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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "A top-down assessment using OMI NO2 suggests an underestimate in the NOx emissions inventory in Seoul, South Korea during KORUS-AQ" *by* Daniel L. Goldberg et al.

**Anonymous Referee #2**

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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).



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Discussion paper

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

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

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

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.

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.

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.

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.

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

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

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

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

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

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

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.

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

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.

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).

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

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

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

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.

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.

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

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.

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