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Atmospheric Chemistry and Physics Discussions

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

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The authors would like to thank the reviewer for the comprehensive and constructive comments received. We have to our best knowledge replied to all issues raised in detail below, as well has having rewritten and revised – in parts substantially – those sections that have been identified to be lacking clarity or conclusiveness. We hope that in particular updating the datasets used to the year 2005 and using solely officially reported emissions in the EMEP inventory comparison are contributing to make our paper more useful for current scientific discussions. The section on the People's Republic of China has been significantly revised and many of the recent studies mentioned by the reviewer taken into account both in figures and in the discussion.

1. I recommend a summary table to list the databases discussed in this paper. The

table should include the name and reference of the database, representing year, containing species, as well as the emissions. Summarize three regions in separate tables, if necessary This has been addressed by adding a table with all datasets used in an ANNEX, providing an overview over the datasources.

2. Although this paper is focused on three regions, an overview of global emissions would be very helpful for understanding atmospheric N budgets. It will add the value of this review paper. I would suggest adding a section to discuss global inventories before the three regional sections. EDGAR is not the only global inventory dataset addressing reactive nitrogen species. Global datasets can be introduced in this section, and comparisons between global inventories and regional inventories can be then discussed in the following sections. A new table has been introduced (Table 2) to put the emissions (in Tg N) of the three regions into the global perspective of the literature discussed in the first part of the paper. We feel that it is beyond the scope and coverage of this paper to properly address global aspects in a sufficient level of detail, however.

3. The three regional sections were organized in different way, which makes the structure of this manuscript unclear. The differences between EDGAR data and European inventory were discussed in Sect. 2, and Sect. 3.2, but EDGAR data was not mentioned in Sect. 3.1, 3.3, and the whole Sect. 4. Emission trends were discussed in Sect. 3.2, 3.3, and 4.2, but missing in other sections. I suggest that the authors report each of three regions in a similar way. As far as possible, the revisions made have taken this into account. The different inventories available for each region – in particular for the PR of China – make it difficult however to follow through a fully consistent and equally structured format. We hope that the reviewer will find the additions and changes made useful to make the structure and discussion more clear and easier to follow.

4. As EDGAR 4 is fresh (only GHG gases are available by now) and rarely validated, and previous EDGAR dataset has been extensively used in scientific communities, I suggest that the authors also compare EDGAR 3 or EDGAR FT2000 with regional in-

ventories. It would be also interesting to see the differences between EDGAR FT2000 and EDGAR 4. This is a valid point, however, in communication with the EDGAR team during the research for this paper, we learned that the methodology and approach taken to compile EDGAR 4 has been heavily based on the previous versions of EDGAR 3.2 and prior. As scientific understanding improved and more measurements became available over time, v4 of EDGAR is notably different in some areas (substances, sectors), hence a direct comparison of different EDGAR versions would more hint at progressing scientific understanding of the methods and new EFs emerging etc. While this would be an interesting endeavour, it is outside of the scope we had for our paper and would most likely best conducted by those with innate knowledge of and access to all underlying datasets that have been used to compile the EDGAR versions.

5. NO emissions from soil are now of great concern. These emissions are usually missing from official inventories, but there are several scientific inventory studies addressing this. I recommend the authors be aware of this part because it might be significant to global and regional NOx budget. The authors are aware of this and some of the ongoing work mentioned (NitroEurope, Skiba et al., etc.) address this issue. The new Fig. 7 actually indicates that NOx emissions from soils are covered both by EDGAR v4 and EMEP for the year 2005.

6. I agree with Reviewer #1 that all the units in this manuscript should be converted to Tg or Gg N. It's difficult to me to follow the numbers with different units. All relevant figures and numbers have been converted to Gg N/Tg N as suggested.

7. P12419, L8: I am surprising that RAINS and GAINS database were not mentioned here. These two databases should be discussed in Sect. 2. In the present manuscript, RAINS dataset was mentioned in Sect. 2.2, but missing in Sect 2.3 and 2.4. The RAINS/GAINS datasets have been included in the graphs in all sections 2.x now, however it needs to be stated, that the data on which RAINS/GAINS datasets are based is not substantially different from the EMEP submissions by countries. Hence, the main focus of this paper is not to compare in detail two datasets which are based on the

same underlying data.

8. P12419, L25: EDGAR FT2000 should be mentioned here. A reference to EDGAR 3.2/FT2000 has been added.

9. P12420, L8: It should be stated that which dataset is used in the comparison (Official submissions or model use). This has been changed and clarified in the text, in addition, all datasets investigated have been updated to 2005 instead of using 2000 data.

10. P12421, L1: I would like to see a percentage difference presented here. This section was completely rewritten following the update to 2005 emissions and EMEP official country submissions only, as requested by Rev 1. And is hopefully more clear now.

11. P12421, L9-14: Here EDGAR's estimates were lower than that of EMEP. It's conflict with the previous statement of "EDGAR emissions are (much) higher for all EU27 countries". This section was completely rewritten following the update to 2005 emissions and EMEP official country submissions only, as requested by Rev 1, it is in line with the figures now.

12. P12421, L17: Again, RAINS and GAINS database should be discussed. GAINS figures for 2005 have been added to the comparison

13. P12422, L5: Please compare with IIASA data. Winiwarter W. (2005) The GAINS Model for Greenhouse Gases - Version 1.0: Nitrous Oxide (N2O) IIASA Interim Report IR-05-55 GAINS figures for 2005 have been added to the comparison, based on online-accessible datasets for NEC2007 baseline scenario.

14. P12422, L27: which inventory is discussed here? This section has been revised and rewritten, clarifiying this aspect.

15. P12423, L1-3: I would suggest adding GAINS data to this comparison. Also, compare at the sector level if possible, to make this conclusion more convincible. IIASA GAINS emissions have been added to all European figures (and China) for comparison

reasons. A new figure has been added comparing the main contributing sectors to allow for a direct comparison of EDGAR and UNFCCC data for agricultural soils and manure management to hightlight the difference.

16. P12423, L10: Which year is used? 1999, 2002, or 2005? 2005, this has been added in the beginning of the paper and for the figures to clarify.

17. P12423, L19: How the NEI estimates compared to EDGAR? We have added this comparison into section 4. Note the updated Figure 5 and additional text.

18. P12424, L7: Discussions on regional trend in NOx emissions would be interesting, e.g., Ohio River region, California, etc. . . While interesting, a complete comparison of the trends for each region of the US is outside of the scope of this paper.

19. P12424, L26: Add the following reference: Kim, S.-W., A. Heckel, G. J. Frost, A. Richter, J. Gleason, J. P. Burrows, S. McKeen, E.-Y. Hsie, C. Granier, and M. Trainer (2009), NO2 columns in the western United States observed from space and simulated by a regional chemistry model and their implications for NOx emissions, J. Geophys. Res., 114, D11301, doi:10.1029/2008JD011343. We have added this reference.

20. P12425, L14: Compare with EDGAR data. We have added this comparison into Section 4. Note the updated Figure 5 and additional text.

21. P12426 L3: The authors should revisit this section carefully. Many of recent studies were missing.

22. P12426, L5: The following inventories should be discussed in this section: EDGAR; TRACE-P inventory: Streets D. G., et al., An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, J. Geophys. Res., 108 (D21), 8809, doi:10.1029/2002JD003093, 2003. REAS inventory: Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, Atmos. Chem. Phys., 7, 4419-4444, 2007. Klimont, Z., Cofala, J., Schopp, W., Amann, M., Streets,

D.G., Ichikawa, Y., and Fujita, S.: Projections of SO2, NOx, NH3 and VOC emissions in East Asia up to 2030, Water Air Soil Pollut., 130, 193-198, 2001 TRACE-P and REAS, as well as other references quoted in Ohara et al. have been added for NOx and included in the assessment of trends as well; the Klimont et al. 2001 paper was unfortunately not available to the authors in the course of the revision, but figures from GAINS for China (online version, 2009) have been added to the trends, which are based on the findings presented in that paper.

23. P12427, L1: Almost all recent studies are missing in this section. This section should be rewritten carefully. EDGAR4; REAS inventory: Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, Atmos. Chem. Phys., 7, 4419-4444, 2007. Zhang, Q., Streets, D.G., He, K., Wang, Y., Richter, A., Burrows, J.P., Uno, I., Jang, C.J., Chen, D., Yao, Z., and Lei, Y.: NOx emission trends for China, 1995-2004: The view from the ground and the view from space, J. Geophys. Res., 112, D22306, doi:10.1029/2007JD008684, 2007. INTEX-B inventory: Zhang, Q., Streets, D. G., Carmichael, G. R., He, K., Huo, H., Kannari, A., Klimont, Z., Park, I., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L., and Yao, Z.: Asian emissions in 2006 for the NASA INTEX-B mission, Atmos. Chem. Phys. Discuss., 9, 4081-4139, 2009. Gains-China: Z. KLIMONT, J. COFALA, J. XING, W. WEI, C. ZHANG, S. WANG, J. KEJUN, P. BHANDARI, R. MATHUR, P. PUROHIT, P. RAFAJ, A. CHAMBERS, M. AMANN, J. HAO, Projections of SO2, NOx and carbonaceous aerosols emissions in Asia, TellusB, DOI: 10.1111/j.1600-0889.2009.00428.x, in press, 2009 (available online). References for Ohara et al., Zhang et al., have been added; GAINS-China reference has been added as well, but drawn from the online-accessible version of GAINS (2009). The Chen et al. 2007 reference has been viewed, but did not add more or different information to the already well covered NOx trends and hence was omitted from the graph.

24. P12427, L1: I agree with Reviewer #1 that the satellite observed NOx trends

should be discussed in this paper, not only for China, but for all three regions. Therefore, maybe this part can be placed to the Sect. 5.3. Several additional references for satellite-based NOx emission trends among the world: Richter, A., Burrows, J.P., Nüß, H., Granier, C., and Niemeier, U.: Increase in tropospheric nitrogen dioxide levels over China observed from space, Nature, 437, 129-132, 2005. Kim, S.-W., A. Heckel, S.A. McKeen, G.J. Frost, E.-Y. Hsie, M.K. Trainer, A. Richter, J.P. Burrows, S.E. Peckham, and G.A. Grell (2006), Satellite-observed U.S. power plant NOx emission reductions and their impact on air quality, Geophys. Res. Lett., 33, L22812, doi:10.1029/2006GL027749. van der A, R.J., Peters, D.H.M.U., Eskes, H., Boersma, K.F., Van Roozendael, M., De Smedt, I., and Kelder, H.M.: Detection of the trend and seasonal variation in tropospheric NO2 over China, J. Geophys. Res., 111, D12317, doi:10.1029/2005JD006594, 2006. Stavrakou, T., Müller, J.-F., Boersma, K.F., De Smedt, I., and van der A, R.J.: Assessing the distribution and growth rates of NOx emission sources by inverting a 10-year record of NO2 satellite columns, Geophys. Res. Lett., 35, L10801, doi:10.1029/2008GL033521, 2008. Konovalov, I. B., Beekmann, M., Burrows, J. P., and Richter, A.: Satellite measurement based estimates of decadal changes in European nitrogen oxides emissions, Atmos. Chem. Phys., 8, 2623-2641, 2008. As indicated before, we are aware of a wealth of information being available both regarding groundbased measurement programmes (long-term and intensive campaigns) and remote sensing data, which can be used for verification and validation of emission inventory data. However, as the amount of literature listed indicates already, a thorough and scientifically sound evaluation of these datasets and discussion in this paper would easily extend the paper substantially. Yet, this would be a relevant undertaking and would merit a paper of its own.

25. P12427, L23: Bai, 1996 and Ma and Zhou, 2000. These two references are not appropriate. They did not address NOx emission trends over China. Checked, they did not address trends, but contributed to the overall studies described in the section

26. P12428, L12: Discuss the following studies: EDGAR; Yan, X., Akimoto, H., and

Ohara, T: Estimation of nitrous oxide, nitric oxide and ammonia emissions from croplands in East, Southeast and South Asia, GBC, 9, 1080-1096, 2003 REAS inventory: Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, Atmos. Chem. Phys., 7, 4419-4444, 2007. Gu, J., Zheng, X., and Zhang, W.: Background nitrous oxide emissions from croplands in China in the year 2000, Plant and Soil, 320, 302-320, 2009. The paper by Gu et al, 2009 has been assessed due to its focus on testing a new method to estimate N2O emissions (and achieving 26-30% of total cropland N2O), does not seem to contribute additional data, as the EFs used and other parameters are in a similar way addressed by other articles already discussed. P12430, L2: NARSTO assessment report should be mentioned in this section. A sentence and reference for NARSTO 2005 has been added.

27. P12434, L13: Satellite observed trends can be discussed here. Also, add the following references: REAS inventory: Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, Atmos. Chem. Phys., 7, 4419-4444, 2007. Gains-China: Z. KLIMONT, J. COFALA, J. XING, W. WEI, C. ZHANG, S. WANG, J. KEJUN, P. BHANDARI, R. MATHUR, P. PUROHIT, P. RAFAJ, A. CHAMBERS, M. AMANN, J. HAO, Projections of SO2, NOx and carbonaceous aerosols emissions in Asia, TellusB, DOI: 10.1111/j.1600-0889.2009.00428.x, in press, 2009 (available online). This is a valid comment and has been discussed, however, we think that do to this in detail would substantially extend the papers size and add a new layer of discussion that we feel is outside of the scope of this paper. Nevertheless, the aspect of using ground-based measurements and remote sensing for the verification and validation of inventories would make a very interesting publication and merit an individual paper in itself.

28. P12438, L20: This section looks weird to me. I could not understand the means of “uncertainties” here. Uncertainties in emission projections? Also, historical emission

trends have been discussed in Sect. 5.3. This section and Sect. 5.3 should be restructured. The title was indeed not clear and has been changed to “Trend analysis”. This section aims at looking at the trends discussed in Sect. 5.3 and draw conclusions, thus not trying to reiterate the discussion, but summarizing the main issues.

29. P 12450: I suggest merging Table 2, 3, and 4 into one table. This suggestion has been taken up and a full table with all data used in the assessment combined in an overview table and placed in an ANNEX has been added.

30. Fig. 2, Fig.3 and Fig.7, add IIASA’s data. This has been done for Fig 2 and 3, for Fig. 7, where the focus is on showing the difference between inventories regarding the sectoral structure, adding a third inventory would not add significantly novel information and rather make the figure less easy to read.

31. Fig. 8, Some references are not complete, (Klimont et al., Tian et al.). Re-draw this figure after including the missing studies mentioned above. The references have been completed and the figure redrawn.

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