

# ***WORKING PAPER***

## **NITROGEN DEPOSITION CALCULATIONS FOR EUROPE**

*Joseph Alcamo  
Jerzy Bartnicki*

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WP-88-025

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## Preface

Thus far, the emphasis in the development of the Regional Acidification Information and Simulation (RAINS) model has been on sulfur. However, the long range atmospheric transport and deposition of nitrogen and associated pollutants such as oxidants may also have important effects upon forests and soils. In the Acid Rain Project at IIASA, work has already been carried out in estimating emissions of nitrogen oxides in Europe. This Working Paper presents another important step in that nitrogen from  $\text{NO}_x$  and  $\text{NH}_3$  emissions is incorporated into the atmospheric transport and deposition submodel of RAINS and is applied to an assessment of the effectiveness of several proposed strategies for reducing  $\text{NO}_x$  emissions.

R.W. Shaw  
Leader, Acid Rain Project

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## Abstract

Nitrogen deposition calculations for Europe were performed by separate models describing the long range transport of ammonia and oxidized nitrogen. Tests of model results against observations are briefly described.

Nitrogen deposition was computed for several  $\text{NO}_x$  emissions reduction scenarios. These reductions were adapted from an OECD study and applied to the 27 largest countries in Europe. They do not refer to a particular target year. A "Maximum feasible  $\text{NO}_x$  emissions reduction" scenario reduced overall European emissions by 62% relative to 1980 emissions. Other scenarios, such as point source reduction or traffic limits, reduced European emissions by 9 to 35%. Most reduction scenarios affected the pattern of  $\text{NO}_x$ -nitrogen deposition, but did not change very much the overall pattern of total ( $\text{NO}_x$  plus  $\text{NH}_3$ ) nitrogen deposition. Depending on the desired level of environmental protection, it may be necessary to reduce ammonia emissions in addition to  $\text{NO}_x$  emissions in order to reduce nitrogen deposition in Europe.

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# Nitrogen Deposition Calculations for Europe

*Joseph Alcamo and Jerzy Bartnicki*

## Introduction

Results from studies of lakes, soil, and forests point to the increasing importance of atmospheric nitrogen as a source of environmental impacts in Europe. Lakes in remote areas may be over-fertilized or acidified by nitrogen in runoff that originates as deposition from the atmosphere. Perhaps of greater consequence are the effects of nitrogen deposition on forests. Though nitrogen is an important nutrient for trees, researchers now believe that some forests may be receiving an excess of required nitrogen (see, e.g. Nilsson (1986)). This so-called "nitrogen saturation" problem can physiologically affect trees because they cannot process the excess nitrogen, or the nitrogen may pass into soil water and contribute to soil acidification which may interfere with the ability of trees to assimilate nutrients through their roots.

$\text{NO}_x$  also has an important role in production of ozone in the atmosphere; ozone in turn has been linked to forest damage (see e.g., McLaughlin, 1985). In this paper, however, we will focus only on nitrogen deposition.

Nitrogen deposition originates from both oxidized nitrogen ( $\text{NO}_x$ ) and ammonia/ammonium-nitrogen ( $\text{NH}_x$ ) in the atmosphere. Ammonium actually helps neutralize acidity due to sulfate and nitrate in precipitation. However, ammonium can also be oxidized to nitric acid in soil water and thereby contribute to soil acidification. Also, ammonia as a form of nitrogen contributes to the overall problem of nitrogen saturation.

We now turn to the question of where this nitrogen originates from. The residence time of oxidized nitrogen in Europe's atmosphere is about 1 to 4 days, which implies a transport distance of from several hundred kilometers to about 2000 km (e.g. ECE, 1987). It is clear, then, that nitrogen deposition to forests and lakes is at least partly due to  $\text{NO}_x$  and perhaps  $\text{NH}_3$ , transported over long distances. This paper presents calculations of this long range transport and deposition. These calculations are performed by a submodel of RAINS (Regional Acidification INFORMATION and Simulation) which is an integrated model used to evaluate strategies for controlling transboundary air pollution in Europe (see, e.g. Alcamo, et al, 1987). We also present some preliminary results from these calculations.

In the first part of the paper we provide a very brief overview of key atmospheric processes of nitrogen; we next describe the principles behind the models we use for nitrogen calculations and how they were tested. Then we review how these models were adapted for routine use in RAINS and some preliminary results of using them to evaluate  $\text{NO}_x$  emission control scenarios in Europe.

### Atmospheric Processes

It is well known from monitoring data that both ammonia and oxidized nitrogen contribute to wet nitrogen deposition throughout Europe. (Measurements of dry deposition are not extensive enough to draw such firm conclusions.) The data in Table 1, for example, indicate that nitrate contributes about, one quarter to one-half of wet nitrogen deposition, and ammonia the remainder.<sup>1</sup> Therefore we are concerned with the processes of both in the atmosphere.

Table 1. Selected measurements of nitrate and ammonium in precipitation. \* Both,  $NO_3$  and  $NH_3$ , are expressed in  $mg$  nitrogen  $l^{-1}$ .

Station	April-September 1980			April-September 1982		
	$NO_3$	$NH_3$	$\frac{NO_3}{NO_3 + NH_4}$	$NO_3$	$NH_3$	$\frac{NO_3}{NO_3 + NH_4}$
DK3 Tange	0.44	0.69	0.39	0.48	0.79	0.38
F1 Vert-le-Petit	1.68	4.77	0.26	1.05	1.75	0.38
H1 K-puszta	0.62	1.40	0.31	0.56	1.12	0.33
N1 Birkenes	0.54	0.69	0.44	0.56	0.77	0.42
NL5 Rekken	0.72	1.82	0.28	0.90	1.80	0.33
PL 1 Suwalki	0.4	0.99	0.31	0.70	1.40	0.33
S 3 Velen	0.36	0.44	0.45	0.31	0.42	0.42
SF 4 Antäri	0.21	0.33	0.39	0.34	0.46	0.27

\* Precipitation-weighted concentrations compiled by Norwegian Institute for Air Research (1984), Summary Report from the Chemical Coordinating Center for the second phase of EMEP, EMEP/CCC Report 2/84, Postboks 130, N-2001 Lillestrøm, Norway.

**Oxidized Nitrogen.** The chemistry of  $NO_x$  in the atmosphere involves the formation and destruction of many different forms of oxidized nitrogen, many of which are interconverted in a few minutes or hours. The  $NO_x$  in Western Europe's atmosphere originates primarily from vehicle emissions (53%) and secondarily from power plants (24%) and industry (12%) (Lübker, 1987). Since the solubility of nitric oxide ( $NO$ ) and nitrogen dioxide ( $NO_2$ ) is low, they are not effectively removed by precipitation near emission sources. Knowledge about the dry deposition of  $NO$  and  $NO_2$  is limited but it is thought that this is an inefficient removal pathway because of the low reactivity of these gases. Hence, these species are commonly converted to nitric acid ( $HNO_3$ ) vapor or nitrate ( $NO_3^-$ ) aerosol before being removed from the atmosphere. Nitric acid can be removed by precipitation after dissolving in cloud droplets or raindrops, or may be adsorbed onto surfaces of the earth. Nitrate aerosols serve as condensation nuclei in clouds and can then be removed by precipitation. Sometimes nitrate-containing droplets evaporate and re-release nitrate aerosol to the air stream which then transports the nitrate still further from its  $NO_x$  source.

<sup>1</sup>It is usually assumed that wet nitrogen deposition consists entirely of either  $NO_x$  or  $NH_3$ .

**Ammonia Nitrogen.** Some ammonia in the European atmosphere comes from emissions of fertilizer and certain industrial processes. However, its principal source is the decomposition of livestock wastes (Buijsman, et al. 1987). Since these emission sources cover wide areas, the locations of ammonia emissions largely coincide with areas of deposition. Because the rate of dry deposition flux of a gas depends on its concentration difference near the earth's surface (with the stronger the gradient the stronger the flux) the emission of  $\text{NH}_3$  tends to inhibit the rate of its own deposition in the vicinity of the source region. This is an important cause of long range transport of ammonia in Europe. Nevertheless, some ammonia gas is dry deposited in the vicinity of its source. The fraction not dry deposited will travel some distance before converting to ammonia aerosol. This aerosol in turn may be dry deposited, though this is not an important removal pathway because of the low deposition velocities of aerosols in general. Greater removals are accomplished by the capture of ammonium aerosol in precipitation. Ammonia gas will also be efficiently removed from the atmosphere by dissolving in cloud droplets or dry depositing in low emission regions.

**Interaction of oxidized and ammonia nitrogen.** Ammonia and oxidized nitrogen interact in an important way in the atmosphere. When  $\text{NH}_3$  is not present, and depending on climatic conditions, the  $\text{NO}_2$  can be converted to  $\text{HNO}_3$  vapor and rather quickly removed from the atmosphere by dry deposition. However, if  $\text{NH}_3$  is present in sufficient quantities,  $\text{HNO}_3$  can combine with  $\text{NH}_3$  to form ammonium nitrate aerosol which has a relatively low dry deposition rate. The question arises, whether this phenomenon affects the large-scale patterns of nitrogen deposition in Europe. In model experiments, Derwent and Nodop (1986) showed that the presence of ammonia enhanced the wet deposition of  $\text{NO}_x$  in northwest Europe, but not the total of wet plus dry (see Figure 4). However, further investigation is needed with models containing both  $\text{NO}_x$  and  $\text{NH}_x$  species in order to understand how closely these species are coupled in the atmosphere.

### The Models: Similarities

Nitrogen calculations in this paper are based on separate models for ammonia and oxidized nitrogen; this approach simplifies and speeds calculations of total nitrogen deposition. However, by using separate models we imply that ammonia is not coupled with oxidized nitrogen. As noted above, we cannot yet say whether this is a good assumption or not.

Calculations for oxidized nitrogen are based on a model developed by R. Derwent at the Harwell Laboratory (Derwent, 1986 and Derwent and Nodop, 1986). (This model will be referred to as the "Harwell model".) Ammonia calculations use a model of Asmann and Janssen (1987) developed at the University of Utrecht in the Netherlands and the Netherlands Energy Research Foundation, Petten, Netherlands. (We will refer to this model as the "IMOUECN model".) Each model describes how emissions from a European grid are redistributed by winds and precipitation to other parts of Europe. The main inputs to the models are meteorological data such as wind velocity and precipitation, and European emissions on the EMEP grid with a  $150 \times 150$  km resolution (Figure 1). Output includes wet, dry and total deposition and air concentration. Considering the grid's coarse spatial resolution, these models are more suitable for long range computations and less so for local ( $< 150$  km) calculations.

Both the  $\text{NO}_x$  and  $\text{NH}_x$  models are of the Lagrangian type in that they have moving frames of reference. The basic idea of this type of model is to track the behavior of a parcel of air as it moves under the influence of winds over emission areas of Europe (see Figure 2). The paths of these trajectories are computed backwards for 96 hours from the centers of grid elements by using wind velocity data. The conceptual size of the air parcel is that of the spatial grid of the model ( $150 \times 150$  km). The height of the parcel is the "mixing height"; pollutants are assumed to be well mixed up to this height.



As an air parcel travels over a grid element, the concentration of nitrogen may decrease by dry or wet deposition, or increase by added emissions which are assumed to be instantaneously mixed throughout the air parcel.

### Principles and Testing of the $\text{NO}_x$ model.

*The Harwell Model.* To calculate the atmospheric transport of  $\text{NO}_x$ , the Harwell model assumes straight trajectories with a constant transport wind of  $8.0 \text{ m s}^{-1}$  and mixing height of 1 km.

The physical-chemical processes represented in the model are shown in Figure 3. The main inputs are NO emissions in the EMEP grid and wind roses at several locations in Europe. The chemical scheme and parameter values of the model imply that NO is rather quickly oxidized to  $\text{NO}_2$  in the atmosphere. The  $\text{NO}_2$  is in turn converted to nitric acid and nitrate aerosol. Both  $\text{NO}_2$  and  $\text{HNO}_3$  are removed by dry deposition, while  $\text{HNO}_3$  and nitrate aerosol are removed by wet deposition. Removal rates by precipitation are statistically-averaged so that each parcel is in effect subject to a constant "slow drizzle". Annual precipitation is set constant at  $1000 \text{ mm yr}^{-1}$ . As we see later in this paper, these simple meteorological assumptions produce smooth deposition patterns.

Details of the model can be found in Derwent (1986).

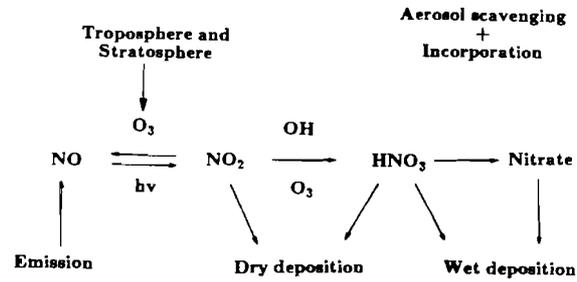
*Testing of the Harwell Model.* The computed pattern of  $\text{NO}_3^-$  in rain is compared with the observed pattern in Figure 4. Derwent notes that a modified version of his model, which includes  $\text{NH}_3$  and night-time chemistry, gives better results for wet deposition calculations. However he also notes that computed total (wet + dry) deposition is unchanged between these model versions. For all model versions the coefficient of determination ( $r^2$ ) between computed and observed  $\text{NO}_3^-$  in rain was greater than 0.75 (Derwent and Nodop, 1986).

Derwent (1987b) also estimated the model's parameter uncertainty. Frequency distributions were assigned to model parameters, and these uncertainties were propagated through the model equations by use of the so-called "latin hypercube" sampling technique (McKay, et al, 1976). His conclusion was that wet and dry deposition can only be estimated individually within an order of magnitude, whereas total (wet plus dry) deposition errors compensate so that one standard deviation of calculations was about 40 to 50%. For more details about the model's testing the reader is referred to Derwent (1987b).

*IIASA Simplified Version of the  $\text{NO}_x$  Model.* The  $\text{NO}_x$ -nitrogen model described above has only been applied to the north-western part of Europe. In order to speed calculations for all Europe, it was desirable to further simplify the model. This simplification, however, retained its basic behavior. The procedure for doing so was as follows:

1. Based on simulation results from the full non-linear Harwell model Derwent (1987a) established the typical profile of total (wet plus dry)  $\text{NO}_x$ -nitrogen deposition versus distance from a source. Figure 5 shows the initial increase of nitrogen deposition near the  $\text{NO}_x$  source owing to deposition of secondary emission products. This is followed by a steady decrease with distance as removal processes deplete nitrogen from the air parcel. Twenty different deposition profiles are shown in this figure, each based on a different set of parameter values from a probability distribution of each parameter. The range of these calculations is an estimate, therefore, of the range of model uncertainty due to uncertain parameters. Note that the uncertainty, after reaching a maximum at about 75 hours travel time, decreases with distance from the source because of the compensation of different parameter errors. This adds validity to the usage of a simple approximation for long range transport of  $\text{NO}_x$ . (Though Figure 5 still indicates a factor of two uncertainty even at a large distance from the emission source).

(a) Oxidized nitrogen model



(b) Ammonia model

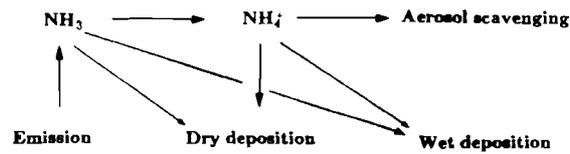


Figure 3. Model processes and constituents. (a) Harwell  $NO_x$  model, diagram from Derwent (1986), (b) IMOUECN  $NH_3$  model, based on information in Asman and Janssen (1987).

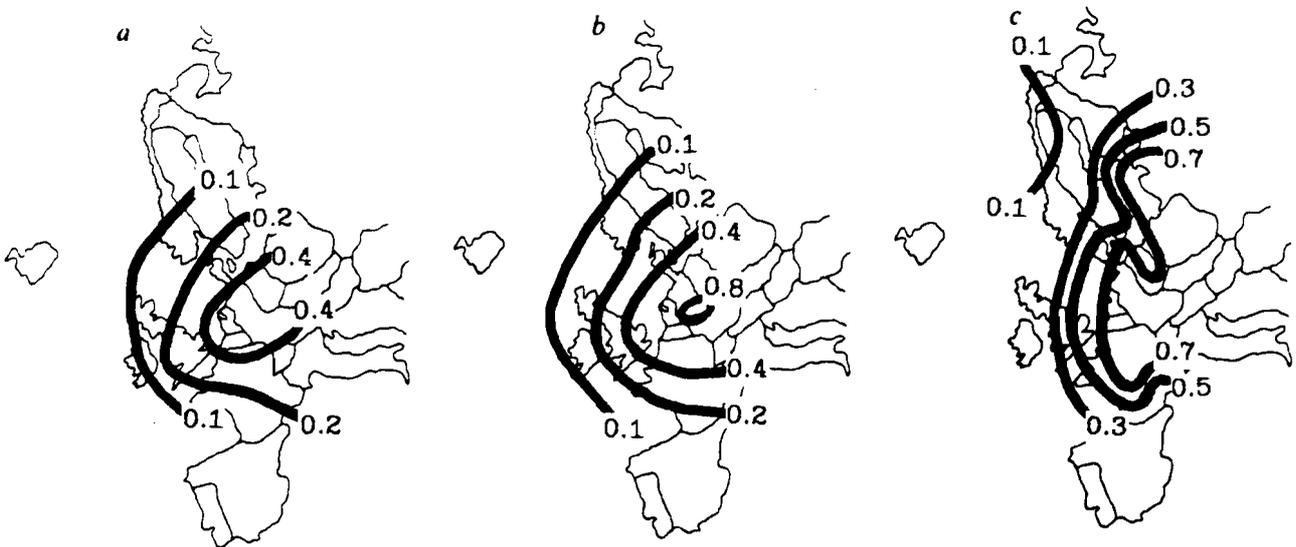


Figure 4. Nitrate concentration in rain. Units:  $mg\ l^{-1}nitrogen$ . (a) Harwell model without ammonia, (b) Harwell model with ammonia, (c) Observed concentration. All figures from Derwent and Nodop (1986).

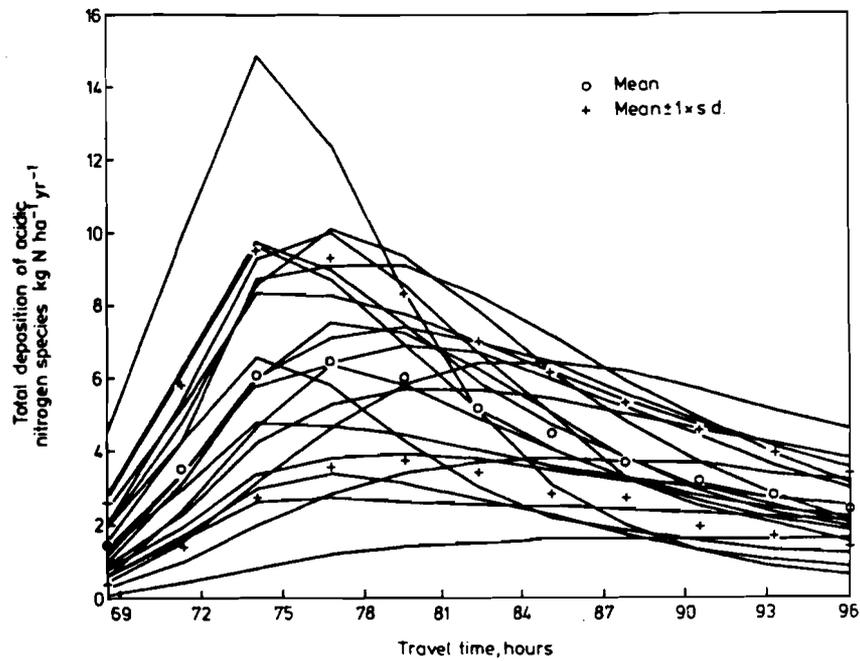


Figure 5. Total NO<sub>x</sub>-nitrogen deposition profile downwind of a single source. Different profiles are due to different sets of parameters selected during an uncertainty analysis. From Harwell model (Derwent, 1987a).

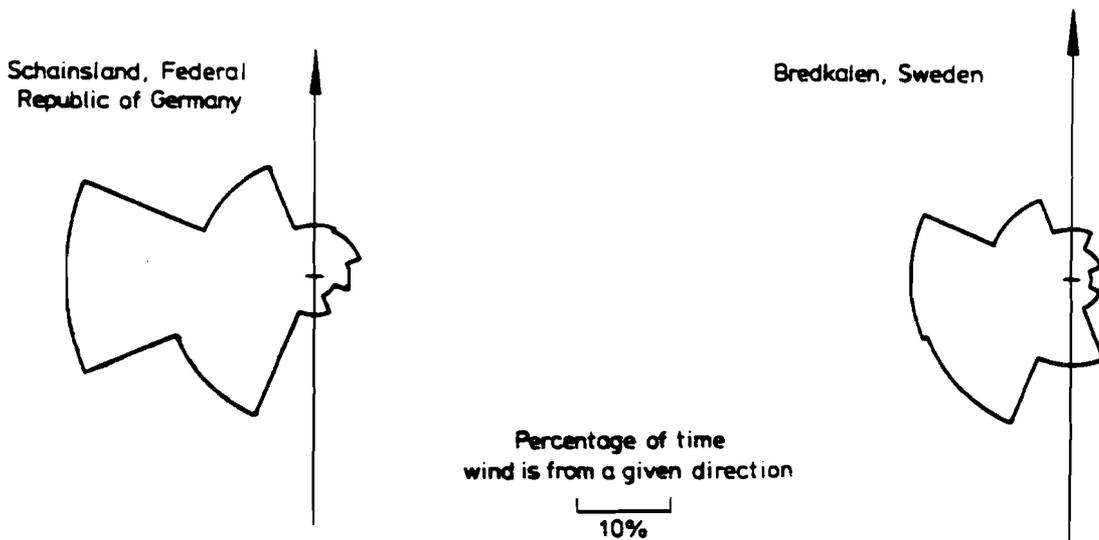


Figure 6. Two examples of wind roses used in NO<sub>x</sub> deposition calculations.

An equation that fits this profile (Derwent, 1987a) serves as the basis of deposition calculations:

$$d_n = E \frac{f}{x} (e^{-0.0017x} - e^{-0.0057x}) \quad (1)$$

where  $d_n$  is the total nitrogen deposition ( $\text{gm}^{-2} \text{yr}^{-1}$ ),  $E$  is the grid NO emissions ( $\text{kt yr}^{-1}$ ),  $f$  is an integrating factor obtained by assuming that emissions originate from the center of grid elements.

Incorporated in this model are all the assumptions of the full non-linear model (straight trajectories, constant wind speed, and so on).

2. We use wind roses based on 1981 and 1982 sector analysis to allocate contributions of different source areas to a particular receptor in Europe. Typical wind roses are shown in Figure 6. Wind roses were also used in calculations of the Harwell model, although fewer are used in the IIASA simplified version and, they are, consequently applied over greater areas. Unlike the Harwell model, we apply these wind roses to the emission grid elements rather than the receptor grid elements. This, we believe, yields a more mass-consistent calculation for the simplified model.<sup>2</sup>

3. Using the information in (1) to (3) above, we calculate the annual contribution of every grid element to deposition in every other grid element. This information is stored in the form of a country-to-grid transfer matrix.

To do so we use  $\text{NO}_x$  country emissions from Semb and Amble (1981) with the same spatial distribution as for  $\text{SO}_2$  in Dovland and Saltbones (1979). These emission estimates cannot be related to a particular year, but rather refer to emissions in the "mid- to late 70s". More recent information is available for emissions but we used these emissions to test the model so that our calculations are consistent with the Harwell model calculations (Derwent, 1986). When we analyze emission control scenarios later in this paper, however, we use more recent estimates of 1980  $\text{NO}_x$  emissions as a reference case.

*Testing of the Simplified  $\text{NO}_x$  Model.* As a first step in examining model results, we compare computations of the simplified model with those of the full non-linear Harwell model (Figure 7).

Though the simplified model covers all Europe, only the part that coincides with the north-west European study area of the Harwell model is shown in this figure.  $\text{NO}_x$ -N deposition patterns are similar except for the eastern part of the Nordic countries where the simplified model computes higher deposition. This discrepancy may not be too important since it occurs largely outside of the Harwell model's study area. Derwent (1987a) also found good agreement between deposition calculated by the full and simplified models downwind of a single source (Figure 8).

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<sup>2</sup>The reason is that in the Harwell model calculations are performed at 51 receptors, each having its own wind rose. Since calculations at each point are based on actual meteorological measurements, there is small chance that they will "double count" emission areas. However the simplified model covers all of Europe (approximately 700 receptors) with 8 wind roses which would require that we apply a particular wind rose to several receptor points. Under this situation we may incorrectly estimate that several receptors receive a large fraction of their deposition from the same source area. This may lead to incorrect mass calculations. For example it is possible that if we add up the contributions of a particular source area to all receptors, the sum might exceed the emission rate of the source area by a factor of two. This problem is avoided in the Harwell model because deposition is only calculated at receptors where wind roses are available. To circumvent this problem in the simplified model, we assign the wind roses to the emissions grid elements rather than the receptor grid elements. By doing so we guarantee that we do not assign more mass to receptors than is being emitted by the grid elements.

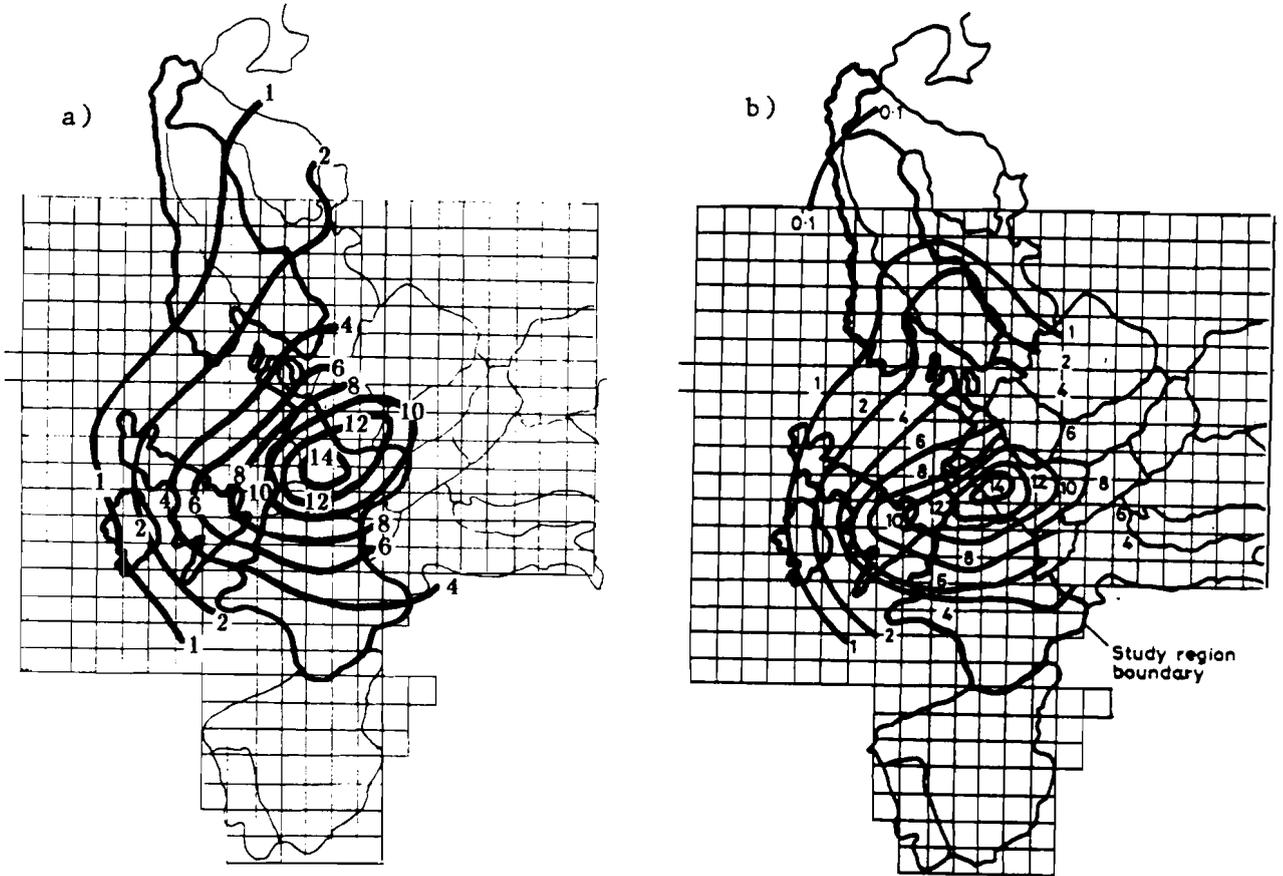


Figure 7. Computed NO<sub>x</sub>-nitrogen deposition using emissions from "late 70s" (see Table 3). Units: gm<sup>-2</sup>yr<sup>-1</sup> nitrogen. (a) Simplified model, (b) Harwell model.

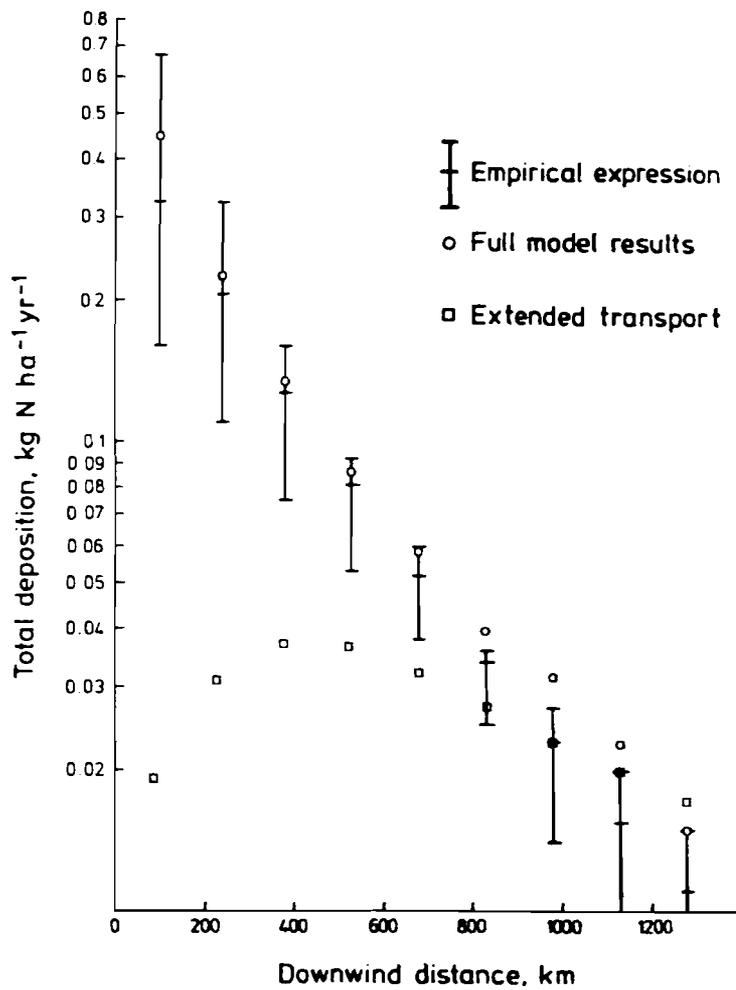


Figure 8. Comparison of simplified NO<sub>x</sub>-nitrogen model results with full non-linear model for a single trajectory and source. From Derwent (1987a).

In Figure 9 we compare computations of 1980  $\text{NO}_x$ -nitrogen deposition from the simplified model with observations of wet  $\text{NO}_3^-$  deposition. For these calculations we use 1980  $\text{NO}_x$  emission estimates from the RAINS model, as will be explained later in this paper.<sup>3</sup> It is important to note that we compare total nitrogen calculations with wet-deposited nitrogen observations (rather than total wet plus dry) because: (1) the simplified model cannot calculate wet deposition owing to its simplifications, and (2) total nitrogen observations are unavailable because of the difficulty in routinely measuring dry deposition.

Since we are comparing total with wet deposition, we expect model calculations to exceed observations in Figure 9, as they indeed do. The overestimate is smaller in the Nordic countries, where dry deposition probably contributes a smaller amount to total nitrogen deposition, than in Central and Western Europe where higher airborne levels of oxidized nitrogen promote dry deposition. As can be seen in Figure 9, the computed total nitrogen does not exceed the measured wet deposition in Central Europe as much as it does in the North. In any event, the spatial pattern of calculated deposition is consistent with the observed pattern though the computed peak is north of the observed peak.

As noted above, for these calculations we applied eight wind roses to all Europe. Because of this simplification, it is important to examine the sensitivity of calculations to the selection of wind roses. Figures 10(a) and (b) were obtained by applying a single wind rose to all grid elements. When we compare these results with each other and with our base calculations which used eight wind roses (Figure 7(a)), we observe that calculations are not too sensitive to these wind rose assumptions. The spatial patterns are similar, though peak locations shift. This agreement may be fortuitous or could indicate that for long time and space scales total N is not very sensitive to the spatial variability of long-term meteorological patterns or, in fact, that wind roses are not very different for different parts of Europe.

As noted above, Derwent (1987b) estimates that one standard deviation of computed total  $\text{NO}_x$ -N deposition in the fully non-linear Harwell model is roughly  $\pm 40\text{-}50\%$  (due to parameter uncertainty). We may speculate that this is a lower limit of accuracy for the simplified model.

It should also be noted that the smooth spatial pattern of oxidized nitrogen deposition (e.g. Figure 9) is an artifact of the model. This is because both wet and dry deposition rates are constant along the trajectories and because the assumption of straight line trajectories smooths out irregularities of the wind. We will see in the next section that the  $\text{NH}_3$  model, which contains more refined meteorology, produces more irregular spatial gradients of deposition.

### Principles and Testing of the Ammonia Model.

*The IMOUECN Model.* The model used to compute  $\text{NH}_x$  deposition is an EMEP-type Lagrangian model developed by Asman and Janssen (1987). Processes contained in the model are summarized in Figure 3(b). The model assumes simple conversion of ammonia to ammonium. This is similar in treatment to the sulfur dioxide - sulfate conversion in the EMEP model. In comparison to the Harwell model, curved irregular air trajectories are computed from actual wind data. Dry deposition flux is calculated by multiplying airborne concentration of ammonia by a constant deposition velocity. Rather than using a constant wet deposition rate as in the Harwell model, the ammonia model accounts for wet deposition only when data indicate the occurrence of precipitation. Calculations are based on 1980 wind and precipitation data at 6 hour intervals.

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<sup>3</sup>Strictly speaking, this is not "1980  $\text{NO}_x$ -nitrogen deposition" because 1980 meteorology was not used for these calculations.

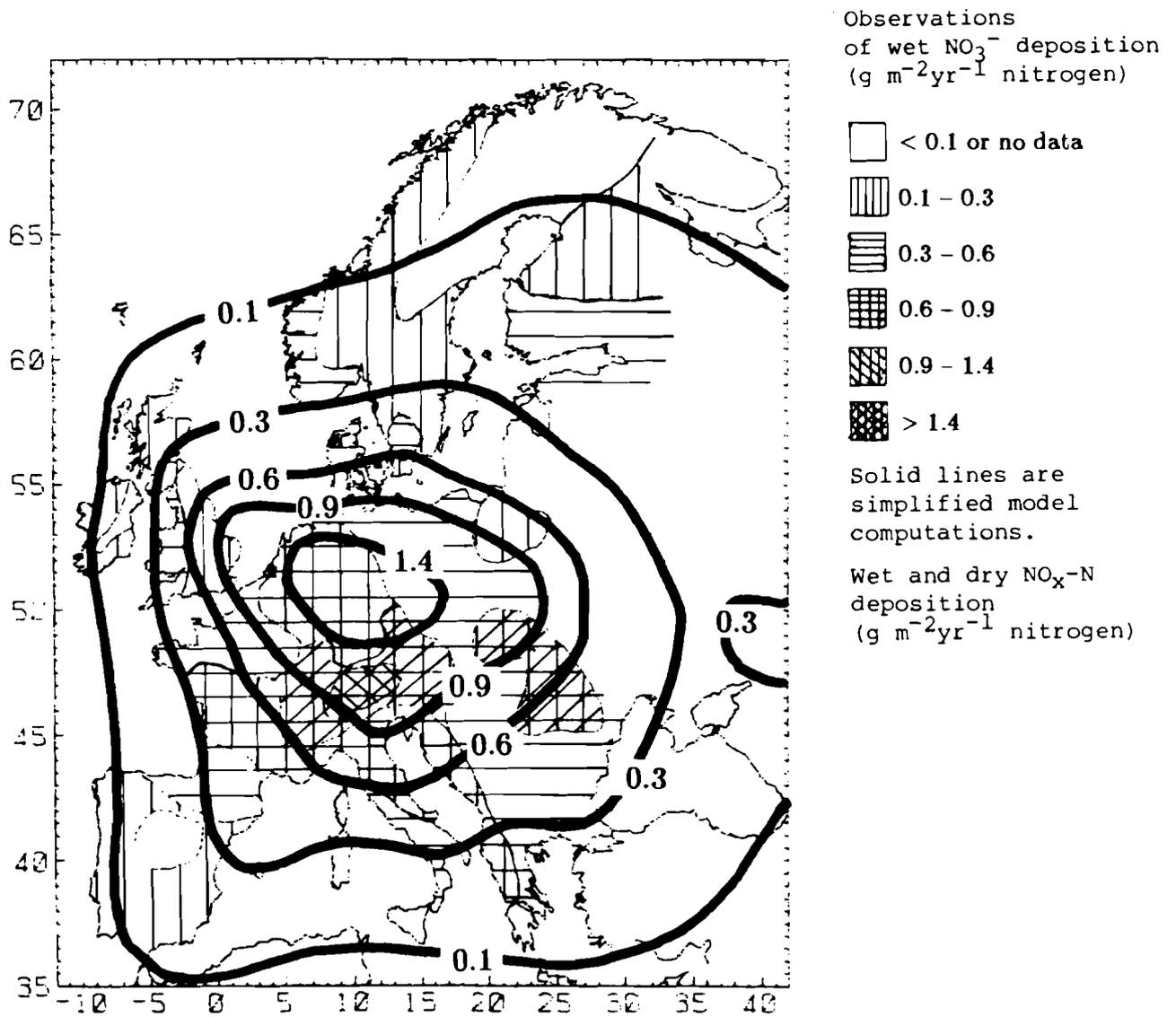


Figure 9. Comparison of computed 1980  $\text{NO}_x$ -nitrogen deposition (solid lines = simplified model) with observations (areal patterns) from 1982. Data reported by Nodop (1987).

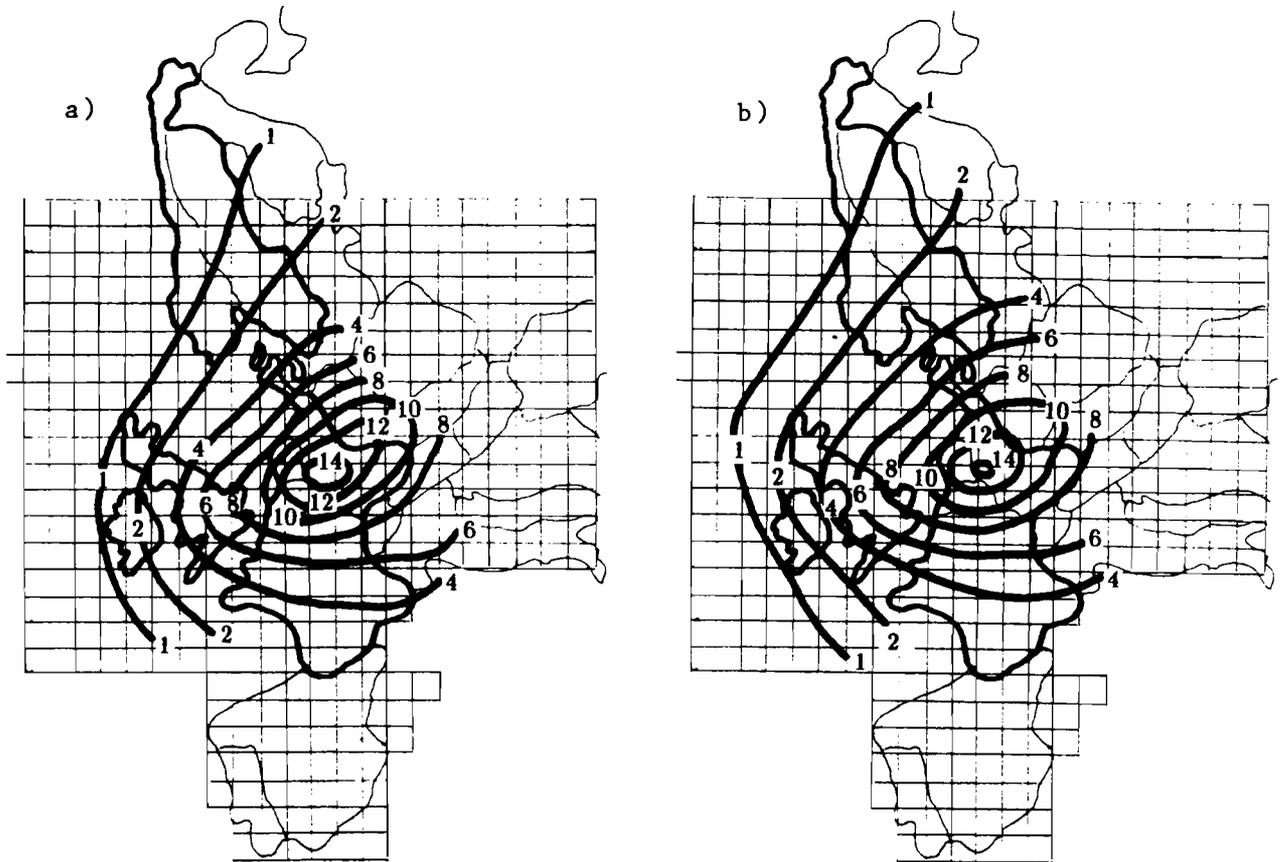


Figure 10. Sensitivity of computed NO<sub>x</sub>-nitrogen deposition ( $\text{g m}^{-2} \text{yr}^{-1}$  nitrogen) to wind rose selection. (a) Schauinsland, FRG wind rose applied to all Europe, (b) Bredkalen, Sweden wind rose applied to all Europe.

This should be kept in mind when the  $\text{NH}_x$  and  $\text{NO}_x$  models are used later in this paper to evaluate emission control strategies. Though the  $\text{NO}_x$  model invokes crude meteorological assumptions, it also reflects a long-term meteorological average. Of course we cannot say whether or not this long-term average corresponds to an actual long-term meteorological average.

Results of the  $\text{NH}_x$  model are expressed as a country-to-grid matrix as in the  $\text{NO}_x$  model. A preliminary version of this matrix was obtained by Asman and Janssen.

*Testing of The Ammonia Model.* To test the model, calculations were compared to measured  $\text{NH}_4^+$  aerosol and  $\text{NH}_4^+$  in precipitation. Measurements were not all from 1980, nor were they distributed evenly throughout Europe. The coefficient of determination ( $r^2$ ) between calculations and the limited number of aerosol readings ( $n=16$ ) was 0.50. For the more plentiful precipitation measurements ( $n=90$ ), the  $r^2$  value was 0.53. For further details about the ammonia model testing of the reader is referred to Asman and Janssen (1987).

We present the calculated 1980  $\text{NH}_x$  deposition in Figure 11. This corresponds to results presented in Asman and Janssen (1987). Note that the pattern in this figure is much more irregular than the pattern of  $\text{NO}_x$  deposition (Figure 9). As noted above, this is because the  $\text{NH}_x$  model incorporates curved wind trajectories based on actual wind data, as well as a wet deposition rate which depends on actual precipitation data of the previous 6 hours. Consequently, deposition processes are much more spatially dependent than in the  $\text{NO}_x$  model. Another reason is that dry deposition of  $\text{NH}_3$  is more important than dry deposition of  $\text{NO}$  or  $\text{NO}_2$  because of the greater reactivity of  $\text{NH}_3$ . Therefore the spatial pattern of  $\text{NH}_x$  deposition reflects the irregularity of the  $\text{NH}_3$  emission pattern.

Figure 11 also notes that  $\text{NH}_x$  deposition is much higher than  $\text{NO}_x$  deposition (Figure 9) as is confirmed by wet deposition data (Table 1).

### Use of the Models for Routine Calculations

It is impractical to routinely run the above models to investigate the effect of emission control scenarios because of the amount of input data and computations required. Instead we use *transfer matrices* which describe the deposition and concentration in each EMEP grid element due to a unit emissions from each of 27 European countries.

Use of transfer matrices implies certain key assumptions about how emissions and deposition are related:

(1) *The relationship between a country's emissions and its contribution to deposition at a distant receptor is assumed to be linear.* Our understanding of nitrogen linearity/nonlinearity is not as advanced as it is for sulfur where it is generally considered that emissions and total deposition are linear over the long time and space scales being considered in this paper (see, e.g. findings in Alcarno, et al, 1987). The equations of the  $\text{NH}_x$  model do, however, imply a linear relationship between emissions and deposition. Therefore, use of an ammonia transfer matrix does not conflict with model assumptions.

In comparison, the equations of the Harwell  $\text{NO}_x$  model are nonlinear. However Derwent (1987) found that these non-linear equations produced an effective linear relationship between emissions and deposition when the model was applied to real-world data at a single remote site. It is possible that although  $\text{NO}_x$  emissions and deposition are non-linear in nature over the short time and space scales (see e.g. Builtjes, 1987) the relationship may be linear over long distances ( $> 150$  km), long time periods (one year or longer), and for total (rather than just wet or dry) deposition.

The situation is different with the IIASA simplified version of the Harwell model in that the model equation itself (eq. 1) results in a linear relationship between  $\text{NO}_x$  emissions and total nitrogen deposition. Therefore the assumption of linearity in the transfer

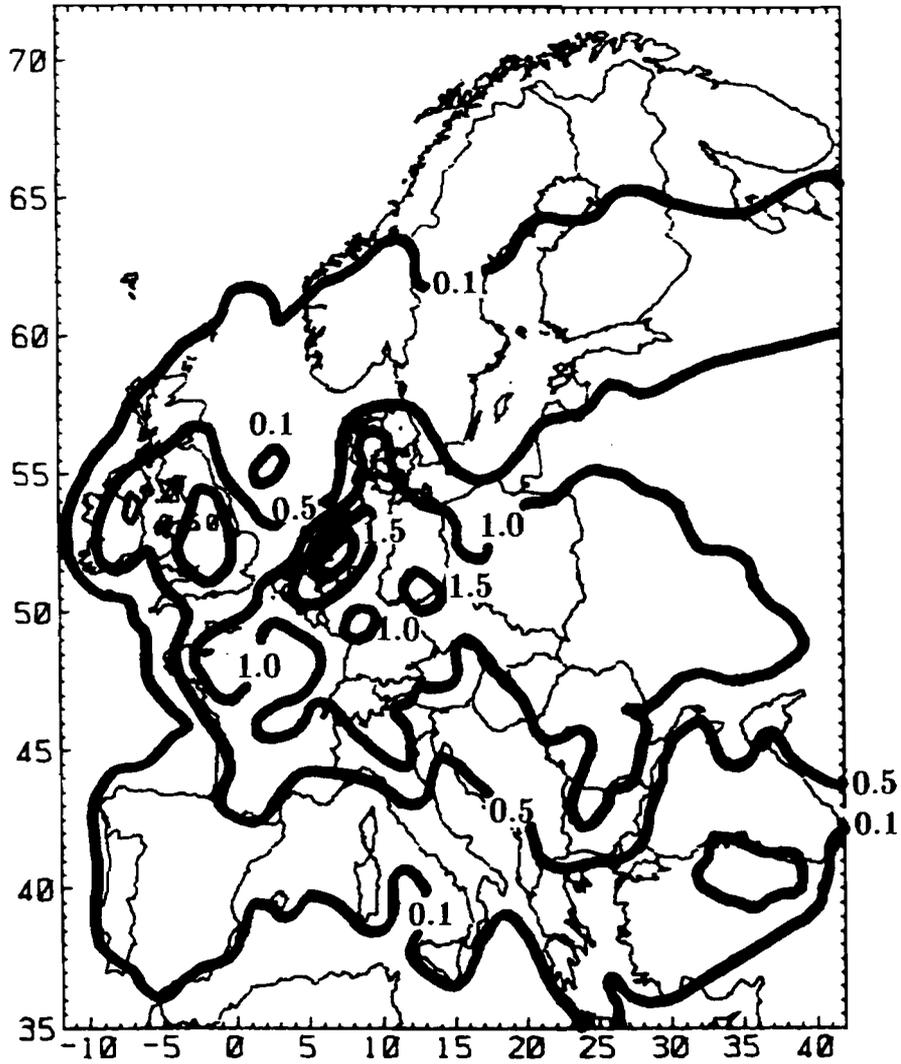


Figure 11. Computed  $\text{NH}_3$ -nitrogen deposition for 1980. Units:  $\text{g m}^{-2} \text{yr}^{-1}$  nitrogen.

matrix is at least consistent with the model formulation.

(2) *If total emissions in a country change in the future it is assumed that the spatial distribution of emissions within a country changes proportionately.* For example, if the NO<sub>x</sub> emissions in Poland are reduced by 50%, it is assumed that emissions in both northern and southern Poland are also reduced by 50%. As one can imagine, this is probably not a good assumption for large countries such as the USSR. Nevertheless, it has been shown elsewhere (Alcamo, 1987) that this assumption did not result in a large error for SO<sub>2</sub> deposition calculations in the EMEP model which had the same time and space scales as those considered in this paper. In addition, NO<sub>x</sub> emissions are more evenly distributed in a country compared to SO<sub>2</sub> emissions because much of NO<sub>x</sub> comes from vehicle emissions. These vehicles are much more widely distributed in a country than are power plants which are the principal source of SO<sub>2</sub>.

These assumptions should be kept in mind as we use the N models to evaluate emission reduction scenarios in the next section.

### Emission Reduction Scenarios

With the models described above, we can now investigate the consequences of various emission reduction scenarios. NO<sub>x</sub> emission estimates for 1980 plus five NO<sub>x</sub> emission scenarios described in OECD (1987) were revised and adapted in a consistent manner to each of the 27 largest countries in Europe. The assumed reductions in each emission sector are given in Table 2. Note that the same sector reductions were assumed for each country. However, since sector-by-sector emissions vary for each country, the net emission reduction for each country will be different. As a reference for these scenarios we use estimated 1980 emissions. Resultant country emissions for the six scenarios (reference plus five control scenarios) are presented in Table 3. NH<sub>3</sub> emissions were held constant for these calculations at their 1980 values reported in Asman and Buijsman (1987). We now briefly describe each of the NO<sub>x</sub> emission scenarios:

Table 2. NO<sub>x</sub> scenario assumptions (% reduction)

Sector	Maxnox *	EEC NO <sub>x</sub>	Lux. Agree.	U.S. Traffic	Max PS
Conversion	55	25	0	0	55
Power Plants	75	75	0	0	75
Domestic	30	6	0	0	30
Transport Gasoline	80	0	50	80	0
Diesel	50	0	0	32	0
Industry	55	25	0	0	55
Processes	0	0	0	0	0

\* For the meaning of this and other abbreviations, see text.

Table 3. Summary Table -  $NO_2$  Emissions kt/yr as  $NO_2$

Country	Scenario						
	1980 (3)	Late 70s (1)	Maxnox (3)	Lux Agree (3)	maxPS (3)	EECnox (3)	US traffic (3)
Albania	27	9	12	26	17	21	23
Austria	196	150	82	165	166	176	124
Belgium	425	290	167	390	294	323	324
Bulgaria	291	200	108	276	162	191	248
Czechoslovakia	643	500	228	568	283	356	558
Denmark	271	180	97	245	167	180	211
Finland	243	150	97	227	164	181	191
France	2014	1300	808	1805	1487	1616	1459
Fed Rep Germany	2822	2200	1126	2551	1895	2003	2029
German Dem Rep	523	570	171	504	211	256	486
Greece	217	150	85	193	171	181	148
Hungary	218	185	85	207	134	151	180
Ireland	84	60	31	71	66	71	54
Italy	1501	1000	634	1356	1167	1252	1076
Luxembourg	39	20	17	36	26	33	31
Netherlands	545	400	246	491	428	455	397
Norway	180	100	85	163	164	173	119
Poland	1490	840	462	1455	571	657	1400
Portugal	148	76	62	135	123	131	101
Romania	368	390	129	350	175	216	328
Spain	951	560	383	874	735	780	682
Sweden	331	250	158	290	276	300	232
Switzerland	161	125	62	126	137	150	92
Turkey	357	175	157	336	296	316	256
United Kingdom	2457	1900	826	2212	1481	1586	1897
USSR	2790 (emep) (4)	4533 (2)	736 (4)	2081 (4)	1870 (4)	2014 (4)	1656 (4)
Yugoslavia	338	190	112	286	218	245	240
Sum (w/USSR)	19630	16503	7166	17412	12883	14018	14543
Sum (w/out USSR)	16840	11970	6430	15331	11013	12004	12887
Reduction %		-29	-62	-9	-35	-29	-23

Notes:

- (1) These emissions are used to compare simplified  $NO_x$  model results with results from Harwell model. From Semb and Amble (1981).
- (2) This is the figure from Semb and Amble (1981)  $\times 2/3$  to account for fraction of total USSR emissions originating from Europe.
- (3) RAINS estimates except for USSR. See note (4).
- (4) The EMEP estimate from U.N. (1987, ECE/EB.AIR/14) is used instead of the RAINS estimate because of the large discrepancy between estimates. Emissions for different reduction scenarios are first calculated by RAINS (because reduction scenarios are sector specific and EMEP does not provide sector breakdown of emissions) and are then scaled down to correspond with the year 1980 USSR emissions from EMEP.

(1) *1980 NO<sub>x</sub> Emissions.* These emissions are taken from the RAINS model which computes emissions by multiplying sectoral energy use by sector-specific emission factors from Lübker (1987). Though the RAINS estimates do not correspond exactly with other country estimates, (e.g. U.N. (1987), Lübker and de Tilly (1987)) they were used in this paper because they are the most complete data set available with a sector-by-sector breakdown of emissions for each country in Europe. We require a sectoral breakdown of emissions because emission reduction strategies are sector-specific.

Despite their disagreement in a country-by-country comparison with official estimates, RAINS estimates for total European emissions in 1980 without the USSR (16.5 MT NO<sub>2</sub> yr<sup>-1</sup>) are close to EMEP estimates reported in U.N. (1987) (15.9 MT NO<sub>2</sub> yr<sup>-1</sup>).

The 1980 RAINS estimates are used as a reference for the following scenarios.

(2) *Maximum NO<sub>x</sub> Reductions ("Maxnox").* These are defined by OECD as the "maximum emission reductions technically achievable" for each source category. Since a detailed analysis of sector-by-sector emissions for each country was outside the scope of this paper, we approximate the OECD scenario as follows: 80% reduction of gasoline-transportation emissions, 50% from diesel-transportation, 75% from power plants, 55% from industry and conversion, 55% from domestic emissions, and no reduction applied to process emissions.

(3) *EEC Large Combustion Directive ("EEC Directive").* In this scenario we apply reductions called for by this EEC-specific directive to all European countries. As in the above scenario, the Directive is not applied exactly as written. We assume instead a 75% reduction of emissions from the power plant sector, 6% from the domestic sector (accounting for reductions at district heating plants), 25% from industry, and 25% from the conversion sector.

(4) *Luxembourg Agreement.* This agreement specifies a 50% reduction of NO<sub>x</sub> emissions for passenger cars and light trucks. We use a 50% reduction for the entire gasoline-transportation sector.

(5) *U.S. Traffic Limits.* These limits call for 35% reduction of emissions from diesel trucks. As an approximation we apply a 32% reduction to the entire diesel-transportation sector. For gasoline-transportation an 80% reduction is assumed.

(6) *Maximum Point Source Reductions ("max PS").* This scenario is similar to the Maxnox scenario except that the reduction of domestic emissions is taken to be 30% rather than 55% (assuming some amount of energy conservation in district heating plants), and no reductions are assumed for the transportation sector.

## Nitrogen Deposition Results

We now use the simplified NO<sub>x</sub> model and the IMOUECN NH<sub>x</sub> model to evaluate the above emission reduction scenarios. We have already examined the 1980 NO<sub>x</sub> and NH<sub>x</sub> deposition, so we now turn our attention to the sum of these species, total nitrogen deposition. The computed 1980 deposition is shown in Figure 12(a). The irregularity of the spatial pattern follows that of NH<sub>x</sub> deposition (Figure 11). In Figure 13 we compare these calculations with wet deposition data from the Nordic countries. Again we are comparing the computed total (wet plus dry) deposition with wet deposition measurements, as we did for the NO<sub>x</sub> model.<sup>4</sup> The model seems to underestimate deposition in this area.

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<sup>4</sup>Since the data in Figure 13 may be a part of the data base used in Figure 9, the tests of the NO<sub>x</sub>-nitrogen and total nitrogen calculations in this paper may not be independent.

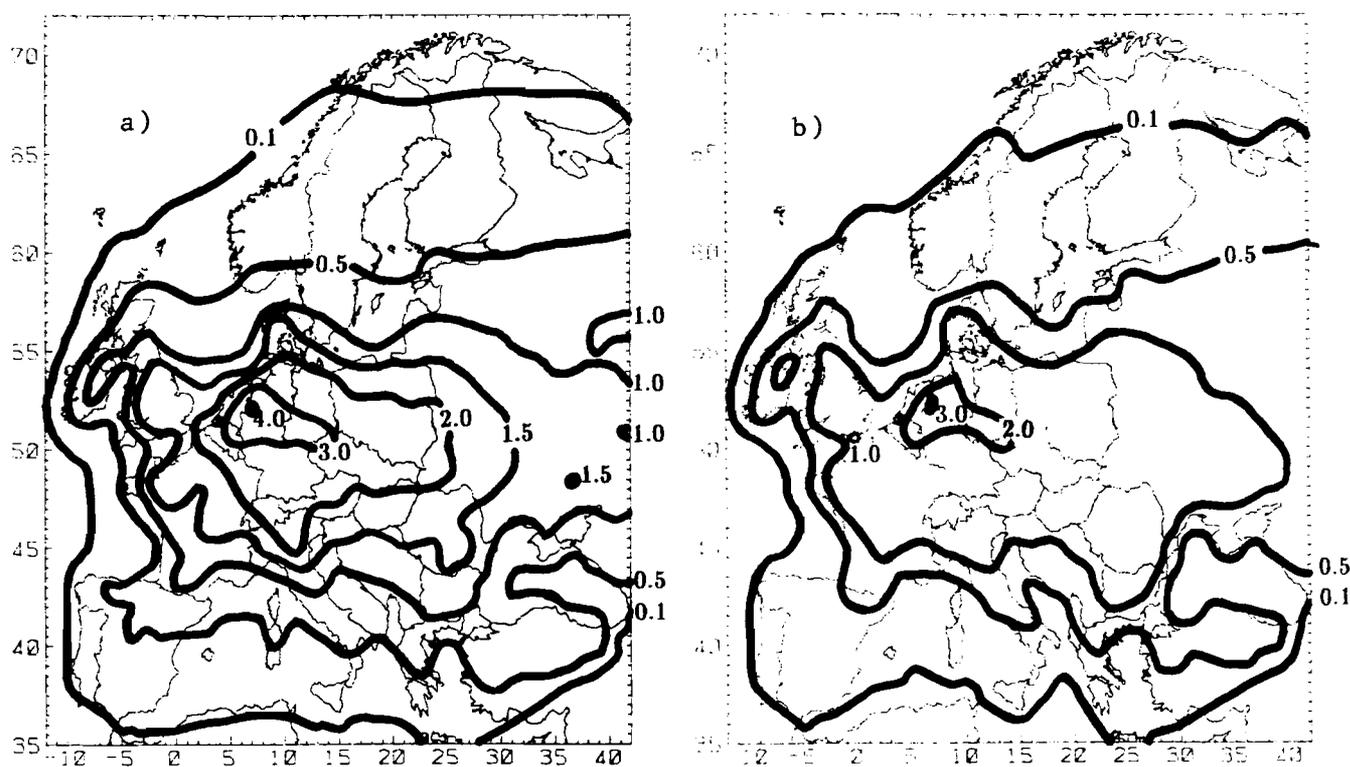


Figure 12. Computed total N ( $\text{NO}_x$ -nitrogen +  $\text{NH}_x$ -nitrogen) deposition: (a) 1980 no controls, (b) Maxnox Scenario. Units:  $\text{g m}^{-2} \text{yr}^{-1}$  nitrogen.

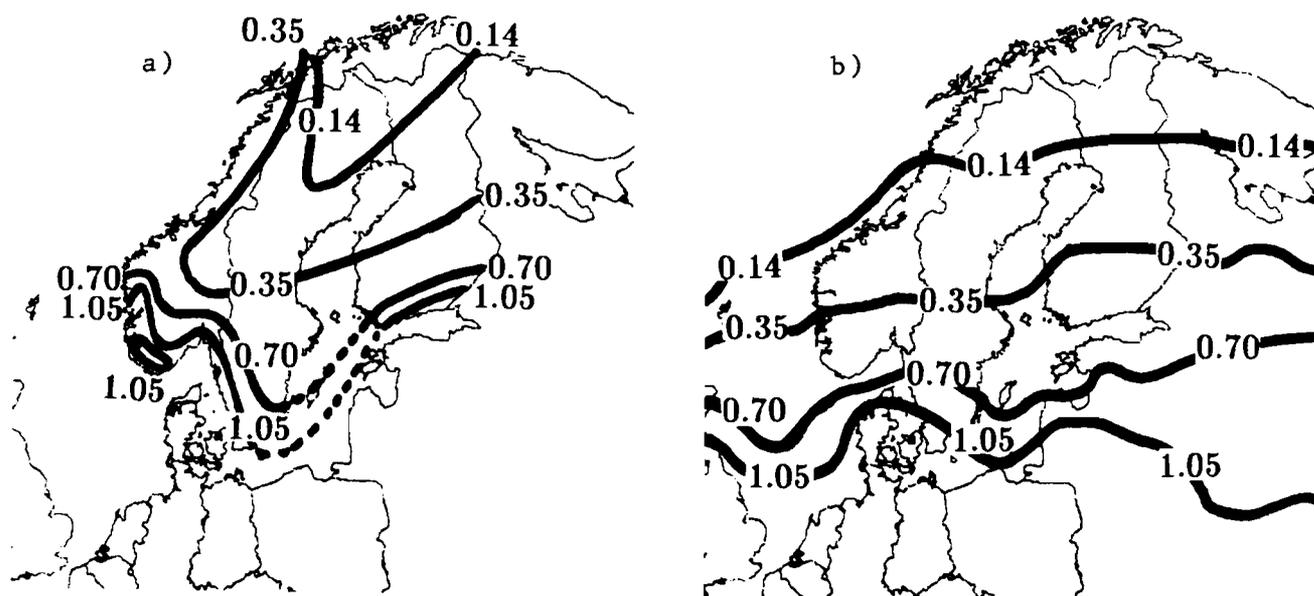


Figure 13. Comparison of model calculations vs observations in Nordic countries. (a) Observed wet nitrogen ( $\text{NO}_x$ -nitrogen +  $\text{NH}_x$ -nitrogen) from Andersen (1986). (b) Computed total (wet plus dry,  $\text{NO}_x$ -nitrogen +  $\text{NH}_x$ -nitrogen) deposition. Units:  $\text{g m}^{-2} \text{yr}^{-1}$  nitrogen.

The higher observed deposition along the Norwegian coast could result from intense precipitation due to orographic effects. This more "local" phenomena cannot be well taken into account by the interregional-scaled  $\text{NO}_x$  model. Deposition in Finland is probably underestimated by the model because of inaccurate representation of  $\text{NO}_x$  emissions in the Northwest USSR and Poland. All in all, however, both Figures 13 and Figure 9 show that the model computes N deposition in the correct order of magnitude.

**Critical Load of Nitrogen.** It is, of course, difficult to assess the ecological significance of the nitrogen deposition picture in Figure 12(a). One point of reference is presented in Table 4, which is a summary of the Nordic Council's recommended "critical loads" of nitrogen deposition in Europe. As stated in their report, the critical load is "The highest load that will not cause chemical changes leading to long-term harmful effects on the most sensitive ecological systems" (Nilsson, 1986; p.4). But these figures must be used with extreme caution because the impact of nitrogen deposition on soil, surface water, groundwater, or forests certainly depends on site-specific dynamic processes. As an example, nitrogen deposition may or may not acidify a lake depending on the resilience of a particular watershed to deposition as well as the time history of deposition -- how fast and for how long deposition occurs. These dynamic processes cannot be captured by a static critical load.

Table 4. Estimated critical loads for nitrogen based on different concepts. From Nilsson (1986).

Concepts	Critical Load $\text{kg N} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$	Remarks
<b>Basic concept</b>		
1. Net removal in forestry operation -low to medium productivity -high productivity	5-20 20-45	Total
<b>Concepts for validation</b>		
2. Input/output studies	10-15	Wet
3. Vegetational changes	10-20 (30)	Total
4. The nitrogen productivity concept	15-35	Total
5. Surface Water	15	Total
6. Nutrient balance in the soil	< 20	Total

With these qualifications in mind, we note that a commonly cited critical load of total nitrogen ( $\text{NO}_x$  plus  $\text{NH}_x$ , and wet plus dry deposition) is about  $1.0 \text{ g m}^{-2} \text{ yr}^{-1}$ , or lower. Figure 12 (1980 deposition) indicates that a large area of Europe receives in excess of this amount. This includes nearly all of Europe south of the Nordic countries and the northern part of the USSR, and north of Spain, Greece and Turkey. Deposition is greater than  $3.0 \text{ g m}^{-2} \text{ yr}^{-1}$  in the Benelux countries and in a large part of the FRG and GDR. Deposition decreases to  $0.1 \text{ g m}^{-2} \text{ yr}^{-1}$  on the fringes of Europe.

**$\text{NO}_x$ -N Deposition Scenarios.** Since the reduction scenarios described above pertain only to  $\text{NO}_x$  emissions, we now focus on the deposition of  $\text{NO}_x$ -nitrogen deposition. We will return shortly to total N calculations. For reference we again present the 1980

$\text{NO}_x$ -nitrogen deposition in Figure 14(a). Note that the  $0.1 \text{ g m}^{-2} \text{ yr}^{-1}$  slices through the Nordic countries and  $1.0 \text{ g m}^{-2} \text{ yr}^{-1}$  covers a large area of Central Europe.

We begin with the *EEC Directive* (Figure 14(b)) which moves the  $0.1 \text{ g m}^{-2} \text{ yr}^{-1}$  isoline slightly southward compared to the 1980 deposition, and reduces by two-thirds the area covered by  $1.0 \text{ g m}^{-2} \text{ yr}^{-1}$  to a large part of the Benelux countries, together with FRG, GDR, and a small part of Czechoslovakia.

The *Luxembourg Agreement* (Figure 14(c)) has a very small effect on the 0.1 and 1.0 isolines, but does reduce by one-half the area covered by  $\geq 1.5 \text{ g m}^{-2} \text{ yr}^{-1}$  deposition.

Interestingly, the application of *U.S. Traffic Limits* (Figure 14(d)) to European  $\text{NO}_x$  emissions "flattens out" and reduces by one-half the area covered by  $1.0 \text{ g m}^{-2} \text{ yr}^{-1}$ , and concentrates the highest levels of deposition on the center of Europe.

The *Maximum Point Source* scenario (Figure 14(e)) reduces the area of the  $1.0 \text{ g m}^{-2} \text{ yr}^{-1}$  isoline to much greater extent than the previous scenarios (by three-quarters relative to the 1980 reference case), but the isoline of  $0.1 \text{ g m}^{-2} \text{ yr}^{-1}$  is virtually unchanged.

We now come to the most extreme of the reduction plans, the "*Maxnox*" scenario (Figure 14(f), which calls for a 62% reduction in European  $\text{NO}_x$  emissions. We see in Figure 14(f), that the  $1.0 \text{ g m}^{-2} \text{ yr}^{-1}$  isoline disappears. Also in this scenario we finally see movement of the  $0.1 \text{ g m}^{-2} \text{ yr}^{-1}$  isoline, as it retreats south to southern Sweden, and north towards the Spanish-French and Greek-Yugoslavian borders. The area covered by this isoline is only about one-half the area it covers in the 1980 reference scenario.

*Total N Deposition Results.* The situation is different, however, when we examine the effect of the Maxnox scenario on total ( $\text{NO}_x$  plus  $\text{NH}_x$ ) nitrogen rather than  $\text{NO}_x$  alone (Figure 12(b)). The 0.1 isoline re-appears in nearly the same location it had for the 1980  $\text{NO}_x$ -nitrogen deposition (Figure 14(a)). Also the 1.0 isoline still covers much of Europe south of the Nordic countries, and north of Central France, Italy and Yugoslavia. The peak deposition level has been reduced, however: The area of Central Europe which had greater than  $3.0 \text{ g m}^{-2} \text{ yr}^{-1}$  in the 1980 deposition picture (Figure 12(a)) now has  $2.0 \text{ g m}^{-2} \text{ yr}^{-1}$ . Also, the area covered by  $1.0 \text{ g m}^{-2} \text{ yr}^{-1}$  has decreased by 22%.

It seems that reductions of  $\text{NO}_x$  emissions alone may not adequately reduce the total nitrogen deposition in Europe. As pointed out at the beginning of this paper, measurements indicate that ammonia makes up about 60% or more of the total nitrogen in wet deposition. Of course the word "adequately" depends on acceptance of the critical load concept for nitrogen deposition which, as pointed out earlier, has major drawbacks.

The next step in the assessment of nitrogen deposition in Europe should be to link computed nitrogen deposition with mechanistic models of forests, lakes and soil. For example, Agren and Kauppi (1983) proposed a regional-scale model for studying nitrogen saturation in forests. This type of model allows us to better assess the significance of observed or predicted deposition levels. We would be able to take into account the dynamic processes of ecological impacts of nitrogen and would not have to rely on the static critical load concept. Also, the underlying assumptions of the nitrogen deposition calculations, such as uncoupled  $\text{NO}_x$  and  $\text{NH}_x$  chemistry, should be examined with model experiments using more complicated models.

## Summary and Conclusions

1. Separate models are used to calculate  $\text{NO}_x$  and  $\text{NH}_x$  nitrogen deposition. We have based our  $\text{NO}_x$  calculations on a simplified version of a non-linear  $\text{NO}_x$  model. The non-linear  $\text{NO}_x$  model has been previously checked against  $\text{NO}_3^-$  precipitation data. Since the simplified model only calculates total (wet plus dry)  $\text{NO}_x$  deposition, we were unable to check it decisively against data because dry deposition data of the necessary time scale

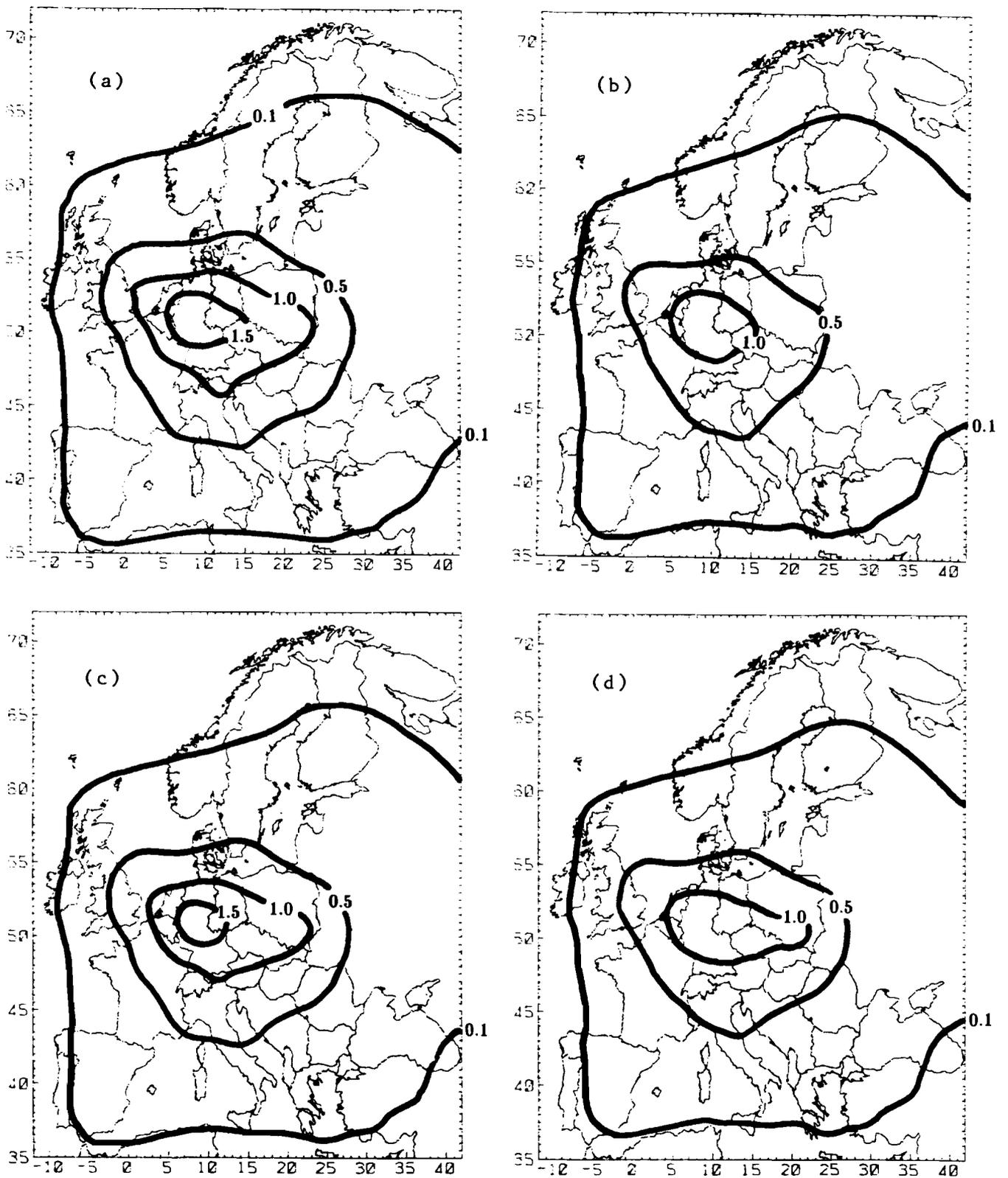


Figure 14. NO<sub>x</sub>-nitrogen deposition for reduction scenarios: (a) 1980 emission - no control, (b) EEC Directive, (c) Luxembourg Agreement, (d) US Traffic Limits. Units: g m<sup>-2</sup> yr<sup>-1</sup> nitrogen. Continued on next page.

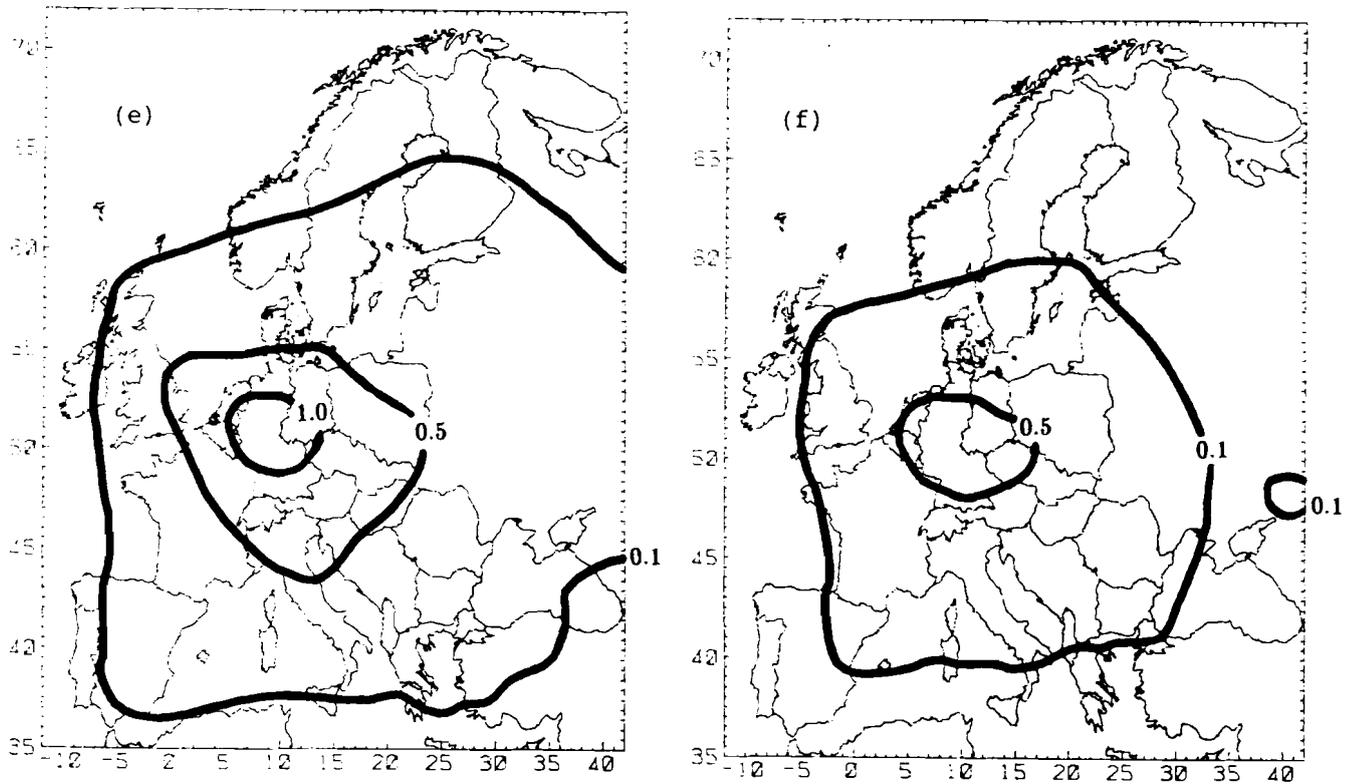


Figure 14. (Continued) (e) Max. Point Source Controls, (f) Maxnox. Units:  $\text{g m}^{-2} \text{yr}^{-1}$  nitrogen.

and spatial coverage were unavailable. However we have compared simplified model output with wet deposition patterns in Europe and found it to be at least consistent with these data.

Our  $\text{NH}_x$  calculations are based on results from a model that has been tested against ammonium aerosol and ammonia precipitation data.

2. Results from the  $\text{NO}_x$  and  $\text{NH}_x$  models are summed to obtain total nitrogen deposition. These calculations implicitly assumed that the relationship between emissions and deposition is linear, that the proportional distribution of emissions within a country will not change even if total country emission changes, and that the behavior of  $\text{NO}_x$  and  $\text{NH}_x$  in the atmosphere is uncoupled. These crude assumptions may be appropriate for the long time and space scales considered in this paper, though they should be more closely studied.

3. We evaluated the effect of five  $\text{NO}_x$  emission reduction scenarios which were adapted from the OECD and applied in a consistent fashion to each of 27 European countries. The most extreme plan, "Maxnox", reduced total European emissions by 62%. The others reduced emissions from 9 to 35%.

4. The scenarios had the following effect on *oxidized* nitrogen deposition: Only the Maxnox scenario appreciably affected the isoline of  $0.1 \text{ g m}^{-2} \text{ yr}^{-1}$ . All scenarios with the exception of the Luxembourg Agreement significantly reduced the area covered by  $1.0 \text{ g m}^{-2} \text{ yr}^{-1}$ . For the Maxnox scenario, the area of  $1.0 \text{ g m}^{-2} \text{ yr}^{-1}$  disappeared.

5. The scenarios had the following effect on *total* ( $\text{NO}_x + \text{NH}_x$ ) nitrogen deposition: The most extreme  $\text{NO}_x$  reduction plan, the Maxnox scenario, reduces peak deposition levels (which extend from the Benelux countries to the GDR) by one-third. However, it only slightly alters the area covered by 0.1 and  $1.0 \text{ g m}^{-2} \text{ yr}^{-1}$  (compared to the 1980 reference case). Overall, the pattern of total nitrogen deposition is not changed very much. This emphasizes the need to reduce  $\text{NH}_x$  emissions, as well as  $\text{NO}_x$  emissions, to accomplish reductions of total nitrogen deposition in Europe.

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