

# Reactive and Non-reactive Species of Nitrogen in Atmospheric Emission Inventories – A Review

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## ABSTRACT

Nitrogen in different forms is naturally available in abundance in the atmosphere. Excess nitrogen, however, has gradually become the one cause for the most pressing environmental problems we currently face in various regions of the world, since the *Haber-Bosch* process has made the industrial scale production of nitrogen fertilisers possible. Interactions between the drivers, namely emissions of ammonia, nitrogen oxides and nitrous oxide and pressures such as air pollution, acidification and eutrophication of soils and freshwater ecosystems, ecosystem damages and biodiversity impacts, leaching of nitrates into groundwater and global warming are very complex (see Galloway et al., 2003). Furthermore, trade-offs between effects and the control of emission sources lead to a need to balance the management of nitrogen at all stages of the nitrogen cycle.

This paper aims to review, how emissions of different nitrogen species, oxidised and reduced, reactive and non-reactive, are currently treated in emission inventories. While the paper and presentation here will focus on a European perspective and discuss some aspects of emissions in China, it forms part of a journal paper currently in preparation which will investigate how the situation of N species in inventories is addressed in *Europe*, the *United States of America*, and *The People's Republic of China*. Focusing on anthropogenic sources, the aim of the work presented here is to discuss the current state-of-the-art of quantifying the emissions of  $\text{NH}_3$ ,  $\text{NO}_x$  and  $\text{N}_2\text{O}$  and to assess, if current inventories provide a solid data basis for atmospheric dispersion models. In addition to that, aspects such as the potential synergies and trade-offs of attempts to manage different substances with often contradicting objectives and results are taken into account.

For the purpose of this conference, the paper will focus on Europe and China, and specifically investigate aspects of agreement between different inventories, temporal and spatial resolution, and observed trends in emissions in the past and anticipated future developments.

**Keywords:** nitrogen, ammonia, nitrogen oxides, nitrous oxide, emission inventories

## INTRODUCTION

Nitrogen gas (N<sub>2</sub>) presents more than 99.99% of nitrogen present in the atmosphere, and of the rest, again 99% are N<sub>2</sub>O (Wallace and Hobbes, 2006). Other N species are thus only present in trace concentrations, but play a vital role in atmospheric chemistry nonetheless. Ammonia is the only basic gas in the atmosphere and is solely responsible for neutralising acids formed through the oxidation of SO<sub>2</sub> and NO<sub>2</sub>, creating ammonium salts of sulphuric and nitric acid, which become atmospheric aerosols. NO and NO<sub>2</sub> play crucial roles both in tropospheric and stratospheric chemistry, for instance in the formation of tropospheric ozone.

At the same time, all N containing gases in the atmosphere are involved in biological nitrogen fixation and denitrification. Fixed nitrogen, however, can be returned into the atmosphere, for instance by biomass burning.

**Table 1.** Overview of the main sources and sinks of nitrogen-containing species

Sources	N <sub>2</sub>	NH <sub>3</sub>	NO <sub>x</sub>	N <sub>2</sub> O
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 nitrate fertilisation				X
Lightning			X	
<b>Sinks</b>				
Wet deposition		X	X <sup>1</sup>	
Dry deposition		X	X	
Chemical breakdown in the stratosphere				X

Anthropogenic activities have a significant impact on the magnitude of N cycled and released into the atmosphere, for instance, about 26 Tg N were produced by fossil fuel combustion in 1990 (Galloway 1998). And for the year 2050, Galloway et al. (2004) calculate a global rate of annual creation of reactive nitrogen (N<sub>r</sub>) of 221 Tg N yr<sup>-1</sup>, compared to approximately 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 has led to the establishment of more stringent critical loads and dynamic modelling approaches are explored to assess the ability of and time required for the recovery of ecosystems. In addition to that, the relevance of N<sub>2</sub>O as a potent driver of global warming has been acknowledged and emission control strategies do not focus solely on the CO<sub>2</sub> anymore.

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 do reflect the current knowledge on emissions of these trace gases. Furthermore, the somewhat parallel 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. In this paper, we want to critically review the way nitrogen in its different forms is accounted for and if the quality of current emission inventories is sufficient to support integrated strategies for N management, which are emerging in the US and Europe. For this purpose, existing inventories are assessed 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 and China will be discussed with the focus on how emissions of NH<sub>3</sub>, NO<sub>x</sub> and N<sub>2</sub>O are covered. Aspects of spatial and temporal resolution, sectoral detail on emission sources and completeness of reporting will be taken into account as well as 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, which was presented to the *United Nations Environment Programme* (UNEP) in Nanjing, China on October 16, 2004.

<sup>1</sup> as NO<sub>3</sub><sup>-</sup>

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.
- 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 will investigate in this paper, if our current methods to account for the emissions of different nitrogen species in emission inventories provide a sufficiently robust data basis for in depth assessments.

## EUROPEAN EMISSION INVENTORIES OF NITROGEN SPECIES

### 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 and EMEP *Meteorological Synthesizing Centre West* (MSC-West) in Norway currently hosts the inventory datasets submitted by countries in an online accessible database<sup>2</sup>.

Nitrous oxide on the other hand is not covered by the CLRTAP, but has to be reported under the UNFCCC by ANNEX I countries. Inventory submissions under the UNFCCC can be accessed online<sup>3</sup>.

In addition to these inventories which are generated based on obligatory reporting of national emissions, the EDGAR<sup>4</sup> 3.2 database provides global annual emissions per country and on a 1×1 degree grid for 1990 and 1995 for direct greenhouse gases CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O and HFCs, PFCs and SF<sub>6</sub> and the precursor gases CO, NO<sub>x</sub>, NMVOC and SO<sub>2</sub>. Similar inventories were compiled for acidifying gases, NH<sub>3</sub>, NO<sub>x</sub> and SO<sub>2</sub> and Ozone Depleting Gases (EDGAR 2.0). For the year 2000, the *EDGAR 32 Fast Track 2000* (32FT2000) dataset is 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. However, a trend projected dataset for NH<sub>3</sub> emissions for the year 2000 has not been compiled yet, hence EMEP NH<sub>3</sub> emissions will be compared to IIASA-GAINS data for the time being.

### Ammonia

In the case of NH<sub>3</sub>, the vast majority of EU27 emissions originate from agriculture (93%), with some small contributions from waste management (2.5%), industrial production processes (2%) and road transport (1.8%). 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.

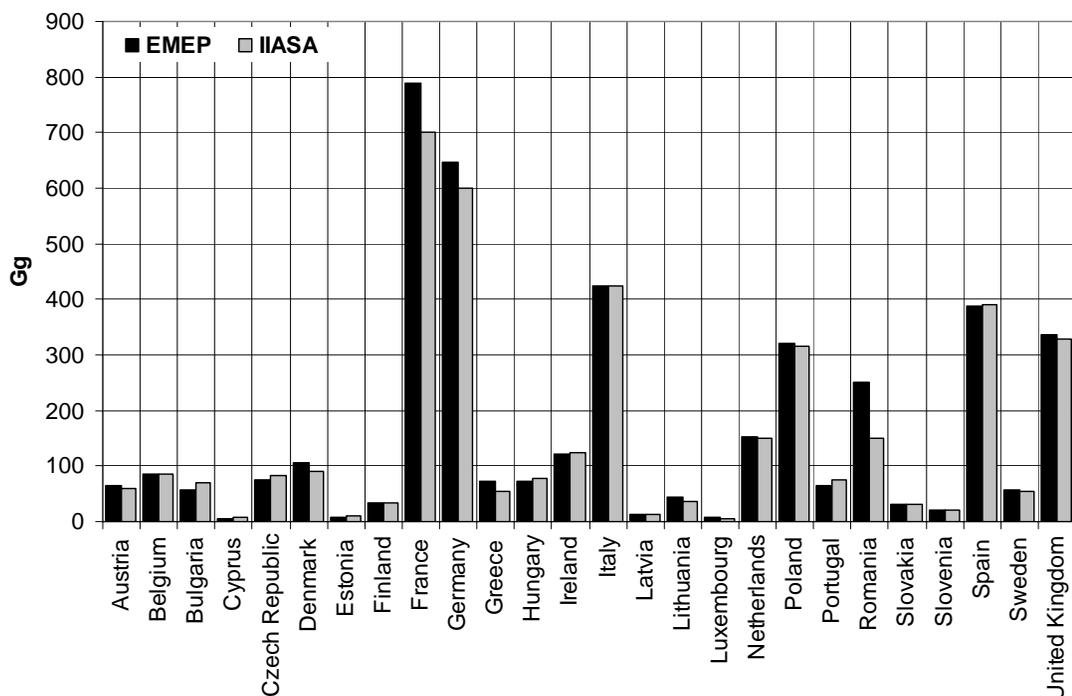
The comparison of datasets reported to EMEP and used by IIASA for integrated assessment modelling should not reveal significant difference, as input data (mainly animal numbers and agricultural practises) stem from the same information sources, national experts in most cases. However, a few differences seem to exist (France, Romania), which are most likely based on revisions of livestock numbers during bilateral consultations that have found their way into the IIASA dataset, but were not reported as recalculated emissions to EMEP. A more meaningful intercomparison will be made once the 2000 dataset of EDGAR is released, as the NH<sub>3</sub> figures there will be based on centrally available data and allow for an assessment, in how far basic assumptions underlying these numbers are robust.

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<sup>2</sup> EMEP WEBDAB, <http://webdab.emep.int>

<sup>3</sup> UNFCCC GHG inventory, [http://unfccc.int/ghg\\_emissions\\_data/items/3800.php](http://unfccc.int/ghg_emissions_data/items/3800.php)

<sup>4</sup> Emission Database for Global Atmospheric Research, <http://www.mnp.nl/edgar>



**Fig. 1.** Comparison of EU27 (not showing Malta and Cyprus) emissions of ammonia reported to EMEP and compiled by IIASA for the year 2000

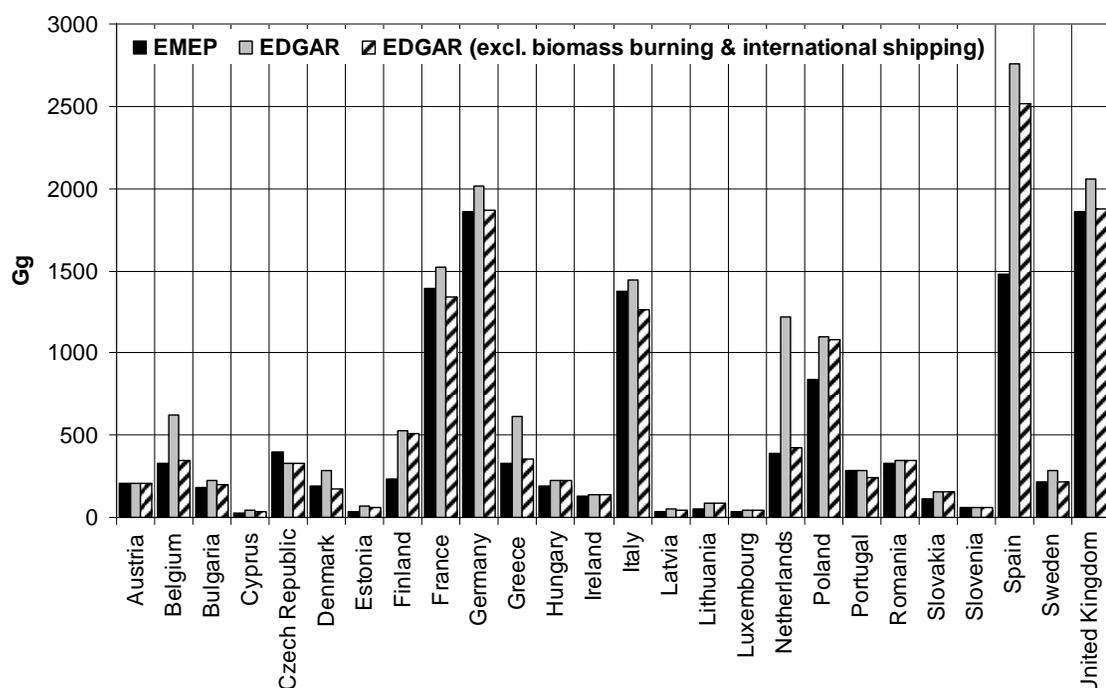
Fig. 1 illustrates that differences between the EMEP dataset and data used for model calculations with the RAINS model by IIASA are marginal. One reason for this is most likely that the EMEP emissions displayed are based on gap-filled expert estimates to complete the submissions by countries, which are often incomplete or comprise errors. In a similar way, the IIASA dataset was compiled centrally by emission experts and subsequently validated and revised in extensive bilateral consultations with country experts. For some countries, the IIASA data are slightly lower, overall, for the EU27 6% below the EMEP figures.

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, which 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 the 1990 data from EDGAR (4.02 Tg for the sum of Western Europe and Eastern Europe). 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  $\text{Nyr}^{-1}$ , with FSU emissions in 1990 at 3.4 Tg according to the EDGAR dataset.

### Nitrogen Oxides

The case of nitrogen oxides has been the focus of significant emission control activities in the recent decades, both from stationary sources (mainly large combustion plants) and mobile sources (road transport in particular). 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. However, comparing the inventories as they are, some emission sources included in the EDGAR dataset are not included in EMEP, such as biomass burning in forest and grassland fires and international shipping activities.

By excluding these emissions, which account for approx. 2.5 Tg in Europe in 2000, the EDGAR dataset still shows 12.6% more emissions in the EU27 than EMEP, with significant differences for individual countries. Finland, for instance, has more than 300 Gg of  $\text{NO}_x$  emissions from non-ferrous metal production in EDGAR, which makes up about 30% of the total global  $\text{NO}_x$  emissions from this sector. A similar case is Spain, where non-road transport emissions amount to 1.3 Tg, which is more than a factor of 10 higher than the same sectors emissions in Germany, for instance. For other countries, differences are less pronounced and hence potential errors harder to identify. It has to be stated, however, that the EMEP emission figures for  $\text{NO}_x$  as well have significantly changed over time, as only in recent years for instance emission factors for  $\text{NO}_x$  emissions from heavy duty vehicles had to be revised and increased by about 30%. Hence, inventories had to be recalculated, resulting in substantially higher emissions of  $\text{NO}_x$  than anticipated for instance when negotiating the ceilings for the NEC Directive.



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

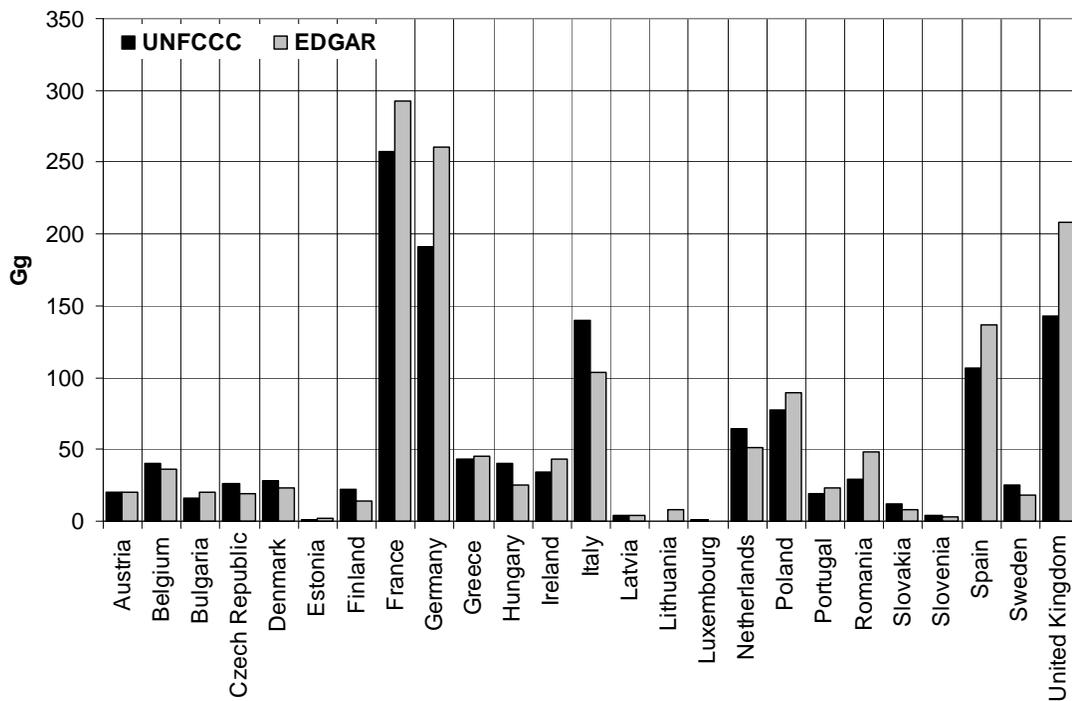
**Table 2.** NO<sub>x</sub> Emissions of ECCA countries and non-EU member states in EMEP and EDGAR

	<b>EMEP</b>	<b>EDGAR</b>	<b>Difference</b>
Croatia	76.81	100.30	30.6%
Macedonia (FYROM)	38.83	35.41	2.7%
Turkey	942.41	968.06	-8.8%
<b>Subtotal EU Candidate Countries</b>	<b>1058.04</b>	<b>1103.78</b>	
Norway	223.79	191.29	-14.5%
Switzerland	101.30	121.95	20.4%
<b>Subtotal Non-EU Countries</b>	<b>325.09</b>	<b>364.85</b>	

This comparison indicates that while NO<sub>x</sub> emissions are undoubtedly better understood 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.

### Nitrous Oxide

Finally, N<sub>2</sub>O emissions are not reported to EMEP under the CLRTAP, but subject to reporting obligations to the UNFCCC for ANNEX 1 countries. The following comparison is hence conducted 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%.



**Fig. 3.** Comparison of EU27 (not showing Malta and Cyprus) emissions of N<sub>2</sub>O reported to UNFCCC and presented in the EDGAR database for the year 2000

**Table 3.** N<sub>2</sub>O Emissions of ECCA countries and non-EU member states in UNFCCC and EDGAR

	UNFCCC	EDGAR	Difference
Croatia	10.59	10.68	0.8%
Macedonia (FYROM)	N/A	3.05	N/A
Turkey	18.52	136.39	636.6%
<b>Subtotal EU Candidate Countries</b>	<b>1058.04</b>	<b>1124.26</b>	
Norway	14.59	10.31	-29.4%
Switzerland	10.53	9.38	-10.9%
<b>Subtotal Non-EU Countries</b>	<b>25.12</b>	<b>19.69</b>	

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 remarkably 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 6 times higher than UNFCCC figures. At this stage, it is difficult to fully assess the uncertainties 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.

## UNITED STATES OF AMERICA

The US perspective is not covered at this stage. However, as will be indicated in the summary and conclusions, trends in emissions of NO<sub>x</sub> and NH<sub>3</sub> are similar in the US and Europe. While this will be elaborated on in the paper currently in preparation, on a general scale, this can be explained by control regimes which have focused on NO<sub>x</sub> and SO<sub>2</sub> emissions (leading to a considerable reduction in NO<sub>x</sub> emissions from stationary and mobile fossil fuel combustion). NH<sub>3</sub> emissions, mainly driven by mineral fertiliser application and livestock numbers, have not fallen markedly in the past decades.

## CHINA

### 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 TgN yr<sup>-1</sup>) to 1987 (3.25 TgN 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, 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 TgN 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 TgN 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 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 NO<sub>x</sub> emissions in China will continue to grow rapidly. However, other researches' results using latest statistical data (e.g., Tian et al., 2001) indicate that NO<sub>x</sub> emissions began to decrease somewhat, with total emissions in China amounting to 11.3 Tg, 12.0 Tg, 11.7 Tg and 11.2 Tg, respectively, from 1995 to 1998. In analysis by Tian et al., this may be the result of 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 NO<sub>x</sub> emissions in China are not growing as rapidly as some research had previously projected (e.g., Aardenne et al., 1999).

### Nitrous Oxide

The UNDP/GEF ECPINC Project<sup>5</sup> (*Enabling China to Prepare Its Initial National Communication*) 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<sub>2</sub>O emissions inventory for China. Directly following the IPCC concept and method (IPCC, 1997; 2000), Zhen et al (2004) estimated the direct N<sub>2</sub>O emissions in China at approximately ~75% of the annual total N<sub>2</sub>O released from anthropogenic reactive nitrogen input into croplands of China presently occurring as direct emissions. Accordingly, the estimated uncertainty for the N<sub>2</sub>O emission inventory of crop production is reasonably due to that of the direct emission. Zhen et al (2004) measured 54 direct N<sub>2</sub>O emission factors (EF<sub>d</sub>'s) obtained from 12 sites of Chinese croplands; of these 60% are underestimated by 29% and 30% are overestimated by 50% due to observation shortages. The biases of EF<sub>d</sub>s are corrected and their uncertainties are re-estimated. Of the 31 site-scale EF<sub>d</sub>s, 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<sub>d</sub>. The direct N<sub>2</sub>O emissions from Chinese croplands are estimated at 275 Gg N<sub>2</sub>O-N yr<sup>-1</sup> in the

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<sup>5</sup> <http://www.ccchina.gov.cn/en/NewsInfo.asp?NewsId=5392>

1990s, of which  $\sim 20\%$  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  $129 \pm 62\%$ ). And 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  $58.22 \text{ Gg N yr}^{-1}$  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  $0.41\text{--}0.90 \text{ Gg N yr}^{-1}$ . The total emission of  $N_2O$  as a result of fertilizer application was  $342.5 \text{ Gg N}$  in 1990, being the most important contributor to  $N_2O$  emission from agricultural soils.

While the IPCC methodology (IPCC, 1997; 2000) emphasizes anthropogenic effects, it neglects background emissions. If these are included, total emissions may be up to  $70\%$  higher than when using the IPCC methodology (Yan et al., 2003). Most of the current estimates following the IPCC methodology do not include background  $N_2O$  emissions; some even consider  $N_2O$  emissions from paddy fields to be zero. Yan et al. (2003) demonstrate some modifications to the IPCC method, considering background emissions in the process of estimating direct  $N_2O$  emission from cropland. Their results estimated  $N_2O$  emissions from croplands at  $476.3 \text{ Gg N}$  in 1995, including background emissions of  $127.1 \text{ Gg N}$ . Background emission values were derived from measurements in studies that compared emissions from fertilized and unfertilized fields.

On the basis of available field measurements of  $N_2O$  emissions and statistic data (China Agricultural Yearbook, 1996), Xing et al (1998) present a preliminary estimation of  $N_2O$  emission in year 1995 from various cropping systems. Although current field data are not enough to quantify emissions per crop, they suggest that the emission fluxes of this gas from upland fields could differ significantly from those of paddy fields. As calculated the average  $N_2O$  emission flux from each region based on the available field measurements from different region in different cropping season, it shows that  $N_2O$  emission from cropland in China in 1995 was  $398 \text{ Gg N}$ , in which,  $310 \text{ Gg N}$ , accounting for  $78\%$  of the total, was emitted from upland and  $78 \text{ Gg N}$  from paddy fields. Another estimation result is  $340 \text{ Gg N}$ , based on DNDC model simulations (Li et al., 2001). 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 its great temporal and special variations of fluxes from cropland.

## 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 and livestock, representing in the 1990's about  $52\%$  and  $41\%$  of total emissions, respectively (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  $9.7$  and  $11.7 \text{ Tg}$  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  $100 \text{ Gg NH}_3$  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  $1.65 \text{ Tg N}$  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  $5.8 \text{ TgN}$  for the area of China.

Because of the higher application rate of synthetic N-fertilizers needed in order to increase crop production efficiency, the ammonia emission is expected to increase in the future.

## EVALUATION

### General uncertainty assessment

For all emission inventories portrayed here, the assessment of uncertainties is a key aspect (Lee et al., 1997). For national inventories, issues of compliance with reduction targets and emission ceilings is 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<sup>6</sup>, covering aspects of completeness, an analysis of key sources and uncertainties. In addition to this, regular desk and 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”<sup>7</sup> 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. Oliviver et al. 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 4).

**Table 4.** Indication of uncertainty estimates 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
<b><i>Fossil fuel use</i></b>							
Fossil fuel combustion	S	S	M	M	S	M	M
Fossil fuel production	S	M	M	-	M	M	-
<b><i>Industry/solvent use</i></b>							
Iron & steel production	S	-	S	-	-	S	-
Non-ferro production	S	-	S	-	-	S	-
Chemicals production	S	-	S	L	-	S	M
Cement production	S	S	-	-	S	-	-
Solvent use	M	-	-	-	-	-	-
Miscellaneous	V	-	-	-	-	-	-
<b><i>Landuse/waste treatment</i></b>							
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	-	-	-	-	-	-
<b>All sources</b>	-	-	-	-	<b>S</b>	<b>M</b>	<b>L</b>

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

### Inventory compilation

#### General aspects

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 NO<sub>x</sub>, 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 above, in particular the

<sup>6</sup> see <http://www.emep.int/emis2007/reportinginstructions.html> for the current reporting guidelines

<sup>7</sup> <http://www.ipcc-nggip.iges.or.jp/public/gp/english/>

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 N<sub>2</sub>O emission data reported under UNFCCC with EDGAR data. Here, a more in-depth review of the definitions of sectors and the methodologies for calculating emissions in particular for those sources with large uncertainties (e.g. land-use, natural and biogenic sources) is required to finally identify where these differences may stem from.

### Sectoral structure and inventory compilation

The EDGAR inventory is different from both EMEP and UNFCCC in the way it is compiled based on independent expert assessments regarding emission factors and activity rates. This approach has the distinct advantage of providing a harmonised, streamlined inventory across different countries using the same methodology and most often based on openly available datasets. A downside of this approach is obviously the lack of intimate and detailed knowledge of national or regional aspects that may well influence emissions substantially.

Both EMEP and UNFCCC inventory compilations are conducted through submissions from signatories to the relevant protocols (UNECE Convention on Long Range Transboundary Air Pollution, respectively the United Nations Framework Convention on Climate Change). National experts compiling the submissions for these inventories often have unique access to real-time (measure or calculated) activity data and better knowledge of technologies currently applied, which is not always accessible from the outside. On the other hand, as the reported emission figures can be highly political, when compliance with emission targets is concerned, it may occur that not in all cases the emission calculation is based on the state-of-the-art of scientific findings with regard to emission factors. In addition to that, as new findings emerge, the very basis for the emission targets that have been agreed may be changed, where changing the methodology for emission reporting would require significant recalculations of historic emissions. As an example for this, research indicated and later confirmed that heavy goods vehicles (HGV) had significantly larger (in the order of 30%) NO<sub>x</sub> emissions in real-life operations compared to test cycle measurements in Europe in the late 1990s. This information was finding its way into different national inventories with a substantial time lag and it is yet unclear if the new emission factors are currently applied in all countries. Last but not least, it is still the case that in some countries emission reporting and energy policy (including projections) or other sectoral policy areas are not linked and follow different agendas. In the same way, reporting of mainly air pollutant emissions to EMEP is not always integrated with or conducted by the same team that reports greenhouse gases to UNFCCC, at times using different methodologies or underlying activity rates, both current and projected. Last but not least, a major caveat of officially reported emission data is a time-lag of often several years, as well as inventories suffering from incomplete reporting for individual countries (both missing sectors and late submissions of full inventories). This is a serious problem for modelling activities, as it is impossible to conduct any model experiment without a complete dataset.

Where the sectoral structure is concerned, the EMEP and UNFCCC inventories have started on very different grounds. While EMEP has a clear focus on air pollutants, including acidifying substances and nutrients, heavy metals and photo-oxidant precursors, UNFCCC includes greenhouse gases only. Hence, the initial sectoral structure of the EMEP inventories was marked by a dominance of combustion sources, on a very detailed technical level (as different fuels and combustion technologies have very distinct NO<sub>x</sub> or SO<sub>2</sub> emissions). In recent years, harmonisation efforts with regard to the reporting guidelines to EMEP have resulted in a sectoral format that is similar to that of UNFCCC. And since greenhouse gas emissions are largely emitted from fossil fuel combustion sources, this harmonisation has on the one hand facilitated a more streamlined reporting of emissions to both conventions. However, in particular non-combustion emissions require different methodologies and thus additional sectors/headings needed to be (re)introduced over time. The current “Nomenclature for Reporting” (NRF) used by EMEP in its 2002 version has been widely harmonised with the UNFCCC CRF (Common Reporting Format), but was amended to include sources e.g. of particulate matter (from tyre and break wear, suspension of road dust) or NMVOCs (from using organic solvents) etc.

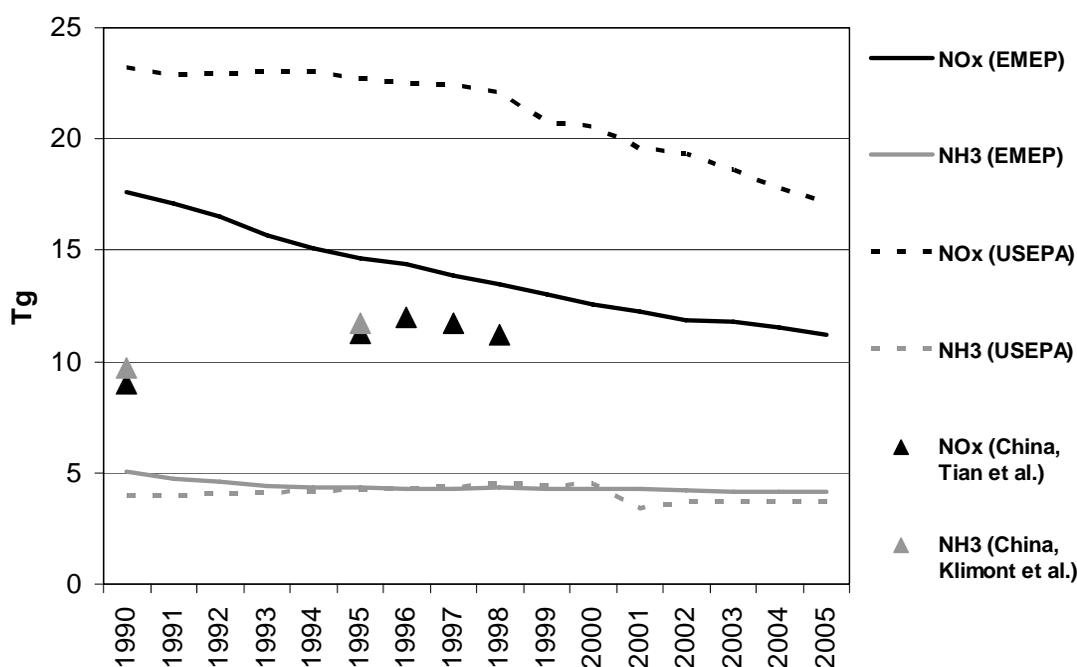
### Different regional trends

In general, marked differences exist between the emission situation in Europe and China, mainly with regard to the control and management regimes of emissions and the current projections for future development. In Europe, NO<sub>x</sub> and NH<sub>3</sub> emissions have been subject to regulation under both European Commission legislation and – even longer term – to the protocols under the UNECE Convention on Long Range Transboundary Air Pollution. This has not only led to coordinated reduction efforts, but has as well created reporting and inventorying efforts, supported by research activities, which allow for a detailed investigation of sources, pollutants and trends. For China, emission inventories are at this stage more driven by scientific interest and are compiled and evaluated by researchers (see for instance Wang et al. 2005, Ohara et al. 2007).

While in Europe, trends for NO<sub>x</sub> emissions between 1990 and 2005 have been steadily pointing downwards due to legislation to reduce emissions from stationary sources (e.g. the Large Combustion Plants Directive) and mobile sources (mainly the EURO emission standards for vehicles) amounting to

approx. 34% reduction of EU27 emissions, NH<sub>3</sub> emissions have only been reduced slightly (-11.7% for the EU15, no EU27 data available). These trends are roughly comparable with the situation in the US (NO<sub>x</sub> -26% and NH<sub>3</sub> -6% between 1990 and 2005 based on US EPA figures).

The situation in China, however, is different, both due to trends of strong economic growth (and the accompanying effects on transport and energy demand increasing significantly) and emissions not (yet) being subject to regulations enforcing control measures. Recent findings indicate though that a gradual decoupling of growing energy demands and the increase in NO<sub>x</sub> emissions can be observed. The case of NH<sub>3</sub> emissions in China is – similar to Europe or the US – driven by agricultural production primarily (livestock numbers and fertiliser production/application).



**Fig. 4.** Different regional trends in NO<sub>x</sub> and NH<sub>3</sub> emissions based on EMEP (gap-filled ‘expert emissions’ for the EU27), US EPA (national Tier1 emissions) and literature assessments for China

Where N<sub>2</sub>O is concerned, the UNFCCC and the guidelines of IPCC provide a more harmonised, common framework. However, as stated above, the IPCC methodology for calculating emissions e.g. from soils and other sources is subject to vast uncertainties. Both in Europe and China, significant efforts will still need to be undertaken in order to reduce these uncertainties and to quantify N<sub>2</sub>O emissions as well as an overall N budget validated by measurements. Research projects are currently underway in both regions to address some of the issues related to this (see for instance NitroEurope IP<sup>8</sup>, ChinaFlux<sup>9</sup> or a project on “Continuous field measurements and process-oriented modeling of N<sub>2</sub>O emission from the typical irrigated croplands in the North China”). In spite of the different targets for reduction under the UNFCCC and the opposite trends in energy demand and agricultural production, the use of common methodologies and approaches under the convention will make it easier to access and evaluate information on N<sub>2</sub>O and other GHG emissions in the future.

#### Temporal and spatial resolution

For both the EMEP and the EDGAR emission inventories, gridded datasets are available for modellers, with a 50 × 50 km resolution for EMEP and 1° × 1° for EDGAR. In both cases, the main source sectors are distinguished. For N<sub>2</sub>O, gridded emissions are only available from EDGAR, as spatial aspects of emissions are not taken into account under UNFCCC below the country scale.

At this stage, the temporal resolution is not addressed in any of the inventories investigated, while in some cases, emissions may have been calculated with comprehensive, process-based models driven by time-dependent factors, both anthropogenic (e.g. manure spreading on farmland) and biogenic/natural (e.g. meteorological parameters). All nitrogen species considered here are, however, subject to significant variations over time, showing marked diurnal and seasonal cycles. These temporal patterns are in some cases handled by atmospheric dispersion models, with built-in distribution patterns,

<sup>8</sup> <http://www.nitroeuropa.eu>

<sup>9</sup> <http://www.chinaflux.org/en/index/index.asp>

which are mainly static time-curves for anthropogenic sources and are to some extent driven by meteorological parameters (e.g. temperature, precipitation) for natural and biogenic sources.

## SUMMARY AND CONCLUSIONS

We have investigated different emission inventories and highlighted some of the most relevant similarities and differences observed. While it is clear, that different inventories are compiled and used for specific purposes and thus are subject to at times conflicting aims and objectives, one of the questions raised in this paper was, in how far existing inventories are able to provide robust and comprehensive datasets for the modelling of different nitrogen species. This question is of particular interest for efforts aiming at the calculation of closed nitrogen budgets. In this context, verification and validation of emission figures through modelling exercises and intercomparisons can only make a meaningful contribution to closing existing gaps in these budgets, if the main input data set – emissions – has known and quantifiable uncertainty ranges.

Where ‘official’ inventories compiled by national experts to fulfil reporting obligations to international bodies are compared to inventories calculated by independent experts for modelling purposes, the discussion is often marked by which of these are ‘better’. Based on our assessment, this argument is flawed and a more sensible concept is to use both types of inventories for the purposes they are compiled for. Independent inventories can provide a valuable source for modelling activities, as they provide a complete dataset without gaps and based on a transparent, harmonised methodology. Furthermore, they can help to identify potential gaps or inconsistencies in national ‘official’ inventories by stimulating scrutiny of potential missing sources, respectively invite national expertise to contribute to revising the independent assumptions regarding specific emission factors or activity rates for a country/region.

The temporal and spatial resolution provided by regional inventories is not sufficiently detailed for high-resolution modelling of the environmental fate of nitrogen species. This is particularly true for NH<sub>3</sub> emissions, where a significant share of emissions is often deposited near the source. However, the amount of input data required to develop emission inventories with a spatial resolution of e.g. 5 × 5 km or even 1 × 1 km makes it infeasible on a regional or global scale. For the purpose of verification/validation experiments, dedicated efforts to create inventories on the field or landscape scale should be undertaken to provide emission data input to atmospheric dispersion models with sufficient spatial resolution to catch near-source effects.

Long-term trends in emissions are similar in Europe and the US, where NO<sub>x</sub> emissions have fallen markedly due to regulations introduced for stationary and mobile combustion sources aimed at reducing the impacts of acidification and eutrophication (and tropospheric ozone). In a similar way, ammonia emissions in both regions have not reduced much in the course of the last two decades, and are to date mainly driven by mineral fertiliser application and livestock numbers. While there is further potential for reductions, future trends cannot be derived from the observed past trends in a straightforward way. The main reason for this is, in Europe, significant uncertainties regarding the future energy mix, the impact of increased shares of renewable energy sources/biofuels and the growing importance of domestic coal for electricity generation in the view of increasing oil and gas prices, as well as the phasing out of nuclear power plants in some European countries. The European situation for NH<sub>3</sub> and NO<sub>x</sub> is likely to change subject to the implementation of the revision of the EC National Emission Ceilings Directive (NECD), which is currently under development. This directive will set emission ceilings for each EC member state for the year 2020 and more stringent targets for NH<sub>3</sub> are expected to be established. The main reason for this is that while emissions of acidifying substances have markedly fallen due to reductions in NO<sub>x</sub> and SO<sub>2</sub> emissions, remaining NO<sub>x</sub> and NH<sub>3</sub> emissions still contribute significantly to exceedances of nutrient critical loads in ecosystems across Europe. Ammonia has become a major contributor to this in the past decade.

In China, a steady growth in emissions has been observed, and with levels of economic growth anticipated for the near future, can be expected to further increase. However, first signs of a decoupling of emission and e.g. power generation capacity increases may indicate a less steep increase of emissions, most likely depending on efficiency gains in power plant technologies at this stage. This may be easily off-set by increasing vehicle fleets and it is yet to be seen how air quality objectives will drive emission control efforts, in particular with regard to NO<sub>x</sub> emissions in urban areas. For both NH<sub>3</sub> and N<sub>2</sub>O, the growing food demand in China is likely to lead to steady and substantial increases in emissions.

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