Uncertainties in emission inventories

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Stellingen

1. Bouwers van atmosferisch-chemische modellen moeten aangeven in hoeverre de inaccuraatheid van de emissie inventarisatie terug te vinden is in resultaten van hun modellen. *(dit proefschrift)*

2. De door het IPCC voorgestelde methoden voor het kwantificeren van onzekerheden in nationale schattingen van broeikasgasemissies gaan voorbij aan het feit dat naast emissie factoren en activiteiten data, ook de structuur van de inventarisatie een bron van onzekerheid is. *(dit proefschrift)*

3. Twijfel aan de mogelijkheid van klimaatverandering is vaker ingegeven door angst voor economische gevolgen van preventie maatregelen dan door een overtuiging gebaseerd op wetenschappelijke feiten.

4. Onzekerheden in emissie inventarisaties kunnen zo groot zijn, dat het in sommige gevallen wetenschappelijk gezien beter is om af te zien van kwantificering van emissies ook al is dit vanuit beleidsoogpunt onacceptabel.

5. Als talenten niet in de basisformatie van een team worden opgenomen maar op zijn best korte invalbeurten krijgen, zullen ze hun toekomst elders zoeken. Dit geldt zowel voor sportclubs als voor universiteiten.

6. Nederland kent een unieke vorm van grensoverschrijdende luchtverontreiniging omdat het wordt toegestaan dat inwoners uit buurlanden stankoverlast veroorzaken in Nederland door het roken van wiet in koffieshops, het openbaar vervoer en op straat.

7. De honkbalsport is een goede training voor het werken in instituten en organisaties. Het team moet vaak lijdzaam afwachten welke klappen het moet verwerken wanneer de speler met de grootste verantwoordelijkheid niet in staat is om een goed gericht balletje op te gooien.

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Het schrijven van deze tekst betekent het einde van een periode in mijn leven van zo'n 4,5 jaar. De periode dat ik onderzoek heb mogen doen naar onzekerheden in emissie inventarisaties is een leerzame en mooie tijd geweest. Het is een goede mix geweest van leren, lesgeven, reizen, hard werken en plezier. Het is niet altijd even makkelijk geweest en het is dan ook jammer dat mijn vader, oma Van Rey, oma Rutten en Emily (Pietje) dit proefschrift niet zullen zien. Graag wil ik mijn familie en vrienden bedanken voor hun steun en begrip gedurende de afgelopen tijd.

Lieve Esther, de afgelopen jaren stonden in het teken van analyse van onzekerheden. Echter, de enige zekerheid die voor mij belangrijk was om te mogen vinden was jouw liefde en steun.

Table of contents

Chapter 1

Introduction

1.1 Emission of air pollutants

The emission of air pollutants are characterised by four dimensions: (i) variety of emitting sources, (ii) variability in time, (iii) variability in space and (iv) their environmental impact.

(i) Variety of emitting sources. Air pollutants are emitted into the atmosphere as a result of different processes. These processes can be divided into anthropogenic activities and natural processes while also a distinction between biogenic and abiogenic processes can be made. **Anthropogenic** (human) activities lead to the emission of a variety of compounds to the atmosphere. An illustration of different anthropogenic activities and their importance for emissions of –selected- air pollutants is presented in Table 1.1. The table shows the contribution of different anthropogenic activities such as energy use, industrial processes, agricultural practices, biomass burning or waste handling to the emissions of a selection of important air pollutants. As presented in the table, some air pollutants are mainly the result of energy related processes, while other emissions are mainly resulting from agricultural practices. For example carbon dioxide (CO_2), sulphur dioxide (SO_2) or nitrogen oxides (NO_x) emissions result mainly from the combustion of fossil fuels for electricity generation, heat production, or in the transport sector. Methane (CH_4), nitrous oxide (N_2O) and ammonia (NH_3) are to a large degree the result of agricultural practices such as rice cultivation (CH_4) or animal production (N_2O and NH_3). Non methane volatile organic compounds are to a large degree emitted as the result of industrial processes. Beside anthropogenic activities, **natural** processes also result to the emission of a variety of air pollutants. Examples of natural emissions are SO_2 from volcanoes, and NO_x and N_2O formation during lightning. It is important to realise that the distinction between anthropogenic and natural emissions is sometimes not clear. The emission of carbon monoxide (CO), non-methane volatile organic compounds (NMVOC) and other pollutants are amongst other sources caused by the burning of savannahs and forests. The burning of savannahs and forest is both induced by human (slash-and burn farming, accidents) or natural activities (lightning). Besides anthropogenic or natural processes, emissions can vary between abiogenic and biogenic origin. For example, emissions of carbon dioxide and other compounds resulting from the burning of fossil fuels are **abiogenic** emissions while emissions of methane from rice cultivation are **biogenic** emissions.

Table 1.1 Overview of anthropogenic sources of atmospheric compounds and their estimated related contribution to worldwide emissions in 1990. (Van Aardenne et al., 2001). The information presented in Table 1.1 is based on 1990 emission numbers from the Emission Database for Global Atmospheric Research (EDGAR) (Olivier et al., 1999a,b).

Categories	Importance of category for emission of compound							
	CO_2	**CO**	**CH_4**	**NMVOC**	**SO_2**	**N_2O**	**NO_x**	**NH_3**
Energy								
Fossil fuel combustion	++	+	--	+	++	-	++	--
Fossil fuel production	--	o	+	+	o	o	o	o
Biofuel combustion	(+)	+	-	+	--	--	-	-
Industry								
Industrial processes	--	--	--	++	+	+	-	--
Agriculture								
Agricultural land	o	o	+	o	o	++	+	+
Animals	o	o	+	o	o	++	o	++
Biomass burning								
Savannah burning	o	+	--	-	o	--	-	-
Deforestation	-	+	--	--	o	--	-	-
Waste								
Agricultural waste burning	o	+	--	-	--	--	-	-
Landfills	o	o	+	o	o	o	o	-

contribution: >30% of total [++], 10-30% of total [+], 5-<10% of total [-], <5% of total [--],
not included, no source [o]

(ii) Variability in time. Both anthropogenic and natural emissions of air pollutants are usually variable in time. For instance, emissions from the combustion of fossil fuels (e.g. CO_2) are larger in the winter than in the summer because of the increased burning of fossil fuels for domestic heating purposes in the winter period. Variability in time may be **intra-annual**, **seasonal**, **diurnal** or even **within-hour**. Also natural emissions show this temporal (seasonal and diurnal variation); for example NMVOC emissions from forests or SO_2 emissions from volcanoes.

(iii) Variability in space. Emissions to the atmosphere may also show spatial variability. Emissions of N_2O or NH_3 from the application of manure on agricultural fields are being emitted from agricultural areas (**area** source), whereas the largest part of NO_x emissions resulting from fossil fuel combustion in the transport sector are being emitted by **mobile** sources in urban areas. **Point** source emissions (e.g. stacks) are not variable in space, unless the location of the stack is being changed. Also natural emissions show this spatial variability; for example, NO_x emissions by lightning are highly variable.

(iv)Environmental impact. Because of their chemical and/or physical properties, the emissions of air pollutants result in different environmental problems. The effects of air pollutants occur on different spatial and temporal scales. A **global** problem, occurring on time scales of decades to centuries is caused by the emissions of CO_2, CH_4, N_2O and other so-called greenhouse gases. The increased concentration of these gases in the atmosphere enhance the natural greenhouse effect, which is expected to result in global climate changes. On a **regional** scale, the emissions of SO_2, NO_x and NH_3 cause acidification and eutrophication.

After atmospheric transport and chemical transformation, oxidised sulphur and nitrogen are deposited on soils and waters, leading to acidification and eutrophication of the terrestrial and aquatic environment. These problems occur on time scales of weeks to years. An example of a **local** air pollutant problem is urban ozone pollution which is a result of emissions of NO_x and CO. Typical timescales for local air pollution problems are hours to days.

1.2 Emission inventories

To study the environmental impact of emissions of air a pollutant to the atmosphere information is needed on why, where and when air pollutants are being emitted. Therefore, an assessment has to be made of the quantity of a certain compound that is emitted at a certain geographic location at a certain time caused by a specific activity. This assessment is performed through the compilation of emission inventories. Emission inventory calculation is based on various approaches, for instance, (1) direct monitoring at individual sources, (2) emission factor approaches using emission factors from measurements, extracted from literature or calculated based on mass balance approaches or (3) process based models.

Based on Pulles and Builtjes (1998) we define emission inventory compilation as:
The calculation – using a certain methodology and reporting in a specific format – of a collection of numbers representing the quantity of a certain pollutant being emitted to the atmosphere, caused by an economic, social or natural activity, emitted at a certain geographic location at a given time (-in the past, present or future).

Emission inventories can be used for either policy or scientific purpose and a variety of emission inventories exist, each with different characteristics (Pacyna and Graedel, 1995).

For **policy purposes**, emission inventories can be used to monitor the progress of environmental policy by revealing trends in emissions over time. Furthermore, emission inventories can be used for checking compliance to national and international conventions and protocols, or national and international emission targets. In order to monitor the progress of environmental policy in the Netherlands, the national Pollutant Emission Register (PER: Koch et al., 2001) presents emissions such as greenhouse gases, ozone depleting substances and acidifying compounds. The data information from PER is used in the Dutch Environmental Outlook which describes the expected developments in the environmental quality in The Netherlands over the period 2000-2030 (RIVM, 2001a) or the Environmental Balance (RIVM, 2001b) which describes the state of the environment in The Netherlands and the effectiveness of the Dutch environmental policies. Countries that signed the United Nations Framework Convention on Climate Change (UNFCCC) are obliged to report their national emissions of greenhouse gases to the climate secretariat. The National Communication of the Netherlands (VROM, 1998) is an example of such a report. In relation to the Convention on Long Range Transboundary Air Pollution (CLRTAP), the CORINAIR 1994 inventory presents officially reported national emission estimates for acidifying compounds and ozone precursors for 20 European countries (ETCAE, 1997).

For **scientific purposes**, emission inventories are used as input into atmospheric dispersion models that aim at understanding the chemical and physical processes and the behaviour of air pollutants in the atmosphere. A global emission inventory that is developed for this purpose is the Global Emission Inventory Activity (GEIA) (Benkovitz et al., 1996). The data included in the GEIA inventories is taken from published emission inventories. A large part of the GEIA data can also be found in the Emission Database for Global Atmospheric Research (EDGAR) (Olivier et al., 1999a) in which also regional and national emission inventories are available. On the continental scale the LOTOS model (Builtjes, 1992) uses emission inventories for Europe that have been constructed on the base of national emission estimates as part of the EMEP/CORINAIR activity (McInnes, 1996). The LOTOS emission inventory (Builtjes, 1992) for example consists of the CORINAIR 1994 inventory, supplemented with data for countries not included in the 1994 CORINAIR database and a spatial desegregation of the data onto a 0.5^0 x 1^0 grid.

Although the purpose of applying emission inventories might be different (policy vs. science), a strict separation between policy and scientific oriented emission inventories is not always possible. For example, the CORINAIR 1994 emission inventory (policy) is used as basis for the LOTOS emission inventory (science).

An overview of typical characteristics of emission inventories is shown in Table 1.2. The table presents information on characteristics of emission inventories that are described above, as gathered by Ignaciuk et al. (in press). These characteristics are related to compounds and processes included, their temporal and spatial scale, the methodology used for construction of the emission inventory and its intended purpose. As shown in the table, most of these inventories consist of a variety of compounds and emission sources included. The inventories that are used for policy purposes (EDGAR, CORINAIR 1994, National communications and PER) use an annual temporal scale while a typical scientific emission inventory such as GEIA provides emission information on different temporal scales. Inventories used for policy purposes use as temporal scale the border of countries (National Communications) or region (CORINAIR 1994), while inventories applied in scientific studies also use geographical grid cells (EDGAR, GEIA). The methodology used to construct the emission inventories is for typical policy inventories often based on instructions or default methodologies that are based on an emission factor approach (CORINAIR 1994, or National Communications). Other inventories used a mixture of different methodologies that can applied (e.g. emission factor approach (PER), process based models (EDGAR).

Table 1.2 Characteristics of a selection of emission inventories.

	EDGAR 2.0 [a]	**CORINAIR 1994**[a]	**National communications to UNFCCC** [a]	**PER** *	**GEIA**[b]
Compounds included	CO_2, CH_4, N_2O, Fluorinated compounds, SO_2, NO_x, CO, NMVOC, NH_3	SO_2, NO_x, CO, NH_3, CH_4, NMVOC, N_2O, CO_2, 9 heavy metals, 10 POPs	CO_2, CH_4, N_2O, HFCs, PFCs, SF_6, NMVOC, NO_x, CO, SO_2	CO_2, CH_4, N_2O, CF_s, SF_6, SO_2, NO_x, NH_3, and many others (~170)	CO_2, CO, black carbon, CH_4, VOC, N_2O, NO_x, NH_3, SO_2, CFCs, reactive chlorine
Emission source included	25 anthropogenic and 4 natural source categories	10 Anthropogenic and 1 natural source categories	5 major sectors subdivided into sub sectors	8 sectors	Anthropogenic and natural sources
Temporal scale	Annual totals, *1890 – 1995*	Annual totals, *1994*	Annual totals, *1990 – 2010*	Annual totals, *1995 – present*	Annual, partly seasonal or monthly, 1990
Spatial scale	World, countries, 1x1 grid	Europe, countries	Countries	The Netherlands, provinces	World, 1x1 grid
Methodology	Combination of emission factor approach, regression analysis, and process based models	Emission factor approach following EMEP/ CORINAIR Guidebook	Partly IPCC/partly national method	Emission factor approach combined with statistical analysis	Selection of published inventories for specific pollutants and source categories
Purpose	Primarily scientific	Policy	Policy	Policy	Scientific

[a] Modified from Ignaciuk et al. (in press), [b] Benkovitz et al. (1996), * Pollutant Emission Register

1.3 Uncertainty and emission inventories

It is practically not possible to measure emissions at each individual source. Therefore, emissions need to be quantified in another way. In many studies, the "emission factor approach" has been adopted to quantify emissions of air pollutants at higher aggregation levels than the individual source of emissions. This approach aggregates information on sources, in time and space. Consequently, the resulting emission estimate is inevitably an inaccurate representation of the emission that has actually occurred. This inaccuracy has consequences for the proper use of emission inventories for policy or scientific purposes. Analogue to Gardners (1990) discussion on environmental modelling, we may argue that the usefulness of any emission inventory depends on its accuracy and reliability. There is uncertainty about the emission inventory when the accuracy and reliability of the emission estimates are not known. In order to make an assessment of uncertainty in emission inventories both identification, qualification and quantification of the different sources of uncertainty is needed.

The emission inventory community has seen a shift from not considering uncertainty analysis towards uncertainty analysis being a focal point of attention. This is illustrated by the activities of the Intergovernmental Panel on Climate Change (IPCC) in which uncertainty has become a key issue in the construction of national emission inventories that report emissions to the Climate

Convention (Subak, 1999; IPCC/OECD/IEA, 2000). Before this, as argued by Benkovitz (1999), little attention has been paid to the assessment of uncertainty in emission inventories. Furthermore, Rypdal and Winiwarter (2001) argue that at this moment there is little experience in assessing inventory uncertainties. One of the first studies on assessment of uncertainty in emission inventories is to our knowledge the work performed by Benkovitz and Oden (1985). They used a statistical approach (error propagation) to quantify the uncertainty in emission estimates of NO_x, SO_2 and VOC in the NAPAP emission inventories. The work by Egglestone (1988) on accuracy of the United Kingdom annual emission inventory of CO, NO_x, SO_2 and VOC is another early example of studies on uncertainty in emission inventories. A large activity on the analysis of the accuracy of emission inventories is the work done by the European Task Force on Emission Inventories (McInnnes, 1996) together with United States Environmental Protection Agency (EPA, 1996a). They discuss the concept of verification of emission inventories and a variety of methods that can be used for that purpose. Other work on uncertainty analysis has been performed by U.S. EPA in the Emission Inventory Improvement Program (EPA, 1996b). Based on this we observe that different methods for the assessment of uncertainties in emission inventories have been proposed. However, no systematic approach that distinguishes between different sources of uncertainty and - even more important-quantification of the resulting uncertainty in the emission estimate have been finalised/developed yet.

To identify different sources of uncertainty, an overview of the potential sources of uncertainty is needed. Although a variety of sources of uncertainty has been mentioned in inventory studies no extensive distinction between different sources of uncertainty has been proposed yet. In the field of environmental modelling several classifications have been proposed which could be useful for application of emission inventory studies. Interesting classifications have been proposed by Morgan and Henrion (1990), Van der Sluijs (1997) and Van Asselt (2000). Morgan and Henrion (1990) distinguish between (i) uncertainty about **quantity** caused by incomplete information and variability, (ii) structural uncertainty about **model form** and (iii) uncertainty by **simplification** and approximation caused by disagreement among experts, about quantity or form. Van der Sluijs (1997) proposes a classification of uncertainty in (i) uncertainty in **input data** arising from the quality or appropriateness of the data used as inputs to model; (ii) uncertainty in both conceptual and technical **model structure**, with conceptual uncertainty arising from the lack of understanding of the modelled system and technical uncertainty arising from simplification and errors in software and hardware; (iii) uncertainty about **model completeness** caused by all omissions due to lack of knowledge. Finally, Van Asselt (2000) proposed a classification of the modellers' view on uncertainty. This classification consists of (i) technical uncertainty meaning uncertainty in model quantities due to uncertainties in **input data** and **parameter** uncertainties; (ii) methodological uncertainty meaning uncertainty about **model form** due to uncertain equations and model structure uncertainties and (iii) epistemological uncertainties meaning uncertainty about **model completeness** due to uncertain levels of confidence or uncertainty about model validity. A systematic approach to classify uncertainty does not exist for emission inventories at this moment. Such a classification of uncertainty would help the emission inventory community to identify potential causes of uncertainty in emission inventories.

Several methods for qualitative or quantitative uncertainty assessment have been used or proposed, each of them with their specific advantages and disadvantages. In the IPCC Guidelines for instance, a method for uncertainty analysis is proposed that is based on error propagation (TIER 1) or Monte Carlo analysis (TIER 2). Both approaches result in an estimate of the uncertainty, however, they do not allow for actual assessment of accuracy (verification). The Emission Inventory Improvement Program (EPA, 1996b) proposes methods with as aim to reduce the uncertainty rather than an assessment of the accurate emission value. As preferred method for EIIP activities, EIIP proposes to use the Data Attribute Rating System (DARS). The basis for DARS is the assignment of numerical scores to emission factors and activity data to provide an overall confidence rating. Although numerical scores are used, DARS does not result in a quantitative assessment of uncertainty. Other studies like McInnes (1996) or (1996a) propose to use ground truth verification with as basis the comparison between the emission inventory and some other known quantity directly or indirectly related to the emission sources such as field measurement or results from atmospheric dispersion modelling.

Based on the above, we can state that proper use of emission inventories for policy or scientific purposes requires an assessment of the uncertainties in emission estimates. This requires both identification of the different sources of uncertainty and the assessment (qualitative or quantitative) of the uncertainties in emission estimates. A systematic approach to classify uncertainty does not exist for emission inventories at this moment and although different methods for the assessment of uncertainty in emission inventories have been proposed no systematic approach for both quantification of uncertainty exists. Therefore, we define the following research objectives.

1.4 Research objectives

The objective of this thesis is to develop a systematic approach for the assessment of uncertainty in emission inventories of air pollutants. The focus will be on large-scale emission inventories such as national inventories or international emission databases. There is uncertainty about the emission inventory when the accuracy and reliability of the emission estimates are not known. In order to make an assessment of uncertainty in emission inventories both identification, qualification and quantification of the different sources of uncertainty is needed. Therefore, the procedure for uncertainty assessment should allow for the identification of the sources of inaccuracy as well as a qualitative or quantitative assessment of the accuracy of an emission inventory. In order to meet the objective of this thesis we have identified the following three research questions.

I. What are the potential sources of uncertainty in emission inventories?

II. Which methods can be followed for assessment of the uncertainty?

III. To what extent can uncertainty in emission inventories be identified, qualified or quantified?

The research questions have been answered as follows.

First, two different types of emission inventories have been constructed to illustrate the methodology of emission inventory compilation and for providing examples of different sources of uncertainty. In Chapter 2 a time series of past worldwide emissions (period 1890 – 1990 is constructed. Emissions estimates for the earlier years are generally regarded as highly uncertain. The second inventory (Chapter 3) projects emissions of NO_x in Asia for the period 1990 –2020. The results are a possible scenario for the future. Information from these two emission inventories, complemented the results of a literature research to develop an overview of the potential sources of uncertainty (research question I, Chapter 4).

Second, based on literature review and own work an overview of promising approaches that can be used to identify, qualify and quantify uncertainty in emission inventories has been made. Based on this overview a systematic approach (framework) for the assessment of uncertainty has been constructed (research question II, Chapter 4). Different tools identified in the framework have been applied in two illustrative case studies to identify the advantages and disadvantages of these different tools. In the first case study an uncertainty assessment has been performed based on the information included in the emission inventory (Chapter 5), while the second case study (Chapter 6) is based on information from other tools for air quality studies (atmospheric dispersion model, field measurements).

Finally, based on a discussion of the information gained from answering questions I and II as described above, conclusions are drawn about the extent to which uncertainty in emissions inventories can be quantified using the framework for uncertainty assessment identified in this study (Chapter 4 and 7).

Chapter 2

Historical emissions

A 1° x 1° resolution data set of historical anthropogenic trace gas emissions for the period 1890 - 1990

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2.1 Introduction

To quantify current developments and assess possible future scenarios, it is essential to try to understand past anthropogenic changes. No doubt, human activities have always modified the natural environment; however, during the past century the intensity and scale of these modifications have increased dramatically. Emissions of greenhouse gases and their precursors have been identified as significant driving forces of global changes (Intergovernmental Panel on Climate Change (IPCC), 1995) that occur on a wide range of spatial and temporal scales and dimensions and often differ among regions. In spite of their importance, there have been few attempts to estimate long-term historical emission time series (especially before 1970), let alone on a relatively detailed sectoral basis and on a high-resolution grid basis. The Carbon Dioxide Information and Analysis Center (CDIAC) presents a good estimate of historical CO_2 emissions from fossil fuel combustion for the period 1950–1990 on a 1° x 1° grid based on United Nations (UN) energy data (Andres et al., 1997) (In a recent publication, Andres et al. (1999) report on an exercise similar to that described in this paper to extend their data sets of energy consumption on country levels to 1751.) Gschwandtner et al. (1985) estimated emissions of sulphur and nitrogen oxides by the United States for the period 1900–1980, partly on a sub national (state) level. In 1996 the Environmental Protection Agency (EPA) (1996c) presented an overview of historical emissions for several greenhouse gases and other pollutants for the United States in the period 1900–1995. Mylona (1996) presented sulphur emissions for several European countries (including Russia and Turkey) for the period 1880–1990. A detailed global study of sulphur emissions from 1850 to 1990 with data per country and for some sectors has been presented by Lefohn et al. (1996, 1999).

The information presented in this paper is to our knowledge the first attempt to construct a global gridded trace gas emission database on a sectoral basis with a time frame of 100 years using a consistent and transparent methodology for a set of trace gases that are relevant for global atmospheric chemistry. However, one should be aware of the limitations of such a large-scale historical emission inventory. Information on activities and emission factors in the past is limited and uncertain and sometimes nonexistent, leading in some cases to scaling back of current activity rates using indicators and the application of global aggregated emission factors.

This study includes emissions of three groups of anthropogenic sources: (1) energy/industry, (2) agriculture/waste, and (3) biomass burning, and builds on knowledge that is currently available on these three source categories. We focused on construction of a detailed sectoral energy/industry data set; in addition, a more aggregated agriculture/waste data set was compiled. For completeness we included a data set on biomass burning, using a simplified, however, transparent method that can be improved when more information becomes available. A further discussion of limitations of this emission inventory can be found in section 2.4.2. Finally, we note that the distinction between anthropogenic and natural sources is not always clear. We have neglected secondary effects of human activities on natural sources such as methane emissions from wetland changes.

2.2 Methodology

This study builds on the data and methodology of the Emission Database for Global Atmospheric Research (EDGAR 2.0) (Olivier et al., 1999a). In EDGAR, emissions are calculated on the basis of information stored in the system: activity data, emission factors, and other explanatory variables. The underlying information is organized by (sub)source category, by country or region, or as gridded maps, for a number of sources by season. The database has been designed in a modular fashion using a so-called process approach (Laan and Bruinsma, 1993). In general, emissions are first calculated on a country basis by multiplying activity levels by compound-specific emission factors. These emission factors define the source strength as emission per unit time and per unit activity of the process. The process approach allows the required level of detail to be included through defining a tree of subprocesses in which emission factors are adopted from the parent process if no factor is explicitly specified at the lower level. This inference of emission factors (either through the process tree or the location tree or, subsequently, from a previous year) and related maps (via the process tree) efficiently and transparently defines emission factors and spatial allocation functions. Using specific definitions of sources and regions as groups of subprocesses and countries, respectively, for each compound, emission tables per region and source type can be generated. In addition, thematic maps on a 1° x 1° grid are used by relating a specific grid map to each subprocess defined as the spatial allocation function to convert per process total country emissions to gridded emissions per process involved.

For EDGAR 2.0, 1990 data on national activities were selected on the basis of generally accepted statistical data assembled by international organizations that have performed consistency checks on the data. Thus activity data have been derived, for example, from the International Energy Agency (IEA, energy data), the UN (industrial production and consumption), and the Food and Agriculture Organization (FAO, agricultural data). For biomass burning, agricultural waste burning, and biogenic land-related sources, gridded data were used as basic activity data. Emission factors are either defined uniformly for all countries, for example, for CO_2, or are evaluated for individual countries or groups of countries (regions). In some cases, such as for road traffic, emission estimates for individual countries were used as well as independently defined activity levels to derive country-specific emission factors. When available, major point sources are included in version 2.0 as distribution parameters by combining these per source categories in so-called thematic maps. A population density map was used as the default when no source-specific map was available. Unless stated otherwise, the population map provided by J. A. Logan (personal communication, 1993) was used as a default when no source-specific map was available or when point source data were only available for a few countries. A more detailed description of the data sources used is given by Olivier et al. (1996,1999a, 1999b).

The emissions of CO_2, CO, CH_4, nonmethane volatile organic compounds (NMVOC), SO_2, NO_x, N_2O, and NH_3 for the period 1890–1990 with 10 year intervals presented in this publication have been computed using an emission factor approach. The activity data were taken from international statistics included in the Hundred Year Database for Integrated Environmental Assessments (1890–1990) (HYDE) (Klein Goldewijk and Battjes, 1997) supplemented with other data and our own estimates. Historical emission factors per process are based on the emission factors for uncontrolled sources in EDGAR 2.0 for 1990. The databases describe anthropogenic source categories such as fossil fuel production and combustion, industrial production, agricultural practices, waste handling, and land use-related activities. An overview of these categories and their importance for present emissions can be found in Table 2.1. Please note that the emission inventories for 1990 in EDGAR 2.0 were compiled using more complete and more detailed source categories (see also section 2.6).

Table 2.1 Overview of anthropogenic source categories distinguished in this study together with an overview of their importance for emissions of gases considered here [a].

	CO_2	CO	CH_4	NMVOC	SO_2	N_2O	NO_x	NH_3
				Energy				
Fossil fuel combustion	>30	10-30	<5	10-30	>30	5-<10	>30	<5
Fossil fuel production	<5	NI/NS	10-30	10-30	NI/NS	NI/NS	NI/NS	NI/NS
Biofuel combustion	10-30	10-30	5-<10	10-30	<5	<5	5-<10	5-<10
				Industry				
Industrial processes	<5	<5	<5	>30	10-30	10-30	5-<10	<5
				Agriculture				
Agricultural land	NI/NS	NI/NS	10-30	NI/NS	NI/NS	>30	10-30	10-30
Animals	NI/NS	NI/NS	10-30	NI/NS	NI/NS	>30	NI/NS	>30
				Biomass burning				
Savannah burning	NI/NS	10-30	<5	5-<10	NI/NS	<5	5-<10	5-<10
Deforestation	5-<10	10-30	<5	<5	NI/NS	<5	5-<10	5-<10
				Waste				
Agricultural waste burning	NI/NS	10-30	<5	5-<10	<5	<5	5-<10	5-<10
Landfills	NI/NS	NI/NS	10-30	NI/NS	NI/NS	NI/NS	NI/NS	5-<10

[a] Importance determination is based on EDGAR 2.0 1990 emission numbers (Olivier et al., 1999a, 1999b). Contribution given as percent of total; NI/NS indicates not included, no source.

2.2.1 Energy use

Within the energy sector, three emission source categories are distinguished (Table 2.1): production and combustion of fossil fuels and burning of biofuels. In the remainder of this paper we define important sources as those that contribute more than 30% to the total emission and significant sources as those that contribute between 10 and 30% to the total emission. Fossil fuel combustion is an important source of CO_2, SO_2, and NO_x and a significant source of CO and NMVOC. Fossil fuel production contributes significantly to CH_4 and NMVOC emissions, and biofuel combustion is a significant source of CO_2 (gross), CO, and NMVOC emissions (see Table 2.1). However, not all sectors contribute to the emission of each compound. For instance, the analysis by Olivier et al. (1999a) showed that the emission factors for combustion differ not only between regions but also between sector and fuel-type combinations. Therefore a further breakdown was made in the fossil fuel combustion sector (i.e., use of coal, oil, and gas for power generation, industry, transport, and residential energy use including services) and the biofuel combustion sector (industrial and residential sectors). In the fuel production sector we distinguish between the production of hard and brown coal, oil, and gas.

Fossil fuel combustion. Fossil fuel combustion activity data were included for three periods (1890–1920, 1930–1960, and 1970–1990). For the years 1970, 1980, and 1990, IEA energy statistics were used that include annual energy statistics on a country level followed by the breakdown into many detailed fuel types and sectors (IEA, 1994). For the years 1930, 1940, 1950, and 1960 an energy consumption data set was constructed on the basis of a study by Darmstadter (1971). Darmstadter provided annual consumption statistics for three main fuel types (solids, liquids, and gas) and totals of electricity and hydroelectricity production by region and for some countries without sectoral information for selected years only; (i.e., 1925, 1929, 1930, 1933, 1937, 1938, 1950, 1953, 1955, 1957, 1960, 1961, 1962, 1963, 1964, and 1965). The amount of fossil fuels (coal, oil, and gas) needed in the power generating sector to meet electricity production was determined by subtracting hydroelectricity production from electricity data taking into account the efficiency in electricity production in the past from Etemad et al. (1991). Darmstadter presented the mixture of coal, oil, and gas (in percentages) in the years 1929 and 1965 that was used to produce electricity. The fuel mix applied in the power generation sector was estimated for each year by linear extrapolation of the fuel mix in 1929 and 1960 as presented by Darmstadter. The amount of coal, oil, and gas used for other purposes than power generation (transport, industry, and residential) was determined by subtracting the amount and type of fuel used for power generation from the total fuel consumption of each fuel type. For 1930 and 1940, no ''year-specific'' data were included in the Darmstadter (1971) study. Linear extrapolation of the 1929 and 1933 values yielded the data for 1930, and likewise, 1940 is based on linear extrapolation of 1938 and 1950 values. Detailed sectoral data were available in the IEA data used for the period 1970–1990. The sectoral split of 1970 was scaled back in time for the years 1930, 1940, 1950, and 1960 by using indicators per sector that can be associated with the fuel use in the sector industry (value-added industry), transport (number of vehicles), and residential (gross domestic product (GDP) per capita). The procedure used for this was to divide the consumption of fuel type per sector in 1970 by the indicator value of 1970 for each region and multiply the result by the indicator value for the years 1930, 1940, 1950, and 1960. The indicators from HYDE were used as driving factors. Value-added industry is an indicator for the contribution of the industrial sector to the GDP of a country. The sum of the value added of the economic sectors agriculture, industry, and service together form the total GDP of a country. Value-added industry and GDP per capita are based on World Bank (1993), which presents time series for most of the world from 1970 until 1990. For the period 1890– 1970 the historical estimates from Maddison (1994) were used. The numbers of vehicles were derived from Mitchell (1992, 1993, 1995). For the years 1890, 1900, 1910, and 1920 the energy consumption per fuel, per sector, and per region were scaled back in time by using UN coal, oil, and gas production used by Darmstadter (1971) as an indicator for the fuel consumption. Etemad et al. (1991) also presents fuel production figures, which are comparable with the data included by Darmstadter. Energy consumption per fuel type per sector was scaled by multiplying the average fraction of fuel used in each sector (electricity generation, transport, industry, and residential) in the period 1925–1929 with fossil fuel production in the years 1890–1920. Emissions in the pre-1970 period were calculated by using per sector per fuel type globally uniform emission factors based on emission factors in 1990 in EDGAR 2.0 (Olivier et al., 1999a) in those regions without emission control. We assumed that these values reflect the uncontrolled emission factors of equipment in the pre-1970 period. Fuel- and sector-specific emission factors for CH_4, NO_x, CO, and NMVOC from stationary sources in EDGAR 2.0 were adopted from the Long-Term Ozone Simulation (LOTOS) database developed for European countries as described by Builtjes (1992) and from EPA (1996c) data for the United States. For the rest of the world we assumed that emissions were essentially unabated, and we used emission factors from LOTOS valid for eastern Europe. For road transport in 1990 country-specific emission factors for CH_4, NO_x, CO, NMVOC, and SO_2 were based on IPCC (1994), which are essentially the same as those used by Andres et al. (1996). The factors for N_2O were also from IPCC (1994), except for those for road transport, which were based on Olivier (1993) and De Soete (1993). For more details on the data sources for the 1990 emission factors, see Table 2.2. Since SO_2 emissions from coal largely depend on the fuel sulphur content (for which regional information is available), regional emission factors were used, which represent uncontrolled emission of SO_2. For the period 1970–1990, per sector per fuel/type the non-CO_2 emission factors were interpolated between the 1970 regional emission factors for uncontrolled equipment and the 1990 regional per country values estimated in EDGAR 2.0 as described by Olivier et al. (1999a).

Fossil fuel production. Activity data on the production of hard coal, brown coal, crude oil, and natural gas were included on a country basis using input from Etemad et al. (1991). CH_4 emissions from coal mining were calculated with the emission factors used for the 1990 coal production in EDGAR 2.0, which are based on Smith and Sloss (1992) (see Table 2.2). A distinction between surface and underground mining was made. Since further historical information for the period 1890–1990 is lacking, we assumed a constant ratio between underground and surface mining, with values equal to the ratio per country in the year 1990 in the EDGAR 2.0 data set (Olivier et al., 1999a). CO_2, CH_4, and NMVOC emissions from oil production were calculated with emission factors included in the EDGAR 2.0 data set for 1990. The CO_2 country-specific emission factors were based on estimated emissions from Marland et al. (1994). The aggregated NMVOC emission factors were calculated from emission estimates by Little (1989). CH_4 emissions from natural gas production and transmission are calculated with emission factors taken from Ebert et al. (1993), as applied in EDGAR 2.0.

Table 2.2 Emission factors in rounded figures as used for energy-related emissions 1890-1970[a].

	CO_2	CO	CH_4	NMVOC	SO_2	N_2O	NO_x	NH_3
	kg C/GJ	g C/GJ	g C/GJ	g C/GJ	g S/GJ	g N/GJ	g N/GJ	g N/GJ
				Fossil fuel combustion				
Power plants								
Coal	26	9	1	2	450	0.6	122	...
Oil	19	9	2	3	600	0.4	67	...
Gas	15	9	1	5	10	0.1	46	...
Domestic								
Coal	26	2100	225	200	Region[b]	0.9	25	...
Oil	19	13	8	3	200	0.4	15	...
Gas	15	27	...	10	5	0.1	15	...
Industry								
Coal	26	60	8	20	550	0.9	82	2.5
Oil	19	9	2	2	400	0.4	18	...
Gas	15	13	4	5	10	0.1	34	...
Transport								
Coal	26	64	8	20	450	0.9	82	...
Oil	19	4300	15	1300	100	0.4	183	0.1
Gas	15	...	...	10	10	0.1	...	...
				Fossil fuel production				
Brown coal	...	...	24	...	...	...	...	...
Hard coal - surface	...	...	840	...	...	...	...	...
Hard coal - undergr.	...	...	77	...	...	...	...	...
Oil	Country[c]	...	...	1800	...	...	...	...
Gas	...	...	660	...	...	...	...	...
				Biofuel combustion				
Industry	30	510	8	65	5	0.9	31	45
Domestic	30	2000	300	800	5	1.3	24	45

[a] Resulting aggregated regional emission factors for 1990 presented by Olivier et al. (1999a, 1999b, 2000). Primary data sources for the 1990 emission factors: CO_2 , IPCC (1994) and Andres et al. (1996), also see Marland et al. (1999); N_2O, Bouwman et al. (1995, and references therein) and Olivier (1993); petrol-fuelled cars equipped with catalytic converters, De Soete (1993); NH_3 , Bouwman et al. (1997); SO_2 : J. Berdowski (personal communication,1995)(except Japan: Kato and Akimoto (1992)). Primary data sources for other factors: all sectors road and aircraft, Builtjes (1992, and references therein) (except United States: EPA (1996c)); NO_x in Japan, Kato and Akimoto (1992); and road transport: Samaras and Veldt (1993) (except United States: EPA (1996c)).

[b] Domestic coal (kg SO_2 -S GJ^{-1}): Canada, 550; United States, 350; Latin America, 500; Africa, 300; OECD Europe, 450; eastern Europe, 450; FSU,400; Middle East, 700; India region, 300; China region, 500; east Asia, 350; Oceania, 400; and Japan, 250.

[c] Country-specific emission factors (Olivier et al., 1999b).

Biofuel combustion. Reliable activity data of historical biofuel combustion are not included in HYDE and are rarely found in literature. Even for the present data, the information is very uncertain. Therefore we applied a simple approach for this source (assuming that biofuel use in industrialized countries was not substantially higher in the past century than at present). To estimate emissions from biofuel use in the period 1890–1980, the EDGAR 1990 values were extrapolated back in time by dividing the biofuel use in 1990 per country by the rural population per country in 1990 and multiplying this with the rural population per country for the years 1890–1980.

For most countries the total biomass use per country in 1990 has been taken from Hall et al. (1994), resulting in a global total of 50 EJ for 1990, which is considerably higher than most FAO estimates. A possible explanation for this discrepancy is that FAO statistics are based on market figures, whereas in many countries these may not be representative. Rural population data for the years 1890–1980 were taken from the HYDE database, based on United Nations (UN) (1995), Urquhart and Buckley (1965), U.S. Bureau of Census (1990), Grigg (1997), and our own estimates. The result of scaling biofuel with rural population data was consistent with results obtained in Africa by Marufu et al. (1999). We distinguish between industrial and domestic emissions. The calculation of emissions from biofuel combustion is based on globally uniform emission factors (Table 2.2). Emission factors are taken from EDGAR 2.0: CO, CH_4, and NMVOC from Veldt and Berdowski (1995), N_2O from fuel wood from Smith et al. (1993), and NO_x and NH_3 from LOTOS (Builtjes, 1992). For CO_2, gross emission factors are from IPCC (1994), and the SO_2 emission factors are provided by J. Berdowski (personal communication, 1995).

2.2.2 Industrial processes

The following industrial processes are included in this study: the production of iron, steel, copper, nitric acid, adipic acid, and cement and solvent use. For 1890–1980, production data for iron, steel, copper, and cement by country are taken from the HYDE database. Additional estimates were derived from activity data pertaining to solvents, adipic acid production, and nitric acid production. As shown in Table 2.1, industrial processes are an important source of NMVOC emissions (solvents) and a significant source of SO_2 and N_2O emissions (copper and adipic/nitric acid production, respectively).

Data in HYDE on iron production are taken from Mitchell (1992, 1993, 1995). Mitchell (1992, 1993, 1995) and Organization for Economic Co-operation and Development (OECD) (1972, 1975a, 1975b, 1992) provided data for steel production figures as used in HYDE. Emissions from iron production were calculated using default world emission factors for CO, CH_4 (Builtjes, 1992; Olivier et al., 1999b), and NMVOC (Olivier et al., 1999a). Table 2.3 presents emission factors used for the calculation of industrial emissions. CO and NO_x emissions from steel production are calculated with regional emission factors based on the LOTOS database (Builtjes, 1992), taking into account steel production in different furnace types. NMVOC emissions from steel production were calculated with a global default emission factor from Olivier et al. (1999a). Copper production data in the period 1890–1955 are based on Schmitz (1979), and those for the period 1960–1990 on Metallgesellschaft (1991). SO_2 emission factors for copper production were taken from Olivier et al. (1996, 1999a). Finally, historical data on cement production are based on Woytinski and Woytinski (1953), Marland et al. (1994), and Solomon (1994); the emission factor for CO_2 emissions from cement production is from Marland and Rotty (1984).

Table 2.3 Emission factors in rounded figures as used for industrial emissions.

	CO_2, kt C /kt	**CO, kg C/kt**	**CH_4, kg C/kt**	**NMVOC kg C/kton**	**SO_2, t S/kton**	**N_2O, kg N/kton**	**NO_x, kg N/kton**	**NH_3, kg N/kton**
Iron	...	4	675	100	...	...	...	...
Steel								
OHF	...	21	...	100	...	...	...	...
BOF	...	6	...	...	...	...	30	...
EAF	...	4	...	200	...	...	61	...
Nitric Acid	...	...	...	...	...	.16	...	...
Adipic acid								
Global	...	...	...	...	...	147	...	...
United	...	...	...	...	...	110	...	...
States	...	...	...	...	...	185	...	...
Canada								
Cement	136	...	...	...	...	...	...	...
Solvents	....	....	...	1000000	...	...	...	...

Copper, SO_2 (ton S/kton)

Canada	79	OECD Europe	641	Middle East	1470	East Asia	1060
United States	9	Eastern Europe	1100	India region	470	Oceania	560
Latin America	1060	FSU	1100	China region	960	Japan	11
Africa	910						

Adipic acid and nitric acid. The production of adipic acid became significant after World War II. Scaling the 1990 country data from EDGAR 2.0 to the year 1950 (for which no production was assumed), using population numbers as an indicator yields a first estimate of adipic acid production. Nitric acid is mainly used in fertilizer production. The production of nitric acid has become significant since 1930 (Mitchell, 1992, 1993, 1995). During the construction of EDGAR 2.0, Olivier et al. (1999a) found that these statistics and data provided from industry are inconsistent. Therefore statistics of N fertilizer production FAO (1991) were adopted as a proportionality factor for nitric acid production. FAO presents data for the period 1961–1990; the 1970 value was extrapolated to zero in 1930. The emission factors for N_2O emissions from adipic and nitric acid production were based on Reimer et al. (1992).

Solvent use. Historical data on solvent use are not available. The 1990 data included in EDGAR 2.0 were adopted to estimate the solvent use in the period 1890–1970. The 1990 solvent data from EDGAR 2.0 suggest a relationship between GDP and solvent use. Therefore historical solvent use was scaled to GDP in time. Historical GDP data are presented in HYDE on a regional scale, being used to estimate the regional historic solvent use production by each category. The solvent use by country is based on the 1990 country share to the total regional consumption of solvents.

Table 2.4 Emission factors in rounded figures as used for agricultural emissions.

	CH_4	**N_2O**	**NO_x**	**NH_3**
	ton C/km^2	**kg N /kton fertilizer**	**kg N /kton fertilizer**	**kg N /kton fertilizer**
		Agricultural land		
Rice 1890 - 1970	34	...	...	...
Rice 1970 - 1990	regional [a]	...	...	...
Fertilizer	...	12500	description in text	*Bouwman et al. (1997)*

	CH_4	**N_2O**	**NO_x**	**NH_3**
	kg C/ 1000 head	**kg N /1000 head**	**kg N /1000 head**	**kg N /1000 head**
		Livestock		
Buffaloes	39800	286	...	7160
Camels	460	350	...	870
Poultry	regional [b]	3	...	1480
Goats	regional [b]	57	...	7410
Horses	regional [b]	286	...	7160
Pigs	regional [b]	70	...	3260
Sheep	regional [b]	64	...	820
Non-dairy cattle	regional [b]	regional [c]	...	6410
Dairy cattle	regional [b]	regional [c]	...	14300

	Canada	**United States**	**Latin America**	**Africa**	**OECD Europe**	**Eastern Europe**	**FSU**	**Middle East**	**India region**	**China region**	**East Asia**	**Oceania**	**Japan**
a)						*Rice (1970 - 1990) ton C/km^2*							
Rice	34	34	29	17	34	34	34	34	30	33	30	34	34
b)						*CH_4, kg C / 1000 head*							
Poultry	200	100	90	86	120	100	110	58	74	43	95	110	270
Goats	4070	4070	3920	3910	4190	4330	4520	3980	3870	4010	4070	4080	4070
Horses	16300	16400	16400	15100	18800	24100	15900	17800	14400	15500	16300	16300	16300
Pigs	5360	15300	2970	2500	7640	8180	2570	750	1300	4000	6200	30400	12500
Sheep	6300	6300	4000	3910	6440	6580	6770	3980	3900	4000	4100	4080	6300
Non-dairy cattle	42700	42100	38300	24900	58500	55800	54500	27300	23000	34200	26200	41800	52400
Dairy cattle	51100	95200	39000	25600	72300	53500	54400	29200	24600	43600	72500	44400	103000
c)						*N_2O, kg N / 1000 head*							
Non-dairy cattle	290	290	250	250	290	290	290	250	250	250	250	250	290
Dairy cattle	510	510	380	380	510	510	510	380	510	380	380	510	510

2.2.3 Agriculture (Nonburning)

We distinguish between agricultural emissions from rice cultivation, fertilizer use, and domestic animals. According to Table 2.1 these sectors are important sources of N_2O and NH_3 (fertilizer use and animals) and significant sources of CH_4 (rice). The area under rice cultivation was adopted from the HYDE database, as based on Mitchell *(*1992, 1993, 1995*)*. Data gaps between years were filled by linear extrapolation. For the period 1890–1970 a world default emission factor was applied to calculate CH_4 emissions from rice fields, and for the period 1970–1990, regional emission factors were used. Both global and regional emission factors were adopted from Kreileman and Bouwman (1994).

Fertilizer consumption was taken from FAO statistics (FAO, 1991) for the period 1961–1990. Since chemical fertilizers were not widely used before 1950, the 1965–1961 FAO trend is extrapolated to the year 1950. Emissions of N_2O and NH_3 result from fertilizer use. The global emission factors for N_2O and NH_3 emissions were adopted from Bouwman et al. (1995, 1997). NO_x emissions from soils can be enhanced because of fertilization. We used the 1990 Global Emissions Inventory Activity (GEIA) estimate for above-canopy soil emissions compiled by Yienger and Levy (1995). According to Yienger and Levy, ~2 out of 5.5 Tg N yr˙1 present-day soil emissions result from agriculture and conversion of forests to grasslands. Using fertilizer use as an approximation for the regional development of agriculture, soil-NO_x emissions were scaled with time. Thus, following the estimate of Yienger and Levy, preindustrial soil NO_x emissions were assumed to amount to 2.7 Tg N yr˙1.

Country statistics for livestock are available for the period 1961–1990 from FAO (1991), and regional estimates for the period 1890 – 1960 are presented in the HYDE database, based on Mitchell (1992, 1993, 1995). Animals included in this study are cattle, goats, chicken, pigs, sheep, horses, buffaloes, and camels. To derive country data on animal numbers in the period 1890– 1990, we combined the FAO country figures of 1961 with the regional trends from the HYDE database. Emission factors for CH_4, N_2O, and NH_3 from animal waste are taken from Kreileman and Bouwman (1994) and Bouwman et al. (1995, 1997) and are applied to the whole period (see Table 2.4). Global emission factors for emissions by buffaloes and camels and regional emission factors for other ruminants were taken from Gibbs and Leng (1993).

2.2.4 Biomass burning

In this study we define biomass burning as savannah burning and deforestation, although formally, organic waste burning (municipal, industrial, and agricultural) and residential biofuel burning also contribute. In this study municipal and industrial waste burning are considered to be negligible, and agricultural waste burning and biofuel burning are treated as separate source categories (see sections 2.5 and 2.1). Savannah burning and deforestation are mostly human-induced but can also be caused by lightning. A problem with determining the emissions from biomass burning is the lack of information, especially about the amount of biomass burned, which applies to both recent and historical data. A further complicating factor is that even emission factors may have changed since at present, vegetation burns more frequently than in the past; young vegetation contains more nitrogen than mature vegetation. Both savannah burning and deforestation are large sources for CO, and deforestation is a significant source for CO_2 (Table 2.1). Note that we assume that savannah burning is not a net source of CO_2 since this vegetation regrows on the timescale of a year or so.

Present-day savannah-burning emissions were taken from the compilation by Hao et al. (1990), which was interpolated to a 1° x 1° grid. This compilation was augmented by an estimate for savannah burning in tropical north Australia according to Bouwman et al. (1997). To estimate the human contribution to savannah burning, we assumed that in the tropical wet savannah (arbitrarily defined as that exposed to an annual rainfall amount in excess of 1000 mm yr^{-1}) all biomass burning is induced by human activities (J. Lacaux, personal communication, 1998). For dry savannas we assumed that natural fires lead to half of the biomass-burning emissions.

Deforestation is related to both slash-and-burn agriculture and, especially in more recent years, large-scale logging. This presents the difficulty that whereas the first practice leads to direct trace gas emissions in the respective regions, the latter activity only partly leads to reactive trace gas emissions. In, for example, the Amazon region, deforestation was strongly related to the building of new roads, opening up the primary forest, which led to the migration of slash-and-burn farmers. Therefore we assumed the development of a rural population, for which we have a gridded database available, as an indicator for emissions resulting from deforestation fires. We compared the temporal development of these emissions with deforestation data provided by FAO (1991) and Richards (1990) and infer that as a first approximation, the relation between rural population and deforestation emissions can be used. The above mentioned approach only refers to tropical regions. We have assumed that deforestation in industrialized regions occurred primarily before 1890, and we have neglected for these regions both deforestation and afforestation activities after 1890. Clearly, our simplified approach for tropical as well as for temperate regions should be improved by other studies on global biomass burning, which were not accessible during the construction of our data set. We note, however, that at present, there is no consensus on the net flux of CO_2 from industrialized countries. Since most CO_2 flux models are process-based, one cannot easily derive national total and spatial distributions of large-scale biomass burning related to deforestation. According to Hougthon (1999) the nontropical countries contribute only about 25% to the global total net flux for the past 100 year period. Using an alternative method than the so-called bookkeeping methods used by Houghton and others, DeFries et al. (1999) tried to quantify this integrated net CO_2 flux and found that regional estimates deviate considerably from the other methods, notably in the temperate regions.

Since the amount of biomass burnt at present and the spatial distribution are already highly uncertain, and in addition subject to a large interannual variability, we feel that it is not justified to add too much detail to the database. Therefore, in future work a combination of satellite retrieval and modelling may provide more reliable estimates on the present and past emissions by biomass burning.

2.2.5 Agricultural waste burning and landfills

Agricultural waste burning is a large source of CO emissions and significant for NO_x and NMVOC emissions. Landfills are a significant source of CH_4 (Table 2.1). The amount of arable land is used as a proxy for historical agricultural waste burning and has been derived from the HYDE database and Houghton et al. (1983). Global emission factors were applied to calculate emissions of CH_4, CO, N_2O, NH_3, NMVOC, NO_x, and SO_2 (see Table 2.5). N_2O emission factors were taken from Crutzen and Andreae (1990); the NMVOC, CO, and CH_4 emission factors are from Veldt and Berdowski (1995); the SO_2 and NO_x emission factors are from Andreae (1991); and that for NH_3 is from Bouwman et al. (1997). As a first approximation for landfill emissions, we assumed a linear relationship with urban population numbers (Klein Goldewijk and Battjes, 1997; UN, 1995). Regional emission factors for CH_4 were derived from Subak et al. (1992).

Table 2.5 Emission factors in rounded figures as used for waste-related emissions.

	CO_2 kg C/GJ	CO kg C/GJ	CH_4 kg C/GJ	NMVOC kg C/GJ	SO_2 kg S/GJ	N_2O kg N/GJ	NO_x kg N/GJ	NH_3 kg N/GJ
Agricultural waste burning	...	100000	10000	1600	710	110	2500	1500

Landfills, CH_4 (kg C/GJ)

Canada	31	OECD Europe	13	Middle East	7	East Asia	2
United States	30	Eastern Europe	9	India region	1	Oceania	34
Latin America	5	FSU	8	China region	2	Japan	4
Africa	2						

2.2.6 Distribution of emissions on a 1° x 1° grid

The calculated country-by-country anthropogenic emissions in this study are distributed on a 1° x 1° grid using spatial allocation functions in the form of thematic maps on this grid scale. Grid maps used for distributing the per country emission for 1990 were also applied for the whole 100 year period. Obviously, this is only valid as a proxy since many human activities have spatially shifted in time, sometimes even substantially. However, since accurate historical information is lacking, this approach is a transparent method to estimate the spatial distribution of historical emissions within a country and is also used in other studies (e.g., Andres et al., 1996, 1999). Thus the method can be easily improved as more information becomes available. For fossil fuel distribution and combustion, industrial processes, and landfills the emissions were distributed according to population density (J. Logan, personal communication, 1993). Emissions from fossil fuel production were allocated using point source maps for coal mining and for oil and gas production, respectively (Olivier et al., 1996, 1999a). In the agricultural sector, emissions from fertilizer use were allocated according to an arable land map (Bouwman et al., 1995). Emissions from rice cultivation were distributed using a wetland rice cultivation map at 5° x 5° (Aselman and Crutzen, 1989), while emissions from animals were distributed according to livestock density Lerner et al., 1988). Biomass-burning emissions were allocated to a 1° x 1° grid by a deforestation and savannah-burning map (Hao et al., 1990). Finally, agricultural waste-burning emissions were distributed according to Bouwman et al. (1995).

2.3 Results

By using the activity data and emission factors as described in section 2, emissions were calculated for the period 1890–1990 by country and interpolated to a 1° x 1° longitude/latitude grid. Global emissions for the eight compounds by source category for the years 1890–1990 are presented in Figure 2.1 and Table 2.6. In Figures 2.2a–2.2d we present as an example the temporal and spatial development of NO_x emissions in 1890, 1940, 1970, and 1990. The strong increases in emissions in Europe and North America are clearly visible in the 1890–1970 plots, whereas emissions in, for example, Asia become increasingly important after 1970.

2.3.1 Carbon Dioxide (CO_2)

According to this study the overall global anthropogenic CO_2 emissions have increased from 1.2 in 1890 to 7.9 Pg CO_2-C in 1990, which is a more than six fold increase (Figure 2.1a). The contributions by biofuel and fossil fuel combustion emissions increased by factors of 2 and 13, respectively. It is clear that energy use has been the major contributor to the total anthropogenic carbon dioxide emissions throughout this century (85% in 1890 and 92% in 1990). Within the energy sector the gross share of biofuel combustion has declined from 64% in 1890 to 21% in 1990, (net: from 15 to 3% when assuming 10% unsustainable production), while the contribution of fossil fuel combustion increased conversely. CO_2 emissions caused by deforestation, fossil fuel production, and industrial activity (cement production) also show a relatively large increase, but these sectors contribute little to the total CO_2 emissions. In 1890 the regions with the largest contribution to CO_2 emissions are the United States (22%), OECD Europe (19%), China (16%), Latin America (12%), and India (11%). In 1990 the four main regions are the United States (18%), China (16%), Latin America (12%), and India (11%).

2.3.2 Carbon Monoxide (CO)

Anthropogenic CO emissions have increased by a factor of 3, from 121 Tg C in 1890 to 423 Tg C in 1990. Main source categories in 1890 were agricultural waste burning (46 Tg, 38%), biomass burning (savannah burning and deforestation; 38 Tg, 31%), and biofuel combustion (31 Tg, 26%). Throughout the years these sources have increased by factors of 2, 2, and 3, respectively. Fossil fuel combustion has increased from a small CO source in 1890 to the largest CO emission category in 1980 and 1990 (112 Tg in 1990, 26% of the total anthropogenic CO emissions). CO emissions from industrial activities (iron and steel production) show the largest increase in 100 years (a factor of 62), but this sector does not contribute significantly to the total CO emissions (<4% in 1990). Africa, Latin America, India, and China are the regions with the largest CO emissions over the entire time period, which is remarkably different from the emissions of CO_2. This difference can be explained by the fact that waste and biomass-burning activities (mostly applied in these regions) contribute significantly to total CO emissions, while CO_2 emissions are dominated by fossil fuel combustion.

2.3.3 Methane (CH_4)

Global anthropogenic CH_4 emissions in the period 1890–1990 show a threefold increase, from 71 Tg C in 1890 to 240 Tg in 1990. In 1890 the agricultural emissions were the largest CH_4 source (47 Tg, 66% of the total anthropogenic emissions), with equal contributions by domestic ruminants and agricultural land (anaerobic processes in rice fields). Over the period of study the agricultural sector remained the largest CH_4 source (113 Tg in 1990, 47%). From 1950 onward, livestock was the largest contributor to the sector total. After 1960, fossil fuel production became a significant source (67 Tg in 1990, 28%), mainly caused by leakage during transmission and distribution of natural gas.

Because of the dominance of methane emissions by the agricultural sector, it is no surprise that regions with important agricultural activity are the largest CH_4-emitting regions. In 1890 the regions with the largest contribution to CH_4 emissions were India (36% of total emission), OECD (15%), China (14%), and the United States (11%). India (18%), China (15%), Former Soviet Union (FSU; 15%), the United States (13%), and Latin America (11%) were the major contributors to the global CH_4 emissions in 1990.

2.3.4 Nonmethane Volatile Organic Compounds (NMVOC)

In the period 1890–1990, anthropogenic NMVOC emissions increased by a factor of 7, from 25 Tg NMVOC in 1890 to 181 Tg in 1990 (note that the units are Tg NMVOC). In 1890, biofuel combustion (11 Tg, 44% of total emission) and agricultural waste burning (7 Tg, 28%) accounted for 72% of the total NMVOC emissions. During the period of study there has been a shift from biofuel combustion and agricultural waste burning toward industrial and energy-related processes. In 1990, industrial processes (solvent use and iron and steel production) emit 56 Tg NMVOC, which is 31% of the total emissions. Fossil fuel combustion accounts for 42 Tg (23%). The 32 Tg from biofuel combustion and the 26 Tg from fossil fuel production account for 18 and 14%, respectively. The largest emitters in 1890 were Africa (24%), China (18%), India (16%), and Latin America (15%) related to biofuel, savannah, and agricultural waste burning. Over the period 1890–1990, energy and industrial processes increasingly contributed to NMVOC emissions, reflected in the contribution of OECD (11% in 1990) and the United States (13%) as important emitters in 1990.

2.3.5 Sulphur Dioxide (SO_2)

Anthropogenic SO_2 emissions have increased from 6 Tg S in 1890 to 70 Tg in 1990, an increase of a factor of 12. Throughout the period considered in this study, fossil fuel combustion remained by far the major contributor to SO_2 emissions (81% in 1890 and 81% in 1990). Over the years, emissions from industrial production (copper smelting) became a significant source of SO_2, with an increase from 0.3 Tg in 1890 (5% of the total) to 11 Tg in 1990 (16%). From 1890 to 1940, world SO_2 emissions mainly originate from the regions the United States (~3 Tg in 1890, 43% of total emission) and OECD Europe (~2 Tg in 1890, 28%). From 1940 onward the contribution of the FSU region increased, and from 1960 the emissions from the China region became important.

2.3.6 Nitrogen Oxides (NO_x)

The calculated NO_x emissions show an increase from 1890 to 1990 by a factor of 5 from 7 Tg N in 1890 to 35 Tg in 1990. In 1890, NO_x was mainly emitted from soils and savannah burning; in fact, these sources represent to a large degree the ''natural'' fraction of these emissions. Within only a few decades the emissions from fossil fuel combustion became more important, and from 1930 onward it has been the largest NO_x source, resulting in 60% of the global NO_x emissions in 1990. Africa, Latin America, and the United States together accounted for 60% of the global NO_x

emissions in 1890. Later in the period of study, emissions from Europe, FSU, and China also contributed significantly to the total NO_x emission. In Figures 2.2a–2.2d we present the temporal and spatial development of NO_x emissions in 1890, 1940, 1970, and 1990. In 1890, parts of northern Europe and the east coast of the United States have emissions larger than 1000 mg N m^{-2} yr^{-1}; in the rest of the world, emissions are generally below 300 mg N m^{-2} yr^{-1}. In 1940 the regions of high emissions are still found in northern Europe and the eastern United States. The development and further spread of anthropogenic emissions in the United States and Europe are evident in 1970. In Africa and Latin America, biomass-burning emissions become more important, whereas the first signs of strong economical development in Asia become apparent. The further increase of Asian emissions is visible in 1990.

2.3.7 Nitrous Oxide (N_2O)

Nitrous oxide emissions increased from 0.4 Tg N_2O-N in 1890 to 2.8 Tg in 1990, which is an increase of a factor of 7 (excluding indirect emissions from deforestation). Animals were the main source of N_2O emissions until World War II, ranging from 0.3 Tg (70%) in 1890 to 0.5 Tg (70%) in 1940. In 1950, after the introduction of synthetic fertilizers, N_2O emissions from agricultural land and industry became significant. Regions with the largest N_2O emissions in 1890 were India (34%), the United States (15%), and OECD Europe (11%). In the period between 1890 and 1990, emissions from China and Latin America contributed significantly to the global budget, while the contribution of the Indian region became less dominant (17% in 1990).

2.3.8 Ammonia (NH_3)

Ammonia emissions increased from 9 Tg NH_3-N in 1890 to 43 Tg in 1990 (almost a fivefold increase). Throughout the period of study, NH_3 emissions are dominated by agricultural emissions. Emissions by animals account for 65% of total NH_3 emissions in 1890. In 1990, 79% of the NH_3 emissions originate from the agricultural sector, with 21 Tg from animals (~50%) and 13 Tg from agricultural land (~30%).

Although the hundred year historical emission data set is based on the EDGAR 2.0 methodology, as presented by Olivier et al. (1999a), some differences between our 1990 emission estimates and the 1990 emission estimates in this report are present. The use of aggregated emission factors will lead to some differences in situations where EDGAR 2.0 applied detailed emission factors. Furthermore, the spatial emissions from aircraft were not explicitly considered in this study but were partially included in the total transport sector. These emissions are mainly important for CO_2 (~2–3% of present-day fossil fuel-related emissions) and NO_x (~1.5% of present-day NO_x emissions, mainly emitted at upper tropospheric and lower stratospheric altitudes, where NO_x is very efficient in O_3 production). We recommend future users of this database to include separately the aircraft emissions.

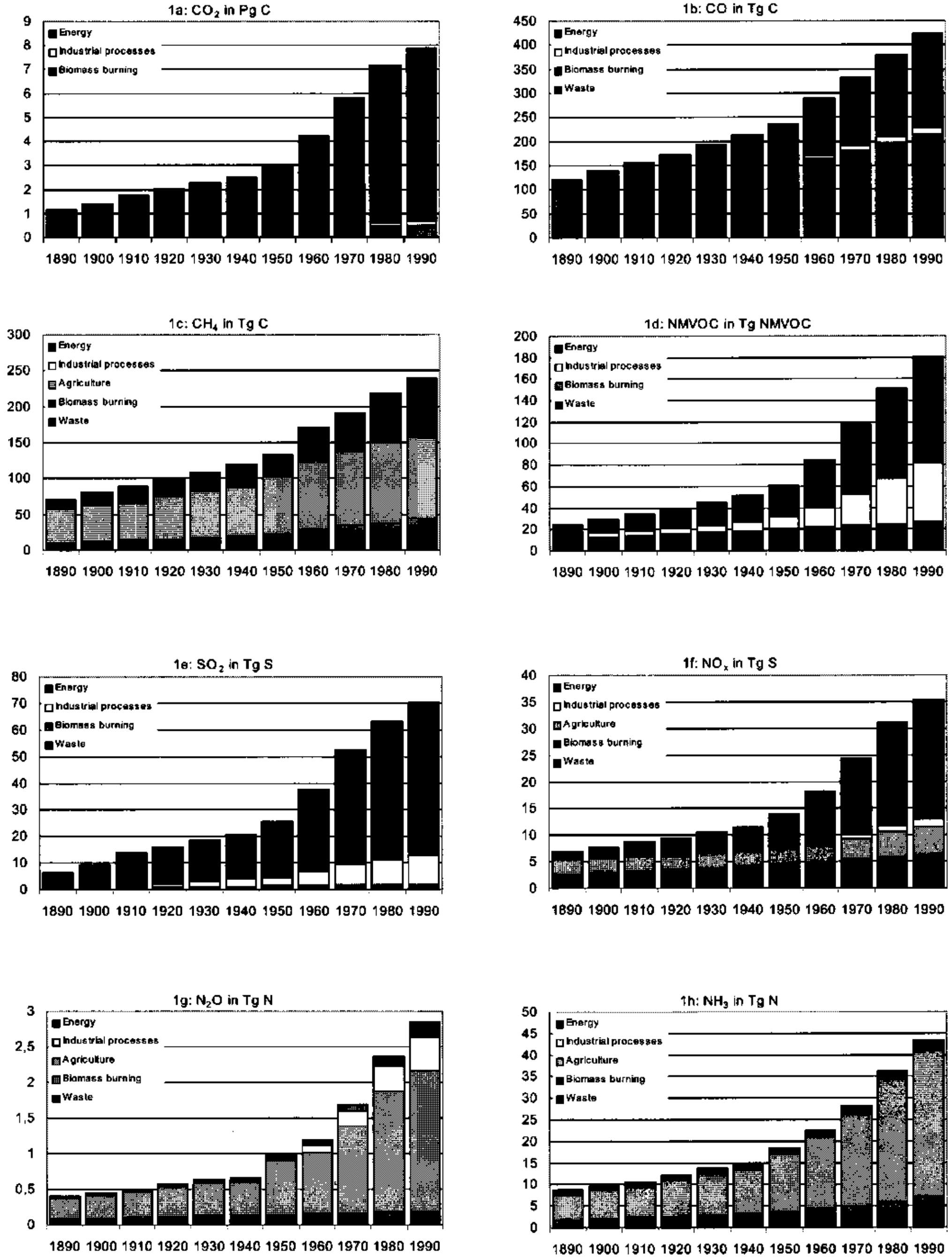

Figure 2.1 Estimated anthropogenic emissions in the period 1890 - 1990: (a) CO_2 emissions in Pg CO_2-C, (b) CO emissions in Tg CO-C, (c) CH_4 emissions in Tg CH_4-C, (d) NMVOC emissions in Tg NMVOC, (e) SO_2 emissions in Tg SO_2-S, (f) NO_x emissions in Tg NO_2-N, (g) N_2O emissions in Tg N_2O-N, and (h) NH_3 emissions in Tg NH_3-N.

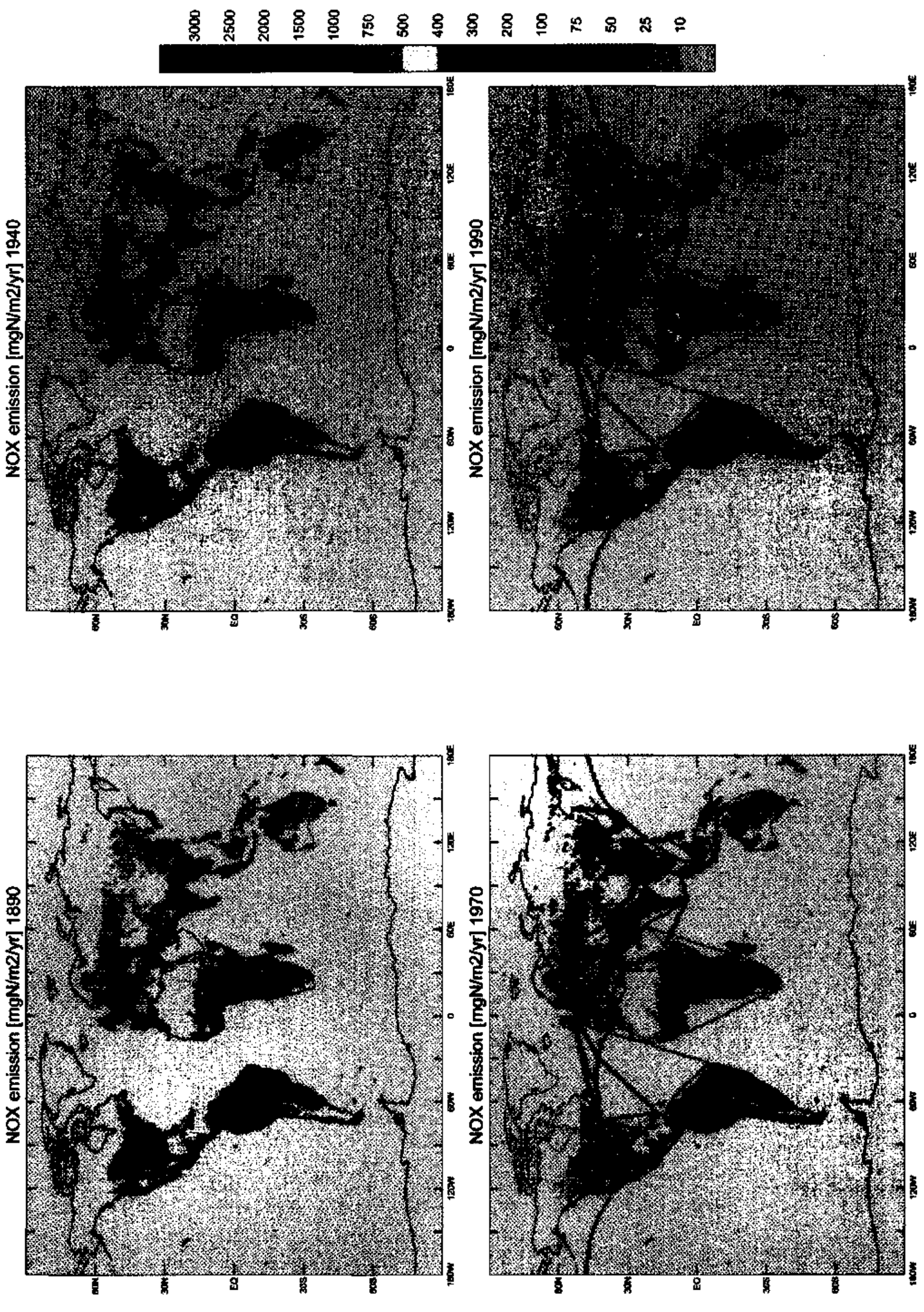

Figure 2.2 Spatial distribution of average NO_x emissions in (a) 1890, (b) 1940, (c) 1970, and (d) 1990. Note that emissions from ships prior to 1970 are represented through continental emissions.

Table 2.6 Anthropogenic emissions by compound and sector for the period 1890 – 1990.

	1890	**1900**	**1910**	**1920**	**1930**	**1940**	**1950**	**1960**	**1970**	**1980**	**1990**
CO_2, Pg C											
Energy	1.0	1.2	1.5	1.7	2.0	2.2	2.7	3.8	5.3	6.6	7.2
Fossil fuel comb.	0.4	0.5	0.8	1.0	1.2	1.3	1.8	2.8	4.1	5.2	5.7
Fossil fuel prod.	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1
Biofuel comb.	0.6	0.7	0.7	0.8	0.8	0.9	0.9	1.0	1.2	1.3	1.5
Industrial processes	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.2
Biomass burning	0.2	0.2	0.2	0.2	0.3	0.3	0.3	0.4	0.4	0.5	0.5
Savannah burn.	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Deforestation	0.2	0.2	0.2	0.2	0.3	0.3	0.3	0.4	0.4	0.5	0.5
Total[a]	**1.2**	**1.4**	**1.8**	**2.0**	**2.3**	**2.5**	**3.0**	**4.2**	**5.8**	**7.2**	**7.9**
CO, Tg C											
Energy	35.9	40.5	46.8	52.9	62.5	70.0	82.6	118.1	143.0	169.1	194.9
Fossil fuel comb.	4.8	7.4	11.3	14.7	21.7	26.4	37.0	65.7	80.6	96.1	112.0
Biofuel comb.	31.0	33.1	35.5	38.1	40.7	43.6	45.6	52.4	62.4	73.0	82.9
Industrial processes	0.2	0.5	1.2	1.4	1.8	2.8	3.6	6.7	10.2	12.6	14.9
Biomass burning	38.4	48.8	54.1	60.4	66.8	72.1	76.2	83.4	94.9	110.6	124.4
Savannah burn.	23.3	31.5	34.1	37.2	40.9	44.3	47.4	50.0	56.2	67.3	76.6
Deforestation	15.0	17.4	20.1	23.2	25.9	27.8	28.8	33.4	38.6	43.3	47.8
Waste	46.0	49.3	53.2	57.1	61.9	67.9	73.7	81.4	85.1	86.9	88.7
Agr. waste burn.	46.0	49.3	53.2	57.1	61.9	67.9	73.7	81.4	85.1	86.9	88.7
Total	**120.5**	**139.2**	**155.3**	**171.7**	**193.0**	**212.8**	**236.2**	**289.6**	**333.1**	**379.3**	**422.9**
CH_4, Tg C											
Energy	13.0	17.3	23.4	24.5	26.4	30.7	31.8	45.7	55.0	68.8	81.7
Fossil fuel comb.	0.5	0.7	1.1	1.3	1.5	1.6	2.0	3.1	3.6	3.2	3.3
Fossil fuel prod.	8.4	12.2	17.6	18.1	19.4	23.2	23.6	35.5	42.8	55.4	66.8
Biofuel comb.	4.1	4.4	4.7	5.1	5.5	5.9	6.2	7.1	8.7	10.2	11.6
Industrial processes	0.0	0.0	0.0	0.1	0.1	0.1	0.1	0.3	0.4	0.5	0.6
Agriculture	46.8	51.5	52.4	60.7	65.0	68.2	77.5	95.3	102.3	111.2	113.1
Agr. land	26.7	28.1	27.9	31.7	32.8	34.5	35.2	43.6	45.0	46.2	44.4
Animals	20.1	23.3	24.5	29.1	32.2	33.7	42.3	51.7	57.2	65.0	68.7
Biomass burning	3.2	3.5	3.9	4.4	4.9	5.2	5.5	6.2	7.1	8.0	9.0
Savannah burn.	1.8	2.0	2.2	2.4	2.6	2.8	2.9	3.3	3.7	4.2	4.8
Deforestation	1.3	1.5	1.8	2.0	2.3	2.4	2.5	2.9	3.4	3.8	4.2
Waste	7.5	8.5	9.6	11.0	12.7	14.9	18.5	22.6	26.8	30.6	35.4
Agr. waste burn.	4.6	4.9	5.3	5.7	6.2	6.8	7.4	8.1	8.5	8.7	8.9
Landfills	2.9	3.5	4.3	5.3	6.5	8.1	11.2	14.4	18.3	21.9	26.5
Total	**70.5**	**80.8**	**89.4**	**100.7**	**109.1**	**119.1**	**133.4**	**170.1**	**191.6**	**219.1**	**239.8**
NMVOC, Tg mass											
Energy	12.2	13.5	15.3	17.6	21.2	24.4	29.8	45.3	66.1	84.2	98.9
Fossil fuel comb.	0.8	1.3	2.0	2.8	4.7	6.1	8.8	17.7	25.4	34.2	41.5
Fossil fuel prod.	0.1	0.2	0.3	0.8	1.5	2.2	4.0	8.1	17.1	22.3	25.8
Biofuel comb.	11.3	12.1	13.0	14.0	15.0	16.1	16.9	19.5	23.6	27.7	31.6
Industrial processes	1.8	3.5	4.5	5.5	6.6	8.2	11.2	17.5	28.7	42.0	55.7
Biomass burning	3.6	5.0	5.8	6.6	7.4	8.0	8.4	9.1	10.4	11.7	12.7
Savannah burn.	1.2	2.2	2.5	2.8	3.2	3.5	3.6	3.7	4.1	4.6	4.9
Deforestation	2.5	2.8	3.3	3.8	4.2	4.6	4.7	5.5	6.3	7.1	7.8
Waste	7.2	7.7	8.3	8.9	9.6	10.6	11.5	12.7	13.3	13.5	13.8
Agr. waste burn.	7.2	7.7	8.3	8.9	9.6	10.6	11.5	12.7	13.3	13.5	13.8
Total	**24.8**	**29.7**	**33.8**	**38.5**	**44.9**	**51.2**	**60.8**	**84.6**	**118.4**	**151.5**	**181.0**

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[a] Gross CO_2 from biofuel combustion, i.e. assuming 100% unsustainable production. In practice, this value may be as low as 10%, which could reduce the net CO_2 emission from biofuels by 90%.

Table 2.6 Continued

	1890	1900	1910	1920	1930	1940	1950	1960	1970	1980	1990
						SO_2, Tg C					
Energy	5.3	7.9	12.0	14.0	15.7	16.6	21.4	30.8	42.8	52.0	57.5
Fossil fuel comb.	5.2	7.8	11.8	13.9	15.6	16.5	21.2	30.6	42.7	51.7	57.2
Biofuel comb.	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.2	0.2	0.2
Industrial processes	0.3	0.5	0.9	1.0	1.8	2.8	3.0	5.2	7.8	9.3	10.8
Biomass burning	0.5	0.6	0.6	0.7	0.7	0.8	0.8	0.9	1.1	1.2	1.4
Savannah burn.	0.4	0.4	0.5	0.5	0.6	0.6	0.6	0.7	0.8	0.9	1.0
Deforestation	0.1	0.1	0.1	0.2	0.2	0.2	0.2	0.2	0.3	0.3	0.4
Waste	0.3	0.4	0.4	0.4	0.4	0.5	0.5	0.6	0.6	0.6	0.6
Agr. waste burn.	0.3	0.4	0.4	0.4	0.4	0.5	0.5	0.6	0.6	0.6	0.6
Total [a]	**6.4**	**9.3**	**13.8**	**16.1**	**18.7**	**20.7**	**25.8**	**37.5**	**52.3**	**63.1**	**70.2**
						NO_x, Tg N					
Energy	1.5	2.0	2.7	3.3	4.1	4.7	6.4	10.0	14.3	19.4	22.5
Fossil fuel comb.	0.9	1.4	2.1	2.6	3.4	3.9	5.6	9.1	13.3	18.2	21.2
Biofuel comb.	0.6	0.6	0.6	0.7	0.7	0.8	0.8	0.9	1.0	1.1	1.3
Industrial processes	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.4	0.7	1.1	1.5
Agriculture	2.7	2.7	2.7	2.7	2.7	2.7	3.0	3.1	3.9	4.7	5.2
Agr. land	2.7	2.7	2.7	2.7	2.7	2.7	3.0	3.1	3.9	4.7	5.2
Biomass burning	1.5	1.7	1.9	2.1	2.3	2.4	2.5	2.9	3.3	3.7	4.2
Savannah burn.	1.2	1.3	1.4	1.5	1.7	1.8	1.9	2.1	2.4	2.7	3.1
Deforestation	0.3	0.4	0.5	0.5	0.6	0.6	0.7	0.8	0.9	1.0	1.1
Waste	1.1	1.2	1.3	1.4	1.5	1.7	1.8	2.0	2.1	2.1	2.2
Agr. waste burn.	1.1	1.2	1.3	1.4	1.5	1.7	1.8	2.0	2.1	2.1	2.2
Total	**6.9**	**7.6**	**8.7**	**9.5**	**10.6**	**11.6**	**13.9**	**18.3**	**24.3**	**31.0**	**35.4**
						N_2O, Tg N					
Energy	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.1	0.1	0.1	0.2
Fossil fuel comb.	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.2
Biofuel comb.	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1
Industrial processes	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.2	0.3	0.5
Agriculture	0.3	0.3	0.4	0.4	0.5	0.5	0.8	0.9	1.2	1.7	2.0
Agr. land	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.4	0.8	1.0
Animals	0.3	0.3	0.4	0.4	0.5	0.5	0.6	0.7	0.8	0.9	1.0
Biomass burning	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Savannah burn.	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1
Deforestation	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Waste	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Agr. waste burn.	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Total	**0.4**	**0.4**	**0.5**	**0.6**	**0.6**	**0.7**	**1.0**	**1.2**	**1.7**	**2.4**	**2.9**
						NH_3, Tg N					
Energy	1.0	1.0	1.1	1.2	1.3	1.3	1.4	1.6	1.8	2.1	2.3
Fossil fuel comb.	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1
Biofuel comb.	1.0	1.0	1.1	1.1	1.2	1.3	1.3	1.5	1.7	2.0	2.2
Industrial processes	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.2	0.2
Agriculture	5.9	6.7	7.1	8.4	9.5	10.0	13.3	16.4	21.1	28.1	33.7
Agr. land	0.3	0.3	0.4	0.5	0.6	0.8	1.8	2.4	4.7	8.8	12.6
Animals	5.6	6.4	6.7	8.0	8.9	9.2	11.5	14.0	16.4	19.3	21.1
Biomass burning	0.9	1.0	1.2	1.4	1.5	1.7	1.8	2.1	2.4	2.8	3.2
Savannah burn.	0.4	0.5	0.6	0.7	0.8	0.8	0.9	1.1	1.3	1.5	1.8
Deforestation	0.4	0.5	0.6	0.7	0.8	0.8	0.8	1.0	1.1	1.3	1.4
Waste	0.9	1.0	1.1	1.2	1.3	1.6	1.9	2.3	2.7	3.3	4.0
Agr. waste burn.	0.7	0.7	0.8	0.9	0.9	1.0	1.1	1.2	1.3	1.3	1.3
Landfills	0.2	0.2	0.3	0.3	0.4	0.5	0.7	1.1	1.4	1.9	2.7
Total	**8.6**	**9.7**	**10.5**	**12.2**	**13.7**	**14.6**	**18.4**	**22.4**	**28.1**	**36.3**	**43.4**

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[b] Direct effects only. Total N_2O emissions from deforestation including direct effects resulting from delayed emissions are estimated at about ten times the level of the direct emissions presented here.

2.4 Discussion

2.4.1 Results compared to other studies

Figure 2.3 presents an overview of CO_2 emissions from fossil fuel combustion and cement production computed in this study and estimates by Keeling (1994) and Marland et al. (1994). For the period 1890–1960 the global emissions calculated by Keeling and from this study are in good agreement (<5% difference). For the years after 1960 the emissions calculated by Marland et al. are somewhat higher than our estimates. This discrepancy can be partly explained from the fact that we excluded emissions from international air transport, which accounts for about 2% in 1990, and we also used different input data sets (United Nations energy data by Marland et al. versus IEA (1994) and Darmstadter (1971) energy data in our study). A detailed study by Marland et al. (1999) showed that for 1990 the data sets showed very little differences on a country level, generally <1%. The smallest countries, for example, in Africa, had the largest differences, but the largest contributions to uncertainties originate from the largest emitting countries, where low relative uncertainties lead to high absolute uncertainties.

Global CH_4 emissions are presented in Table 2.7. The results of this study are in fair agreement with the estimates of Stern and Kaufmann (1995). For the years 1980 and 1990 the estimates of Stern and Kaufmann are higher than in this study. The sector emissions are different in a few cases: our estimates of energy related emissions are 25% or higher throughout the period of study. This difference can be partly explained by the fact that biofuels are included in the energy sector, while they are not included in the Stern and Kaufmann study. Biomass-burning emissions are in good agreement for the period 1890–1960. From 1970 onward our estimate is about 30% lower. Emissions by animals are for most years comparable, with the exception of 1950 and 1960 (our numbers indicate a 10% higher emission) and 1990 for which we calculate a 15% lower emission by domestic ruminant. The emissions from agricultural land (mainly rice cultivation) calculated by Stern and Kaufmann are higher than our estimates with the difference increasing over the years from 20 to 40%. Finally, emissions from landfills are in good agreement.

We compared our SO_2 emissions with global estimates by Stern and Kaufmann (1996), Örn et al. (1996), Lefohn et al. (1996, 1999), Möller (1984) and Ryaboshapko (1983). The results are presented in Figure 2.4. Figure 2.4 illustrates the apparently large uncertainty in the historical emission estimates. Furthermore, our global estimates are in the lower part of the range compared to previous studies. However, when looking at two regional studies, European SO_2 emissions according to Mylona (1996) and U.S. emissions from Gschwandtner et al. (1985), as presented in Figure 2.5, our emission estimates and trends are in rather close agreement. The year 1990 is an exception for which our estimates for the United States are significantly higher than the estimate by Gschwandtner et al., which is probably caused by differences in the assumed effectiveness of emission abatement. Note that Örn et al. use the Mylona and Gschwandtner et al. estimates for Europe and the United States, respectively. This indicates that regions other than Europe and the United States primarily cause the difference between our estimates and the other estimates. Discrepancies in emission estimates in these regions are probably caused by uncertainties in activity data and emission factors.

In Figure 2.6, global NO_x emissions for the years 1960, 1970, and 1980 are compared with Hameed and Dignon (1988). Regional estimates for the United States (Gschwandtner et al., 1985) are compared with our NO_x calculations for this region. Our global estimates for the years 1960, 1970, and 1980 are substantially lower, which might be related to differences in the methodology. Compared to the study by Gschwandtner et al. (1985), our estimates of the U.S. NO_x emissions are in close agreement for the period 1900–1960. For the period 1970–1990 our estimates are slightly different. Comparison of our gridded historical emissions of N_2O, NH_3, and CO prior to 1970 was not possible because of a lack of comparable data sets. For comparison of the 1990 data we refer to Bouwman et al. (1995, 1997) and Olivier et al. (1996, 1999b).

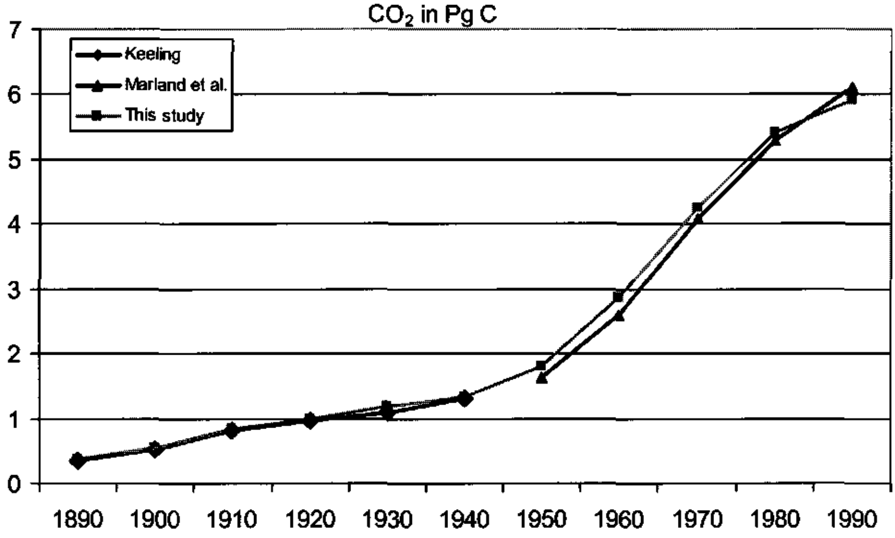

Figure 2.3. Fossil fuel and industrial CO_2 emissions from this study compared with Keeling (1994) and Marland et al. (1994)

Table 2.7 CH_4 emission estimates from this study compared with Stern and Kaufman (1995)[a].

		Energy Tg C	Biomass burning Tg C	Animals Tg C	Rice Tg C	Landfills Tg C	Total Tg C
1890	Stern and Kaufmann	6	10	23	33	3	74
	This study	13	8	20	27	3	71
1900	Stern and Kaufmann	9	10	25	35	3	82
	This study	17	8	23	28	4	81
1910	Stern and Kaufmann	14	12	27	37	4	94
	This study	23	9	24	28	4	89
1920	Stern and Kaufmann	16	11	29	38	5	99
	This study	24	10	29	32	5	101
1930	Stern and Kaufmann	17	12	32	40	7	109
	This study	26	11	32	33	7	109
1940	Stern and Kaufmann	20	12	35	43	9	118
	This study	31	12	34	34	8	119
1950	Stern and Kaufmann	23	13	38	45	11	130
	This study	32	13	42	35	11	133
1960	Stern and Kaufmann	33	18	46	51	15	162
	This study	46	14	52	44	14	170
1970	Stern and Kaufmann	48	22	56	59	19	204
	This study	55	16	57	45	18	191
1980	Stern and Kaufmann	56	23	67	67	24	238
	This study	69	17	65	46	22	219
1990	Stern and Kaufmann	61	29	80	75	31	275
	This study	82	18	69	44	26	239

[a] Biomass includes waste burning

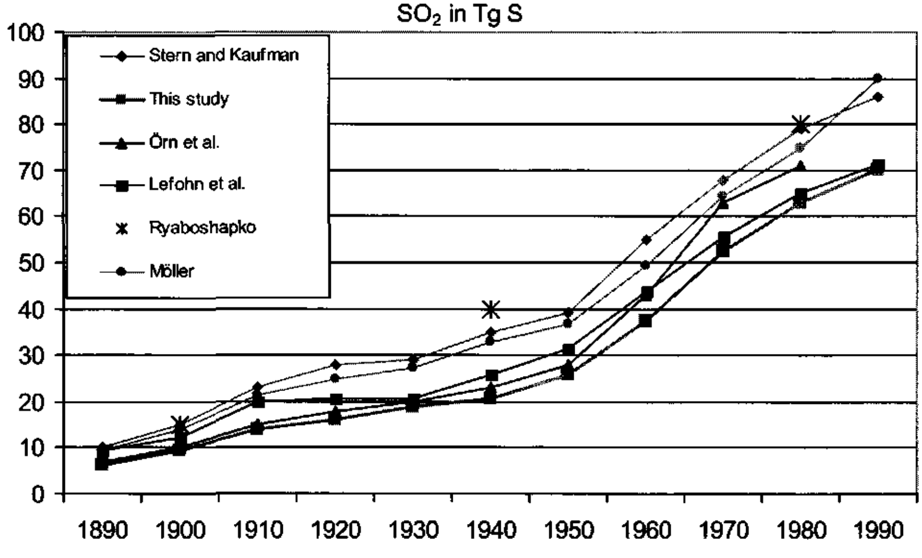

Figure 2.4 Comparing our estimated global SO_2 emissions with Stern and Kaufmann (1995), Örn et al. (1996), Lefohn et al. (1996), Ryaboshapko (1983), and Möller (1984)

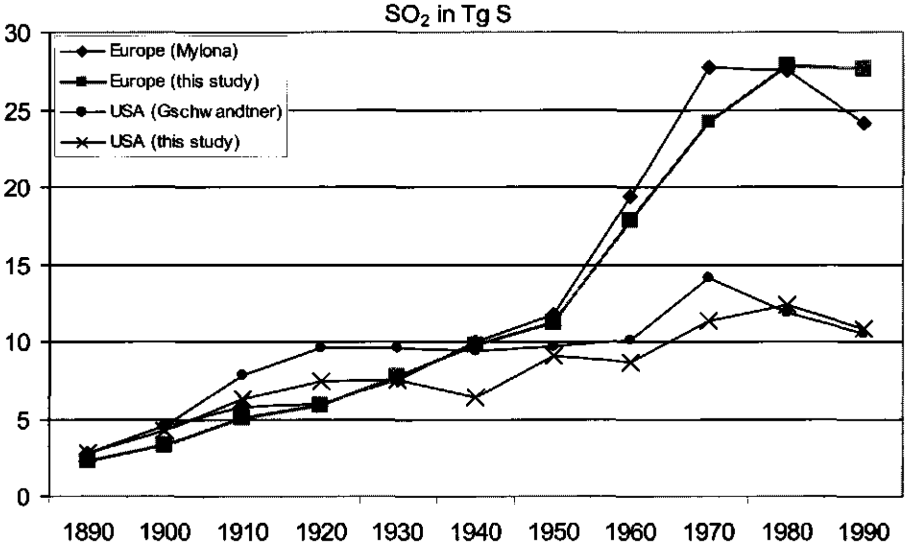

Figure 2.5 Comparing our estimated European SO_2 emissions with Mylona (1996) and U.S.A. emissions with Gschwandtner et al. (1985)

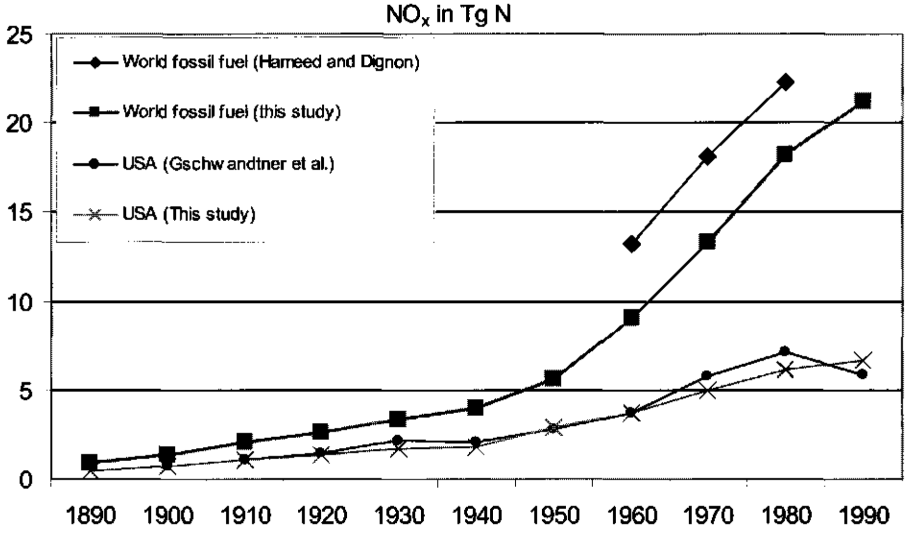

Figure 2.6 Comparing our estimated global NO_x emissions with Hameed and Dignon (1988) and U.S.A. emissions with Gschwandtner et al. (1985)

2.4.2 Uncertainties

The results of this study are associated with significant uncertainties. Even for present-day emissions, estimates, based on relatively more reliable activity data and emissions factors, are sometimes highly uncertain (see, e.g., Olivier et al. (1999a, 1999b, and references therein) and for CO_2, Marland et al. (1999)). We refer to the various cited papers on 1° x 1° grid emission studies for an extensive discussion of the variability of the emission factors, both within and between countries. Historical activity data, as presented in EDGAR and HYDE, are mostly based on studies using data from national or international statistics agencies. Although the quality of these data is difficult to assess, this information is probably the best available, with consistent source definitions across countries. Where no activity data are available, assumptions were made on processes leading to the activity or process developments influencing the activity. Evidently, this is an important source of uncertainty, also influencing the sector weighted emission factors for fuel combustion and the historical spatial distribution of total emissions. Improvements in the activity data and, in particular, historical land use maps (e.g., biomass burning!) are needed and collaborations with other research disciplines (socio-economic) could be helpful.

Using constant aggregated emission factors for the period 1890– 1970 instead of representative emission factors for emitting processes in the past is, of course, a major source of uncertainty; however, it cannot be avoided in studying historic emissions. To our knowledge, verified emission factors prior to 1970 are hardly available; thus one can only make assumptions on the value of emission factors in the past. For example, many emission factors for fossil fuel combustion and industrial activities during the past 10–15 years (on which most emission factors in this study are based) were probably lower compared with technology used prior to 1970, in particular in the more industrialized countries. For fossil fuel combustion we tried to incorporate this by using globally uniform emission factors for combustion emissions in the pre-1970 period based on emission factors for 1990 in EDGAR 2.0 in regions without emission controls, reflecting the higher uncontrolled emission factors in less industrialized regions in 1990. Olivier et al. (1999a, 1999b) specified the regional average emission factors, as used for 1990 for the compounds considered here. For other activities, such as industrial processes and agriculture, this was not done as we did not have a ratio for selecting specific factors. Hence, for years prior to 1970, constant default aggregated emission factors of the latter categories will probably lead to the underestimation of these emissions. Another source of uncertainty is the interpolation of the emissions to a 1° x 1° grid. The location of the release of pollutants to the atmosphere is dependent on the location of the activity leading to the emission. Since we used 1990 maps for population, etc., migration of people within a country, animals, and economic activities in the past are not taken into account in this study. Although this does not influence the total emissions flux on a country level, it causes additional uncertainties in the spatial emission distribution at 1° x 1° resolution, which increases going back in time. A similar problem was faced by Andres et al.'s (1996,1999) focusing on CO_2 emissions from fossil fuel use and cement production. The largest uncertainties may be expected in large countries with substantial internal migration (e.g., the United States) but may be less for, for example, northern Europe. To give an indication of the uncertainty in the activity data the emission factors and 1990 grid maps, a broad classification of data quality is applied (Table 2.8).

For activity data we use the quality ratings *''acceptable''*, *''poor''*, and *''very poor''*. Data rated acceptable are based on national/international statistics or on publications where activity data are presented. Acceptable is applied here because this category of data is probably the best available at this moment as they are used and tested for many different applications. If data from national/international statistics or other publications were not sufficient for use over the entire period of study or for application to all countries or regions, we used extrapolations or assumptions based on these data to estimate these activities. This category of activity data is rated poor. For activities where no data at all were available the scaling of known information was a means to achieve an estimate of the activity in the past. The rating very poor is applied to these data.

For emission factors we used three types of emission factors: *''detailed''* emission factors, *''regional''* aggregated emission factors, and *''global''* averaged emission factors. Detailed emission factors are country-specific emission factors as used in the EDGAR 2.0 database. Regional aggregated emission factors are based on aggregation of detailed emission factors to a certain region-specific emission factor or on studies where only regional-specific emission factors are presented. Global emission factors are the result of aggregation of detailed and/or regional emission factors to global emission factors or are used because only one emission factor for a certain sector is known. The nature of the inherent uncertainties associated with emission factors, the variability of the 1990 emission factor figures over different countries, and the increased uncertainty from applying 1990 emission factor figures for the more distant past result in a qualitative picture of uncertainty varying from source to source in 1990 and in all cases increasing when going farther back in the past.

The quality of spatial distributions using the 1990 grid maps for the whole 100 year period could generally be classified as acceptable or poor except for a few sources (Table 2.8). This refers both to the quality (applicability) of the grid maps used for distribution of 1990 emissions and to the facts that spatial distributions of emission sources within a country shift in time (e.g., because of urbanization) and that we have neglected this feature by applying the same 1° x 1° distribution for maps for the whole 100 year period.

In particular, for biomass burning due to the simplified approach, and also for point sources like power plants and industrial process locations, we note the important large uncertainties associated with the methods used. Nevertheless, these uncertainties of the within country distribution are partly compensated by the presumably more accurate emission estimates at the country total level, which are, in fact, the first step in locating emissions on the world map. The highest uncertainties can therefore be expected in the largest countries as well as in the gridded emissions of biomass burning. This evaluation indicates that although the uncertainty in the gridded inventories for the distant past may be considerable at the 1° x 1° resolution (also because shifts in boundaries between countries have not been considered), it is likely that the uncertainty in the spatial distribution is smaller when the aggregated data are used at a lower resolution, for example, in models with a 5° x 5° grid spacing. The result of this data quality rating is presented in Table 2.8 and provides an impression of the uncertainty in the emission estimates. Very uncertain emission estimates are the result of applying very poor activity data with global aggregated emission factors. Probably the best emission estimates are based on acceptable activity data and detailed emission factors. This qualitative discussion will be used to prioritise improvements on the historical emission data. Unfortunately, we are at this point not able to give a more quantitative statement on data quality, although this appears to be very important for, for example, inverse modelling studies (e.g., Houweling et al., 1999).

2.5 Concluding remarks and recommendations

We constructed a data set of historical anthropogenic trace gas emissions for the period 1890–1990, which can be useful in trend studies of tropospheric trace gases (e.g., Lelieveld and Dentener, 2000; Houweling et al., 1999) and also in environmental assessments. Examples are the analyses of historical contributions of regions and countries for environmental problems such as the enhanced greenhouse effect, acidification, and eutrophication. In general, the global and regional emission trend estimates show a rather good agreement with previous studies. Nevertheless, some differences in the total emission fluxes are evident, although there is no absolute reference to (in)validate the different estimates. One advantage of our data set is that greenhouse and reactive trace gas emission estimates are calculated consistently using the EDGAR system so that future improvements concerning activities and emission factors can be easily implemented. Validation and further development of the emissions inventory will rely to a large degree on the use of the data in global models and comparison of the results with field observations.

We started with 1990 data, which are relatively robust, focusing on energy/industry and including biomass burning for completeness in a simple, transparent way, and worked back in time with activity data and emission coefficients, realizing that the data quality of all aspects considered (statistics, emission factors, and country definition/spatial distribution) becomes increasingly uncertain when going farther back in time. Thus the results should be viewed as a first attempt to estimate historical emissions for a time span of 100 years in a spatial explicit way using a consistent approach, i.e., taking into account the relative uncertainties for various source categories in both present and past emission inventories. In section 4.2 several elements were identified that could be recommended for improvement of the data set: (1) improvement of estimates of uncontrolled emissions factors for 1970 for many trace gases, if possible region-specific, to reflect better the average age and maintenance levels of applied technologies; (2) inclusion of trends in emission factors for the period 1890–1970 in cases where significant shifts can be expected, for example, in the type of coal mining (surface versus underground) and rice production regimes (irrigated versus other) and in autonomous trends in emission factors, for example, for road transport; (3) improvement in procedures to estimate activity data, notably fuel consumption and fuel mix per key economic sector and the amount of biomass burning in all regions; (4) inclusion of separate data sets for air traffic and international ship traffic; (5) improvement of grid maps for older historical years, in particular, for key maps like population, for example, by taking into account shifts due to ongoing urbanization, and like large-scale biomass burning. However, the results of the present study can be used as an a priori emission data set for atmospheric models that investigate the effect of long-term trends in the emissions of trace gases and require global emissions with an explicit spatial distribution at a model resolution lower than or equal to 1° x 1°.

Table 2.8 Qualitative analysis of activity data, emission factors and 1990 grid maps used to calculate historical emissions.

Emission Source	Activity data (Quality [a])	Emission factors [b]								1990 grid map [c]	
		CO_2	CO	CH_4	NMVOC	SO_2	N_2O	NO_x	NH_3	Type	Quality
Fossil fuel combustion											
Power plants	1890 – 1920: scaling (3)	G	G	G	G	G	G	G	...	pop	3
	1930 – 1960: literature, assumptions (2)										2
	1970 – 1990: detailed statistics (1)										1
Domestic	as above	G	G	G	G	G, R for coal	G	G	...	pop	1
Industry	as above	G	G	G	G	G	G	G	G	pop	2
Transport	as above	G	G	G	G	G	G	G	G	pop	1
Fossil fuel production											
Brown coal + hard coal	international statistics + assumptions (2)	...	...	G	...	...	...	...	...	ps	1
Oil	international statistics (1)	D	...	G	G	...	...	...	...	ps	1
Gas	international statistics (1)	...	...	G	...	...	...	...	...	ps	1
Biofuel combustion											
Industry + domestic	scaling of 1990 data (3)	G	G	G	G	G	G	G	G	pop	2
Industrial processes											
Iron	international statistics (1)	...	G	G	G	R	...	...	...	pop	2
Steel	international statistics (1)	...	R	...	R	...	...	R	...	pop	2
Copper	international statistics (1)	...	...	...	...	R	...	...	...	ps	1
Nitric acid	scaling of 1990 detailed data (2)	...	...	...	...	...	G	...	G	ps/pop	2
Adipic acid	scaling of 1990 detailed data (1)	...	...	...	...	...	G,R: United States, Canada	...	...	ps	1
Cement	1890 – 1960: literature (3) 1970 – 1990: international statistics (1)	G	...	...	...	...	...	...	...	pop	3
Solvents	scaling of 1990 data (3)	...	...	...	G	...	...	...	...	pop	2
Agriculture											
Rice	international statistics (1)	...	...	G: 1890 – 1970 R: 1970 - 1990	...	...	...	...	...	s-s	2
Fertilizer	pre-1960: scaling (3) 1960 – 1990: international statistics (1)	...	...	...	...	...	G	G	G	s-s	1
Animal types	1890 – 1960: literature (1) 1970 – 1990: international statistics (1)	...	...	G,R	...	...	G,R	...	G,R	s-s	1
Biomass burning											
Savannah burning	educated quess (3)	...	G	G	G	G	G	G	G	s-s	2
Deforestation	scaling of 1990 data (3)	G	G	G	G	G	G	G	G	s-s	3
Waste											
Agr. waste burning	scaling of 1990 data (3)	...	G	G	G	G	G	G	G	s-s	1
Landfills	scaling of 1990 data	...	...	R	...	...	...	...	...	s-s	2

[a] Quality ranking: 1: Acceptable, 2: Poor, 3: Very poor, [b] Emission factors: G: Global aggregated, R: Regional aggregated, D: Detailed (country-specific), [c] Map type: pop: population as surrogate, ps: point sources, s-s: source-specific map

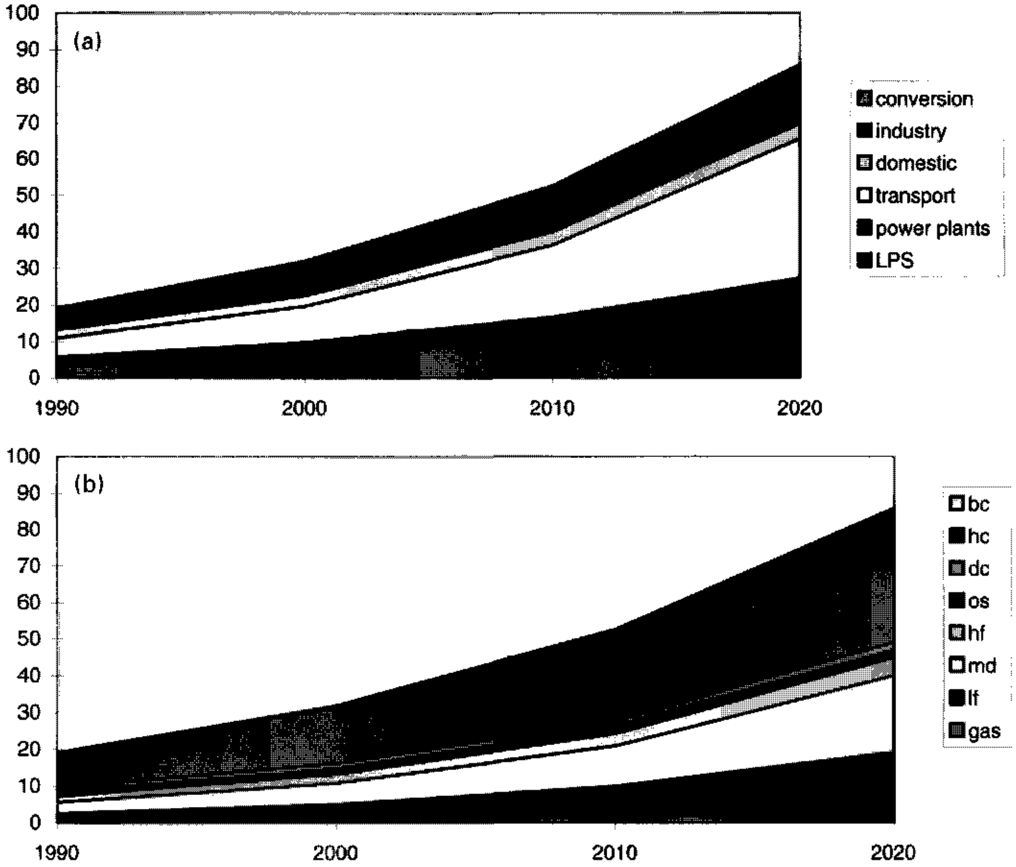

Figure 3.2 NO_x emissions by economic sector and fuel type in the period 1990 - 2020 (NO_2 yr^{-1}).

The calculated emissions by country, region and LPS location were allocated to a 1° x 1° grid based on the methodology used in RAINS-ASIA (Foell et al., 1995). In this procedure each individual emission source (LPS) is assigned to its appropriate 1° x 1° grid, with the remaining emissions shared out to grid cells on the basis of population. While this process has potential errors (for example the gridding of emission from 'fixed' small power plants by population may not be totally justified), it must suffice until more detailed information on source location becomes available.

Figure 3.3 presents the 1°x1° grid emissions for 1990 and Figure 3.4 presents the ratio between 2020 and 1990 emissions. In 1990 areas with the highest emission intensities (emissions '0.1 Tg NO_2 per grid annually) are found in Bangkok, Singapore, Hong Kong, South Taiwan, parts of east China (including Shanghai, Beijing, and Tianjin), Seoul, Pusan, and in Japan around Tokyo, Osaka, and Kyoto.

In 2020 many individual grids show emissions larger than 0.1 Tg NO_2. These areas are found in parts of India (Bombay, Madras, Delhi, Calcutta), Bangkok, Kuala Lumpur, Singapore, parts of Java (including Jakarta), Manila, Taiwan, Hong Kong, large areas of eastern China (including Shanghai, Tianjin, Beijing), Seoul, Pusan and parts of Japan (Tokyo, Osaka, Kyoto). The magnitude of the increase in emissions is clearly shown in Figure 3.4 where the 2020 emissions in all of Asia except Japan and Myanmar are more than five times larger than the 1990 emissions. Emissions in India, Indonesia, Vietnam, South China, Philippines and Malaysia are between five to ten times the 1990 emissions, and emissions in parts of Pakistan, Cambodia and Thailand are more than ten times the 1990 levels. The megacities (e.g. Jakarta, Madras, Delhi, Manila, Bangkok, Calcutta and Seoul) also show tremendous growth as will be discussed in more detail below.

3.4 Discussion of results

The above results illustrate the fact that NO_x emissions in Asia are growing rapidly, and will continue to do so over the decades to come. Because the region is changing so rapidly, the results for the later years are subject to a high degree of uncertainty and should be viewed simply as a possible endpoint of present-day practices. The actual emissions trajectory will change (and lower) if the region makes more use of cleaner fuels, implement (further) control policies, and utilizes its energy more efficiently. However, it is informative to look more closely at these projections since they provide a baseline for future emissions in the region.

3.4.1 Economic restructuring

The present and future NO_x emissions reflect the diversity in Asian economies and the growth and restructuring that is taking place within the region. This is illustrated in Table 3.9, where the emissions by economic sector are presented for the largest emitting countries for the period 1990–2020. Also shown for reference is the Asia-wide composite breakdown. Japan represents one extreme of the economy-emission relationship in Asia. Japan has a low energy intensity, reflecting an economy built on finished goods and services, which are less energy intensive than heavy industries such as steel and cement production. Japan also has stringent environmental regulations which require widespread use of advanced control technologies. Emissions in Japan in the period 1990–2020 show the smallest increase due to the facts that economic growth in Japan is not expected to be as fast as in other Asian countries, and Japan is already applying energy efficiency and emission control technologies (which are reflected in our energy scenario).In 1990 Japan's NO_x emissions are dominated by the transport sector (55%), followed by the industrial (22%) and power (18%, including LPS) sectors. In 2020, the largest growth in emissions arises from the power sector (increasing to 31%). This growth is compensated by decreasing shares (in terms of percentages) from the industrial and transport sectors.

A similar sectoral picture emerges for South Korea, with the major difference being that its growth in NO_x emissions is double that for Japan. One interesting point for South Korea is that the emissions from the power sector are well below the Asian average, reflecting their use of nuclear power (48% of total power generation) (Foell et al., 1995). The situation is markedly different for the sectoral contributions in China's emissions. Compared to Japan and to the Asian composite, the

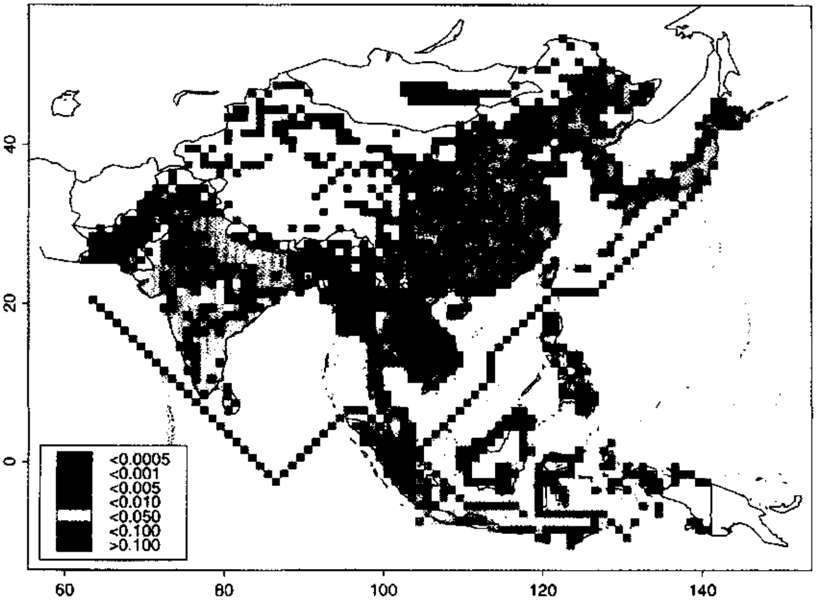

Figure 3.3 1990 emissions on 1 x 1 grid (Tg NO_2 per grid).

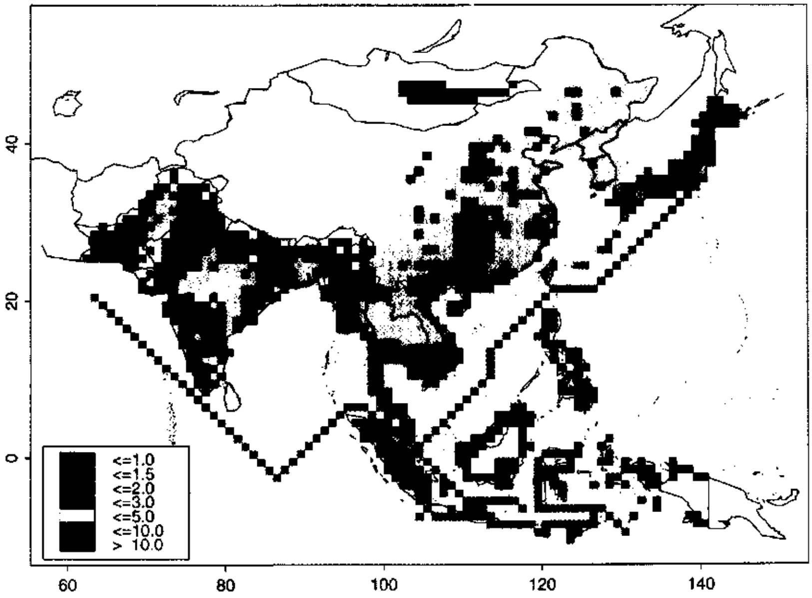

Figure 3.4 Ratio between 2020 and 1990 emissions on 1 x 1 grid.

industrial sector contribution is very high while the contribution from transportation is very low. Over the next few decades the situation changes, with a dramatic increase in the transport sector (growing to 33%). The contribution from the domestic sector decreases by a factor of 2 in China (and throughout Asia). This reflects the fact that throughout Asia much of the present energy is used for domestic purposes (cooking, heating and agriculture), but as the economies in the region industrialize, the relative importance of the domestic sector falls. It is also important to recognize when discussing China that there are significant regional differences within China. As already discussed in regards to Figure 3.4, growth in NO_x emissions can vary by a factor of 3 or more from region to region within China.

India presents another energy-emission mix. India has a very high contribution from the domestic sector (25%), which is 2 times higher than the Asia-wide value. This reflects the reliance on dung and fuel wood for cooking and heating. The contribution of industrial emissions are low compared to the Asian average, indicating that the industrial sector in India is a smaller economic factor compared to countries such as China, Korea, and Taiwan. NO_x emissions in India are projected to increase by more than a factor of 5, with the largest increase in the transport sector, which accounts for 50% of the NO_x emissions in 2020.

In Indonesia the transport sector already dominates NO_x emissions, accounting for 51% of 1990 emissions. Indonesia also has a high contribution from the domestic sector, but relatively low contributions from the power and industry. Indonesia's NO_x emissions are estimated to increase six-fold by 2020, with the largest increase in the power sector.

Thailand has the largest contribution from the transport sector at present (64%). This is due in part to relatively low emissions from the industrial and power sectors. However, Thailand's NO_x emissions are projected to increase by a factor of 10 by 2020, with the largest growth in the power sector.

As discussed above, transport becomes the largest NO_x emitting sector by the year 2020. In 1990, emissions from the transport sector account for 27% of the total Asian NO_x emissions, with the smallest contribution to total emissions by country for China (7%) and the maximum contribution of 67% found in the Philippines and Sri Lanka. The average contribution of the transport sector to NO_x emissions in Asia in 2020 grows to 44%, with a minimum contribution in North Korea (10%) and a maximum in Cambodia (86%). The importance of the transport sector means that particular attention must be directed to improving these emission estimates. This will be very challenging in Asia, and will require more detailed information on vehicle counts and Asian-specific vehicle emission factors.

Table 3.9 Contribution of economic sectors to total NO_x emission (unit: % of total).

		Conversion	**Industry**	**Domestic**	**Transport**	**Power plants**	**LPS**
China	1990	2	44	9	7	18	20
	2020	1	30	5	33	6	25
India	1990	0	17	25	25	2	31
	2020	0	7	7	58	8	19
Indonesia	1990	4	11	21	51	5	9
	2020	3	8	4	47	19	9
Japan	1990	1	22	4	55	14	4
	2020	1	13	4	51	23	8
South Korea	1990	0	27	7	49	10	7
	2020	1	19	2	42	25	11
Thailand	1990	1	10	9	64	11	5
	2020	1	6	2	55	4	36
Total Asia	1990	2	30	12	27	13	17
	2020	1	18	5	44	13	19

3.4.2 Emissions and socio-economic factors

Table 3.10 presents the NO_x emission, Gross Domestic Product (GDP), and emission per million inhabitants for each country (sorted by NO_x emissions in 2020). In general, there is a close association between emissions and GDP, with countries with high GDP also having large emissions. In 1990 the top emitting countries (China, India, Japan, and South Korea) also have the highest GDPs. The largest emitting countries in 2020 also have the highest GDP, with the exceptions being Japan and North Korea. Although Japan has a high GDP, the emissions are smaller than countries with 4–10 times lower GDP. These low emissions together with a high GDP occur because of the application of abatement technologies in Japan. North Korea is a country with a low GDP but has emissions as large as countries with GDPs which are much higher (4-10 times). This is the result of the prevalence of small, energy inefficient factories and technologies in North Korea.

Examination of emissions normalized by GDP presents a useful framework for discussion. This information is also presented in Table 3.10. The average value for Asia in 1990 is ~ 5 Gg NO_2 per billion US$ of GDP. Japan has the lowest NO_x emissions per US$ GDP (~ 1 Gg NO_2 per billion US$ of GDP), while China has the highest (5 times greater than the Asia average) and India the second highest value (~4 times the Asian average). The Asia-wide average emissions per US$ GDP increase by ~15% by the year 2020. However, China's emissions per US$ of GDP are predicted to decrease by a factor of 2, while India's value remains constant. China's projected decline in emissions per GDP is due to a combination of two factors. The numerator (i.e. the emissions) is growing less rapidly than in other countries. This is because a large portion of energy use in China remains in the industrial sector, where energy efficiency improvements are continually being made, and relatively less energy is going to the transportation sector (see Table 3.9). Second, the denominator is growing much faster than in other countries, because of the dramatic growth of the Chinese economy. Large increases (more than a factor of 2) are projected for Indonesia, Thailand, North Korea and Cambodia, while substantial decreases are estimated for Japan, Malaysia, Hong Kong and Singapore.

Emissions normalized by population are also presented in Table 3.10. The Asia-wide average value in 1990 is 7 Gg NO_2 per 1 million inhabitants. On this basis, Japan, Hong Kong, Singapore, South Korea, and Taiwan, show the highest values (3–7 times higher than the Asia average). In contrast, China, India, and Thailand have emission values at or below the average. The situation changes dramatically by the year 2020. The emissions per capita increase by nearly a factor of 3, with some large emitting countries like India, Indonesia, Thailand and South Korea, increasing by factors of 4–5. The per capita emissions in China in 2020 are projected to be similar to Japan's 1990 levels. The more rapid growth in emissions per capita compared to emissions per unit GDP shows that economic growth in Asia greatly outpaces population growth. These statistics also point out the fact that without aggressive use of emission control technologies and more efficient use of energy, the emissions per capita in Singapore, Taiwan, South Korea, and Hong Kong could exceed 100 Gg NO_2 per million inhabitants.

Table 3.10 NO_x emission and socio-economic factors.

	Emission Gg NO_2		**GDP (billions of 1990 U.S. $)**		**Gg NO_2 per 10^6 capita**		**Emission/GDP**	
	1990	2020	1990	2020	1990	2020	1990	2020
China	8273	32364	321	2376	7	21	25.77	13.62
India	3481	22824	209	1255	4	14	16.65	18.19
Indonesia	793	5090	133	439	4	22	5.96	11.59
Thailand	494	4083	86	448	9	52	5.74	9.11
South Korea	977	4590	253	1228	23	91	3.86	3.74
Japan	2468	4271	2405	7388	20	34	1.03	0.58
Taiwan	520	2176	155	551	26	85	3.35	3.95
Pakistan	271	2056	35	203	2	9	7.74	10.13
North Korea	518	2429	28	51	24	76	18.50	47.63
Malaysia	283	1708	29	236	16	43	9.76	7.24
Philippines	166	994	37	185	3	10	4.49	5.37
Hong Kong	245	674	70	280	42	98	3.50	2.41
Vietnam	128	734	13	164	2	7	9.85	4.48
Bangladesh	102	474	22	97	1	3	4.64	4.89
Cambodia	29	389	2	10	3	20	14.50	38.90
Singapore	101	383	31	189	37	109	3.26	2.03
Sri Lanka	39	132	8	19	2	6	4.88	6.95
Myanmar	41	93	23	50	1	1	1.78	1.86
Mongolia	30	85	2	9	14	20	15.00	9.44
Nepal	29	67	3	11	2	2	9.67	6.09
Brunei	16	43	2	8	64	90	8.00	5.38
Laos	4	15	1	3	1	2	4.00	5.00
Bhutan	2	8	0	2	1	3	.	4.00
Total Asia	19158	85987	3860	15100	7	19	4.96	5.69

3.4.3 Megacities

One important feature in Asia is the rapid rate of urbanization which is taking place. It is expected that the economic growth in Asia will have its largest impact on the environmental quality in large cities. To examine this situation more closely NO_x emissions by sector for seven megacities (Bangkok, Delhi, Jakarta, Manila, Beijing, Shanghai and Seoul) were estimated and are presented in Table 3.11. In both 1990 and 2020 emissions from the transport sector are the largest single source in these cities.

4.2 Types of uncertainty

Although the term uncertainty is commonly used, people (including scientists) use it in different ways. In most dictionaries like for example Chambers (1988) and Webster's (1971), uncertainty is related to something that is doubtful, not definitely known or variable. In the field of emission inventories the term uncertainty has been defined by McInnes (1996) as *"a statistical term that is used to represent the degree of accuracy and precision of data"*. IPCC/OECD/IEA (2000) presents both a statistical definition and an inventory definition. The statistical definition relates uncertainty to sample variance of the coefficient of variation while the inventory definition describes uncertainty as *"a general and imprecise term which refers to the lack of certainty (in inventory* components) resulting from any causal factor such as unidentified sources and sinks, lack of transparency etc". In this thesis we define uncertainty as lack of knowledge of accuracy (section 4.2.1) and lack of knowledge of reliability (section 4.2.2).

4.2.1 Uncertainty about accuracy

According Chambers (1988) and Webster's (1971) the word accuracy can be used to express that something is exact. Based on this dictionary description we define the accuracy of an emission inventory as the extent to which an emission inventory is an exact representation of the emission that has occurred in reality.

It is common practice that large-scale emission inventories such as the inventories of worldwide historical emissions (chapter 2) or NO_x emissions in Asia (chapter 3) are not an exact representation of the emission that has occurred in reality. An important reason for this is the need to extrapolate and aggregate available information both in time and space. Emissions of air pollutants from anthropogenic origin are caused by a variety of small and large individual sources such as power plants, industries, motor vehicles or animals. The emissions from these individual sources are usually variable both in time and space. It is practically not possible to monitor each of the emission sources individually and therefore the emission factor approach has been adopted in many studies to quantify the emissions of air pollutants at higher aggregation levels than the individual source of emissions and by extrapolation of existing data. The resulting emission inventory is therefore inaccurate.

The fact that emission inventories are inaccurate representations of the emissions that have occurred in reality can be formalized by equation (1), where E_{real} is the emission that has actually occurred and $E_{inventory}$ the estimate of the real emission. The term ε_i represents the inaccuracy (ε) caused by a source of inaccuracy *i*. The overall inaccuracy $\cup$ is the result of N sources of inaccuracy.

$$E_{real} = E_{inventory} + \bigcup_{i=1}^{N} \varepsilon_i \qquad (1)$$

Quantifying the inaccuracy using equation (1) for a specific emission inventory is not a straightforward exercise. In order to determine the inaccuracy $\cup\varepsilon_i$ of an emission inventory, the emission that actually has occurred (E_{real}) needs to be known. This establishment of the truth (the true emission) is called verification (Webster, 1989; McInnes, 1996). However, according to Oreskes et al. (1994) verification is only possible in closed systems in which all of the components of the system are established independently and are known to be correct. This means that verification of large-scale emission inventories (e.g. total anthropogenic NO_x emissions in China in 1990) is not possible, because it is practically impossible to perform a continuous emission monitoring on each emission source at such a large scale. As a result, the real emission cannot be exactly known. This means that we do not know the exact size of the inaccuracy and this inaccuracy can then only be roughly estimated. In order to gain insight into the inaccuracy of emission inventories we need to know the different sources of inaccuracy *i*. In many cases this is not known. Not knowing the accuracy or the sources of inaccuracy in emission inventory can be defined as uncertainty about accuracy. Or in other words:

Uncertainty about accuracy is the lack of knowledge of the sources and the size of inaccuracy.

4.2.2 Uncertainty about reliability

According to Chambers (1988) and Webster's (1971) the word reliability can be used to express that one relies or depend upon something with confidence (in this study the emission inventory). In the field of emission inventories the word reliability has been defined by McInnes (1996) as trustworthiness, authenticity or consistency. IPCC/OECD/IEA (2000) does not define reliability. We define reliability as the extent to which one can rely on or trust the emission inventory. Unreliability is lack of knowledge of the extent to which one can rely on or trust an emission inventory.

The important question that needs to be addressed is: when does one rely on or trust an emission inventory? First of all, there is no uncertainty about the reliability of an emission inventory when the inventory is found to be accurate. However, when the accuracy is not known -or even more- when the inventory is found to be inaccurate, does this mean that one cannot rely upon or trust an emission inventory?

The reliability of an emission inventory and the role of accuracy in this is dependent on the intended purpose of an emission inventory. In order to understand the chemical and physical processes and the behaviour of air pollutants in the atmosphere, atmospheric modelling studies require accurate estimates of emission of air pollutants. This means that for scientific purposes the reliability of an inventory is directly related to accuracy. When emission inventories are used for policy purposes the reliability of the emission estimate in relation to the accuracy of the emission estimate is different. For example, the Dutch Ministry of Environment formulated reliability of the Netherlands national pollutant register (PER) as *"the whole process of construction of emission figures and inclusion of the figures in the PER should be traceable"*(VROM, 1997). This definition of reliability does not include accuracy as a condition for reliability of an inventory. The reason for this is that when emission inventories are used for policy purposes, users are more interested in the transparency of the emission calculations and the compliance with agreed upon methodologies or reporting formats (Pulles and Builtjes,1998; Lim and Boileau,1999). In order to achieve this transparency (i.e. use of agreed upon methodologies) several quality criteria have been defined that are to be met by the emission inventories. In this sense, the reliability of an emission inventory is related to the quality criteria. These quality criteria could include accuracy of the emission estimate but this is not always the case (such as for PER as defined in VROM, 1997). For the UN Framework Convention on Climate Change (UNFCCC) accuracy is one of the quality criteria and therefore of the reliability of greenhouse gas emission inventories. The IPCC good practice guidelines (IPCC/OECD/IEA, 2000) include five quality criteria that should be met (1) transparency, (2) consistency, (3) comparability, (4) completeness and (5) accuracy. Using the IPCC definitions, this means that (1) documentation should allow for a reconstruction of the emission inventory, (2) the same methodologies and consistent data sets should be used for calculation of emission in subsequent years, (3) emission inventories from different countries can be compared with each other, (4) the emission inventory should include all sources and sinks of greenhouse gases and (5) emission estimates are systematically neither over nor under the true emissions or removals, as far as can be judged and that uncertainties are reduced as far as practically possible.

Based on the discussion above, the lack of knowledge of the extent to which one can rely on or trust an emission inventory is dependent on the criteria set by the users of an emission inventory. In the case of scientific use of inventories the criterion is accuracy. In the case of policy applications different user-specified criteria defined such as for example transparency, consistency and accuracy. These criteria can differ from case to case such as the criteria set by VROM (1997) or IPCC/OECD/IEA (2000). Uncertainty about reliability can therefore be defined as follows:

Uncertainty about reliability is the lack of knowledge of the degree to which the emission inventory is meeting user specified quality criteria.

4.3 Sources of uncertainty

4.3.1 Introduction

As shown in the previous section, we make a distinction between uncertainty about accuracy and uncertainty about reliability. These two types of uncertainty are related because in most cases the accuracy of an inventory is one of the quality criteria for the inventory.

There are two main reasons why there is uncertainty about reliability. First, the accuracy of an emission inventory is not known (uncertainty about accuracy). Second, the documentation of the emission inventory is inadequate and incomplete which prevents to determine whether the inventory meets quality criteria such as transparency, consistency and comparability. There can be several causes for the documentation to be inadequate ranging from inexperience to unwillingness of countries to comply with quality criteria such as set by UNFCCC.

In the following two sections we will only discuss the various types and sources of inaccuracy. However, in section 4.4 we will come back to uncertainty about reliability when discussing the assessment of the causes of uncertainty.

4.3.2 Sources of inaccuracy

To gain insight in the uncertainty about accuracy we need an overview of the potential sources of inaccuracy that can exist in an emission inventory. The variables *i* in equation (1) represent the sources of inaccuracy. In the emission inventory literature different sources of uncertainty in emission inventories have been described. Uncertainties in the accuracy of emission inventories can be subdivided in inaccuracies in (i) the emission inventory structure: **structural inaccuracy** and (ii) the values of activity data and emission factors: **input value inaccuracy**. Within each category different types of either structural or input value inaccuracy can be defined.

Figure 4.1 presents an overview of the categorisation of inaccuracy. Section 4.3.2.1 and 4.3.2.2 describe structural inaccuracy and input value inaccuracy, respectively. In Table 4.1 we present an overview of the different sources of inaccuracy as mentioned by several authors, using the categories that will be described below.

The categorisation into structural and input value inaccuracy is comparable with the classification of sources of uncertainty in risk and policy analysis of Morgan and Henrion (1990), however the different types of structural and input value inaccuracy are typical for the field of emission inventories.

4.3.2.1 Structural inaccuracy

The structure of the emission inventory is defined by the selected source categories, the spatial scale (local, national, global or by grid), the temporal scale (year, season, month, day, hour) and the equations to calculate the emissions for the specific source categories on the selected spatial and temporal scale. **Uncertainty about structural accuracy** (ε_S) is the lack of knowledge of the extent to which the structure of an emission inventory allows for an accurate calculation of the 'real' emission. Three important causes of structural inaccuracy are aggregation error, incompleteness, and mathematical formulation error.

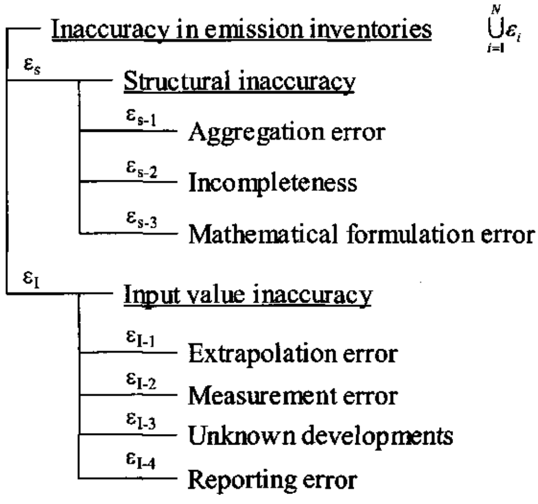

Figure 4.1 Categorisation of inaccuracy in emission inventories. See text for detailed description.

ε_{S-1}: Aggregation error

Inaccuracy due to aggregation is caused by the fact that emissions are calculated on a spatial scale, a temporal scale and for emission source categories that are different from the scale on which the emissions in reality occur. In many cases there is lack of information about the emission processes and the variability of emissions on the required spatial and temporal aggregation level. In such cases, aggregation of the limited data available is needed, introducing inaccuracy. We illustrate this type of inaccuracy with the following example. During the construction of the historical emission database (chapter 2) a selection of source categories was made for which the emissions of a specific compound were calculated. Because information about activities in the past was limited, and sometimes not existing, aggregated source categories were used. An example of this is the calculation of transport emissions. In other emission inventories, including the EDGAR methodology (Olivier et al., 1996), present-day emissions from transport are calculated for the sectors road, rail, inland water, land non-road, domestic air, international air and international shipping. For the calculation of historical emissions in the period 1890 – 1970 we treated transport as a single sector because no activity data or emission factor values for sub-sectors were available. Table 4.1 contains several other examples of structural inaccuracy due to aggregation.

ε_{S-2}: Incompleteness

An emission inventory may be inaccurate due to missing emission sources. This can be caused by incomplete scientific understanding of the emission processes. This source of structural inaccuracy can be expected in -but is not limited to- first time inventories of emissions. For example in the problem of acidification it was clear from the beginning that sulphur dioxide emissions from fossil fuel combustion were a large source of emissions. However, in many inventory studies the emissions from ships were neglected because it was felt that emissions from ships were small. In more recent years, it became clear the sulphur dioxide emissions from ships are a substantial source of sulphur dioxide emissions. Emissions inventories that –due to incomplete understanding of the emission process- excluded emissions from ships are inaccurate. Another example of incompleteness can be found in early N_2O emission inventories. For a long time, estimates of N_2O from agriculture considered only direct N_2O emissions from agricultural fields. Recently, however, it became clear that agricultural activities may also result in indirect N_2O formation, far away from the agricultural fields (e.g. after leaching) (Mosier et al., 1998). AEAT (1998), Amann (1992) and Rypdal and Winiwarter (2001) mention incompleteness as source of uncertainty.(Table 4.1).

ε_{S-3}: Mathematical formulation error

At a given aggregation level, calculation errors can be a source of structural inaccuracy. If errors are made in the equations of an emission inventory the result of the emission inventory calculation is a wrong representation of the real emission. Calculation errors may be simply mistakes. Baars et al. (1992) and U.S. EPA (1996a) have mentioned simple calculation errors as source of uncertainty (Table 4.1). More importantly, the mathematical formulation used may be wrong. For example, the emission factor calculation may wrongly assume that there is a linear relationship between emissions and activities, while in some cases this relation is not linear. Inappropriate mathematical formulation can be a result of lack of data or missing information, but it can also result from incomplete understanding of the processes involved.

4.3.2.2 Input value inaccuracy

As shown above, the content and structure of an emission inventory can be defined by the selection of source categories and the spatial and temporal scales for which the emissions are calculated. The equations that are used to calculate the emissions for an inventory of a given structure contain parameters and variables such as emission factors and activity data of emission sources for which input values are needed. We define uncertainty about **input value accuracy** (ε_I) as the lack of knowledge of the values of activity data and emission factors. This type of inaccuracy can be subdivided into four different types: extrapolation, measurement error, unknown developments and reporting.

ε_{I-1}: Extrapolation error

The term extrapolation means *'estimate from observed tendencies the value of any variable outside the limits between which these are known'* (Webster, 1988). Due to lack of measurements of emission rates or activity data, readily available measurements (not specific for the source category, spatial or temporal scale) are extrapolated leading to inaccurate input values. One of the reasons that specific measurements are missing is the variability of emissions in time, space and source. Inaccuracy due to extrapolation is illustrated by the following examples. Due to lack of data on biofuel consumption in the past, the calculation of historical emissions from biofuel burning for the years 1890 -1980 was performed by scaling the available information on biofuel use per country per capita in rural areas in 1990 with the rural population per country for the years 1890 – 1980 (chapter 2). Another example can be found in chapter 3 on NO_x emissions in Asia where we needed emission factors by vehicle and fuel. Besides emission factors for Japan (OECD/IEA, 1991) and detailed information about vehicle fleet composition, miles driven and emission factors in the Manila region (Urbair, 1995; VECP, 1992) no specific emission factors were available. For the whole Asian region, except Japan, the emission factors have been calculated based on the Manila data. Application of these emission factors leads to inaccuracies because the composition of the vehicle fleet and the miles driven in other Megacities will be different from the situation in Manila. Furthermore, the vehicle composition and miles driven in remote areas with unpaved roads will also be very different from Manila. Other emission inventory studies have mentioned extrapolation as source of inaccuracy in emission inventories (see Table 4.1).

ε_{I-2}: Measurement error

Errors in the available measurements can lead to inaccurate values of emission factors or basic socio-economic activity data. Baars et al. (1992) describe an example of measurement error due measurement conditions: When measuring NO_x from stationary industrial combustion using the chemiluminescence's technique, the presence of other components like CO_2, H_2O or O_2 in the air sample can cause interference with the NO_x measurement. Another example of measurement error can be the fact that the concentration of a pollutant in an air sample may be below the detection limit of the analysis equipment or that emission measurements are performed while the emitting equipment is not operating properly. As shown in Table 4.1, several studies mention measurement error as source of inaccuracy.

ε_{I-3}: Unknown developments

When constructing emission scenarios to analyse possible future trends, information is needed about future emission factors and future socio-economic developments. Due to incomplete understanding of developments in the future, these emission scenarios can never be accurate. In our study of NO_x emission scenarios for Asia (chapter 3) there is uncertainty about the accuracy of future activity data. The reason for this inaccuracy is that the estimates of Asian NO_x emissions in the future are based upon on energy pathway and a control strategy. The energy pathway estimates the change in energy use based upon regional assumptions of growth in GDP, population and other socio-economic data. These future developments in economic growth, energy efficiency improvement or use of cleaner fuels cannot be exactly known at the moment of constructing the emission inventory. For this reason, the emission inventory is regarded as uncertain because the accuracy of the activity data could not be established.

ε_{I-4}: Reporting error

Just as calculation errors are a type of structural inaccuracy, reporting errors can lead to inaccurate emission inventories. When values of emission factors or activity data are accurately known but erroneously reported due to for example typing errors, the input values that are used for the emission inventory calculations will become inaccurate. Since most emission inventory studies are subject to review, reporting errors should be identified during the emission inventory construction process. This might be the reason why only two emission inventory studies mention this source of inaccuracy (Baars et al., 1992; U.S. EPA, 1996b) (Table 4.1).

Table 4.1 Overview of different sources of uncertainty as mentioned in selected emission inventory studies and a classification of these uncertainties. See text for description of ε_S and ε_I. The presence of a question mark (?) means that the example could fit into different sources of uncertainty.

Reference	**Source of uncertainty**
AEAT (1998)	*Inventory of CO_2, CH_4 and N_2O in United Kingdom in 1990 and projections for 2010* 1. Experimental uncertainties in measured emission rates (ε_{I-2}) 2. Measured sources may not be representative of the wide range in the country (ε_{I-1}) 3. Simplifications by treating emissions as coming from a small number of source types with average emissions behaviours (ε_{S-1}) 4. Uncertainties in the basic socio-economic activity data (ε_{I-2}, ε_{I-4}) 5. Uncertainties in the basic understanding of sources and removal mechanisms (ε_{S-2}) 6. Lack of information on the nature of certain sources (ε_{S-2})
Amann (1992)	*Emission inventory of SO_2, NO_x and NH_3 in Europe in 1985* 1. Inaccuracy in emission factors (ε_I) 2. Inaccuracy in spatial distribution (ε_{I-1}) 3. Completeness of the inventory (ε_{S-2})
Baars et al. (1992)	*SO_2 and NO_x emissions in The Netherlands for the year 1985* 1. Variance of the emissions in time and between comparable units (?) 2. Variability in the external conditions in which the units are working (?) 3. Uncertainties in measurements of emissions, emission factors and activity data (ε_{I-2}) 4. Possible errors in the databases itself (ε_{S-3})
Benkovitz (1985)	*Emissions of particulates, SO_x, NO_x, HC and CO, USA in 1980's.* 1. Estimates using stack tests, material balance or engineering judgement (ε_{I-2}) 2. Federal and local emission factors (ε_I) 3. Ash and sulphur content (ε_I) 4. Point source fuel or process rates (ε_I) 5. Efficiency of control equipment (ε_I) 6. Area sources activity rates and category adjustment factors (ε_I)
Benkovitz et al. (1996)	*Global emissions inventory of SO_2 and NO_x for 1985 conditions* 1. Inaccuracy when missing values for emission estimation parameters are assigned based on values from a 'similar' sector (ε_{I-1}) 2. Inaccuracy in level of regional detail in method and data (ε_{S-1}, ε_{I-1}) 3. Inaccuracy due to the gridding process (ε_{S-1})
Bouwman et al. (2000)	*Global emissions of N_2O* 1. Inaccuracy in the geographic data to allocate emissions (ε_{I-1}) 2. Inaccuracy in the economic data (ε_I) 3. Inaccuracy in the emission factors (ε_I) 4. Inaccuracy due to aggregations (ε_{S-1})
Egglestone (1988)	*Emission inventory of CO, NO_x, SO_2 and VOC in the United Kingdom* 1. The fact that emission inventory is estimation process (?) 2. Inaccuracy due to emission measurement error (ε_{I-2}) 3. Inaccuracy in activity statistics (ε_I) 4. Inaccuracy in emission factors due to measurement error or limited amount of measurements (ε_{I-1}, ε_{I-2})
IPCC/OECD/ IEA (1997)	*IPCC Guidelines for construction of national greenhouse gas inventories* 1. Different interpretations of sources, sinks or other definitions (ε_{S-1}, ε_{S-2}) 2. Simplification by using averaged values (emission factors) (ε_{I-1}) 3. Inaccuracy in activity data (ε_I) 4. Non-complete scientific understanding of the processes involved (ε_{S-2})

Table 4.1 Continued.

Reference	**Source of uncertainty**
IPCC/OECD/ IEA (2000)	*IPCC Guidance on uncertainty management in national greenhouse gas inventories* 1. Uncertainties in continuous monitoring of emissions (ε_{I-2}) 2. Uncertainties associated with direct determination of emission factors (ε_{I-2}) 3. Uncertainties associated with emission factors from published references (ε_{I-1}, ε_{I-4}) 4. Uncertainties in activity data (ε_{I})
Janssen et al. (1995)	*NH_3 emissions inventory for The Netherlands in 1992* 1. Errors in specification of activity data (ε_{I}) 2. Errors in specification of emission factors (ε_{I}) 3. Errors in spatial distribution (ε_{I-1})
Kuhlwein and Friedrich (2000)	*NO_x and NMHC emissions from road transport in Germany* 1. Statistical errors (variation in measurements of emissions) (ε_{I-2}) 2. Systematic errors (erroneous fleet composition and errors in method for determination of emission factors) (ε_{I})
Langstaff and Wagner (1986)	*Emissions of NO_x, SO_2, CO, VOC, and NH_3 in the United States of America in 1980* 1. Emission factors (incl. Sulphur retention in ash) (ε_{I}) 2. Throughput of activity (ε_{I}) 3. Knowledge on control penetration (ε_{S-2}, ε_{I-3}) 4. Temporal allocation (ε_{S-1}, ε_{I-1}) 5. Pollutant species allocation (e.g. mix of VOC) (ε_{S-1}) 6. Spatial allocation factors (ε_{S-1}, ε_{I-1})
McInnes (1992)	*Review of inventories for long range transboundary air pollutant studies e.g. CORINAIR90, and IPCC/OECD* 1. S retention in fuel ash (ε_{I}) 2. Difference in de definition of the volatile organic compounds (ε_{S-1}) 3. Distribution and use of solvents (ε_{I}) 4. Assessment of CH_4, NH_3, N_2O and VOC emission factors (ε_{I}) 5. CO_2 emission from landfills (?) 6. Transport data like trip length with cold engines or the state of vehicle fleet related to the optimum operating conditions (ε_{I})
Mylona (1996)	*European SO_2 emission inventory for the period 1880-1991* 1. Inaccurate official or estimated annual emission figures (?) 2. Errors in spatial disaggregation of national emissions (ε_{I-1}) 3. Inadequate information on natural emissions (ε_{S-2}) 4. Assumed temporal variability of annual emissions (ε_{S-1}) 5. Disregard of emissions source height (ε_{S-1}) 6. Simplified approach for calculating emissions (?) 7. The data coverage of activity statistics (ε_{I-1}) 8. Inaccurate sulphur contents and emission factors (ε_{I-1}, ε_{I-2}) 9. Inaccurate fractions of sulphur retained in the ash (ε_{I-1}, ε_{I-2})
Olivier et al. (1999a)	*EDGAR emission database* 1. Inaccuracy in activity data (ε_{I}) 2. Inaccuracy in emission factors (ε_{I}) 3. Inaccuracy in spatial distribution of emissions (ε_{I-1})
Pulles et al. (1996)	*NO_x, SO_2, CO and NMVOC in The Netherlands and Slovak Republic in 1990* 1. Inaccuracy due to (stochastic) deviations of assumed averaged values in time, space or activity (ε_{S-1}) 2. Simplifications that introduce errors or uncertainties in parameterisation (ε_{S-1}) 3. Errors in measuring results of values or parameters or inputs used (ε_{I-2})

Table 4.1 Continued.

Reference	Source of uncertainty
RIVM (1999)	*Emission inventory of air pollutants in The Netherlands* 1. Inaccuracy in statistics (ε_I) 2. Inaccuracy in emission factors (ε_I) 3. Inaccuracy due to temperature correction (ε_{S-1})
Rypdal and Winiwarter (2001)	*Emission inventories of CO_2, CH_4 and N_2O from Austria, Norway, The Netherlands, United Kingdom and United States of America* 1. Variability of emitting process in time and space (ε_{S-1}, ε_{I-1}) 2. Poor understanding of emitting processes (ε_{S-2}) 3. Missing data leading to approximations (ε_{I-1}) 4. Errors in data processing (ε_{S-3}, ε_{I-4}) 5. Model error (inadequate equations to estimate the emissions) (ε_{S-1})
USEPA (1996b)	*Emission inventory improvement program* 1. Inaccuracy due to variation from source to source (?) 2. Inaccuracy due to variation within an emitting source (?) 3. Measurement error in activity data and emission factors (ε_{I-2}) 4. Sampling (random) error in activity data and emission factors (ε_{I-2}) 5. Systematic errors (bias) (ε_{S-1}, ε_{I-1}, ε_{S-3}, ε_{I-4}) 6. Use of surrogate variables (ε_{I-1}) 7. Model simplification/over-simplification (ε_{S-1})
Van Aardenne et al. (1999)	*Anthropogenic NO_x emissions in Asia in the period 1990 – 2020* 1. Inaccuracy in activity data (ε_{I-1}) 2. Inaccuracy in emission factors (ε_{I-1}) 3. Inaccuracy in spatial distribution (ε_{I-1})
Van Aardenne et al. (2001)	*Historical emissions in the period 1890 – 1990* 1. Inaccuracy in source category specification (ε_{S-1}) 2. Inaccuracy in activity data (ε_{I-1}) 3. Inaccuracy in emission factors (ε_{I-1}) 4. Inaccuracy in spatial distribution (ε_{I-1})

4.3.2.3 Sources of inaccuracy mentioned in the literature

Table 4.1 presents sources of inaccuracy as mentioned in chapter 2 and 3 of this thesis and in a variety of emission inventory studies. The examples from table 4.1 have been interpreted and have been assigned to the sources of inaccuracy as defined in this thesis. Figure 4.2 presents the frequency counts of the appearance of sources of inaccuracy in the literature. Because Table 4.1 is not a complete overview of the emission inventory literature, Figure 4.2 should be regarded as an illustrative example rather than an indication of the relative importance of sources of inaccuracy. As with every categorisation, there will be exceptions and probably one type of inaccuracy is more obvious for an inventory developer than another type.

Figure 4.2 also shows that input value inaccuracy (ε_I) is mentioned more often as a source of inaccuracy than structural inaccuracy (ε_S). This could be an indication that the inaccuracy in the values of activity data and emission factors is considered a larger problem than the methodology that is used to calculate the emissions. The methodology to calculate the emissions is sometimes given by international agreements as is the cases in the application of default IPCC guidelines

(IPCC/OECD/IEA, 1997). In ten cases, the example of uncertainty could not be fitted into our categorisation. For example, Baars et al. (1992) mention the variance of the emissions in time and between comparable units as a source of uncertainty. We could not determine whether this variance is leading to inaccuracy because the structure of the inventory is an aggregation of the actual emitting process ($\varepsilon_{S\text{-}1}$) or that extrapolation ($\varepsilon_{I\text{-}1}$) is applied because the measurements cannot capture the variance.

As far as input value inaccuracy (ε_I) is concerned, Figure 4.2 shows that one third of the examples of input value inaccuracy do not distinguish between the different types of input value inaccuracy ($\varepsilon_{I\text{-}1,2,3,4}$). For example, RIVM (1999) mentions '*uncertainty in statistics*' as source of uncertainty. This could be an indication that these examples are used only to express the component of the emission inventory that one is uncertain about or that the underlying cause of the input value inaccuracy has not been recognized. Inaccuracy due to extrapolation ($\varepsilon_{I\text{-}1}$; ~25 times) and measurement error ($\varepsilon_{I\text{-}2}$; ~15) are the sources of inaccuracy that are mentioned very often. Inaccuracy due to unknown developments ($\varepsilon_{I\text{-}3}$) and reporting errors ($\varepsilon_{I\text{-}4}$) are mentioned less frequent. The low score for $\varepsilon_{I\text{-}3}$ could be caused by the fact that most of the studies included in Table 4.1 do not deal with emission scenarios. The likely reason that reporting errors are mentioned not often is that most emission inventory studies are subject to peer review so that reporting errors are identified and minimized in that process. Inaccuracy due to aggregation ($\varepsilon_{S\text{-}1}$) is the most common identified type of structural inaccuracy in the literature (Figure 4.2), while incompleteness ($\varepsilon_{S\text{-}2}$) and mathematical formulation error ($\varepsilon_{S\text{-}3}$) are mentioned less frequently.

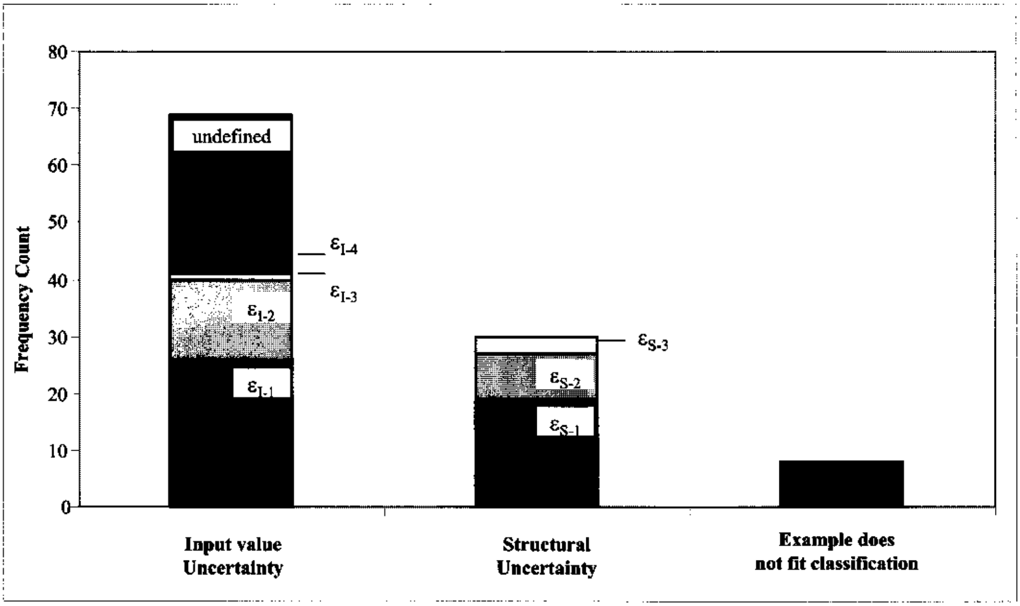

Figure 4.2 Frequency count of sources of inaccuracy (i) as mentioned in the literature (studies included in Table 4.1) and the classification of inaccuracy as presented in this thesis.

When using the categorization of scientific inaccuracy one should realize that there is a relation between the different types of inaccuracy and that the distinction between different sources of inaccuracy is not always very strict. For example, structural inaccuracy due to aggregation can be caused by lack of information or the variability of the emissions in space, time and emitting sources. This makes it impractical to calculate the emissions on the scale on which the emission process occur in reality. At the same time lack of information and variability of the emissions could lead to extrapolation of available activity data and emission factors. This means that for the same reasons (lack of information, variability) different aspects of the emission inventory become inaccurate (structure, input values).

The categorisation presented here allows for a systematic discussion of the sources of inaccuracy. It focuses on the question: *which part of the emission inventory is for what reason inaccurate?* This categorisation can furthermore be useful to determine which assessment method for the size of the inaccuracy is dealing with what source of inaccuracy. This will be shown in the next sections.

4.4 Assessment of uncertainty in emission inventories

4.4.1 Introduction

In section 4.3 we have argued that uncertainty about accuracy is caused by structural inaccuracy and input value inaccuracy. Uncertainty about reliability is dependent on the user-specified quality criteria and could be caused by lack of knowledge of accuracy or by inadequate and incomplete documentation of emission inventories. In chapter 1 we discussed that we have seen a shift from not considering uncertainty analysis towards uncertainty analysis being a focal point of attention and that several methods for assessment of uncertainty have been proposed. However, at this moment, no approach is available that:

a) Distinguishes between assessment of uncertainty with as sole purpose the estimation of inaccuracy versus the judgement whether an inventory meets specific user-defined quality criteria (accuracy versus reliability)
b) Allows for a systematic assessment of uncertainty about accuracy through both an identification, qualification and quantification of inaccuracy.

In this section we present a framework that can be applied to emission calculations, typically used in large-scale emission inventories. The framework addresses both the different types and sources of uncertainty as identified in section 4.2 and 4.3 taking into account the different tools that are available for the assessment of uncertainty. The ultimate goal of this framework is to make the accuracy and reliability of an emission inventory known.

The framework as depicted in Figure 4.3 consists of two paths that can be followed. **Path A** addresses uncertainty about reliability and results in the judgement whether the emission inventory meets the criteria that are set by the users of an emission inventory. **Path B** addresses uncertainty about accuracy through identification of the sources of inaccuracy in an inventory and through qualification or quantification of the inaccuracy of an emission inventory. In the situation that accuracy is one of the criteria set by the users of an inventory, the results from path B are used as information source for the assessment of reliability. Application of these two paths results in both an assessment of the accuracy and reliability of an emissions inventory. This may reveal how the inventory can be improved with respect to reliability and/or accuracy.

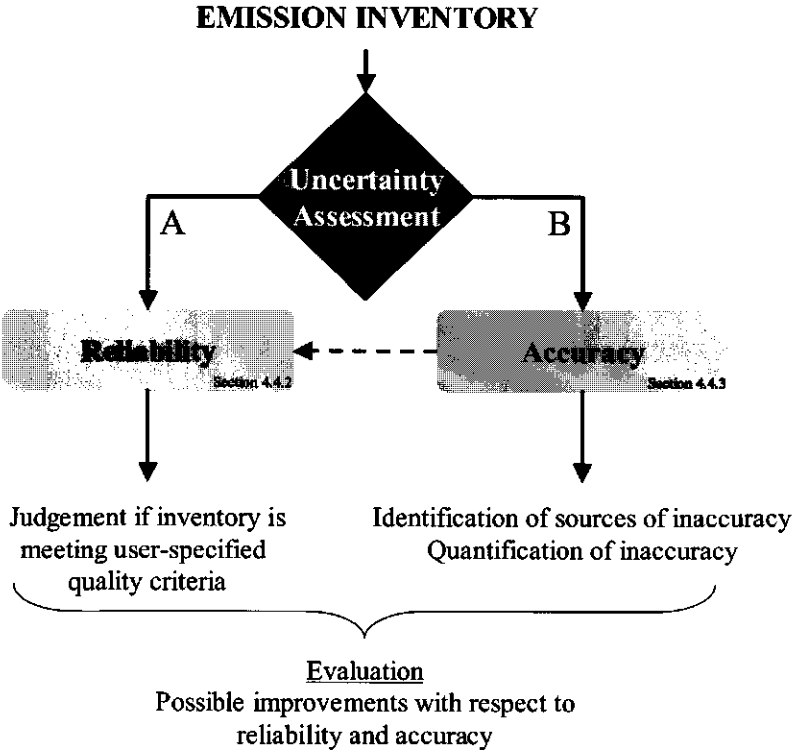

Figure 4.3 Illustration of the paths that can be followed to assess uncertainties in emission inventories. Uncertainty about reliability is assessed using path A while uncertainty about accuracy is assessed using path B. In the case where assessment of accuracy is one the quality criteria of the users of the inventory, information from path B is used as input into path A (marked in --).

4.4.2 Assessment of uncertainty about reliability (path A)

The purpose of assessing the uncertainty about reliability is to judge whether the emission inventory is meeting the user-specific quality criteria that define the reliability of the emission inventory. The outcome of this assessment can be used to improve the emission inventory when these criteria have not all been met (see figure 4.3).

The user-specific quality criteria may differ for different users of inventories. For instance, the national greenhouse gas inventories prepared for the UN Framework Convention on Climate Change must meet quality criteria as specified in the IPCC report on good practice guidance and uncertainty management (IPCC/OECD/IEA, 2000). These criteria refer to the transparency, consistency, comparability, completeness and accuracy of the inventories (see also section 4.2.2).

Other inventory users may specify different criteria, depending on the specific characteristics of the inventories and the specific purpose for which they were compiled. In general expert judgement through peer review is used to judge whether the quality criteria have been met (EPA, 1996; IPCC/OECD/IEA, 2000). Peer review is an independent review of calculations, assumptions and documentation performed by experts. It is important to realize that the assessment of accuracy of an emission inventory can also be one of the criteria for judging the quality of an emission inventory. In such a case, information from path B is used as input into path A (Fig. 4.3).

4.4.3 Assessment of uncertainty about accuracy (path B)

The purpose of assessing the uncertainty about accuracy is to provide insight in the different sources of inaccuracies (ε_S, ε_I), and if possible, a qualitative or quantitative assessment of the inaccuracy. The information from the uncertainty assessment can help the emission inventory developer to improve the most inaccurate aspects of the inventory and to communicate to the users an estimate of the extent to which the emission inventory is inaccurate. In this section several tools will be described that can be used for the analysis of inaccuracy. We distinguish between internal and external assessment of uncertainty about accuracy. In an **internal assessment** the methodology and information used to construct an emission inventory form the basis for the identification and the qualitative or quantitative assessment of the inaccuracy. In an **external assessment** information from other studies than the emission inventory itself are used to identify, qualify or quantify the inaccuracy in the emission estimates. The external information may be from atmospheric dispersion modelling, atmospheric measurements or other emission estimates.

In the literature several methods that can be applied for the assessment of uncertainty in emission inventories can be found. In our framework we have identified methods (tools) that can be applied for either internal or external uncertainty assessment. This identification of tools is based on several studies of uncertainty, sometimes with a different focus (EPA, 1996a,b and IPCC/OECD/IEA, 2000).

The Emission Inventory Improvement Program of the USA (EPA, 1996b) includes methods for qualitative, semi-quantitative and quantitative uncertainty analysis of state and national emission inventories of so-called criteria pollutants. EPA (1996b) aims at a reduction of uncertainty rather than an assessment of the 'accurate' or 'true' emission value. Qualitative uncertainty analysis is defined as the discussion of all known or suspected sources of uncertainty. The category semi-quantitative analysis refers to methods to translate a qualitative assessment of uncertainty into subjective quantitative information. Quantitative methods identified by EIIP include error propagation and techniques such as Monte Carlo analysis. Other methods include a comparison of the estimated emissions to real-world measurements, by for instance direct and indirect measurements, receptor modelling and inverse air quality modelling.

The methodologies described in EPA (EPA, 1996a) form the basis for a chapter on procedures for verification of emission inventories in the European Emissions Inventory Guidebook (McInnes, 1996). In the guidebook the term verification is used to express both accuracy and reliability of an emission inventory and three approaches for verification are mentioned. These are (i) comparison of emission estimates to alternative estimates, (ii) uncertainty estimates, and (iii) ground truth verification. Examples of data comparison are statistical comparison of aggregate emissions between countries or regions with similar population and economic status. Error propagation and the application of quality ratings are mentioned as tools for uncertainty assessment. The basis for ground verification is the comparison between emission estimates and some other known quantity that is directly or indirectly related to the emission source. One of the methods that can be applied is monitoring analysis, which compares the emission inventory result with direct measurements or indirect measurements.

The good practice guidance and uncertainty management report of IPCC/OECD/IEA (2000), aims at the construction of reliable greenhouse gas emission inventories at the national level. Two methods that can be used for the assessment of uncertainty are proposed: TIER I and TIER II. The concept of the TIER I approach is sum or squares and the concept of the TIER II approach is Monte-Carlo analysis. These techniques will be discussed below in more detail.

4.4.3.1 Internal assessment

In an internal assessment, the methodology and information used to construct an emission inventory form the basis for the identification and the qualitative or quantitative assessment of the inaccuracy ($\cup \varepsilon_i$). We distinguish six tools for internal uncertainty assessment: (i) qualitative discussion, (ii) data quality ratings, (iii) calculation check, (iv) expert estimation, (v) error propagation, and (vi) importance analysis.

(i) Qualitative discussion

The tool of qualitative discussion includes a discussion of the sources of inaccuracy that are known or expected to be occurring in an emission inventory. This can serve as a basis for a further identification of different causes of inaccuracy in an emission inventory (ε_S and ε_I, see section 4.3). It is not primarily used for the qualitative or quantitative assessment of inaccuracy. An example of a qualitative discussion is found in IPCC/OECD/IEA (2000) in the section on agriculture: *"The uncertainty in livestock population data is larger than typically recognised. There may well be systematic biases in the reporting of the livestock population to national census takers (positive and negative). The migration of livestock within or between countries may lead to double counting or under counting of some animals. Seasonal changes in populations may not be adequately reflected in annual census data"*. Another example of a qualitative discussion can be found in the work on historical emission as presented in Chapter 2. The application of this tool for internal uncertainty assessment is limited by the knowledge of the emission processes and does not allow for identification of all the sources of inaccuracy in an emission inventory. Missing sources ($\varepsilon_{S\text{-}2}$) due to the lack of information cannot be identified by a qualitative discussion.

(ii) Data quality ratings

In data quality rating, alphabetical or numerical scores are assigned to emission factors and activity data, to express the inaccuracy in a qualitative way (low versus high inaccuracy). Several types of data quality ratings exist, ranging from simple ad-hoc ratings to more systematic approaches.

An example of a simple quality rating has been presented in chapter 2. The activity data are qualified as being *acceptable* (national and international activity data), *poor* (extrapolation of activity data or *very poor* (no activity data, application of proxy). The emission factors have been qualified as *detailed* (country specific), *regionally aggregated* (aggregation of detailed emission factors), or *globally aggregated* (aggregation of regional factors or application of a single emission factor). Based on these ratings we judged emission estimates based on application of *globally aggregated* emission factors and *very poor* activity data as most uncertain. The application of *acceptable* activity data to *detailed* emission factors provides probably the best emission estimate possible.

There are more systematic approaches for the performance of data quality rating. In EPA (1996b) the Data Attribute Rating System (DARS) is mentioned as the preferred method for EIIP activities and the AP-42 emission factor rating system is mentioned as alternative. DARS and AP-42 are tools that are often used in emission inventory studies and a description is presented below.

DARS

The Data Attribute Rating System DARS is based on the assignment of numerical scores to emission factors and activity data in order to provide an overall confidence rating for the emission inventory. The DARS approach has been derived from procedures to compare household products on a number of attributes (Beck, personal communication, November 1999). The purpose of DARS is to identify the weak and strong parts of an emission inventory in relation to the accuracy of the emission estimate. Its principle is to evaluate four different attributes that are believed to influence the accuracy of the emission estimate. These attributes are (i) measurement technique, (ii) source specificity, (iii) temporal agreement and (iv) spatial agreement. For each of these four attributes a score from 1 to 10 is assigned with a score of 10 being the highest. By defining four attributes, a systematic discussion of the sources of inaccuracy in emission inventories becomes possible. The use of relative scores for emission factors and activity data allows for the user to distinguish relative accurate from inaccurate components in the inventory (high versus low DARS score). This can be used to prioritise emission inventory improvement. DARS can be used to identify uncertainty in emission inventories in relation to input value inaccuracy such as extrapolation error (ε_{I-1}) and measurement error (ε_{I-2}). One of the limitations of DARS is that it only provides relative scores and no absolute assessment of inaccuracy. Moreover, the assignment of scores is to a certain extent subjective. Therefore, DARS must be considered a qualitative tool for assessment of inaccuracy.

AP-42's emission factor rating

AP-42 stands for Compilation of Air Pollutant Emission Factors, and it is the main emission factor reference in the U.S.A. (EPA, 1995). Each emission factor is given a 'factor rating' to indicate its accuracy or appropriateness. The methods that have been use to develop the emission factors are given scores based on an A to D ratings on the emission measurements with A being the best ("sound methodology" (EPA, 1995) and D the worst ("generally unacceptable method" (EPA, 1995). The scores have been combined with an evaluation on the representativeness of the emission factor. The result is an A to E rating system. A score of 'A' means that the emission factor is based on A to B rated emission measurements and that the emission factor is representative for the source category for which the emission factor has been developed. The lowest score of 'E' means that the emission factor is based on C to D rated emission measurements and that the factor is probably not representative for the specific source category.

The AP-42 rating system can be used to identify inaccuracy in emission inventories in relation to input value inaccuracy such as extrapolation error (ε_{I-1}) and measurement error (ε_{I-2}) and to prioritise the improvement of the emission inventory. As is the case with DARS, AP-42 is a qualitative tool for assessment of inaccuracy.

(iii) Calculation check and evaluation of mathematical formulation

A calculation check (EPA, 1996a) can be used to assess inaccuracy in the emission inventory resulting from typing errors or other human errors in for example spreadsheets. Application of this straightforward method for assessment of inaccuracy will result in the reduction of uncertainty about accuracy because it allows for repairing errors that are found in the calculation. A calculation check may also reveal inaccuracies due to incorrect mathematical formulation, when an evaluation of the equations and assumptions is part of the check. A full assessment of inaccuracies associated with mathematical formulation will, however, be difficult to achieve, because these inaccuracies may in part be caused by incomplete understanding of processes involved.

(iv) Expert judgement

This assessment consists of asking experts to give a qualitative or quantitative assessment of either the overall inaccuracy of an emission inventory, or the structural (ε_S) or input value inaccuracy (ε_I). It is different from other tools for assessment of inaccuracy because the estimates are the result of reasoning by the expert rather than the application of systematic approaches such as error propagation, importance analysis or the tools for external assessment. Expert judgement is often used when other methods can not be used due to lack of information or methods. Examples are Van Amstel (1999) on greenhouse gas emissions in The Netherlands and RIVM (1999) on different air pollutants and sectors in The Netherlands. The expert judgement is based on personal expertise or combination of expertise from several experts on the emission of a specific pollutant or specific source category. The problem with expert judgement is that often a clear rationale for the quantitative estimate of inaccuracy is missing. When more advanced methods for expert judgement such as expert panel or Delphi methods are applied the rationale for the estimation can be made visible. There is a relation between expert judgement and other tools for uncertainty assessment. The results of an expert judgement are sometimes used as input in error propagation and importance analysis. Furthermore, the results from applying other tools uncertainty assessment can be used as information for the expert to provide his estimate of inaccuracy.

(v) Error propagation

Based on Morgan and Henrion (1990), error propagation in emission inventories can be seen as the calculation of inaccuracy ($\cup\varepsilon_i$) in an inventory that is caused by the inaccuracies in the input values (ε_l). The principle of error propagation is that the emission estimate and its inaccuracy are treated from a probabilistic perspective. This means that in equation (1), the inaccuracy is seen as a variation around its mean value $E_{inventory}$. Two approaches can be used: (1) sum of squares and (2) distribution sampling.

Sum of squares

Using the sum of squares, the variance of the output is estimated as the sum of the squares of the contributions from each input (Morgan and Henrion, 1990). This means that the inaccuracy is estimated as the sum of squares of each inaccuracy in the emission inventory. This principle has been applied by Benkovitz and Ogden (1985) and extended by Langstaff and Wagner (1986) in an assessment of uncertainty in the 1980 USA National Acid Precipitation Assessment Program. In their study Langstaff and Wagner (1986) mention that the sum of squares approach uses the assumption that the emission inventory parameters (activity data and emission factors) are independent and that they are random variables that are approximately normally distributed. Furthermore they conclude that the usefulness of the method depends on the quality of the data available to characterise the variability (inaccuracy). The approach of sum of squares has been adopted by IPCC/OECD/IEA (2000) as one of two standard methodologies for assessment of scientific uncertainty (the so-called TIER I method). The TIER 1 method has been used by Olivier et al. (2001) for the greenhouse gas emission inventory of The Netherlands for the period 1990-1999.

Distribution sampling

The most common used approach for error propagation is distribution sampling, by which the inaccuracy of a certain emission inventory parameter is expressed through a probability density function (PDF). This PDF is a representation of the best available knowledge of the range of values (variance) of an inaccurate parameter. Several publications provide guidance on how to perform this type of error propagation that is also known as Monte Carlo analysis (Morgan and Henrion, 1990; Janssen et al., 1992; NCRP, 1996; EPA, 1997; IPCC/OECD/IEA, 2000). The method of distribution sampling does not require conditions such as normality of the variance or independent emission factors and activity data, as is the case in sum of squares. By sampling over the probability density functions a set of input values is generated which is used to propagate the uncertainty about inaccuracy through the emission inventory calculation. Several techniques for sampling can be used, but the most common techniques are random sampling and Latin Hypercube Sampling. By random sampling a value is randomly selected from the probability density function while in Latin Hypercube Sampling the probability density function is first divided into equiprobable intervals followed by a sampling of each of these intervals (Morgan and Henrion, 1990). The advantage of this stratified way of sampling is that the number of samples needed for a good representation of the mean and variance of the probability density function is smaller than in random sampling. Several studies on uncertainties in emission inventories have applied distribution sampling as technique for quantitative assessment of inaccuracy (e.g. AEAT, 1998; Van Aardenne et al., 1998; Frey et al., 1999; Brown et al., 2001, Winiwarter and Rypdal, 2001). Furthermore, distribution sampling has been adopted by IPCC/OECD/IEA (2000) as one of two standard methodologies for quantification of uncertainty in national greenhouse gas inventories (the so-called TIER II method). As for error propagation, the usefulness of distribution sampling in the assessment of inaccuracy depends on the information available to construct the PDF for each uncertain parameter. Several studies indicate that because of lack of empirical data assumptions needed to be made to define PDFs (e.g. Van Aardenne et al., 1998; Brown et al., 2001; Winiwarter and Rypdal, 2001).

(vi) Importance analysis

Importance analysis is the calculation of the relative importance of inaccuracies in emission inventory parameters (ε_I) in the overall inaccuracy of an emission inventory (based on Morgan and Henrion, 1990). Importance analysis is often applied after the performance of error propagation (e.g. Van Aardenne et al., 1998; Brown et al., 2001). There are several statistical methods to determine the relative importance of inaccurate parameters (e.g. Janssen et al., 1990 or Saltelli, 1999). The basis of an importance analysis is the selection of a measure that relates the overall inaccuracy of the emission inventory to the inaccuracy of the individual components of an emission inventory. Two commonly used approaches are (i) Spearman's rank correlation coefficient and (ii) Standardised regression coefficient determined through multiple linear regression analysis. The Spearman's correlation coefficient is a measure of the monotony between the overall inaccuracy of an emission inventory and the inaccurate emission inventory component (Janssen et al., 1990). It is a relative measure that can be applied under the assumption that there no correlations exist between the different uncertain variables. The standardised regression coefficient expresses the linear contribution of an inaccurate inventory parameter to the overall inaccuracy under the condition that the different inaccurate parameters are not correlated (Janssen et al., 1990). Based on the value of the standardised regression coefficient, the absolute contribution of each uncertain parameter to the inaccuracy in the emission estimate can be calculated.

4.4.3.2 External assessment

In an external assessment of inaccuracy, the differences between the emission inventory and other, independent, information is used to identify or quantify inaccuracies in the emission inventory. As discussed in section 4.2.1, the emission that has actually occurred (E_{real}) needs to be known to determine the inaccuracy ($\cup \varepsilon_i$) of an emission inventory ($E_{inventory}$). In an external assessment, the external emission estimate ($E_{external}$) is treated as an independent estimate of the real emission and the inaccuracy of the emission inventory can be estimated from the difference between $E_{external}$ and $E_{inventory}$. Given the fact that E_{real} is unknown by nature, the strength of external assessment tools largely depend on our confidence in $E_{external}$ as an indicator of the real world emissions. Four tools for the external assessment of inaccuracy are distinguished here: (i) comparison with other emission inventories, (ii) comparison with direct or indirect measurements, (iii) performance of forward air quality modelling studies, (iii) comparison with results from inverse modelling studies.

(i) Comparison with other emission inventories

The difference between one emission estimate and another can be used as indication of the inaccuracy of both estimates. The two emission inventories need to be independent from each other. The reason for this is that agreement is easily found between two emission estimates when the same activity data and emission factors are used. Usually the causes and size of inaccuracy are not available for both estimates. Without this information, the difference between the two estimates can be caused by inaccuracies in either emission inventory. Although inventory comparison will in general not reveal the inaccuracy of emission estimates, it can be used to point the attention to parts of the inventories that show large differences. This approach has been applied by Van Amstel et al. (1999). In their study, national emission estimates of greenhouse gases for the year 1990 from the EDGAR database have been compared to National Communications of several countries. In some cases the reasons for the differences were clear and led to conclusions about either the EDGAR or national emission estimates (e.g. different emission factors, different activity data, gaps in inventories). In many cases, the differences are a basis for further research. The work by Van Amstel et al. (1999) shows that comparison between inventories can be used to identify sources of inaccuracy related to method (ε_s) or data (ε_s). However, it does not allow for a full assessment of accuracy. The reasons for this are that (i) in fact we compare two inaccurate inventories, and (ii) it is almost never entirely clear to what extent the two estimates are really independent. The same problem is present in the comparison of the results of the historical emission database presented in chapter 2 to other historical emission estimates.

(ii) Comparison with direct or indirect measurement

As argued in section 4.2.1 the accuracy of an emission inventory can only be assessed through accurate and continuous emissions monitoring of a single sources. The difference between the accurate measurement and the emission estimate can give insight in the accuracy of the emission estimate or the accuracy of a specific emission factor or activity. Due to the fact that emissions are variable between sources, and in space and time, continuous emission measurements are impractical and expensive. A good overview of different types of measurements and the application for assessment of accuracy of emission inventories is provided by EPA (1996a). Basically, the measurements of emissions can be either direct or indirect.

Direct measurements

Direct measurements are taken directly at the emitting source through either continuous emission monitoring or monitoring during restricted time periods. Measurements over restricted time periods determine the emissions during specific periods that are considered to be representative for the emitting process. An alternative to direct emission measurements is the measurement of emission process conditions. When it has been proven that emission estimates can be derived from the operating conditions, the measurement of these operating conditions can provide accurate information on the emissions.

Indirect measurements

Indirect measurements can be an alternative to direct emission measurements. For instance, a comparison of the emission estimates with the results of indirect air quality measurements can be used as an indicator for inaccuracy of the emission estimate. Indirect measurements are not taken directly at the emitting source. For example, ambient measurement may include sampling of air down-wind and up-wind of an emission source. The difference between the down-wind and up-wind concentration is caused by the emission of the air pollutant. Often the interpretation of these measurements requires the use of atmospheric models. Another example is remote sensing. Remote sensing through LIDAR (light detection and ranging) measures concentration of an air pollutant along a specific path through spectroscopy.

(iii) Forward air quality modelling

In forward air quality modelling an emission inventory is used as input into an atmospheric dispersion model which calculates the atmospheric concentration of the pollutant. When accurate atmospheric concentration measurements are available, and the model is considered accurate, the difference between model result and observation can be used as an indicator for the inaccuracy of the emission inventory. An example of such a study is the work performed by Iversen (1993). Iversen attempted to diagnose model error, emission error or inaccurate measurement as cause for the difference between EMEP/MSC-W acid deposition model calculations and EMEP measurement network observations for NO_2, SO_2 and sulphate in the period 1985-1989. Iversen used (i) scatter plots of measured versus calculated concentrations, (ii) comparison of yearly averaged modelled and measured concentrations, (iii) comparison of both measured and modelled concentrations with emissions estimates per grid cell, (iv) and calculation of variation in measured concentrations from year to year.

Iversen concluded that the SO_2 emissions from the inventory were probably overestimated, but that for some measurement locations, it was not clear whether measurement errors or inaccurate emission estimates were responsible for the deviation between model and measurement. However, the results of the study did not allow for a quantification of the overall inaccuracy of the emission inventory. This example illustrates that although forward air quality modelling provides insight in the accuracy of emission estimates, it is difficult to distinguish between inaccuracies in the model, the measurements and the emission inventory. Forward air quality can be applied on different temporal and spatial scales. The appropriate scale depends on the type of atmospheric dispersion model, the spatial and temporal resolution of the atmospheric concentration measurements, and the chemical and physical characteristics of the compounds considered.

(iv) Inverse air quality modelling

In inverse modelling atmospheric concentrations are used as input into an atmospheric dispersion model to calculate the emissions needed to reproduce the observed concentrations. Comparison of the 'back-casted' emission estimates with the emission inventory can be used as indicator for the accuracy of the emission inventory. Inverse modelling studies have been applied both on a global, regional, national, and local scale. For instance, Hein et al. (1997) used inverse modelling by a three-dimensional transport model to analyse the global methane budget. They describe inverse modelling as an optimisation problem, minimising the difference between calculated and observed concentrations. The solution to this optimisation provides emission patterns that result in an optimal agreement between calculated and observed concentrations, the so-called 'back-casted' emission estimates. An example of regional inverse modelling is the work by Seibert (2000) on sulphur emission in Europe. One a national scale Vermeulen et al. (1999) have used inverse modelling to estimate methane emissions in several countries in NW-Europe including the Netherlands. A problem in application of inverse modelling can be that a good reconstruction of the emissions is hampered by the accuracy of the dispersion model and representativeness of the atmospheric measurements (Seibert, 2000). Moreover, information is needed on the accuracy of the emission estimates to constrain the a-priori information on the emissions in the optimisation problem (Houweling, 2000).

4.5 FRAULEIN: FRamework for the Assessment of Uncertainty in Large-scale Emission INventories

In the previous sections of this chapter two types of uncertainty in emission inventories have been defined (section 4.2) followed by an overview of sources of uncertainty about reliability and accuracy (section 4.3). In section 4.4 a distinction between the assessment of uncertainty about accuracy and reliability has been presented together with an overview of different methods that can be used for this purpose. In this final section the information on types and source of uncertainty and the characteristics of methods for uncertainty assessment are combined into a (framework) for uncertainty assessment.

The different methods for assessment of uncertainty about reliability and uncertainty about accuracy have different capabilities to identify, qualify or quantify the uncertainty. Some methods are only able to judge whether quality criteria have been met (peer review) while other tools are only able to estimate the overall inaccuracy of an inventory and not of the inventory components (e.g. inverse modelling). The capabilities of the different methods are presented below and a schematic overview of which method can be used for which part in the assessment of inaccuracy is presented in Figure 4.4. This combined information forms a FRamework for the Assessmnet of Uncertainties in Large-scale Emission INventories (FRAULEIN). The framework may help to identify the appropriate methods for uncertainty assessment.

Path A: assessment of uncertainty about reliability

The assessment of uncertainty about the reliability of emission inventories, depends on quality criteria defined by the user of the inventory, which in turn are associated with the specific purpose of the inventory. For emission inventories that are used for policy purposes usually a set of quality criteria can be, or have been, defined. The approach for assessment of reliability depends on these criteria as defined by the users of the emission inventory. Reliability or the extent to which the inventory is meeting the quality criteria can be assessed through peer review. This method is an independent review of the inventory and results in the judgement whether the inventory meets the criteria or not and for what reason the criteria are not met. For emission inventories prepared for scientific purposes, the accuracy of the emission inventory is typically considered the most important (if not only) quality criterion. In such cases,, information from Path B is needed in order to judge to what extent the inventory is inaccurate.

Path B: assessment of uncertainty about accuracy

Following path B results in the identification or qualification or quantification of the uncertainty about accuracy in an emission inventory. The different tools for the assessment of inaccuracy have different capabilities as discussed below and presented in Figure 4.4. Here, a distinction is made between identification of different sources of input value inaccuracy (ε_I) and structural inaccuracy (ε_S), the qualification or quantification of inaccuracy caused by input value inaccuracy (ε_I) or structural inaccuracy (ε_S), and the qualification or quantification of the inaccuracy of the emission inventory ($\cup\, \varepsilon_i$).

Tools for internal assessment of inaccuracy

A qualitative discussion of sources of inaccuracy in an emission inventory can result in identification of the different sources of inaccuracies in input values (ε_I) and structural inaccuracy (ε_S). One could argue that qualitative discussions will not identify incompleteness of inventories due to lack of knowledge ($\varepsilon_{S\text{-}2}$).

Expressing the relative accuracy of emission factors and input data by assignment of alphabetical or numerical scores (Data quality rating) results in identification of inaccurate input vales (ε_I) that are caused by extrapolation error ($\varepsilon_{I\text{-}1}$) or measurement error ($\varepsilon_{I\text{-}2}$). It can also result in a qualification of those aspects of an inventory that are expected to be more inaccurate than other parts.

Performing a calculation check on the emission inventory could result in identification of errors in the equations of the inventory (e.g. plus instead of minus; $\varepsilon_{S\text{-}3}$) or in the identification of reporting errors ($\varepsilon_{I\text{-}4}$). An evaluation of the mathematical formulation may reveal errors due to inappropriate formulation ($\varepsilon_{S\text{-}3}$). However, these checks will not identify errors ($\varepsilon_{S\text{-}3}$) that are caused by incomplete understanding.

Asking experts to give a qualitative or quantitative estimate of the inaccuracy in input values (ε_I), structure (ε_I) or overall inaccuracy ($\cup\varepsilon_i$) of an emission inventory (expert judgement) results in both an identification, qualification and quantification of inaccuracies. The basis of the expert estimate is the available knowledge of one expert or a group of experts.

Calculation of inaccuracy in the emission inventory induced by inaccuracy in input values (error propagation can be used to quantify the inaccuracies in emission inventories as result of different input value uncertainties (ε_I). The results of error propagation can be used to qualify whether there is high or low inaccuracy with the input values of an emission inventory. Although in practice seldom used for that purpose, error propagation can in theory be used to address structural inaccuracy (ε_S).

The results from error propagation are used to calculate the importance of inaccuracy in input values (importance analysis). This tool allows for both a qualification and quantification of those input values (ε_I) that have a large contribution to the inaccuracy of an emission inventory.

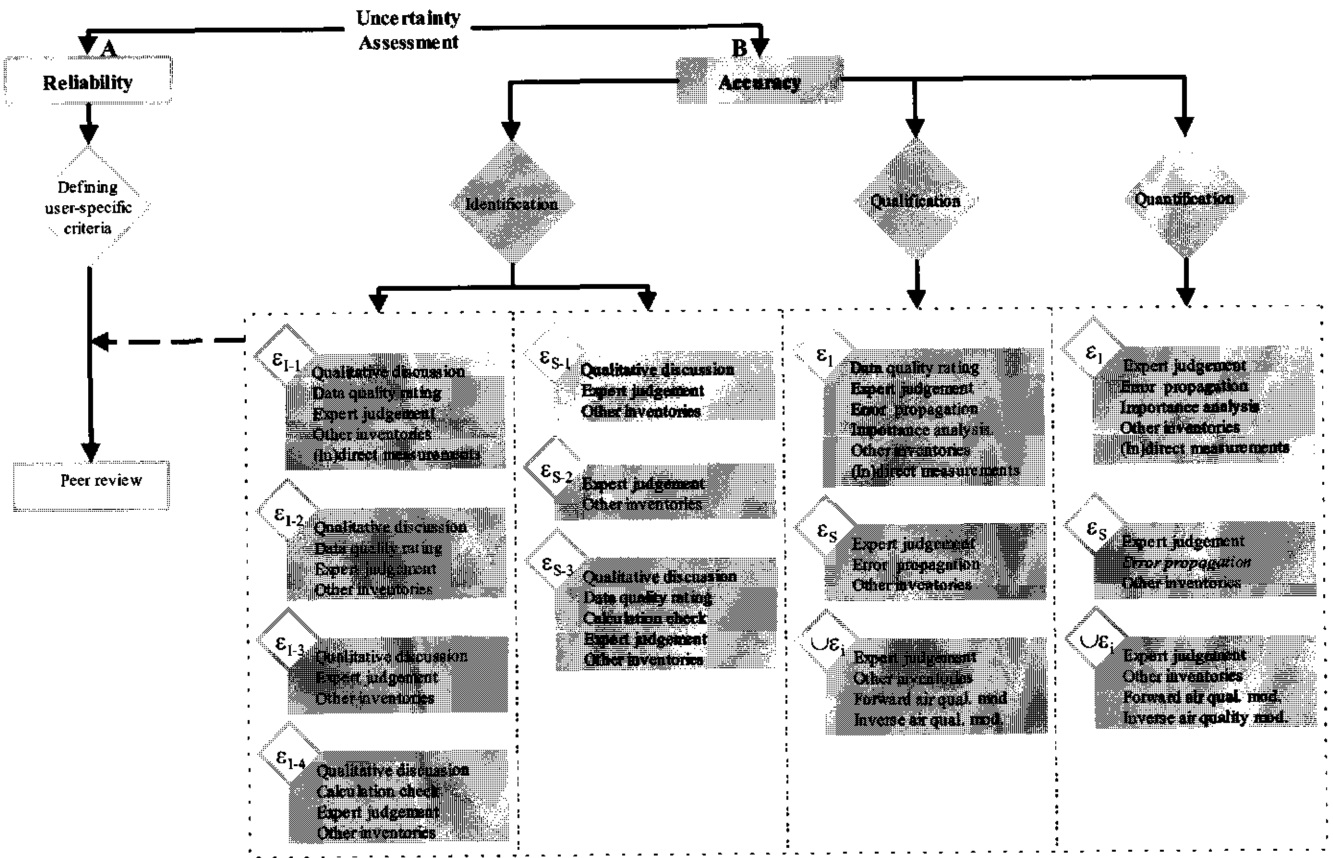

Figure 4.4 A systematic framework for the assessment of uncertainty in emission inventories. Uncertainty about reliability is assessed using path A while uncertainty about accuracy is assessed using Path B. In the case where the assessment of accuracy is one of the quality criteria of the users of the inventory, information from path B is used as input into path A (marked - -). See the text for a description of ε_{x-y}.

Tools for external assessment of inaccuracy

The comparison of an emission inventory with another independent emission inventory (other inventories) could be used to identify possible inaccuracies associated with input values (ε_I) or the structure (ε_S) of an emission inventory. When the accuracy of the independent emission inventory is known, qualification and quantification of ε_S, ε_I and $\cup\varepsilon$ becomes possible.

The comparison of input values of an emission inventory with accurate measurements ((in)direct measurement) allows for the qualification and quantification of the inaccuracy of input values (ε_I) or emission inventory ($\cup\varepsilon_i$). The sources of inaccuracy that can be identified by application of this tool are limited to inaccuracy due to extrapolation ($\varepsilon_{I\text{-}1}$) and measurement error ($\varepsilon_{I\text{-}2}$).

Comparing modelled atmospheric concentrations that have been calculated using an emission inventory with atmospheric concentrations measurements (forward air quality modelling) could be used to qualify and quantify the overall inaccuracy of an emission inventory ($\cup\varepsilon_i$). A prerequisite, however, is that the accuracy of atmospheric concentration measurements and the model is known.

Comparison of an emission inventory with emissions calculated by using atmospheric concentrations as input into an atmospheric dispersion model (Inverse air quality modelling) allows for the qualification and quantification of the overall inaccuracy of the emission inventory ($\cup\varepsilon_i$). Just as with forward air quality modelling both the model and measurement should be accurate.

Final remarks

The framework for uncertainty assessment can be used for a systematic analysis of the uncertainty in emission inventories. It combines information from different fields of study such as other emission inventories, atmospheric modelling, air quality measurements and statistics. The framework can be used to perform a four-step assessment of inaccuracy: identification, qualitative assessment, quantitative assessment, and prioritising further research. Starting point of the four-step approach is the framework as shown in Figure 4.4 which presents a complete overview of the use of all tools in the various parts of the analysis. From Figure 4.4 it is clear that most tools can be used in more than one part of the assessment. Therefore, in the four-step approach we have chosen to assign each tool to one specific step only. This choice was based on our understanding of the tools and their use in practice in the literature. Obviously this does not lead to a unique choice, but we judge that the four steps and the tools assigned to each of them will help scientists and practitioners in achieving a good assessment of uncertainty about inaccuracy in emission inventories.

Step I: Identification

In the first step major causes of inaccuracy can be identified using expert views, qualitative discussion and other inventories. These three tools will enable the user to identify inaccuracies in the structure and in the input values of the emission inventory. In this step both uncertainties in sources of emission and estimation method can be traced.

Step II: Qualification

The second step takes the analysis to a qualitative assessment and eventually a ranking of the inaccuracies. This takes us one step further in the direction of full quantification of the inaccuracy. The tools used in the first step may be used again in an attempt to derive more than just identification from them. In addition the tools data quality ranking and direct and (in)direct measurements can now be used.

Step III: Quantification

The third step aims at a full quantification of the inaccuracies. Again, tools used in earlier steps may be stretched to gain a beginning of an insight in the quantitative value of the inaccuracies. In addition, the tools error propagation, importance analysis and forward and inverse air quality modelling can be used.

Step IV: Evaluation

The nature this final step of the assessment differs from earlier steps. Whereas steps 1-3 gradually improve the insight in the cause and size of the uncertainty about accuracy, step 4 uses these results to assist in prioritising future research. The most important uncertainties could then be reduced first, taking into account that it is sometimes very difficult if not impossible to reduce a large uncertainty.

There is one question that remains unanswered at this point: the framework does not discuss the advantages or disadvantages of the different methods in great detail. In the following chapters, some of the tools are applied in practice in two case studies. Based on the results of these analyses we can discuss the advantages and disadvantages of some (combinations of) tools for the assessment of inaccuracy. The first case study includes application of tools for internal assessment of inaccuracy (error propagation and importance analysis (chapter 5). The second case study combines information from atmospheric measurement, atmospheric dispersion modelling and emission inventories (chapter 6).

Chapter 5

Internal assessment of inaccuracy

Error propagation and importance analysis in practice - a case study of N_2O emissions from agriculture

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J.A. van Aardenne, C. Kroeze, M.P.J. Pulles and L. Hordijk. Uncertainties in the calculation of agricultural N_2O emissions in The Netherlands using IPCC Guidelines: In: J. van Ham et al (Eds.) Non-CO_2 greenhouse gases: scientific understanding, control and implementation. Kluwer Academic Publishers, pp. 493-498, 2000.

5.1 Introduction

Air pollutants are emitted from a variety of individual sources. Anthropogenic emissions of the greenhouse gas nitrous oxide (N_2O), for instance, are caused by agricultural practices (animal husbandry, fertiliser), fossil fuel combustion, waste treatment and industrial processes (Mosier et al., 1998). Most of these emissions show large spatial and temporal variability, so that it is practically impossible to measure each emission source individually. Quantifying emissions at relatively high aggregation levels (e.g. for national emission inventories) is therefore often based on so-called emission factor approaches. These emission factor approaches aggregate information on sources, in time and space and combine this information with emission factors that are often extrapolations of existing data. Consequently, the resulting emission estimates are inevitably inaccurate representations of the actual emissions. Although we know that emission inventories are inaccurate, we do not know exactly the sources and size of these inaccuracies.

There are different methods available for the assessment of the sources of inaccuracy in an emission inventory and to identify, qualify or quantify inaccuracies in emission inventories. We distinguish between internal and external assessment of inaccuracy (chapter 4). In an *internal assessment* the methodology and information used to construct the inventory form the basis for the assessment of inaccuracies. In an *external assessment* information from other studies than the emission inventory itself is used as a basis for the assessment of inaccuracies. In this paper we focus on an internal assessment of the inaccuracies in a national inventory of N_2O emissions. Two of the tools that can be used for the performance of an internal assessment of inaccuracies are error propagation and importance analysis. *Error propagation* is the calculation of inaccuracy in an inventory that is caused by the inaccuracies in the inventory parameters. *Importance analysis* is the calculation of the relative importance of inaccuracies in emission inventory parameters in the overall inaccuracy of an emission inventory (based on Morgan and Henrion, 1990). Several studies have shown the usefulness of error propagation and importance analysis in analyses of the uncertainties in large-scale emissions inventories that are based on emission factor calculations (e.g.

Benkovitz and Ogden, 1985; AEAT, 1998; Van Aardenne et al., 1998; Brown et al., 2001 and Winiwarter and Rypdal, 2001). Statistical uncertainty analyses have proven to be useful in prioritising future research aiming at reducing the uncertainties in emission inventories. However, the statistical information on individual emission inventory components (emission factors and activity data) needed to perform error propagation and importance analysis is often limited or not always well defined. This could influence the applicability of error propagation and importance analysis in practice.

Emissions of N_2O are not easily quantified at the national level. Nevertheless, countries that are party to the UN Framework Convention on Climate Change are expected to report their national emissions on a regular basis. The Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC/OECD/IEA, 1997) have been developed to assist countries in preparing their emission inventories, and include default methods to estimate national emissions. These guidelines also include a default methodology for estimating N_2O emissions from agriculture. As mentioned above, agricultural emissions of N_2O are relatively uncertain. Moreover, country-specific information on N_2O is limited. As a result, many countries use the Revised IPCC Guidelines for the emission calculations.

This study focuses on an assessment of the inaccuracies in estimates of agricultural N_2O emissions that are based on the Revised IPCC Guidelines described by Mosier et al. (1998). This will be done through error propagation and importance analysis. To this end, probability density functions of the inventory parameters (emission factors and activity data) are needed.

The IPCC default values for emission factors and other inventory parameters have been described by Mosier et al. (1998). They also provide uncertainty ranges for the most important emission factors. These default values and uncertainty ranges were defined by an IPCC Expert Group, and therefore largely based on expert judgement. The information on these IPCC default values and uncertainty ranges is not sufficient for the performance of error propagation and importance analysis for a number of reasons. First, specific probability density functions for inventory parameters are not available. Second, it is unclear whether the IPCC "default" values should be interpreted as the average value (mean), the most probable value (modus) or as the median value. Furthermore, the range of uncertainty can be interpreted either as absolute minimum and maximum or as the boundaries of a confidence interval. As a result, a choice has to be made between the possible interpretations when performing an uncertainty assessment through error propagation. For instance, Brown et al. (2001) assumed that the default value is the most likely value (mode) and that the range of uncertainty reflects the absolute minimum and maximum of a given parameter. Based on this, Brown et al. (2001) used a beta-pert distribution for their uncertainty assessment.

As discussed above, it is possible to interpret the IPCC default values and accompanying uncertainty ranges for inventory parameters in various ways, leading to different probability density functions needed for the performance of the error propagation and importance analysis. The question arises whether the results of the assessment of inaccuracies would be influenced by these interpretations. In this study, we have analysed whether the statistical interpretation of the IPCC Guidelines affects the assessment of the inaccuracy or the conclusions about the relative contribution of inventory parameters to the inaccuracy of the emission estimate as a results of inaccuracies in the input values.. We have focussed on the estimates of N_2O emissions from agriculture in The Netherlands in 1990 calculated using the Revised IPCC Guidelines. Section 5.2 describes the application of the IPCC method to calculate the N_2O emission. Section 5.3 presents the methodology for performance of error propagation and importance analysis. In section 5.4 the results are discussed and in section 5.5 we will present our conclusions.

5.2 IPCC default methodology to calculate N_2O emissions from agricultural soils

The Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC/OECD/IEA, 1997) provide a default methodology to calculate the emissions of N_2O from agriculture on a national scale. The methodology as described by Mosier et al. (1998) includes three sources of N_2O: (i) direct emissions from agricultural fields, (ii) emissions from animal production and (iii) indirect emissions. **Direct** emissions are increased N_2O fluxes from agricultural soils resulting from application of fertiliser, animal waste, or crop residues, an increase in biological N-fixation or the cultivation of organic soils. Emissions from **animal production** are from animal waste management systems. These N_2O emissions are a function of the number and type of animals, the amount of nitrogen (N) excretion per animal type and the type of waste management system. **Indirect** emissions are N_2O emissions induced by agricultural activities but in fact taking place at remote sites. These can result from (a) atmospheric deposition of agricultural nitrogen oxides (NO_x) and ammonia (NH_3), (b) trough leaching and runoff of agricultural N to aquatic systems, or (c) from human consumption of N containing food, followed by sewage treatment.

The equations, variables and parameters used to calculate N_2O emissions from agriculture in are presented in Box I. We define variables as model input values which vary over time that are relatively well known for the specific situation (e.g. number of dairy cattle). Parameters are model values that do not vary over time but show variability due to uncertainty (for example the emission factor for direct soil emissions EF_1). In order to calculate N_2O emissions from agriculture in the Netherlands in 1990, the input variables were taken from literature and parameter values from Mosier et al. (1998). Table 5.1 and Table 5.2 presents these input variables, parameter values and the references for these values. The table also provides information on inaccuracy, which will be discussed in section 3. Using the input data as shown in Tables 5.1 and 5.2 a and the default IPCC methodology as described in Box I, the N_2O emissions from agriculture in The Netherlands in 1990 are calculated to be 30 Gg N. Direct emissions account for 13 Gg N, emissions from animals are 5 Gg N and 12 Gg N is resulting from indirect emissions.

Table 5.2 Values of AWMS(T): Fractions of N excretion (NEX) in 1990 that is managed in the different Agricultural Waste Management Systems (AWMS) for different animal types (T), as used for calculations of 1990 N_2O emissions from agriculture in The Netherlands (Source IPCC, 1997).

	Anaerobic lagoon	**Liquid systems**	**Daily spread**	**Solid, storage, dry lot**	**Pasture, range, paddock**	**Used as fuel**	**Other systems**
Dairy Cattle	0	0.46	0.24	0.21	0.08	0	0.01
Non-dairy cattle	0	0.55	0	0.02	0.33	0	0.09
Poultry	0	0.13	0	0.01	0.02	0	0.84
Sheep	0	0	0	0.02	0.87	0	0.11
Swine	0	0.77	0	0.23	0	0	0
Other	0	0	0	0	0.96	0	0.04

5.3 Assessment of inaccuracies in N_2O emissions from agricultural soils in The Netherlands in 1990

The concept of uncertainty assessment through error propagation and importance analysis is described in section 4.4. In our analysis we have performed error propagation through application of Latin Hypercube Sampling (LHS) using the statistical software package Analytica® (Lumina, 1999). Results of the error propagation have been used as input into an importance analysis on the 23 inaccurate IPCC parameter through calculation of the Spearman's rank correlation coefficient using Analytica followed by calculation of the standardised regression coefficient using SPSS.

To perform a LHS, probability density functions (PDFs) have to be defined. An overview of commonly used probability density functions and a description of their important characteristics are shown in Table 5.3. As far as continuous probability density functions are concerned we distinguish between Uniform, Triangular, Normal, Lognormal, Exponential, Gamma, Weibull and Beta distributions.

For the assessment of inaccuracy in the 1990 N_2O emission estimates for The Netherlands using the default IPCC Guidelines, information is needed on the input variables, parameter values and their inaccuracies. We assume that the input variables are relatively well known from national and international statistics and that the inaccuracies in these values can be considered negligible compared to the inaccuracy in the IPCC default parameter values. To determine the PDF for the parameters distinguished in the IPCC methodology, we have used the default values and uncertainty range as presented in Table 5.1.

For two parameters ($EF_{3\text{-daily spread}}$) and $Frac_{GRAZ}$ we did not define an uncertainty range. For $EF_{3\text{-daily spread}}$ the default value is zero and for $Frac_{GRAZ}$ information was taken from another source than IPCC default guidelines and we assumed arbitrarily no range of uncertainty.

Of the parameters distinguished in the IPCC methodology 23 will be used in the statistical uncertainty analysis.

Table 5.3 Brief description of most common used probability density function (PDF) types as distinguished by Morgan and Henrion (1990) and Lumina (1999).

PDF type	**Description / usage**
Uniform	A Uniform distribution can be selected for an uncertain quantity when nothing is known excepts its bounds.
Triangular	Applying a Triangular distribution can be appropriate when values towards the middle of the uncertainty range are more likely to occur than values near extremes. Using the triangular distribution can be a way to communicate that the shape of the distribution is not precisely known. The triangular distribution is bounded by the minimum and maximum value of the uncertainty range and depending on the value of the modus compared to its range the shape can be both symmetric or asymmetric.
Normal	The *Normal* distribution can be applied for uncertainty quantities that show a symmetric distribution of the uncertainty range around the mean. The uncertainty range is unbounded (-∝, + ∝).
Exponential	The *Exponential* distribution can be used for uncertain quantities which have a modus value of zero and for which the probability is decreasing with increased values ranging from zero to + ∝.
Lognormal, Gamma, Weibull	Both the *Lognormal, Gamma and Weibull* distributions can be used to describe an uncertain quantity that is being constrained to non-negative values, which is positively skewed towards a large upper uncertainty range (0, + ∝) and which has a single mode. The Gamma distribution is an alternative for the Lognormal distribution being less positively skewed and giving lower probability to extreme values. The Weibull distribution is similar to the Gamma distribution but is less skewed and gives lower probability to extreme values.
Beta	The *Beta* distribution can be used for uncertain quantities that are bounded by a minimum and maximum value. It allows a flexible way to represent uncertainty. It is often used to represent variability over fractions (between 0 and 1).

Case Description

The information on uncertainty as presented in Mosier et al. (1998) does not mention a specific probability density function and it remains unclear whether the default values should be interpreted as the averaged value (mean), the most probable value (modes) or as the median value. Furthermore, the uncertainty ranges can be interpreted either as absolute minimum and maximum or as the boundaries of a confidence interval. This means that based on the interpretation of the uncertainty information different PDFs can be defined. In order to analyse whether different interpretations of the uncertainty information is influencing the results of the uncertainty analysis we have analysed four cases using different PDFs. In each case we combine an interpretation of the IPCC default parameter value with an interpretation of the IPCC uncertainty range to select a PDF.

Case I: In Case I, a uniform distribution was used for all PDFs. When selecting the Uniform frequency distributions for parameters, there is no need to interpret the IPCC default values. The minimum and maximum values of the Uniform distributions are defined by the bounds of the IPCC uncertainty ranges as given in Table 5.1. This means that we interpret the IPCC uncertainty range as the absolute minimum and maximum. The definition of the Uniform distribution for each uncertain parameter is shown in Table 5.4.

***Case II*:** In Case II, triangular distributions were chosen. The IPCC default value is in this case interpreted as the mode of a triangular distribution that is bounded by the IPCC uncertainty range for each parameter (Table 5.1). When the uncertainty range is symmetric to the default value this resulted in a symmetric triangular distribution and in other cases this resulted in the selection of an asymmetric triangular distribution (see Table 5.4).

***Case III*:** The IPCC default value is in this case interpreted as the mean of a distribution that provides the best fit with the IPCC uncertainty range. For parameters where the uncertainty range is symmetric to the default value we have selected a normal distribution with a standard deviation that provides the best fit with the uncertainty range. Since Analytica truncates the normal distribution at 3 times the standard deviation, the uncertainty range is interpreted as a 99% confidence interval. In case the IPCC uncertainty range is a-symmetric to the default value we have selected a beta distribution with a mean equal to the default value and which provides the best with absolute minimum and maximum of the uncertainty range. In one case (EF_3_ls) both the normal and the beta distribution did not provide a good fit with the given uncertainty range. We concluded that for this parameter, the uncertainty information can only be interpreted to be uniformly or triangularly distributed and it was decided to use the triangular distribution as used in Case II. The characteristics of the PDFs for the different parameters are shown in Table 5.4.

***Case IV*:** In this case the IPCC default value is, for each parameter, interpreted as the median of a distribution that provides the best fit with the uncertainty range. For parameters having an uncertainty range symmetrical around the default value, we have used the normal distribution as in Case III. By definition, the median, mode and mean are equal for the normal distribution. For parameters having uncertainty ranges that are asymmetrical to the default value we have defined a lognormal distribution. Since Analytica truncates the lognormal distribution at the quotient and product of the median and $gsdev^3$ a, the uncertainty range is interpreted as the 99% confidence interval. For cases where the lognormal distribution did not result in a good fit to the uncertainty range we used a beta distribution (see Table 5.4).

The Uniform, Triangular, Normal, Lognormal and Beta-distributions include the most commonly used PDF types as described in Table 5.3. Although alternative PDFs can be defined, the four cases described above provide a sufficient basis to analyse the influence of PDF selection on results of the uncertainty analysis. As described above, we have interpreted the IPCC uncertainty ranges as the absolute minimum and maximum values (Cases I en II) or closely to these value (99% confidence interval in Case III and IV). This was an arbitrary choice. An interpretation of, for example, the 95% confidence interval would also have been possible. However, this likely only results in other bounds for the PDFs, resulting in a smaller uncertainty estimate, but not necessarily in new insights on the effect of PDF selection on the results of the uncertainty analyses. Finally, we assumed that no correlations between the different uncertain parameters exist.

Table 5.4 Overview of Case I-IV, including different interpretations of the uncertainty information provided by IPCC. The table summarizes the interpretation of the IPCC default and IPCC uncertainty range, followed by the definitions probability density functions for the parameters assuming Uniform (U), Triangular (T), Normal (N), Beta (B) or Lognormal (L) distributions.

Parameter[1]	**Case I:** • **default: -** • **range: min and max of Uniform distribution**	**Case II:** • **default: modus** • **range: minimum and maximum of Triangular distribution**	**Case III:** • **default: mean** • **range:** [2] **m: 10^{-3}u: 10^{-6}**	**Case IV:** • **Default: median** • **Range** [3]
Distribution type	**U(min, max)**	**T(min, modus, max)**	**N(mean, stdev) or B(x,y, lower, upper)**	• **L(median, gsdev)**[4]
EF1	U(0.0025-0.0225)	T(0.0024, 0.0125, 0.0226)	N(0.0125, 2.57m)	N(0.0125, 2.57m)
EF2	U(2-8)	T(1.97, 5, 8)	N(5,0.77)	N(5,0.77)
EF3 anaer. lagoon	U(0-0.002)	T(0, 0.001, 0.00201)	N(1m, 255u)	N(1m, 255u)
EF3 daily spread	-	-	-	-
EF3 liquid systems	U(0-0.001)	T(0, 0.001, 0.001)	T(0, 0.001, 0.001)	T(0, 0.001, 0.001)
EF3 other systems	U(0-0.01)	T(0, 0.005, 0.01)	N(5m, 1.28m)	N(5m, 1.28m)
EF3 pasture range paddock	U(0.005-0.03)	T(0.00485, 0.02, 0.03)	B(7.5, 5, 3m, 0.031)	B(7.5, 5, 3m, 0.031)
EF3 solid storage, dry lot	U(0.005-0.03)	T(0.00485, 0.02, 0.03)	B(7.5, 5, 3m, 0.031)	B(7.5, 5, 3m, 0.031)
EF4	U(0.002-0.02)	T(0.00191, 0.01, 0.02)	B(8, 10, 1.2m, 0.021)	B(8, 10, 1.2m, 0.021)
EF5	U(0.002-0.12)	T(0.00162, 0.025, 0.121)	B(2.2, 10, 1.9m, 0.13)	L(0.025,1.96): truncated at 0.12
EF6	U(0.002-0.12)	T(0.00177, 0.01, 0.12)	B(1.35, 20, 2m, 0.13)	L(0.01,1.94): truncated at 0.002
FRACGASF	U(0.01-0.2)	T(0.0091, 0.1, 0.2)	N(0.1,0.0232)	N(0.1,0.0232)
FRACGASM	U(0.01-0.3)	T(0.0083, 0.2, 0.3)	B(3.9, 2, 2.4m, 0.3)	B(3.9, 2, 2.4m, 0.3)
FRACGRAZ	-	-	-	-
FRACLEACH	U(0.1-0.8)	T(0.097, 0.3, 0.8)	B(2.9, 8, 0.1, 0.85)	L(0.3, 1.31)
FRACNCR0	U(0.005-0.025)	T(0.0049, 0.015, 0.025)	N(0.015,2.57m)	N(0.015,2.57m)
FRACNCRBF	U(0.02-0.04)	T(0.0199, 0.03, 0.04)	N(0.03,2.55m)	N(0.03,2.55m)
FRACNPR	U(0.1-0.2)	T(0.99, 0.16, 0.2)	N(0.16,0.0139)	N(0.16,0.0139)
FRACR	U(0.3-0.7)	T(0.298, 0.45, 0.7)	B(3, 5, 0.3, 0.7)	L(0.45,1.12)
NEX(dairy)	U(67-133)	T(67, 100, 133)	N(100, 8.5)	N(100, 8.5)
NEX(non-dairy)	U(47-93)	T(47, 70, 93)	N(70, 5.9)	N(70, 5.9)
NEX(other animals)	U(17-33)	T(16.9, 25, 33)	N(25, 2.05)	N(25, 2.05)
NEX(poultry)	U(0.4- 0.8)	T(0.4, 0.6, 0.8)	N(0.6, 0.051)	N(0.6, 0.051)
NEX(sheep)	U(13 - 27)	T(12.9, 20, 27)	N(20, 1.8)	N(20, 1.8)
NEX(swine)	U(13 - 27)	T(13, 20, 27)	N(20, 1.8)	N(20, 1.8)

[1] See Table 5.1 and Box 5.1 for a description; [2] The normal distributions are unbounded, but since Analytica truncates the distribution at 3x standard deviation (99% confidence interval), the uncertainty range is defined to equal the 99% confidence interval.. [3]The Lognormal distribution is unbounded to the right, but since Analytica truncates the distribution at medain/gsdev3 and median x gsdev3 (99% of the confidence interval), the uncertainty range is defined equal to the 99% confidence interval. [4] For case where the uncertainty is symmetric to the median a normal distribution was selected, for case where the lognormal distribution could not fit the uncertainty closely, a beta distribution was used.

5.4 Results

5.4.1 Error propagation

The result of the error propagation are used to estimate the inaccuracy in the inventory of agricultural N_2O emissions in the Netherlands in 1990 caused by inaccuracies in input values. The IPCC Good Practice Guidance Report (IPCC/OECD/IEA, 2000) suggests to use the 2.5 and 97.5 percentile of the cumulative distribution function as indication of the uncertainty range and we use this 95% confidence interval as expression for the estimate of inaccuracy.

Figure 5.1 and Figure 5.2 present the results of the error propagation through distribution sampling. As shown in section 2, the N_2O emissions from agriculture in The Netherlands in 1990 using the IPCC default guidelines – assuming no inaccuracy- are calculated to be 30 Gg N. Direct emissions contribute 13 Gg, with emission s from animal production of 5 Gg N and indirect emissions of 12 Gg N.

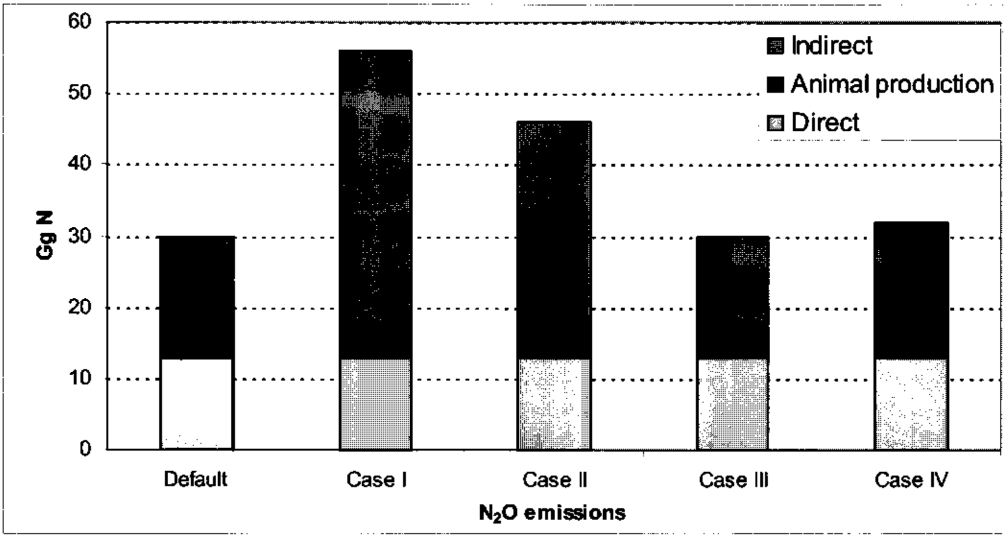

Figure 5.1 N_2O emission from agriculture in The Netherlands in 1990 as calculated using the IPCC default parameter values. The figure shows the mean values for direct emissions from agricultural fields (Direct), animal production, and indirect agricultural emissions (Indirect).

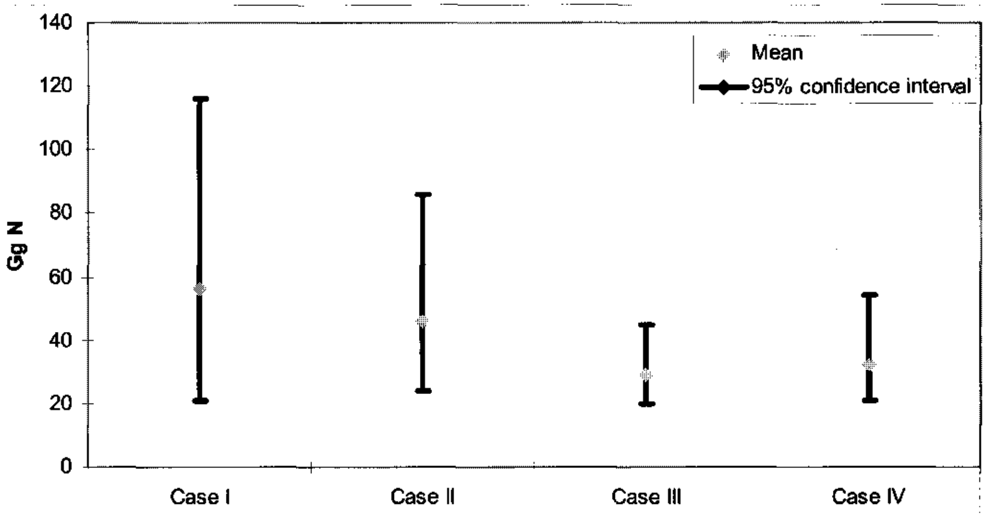

Figure 5.2 Results of the error propagation through distribution sampling. Shown are the mean value and the 95 % confidence interval for each case.

Cases I-IV result in mean values for total agricultural N_2O emissions that are not always the same as the IPCC Default of 30 Gg N (Fig. 5.1). The largest difference is calculated for Case I, in which we assume that that the IPCC uncertainty ranges are the minimum and maximum values of an uniform distribution This case results in a mean value for agricultural N_2O emissions of 56 Gg N, of which 13 Gg are from direct emissions, 4 Gg from animal production and 39 Gg from indirect emissions. The calculated total inaccuracy is relatively large for Case I. The 95% confidence interval is calculated to range from 21 Gg N (-63% compared to the mean) to 116 Gg N (+107%).

For Case II we also calculate a mean N_2O emission of 46 Gg N, which is considerably higher than the IPCC Default of 30 Gg N. Case II assumes that the default value is the mode of a triangular distribution with the uncertainty ranges as boundary, the Latin Hypercube Sampling. Of the 46 Gg N, 13, 5 and 28 Gg are from direct soil emissions, animal production and indirect emissions, respectively. The calculated 95% confidence interval ranges from 23 Gg N (-50%) to 87 Gg (+87%).

For Case III, in which we characterised the default value as the mean of a distribution that provided the best fit with the given uncertainty range, we calculate a mean emission of 29 Gg N, with 13 Gg N from direct soil emissions, 5 Gg N from animal production and 12 Gg from indirect emissions. The uncertainty in the emission estimate expressed as the 95% confidence interval ranges from 20 Gg N(-31%) to 45 Gg N (+55%).

In Case IV the IPCC default values are interpreted as the medians of a distributions that provide the best fit with the uncertainty ranges. The mean N_2O emissions from Dutch agriculture under this assumption are calculated to be 32 Gg N with 13, 5 and 14 Gg N from direct, animal production and indirect emissions, respectively. The 95% confidence interval ranges from 21 Gg N(-34%) to 54 Gg N (+71%).

5.4.2 Importance analysis

Next, we analysed the influence of the different statistical interpretations of the IPCC information on default values and uncertainty ranges (Case I to IV) on the results if importance analysis. We quantified the contribution by individual parameters to the overall inaccuracy in the N_2O emissions for each of the four cases. To this end, we calculated the Spearman's rank correlation coefficient and the standardised regression coefficient.

Table 5.5 shows the different Spearman's rank correlation values for the 23 inventory parameters used in the uncertainty assessment. A higher value for the Spearman's Rank Correlation Coefficient (RCC) can be considered an indication for a larger contribution of a specific parameter to the overall inaccuracy in the N_2O emission estimate. The results show that six (out of 23) different parameters have a relatively high RCC, indicating that they have a relatively large share in the total inaccuracy in agricultural N_2O emission estimates. These results are similar for all four cases. These influential parameters are the emission factor for indirect emissions (EF5), the fraction of agricultural N that is lost through leaching (Fracleach), the emission factor for direct soil emissions (EF_1) the emission factor for cultivated histosols (EF_6), and N excretion by swine (Nex(swine)). In all four cases EF_5 is the most important uncertain parameter. In case I, II, and IV Fracleach is the second most important parameter, while EF1 is the third most important parameter. In case IV EF1 is the second most important uncertain parameter followed by Fracleach. EF_6 and Nex(swine) are the fourth and fifth most important parameters in all cases, with Nex(swine) the fourth in case I and case III.

In order to derive a more clearer and quantitative expression of the contribution of uncertain parameters to overall inaccuracy in the agricultural N_2O emission estimate, we performed a multiple linear regression analysis (Table 5.5). The values of R^2 for the four cases illustrate that more than 90% of the variation (inaccuracy) in the agricultural N_2O emission estimate can be explained by the linear regression model. Based on the value of R^2 and the square of the standardised regression coefficient (SRC), the percentage contribution of each inaccurate parameter to the inaccuracy in the regression model can be calculated. For R^2 close to 1, this percentage can be used to express the relative contribution of the uncertain parameters to the inaccuracy in the N_2O emission estimate (Saltelli, 1999).

The regression analysis using the results from the numerical simulation in Case I results in an R^2 of 0.896 (Table 5.5). This means that ~90% of the overall inaccuracy can be explained by uncertainty in the inventory parameters. The relative contribution of EF5 to the regression results for Case I is 55% ($SRC^2 / R^2 : 0.704^2 /. 0.896 = 0.55$). This indicates that EF_5 has a large influence on the inaccuracy in the N_2O emission estimate. Figure 5.3 presents the contribution of the uncertain parameters to the regression results. The results in Figure 5.3 clearly indicate that in all four cases only three of the 23 parameters (EF_5, $Frac_{LEACH}$ and EF_1) have a relatively large share in the total inaccuracy in the N_2O emission estimate. These results as presented in Figure 5.3 show that independent of the PDF type selection, only three parameters have a large impact on the overall uncertainty estimate. However, there are some differences in the contribution of each of these parameters in the different cases. Parameter EF5 shows that its contribution increasing depending on the interpretation of the default value from uniform (case I), mode (case II), mean (case III) and median (case IV).

Table 5.5 Results from the rank correlation and regression analysis. For Case I to IV are presented the Spearman's rank correlation coefficient (RCC) and the results of the regression analysis (R^2 and standardised regression coefficient (SRC)).

	Case I		**case II**		**case III**		**case IV**	
	Rank correlation	Regression $R^2 = 0.896$	Rank correlation	Regression $R^2 = 0.922$	Rank correlation	Regression $R^2 = 0.938$	Rank correlation	Regression $R^2 = 0.943$
	RCC	SRC	RCC	SRC	RCC	SRC	RCC	SRC
EF$_1$	0.26	.209	0.26	.235	0.40	.364	0.33	.271
EF$_2$	0.01	.021	0.02	.021	0.03	.032	0.03	.024
EF$_3$ *anaerobic lagoon*	0.00	-.007	0.00	.000	0.00	.000	0.01	-.001
EF$_3$ liquid systems	0.01	.002	0.01	.014	0.02	.022	0.02	.008
EF$_3$ other systems	0.02	.009	0.02	.008	0.02	.013	0.01	.012
EF$_3$ pasture, range, paddock	0.03	.028	0.05	.032	0.08	.062	0.06	.049
EF$_3$ solid storage, dry lot	0.04	.033	0.06	.037	0.09	.066	0.07	.049
EF$_4$	0.04	.033	0.06	.040	0.09	.069	0.08	.051
EF$_5$	**0.72**	**.704**	**0.76**	**.740**	**0.75**	**.751**	**0.82**	**.848**
EF$_6$	**0.10**	**.103**	**0.14**	**.128**	**0.08**	**.087**	**0.10**	**.087**
FRAC$_{GASF}$	0.01	-.001	0.00	-.002	0.00	-.004	0.00	-.004
FRAC$_{GASM}$	0.00	.000	0.00	-.006	0.01	-.016	0.01	-.014
FRAC$_{LEACH}$	**0.54**	**.568**	**0.51**	**.526**	**0.42**	**.442**	**0.33**	**.351**
FRAC$_{NCR0}$	0.01	.007	0.03	.004	0.03	.009	0.02	.005
FRAC$_{NCRBF}$	0.00	-.001	0.01	.001	0.01	.002	0.00	.000
FRAC$_{NPR}$	0.03	.031	0.03	.030	0.02	.013	0.02	.011
FRAC$_R$	0.01	-.007	0.01	-.006	0.01	-.008	0.01	-.003
N$_{EX}$(dairy)	0.07	.077	0.05	.061	0.06	.066	0.07	.054
N$_{EX}$(non-dairy)	0.06	.063	0.05	.055	0.06	.061	0.07	.054
N$_{EX}$(other animals)	0.01	.006	0.01	-.001	0.01	.000	0.02	.004
N$_{EX}$(poultry)	0.01	.018	0.04	.021	0.04	.020	0.03	.011
N$_{EX}$(sheep)	0.00	.017	0.01	.009	0.02	.013	0.02	.016
N$_{EX}$(swine)	**0.12**	**.090**	**0.08**	**.080**	**0.09**	**.084**	**0.08**	**.071**

Parameter Fracleach has its largest contribution to uncertainty when assuming a uniform distribution (case I), while EF1 has the largest contribution in case III compared to the other cases. These results are comparable with the results found by Brown et al. (2001). Although they applied a different probability distribution to calculate the uncertainty in the N_2O emission estimate in the United Kingdom, Brown et al. (2001) concluded that the parameters EF5, Fracleach and EF1 were the most influential parameters on the total emission estimate for the UK.

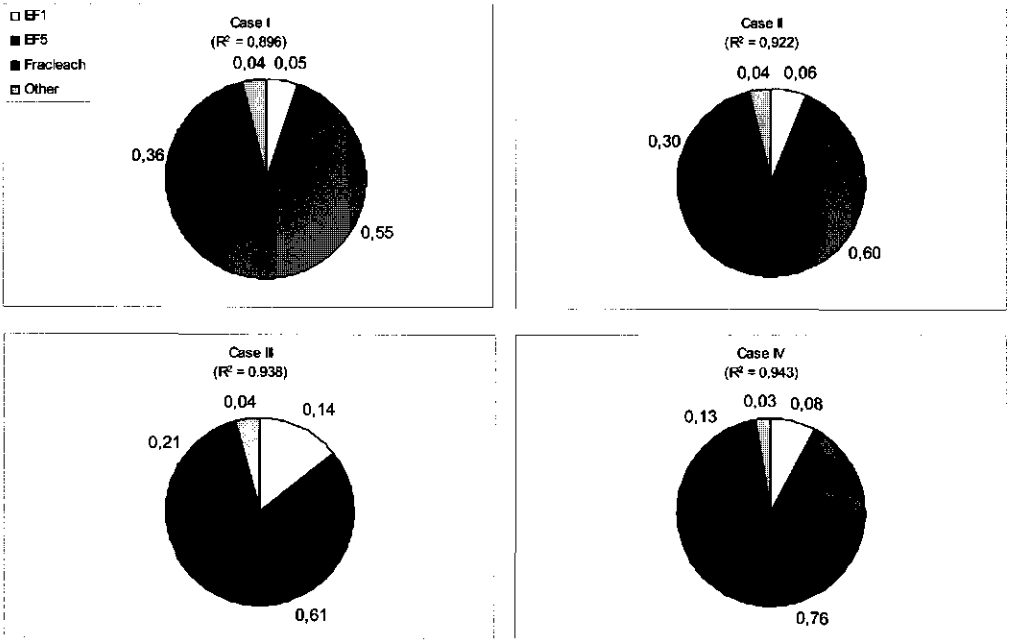

Figure 5.3 Relative share of inventory parameters in the regression results and therefore in the total inaccuracy of the N_2O emissions from agriculture in The Netherlands in 1990. Results are shown for the emission factors for direct soil (EF_1) and indirect (EF_5) emissions, and for the fraction of agricultural N leaching from soils ($FRAC_{LEACH}$), as well as for the other 20 parameters. Unit: contribution to regression result (SRC^2/R^2).

5.5 Conclusions

In this study we have analysed the influence of statistical interpretation of limited information about inaccuracy in IPCC parameters on the results of a statistical analysis of inaccuracies in estimates for agricultural N_2O emissions from the Netherlands in 1990. To this end, we defined four cases reflecting various interpretations of the IPCC information on parameter values and their uncertainty ranges. These interpretations resulted in the selection of different probability density functions, which were used in the statistical analysis. From our analysis we can draw two important conclusions.

First, our results indicate that the calculation of the overall inaccuracy depends considerably on the statistical interpretation of the IPCC information on the inventory. We conclude this from the error propagation through Latin Hypercube Sampling. The results gave insight in the overall inaccuracy of the emission estimate. Our result show that for the four different cases, the mean values of the N_2O emissions calculated in Case I-IV differ by almost a factor of two, and range between 29 and 56 Gg N. We expressed overall uncertainty in two ways, and conclude for both that the differences between the four cases are considerable. We first expressed the inaccuracy by the 2.5 and 97.5 percentile of the cumulative distribution function resulting from the Latin Hypercube Sampling. The resulting in confidence intervals for Case I-IV have lower bounds varying between -33% and -63 % of the mean and higher bounds ranging from +55 % to +107% of the mean.

Second, our results indicate the statistical interpretation of the IPCC information on the inventory does not have a large impact on the identification of the parameters having a largest share in the overall inaccuracy. We applied rank correlation and regression analysis to analyse the relative contribution of uncertain parameters to the overall inaccuracy in the emission estimate. This information allows for prioritising future research aiming at further improvement of the methodology to estimate emissions. It may help to guide experimental research towards the most uncertain aspects in the emission inventory. The results of applying rank correlation and regression analysis show that only a small number of uncertain parameters (EF_5, $Frac_{leach}$, EF_1, EF_6 and Nex(swine)) have a relatively large share in the overall inaccuracy. The regression analysis indicates that three out of 23 parameters (EF_5, $Frac_{leach}$ and EF_1) have a relatively large share in the inaccuracy. These results are similar for all four cases. This conclusion is strengthened by the study of Brown et al. (2001) who also concluded that EF5, Fracleach and EF1 are the most influential uncertain parameters.

All parties to the UN Framework Convention on Climate Change will have to assess the inaccuracies in their national greenhouse gas emission inventories. Our analysis indicates that the current IPCC Guidelines do not provide the information needed to perform such analyses. Although our results are representative for the calculation of agricultural N_2O emissions in The Netherlands in 1990, the conclusions about the applicability of uncertainty propagation and importance analysis hold for other countries as well. Any country, calculating agricultural N_2O emissions based on the IPCC default values will face the problems associated with interpreting the IPCC parameter information for calculation of the uncertainty in the emission estimate. This will have an effect on the use of national communications reported to the Climate Convention. When different countries report uncertainty based on various interpretations of the same default values the check for reliability and quality will become difficult.

Based on our analysis, we conclude that for the calculation of the overall inaccuracy in the emission estimate for N_2O emissions from agriculture more information is needed on the statistical interpretation of the IPCC default values and their uncertainty ranges. This could be achieved by developing guidelines on the interpretation of the default values as either the averaged values of measurements (mean), a median value or the most probable (mode). Furthermore, additional information is needed on the meaning of the uncertainty range of parameter values. This could be achieved by specifying whether the range is an absolute minimum or maximum of possible values or the boundaries of a certain probability function.

Although there is uncertainty about the statistical interpretation of the IPCC parameter defaults and uncertainty ranges, the available information was found to be sufficient to allows for conclusions with respect to the identification of parameters having the largest influence on the overall inaccuracy in the emission estimate. In the case of N_2O emission from Dutch agriculture the inaccuracy in the estimate can be reduced by improving the methodology for estimating indirect N_2O emissions (EF_5 and $Frac_{leach}$) and direct soil emissions (EF_1).

Chapter 6

External assessment of inaccuracy

Using wind-direction-dependent differences between model calculations and field measurements as indicator for the inaccuracy of emission inventories

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6.1 Introduction

Emissions of air pollutants within a country or region are the result of a variety of individual sources. Since it is not practical to measure each emission source individually, the estimation of large-scale emissions is in most cases based on calculation of emissions using an emission factor approach. This emission factor approach aggregates information of sources in both time and space, which will (amongst other reasons, e.g. error in emission measurement) lead to an inaccurate representation of the real emission. Although we know that emission inventories are inaccurate, we do not know exactly the specific sources or the size of the inaccuracy. In other words, we there is uncertainty about the source or size of the inaccuracy. This is what we define as the uncertainty of the emission inventory.

By performing an uncertainty assessment an attempt is made to identify the sources of inaccuracy that we are ignorant about and to quantify their impact on the accuracy of the emission estimate. We distinguish between internal and external assessments. In an *internal assessment* the methodology and data used to construct the inventory are used while in an *external assessment* the differences in methodology, data or emission derived from other studies than from which the emission inventory was derived are used to identify or quantify the inaccuracy of the emission inventory. In this paper, we focus on an external assessment. Examples of external assessments are forward air quality modelling and inverse air quality modelling. In forward modelling, an emission inventory is used as input into an atmospheric dispersion model, which calculates atmospheric concentrations of the pollutant. The deviation between modelled and observed concentrations can be an indicator for the inaccuracy in the emission inventory (e.g. Iversen, 1993). In inverse modelling, measured concentrations are used as input into an atmospheric dispersion model to calculate the emissions needed to reproduce the observed concentrations. Differences between the 'back-casted' emission estimates and the emission inventory can be considered as indicator for the inaccuracy of the emission inventory (e.g. Seibert, 2000).

The problem with the performance of an external assessment is that it is not easy to pinpoint emission inventory as single cause for the difference between model and observations. The reasons for this are the following: First, every emission inventory, dispersion model and measurement contains inaccuracies which means that deviations between model and observations can be caused by either erroneous model formulation, inaccuracies in emissions or meteorological data or inaccuracies in the measurements (Lehmhaus et al., 1986). Second, measurement data are often taken from an inhomogeneous monitoring network, which means that measurements are usually not available on the desired spatial and temporal scale to allow for a good comparison between model and observations. Consequently, modelled grid cell averages are often compared with point observations. By using forward and inverse modelling in the assessment of uncertainty in emission inventories, the implicit assumption is that both the dispersion model and observation contain no major inaccuracies.

Pulles et al. (1996) used an approach to pinpoint emission inventory as the cause for difference between measurement and calculation consisting of studying the differences between modelled and observed concentrations by wind direction sector. The approach was applied to the CORINAIR 1990 emission inventory for emissions of nitrogen oxides (NO_x), sulphur dioxides (SO_2) and carbon monoxide (CO) from The Netherlands and the Slovak Republic. Although the study was limited to the Netherlands and Slovakian monitoring networks, one of the conclusions was that NO_x and CO concentrations in the Netherlands seemed to be underestimated by the model at southeastern winds importing emissions from Germany.

In this chapter we will expand the analysis by Pulles et al. (1996). We will analyse wind-direction-dependent differences between calculated and modelled concentrations in more detail and on a larger scale. The objective of this study is to analyse whether wind-direction-dependent differences between calculated and measured concentrations can be used to identify and quantify the sources and size of inaccuracy in a large-scale emission inventory such as an SO_2 emission inventory for Europe in 1994. We will attempt to determine (1) whether this type of analysis is a useful tool in the assessment of uncertainty about accuracy in large scale emission inventories, (2) the information needs for such an assessment, and (3) the limitations of this type of analysis. To meet these objectives we will analyse differences between calculated concentrations of SO_2 in Europe and SO_2 field measurements for Europe in 1994.

6.2 Methodology

In this study we will analyse wind-direction-dependent differences between measured SO_2 concentrations for 1994 from the EMEP network with calculated SO_2 concentrations from the LOTOS model (Builtjes, 1992). The calculated SO_2 concentrations from the LOTOS model were derived using a 1994 SO_2 emission inventory as model input.

EMEP SO_2 measurements.

For our analysis of SO_2 in 1994 we used data from the Co-operative Programme for Monitoring and Evaluation of Long Range Transmission of Air Pollutants in Europe (EMEP). The monitoring stations included in EMEP are measuring deposition and background concentrations of air pollutants in Europe. The 1994 SO_2 data set is described in Hjellbrekke et al. (1996 a, b) and consists of daily averaged SO_2 concentrations based on a sampling period of 24 hours from 91 stations within the EMEP monitoring network.

LOTOS SO_2 calculations.

The Long Term Ozone Simulation (LOTOS) model is an eulerian grid model that uses a 3-D time dependent atmospheric diffusion equation for multiple chemical species including SO_2 (Builtjes, 1992). The modelling domain of LOTOS is Europe between a latitude of 10° West to 60° East and a longitude of 35° to 70° North with a grid cell size of 0.5° latitude and 1° longitude. The LOTOS model uses a chemical kinetics package called CBM-IV to model atmospheric chemistry within four vertical dynamic layers (surface, mixed, lower inversion, upper inversion). The vertical domain of these layers is between 0 and 2600 m altitude. The layer mean values of SO_2 per grid cell in 1994 are calculated based on emission and meteorological input data. Furthermore, the model uses a landuse database to calculate, using the resistance analogue, the dry deposition. Wet deposition is included as well. The boundary conditions for SO_2 are taken from the 2-D global Isaksen-TNO model. The model data used in this study consists of gridded hourly averaged SO_2 concentrations ($\mu g\ SO_2$) at a height of 3.5 (m), and wind direction (degrees) at the surface layer level.

SO_2 emission inventory. The emission inventory that is used by LOTOS as input for modelling studies and which will be analysed in this study is based on the CORINAIR 1994 inventory (ETCAE, 1997). This inventory contains the officially reported national totals for 20 European countries (EU 15, Norway, Switzerland, Malta, Iceland and Croatia) divided into several emission source categories. An earlier version of the CORINAIR database, CORINAIR 1990, (ETCAE, 1996) contains data at a higher spatial resolution for 1990. This data set was used to obtain emission data for countries not included in the 1994 database and to spatially disaggregate the 1994 data. Before these CORINAIR data could be used as input to the LOTOS model, the following three tasks have been performed. Firstly, calculation of the emissions per grid cell of 0.5 x 1.0 latitude-longitude by using population density and information on the location of the activities/industries. The basis for this allocation of emissions is information for the year 1990 as described above. Secondly, addition of the emissions for countries not included in CORINAIR 1994 or 1990 (mainly former Soviet Union countries). To do this, the former LOTOS-structure (Builtjes, 1992) is used, following as close as possible the CORINAIR-approach. Thirdly, addition of the known time dependency (e.g. season) of the specific source categories, following the former LOTOS-structure. It should be noted that the biogenic S-emissions of DMS are not included in the emissions, and that volcanic emissions are excluded. Figure 6.1 shows the total SO_2 emission per grid cell in 1994.

The following five-step methodology was used:

A. Selection of EMEP stations to be included in the analysis.

We used EMEP measurements from stations for which SO_2 measurement data are available and that are located within the domain of the LOTOS model. Furthermore when using measurement data for comparison with model results, one should take into account that the comparison result is unreliable when (i) measurements show rather low SO_2 concentrations (possibly near the detection limit), (ii) measurements data are erroneous, or (iii) the data coverage (number of days for which measurements are available) is low. For this reason, stations that show low SO_2 concentrations (well below 1 $\mu g\ SO_2$) or have a low temporal coverage (less then a few months within a year) were excluded from the analysis.

By comparing measurements within EMEP, (Hanssen and Skjelmoen, 1995) insight was gained about the measurement error. This lead, for example, to the exclusion of the Slovakian measurement stations. Finally, we assume that all EMEP measurement stations are representative background stations. Out of 91 EMEP measurement stations from the 1994 EMEP database, 70 were included in the analysis.

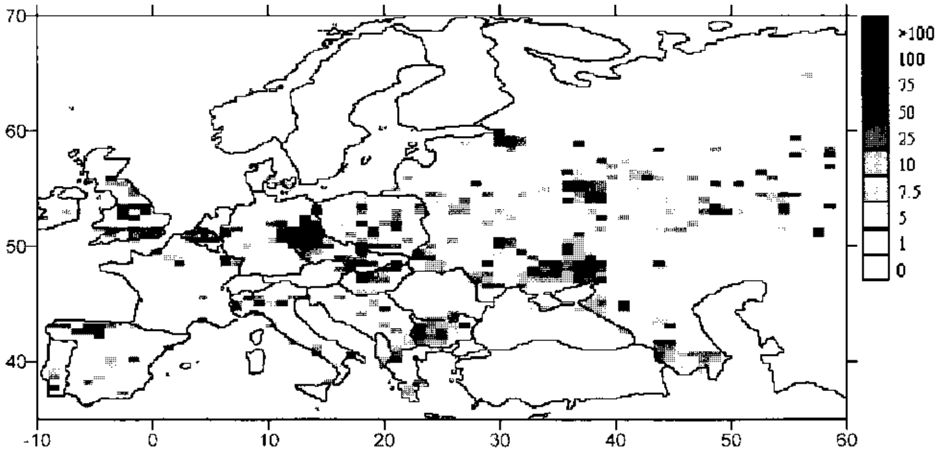


Figure 6.1 Total SO_2 emission per grid cell in 1994 in Europe (unit: kt SO_2).

B. Selection of grid cell data from LOTOS and calculation of daily averaged data.

Based on information (Hjellbrekke et al., 1996a) about longitude and latitude of the measurement stations we determined in which LOTOS grid cell the selected EMEP stations are situated. For these grid cells we used the grid cell average data on surface level SO_2 concentration and wind direction. Because of the different temporal scale of the LOTOS calculations (hour) and the EMEP observations (day), we calculated daily averaged SO_2 concentrations and wind directions from the LOTOS data per grid cell. The wind direction per day was calculated based on averaging the hourly vertical and horizontal wind components (u and v) and transformation of the averaged u and v into degrees. Because of the different spatial scales between the LOTOS calculation (grid cell) and EMEP observations (point), we have to assume that within a LOTOS grid cell size of 1° x 0.5° the concentrations are well mixed.

C. Selection of days included in the analysis and classification in wind sectors.

The daily averaged EMEP and LOTOS SO_2 concentrations were compared by wind direction. Calculating daily averaged wind concentrations from hourly data holds a risk that the most frequent wind direction might be averaged out on days where the wind direction change is large (e.g. from North to South). To avoid misinterpretation of wind-direction-dependent differences, the standard deviation of the hourly wind directions over one day were used as an indicator for large changes in wind direction over a day. This was performed by calculation of the standard deviation based on hourly wind direction in degrees (transformed from hourly u and v) and the averaged wind direction as calculated under section B. For days with a standard deviation larger than 30 degrees, the EMEP and LOTOS data for that day were excluded from the analysis. Days for which no EMEP data was available were also excluded from the analysis. The daily averaged EMEP and LOTOS SO_2 concentrations that met the criteria of standard deviation of wind directions and availability of measurement data were classified in wind direction sectors of 30 degrees. These means, for example, that wind directions ranging from 345° to 15° are classified in a category with a class midpoint of 0°.

D. Graphical display of differences between model and measured SO_2.

The wind-direction-dependent differences between modelled and measured SO_2 concentrations were analysed by displaying the data in three different graphs as shown in Figure 6.2. First, a concentration rose illustrates the wind-direction-dependent differences between LOTOS and EMEP SO_2 concentrations. Figure 6.2a is an example of this for EMEP station NL09. Second, a histogram of observed concentrations is plotted beside the concentration rose (Figure 6.2b). Differences between LOTOS and EMEP can only be an indicator for emission inaccuracy if they are based on more than a single observation: a single observation cannot distinguish between mere coincidence, measurement error or local source. Finally, a further discussion on the differences found in the concentration rose (e.g. on seasonality) is based on Figure 6.2c, which shows the daily averaged SO_2 concentrations of both LOTOS and EMEP in the year 1994.

E. Interpretation of graphical displays.

As an example of the analysis of wind-direction-dependent differences between model and measurement we will discuss the results for EMEP station NL09. As shown in Figure 6.2, the name of the station is Kollumerwaard, situated in the Netherlands and the measurements are based on instrumental UV/fluorescence. The results for 248 days are used in the analysis, for 50 days no measurement data were available and on 67 days the wind was highly variable (standard deviation wind direction larger than 30 degrees). For winds from north to northeast (330°- 90°) there is a good agreement between (LOTOS) modelled and measured (EMEP) concentrations (Figure 6.2a) With wind from southeastern direction (classes 120^0 and 150°) the measurement shows higher concentrations with a clear discrepancy in wind direction 150^0 where the difference is a factor 1.7. With winds from southwest to northwest (210°-330°) the model shows higher SO_2 concentrations than the measurements with about a factor of two. The frequency histogram for wind direction categories (Figure 6.2b) indicates that the wind-direction-dependent difference plot (Figure 6.2c) is based on results from several days per wind direction. Figure 6.2c shows that the higher measurement values from southeastern wind directions are found on several days in February and October, and that the higher model values from southwest to northwest seem to occur especially in the winter months November to January. Analysing the result for only the one station NL09 does not allow a clear distinction between emission error, model error or measurement as cause for the discrepancy between modelled and measured SO_2 concentrations. However when a systematically model or measurement error was occurring it is unlikely that a wind direction dependent difference as shown in Figure 6.2awas found. Based on the results of a single measurement station it is not possible to distinguish emissions and their originating region as being inaccurate/uncertain. For that we would need information from other stations. If other stations in the Netherlands and neighbouring countries show a comparable wind direction dependent difference (higher measurement values in South-eastern direction or higher model values in Southwest-North-western direction) the conclusion can be drawn that the discrepancy is caused by a source region that is influencing the LOTOS grid cell of station NL09. In the same manner as discussed for station NL09 we analysed the results for the other stations. We used as criterion for selecting inaccuracy in the emission estimate for a certain region as possible cause for the discrepancy between model and measurement that the wind-direction-dependent differences from several measurement stations from different countries point towards that specific region. If this criterion is not met, we did not distinguish between model error, emission error or measurement error.

Station:	NL09	**Name:**	Kollumerwaard	**Country:**	Netherlands
Lat:	53,33			**Observations included:**	248
Lon:	6,28			*Excluded:*	
Height (m):	0			**No EMEP data:**	50
				Measurement error:	0
Measurement method:		Instrumental UV fluorescence		**STdev > 30 degr.:**	67
					365

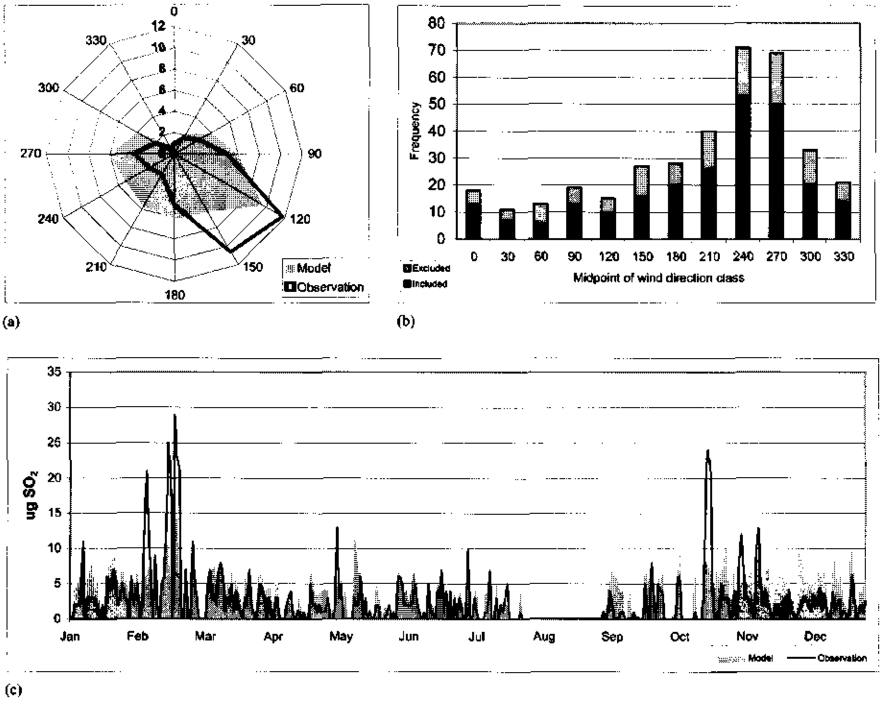


Figure 6.2 Graphical display of differences between modelled and observed SO_2 concentrations. (a) The wind-direction-dependent modelled and observed SO_2 concentrations in 1994. (b) The frequency diagram for wind direction classes of 30°. Shown are days that are included or excluded from the analysis. (c) The modelled and observed SO_2 concentrations per day in 1994.

The methodology applied in this study is only valid under certain assumptions. First, when information on measurement error in the EMEP measurements is not available we assume that the measurements are non-erroneous. Second, we assume that when we find a large discrepancy between model and measurement without a specific wind-direction-dependency the cause for discrepancy is systematic model error. Systematic model error can occur on the boundaries of the modelling system (e.g. boundary concentrations/emissions) or in regions where the model is unable to simulate the atmospheric processes (mountainous areas), or when the process description in the model is erroneous like calculation of deposition values or pollutant lifetime. This does not mean that when a specific-wind-direction dependency is found systematic model error is excluded. For example a measurement station located in the mountains can show specific wind-direction-dependent concentration when the mountain is on one side of the station. Furthermore, measurement stations located near coastal areas can measure recirculation of concentrations due to land-sea breeze, a feature that can be captured by the model resulting in wind-direction-dependent differences due to systematic error. The combination of different measurement stations from different countries can exclude these occurrences. Third, we assume that within a LOTOS grid cell (1° x 0.5°) the concentrations are well mixed so that a point measurement can be compared with a grid cell average.

6.3 Results

The graphical displays as described above show a coherent behaviour of a number of stations in several regions within the study area. These regions are: Sachsen/Branderburg (Germany), Western Russian Federation, Central England, Spain and Alpine region. For each of these regions the differences between calculated and observed concentrations were inspected. Below we will describe the results for each of the regions. Detailed graphical information, as shown in Figure 6.2, can be found at the Internet address: www.dow.wau.nl/msa/e-a/lotos.htm. Table 6.1 provides a summary of the analysis of wind-direction-dependent differences for all five regions.

6.3.1 Sachsen/Brandenburg

The group of measuring stations surrounding the German "Länder" Sachsen and Brandenburg (Figure 6.3) show a clear overestimation of calculated SO_2 concentrations at wind directions from these "Länder" towards the stations. At southeastern (SE) winds, calculated concentrations are higher at the stations NO01, NO08, DK03, DK05, DK08 and DE09. The same occurs at southern (S) wind directions at the stations NO41, SE05 and DE07. At northern (N) wind directions overestimation occurs at DE05 and to a lesser extent CS03. The station PL03 shows a clear overestimation by the calculation at northwestern (NW) wind directions. These observations can consistently be explained by the assumption that the emissions in Sachsen and Brandenburg might be overestimated. At some other stations in this area this is less clearly visible, but in none of the stations surrounding the area observations were made that contradict this assumption.

Table 6.1 Summary of the analysis of wind-direction-dependent differences between modelled and measured SO_2 concentrations in Europe [a]

Regions	Information from	Conclusion
1. Sachsen/Brandenburg	*Czech Republic*: CS03 *Denmark*: DK03, DK05, DK08 *Germany*: DE02, DE05, DE07, DE08, DE09 *Norway*: NO01, NO08, NO41 *Poland*: PL03, PL04 *Sweden*: SE02, SE11	*Emission inventory inaccuracy*: Inaccurate spatial distribution of SO_2 emissions in Germany leads to overestimation of emissions in this region. At specific locations the overestimation might be 200 - 400%.
Nordrhein-Westphalia	*Germany*: DE01, DE02, DE04, DE08 *Netherlands*: NL09, NL10	Possible underestimation of emissions in this region due to inaccuracy about spatial distribution but conclusion is based on not enough measurements.
2. Russian federation	*Finland*: FI04, FI09, FI17, FI22 *Latvia*: LV10 *Lithuania*: LT15 *Poland*: PL02, PL04, PL05 *Russia*: RU01, RU13, RU14 *Sweden*: SE08, SE12	*Emission inventory inaccuracy*: The budget of SO_2 emissions in Russia lead to underestimation of emissions. At specific areas this underestimation might be more than 500 %.
3. Central England	*Denmark*: DK03 *France*: FR05 *Germany*: DE01 *Netherlands*: NL09 *Norway*: NO01, NO08 *United Kingdom*: GB02, GB04, GB06, GB07, GB13, GB14, GB15, GB16	*Emission inventory inaccuracy*: Inaccurate spatial distribution of emissions within the United Kingdom leading to overestimation of emissions at specific location with +/- 200%.
4. Spain	*France*: FR12 *Portugal*: PT04 *Spain*: ES01, ES02, ES03, ES04, ES05	*No distinction possible*: Discrepancy could be either caused by not including ship emissions, overestimation of emissions in Northern Spain, modelling error like boundary conditions or measurement error.
5. Alpine region	*France*: FR08, FR11 *Germany*: DE03 *Switzerland*: CH03, CH04, CH05	*No clear conclusion*: Either the model performance in mountainous regions or inaccurate emission inventory for large point source emissions.

[a] The table shows five regions towards which country the discrepancy points, the measurement stations on which the analysis is based and the conclusions drawn from the analysis. See www.dow.wau.nl/msa/e-a/lotos.htm for details.

The graphs of the daily averaged SO_2 concentrations from LOTOS versus those from EMEP (see Figure 6.2c for an example) for all of the stations discussed above do not show a deviation in temporal patterns between measured and observed concentrations over the year. The apparent overestimation of emissions might be caused by inaccuracy in the spatial distribution (emission per grid cell) of the national German SO_2 emission budget due to changes since 1990 in relative importance of sources in the former GDR compared to the western parts of the country. Since the spatial distribution of emissions for 1994 (after the German unification) was based on the one for 1990 (before the German unification), the shut down of major parts of the industry in the former GDR might not be reflected in the emission inventory. If this conclusion is correct, we expect an underestimation of the emissions in other parts of the country. This is consistent with the observations in the Nordrhein-Westphalia region: The measurements from the Dutch stations NL09 and NL10 show that with southeastern (SE) winds the model is underestimating SO_2 concentrations by about 50 to 70%. This discrepancy could be caused by an underestimation of emissions from the Ruhr area located in the German state Nordrhein Westphalia. The same indication can be found in the result for German station DE01 where an underestimation of SO_2 concentrations by a factor of 2 is found at southern (S) winds but this measurement station can also be influenced by emissions from the Hamburg/Bremen area. Consequently, information from other stations is needed. The German stations DE02, DE04 or DE08 do not confirm the underestimation of emissions from the Ruhr area. Since no data from other measurement stations than NL09, NL10 and DE01 show the same trend, the cause for this wind-direction-dependent discrepancy remains unclear.

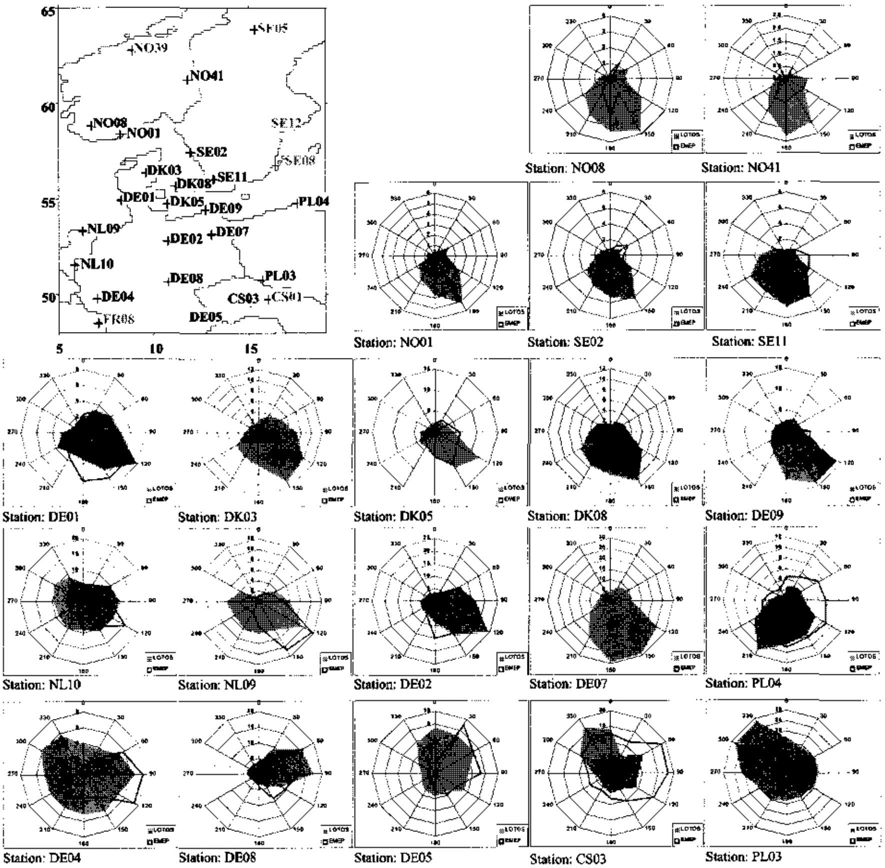


Figure 6.3 Wind-direction-dependent modelled (LOTOS) and observed (EMEP) SO_2 concentrations (in µg SO_2) at measuring stations surrounding the Sachsen/Brandenburg region and stations surrounding the Nordrhein-Westphalia region. The stations included in the analysis are marked in bold on the map while the plots per stations are sorted by longitude-latitude position.

6.3.2 Russian Federation

The results from stations located in Finland, Russia, Sweden, Latvia, Lithuania and Poland indicate an underestimation of modelled SO_2 concentrations originating from the Russian Federation (Figure 6.4). The Finnish stations FI04, FI09, FI17 and FI22 indicate an underestimation of modelled SO_2 concentrations from northeastern (NE) wind directions with the modelled concentrations about 2-5 times lower than the measurements. The Polish stations PL02, PL04 and PL05 show 2-3 times lower modelled concentrations with winds coming from the east (E). In Sweden the model concentrations at stations SE08 (E-NE) are a factor 4 lower than the measurements and at location SE12 (E) this discrepancy is also visible (however less clear with 40 % lower model concentrations). The Latvian station LV10 (E with a factor of 2) and Lithuanian station LT15 (NE-E, factor 2-4) point towards an underestimation of modelled SO_2 concentrations coming from the direction of the Russian Federation. Within the Russian Federation the radar plots of RU01 and RU13 and RU14 show a large underestimation of the modelled concentrations at all wind directions. As was the case with the emission from Sachsen/Brandenburg, several stations from different countries show a discrepancy between calculated and measured concentrations originating from the Russian Federation. Therefore, we conclude that the emissions from the Russian Federation are underestimated. The graphs of the daily averaged SO_2 concentrations from LOTOS versus those from EMEP for the stations in Finland, Sweden and Poland show that especially in February the differences are the largest. This is probably caused by the fact that eastern winds are dominant in this period of the year at these stations and not so much by inaccuracy in the spatial distribution of emission in the Russian Federation. Since the Russian, Latvian and Lithuanian stations show the discrepancy throughout the year, the inaccuracy in the emission budget of the Russian Federation is the most likely cause for the difference between model and measurement. The factor of difference between model and measurement found at stations outside the Russian Federation are in the range of a factor 2 to 5 and within the Russian Federation they range from 2 to 10. It is difficult to assign a quantitative value based on these factors but probably the inaccuracy in the Russian emission budget is about 200 to 500% which probably means that several emission sources (e.g. large point sources) are excluded from the Russian emission inventory. The exclusion of several large emission sources near the region of station RU01 can be an explanation for the factor 10 difference found at that site.

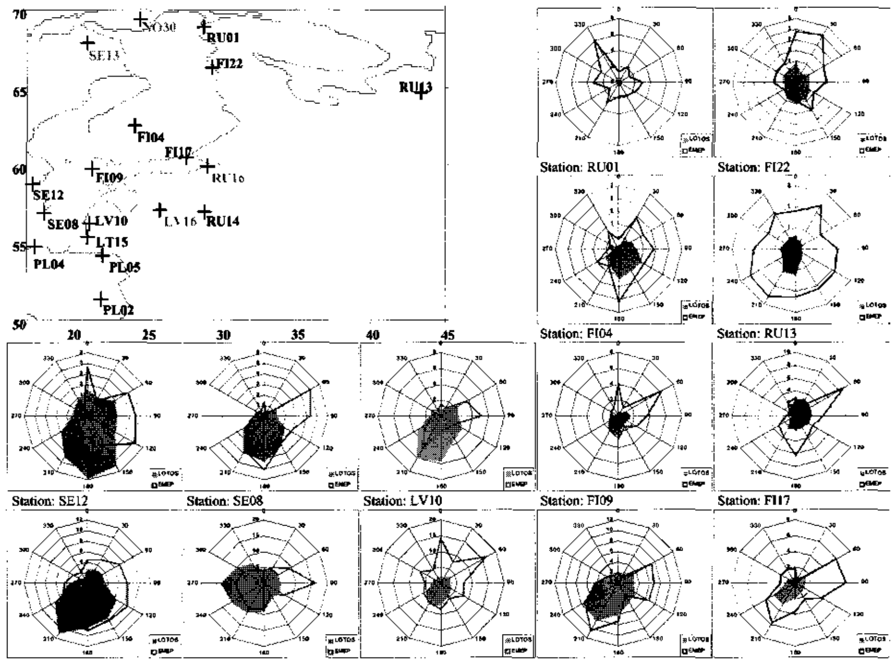


Figure 6.4 Wind-direction-dependent modelled (LOTOS) and observed (EMEP) SO_2 concentrations (in µg SO_2) at measuring stations surrounding the western part of the Russian Federation.

6.3.3 Central England

The comparison of calculated and measured concentrations within the United Kingdom (Figure 6.5) shows that several stations point towards an overestimation of modelled SO_2 concentrations from the direction of Central England. Stations with a clear wind-direction-dependent discrepancy are: GB04 (SE-S, SW-W), GB14 (S, SW), GB15 (SW), GB16 (S-SW). These stations show that the modelled values are 2-3 higher than measured values from these wind directions. The results for stations GB02, GB06 and GB07 and GB13 do not show a clear discrepancy between model and measurement from Central England. Station GB02 shows a good fit, except for slightly higher modelled values from southeastern directions (20-30%). The results for station GB06 also show a good fit between model and measurement. Since GB06 is situated in Northern Ireland, this station is not influenced by emissions from Central England. As shown in Figure 6.5 the radar charts for stations GB07 and GB13 show that wind direction data from Central England are not included in the analysis because of missing data. At station GB07, the modelled and measured concentrations show a rather close agreement. At station GB13 the measurement data from Western winds are a factor 4 higher than the modelled data. The explanation for the discrepancy at station GB13 can be found by looking at Figure 6.1. The emissions per grid cell show that within the model, the emissions in the Cornwall region are located in the sea and not the mainland, which means that grid cell allocation of emissions is the cause of the higher measurement vales from western wind direction. The data from the British measurement network only represent wintertime SO_2 concentrations since no measurements from the months April to October are available. This could indicate that the measurements from the United Kingdom might be erroneous. This is unlikely since stations from other countries also point towards an overestimation of modelled SO_2 concentrations from wind directions that transport SO_2 from the United Kingdom to other areas. These stations are NL09, DE01, NO01, NO08, DK03 and FR05. Based on this, we conclude that the emissions from Central England are overestimated. The graphs of the daily averaged SO_2 concentrations from LOTOS versus those from EMEP do not show a temporal pattern of differences. Therefore, the emission inventory of the United Kingdom is inaccurate because of the SO_2 budget or because of inaccuracies in the spatial distribution of the emissions. The stations within the United Kingdom show that the modelled values are approximately two times higher than the measured concentrations from the direction of Central England and that at other wind directions the model and the measurements agree except for station GB13 which shows an underestimation of modelled values because of dislocation of emissions (see Figure 6.1). It might therefore be reasonable to assume that as with the case of station GB13 some emission sources are not correctly positioned in the emission inventory of the UK leading to overestimation of emissions at specific location by approximately 200%.

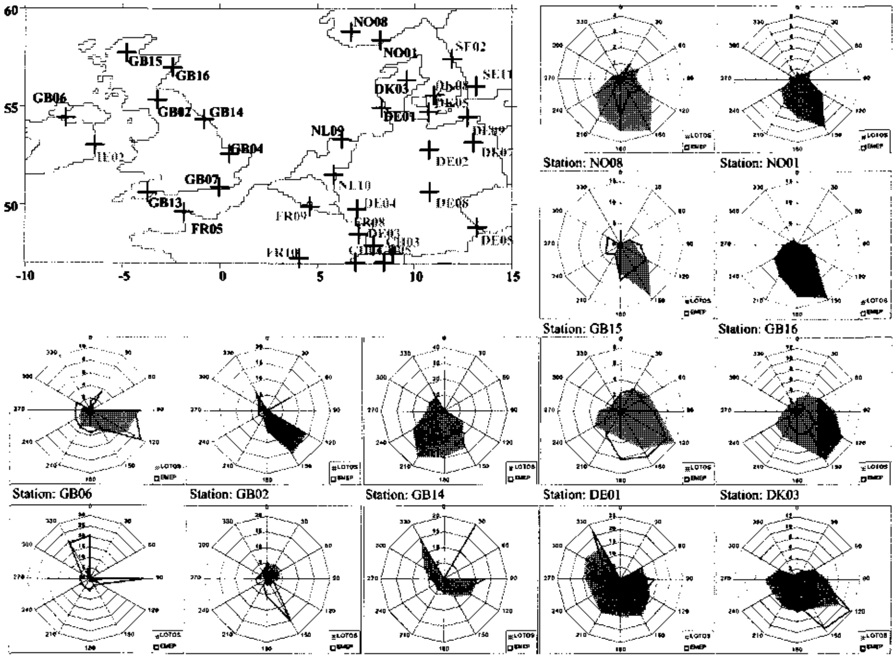
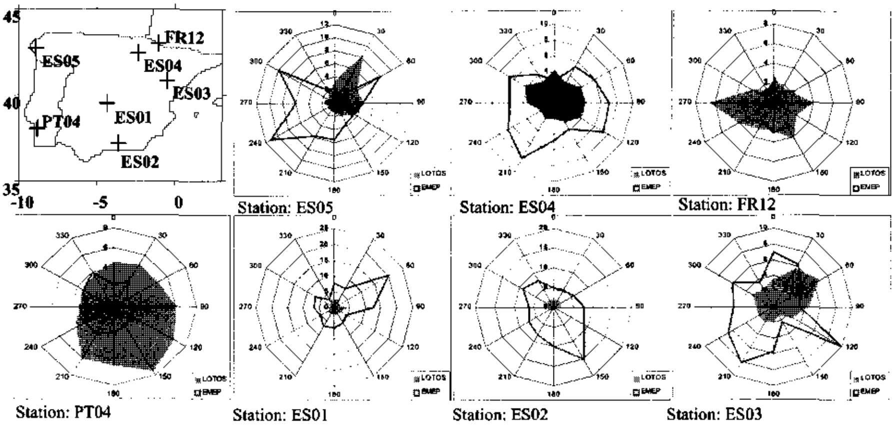


Figure 6.5 Wind-direction-dependent modelled (LOTOS) and observed (EMEP) SO_2 concentrations (in μg SO_2) at measuring stations surrounding the United Kingdom.

6.3.4 Spain

The Spanish stations ES01, ES02, ES03, ES04 and ES05 (Figure 6.6) show that the measured SO_2 concentrations are high compared to the calculated concentrations. For the stations ES01 and ES02 the boundary conditions of the LOTOS model could cause this discrepancy. However, the results for station PT04 do not support this hypothesis. Another cause of the difference between model and measurement could be an underestimation of emission of Spain. The radar plots of stations ES02 and ES05 show that with wind coming from the sea the underestimation of the model is the largest (factor 14 at stations ES02 and a factor 4 to 9 at ES05). This could mean that emissions from ships are underestimated in the emission inventory, but this does not explain the differences found at stations ES01, ES03 and ES04. The radar plot of ES05 also shows that with winds from northeastern direction the modelled concentrations are a factor 6 higher than the measured values. This difference is not found at station ES04, but measurement station FR12 in France shows that with Western winds the modelled concentrations are 3 times higher than the measured concentrations. However, since station FR12 is located in the Pyrenees (mountain area, where LOTOS does not perform well) no conclusion could be drawn. The results for Spain show that there are several discrepancies found between model and measurement and that not enough information is available to determine ship emissions, overestimation of emission in Northwest Spain or boundary conditions as the cause of the discrepancy.



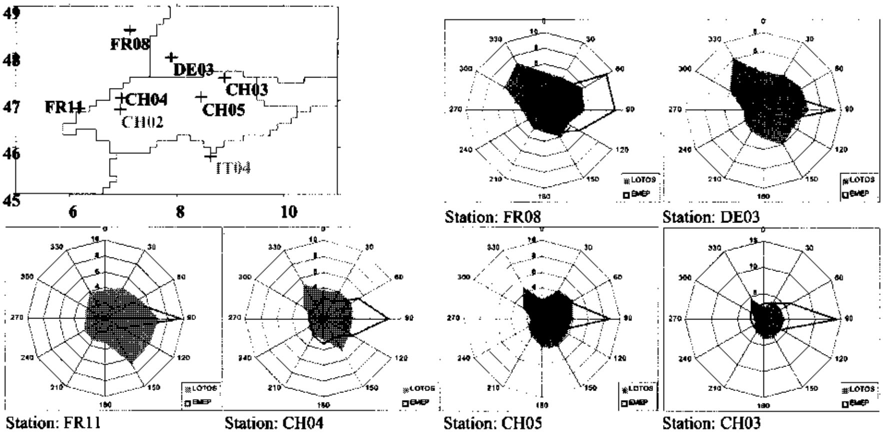
Station: ES05 Station: ES04 Station: FR12

Station: PT04 Station: ES01 Station: ES02 Station: ES03

Figure 6.6 Wind-direction-dependent modelled (LOTOS) and observed (EMEP) SO_2 concentrations (in μg SO_2) at measuring stations surrounding Spain.

6.3.5 Alpine region

The results for the Swiss stations CH03, CH04, CH05, the German stations DE03 and the French stations FR08 and FR11 (Figure 6.7) show that with an easterly wind the model underestimates SO_2 concentrations. These stations are all located at higher altitude, so that the discrepancy can be caused by improper model performance at higher mountainous regions. However, the graphs with the daily modelled and measured concentrations for the stations CH03, CH04, CH05 and DE03 show a clear peak in measured SO_2 concentrations in mid-February and at the end of December. This is less clear for the stations FR08 and FR11. Since the wind-direction-dependent difference is clearly present only in the wind direction 90° at a specific period within the year one could think of influences of a single large point source that is not accounted for in the emission inventory. Measurement error as cause for this wind dependent difference seems unlikely since measurements are available from different countries. Nevertheless, we conclude that for the observed wind-direction-dependent differences in the Alpine region no clear explanation is possible. Either the model performance in mountainous regions or emission inventory error caused by uncertainty of large point source emission can be the cause for the observed differences.



Station: FR08 Station: DE03

Station: FR11 Station: CH04 Station: CH05 Station: CH03

Figure 6.7 Wind-direction-dependent modelled (Model) and observed (Obs.) SO_2 concentrations (in μg SO_2) at measuring stations surrounding the Alpine region.

6.4 Conclusion

The objective of this study was to analyse whether wind-direction-dependent differences between modelled and observed concentrations could be used to identify and quantify sources and size of inaccuracy in large-scale emission inventories like an emission database for European SO_2 emission in 1994. We have studied the wind-direction-dependent differences between measured SO_2 concentrations for 1994 in Europe from the EMEP network with calculated SO_2 concentrations from the LOTOS model that were derived using the SO_2 emission inventory as input.

When the wind-direction -dependent differences from several measurement stations from different countries point towards a specific region inaccuracy in the emission estimate is the likely cause for the discrepancy between model and measurement. If this critierion is not in agreement, then we cannot distinguish between model error, emission inventory error or measurement error. Based on this and as discussed in section 3, we identified five regions where the difference between model and measurement showed a wind-direction-dependent pattern. In three cases (Sachsen/Brandenburg, Russian Federation and Central England) the conclusion could be drawn that these discrepancies are caused by inaccuracy in the emission inventory with as likely source the inaccuracy of the spatial distribution of emissions (Sachsen/Brandenburg, Central England) or the emission budget (Russian Federation). In two cases (Spain and Alpine region), no clear conclusion could be drawn because the available measurements did not allow distinguishing between either model, emission inventory or measurement error.

Our results show that this type of external uncertainty analysis is a useful tool in the assessment of uncertainty in large scale emission inventories, provided that atmospheric measurements from different countries are available for the analysis and that the inaccuracy in the emission inventory has a single cause (budget, spatial distribution or temporal distribution). This was the case for the regions Sachsen/Brandenburg, Central England, and Russian Federation. When more sources of inacuracy are probable (as was the case for Spain), this method does not allow for distinguishing between them. Also in areas where the dispersion model is known to be less effective (mountain regions) a distinction of a single cause of inaccuracy is not possible.

The results also show that sufficient prior information is needed about the quality of the measurements and about the model performance in certain regions. In our case information on measurement quality was available from Hanssen and Skjelmoen (1995). However, they did not provide information on all measurement sites, so we arbitrarily assumed that at several stations the measurement error was negligable (just as assuming an error of zero, a typical or averaged value of error would introduce assumptions, not facts). As far as model performance is concerned, the model has been evaluated for ozone (Hass et al., 1997) which showed that the model performance is limited in mountain regions but that at other areas the model performed well.

Although we were able to identify several areas within Europe where the emission database is concluded to be responsible for the discrepancy between model calculation and observations, this type of external assessment of inaccuracy has several limitations.

First, this type of analysis is not possible without a number of assumptions introducing uncertainty: (i) When information on measurement quality is lacking we assumed that the measurements are non-erroneous. (ii) When we find a large discrepancy between modelled and observed values without a specific wind-direction-dependency, the cause for the discrepancy is considered a systematic model error. (iii) Within a LOTOS grid cell size of 1^0 x 0.5^0 the concentrations are assumed to be well mixed so that a point measurement can be compared with a grid cell average.

Second, we had to aggregate the modelled SO_2 concentrations and wind directions to daily averaged values because of the difference in temporal resolution with the measurement data. This introduces additional uncertainties, especially in the determination of the daily averaged wind direction. As far as spatial resolution is concerned, the analysis was limited by the availability of measurement data in certain regions (e.g. Nordrhein Westphalia) or not possible for the southeastern part of Europe.

Third, despite the quantitative information used to construct the graphical displays of wind-direction-dependent differences, this study is more a qualitative than a quantitative study. Identifying areas where inaccuracy in the emission inventory leads to observed discrepancy between model and measurement is largely a qualitative (visual interpretation) exercise. Quantitative information about the difference between model and measurement in certain wind directions is used solemnly to give a first estimate of the amount of inaccuracy in the emission inventory.

Therefore we conclude that the method described in this study is a tool for identification of sources of inaccuracy in an emission inventory and not so much a tool for quantifying the size of the inaccuracy.

The limitations mentioned above indicate that our method may not be as powerful as other methods such as trajectory analysis and inverse modelling. However, it should be noted that our type of analysis is relatively simple and easy to apply. By using a rather straightforward methodology, inhomgeneous and incomplete measurement data can be used to compare model calculations with measurement data to determine whether the differences between modelled and observed concentrations of air pollution are caused by inaccuracies in the emission inventory. Complicated techniques like data assimilation (e.g. Heemink, 2000) or kriging (e.g. Schaug et al., 1993) that introduce additional assumptions and uncertainties are not needed in this approach. Furthermore, in this study, we were able to conclude about uncertainty about accuracy in the emission budget of a certain country, in the spatial distribution of the emissions and - although not found in the cases presented above- in the temporal distribution of emissions. We were not able to identify certain economic sectors or specific large point sources that cause the emission inventory inaccuracy, but additional research through an internal assessment might provide more insight into the sources of the inaccuracy. Identifying emission inventory inaccuracies as cause for the discrepancy between model and measurement in the cases described above (Sachsen/Brandenburg, Central England and Russian Federation) can be used to further improve the emission estimate and therefore, reduce the ignorance (uncertainty) about the sources and size of inaccuracy in the emission inventory.

Chapter 7

Discussion and conclusions

7.1 Introduction

In this thesis methods to assess the uncertainties in large-scale emission inventories of air pollutants have been studied. Although the term uncertainty is commonly used, different meanings are given to it and a variety of different sources of uncertainty are mentioned in the literature. Furthermore, several methods to qualify or quantify uncertainty have been used or proposed (e.g. EPA, 1996a,b; IPCC/OECD/IEA, 2000). However, there is little experience in the assessment of uncertainties in emission inventories. What is missing in particular is a systematic approach that allows for the identification of the different sources of uncertainty as well as for a qualitative or quantitative assessment of uncertainty in emission inventories. That has been the objective of this thesis. To achieve this objective, three research questions were answered in the different chapters of the thesis.

The methodology of large-scale emission inventory compilation has been presented in chapter 2 (time series of past worldwide emissions of trace gases) and chapter 3 (emission scenarios for NO_x in Asia). These two inventories provided different examples of sources of uncertainty. This information, together with the results of literature research (chapter 4) was used to provide an overview of the potential sources of uncertainty in large-scale emission inventories (research question I). Further, we provided an overview of the different methods that can be used to either identify, qualify or quantify the uncertainty in emission inventories (research question II). The combination of different types and sources of uncertainty and methods for the assessment of these uncertainties provides a systematic approach for uncertainty assessment. This approach is systematic and unique because it allows answering the question what source of uncertainty can be assessed using which tool. Finally, case studies have been performed to test the usefulness of a selection of methods for uncertainty assessment in practice and to determine to what extent uncertainty in emission inventories can be assessed (research question III).

This concluding chapter presents a discussion of the three research questions (sections 7.2, 7.3 and 7.4) followed by a discussion of the framework for potential users (section 7.5). The chapter concludes with general conclusions and recommendations.

7.2 Sources of uncertainty (research question I)

In order to discuss the sources of uncertainty, it should be made clear what is meant with the term uncertainty. In section 4.2 it has been argued that two types of uncertainty in emission inventories can be distinguished: uncertainty about accuracy and uncertainty about reliability. Accuracy is the extent to which an emission inventory is an exact representation of the emission that has occurred in reality. **Uncertainty about accuracy** is the lack of knowledge of the sources and size of the inaccuracy. Reliability means the extent to which one can rely on or trust the outcome of an emission inventory. This reliability is dependent on the purpose of the inventory. For scientific purposes such as atmospheric modelling the reliability is defined by the accuracy of the inventory. For policy purposes the reliability is related to user-specified quality criteria such as transparency or application of agreed upon methodologies. **Uncertainty about reliability** is the lack of knowledge of the degree to which the emission inventory is meeting these user-specified quality criteria.

The uncertainty about reliability and the uncertainty about accuracy exist for different reasons. As described in section 4.3, uncertainty about reliability exists when either the accuracy of the emission inventory is not known or when the documentation of the inventory is inadequate and incomplete, which prevents to determine whether the inventory is for example, transparent, consistent or comparable. The underlying causes for this incompleteness are not within the scope of this study. In this thesis we emphasise uncertainty about accuracy. Uncertainty about accuracy exists because in the field of large-scale emission inventories, such as the work presented in chapters 2 and 3, the variation of emissions over sources, time and space makes it impossible to monitor the emissions at each source continuously. Consequently, an emission factor approach has been adopted to quantify the emissions at higher aggregation levels than the individual source of emissions and by extrapolation of existing data. In section 4.3 a categorisation of the sources of inaccuracy has been presented which is based on information from a variety of emission inventory studies such as presented in Table 4.1 and own experience. This categorisation consists of **structural inaccuracy** and **input value inaccuracy**. Uncertainty about structural inaccuracy (ε_S) is the lack of knowledge of the extent to which the structure of an emission inventory allows for an accurate calculation of the 'real' emissions. Structural inaccuracy has three important causes: aggregation error (ε_{S-1}), incompleteness (ε_{S-2}) and mathematical formulation error (ε_{S-3}). Uncertainty about input value inaccuracy (ε_I) is the lack of knowledge of the values of activity data and emission factors. This type of inaccuracy can have four different causes: extrapolation error (ε_{I-1}), measurement error (ε_{I-2}), unknown developments (ε_{I-3}), and reporting error (ε_{I-4}).

7.3 Methods for uncertainty assessment (research question II)

In section 4.4 we presented a distinction between the assessment of reliability and the assessment of accuracy. The **assessment of uncertainty about reliability** is the judgement whether the emission inventory is meeting the user-specific quality criteria that define the reliability of the emission inventory. The **assessment of uncertainty about accuracy** is the identification of the different sources of inaccuracy and, if possible, a qualitative or quantitative assessment of the inaccuracy of the emission inventory.

Uncertainty about reliability can be assessed through an independent review of calculations, assumptions and documentation. It is important to realize that when the assessment of accuracy is one of the user defined quality criteria, the review process needs to include the accuracy assessment.

For the assessment of uncertainty about accuracy, we distinguished between internal and external uncertainty assessment. In an **internal assessment**, the methodology and information used to construct an emission inventory forms the basis for the assessment of inaccuracy. In an **external assessment** the difference between the emission inventory and external sources of information is used to identify, qualify or quantify inaccuracy in the emission inventory. Different methods for assessment of uncertainty have been presented or discussed in the literature, sometimes with a different focal point. As a result, a variety of methodologies are available for this purpose (EPA, 1999a,b, and IPCC/OECD/IEA, 2000). In this thesis different methods have been identified for application in a framework for uncertainty assessment. For an internal assessment of inaccuracy six methods can be used: (i) qualitative discussion, (ii) data quality rating, (iii) calculation check and evaluation of mathematical formulation, (iv) expert judgement, (v) error propagation and (vi) importance analysis. Four methods can be used for an external assessment of inaccuracy: (i) comparison with other emission inventories, (ii) comparison with direct or indirect measurements, (iii) forward air quality modelling and (iv) inverse air quality modelling.

The information on different capabilities and different sources of inaccuracy in an emission inventory have been combined into a FRamework) for Assessment of Uncertainty in Large-scale Emission INventories (FRAULEIN). FRAULEIN is presented in figure 7.1 and the systematic of the approach is found in the distinction between assessment of uncertainty about accuracy versus uncertainty about reliability and the possibility to select which methods can be used to identify, qualify or quantify what source of uncertainty. Two paths can be followed in the framework. **Path A** addresses uncertainty about reliability and by using peer review a judgement can be given whether user-specified quality criteria have been met. **Path B** addresses uncertainty about accuracy by identification of the sources of inaccuracy in an emission inventory and by qualification or quantification of the extent to which an inventory is inaccurate. In the case where accuracy is one of the quality criteria (path A) the results from the assessment of accuracy (path B) should be used as information source for the assessment of uncertainty about reliability. The outcome of the two assessment paths can for a basis for an evaluation of the possible improvements that can be made with respect to reliability and accuracy.

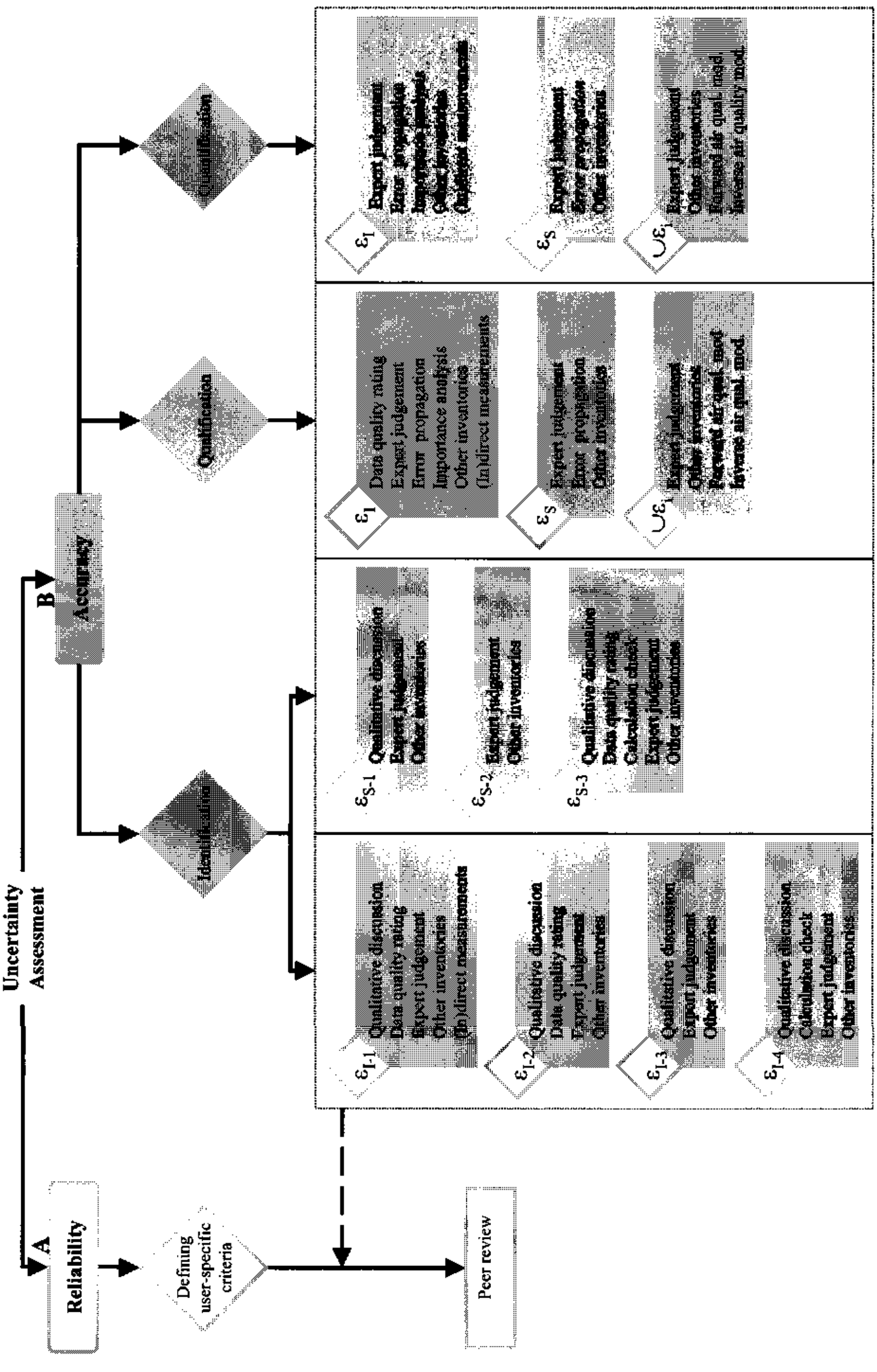


Figure 7.1 A systematic framework for the assessment of uncertainty in emission inventories. Uncertainty about reliability is assessed using path A while uncertainty about accuracy is assessed using Path B. In the case where the assessment of accuracy is one of the quality criteria of the users of the inventory, information from path B is used as input into path A (marked - -).

7.4 The extent to which uncertainty can be assessed (research question III).

The different methods for uncertainty assessment have different capabilities to identify, qualify or quantify uncertainty. Table 7.1 shows these differences and the extent to which application of these methods will result in an assessment of uncertainty about accuracy (see section 4.4 and section 4.5 for more detail). For example, based on Table 7.1, expert judgement may seem to be capable to perform all tasks of an uncertainty assessment, however, the limitation of this tool is that often a clear rationale is missing and that the judgement is subjective. An interesting development in this context is the work by Risbey et al. (2001) who defined a protocol that could enable an indication of the scientific status of the knowledge using a so-called pedigree matrix (see also Van der Sluys, 1997). This does however, not prevent that the expert judgement is sometimes more a guess than based on facts. Another example is the method of inverse air quality modelling. It could be a powerful tool for the quantification of the overall inaccuracy of an emission inventory if not for limitations such as the difficulties in distinguishing between measurement error, emission error or model error or the fact that measurements are often not representative for use in an inverse study. It should be noted that no method exists that can exactly quantify the inaccuracy. Inaccuracies in emission estimates can only be approached. To what extent it can be approached may differ for the different methods.

In theory, some methods may seem to be the best choice to analyse a particular emission, however in practice the method may seem to be a second-choice option or requires information from other methods for uncertainty assessment. For example, the results of performing an *error propagation* on an inventory of N_2O emissions in The Netherlands in 1990 (chapter 5) showed that the quantification of input value inaccuracy (ε_I) is dependent on the statistical interpretation of the available information on uncertainty. This illustrates how the suitability of a specific method for uncertainty assessment is not only determined by the characteristics of the method but also by the availability of information about uncertainty in input value or inventory structure.

The availability of external information is also influencing the selection of the most appropriate method. This is illustrated by the work on time series of past worldwide emissions of trace gases (chapter 2). Our historical SO_2 emission estimates for areas outside Europe and North America were lower compared to other studies (*comparison with other inventories*). However, this difference in emission estimates did not allow for a conclusion that one of the studies is inaccurate because: (a) it is doubtful to what extent the different emission inventories are independent because for historical data limited information sources are available, and (b) the emission inventories that were used for comparison where not subject to an assessment of accuracy. Other methods for the assessment of inaccuracy such as *error propagation and importance analysis*, *(in)direct measurements* or *forward and inverse air quality modelling* were difficult to apply to the historical emission inventories, because of lack of empirical data on emissions or atmospheric concentrations. As a result, an uncertainty assessment of the work on historical emissions is limited to the performance of a *qualitative discussion*, *data quality ratings* and *calculation check* and *expert judgement*. Likewise, the methods that can be used for the assessment of inaccuracy in our work on projections of NO_x emissions in Asia for the period 1990-2020 (chapter 3) is also limited to a *qualitative discussion* and a *calculation check* due to lack of information.

Table 7.1 Overview of different methods for the assessment of uncertainty about accuracy. Shown are a brief description of the different methods, the capability to identify, qualify or quantify inaccuracy associated with input values (ε_I), the structure (ε_S) or the overall inaccuracy of the emission inventory ($\cup\varepsilon_i$) and important limitations. See section 4.3 for explanation of $\varepsilon_{x\text{-}y}$

Method	Description	Capabilities	Limitations
Qualitative discussion	Discussion of known or expected sources of inaccuracy	- Identification of different causes of input value and structural inaccuracy (except $\varepsilon_{S\text{-}2}$)	- Provides only preliminary insight into sources of inaccuracy - Limited by understanding of emitting process - No qualification or quantification
Data quality rating	Expression of relative accuracy of input values by assignment of alphabetical or numerical scores	- Systematic identification of $\varepsilon_{I\text{-}1}$ and $\varepsilon_{I\text{-}2}$ - Qualification of most inaccurate parts of inventory	- Only relative scores - Assignment of scores is subjective - Only assessment of input value inaccuracy
Calculation check and evaluation of mathematical formulation	Check on calculation and mathematical formulation	- Identifying $\varepsilon_{I\text{-}4}$ and $\varepsilon_{S\text{-}3}$ - Correct inventory for $\varepsilon_{I\text{-}4}$ and $\varepsilon_{S\text{-}3}$	- Only assessment of $\varepsilon_{I\text{-}4}$ and $\varepsilon_{S\text{-}3}$ - Limited by knowledge of emitting process
Expert judgement	Asking experts to give estimate of inaccuracy	- Identification of ε_I, ε_S - Qualification/quantification of ε_I, ε_S, $\cup\varepsilon_i$	- Clear rationale for assessment is often missing - Assessment is subjective
Error propagation	Calculation of inaccuracy in inventory induced by inaccuracy in input values (in theory also for structure)	- Quantification of ε_I, ε_S - Based on calculation results, qualification of ε_I, ε_S	- Only assessment of input value inaccuracy - Lack of empirical data
Importance analysis	Calculation of (relative) importance of inaccuracy in input value to inaccuracy of inventory	- Qualification and quantification of ε_I	- Only assessment of input value inaccuracy - Lack of empirical data
Other inventories	Comparison of different (independent) inventories	- Identification of ε_I, ε_S - Qualification and quantification of ε_I, ε_S, $\cup\varepsilon_i$	- Availability of independent emission estimates - Uncertainty about accuracy in other inventory
Direct and indirect measurements	Comparison of input values of inventory with measurement results	- Identification of $\varepsilon_{I\text{-}1}$ and $\varepsilon_{I\text{-}2}$ - Qualification of ε_I - Quantification of ε_I and $\cup\varepsilon_i$	- Only identification of $\varepsilon_{I\text{-}1}$ and $\varepsilon_{I\text{-}2}$ - Lack of continuous monitoring of each emission source
Forward air quality modelling	Comparison of modelled atmospheric concentrations with atmospheric concentrations measurements	- Qualification and quantification of $\cup\varepsilon_i$	- No identification of ε_I, ε_S - Difficulties in distinguishing between measurement error, emission error and model error - Representativeness of measurements
Inverse air quality modelling	Comparison of emission inventory with emissions calculated by an atmospheric dispersion model using atmospheric concentration measurements as input	- Qualification and quantification of $\cup\varepsilon_i$	- No identification of ε_I, ε_S - Difficulties in distinguishing between measurement error, emission error and model error - Representativeness of measurements

Forward air quality modelling may in theory be used for the quantification of inaccuracy of an emission inventory. This is illustrated by an assessment of uncertainty about accuracy of a European emission inventory of SO_2 using the atmospheric transport model LOTOS and measured atmospheric concentrations from EMEP (chapter 6). This analysis illustrates that the applied method resulted in a qualitative rather than a quantitative assessment of inaccuracy by identifying for which regions within the study area the inventory is inaccurate. However, the inaccuracies could not be quantified using the approach. Information from for example, an *error propagation* analysis on the regions that where found to be inaccurate might provide a quantification of inaccuracy.

7.5 Conclusions

In this thesis in emission inventories has been studied. The objective was to develop a systematic approach for the assessment of uncertainty in emission inventories of air pollutants. Different types and sources of uncertainty have been analysed together with the methods that can be used for the assessment of these uncertainties. Based on the analysis a systematic approach, a framework, has been developed for the assessment of uncertainty in emission inventories and the three research questions have been answered. Based on the analysis, the following conclusions can be drawn.

Sources of uncertainty

I. A systematic assessment of uncertainties in emission inventories requires a clear definition of uncertainty. Two types of uncertainties can be distinguished: uncertainty about accuracy and uncertainty about reliability. These have been defined, respectively, as lack of knowledge of the sources and the size of inaccuracy, and lack of knowledge of the degree to which the emission inventory is meeting user-specified quality criteria. This thesis focuses in particular on uncertainty about accuracy.

II. Uncertainties in emission inventories arise from a variety of sources. Sources of inaccuracy include structural inaccuracy and input value inaccuracy. Uncertainty about structural inaccuracy is the lack of knowledge of the extent to which the structure of an emission inventory allows an accurate calculation of the 'real' emission. Uncertainty about input value inaccuracy is the lack of knowledge of the values of activity data and emission factors.

III. Both structural inaccuracy and input value inaccuracy have a variety of causes. Aggregation error, incompleteness and mathematical formulation error. Input value inaccuracy is caused by extrapolation error, measurement error, unknown developments and reporting error.

Method for uncertainty assessment

IV. Assessment of the reliability of an emission inventory is particularly important for inventories that are used for policy purposes. Often, inventory users have defined clear quality criteria. The assessment of uncertainty about the reliability can be based on an independent review of the emission inventory.

V. Assessment of the accuracy of an emission inventory is particularly important for inventories that are used for scientific purposes. A variety of tools are available for the assessment of uncertainty about inaccuracy. We distinguish between approaches that analyse information that has been used to compile the inventory (internal assessment) and approaches that use external sources of information to assess the inaccuracy (external assessment).

VI. A Framework for Assessment of Uncertainties in Large-Scale Emission Inventories (FRAULEIN) has been developed, which may serve as a guide for uncertainty assessment. The framework includes different methods for uncertainty assessment and explains the sources of inaccuracy, and how these can be identified, qualified or quantified.

VII. There are three ways to use FRAULEIN in practice. First, in situations where the method for uncertainty assessment is prescribed, FRAULEIN clarifies the sources of uncertainty that can be identified, qualified or quantified. Second, if the objective of a study is to assess a specific source of uncertainty, FRAULEIN may serve as a guide for selection of the appropriate methods. Third, if the aim is to perform a full uncertainty assessment, FRAULEIN forms the basis of a four-step approach, including (1) identification, qualification (2) and quantification (3) of the sources of inaccuracy, followed by an evaluation to prioritise further research (4).

VIII. The ultimate goal of assessment of uncertainty about accuracy is to reduce the uncertainty as much as possible. In the field of emission inventories continuous emission monitoring of all emission sources without measurement error is practically not possible, and therefore uncertainty about accuracy can only be reduced in a limited way.

The extent to which uncertainties can be assessed.

IX. The extent to which the reliability of emission inventories can be assessed depends on the quality criteria that have been defined by the users of an emission inventory, the documentation of the construction of the emission inventory or –in the case when accuracy is one of the criteria- the results and documentation of the assessment of accuracy.

X. The extent to which inaccuracies in emission inventories can be assessed depends on the source of inaccuracy that needs to be assessed, the capability and limitations of the selected method for uncertainty assessment and the amount of information about the inaccurate aspects of the emission inventory.

Lessons learned from the case studies

The two case studies provided information on the uncertainty of accuracy within the specific emission inventories, and insights in advantages and disadvantages of the different methods for assessment of uncertainty.

XI. The assessment of uncertainty of an inventory of N_2O emissions from agriculture in The Netherlands in 1990 through the combination of error propagation and importance analysis showed that only three uncertain input values have a large influence on the uncertainty in the emission inventory. These are the emission factor for indirect N_2O emissions (EF_5), the fraction of N leaching from agricultural soils ($Frac_{LEACH}$) and the emission factor for direct soil emissions (EF_1).

XII. From a methodological point of view, the results of the N_2O case study shows that quantification of input value inaccuracy (ε_I) through error propagation was influenced by the statistical interpretation of the available information by the IPCC Guidelines (default values, and uncertainty ranges of emission factors in particular). This clearly illustrates that the extent to which inaccuracies can be assessed depends not only on the characteristics of the method used for uncertainty assessment but also on the available statistical information on inventory parameters and that there is a need of a more explicit description of such information in the IPCC Guidelines and in other publications.

XIII. The assessment of uncertainty of an inventory of SO_2 emissions in Europe in 1994 using measurement data and forward air quality modelling, shows that in three regions within the inventory domain inaccuracy in the emission inventory is the most likely cause for the discrepancy between model calculations and measurements. These regions are Sachsen/Brandenburg (Germany), Central England and the western part of the Russian Federation. In Sachsen/Brandenburg and Central England the regional distribution of the emissions seems to be inaccurate and in the western part of the Russian Federation the total emission estimate seems to be inaccurate.

XIV. From a methodological point of view the case study on SO_2 emissions shows that a rather straightforward method such as the analysis of wind-direction-dependent differences is able to identify inventory inaccuracy from inaccuracies in the air quality model and atmospheric measurements. However, it is rather a tool for identifying parts of the inventory that are inaccurate than a tool to quantify the inaccuracy of the emission inventory.

7.6 Recommendations

Developers and users of emission inventories can apply FRAULEIN in the following way. First, it should be determined whether the uncertainty about reliability or uncertainty about accuracy is to be addressed. For assessment of uncertainty about reliability the process of peer review should be used (path A in figure 7.1). In the case of assessment of accuracy path B should be selected. The framework can be used in different ways:

(i) When the methods for uncertainty assessment have been prescribed (e.g. due to requirements or availability of information) the framework shows which sources of uncertainty about accuracy can be identified, qualified or quantified and even more important which sources cannot (Figure 7.1 and Table 7.1). Error propagation is a widely used tool for the assessment of inaccuracies in emission inventories. Furthermore, the IPCC Good Practice Guidance (IPCC/OECD/IEA, 2000) prescribes error propagation as a tool to assess the accuracy of national greenhouse gas inventories. It is important to realize that when error propagation is used to quantify the inaccuracy due to input value uncertainty (ε_I), the overall inaccuracy of the inventory $\cup\varepsilon_i$ is not quantified. The overall inaccuracy is resulting from inaccuracies associated with both input values and inventory structure.

(ii) Given a source of inaccuracy (e.g. uncertainty due to extrapolation), the framework can be used to determine the methods that could be used for uncertainty assessment (Figure 7.1 and Table 7.1).

(iii) For a full assessment of the inaccuracy of the inventory resulting in guidance on reduction of uncertainties, a four step approach can be followed:

Step 1, Identification: Major causes of inaccuracy can be identified using expert views, qualitative discussion and comparison with other inventories. In this step both uncertainties in sources of emission and estimation method can be traced.

Step 2, Qualification: A qualitative assessment and eventually a ranking of the inaccuracies. The tools used in the first step may be used again in an attempt to derive more than just identification from them. In addition the tools data quality ranking and direct and indirect measurements can now be used.

Step 3, Quantification: Tools used in earlier steps may be stretched to gain a beginning of an insight in the quantitative value of the inaccuracies. In addition, quantification of inaccuracy becomes possible through application of error propagation, importance analysis and forward and inverse air quality modelling.

Step 4, Evaluation: Whereas steps 1-3 gradually improve the insight in the cause and size of the uncertainty about accuracy, step 4 uses these results to assist in prioritising future research. The most important uncertainties could then be reduced first, taking into account that it is sometimes very difficult if not impossible to reduce a large uncertainty.

There are several issues not addressed within this thesis which could provide a starting point for further research.

First, it may be interesting to investigate to what extent FRAULEIN may be applicable to other research fields. FRAULEIN provides a framework for the assessment of uncertainty in large-scale emission inventories. However, the emission inventory community is not the only community that has to deal with uncertainty assessment. There is a growing interest in uncertainty issues, as illustrated by the work on integrated assessment studies such as Van der Sluijs (1997) and Van Asselt (2000), and workshops on uncertainty assessment such as (EFIEA, 1999). FRAULEIN may contain useful elements to be used by other research fields such as integrated environmental assessment modelling.

Second, this thesis does not address the question to what extent it is needed to reduce inaccuracies in emission inventories. The answer to this question depends on the use of the emission inventory. Obviously, when an inventory is used for compliance checking, the inventory does not need to be of higher quality than is needed to check the targets. The required accuracy might be very well dependent upon whether the targets are set as absolute values (as is the case in the Convention on Long Range Transboundary Air Pollution) or set as reduction targets as is the case for the Kyoto Protocol under the Framework Convention on Climate Change. In air quality modelling applications, the required accuracy of the inventory should be derived from the accuracy of the model. In cases where the inventory itself is the final goal of the research project, one could argue that the accuracy should be as high as possible.

Third, the communication of uncertainty to inventory users may need further attention. In most studies uncertainty is quantified through the values of a confidence interval. For scientific purposes this might be the best way to express the accuracy. Whether or not this is the best way to communicate uncertainty to users in the policy field could be questioned, because of the risk of misinterpretation of statistical information.

Finally, it is important to realize that most inventories are prepared for policy purposes. An inventory that is considered to be reliable for policy purposes, however, may not be accurate enough for scientific analyses. This raises the question if and when emission inventories made for policy purposes can be used for scientific purposes.

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Summary

Emission inventories provide information about the amount of a pollutant that is emitted to the atmosphere as a result of a specific anthropogenic or natural process at a given time or place. Emission inventories can be used for either policy or scientific purposes. For policy purposes, emission inventories can be used to monitor the progress of environmental policy or to check compliance with conventions and protocols. For scientific purposes, emission inventories can be used as input into atmospheric dispersion models that are aimed at understanding the chemical and physical processes and the behaviour of air pollutants in the atmosphere. A strict separation between policy and scientific oriented emission inventories is not always possible. The usefulness of emission inventories for policy or science depends on the accuracy and the reliability of the inventories. There is uncertainty about an emission inventory when the accuracy and reliability of the emission estimates are not known. Proper use of emissions inventories requires an assessment of the uncertainties, including identification, qualification and quantification of the uncertainty. Although different methods for the assessment of uncertainty in emission inventories have been proposed, a systematic approach for identification, qualification and quantification of uncertainty does not exist. The objective of this thesis is to develop such a systematic approach for large-scale inventories. In order to meet this objective three research questions have been formulated:

(i) What are the potential sources of uncertainty in emission inventories
(ii) Which methods can be followed for the assessment of uncertainty
(iii) To what extent can uncertainty in emission inventories be identified, qualified or quantified.

The methodology of emission inventory compilation typical for large-scale emission inventories has been illustrated by two emission inventories. In chapter 2, time series of past worldwide emission of anthropogenic trace gases for the period 1890 – 1990 are described. Chapter 3 presents projections for NO_x emissions in Asia for the period 1990 –2020. The construction of these emission inventories was hampered by the lack of experimental data on the different sources of emission. As a result, the emissions were calculated on another scale than on which the emission processes occur in reality. The activity data and emission factors were based on extrapolation of existing information. Due to these aggregations and extrapolations, the emission inventories are inaccurate representations of the actual emissions.

Chapter 4 describes the theoretical basis for our definitions of uncertainties, followed by a categorisation of uncertainties in emission inventories. It is argued that two types of uncertainty in emission inventories exist. *Uncertainty about accuracy* is the lack of knowledge about the sources and size of the inaccuracy. *Uncertainty about reliability* is the lack of knowledge about the degree to which the emission inventory is meeting user-specified quality criteria. These user-specified criteria depend on the purpose of the emission inventory. For scientific purposes the reliability is defined by the accuracy of the inventory. For policy purposes, quality criteria can be related to transparency, application of agreed upon methodologies or sometimes also to the assessment of accuracy. Uncertainty about reliability exists when either the accuracy of the emission inventory is not known or when the documentation of the inventory is inadequate and incomplete. Uncertainty about accuracy exists when the different sources of inaccuracy or the extent to which the inventory is inaccurate is not known. A categorisation of uncertainty about different sources of inaccuracy has been presented. Uncertainty about *structural inaccuracy* is the lack knowledge about the extent to which the structure of an emission inventory allows for an accurate calculation of the 'real' emission. Three causes for structural inaccuracy have been defined. These are aggregation error, incompleteness and mathematical formulation error. Uncertainty about *input value inaccuracy* is the lack of knowledge about the values of activity data and emission factors. Four causes for input value inaccuracy have been identified. These are extrapolation error, measurement error, unknown developments and reporting error.

Uncertainty about reliability can be assessed through peer review. For the assessment of inaccuracy, a distinction is made between internal and external assessment of uncertainty. In *an internal assessment*, the methodology and information to construct an emission inventory form the basis for the assessment of inaccuracy. Based on review of available methodologies six methods for internal assessment are proposed: (i) qualitative discussion, (ii) data quality rating, (iii) calculation check and evaluation of mathematical formulation, (iv) expert judgement, (v) error propagation and (vi) importance analysis. In an *external assessment,* the difference between the emission inventory and external sources of information is used to identify, qualify or quantify inaccuracy in the emission inventory. Four methods can be used: (i) comparison with other emission inventories, (ii) comparison with (in)direct measurements, (iii) forward air quality modelling and (iv) inverse air quality modelling.

Against this background we developed a systematic approach for the assessment of uncertainty in emission inventories. This framework, FRAULEIN (FRamework for the Assessment of Uncertainty in Large-scale Emission INventories) can be used to assess uncertainty about reliability and uncertainty about accuracy. It provides guidance for selection of the methods that can be used to identify, qualify or quantify different sources of uncertainty.

Several methods included in the framework have been analysed in more detail to identify the advantages and disadvantages of these methods in practice. Chapter 5 presents the results of assessment of uncertainties in estimates of 1990 N_2O emissions from agriculture in The Netherlands using the methods of error propagation and importance analysis. The results indicate that only a small number (three out of 23) of uncertain inventory parameters have large share in the inaccuracy of the emission inventory. These parameters include emission factors for indirect N_2O emissions (EF_5), the fraction of N leaching from agricultural soils (Fracleach) and the emission factor for direct soil emissions (EF_1). Reducing the inaccuracy in the inventory should therefore focus on improved quantification of indirect emissions (based on EF_5 and Fracleach) and direct soil emissions (EF_1). From a methodological point of view, the results of the N_2O case study show that quantification of input value inaccuracy through error propagation is influenced by the statistical interpretation of the available information in the IPCC Guidelines (default values, and uncertainty ranges of emission factors in particular). This result provides an indication that the extent to which inaccuracies can be assessed depends not only on the characteristics of the method used for the assessment but also on the available information on inventory parameters. Identification of inventory parameters having the largest share in the inaccuracy, on the other hand, was not influenced by the statistical interpretation of IPCC information.

Chapter 6 describes the results of assessment of uncertainty in a European emission inventory of SO_2 in 1994 using forward air quality modelling and atmospheric measurements. The problem with this type of assessment is that it is not easy to pinpoint emission inventory inaccuracy as single cause of the deviation between measurements and model results. Inaccuracies exist in both the inventory, model and measurements. In the case study it has been analysed whether wind-direction-dependent differences between calculated and measured concentrations can be used to assess inaccuracies in emission inventories. The results indicate that in three regions within the study domain inaccuracy in the emission inventory is the most likely cause for the discrepancy between modelled and observed SO_2 concentrations. These regions are Sachsen/Brandenburg (Germany), Central England and the western part of the Russian Federation. In Sachsen/Brandenburg and Central England the spatial distribution of the emissions seems to be inaccurate while in the western part of the Russian Federation the total emission estimate seems to be inaccurate. We developed a relatively simple method to identify inventory inaccuracies based on differences between the air quality model and atmospheric measurements. However, it was also shown that the method is primarily a tool for identifying relatively inaccurate parts of the inventory. The method cannot be used to analyse causes of the inaccuracies, such as inaccurate structure or input values. Furthermore, it was concluded that the method is more a qualitative than a quantitative approach.

There are three ways to use FRAULEIN in practice. First, in situations where the method for uncertainty assessment is prescribed, FRAULEIN clarifies the sources of uncertainty that can be identified, qualified or quantified. Second, if the objective of a study is to assess a specific source of uncertainty, FRAULEIN may serve as a guide for selection of the appropriate methods. Third, if the aim is to perform a full assessment of inaccuracy, FRAULEIN forms the basis of a four-step approach: (1) identification, qualification (2) and quantification (3) of the sources of inaccuracy, followed by evaluation to prioritise further research (4).

Samenvatting

Emissie inventarisaties bevatten kwantitatieve informatie over de hoeveelheden van milieuverontreinigende stoffen die in het milieu terecht komen als gevolg van een specifiek antropogeen of natuurlijk proces op een bepaalde plaats of een bepaald moment. In dit proefschrift staan inventarisaties van emissies naar de lucht centraal. Deze emissie inventarisaties worden zowel voor beleids- als voor wetenschappelijke doeleinden ontwikkeld. Zo kunnen ze gebruikt worden bij de evaluatie van milieubeleid of bij het controleren van naleving van verdragen en protocollen. In wetenschappelijke onderzoek worden emissie inventarisaties gebruikt als invoer in atmosferische verspreidingsmodellen. Met deze modellen wordt onderzoek gedaan naar de chemische en fysische processen in de atmosfeer, alsmede het gedrag van luchtverontreinigende stoffen. Een strikte scheiding tussen beleids- en wetenschappelijk georiënteerde emissie inventarisaties is niet altijd mogelijk. De bruikbaarheid van emissie inventarisaties voor beleid of wetenschap is afhankelijk van de nauwkeurigheid en betrouwbaarheid van de emissie schattingen. Er is spake van onzekerheid wanneer de nauwkeurigheid en de betrouwbaarheid van emissie schattingen niet bekend zijn. Een goed gebruik van emissie inventarisaties behoeft vaststelling van deze onzekerheden. Ondanks het feit dat verschillende methoden voor het vaststellen van de onzekerheid in emissie inventarisaties zijn beschreven, bestaat er nog geen systematische benadering voor het identificeren, kwalificeren en kwantificeren van onzekerheid. Het doel van dit proefschrift is om een dergelijke systematische benadering te ontwikkelen. Hiertoe zijn drie onderzoeksvragen geformuleerd:

(i) Wat zijn de potentiële bronnen van onzekerheid in emissie inventarisaties.
(ii) Welke methoden kunnen gebruikt worden bij het vaststellen van onzekerheid.
(iii) In welke mate kan onzekerheid in emissie inventarisaties worden geïdentificeerd, gekwalificeerd of gekwantificeerd.

In dit proefschrift worden twee voorbeelden gegeven van inventarisaties op een hoog aggregatieniveau. In hoofdstuk 2 wordt een inventarisatie van mondiale antropogene emissies voor de periode 1890 – 1990 beschreven. Vervolgens wordt in hoofdstuk 3 een projectie van emissies van stikstofoxiden (NO_x) in Azië voor de periode 1990 – 2020 beschreven. Deze twee hoofdstukken geven een aantal voorbeelden van onzekerheden in emissie schattingen. Zo bleek gebrek aan experimentele gegevens over de verschillende bronnen van luchtverontreinigende stoffen in verleden en toekomst een belemmering bij het schatten van emissies. Een gevolg hiervan is dat

emissies veelal zijn berekend op een ander schaalniveau dan de schaal waarop de stoffen in werkelijkheid vrijkomen en dat de gegevens over menselijke activiteiten en de gebruikte emissiefactoren veelal zijn gebaseerd op extrapolatie van de bestaande informatie. Als gevolg van deze aggregaties en extrapolaties zijn veel emissieschattingen onnauwkeurige representaties van de feitelijke emissies.

In hoofdstuk 4 staat de theoretische basis voor onze definities van onzekerheid beschreven, gevolgd door een categorisering van onzekerheden in emissie inventarisaties. Er worden twee typen van onzekerheden in emissie inventarisaties onderscheiden. *Onzekerheid over nauwkeurigheid* is het gebrek aan kennis over de bronnen en grootte van de onnauwkeurigheid. *Onzekerheid over betrouwbaarheid* is het gebrek aan kennis over de mate waarin de emissie inventarisatie voldoet aan gebruikersspecifieke kwaliteitscriteria. Deze kwaliteitscriteria zijn afhankelijk van het gebruiksdoel van de emissie inventarisatie. Voor wetenschappelijke doeleinden wordt de betrouwbaarheid veelal bepaald door de nauwkeurigheid van de inventarisaties. Voor beleidsdoeleinden zijn de kwaliteitscriteria gerelateerd aan bijvoorbeeld de transparantie van de informatie, de mate waarin afgesproken schattingsmethoden zijn toegepast, maar soms ook aan de nauwkeurigheid. Er bestaat onzekerheid over de betrouwbaarheid wanneer de nauwkeurigheid van een emissie inventarisatie onbekend is, of wanneer de achterliggende documentatie ontoereikend of incompleet is. Er bestaat onzekerheid over nauwkeurigheid wanneer de verschillende bronnen of de omvang de onnauwkeurigheid onbekend zijn.

Verschillende bronnen van onnauwkeurigheid kunnen worden onderscheiden. Er is sprake van *structurele onnauwkeurigheid* wanneer de structuur van een emissie inventarisatie resulteert in een onnauwkeurige kwantificering van de emissie. Er kunnen tenminste drie oorzaken van structurele onzekerheid worden onderscheiden: aggregatiefouten, onvolledigheid en fouten in de wiskundige formulering. De *onnauwkeurigheid van invoerwaarden* vloeit voort uit gebrek aan kennis over de activiteiten data en emissie factoren. Vier oorzaken van onnauwkeurigheid van invoerwaarden kunnen worden onderscheiden: extrapolatiefouten, meetfouten, onbekende ontwikkelingen en rapportagefouten.

Onzekerheid over betrouwbaarheid kan worden vastgesteld op basis van "peer reviews". De onnauwkeurigheid van een inventarisatie kan worden bepaald op basis van een interne of een externe vaststelling van onzekerheid. In een *interne analyse* vormt de methodiek en informatie die ten grondslag ligt aan de emissie inventarisatie de basis voor de vaststelling van onnauwkeurigheid. Er bestaan zes manieren om een interne analyse uit te voeren: (i) kwalitatieve discussie, (ii) kwaliteitsbeoordeling van gegevens, (iii) controle van berekeningen en wiskundige formulering, (iv) beoordeling door experts, (v) foutenpropagatie en (vi) analyse van het relatieve belang van verschillende onnauwkeurigheden. In een *externe analyse* wordt een externe bron van informatie gebruikt voor identificatie, kwalificatie of kwantificatie van onnauwkeurigheid in de emissie inventarisatie. Vier methoden kunnen worden gebruikt: (i) vergelijking met andere emissie inventarisaties, (ii) vergelijking met directe en indirecte metingen, (iii) voorwaartse luchtkwaliteitsmodellering and (iv) inverse luchtkwaliteitsmodellering.

Tegen deze achtergrond hebben we een systematische methode voor de vaststelling van onzekerheden in emissie inventarisaties ontwikkeld: FRAULEIN (FRamework for the Assessment of Uncertainty in Large-scale Emission INventories). Deze methode kan gebruikt worden voor het bepalen van de onzekerheid over betrouwbaarheid en nauwkeurigheid. Het framework vormt een leidraad voor het selecteren van methoden, die gebruikt kunnen worden voor het identificeren, kwalificeren of kwantificeren van onzekerheid.

Een aantal methoden die zijn opgenomen in FRAULEIN toegepast, om mogelijke voor- en nadelen ervan in de praktijk te illustreren. De eerste toepassing betreft een analyse van onnauwkeurigheid in de schatting van de uitstoot van het broeikasgas lachgas (N_2O) uit de Nederlandse landbouw in 1990 (hoofdstuk 5). De onnauwkeurigheid is bepaald op basis van een foutenpropagatie en een analyse van het relatieve belang van van individuele onnauwkeurigheden. Uit de analyse blijkt dat slechts een klein aantal (3 van 23) onzekere parameters een relatief grote bijdrage leveren aan de totale onnauwkeurigheid van de emissie inventarisatie. Deze parameters zijn de emissiefactor voor de zogenaamde indirecte N_2O emissies (EF_5), de veronderstelde fractie mest die uitspoelt uit landbouwbodems ($Frac_{leach}$) en de emissiefactor voor directe emissie uit landbouwgrond (EF_1). Deze resultaten kunnen gebruikt worden voor het stellen van prioriteiten voor onderzoek. Onderzoek ter vermindering van de onnauwkeurigheid van de inventarisatie zal zich bij voorkeur richten op de indirecte emissies (gebaseerd op EF_5 en $Frac_{leach}$) en de directe bodememissies (EF_1). De foutenpropagatie vereiste een statistische interpretatie van de beschikbare informatie in de IPCC richtlijnen voor het schatten van nationale broeikasgasemissies. In deze richtlijnen zijn standaardwaarden voor emissiefactoren en onzekerheidsranges van emissiefactoren opgenomen. De statistische interpretatie van deze factoren en ranges bleek van invloed op de resultaten van de foutenpropagatie. Dus niet alleen de gebruikte methode voor onzekerheidsanalyse is bepalend voor de mate waarin de onnauwkeurigheid kan worden vastgesteld, maar ook de beschikbare informatie over de gebruikte parameters bij het schatten van de emissies. Het identificeren van de parameters met de grootste bijdrage aan onnauwkeurigheid werd niet beïnvloed door de statistische interpretatie van de IPCC informatie.

De tweede toepassing betreft het gebruik van voorwaartse luchtkwaliteitsmodellering en atmosferische metingen in de analyse van onnauwkeurigheden in emissieschattingen. In hoofdstuk 6 worden de resultaten beschreven van een analyse van de onnauwkeurigheden in een inventarisatie van zwaveldioxide (SO_2) emissies in Europa in 1994. In de studie is geanalyseerd of verschillen (per windrichting) tussen berekende en gemeten SO_2 concentraties kunnen worden gebruikt om onnauwkeurigheden in de emissie inventarisatie vast te stellen. Een probleem met een dergelijke analyse is dat het niet eenvoudig is om de onnauwkeurigheid van de emissie inventarisatie aan te wijzen als enige oorzaak voor de afwijking tussen de gemeten en berekende atmosferische concentraties. Onnauwkeurigheden bestaan zowel in de inventarisatie, het model als in de metingen, of een combinatie daarvan. Toch bleek het mogelijk in een aantal gevallen conclusies te trekken over de nauwkeurigheid van de emissieschattingen. Voor drie regio's is geconcludeerde dat onnauwkeurigheden in de emissie inventarisatie de meest waarschijnlijke oorzaak zijn voor de afwijking tussen berekende en gemeten SO_2 concentraties. Deze regio's zijn Sachsen/Brandenburg (Duitsland), centraal Engeland en het westelijk gedeelte van de Russische Federatie. In

Sachsen/Brandenburg en centraal Engeland lijkt vooral de ruimtelijke verdeling van de emissies een bron van onnauwkeurigheid te zijn. In het westelijk gedeelte van de Russische Federatie lijkt de schatting van de totale emissies onnauwkeurig te zijn. De hier gehanteerde methode om onnauwkeurigheid in de emissie inventarisaties te identificeren is relatief eenvoudig. Deze methode moet echter beschouwd worden als een instrument voor het identificeren van relatief onnauwkeurige onderdelen van de inventarisatie. De methode kan niet gebruikt worden om de onderliggende bronnen van onnauwkeurigheid vast te stellen. Het is daarom meer een kwalitatieve dan een kwantitatieve benadering.

Er zijn drie manieren waarop FRAULEIN in de praktijk toegepast kan worden. Wanneer een methode voor onzekerheidsvaststelling is voorgeschreven, kan met behulp van FRAULEIN inzichtelijk worden gemaakt welke bronnen van onzekerheid kunnen worden geïdentificeerd, gekwalificeerd en gekwantificeerd. Wanneer men een specifieke bron van onnauwkeurigheid wil bepalen, kan FRAULEIN dienen als een wegwijzer voor het kiezen van de meest geschikte methode daarvoor. Tot slot, kan in vier stappen de onnauwkeurigheid van een emissie inventarisatie worden bepaald met behulp van FRAULEIN: (1) identificatie, (2) kwalificatie en (3) kwantificering van de bronnen van onnauwkeurigheid, gevolgd door (4) een evaluatie om prioriteiten te stellen voor vervolgonderzoek

Curriculum vitae

John Albert van Aardenne was born on February 15th, 1970 in Rijswijk, The Netherlands. He completed his secondary school education (VWO) at the Sint Willibrord College in Goes in 1990. In the same year he started his study in Environmental Sciences at Wageningen Agricultural University with specialisation in air quality management, and graduated in 1996. During his study he spent six months at the Center for Global and Regional Environmental Research of the University of Iowa in Iowa City (United States of America). In 1997 he worked at the Institute for Marine and Atmospheric research Utrecht (IMAU) of Utrecht University. Later that year he started his PhD research on 'uncertainties in emission inventories' at the Environmental Systems Analysis Group of Wageningen University. The research was sponsored by the TNO institute of Environmental Sciences, Energy and Process Innovation as part of the TNO and Wageningen University Expertise Centre for Emissions and Assessment. During the PhD research, NWO sponsored in 1999 a two month visit to United States Environmental Protection Agency in North Carolina (Emission Factors and Inventories Group) and Washington D.C. (Climate Policy and Programs Division). In 2001 he joined the Emission and Assessment Group of TNO. As of May 1, 2002 he works for the Max Planck Institute for Chemistry at the Department of Atmospheric Chemistry (Mainz, Germany).