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**Reactive nitrogen in  
atmospheric  
emission inventories**

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# Reactive nitrogen in atmospheric emission inventories – a review

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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 The People's Republic of China. The focus is on anthropogenic sources, assessing the state-of-the-art of quantifying emissions of Ammonia (NH<sub>3</sub>), Nitrogen Oxides (NO<sub>x</sub>) and Nitrous Oxide (N<sub>2</sub>O), 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 robustness 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<sub>x</sub> emissions and only slight reductions for NH<sub>3</sub> 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 basic 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 basic 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).

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 consequences of both these changes include impacts of air pollution human health, acidification and eutrophication of soils, alteration of biodiversity

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in terrestrial ecosystems, nitrate eutrophication in freshwater and marine ecosystems and alteration of the global greenhouse balance.

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.nitroeuropa.eu>) is working toward 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. Emission inventories are a critical source of information in this process. However, inventories are most often compiled for compliance monitoring purposes (e.g. EMEP under the UNECE Convention on Long-Range Transboundary Air Pollution or IPCC/UNFCCC), using officially submitted data. On the other hand, inventories compiled by researchers with the main aim to provide a consistent and comprehensive set of input data for modelling purposes (e.g. the EDGAR dataset) often use a bottom-up approach with information on emission factors and activity data independent of official (often not readily accessible) data.

In this paper, we focus on regional scale nitrogen emission inventories, 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 of our conclusions for the use of emission datasets for modelling purposes.

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## 1.2 Scope

Anthropogenic activities have a significant impact on the magnitude of N cycled and released into the atmosphere, for instance, it has been estimated that about 26 Tg N were attributed to fossil fuel combustion in 1990 (Galloway, 1998). For the year 2050, Galloway et al. (2004) calculated a global rate of annual creation of reactive nitrogen ( $N_r$ ) of 221 Tg N yr<sup>-1</sup>, compared to around 163 Tg N yr<sup>-1</sup> in the early 1990s and 125 Tg N yr<sup>-1</sup> around 1860.

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<sub>2</sub>O as a potent driver of global warming has been acknowledged and emission control strategies no longer focus solely on the CO<sub>2</sub>, which is reflected e.g. by a more detailed integration of N<sub>2</sub>O sources and measures in GAINS or MITERRA (Velthof et al., 2009).

Given the importance of nitrogen-containing species for air quality and climate change, the question emerges if the current inventories of NH<sub>3</sub>, NO<sub>x</sub> and N<sub>2</sub>O 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) has led to different accounting systems with often differing national budgets for the same trace gas. We therefore critically investigate the way nitrogen in its different forms is accounted for in these processes. 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

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this purpose, we here assess existing inventories with regard to their sectoral structure, temporal and spatial resolution, speciation and accessibility.

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<sub>3</sub>, NO<sub>x</sub> and N<sub>2</sub>O 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.

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 16 October 2004. With the aim to optimise nitrogen management in food and energy production on a local, regional and global scale, the declaration calls upon the national governments for instance to

- support the further assessment of the nitrogen cycle,
- focus efforts on increasing the efficiency and effectiveness of agricultural production and energy use, while decreasing the adverse effects of reactive nitrogen, and
- take action to enhance the availability of reactive nitrogen as food, fibre and other basic needs in regions of nitrogen deficiency and avoid nitrogen pollution.

For this last step, three key approaches are identified:

1. A code of good agricultural, forestry and aquacultural practise.
2. Strategies for sustainable energy use to prevent the formation of nitrogen oxides in fossil fuel combustion.

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### 3. Application of emission control technologies.

With regard to the requirements for an integrated nitrogen management and the need to fulfil different obligations under e.g. the UNECE CLRTAP or the UNFCCC, we investigate here if the current methods to account for the emissions of different nitrogen species in emission inventories provide a sufficiently robust data basis for such in depth integrated assessments.

## 2 European emission inventories of reactive nitrogen species

### 2.1 Overview

In Europe, emissions of ammonia and nitrogen oxides are covered by several regulatory regimes, both under the UNECE CLRTAP and directives of the European Commission. Member states of the European Union and signatories 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.emep-emissions.at/emission-data-webdab/>)

Nitrous oxide on the other hand is not covered by the CLRTAP, but is required to be reported under the UNFCCC by ANNEX I countries. The UNFCCC GHG inventory submissions can be accessed [http://unfccc.int/ghg\\_emissions\\_data/items/3800.php](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  $1 \times 1$  degree grid for 1990 and 1995 for direct greenhouse gases  $\text{CO}_2$ ,  $\text{CH}_4$ ,  $\text{N}_2\text{O}$  and HFCs, PFCs and  $\text{SF}_6$  and the precursor gases  $\text{CO}$ ,  $\text{NO}_x$ , NMVOC and  $\text{SO}_2$ . Similar inventories have been compiled for acidifying gases,  $\text{NH}_3$ ,  $\text{NO}_x$  and  $\text{SO}_2$  and Ozone Depleting Gases (EDGAR, 2008). For this paper, the new EDGAR dataset version 4 (v4 in the following text) is used, which has recently been officially released

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(<http://edgar.jrc.ec.europa.eu/>). EDGAR v4 (EDGAR, 2008) provides emissions for all relevant air pollutants and GHGs with an improved spatial resolution and until 2005.

## 2.2 Ammonia

In the case of NH<sub>3</sub>, 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, 2008). 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 that noted above for the EU27 as a whole, which clearly indicates that a more comprehensive reporting of NH<sub>3</sub> emissions is needed.

Figure 1 illustrates that differences between the EMEP dataset and data used for model calculations with the RAINS model by IIASA are marginal. This was anticipated, as the EMEP emissions displayed are based on expert estimates (gap-filled to account for missing data submissions) to complete official submissions by countries, which are often incomplete or comprise errors. In a similar way, the IIASA dataset has been compiled centrally by emission experts and subsequently validated and revised in extensive bilateral consultations with country experts. For some countries, however, the IIASA data are slightly lower, with overall emissions for the EU27 being 6% 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 and more accurate 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

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EDGAR v4 dataset (EDGAR, 2008) shows that EDGAR emissions are (much) higher for all EU27 countries, with only values for Croatia, Macedonia and Switzerland being below the EMEP/IIASA figures. 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 cannot be easily conducted, as the available EMEP emission dataset does not distinguish between the individual emission sources (in particular within the agricultural sector) in the same level of detail EDGAR v4.

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 4.8 Tg of  $\text{NH}_3$  for the year 2000. This is comparable with an estimate of 4.1 Tg for the whole of Europe made by Bouwman et al. (1997) for the year 1990 and an estimate of 4.02 Tg (Western and Eastern Europe) by EDGAR (2008). Galloway et al. (2004) only give a combined figure for Europe and the former Soviet Union (FSU), with atmospheric emissions of ammonia calculated at  $8 \text{ Tg N yr}^{-1}$ , with FSU emissions in 1990 at 3.4 Tg according to the EDGAR dataset.

### 2.3 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). While in general it should be anticipated that  $\text{NO}_x$  emission figures are less uncertain than those of  $\text{NH}_3$  or  $\text{N}_2\text{O}$ , the initial comparison between EMEP and EDGAR datasets for 2000 results in a significant difference both in total emissions for the EU27 and for individual countries (see Fig. 2). Total  $\text{NO}_x$  emissions (expressed as  $\text{NO} + \text{NO}_2$ ) for Europe are estimated at 13 Tg by EDGAR and 13.9 Tg by EMEP (EDGAR, 2008; EMEP, 2008).

This comparison indicates that while  $\text{NO}_x$  emissions are undoubtedly better understood than for  $\text{NH}_3$ , in general, inventory figures still seem to be prone to large variations both between inventories and within inventories over time. 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<sub>x</sub> inventory datasets in the future.

## 5 2.4 Nitrous oxide

Since N<sub>2</sub>O emissions are not reported under the CLRTAP, but subject to reporting obligations to the UNFCCC for ANNEX 1 countries, the comparison is made between UNFCCC data and the EDGAR inventory. For most countries, figures in both inventories are quite comparable, with EDGAR showing an overall higher emission for the EU27 of about 12%.

While EDGAR shows substantially higher emissions from some large European countries in particular (France 13.5%, Germany 36.3%, Spain 28.1%, United Kingdom 46.2%), other figures are substantially lower than UNFCCC data, for instance Finland (-35.2%), Italy (-25.8%) or Sweden (-26.9%). One remarkable difference has to be noted for Turkey, where EDGAR emissions are more than 3 times higher than UNFCCC figures. At this stage, it is difficult to assess fully 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<sub>2</sub>O emissions from soils, one of the main sources of N<sub>2</sub>O, which results in higher emissions than the current UNFCCC established methodology. Within the EU27, 53.8% of N<sub>2</sub>O emissions in the EDGAR inventory stem from agricultural sources (37.9% from agricultural soils, 15.8% from manure management and 0.1% from agricultural waste burning). Shares of emissions from agricultural soils in individual countries vary significantly, between 10.3% (Luxembourg) and 65.5% (Denmark). Figure 4 displays the variation of relative contributions of agricultural soils and manure management to N<sub>2</sub>O emissions in the EU27 countries. A third contributing source to N<sub>2</sub>O emissions in the EU27 is the chemical industry sector, contributing about 34.6% of total N<sub>2</sub>O emissions in 2000. It is likely that emissions from the chemical industry are well understood and hence emission factors and activity rates can be assumed to be

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less uncertain than those from agricultural soils or manure management. Hence, the differences between UNFCCC and EDGAR data will mainly arise from the application of different emission factors (or activity rates) in these two sectors.

However, even in those cases where 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<sub>2</sub>O 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, <http://www.epa.gov/ttn/chief/eiinformation.html>) in support of the Clean Air Act and subsequent amendments. The NEI includes an accounting of pollutants that impact air quality, including NO<sub>x</sub> and NH<sub>3</sub>. These efforts have recently been reviewed in an assessment report by NARSTO (2005). In addition, the EPA prepares an estimate of N<sub>2</sub>O emissions in the US. Inventory of Greenhouse Gas Emissions and Sinks (USEPA, 2008), 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 slip from mobile and stationary combustion sources equipped with catalytic converters, which comprises approximately 10% of the inventory. Because of the operational challenges in measuring ammonia emissions and a lack of detailed animal husbandry practices data, ammonia emission estimates have high uncertainty.

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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<sub>x</sub> emissions include on-road vehicles (36%), electricity and industrial power generation (35%), and off-road vehicles (22%), such as ships, aircraft, and construction equipment. Emissions from on-road and off-road vehicles are calculated using the National Mobile Inventory Model (<http://www.epa.gov/otaq/nmim.htm>). Despite increased diesel and gasoline consumption, NO<sub>x</sub> 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<sub>x</sub> 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<sub>x</sub> 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).

In Fig. 5, EDGAR v4 and USEPA NO<sub>x</sub> emission data are aggregated by sector to make them directly comparable. Where stationary combustion sources are concerned,

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EDGAR has markedly larger emissions, while industrial emissions are substantially larger in the USEPA figures. This is most likely due to a difference in labelling or allocating emissions from industrial combustion sources to either power generation or industry. Road transport emissions are similar in both datasets, but interestingly other mobile sources (which include off-road vehicles, rail, air and shipping) are significantly larger in the USEPA dataset. This could be caused by including all/part of international shipping emissions, which have been omitted from the EDGAR inventory and are estimated in EDGAR v4 at about 1.930 Gg 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<sup>-1</sup> reduction (NEI: 4% yr<sup>-1</sup>), but for power generation and industrial sources, the EDGAR inventory has little trend (NEI: 6% yr<sup>-1</sup> reduction).

### 3.3 Nitrous oxide

Total US N<sub>2</sub>O emissions for 2006 are estimated to be 1.2 Tg N<sub>2</sub>O (USEPA, 2008). The largest source of US N<sub>2</sub>O emissions is agricultural soils (72%), followed by fossil fuel combustion in vehicles (9%), industrial processes (7%), fossil fuel combustion for electricity generation (4%) and livestock manure management (4%). Enhancements of N<sub>2</sub>O 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<sub>2</sub>O 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.

The physical and chemical processes that drive emissions of NH<sub>3</sub>, NO<sub>x</sub>, and N<sub>2</sub>O 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 models and data sources used in the US to quantify emissions of the air quality pollutants are not the same as those used to quantify the N<sub>2</sub>O emissions. These

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shortcomings impede efforts to devise strategies that both mitigate climate change and improve air quality.

## 4 Emission inventories of reactive nitrogen species in the People's Republic of China

### 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  $\text{NH}_3$  emissions in China, the major contribution comes from N-fertilizer application and livestock, representing in the 1990's about 52% and 41% of total emissions, respectively (Klimont, 2001). 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, 2007), and as far as available takes into account information about China-specific characteristics. Klimont (2001) estimated the total ammonia emissions in China at 9.7 and 11.7 Tg in 1990 and 1995. Spatial distribution of emissions was also presented at a  $1 \times 1^\circ$  grid resolution. In 1995 the highest ammonia emission density which exceeds 100 Gg  $\text{NH}_3$  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,  $\text{NH}_3$  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 TgN for the area of China.

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.

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## 4.2 Nitrogen oxides

East Asia is a region of the world with large and rapidly increasing anthropogenic emissions, NO<sub>x</sub> emissions have increased by 58% from 1975 (2.05 Tg N yr<sup>-1</sup>) to 1987 (3.25 Tg N yr<sup>-1</sup>) (Kato and Akimoto, 1992), and Van Aardenne et al. (1999) anticipates an almost fourfold increase in NO<sub>x</sub> 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). However, some recent findings do not agree with the projections of a continuous increase of emissions.

Using data from the China Statistical Yearbook (Press, 1996), Akimoto et al. (1994) and Kato and Akimoto (1992) estimated NO<sub>x</sub> 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<sub>x</sub> emission inventory for the year 1992 using more detailed data from statistical yearbooks on a provincial level and emission factors measured in Chinese installations.

In Bai's 1° × 1° inventory, the highest grid value is >0.1 Tg N yr<sup>-1</sup>, 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<sup>-1</sup>, which is consistent with the distribution of industrial installations and population in these regions.

The rapid growth of NO<sub>x</sub> emissions in China (Bai, 1996; Ma and Zhou, 2000; Streets et al., 2000), with an increase from 9.5 to 12.0 Tg (calculated as NO<sub>2</sub>) 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

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group, contributing approx. 42% of total emissions (5.0 Tg). From 1995 to today, some studies (e.g., Aardenne et al., 1999; Streets et al., 2000) estimated that  $\text{NO}_x$  emissions in China will continue to grow rapidly. However, other researches' results using later latest available statistical data (e.g., Tian et al., 2001) indicate that  $\text{NO}_x$  emissions began to decrease somewhat, with total emissions in China amounting to 11.3 Tg  $\text{NO}_x$  as  $\text{NO}_2$ , 12.0 Tg, 11.7 Tg and 11.2 Tg, respectively, from 1995 to 1998. In the analysis by Tian et al. (2001), this is explained on the basis of changing energy management in China. According to Streets et al. (2003) Chinese emissions in the year 2000 has slightly increased again (11.3 Tg). There is a clear indication that the total  $\text{NO}_x$  emissions in China are not growing as rapidly as some research had previously projected (e.g., Aardenne et al., 1999).

### 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  $\text{N}_2\text{O}$  emissions inventory for China. Zheng et al. (2004) have shown that up to most (up to 75%) of cropland  $\text{N}_2\text{O}$  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  $\text{N}_2\text{O}$  emission factors ( $EF'_d$ s) 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  $EF'_d$ s are corrected and their uncertainties are re-estimated. Of the 31 site-scale  $EF'_d$ s, 42% are lower by 58% and 26% are higher by 143% than the IPCC default values. Periodically wet-

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ting/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  $275 \text{ Gg } N_2O\text{-N yr}^{-1}$  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  $EF_d$ s ( $-78\pm 15\%$  to  $129\pm 62\%$ ). 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 not the only source of  $N_2O$  emissions in China. As the statistical results by Li Yu'e et al. (2000), total  $N_2O$  emissions from fuel combustion amounted to  $58.22 \text{ Gg } N \text{ yr}^{-1}$  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  $0.41\text{--}0.90 \text{ Gg } N \text{ yr}^{-1}$ . The total emission of  $N_2O$  as a result of fertilizer application was  $342.5 \text{ Gg } N \text{ yr}^{-1}$  in 1990, being the most important contributor to  $N_2O$  emission from agricultural soils.

While both Li and Xing's estimations did not consider permanent croplands, Lu et al. (2006) established an empirical model to develop a spatial inventory at the  $10\times 10 \text{ km}$  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  $198.89 \text{ Gg } N_2O\text{-N}$  in 1997 and background emissions of  $N_2O$  from agriculture was estimated to be  $92.78 \text{ Gg } N_2O\text{-N}$  and the annual  $N_2O$  emission totalled  $291.67 \text{ Gg } N_2O\text{-N}$ .

All  $N_2O$  emission measurements are subject to significant uncertainty due to their great temporal and spatial variations of cropland fluxes.

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## 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 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 (<http://www.emep.int/emis2007/reportinginstructions.html>), covering aspects of completeness, an analysis of key sources and uncertainties. In addition to this, regular in-country reviews are conducted for selected countries, with the aim to improve the quality and accuracy of reported emission data.

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.”

In addition to these institutionalised approaches to uncertainty assessment, 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. Olivier et al. (1999) have discussed uncertainties of GHG emissions on a sectoral level, distinguishing between uncertainties in activity datasets, emission factors and the resulting total emissions (see Table 5).

Olivier et al. (1999) conclude that emissions from fossil fuel production and combus-

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tion 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 land-use are viewed as significantly more uncertain. In a similar way, emission estimates of  $\text{N}_2\text{O}$  and  $\text{CH}_4$  are generally less robust, because  $\text{CO}_2$  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  $\text{NO}_x$  emissions can be seen as similarly robust as  $\text{CO}_2$ , stemming primarily from combustion sources. On the other hand,  $\text{NH}_3$  and even more so,  $\text{N}_2\text{O}$  emissions are likely to be more uncertain because their main sources are related to agriculture.

## 5.2 Inventory comparison – analysing differences and similarities

### 5.2.1 General observations

A full and detailed intercomparison of the inventories described above 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  $\text{NO}_x$ , the differences between EMEP and EDGAR figures are substantial for some countries and remarkably different in total. Some of the potential sources of these discrepancies have been highlighted in Sect. 3.3, in particular the different system boundaries including some emission sources that are not reported under EMEP in the EDGAR dataset.

A similar picture emerges when investigating the differences in  $\text{N}_2\text{O}$  emission data reported under UNFCCC with EDGAR data.

The overview comparison of sectoral emission shares between the three regions

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investigated (Fig. 6) indicates some substantial differences between the emission profiles of Europe and the US on one side and China on the other. In particular the high contribution of emissions from agriculture to total  $\text{NH}_3$  emissions in general is evident. In the case of  $\text{NO}_x$  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  $\text{N}_2\text{O}$  emissions and – to a smaller extent –  $\text{NH}_3$  emissions in the US compared to Europe.

This can only be explained by significantly different assumptions regarding emission factors for  $\text{N}_2\text{O}$  and  $\text{NH}_3$  from vehicles. However, vehicle technologies are unlikely to be that different and driving conditions, split between diesel and gasoline operated vehicles and suchlike are not enough to generate a difference of this magnitude.

To explore differences and similarities in more detail, it would require not only emission inventories to be split in highly detailed source sectors, but as well the availability and transparency of activity data and emission factors used for their compilation. Without these, it is merely possible to identify and indicate difference, yet difficult to pinpoint the reasons for them.

### 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 NFR02) which is closely aligned with the UNFCCC common reporting format (CRF) and thus mainly

driven by fossil fuel combustion sources, REF) and provides much less detail and no distinctions are made for instance with regard to the type of fuel used in power plants or road transport modes. EDGAR uses a more processed based sectoral split, which is not always directly comparable to NFR sectors, but in most cases, an equivalent source can be found.

Table 6 provides a detailed comparison of source sectors split into the smallest categories provided in the NAEI (these are grouped by NFR sectors) for NO<sub>x</sub>, NH<sub>3</sub> and N<sub>2</sub>O. 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 in power plants are quite similar, but NO<sub>x</sub> emissions in EDGAR v4 are 17% lower, N<sub>2</sub>O from this source group is less than a third than NAEI has. For road transport, the picture is similar (17% lower NO<sub>x</sub> emissions, 87% lower N<sub>2</sub>O emissions), while NH<sub>3</sub> from road transport is 47% higher in EDGAR v4. NO<sub>x</sub> 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<sub>x</sub> emission factors in general. Thus, magnitude of differences in these sectors between NAEI and EDGAR v4, however, is larger than anticipated.

On the other hand, total NH<sub>3</sub> 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 significantly higher emissions of N<sub>2</sub>O 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 sub-sectors. In addition to that, NH<sub>3</sub> 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.

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In summary, the difference between country total emissions between both inventories is within the uncertainty margins expected, albeit somewhat large for  $\text{NO}_x$  emissions (EDGAR v4 20.9% below NAEI).  $\text{NH}_3$  emissions in EDGAR v4 are 36.9% higher than in the NAEI and  $\text{N}_2\text{O}$  emissions agree very well with regard to total emissions (2.6% difference). However, for all three substances, sectoral allocations can be very different between inventories, which of course has 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.

### 5.3 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.

Figure 8 displays trends in  $\text{NO}_x$  emissions for the period 1990–2005, based on data from EMEP and the USEPA, and additional values from literature for specific years or periods. 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 PR of China, no comprehensive 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) conclude that  $\text{NO}_x$  emissions in China from 1995–2004 likely increased by 70%. This is of particular relevance due to the amount of emissions, which – if emissions in China were set to further increase –

may well exceed those of the US and Europe, especially while these show stagnating ( $\text{NH}_3$ ) or downward trends ( $\text{NO}_x$ ).

The picture for  $\text{NH}_3$  emissions (Fig. 6) shows no significant reductions in the 15 year period indicated. A moderate downward trend in Europe can be observed ( $-18\%$ ), which 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. 6 indicate a small ( $6\%$ ) reduction below 1990 for 2005, this is due to a change in the calculation methodology in 2000, which 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. Again, Chinese emissions of  $\text{NH}_3$  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, 2007) for the year 1995. This latter figure would indicate an increase of  $21\%$  within these five years. This figure is attributed in Klimont's estimates to a  $27\%$  increase in  $\text{NH}_3$  emissions from livestock and a  $19\%$  increase in fertilizer related emissions, reflecting increased consumption of livestock products in China, especially poultry ( $57\%$  increase).

Emission trends for  $\text{N}_2\text{O}$  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; adjusted to Olivier and Berdowski, 2001) inventory of historic emission trends and EDGAR v4 (EDGAR, 2008) emissions for the year 2000, which also include China make a rough trend assessment possible. 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. For the US, EDGAR Hyde 1.4 pictures a slow increase for the period 1990 to 1995, with 2000 emissions remaining at approximately the same level, while UNFCCC figures are slightly above  $1.3 \text{ Tg } \text{N}_2\text{O yr}^{-1}$ . A significant increase of  $\text{N}_2\text{O}$  emissions

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from the PR of 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. Hence some caution is needed in considering the overall trends for the period 1990 to 2000.

## 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 CLR-TAP 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. 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. 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 significant time lag due to the time it takes to compile them based on official statistics and to undergo validation and error

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checking, before they are officially submitted. Independent inventories however do not always have access to the latest, most accurate and detailed statistics for each country and hence are prone to over/underestimate emissions from individual sectors.

At the same time, there are clearly missing sources 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<sub>3</sub> from sea birds, NO<sub>x</sub> from lightning and forest fires etc.). Compared to the main contributing sectors of anthropogenic emissions, the contributions of these missing sources to date is 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<sub>2</sub>O 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<sub>2</sub>O, 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 evidence and the level of detail provided allows for an in-depth assessment of the methodology used and factors and values applied.

The coverage of regions and substances for instance in the EDGAR inventory is comprehensive, but the fact that the NH<sub>3</sub> inventory for the time after 1990 is not yet available 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 pol-

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lutant 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  $1 \times 1$  or  $0.5 \times 0.5$  degree is sufficient to capture the general trends and concentrations/depositions. Yet, for regional and local modelling, inventories with a resolution of more than  $10 \text{ km} \times 10 \text{ km}$  are likely to miss vital distribution patterns and thus lead to mismatches between model results and observations. And while national emission inventories with a spatial resolution of e.g.  $1 \text{ km} \times 1 \text{ km}$  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 in Sect. 6.2 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 Trends and uncertainties

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.  $\text{NO}_x$  emissions in Europe and the US have fallen markedly, reflecting successful emission control policies especially since the early 1990s.  $\text{NO}_x$  emissions from the PR of 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  $\text{NO}_x$  emissions can be anticipated.

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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.

5 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 2 to 3 times higher than European or US emissions in this period. In addition to that, Klimont (2001) projected a steep increase in  $\text{NH}_3$  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 to 19.9 Tg  $\text{NH}_3$  for 2030.

Trends for nitrous oxide emissions finally indicate a slow decrease of emissions in Europe, a slight increase in US  $\text{N}_2\text{O}$  emissions, and a substantial growth in emissions in China. However, the trends depicted by data from the EDGAR Hyde 1.4 project and 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.

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For European and US emissions,  $\text{NO}_x$  emission values can be seen as quite robust and well understood, while in particular the trend of  $\text{NO}_x$  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  $\text{NO}_x$  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  $\text{NO}_x$  emissions from the power generation sector beyond 2010 (Vestreng et al., 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 (IPCC) directives will introduce further stringent measures to reduce  $\text{NH}_3$  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  $\text{N}_2\text{O}$ , 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 of

ten 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<sub>3</sub> from seabirds and other animals (Blackall et al., 2007) need to find their way into the guidelines for emission reporting and become a regular part of emission inventories. 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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**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 consumption	(X)	X	X	
Agricultural mineral Nr fertilisation and denitrification	(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

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**Table 2.** NH<sub>3</sub> Emissions of Candidate Countries and non-EU member states in EMEP (EMEP, 2008) and EDGAR (EDGAR, 2008) (emissions in Tg NH<sub>3</sub> for the year 2000).

	EMEP	EDGAR	Difference
Croatia	53.21	48.32	−9.2%
Macedonia (FYROM)	14.35	11.26	−21.5%
Turkey	402.95	423.36	5.1%
Subtotal EU Candidate Countries	470.51	741.56	
Norway	23.16	45.60	96.9%
Switzerland	60.38	56.35	−6.7%
Subtotal Non-EU Countries	83.55	101.95	

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**Table 3.** NO<sub>x</sub> Emissions of Candidate countries and non-EU member states in EMEP and EDGAR v4 (EMEP, 2008; EDGAR, 2008).

	EMEP	EDGAR	Difference
Croatia	76.81	93.57	21.8%
Macedonia (FYROM)	38.83	31.95	−17.7%
Turkey	942.41	1054.90	11.9%
Subtotal EU Candidate Countries	<i>1058.04</i>	<i>1180.42</i>	
Norway	223.79	191.29	−14.5%
Switzerland	101.30	121.95	20.3%
Subtotal Non-EU Countries	<i>325.09</i>	<i>271.74</i>	

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**Table 4.** N<sub>2</sub>O Emissions of candidate countries and non-EU member states in UNFCCC and EDGAR>.

	UNFCCC	EDGAR	Difference
Croatia	10.59	6.92	−34.6%
Macedonia (FYROM)	N/A	1.22	N/A
Turkey	18.52	66.95	261.5%
Subtotal EU Candidate Countries	N/A	75.09	
Norway	14.59	6.64	−54.4%
Switzerland	10.53	5.46	−47.9%
Subtotal Non-EU Countries	25.12	12.10	

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**Table 5.** Indication of uncertainty estimate for greenhouse gases. Source: Olivier et al. (1999).

Emission source category	Activity data	Emission Factor			Total Emissions		
		CO <sub>2</sub>	CH <sub>4</sub>	N <sub>2</sub> O	CO <sub>2</sub>	CH <sub>4</sub>	N <sub>2</sub> O
Fossil fuel use							
Fossil fuel combustion	S	S	M	M	S	M	M
Fossil fuel production	S	M	M	–	M	M	–
Industry/solvent use							
Iron & steel production	S	–	S	–	–	S	–
Non-ferrous production	S	–	S	–	–	S	–
Chemicals production	S	–	S	L	–	S	M
Cement production	S	S	–	–	S	–	–
Solvent use	M	–	–	–	–	–	–
Miscellaneous	V	–	–	–	–	–	–
Landuse/waste treatment							
Agriculture	S	–	L	L	–	L	L
Animals (excreta/ruminants)	S	–	M	L	–	M	L
Biomass burning	L	S	M	L	L	L	L
Landfills	L	–	M	–	–	L	–
Agricultural waste burning	L	–	L	L	–	L	L
Uncontrolled waste burning	L	–	–	–	–	–	–
All sources	–	–	–	–	S	M	L

S=small (10%); M=medium (50%); L=large (100%); V=very large (>100%) “–” not applicable/negligible

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**Table 6.** Detailed comparison between source sectors in the UK National Atmospheric Emissions Inventory (NAEI, 2008) and EDGAR v4 (EDGAR, 2008).

NFR code	NFR Name	NAEI			EDGAR v4		
		NO <sub>x</sub>	NH <sub>3</sub>	N <sub>2</sub> O	NO <sub>x</sub>	NH <sub>3</sub>	N <sub>2</sub> O
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	<i>714.40</i>	<i>1.10</i>	<i>9.48</i>	<i>591.26</i>	<i>0.75</i>	<i>3.02</i>
1A3aii(i)	Civil Aviation - Domestic Take-Off and Landing	1.80		0.02			
1A3aii(ii)	Civil Aviation - Domestic Cruise	7.10		0.05			
	Subtotal stationary combustion	8.80		0.07			
1A3bi	Road Transport – Passenger Cars	215.10	9.20	14.13			
1A3bii	Road Transport – Light Duty Vehicles	58.50	0.20	1.19			
1A3biii	Road Transport – Heavy Duty Vehicles	274.00	0.10	1.09	457.04	13.98	2.14
1A3biv	Road Transport – Mopeds & Motorcycles	1.30		0.01			
	Subtotal road transport	<i>548.90</i>	<i>9.50</i>	<i>16.42</i>	<i>457.04</i>	<i>13.98</i>	<i>2.14</i>
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	<i>134.60</i>		<i>1.09</i>	<i>91.06</i>	<i>0.03</i>	<i>0.05</i>
1A4a	Commercial/Institutional Combustion Plants	20.90		0.08			
1A4bi	Residential Combustion Plants	108.30	1.50	0.37	110.14	0.77	0.51
	Subtotal residential/commercial combustion	<i>129.20</i>	<i>1.50</i>	<i>0.45</i>	<i>110.14</i>	<i>0.77</i>	<i>0.51</i>

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Table 6. Continued.

NFR code	NFR Name	NAEI			EDGAR v4		
		NO <sub>x</sub>	NH <sub>3</sub>	N <sub>2</sub> O	NO <sub>x</sub>	NH <sub>3</sub>	N <sub>2</sub> O
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	

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Table 6. Continued.

NFR code	NFR Name	NAEI			EDGAR v4		
		NO <sub>x</sub>	NH <sub>3</sub>	N <sub>2</sub> O	NO <sub>x</sub>	NH <sub>3</sub>	N <sub>2</sub> O
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		<i>248.40</i>	<i>3.80</i>		<i>233.65</i>	<i>22.04</i>
4D1	Direct Emissions from Agricultural Soils		<i>35.60</i>	<i>81.31</i>	<i>29.34</i>	<i>190.81</i>	<i>26.07</i>
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	<i>0.20</i>	<i>1.90</i>	<i>0.20</i>	<i>2.69</i>	<i>0.92</i>	<i>0.05</i>
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	<i>2.00</i>	<i>9.60</i>	<i>4.08</i>		<i>0.50</i>	<i>5.87</i>
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	1619.80	323.00	129.21	1281.53	442.08	125.89
					-20.9%	+36.9%	-2.57%

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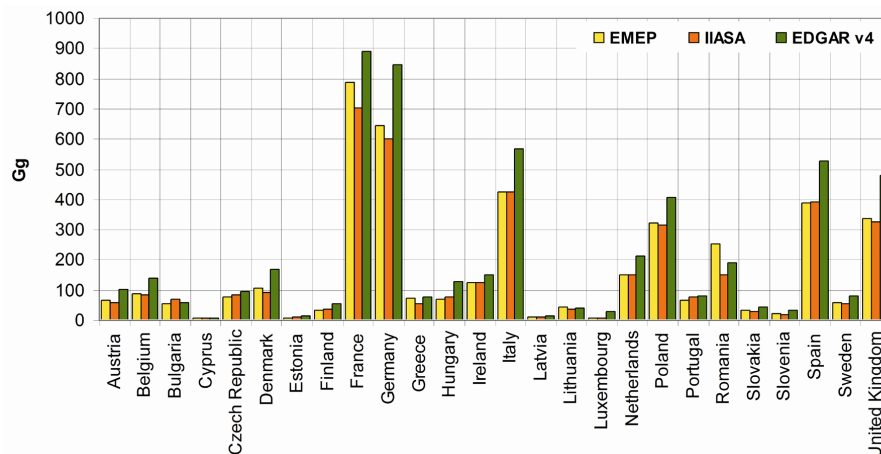
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**Fig. 1.** Comparison of EU27 (not showing Malta and Cyprus) emissions of ammonia reported to EMEP and compiled by IIASA and the EDGAR v4 database for the year 2000. The slight differences between IIASA estimates and EMEP figures arise most likely 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.

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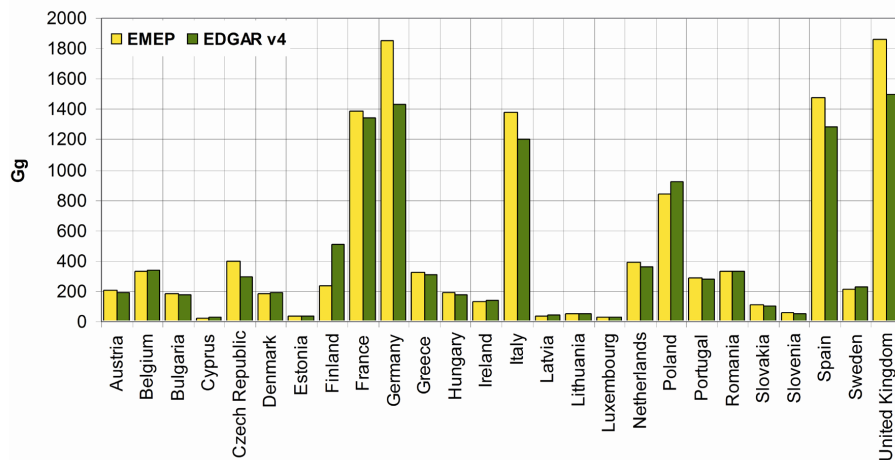
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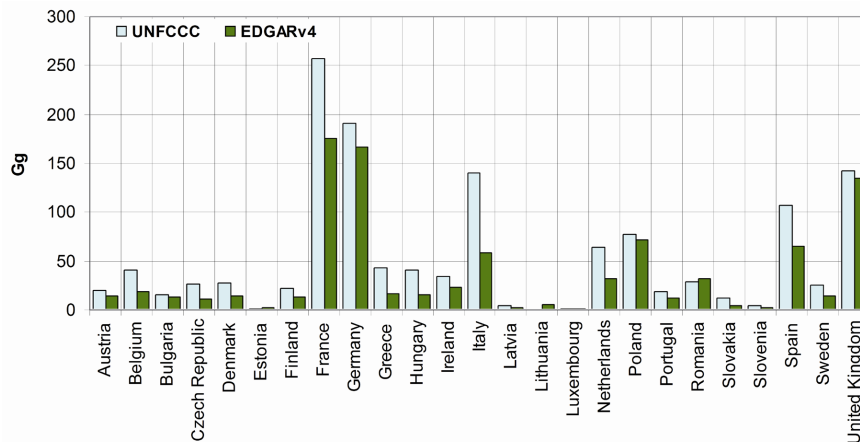


**Fig. 2.** Comparison of EU27 (not showing Malta and Cyprus) emissions of NO<sub>x</sub> (expressed as Gg NO<sub>2</sub>) reported to EMEP (EMEP, 2008) and presented in the EDGAR database (EDGAR, 2008) for the year 2000.

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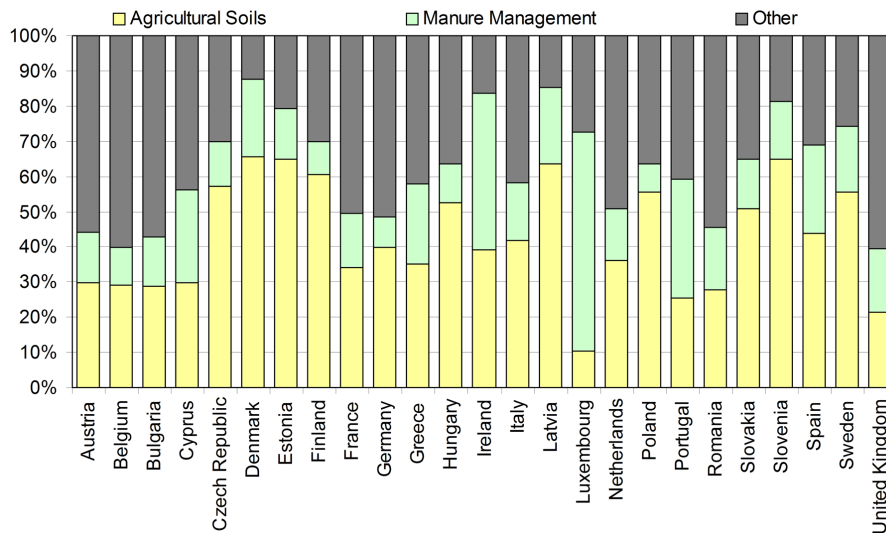


**Fig. 3.** Comparison of EU27 (not showing Malta and Cyprus) emissions of N<sub>2</sub>O reported to UNFCCC (without LULUCF, UNFCCC, 2008) and presented in the EDGAR database (EDGAR, 2008) for the year 2000.

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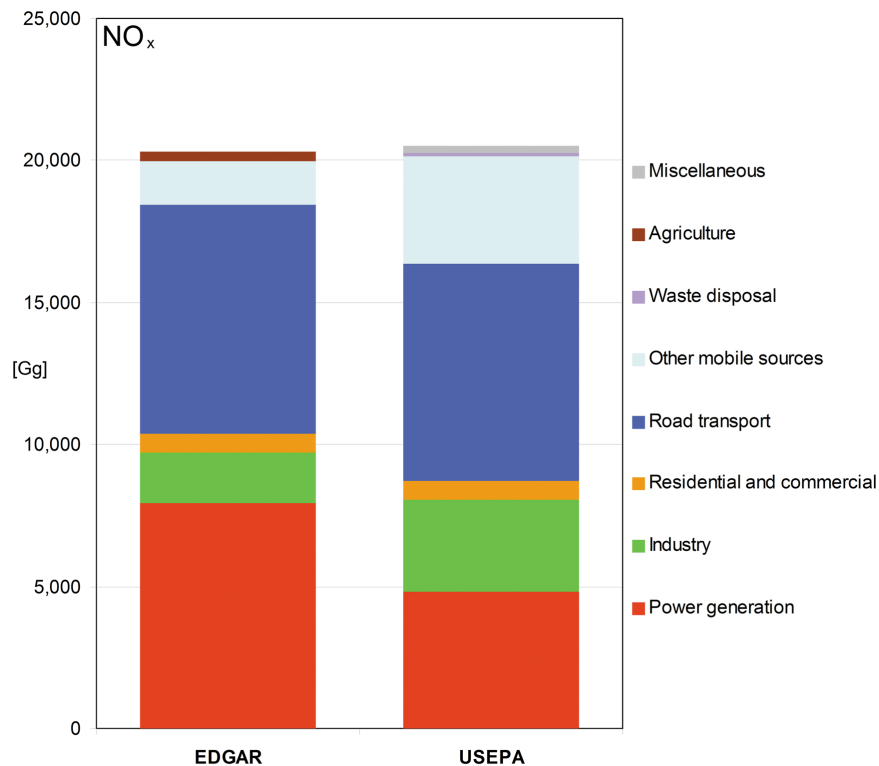
**Fig. 4.** Relative contributions of N<sub>2</sub>O emissions from agricultural soils and manure management in the EU27 countries according to the EDGAR v4 inventory (EDGAR, 2008).

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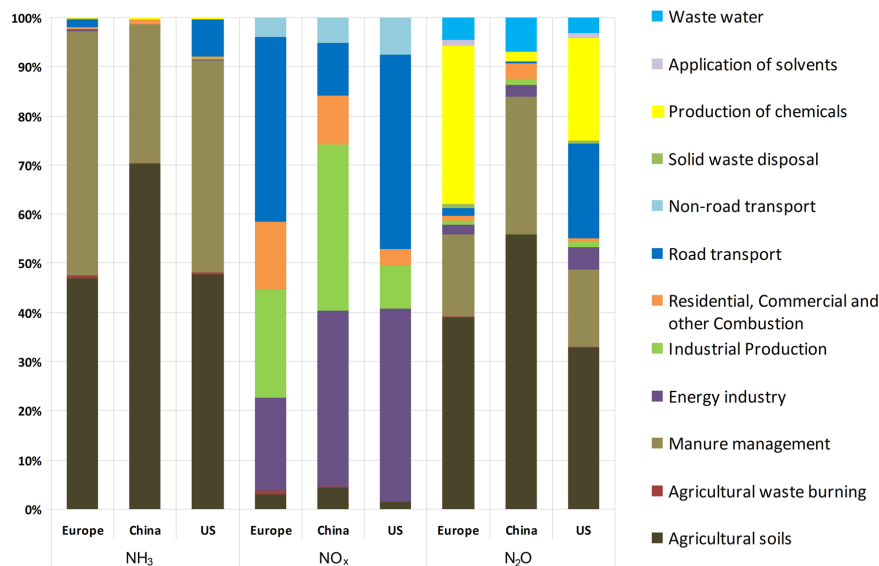


**Fig. 5.** Comparison of US EPA emissions of NO<sub>x</sub> with the EDGAR database for the year 2000 (USEPA, 2008; EDGAR, 2008).

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**Fig. 6.** Overview comparison of sectoral emission shares in EDGAR v4 (EDGAR, 2008) for the three regions Europe, China and the US, for NH<sub>3</sub>, NO<sub>x</sub> and N<sub>2</sub>O. The graphs highlight the quite different contributions of emission sectors to total emissions in different regions, as well as the quite dominant contribution of agricultural sources to both NH<sub>3</sub> and N<sub>2</sub>O emissions.

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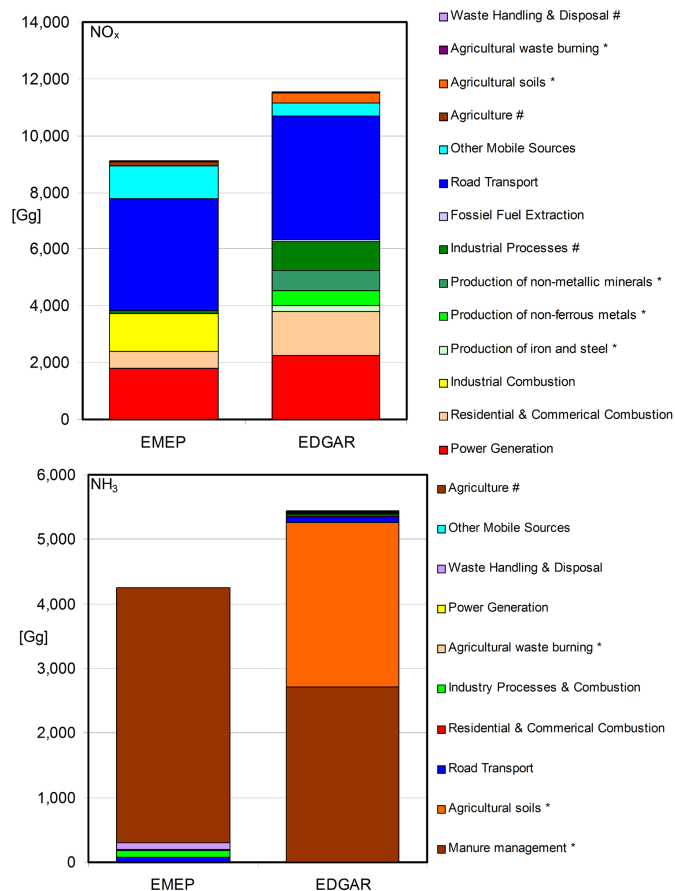
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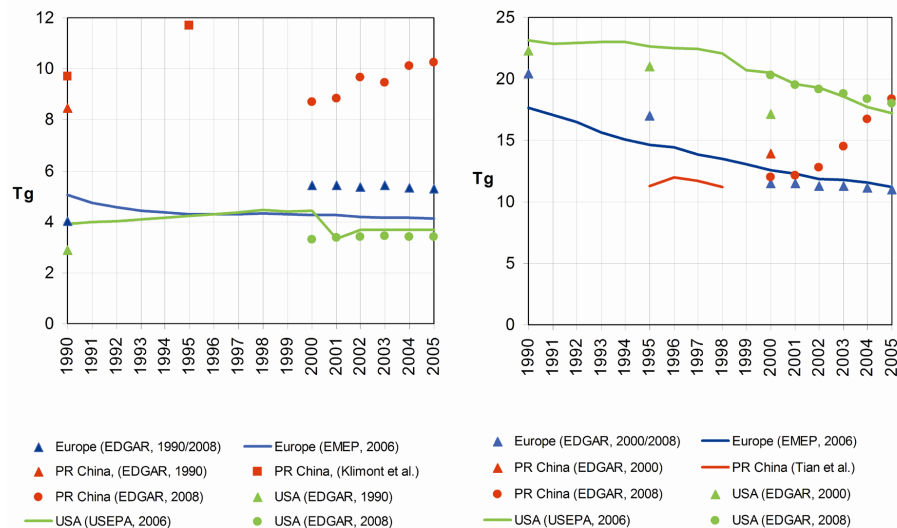


**Fig. 7.** Sectoral comparison of emissions for the EU27 countries for EMEP (EMEP, 2008) and EDGAR v4 (EDGAR, 2008) emission data for the year 2000. (# indicates sectors provided by EMEP only, \* indicates sectors only detailed in EDGAR).

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**Fig. 8.** Estimated trends in NH<sub>3</sub> (left) and NO<sub>x</sub> (right) emissions in Europe, the US and the PR of China for the period 1990–2005.

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