Atmos. Chem. Phys. Discuss., 9, C2745–C2758, 2009
www.atmos-chem-phys-discuss.net/9/C2745/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Reactive nitrogen in atmospheric emission inventories – a review" *by* S. Reis et al.

Anonymous Referee #1

Received and published: 13 July 2009

General comments

This paper aims at assessing (and I roughly quote from the paper itself): 1. the state of the art of emissions estimation of NH3, NOx, N2O in the regions of US, China and Europe including allocation of source categories, temporal and spatial resolution, speciation and accessibility; 2. how inventories developed for different purposes (compliance checking or science) differs, 3. to which extent the inventories meet the modelling requirements hereunder how different Guidelines for emission estimation under e.g. the LRTAP and UNFCCC Conventions provide a sufficiently robust data basis for integrated assessment modelling, and to discuss 4. uncertainties in emission data and 5. future trends.

Further it is stated (p. 12414, l. 21) that: 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.

This last paragraph gives me in addition the expectations that 6. directions for future research needs and 7. how existing gaps between modelled and measured concentration or deposition rates could be most efficiently addressed, will be part of the paper's conclusion, which is only vaguely the case.

It is clearly a huge task to assess and address the above 7 important topics, and in my opinion the present paper only partly succeed in doing so. One reason could be that, even though the paper is rather well structured, mostly clear and generally well written, the section 6, discussion and conclusions, does not address the above topics explicitly to summarize the main findings from this paper. In order to increase the value of the paper, I would recommend to spell out the author's opinions on these issues based on findings from this paper.

The subject of the paper is scientifically highly interesting, both with respect to the nitrogen cycle research, interpretation of model results, and to some extent also to improve emission inventories in the future. The major difficulty in reviewing this paper is that the EDGAR data on NOx and NH3 referenced therein as EDGAR 2008 is not yet readily available to me. The EDGAR v4 (EDGAR, 2008) GHG data was published online in May 2009, thus the paper includes some rather fresh data. With the respect to the lack of EDGAR data accessibility for other scientists, but I underline, also because I cannot remember to have seen such a complete discussion of reactive nitrogen inventories before, the novelty aspect of this paper is certainly assured. I recommend publication in ACP when at least some more work has been put into the analysis of data and the rational behind conclusions are better founded.

I am however not so sure that the paper, at least in its present form, merits the form of

a review - as indicated in the title. Several sources of information, e.g. NO2 trends from satellite observations, ground based measurements in relation to emission trends, and certainly many papers and databases (e.g. GAINS) dealing with NOx, NH3 and N2O emission inventories are for instance not mentioned yet say discussed. Further it is stated (on p. 12431, l 14) that: "A full and detailed intercomparison of the inventories described above is beyond the scope of this paper." However, there are statements made and conclusions drawn based on sparse evidence and or the rational for the conclusions is not evident to me. I find several places both a lack of analysis of the emission data, and when an analysis is provided, it is sometimes rather superficial and the arguments are not checked against relevant information even though this can readily be done in many instances as pointed out below.

The paper also suffers from several errors and the fact that many figures and tables are not referenced or discussed in the text.

This paper has the potential of becoming much more useful source of information for modellers attempting to interpret why their results differ as they do, and for emission inventory people to increase further the level of accuracy in their inventories. Hopefully my detailed comments below might help to improve the final version of this paper.

Specific comments

1. In order to justify more clearly the choice of regions, please state how much of the global emissions per compound that are covered by the selected three regions according to the reference of your preference.

2. I would also recommend to spend one paragraph explaining the topic of lack of closure of the N budget to put this paper into perspective.

3. I would recommend to include in the beginning of the paper a section on data sources to increase the transparency of the paper and in particular with respect to e.g. EDGAR (2008), EDGAR fast track, EDGAR (1990), EMEP model data vs EMEP

officially reported data, data used for China etc.

4. It would be helpful with a table with at least total emissions for the different pollutants, at least in the section concerning emission from China, as this section is now rather obscure.

5. It would also increase the transparency if all emissions were referred to as e.g. Tg og Gg N (not NH3, NOx, NO2, N2O). This is also fits the spirit of this paper which is interested in telling us something about reactive nitrogen as such and not really in N2O, NO2 or NH3.

p. 12414, l.21: What is meant by: robustness (of emission estimates)?

p. 12415, l. 7: Is basic gas a well defined term? How is it defined?

P 12416 l. 18: I have a problem in seeing the contradiction put up between the bottom up inventories used for compliance monitoring (also scientists are working with those), and the bottom up inventories e.g. EDGAR. One main difference is that while many countries have access to country specific EFs, detailed and up to data activity data, good knowledge of penetration of measures and technologies, this is not always the case for "scientific" inventories. On the other hand, it can be argued that e.g. the EDGAR dataset is more comparable over countries and regions, since the same methodology are apparently applied. However, this might not be the case as availability of input data to the calculations differs for different countries, and the Guidelines/guidebooks followed by many national emission experts to a large extent assure a common methodological framework in the national submissions under different international obligations.

p. 12417, l. 25: We therefore critically investigate the way nitrogen in its different forms is accounted for in these processes. [I read processes as the reporting obligations or the emission inventories resulting from them]

I do not agree that the paper does investigate the difference between the UNECE and

the UNFCCC reporting obligations, and how they might be defective in order to do proper nitrogen management. It is however a very interesting subject, so I encourage you to look further into the reporting guidelines in order to highlight differences of importance for the work on the nitrogen cycle.

p. 12419 l. 8: See my recommendation for a session on data sources

p. 12419 l. 26: The air pollutants data are not officially released

p 12429, l. 2: What is improved in the spatial distribution in EDGAR v4?

Does this inventory have the sufficient spatial resolutions for all studies relevant to your paper? What is the resolution of the other inventories applied in this paper? Is there a difference in the spatial allocation of sources between the inventories included in your paper?

p. 12420, l. 25: 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.

I disagree. You have used outdated year 2000 EMEP emissions prepared for modelling in 2006. If you check the web site for officially reported data: `http://www.ceip.at/emission-data-webdab/emission-as-reported-by-parties/`, you will see that countries have reported recalculated emissions for year 2000, which are not reflected in the data you have used. For the countries which displays largest difference with IIASA data:

Diff reported data – model data

France Germany Romania

7.8 -18.8 -46:

Sector: N08 4 D 1 a: Synthetic N-ferilizers						
	Pollutant: NH3					
	Austria	Belgium	France	Germany	Spain	United Kingdom
2000	3.76870571	10.0269415	156.718325	80.9306908	261.564873	35.0905059

Thus my conclusion is that the EMEP trends should be updated with the most recent submission form countries.

p. 12421, l. 6: I disagree that this assumption cannot be checked. In the officially reported data you can compare the emissions from agricultural soils. I do not have the EDGAR data, but the data officially reported to EMEP is:

p. 12421, l. 9-14: What is your conclusion? You have stated that EDGAR emissions are much higher than EMEP for EU27 countries. For Europe (seems to be defined as EU27, TK, MK, HR, NO, CH in the case of EMEP inventory), they are comparable. Which countries do the EDGAR emissions of 4.02 Tg NH3 include?

p. 12421, l. 28: Overall lack of country specific emission factors measurement programs?

p. 124322, l. 4: ALSO in the future?

p. 12422, l. 8-10. Something is wrong with Fig 3, otherwise swap higher with lower. Also insert a reference to the figure in the text.

p. 12422, l. 15 Turkey (not show in Figure 3)

p. 12422, l. 16: It is fully possible to compare the emissions of N2O from agricultural soils in the UNFCCC database with those of EDGAR. I would recommend that you do so, for all countries and regions that have reported to UNFCCC in order to strengthen you argumentation.

p. 12424, l. 12, It might be worth including a note that US is moving from Mobile

6 to MOVES, and possible consequences of that : http://www.epa.gov/otaq/models/moves/: EPA has performed a preliminary comparison of Draft MOVES2009 to MOBILE6.2 using local data for several different urban counties, vary-ing the local data used by fleet age distribution, fraction of light- and heavy-duty VMT, local fuel specifications, meteorology, and other input factors. The results described here are based on the most recent data available and will vary depending on actual local inputs. EPA's findings are described below, by criteria pollutant.

For oxides of nitrogen (NOx), EPA has found that emissions from both light- and heavy-duty trucks are higher than previously estimated. In Draft MOVES2009, emissions estimates are based on EPA's analysis of I/M testing data, which incorporates in-use emissions data on a very large number of vehicles with model years from the mid-1990s to 2004. For heavy-duty trucks, Draft MOVES2009 incorporates newer "real world" data from on-road testing. In Draft MOVES2009, uncontrolled extended idle emissions from heavy-duty vehicles are projected to become a significant share of the mobile source NOx inventory in the future, assuming no change in extended idle activity as a fraction of total activity.

p. 12425, l. 2: Does your argument about difference in allocation between EDGAR and USEPA in your opinion explain the fact that EDGAR emissions for the sum of PP and IND are substantially higher that the sum in USEPA?

p. 12425, l. 4: I find it strange that the USEPA includes international shipping in their inventory. This can be checked however. According to the CEIP online database, the US reports 3780 Gg NOx in the off-road sector to EMEP for the year 2003. These data should be without international shipping.

p. 12425, l. 27: However, the models and data sources used in the US to quantify emissions of the air pollutants are not the same as those used to quantify the N2O emissions. Please reference or explain the difference.

p. 12426, l. 5: According to what you write: Klimont estimated 5 Tg (N?) for N-fertilizer

application in 1990, while Li Yu estimated 1.65 Tg N. Yan estimated 5.8 Tg N and Klimont 9.7 for the total ammonia emissions. What is your analysis of the difference between different ammonia inventories in China?

p. 12427, l. 1: for this section I recommend a table, to keep track of all the different emission numbers and references, and to possibly draw some conclusions. What is the total Bai emissions? Reference the recent findings line 9 (to Titan et al., 2001?) Reference the 9.5 and 12 Tg N numbers.

You might also like to include more information from space observation e.g.
Satellite derived trends in NO2 over the major global hotspot regions during the past decade and their inter-comparison
Author(s): Ghude SD, Van der A RJ, Beig G, et al.
Source: ENVIRONMENTAL POLLUTION Volume: 157 Issue: 6 Pages: 1873-1878 Published: JUN 2009

Adjoint inverse modeling of NOx emissions over eastern China using satellite observations of NO2 vertical column densities
Author(s): Kurokawa J, Yumimoto K, Uno I, et al.
Source: ATMOSPHERIC ENVIRONMENT Volume: 43 Issue: 11 Pages: 1878-1887 Published: APR 2009

Global distribution pattern of anthropogenic nitrogen oxide emissions: Correlation analysis of satellite measurements and model calculations
Author(s): Toenges-Schuller N, Stein O, Rohrer F, et al.
Source: JOURNAL OF GEOPHYSICAL RESEARCH-ATMOSPHERES Volume: 111 Issue: D5 Article Number: D05312 Published: MAR 11 2006

Evaluation of long-term tropospheric NO2 data obtained by GOME over East Asia

in 1996-2002
Author(s): Irie H, Sudo K, Akimoto H, et al.
Source: GEOPHYSICAL RESEARCH LETTERS Volume: 32 Issue: 11 Article Number: L11810 Published: JUN 14 2005

p. 12429, l. 17: Reference to Xing is not in ref list.

p. 12430, l. 13: Replace in country by centralized

p. 12431, l. 9: Depending on what ”robust” means. I tend to disagree. According to Rypdal et al the uncertainty in total NOx emissions are 4 times higher than in CO2. Uncertainty in sector data could certainly be higher.

p. 12431, l. 17: Repetition of what is said before. I disagree. See my comments above on this issue.

p. 12431, l. 23: What is meant by system boundaries? Difference in allocation of emissions in different sectors?

p. 12432, l. 9 and Fig 6: The “hypothesis” that the reason why N2O emissions from road transport is larger in the US than in Europe, should be rather easy to check in e.g. one of the SYNTHESIS AND ASSESSMENT REPORT ON THE GREENHOUSE GAS INVENTORIES reports available from e.g. http://unfccc.int/resource/webdocs/sai/2008.pdf

reports or national communications http://unfccc.int/national_reports/annex_i_natcom/submitted_natcom/items/3625.php

In this way the arguments can be strengthened.

p. 12433, l. 1-4: The CEIP database also contain activity data reported by countries.

p. 12433, l. 11: delete power plants.

p. 12433, l. 16: I recommend to insert information from the NAEI about the uncertainty in the UK NOx inventory. The total uncertainty is something like 8% which is much lower than the difference in inventories that you show.

p. 12433, l. 24: This may hint at a difference in allocation between these sub-sectors.

I recommend to check if this is truly the case.

p. 12434, l. 1: In summary, the difference between country total emissions between both inventories is within the uncertainty margins expected, albeit somewhat large for NOx emissions (EDGAR v4 20.9% below NAEI).

Based on what information in this paper is the expectation about what the differences in inventories should be? I do not find that Table 5 is of much help. It shows that uncertainty in total N2O is large, and larger than CO2 and CH4. Please summarize your findings to show the reader how you arrive at your conclusion.

p. 12434, l. 10: Is the EDGAR v4 more detailed than the UK NAEI? I cannot believe that.

I also think than the more than 100 sectors reported to the LRTAP convention, plus activity data, plus the reports about the inventory you mentioned is more information than what is included in EDGAR. Also all inventories are accessible online from the CEIP web site.

Also, does this sentence imply that you can recommend the EGDAR v4 for nitrogen management purposes above the other inventories?

p. 12435, l. 20: Please see my comment about a section on data sources. The references used in Fig 8 of which many of them are incomplete and not in the reference list, needs to be better explained and that is also the case in the text here. e.g. F.J. Dentener and et al., Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, Atmos. Chem. Phys. 6(2006), pp. 4321-4344 could be included for NH3 emissions year 2000 here. The EDGAR 2000

fast track data are just scaled by activity level. This should be mentioned, as it explains the trend.

p. 12435, l. 21: What does “adjusted to Olivier and Berdowski” means?

p. 12436., l25: officially reported data are available for the actual year minus 2. I do not cal that a significant time lag compared to “scientific” inventories, as you point out yourself with respect to ammonia for instance. Also, the officially reported data are reviewed, both under the UNFCCC and under the UNECE. There is to my knowledge no such procedural review of (unpublished) scientific inventories where they are often used a long time before some modeller applying these emissions starts to wonder about his results, and find that the emission data is not of sufficient quality, e.g. cross pollutant ratios do not make sense or emission are not correctly allocated in space.

p. 12437, l. 2, instead of statistics I propose country specific EFs, penetration of measures and technology

p. 12437, l. 5 Please add reference for identification of missing sources.

Also I recommend that you make it clear that the inventories reported to different international bodies and even national inventories exclude natural emissions because it is out of the scope for policy making. In my opinion, natural emission inventories should be funded over different budgets than the national inventories. It is somewhat unclear to me if you regard the natural emissions as so important for the closure of the N gap, that it should be a (first) priority. In that case I would recommend that you add to your conclusion a call upon funding for such inventory development from relevant funding organizations.

p. 12437, l. 22: Are you saying that you could not identify systematic error and gas due to too low level of detail in the inventories assessed. I do not disagree that this is the case, however, I recommend that you more strongly spell out the “sufficient evidences” you talk about as I read as flagging of sectors and pollutants for which such

investigations are needed in order to improve the level of confidence in the emission data.

p. 12437, l. 27, Please also check out Dentener et al (2006)

P. 12438, l. 7: Here it might be relevant to mention also the work undertaken by EMEP (e.g EMEP report 1/2008, http://www.emep.int/publ/reports/2008/status_report_1_2008.pdf) on the issue of spatial resolution.

p. 12439, l. 22: Trends in N2O are not shown in Figure 8 or elsewhere. Please consider a figure or a table to show the trend.

p. 12441, l. 6: I am not so sure natural emissions should be a part of the obligations under eg UNECE and UNFCCC, but these bodies could (continue to) develop methodology also for natural emissions. As stated before, I think that given that these sources are important, and might become even more important in the future, more resources should be spent on research projects concerning such emissions.

Figures and tables:

Tables 1-4 are not addressed and referenced in the text. If Tabs 1-4 are commented in the text that would probably clarify my question to p. 12421, l. 9-14

Table 1. replace consumption by combustion. Add X for NOx emissions from Agricultural mineral N fertilisation

Table 5. If NH3 and NOx is not included I find this table obsolete.

Table 6: Please make the sub total more visible in this table

Fig 1. Cyprus is shown. I disagree to conclusion mention in caption as stated above. Also seethe difference in the EMEp data you have used for ammonia 2000 and the reported data.

Fig 2. Cyprus is included. I disagree to conclusion mention in caption as stated above.

DIFF officially reported-EMEP model data NO2 Germany is -62.93582021 Gg NO2.

My conclusion is stated above.

Fig 3 is probably wrong, and not referenced in the text.

Figure 7 is not referenced in the text. EDGAR is wrongly spelt.

Figure 8: Please add N2O if possible. Check fig carefully. Reference properly the data sources. NOx as NO2? EMEP (2006) or (2008)?

Technical/editorial corrections:

P 12415, l. 13: role

P 12415, l.27 consider: The alteration in agricultural practice and increase in fossil fuel combustion have impacts human health, ...

P 12416, l. 3: What is meant by alteration of the GHG balance?

P 12416, l. 17: insert in (LRTAP) after Pollution, delete IPCC and spell out United Nation Framework Convention (UNFCCC)

P 12416 l. 27: implications for – conclusions on

p. 12417, l. 17: Explain and reference GAINS (and MITERRA)

p. 12418, l. 24: Does the document read prevent formation of NOx, or rather minimize???

p. 12420, l. 3: Fulfil

p. 12419, l. 11 and so forth: Parties not signatories.

p. 12421, l. 8: as EDGAR

p. 12421, l. 27: than delete for

p. 12429, l. 5: The direct N2O emission intensities significantly depend upon the economic situation of the region, implying potentially higher emission in the future.

P. 12433, l. 1: driven by fossil fuel combustion sources, REF. Do you intend to insert a reference?

p. 12434, l. 19. from the literature

p. 12434, l. 24. PR of China spell out People's Republic. No comprehensive or official??

p. 12435, l. 3 fig 8

p. 12435, l. 11: Provide a reference to the increase of 22%.

p. 12439, l. 5: estimated in

p. 12439, l. 24: However, the trends depicted by data from the EDGAR Hyde 1.4 project and

THOSE reported to UNFCCC. Also see my comment about the EDGAR fast track above.

p. 12440, l. 8: Vestereng et al. (2009) is probably not the correct reference here, but rather a reference for the EMEP NOx emissions.

p. 12440, l. 13: IPPC not IPCC.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 12413, 2009.