

Reply to reviewer #3

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.

Nice job with this new version of the algorithm. All comments are minor.

We thank the reviewer for the positive assessment of our study. Below, we refer to the specific comments one by one.

One overarching comment is in the lack of literature on the realistic nature of both the effective plume heights and the wind speed used. I am glad you performed a sensitivity analysis in Section 3.11 showing the effects of wind speeds on emission estimates, but I do think some literature supporting this analysis is warranted. I am not requesting additional analyses, but instead a longer discussion, including additional literature, on both any potential biases in the ERA5 wind speed/direction and how realistic a 500-m effective plume height is. For example is there a global bias or any regional variation in a wind bias? How is performance of the wind product near coastlines? Are there any references - using in situ aircraft observations - demonstrating that a 500-m plume height is approximately correct mean value?

We agree that our choice of a 500 m effective plume height needs further motivation and discussion. Thus, we extended the discussion of the plume height in a new subsection 3.2 in the revised manuscript as follows:

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 CO₂ 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)

Other minor comments:

Line 39. “far higher and thus more realistic” —> “larger and more realistic”. The word “higher” could refer to height in the atmosphere.

Done.

Line 63. Mention that the 500-m wind is used, and that a sensitivity analysis is performed with the 300-m wind.

In the revised manuscript, we added this information to the end of section 2.2.

Line 74. I see in Line 154 that you provide a range of NO_x/NO₂ ratios that were used range of NO_x/NO₂ ratios that were used, but maybe include here also?

Section 2.3 (line 74) is meant to describe the input data, here the ozone climatology. The actual calculation of the NO_x/NO₂ ratio is described later in section 3.3 (line 154), thus we provide the range of NO_x/NO₂ ratios in section ~~3.3~~ 3.4.

And quickly discuss (in either section) where ratios may be higher and lower?

The calculation of the NO_x/NO₂ ratio is the same as in v1 of the catalog (Beirle et al., 2021). In Beirle et al., 2021, some additional information is provided, in particular a map showing the spatial distribution of the NO_x/NO₂ ratio (Fig. 2 therein). In the revised manuscript, we have added this information and the respective reference to Beirle et al., 2021 to section ~~3.3~~ 3.4.

Line 86. Remove “Basically”

Done

Line 105. In the US, the CAMPD is used most often (<https://campd.epa.gov/>). I am assuming eGRID and CAMPD are identical. Can you confirm? And if so, can you add a sentence in the text mentioning this?

According to eGRID FAQs (<https://www.epa.gov/egrid/frequent-questions-about-egrid#egrid1>), “eGRID uses data from the [Energy Information Administration \(EIA\) Forms EIA-860 and EIA-923](#) and [EPA’s Clean Air Markets Program Data](#).” We have added this information to the revised manuscript.

Line 121. Are there any references - using in situ aircraft observations - demonstrating that a 500-m plume height is approximately correct mean value? It’d be great for you to check out the literature and see if you can find anything.

We have extended the discussion of the plume height and the reasons for choosing 500 m, see the reply above to the overarching comment.

Line 138. This is great, thank you for adding! But can you add a bit more detail? I’m not fully following how the new AMF is calculated. This section would benefit greatly from an illustrative figure showing a standard a priori profile vs an a priori profile with the “excess” at 500-m.

The assumed profile shape of the excess plume is just a δ -peak at 500 m.
We have extended the respective section in the revised manuscript as follows:

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. I.e., c_{AMF} reflects how much higher the plume AMF is compared to the a-priori value.

Line 178. When you say “this procedure” are you referring to de Foy and Schauer? I think yes, but can you make this clearer?

We reformulated line 178 to “The main advantage of taking derivatives directly on TROPOMI grid is...”

Line 220. Local maxima during the May 2018 - Nov 2021 average? Or daily average? I can imagine the latter is much harder.

We have added “... *the temporal mean (May 2018 - Nov 2021) advection map*” to line 220.

Line 255. Define “area” source. Does this include mobile/vehicle emissions? Or something else?

Within the candidate classification algorithm, local maxima in the advection map are classified by different criteria. Peaks covering a larger area are labelled as “area source” (lines 247-248); this term just refers to the finding of a spatially extended advection maximum. There might be different reasons causing such broad peaks in the advection map, e.g. cities (vehicle emissions) as well as extended industrialized areas, or multiple power plants within about 10-20 km distance. In the revised manuscript, we have extended the candidate classification description and clarified what is meant by “area source”:

Such broad peaks in the advection map might be caused by cities (vehicle emissions) as well as extended industrialized areas, or multiple interfering point sources within about 10-20 km distance.

Line 291. The NO₂ lifetime is also function of the atmospheric composition (both total NO₂ and VOCs) as well. I could imagine two locations at the same latitude and with similar wind speeds having differing NO₂ lifetimes based on ambient atmospheric composition (Figure 1; <https://www.science.org/doi/10.1126/science.aax6832>). It seems like you are not accounting for this in your lifetime derivation. Is that a correct assumption? If so, please state explicitly, and discuss in here or in the Discussion section that this would be an opportunity for further improvement of the methodology.

We thank the reviewer for raising this issue. In fact, we have tried to perform individual lifetime estimates for each point source based on an approach similar to Beirle et al., 2011. However, the associated uncertainties were found to be considerably large, and for several point sources (in particular the weaker ones) the algorithm fails. Thus we made use of the simple dependency of latitude reported in Lange et al. (2022). We address the variability of NO_x lifetimes at same latitude, as reported in e.g. Laughner and Cohen, 2019, by assuming an uncertainty of 50% for τ and added this information to section ~~3.9.2.~~ 3.10.2.

In addition, we added a new subsection to the discussion of systematic errors dedicated to the lifetime:

5.3.6 Lifetime correction

The lifetime correction (section 3.9.2 3.10.2) is based on a simple parameterization of τ as function of latitude. However, the OH concentration depends on several parameters like VOC concentrations, as well as on NO_x concentration itself, and Laughner and Cohen (2019) report on systematically different lifetimes for locations at comparable latitude.

Thus we assumed a rather large uncertainty of 50% for τ . Still, the lifetime correction might be biased for locations where τ deviates systematically from the parameterization proposed by Lange et al., 2022. In future studies, uncertainties might be reduced by accounting for the actual lifetime for each individual power plant. However, this will be particularly challenging for the weaker sources.

Line 417. In the Supplement, I noticed that some of the power plants (the ones in Canada and the US at least) didn't match exactly. Instead of saying "First match only", perhaps say "Potential match"

We agree that the phrase "first match only" might be misleading and modified the footnote to "GPPD power plants / WCD cities within 15 km. Here, only the first match is listed. In the original catalog, all matches are included. Note that the listed power plant or city does not need to represent the actual dominating NO_x source."

Line 421. I think you meant to say in the "Supplemental Material"

We corrected line 421 to

"Additional tables ... are provided in the Supplement for various regions."

Line 425. Tab 2? I think you meant to say "For five of the point sources listed in Table 2...".

We corrected line 425 accordingly.

Line 458. Seems like more than 41 points are in Figure 13. I could be wrong though. Please double check

Figures 12 & 13 display annual means. As PRTR and eGRID data is considered for the years 2018-2020, there are up to 3 data points shown in Figures 12 & 13 per identified point source. We have clarified the figure captions accordingly.

Additional references

Brunner, D., Kuhlmann, G., Marshall, J., Clément, V., Fuhrer, O., Broquet, G., Löscher, A., and Meijer, Y.: Accounting for the vertical distribution of emissions in atmospheric CO₂ simulations, *Atmos. Chem. Phys.*, 19, 4541–4559, <https://doi.org/10.5194/acp-19-4541-2019>, 2019.

Laughner, J.L., Cohen, R.C., Direct observation of changing NO_x lifetime in North American cities. *Science* 366, 723-727. DOI:10.1126/science.aax6832, 2019.

Kuhn, L., Kuhn, J., Wagner, T., and Platt, U.: The NO₂ camera based on gas correlation spectroscopy, *Atmos. Meas. Tech.*, 15, 1395–1414, <https://doi.org/10.5194/amt-15-1395-2022>, 2022.