Authors' reply to 'Comment on egusphere-2022-1473'

The authors would like to thank the Referee #3 for the constructive and encouraging review of the manuscript.

It must be stressed, that in 'Reviewer comments on egusphere-2022-1473' by Referee #2 several major revisions were requested, upon which the paper has been fundamentally restructured, including additional sensitivity studies to various simulation parameters. The content described by Referee #3 (mainly, the tuning of temporal NO_x emission profiles) remains in the revised manuscript, but is no longer considered the main finding.

Below, the authors respond to the review point by point. The referee's comments are printed in blue, and the authors' responses in black.

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While most of the NOx emission is set to be in form of NO, why the model simulated NO does not show significant changes and the model still misses its peaks systematically?

The simulation with tuned temporal profiles does show differences in modelled NO (see Fig. 3 (f) in the original manuscript): NO concentrations are approximately 50 % higher at daytime and 50 % lower at nighttime compared to the simulation with temporal emission profiles from Kumar et al. (2021). This agrees well with the temporal redistribution of the emissions.

The morning peak at ~ 05:30 shows almost no response to the change in emission profiles, but coincides with sunrise (see e.g. Fig. 5 in the revised manuscript). It can be expected, that photolysis of nighttime NO_x reservoir species (e.g. NO_3 and N_2O_5) contributes strongly to this morning peak, hence optimization of emission profiles can not fully account for it.

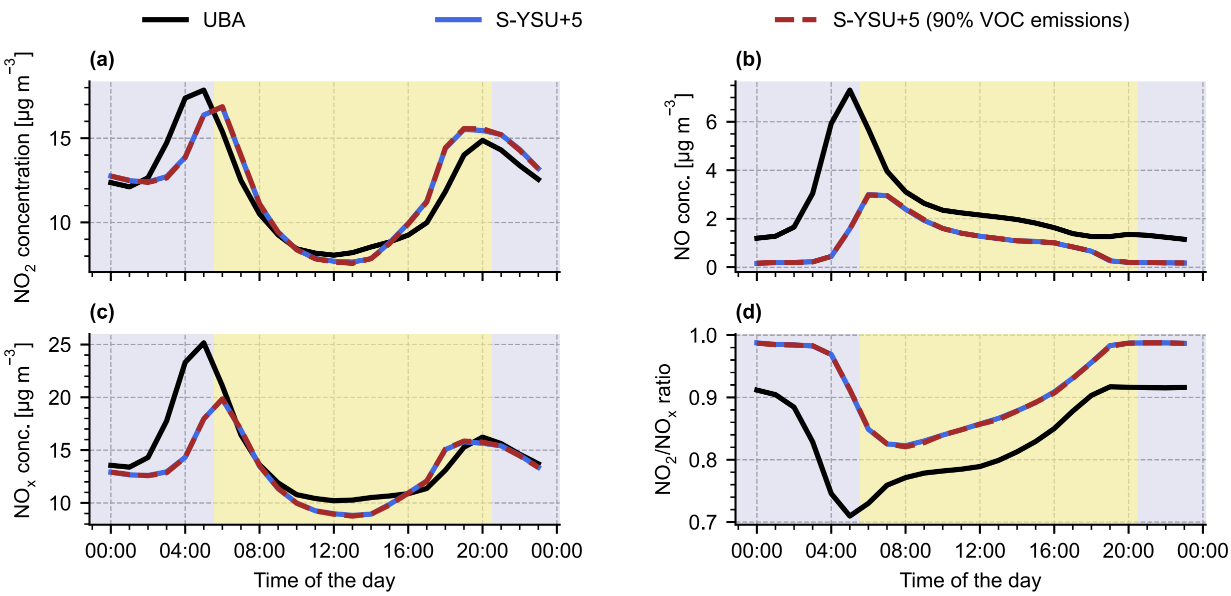
The Referee has further pointed out, that the model could be overestimating the conversion rate of NO to NO_2, which the authors address below.

The overestimation of NO2/NO ratio suggests (authors have also pointed this out) that model is having a limitation in capturing the chemistry accurately. This needs investigation why the NO is getting converted to NO2 more rapidly than reflected in the observational data. Sensitivity simulations should be conducted with say +- 10% VOC emissions to gain insight into the likely causes and probably to reach better estimation of the diurnal variability in NOx emissions over this region.

Following the Referee's suggestion, the model run „S-YSU+5" (explained in the revised manuscript) is repeated with 10 % reduced VOC emissions. This can also be motivated by the overestimation of the HCHO column (see the authors' reply to 'Reviewer comments on egusphere-2022-1473' by Referee #2), which could indicate an overestimation of VOC emissions.

The figure below shows the diurnal cycles of modelled surface NO_2, NO, NO_x, as well as the NO_2/NO_x ratio. The reduction of VOC emissions has minimal to no influence on the model results. In the original manuscript, it was shown that the reduction of O_3 boundary conditions by 15% results in similarly minimal changes. In consequence, it can be assumed that the oxygenation of NO to NO_2 (either via O_3 or VOCs) is not the reason for the faulty NO_2/NO ratios in the model. Instead, the problem could be caused by inaccurate representations of night- or daytime chemistry in the chemical mechanism, the photolysis scheme, or the emission speciation. These points can be investigated in the future.

In the revised manuscript, the topic of NO_2/NO ratios is discussed and possible solutions are mentioned.



UBA
S-YSU+5
S-YSU+5 (90% VOC emissions)
(a)
NO2 concentration [µg m−3]
(b)
NO conc. [µg m−3]
(c)
NOx conc. [µg m−3]
(d)
NO2/NOx ratio
Time of the day