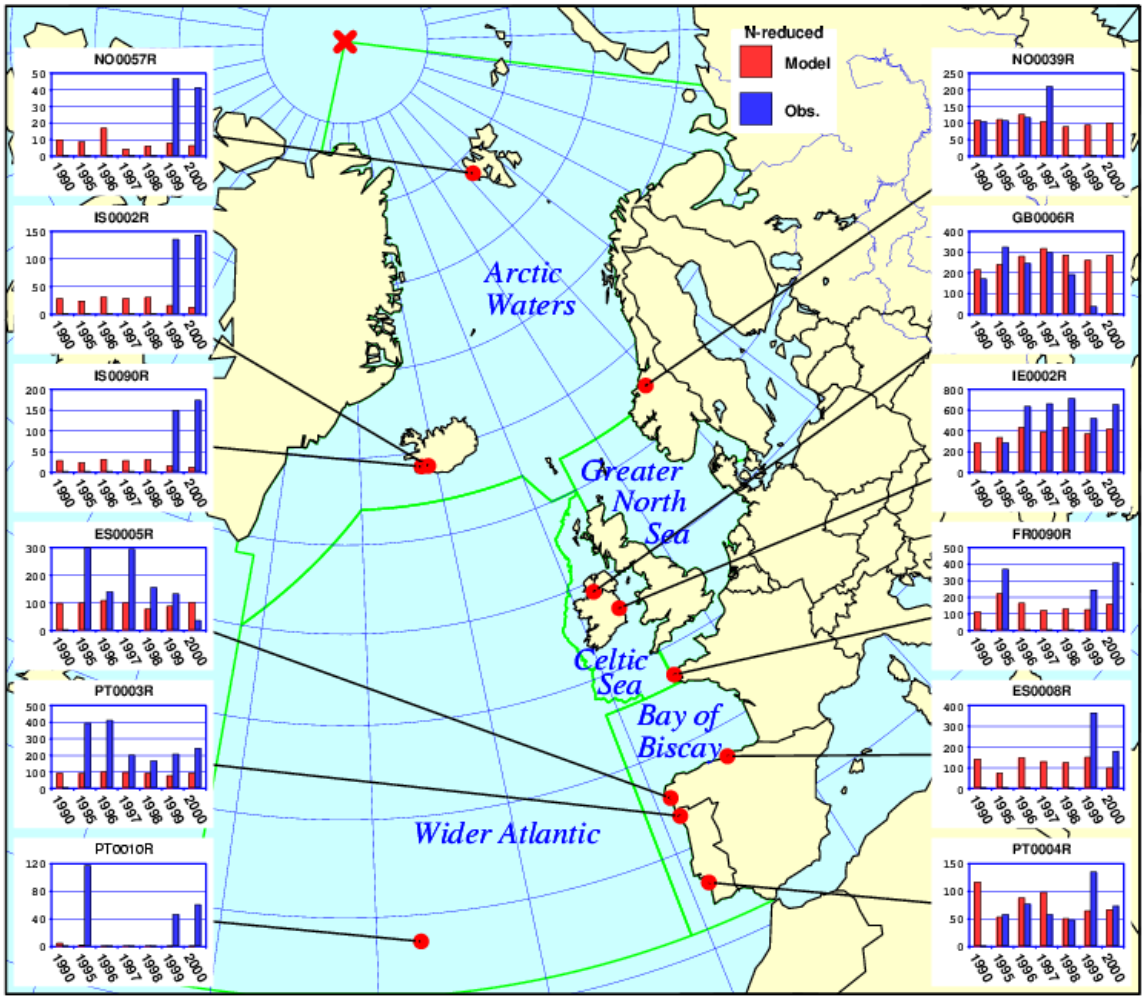

Figure 26. Map with time series of modelled versus measured annual reduced nitrogen depositions at the stations in the five main OSPAR regions. Unit: mg N m^{-2}.

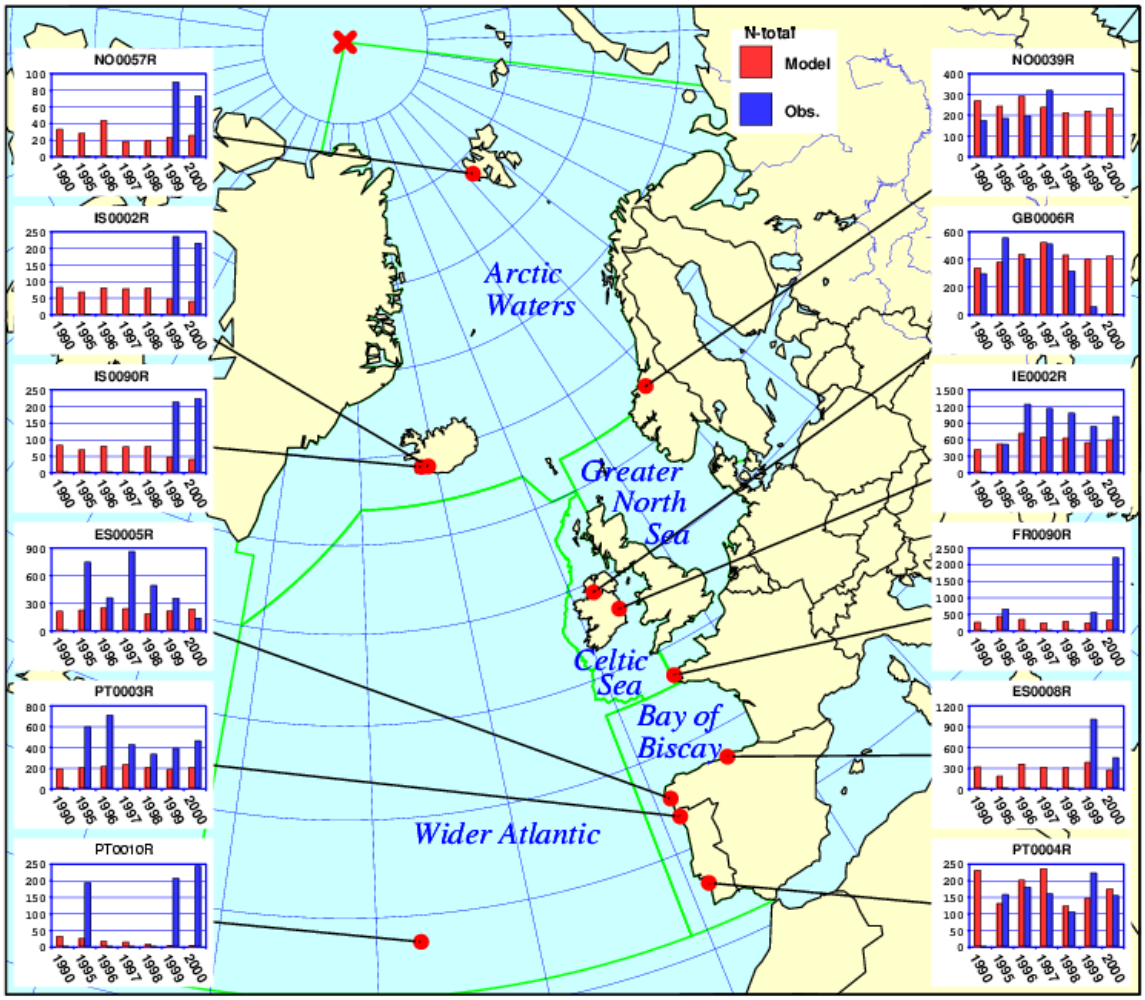

Figure 27. Map with time series of modelled versus measured annual total nitrogen (oxidized + reduced) depositions at the stations in the five main OSPAR regions. Unit: mg N m^{-2}.

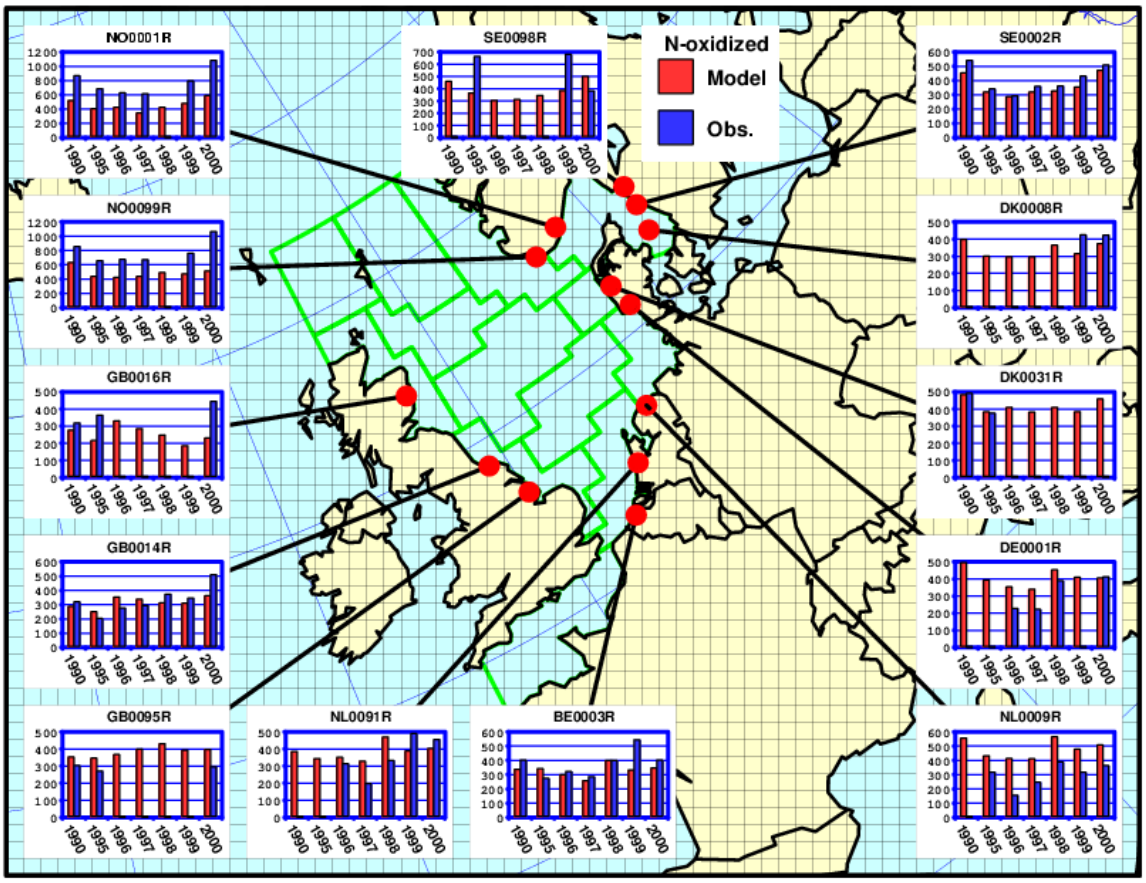

Figure 28. Map with time series of modelled versus measured annual oxidized nitrogen depositions at the stations in the OSPAR Region 2 (Greater North Sea) . Unit: mg N m^{-2}.

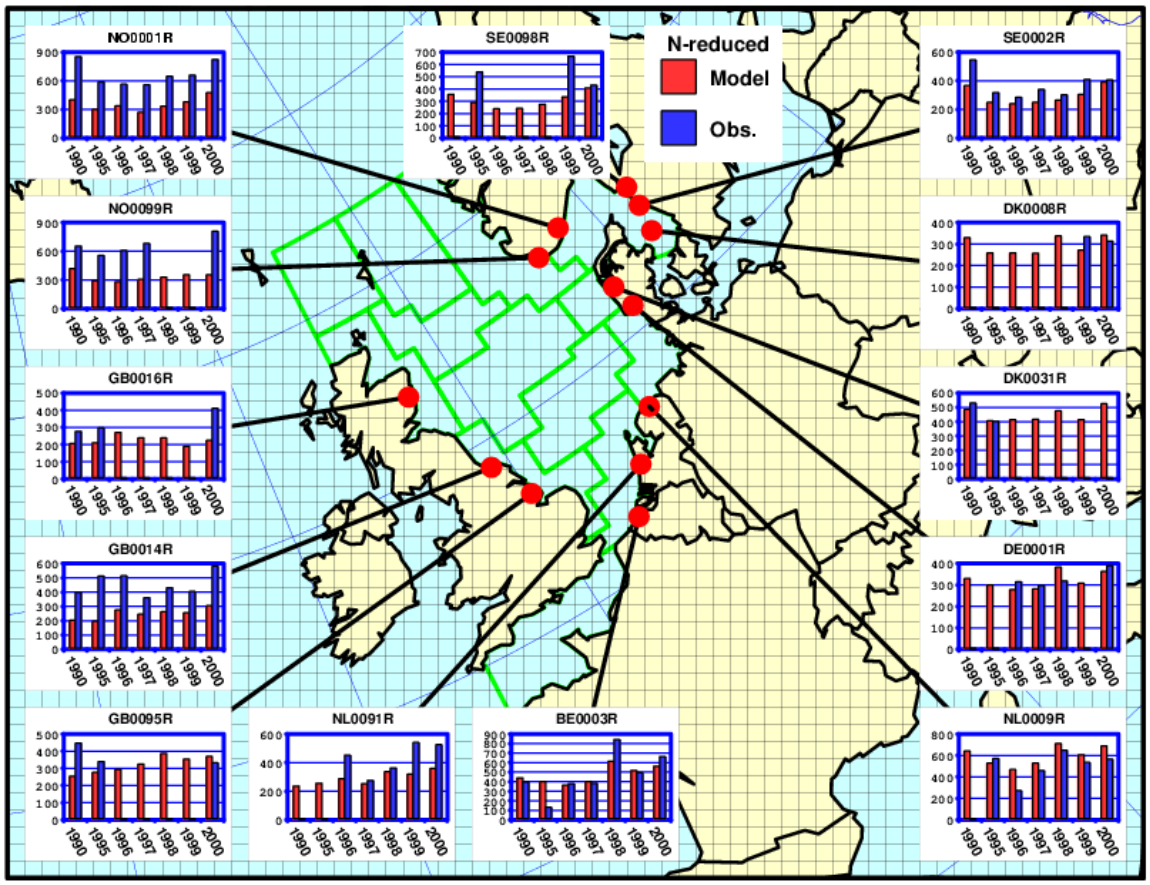

Figure 29. Map with time series of modelled versus measured annual reduced nitrogen depositions at the stations in the OSPAR Region 2 (Greater North Sea) . Unit: mg N m^{-2}.

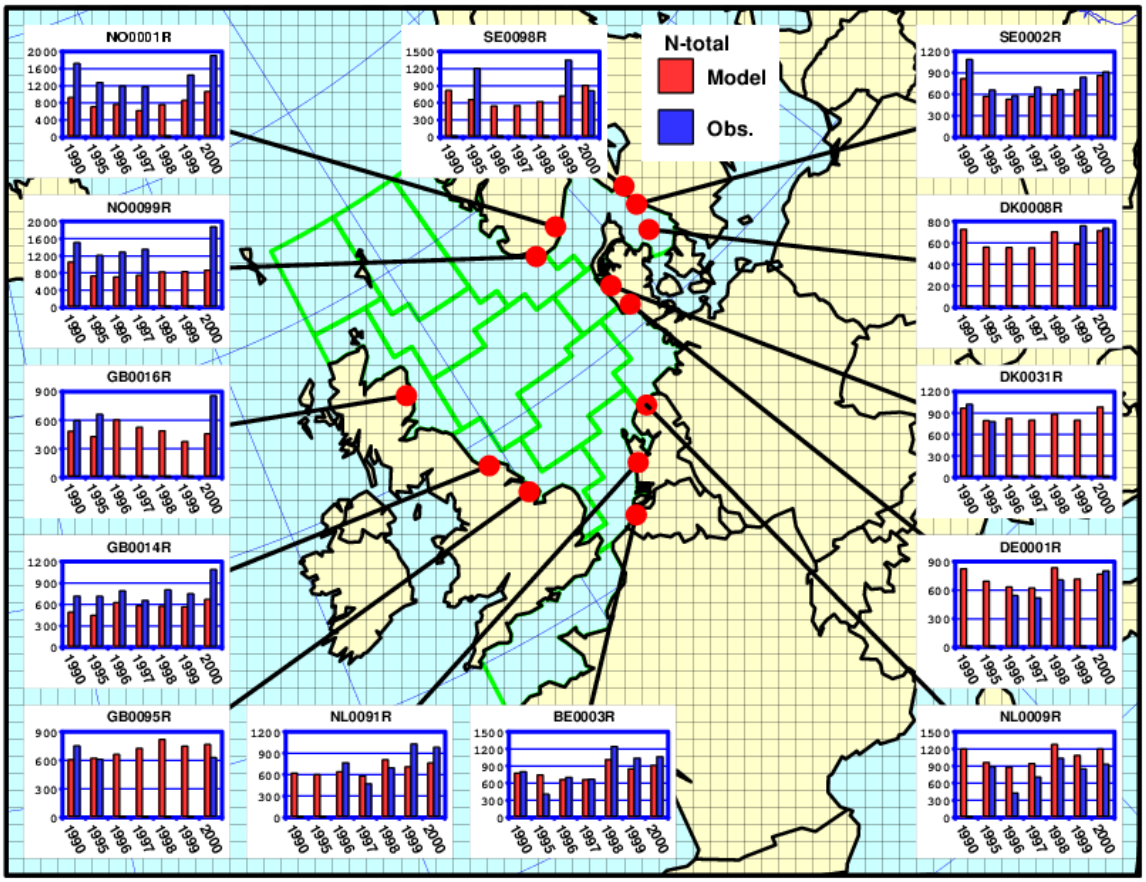

Figure 30. Map with time series of modelled versus measured annual total nitrogen (oxidized + reduced) depositions at the stations in the OSPAR Region 2 (Greater North Sea). Unit: mg N m^{-2}.

Table 1. Annual nitrogen oxides (NO_2) emissions from the OSPAR Contracting Parties and three main contributors to the deposition. Unit: kt N a^{-1}.

Country	Year											
	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
Belgium	102	99	102	100	101	109	96	93	95	88	100	96
Denmark	84	99	85	86	87	82	95	82	74	69	64	62
Finland	91	88	86	86	86	79	82	79	77	75	72	68
France	577	598	584	547	532	520	511	490	485	462	439	429
Germany	830	765	707	672	625	604	577	543	510	493	482	485
Iceland	8	8	9	9	9	9	9	9	9	9	9	9
Ireland	36	37	40	33	35	35	37	36	37	36	38	38
Luxembourg	7	7	7	8	7	6	7	5	5	5	5	5
Netherlands	173	173	169	163	155	148	152	138	130	131	126	125
Norway	68	65	65	68	67	67	70	71	72	72	68	67
Portugal	83	87	94	92	93	97	96	98	105	111	117	115
Spain	367	380	389	381	383	387	375	389	387	400	406	397
Switzerland	47	44	42	39	38	37	34	33	32	30	29	28
Sweden	102	102	97	93	97	90	90	85	81	79	77	75
UK	840	801	771	720	700	662	659	612	584	551	529	511
Italy	590	604	612	606	544	538	531	506	485	452	418	418
Poland	390	367	344	341	336	341	351	339	302	289	255	245
Russia	1096	1045	950	929	812	782	751	724	757	759	717	717

Table 2. Annual ammonia emissions (NH_3) from the OSPAR Contracting Parties and three main contributors to the deposition. Unit: kt N a^{-1}.

Country	Year											
	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
Belgium	82	77	77	80	79	82	82	82	84	82	67	67
Denmark	110	106	105	102	99	93	90	90	91	86	86	84
Finland	31	33	34	32	30	29	29	31	31	29	27	27
France	642	637	630	623	628	631	640	645	646	648	646	642
Germany	606	538	524	506	490	497	501	493	497	497	491	500
Iceland	2	2	2	2	2	2	2	2	2	2	2	2
Ireland	92	95	96	96	98	99	100	101	105	105	100	100
Luxembourg	6	6	6	6	6	6	6	6	6	6	6	6
Netherlands	191	188	48	157	137	159	120	155	140	137	125	122
Norway	19	19	21	20	21	21	22	21	21	21	21	21
Portugal	87	84	88	82	77	82	80	79	81	85	84	84
Spain	269	260	259	244	260	250	278	278	293	303	303	313
Switzerland	59	58	58	58	58	57	57	57	56	56	56	56
Sweden	44	45	45	51	51	51	51	49	49	47	47	44
UK	281	282	270	270	271	263	265	268	264	260	245	239
Italy	384	371	362	370	378	380	354	365	361	369	360	360
Poland	418	371	368	315	316	313	300	288	306	281	265	254
Russia	981	956	893	744	636	679	617	601	556	541	535	535

Table 3. Annual modelled oxidized nitrogen depositions to the main regions of the OSPAR maritime area. Unit: t N a^{-1}.

Year	OSPAR Region				
	1. Arctic Waters	**2. Greater North Sea**	**3. Celtic Sea**	**4. Bay of Biscay**	**5. Wider Atlantic**
1990	292564	336108	75928	127986	429163
1995	253791	290341	81957	117441	423612
1996	335480	341975	104599	136082	493623
1997	266470	303393	87920	112022	455901
1998	199275	303801	68760	116728	364924
1999	223288	265423	60049	112508	362455
2000	230034	307430	61123	105985	341703
2001	219990	286368	70778	112894	406477

Table 4. Annual modelled reduced nitrogen (NH_3-N + NH_4^+-N) depositions to the main regions of the OSPAR maritime area. Unit: t N a^{-1}.

Year	OSPAR Region				
	1. Arctic Waters	**2. Greater North Sea**	**3. Celtic Sea**	**4. Bay of Biscay**	**5. Wider Atlantic**
1990	116220	198289	52213	66181	106737
1995	110635	192534	64090	65434	103163
1996	169805	219973	75331	76454	154243
1997	111158	209527	67192	66514	134577
1998	83814	219440	60242	61396	83789
1999	100138	204010	54654	64570	84169
2000	89388	242805	55282	56012	74937
2001	81483	199678	55135	59194	90193

Table 5. Annual modelled total (NO_x-N + NH_4-N) nitrogen deposition to the main regions of the OSPAR maritime area. Unit: t N a^{-1}.

Year	OSPAR Region				
	1. Arctic Waters	**2. Greater North Sea**	**3. Celtic Sea**	**4. Bay of Biscay**	**5. Wider Atlantic**
1990	408784	534397	128140	194168	535900
1995	364426	482875	146047	182875	526774
1996	505285	561948	179931	212536	647866
1997	377628	512920	155112	178536	590479
1998	283089	523241	129002	178124	448714
1999	323426	469433	114703	177078	446624
2000	319422	550235	116405	161997	416640
2001	301473	486047	125913	172089	496670

Table 6. Annual modelled oxidized nitrogen deposition to the sub-regions of the Region 2 (Greater North Sea) of the OSPAR maritime area. Unit: t N a^{-1}.

Year	Sub-regions of the OSPAR Region 2												
	1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**
1990	24901	10343	19808	12557	18776	27653	10541	21062	42967	46008	38155	35139	28103
1995	23081	11147	16331	11546	16966	25217	8205	17171	35704	35884	30119	27282	31827
1996	28592	15961	21010	16700	20136	25906	9386	17038	38670	44629	38713	28203	37062
1997	24853	13433	18881	13789	17687	22572	8421	15643	34976	43312	35177	24249	30409
1998	21687	9850	16971	10866	19104	28749	8567	18868	36723	40488	35594	27391	28880
1999	18682	7434	13940	9349	16972	24512	7472	16590	32888	33640	30845	27460	25578
2000	22751	8808	17114	10939	18798	26478	9844	17827	34966	42616	37086	32015	28125
2001	20928	10960	16399	12798	18463	27778	7353	19076	30575	35421	32109	26383	28116
%	16.0	-6.0	17.2	-1.9	1.7	-0.5	30.2	9.4	28.8	23.0	15.8	24.9	0.0

Table 7. Annual modelled reduced nitrogen deposition to the sub-regions of the Region 2 (Greater North Sea) of the OSPAR maritime area. Unit: t N a^{-1}.

Year	Sub-regions of the OSPAR Region 2												
	1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**
1990	9993	4193	8998	8993	12582	20759	6047	18198	20054	19912	16742	26997	24743
1995	10269	5265	8285	8955	12639	20665	5357	16492	18531	17845	15672	21577	31168
1996	14556	8326	10963	11727	13374	19948	6391	16052	20436	22543	20323	21583	33844
1997	10964	6318	9423	10088	13596	21138	5984	16877	18805	22757	20025	19627	34077
1998	11013	5275	9242	9277	15645	24549	6188	19457	20513	21589	19398	23593	33660
1999	10005	3763	8846	8876	15026	22233	5089	16935	20132	19811	18310	23313	31668
2000	11650	4014	9983	9521	17048	25733	7570	20744	20626	26031	24255	28559	37057
2001	10103	5458	8547	9625	14042	22200	5340	19273	15684	18327	18396	21574	31150
	-1.1	-30.2	5.0	-7.0	-11.6	-6.9	11.7	-5.9	21.8	8.0	-9.9	20.1	-25.9

Table 8. Annual modelled total nitrogen deposition to the sub-regions of the Region 2 (Greater North Sea) of the OSPAR maritime area. Unit: t N a^{-1}.

Year	Sub-regions of the OSPAR Region 2												
	1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**
1990	34894	14535	28805	21550	31358	48412	16588	39260	63021	65919	54898	62137	52845
1995	33350	16412	24615	20501	29605	45882	13563	33662	54234	53728	45790	48859	62995
1996	43148	24287	31972	28427	33510	45854	15776	33090	59105	67172	59035	49786	70906
1997	35818	19750	28305	23877	31284	43710	14405	32520	53782	66069	55201	43876	64486
1998	32701	15125	26213	20143	34749	53298	14755	38325	57235	62078	54992	50984	62540
1999	28687	11197	22786	18225	31997	46745	12560	33525	53020	53451	49155	50773	57247
2000	34401	12822	27097	20460	35846	52211	17414	38572	55593	68647	61341	60575	65182
2001	31031	16418	24946	22423	32505	49978	12693	38349	46259	53748	50505	47957	59267
	11.1	-13.	13.4	-4.1	-3.7	-3.2	23.5	2.3	26.6	18.5	8.0	22.8	-12.