

This paper by Beirle and Wagner presents a new method for estimating NO<sub>x</sub> emissions and lifetimes for 100 large cities from TROPOMI observations. The estimate is based on the downwind plume evolution for different wind directions separately. A simultaneous fit of downwind patterns for opposing wind directions makes the estimate more robust than previous studies using a single fit.

Results are compared to EDGAR. Seasonal NO<sub>x</sub> emissions and lifetimes are provided. The assumption of point-source like cities, the background dependency on wind direction, and the benefits of the described method are discussed.

The paper is well-written, falls into the scope of AMT and is scientifically relevant. Therefore, I recommend publication in AMT after addressing the following comments.

We would like to thank the reviewer for the positive evaluation of our study and for the valuable feedback. Below we refer to the reviewer's comments one-by-one.

General comments:

Line 91: You wrote: "As the NO<sub>2</sub> photolysis rate is driven by the SZA, which shows a seasonality with minimum and maximum close to the solstices, seasons are defined accordingly as winter (NDJ), spring (FMA), summer (MJJ) and autumn (ASO) in this study." This definition of seasons is not in line with the meteorological and more common definition of seasons as winter (DJF), spring (MAM), summer (JJA), and autumn (SON) and might cause confusion when comparing the presented results with other studies. It may lead to wrong seasonality in the presented emissions and lifetimes. I understand that adapting the seasons to the meteorological and more common definition is probably a lot of work and not easily possible. Therefore, I would recommend pointing out your definition of seasons at important points. This would hopefully avoid confusion and inconsistency in future comparisons for readers not reading the manuscript in detail.

We agree that the unusual definition of seasons in our study is potentially confusing. In order to clarify this point, we further motivate our definition of seasons in subsection 3.2 in the revised document. In addition, we repeat our definition of seasons within all figure captions or labels.

It is possible to do your analysis on a seasonal basis, even if for many cities only one or two seasons are possible to analyze. But is it also necessary to do the analysis on a seasonal basis, as wind directions and speed have a seasonal dependence, or would your "Selection and averaging of fit results", that is described in Sect. 3.6, consider this? Please comment on this.

The TROPOMI data is sorted for wind direction before averaging. For each season, the TROPOMI pixels are averaged separately for each wind condition. The subsequent fits are based on the seasonal mean TROPOMI columns combined with the respective seasonal mean wind properties. Thus potential seasonal dependencies of wind conditions are accounted for in the gridding & fitting process.

In the revised manuscript, we have now explicitly specified that seasonal means are calculated for NO<sub>2</sub> **and wind fields** in section 3.2.

If you are not separating the data set into seasons, you wouldn't need to work on such long-term temporal averages (Line 231).

This is true, but our intention was to check for potential seasonalities in emissions and/or lifetimes, which is now explicitly stated in section 3.2. Considering total means might be misleading in cases where some seasons (in particular winter for Northern mid-latitudes) are not contributing valid measurements. Thus we would like to stick to

the seasonal analysis.

Specific comments:

Abstract: Add that your study is based on satellite data, maybe even more specifically, 3.5 years of TROPOMI NO<sub>2</sub> data.

We have added the following information to the first sentence of the abstract:

*"...based on NO<sub>2</sub> measurements from TROPOMI (PAL dataset, May 2018-November 2021)."*

Line 65: There is a newer, consistently reprocessed data product than the used v2.3.1. Can you comment on how the latest reprocessed data product v2.4 may influence your results?

To our understanding, the processor version 2.3 is a major improvement compared to v1.x, as cloud altitudes are now more realistic. Thus, the reported low-bias of NO<sub>2</sub> columns (e.g. Judd et al., 2020, <https://doi.org/10.5194/amt-13-6113-2020>; Lange et al., 2023, <https://doi.org/10.5194/amt-16-1357-2023>) is expected to be reduced. This aspect is now discussed in section 5.2 of the revised manuscript.

Compared to this major improvement, the updated from v2.3 to v2.4 have only minor impact on tropospheric NO<sub>2</sub> columns, thus we would expect that the emission estimates based on v2.4 would be similar to the results presented in this study.

Line 66: How are the good viewing conditions determined? Have you tested for larger SZA? You mentioned that this may influence the lifetime estimates.

This study is based on the same data processing as in Beirle et al., 2023. Therein, the quite strict selection criteria were applied for two reasons:

- SZA: NO<sub>2</sub> is scaled to NO<sub>x</sub> based on a parameterization of the photolysis frequency as function of SZA proposed by Dickerson et al., 1982, which is recommended to be applied for SZA < 65°.
- VZA: Large VZAs (> 56°) are skipped in order to avoid the large ground pixels at the TROPOMI swath edges.

In the revised manuscript, we provide additional information about why the angles where selected that strictly for this study.

Line 69: Are ozone concentrations from ERA5 as well or only the temperature data?

Ozone concentrations were not taken from ERA5. In the revised, we avoid this potential misunderstanding by changing the order of the listing of temperature (ERA5) and ozone.

Line 76: You wrote ERA5 wind data were interpolated on a regular horizontal grid with a resolution of 1° and stored in intervals of 6 hours. This means the closest interval to the TROPOMI overpass is chosen? Can it still represent the wind conditions around the TROPOMI overpass? ERA5 data are available in intervals of one hour. Is horizontal variability a problem when interpolating the original 0.25° resolution of ERA5 to 1° resolution? This is significantly larger than TROPOMI pixel sizes. The study would probably benefit from using finer temporal and spatial grids.

We have compiled an intermediate ERA5 dataset on 1° grid and 6 hour sampling. During data processing, these intermediate wind fields are interpolated in space and time matching to the TROPOMI measurements. We have clarified this procedure in the revised manuscript.

We agree that 1° is quite coarse, and we are currently preparing the implementation of high-resolution wind data for future studies (compare the reply to reviewer 1 on this issue).

In addition, we add the aspect of horizontal wind fields taken at one selected altitude as general simplification of our method to the conclusions.

Fig. 2/3: Please add the used data period. Are these figures based on data from May 2018 to November 2021 or a specific season?

As specified in the caption, the figures are showing seasonal means for winter (Fig. 2) and summer (Fig. 3). In order to clarify, we have modified the caption of Fig. 2 to “Mean NO<sub>x</sub> distribution for Riyadh depending on wind conditions for winter months (NDJ) in the PAL period (May 2018-November 2021)”, and Fig. 3 accordingly.

Some wind directions show distinct outflow patterns (Riyadh: North-West-wards and South-East-wards), some do not (Riyadh: East-wards) or are not very pronounced. Can you comment on this?

We thank the reviewer for this very detailed investigation of the presented data. The East-ward transport for Riyadh in winter, as shown in Fig. 2, is indeed less pronounced than for other wind directions. This corresponds to the findings shown in Fig. 4 (left): The observed line density for West to East (straight green line) cannot be reproduced well by the forward model (dashed green line). Actually, the reduced chi<sup>2</sup> for this wind axis is quite high (4.8) such that this wind axis is skipped from the calculation of mean emissions and lifetimes for Riyadh in winter (see section 3.6). In the revised manuscript, we now motivate the chi<sup>2</sup> selection criterium in section 3.6 for the Riyadh case.

The reason for the poor match of the fitted line densities is probably that ERA5 wind speeds and/or directions are not matching well to the actual transport for the observations sorted into the West-to-East direction.

How is the data availability? Is there a way to provide information on data availability for the different wind sectors?

We considered the question how to best provide information on data availability, but since the processing is quite complex, there are different levels of availability (available data per wind sector, available wind conditions per axis, available fit results per city, available mean results per season) and it is difficult to condense this information e.g. in a table. In the revised manuscript in section 3.6, further information is now provided on the net data reduction due to the different selection criteria. Concerning individual cities, we would like to refer to the Supplement: In the plots for mean columns and line densities for each city and each season, the different levels of data availability can be assessed. In order to allow for a simple identification of valid fit results, we changed the labels for discarded wind axes to grey in Fig. 4 and the respective figures in the Supplement.

Line 100: You wrote you consider distances of +/-50km in all directions. I agree that this is enough in the across-wind direction, but is it enough in the outflow direction? For Riyadh, plumes cover more than 50 km in the outflow direction.

We apologize for the confusion of the 50 km vs. 150 km distance:

- The +/-50km are applied for the *calculation of emissions*, i.e. spatial integration.
- For the *fit of the outflow pattern*, larger distances are considered, as stated in line 122.

We have clarified the usage of 50 km for emission estimates vs. larger distances in upwind/downwind direction for the fit in the revised manuscript.

As the line densities are binned with 10 km resolution, the selection of pixels within 150 km actually keeps the bin from -145 km to -135 km as first element, and 135 km to 145 km as last. Thus, we actually considered distances from -145 km to 145 km, which can also be seen in the line density plots (Fig. 4). We have modified this in the revised document.

Line 104: You wrote “the mean line density is calculated for calm, forward and backward wind direction”. Maybe replace with: calculated for calm wind conditions and forward and backward direction for windy conditions. Or replace direction in condition. Are windy conditions considered as  $\geq 2$ ? I think this was not specified yet, maybe obvious, but it is better to add it when introducing the criterion for calm conditions.

We thank the reviewer for this proposal and now use the term “condition” instead of “direction” whenever we refer to the set of forward/backward/calm wind conditions.

Windy conditions are considered as wind speeds  $\geq 2$  m/s, which is now explicitly specified in section 3.1.

Fig. 4: “The line densities for calm (blue), forward (green) and backward (purple) wind directions” See comment above “calm” is not a wind direction.

We changed “calm” into “calm conditions” in the figure caption.

Line 115 & Fig 4: You wrote: “E(x) represents the spatial density of emissions. It is considered to be the same for all 3 wind conditions.” Is this the case, I see that it is sometimes very similar to one of the wind conditions. Which E(x) is plotted in Fig. 4, the result from the simultaneous fit?

E(x) represents the spatial distribution of emissions and is thus considered to be the same for all wind conditions (i). We have clarified this in the extended description of the forward model. For calm conditions, L(x) is generally most similar to E(x), as almost no transport is taking place.

Line 122: You wrote: “observed line densities within 150 km of the city center”. This is not in line with the statement in line 100 that you consider distances of  $\pm 50$  km in all directions, please clarify. The resulting emission estimates are determined within  $100 \times 100$  km<sup>2</sup>, but line densities are fitted within 150 km of the city center?

Yes (see also comment/reply above). We have clarified this in the revised manuscript.

Line 132: You wrote: “Fit results for a wind axis are considered only if at least 2 directions have sufficient data”. This is not clear to me. One wind axis, e.g., North-South, only has three wind conditions: calm, forward, and backward. Do you mean the wind conditions, or is it something else?

Yes, meant is wind condition, which is now modified in the manuscript.

Line 138: You wrote “weighted by the number of contributing directions for each axis”. If the weighting considers calm, forward, and backward wind conditions, I would prefer to replace “directions” maybe with “wind conditions”. The same is true for the word direction in lines 140 & 141

We changed “directions” into “conditions”.

Fig. 5: Please provide error bars for the TROPOMI based emission estimates.

As discussed in section 5.2, we estimate the uncertainty of the TROPOMI based emission estimates to be about 30%-50%, which is still considerably large. Including these uncertainties as error bars would degrade readability of Fig. 5. Instead, we

state the uncertainty of the emission estimates in the figure caption and refer to section 5.2 for details.

Line 153: You mentioned that for most cities, valid emission estimates could only be derived for 1 or 2 seasons. How do you think this influences this comparison? For most cities, emission estimates are only derived for summer and autumn, but EDGAR considers data from all seasons.

We agree that the comparison of the mean of seasonal values to an annual mean is problematic due to the many gaps. In the revised manuscript, we thus now provide a comparison of seasonal emission estimates, based on the monthly gridded EDGAR NO<sub>x</sub> emissions. The comparison to seasonal mean EDGAR yields a correlation  $R=0.72$  and a ratio of means of 0.86, which is similar to the former values based on annual mean EDGAR emissions ( $R=0.76$ , ratio 0.84). Interestingly, correlation did not improve by accounting for seasonality in EDGAR.

Do the estimated emissions show seasonality for cities for which estimates are available for all seasons?

In the revised manuscript, we now present and discuss the seasonality of emissions for those 9 cities where the fit worked for all seasons. This is complemented by EDGAR emission data which is now processed on seasonal basis as well. We have extended section 4.1 by a new figure showing the seasonality of emissions. Some cities (e.g. Tripoli) show hardly any seasonality, while for others (e.g. Algiers) the seasonal emission estimates vary considerably.

See your discussion in Line 200. If higher emissions are expected for winter months, TROPOMI based estimates are low biased because most of them do not consider winter months, and EDGAR does. In your comparison, most cities are close to the 1:1 line.

We agree that the comparison of mean of seasonal means vs. annual means is problematic and could cause a bias in case of gaps, mostly affecting winter. In the revised manuscript, we account for seasonal mean EDGAR emissions, thus this issue is resolved. Interestingly, the ratio of seasonal mean emissions from TROPOMI vs. EDGAR is not much different from that derived for annual means before.

Line 214: Which seasons are considered for Jeddah, Vishakhapatnam, and Copenhagen? Could this explain the overestimation of EDGAR compared to the TROPOMI based emissions? I think it is not given that EDGAR is wrong (too high) here.

For Jeddah and Vishakhapatnam, emission estimates could be derived only for summer and autumn, respectively. For Copenhagen, summer and autumn are available. With the updated comparison based on seasonal emissions, EDGAR values are still considerably larger than the TROPOMI based estimate (for Jeddah by a factor  $>5$ ). As the respective fit results look reasonable, and the discrepancy is already evident in the spatial maps of EDGAR emissions vs. mean VCD, we still conclude that for these locations the EDGAR emissions are probably too high. We have updated the discussion in section 5.3 accordingly.

Line 219: Add mean lifetime with standard deviation.

Done.

Technical corrections:

Line 51: I think m/s should be in exponential notation, please check throughout the manuscript

We have formatted units according to Copernicus guidelines in the revised document.

| Line 68: Change Ozone to ozone

Done.

| Line 83: Add a reference/link and add that you use NOx data

We added a link to EDGAR and specify that we use NOx data.

| Line 86: Change ERA-5 to ERA5 to be consistent

Done.

| Line 94: Change Winter into winter.

Done.

| Line 105/6: Change calm to calm wind conditions

Done.

| Line 110: Delete "the patterns"

Done.

| Equation 1: multiplication should be a dot and not x.

Modified accordingly.

| Line 146: Add NOx

Done.

| Data availability: You could add links to used data like EDGAR, ERA5 wind data, ozone data, and the World Cities Database.

We will provide links and references to the external datasets used in this study in the revised manuscript.