

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 used the GEOS-Chem model to investigate the uncertainties associated with both meteorology and chemistry in simulating tropospheric NO₂ column and surface ozone. Authors identified several important meteorological fields and chemical processes that may affect the simulated NO₂ column and conducted and analyzed multiple sensitivity simulations by perturbing the selected parameters within the reasonable ranges to their best knowledge. They quantified errors in simulated NO₂ columns associated with the uncertainties of 10 selected parameters. The uncertainty in simulating NO₂ column could have implication for uncertainty of top-down NO_x emission estimate using inverse modeling methodology based on GEOS-Chem model and satellite retrievals.

I find this topic about uncertainty quantification is interesting and important. The uncertainty quantified could be used to improve the corresponding model processes and also used for top-down emission inversion. It highlights the importance to understand model biases before deriving top-down emissions with traditional inversion methodology. However, this paper is not well presented. I do have some specific comments and questions about the results as listed below. I would only suggest this paper for publication if the authors address these comments.

Specific Comments

1. The title is not appropriate. This study focuses on the uncertainties in simulating NO₂ columns instead of inverse modeling although it has implication for inverse modeling studies. The current title is misleading. In addition, the model uncertainty is always related to a specific model, although it may have implication for other models. Therefore, the title should be something like “modeling uncertainties of tropospheric NO₂ columns in GEOS-Chem”.

2. In the abstract, authors suggest “a possible systematic model bias such that the top-down emissions will be overestimated by the same magnitudes if the model is used for emission inversion without corrections”. I don’t agree with this. In the inverse modeling methodology (e.g., Martin et al., 2003; Zhao and Wang, 2009), the model error is accounted for, therefore, as long as the model error is estimated correctly, the inversed emission is still valid.
3. In section 2.1, the meteorological parameters investigated in this study are only compared with surface measurements to estimate uncertainties. It has limit, since some parameters such as RH may have significant vertical gradient. Please discuss it.
4. One of my major concerns about tuning meteorological parameters off-line in GEOS-Chem is that offline tuning may cause meteorological inconsistency, since all these meteorological parameters are not independent. For example, authors tuned surface air temperature in section 4.1, it actually very likely affects the PBLH, water vapor and RH, which are fixed by authors artificially. However, authors again tuned water vapor and PBLH in other sensitivity simulations. Therefore, the NO₂ sensitivities due to these tunings should not be added linearly as that authors formulated (equation 1) in section 6. The interaction effect among parameters should be accounted to correctly estimate the overall errors of model.
5. In section 2.3, MODIS AOD is known having some problems over land and overestimated compared with MISR. It’s better to evaluate model aerosol simulation using MISR instead of MODIS over land.
6. In section 4, line 3 of page 8, you may not want to say that precipitation is not important for simulating NO_x, since precipitation is important for wet scavenging of HNO₃ and aerosols.
7. Line 29 of page 10, please give the reference that GEOS-5 simulated PBLH is overestimated in the daytime. Is your conclusion of GEOS-5 biases based on studies over the US? Can you apply it over China? Any difference between the US and China?
8. In section 4.5, lightning in GEOS-Chem is parameterized based on the lightning observations over the U.S. and it’s very sensitive to regions and years. Authors should be very careful when discussing GEOS-Chem simulated lightning NO_x over East

Asia unless they can constrain GEOS-Chem lightning production over East Asia using observations.

9. When authors discuss the relative importance of uncertain parameters, it should be pointed out that the relative importance of parameters are related to their uncertain ranges assigned by authors. The uncertain ranges are also quite uncertain. So please discuss it.
10. One of the important points of this study is to estimate the overall uncertainty of GEOS-Chem in simulating NO₂ column, which can be used for future inverse modeling study using GEOS-Chem. However, authors didn't report an overall error based on their estimate. How to use the estimated model uncertainties for top-down inversion? Martin et al. [2003] estimated 30% relative error of GEOS-Chem simulated NO₂ column. How is it compared with authors' estimate? Which uncertainty value should people use for future inverse modeling using GEOS-Chem?

Technical Comments

1. Line 30 of page 24, "about 0-20%" to "up to 20%".
2. Figure 1 is not necessary to be included in this paper. It's not well described and discussed.
3. Remove figure 13-16, it's better to just summarize the statistical metrics from all these figures in a table. The figures are not very informative. This paper includes too many figures and some of them are not in good quality, such as Figure 5 and 7. The contour colors can be improved to have less blue.
4. In Figure 2, why is R_square not improved in the modified model? Does it mean that modification doesn't improve model in terms of capturing variance of satellite retrievals?