

The paper by Stavrou et al. evaluates uncertainties in several chemical parameters affecting the simulation of NO₂ in linking satellite NO₂ data to NO_x emissions. The paper further compares the modeled NO₂ and retrieved NO_x emissions with aircraft data, satellite NO₂ data and bottom-up emission inventories. The analysis, however, has various issues that prevent its publication in the current form.

Model uncertainties have been evaluated by Lin et al. (2012) in a comprehensive manner for China. Lin et al. examined (almost) all chemical and meteorological parameters/processes that are important to NO₂ simulations at a fine resolution (0.5 degree x 0.667 degree) using GEOS-Chem, including chemical parameters analyzed by the current paper and those not evaluated by the current paper. The current paper extends the analysis for a global simulation (IMAGE), but only evaluating a subset of chemical parameters by Lin et al. and only on a relatively coarse resolution (2x2.5 degree). Therefore, the current paper should state clearly how it builds upon Lin et al. throughout the text (abstract, introduction, Sect. 2, results, conclusion). More specifically,

1. The paper focuses on OH+NO₂ reaction rate, NO+HO₂->HNO₃, N₂O₅ uptake on aerosols and OH regeneration from isoprene chemistry. However, other chemical processes are important too, including the HO₂ uptake on aerosols, anthropogenic aromatics, organic (isoprene) nitrates, PAN formation and thermal-decomposition, and emissions of CO and VOC affecting OH. The importance of these processes has been found for various months and regions by Lin et al. For example, HO₂ uptake is critical for all seasons, anthropogenic aromatics is important for winter, isoprene nitrates are important for summer in vegetated areas. The paper should better justify the choice of not including these additional chemical uncertainties and their potential consequences.
2. The role of aerosols is important because aerosols affect both radiation and uptake of HO₂ and N₂O₅. There are no analyses of model aerosol performance for the reader to judge the validity of the model sensitivity to N₂O₅ and HO₂ uptake rate.
3. Much larger HO₂ uptake rate on aerosols than previously thought is recently established, due to the findings of significant transition metal ions in aerosols catalyzing the uptake. HO₂ uptake on aerosols is found to be extremely important for OH, ozone and NO₂ (Mao et al., 2012, ACP; Lin et al., 2012). The current paper appears to suggest the opposite without clear explanation and comparison. Is it because of low aerosol loadings or other model issues? It is very hard to believe the HO₂ uptake is not important. Macintyre and Evans (2011) parameterization does not include the effect of transition metal ions. However, they did show significant effects on HO_x and ozone when the HO₂ uptake rate reduces from 0.2 to 0, and they further recognized the potential importance of transition metal catalysis in the conclusion.
4. Model meteorology is also important. For example, Lin et al. found errors in PBL mixing, cloud optical depth, and water vapor to be very important for their GEOS-Chem simulation. How about IMAGE? Although the current paper tries to focus on chemical uncertainties, errors in IMAGE met fields may have important effects on the model performance and NO_x emission inversion, and may affect to some extent the sensitivity of model NO₂ to chemical uncertainties (e.g., relative humidity affects aerosol scattering). This is critical since the current paper appears to suggest an upper/lower bound of model NO₂ and NO_x emission

inversion in comparison with aircraft/satellite/emission inventories. The potential effect of met field errors should be discussed more clearly.

5. How about the effect of model resolution affecting the nonlinear chemistry?
6. Sensitivity to model chemistry/meteorology uncertainties may be model dependent. For example, if the model only produces small amounts of aerosols, the effect of HO₂/N₂O₅ uptake rate would be less important. The importance of isoprene nitrate depends on the assumed yield and recycling ratio. The met field errors may be highly model and region dependent.
7. There are many over-statements without justifications throughout the paper. For example, 'four key parameters believed to be of primary importance' and 'largely overlooked in previous inversion exercises' in the abstract. These statements are against the fact that Lin et al. have done lots of work in this regard and that they found many other parameters to be important as well.
8. Section 2 provides a useful analysis of the parameters being investigated. However, it should be made more explicitly how the choice of these parameters is built upon Lin et al.
9. The current paper should state clearly how it proceeds beyond Lin et al. throughout the text (abstract, introduction, Sect. 2, results, conclusion).

Model simulations and inversion procedures are not clearly defined in the current paper. More specifically,

1. What is the PBL scheme? What is the convection scheme? How about aerosol simulations? How about aromatics? How about the vertical distribution of lightning emissions in comparison with recent development by Ott et al. (2010)? The vertical distribution is very important since the averaging kernel depends on height. The authors should describe/analyze these model aspects.
2. The time step of 4 hours raises questions about the model performance, particularly when the lifetime of NO_x in midday is shorter than 4 hours and that the radiation (and photochemistry) changes rapidly during a 4-hour period. The authors should justify the choice of the particular time step.
3. The use of OMI data is not clearly described. How was the row anomaly treated? What do you mean by 'cloud free'? Did you match OMI data with model for each day before obtaining the monthly mean? How to deal with gridding of averaging kernels to model gridcells? What is 'super-observation'? Same issues for comparison with SCIAMACHY data.
4. Furthermore, quantitative uncertainty values in emission sources are given in Sect. 3 without justification. These values are also inconsistent with Stavrakou et al. (2008) without further explanations of the change. The error values are very important because they determine whether the 'optimized' emissions are closer to top-down emissions or to a priori emissions, with further consequences in comparison with other emission inventories.
5. The importance of individual chemical uncertainties may be different before and after the averaging kernel is applied, because these uncertainties may affect the vertical distribution of NO₂.

Section 4 and Figs. 2,3. The analysis of the effects of chemical uncertainties is unclear/incorrect. For example, how does increased OH+NO₂ reaction rate (from Henderson et al. to Mollner et al.

and to JPL) result in little change/reduction in NO₂ loss rate in most regions? How does additional NO+NO₂->HNO₃ result in little change/reduction of NO₂ loss rate in East U.S., EU and E. China? How does increased N₂O₅ uptake rate (from Brown et al. to DAVIS) lead to reduction in NO₂ loss rate in several continental and oceanic regions and little change/reduction in North China in January? How does the OH regeneration from isoprene chemistry not have effects on NO₂ loss rate in vegetated EU and S. China? How does HO₂ uptake on aerosols ($\gamma = 0.2$) not have large impacts on NO₂ even in those areas with high aerosol loadings? In any case, the section should be expanded with more analyses over the polluted areas, and the results from all these chemical sensitivity tests should be compared with Lin et al. and differences should be explained. Also, the color scheme in Figs2-3 is confusing. It is much better to use warm colors for positive values and cool colors for negative values, and to separate negative from positive values.

The lifetime analyzed in most of the text and in Fig. 4 and Table 1 is somehow misleading. It represents the average lifetime of NO_x throughout the day rather than the 'local' lifetime for early afternoon which is more relevant to linking OMI NO₂ with NO_x emissions. In addition, the impacts of chemical uncertainties on the average lifetime (sink rate) shown in Fig. 4 and Table 1 are not the same as the impacts on the local lifetime (sink rate) relevant to early afternoon NO₂. For example, the effect of N₂O₅ uptake is much less important for early afternoon NO₂ than NO₂ at night. Also, in Fig. 4, the lifetime are longer than 6 hours, as opposed to the lifetime shown in Fig. 13 (i.e., less than 6 hours).

The definition of lifetime should be made clear throughout the text, i.e., daily average lifetime or daytime lifetime or local lifetime (most relevant to OMI NO₂). The discussion of lifetime is quite confusing currently.

The current paper compares model NO₂, model lifetime and retrieved NO_x emissions with other datasets. While these comparisons are interesting to some extent, they are complicated by the model/retrieval issues raised above. More comments specific to individual comparisons are as below:

1. The good comparison of model NO₂ (driven by 'optimized' NO_x emissions) with OMI data appears to suggest that the emission retrieval simply finds NO_x emissions that best fit OMI data in the first place. This is a result of much larger biases assumed for a priori emissions than the biases assumed for the model-retrieval system -- in this case, the 'optimized' NO_x emissions simply reflect the 'top-down' emissions. It is not surprising that using such emissions, model can reproduce satellite data, especially at the coarse resolution.
2. The comparison with SCIAMACHY does not tell much about the model performance nor about emission inversion. This is because satellite data are always subject to errors and thus do not represent the true state of atmosphere. In addition, SCIAMACHY is further subject to data coverage such that the data representativeness is an issue.
3. The comparison with bottom-up inventories is not a good constraint of model/inversion either. It is well known that bottom-up inventories contain large uncertainties, which is the primary reason of the efforts to developing top-down emission constraint.
4. The comparison with aircraft data is potentially useful for model constraint. However, inadequate information is provided to justify the comparison. For example, is the mean or

median of aircraft measurements used? This makes a huge difference. In addition, these aircraft measurements do not sample 0-500m above the ground. This causes a lot of problems interpreting the model-aircraft comparison for NO₂ which is concentrated near the ground. Furthermore, the comparison should focus on the lowest 2-3 km at higher vertical resolution. The current 1-km resolution may skew the comparison especially considering the coarse model vertical resolution. Only on a high vertical resolution can the model PBL mixing be evaluated properly.

5. The comparison of lifetime with Beirle et al. (2011) should be interpreted with caution. Beirle et al. shows the local (early afternoon) lifetime corresponding to the OMI NO₂ data, on the local spatial scale. The model lifetime is however for the 2x3 degree resolution. Such differences should be made explicit and discussed, especially in light of Valin et al. (2012) showing the dependence of NO_x lifetime on resolution.

Table 2. The total source is not equal to sink. The authors explain the difference (1.9 – 5.4 Tg) as the effect of HNO₃+OH. This may not be true given the low oxidation rate. Please double check. In any case, this imbalance should be clarified in a more visible place.

Page 2893, L20. The explanation for the weak sensitivity of top-down emissions to NO_x sink rates is unclear. I suspect these results are related to the strange results in Sect. 4 and Fig. 2-3 (see above point).

More specific comments:

P7885. Last paragraph. Descriptions of natural emissions could be placed in a separate paragraph.

P7886, L1. Discussion on aerosols is out of place. Should place it somewhere else. Analyses of aerosol simulations should be largely extended.

P7889, L5, MOLLNER has weaker T dependence than Henderson et al.

In several places, the test on HO₂ uptake is forgotten to mention.

Fig. 10. Quantitative errors in SCIMACHY are not described here nor in the text.