

Interactive comment on “Improving NO₂ and ozone simulations through global NO_x emission inversions” by Zhen Qu et al.

Anonymous Referee #1

Received and published: 29 April 2020

Review of ‘Improving NO₂ and ozone simulations through global NO_x emission inversions’ by Zhen Qu et al.

General comments

Qu et al. have studied the impact of top-down NO_x emission estimates derived from two OMI NO₂ satellite data sets (NASA SP v3 and DOMINO v2) on NO₂ and O₃ simulations with the GEOS-Chem model. Previous work already showed (e.g. Verstraeten et al. [2015], studies by Miyazaki et al.) that O₃ in the troposphere is generally better understood when NO_x emissions are derived from satellite NO₂ data than when taken from emission inventories.

Here, Qu et al. find substantial differences in the agreement of NO₂ and O₃ simulations

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against independent measurements depending on whether data set NASA or data set DOMINO is used. This was to be expected given that it is well-known that the NASA and DOMINO datasets have considerable differences. A useful aspect of the study is that the authors now quantify the consequences of these differences, which is relevant because satellite data is increasingly used to improve model understanding of atmospheric composition.

What is disappointing however is that we do not learn much new. Simulations with the NASA emissions compare better to some metrics, and worse to others, but the authors do not explain why. This makes the manuscript a technical document, where it is left to the reader to figure out what emissions could work best for his/her particular purpose, without actual guidance on why that would be. The authors should do more to investigate why using one dataset leads to better agreement e.g. for surface O₃ at remote sites, and the other for polluted sites. Aspects of spatial resolution, temporal representativeness, and vertical sensitivity should be taken into account when providing this guidance to potential users.

Another criticism is that the chain of technicalities is very long and that the experiments are set-up in a sub-optimal manner (for example comparing 2.5 degree simulations of surface NO₂ to surface stations that are representative for much smaller domains). A major concern I have is with the lack of detail and clarity on how the adjoint incorporates the information from the satellite retrievals. From the manuscript I first suspected that monthly mean column NO₂ data was simply used to estimate the emissions, suggesting that the highly variable and non-linear vertical sensitivities of the retrievals have not been used to interface the model with the satellite data. There are various studies pointing out how crucial it is to account for the vertical sensitivity of the NO₂ retrievals, e.g. Miyazaki et al. [2017], Boersma et al. [2016] to name a few. Then I read the supplementary material and there the impression was given that at least the a priori profile shapes are made consistent between the NASA and DOMINO retrievals, but it remains unclear to what extent this has harmonized the data, and to what extent

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vertical sensitivities between the two datasets are still fundamentally different.

Specific comments

P2, L42-43: the formation depends not only on the local NO_x and VOC concentrations, but also on the radiative regime in which these occur.

P2, L65: different → differ

P3, L72: import → importer

P3, L78-81: Zhang et al. [2008] and Verstraeten et al. [2015] already showed that through optimizing NO_x emissions in China, the simulated O₃ over the Pacific and over the western US indeed improved.

Section 2.1 It is unclear in this manuscript how the adjoint accounts for (a) vertical sensitivity of the satellite retrievals, and (b) the diurnal cycle of NO_x emissions. These aspects are important enough to describe in the manuscript, for (a) useful information is provided in the supplement but it is not clear whether the replacing of the a priori profiles by GEOS-Chem prior profiles was also applied in the research to estimate the emissions. The authors should clarify this in section 2, and also briefly quantify to what extent the differences in prior simulations have been minimized by this approach. For (b), some info is given but only late in the game (P7: The diurnal variations of NO_x emission are constrained to be those of the prior emissions), and we do not learn what the diurnal cycle is in the first place. Please revise section 2 thoroughly with this in mind.

Then I have other questions:

- how does the adjoint approach account for other relevant aspects of data assimilation?
- how is the OMI data averaged spatially to the grid of GEOS-Chem, and how are superobservation errors incorporated?

- did the authors only use the mostly cloud-free OMI retrievals?

Section 2.2: OMI is suffering from the so-called row anomaly, which was absent until mid-2007, and then became gradually more important. How did the authors ensure that the growing impact of the row anomaly did not unduly affect their trends in NO_x emissions?

Section 2.3: it remains unclear what type of surface station was used for the GEOS-Chem surface evaluation. Using urban background and regional stations seems appropriate to evaluate the large GC grid cells, but urban street stations should be excluded.

P5, L152-154: what explains the OMI-driven differences between the posterior NO_x emissions, differences in tropospheric slant columns or in the AMFs? Presumably the latter, but since the a priori profile differences have been “minimized”, the differences must be in the assumptions on surface albedo and clouds. It would be best if the authors could shed more light on how the scattering weights or averaging kernels are different between the OMI NO₂ retrievals. Please clarify.

P6, L173-174: the statement that “NO₂ column simulations at 2° × 2.5° in this study are likely to be underestimated and lead to high biases of posterior NO_x emissions to match satellite NO₂ column concentrations” needs more evidence. The hypothesis that instant dilution leads to too much OH (by Valin et al. [2011]) may be valid for isolated NO_x sources in otherwise pristine areas, but instant dilution of NO_x emissions situated in high-background NO₂ regions such as the eastern US or western Europe is probably of less concern.

P6, L193: what is the magnitude of the correction factors over China and the US? How do they vary by season?

P7, L195-199: this part is rather inconclusive. The GEOS-Chem simulations have been corrected for resolution (an increase) and surface measurements have been corrected down for molybdenum interference, and still GEOS-Chem with posterior emissions is

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biased low by 20%-50%. What explains the persistent low bias?

P7, L224-225: OMI measurements frequently miss the high values of NO₂ column densities that occur before or after its overpassing time. OMI was never designed to measure NO₂ before or after its overpass time, so to say that OMI misses these high values is misleading. Please rephrase.

P7, L226: twice-per-day constraints on NO_x emissions have been achieved in earlier studies based on SCIAMACHY + OMI (Boersma et al. [2008], GOME-2 + OMI [Lin et al., 2011], including via sophisticated assimilation schemes [Miyazaki et al., 2017].

P8, L237: the June peak in NO₂ over China can be easily traced back to crop residue burning in that month – e.g. Stavrou et al. [2016].

P8, L238-240: can you explain more why the DOMINO product would be more sensitive to soil NO_x emissions? It's not because of the different a priori profiles assumed in the NASA and DOMINO retrievals?

P8, L243-244: please see my previous comment. The authors seem to know something very interesting here, but they don't show it. Is there any evidence that one retrieval would be more sensitive to NO_x sources than the other? That would be extremely relevant to know more about. Since the satellite measurements are identical for the NASA and OMI retrievals, it must have to do with AMF differences, see e.g. Lorente et al. [2017]. But what drives the apparent difference in sensitivity – albedo, cloud fraction, cloud pressure?

P8, L246-256: Figure 5 – the daytime O₃ simulations in China all seem strongly low biased relative to the observations. The other ozone metrics in China and all in the US match much better. Why is this?

P9, L271-272: “also not reflected”?

P9, L276: no reduction of NO_x emissions in Europe? This is strange – NO₂ tropospheric columns are decreasing over Europe, and Miyazaki et al. [2017] showed reduc-

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tions in for NO_x emissions. Overall, Figure 6 looks very odd to me. DOMINO NO₂ columns are 40% higher than NASA, but the NO_x emissions inferred from DOMINO are more than 40% higher than the emissions inferred with NASA (L278-281). Also, Miyazaki et al. [2017] (Figure 9) still find reductions in NO_x emissions over Europe between 2005 and 2014 based on the same DOMINO data, so how can you find increases? Please clarify.

P10, L295-297: I'm missing an explanation or hypothesis why NO_x emissions from one dataset would do better than the other for different ozone metrics.

P10, L304 and L315: please clarify how the impact of meteorology and non-NO_x sources on O₃ changes was evaluated.

L306-307: "The trends of simulated MDA8 ozone are similar when using the NASA and the DOMINO posterior NO_x emissions as inputs" – yes, but please also explain why the magnitude of the NASA-derived MDA8 O₃ levels are biased high then.

P11, L332-333: the prior simulated O₃ profiles in Figure 10 agree much better with the O₃ sondes between 800-400 hPa than the assimilated profiles. I don't understand why that is, since the effect of the updated NO_x emissions should be mostly felt in the lower 2 kms of the atmosphere. Or is this the impact of changes in background O₃ in response to changing Asian emissions?

P13, L394-395: one important difference between this research and the work done by Miyazaki in a number of papers, is that the latter assimilates also other species relevant for NO_x inversions and O₃ simulations (e.g. CO, HNO₃, SO₂). It would be interesting to also discuss to what extent these additional constraints can help explain the "remaining differences between simulated and measured ozone".

P13, L398-400: the statement "Both OMI NO₂ retrievals employed in this study use NO₂ vertical shape factors from coarse resolution simulations, and therefore are biased low compared to in-situ measurements [Goldberg et al., 2017]." Brought up the

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question (again) whether both OMI NO₂ retrievals are at least consistent now in their use of the same coarse-resolution vertical shape factors (i.e. those from GEOS-Chem).

P13, L401: “retrievals also have not explicitly accounted for the aerosol optical effects, which are demonstrated to degrade the accuracy of NO₂ column concentrations”. This is an overstatement. Only when AOD is very high (>0.5-1.0) there are indications that implicit corrections break down. Even in Liu et al. [2019] accounting for AOD did not solve the low bias in tropospheric NO₂ which was not apparent in the DOMINO scheme without an explicit aerosol correction.

References

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