Comment on acp-2021-642

Anonymous Referee #1

Referee comment on "A new method for inferring city emissions and lifetimes of nitrogen oxides from high-resolution nitrogen dioxide observations: a model study" by Fei Liu et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-642-RC1, 2021

This manuscript describes an enhanced method to infer NOx emissions from urban areas using satellite data. This manuscript is a proof of concept and validation study tested on 60 US cities. This manuscript is a nice advancement. I recommend minor revisions based on my comments below.

The mention of "TROPOMI overpass time" initially added some confusion, since the mention of TROPOMI implied that TROPOMI data was somehow used. After re-reading, it is clear that TROPOMI data was not used and instead this method is applicable to any satellite with an early afternoon overpass time (e.g. OMI). Lines 120, 156 and figure captions of Figure 2, 5, and 6 should be revised to remove the word "TROPOMI" and instead use the actual model time, presumably 13:00 or 14:00 local time. Perhaps Line 120 should be revised to say ... "sampled at 13:00 local time, which approximately corresponds the early afternoon overpass time of OMI and TROPOMI", and then 13:00 (or whatever the exact time of the model output used) should be used in Line 167, and the figure captions.

Section 2.1 could use a bit of reorganization. For example, "tau" is first discussed in Lines 144 - 147, and then other variables are mentioned and then "tau" is discussed again in Lines 162 - 173. Lines 144 - 147 should be discussion in succession with Lines 162 -173. This is also true of the "ratio" and "b" variables. They are first discussed in Line 137, and then again in Lines 148 - 154. This makes it hard to follow.

In the paragraph starting at Line 376 the authors discuss the uncertainties caused by clouds (which is good), but it's unclear if cloudy days were filtered out in the analysis. If not, then it would be important to mention this, perhaps near Line 188 of Section 2.2. Also, please mention that the NO2 lifetime during a day with a true satellite observation will likely be smaller than the values reported herein since sunny days yield faster photolysis rates.

Line 285 - 288: r=0.01 is quite poor performance of the Beirle et al. 2011 method and a bit surprising. I wonder how the correlation would be if you eliminated "poor fitted" results (tau <1 hour and tau>5) in a similar manner to how certain cities were "eliminated" for the method described herein (Lines 238 - 241). It seems like the especially low correlation is driven by six outlier points, that if removed, might give better correlation. It'd be fair to do this if you are filtering out cities in your own method! This is not to say that the Beirle et al. method is equally good as the new method described herein, but it's probably not as bad as implied by the low correlation. I think it'd be fair to say that the Beirle et al. 2011 method might only work in a narrower range of cities (i.e., needs a stricter filter) as opposed to implying that it has almost no correlation in most circumstances.

Other minor suggestions:

Line 52: exploit —> use

Line 69: Perhaps mentioning validation field campaigns (e.g. DISCOVER-AQ, KORUS-AQ, CINDI-2) as being helpful to better quanitfy errors in the satellite data, and therefore reduce uncertainties in the applications of satellite data such as this project.

Figure 1: This figure is a bit confusing to me. I've attached an image in the supplement that is a bit more intuitive to me based on my understanding. Please feel free to discard if not correct or helpful.

Line 108: Please be more specific about how the emissions were adjusted. Were they projected to a different year? If so, can you give a ballpark number as to how they are different than the 2011 NEI (10% lower? 20% lower? etc.)

Line 114: Appears that the simulation is 6 months. Does it correspond to a specific 6 months in time? Presumably it is Apr - Sept, as implied in Line 159?

Line 114: Mention here that the domain is shown in Figure 2, and covers the Continental US.

Line 115: This is nitpicky, but tropopause is closer to ~15 km during midsummer in most cases. How are you determining tropopause? From WRF or something else? Or are you assuming tropopause is consistently at the model top (50 hPa)?

Line 127: Can you list 60 cities in the supplementary? Also can you briefly comment on why only 26 cities are shown on Figure 2? (I see a longer explanation in Section 3.1)

Line 130: Would be helpful to mention here that a comparison to these two methods is forthcoming in Section 3.2.

Line 131: What does "Spatial emissions patterns" actually mean? This term is confusing to me. If I interpret Line 161 correctly, is it the instantaneous emissions rate (since the integral of it gives the emissions)?

Figure 4: Can you display the number of days included in each average? Presumably there are fewer days with easterly winds than westerly winds (for example).

Figure 5 (and S2): I very much like these figures, but wonder what information is missing by rounding the speed to 5 knots. Could you display the mean wind speed at each hour in a box on the bottom right of each of the nine panels?

Line 201: This sentence appears to be a bit contradictory with Line 202. For New York City, easterly winds don't do as well, but Line 201 seems to imply that all wind directions are OK. This should be re-phrased. More generally though, is there worse performance with easterly winds? That would be interesting information for the reader to know.

Line 255: I think you mean that there is "increasing agreement" over the 1 hr lifetime. Might also want to note that tau=3,6,9 are all similar, and that tau=12 does seem worse then 3,6,9, but better than 1 h (and this make sense since -12 h includes overnight!)

Line 301: Might be better to re-phrase to say that MISATEAM is better than Liu et al., 2016

Line 346: "layer height" —> "top wind layer height" or ""wind layer depth"

Line 392: It's probably worthwhile to also mention these uncertainties in the abstract (Line 27) and conclusions (Line 413) in addition to the 15% / 20% values currently mentioned.

Line 411: Maybe add a short comment to say that diurnal NO2 lifetime differences will need to be investigated before applying this method to hours outside of the early afternoon timeframe. Or perhaps you are even willing to say that the method can only be applied in the early afternoon based on work applying this method to timeframes with longer NO2 lifetimes (Line 354)

Line 414: Maybe be more explicit and mention a current low bias in TROPOMI NO2 (Verhoelst et al. 2021) that would likely yield low satellite-derived NOx emissions if not bias-corrected.

Please also note the supplement to this comment:
https://acp.copernicus.org/preprints/acp-2021-642/acp-2021-642-RC1-supplement.pdf