

The authors present a new method for estimating NO<sub>x</sub> emissions and effective lifetimes from large cities. They combined observations for calm as well as two opposite wind directions to shorten the period of data needing for estimation. The method is sound, and the paper is well written. I would recommend minor revisions before publication.

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 comment

1. Additional details for the fitting procedure would be helpful for readers to follow up. For instance, Line 113, "E(x) has the same unit as L(x) (amount per length unit) and corresponds to the line density that would be observed if no wind transport would occur." Does it indicate that  $E(x)=L(x)$  when  $w=0$ ? Does this condition be used for the fit? If not, in Line 120, "the distribution of emission densities E(x), lifetime  $\tau$  and backgrounds b are fitted simultaneously." It looks like too many parameters, each item in E(x), are required to be fit here. How it could be achieved?

We thank the reviewer for the in-depth discussion of our method. Concerning the raised questions:

- E(x) is the distribution of emission as function of x. In case of w being exactly zero, the *calculated* line density L(x) would indeed equal E(x), if there would be no additional smoothing ( $\sigma=0$ ) nor background ( $b=0$ ). The *measured* line densities for calm wind conditions (blue lines in Fig. 4) differ from E(x) (red lines) due to the additional smoothing ( $\sigma=7$  km), additional background b, and wind speeds for calm conditions not being exactly 0, but below 2 m/s.
- E(x), tau and b are fitted to match the observed line densities for forward, backward and calm wind conditions at the same time. The number of parameters (tau, b\_calm, b\_forward, b\_backward, and each item in E(x) (29), i.e. 33 fit parameters in total) are outnumbered by the simultaneous fit of 3 different wind conditions (up to  $3 \times 29 = 87$  constraining data points), resulting in an overdetermined system allowing for a least-squares fit.

In the revised manuscript, we have clarified this procedure by providing more detailed explanations about e.g. the meaning of E(x) and the number of fit parameters vs. number of measurements used for the fit.

2. Seasonal analysis. The big improvement of the method is to shorten the required data period from annual to seasonal. A seasonal analysis of derived emissions will help to clarify this improvement.

The seasonal results for fitted emissions and lifetimes are provided in Table 1. As there are seasons missing for most cities, a combined seasonal analysis (like the mean seasonality of all cities, or cities within a latitude range) is not meaningful. Thus, in the revised manuscript, we 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. However, the resulting seasonal cycles differ with respect to amplitude and patterns amongst the cities as well as between TROPOMI and EDGAR and do not allow for clear conclusions.

3. Section 5.3.1. I'm wondering how much the bias of TROPOMI NO<sub>2</sub> over urban areas contribute to the lower NO<sub>x</sub> emissions?

For v1 of the TROPOMI NO<sub>2</sub> column densities, a low-bias was reported over urban areas (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>). In the PAL dataset, based on the

NO2 processor version v2.3.1, many retrieval steps have been improved. In particular an improved cloud algorithm has been used, resulting in higher cloud altitudes, thus lower air-mass factors and higher NO2 columns. Thus, the reported low-bias is expected to be reduced. However, any remaining bias in the NO2 column would directly affect the presented (as well as any other TROPOMI-based) emission estimate. In the revised manuscript, we have extended section 5.2 accordingly.

| Specific comment

1. Line 74. Too many full stops here?

Lines 74-77 contain a direct quote, where the triple dot is meant to indicate an omission. We will clarify with Copernicus how this should be formatted properly.

| The original ERA5 data is 0.3 degree, but the intermediate meteorological dataset is 1 degree. I suppose additional errors have been introduced by this interpolation. A case study using both the original and interpolated datasets for a few cities would be useful to quantify the level of uncertainties.

We agree that 1° is quite coarse, and we are currently preparing the implementation of high-resolution wind data for future studies. So far, we applied hi-res wind data for the calculation of advection of SO2 fluxes (Adrian Jost et al., in preparation) based on the algorithm developed for NOx (Beirle et al., 2023). For most locations, the resulting emission estimates only change slightly for high vs. low resolution wind fields, but for some it can be significant (up to 20%). On average, emission estimates for high resolution wind fields are higher by about 5%. This systematic difference can be understood, as the analysis considers the wind speed  $w_{\text{model}}$  in wind direction  $\alpha_{\text{model}}$ , while the actual transport in direction  $\alpha_{\text{true}}$  is  $w_{\text{true}} \cdot \cos(\alpha_{\text{model}} - \alpha_{\text{true}})$ . I.e. an error in input wind data (systematic or statistic) results in an underestimation of the actual wind speed, thus an overestimation of  $\tau$ , and an underestimation of  $E$ .

In the revised manuscript, we discuss the impact of uncertainties of wind fields in detail within section 5.2 about errors and limitations. In addition, we add the aspect of horizontal wind fields taken at one selected altitude as general simplification of our method to the conclusions.

1. Figure 2. The period used for averaging is missing in the caption.

We have added the respective time period to the figure caption.

2. Line 133. How many fitted lifetimes are not plausible? A ratio of the failed values is helpful for readers to understand the results.

We thank the reviewer for raising this issue.

From the 11200 combinations (700 cities, 4 seasons, 4 axes), a fit was performed for 2154 cases with sufficient data coverage and appropriate mean wind speeds. After applying the filter for  $\chi^2$ , 1862 valid fit results remain. From these results, 151/34 fits are discarded due to too short/too long lifetime.

While checking the number of skipped cases, we noticed that a further selection criterium was not described in the manuscript so far: Cities are skipped if interfering emissions ( $E(x)$  for  $|x| > 50$  km) are found.

In the revised manuscript, we have revised and extended section 3.6 accordingly.

3. Figure 5. The period used for comparison is missing in the caption.

We have added the respective time period to the figure caption.

4. Section 5.3.1. It is surprised to see the large power plants are missing from TROPOMI NO<sub>2</sub> maps. Could you confirm the locations of power plants by Google map to support the guess that emissions are shift?

We thank the reviewer for this suggestion. We had a closer look to the locations of enhanced NO<sub>x</sub> emissions in EDGAR shown in Fig. 7, and could indeed identify potential NO<sub>x</sub> sources at the respective coordinates:

- The enhanced emissions West from Algiers correspond to the Hadjret en Nouss gas-fired combined cycle power plant at 2.0797°E, 36.5767°N, with a capacity of 1.2 GW.
- The enhanced emissions South from Chelybinsk correspond to power plants in the cities Yuzhnouralsk (54.4552°N, 61.2358°E, [https://www.gem.wiki/Yuzhnouralskaya\\_GRES\\_power\\_station](https://www.gem.wiki/Yuzhnouralskaya_GRES_power_station)) and Troitsk (54.0361°N, 61.6512°E, [https://www.gem.wiki/Troitskaya\\_GRES\\_power\\_station](https://www.gem.wiki/Troitskaya_GRES_power_station)).

It is unclear to us why these power plants are not visible in the TROPOMI NO<sub>2</sub> data, while several other power plants show up as point sources in Algeria as well as in Russia (Beirle et al., 2023). This finding might indicate that those facilities are not running on full capacity for the last years.

Due to these findings, however, we cannot any longer support the hypothesis that the observed mismatch is just caused by a simple displacement of point sources in EDGAR, and skip this part in the revised manuscript.