

## Reply to reviewer #1

The original review is included in grey. Text changes in the revised manuscript are indicated in italic font. References to section numbers are given for both the discussion version of the manuscript (in strike-through mode) as well as the revised manuscript.

This paper presents an improved (version 2) catalog of global NO<sub>x</sub> point source emissions from TROPOMI NO<sub>2</sub> data. Point source rates are estimated on the basis of flux divergence applied to TROPOMI gradients. The paper includes detailed discussion of errors and of improvements relative to version 1, as well as evaluation with power plant data in Germany and the US. The dataset is a very useful compilation, representing a significant advance over version 1, and the error analysis is clearly presented. The Abstract and Conclusions summarize the paper well. The presentation is very good. I recommend publication after consideration by the authors of the following comments.

We thank the reviewer for his/her positive feedback. Below we reply to the specific comments point by point.

1. Figure 2: I found the presentation of AMF, NO<sub>2</sub>/NO<sub>x</sub>, and lifetime as scaling factors not helpful at all. I'm sure that these scaling factors are of interest to the authors but they are not to the reader. I recommend showing the actual quantities in Figure 2, and the scaling factors can be given as statistics in the text.

We thank the reviewer for this critical feedback, pointing out the need for clarification of our motivation for displaying the “scaling factors”, which might seem to be rather abstract quantities.

However, we still consider these scaling factors to be an essential part of the update, as they actually explain the overall higher emission values in v2 by a factor of about 3, and quantify the impact of the different corrections applied:

- For the AMF, the scaling factor indicates how much higher the corrected AMF (for a delta peak at 500 m agl) is over the a-priori value.
- For the NO<sub>x</sub>/NO<sub>2</sub> ratio, the scaling factor directly represents the “actual quantity”, i.e. [NO<sub>x</sub>]/ [NO<sub>2</sub>].
- For the lifetime correction, the scaling factor compensates for the loss of NO<sub>x</sub> within the integrated area (circle with 15 km radius). E.g., a scaling factor of 1.25 would compensate a reduction to 80% of the emitted NO<sub>x</sub> due to chemical loss within the residence time.

In the revised version of the manuscript, we have clarified the meaning of the scaling factors as follows:

Section ~~3.2~~ 3.3:

...  
*Hence, we apply an AMF scaling factor  $c_{AMF} = AMF_{plume}/AMF_{PAL}$ , where  $AMF_{PAL}$  is the tropospheric AMF applied in the PAL product, and  $AMF_{plume}$  is calculated from the AK based on a delta-peak profile at plume height (default 500 m). I.e.,  $c_{AMF}$  reflects how much higher the plume AMF is compared to the a-priori value.*

Section ~~3.3~~ 3.4:

... *by the scaling factor  $c_{NO_x}$  which is calculated based on photo-stationary state ...*

In addition, we now clarify at the end of section 3.3.4 that  
*Below, we consider TVCDs of NO<sub>x</sub> (denoted as V) which are derived from the PAL NO<sub>2</sub> TVCD multiplied by scaling with c<sub>AMF</sub> and c<sub>NO<sub>x</sub></sub>.*

Section 3.10.2:

...

*A scaling factor of e.g. c<sub>τ</sub>=1.25 thus compensates a reduction to 80% of the emitted NO<sub>x</sub> due to the chemical loss within the residence time.*

In the caption of Fig. 2, we now refer to the main text for further details about the scaling factors. In addition, instead of “scaling factor” we now use the more specific terms c<sub>AMF</sub>, c<sub>NO<sub>x</sub></sub> and c<sub>τ</sub> as labels of the subplots.

2. Section 3.3: Inference of the NO<sub>2</sub>/NO<sub>x</sub> ratio is crude and could be a significant source of error needing to be mentioned.

We inferred the NO<sub>x</sub>/NO<sub>2</sub> ratio based on photo-stationary state for each individual TROPOMI pixel. We consider this being far less crude than using a constant value as was done in our earlier studies (Beirle et al., 2011; 2019) as well as in several other recent publications (e.g. de Foy and Schauer, 2022; Dix et al., 2022; Goldberg et al., 2022; Sun, 2022).

Statistical fluctuations of the photolysis frequency, the rate constant, or the ozone concentration are eliminated by the consideration of the temporal mean. Any systematic deviation of J, k or [O<sub>3</sub>] from the used parameterizations/climatologies would indeed affect the resulting NO<sub>x</sub> values. However, a systematic bias of e.g. the ozone concentration of 10% would affect the NO/NO<sub>2</sub> ratio by about 10%, but the NO<sub>x</sub>/NO<sub>2</sub> ratio would change by only 3%. Thus we do not consider the calculated NO<sub>x</sub>/NO<sub>2</sub> ratios to be a general major source of uncertainty for the presented results.

*J* should depend on surface reflectivity and it's a bit embarrassing that this is not recognized since it is key to the NO<sub>2</sub> retrieval.

We agree that ignoring the surface albedo within the parameterization of *J* is a simplification that systematically affects the results for scenes with high albedo (deserts). According to Augustsson, 1981, a change of the surface albedo from typically 5% to 25% over deserts increases *J* (and thus the NO/NO<sub>2</sub> ratio) by a factor of about 1.3. This increase corresponds to an increase of the NO<sub>x</sub>/NO<sub>2</sub> ratio of about 5% (based on an a-priori NO<sub>x</sub>/NO<sub>2</sub> ratio of about 1.2 over deserts, see Fig. 2 in Beirle et al., 2021).

In the revised version of the manuscript, we shortly discuss this systematic error at the end of a new subsection 5.3.1 discussing the PSS assumption:

*Additional systematic errors might be introduced by the parameterization of J as function of the SZA, which so far ignores the impact of the surface albedo, causing a low bias of the [NO<sub>x</sub>]/[NO<sub>2</sub>] ratio over deserts of about 5%. This parameterization might be improved in future studies.*

The ozone climatology likely does not apply to the plume where ozone would be titrated at least in the near-field (and 15 km is close to that). I see no support for the authors' claim that PSS is a reasonable assumption on the 15-km scale. Plume chemistry can be very weird. It would be good to discuss the literature for direct measurements of the NO<sub>2</sub>/NO<sub>x</sub> ratio in plumes.

We thank the reviewer for raising this issue, and agree that PSS might not always be reached within 15 km, in particular for the strongest emitters. In the revised manuscript, we have added a new subsection to the discussion of systematic errors:

#### *5.3.1 Photostationary state*

*The scaling of NO<sub>2</sub> observations to NO<sub>x</sub> is based on PSS assumption. This is typically not fulfilled directly at a strong point source due to the added emissions which take place largely in form of NO. These NO emissions are converted to NO<sub>2</sub> during plume travel and will thus be detected by the spatial gradient not at, but downwind from the source, which would cause a smearing out of the peak in the advection map. Note, however, that the width of the advection plume of about 5 km (Beirle et al., 2021) is of the order of the TROPOMI pixel size, and we do not observe a significant downwind shift in the advection (or divergence) signal.*

*Nevertheless, we cannot rule out that PSS is not always reached completely within 15 km. Janssen et al., 1988 parameterized the deviation from PSS as function of downwind distance based on actual aircraft measurements of power plant plumes. The deviations from PSS at 15 km distance have been found to be about 2% for summer (based on  $\alpha=0.25/\text{km}$ , see Table 4 in Janssen et al., 1988) up to 24% in spring and autumn for low background ozone concentrations (based on  $\alpha=0.1/\text{km}$ , see Table 3 in Janssen et al., 1988), which would cause a corresponding low bias in the estimated emissions.*

*Increasing the considered radius to e.g. 20 km would reduce the possible bias of the emission estimate due to non PSS. On the other hand, this would have other negative impacts:*

- some of the detected point sources could not be separated any more,*
- the interference with other sources around the point source would increase, and*
- the uncertainty of the lifetime correction, which is based on the residence time derived from the wind speed at the point source, would increase.*

*Thus, we stick to the choice of the 15 km radius in this study.*

In addition, we also list this effect in the concluding discussion in section ~~5.3.5~~ 5.3.7, and increase the estimated overall low bias to “up to 40%”.

3. Section 3.8: I don't understand the iterative nature of the procedure, and this is worth explaining better because it's brought up in a number of places including the abstract. As I (maybe incorrectly) see it, the procedure applies successively lower thresholds to the flux divergence thresholds. It's not really iterative except that previously identified plumes are removed from the dataset. I may not have it right, which is the point. Clarify.

We use the term “iteration” as “the repetition of a process in order to generate a ... sequence of outcomes. Each repetition of the process is a single iteration, and the outcome of each iteration is then the starting point of the next iteration.” (Wikipedia). The “sequence of outcomes” is the list of candidates, and the “starting point of the next iteration” is the advection map where the previous candidate has been removed. We point out the iterative nature of the plume identification in contrast to other algorithms like e.g. pattern recognition.

4. Section 3.9.2: do we understand the chemistry behind this NO<sub>x</sub> lifetime of 2 hours at low latitudes, 4-6 hours at higher latitudes? My recollection is that the mechanism for

fast NO<sub>x</sub> oxidation in power plant plumes remains a bit of a mystery considering that ozone titration would be expected. It would be good to review some of that literature.

Several studies report on a generally short NO<sub>x</sub> lifetimes of about few hours for power plant plumes based on satellite measurements (Goldberg et al., 2019, 2022; Lange et al., 2022). Similar short lifetimes of about 3 hours have been reported in Ryerson et al., 1998, based on aircraft measurements.

This is consistent with enhanced OH concentrations reported in power plant plumes (Kim et al., 2016; de Gouw et al., 2019). Thus we see no fundamental contradiction in atmospheric chemistry with respect to short NO<sub>x</sub> lifetimes.

Surely the lifetime parameterization should be a function of season as well as latitude.

We added the following paragraph to section ~~3.9.2~~ 3.10.2:

*Note that the seasonal dependency of the NO<sub>x</sub> lifetime has been found to be rather weak (probably due to the focus on cloud free conditions around noon), while seasonal estimates have larger uncertainties due to reduced statistics (Lange et al., 2022). Thus, we do not consider a possible seasonal dependency of the NO<sub>x</sub> lifetime explicitly. In addition, high variability of lifetimes at different locations of similar latitude has been reported e.g. in Laughner and Cohen (2019). Thus we assume a rather large uncertainty of 50% for  $\tau$  (see Section 3.12.1).*

5. Equation (10): I don't understand the double integral. Shouldn't it be a contour integral?

The point source emissions are derived from the advection map, which provides rate densities (mass per time per area), by spatial integration over the considered area of a circle with 15 km radius (yielding mass per time). Thus Eq. 40 11 does not describe a contour integral, but a double (area) integral.

We see the potential confusion by Eq. 40 11 due to the complicated integral bounds, while the corresponding procedure in the code is a simple summation of advection values of the grid pixels within 15 km, multiplied with the respective pixel area. We tried to reduce the confusion by replacing the explicit integral bounds by a more symbolic notation indicating the "circle area".

6. Section 3.11.1: I don't understand the 'statistical error' terminology in that section. Are you referring instead to variability, such as standard deviation?

The "statistical error" refers to the standard error of the temporal mean, i.e. the temporal standard deviation divided by the square root of the sample size. This is clarified in the revised manuscript.

7. Section 3.11.3: a plume height of 300 m vs. 500 m is not enough to characterize the uncertainty in the AMF. In early afternoon when TROPOMI observes, vertical mixing up to the PBL depth (typically 2 km) can take place within 1 hour. That may give you an AMF error of more like 10%.

Within the error formalism, the statistical uncertainty of the AMF is estimated from the temporal variability of the AMF scaling factor and is found to be of the order of up to 5% (section ~~3.11.1~~ 3.12.1, Fig. 6 (b)).

With respect to the assumed plume height, the focus of this study is set to the

horizontal transport close to the point source, where spatial gradients (and thus the advection) are largest. Thus we consider a plume height representative for power plant plumes shortly after release rather than a completely mixed PBL.

In response to this comment as well as the comments raised by reviewer #3, we have extended the discussion of the plume height in the revised manuscript in a new dedicated subsection:

### 3.2 Effective plume height

*In this study, horizontal transport is described by horizontal wind fields at a fixed “plume height”. This is a simplifying assumption, as the emissions take place at stack height of about 200 m, but are uplifted and vertically mixed within the boundary layer during downwind transport.*

*For the quantification of point source emissions, the focus of this study is set to the horizontal transport close to the point source, where spatial gradients are largest. As shown in Kuhn et al., 2022, power plant emissions at 200 m stack height quickly rise to about 500 m within the first hundred meters.*

*Brunner et al. (2019) investigated the effective height of CO<sub>2</sub> emissions for atmospheric transport simulations. This is closely related to the question which altitude has to be considered in order to describe horizontal transport of a fresh power plant plume appropriately. For summer around noon, they report mean effective heights of about 450 m (with a long tail towards larger values).*

*In this study, we assume an effective plume height of 500 m above ground level (agl). For individual stations and specific meteorological situations, systematic deviations might occur. In order to quantify the impact of this assumption, we thus also performed the analysis for a plume height of 300 m (see section ~~3.11.3~~ 3.12.3).*

*ERA5 wind fields are vertically interpolated to the assumed plume height. In addition, the AMF correction is applied consistently for the same height (see section ~~3.2~~ 3.3).*

#### 8. Line 421: Appendix ??

We corrected line 421 to

*“Additional tables ... are provided in the Supplement for various regions.”*

#### 9. Table 1: the last column is key to understanding the factor of 3-4 increase relative to version v1, so I would give it a more helpful title and I would identify the principal contributors to the factor of 3-4 increase in the text.

We thank the reviewer for this proposal and modified the column title to “*impact on v2 emission estimate compared to v1*”. In addition, we extended the discussion in section ~~3.13~~ 3.14 by including the numbers of the main contributors to the factor of ~3.

## Additional references

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