

Comments from anonymous Referee #3:

We would like to thank the reviewer for his/her helpful comments. We hope that we could address all questions and unclear points satisfactorily.

In the course of the revision, we have made the following important changes to the analysis: We updated our wind and ozone data and are now using ERA5 reanalysis data averaged about the boundary layer instead of a fixed height to include possible seasonality in these input data. This led to changes in all our estimates but not in any fundamental way. We also applied our method to a power plant for which hourly NO_x emissions are reported, and analyzed the temporal variability over the course of the year in more detail. From this comparison we conclude that the EMG method applied to TROPOMI data at least for this power plant can reproduce the temporal variability reasonably well and does not show a clear seasonal bias. To get a better representation of source areas from Southern Hemisphere, we included five new target areas. The results fit well into the overall result of the study.

Legend: Referee comments in black, author comments in blue

The study "Variability of nitrogen oxide emission fluxes and lifetimes estimated from Sentinel-5P TROPOMI observations" by Lange et al. presents emission and lifetime estimates from TROPOMI NO_2 measurements for a selection of source regions around the world, and investigates weekly & seasonal cycles as well as the impact of recent lockdown measures.

The manuscript is clearly written, provides a detailed description of method & error calculation, and discusses the results adequately, also in context with previous studies. I recommend publication after dealing with the comments below.

NO_x : x should be subscript throughout the manuscript.

NO_x was changed to NO_x throughout the manuscript.

Line 3: this resolution was not provided in the first months of operation.

We changed the sentence to: "In this study, two years of TROPOMI NO_2 data, having a spatial resolution of up to 3.5km x 5.5km, have been analyzed together with wind data." More details about the change of resolution are given in section 2.1.

Line 7: "dessert" should be "desert"

Changed.

Lines 14&15: 2x "In the atmosphere". I would skip the first one.

We skipped the first one.

Line 123: Please specify that for the highest latitudes considered here, up to two overpasses are available. Three overpasses only occur for higher latitudes.

For some days there are three overpasses available, not every day and two of them are then also only on the outer edge of the orbit but for example on 17 July 2020 there are three overpasses available for Saint Petersburg:

Date	UTC	Satellite azimuth	Satellite elevation	Viewing zenith	Solar zenith	Satellite direction	Distance to site (km)
17.07.2020	08:41	47.23	25.65	64.35	41.41	ascend	1287.63
17.07.2020	10:20	69.08	83.41	6.59	38.82	ascend	85.91
17.07.2020	12:02	270.67	25.23	64.77	43.94	ascend	1305.12

We rewrote the sentence to make clear that two of the three measurements are then on the edge of the orbit: “At higher latitudes, on some days up to three overpasses, with approximately 100 minutes in between the measurements, are available. In the rare case of three overpasses, two of them are only with the outer edge of the swath.”

Lines 126-129 (up to "Seo et al."): This is off-topic and should be skipped.

We skipped the part about the other available products of TROPOMI.

Line 135: Please explain why the difference in resolution is more extreme at the edges.

The larger pixels are a geometric effect due to the slanted view to the ground. To avoid this, for TROPOMI, in the middle part of the scan always two pixels are averaged but not at the edges. As a result, the resolution decreases towards the edges, improves by a factor of two and then decreases again. We added a short comment to the manuscript.

Lines 145ff: Please explain why versions up to 1.03 are mixed in your analysis, but 1.04 is excluded. Shortly mention what has changed in 1.04 and refer to the discussion of the low bias of tropospheric NO₂ in section 4.6.

We made some changes in the text to better explain the version changes:

“Changes between the versions before 1.04. are only minor and data can be mixed. 1.04 was activated on 02 December 2020 and implemented major changes, which led to a substantial increase in the tropospheric NO₂ column over polluted areas for scenes with small cloud fractions. We only use the data up to end of November 2020 for our analysis to ensure better comparability, since mixing data from before version 1.04. and after is not recommended. A complete mission reprocessing will be performed to get a harmonized dataset (Eskes and Eichmann, 2021). “

We also added some more discussion in section 4.6 about the version changes and possible influence on the analysis when using the reprocessed dataset.

Line 153: Do you have a reference for 50% CRF ~ 20% GCF?

We do not have a reference and deleted the last part about the GCF, as it is not important for the study.

Line 154: I am confused by the different time intervals.

In line 144: March 2018 to November 2020.

In line 197: March 2018 to February 2020 general January 2019 to November 2020 Covid 19 Why is yet another interval chosen for Fig. 1? Why not March 2018 to February 2020?

We changed the time interval for Fig1 to the complete time interval March 2018 to February 2020 of the main analysis.

For the main part we used data from March 2018 to February 2020 to exclude the influence of COVID-19 regulations. Following a comment from referee 2, we now shortened the time interval for the two analyzed cities in China (Wuhan, Xian) since lockdown in China started on 23 January 2020. All other cities should not be influenced since all following lockdowns were after February, the next one was in northern Italy on 8 March.

For the Covid analysis we used January 2019 to November 2020 data.

Line 155: The criterion "higher NO₂ than their surrounding" would result in a quite different selection of hot spots visible in the mean map. I propose to skip the reference to Fig. 1 in this section, but add it section 3.1, where the selection of source regions is explained in detail.

Thank you for the comment, we skipped the reference in this section and added the figure to section 3.1.

Caption Fig. 1: Add a reference to table A1 for further details.

Done

Line 162: Please quantify "low".

We added more detail here: "The resulting emissions and lifetimes change less than 2% (5%) on average when using 200m (1000m) instead of 500m and less than 15% for individual sources."

Section 2.3: Using model input in order to correct for the NO₂/NO_x ratio is probably an improvement compared to just taking a constant value. However, I don't understand why the calculation in Eq. 3 was done for a fixed pressure, since the model would also provide the actual pressure at the altitude of interest (probably the same as chosen for the wind vector). The considered source regions include some elevated locations like Las Vegas, Medupi, or Colstrip (1 km asl!), where 950 hPa is not appropriate.

We updated the ozone data and are now also using the ERA5 reanalysis data. We use hourly ozone volume mixing ratios with a horizontal resolution of 0.25° in model levels averaged over the boundary layer and interpolated to TROPOMI overpass time and oversampled to the same 0.01° resolution as the TROPOMI data.

Line 219: How far does the - somehow arbitrary - selection by visual inspection affects the generality of the results? Could there be some "selection bias"?

This is of course hard to quantify. Of course, we tried to select a good mix of sources from different regions, we extended our analysis region as far as possible north. Referee 2 suggested to add more source regions from the southern hemisphere, and we added 5 new target regions. We analyzed urban sources, isolated power plants, oil fields, industrial regions and some mixed sources. Our results show a wide range of mean emissions (4.6mol/s – 272.9mol/s) and lifetimes (1.2h – 8.2h). Nevertheless, some selection bias could have been introduced, for example by not analyzing regions with low wind speed or areas in regions with many emission sources and

higher background levels as for example in eastern China. We added a comment about this possibility in this section and also in the conclusion.

Line 224: What amount of TROPOMI observations is needed in order to get robust results from this method?

The method works in principle on a single TROPOMI overpass as shown in Lorente et al. (2019) and Goldberg et al. (2019). If the conditions are favorable (e.g. clear sky, homogeneous, strong wind conditions...) a single overpass can deliver good results.

But of course, it depends on the goal of the analysis. We wanted to analyze variability due to seasonal or weekday effect in a more general way, therefore we need better statistics to not for example introduce a bias in the seasonal analysis by weekday variability. We used two years of data for our analysis, to have for example at least two times three months for seasonal analysis. This also makes it possible to analyze regions which are covered by clouds more frequently. We added a comment to the text, that shorter time intervals are possible depending on the goal of analysis.

Line 293: What is "dispersion" meaning in this context?

“effective mean dispersion lifetime” – in this case we mean dispersion (e.g. dilution) by wind.

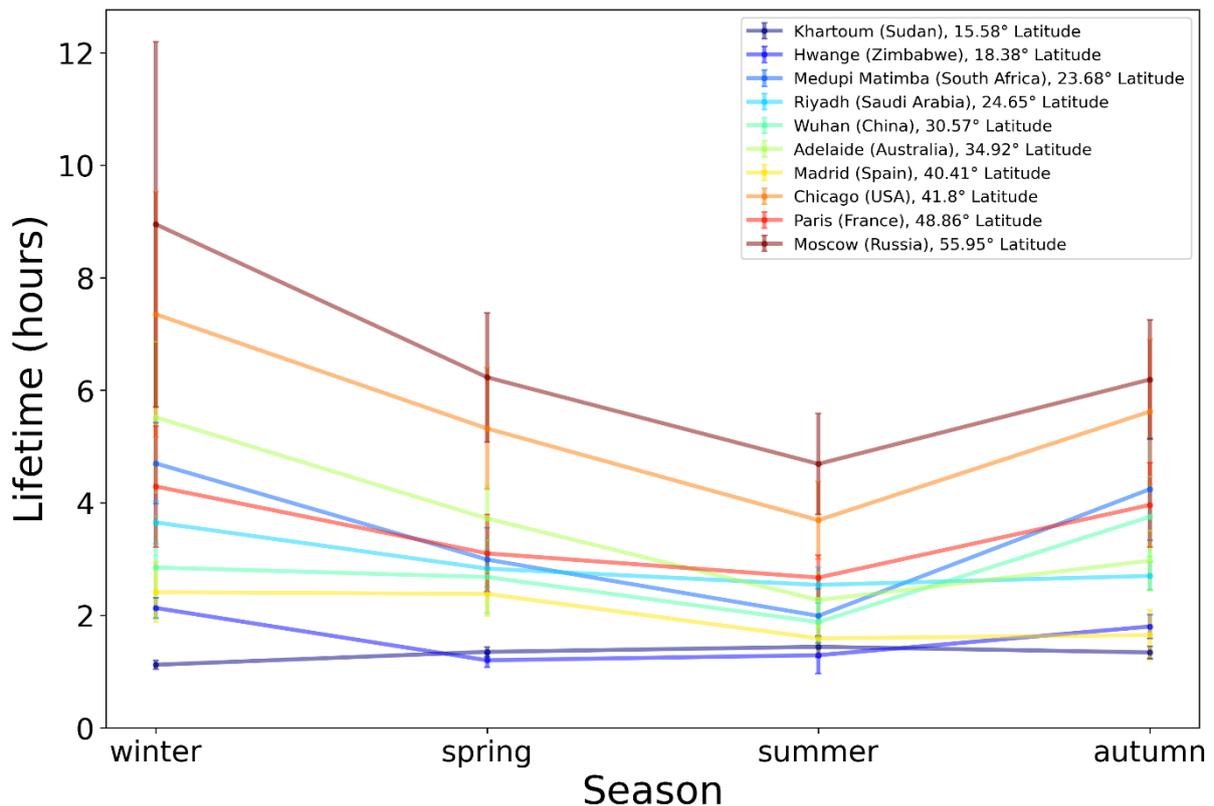
Line 294: or the effects of non-linearities in the NO_x lifetime.

Added to the text.

Figure 5: This figure is very hard to read. I propose to add one further panel, highlighting the seasonal dependence. E.g. show the lifetime for different locations as function of season.

- arrange the 4 panels 2x2
- enlarge

We created an additional figure, which shows the lifetime for different locations as function of season and arranged all 4 plots in 2x2. The additional figure is showing the seasonal dependence in more detail and the new arrangement makes everything better to read. In order to keep the additional figure easy to read, we could only include a selection of the sources we analyzed. We tried to find a mix of regions from southern and northern latitude, cities as well as powerplants and well distributed over many latitudes



Line 430: One important difference is that the tropospheric columns also contain the upper tropospheric background, which is removed in the EMG method by fitting the background B.

Thank you for pointing this out, we added this comment to the text.

Lines 507ff: This section is quite vague. One main reason for the low bias was identified as the FRESCO cloud height which is biased low (Compernelle et al., <https://amt.copernicus.org/articles/14/2451/2021/>) causing high-biased AMFs. This was changed with v1.04 of the operational NO2 product. Please extend this discussion accordingly.

We added more details and discussion about the version changes and possible influence on the analysis when using the reprocessed dataset in section 4.6.

Line 613: Please specify "short time periods": how many TROPOMI overpasses are needed?

We added the following comment about possible time periods:

“Depending on the goal of the analysis, already a few days of measurements can be sufficient; for seasonal studies, depending on the local meteorological conditions, one to two seasons are sufficient, and it is not anymore necessary to average over several years.”