

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2021-273

Anonymous Referee #1

Referee comment on "Variability of nitrogen oxide emission fluxes and lifetimes estimated from Sentinel-5P TROPOMI observations" by Kezia Lange et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-273-RC1>, 2021

This manuscript estimates the NO_x emissions from cities and large sources around the globe using TROPOMI NO₂ satellite data. I appreciate that the manuscript is thorough and well-written.

I am concerned about the derived NO₂ lifetimes by season and month. The magnitude of the seasonal cycle of emissions reported here are in disagreement (and sometimes strong disagreement) with previous literature. Please see Crippa et al., 2020 which shows that CO₂ emissions (which can be used as a rough surrogate for NO_x in this instance) don't have a very strong seasonal cycle. Using data reported in Crippa et al., 2020... CO₂ emissions are maybe 10% lower in summer in the US and China as compared to winter. In Spring and Fall, CO₂ emissions are lowest, perhaps 15-20% lower than the winter peak. The summer-to-winter ratios reported in this manuscript, such as ~0.5 in New York City, ~0.4 in Chicago, ~0.25 in Wuhan are simply not reasonable. Even in Europe, Crippa et al., report ratios of ~0.8, and the values reported here are significantly lower.

My guess is that the EMG method is having trouble discerning the true NO₂ lifetime during winter. This is a known issue and is why previous literature focus on summer time emissions (Lu et al., 2015; Goldberg et al., 2019). To prove whether your method is reasonable during the winter, I suggest two potential strategies: 1.) You can apply the method to a model simulation that has data during both the winter and summer; since the emissions in the model are known, the method should hopefully reproduce the seasonal cycle of the emissions that are input into the model (whatever they are). 2.) You can apply the method to power plants which report their NO_x emissions for all seasons such as all the large power generation facilities in the US. This will hopefully provide insight on whether the seasonal effects on the NO₂ lifetime and therefore NO_x emissions are being correctly accounted for.

With the aforementioned said, I think that Sections 4.1 and 4.4 are useful advancements to the literature. I recommend that Sections 4.2, 4.3 and 4.5 are excluded, and included

in a follow-up manuscript that more rigorously evaluates the top-down NO₂ lifetime by season.

All suggestions:

Line 39: in-homogeneous → heterogeneous

Line 43: Missing comma between winter and residential

Line 62: states → countries

Line 147: Availability of data in March and April?

Figure 1: Suggestion to remove lat/lon lines on the plot. The lines obscures a few cities.

Line 163: Wind direction likely similar between heights, but wind speed can be different, especially if comparing near surface to 1000 m. Maybe something to note.

Line 163: Might also want to mentioned ease of use of 100-m wind speed since it is a standard variable output in the ERA5 re-analysis.

Line 172: Can you clarify? Do you mean every three hours (0Z, 3Z, etc.)? I originally interpreted three hourly estimates to mean three different outputs each hour.

Line 173: Presumably the interpolation is location specific (Europe may use 12Z model data but China may use 3Z for example). Please clarify.

Line 207: earth → globe

Line 213: Modify "low cloud coverage" to "few clouds". "Low cloud" could be interpreted to mean low in the atmosphere.

Line 318: Verhoelst et al., 2021 and Judd et al. 2020, which you already cite, show a low bias in polluted areas of ~20-30%. I see no problem with using this to scale the final estimates by the value, while also noting the large uncertainty in this conversion factor.

Line 336: Early studies, published in 2018 & 2019 used an algorithm that has since been re-processed. Also there was a different rotation in Goldberg et al., 2019; it was done manually.

Line 350: I am not convinced that these ratios should be this small. Traffic pollution and industrial/manufacturing pollution, is relatively constant year-round and represents a large fraction of emissions. If anything traffic and industrial emissions would be biased to be larger in the summer (traffic peaks in July and lowest in February: https://www.fhwa.dot.gov/policyinformation/travel_monitoring/tvt.cfm). NO₂ lifetime, in theory, should be varying much more than you found. So I'm thinking the summer/winter ratios are an artifact of an erroneous lifetime fit. See Zheng 2018 as an example. They report that residential emissions in China (the sector causing the most intraseasonal variation in emissions) is ~5% of Chinese emissions. Even if this percentage is off by a factor of two and power generation varies much more than we suspected (Crippa et al. 2020 show this doesn't vary much by season in China), it would be very unlikely to get ratios larger/smaller than 0.8-1.2.

Line 381: I'm glad you are transparent with this, but these don't seem to be reasonable.

NO₂ lifetime should be shortest in summer, similar in Spring & Fall, and longest in Winter.

Line 391: Can't you go a step further and estimate OH for a few cities, perhaps from a model or previous literature, and see if the NO₂ lifetimes you derive are approximately similar?

Line 415: Often, Saturday is different than Sunday... Saturday emissions can be quite larger than Sunday. See Goldberg et al., 2021 and Crippa et al., 2020 as an example

Line 420: Also see Goldberg et al., 2021

Section 4.5 and Figure 7: If seasonal emissions are biased high in winter, then these drops attributed to COVID will be overestimated. Sections 4.2 & 4.3 need to be revised in order for me to have more confidence in the results presented in this section.

Section 4.6: Please more explicitly differentiate between random errors, which should mostly cancel out since you are averaging over many days of observations, and systematic errors, which would not cancel out.

Line 564: Perhaps you may even want to say that re-gridding TROPOMI data to a resolution of 0.1 degrees (~10 km) might be better than a higher resolution (~1 km) for this specific purpose only.

References:

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