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Reactive nitrogen in atmospheric emission inventories

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Abstract. Excess reactive Nitrogen (Nr) has become one of the most pressing environmental problems leading to air pollution, acidification and eutrophication of ecosystems, biodiversity impacts, leaching of nitrates into groundwater and global warming. This paper investigates how current inventories cover emissions of Nr to the atmosphere in *Europe*, the *United States of America*, and *China*. The focus is on anthropogenic sources, assessing the state-of-the-art of quantifying emissions of Ammonia (NH_3), Nitrogen Oxides (NO_x) and Nitrous Oxide (N_2O), the different purposes for which inventories are compiled, and to which extent current inventories meet the needs of atmospheric dispersion modelling. The paper concludes with a discussion of uncertainties involved and a brief outlook on emerging trends in the three regions investigated is conducted.

Key issues are substantial differences in the overall magnitude, but as well in the relative sectoral contribution of emissions in the inventories that have been assessed. While these can be explained by the use of different methodologies and underlying data (e.g. emission factors or activity rates), they may lead to quite different results when using the emission datasets to model ambient air quality or the deposition with atmospheric dispersion models. Hence, differences and uncertainties in emission inventories are not merely of academic interest, but can have direct policy implications when the development of policy actions is based on these model results.

The level of uncertainty of emission estimates varies greatly between substances, regions and emission source sectors. This has implications for the direction of future research needs and indicates how existing gaps between modelled and measured concentration or deposition rates could be most efficiently addressed.

The observed current trends in emissions display decreasing NO_x emissions and only slight reductions for NH_3 in both Europe and the US. However, in China projections indicate a steep increase of both.

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1 Introduction

1.1 Aims and objectives

Nitrogen gas (N_2) accounts for more than 99.99% of all the nitrogen present in the atmosphere, while of the rest, again 99% is accounted for by nitrous oxide (N_2O) (Wallace and Hobbes, 2006). Other N species are thus only present in trace concentrations, but nonetheless play a vital role in atmospheric chemistry. Ammonia (NH_3) is the most abundant alkaline gas in the atmosphere and is responsible for neutralising acids formed through the oxidation of sulphur dioxide (SO_2) and nitrogen oxides (NO_x), creating ammonium (NH_4^+) salts of sulphuric and nitric acid, which become atmospheric aerosols. The only other alkaline gases in the atmosphere are also reduced nitrogen, such as volatile amines, though these are present in much smaller quantities. Oxidized nitrogen, mainly originating as nitric oxide (NO) and nitrogen dioxide (NO_2) plays a crucial roles both in tropospheric and stratospheric chemistry, for instance in the formation of tropospheric ozone (see Bradshaw et al., 2000; Brasseur et al., 1999).

Nitrogen is thus present in the atmosphere in a multiplicity of chemical forms, contributing both the majority (78%) of the atmosphere as N_2 and a plethora of trace N components that are fundamental to the atmosphere's chemical and radiative properties. It is convenient to distinguish these N forms into two main groups: N_2 being termed non-reactive (or "fixed") nitrogen, with the sum of all other N forms present being termed "reactive nitrogen" (Nr) (Galloway et al., 2003, 2008).

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The amounts of Nr in the world are of fundamental importance to society. Since the invention of the Haber-Bosch process it has become possible to synthesise huge amounts of NH_3 directly from N_2. The key benefit has been to feed the increasing world population, which is offset by the consequent increases in Nr losses to the environment (e.g. Erisman et al., 2008). In parallel, high temperature combustion of fossil fuels oxidizes atmospheric N_2, causing a huge increase in NO_x emissions in the atmosphere (Lee et al., 1997). The alteration in agricultural practice and increase in fossil fuel combustion impacts human health, acidification and eutrophication of soils, biodiversity change in terrestrial ecosystems, nitrate eutrophication in freshwater and marine ecosystems and affects the global radiative balance. Furthermore, N_2O emissions are not only contributing to global warming with a significantly higher global warming potential than CO_2 and CH_4, but have recently been identified to substantially contribute to stratospheric ozone depletion on a global scale (Ravishankara, 2009). And as N_2O emissions from agricultural soils are closely linked to NH_3 emissions from fertilizer and manure application, it is sensible to address N_2O emissions in the context of this study, even though N_2O is typically not classed as reactive nitrogen (N_r).

While there is sufficient evidence to demonstrate the substantial perturbation of the global nitrogen cycle, the exact quantification of the magnitude and spatial distribution of this perturbation is presently subject to in-depth research. The NitroEurope research project (Sutton et al., 2007; http://www.nitroeurope.eu) is working towards deriving more precise nitrogen balances from local to regional scales. A major challenge in compiling nitrogen budgets and quantifying pools and fluxes of nitrogen in the atmosphere and biosphere is that models and measurements need to be of good and known quality to allow for a validation of results and ultimately provide the scientific understanding of processes on all relevant spatial scales. The variety of sources of different forms of nitrogen being emitted into the atmosphere, soils and water bodies, their heterogeneous distribution in space and the often high uncertainties regarding specific N fluxes creates a challenge for the validation and verification of models. At present, closing the gaps in the nitrogen budgets is the aim of major efforts on a global scale. One example for this is the European Nitrogen Assessment Report (compiled under the auspices of the ESF Programme Nitrogen in Europe and the International Nitrogen Initiative), which is due to be completed by early 2011 and which aims to improve the understanding of the European N cycle and move towards closing the gaps. Emission inventories are a critical source of information in this process. They are most often compiled for compliance monitoring purposes, e.g. EMEP under the UNECE Convention on LRTAP or the United Nations Framework Convention on Climate Change (UNFCCC), using officially submitted data compiled by country experts. Other inventories are compiled by researchers with the main aim to provide a consistent and comprehensive set of input data for modelling purposes (e.g. the EDGAR datasets, IIASA GAINS), often using a bottom-up approach with information on emission factors and activity data that are publicly accessible.

In this paper, we focus on regional scale nitrogen emission inventories for the year 2005, with the aim to assess how different methods and data sources influence the resulting inventory datasets. In a second step, we analyse the main drivers for differences identified, and consider how these may affect the usefulness of datasets for atmospheric modelling. Finally, we briefly discuss emission trends from inventory datasets and scientific literature, considering the implications for our conclusions on the use of emission datasets for modelling purposes.

1.2 Scope

Anthropogenic activities have a significant impact on the magnitude of N cycled and released into the atmosphere (see Table 1), for instance, Galloway et al. (2004) calculated a global rate of annual creation of reactive nitrogen (Nr) of 163 Tg N yr^{-1} in the early 1990s, compared to 125 Tg N yr^{-1} around 1860.

The following Table 2 displays the total N emissions in Tg for the three regions on which this paper focuses. It illustrates both the similarities and differences between different inventories and the contribution to the atmospheric domain of global N emissions, which based on the EDGAR v4 inventory (EDGAR, 2009) amounts to approx. 32 Tg N in the year 2005.

At the same time, the scientific understanding of many environmental effects of excess nitrogen in the atmosphere has significantly advanced in recent years. Current research into the critical loads of N deposition both for acidification and eutrophication (see Hettelingh et al., 2008) has led to the establishment of more stringent critical loads, with dynamic modelling approaches being explored to assess the timescales of ecosystem damage and recovery. In addition, the relevance of N_2O as a contributor to global warming has been acknowledged and emission control strategies no longer focus solely on CO_2, which is reflected e.g. by a more detailed integration of N_2O sources and measures in the GAINS model (Greenhouse Gas – Air Pollution Interaction an Synergies, http://gains.iiasa.ac.at/) or MITERRA (Velthof et al, 2009).

Given the importance of nitrogen-containing species for air quality and climate change, the question emerges if today's inventories of NH_3, NO_x and N_2O reflect the current knowledge on emissions of these trace gases. Furthermore, the parallel but rather separated development of emission inventories under air quality related activities (such as the UNECE *Convention on Long Range Transboundary Air Pollution*) and under the climate change focus of the *Intergovernmental Panel for Climate Change* (IPCC) and the *United Nations Framework Convention on Climate Change* (UNFCCC)

Table 1. Overview of the main sources and sinks of atmospheric nitrogen-containing species. N_2 has been listed for completeness, but is not discussed further, as the paper focuses on reactive N species only.

Sources	(N_2)	NH_3	NO_x	N_2O
Biogenic emissions from the terrestrial and marine biosphere		X	X	X
Decomposition of proteins and urea from animals		X		
Biomass burning and fossil fuel combustion	(X)	X	X	
Agricultural mineral Nr fertilisation and denitrification	(X)	X	X	X
Lightning			X	
Sinks				
Wet deposition (as NH_4^+ and NO_3^-)		X	X	
Dry deposition		X	X	
Biological nitrogen fixation	(X)			
Industrial nitrogen fixation (mainly Haber-Bosch process)	(X)			
Chemical breakdown in the stratosphere				X

Table 2. Emissions in Tg Nitrogen (Tg N) for the three regions analysed in this paper based on a global inventory (EDGAR v4) and national submissions (to EMEP, IPCC) for the year 2005. In the case of the China, no official national inventory was available; NH_3 emissions as estimated for 2006 by Yan et al. (2003) and other data from the IIASA GAINS China model have been used for the comparison. The last column shows global figures of Tg N emitted based on the EDGAR v4 dataset, as well as the share of emissions of the three regions investigated contributing to global emissions in brackets and *italics*.

	EU25[a]		USA		China		Global
	EDGAR v4	*EMEP*[1]*/IPCC*[2]	*EDGAR v4*	*US EPA*	*EDGAR v4*	*IIASA*[3]*/Yan et al. 2003*[4]	*EDGAR v4*
NH_3	4.35	3.30[1]	2.81	3.02	8.43	5.80[4]	33.4 *(46.7%)*
NO_x	3.51	3.49[1]	5.48	5.61	5.59	5.20[3]	28.1 *(51.8%)*
N_2O	0.63	0.82[2]	0.46	0.65	0.68	1.18[3]	3.9 *(45.1%)*
	8.48	7.61	8.75	9.28	14.69	12.18	65.4 *(48.8%)*

[a]EU27, except Malta and Cyprus, omitted for consistency with figures.

has led to different accounting systems with often differing national budgets for the same trace gas. In this paper, we compare how different approaches to inventory compilation may lead to similar or quite different results and – where possible – discuss likely reasons for differences observed. Specifically, we assess whether the quality of current emission inventories is sufficient to support integrated strategies for N management, which are emerging in the US and Europe (Erisman et al., 2007). For this purpose, we here assess existing inventories with regard to their total numbers and *sectoral structure*. The *temporal* and *spatial resolution*, *speciation* and *accessibility of inventories* are not the focus of this paper, but will be addressed where necessary.

In the following sections, the current situation of emission inventories in Europe, the US and China is discussed with the focus on how emissions of NH_3, NO_x and N_2O are estimated and allocated to source sectors. Aspects of spatial and temporal resolution, sectoral detail on emission sources and completeness of reporting are addressed for the legislative and regulatory regimes under which data are compiled. Where different inventories are compiled, a comparison and analysis of potential variations will be conducted, alongside uncertainty assessments.

For this purpose, the following inventories and data sources have been analysed in detail:

- National submissions of NO_x and NH_3 emission date to the EMEP programme
- National submissions of N_2O emission data to the UNFCCC/IPCC
- The US National Emissions Inventory (NEI) dataset for NO_x and NH_3
- Inventory of US Greenhouse Gas Emissions and Sinks for N_2O
- The EDGAR global emission inventory dataset for NO_x, NH_3 and N_2O (version 4 representing the most up-to-date emissions; EDGAR Fast Track 32 and EDGAR HYDE for trends and for comparison purposes)

– Literature data in particular for the China and for global/regional comparisons.

The datasets listed are openly accessible, in most cases directly online. The EDGAR v4 inventory (EDGAR, 2009) has only been partly published at this stage (greenhouse gases). However, the authors had access to a preliminary version of the dataset for air pollutants, which will be published in the near future, subject to error corrections to which the comparisons made in this paper may contribute (http://edgar.jrc.ec.europa.eu/). For an overview over all data sources and URLs, see Table 3.

On a global scale, the need for nitrogen management has been formulated in the 2004 Nanjing Declaration (Erisman 2004, UNEP 2004), which was presented to the *United Nations Environment Programme* (UNEP) in Nanjing, China on October 16, 2004, with the aim to optimise nitrogen management in food and energy production on a local, regional and global scale.

2 European emission inventories of reactive nitrogen species

In Europe, emissions of ammonia and nitrogen oxides are covered by several regulatory regimes, both under the UN-ECE CLRTAP and directives of the European Commission. Member states of the European Union and parties to the protocols under the CLRTAP are subject to mandatory emission reporting. For the UNECE, the EMEP Centre on Emission Inventories and Projections (CEIP) at the Umweltbundesamt Vienna, Austria, hosts inventory datasets (both official submissions of signatories to the different protocols of the CLRTAP and emissions for modelling purposes) in an online-accessible database (http://www.ceip.at/).

Nitrous oxide on the other hand is not covered by the CLRTAP, but is required to be reported under the UNFCCC by Table 3 countries. The UNFCCC GHG inventory submissions can be accessed http://unfccc.int/ghg_emissions_data/items/3800.php online.

In addition to these inventories which are generated based on obligatory reporting of national emissions, the EDGAR database provided global annual emissions per country and on a 1×1 degree grid for 1990 and 1995 for direct greenhouse gases CO_2, CH_4, N_2O and HFCs, PFCs and SF6 and the precursor gases CO, NO_x, NMVOC and SO_2. Similar inventories have been compiled for acidifying gases, NH_3, NO_x and SO_2 and Ozone Depleting Gases (EDGAR v4 as well as EDGAR 3.2/FT 2000, see EDGAR, 2009). For this paper, the new EDGAR dataset version 4 (v4 in the following text) is used, which has recently been (partly, for GHGs) officially released and of which a preliminary version was made available to the authors by the EDGAR team (http://edgar.jrc.ec.europa.eu/). EDGAR v4 (EDGAR, 2009) provides emissions for all relevant air pollutants and GHGs with an improved spatial resolution and until 2005.

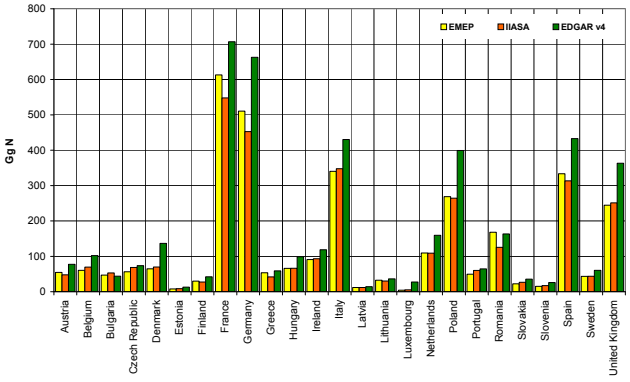

Fig. 1. Comparison of EU27 (not showing Malta and Cyprus) emissions of ammonia (expressed in Gg N) reported to EMEP and compiled by IIASA and the EDGAR v4 database for the year 2005. The slight differences between IIASA estimates and EMEP figures arise most likely from bilateral consultations with country experts, which led to corrections in agricultural emissions that had not (yet) been reflected in the EMEP inventories by recalculations of the year 2000 emissions submitted.

2.1 Ammonia

In the case of NH_3, the vast majority of emissions from the 27 member states of the European Union (EU27) originate from agriculture (93%), with some small contributions from waste management (2.5%), industrial production processes (2%) and road transport (1.8%) (EMEP, 2009). This sectoral distribution is valid for most countries, with slight difference depending on the state of the art of agricultural production and, for instance, livestock intensity. For ammonia, a large number of non-agricultural sources contribute a small amount of emission (Sutton et al., 2000). Because the individual contributions are small for these sources (e.g. wild animals, direct emissions from humans, sewage management) many countries do not report emissions for all these terms. For example, detailed analysis of these non-agricultural emissions for the UK showed that they contribute around 15% of total ammonia emissions (Sutton et al., 2000; Dragosits et al., 2008). This is double the share noted above for the EU27 as a whole, which clearly indicates that a more comprehensive discussion of NH_3 emissions and sources is needed.

Figure 1 illustrates that differences between the EMEP dataset and data used for model calculations with the GAINS model by IIASA (IIASA, 2009) are marginal for most countries. This was anticipated, as the EMEP emissions displayed represent official submissions by countries, which, in ultimately, form the basis for the IIASA data through a validation process by extensive bilateral consultations with country experts, often the same experts preparing the inventories reported to EMEP.

For some countries, however, the IIASA data are slightly different, with overall emissions for the EU27 being 4.4%

Table 3. Overview over all regional emission inventories used in this paper.

Inventory	Substances covered	Time scale covered	Geographical coverage	Spatial disaggregation	URL/Reference
EMEP	NH_3, NO_x, CO, SO_2, NMVOC, PM, Heavy Metals, POPs	1980–2007 (reported) projections to 2030	Europe, Russian Federation, Belarus, USA, Canada *(area covering all parties to the CLRTAP)*	50 km×50 km (10 km×10 km planned)	http://www.ceip.at
	Parties to the Convention on Long-Range Transboundary Air Pollution (CLRTAP) are legally required to report emission data to the European Monitoring and Evaluation Programme (EMEP) on an annual basis. This includes spatially disaggregated data every 5 years (2000, 2005, . . .) as well as supporting information (activity data, Informative Inventory Report IIR). Theoretically, a full inventory should be available with approx. a two year time lag (e.g. data for 2007 have to be reported in 2009), however, individual parties do not always report on time or fully complete datasets. While the EMEP Centre for Emissions and Projections (CEIP) monitors reporting compliance and supports inventory reviews, datasets for modelling have to be gap-filled frequently to allow for modelling over the full spatial domain, without data gaps.				
EDGAR v4	CO_2, N_2O, CH_4, F-Gases, CO, NO_x, NMVOC, NH_3, SO_2, $PM_{2.5,10}$, BC, OC, POPs	2000–2005	Global *(Regions/Countries)*	0.1°×0.1°	http://edgar.jrc.ec.europa.eu
	The EDGAR 4 dataset is thelatest version of a series and is currently maintained and hosted by the Joint Research Centre of the European Commission at Ispra/Italy. With its global coverage, comprehensive set of substances and high spatial resolution, the EDGAR 4 inventory is designed for use as input for global/regional atmospheric and climate models. It is not based on reported emissions, but calculated bottom-up using emission factors (EFs) and estimated activity rates for each source sector.				
EDGAR32 FT2000	CO_2, CH_4, N_2O, F-Gases, CO, NMVOC, NO_x and SO_2	2000	Global *(Regions/Countries)*	1°×1°	http://www.mnp.nl/edgar/model/v32ft2000edgar/
	The Fast Track (FT) 2000 dataset has been published in 2005, based on the EDGAR 3.2 estimates for 1995 and prepared by trend analyses at country level for each standard source category of EDGAR 3.2. Thus, the sectoral structure and other features for EDGAR 3.2 and FT 2000 are the same. As one caveat, NH_3 emission data have not been calculated for the year 2000 in either EDGAR 3.2 or FT 2000 and have only been available for 1990 based on GEIA/EDGAR 2.1 data.				
EDGAR Hyde 1.3/1.4	CO_2, CH_4, N_2O, CO, NO_x, NMVOC, SO_2 and NH_3	1890–1990 *(10 year time steps)*	Global *(Regions/Countries)*	1°×1°	http://www.mnp.nl/edgar/model/100_year_emissions/edgar-hyde-1.3/ http://www.mnp.nl/edgar/model/100_year_emissions/edgar-hyde-1.4/
	Based on EDGAR v2 and the History Database of the Global Environment (HYDE) global anthropogenic emissions for the period 1890–1990 by 13 world regions and on 1 x 1 degree grid have been constructed (Van Aardenne et al., 2001). EDGAR Hyde has been developed by the National Institute for Public Health and Environment (RIVM) and Utrecht University, The Netherlands.				
UNFCCC	CO_2, CH_4, N_2O, PFCs, HFCs, SF_6	1990–2006	Global *(Parties to the UNFCCC)*	N/A	http://unfccc.int/ghg{_}data/items/3800.php
	Parties to the United Nations Convention on Climate Change (UNFCCC) report greenhouse gas (GHG) emissions annually to UNFCCC. The sectoral structure of the Common Reporting Format (CRF) is similar to the NFR used for EMEP reporting, but mainly tailored for fossil fuel combustion sources.				
IIASA GAINS	SO_2, NH_3, NO_x, VOC, PM (TSP, PM_{10}, $PM_{2.5}$ and PM_1), BC, OC, CO_2, CH_4, N_2O, F-Gases	2000–2030	Europe (EU27 and Central and Eastern European Countries), Russian Federation, China, South-Asia, Rest of the World	N/A (mapping of model output)	http://gains.iiasa.ac.at
	The GAINS model has been implemented for the analysis of air pollution control and greenhouse gas mitigation strategies for a number of regions. The online version of GAINS provides access to all input datasets, including emissions in year time steps. For comparing with European inventory datasets, the NEC2007 baseline scenario has been selected.				

below the EMEP figures. The reason for this small difference in emissions that have the same underlying data sources is most likely revised animal numbers or more detailed emission calculations that have been available to the experts during the consultations, but have not yet been used to submit recalculated inventory figures to EMEP. The comparison with the EDGAR v4 dataset (EDGAR, 2009) shows that EDGAR emissions are (consistently) higher for the bulk of EU27 countries (32%). As a major difference, the EDGAR emissions for agricultural sources are significantly higher than those reported to EMEP, with emissions from agricultural soils being most likely the main contributor to the difference observed. A more detailed analysis is not straightforward due the degree of completeness of EMEP emissions reported based on the current reporting format (Nomenclature for Reporting, NFR08), which distinguishes in sufficient detail emissions within the agricultural sector (currently available for 10 countries). An in-depth assessment is thus conducted for the case of the UK in Sect. 6.2.2.

For the European region, including the EU27, the accession countries (Turkey, FYR of Macedonia and Croatia), as well as Norway and Switzerland, the EMEP inventory amounts to 4.5 Tg of NH_3 for the year 2005 (3.3 Tg N). This is comparable with an estimate of 4.1 Tg NH_3 for the whole of Europe made by Bouwman et al. (1997) for the year 1990 and an estimate of 5.3 Tg NH_3 (4.3 Tg N) by EDGAR (2009) for the same set of European countries. Galloway et al. (2004) only give a combined figure for Europe and the former Soviet Union (FSU), with atmospheric emissions of ammonia calculated at 8 Tg N yr^{-1}, with FSU emissions in 1990 at 3.4 Tg N yr^{-1} according to the EDGAR dataset.

2.2 Nitrogen oxides

Nitrogen oxides have been the focus of significant emission control activities in recent decades, both for stationary sources (mainly large combustion plants) and mobile sources (especially road transport). In general it should be anticipated that NO_x emission figures are less uncertain than those of NH_3 or N_2O and the initial comparison between EMEP and EDGAR datasets for 2005 confirms this with only slight differences for individual countries (see Fig. 2). Total NO_x emissions (expressed as $NO+NO_2$) for Europe are estimated at 11.5 Tg NO_x by both EDGAR v4 and EMEP (EDGAR, 2009; EMEP, 2009) for the year 2005 (3.5 Tg N).

This comparison indicates that NO_x emissions are better understood than for instance NH_3, in general. National totals do not display large variations between inventories; however, sectoral differences can be significant for individual countries. Issues such as an overall lack of measurement programmes, for instance for new vehicle technologies in road transport, and the uncertainties in the effects of decentralised power generation in a liberalised energy market on power plant emissions are likely to have an effect on the quality of NO_x inventory datasets also in the future. For an in-depth assessment of e.g. sectoral differences between inventories, up-to-date and documented national emission factors based on measurements for different technologies would be vital.

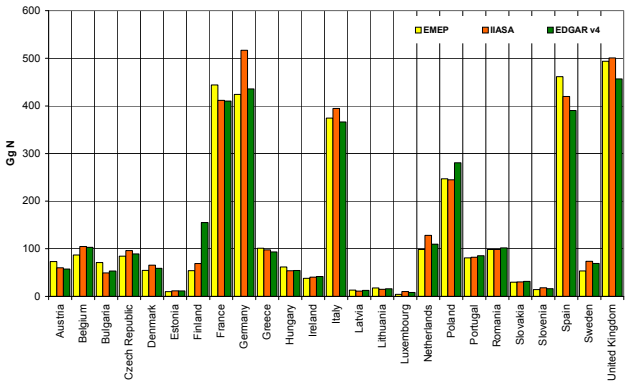

Fig. 2. Comparison of EU27 (not showing Malta and Cyprus) emissions of NO_x (expressed as Gg N) reported to EMEP (EMEP, 2009) and presented in the EDGAR database (EDGAR, 2009) for the year 2005.

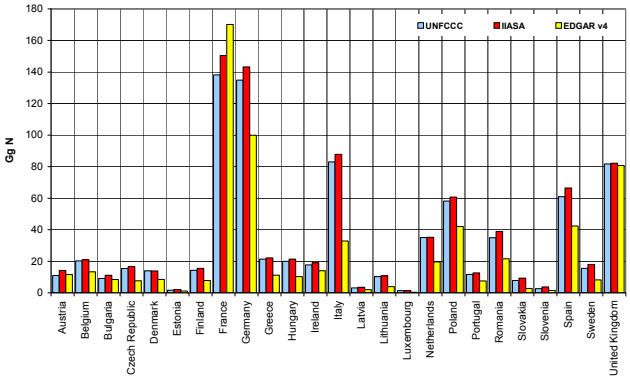

Fig. 3. Comparison of EU27 (not showing Malta and Cyprus) emissions of N_2O (expressed as Gg N) reported to UNFCCC (without LULUCF, UNFCCC, 2009) and presented in the EDGAR v4 database (EDGAR, 2009) for the year 2005.

2.3 Nitrous oxide

Since N_2O emissions are not reported under the CLRTAP, but subject to reporting obligations to the UNFCCC for Table 3 countries, the comparison is made between data collected under the UNFCCC and the EDGAR v4 inventory (Fig. 3).

For most countries, figures in both inventories are quite similar, with EDGAR showing an overall lower emission for the EU27 of about 24%. For comparison, figures from the IIASA GAINS model are displayed in Fig. 3 as well. While EDGAR shows higher emissions or France (23%), most other figures are substantially lower than UNFCCC data, for instance Finland (−45%), Italy (−60%) or Sweden (−47%). At this stage, it is difficult to assess fully

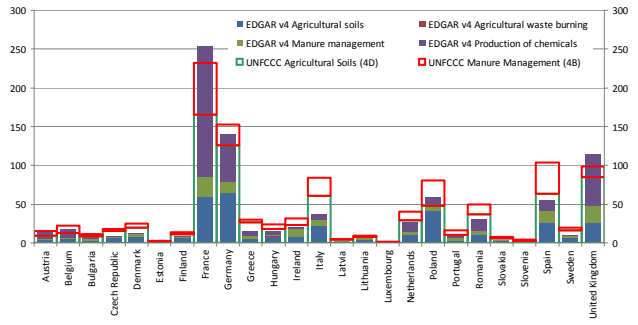

Fig. 4. Relative contributions of N_2O emissions (in Gg N_2O) from agricultural sources soils and manure management in the EU27 countries (not displaying Cyprus and Malta) according to the EDGAR v4 inventory (EDGAR, 2009) and compared to major agricultural sources as reported to UNFCC (Agricultural Soils, NRF 4D; Manure Management, NRF 4B) (UNFCCC, 2009).

the uncertainty in either dataset, but it should be stated that recent findings of Skiba et al. (2001) provide a methodology for the calculation of N_2O emissions from soils, one of the main sources of N_2O, which results in higher emissions than the current UNFCCC established methodology. Within the EU27, 48% of N_2O emissions in the EDGAR v4 inventory stem from agricultural sources (34.2% from agricultural soils, 13.9% from manure management and 0.1% from agricultural waste burning). Shares of emissions from agricultural soils in individual countries vary significantly, between 9.6% (Greece) and 67.6% (Denmark). Figure 4 highlights the variation of relative contributions of agricultural soils and manure management to N_2O emissions in the EU27 countries for a comparison between EDGAR v4 and UNFCCC. A third contributing source to N_2O emissions in the EU27 is the chemical industry sector, contributing about 28% of total N_2O emissions in 2005 in EDGAR v4. It is likely that emissions from the chemical industry are well understood and hence emission factors and activity rates can be assumed to be less uncertain than those from agricultural soils or manure management. Hence, the differences between UNFCCC and EDGAR data most likely mainly arise from the application of different emission factors (or activity rates) in these two sectors.

Figure 4 illustrates the difference between the main sources of N_2O emissions with a focus on emissions from agricultural sources. There is a clear indication that emissions reported to UNFCCC under CRF (Common Reporting Format) categories 4B (Manure Management) and 4D (Agricultural Soils) are substantially higher than those estimated in EDGAR v4 under the same headings.

However, even in those cases were both inventories provide similar figures, a more thorough investigation of emissions from agricultural soils may be required in the light of the findings of Skiba et al. (2001), Crutzen et al. (2008) and Mosier et al (1998), which indicate a potential underestimation of N_2O emissions from soils in current inventories. The increasing demand for bio fuels could even lead to a larger underestimation, unless future emission factors take these findings into account.

3 Emission inventories of reactive nitrogen species in the United States of America

The US Environmental Protection Agency is charged with developing the National Emission Inventory (NEI, US EPA, 2009a) in support of the Clean Air Act and subsequent amendments. The NEI includes an accounting of pollutants that impact air quality, including NO_x and NH_3. These efforts have recently been reviewed in an assessment report by NARSTO (2005). In addition, the EPA prepares an estimate of N_2O emissions in the US Inventory of Greenhouse Gas Emissions and Sinks (US EPA, 2009b), in accordance with the UNFCCC.

3.1 Ammonia

The NEI estimates that more than 80% of total USA ammonia emissions are from livestock manure management and application of chemical fertilizers. The next largest source is ammonia from vehicles equipped with catalytic converters, which comprises approximately 7% of the inventory. While the NEI and EDGAR database have similar total agricultural and vehicle emissions, 14% of the EDGAR NH_3 emissions are from industrial combustion, while this source is less than 1% in the NEI.

Because of the operational challenges in measuring ammonia emissions and a lack of detailed animal husbandry practices data, ammonia emission estimates have high uncertainty. Independent efforts to quantify the seasonal variability have shown agreement for winter and summer emissions, but differ for the spring and fall (Gilliland et al., 2006: Pinder et al., 2006: Henze et al., 2008). Because atmospheric agricultural emissions are rarely regulated, the trend in emissions is expected to be proportional to the increase in livestock population and acres under cultivation. Both are expected to increase in coming years (USDA, 2007).

3.2 Nitrogen oxides

The NEI estimates that the largest contributors of US NO_x emissions include on-road vehicles (32%), off-road vehicles (30%), such as ships, aircraft, and construction equipment, and electricity and industrial power generation (27%). Emissions from on-road and off-road vehicles are calculated using the National Mobile Inventory Model (NMIM) (http://www.epa.gov/otaq/nmim.htm). While not used in this study, a more advanced mobile source emissions model, MOVES (http://www.epa.gov/otaq/models/moves/), is currently under development and a draft version is available for public use. Important improvements include more detailed

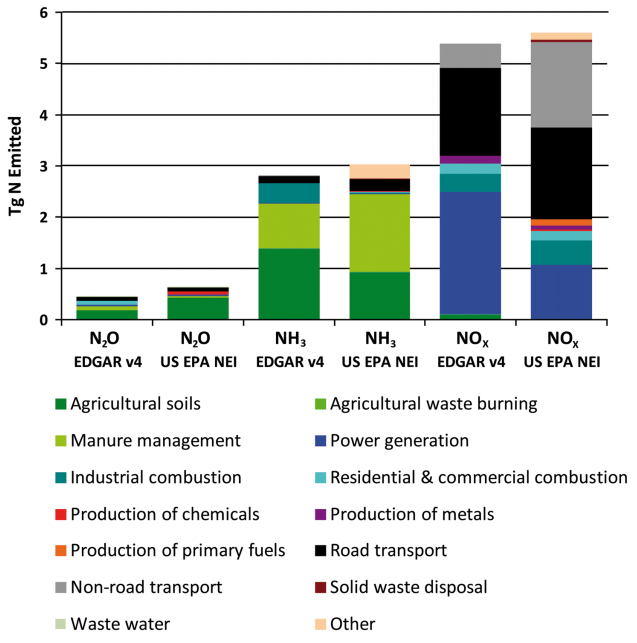

Fig. 5. Comparison of US EPA emissions of N_2O, NH_3, and NO_x with the EDGAR database for the year 2005 (USEPA, 2009a, b; EDGAR, 2009).

inspection and maintenance data and better representation of extended idling emissions.

Despite increased diesel and gasoline consumption, NO_x emissions from on-road sources are estimated to have decreased by on average 5% per year since 2002 due to the introduction of stricter emission standards. Parrish (2006) used ambient data to show that total on-road emissions may have increased from 1990–2000; however, in a multi-city, multi-year study, Bishop and Stedman (2008) have demonstrated that per-car emission factors have decreased from 2000 to 2006. The NEI reports little year-to-year change in off-road vehicle emissions. Many electricity and industrial power generation facilities have been subject to several recent regulated emission reductions including the NO_x Budget Trading Program. Because most of these facilities are equipped with Continuous Emission Monitors, the emission magnitude and trend is well quantified. Since 1999, this sector has reduced emissions by an average of 6.4% per year. The combined effect of the reductions in mobile and stationary source NO_x emissions has been observed in the surface concentration monitoring data (Godowitch et al., 2008) and from space-based remote sensing methods (Kim et al., 2006; Kim et al., 2009).

In Fig. 5, EDGAR v4 and US EPA NO_x emission data are aggregated by sector to make them directly comparable. Where stationary combustion sources are concerned, EDGAR has markedly larger emissions. The NEI power generation emission rates are likely more accurate since they are measured at the electricity generating units as part of the Continuous Emission Monitoring System. Road transport emissions are similar in both datasets, but interestingly non-road mobile sources (which include off-road vehicles, rail, air and shipping) are significantly larger in the US EPA dataset. The NEI partially includes international shipping emissions near the US coast, which have been omitted from the EDGAR inventory and have been estimated in EDGAR FT32 at about 0.43 Tg N for the US in the year 2000. Another possible explanation is a difference in activity rates and/or emission factors for domestic air transport. From 2000 to 2005, the trends in these two databases also differ. For on-road sources, EDGAR estimates 6% yr^{-1} reduction (NEI: 4% yr^{-1}), but for power generation and industrial sources, the EDGAR inventory has little trend (NEI: 6% yr^{-1} reduction).

3.3 Nitrous oxide

Total US N_2O emissions for 2005 are estimated to be 1.02 Tg N_2O (US EPA, 2009b). The largest source of US N_2O emissions is agricultural soils (67%), followed by fossil fuel combustion in vehicles (12%), industrial processes (8%), fossil fuel combustion for electricity generation (5%) and livestock manure management (4%). Enhancements of N_2O emissions from agricultural soils include practices such as fertilization, application of livestock manure, grazing animals on pasture or feedlots, and cultivation of N-fixing crops. The estimated trend in N_2O emissions for the USA is a gradual reduction of approximately 1% per year since 2000. This is due to a 5% reduction per year in the emissions from vehicles and industrial processes – the other sectors are estimated to have largely remained constant over this period. EDGAR v4 emissions are lower than those reported by EPA. This is almost entirely due to lower emissions from agricultural soils in the EDGAR v4 inventory.

The physical and chemical processes that drive emissions of NH_3, NO_x, and N_2O are often intertwined. This is also true for the human activities that cause these emissions, such as fuel combustion in motor vehicles and agricultural land under cultivation. However, the GHG emission inventory and the NEI do not always use the same models and data sources. For example, direct fertilized crop emissions of N_2O are estimated using the DAYCENT model (Del Grosso et al., 2001), while the temporal pattern of NH_3 emissions are estimated using an emission factor approach (Goebes et al., 2003), and NO emissions are calculated using the BEIS model (http://www.epa.gov/asmdnerl/biogen.html). Because the interactions between these processes are not explicitly considered, these shortcomings impede efforts to devise strategies that simultaneously mitigate climate change and improve air quality.

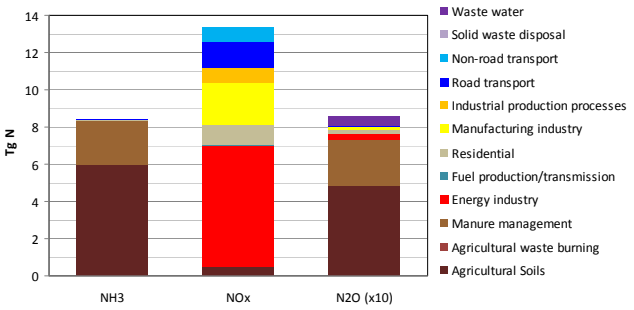

Fig. 6. Emissions of NH_3, NO_x and N_2O (expressed in Tg N) for the China in 2005 according to the EDGAR v4 inventory (EDGAR, 2009) for the main emitting source sectors. N_2O emissions are increased by a factor of 10 for ease of comparison.

4 Emission inventories of reactive nitrogen species in China

The analysis of the emission situation in China is comparatively more difficult than is the case for Europe or the United States. Reasons for this are, among others, the immense growth rates of the economy and thus fast developing emission sources, the lack of an official national inventory programme and no reporting of emissions to international organisations. Figure 6 displays the emission estimates for the year 2005, distinguishing the main source sectors. NH_3 and N_2O emissions are here dominated by agricultural sources (mainly agricultural soils and manure management), while for NO_x, the main difference to Europe and the US is the large share of stationary combustion relative to mobile sources. In the following sections, recent studies are discussed for each of the substances.

4.1 Ammonia

Only a few studies on ammonia emissions in China are available (e.g. Zhao et al., 1994; Yan et al., 2003). With regard to total NH_3 emissions in China, the major contribution comes from N-fertilizer application (52%) and livestock (41%) respectively in the 1990's (Klimont, 2001a). Other sources of ammonia emissions include biomass burning, natural ecosystems, crops and oceans, humans (breath, sweat, excretion) and fossil fuel combustion.

The basic methodology applied to derive these emissions relies on the approach used in Europe (Klaassen, 1994; Klimont, 2001b), and as far as available takes into account information about China-specific characteristics. Klimont (2001a) estimated the total ammonia emissions in China at 9.7 Tg NH_3 in 1990 (11.7 Tg NH_3 in 1995), which translates into 7.98 Tg N (1990) respectively 9.62 Tg N (1995). Emissions were as well spatially disaggregated on a $1^\circ \times 1^\circ$ grid for both years. In 1995 the highest ammonia emission density, exceeding 100 Gg NH_3 per grid, is observed in *Jiangsu* and *Henan* provinces. This corresponds well with the large population of pigs in these regions as well as high cattle density in *Henan* province. Using the IPCC approach, NH_3 emission from synthetic fertilizer and manure application in 1990 was estimated to be 1.65 Tg N by Li Yu'e et al. (2000). Yan et al. (2003) quantified the use of urea and ammonium bicarbonate and the cultivation of rice leading to a high average ammonia loss rate from chemical N fertilizer in East, Southeast and South Asia, and the total emission was estimated to be 5.8 Tg N for the area of China. These values compare reasonably well with the amount of 8.4 Tg N from NH_3 emissions for China in EDGAR v4, with 6.01 Tg N (72%) stemming from agricultural soils and 2.29 Tg N (27%) from manure management.

Due to anticipated increases in synthetic fertilizer application rates and per-capita meat and dairy consumption, future ammonia emissions are expected to continue to rise.

4.2 Nitrogen oxides

East Asia is a region of the world with large and rapidly increasing anthropogenic emissions, NO_x emissions have increased by 58% from 1975 (2.05 Tg N yr^{-1}) to 1987 (3.25 Tg N yr^{-1}) (Kato and Akimoto, 1992), and Van Aardenne et al. (1999) anticipated an almost fourfold increase in NO_x emissions for the period from 1999 to 2020. Especially in China, anthropogenic emissions associated with fossil fuel combustion have grown significantly due to a period of rapid economic development and industrial expansion in the last three decades (e.g., Streets and Waldhoff, 2000).

Using data from the China Statistical Yearbook (Press, 1996), Akimoto et al. (1994) and Kato and Akimoto (1992) estimated NO_x emissions in China for the year 1987. Bai (1996) considered that the inventory should be based on more detailed data than available from the Yearbook, and the emission factors should be modified to be consistent with actual emission factors applicable to the situation in China. Bai (1996) provided a NO_x emission inventory for the year 1992 using more detailed data from statistical yearbooks on a provincial level and emission factors measured in Chinese installations.

Bai (1996) created a spatially disaggregated inventory on $1^\circ \times 1^\circ$ with the highest grid value being >0.1 Tg N yr^{-1}, which occurred only in Shanghai. From the east to the west of China, the values decreased. The emissions in some provinces such as *Inner Mongolia*, *Qinghai* and *Tibet* are even <0.0001 Tg N yr^{-1}, which is consistent with the distribution of industrial installations and population between these regions.

The rapid growth of NO_x emissions in China (Bai, 1996; Ma and Zhou, 2000; Streets et al., 2000), with an increase from 9.5 Tg to 12.0 Tg (calculated as NO_2) between 1990 and 1995 is driven by a significant increase in emissions from the transport sector (increase of 62%). Emissions also increase significantly in the industrial, power generation and

domestic sectors, with increases of 26%, 20% and 21%, respectively. Within these sectors, emissions from industrial installations were the largest individual source group, contributing approx. 42% of total emissions (5.0 Tg NO_x as NO_2). From 1995 to 2000, some studies (e.g., Aardenne et al., 1999; Streets et al., 2000) estimated that NO_x emissions in China will continue to grow rapidly. However, other researches' results using more recent statistical data (e.g., Tian et al., 2001) indicate that NO_x emissions began to remain somewhat stable for a few years with total emissions in China amounting to 11.3 Tg NO_x (as NO_2) in 1995, 12.0 Tg (1996), 11.7 Tg (1997) and 11.2 Tg (1998). In the analysis by Tian et al., this is explained on the basis of changing energy management in China. However, for the same time period (1995-2000), Ohara (2007) and IIASA (2009) estimated a slow increase of emissions from 9.31 Tg NO_x to 11.19 Tg NO_x (Ohara, 2007), respectively 9.38 Tg NO_x to 11.73 Tg NO_x (IIASA, 2009), all calculated as NO_2 (see as well Fig. 9 for the development over the whole period).

According to Streets et al. (2003) Chinese emissions in the year 2000 have only slightly increased (11.3 Tg NO_x). For the year 2005, however, both EDGAR (2009) and IIASA (2009) estimate further increases to 18.35 Tg NO_x (as NO_2) and 17.09 Tg NO_x (as NO_2) respectively. Based on EDGAR (2009), power generation still contributes the lion's share of NO_x emissions in China (48%), while road transport is still comparatively low at 11%. In addition to that, satellite observations of the tropospheric NO_2 column density over Eastern China have increased considerably during 1996 to 2004 (Richter et al., 2005; van der A et al., 2006), suggesting large emission increases over that period

4.3 Nitrous Oxide

The UNDP/GEF ECPINC Project (*Enabling China to Prepare Its Initial National Communication,* http://www.ccchina.gov.cn/en/NewsInfo.asp?NewsId=5392) has been implemented to support China in fulfilling its commitments under the UNFCCC to communicate to the Conference of Parties to the Convention (1) a national inventory of emissions and sinks of greenhouse gases (2) a general description of steps taken or envisaged by China to implement the Convention and (3) any other information China considers relevant and suitable for inclusion in its Communication.

A fair amount of research has been conducted on an N_2O emissions inventory for China. Zheng et al. (2004) have shown that most (up to 75%) of cropland N_2O emissions are direct emissions: immediately from fertilized top-soil rather than denitrification of nitrate leached into sub-surface and groundwater. Zheng et al. (2004) collected 54 direct N_2O emission factors (EF'_ds) obtained from 12 sites of Chinese croplands and found that of these 60% are underestimated by 29% and 30% are overestimated by 50% due to observation shortages. The biases of EF_ds are corrected and their uncertainties are re-estimated. Of the 31 site-scale EF_ds, 42% are lower by 58% and 26% are higher by 143% than the IPCC default values. Periodically wetting/drying the fields or doubling nitrogen fertilizers may double or even triple an EF_d. The direct N_2O emissions from Chinese croplands are estimated at 275 Gg N_2O-N $yr^{|1}$ in the 1990s, of which 20% is due to vegetable cultivation. The great uncertainty of this estimate, −79% to 135%, is overwhelmingly due to the huge uncertainty in estimating EF_ds (−78±15% to 129±62%). The direct N_2O emission intensities significantly depend upon the economic situation of the region, implying a larger potential emission in the future.

However, agricultural activity is the main, but not the only source of N_2O emissions in China. According to results by Li Yu'e et al. (2000), total N_2O emissions from stationary fuel combustion amounted to 58.22 Gg N yr^{-1} in 1990 in China. Within the fuel combustion sector, energy industries, manufacturing industries and residential areas were the main sources of N_2O. Among fossil fuels, hard coal was the main contributor to N_2O emissions. The industrial process sector was a less critical source of N_2O emissions, with the total N_2O emissions from this source group amounting to 0.41–0.90 Gg N yr^{-1}. The total emission of N_2O as a result of fertilizer application was 342.5 Gg N yr^{-1} in 1990, being the most important contributor to N_2O emission from agricultural soils.

While both Li (2000) and Xing's (1998) estimations did not consider permanent croplands, Lu et al (2006) established an empirical model to develop a spatial inventory at the 10×10 km scale of direct N_2O emissions from agriculture in China, in which both emission factor and background emission for N_2O were adjusted for precipitation. As a result, the total annual fertilizer-induced N_2O emission was estimated to be 198.89 Gg N_2O-N in 1997 and background emissions of N_2O from agriculture was estimated to be 92.7 Gg N_2O-N and the annual N_2O emission totalled 291.67 Gg N_2O-N. All N_2O emission measurements are subject to significant uncertainty due to their great temporal and spatial variations of cropland fluxes.

For the years 2000 and 2005, EDGAR (2009) estimates 928 Gg N_2O (2000) and 1065 Gg N_2O (2000). This is consistently lower than figures by IIASA (2009), with 1,747 Gg N_2O (2000) and 1,854 Gg N_2O respectively. It has to be noted, that EDGAR 32 FT 2000 had estimated N_2O emissions for China at 1764 Gg N_2O, which is comparable with IIASA (2009) for that year, but almost twice the amount of what EDGAR (2009) states. One possible explanation for this is the inclusion of sources such as *run-off and leaching*, *forest* and *shrub fires* in EDGAR 32 FT 2000, which are not included in the figures presented here for EDGAR v4.

5 Evaluation of emission inventories for reactive nitrogen

5.1 Uncertainty assessment in general

For all emission inventories portrayed here, the assessment of uncertainties is a key aspect. For national inventories, issues of compliance or non-compliance with reduction targets and emission ceilings are relevant, while in general the quantification of uncertainties of emissions as input data for atmospheric dispersion models is of importance. Aspects of completeness regarding the total amount of emissions accounted for are as important as the spatial distribution, chemical composition and temporal patterns of emission occurrences.

Within the context of emission inventories compiled under the CLRTAP, country submissions are accompanied by so-called informative inventory reports (IIRs, http://www.emep.int/emis2007/reportinginstructions.html), covering aspects of completeness, an analysis of key sources and uncertainties. In addition to this, regular centralised reviews are conducted for selected countries, with the aim to improve the quality and accuracy of reported emission data. For the United States, NARSTO's third assessment report *Improving Emission Inventories for Effective Air Quality Management Across North America: A NARSTO Assessment* (NARSTO, 2005) took stock of the current state of emission inventories for Canada, the United States and Mexico and identified areas for improvement.

Under the UNFCCC, emission reporting is guided by a document called "Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories" (http://www.ipcc-nggip.iges.or.jp/public/gp/english/, see as well IPCC 1997, 2000) in order to *"...to assist countries in producing inventories that are neither over nor underestimates so far as can be judged, and in which uncertainties are reduced as far as practicable."*

These formalised assessments of inventories are relevant sources of information when trying to identify the overall accuracy of emission estimates, for instance, on a national scale or across countries for individual pollutants. In addition, studies such as conducted by Winiwarter and Rypdal (2001) and Rypdal and Winiwarter (2001) provide a methodological analysis of uncertainties for specific trace gases, respectively individual countries. An analysis conducted by Olivier et al. (1999) discusses in detail uncertainties of GHG emissions on a sector level, distinguishing between uncertainties in activity datasets, emission factors (as the main drivers of uncertainties in inventories) and the resulting total emissions.

Olivier et al. (1999) conclude that emissions from fossil fuel production and combustion are generally well understood and prone to small to medium uncertainties only. This can be said as well for industrial production processes or solvent use. In contrast, emissions related to agricultural landuse are viewed as significantly more uncertain. In a similar way, emission estimates of N_2O and CH_4 are generally less robust, because CO_2 is emitted from combustion processes mainly can be calculated more accurately than emissions based on soil microbial processes for instance. These findings have implications as well for the discussion of the different substances in the inventories analysed in this paper, where NO_x emissions can be seen comparatively robust. Yet, the uncertainty associated with NO_x emissions is approx. 4 times higher than for CO_2 emissions, because these are directly linear to fuel input in combustion sources and EFs are not influenced by processes and hence easier to determine. On the other hand, NH_3 and even more so, N_2O emissions are likely to be more uncertain because their main sources are related to agriculture and affected e.g. by soil biochemical processes and meteorological and climatological drivers.

For individual inventories, e.g. the UK NAEI, uncertainty assessments are provided by inventory compilers (http://www.naei.org.uk/emissions/emissions_2007.php?action=notes1). For NO_x, the NAEI assumes $\pm$7%, for NH_3 $\pm$20%, while for N_2O no individual assessment is made, but an overall uncertainty of $\pm$15% is estimated across the 6 greenhouse gases overall.

Following the differences in sectoral uncertainties provided by Olivier et al. (1999), the following sections will specifically investigate differences in sectoral emissions between inventories in order to identify, if current inventory differences are within range of the uncertainty estimates.

5.2 Inventory comparison – analysing differences and similarities

5.2.1 General observations for the situation in Europe

A full and detailed intercomparison of the inventories on the most detailed sectoral level for all countries/regions is beyond the scope of this paper. However, some similarities and differences are worth noting. In the case of ammonia, both the EMEP and the IIASA figures are quite similar, with some countries showing different emission levels, most likely due to recalculations and re-assessments of e.g. animal numbers that have been emerging in the course of bilateral consultations in the preparation of the IIASA dataset and which have not (yet) been incorporated in recalculations of the data reported to EMEP.

With regard to NO_x, the differences between EMEP and EDGAR figures are substantial for some countries and remarkably different in total (EDGAR v4 32% higher than EMEP). Some of the potential sources of these discrepancies have been highlighted in Sect. 3.3, in particular the different sectoral allocations including some emission sources that are not reported under EMEP in the EDGAR dataset.

In addition to that, a likely reason for these large differences are assumptions regarding underlying emission factors for some of the largest contributing sources, e.g. due to the estimated share of power plants equipped with efficient NO_x

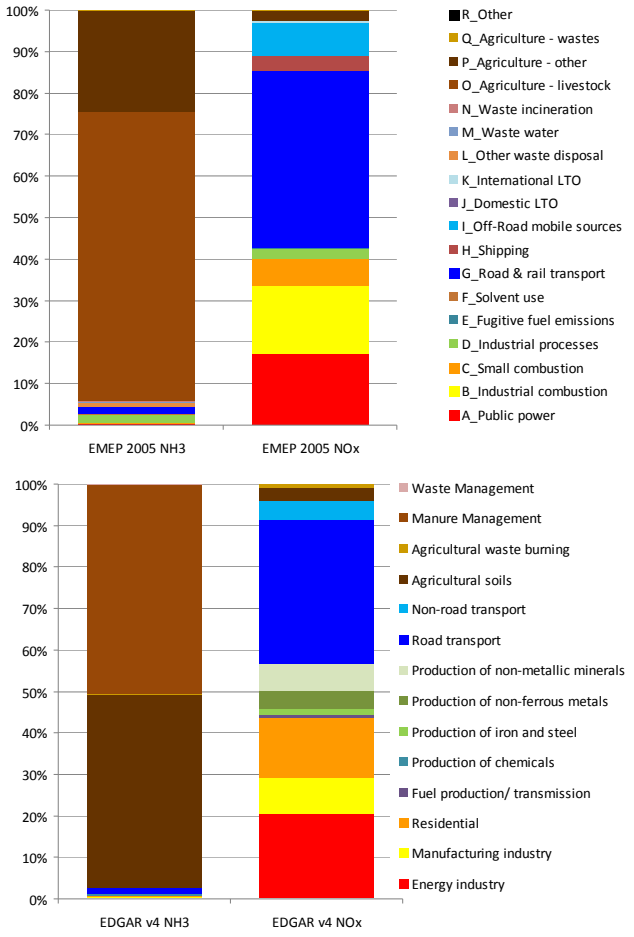

Fig. 7. Sectoral comparison of emissions for the year 2005 between EMEP (top; EMEP, 2009) and EDGAR v4 (bottom; EDGAR, 2009) for the EU27 countries (excluding Malta and Cyprus). It should be noted, that the EMEP sectoral split is based on only 10 countries reporting according to the latest sectoral structure (NFR 08, Level 1), which is best comparable with the EDGAR v4 sector structure.

control equipment, or emission factors and activity rates for road transport sources.

Figure 7 displays a direct comparison of NH_3 and NO_x emissions in the EMEP and the EDGAR v4 inventories by sector for the year 2005. The most recent change in EMEP reporting requirements, adopting a new reporting format termed NFR 08 (Nomenclature for Reporting 2008) which is – even on an aggregated level – better suited for sectoral analysis than its predecessors (e.g. NFR 01, NFR 02) which were based on the UNFCCC CRF format and in some areas lacking a sufficient level of detail. With regard to NH_3, agricultural emissions show a similar pattern in both inventories overall and are dominating emissions for this substance. The situation for NO_x emissions is more diverse and shows a distinct difference for stationary combustion sources, which contribute more than 55% to EDGAR v4 NO_x emissions, but only account for about 42% of EMEP NO_x emissions. A full comparison of individual sectors by country on a European scale would be worthwhile once all countries are reporting emissions based on the new NFR 08 sectoral structure. As this is not feasible at this stage and within the scope of this paper, an analysis is conducted for the United Kingdom in the following section.

5.2.2 Detailed analysis for the United Kingdom

For a thorough investigation of the differences between inventories, it is essential to have access to a very detailed sectoral split, and if feasible, even the emission factors and activity rates that have been used to compile the inventories. In this context, the structure initially applied in the EMEP inventories labelled SNAP (Selected Nomenclature for Air Pollution, EMEP) provided emission data with a detailed level 3 split allowed for a comparison down to process and fuel level. However, not many countries provided the obligatory information in this detailed split and hence inventories were subject to different levels of detail and substantial gaps. The new format for reporting under EMEP has been termed Nomenclature for Reporting (NFR, current version is NFR 08). While NFR has been closely aligned with the UNFCCC common reporting format (CRF) and thus been mainly driven by fossil fuel combustion sources, it provided less detail and no distinctions had been made for instance with regard to the type of fuel used in power plants or road transport modes. The recently adopted NFR 08 better accounts for non-combustion sources and already on the most aggregated level (NFR 08, Level 1) provides a useful split into the main sectors (for details on NFR 08, see http://www.unece.org/env/documents/2008/EB/EB/ece.eb.air.2008.4.e.pdf). The sectoral split used in EDGAR v4 reflects the bottom-up character of the inventory. It is not always directly comparable to NFR sectors, but in most cases, an equivalent source sector allocation – on a more aggregate level – can be found.

Table 4 provides a detailed comparison of source sectors split into categories for which a cross-comparison between the UK NAEI and EDGAR v4 could be sensibly conducted for NO_x, NH_3 and N_2O. As some of the individual source sectors in NAEI and EDGAR do not fully match, the following discussion will focus on the subtotals for sector groups rather than on individual figures.

The figures for stationary combustion are quite similar overall, but NO_x emissions in EDGAR v4 are 17% lower, N_2O from this source group is less than a third than those in the NAEI. For road transport, the picture is similar (17% lower NO_x emissions, 87% lower N_2O emissions), while NH_3 from road transport is 47% higher in EDGAR v4. NO_x emissions from other mobile sources and machinery are about 32% lower in EDGAR v4. These combustion sources, both stationary and mobile, are assumed to be best understood with low uncertainties typically attributed to NO_x emission factors in general. Thus, the magnitude of

differences in these sectors between NAEI and EDGAR v4, however, is larger than anticipated taking into account the uncertainty ranges assumed for the UK NAEI (see section 6.1).

On the other hand, total NH_3 emissions from the sector manure management are quite similar (~6% lower in EDGAR v4), yet some significant differences between the emissions from individual animal types can be observed. EDGAR v4 has substantially higher emissions of N_2O from manure management (about 6 times higher than NAEI), but less than one third of direct emissions from agricultural soils. This may hint at a difference in allocation between these subsectors. In addition to that, NH_3 emissions directly originating from agricultural soils are 5.3 times higher in EDGAR v4.

Finally, NAEI lists a few sectors that are not subject to reporting obligations under EMEP, while EDGAR v4 – as an inventory primarily compiled for modelling purposes – does not make this distinction.

In summary, the difference between country total emissions between both inventories is within the uncertainty margins expected for N_2O at 2.6% (±15% uncertainty range for NAEI, see section 6.1), however larger for NO_x emissions with EDGAR v4 being 20.9% below NAEI, (NAEI assuming a ±7% uncertainty range). NH_3 emissions in EDGAR v4 are 36.9% higher than in the NAEI (uncertainty range for NAEI assumed to be (±20%). However, for all three substances, sectoral allocation are quite different between inventories, which has direct implications for the use of the emissions for atmospheric modelling, e.g. the temporal profiles and spatial distribution of emissions in different sectors. The current structure of the inventories such as NAEI and hence EMEP – which are compiled primarily for reporting and compliance monitoring purposes – is not always detailed enough for modellers to fully assess the quality of the inventory and supporting datasets are often not accessible or documented well enough.

5.2.3 Observations for all regions

The overview comparisons of sectoral emission shares in the US and the China (see Figs. 5 and 6) indicate some substantial differences between the emission profiles of the US (and similarly Europe) on one side and China on the other. In particular the high contribution of emissions from agriculture to total NH_3 emissions in general is evident. In the case of NO_x emissions, mobile sources (road transport and off-road) contribute a larger share in Europe and the US, while power generation based on fossil fuels (coal mainly) in the energy industries of China and the US contribute in a similar magnitude. A remarkable difference can be seen in the contribution of road transport to N_2O emissions and – to a smaller extent – NH_3 emissions in the US compared to Europe. The reason for the difference in transport N_2O emissions can be explained by comparing EFs for running (hot)

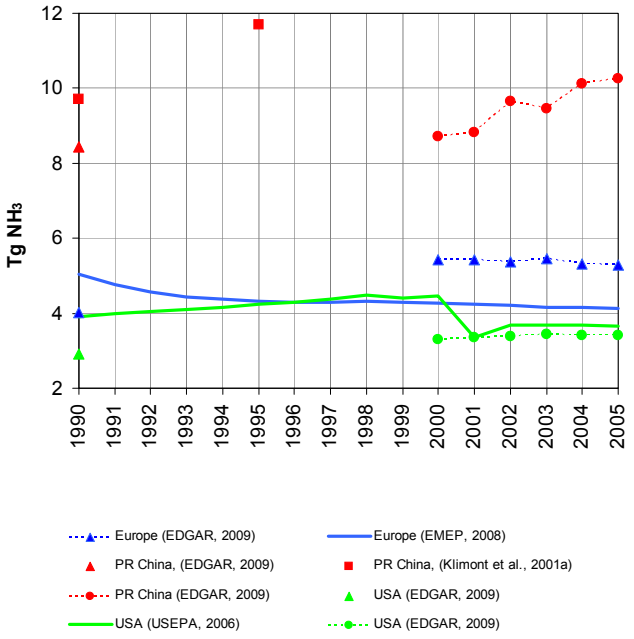

Fig. 8. Estimated trends in NH_3 emissions in Europe, the US and the China for the period 1990–2005.

emissions for passenger cards advanced three-way catalytic converters) for US and European vehicles in the IPCC Emission Factor Database (EFDB, http://www.ipcc-nggip.iges.or.jp/EFDB/main.php). While the EF for US vehicles is given as 9 mg/km, the EFs for European vehicles (based on the COPRT IV model, http://lat.eng.auth.gr/copert/) range from 0.7 mg/km (highway driving) to 2 mg/km (urban driving). It is not straightforward to assess, if all N_2O emissions from road transport in the US and in Europe have been calculated using these factors, but the magnitude of difference suggests substantially different assumptions as to technology and resulting emission factors in both regions.

A comparison between the US NEI and EDGAR v4 indicates overall quite similar total figures, except for N_2O (EDGAR v4 substantially higher than US NEI). However, the sectoral structure for NO_x and NH_3 shows remarkable difference, for instance regarding the split between manure management and agricultural soils (NH_3) and power generation (NO_x).

5.3 Emission trends and implications

Apart from the analysis of emission inventories for specific years, which give a snapshot for a certain point in time, looking at the temporal trends in emissions can give valuable insight in the development of both emissions as well as the methodologies applied for their calculation.

The trend for NH_3 emissions (Fig. 8) shows no significant reductions in the 15 year period indicated. A moderate downward trend can be observed in Europe (−18%), which

Table 4. Detailed comparison between source sectors in the UK National Atmospheric Emissions Inventory (NAEI, 2008) and EDGAR v4 (EDGAR, 2009) for the year 2005.

		UK NAEI			EDGAR v4		
NFR code	NFR Name	NO_x	NH_3	N_2O	NO_x	NH_3	N_2O
1A1a	Public Electricity and Heat Production	372.60	0.70	3.48	410.14	0.57	2.65
1A1b	Petroleum Refining	30.70		0.39	3.20		0.01
1A1c	Combustion in Manufacture of Solid Fuels and Other Energy Industries	58.80		1.28			
1A2a	Combustion in Iron and Steel Manuf. Industry	19.50		0.33	12.19		
1A2f	Combustion in Other Manufacturing Industry	232.80	0.40	4.00	165.73	0.17	0.36
	Subtotal stationary combustion	*714.40*	*1.10*	*9.48*	*591.26*	*0.75*	*3.02*
1A3aii(i)	Civil Aviation – Domestic Take-Off and Landing	1.80		0.02			
1A3aii(ii)	Civil Aviation – Domestic Cruise	7.10		0.05			
	Subtotal aviation	*8.80*		*0.07*			
1A3bi	Road Transport – Passenger Cars	215.10	9.20	14.13	457.04	13.98	2.14
1A3bii	Road Transport – Light Duty Vehicles	58.50	0.20	1.19			
1A3biii	Road Transport – Heavy Duty Vehicles	274.00	0.10	1.09			
1A3biv	Road Transport – Mopeds & Motorcycles	1.30		0.01			
	Subtotal road transport	*548.90*	*9.50*	*16.42*	*457.04*	*13.98*	*2.14*
1A3c	Railways – Mobile Sources	36.90		0.80			
1A3dii	National Navigation (including Inland Waterways and Maritime Activities)	92.10		0.10			
1A3eii	Other Off-Road Mobile Sources and Machinery	5.60		0.19	91.06	0.03	0.05
	Subtotal other mobile sources	*134.60*		*1.09*	*91.06*	*0.03*	*0.05*
1A4a	Commercial/Institutional Combustion Plants	20.90		0.08	110.14	0.77	0.51
1A4bi	Residential Combustion Plants	108.30	1.50	0.37			
	Subtotal residential/commercial combustion	*129.20*	*1.50*	*0.45*	*110.14*	*0.77*	*0.51*
1A4bii	Household and Gardening (Mobile Machinery)	0.90		0.02			
1A4ci	Stationary Combustion Plants (Agriculture/Forestry/Fishing)	0.70		0.01			
1A4cii	Agricultural/Forestry Off-Road Vehicles and Other	52.10		1.59			
1A5b	Other Mobile Sources (including Military)	22.20		0.08			
1B1b	Fugitive Emissions from Fuels – Solid Fuels/Transformation	0.20	0.10	0.00			
1B2ai	Fugitive Emissions from Fuels, Oil – Exploration, Production, Transport	0.70	0.10	0.00			
1B2c	Oil and Natural Gas/Venting and Flaring	2.20		0.14			
2A7	Other Production		0.50				
2B2	Nitric Acid Production	0.60		6.52			
2B3	Adipic Acid Production			2.50			
2B5	Other Chemical Industry Processes	0.40	4.00			0.67	66.14
2C	Metal Production	1.60		0.03			
2D1	Pulp and Paper Production						
2D2	Food and Drink Production		0.90				
3D	Other Solvent Use (including products containing HMs and POPs)		1.20				
	Subtotal miscellaneous sources	*157.50*	*6.80*	*12.59*		*0.67*	*66.14*
4B12	Solid Storage and Drylot			0.15			
4B13	Manure Management – Other		17.90	3.80			
4B1a	Manure Management – Dairy		86.00			52.20	5.72
4B1b	Manure Management – Non-Dairy		63.60			75.92	8.10
4B3	Manure Management – Sheep		12.10			23.49	7.33
4B6	Manure Management – Horses		4.40			0.49	0.08
4B8	Manure Management – Swine		26.70			17.85	0.53
4B9	Manure Management – Poultry		37.70			63.70	0.29
	Subtotal manure management		*248.40*	*3.80*		*233.65*	*22.04*
4D1	*Direct Emissions from Agricultural Soils*		35.60	81.31	29.34	190.81	26.07
4F	Field Burning of Agricultural Wastes		1.90		2.69	0.92	0.05
4G	Other Agricultural (including use of pesticides)			0.19			
5B	Forest and Grassland Conversion	0.20		0.01			
	Subtotal agricultural soils and wastes	*0.20*	*1.90*	*0.20*	*2.69*	*0.92*	*0.05*
6A	Solid Waste Disposal on Land		4.10				
6B	Waste Water Handling		5.50	3.92			5.03
6C	Waste Incineration	1.70		0.16			0.21
6D	Other Waste (incl. Composting and Biogas Prod.)	0.30				0.50	0.62
	Subtotal waste handling	*2.00*	*9.60*	*4.08*		*0.50*	*5.87*
7	Other		0.30				
z_5E	Other Sources and Sinks		8.30				
z_1A3ai(i)	International Aviation -Take-Off and Landing			0.08			
z_1A3ai(ii)	International Aviation – Cruise			1.04			
z_1A3di(i)	International Maritime and Inland Waterway Navigation			0.15			
	Total	1619.8	323.00	129.2	1281.5	442.1	125.9
					−20.9%	*+36.9%*	*−2.6%*

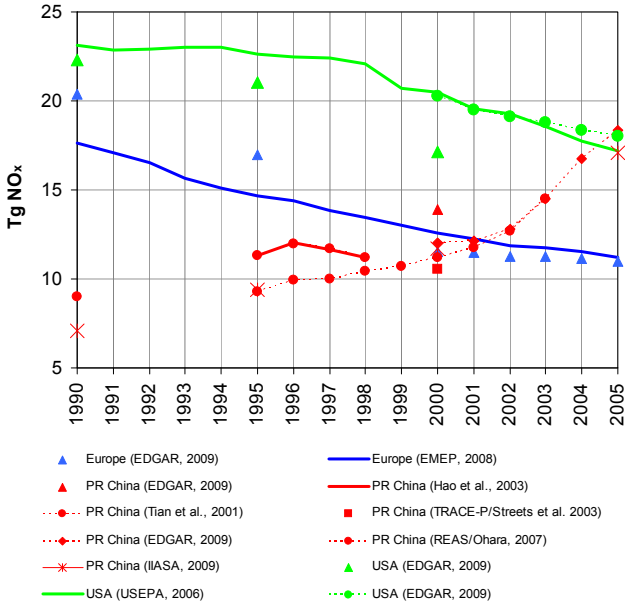

Fig. 9. Estimated trends in NO_x emissions in Europe, the US and the China for the period 1990–2005.

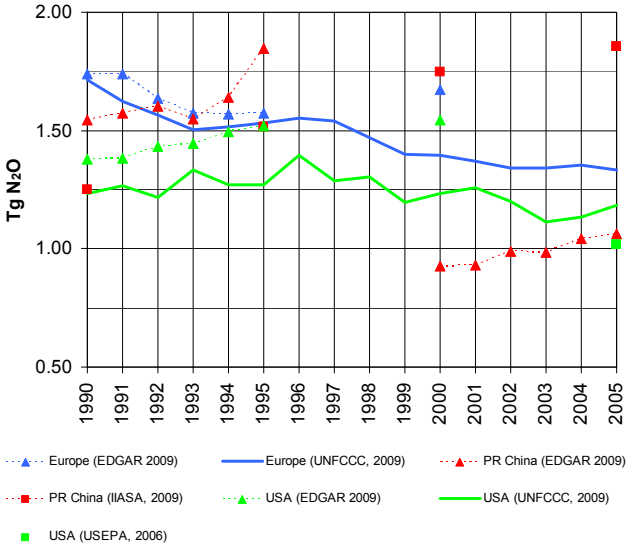

Fig. 10. Estimated trends in N_2O emissions in Europe, the US and the China for the period 1990-2005. The observed break in trend for EDGAR 2009 arises from differences between EDGAR v4 (2000 onwards) and EDGAR 32/EDGAR Hyde 1.3/1.4 for (1990-1995). While there is a consistent upward trend, some natural sources previously included in the analysis have not been included in EDGAR v4; For further detail, see (EDGAR, 2009) for all EDGAR inventory datasets.

is largely attributable to the political restructuring of Eastern Europe after 1989 (Horvath and Sutton, 1998) while US emissions have increased slightly in the period from 1990 to 2000. Although the values shown in Fig. 8 indicate a small (6%) reduction below 1990 for 2005, this is due to a change in the calculation methodology in 2000, which was has not been reapplied to the earlier years.

If the old methodology had been maintained throughout (with 2001 set equal to 2000), this would imply an overall national increase of about 22% between 1990 and 2005 comparing the US NEI values for both years. Chinese emissions of NH_3 have as of yet only been estimated by few authors, with 1990 emission estimates ranging between 8 and 10 Tg, with a figure of 11.7 Tg (Klimont, 2001b) for the year 1995. This latter figure would indicate an increase of 21% within these five years. This figure is attributed by Klimont (2001b) to a estimated 27% increase in NH_3 emissions from livestock and a 19% increase in fertilizer related emissions, reflecting increased consumption of livestock products in China, especially poultry (57% increase). Figure 9 displays trends in NO_x emissions for the same period based on data from EMEP, EDGAR and the USEPA NEI In addition to that, a number of figures from the literature for specific years or periods have been used as well as data from the GAINS model (IIASA, 2009) to discuss the trend in China. A general downward trend can be observed for Europe and the US, reflecting significant emission control activities in particular with regard to stationary and mobile combustion sources. This trend occurs both in the official inventory figures (EMEP, USEPA) and the EDGAR data, with similar slopes, but with different starting points.

In the case of the China, no official dataset for a longer term trend analysis is yet available. Using a synthesis of satellite observations and bottom-up emission inventory development, Zhang et al. (2007) for instance conclude that NO_x emissions in China from 1995–2004 likely increased by 70%. Those studies (Tian et al., 2001; Hao et al., 2003; Streets et al., 2003; Ohara et al., 2007) displayed in Fig. 9 show reasonable agreement for individual years, with a spread of approx. 3 Tg for the year 2000. With a starting point around 7-8 Tg NO_x in 1990 and estimated values around 17–18 Tg in 2005, a further substantial increase of China's NO_x emissions has to be anticipated, which may well off-set the reductions in Europe and the US.

Emission trends for N_2O emissions are subject to significant uncertainties in the same way as the annual emission estimates. UNFCCC reported data for Europe and the US, as well as emission figures from the EDGAR Hyde 1.4 (Van Aardenne et al., 2001) inventory of historic emission trends and EDGAR v4 (EDGAR, 2009) emissions for the year 2000, which also include China make a rough trend assessment possible (Fig. 10.). For Europe, a consistent downward trend for both EDGAR Hyde 1.4 and UNFCCC figures can be observed, but it has to be noted that EDGAR FT32 (Olivier et al., 2005; Van Aardenne et al., 2005) for the year 2000 are significantly above UNFCCC figures, and higher than 1995 emissions from EDGAR Hyde 1.4. (EDGAR, 2009).

For the US, EDGAR Hyde 1.4 indicates a slow increase for the period 1990 to 1995, with 2000 emissions remaining at approximately the same level, while UNFCCC figures remains around 1.25 Tg N_2O yr^{-1} until 2000, then shows a slight downward move with an increase again towards 2005. A significant increase of N_2O emissions from the China is portrayed in EDGAR Hyde 1.4, but EDGAR FT32 emissions for 2000 are lower than EDGAR Hyde for 1995, and not in the pathway that could be interpolated from the sharp increase of Chinese emissions in the period 1990 to 1995. The GAINS model (IIASA, 2009) contains five-yearly figures for N_2O, which support the general upward trend, however less steep than by EDGAR Hyde 1.4, with the GAINS value for 2005 being similar to the HYDE 1.5 for 1995 at about 1.8 Tg N_2O. Due to the small amount of studies available to elicit a trend and significant uncertainties associated to the quantification of N_2O emissions in general, some caution is needed in interpolating beyond 2005 based on the trends observed for the period 1990 to 2005.

6 Discussion and conclusions

6.1 Completeness and coverage of emission inventories

The picture presented in the previous sections is quite diverse across different dimensions. In many cases, emission inventories clearly reflect the purpose they have been designed for, i.e. serving for regulatory purposes (e.g. the UNFCCC and UNECE CLRTAP EMEP inventories), where compliance with international protocols drives the need for a pragmatic accounting system for emissions. On the other hand, inventories such as the EDGAR emission database are bottom-up, science driven compilations of emissions based on emission factors and – typically – publicly available, statistical information on activity rates, emission factors and suchlike. While the legal implications and validation of national submissions are key aspects for the former, the latter have the main objective to provide comprehensive and consistent datasets for (atmospheric) modelling exercises in a timely manner. This is somewhat reflected as well in the sectoral structure in which these inventories are compiled.

In terms of completeness, two aspects need to be considered: firstly, are all known sources of emissions reflected in the inventory, or can missing sources be identified based on the state-of-the-art of scientific research? And secondly, have all known emissions been calculated based on a consistent methodology and no figures are missing or not estimated?

Officially reported emission inventory data are often incomplete with regard to both aspects, respectively are only available with a certain time lag due to the time it takes to compile them based on official statistics and to undergo validation and error checking, before they are officially submitted. Their legal implications for monitoring compliance with international treaties or protocols, however, and the resulting reviews both within the country and by external review processes provides a crucial stage of quality control. Independent inventory compilers on the other hand do not always have access to the latest, most accurate and most detailed country specific EFs, activity rates, penetration of measures and technology etc. for each country and hence are prone to over/underestimate emissions from individual sectors. Yet, by providing consistent (as regarding methodology used), comprehensive (no gaps, spatial coverage and sectoral resolution) and complete (anthropogenic and natural sources, species) datasets for recent years, they provide vital input for modelling studies. There is, undoubtedly, a clear need for validation for both and here inventory compilers and atmospheric modellers need to work in close collaboration to mutually improve model results.

For all inventories, there have clearly been sources missing that have been identified and quantified in recent years, for instance emissions from NO from agricultural and forest soils and other natural and biogenic emissions (NH_3 from sea birds, NO_x from lightning and forest fires etc.). As most of the inventories compiled for regulatory purposes focus on anthropogenic (i.e. regulated and controllable) sources, they often do not calculate and report natural and biogenic emissions.

Compared to the main contributing sectors of anthropogenic emissions, the contributions of these missing sources to date may be small, but with decreasing emissions due to emission control activities, their relative importance will continue to grow in the future. However, there may yet be major missing sources in the inventories. For example low temperature coal and other biomass combustion (Sutton et al., 2008) may be a major additional source of ammonia emissions in China that has yet to be quantified. Although of historical importance (Fowler et al., 2004), coal burning in residential combustion sources is no longer a significant activity for much of Europe and North America. Similarly, there remain major uncertainties in regional nitrous oxide emissions, as indicated by the assessment of Crutzen et al. (2008) who estimated from the global rate of N_2O increase that total agricultural emissions should be 4–5% of the input reactive nitrogen, which is larger than the base emission rate of 1% used in the IPCC methodology. This differential is most likely due to other biogenic sources of N_2O, but it cannot be ruled out that there are missing industrial or combustion source emissions.

In comparing the regulatory and science based inventories, it is possible to identify systematic errors or gaps in both, provided there is sufficient level of detail available with regard to the sectoral disaggregation, emission factors, control technologies and activity rates, allowing for an in-depth assessment of the methodology used and factors and values applied. Documents such as IPCC (2000) providing *Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories* can be essential to systematically check for completeness of inventories, missing sources

and general methodology to assess and quantify uncertainties.

The coverage of regions and substances for instance in the EDGAR inventory is comprehensive, but the fact that the NH_3 inventory for the time after 1990 has not been compiled for a long time hints at the substantial resources and time required to compile these datasets. This is particularly problematic in the case of ammonia, as this is going to be a key pollutant to be targeted in future reduction efforts at least in Europe due to its growing relative importance for the control of secondary aerosol concentrations and its contribution to acidification and eutrophication.

Finally, the spatial and temporal coverage of emission data as input to atmospheric dispersion models is a key issue. On a global scale, a resolution of $1^\circ \times 1^\circ$ or $0.5^\circ \times 0.5^\circ$ is sufficient to capture the general trends and concentrations/depositions. Yet, for regional and local modelling, inventories with a resolution of more than $10\,km \times 10\,km$ are likely to miss vital distribution patterns and thus lead to mismatches between model results and observations. The EMEP modelling team has recently begun testing $25\,km \times 25\,km$ and $10\,km \times 10\,km$ resolutions for the EMEP domain, which will – if established – have implications for the required input data resolution of the EMEP inventory in the long run (see http://www.emep.int/publ/reports/2008/status_report_1_2008.pdf). And while national emission inventories with a spatial resolution of e.g. $1\,km \times 1\,km$ are available for some countries, they will rarely have been compiled using the same datasets and assumptions as the global inventories, hence may create boundary problems when nesting different inventories for modelling purposes (see as well Reis et al., 2008). The differences in the sectoral emissions splits between the inventories described previously have implications as well for the temporal and spatial profiles of these emissions when used as input for atmospheric dispersion models. Ultimately, this may lead to quite different ambient modelled concentrations of primary and secondary pollutants depending on which inventory data are used, in particular for pollutants such as tropospheric ozone, where the time and the location of the precursor emissions matters.

6.2 Trend analysis

The trends for the three gases investigated in this paper show quite different patterns, as well as differences between the countries and regions included in this paper. NO_x emissions in Europe and the US have fallen markedly, reflecting successful emission control policies especially since the early 1990s (see e.g. Vestreng et al., 2008). NO_x emissions from the China, while much lower than those of Europe and the US, do not show a consistent trend due to the lack of data available. However, with the substantial growth of the Chinese economy and hence energy demand, which is mainly supplied by coal fired power plants, a continuing increase of NO_x emissions can be anticipated.

Ammonia emission trends show a quite different picture, with only minor reductions in Europe over the period of 1990 to 2005, which were mainly a side effect of political restructuring around 1990, while the US ammonia emissions have increased by around 20% over the period. In both areas there have been major within-region differences. For example, in Europe, significant ammonia emission reductions were estimated or Denmark and the Netherlands, where control policies have been in place for more than a decade, while conversely emissions in Spain are estimated to have increased substantially over the last 20 years. In Europe there has been substantial debate as to whether estimated emission trends are verified by measurements (e.g. Horvath and Sutton, 1998; Sutton et al., 2003), and a recent assessment by Bleeker et al. (2009) concluded that the trend in reductions in ammonia emission in the Netherlands is supported by the measurements, though debate remains concerning the absolute magnitude of emissions. For the US, the overall increase in ammonia emissions is matched by major regional increases, especially in the Midwest (Sutton et al., 2008, based on Lehman et al., 2007) and in the Eastern US (Bleeker et al., 2009). With only a few data points available for China, it is yet evident that emissions are estimated to be about two to three times higher than European or US emissions in this period. In addition to that, Klimont (2001) projected a steep increase in NH_3 emissions for the period 1990–1995, equally matched between increased livestock numbers and mineral fertilizer use. Future projections for China suggest further increase, with Klimont (2001) estimating emissions in the range 8.2 Tg NH_3 to 19.9 Tg NH_3 for 2030.

Trends for nitrous oxide emissions finally indicate a slow decrease of emissions in Europe, a slight increase in US N_2O emissions, and a substantial growth in emissions in China. However, the trends depicted by data from the EDGAR Hyde 1.4 project and those reported to UNFCCC do not match well with 2000 figures from EDGAR FT32, indicating different methodologies, respectively potentially different source sectors being included/excluded.

Some of the differences hint as well at the varying state of knowledge and certainty with regard to both the current amount and the future direction of emissions and trends. For European and US emissions, NO_x emission values can be seen as quite robust and well understood, while in particular the trend of NO_x emissions from China is highly uncertain due to the unknown developments in the Chinese energy demand and potential efficiency gains and decoupling efforts between economic growth and energy demand. This is to some extent also the case for European NO_x emissions. The phasing out of nuclear power on the one hand, and the plans to increase the use of domestic coal and biomass for power generation on the other hand make it more difficult to predict the development of NO_x emissions from the power generation sector beyond 2010 (EMEP, 2009).

Ammonia emissions are dominated by agricultural production, and here future trends will be heavily influenced by

agricultural policies, such as for instance the Common Agricultural Policy (CAP) Reform in Europe. Livestock numbers and the implementation of measures under the Integrated Pollution Prevention and Control (IPPC) directives will introduce further stringent measures to reduce NH_3 emissions from animal housing. More evidence is yet required to assess if China's ammonia emissions are indeed growing at a rate as the few data points suggest, for example illustrated in the wide range between the 2030 projections of Klimont (2001), the high estimate of which reflected a simple projection of activity statistics, while the low projection assumed full implementation of Best Available Techniques.

For N_2O, both the general uncertainty about the emissions, and their trends, are significantly larger than for the other pollutants. How trends after 2010 evolve will depend on future negotiations and the way how Kyoto and post-Kyoto greenhouse gas emission targets are achieved and how further reductions will be set.

6.3 Future research needs

A first step to reduce the uncertainties identified above and to support the efforts to close the gaps in N budgets on a global or regional scale will be to detect missing sources and to quantify their contribution to overall emissions. This is of particular importance in the field of natural and semi-natural sources of emissions, which are often not yet well covered by emission inventories, which focus on anthropogenic emissions due to their history and heritage. Current research targeting unknown emission sources on the one hand, and recent achievements in quantifying emissions e.g. of NO from agricultural and forest soils (Skiba et al., 2001: Friedrich, 2009), NH_3 from seabirds and other animals (Blackall et al., 2007) may become vital in the future with anthropogenic emissions declining, yet are often not suited for reporting in (static) inventories due to their dependence on meteorological factors. In this context, the inventory compilers and modelling communities need to work in close collaboration to avoid some of these sources being missed or inconsistently treated. At the same time further efforts are needed to better quantify important but highly uncertain emissions such as ammonia from combustion sources (Fowler et al., 2004; Sutton et al., 2008) and apparently missing sources of nitrous oxide (Crutzen et al., 2008). Only if inventories reflect the current state-of-the-art of research can validation and verification experiments, comparing model calculations with observations on different spatial scales (e.g. Bleeker et al., 2009; Bergamaschi et al. 2005; Henze et al., 2008) deliver meaningful results towards a better understanding of the processes through which reactive and non-reactive nitrogen species contribute to air pollution and climate change. In addition, a fully quantified nitrogen cycle is a key requirement for the design of intelligent and effective management options towards reducing the negative impacts of N along the full nitrogen cascade. Complete and accurate emission inventories of all nitrogen species are a crucial building block for this.

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