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 # 2

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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. It seems that are tests are done setting to zero inter-species error correlation. While some justification is provided for this, perhaps an actual figure would help. For example on p. 23 it is said: Finally, 4DEnVar was capable to provide same good results as in Sec 4.1.1 when enabling the cross-variables covariances?. Results are not shown to back this
 - Answer:

statement?

We omitted showing the corresponding Figure since differences were not very significant. For completeness we report here in Fig. 1 the results (RMSE gain) obtained when assimilating all species at once and enabling cross-variables covariances, compared to those already shown in the original manuscript (univariate and independent experiments for each species). We note that differences are quite small. Moreover, summarized scores for both multivariate and univariate DA were already presented in Fig. 10 (Fig. 7 of the revised manuscript), when discussing the impact of multivariate localization. We removed the word "Finally" from the beginning of the sentence to avoid possible misunderstandings, since we just refer there to the previously discussed results (Page 25, line 7).

15 2. It is said that "Only surface observations are considered in this study" (p13, L28). There is a need to acknowledge as well the lack of vertical propagation of information from these surface variables in the context of the "toy" system. This represents a significant challenge in a real system.

Answer:

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We agree with the reviewer about the importance of correctly propagating vertical information in chemical DA, especially for the assimilation of satellite data. QG-Chem could permit vertical propagation of information in DA experiments but it has an over-simplified vertical structure (two layers). Therefore, we think that vertical aspects of chemical DA remain beyond the scope of QG-Chem. We added the following sentence to the conclusions of the revised manuscript:



Figure 1. DA results with a perfect model hypothesis. From top to bottom: the RMSE gain is displayed, respectively, for O_3 , CO, CO₂ and NO₂ assimilation experiments. Blue color means that DA lowered the RMSE, red color means that DA increased the RMSE. Plots on the left are the same as the ones shown in Fig. 4 of the newer version of the manuscript (Fig. 7 in the original manuscript), i.e., done with independent and univariate DA experiments. On the right only one DA experiment is performed perturbing and assimilating all species at the same time. Multivariate corrections are also activated for the right plots. The plots shown here are obtained with the revised version of QG-Chem, which included chemical deposition (c.f. reponse to reviewer n. 1)

Aspects related to the vertical propagation of the information, which have been neglected with QG-Chem, could also represent an additional challenge in real systems.

All minor corrections have been included in the revised manuscript.