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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 #1

Received and published: 17 August 2018

The manuscript by Goldberg et al. is a valuable and timely analysis of NOx emissions during KORUS-AQ. It identifies some potential issues with NOx emissions in the region that are useful for air quality management as well as other works studying pollution during this campaign period. The work also has relevance beyond KORUS-AQ in terms of how OMI data is used to estimate NOx from urban areas, and also how TROPOMI data will be used in such studies in the future. The article is in general quite clear and easy to read, and most figures are useful and essential.

That being said, the work misses a critical opportunity to evaluate one of their main hypotheses, which is that regionally-derived NO2 columns (using air mass factors from

high-resolution WRF-Chem simulations) lead to objectively better NOx inversions. In fact, while they report the difference between these NOx inversions and those based on the standard OMI NO2 data, the differences aren't critically evaluated, which is a shame, as it seems to be a rather easy next step. This would thus be my primary suggestion for revision. A few other aspects such as how using AMFs derived from a model that is clearly inaccurate to begin with affect their analysis, why spatial averaging is presented and then discarded, and why the regionally-derived NO2 columns may be overestimating NO2 in rural areas need to also be addressed.

Details of these comments as well as other are presented below; addressing them likely constitutes major revisions as additional WRF-Chem calculations are required.

Major comments:

Section 3.6: It isn't clear to me why the authors test a doubling of the emissions. The prior bottom-up values are 198, the top-down using standard product are 353 (an increase of x1.78) and the top-down using the regional product are 484 (an increase of x2.44). The test increase of x2 thus does little to distinguish between these two. This is a bit of a disappointment, as a major conclusion from this work is that the regional product (and top-down emissions using this product) are significantly different and better than the standard product. However, the only evidence presented that the regional product is better than standard thus far is the comparison to Pandora data. While encouraging, the authors are missing an big opportunity to make this argument much stronger by performing two model simulations for the entire KORUS-AQ period with top-down emissions that match those derived using the standard product and the regional product, precisely, and not some estimate of x2 that is neither here nor there. These two different model simulations can then be evaluated using the aircraft data.

General: Model values of NO2 column are much lower than regionally-derived OMI NO2 column in most areas, including rural areas (Fig 3). However model values match the aircraft data in rural areas (i.e. the only major discrepancies noted in discussion

of Fig 5 or e.g. the conclusions (12.17-19)). What are we thus to make then of the quality of the regionally-derived OMI values in rural areas? Too high? This should be discussed. If these are too high, will the background values estimated in the EMG value thus be too high, and this error propagate into an error in the urban emissions?

General: If model columns are too low, how does that impact model calculated AMF? How much would AMF change if using posterior emissions in WRF-Chem? An additional calculation of AMFs based on WRF-Chem simulations with adjusted emissions needs to be performed to answer this question. Or perhaps the NO2 profiles in WRF-Chem are adjusted to account for this bias (this is indicated on 4.23, but no details are provided as to what this adjustment is, or how it is derived)? I try to evaluate the WRF-Chem profiles visually, based on Fig 5, but this plot doesn't make that information clearly visible given the way the vertical axis isn't strictly used (i.e. model and aircraft data collected at the same height are not plotted at the same height – which I understand from the perspective of clarity in showing their differences with box-whisker plots, but something else is needed to evaluate profile shapes).

General: if results with spatial ave kernel are not trusted for analysis, they should be removed throughout from the results. Otherwise, it is a bit of a distracting / potentially misleading presentation. For example on page 12, line 5 – this isn't used, so why is it highlighted here? Still, wouldn't there be some data from KORUS-AQ with which wind field estimates in WRF could be evaluated? It just seems a bit subjective here that this source of error is singled out (11.18) as justification for not using this approach, whereas profile shapes that come from WRF-Chem are deemed acceptable, even though WRF-Chem NO2 column values are significantly biased low in urban areas. Further, it seems that comparison to the Pandora data in Fig 6 would indicate that the spatial kernel adjustment is improving, rather than degrading, the column estimates, which is a point in favor of this approach.

9.30-34: Not sure how this statement about NOx diurnal variability contributes to the difference between modeled and observed NO2 columns. Are the authors suggesting

that the diurnal variability of NOx emissions in Korea is incorrect? Simply noting that it is different than modeled diurnal variability in the US is not sufficient evidence and in fact comes across as tangential, unless the authors are claiming that NOx source profiles (EGUs, distribution of diesel vehicles in the transportation fleet) are identical, which seems dubious. So I suggest removing Fig 4, unless this argument can be substantially strengthened. Additionally, I wonder to what extent excessive NO2 deposition in the model might be contributing to the noted differences; this could be driven by e.g. PBL heights in the model that are too low. I suspect there is more information from the KORUS-AQ campaign that could be used to evaluate this.

Fig 5 and associated text: I agree this suggests the differences between WRF-Chem and OMI near Seoul are likely driven by emissions, rather than chemistry, deposition, or PBL heights, as suggested by the authors or myself.

10.20: Thoughts on why bias improves but not correlation? This might suggest that the daily variability of WRF-Chem (which impacts daily AMFs) is not correct, or at least not an improvement upon larger-scale averages.

General: How does the plume analysis / rotation / EMG inversion process work if e.g. there is a large point source whose outflowing plume flows over another source (e.g. a highway) that runs parallel underneath it, replenishing NO2 concentrations that are then going to be ascribed only to emissions at a single point of plume origin? So, related, at 11.10: Yes, but the concern is rather smaller sources within this radius but not at the center that contribute to the plume (i.e. mobile sources).

Minor comments and corrections:

Throughout: "shape profiles" reads a bit strange. Change to "profile shapes"? Or just profiles?

1.25: for the –> for 1.26: "larger near large" rewrite

2.4-5: "another . . . another" rewrite

3.27: trace-gas

Eq. 2: include a proper summation index

4.5-6: It isn't clear here if the authors are discussing how AMFs are calculated in general, in the standard retrieval, or in their own regionally-specific retrieval. Please clarify.

5.1: How big of an assumption is this, that the profiles are constant over this time range?

6.26: I'm pretty sure AOD from geostationary satellites over Korea have been used for forecasting studies. Not sure though how the authors here qualify their study as "near-real time"; all I saw was reanalysis. NRT usually means forecasting. Just because the winds were forecast within the domain doesn't mean this is a chemical forecast, since the observations used span the time period over which the analysis (aircraft obs) are made (considerably, given that satellite data for several more years and months are used). This entire approach would be impossible in an NRT setting, given the data requirements for oversampling.

7.28: plume, –> plume

8.6: Why using wind estimates from a different model than the one used to constrain WRF met a the boundary (NCEP), or different from WRF itself?

8.8: Why 500m? Based on Fig 5 it looks like NO2 plumes extend much higher than that, up to 1 km or possibly above (although a bit hard to tell from this plot, given the manner in which the vertical scale is treated).

Fig 1: content –> concentration

Fig 1: Why showing US domain?

9.4: is in despite of âĂŤ> is despite

Section 3.1: Inclusion of / comparison to the US feels tangential and unnecessary. Suggest focus on Korea domain; remove US from Fig 1 and remove discussion here. This point could be touched on in intro or conclusions but doesn't fit well in the results.

9.17: There are also small decreases in the southern part of the peninsula, as well the SE corner of the domain. Further, the explanation provided for the decreases isn't particularly insightful.

9.21: From the presence of red in panel (c), the statement "in all areas" does not seem to accurately describe the results. Please update text to more precisely reflect the findings.

Section 3.3.1: it's not good style to have only one subsub section in a section. Consider merging this with 3.3 or making 3.3 WRF-Chem evaluation, 3.3.1 comparison to OMI and 3.3.2 comparison to aircraft.