

We would like to thank reviewer 2 for his/her comments and suggestions.

Response Reviewer 2

- 1. Satellite images only contain information of NO₂, but NO_x from the emission inventories includes NO and NO₂. In emission inventory, the ratio of NO_x to NO₂ is different from the ratio in ambient concentration. The conversion of NO to NO₂ changes the ratio. The study uses a factor of 1.32 (based on ambient concentrations) for all sources definitely leading to uncertainties.**

Response:

The reviewer is correct in stating that the 1.32 factor shows some variability typically depending on the atmospheric concentrations of NO₂, NO and Ozone. The choice for 1.32 was based on the value used by Beirle et al., 2019 who in turn based is on the ratio given by Seinfeld and Pandis (2006) over regions under polluted conditions around noontime. Depending on the season and latitude the ratio can shift significantly. More recent studies give values ranging between 1.22 (Riyadh, Beirle et al., 2021, based on modelling) and 1.54 (South Africa, Lange et al., 2022). Furthermore, a study by Griffin et al. (2021) reported NO_x:NO₂ ratios based on aircraft measurements and model simulations near a biomass burning source, and concluded on a ratio between 1.3-1.5 near the source. To test the representativity for Germany as a whole, we used a simulation with the regional transport model LOTOS-EUROS over 2019 to calculate the NO_x:NO₂ regions throughout the year for the hours around the TROPOMI overpass (LOTOS-EUROS version 2.2, Manders et al., 2017, more details on simulation on request). The simulated yearly mean averaged values of NO_x:NO₂ range between 1.3 for northern regions further away from major emissions, and about 1.5 on top of major industrial sources such as power plants and around the more elevated regions. The standard deviation of the daily values (at around the TROPOMI overpass) were also calculated with typical values around 0.1-0.15 and the largest values (<0.3) calculated around the major emission regions (e.g. Powerplants, Ruhr industrial area, Hamburg). The mean values over Germany for 2019 are 1.39 with a standard deviation of 0.16. Both the more recent Beirle et al., (2021) value of 1.41 (for Germany) and our earlier choice of 1.32±0.26 are within agreement with our simulated results. As variations can be expected from year to year we stick with the earlier value of 1.32 and add the standard deviation of 0.26 to our uncertainty estimate.

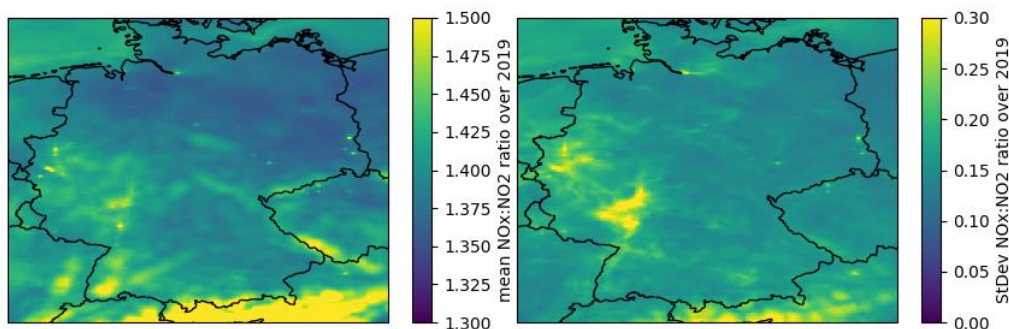


Fig. R2.1 Yearly Mean and StDev of NO_x:NO₂ ratios for 2019.

Changed lines 285-288 to: “TROPOMI is only capable in observing NO₂, therefore an additional correction is needed to account for the NO mass. The NO_x to NO₂ concentration ratio depends on the local chemistry with values commonly falling within the 1.2-1.5 range for polluted regions (Beirle et al., 2011, Beirle et al., 2019, Beirle et al., 2021, Lange et al., 2022). In this study we apply the 1.32±0.26 factor as used by Beirle et al., 2019 and include the standard deviation of 0.26 further into the uncertainty budget account for the variations.”

Lange et al., 2022 (Lange, K., Richter, A., and Burrows, J. P.: Variability of nitrogen oxide emission fluxes and lifetimes estimated from Sentinel-5P TROPOMI observations, *Atmos. Chem. Phys.*, 22, 2745–2767, <https://doi.org/10.5194/acp-22-2745-2022>, 2022.)

Beirle, S., Borger, C., Dörner, S., Eskes, H., Kumar, V., de Laat, A., and Wagner, T.: Catalog of NO_x emissions from point sources as derived from the divergence of the NO₂ flux for TROPOMI, *Earth Syst. Sci. Data*, 13, 2995–3012, <https://doi.org/10.5194/essd-13-2995-2021>, 2021

Manders, A. M. M., Bultjes, P. J. H., Curier, L., Denier van der Gon, H. A. C., Hendriks, C., Jonkers, S., Kranenburg, R., Kuenen, J. J. P., Segers, A. J., Timmermans, R. M. A., Visschedijk, A. J. H., Wichink Kruit, R. J., van Pul, W. A. J., Sauter, F. J., van der Swaluw, E., Swart, D. P. J., Douros, J., Eskes, H., van Meijgaard, E., van Uft, B., van Velthoven, P., Banzhaf, S., Mues, A. C., Stern, R., Fu, G., Lu, S., Heemink, A., van Velzen, N., and Schaap, M.: Curriculum vitae of the LOTOS–EUROS (v2.0) chemistry transport model, *Geosci. Model Dev.*, 10, 4145–4173, <https://doi.org/10.5194/gmd-10-4145-2017>, 2017.

2. **Photochemical reactions are different among seasons and day-night. The life time of 4 hours for NO₂ uniformly seems unreasonable for all days during 2019-2021. Radiation could be a good indicator for the lifetime.**

Response:

As the reviewer points out the lifetime of NO₂ varies throughout the year. The effective lifetime of NO_x depends on both the chemical decay rate and loss to surfaces (dry deposition). Of these two the chemical decay is the dominant factor. While radiation can be a good indicator, the lifetime is typically estimated via the availability of OH and production thereof (typically including radiation). Several studies have explored this route before and either estimate the availability of OH by some basic assumptions on production, or by using modelled OH fields (with the drawback of a potential bias within the simulated concentrations). Either route is possible and estimates for the effective lifetimes end up around 2-5 hours for spring and summertime values (Lorente et al., 2019; Valin et al., 2013). Outer estimates for winter are 12-24 hours (Shah et al., 2020).

Several studies report on effective lifetimes derived from fits to observed plumes from cities and large industrial areas. These values typically give a range between 2-5 hours (Goldberg et al., 2021, Fioletov et al., 2022, Beirle et al., 2011, Lange et al., 2022) with a recent study by Fioletov et al. (2022) giving a value of 3.3 hours representative for larger emissions within the US and Canada (2018-2022). Furthermore, Fioletov et al. (2022) also notes that while lifetime has a large impact on the emission estimates, relative changes do not have a major impact when comparing individual years to one another. They point out that 1h deviations from the 3.3 hour mean only changed the emission estimates between years by about 1%.

Besides estimates of lifetime based on observations, we can also look at simulated lifetimes within chemistry transport models. While our LOTOS-EUROS chemistry model has no option to directly write out lifetime, we can look at earlier studies that used a tagging approach to label emissions from individual hours. An earlier study by Curier et al., 2014 did just that to study the source sector contribution of emissions from individual hours to the OMI NO₂ column at OMI overpass for several industrial regions in Europe. For the region most representative of Germany (Benelux) the study states: "Approximately 50% of the modelled OMI signal results from NO_x emissions in the 3 h prior to OMI overpass.". This statement holds for most the source sectors. Assuming a relatively constant source this translates to a lifetime of about 4 hours (at column level, and assuming basic mass balance).

A potential point of concern remains the representativity for the whole year. Most of the estimates are biased towards spring, summer and autumn as there are typically more observations available within these months. To correct for the representativity bias we already include a seasonal variation factor (1.11), but also remain on the high end of the lifetime estimates by choosing a value of 4.0 hours. The standard deviation of +/-1 hour ensure that common values within 3-5 hours remain within the uncertainty range.

Changed lines 258-259:

The effective lifetime of NO_x depends on both the chemical decay rate and loss to surfaces (dry deposition). Within our domain of interest the chemical decay will be the dominant factor. Earlier studies using the EMG plume functions derived lifetimes between 2-5 hours based on the decay downwind of major sources worldwide \citep{Beirle_2011, deFoy_2015,Goldberg_2021, Lange_2022, Fioletov_2022}. Following those results we assume a mean lifetime of 4 hours +/- 1 hour to account for local and seasonal variations.

Curier, R.L., Kranenburg, R., Segers, A.J.S., Timmermans, R.M.A. and Schaap, M., 2014. Synergistic use of OMI NO₂ tropospheric columns and LOTOS-EUROS to evaluate the NO_x emission trends across Europe. *Remote Sensing of Environment*, 149, pp.58-69.

Fioletov, V., McLinden, C. A., Griffin, D., Krotkov, N., Liu, F., and Eskes, H.: Quantifying urban, industrial, and background changes in NO₂ during the COVID-19 lockdown period based on TROPOMI satellite observations, *Atmos. Chem. Phys.*, 22, 4201–4236, <https://doi.org/10.5194/acp-22-4201-2022>, 2022.

Lorente, A., Boersma, K., Eskes, H., Veefkind, J., Van Geffen, J., De Zeeuw, M., Denier Van Der Gon, H., Beirle, S., and Krol, M.: Quantification of nitrogen oxides emissions from build-up of pollution over Paris with TROPOMI, *Scientific reports*, 9, 1–10, 2019

Shah, V., Jacob, D. J., Li, K., Silvern, R. F., Zhai, S., Liu, M., et al. (2020). Effect of changing NO_x lifetime on the seasonality and long-term trends of satellite-observed tropospheric NO₂ columns over China. *Atmospheric Chemistry and Physics Discussions*, 20(3), 1483– 1495. <https://doi.org/10.5194/acp-2019-670>

Valin, L. C., Russell, A. R., & Cohen, R. C. (2013). Variations of OH radical in an urban plume inferred from NO₂ column measurements. *Geophysical Research Letters*, 40(9), 1856– 1860. <https://doi.org/10.1002/grl.50267>.

3. The comparison of inventory and Tropomi in Figure 3 is not clearly, showing the difference is better for readers.

Response: The requested difference plots between the inventory and TROPOMI based emissions are already given in 5A-F.

4. The abstract needs to be modified as the currently version is not clear about the method and the key results.

Response: Rewrote and shortened the abstract to,

“NO_x is an important primary air pollutant of major environmental concern which is predominantly produced by anthropogenic combustion activities. NO_x needs to be accounted for in national emission inventories, according to international treaties. Constructing accurate inventories requires substantial time and effort, resulting in reporting delays of one to five years. In addition to this, difficulties can arise from temporal and country specific legislative and protocol differences. To address these issues, satellite-based atmospheric composition measurements offer a unique opportunity for independent and large-scale estimation of emissions in a consistent, transparent, and comprehensible manner. Here we test the multi-source plume method (MSPM) to assess the NO_x emissions over Germany in the Corona period from 2019-2021. For the years where reporting is available, the differences between satellite estimates and inventory totals were within 75-100 kt (NO₂) NO_x (<10% of inventory values). The large reduction of NO_x emissions (~15%) related to the COVID-19 lock-downs was observed in both the inventory and satellite derived emissions. The recent projections for the inventory emissions of 2021 pointed to a recovery of the 2021 emissions towards pre-COVID-19 levels. In the satellite derived emissions however, such an increase was not observed. While emissions from the larger power-plants did rebound to pre-COVID-19 levels, other sectors such as road transport did not, likely due to a reduction in the number of heavier transport trucks. This again illustrates the value of having a consistent satellite based methodology for faster emission estimates to guide and check the conventional emission inventory reporting. The method described in this manuscript also meets the demand for independent verification of the official emission inventories, which will enable inventory compilers to detect potentially problematic reporting issues, bolstering transparency and comparability: two key values for emission reporting.”

5. The discussion of uncertainties is qualitative rather than quantitative. A ranking of uncertainties from different assumptions is helpful for assessing the results when this method is used for other cases.

Response: The discussion on the uncertainties (lines 430-479) has been moved and extended to form its own section (2.2.2, from line 290 onward) which can be read in the updated version of the manuscript. Additionally a table (1) has been added to summarize the individual uncertainties/errors. Some further explanation has been given for the individual error terms, linking back to earlier sections in the manuscript where needed. The discussion section has also been shortened to account for the moved section.