

Authors' Response to Reviews of

Estimating NO_x emissions of stack plumes using a high-resolution atmospheric chemistry model and satellite-derived NO₂ columns

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

1. Reviewer #3

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

1.1. Main comments

RC: *Title: The title of the manuscript centered on the estimate of NO_x emissions, but the current manuscript seems to focus less on the inversion/optimization of NO_x emissions, and more on the forward simulations of NO₂ columns and how transport, chemistry, and adaptation to TROPOMI affect the simulations. I may suggest the authors revise the title to better highlight the real novelty of their study.*

AR: We agree with this statement and changed "Estimating" in the title to "Evaluating", since this better reflects what we did:

Evaluating NO_x stack plume emissions using a high-resolution atmospheric chemistry model and satellite-derived NO₂ columns

RC: *Methodology: The definition of plume background area and the choice of prior emissions seem to be unclear. Did the authors use "correct" emissions reported from the CoCO2 project? If not, since emission, mixing, and chemistry can all be uncertain, it would be hard to evaluate the model performance (especially given the end goal of estimating emissions). Also, the "background" or background air has been mentioned but not properly defined. Was there a fixed threshold or a fixed distance from the stack? The background definition may affect the statements about lifetime diagnoses (page 18).*

AR: Concerning the emissions, see our answer to Reviewer #2, point 4. We conclude that we used the best available information as emission estimates. For LIP, these numbers are more uncertain. Concerning the background, we supply boundary conditions from ERA5 (meteorology) and CAMS (chemistry), like outlined in lines 156–161 of the manuscript. In response to a reviewer question on another paper (Meier et al., 2024), we provided this information, which can be found under <https://doi.org/10.5194/egusphere-2024-159-AC1>, Supplementary Figure S2. Thus, we used boundary conditions from reanalysis products, in which our simulations are embedded. We did not test the sensitivity of our results to these boundary conditions.

Concerning the location of the stack, we selected the domain such that we kept some distance from the boundary to allow the (circular) turbulent field to interact with the prescribed tracer profiles at the boundary. Furthermore, we selected the domain such that – with the prevailing wind condition – the plume had a substantial length within the domain.

Since we feel that the information in the paper is already adequate to address the issues raised, we did not modify the manuscript.

RC: *Results: I understand the authors' great effort in adding chemical reactions in LES for several simulations, but the explicit estimation of uncertainties associated with transport/mixing and chemistry seem to be missing from the manuscript, especially when the authors also acknowledged large uncertainties in the chemical model parameterization and atmospheric transport (L43 – L45). Again, if the final goal is to estimate/optimize emissions as suggested by the title, it would be important to acknowledge uncertainties in every model component (using either a simple CSF method or more complex chemical inversions). For example, Wu et al. (2023) (as mentioned on L44) tested the WRF-Chem-based chemical parameterization by estimating the chemical uncertainties with NO_x lifetime and NO₂-to-NO_x ratio and by evaluating against TROPOMI using EPA-reported power plant emissions.*

AR: We fully understand this comment. We evaluated the uncertainties associated with chemistry and mixing in more detail than reported in the manuscript. However, we wanted to avoid the paper becoming too technical and only briefly reported these results. We will now include Supplementary information to present these results. Below and in the Supplement, we present figures that show evaluations of the chemistry using a box model (with different emission scenarios compared to Figure 1 in the original paper). We simulated the chemistry of the full IFS chemistry scheme (IFS) and our simplified version presented in the paper (MHH). The box model considers emissions of NO and hydrocarbons (RH = C₃H₆) at the surface of a 1000 m well-mixed atmospheric box. Diurnal variations in light conditions are accounted for and simulations were run for two days. Figures 1–9 show the results with varying emission strengths, covering both ozone depletion and ozone production regimes. These regimes are expected in our plume simulations. Like mentioned in the original manuscript, results are almost identical when no emissions of hydrocarbons are considered (ERH = 0). Results deteriorate when emissions of hydrocarbons are considered, but remain acceptable (errors smaller than 10% for NO₂) specifically for NO₂ (sampled by TROPOMI) and when only the first day is considered. Here we note that air masses stay relatively short in our domain and that air masses forced by the CAMS boundary conditions that enter the domain.

We also evaluated the effects of resolution on mixing and chemistry in the plume. These results have been obtained with a controlled LES setup with a prescribed west–east wind (10 m s⁻¹ geostrophic wind) and prescribed boundary conditions for temperature and chemical species. The simulation domain size was [$\Delta x, \Delta y, \Delta z$] = [9600 m, 4800 m, 3200 m], i.e. much smaller compared to the simulation domain in the main paper. These simulations have been performed on different resolutions (200, 100, 50, 25 m) to test the effects of resolution on the main findings. The simulations were forced with a surface heat flux of 0.1 K m s⁻¹ and emissions from a 200-m high stack, located 1600 m from the west border. The source magnitude was 50% of the MAT emissions. The simulations were performed for 4 hours (starting at 6 am) and the results were averaged over the last hour of the simulation.

First, Figure 10 shows the NO₂ column averaged over the last hour of the simulation. Although the plumes appear similar at different resolutions, the conversion of NO to NO₂ proceeds slower at a finer resolution. This is corroborated in Figures 11, 12, and 13, which show the y-mean columns of, respectively, NO₂, NO, and O₃. Due to emission, sharp concentration gradients form at the stack location, because O₃ levels are titrated by the high supply of NO. Note that 95% of the NO_x emission is emitted as NO. These sharp gradients are expected to be resolved better in the high-resolution simulation. Results seem to converge at a resolution of 25 m. At a resolution of 100 m, as used in the simulations presented in the paper, a substantial instant dilution error is still present. For instance, at a distance of 6000 m from the stack, y-mean NO₂ columns are ≈20% larger at 100 m resolution, compared to the 25 m resolution simulation. These errors are hard to avoid if you want to address a larger domain of 50–100 km because the calculation time quickly becomes

ERH = 0.0, ENO = 0.1 ppb m/s

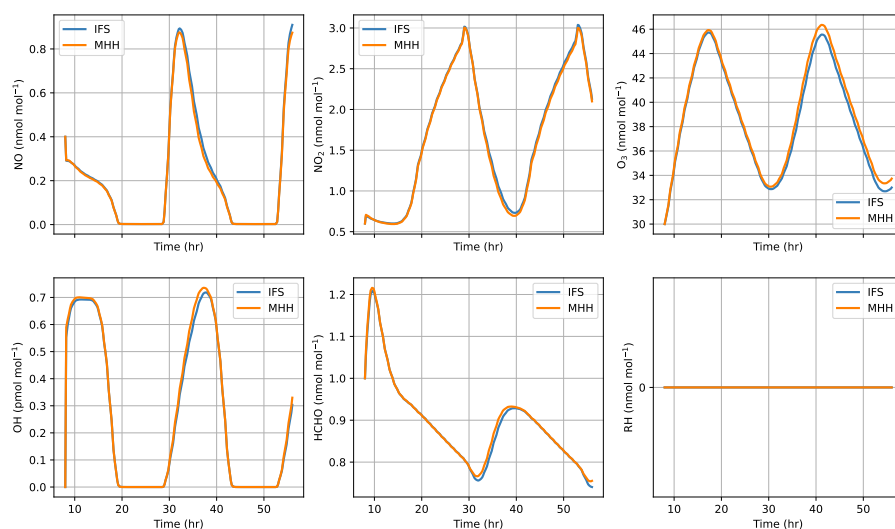


Figure 1: Box model comparison ($h = 1000$ m) of the IFS scheme with the condensed MicroHH (MHH) scheme. Results of a two day simulation are shown, starting at 8 AM. Emission strengths of NO and C_3H_6 are given in the figure title. Time series are plotted for NO, NO_2 , O_3 , OH, HCHO, and RH (C_3H_6).

prohibitive. The 100 m resolution used in the simulations presented in the paper is therefore a compromise, and this issue clearly warrants additional investigation. Concerning the ratio NO_x to NO_2 discussed in the paper, the effect of resolution is also important. Figure 14 shows the calculated ratio downwind of the stack. Although the NO_x to NO_2 ratio shows a clear spike at the emission point, this spike is smaller at 100 m and 200 m resolutions, and the ratio returns faster to background values.

In conclusion, at higher spatial model resolution, the conversion of NO to NO_2 is expected to proceed slower, and the ratios of NO_x to NO_2 are expected to be substantially larger at distances up to 10 km downwind of the stack. These effects likely depend on the emission strength, the background ozone concentration, the wind speed, and the stability of the boundary layer, and warrant further investigation.

We will add a Supplement to the revised manuscript to present these results, and adjust the main text to convey this message more clearly.

1.2. Minor comments

RC: *Eq. 2: What are some other processes? Emission and deposition?*

AR: As written, MicroHH collects tendencies for all processes and uses the RK3 solver to progress in time. Processes include advection and sources. Deposition is treated as part of the chemistry. We modified line 121: "The calculation of the chemistry tendencies is evaluated after the calculation of all other tendencies, including emissions." to:

ERH = 0.0, ENO = 0.3 ppb m/s

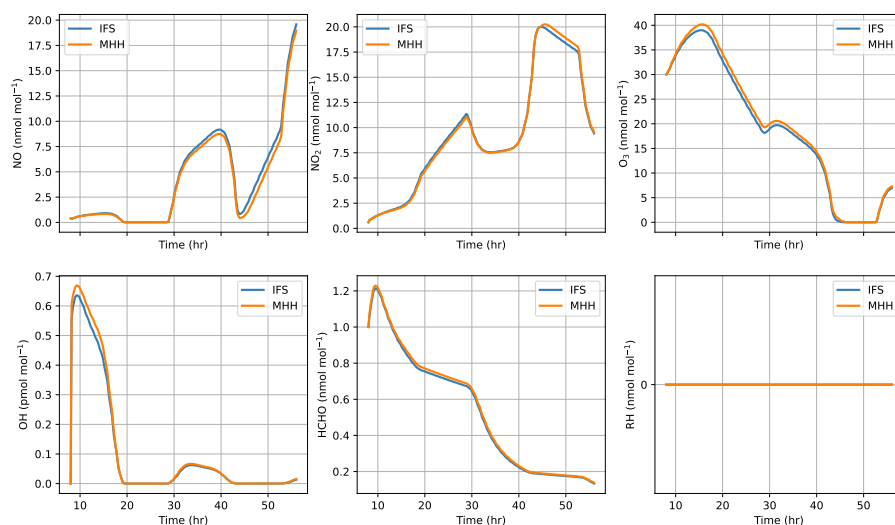


Figure 2: As Figure 1.

The calculation of the chemistry tendencies is evaluated after the calculation of all other tendencies, like advection and emission tendencies.

RC: *There seemed to be some considerations of the injection height (L78) and the use of a plume rise calculation (Table 5). But whether/how both injection height and the plume-rise were implemented in the model is not super clear to me, despite the acknowledgment and discussions in Sect. 4.4. More clarifications would help the readers, as injection height and plume height are important for point sources with intensive emission rates.*

AR: Since not all participating models in the underlying intercomparison study could calculate plume rise in a uniform way, we follow the CoCo2 intercomparison protocols (e.g. line 135, <https://coco2-project.eu/sites/default/files/2021-07/CoCo2-D4.1-V1-0.pdf>). To outline the general approach that led to the profiles in Table 5, we will add to the revised manuscript (above table 5):

Concerning emission height, we prescribed fixed emission profiles that account for plume rise. For JAE and BEL, these were calculated using the empirical equations recommended by the Association of German Engineers, which are based on the original work of Briggs (1984). Typical stack parameters were obtained from Pregger and Friedrich (2009), considering typical power plant capacities and fuel types, and from site descriptions. For LIP and MAT, the emission heights recommended by the CAMS emission dataset (Kuenen et al., 2022) for Industry and Public Power sectors were used.

RC: *Future application: Just out of general curiosity, how is the possibility of applying this approach to urban areas?*

AR: Yes, we are currently working on the implementation of our method to urban areas. We will add a sentence near the end of the revised manuscript to point this out:

ERH = 0.0, ENO = 0.5 ppb m/s

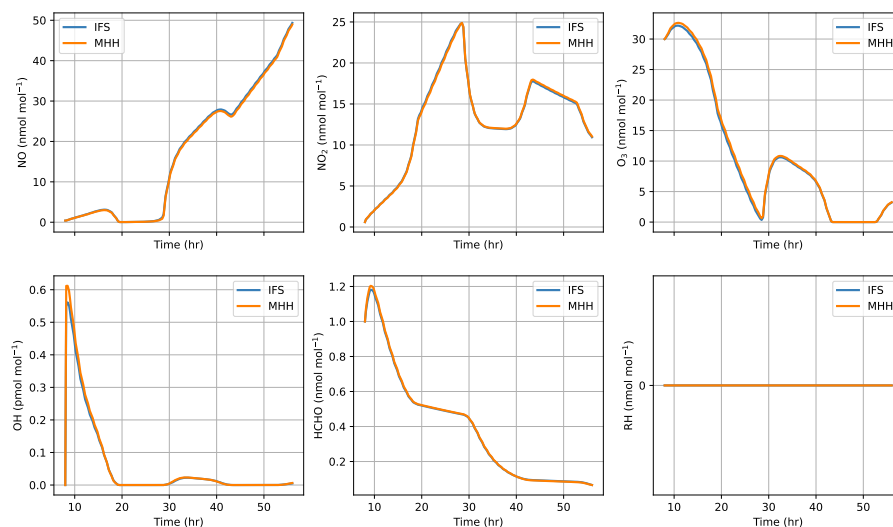


Figure 3: As Figure 1.

Moreover, we are extending our method to more complicated emission hot spots, like cities.

References

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- Pregger, T. and Friedrich, R.: Effective pollutant emission heights for atmospheric transport modelling based on real-world information, *Environmental Pollution*, 157, 552–560, , 2009.

ERH = 0.3, ENO = 0.1 ppb m/s

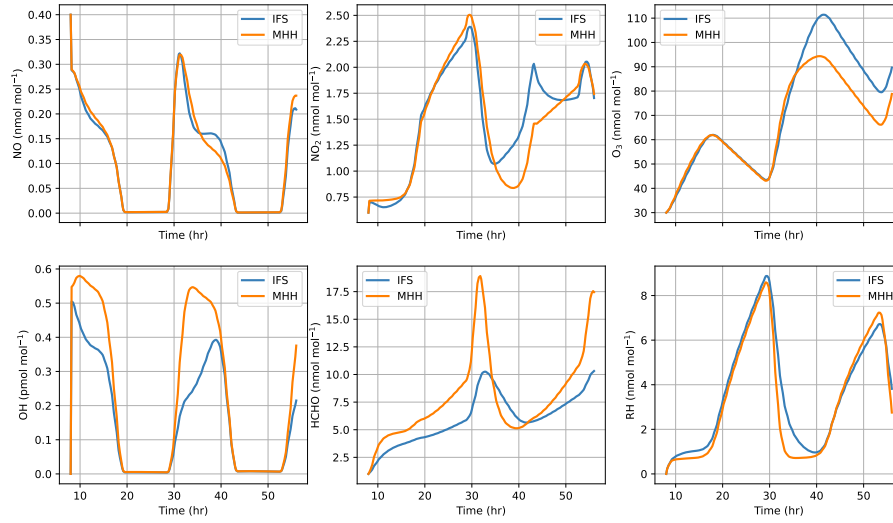


Figure 4: As Figure 1.

ERH = 0.3, ENO = 0.3 ppb m/s

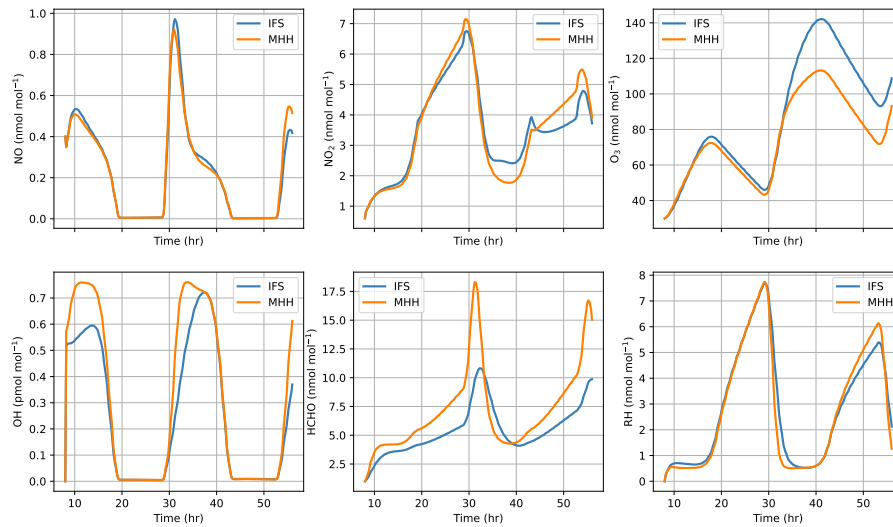


Figure 5: As Figure 1. This figure corresponds to Figure 1 in the main paper.

ERH = 0.3, ENO = 0.5 ppb m/s

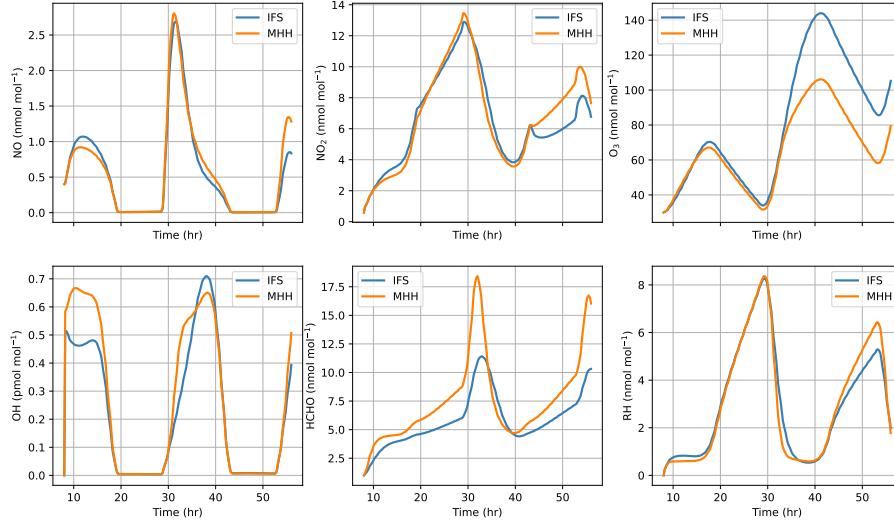


Figure 6: As Figure 1.

ERH = 0.5, ENO = 0.1 ppb m/s

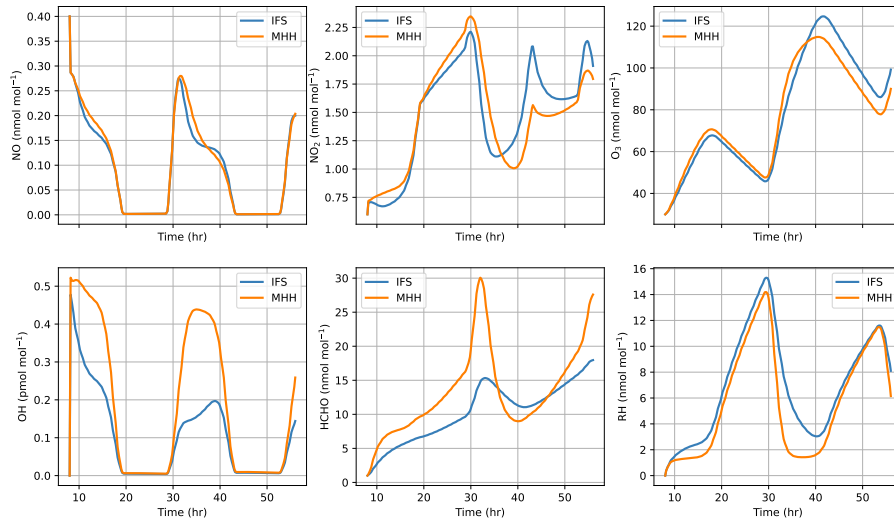


Figure 7: As Figure 1.

ERH = 0.5, ENO = 0.3 ppb m/s

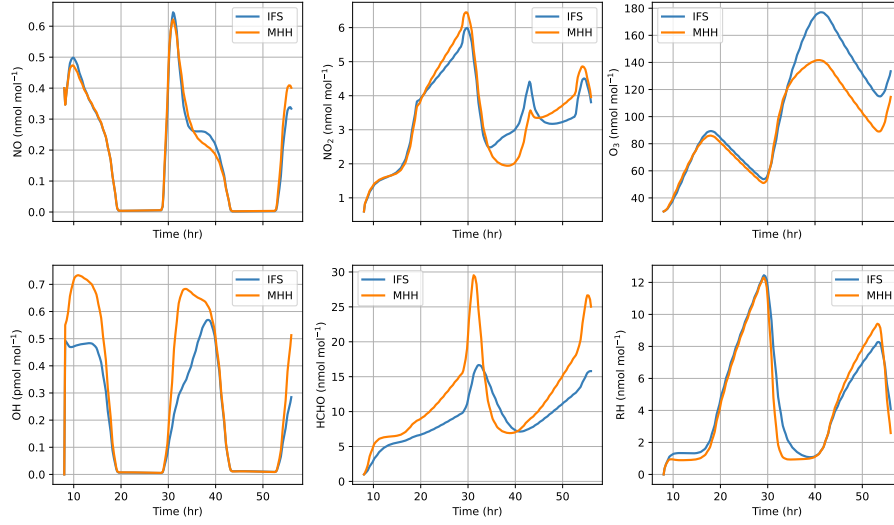


Figure 8: As Figure 1.

ERH = 0.5, ENO = 0.5 ppb m/s

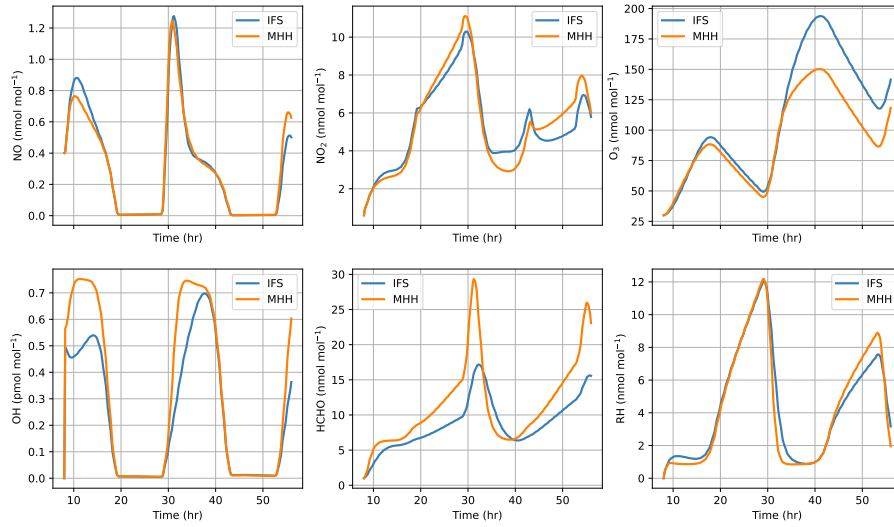


Figure 9: As Figure 1.

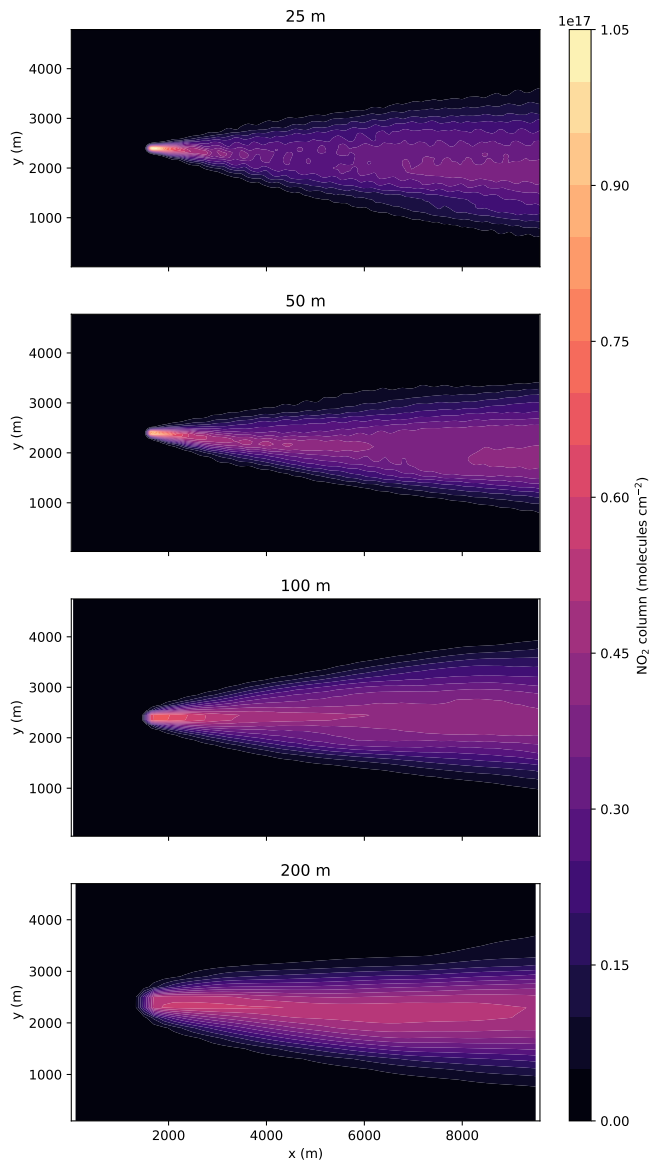


Figure 10: Total column NO_2 averaged over the last hour of a 4 hour simulation on a small domain. Emission with a source strength of half the MAT emissions were added at $[x,y,z] = [1600 \text{ m}, 2400 \text{ m}, 200 \text{ m}]$. Simulations were performed on resolutions of 25 m (top panel), 50 m, 100 m, and 200 m (bottom panel).

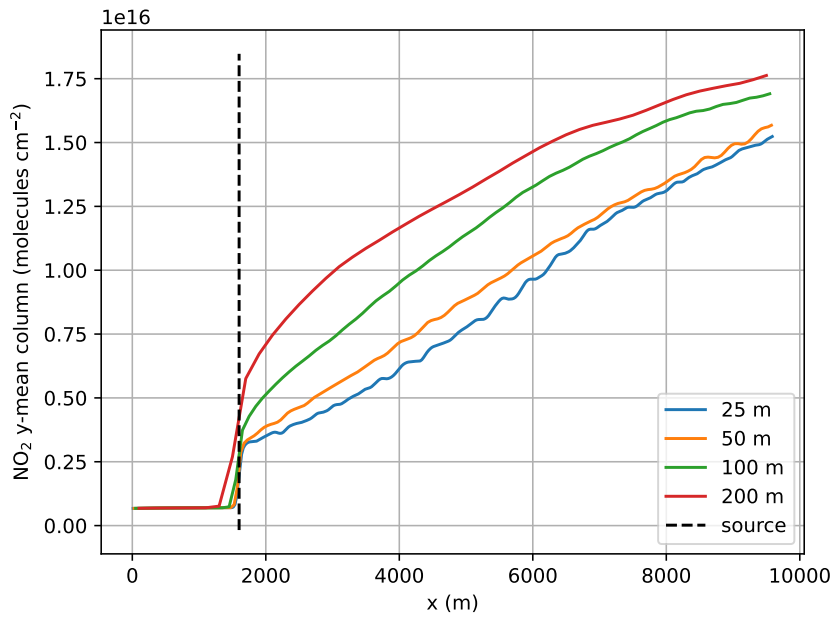


Figure 11: NO₂ y-mean columns from Fig. 10. The dotted line indicates the stack location.

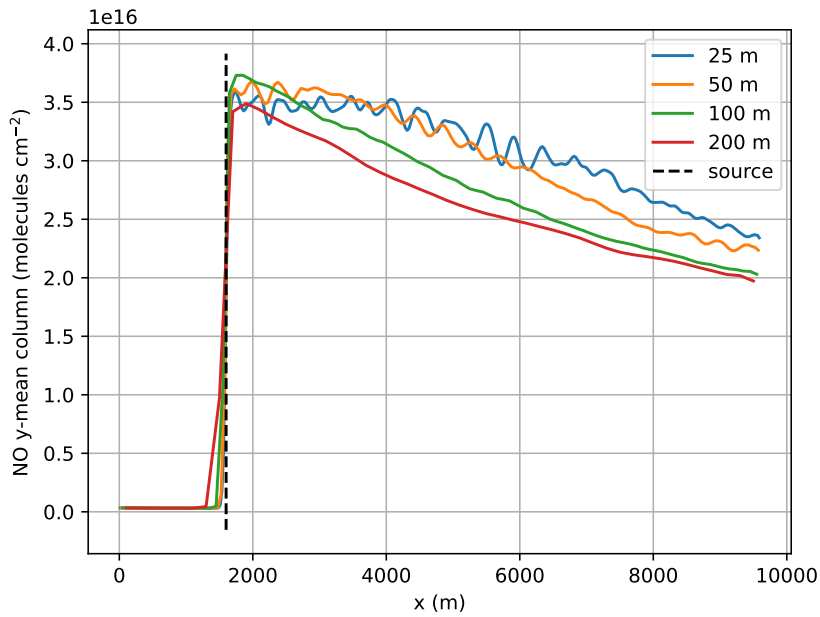


Figure 12: NO y-mean columns from Fig. 10. The dotted line indicates the stack location.

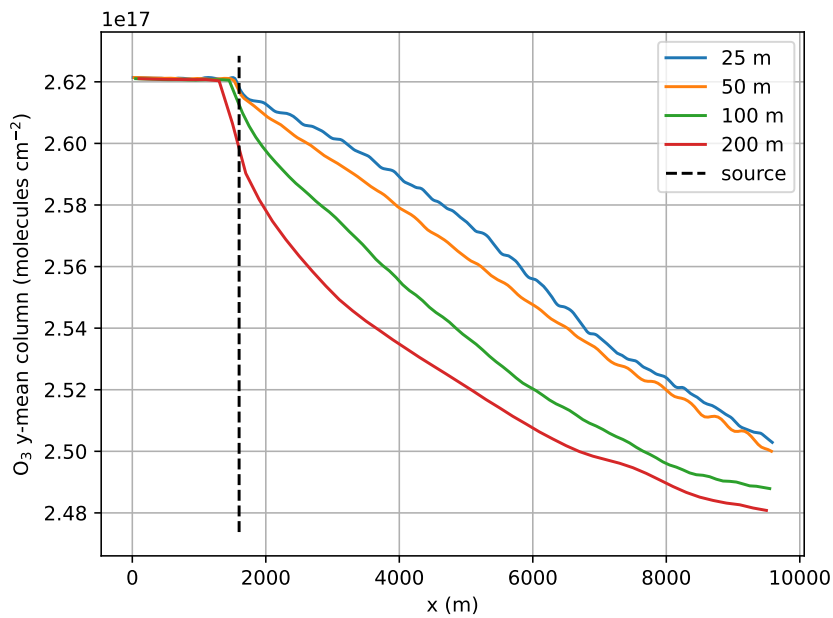


Figure 13: O₃ y-mean columns from Fig. 10. The dotted line indicates the stack location.

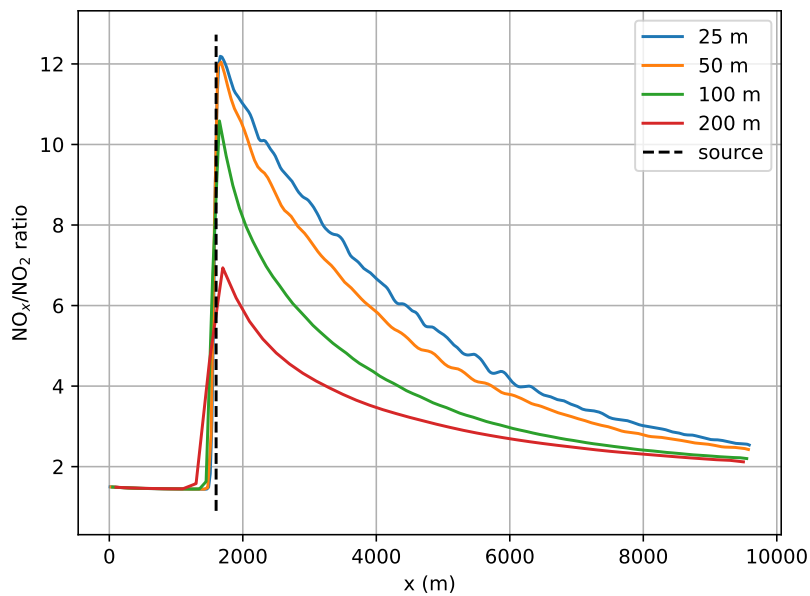


Figure 14: Y-mean NO_x over NO₂ ratio. The dotted line indicates the stack location.