Accounting for model error in air-quality forecasts: an application of 4DEnVar to the assimilation of atmospheric composition using QG-Chem 1.0 . Reply to referee # 3

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1 Reply to general comments

We thank the reviewer for his comments, which helped to improve the manuscript. The detailed replies follow:

1. *I found the last paragraph in section 2.1 to be unclear and somewhat confusing. For instance, it is not clear what was meant by ?The sources of the QG model provided by ECMWF...?. Do the authors mean that the code for the dynamical model was given to them by ECMWF? There are some further examples listed below. I therefore recommend that the authors check this text carefully for its clarity and re-write it.*

   Answer: Yes, the reviewer's interpretation is rigth. The text has been modified as follows:

   The code of the QG model that is distributed with the OOPS DA library have been used for this study (Y. Trémolet, personal communication).

2. *"The boundary conditions are taken cyclic...", but it is not clear what "cyclic" means in this context.*

   Answer: The text has been modified as follows:

   The domain is cyclic in the East-West direction, i.e. the model fields are periodic in this direction. The stream function is set to climatological values at meridional walls (Dirichlet boundary conditions).

3. *"For all the experiments presented in this study a coarse resolution of 16x8 grid points has been used." I have assumed that this is the horizontal resolution, but it is not clear whether 16 refers to the North-South dimension or the East-West. The authors should also write the horizontal resolution in terms of spatial resolution in km.*

   Answer: The sentence has been modified as follows:

   For all the experiments presented in this study a coarse resolution of approximately 750 km (16x8 grid points, respectively, for the East-West and North-South directions) has been used.

4. *Figure 1 would probably be improved with an explanation of what the grid lines represent. One presumes these are the outlines of the grid boxes used in the model, but the authors should state this clearly.*

Answer: In the original manuscript the displayed lines did not correspond to the model grid but were only used for spatial reference. In the revised manuscript all figures showing the model fields have been revised and now display the full model grid. An explanation has been added as well in the legend of Figure 1.

5. *Page 24, lines 1-2. The authors should probably remind readers that only the initial conditions are perturbed during these tests, and therefore NO2 converges to the truth due to the shorter lifetime of NO2 combined with the fact that there are no errors in the emissions.*

   Answer: The following line has been added to remind the reader:

   This happens because, when the model is perfect, the NO_2 field rapidly converges to the truth after few hours.

6. *Page 25, line 15. Can the authors think of any specific reasons why ozone and NO2 show similar behaviour to CO and CO2?*

   Answer:

   We think that such a behavior has been observed because the initial perturbation of NO_2, which is 16% of the average NO_2 concentration ($\sim$0.1 ppbv), is applied at midnight and is quite small compared to the amount of NO_2 that is produced hourly in our experiments as a consequence of NO emissions ($\sim$0.15 ppbv per hour). Hence, the amount of NO_2 available for photochemical reactions and ozone production later in the day is very slightly influenced by the initial perturbation. Cases with larger initial NO_2 perturbations, especially if applied during the active photochemical phase, could show different sensitivities to the initial condition. Additional experiments with a larger variety of initial perturbations should probably be performed to check if the conclusions given in this section remain valid. We included the following sentence in the manuscript to highlight this point:

   A possible reason is that the amount of NO_2 produced hourly by the oxydation of emitted NO is much larger than the applied initial perturbation. Therefore, the O_3 photochemical production, which happens later during the day, is not much influenced by the perturbation of NO_2 at midnight. It would be interesting to verify if similar results also hold when larger perturbations are applied during day-time. A wider exploration of different chemical regimes is left for a future study.

7. *The explanations for the emission perturbations at the beginning of Section 4.2 could benefit from slightly more explanation. It is not stated directly, but I assume from the existing text that the perturbations result in reductions in NO emissions. This should be stated directly. Also, does the size of the perturbations vary with time, and if so at what frequency?*

   Answer:

   The information was given at Page 29, line 4 but was actually not very clear. We updated the manuscript with the following sentence:

Forecast emissions are increased by a multiplicative factor of 2.35, whereas the log-normal distribution has been used to generate emission perturbations for the ensemble of forecasts. The emissions perturbation is constant in time but not in space, due to the geographical variability of emission factors (Fig. 1).

8. *Please can the authors add some further text to the Figure 12 caption to explain what the red and blue colours represent.*

Answer: The color bar has been modified in the revised manuscript (c.f. reply to reviewer n. 1, additional remarks)

All further technical comments have been considered for the revised manuscript.