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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Key chemical NO_x sink uncertainties and how they influence top-down emissions of nitrogen oxides" *by* T. Stavrakou et al.

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We would like to thank the reviewer for his/her positive evaluation of our work. In the following, we address the concerns raised. Reviewer's comments are *italicized*.

This manuscript is a well-conceived and executed investigation of the impact of uncertainties in kinetic parameters and OH levels on top-down constraints on NOx emissions. The authors acknowledge that there are several additional factors that may influence the sinks of nitrogen oxides in the atmosphere, but restrict their analysis to four important factors. In my opinion, the main achievement of the manuscript is to highlight

the sensitivity of the inversion framework to known uncertainties related to NOx sinks. While the authors do carry out model inversions using scenarios with extreme (but plausible) combinations of NOx sinks and compare the optimized NOx sources to the prior estimates, the focus remains on the sensitivity. As the authors point out, the only anthropogenic source region for which the optimized source changes in a consistent direction, regardless of the sink parameterization, is in China. Interestingly, both scenarios also suggest upward revisions for the fire and soil sources of NOx. A recent publication (Lin et al., 2012) explores model sensitivity of nitrogen oxides to a wider range of factors, including meteorological parameters, but using a higher resolution model for a specific region. Given that the current manuscript is carrying out a global analysis and is more directly examining the impacts on the inversion framework, I think the focus on the factors identified by the authors is appropriate and well-motivated in the text. The paper is appropriate for publication in ACP after consideration of the following points.

Specific comments:

1. *I don't follow the logic about chemical feedbacks (P 7891 L 6-10, and P 7893 L 19-25. In the MAXLOSS scenario, why would there by higher levels of NOx in the atmosphere? I can see why increasing the sink would also require increasing the emissions in the inversion framework, but not to the point where the mixing ratios are higher. Please clarify if NOx levels are actually higher in the tropics in the MAXLOSS scenario.*

   We did not mean that NOx levels are higher in the MAXLOSS scenario; but the NOx increment is higher in MAXLOSS than in MINLOSS, relative to the a priori. We provide the following, more detailed explanation regarding the role of chemical feedbacks (Section 5, 4th paragraph):

   "In most tropospheric conditions, a NOx emission increase leads to an increase in OH (through the HO_2+NO $\rightarrow$ OH+NO_2 reaction) and therefore in the NOx sink

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rate. In the context of inverse modelling, this implies that larger emission increments are required in order to match the observations, to compensate for this negative chemical feedback, compared to an emission inversion neglecting feedbacks. Since the NOx emission increments are larger in MAXLOSS compared to MINLOSS, the NOx lifetime decreases (relative to the a priori) are also larger in MAXLOSS compared to MINLOSS, which amplifies the differences between the emission increments in MAXLOSS and MINLOSS."

Over very polluted areas such as Northestern China, characterized by very high NOx levels, this chemical feedback does not operate, because OH levels do not increase much with increasing NOx. In fact, at sufficiently high NOx concentrations, further NOx increases lead to OH levels decreases, due to the lower HO_2/OH ratio and to the growing importance of the OH+NO_2 reaction as a sink for HOx.

2. *P 7884 L 1-15 It would be useful to know what CTM DOMINO v2 uses to determine a priori NO_2 vertical profiles, and how errors in the NOx sinks in this CTM would feed back into the retrieval. In particular, how would the comparisons in Section 5 change if the MINLOSS and MAXLOSS parameters had been used in the CTM that was used in the DOMINO retrieval. Would this make the comparison more internally consistent?*

   The DOMINO v2 algorithm used the model TM4 (Boersma et al., 2011). Differences in NO_2 vertical profiles between IMAGES (using either MAXLOSS or MINLOSS emissions) and TM4 are taken into account through the use of averaging kernels in the comparisons.

3. *P 7891 L 4 - The discrepancies are largest for natural sources only in a relative sense. For example the absolute change optimized anthropogenic emissions for MAXLOSS is larger than for lightning.*

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Agreed. The text has been corrected: “In a relative sense, the largest discrepancies..."

Technical corrections:

1. *P 7873, L 2, “comforting" should be replaced with “consistent with"*

   Corrected as suggested.

2. *P 7892 L 10-11, suggest rewording “Should this be confirmed. . . 1Tg N." as “This lower estimate for NO production efficiency translates to an annual lightning NOx source of 1 Tg N.*

   Corrected as suggested.

3. *P 7892 L24-26, suggest rewording “Exception is made. . . a priori" as “In contrast, both the MINLOSS and MAXLOSS inversions result in an increase in anthropogenic emissions from China to 5.8 and 6.5 Tg N, respectively, significantly higher than the prior of 4.8 Tg N".*

   Due to important model updates compared to the ACPD version of this manuscript (see our reply to Reviewer 1), the optimized anthropogenic emissions over China differ markedly between the MINLOSS and MAXLOSS inversions. The text has been adapted accordingly.

4. *P 7895 L 9-11, suggest rewording to “Comparisons above selected regions between SCIAMACHY and modelled NO_2 columns..."*

   Corrected as suggested.

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Boersma, K. F., H. J. Eskes, R. J. Dirksen, R. J. van der A, J. P. Veefkind, P. Stammes, V. Huijnen, Q. L. Kleipool, M. Sneep, J. Claas, J. Leitao, A. Richter, Y. Zhou, and D. Brunner : An improved retrieval of tropospheric NO_2 columns from the Ozone Monitoring Instrument, Atmos. Meas. Tech., 4, 1905–1928, 2011.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 7871, 2013.