Authors' Response to Reviews of

Estimating NO_x emissions of stack plumes using a high-resolution atmospheric chemistry model and satellite-derived NO_2 columns

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RC: ***Reviewers' Comment***, AR: Authors' Response, ☐ Manuscript Text

1. Reviewer #2

We thank the reviewer for the positive evaluation of our manuscript. Concerning the minor revisions required, we answer point by point below.

RC: ***1) The abstract of this manuscript is quite distracting. All CO2 related remarks should go to the later part of introduction or discussion section. The main topic in this manuscript is NOx emissions from point sources related to LES model simulations and TROPOMI tropospheric NO2 column observations. Furthermore, the sentence "Moreover, results indicate that common assumptions about the NO2 lifetime (4 hours) and NOx:NO2 ratios (1.3) in simplified methods that estimate emissions from NO2 satellite data (e.g. Beirle et al., 2019) need revision" needs to be revised. This targets only specific studies and does not give broad implications and directions.***

AR: The reviewer makes a valid point here. Indeed, the final manuscript is about chemistry-enabled LES, focusing on comparisons to TROPOMI NO_2 satellite data. We will rewrite the abstract and introduction accordingly.

We do not fully grasp what is meant by "This targets only specific studies and does not give broad implications and directions". A recent study (Meier et al., 2024) based on the output of our LES runs develops a general algorithm to account for the NO_x:NO_2 ratio in the analysis of the TROPOMI data. In addition, the role of the NO_2 lifetime seems to be of general importance in the interpretation of NO_2 satellite data, as has been shown in many papers. However, we will omit the reference from Beirle et al. 2019 and simply refer to 'the common assumptions'.

RC: ***2) This manuscript deals with the classical nonlinear relationship between NOx and OH. The authors several times referred to Rohrer et al. (2014) for recycling of OH. Rohrer et al. (2014) is mainly concerning about a new recycling process generating OH under very low NOx and high biogenic VOC condition. I don't think that this manuscript is closely related to Rohrer et al (2014). There would be better references for this. Meanwhile, NOx lifetime estimations and related discussions based on satellite observations can be found in Valin et al (2013) and Laughner and Cohen (2019) and references therein.***

AR: We replaced the Rohrer et al. reference by other references to give proper credit to the historical developments. Specifically, we will refer to: Ehhalt and Rohrer (2000); Lelieveld et al. (2002); Valin et al. (2013).

RC: ***3) It would be beneficial to include the plot for BEL2 from Figure 2 alongside the three plots of the LES model NO2 columns (projected to the TROPOMI pixels) in Figure 11. This addition would facilitate one-to-one comparison.***

AR: Thanks for the suggestion. Below updated figure 11.

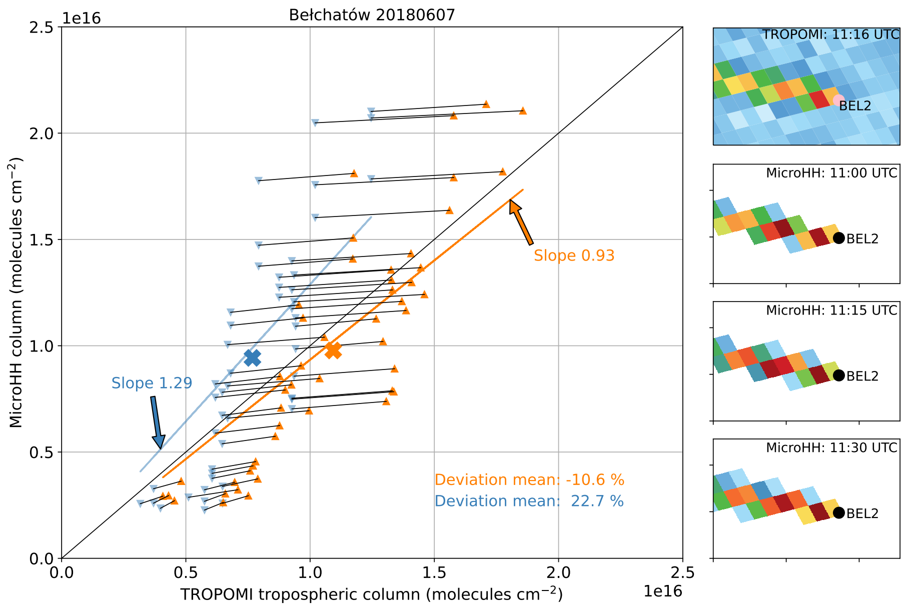

Figure 1: Comparison of simulated and TROPOMI tropospheric NO_2 columns for simulation BEL2. The upper right panel repeats the TROPOMI data from Fig. 2. The three panels below are simulation snapshots 15 minutes apart around TROPOMI overpass time and are used to account for variations in the turbulent field. Domain and color bar as in Fig. 2, but in the range 0–2×10^{16} molecules cm^{-2}. These fields are coarsened to TROPOMI resolution and filtered for enhanced NO_2 mixing ratios to identify the plume (see text). The blue dots and line show comparisons for TROPOMI data that have not been corrected for the AMF (Eq. 4). Moreover, simulated profiles have not been augmented with CAMS NO_2 in the free troposphere (i.e. 0–4 km). The orange dots show the corrected and augmented values, with corrected (orange) and uncorrected (blue) points connected by thin grey lines. Orange and blue lines and slope values represent fits that are forced through the origin. The orange and blue crosses represent the plume-mean columns and mean deviations ((model-tropomi)/tropomi in %) are given in the lower right corner.

RC: ***4) It is not clear that the emission the model used is based on the bottom-up emission inventories and the one measured at each stack. If we consider TROPOMI tropospheric NO2 columns as a ground truth value, the agreement between the model and satellite gives a confidence in the bottom-up emission from this power plant, probably as shown in this study. While the TROPOMI data appear promising, additional validation and calibration would be necessary, particularly for observations near power plants. If the authors utilized the observed NOx emission from the power plants for their simulations, the agreement between the model and satellite data provides insight into the accuracy of the TROPOMI columns. Thus, the story changes, depending on the nature of emission in the model.***

AR: As written in the manuscript, the simulations were performed based on the protocols that were written at the start of the CoCO2 project (`https://coco2-project.eu/sites/default/files/2021-07/CoCO2-D4.1-V1-0.pdf`). Our trust in the emissions used in the simulation depends on the case. As also outlined in Meier et al. (2024), estimated emissions of the European power plants Jänschwalde and Bełchatów are based on yearly data from the European Pollutant Release and Transfer Register (E-PRTR). For these simulations, we used the bottom-up reported emissions to compare to TROPOMI NO_2 columns to detect possible discrepancies. We obtained data as annual NO_x emissions from the Jänschwalde power plant for the year 2018. For the Bełchatów power plant, the data are only available up to 2017, and emissions for 2017 were used.

For the Matimba case, we use the emissions averaged over the year 2018, based on the monthly reports provided by the responsible company Eskom (https://www.eskom.co.za). Earlier studies (Hakkarainen et al., 2021) showed that TROPOMI-based annual NO_x emissions are slightly lower than the annual value reported by the company. However, the difference remained within the errors in the estimation method.

However, for the Lipetsk metallurgical plant, we had to make a rough estimate because no accurate data on emissions are available. We therefore estimated the emission on the basis of an annual report of the operating company NLMK. However, from the report, it is unclear whether the reported emissions are exclusively from the metallurgical plant in Lipetsk only and whether emissions from the captive power plants at the Lipetsk site are included in the reported emissions. Thus for the Lipetsk case, TROPOMI tropospheric NO_2 columns are used as a ground truth value. Reinforcing the accuracy of TROPOMI and the comparison procedure, we find the poorest agreement for the Lipetsk case.

To clarify this issue in the manuscript, we added the following lines to the document

> The emissions are based on the CoCO2 intercomparison protocol: `https://coco2-project.eu/sites/default/files/2021-07/CoCO2-D4.1-V1-0.pdf`. For JAE and BEL,these emissions are based on reported yearly total values in the European Pollutant Release and Transfer Register (E-PRTR). For MAT, we used the average emissions for the year 2018, based on the monthly reports provided by the responsible company Eskom (`https://www.eskom.co.za`). For LIP, emissions are obtained from the 2019 annual report of the NLMK group (`https://nlmk.com/en/ir/reporting-center/annual-reports/`). We have smaller confidence in these LIP emissions, because it remains unclear whether the reported emissions can be fully ascribed to the Lipetsk facility.

RC: ***5) The analysis of Is and NO2 lifetimes for the different power plants is valuable. However, it would be better if Is and NO2 lifetime are also calculated for the pixels of TROPOMI or larger source box (like 100 km x 100 km) for interpretation of real-world problems. Beirle et al (2019) noted the specific condition for which the assumption of NOx/NO2 = 1.3 is valid. It is needed to interpret the results in this study in line with Beirle et al (2019) or similar studies and other research that adopted larger source boxes.***

AR: We tend to disagree with this argument. For the problem of mixing chemicals in a plume (I_s) and the NO_2 lifetime, we considered it useful to analyse what happens as a function of distance from the stack at high spatial resolution, and this is what we did in the paper. The question of how the overall effect is observed by a satellite instrument, such as TROPOMI, is another issue. We consider it, however, instructive to show the implications on TROPOMI resolution. We will show these results in a Supplement to the manuscript. Figures 2, 3, 4, and 5 show results for I_s, the lifetime of NO_2, the lifetime of NO_x, and the NO_x to NO_2 ratio, respectively. Note that for the MAT case, the lifetime of NO_2 in the background gets very long, due to depletion of NO_x by oxidation in the extensive domain. As mentioned in the manuscript, we focused on emission of a single stack, and added no surface emissions of e.g. traffic. Note also that these figures confirm the finding in the paper that the BEL1, MAT1, and MAT2 plumes remain chemically intact for larger downwind distances.

RC: ***6)The recommendations for future calculations of NOx emissions from stack plumes remain unclear in this study. Should the LES method be applied for all power plants worldwide? Can TROPOMI tropospheric NO2 columns sufficiently provide NOx emissions estimations from these sources?***

AR: We believe that our paper, for the first time, applies a very-high resolution model (much finer than TROPOMI resolution) to assess the effects of non-linear NO_x chemistry and mixing on the evolution of chemical plumes. Results of this paper are currently used to derive improved parameterisations for simplified methods, and results are encouraging (Meier et al., 2024). Running LES for all global power plants is obviously not practical. Our method does however provide detailed insights into the roles of chemistry and mixing on quantifying emissions using TROPOMI observations.

We will add towards the end of the manuscript:

> In general, our simulations provide new insights in the factors that are important for the interpretation of satellite-observed NO_2 plumes. Although it is impractical to run the model for each observed plume, it is likely that the main impacts can be parameterised in light-weight methods, like recently shown in Meier et al. (2024).

References

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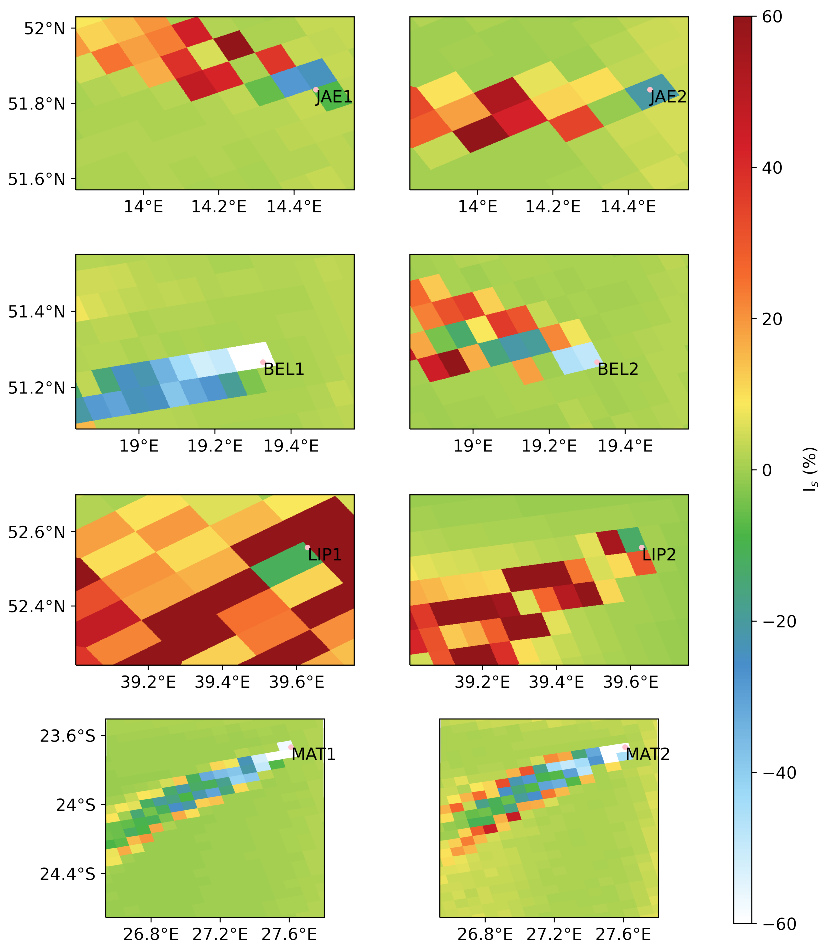

Figure 2: $I_{s,NO_2,OH}$ (in percent) calculated over the entire domain and degraded to TROPOMI resolution. Values are calculated up to the height of the convective boundary layer. These boundary layer heights are respectively 2500 (JAE1), 2000 (JAE2), 1200 (BEL1), 1500 (BEL2), 1800 (LIP1), 1500 (LIP2), 1900 (MAT1), and 1850 m (MAT2).

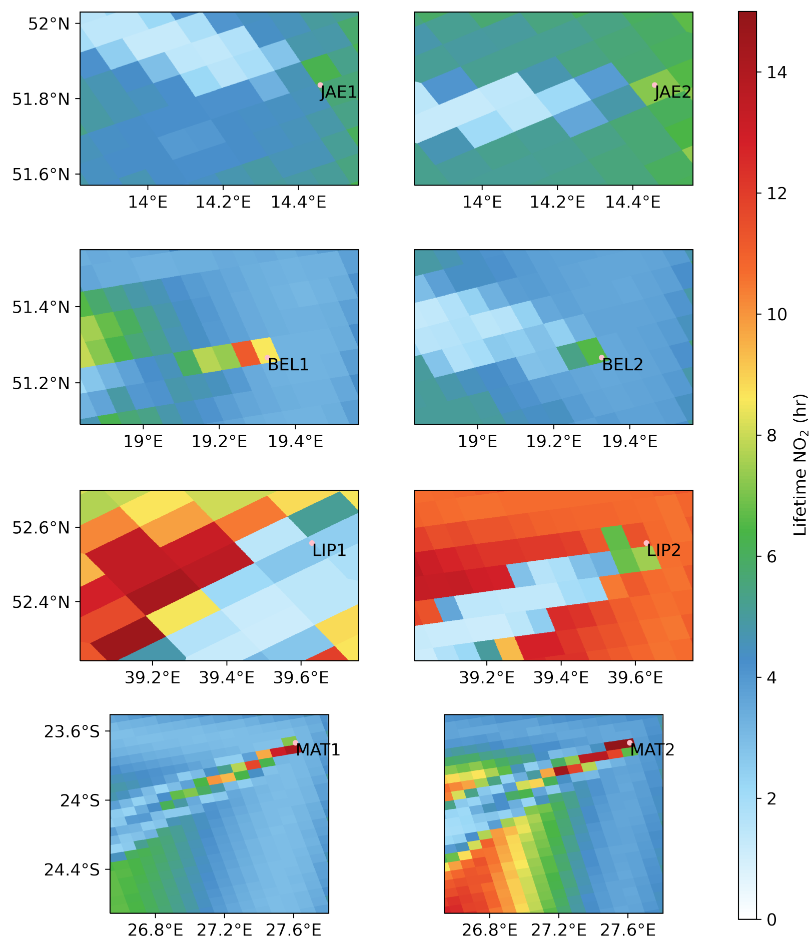

Figure 3: NO_2 lifetime calculated over the entire domain and degraded to TROPOMI resolution. Values are calculated up to the height of the convective boundary layer. These boundary layer heights are respectively 2500 (JAE1), 2000 (JAE2), 1200 (BEL1), 1500 (BEL2), 1800 (LIP1), 1500 (LIP2), 1900 (MAT1), and 1850 m (MAT2).

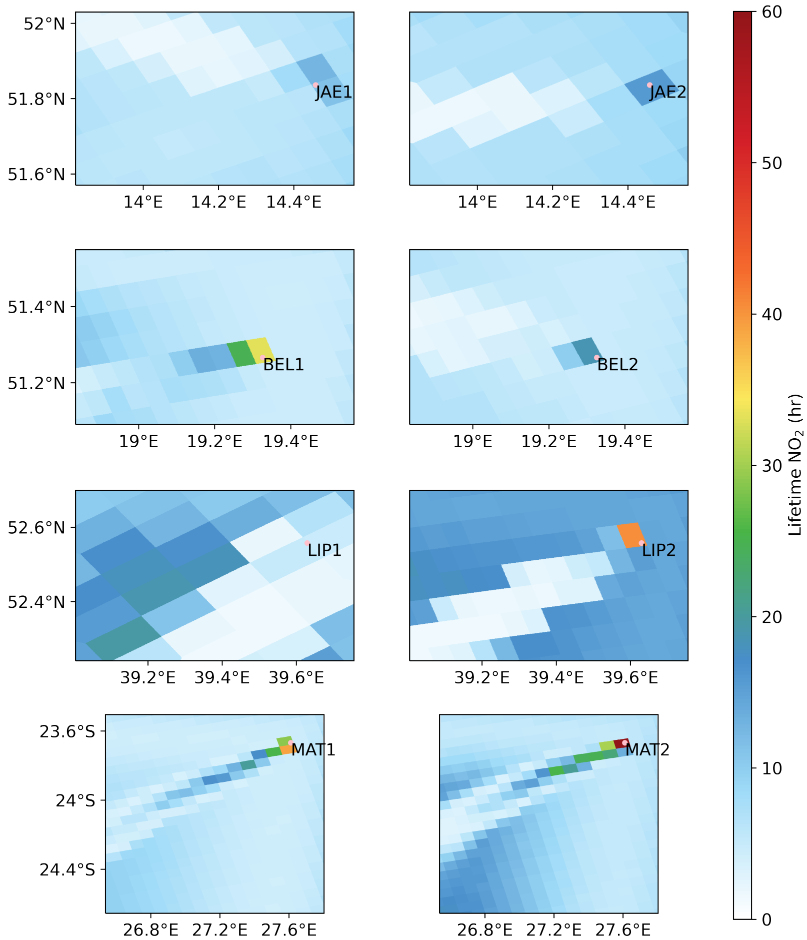

Figure 4: NO_x lifetime calculated over the entire domain and degraded to TROPOMI resolution. Values are calculated up to the height of the convective boundary layer. These boundary layer heights are respectively 2500 (JAE1), 2000 (JAE2), 1200 (BEL1), 1500 (BEL2), 1800 (LIP1), 1500 (LIP2), 1900 (MAT1), and 1850 m (MAT2).

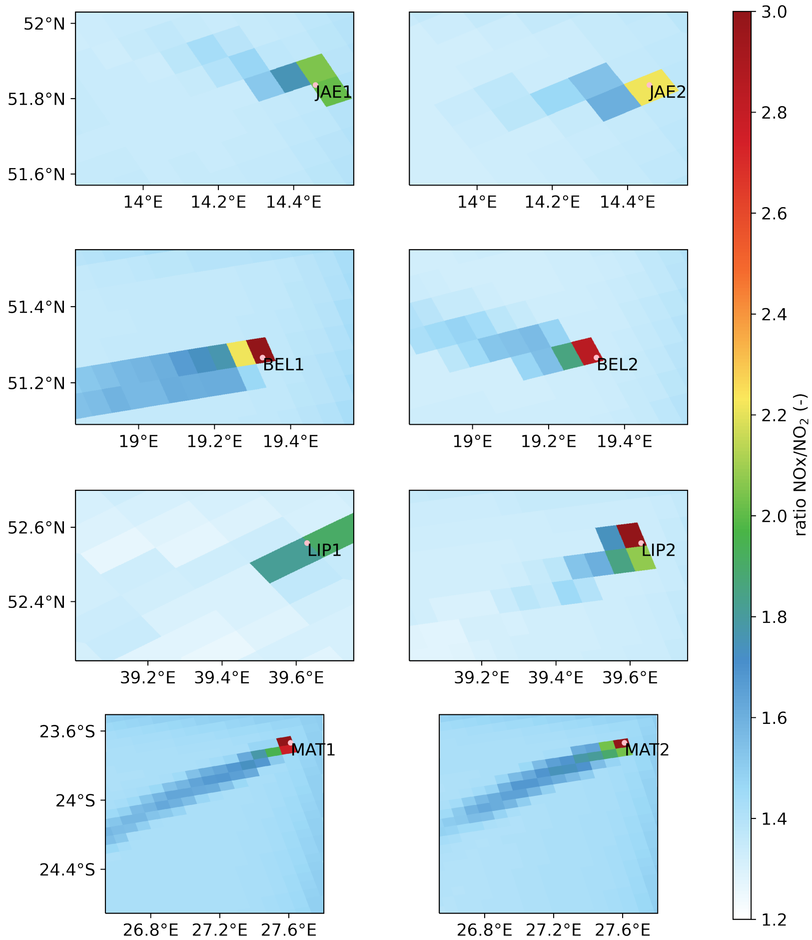

Figure 5: NO_x to NO_2 ratio calculated over the entire domain and degraded to TROPOMI resolution. Values are calculated up to the height of the convective boundary layer. These boundary layer heights are respectively 2500 (JAE1), 2000 (JAE2), 1200 (BEL1), 1500 (BEL2), 1800 (LIP1), 1500 (LIP2), 1900 (MAT1), and 1850 m (MAT2).