WORKING PAPER

A MODEL FOR ESTIMATING NITROGEN OXIDE EMISSIONS IN EUROPE

Barbara Lübkert

December 1987
WP-87-122

International Institute
for Applied Systems Analysis

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PREFACE

IIASA's RAINS model have so far dealt with the emissions and effects of sulphur dioxide. However, acidification is caused by other pollutants as well. In particular, oxides of nitrogen play an important role. Barbara Lübkert joined the Acid Rain Project during the summer of 1986 as a participant in the Young Scientists' Summer Program and developed a method to estimate the emissions of nitrogen oxides. This paper describes the method she used and presents the results.

Her work is very important in that it makes the first step toward a RAINS model that covers nitrogen as well as sulphur.

Roderick W. Shaw
Leader
Acid Rain Project

ACKNOWLEDGEMENTS

The work presented in this paper was supported by the Organisation for Economic Co-operation and Development (OECD) and carried out by the author during several visits to IIASA; she is grateful to Erik Lykke and Pierre Lieben at the OECD Environment Directorate as well as to Leen Hordijk, who made this series of visits possible. The author further thanks Soizick de Tilly, Peter Lübkert, and Steven Beatson of the OECD Pollution Control Division and her colleagues in OECD Member countries who helped in compiling and revising emission data used in this study; thanks are also due to Markus Amann, Stuart Batterman and Wolfgang Schöpp of IIASA's Acid Rain Project for their constructive ideas and Joseph Alcamo for his critical review of this paper.

ABSTRACT

This paper presents a methodology to calculate nitrogen oxide (NO_x) emissions in a consistent way for all European countries. First, the principles of NO_x formation are reviewed, and results of a feasibility study carried out to develop an NO_x emission submodel for inclusion into IIASA's RAINS model to quantify acid deposition in Europe are summarized. It is shown that NO_x emissions are most closely correlated to fossil fuel consumption and that this consumption is, therefore, the best single statistical parameter to estimate a country's NO_x emissions from the various source sectors. Existing NO_x emission inventories and emission factors are analyzed, and a set of comparable data from the OECD emission inventory for major air pollutants (OECD, 1987) is identified which is appropriate for statistical analysis in order to estimate sector- and fuel-specific emission coefficients. Such aggregate coefficients are determined using ordinary least squares (OLS) regression analysis for the sector/fuel combinations presently contained in RAINS to calculate sulfur emissions. Fuel data used in the regression analysis are from the International Energy Agency (IEA, 1987a,b).

Emission coefficients determined in this way are entered into RAINS and NO_x emissions are determined for all 27 European countries. Comparison of the results with reported national totals as well as reported traffic NO_x shows generally good agreement, i.e., within 20 percent. If NO_x emissions calculated by RAINS are compared to EMEP estimates (United Nations, 1987) on a total European scale (USSR excluded), RAINS overestimates the EMEP total by only four percent. Relative contributions as calculated by RAINS for the transportation sector also reflect numbers reported by countries, being on the average 54 percent of total NO_x for OECD Europe and 16 percent for non-OECD Europe (excluding the USSR). A qualitative discussion about the uncertainty of estimated emission coefficients and resulting emission rates is also included. The approach described in this paper is, thus, promising and shows a way in which one can use a set of comparable data in a "top-down" approach to extrapolate the data to other countries where less detailed data are available. The method also allows for testing future emission scenarios with and without assumed emission reduction policies.

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A MODEL FOR ESTIMATING NITROGEN OXIDE EMISSIONS IN EUROPE

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1. INTRODUCTION

The Acid Rain Project at IIASA has developed a set of interactive, computer-based models – RAINS (Regional Acidification Information and Simulation) – to assess long-term acidification in Europe on a regional scale. To date, the RAINS model estimates current and future levels of acid deposition and their impacts on the environment due to sulfur compounds only. The available submodels first estimate emissions of sulfur due to a chosen energy forecast (Economic Commission for Europe (ECE), 1987; International Energy Agency (IEA), 1983), then relate these emissions to atmospheric deposition via a source-receptor matrix (Eliassen and Saltbones, 1983), and lastly predict impacts on forest soils, trees, lakes and groundwater (Kauppi *et al.*, 1986; Mäkelä *et al.*, 1987; Kämäri and Posch, 1987; Holmberg *et al.*, 1987). The model further has the option of evaluating the effect of various emission reduction scenarios and their associated costs (Amann and Kornai, 1987). Since the model is designed to be especially useful to decision makers, particular emphasis has been put on easy-to-use, interactive computer software and good, comprehensible graphical representation of results. The reader is referred to Alcamo *et al.* (1987) for a more detailed description of RAINS and Hordijk (1986) for some policy applications of the model.

To date, we know that acidification of the environment is the result of a combined effect of dry and wet deposition of sulfur and nitrogen compounds. In order to make RAINS more comprehensive it has, therefore, become desirable to also include nitrogen oxides (NO_x) and ammonia (NH_4) into the model. This paper presents the results of a feasibility study carried out to develop an NO_x emission submodel, describes the methodology developed, presents NO_x emissions obtained by this method, and gives a comparison of these results with existing reported nitrogen oxide emission data. Results from this paper can be used to construct an NO_x emissions submodel which will be part of an expanded version of RAINS, as depicted in *Figure 1.*

2. GOALS AND LIMITATIONS OF THE NO_x EMISSION MODEL

The goals of the NO_x emission model are:

1. To provide a consistent method for calculating NO_x emissions in all European countries.
2. To base these calculations on statistical data which are comparable and readily available for all countries.

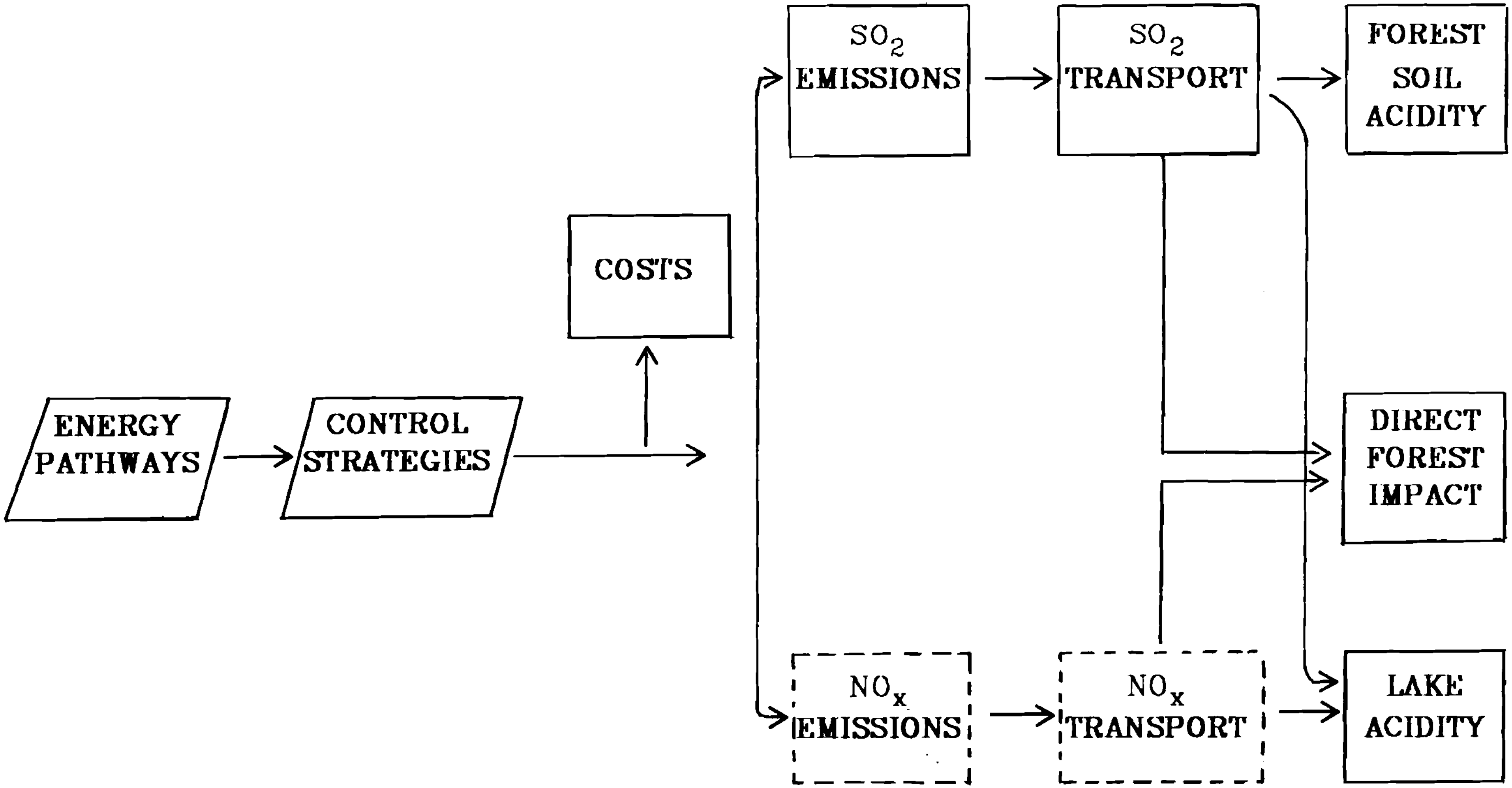

Figure 1. Schematic of the expanded IIASA RAINS model.

3. To be able to calculate emissions for different years in the past, present and future by making use of forecasts of the main variables influencing these emissions.
4. To be able to calculate emissions under different emission reduction scenarios.

Given these objectives a model must be developed by which emissions can be estimated in a relatively simple and aggregated way and therefore one should keep the number of variables that influence predictions to a minimum. Further, these main influencing parameters should be readily available from statistics, preferably collected on an international level which guarantee some compatibility.

The NO_x emission model presented in this paper is not intended to replace any of the detailed "bottom-up" approaches that are used in different countries to inventory past and present NO_x emissions, but is meant to show a way in which one can use a set of comparable data in a "top-down" approach to extrapolate these data to other countries and years.

3. PRINCIPLES OF NO_x FORMATION

Nitrogen oxides are emitted from man-made and natural sources. Man-made emissions are generated during fuel combustion and in industrial processes; they are emitted in the form of nitric oxide (NO) and nitrogen dioxide (NO_2). For most emission source categories, the NO_2 fraction of total NO_x (NO and NO_2 together) emissions is lower than 10 percent with the exception of plants producing nitric acid and/or ammonia (40 percent) and for gas turbines (15 percent) (Bakkum and Veldt, 1986a). Natural nitrogen oxide emissions are predominantly in the non-reactive form of nitrous oxide (N_2O) and are, thus, not believed to contribute to the acid-forming potential of the environment. Of all direct anthropogenic NO_x emissions, those from combustion processes in stationary and mobile sources constitute the overwhelming majority.

Nitrogen oxides are formed during combustion due to three principal mechanisms which result in the so-called (1) fuel NO_x, (2) thermal NO_x, and (3) "prompt" NO_x. Emissions of nitrogen oxides from industrial processes result from processing raw materials that contain nitrogen either as a necessary constituent or as a contaminant. Such emissions depend on the nitrogen content of the raw material and on its volatility. It is shown later that these process emissions are, however, of secondary importance.

3.1. Fuel NO_x

Fuel NO_x formation is a function of the fuel's nitrogen content as well as burner type and firing mode used in the combustion. Fuel nitrogen contents vary typically between 0.5 and 2.0 percent (by weight) for coal and shale, and between 0.5 and 1.5 percent for synthetic fuels derived from coal. The nitrogen content is less than 1 percent for oil and gas, and negligible for natural gas (US Department of Energy, 1983). Fuel NO_x is, however, also dependent on the burner type for which no simple relationship exists. *Figure 2* illustrates this complicated relationship.

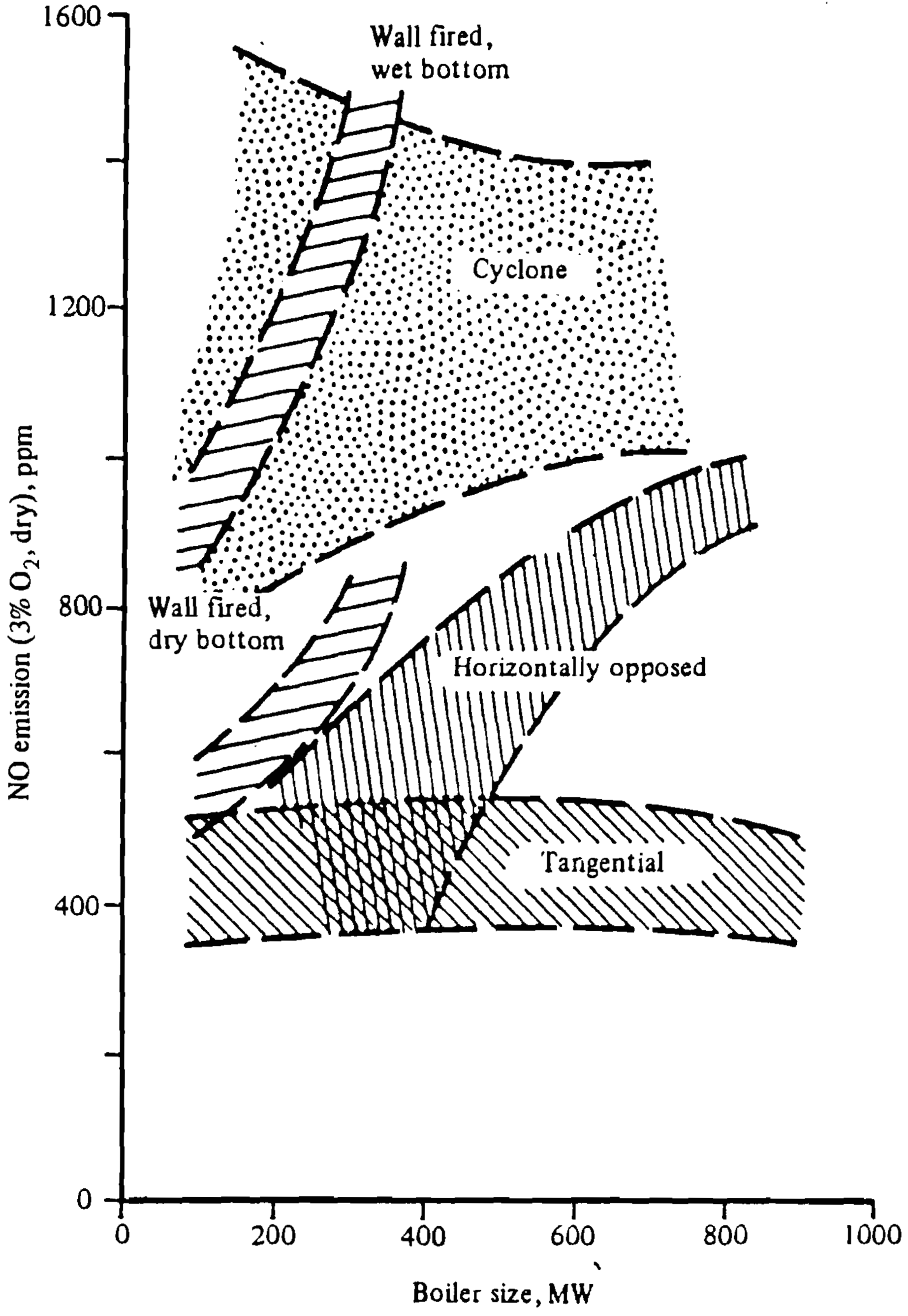

Figure 2. Effects of firing pattern on NO_x emissions (Thompson, 1979; cited in Morrison, 1980).

3.2. Thermal NO_x

Thermal NO_x formation is due to the mechanism discovered by Zeldovich (1946), in which nitrogen and oxygen from the air combine to form NO_x under high temperatures:

$$N_2 + O_2 \rightleftharpoons 2\ NO$$

$$N + O_2 \rightleftharpoons NO + O.$$

This formation process becomes important usually at temperatures above 1400 °C (Rentz, 1984), a temperature which is generally exceeded in most combustion processes. Thermal NO_x formation increases exponentially at higher temperatures, and is also a function of residence time of air in the combustion chamber and of the stoichiometry, i.e., the excess air availability.

To overall NO_x emissions, thermal NO_x contributes around 20 percent in coal combustion, about 50 percent in heavy oil combustion, and the largest portion in light oil and gas combustion (Mason and Herther, 1982).

3.3. "Prompt" NO_x

In the flame reaction zone, significantly higher amounts of NO_x are produced than can be explained by equilibrium calculations. This NO_x is termed "prompt" NO_x, but is generally an order of magnitude lower than thermal and fuel NO_x and is therefore considered negligible in this paper.

4. APPROACHES TO NO_x EMISSION CALCULATIONS

4.1. Theoretical NO_x Emission Calculations

As explained above, total NO_x emission from combustion is the sum of thermal, fuel and "prompt" NO_x emissions:

$$e(NO_x) = e(fuel\text{-}N) + e(thermal) + e(\ prompt\ ) \qquad [1]$$

in which
e(NO_x) = total of the NO_x emissions [tonnes],
e(fuel-N) = f(fuel, firing mode),
e(thermal) = f(combustion temperature, residence time, stoichiometry),
e("prompt") = 0.

Assuming different combustion temperatures, burner types and firing modes in different economic sectors and for different fuels, emissions should, therefore, also depend on the sector in which they are emitted and on the fuel type used:

$$e(NO_x) = \sum_{i,j} e_{ij}(NO_x)\ , \qquad [2]$$

where $e_{ij}(NO_x)$ is the emission rate per sector per fuel, and

$$e_{ij}(NO_x) = n_j{}^*E_{ij}/hv_j{}^*(1-a_i) + \alpha^*E_{ij}{}^*(1-a_i) \qquad [3]$$

in which n_j : nitrogen content of fuel [weight %],
E_{ij} : energy consumed per sector per fuel [PJ/yr],
hv_j : heat content per fuel [PJ/tonne],
a_i : nitrogen removed by flue gas cleaning in each sector [fraction],
α : emission factor due to the thermal NO_x; a function of combustion temperature, burner type and configuration, etc. [tonnes/PJ],
i : emission sector,
j : fuel type.

In practice, it is difficult to use Eq. [3] because the emission factor due to thermal NO_x, α , varies significantly as a function of combustion parameters which are not fuel specific, i.e., α can vary within one sector and fuel as much as between sectors and fuels.

Due to the high variability of NO_x emissions with combustion conditions, it would be necessary to subdivide the emission source categories into various equipment and fuel combinations having distinct emission characteristics. As described by Mason and Herther (1982), such an approach has resulted in over 150 of these equipment/fuel combinations in preparing an NO_x emission inventory for the USA for 1980. That many source categories seem presently infeasible in an NO_x emission calculation model on a European level, in which input data should be comparable between countries.

4.2. Definition of NO_x Emitting Source Categories

Nitrogen oxide emitting sources can be grouped according to economic activities and/or NO_x generating processes; refineries are an example of an economic sector whereas furnaces are an example of the latter category. In preparing emission inventories as well as in making forecasts, it is desirable to aggregate source categories in a way that allows, (1) emissions to be calculated in a uniform manner for the entire source category and (2) emission reduction scenarios to be evaluated. The latter means that one needs to be able to account for control scenarios that appear feasible for a specific economic sector or a particular emission generating process. Broad categories emitting NO_x are (1) mobile sources, including on- and off-road traffic, (2) power plants, (3) industrial and (4) non-industrial combustion, (5) industrial processes, in particular, ammonia and nitric acid plants, (6) agricultural sources, such as open burning, and (7) waste incineration. Of these seven categories, the last three are generally of minor importance at the level of national emissions.

4.3. Use of Emission Factors

Since in almost all countries, emission measurements are not carried out on a routine basis, emission rates are usually based on mass balance calculations and specific point source measurements. This knowledge is translated into an emission factor which represents an extrapolation of point-specific data to an entire emission source category. Therefore, physical verification is not possible of these emission factors nor of the subsequently calculated total emission rates.

It has been stated that, nowadays, knowledge about emission factors for major air pollutants, including NO_x, has accumulated to a level where, for the most important emission source categories, reasonable estimates can be made for total emission rates in these sectors (Bouscaren *et al.*, 1986). Reviewing recent publications and other available lists of

NO_x emission factors shows, however, that ranges of NO_x factors are still so large that their use requires some specific knowledge of the sector and its structure in order to give reasonable estimates. *Tables 1* and *2* (OECD-Air Management Policy Group, 1984; Löblich, 1985) give an overview of such ranges of emission factors for NO_x. It should be noted that these tables are already the result of some extensive literature research trying to present emission factor ranges that are applicable at least to the western European situation.

Table 1. NO_x emission factor ranges found in the literature (OECD - Air Management Policy Group, 1984; Löblich, 1985).

Sector	*Hard Coal*	*Brown Coal*	*Derived Coal*	*Gas*
Transportation	–	–	–	–
Power Plants	280–630	180–290	–	50–140
Domestic	45–100	50–300	70–100	30–120
Industry	200–460	200–300	180–540	40–170

Sector	*Light Fraction of Oil*	*Middle Distillate*	*Heavy Oil*	*Other Solid Fuels*
Transportation	1050 [=44 g/kg]^1	1175 [=50 g/kg]^2	–	–
Power Plants	–	60–150	150–245	120–300
Domestic	50–100	50–160	140–240	50–220
Industry	45–60	60–190	140–240	80–300

Units: tonnes NO_2/PJ input.

Notes:

(1) for gasoline automobiles (see *Table 2*).

(2) for diesel trucks (see *Table 2*).

N.B.: All emission factors proposed by OECD were elaborated in cooperation with TNO (The Netherlands) as average best estimates.

The literature as well as *Tables 1* and *2* show that currently used aggregate NO_x emission factors are fuel-specific (coal, oil, gas, and other solid fuels) and distinguish between different types of coal and oil. Industrial process emissions are generally related to the amount of product manufactured. The use of such emission factors, therefore, requires detailed statistical data on fuel use and production rates in each of the economic sectors. Usually, these data are not readily available from international statistics. When available from national statistics, they are often not comparable between countries.

4.4. Existing Emission Inventories

Some countries have a significant experience in gathering emission data for major air pollutants. These data are usually in the form of emission inventories for particular years and, in many cases, present information about emissions per source category according to the various economic sectors (e.g., power plants, refineries, non-ferrous metal industry, dry cleaning, agriculture, etc.), and/or by type of emission generating process (e.g., combustion, raw material and product storage, industrial processes, etc.). As noted above, a serious problem, however, is the usual lack of comparability or compatibility

Table 2. Emission factors for the transportation sector (road traffic).

	Fuel Type			*Literature Reference*
Vehicle Type	*Gasoline*	*Diesel*	*LPG*	
AUTOMOBILE				
[g/km]	2.54–2.56	0.62–0.74		(1)
	2.17	1.06		(2)
	1.3–3.8	0.9–1.8	1.25–2.5	(3)
[g/kg fuel]	44	20	33	(1)
LIGHT-DUTY TRUCKS				
[g/km]	5.17	0.62–0.74		(1)
	2.0–11.0	1.1–3.8	1.6–3.2	(3)
[g/kg fuel]	65	22		(1)
HEAVY-DUTY TRUCKS				
[g/km]	4.1	11.5–23.1		(1)
	6.0–14.0	2.1–16.0		(3)
	3.03	13.98		(2)
[g/kg fuel]		50		(1)
MOTORCYCLES				
[g/km]	0.3			(1)
[g/kg fuel]	3.0			(1)

(1) OECD – Air Management Policy Group (1984). Note: All emission factors proposed by the OECD were elaborated in cooperation with TNO as average best estimates.

(2) Energiebericht und Energie Konzept der österreichischen Bundesregierung, Austria (1984).

(3) Handbook of Emission Factors, The Netherlands (1980).

between data from different countries.

The Organisation for Economic Co-operation and Development (OECD) together with the Commission of the European Communities (CEC) is currently preparing a coherent, and therefore as far as possible, comparable emission inventory for major air pollutants, i.e., SO_2, NO_x, and volatile organic compounds (VOC's), for OECD-Europe. Together with experts from member countries, the OECD developed a set of general guidelines (OECD-Air Management Policy Group, 1984; Lübkert, 1987) to allow countries to prepare national inventories in a comparable form. Individual member countries have followed these prescribed guidelines and, in early 1987, national emission inventories were available for 12 countries. These inventories have undergone several rounds of verification to assure their compatibility and results have been described by Lübkert and de Tilly (1987a, 1987b).

In general, it can be assumed that these inventories were calculated in a detailed "bottom-up" way, following a set of similar, if not identical, assumptions and definitions, and by making use of the most detailed information available in each country. Major emission sectors – such as those used in RAINS – include the same sub-categories, and are therefore comparable.

4.5. Analysis of the OECD NO_x Emission Inventory

The OECD inventory was compiled for the base year 1980. The 12 countries included at the time of this study were: Austria, Denmark, Finland, France, FRG, Italy, the Netherlands, Norway, Portugal, Sweden, Switzerland, and the UK.

The total national emissions as reported by countries have been correlated with general statistical parameters, including the number of inhabitants, the surface area per country, the gross national product, and the fossil fuel energy consumed. As was expected, NO_x emissions are most closely related to total fossil fuel used (coefficient of determination $r^2 = 0.98$, $t = 36.09$, therefore, confidence level ≥ 99.9 percent), and energy use is, therefore, the best single statistical parameter to correlate NO_x emissions with.

OECD emission data have also been split into the same source sectors contained in the current RAINS SO_2 emission model: (1) transportation, (2) power plants, (3) domestic, (4) industrial combustion, (5) fuel conversion, and (6) industrial processes. The "fuel conversion" sector in RAINS represents refineries and other fuel conversion plants; the only NO_x emissions from this sector are from refinery furnaces and such, resulting from stationary, indirect fuel combustion. The mechanism of generation is, thus, almost identical to industrial combustion, and emissions from the conversion sector in the OECD inventory have been included in "industrial combustion" since they cannot be separated for all 12 countries. The two sectors have, therefore, been added together. Industrial process emissions are not calculated in the sulfur emissions submodel since they are not energy-related, but are added as constants if known, or neglected. In the case of nitrogen oxides, process emissions are generally minor (see *Table 3*). OECD emission data aggregated into the IIASA source sectors are contained in Annex 1. The relative contributions by these sectors to total national NO_x emissions in the 12 OECD countries are shown in *Table 3*. The four main, energy-related emission sectors, which are responsible for almost all NO_x emissions are: (1) transportation, (2) power plants, (3) domestic and (4) industrial combustion.

Table 3. Overall emission contribution by sectors to total NO_x emissions in OECD-Europe in 1980 (Lübkert and de Tilly, 1987b).

	Contribution to Total National NO_x Emissions [%]				
Country	*Transportation*	*Power Plants*	*Domestic Combustion*	*Industrial Combustion*	*Industrial Processes*
AUSTRIA	68	9	5	14	5
DENMARK	33	50	9	7	0
FINLAND	53	37	-	4	3
FRANCE	54	14	11	17	3
GERMANY	55	26	6	9	5
ITALY	61	18	5	12	3
NETHERLANDS	58	15	7	8	11
NORWAY	75	0	4	12	9
PORTUGAL	64	12	-	8	16
SWEDEN	61	3	15	11	10
SWITZERLAND	70	0	4	25	0
UNITED KINGDOM	37	44	3	13	0
AVERAGE	53	24	6	12	4

Until now, only very limited information is available from the countries concerning emissions per fuel type in each sector; on the other hand, internationally comparable information on total energy consumption in 1980 per sector per fuel is available from the OECD IEA statistics.

4.6. Review of SO_2 Emission Calculations in RAINS

The sulfur emission submodel in RAINS calculates SO_2 emissions for each of the above mentioned economic sectors and for each of the following eight fuel types: (1) hard coal, (2) brown coal, (3) derived coal, (4) light fraction of oil (including gasoline), (5) middle distillate (including diesel), (6) heavy oil, (7) gas, and (8) other solid fuels (including wood, etc.). Future emission estimates are therefore dependent on the increased or decreased use of a particular fuel in each sector. If RAINS is to predict the overall impact of sulfur and nitrogen deposition on the environment, it would be advantageous to have emission estimates for both pollutants based on the same statistical variables, i.e., energy forecasts. The advantage of this would be that any mistake in energy predictions would only affect the absolute total emissions and resulting deposition, but would not change the relative contributions to total deposition by the two pollutants.

RAINS contains energy balances for all 27 European countries included in the model; these are based on UN ECE and OECD IEA energy statistics and balances (ECE, 1987; IEA, 1983; IEA, 1984) and form part of the sulfur emission submodel. Generally, the IEA statistics are more detailed, but give data only for OECD-Europe, whereas the ECE statistics give comparable data for all European countries. The total fuel consumption figures are, therefore, taken from the ECE statistics, whereas the allocation to specific coal and oil types (e.g., hard coal, brown coal, and derived coal) is mainly based on IEA information. Unfortunately, the total fuel consumption figures per country are not necessarily identical in the two sets of energy statistics.

In order to use the RAINS energy balances to also predict NO_x emissions, representative, average emission factors for each of these 32 sector/fuel combinations would have to be found. The NO_x emission factor ranges from the literature are, however, too broad, and the proper choice would require too detailed information about each sector in each country.

5. A METHOD FOR CALCULATING NO_x EMISSIONS

From the analysis of the OECD NO_x emission inventory, it can be concluded that this set of data, although small, is appropriate for statistical analysis. The next objective in this study was, therefore, to find an appropriate set of aggregate emission factors by statistical regression analysis for the emission source sectors and different fossil fuels currently used in RAINS.

Nitrogen oxide emissions per sector are the sum of emissions due to the combustion of each of the eight fuels in this sector:

$$e_i = \sum \beta_{ij} E_{ij} \qquad [4]$$

in which e_i : NO_x emissions per sector, $i = 1,\ldots,4$;
β_{ij} : emission factor per fuel per sector, $j = 1,\ldots,8$;
E_{ij} : energy consumed by each of the eight fuels per sector.

In order to get a set of equations which could be estimated for all β_{ij}, it was assumed that emission factors per fuel per sector are the same in each of the 12 European OECD countries. This results in the following equation:

$$e_{ik} = \sum \beta_{ij} E_{ijk} \qquad [5]$$

in which k is the country index; $k = 1,...,12$.

Energy use data E_{ijk} were taken from the OECD IEA energy statistics for 1980 (IEA, 1987a) and the OECD IEA energy balances for "other solid fuels" (IEA, 1987b), which are not included in the energy statistics; total NO_x emission rates per sector e_{ik} were taken from the 1980 OECD emission inventory for NO_x (Lübkert and de Tilly, 1987b). It was decided to estimate the resulting eight unknown parameters by utilizing ordinary least squares (OLS) regression analysis:

$$e_{ik} = \sum \beta_{ij} E_{ijk} + \epsilon_{ij} \qquad [6]$$

in which ϵ is the disturbance term. Before doing so, the absolute amounts of fuel use per country, which are quite different between countries, were converted into their relative shares; i.e., the matrix of independent variables was normalized in order to avoid heteroscedasticity of the disturbance term.

When attempting to solve for eight unknowns with only a maximum of 12 observations, this usually gives implausible results, such as negative coefficients. Therefore, several techniques were used to increase the degrees of freedom in this set of equations. First, all coefficients were eliminated if their corresponding fuel use in a sector was zero in all countries. This reduced the total number of coefficients from 32 to 26. Since most coefficients could be eliminated in one sector (transportation) and, thus, did not help in increasing the degrees of freedom in other sectors, seemingly unrelated regression (SUR) analysis (Zellner, 1962) was tried. This still resulted in some negative emission coefficients.

In the next step, various combinations of aggregations were carried out in the following way:

1. Different sectors were taken together if emission factor ranges shown in *Table 1* for all fuels in these sectors were very similar. As stated earlier, NO_x emissions depend on a variety of combustion parameters and are therefore, in some cases, not necessarily sector-specific but rather fuel-related. In doing so, the number of observations was doubled for the same number of coefficients to be estimated.
2. Different fuel types within one sector were aggregated into the same group if emission factor ranges as shown in *Table 1* were identical or very similar for these fuels. This reduced the number of coefficients to be estimated, and it appears justified because some NO_x emission factors seem to depend more on combustion conditions, such as temperature, etc., than on fuel type.
3. Fuel types were eliminated from regression analysis, also to decrease the number of coefficients to be estimated. This was, however, only done if the remaining fuel mix still covered, on average for all observations, at least 90 percent of all fossil fuel used in the specific sector. In this way, it was tried to keep the error introduced by eliminating explanatory variables from the regression small.

In order to obtain plausible results, the number of independent variables was reduced to a maximum of three if the sample size was 12 or less, and to four if the sample size was larger than 12. In this way, the following sectors or combinations of sectors and fuel types or groups of fuels were selected for final use in the OLS regression:

1.	Transportation	Light oil (i.e., gasoline) and middle distillate (i.e., diesel) as two individual fuel types. These cover together on average in all 12 countries 99 percent of total fossil fuel consumption by this sector, varying from 97 to 100 percent for individual countries.
2.	Power Plants and Industry as a combined sector:	Middle distillate and heavy oil as one group, hard coal, brown coal, and gas as individual fuels. Together these five fuels cover 92 percent on average of total fossil fuel use in these two sectors in all 12 countries. For power plants alone, these fuels cover on average 99 percent in the 10 countries that have emissions from this sector, ranging from a low of 84 percent in Finland to a high of 100 percent in Denmark and the UK. For industry alone, these fuels cover 86 percent on average in all 12 countries, ranging from as low as 60 percent in Sweden to 96 percent in Denmark.
3.	Domestic:	Hard coal, light oil, and gas as one group, and middle distillate and heavy oil as two individual fuels, together covering on average 95 percent of total fossil fuel use in this sector in the 10 countries that reported emissions. For individual countries, these fuels cover between 73 (Austria) and 100 percent (Netherlands).

The sample size was 12 for "transportation", 22 for "power plants and industry" (i.e., 10 countries for "power plants" and 12 countries for "industry"), and 10 for "domestic". In the case of "power plants", two countries (Norway and Switzerland) do not have any major fossil fuel power plants and, thus, no emissions from this sector. In the case of "domestic", Finland and Portugal have not reported emissions from this sector. Emission data used in the "industry" sector include those originating from combustion in industry and fuel conversion such as refineries.

The constant in the regression analysis was always set to zero, i.e., forcing the regression line through the origin. This was based on the assumption that emissions only occur if some fuel is burned.

It should be noted that "other solid fuels" were not included in any final regression even though their use makes up a relatively significant fraction of total fossil fuels in some sectors and some countries, in particular in the "domestic" and "industry" sectors, particularly in Scandinavia. However, no plausible emission coefficients could be derived, probably reflecting the weakness of the energy data. It should be recalled that "other solid fuel" consumption for each country is taken as the difference between all "solid fuels" per sector in the IEA Energy Balances (1987b) and the sum of all coals per sector in the IEA Energy Statistics (1987a); this may be a significant source of error. Further, emission data included in the OECD inventory for "other solids" are most likely also relatively weak; and only few aggregate emission factors are available from the literature for comparison.

6. RESULTS

Emission coefficients as calculated by OLS regression analysis for the fuel types and fuel type aggregations as described above are given per sector in *Table 4*; standard errors and probability values (t-statistics) for each estimated coefficient as well as the coefficients of determination (r^2) for the overall regressions are also presented. Comparison of the calculated coefficients with emission factor ranges of *Table 1* shows that they generally fall within the same ranges; only for gas, the light fraction of oil, and hard coal in the domestic sector, the coefficient tends to be low.

Table 4. Aggregate emission coefficients calculated by ordinary least squares (OLS) method.

Sector *[std. error]* *(t-statistic)*	*Fuels*								
	HC	*BC*	*DC*	*GAS*	*LF*	*MD*	*HO*	*OS*	R^2
Transportation	–	–	–	–	455 [195] (2.3)	1580 [395] (3.9)	–	-	0.29
Power Plants and Industry	460 [65] (7.3)	195 [245] (0.8)	–	70 [80] (0.9)	–	185 [45] (3.9)	185 [45] (3.9)	–	0.57
Domestic	35 [30] (1.2)	–	–	35 [30] (1.2)	35 [30] (1.2)	90 [30] (3.0)	180 [95] (1.9)	–	0.30

Units: tonnes NO_2/PJ input.
LEGEND: HC: hard coal
BC: brown coal
DC: derived coal
LF: light fraction of oil (incl. gasoline)
MD: middle distillate (incl. diesel)
HO: heavy oil
OS: other solid fuels (e.g., wood)

As might be anticipated from the small sample of only 12 countries, the statistical parameters indicate that coefficients as calculated in these regressions are not always significant (i.e., the confidence level is less than 95, or even 90 percent). Since many different sector combinations and fuel aggregations were tried, some further indication about the reliability and thus, the robustness of estimated coefficients was obtained. In those cases where coefficients with high probability values were estimated in one particular aggregation, usually very similar coefficients, varying less than ± 5 percent, were also calculated in other aggregations. Examples are "transportation" coefficients for gasoline and diesel as well as "hard coal" and "heavy oil" emission coefficients for the power plant sector. In the case of "gas", low probability values (below the significance level) were obtained in almost all combinations even though the coefficients themselves were often very similar, i.e., within ± 10 percent of those reported in *Table 4*. Another such example is the "brown coal" coefficient for power plants.

The estimated coefficients represent emission factors for calculating baseline emissions without add-on control equipment in place; combustion modifications are assumed to have only been carried out to improve efficiency in production but not to reduce atmospheric pollution. These emission coefficients can, thus, be considered "uncontrolled", and the use of any emission reduction technology in future scenarios can be figured into the emission calculations by applying the average reduction percentage achievable in a specific sector, as follows:

$$e_{ij} = \beta_{ij} {}^{*} E_{ijk} {}^{*} (1-a). \qquad [7]$$

The next step was to check the reasonableness of this method. The calculated NO_x emission coefficients were entered into an emission factor matrix and incorporated into RAINS; all other emission coefficients were assumed zero. The emission coefficients determined for industrial combustion were used in both RAINS' sectors, "industry" and "conversion". This was appropriate because IIASA's energy balances only include fuels actually consumed in the "conversion" sector.

By multiplication of the NO_x emission coefficients with IIASA's energy balances for 1980, total NO_x emissions per sector and per country were calculated for all Europe. The results of these calculations are shown in *Table 5* for total national NO_x emissions; they are compared to other existing emission inventories such as ECE, EMEP, and PHOXA, and the percentage difference with these inventories is indicated. In many cases (12 out of 27), this difference is smaller than 20 percent, which currently represents the desired accuracy with which modelers would like to have total national NO_x emissions reported (OECD-Air Management Policy Group, 1986). Eggleston and McInnes (1987) showed that, even with very detailed national background data, the uncertainty for British road traffic NO_x emissions, for example, is still as high as 40 percent. If compared on a total European scale (USSR excluded), the RAINS calculations overestimate the EMEP total by only four percent. If compared for all 18 European OECD countries, RAINS overpredicts total NO_x emissions by five percent, and for non-OECD countries (excluding the USSR), the model underpredicts the total by three percent. Large differences are mainly observed between RAINS calculations and ECE or EMEP estimates for non-OECD countries and those countries that show considerable differences between estimates from different inventories. Agreement between RAINS results and PHOXA estimates is within ± 20 percent for all countries, OECD and non-OECD, with the only exception of GDR.

In order to further check the reasonableness of this approach, national emission rates for each economic sector were analyzed and compared with available inventory data. Unfortunately, the availability of sector-specific emission data is limited. Therefore, such comparison was possible only in the case of emissions from transportation. This sector, however, contributes a major share to total NO_x emissions, being 53 percent on the average for the 12 OECD countries used in this analysis. *Table 6* shows the results of this comparison. In general, the over- and underestimations are of the same order of magnitude as for total NO_x emissions when RAINS calculations are compared with OECD and ECE/EMEP figures. In eight cases out of a total of 20, differences are less than ± 20 percent; in 13 cases, they are less than ± 40 percent. In some cases, a comparison between different inventories and RAINS calculations shows that differences for the transportation sector are of opposite direction than for total NO_x; i.e., total NO_x for a specific country may have been underestimated whereas transportation emissions for the same country are overestimated, or vice versa.

Since due to the lack of data, no absolute comparison of estimated emissions is possible at a total European level, the relative shares of transportation to total NO_x emissions as calculated in RAINS are indicated for (a) all Europe, (b) OECD-Europe, and (c) non-OECD Europe. This shows that RAINS calculates on the average 54 percent of total NO_x in OECD-Europe (for all 18 European OECD countries) as originating from the transportation sector, which is in good agreement with the 53 percent on average for the 12 countries that supplied detailed data. For non-OECD countries (excluding the USSR), RAINS calculates on the average 16 percent as coming from transportation, which confirms common knowledge that the relative share by traffic to overall NO_x emissions in eastern Europe is significantly lower than in western countries. Relative contributions by traffic as reported to the ECE, range from 5 to 30 percent in eastern Europe.

Table 5. Comparison of national NO_x emissions (as kt NO_2) for Europe in 1980.

Country	*A* *RAINS* */combustion/*	*B* *ECE/EMEP*[1]	*C* *PHOXA*[2]	*D* *OECD*[3] */combustion/*	$(A-B)/B*100$ */%/*	$(A-C)/C*100$ */%/*	$(A-D)/D*100$ */%/*
ALBANIA	28	9[4]			211		
AUSTRIA	186	216		206	14		-10
BELGIUM	426	442	508		4	-16	
BULGARIA	292	150[5]			95		
CSSR	631	1 204	779		-48	-19	
DENMARK	270	251	253	241	8	7	12
FINLAND	244	280		278	-13		-12
FRANCE	1 976	1 867		1 883	6		5
FRG	2 688	3 100	3 117	2 953	-13	-14	-9
GDR	520	800	1 222		-35	-57	
GREECE	218	127[4]			72		
HUNGARY	220	270[5]	199		-19	11	
IRELAND	84	67	99		25	-15	
ITALY	1 452	1 410		1 435	3		1
LUXEMBURG	40	23	35		74	14	
NETHERLANDS	494	535	465	462	-8	6	7
NORWAY	173	215[6]		106	20		63
POLAND	1 484	840[6]	1 705		77	-13	
PORTUGAL	149	166		166	-10		-10
ROMANIA	383	390[4]			-2		
SPAIN	951	780[6]			22		
SWEDEN	297	328		300	-9		-1
SWITZERLAND	161	196		194	-18		-17
TURKEY	356	175[4]			103		
UK	2 454	1 916	2 642	1 867	28	-7	31
USSR[7]	9 082	2 790			226		
YUGOSLAVIA	339	190[4]			78		
EUROPE	25 598	18 737			37		
EUROPE excl. USSR	16 516	15 947			4		
OECD-EUROPE	12 958	12 284			5		
NON OECD-EUROPE	12 640	6 453			96		
NON OECD-EUROPE excl. USSR	3 558	3 663			3		

Notes and references:

1) United Nations, 1987.

2) Bakkum and Veldt, 1986b.

3) Lübkert and de Tilly, 1987b.

4) Semb and Amble, 1981.

5) Year of the inventory is 1984.

6) Year of the inventory is 1985.

7) Two-thirds of total energy are assumed to be used in the European part of the USSR.

Table 6. Comparison of national NO_x emissions from the transportation sector (as kt NO_2) for Europe in 1980.

Country	*A* RAINS	*B* ECE/EMEP1)	*C* PHOXA2)	*D* OECD3)	$(A-B)/B*100$ [%]	$(A-C)/C*100$ [%]	$(A-D)/D*100$ [%]
ALBANIA	9						
AUSTRIA	131	146		146	-10		-10
BELGIUM	210	242	182		-13	15	
BULGARIA	89						
CSSR	91	60	132		52	-31	
DENMARK	109	75	83	81	45	31	35
FINLAND	115	150		152	-23		-24
FRANCE	1 105	1021		1 056	8		5
FRG	1 313	1 748	1 465	1 693	-25	-10	-22
GDR	57		232			-75	
GREECE	144	91			58		
HUNGARY	85	81 4)	44		5	93	
IRELAND	54	21	54		157	0	
ITALY	893	679		949	32		-6
LUXEMBURG	16	14	15		14	7	
NETHERLANDS	299	321	246	298	5	22	0
NORWAY	137	185 5)		86	-26		59
POLAND	175		290			-40	
PORTUGAL	107	106		106	1		1
ROMANIA	71						
SPAIN	610	340			79		
SWEDEN	185	199		201	-7		-8
SWITZERLAND	108	138		136	-22		-21
TURKEY	252						
UK	1 015	690	978	703	47	4	44
USSR 6)	4 613	698			561		
YUGOSLAVIA	149						
EUROPE	12 142	47% of total NO_x					
EUROPE excl. USSR	7 529	46% of total NO_x					
OECD-EUROPE	6 952	54% of total NO_x					
NON OECD-EUROPE	5 190	41% of total NO_x					
NON OECD-EUROPE excl. USSR	577	16% of total NO_x					

Notes and references:

1) United Nations, 1987.

2) Bakkum and Veldt, 1986b.

3) Lübkert and de Tilly, 1987b.

4) Year of the inventory is 1984.

5) Year of the inventory is 1985.

6) Two thirds of total energy are assumed to be used in the European part of the USSR.

7. DISCUSSION

Some of the differences in total NO_x emission rates between RAINS and those reported by countries can be explained by the fact that some fuel used in individual countries is neglected because no NO_x emission coefficients for these fuels are included in RAINS. No coefficients were estimated for these fuels because their share was minor in the set of countries used in the regression analysis. Using the RAINS energy balances, relative shares of fuels in each sector have been calculated for all 27 European countries and are presented in *Table 7.* Adding the shares for which emission coefficients have been determined shows that only one percent of total fossil fuels remains unaccounted for in the transportation and power plant sectors, whereas 20 and 23 percent remain unaccounted for in all Europe in industrial and domestic combustion. In individual countries, these percentages may be significantly different.

Table 7. Relative fuel consumption by sector and fuel type in all Europe in 1980[%] (RAINS, 1987).

	Fuels:							
Sector	*HC*	*BC*	*DC*	*GAS*	*LF*	*MD*	*HO*	*OS*
Transportation	1	0	0	0	51	48	0	0
Power Plants	35	19	0	21	0	0	24	1
Domestic	7	6	6	26	0	40	4	11
Industry (incl. conversion)	14	2	15	25	4	14	25	1

LEGEND: HC: hard coal
BC: brown coal
DC: derived coal
LF: light fraction of oil (incl. gasoline)
MD: middle destillate (incl. diesel)
HO: heavy oil
OS: other solid fuels (e.g., wood)

The statistical analysis could theoretically be improved either by increasing the number of observations or by reducing the number of coefficients to be estimated. The number of observations could be increased once data from additional countries become available, [e.g., from continuing work in the international air pollution study CORINAIR (Bouscaren *et al.*, 1987)], or if time series for one or more countries were available. The latter would, however, only be of value if calculations of the time series were independent of each other to avoid co-linearity. Many techniques have been tried in this study to reduce the number of coefficients to be estimated, and it appears that any further aggregation of emission coefficients would contradict current engineering knowledge about how NO_x emission factors vary between sectors and fuels.

An attempt has been made in this study to assess the reliability of calculated emission coefficients. Reported coefficients of determination for each regression and standard errors for each estimated coefficient give an indication of the uncertainty of the coefficients and of the quality of the energy data and emission rates used in the analysis. Several additional sources of uncertainty in estimating total emission rates based on calculation by this method can, however, be identified: (1) energy data from international statistics used in RAINS have certain errors associated with them; (2) fuel use patterns may be different in countries other than those used in the regression analysis; (3) fuel use patterns may change over time; and (4) emission factors may change over time independent of add-on emission control technology. The latter two points, therefore, introduce errors into any extrapolation of data in time, i.e., in forecasting. Point (2) introduces errors into geographical extrapolation of data because (1) some significant amount of fuel may be used in a particular sector by a specific country for which no emission coefficient was determined in the regression analysis; and (2) a weak emission coefficient such as for brown coal in "power plants" may introduce a larger mistake in eastern European countries than in OECD Europe because the amount of brown coal used in most European OECD countries in power stations is relatively small (less than 3 percent in 10 out of the 12 countries used in this study) compared to Eastern Europe (on the average for all Europe 19 percent).

Point (1) introduces errors because there are, in fact, several energy estimates available, and the sensitivity of NO_x calculations to these different energy estimates should be investigated. The uncertainty of computed country NO_x emissions can also be estimated by replacing deterministic emission factors with their frequency distributions. The frequency distributions for the emission coefficients can be derived from (1) the errors of the regression analysis used to compute these coefficients, or (2) the range of literature values.

Lastly, differences in the underlying statistics of various countries could be compared and might be explained; examples are different automobile fleet composition and average speeds, or different burner types in power plants, etc. If such distinct differences can be identified, it might be possible to group countries into classes with similar background statistics, e.g., northwestern Europe, eastern Europe, and southern Europe, and the average emission coefficient currently used in RAINS could be scaled up or down by adding a weighting factor to the equation. For countries in which no specific information is available, weighting factors could then be selected for the fuel- and sector-specific average emission factors just by knowing the group to which the particular country belongs.

8. SUMMARY AND CONCLUSIONS

Analysis of the principles of NO_x formation has shown that an attempt to develop an NO_x emission model for RAINS based on these principles is currently infeasible because of the large number of variables upon which nitrogen oxide formation is dependent and the lack of knowledge about these variables. Even if the necessary detail were available for one particular year, information about the variation over time of these variables is available in neither international nor in national statistics; therefore prediction of NO_x emissions is infeasible.

To date, only few NO_x emission inventory data are available for Europe that are comparable with each other. The OECD together with the European Communities are currently developing such a coherent and verified inventory for 1980 in which countries have estimated their total national SO_2, NO_x and VOC emissions for a relatively large number of emission source categories according to a prescribed method. This has shown that the following emission source categories, identical to the ones currently used in the sulfur emissions submodel of RAINS, are also the most relevant to NO_x emission estimations: (1) transportation, (2) power plants, (3) domestic and (4) industrial combustion.

Analysis of the OECD emission data has further shown that overall NO_x emissions are most closely correlated to fossil fuel consumption rates, and aggregate emission factors available from the literature also show that NO_x emissions are fuel-specific. Energy consumption is, therefore, the only statistical parameter that is applicable to calculate emissions in all four sectors. Furthermore, energy predictions are readily available for Europe in a consistent fashion. Basing NO_x estimations on energy balances and forecasts also has the advantage that identical statistical input information is used for sulfur and nitrogen emission calculations; this is important if RAINS is to predict the overall impact of sulfur and nitrogen deposition on the environment because, even if the absolute emission and deposition values are wrong due to wrong energy forecasts, the relative contributions by the two pollutants would remain unchanged.

In this study, the OECD data, currently available for 12 countries, were used in multiple OLS regression analysis to determine average, aggregate, sector- and fuel-specific emission coefficients for those sector/fuel combinations already used in RAINS for sulfur calculations that are most important in calculating total national NO_x. Estimation of such coefficients via regression analysis has the advantage over the use of average emission factors from the literature in that regression coefficients utilize best knowledge of 1980 emission levels.

Emission coefficients determined in this way were used by RAINS to calculate baseline (i.e., uncontrolled) NO_x emissions for all Europe in 1980. Comparison of the results with reported national totals as well as reported traffic NO_x shows generally good agreement, i.e., within 20 percent. If NO_x emissions calculated by RAINS are compared to EMEP estimates on a total European scale (USSR excluded), RAINS overestimates the EMEP total by only four percent. Relative contributions as calculated by RAINS for the transportation sector also reflect numbers reported by countries, being on the average 54 percent of total NO_x for OECD Europe and 16 percent for non-OECD Europe (excluding the USSR). For several countries and sectors, differences between calculated and reported NO_x emissions are, however, still larger than 20 percent and, therefore, some suggestions have been made as to how to improve the method. These include uncertainty analysis of emission coefficients and energy statistics and a more detailed analysis of the underlying statistics of various countries.

This paper describes how a "top-down" approach was used to extrapolate a consistent set of data from one set of countries to other countries and other years. The NO_x emission model derived in this way also allows for the testing of different future scenarios with and without emission reduction technology.

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ANNEX 1. NO$_x$ EMISSIONS IN OECD EUROPE

NO$_x$ emissions per RAINS source category in 1980 [in kt NO$_2$] (OECDMAP– inventory)

Source Sector	AUT	DNK	FIN	FRA	GER	ITA	NLD	NOR	POR	SWE	SWI	UKD	SUM
Transportation	146.0	81.3	151.6	1 055.7	1 692.8	948.8	298.1	87.6	106.3	201.0	136.0	703.0	5 608.0
Powerplants	20.0	121.1	106.3	280.0	803.0	287.0	79.1	0.0	19.7	10.0	0.0	851.0	2 577.3
Domestic combustion	10.0	22.4	–	213.0	191.6	79.3	36.3	4.7	–	48.0	8.6	65.0	678.8
Industrial combustion(1)	30.0	16.0	11.2	334.4	266.1	190.4	42.3	13.4	13.1	35.8	49.4	258.0	1 260.2
Industrial processes	10.0	1.0	9.2	61.9	140.2	50.0	58.0	10.6	26.6	33.7	–	–	401.2
Other	0.0	1.6	5.2	5.1	0.0	0.0	2.9	0.1	0.0	0.0	0.0	47.0	61.9
Total(2)	216.0	243.4	283.6	1 950.1	3 093.6	1 555.5	516.6	116.4	165.6	328.5	194.0	1 924.0	10 587.4

Notes:

(1) Including all indirect process heating (e.g. boilers, furnaces, etc.).

(2) Totals may be incomplete due to unavailable data for certain source categories.