Review #2

In "A top-down assessment using OMI NO2 suggests an underestimate in the NOx emissions inventory in Seoul, South Korea during KORUS-AQ," the authors combine two lines of research by 1) adjusting space-based retrievals of tropospheric NO2 columns with spatially refined model data and then 2) estimating NOx emissions, NOx lifetime and background tropospheric NOx by applying an Exponentially Modified Gaussian fit to the resulting NO2 column field. The authors test their method using model results that had been generate for forecasting purposes (which they determine to be successful by comparing top-down estimate to a bottom-up integration of emissions within what seems to be an arbitrary 40 km radius of Seoul).

√In general, the paper is well-written and is relevant to Atmospheric Chemistry and Physics.

Thank you for your comments; they have substantially improved our manuscript.

√I have two major concerns. The authors ignore impacts of topography and local circulations on the spatial gradients of the NO2 column, the quantity that determines NO2 lifetime and emissions in the analysis. Seoul is in a mountain basin terrain at the coast with further impacts on local atmospheric circulations from urban land use (e.g., https://www.atmos-chem-phys.net/13/2177/2013/acp-13-2177-2013.pdf).

We agree that it is important to reference the complex topography and meteorology of the area as sources of uncertainty, but we do not expect this source of uncertainty to bias our results in any particular direction. We have added a paragraph in the discussion section and now reference the aforementioned study and others.

The complex geography of the region further supports the use of our 4 x 4 km^2 simulation because it will capture topography and mesoscale phenomena better than a coarse global model.

When re-processing the air mass factor we use surface pressure of the WRF-Chem simulation to process the air mass factor, so we are already accounting for topographical differences in surface pressures. This is already discussed in Section 2.1.1.

√Also, the use of KORUS data in this manuscript for understanding the problem is limited, or is non-existent as it relates to understanding the NO2 lifetime and NOy partitioning. The authors can use this opportunity to analyze the KORUS-AQ dataset, to compare observed NOx/NOy partitioning versus the NO2 lifetime inferred in their analysis. The authors state that the NO2 lifetime is not necessarily related to the true chemical lifetime (P12, L22-23), but the theoretical framework for the EMG method assumes that is the case, as NOx lifetime, emissions and background concentration are the only variables affecting total integrated NO2 mass.

We have now included a comparison to NO_y from the DC-8 aircraft. This is now shown in Figures 5 & 9. The large underestimate of NO_y further supports the conclusions of our manuscript and makes it stronger.

It is beyond the scope of this study to do a full analysis of the NO_y partitioning.

√I also recommend that the authors evaluate more than two days of model-observation comparisons in the assessment of the updated CTM simulation and add to the discussion (Fig. 9, P11 L29 – P12 L2) .

As suggested, a CTM simulation with 2.13 × NOx for the entire month of May 2016 is now included.

This paper and the KORUS dataset provides an excellent opportunity to address the above complications and concern and I recommend the authors add more detailed analysis and discussion before publication.

Additional comments:

√P1: L21-22: "regional NASA OMI NO2 product" clarify wording as this is not an official NASA product but rather is regional inputs to a NASA tropospheric SCD product

Updated

√L25-26: Do the reported scalar quantities refer to integrated mass or a scalar difference at a single point?

The scalar quantity for Seoul is within a 40 km radius. This has been clarified in Section 3.2, which is the section this statement is referencing.

√L30-32: Consider clarifying

√P2: L3: "Ideal" – this is strange wording as some ozone production occurs nearly everywhere in the troposphere with sufficient light at wavelengths less than 405 nm. Net production is another question.

Re-phrased to say, "In the presence of abundant volatile organic compounds and strong sunlight, NOx can participate in a series of chemical reactions to accelerate the production of O3".

√L6-7: Consider word choice. Lightning is also a source. Furthermore, there is a large difference between budget and burden, which seems to be confused here. For example, the largest contributor to the atmospheric burden of NO2 is stratospheric N2O photolysis.

Re-phrased to say, "There are some biogenic emissions of NOx (e.g., lightning), but the majority of the NOx emissions are from anthropogenic sources".

√L8: I recommend changing the wording "NO2 is one of the easiest trace gases to observe" to commenting that there is a rich legacy of NO2 measurements by remote sensing that has been validated.

This paragraph has been re-worded as suggested.

√P3: L4: Consider inclusion of Zhou et al. Zhou, Y., D. Brunner, R. J. D. Spurr, K. F. Boersma, M. Sneep, C. Popp and B. Buchmann, Accounting for surface reflectance anisotropy in satellite retrievals of tropospheric NO2. Atmos. Meas. Tech., 3, 1185-1203, 2010. Zhou, Y., D. Brunner, K. F. Boersma, R. Dirksen, and P. Wang, An improved tropospheric NO2 retrieval for OMI observations in the vicinity of mountaineous terrain. Atmos. Meas. Tech., 2, 401-416, 2009.

Included

√P4: L23-24: Please explain in more detail how “vertical profiles are scaled based on a comparison with in situ aircraft observations”

An extra sentence has been added: “For example, if the aircraft observations show that NO_2 concentrations between 0 - 500 m are low by 50%, then we scale the modeled NO_2 in this altitude bin by this same amount.”

As shown now shown in Figure 11, this has a minimal effect on the calculation of vertical tropospheric column contents over the Korean peninsula.

√P5: L1: “We used May 2016 monthly mean values” Please briefly mention here whether data outside of May 2016 will be used in the top-down emissions

Yes, these data are also included in the top-down analysis. A sentence has been added: “In the top-down emissions derivation, we use all nine-months of OMI data for the analysis.”

√P6: L26-27: I am quite skeptical that this is the first time that geostationary products have been used in a forecasting framework. E.g., https://journals.ametsoc.org/doi/abs/10.1175/2008WAF2222165.1

This sentence has been removed.

√L31: Does “enhancements” mean additions to the inventory or does it mean modifications to the inventory? Please clarify

The main enhancement that we made is to add new construction of power plants. Per your question, "addition" would be the right word. This is clarified in the text.

√P7 L2: Why project to 2015 and not project emissions to 2016?

The NIER (National Institute of Environmental Research) of Korea generates a "Present Version Inventory," for their air quality forecasting, by projecting the base year inventory for three years (i.e. 2015). In this simulation, we use that version of the inventory. We have clarified that NIER provided the projected emissions, and that we did not project the emissions.

√P8 L8-10: This statement would be stronger if a reference is cited

Reference to de Foy et al. (2014) is now included.

√L10-12: The ratio of NO2 to NOx is time-dependent and spatially varying, depending primarily on JNO2 and O3. This should at least be noted.

Updated

√L21: please clarify that you are referring to ERA-Interim winds

Clarified

√P9: L19-34: The chemical lifetime of NOx is impacted by uncertainties in the simulations VOC concentrations and type, OH, RO2 and R(O)O2 radicals. As an example, simulated atmospheric concentrations of aromatic compounds in Seoul are much smaller than observed during KORUS-AQ. An underestimate of aromatics would certainly have a large impact on peroxyacyl and alkyl nitrate formation, and should enhance the effective NOx lifetime in the near-field.

This has been updated. Romer et al., (2016) has now been cited to support this.

√Without further justification or analysis, I don't think the comparison of emission timing is all that helpful. We would expect a different time-of-day profile of NOx emissions in Korea and USA given that the source mix of NOx is different. Emissions timing can be listed as an uncertainty without devoting an entire figure to it (Fig. 4).

The emission timing is critical to the conclusion of this paper. We are suggesting that the timing of emissions can yield a large amount of uncertainty when evaluating emission inventories with satellite data. Thus, we still include this figure.

√P10: L20-21: please report the correlation coefficients here and refer to Figure 6.

Included

√L32: If archived, please compute the chemical lifetime of NOx in the model based on all NOy species.

This was not archived.

√P11: L1-4: From where do the uncertainty estimates affecting the analysis originate?

Please reference Lu et al., (2015). We have added the citation here.

√L3-4: "Only the latter three terms are used to calculate the uncertainty of the NO2 lifetime" Why? The NOx to NO2 ratio has a large impact on the NO2 (NOx) chemical lifetime as NO removal tends to be much slower than NO2 removal.

Please reference Lu et al., (2015). We have added the citation here.

√L6-7: Why choose a radius of 40 km when the rotated plume domain width is 250 km? It seems a bit arbitrary as distance can be adjusted to improve the comparison of top-down results with bottom-up inputs.

An assumption with this method is that all of the NOx emissions are clustered near a single point. The radius of 40 km from the city center is chosen because it encompasses an area which includes the highest NOx emission sources, but very little of the emissions from more rural areas, which are contributing to the background NO_2. A radius much larger than 40 km would be inappropriate.

For the calculation of the OMI line densities, we apply a 120 km radius (we are unsure where you saw the number 250 as it is not in the original manuscript). By doing so, we are assuming that emissions between a radius of 40 km and 120 km are contributing to the background. This is an assumption of the top-down method. Figure 7 confirms that this is a valid assumption.

√P11: L30-31: I recommend completing a validation analysis of more than two days' worth of simulations.

This is now included. Please refer to Figures 9 & 10.

√P12: L24-25: "This is because the lifetime calculation is extremely sensitive to the accuracy of the wind direction." Given that top-down inference of lifetime, emissions and estimates of background NO2 are directly linked in the EMG analysis, the above statement is also true for inference of top-down emissions and background NO2. Is this "extreme sensitivity" appropriately characterized by the 30% error estimate reported on P11, L3? Please move this discussion from conclusions section on P12 to error estimate discussion early on P11 and provide a more detailed accounting.

As suggested, this has been moved to Section 3.6.2. A reference is now also cited, de Foy et al. (2014), which describes in detail the uncertainty analysis.