Authors response on the Anonymous Referee #2 review of "Impact of urbanization on gas-phase pollutant concentrations: a regional scale, model based analysis of the contributing factors"

by Huszar et al. (acp-2022-337)

We thank anonymous referee #2 for all his comments and suggestion to increase the quality of our manuscript. We will address each of them and our point-by-point responses follow below. Reviewer's comments are italicized.

The manuscript presents component analysis of the impact of urban areas on air quality in Euorpean cities using several scenario simulations in a regional offline-coupled model system. The manuscript is well written and easy to understand. However there are some issues to be addressed, including the flow and the structure of the manusciprt that can be found below.

Comments:

Section 2.2.

Are fire emissions not taken into account? They can be important episodically for the O3 levels.

Authors response: Fire emissions are not taken into account. We admit that wildfires can significantly contribute to total pollution (not only PM but also gaseous pollutant e.g. NOx and CO) and model biases tend to improve if such emissions are included (Lazaridis et al., 2008), their contribution is important rather over southern Europe and Mediterranean and not on our focus area (central Europe). Moreover, we were interested (among other urbanization impacts) in the impact of urban emissions which not normally include wildfire emissions. We made a short note in the manuscript concerning these emissions with the above argument why we omitted them.

A comparison of the two CAMx versions can be provided in the supplement as difference in the chemical mechanism can have significant impacts on the pollutants considered in the present study? Similar results could point the emissions however different results point both emissions and chemistry. I see this has been discussed in the last section but I think it should also take place, at least partly, here as it now stands detached from each other.

Authors response: We think it would go far beyond the scope of the manuscript to include a graphical comparison (using plots) of two different CAMx versions, although such comparison could be, in general, be very useful. We however added a few further notes on the difference between CAMx v 7.10 vs. 6.50. to the "Models used" section. As emissions in this study were the same as in our previous similarly oriented studies (Huszar et al.2020b, 2021) and the only difference is in the newer CAMx version, the differences in model performance can be explained by model version differences. In this sense, the improvements in CB6r5 vs. to older CB5 are certainly the most important and stand behind the reduction of the model biases. This is detailed in the Discussion section (2nd paragraph) with highlighted improvements in this new mechanism with respect to CB5.

Section 3.1

The plots in Figure 2 are representing cities in the different domains or are they all from the 9 km mother domains? What is the driving meteorology in these plots, WRF or RegCM? Is there a comparison available for the meteorology and associated chemistry over the 9 km grid?

Authors response: Results for Prague are taken from the 1 km domain, otherwise they are extracted from the 9 km one. This is made clear in the revised manuscript. In the original "Discussion" version of the manuscript, the driving meteorology was RegCM, but based on the other Referee's comments, we decided to include the plots also for the WRF driven CAMx runs (here all cities are represented at 9 km x 9 km). This allows us to see what effect has if an alternative driving meteorological model is used. Moreover the 9 km results serve as a good

justification that even at this regional scale resolution, the model performance can be acceptable and the concentrations well represented (except SO2, but this is probably not a resolution issue). Finally, we also included another column to the validation figure showing the diurnal cycles of ozone, as for this pollutant, monthly values are no so policy relevant and the hourly evolution is indeed important (especially the daily maxima). As for the meteorology, we rely on our previous study of Huszar et al. (2020b) which used exactly the same meteorological driving data (from both RegCM and WRF) and provides the validation of the meteorological fields: namely near-surface temperature, precipitation, 10 m wind speed and PBL height, which are all important from air-quality perspective.

Section 3.2

Does Figure 3 provide the ensemble from both the WRF and RegCM simulations? Please explain and modify the caption accordingly.

Authors response: yes, the results represent the ensemble from both simulations. This is clarified in the manuscript.

Section 3.3.4

Can you please a bit more the case for Milan as it really stands out?

Authors response: Milan, Italy has one of the highest emissions among cities and also the worst air quality (https://www.eea.europa.eu/themes/air/urban-air-quality/european-city-air-quality-viewer) which is captured also by the model (see e.g. the validation figure with monthly/daily cycles for NO2). Furthermore it has the warmest climate among the chosen cities and a relatively large size, so it is clear the the urbanization causes strong changes in BVOC emissions, see the figure with BVOC emissions changes (for Isoprene in Fig. 9). Strong decreases of ozone, as detailed in the Discussion, are then result of decreased VOC emissions in a VOC-limited chemical regime. The strong secondary responses of NO2 are the result of these strong ozone changes. We added a note to this in the revised manuscript.

Can you also elaborate a bit on the impact of resolution on these results focusing on the Prague experiment? How much your conclusions would change based on this experiment if you were able to run all the cities on 1 km resolution for example? I am aware this cannot be answered quantitatively without the simulations, but I would like to see a discussion on this.

Authors response: In this regard our discussion should be based especially on the comparison of results for Prague for 1 km vs 9 km resolution. Such comparison shows that the impacts are stronger for the 1 km resolution peaking at the city center, i.e. the largest impact are concentrated in the city core while at 9 km resolution the impact is logically uniformly distributed within the 9 km gridcell, so the peaks are flattened. This is very well seen in each of the four contributors. It can be thus assumed, that had the model been run at 1 km x 1 km resolution for all analyzed cities, the impact would have had a much larger peaks (in absolute sense) concentrated around the cities cores. We added these notes to the revised manuscript (at the Discussion).

Section 3.4 focuses on explaining the diurnal variations but do not discuss much the underlying reasons for these diurnal variabilities. I would expect such a discussion supported with some plots, likely in the supplement. I see these decoupled explanations also in other parts of the manuscript. There is of course not a correct way to provide this information but I think the manuscript would benefit very much if the discussions in the last section could be moved to the corresponding sections explaining the impacts of the different scenarios.

Authors response: We wanted to keep the logical practice that the Result section provides the results without searching for explanations or without deeper discussions, while it is the Discussion section were different findings are explained, interpreted and interconnections are formulated. We would prefer thus to keep the Results section “clean” from deeper discussion. For the diurnal patterns of different contributors, we added a

new paragraph in the Discussion section, which explained the modelled cycles and reflect them to findings in previous studies.

References (not used in the original manuscript before the revision):

Lazaridis, M., Latos, M., Aleksandropoulou, V. et al. Contribution of forest fire emissions to atmospheric pollution in Greece. Air Qual Atmos Health 1, 143–158. **https://doi.org/10.1007/s11869-008-0020-0**, 2008.