Review of "Model Uncertainties affecting satellite-based inverse modeling of nitrogen oxides emissions and implications for surface ozone simulation"

By J.T Lin et al.

General Comments

The authors attempt to improve the agreement of NO_2 column simulated with GEOS-Chem with OMI observations over East Asia. The current study is motivated by a particularly large under prediction of NO_2 columns over polluted regions in China. NO_2 columns are simulated for a range of model parameters to find the localized linear response of NO_2 column to each parameter. Finally simulated columns are modified based on a selected set of parameters and values according to the linear response determined. The authors select a reasonable range of uncertainties. For chemical parameters these ranges are based on recent laboratory and field studies. For meteorological parameters they are largely based on satellite and monitoring networks for meteorological parameters.

The discussion of the uncertainty of simulated NO_2 over a range of model parameters is interesting, but the paper attempts to do more than that. The paper in its current form reads in part as a review, in part an uncertainty analysis, and in part an attempt at a novel means of improving simulated NO_2. I highly recommend removing discussion of ozone chemistry (very little attention is given to it any way), removing discussion of the localized linear response and focusing fully on the uncertainty analysis. I suggest this paper for publication only if the authors respond to the comments above and the more specific comments that follow.

Specific Comments

I. Introduction

P. 14273 L 12; "Even for areas…" to the end of the paragraph seems unnecessary and contrary to the rest of the study. Isn't the purpose of this paper to see how much of a model-satellite discrepancy could be due to parameters other than NO_x emissions?

P 14274 ¶1 The bulk of this paragraph should be moved to section 3

II. Ground and Space measurements

P. 14275 ¶1+2 Are the data filtered for clear-sky conditions? What are biases in these datasets? Is there reason to believe them over GEOS-Chem? Do you compare these datasets or GEOS to surface measurements (e.g., dew point)

P. 14275 ¶3 Please briefly state how NO_2 column is processed (cloud fraction, viewing zenith angle, data quality flags, etc.). For the uninitiated please state basic instrument characteristics (time of day, pixel size, repeats, etc.)

P. 14275 ¶4 What is meant by valid data?

III. GEOS-Chem simulation and comparison with OMI retreivals

Please include a figure that shows simulated and observed NO2, AOD, COD at the product resolution, not just at meteorological stations in a figure. In addition, I would have benefitted in the discussion from a figure of NO_x sinks (see sect. 5 comments)

Throughout the paper, the authors refer to northwest China etc. Please either qualify these descriptions with adjectives (e.g., remote northwest China or urbanized eastern china) or designate 3-4 regional categories in a figure, perhaps the one mentioned above.

IV. Sensitivity of GEOS-Chem simulations to meteorological parameters

In general, many of the effects presented here and in section 5 are relatively uniform across the domain, rather modest and well known (e.g., water vapor). For parameters of these types, I feel the flow of the paper would benefit from fewer figures and a more succinct summary, with a table entry for the range of percentage change.

Also, Please clarify that what parameters independently vary and what do not. (e.g. Sect. 4.1 –Water vapor does not change with temperature in your simulation, but biogenic emissions do)

Heading - Please include "NO_2 column" somewhere in the title.

Sect 4.3 I don't believe sensitivity tests of cloud optical depths are pertinent for NO2 column observed in clear skies unless you have filtered model observations for 1PM clear-sky conditions.

Sect 4.4 Please separately identify effects of NO_2 vertical distribution versus changes in mass due to nonlinear chemistry with regards to PBL analysis.

Sect 4.5 Briefly state where lightning NO_x is most important. Would it affect the remote regions strongly?

Sect 4.5 There is no discussion of current results for lightning, only review. Please include some discussion or remove from analysis.

Sect 4.5 Is northeast China a good location to study lightning NO_x emissions? Are there better, more remote regions with comparable lightning activity? It seems to me that any analysis would be flawed by the affect of boundary layer transport to altitude.

V. Sensitivity of GEOS-Chem simulations to chemical parameters

Please include "NO_2 column" somewhere in the heading title.

Sect 5.1 – I am surprised that the increase of NO_2 across the domain is uniform for a 30% decrease in k_{NO2+OH}. I have found that NO_2 simulated in NO_x suppressed regions does not depend on the rate constant. When NO_x dominates OH reactivity, OH should increase for any decrease in the rate constant, and the product of $k_{NO2+OH}[NO_2][OH]$ should be nearly constant. Please check this result.

Sect 5.3 – Heading should include Isoprene, OH-recycling, PAN and alkyl nitrates

Sect 5.3 – The following combination of findings surprises me.

A 40% increase of OH only leads to a 0-4% decrease of NO_2 column.

No isoprene-OH chemistry results in a 40-50% increase of NO_2 and a 100% increase of OH

What fraction of NO_2 sinks is OH, AN, and PANs?

What happens to isoprene+NO_3 at night?

-- I suggest adding a figure or pie chart in the beginning of the manuscript that shows simulated HNO_3, alkly nitrates, and PANs to provide overview of the various NO_x sinks.

Sect 5.4 – Please state the regional representativeness of the observed Cu aerosol mass fractions reported.

Sect 5.6 Heading, Please change heading title to "Emissions of non-NO_x species"

Sect 5.6 – Propene is not a good proxy for aromatics because of differences in alkyl nitrate formation potential and possible differences in PAN formation. Regardless, the effect seems very small.

Sect 5.7 – Please state a range for the magnitude and the sign of the resolution-dependent biases predicted over urban centers by the Valin et al (2011) modeling study.

What happens for a 50%, 100% increase of NO_x emissions? Many of these parameters tested here depend nonlinearly on NO_x concentration. Please comment on their effects at NO_x concentrations that are more pertinent to what is observed.

VI. Modifying model NO_2 columns accounting for errors in meteorology and chemistry

As mentioned previously, I don't think this section is needed. A simple table of uncertainties for each parameter and scatter plot of simulation and observation are sufficient.

The authors should justify why HO_2 + NO was not selected for this analysis.

Sect 6 –P 14294 L 3. and L4 . Please remove "best estimate"

VII. Implications for surface ozone.

I feel this section is beyond the scope of this paper and no observations are presented for comparison.

VIII. Conclusions

Please comment on the likelihood that the difference between model and observation is simply due to an underestimate of emissions in the context of the range of model errors presented.

Technical comments:

Repeated Instances:

-Please say increase and decrease by 30% instead of scaled by 130% in many examples throughout the manuscript. Also, please check that all captions match the image shown (e.g. Fig. 7 NO_2 increases but the caption says the NO_2 removal rate constant is scaled 130%)

-Readability would be improved by reducing the number of works cited and using (e.g., …) formalism.