Responses to Reviewer # 1

We thank reviewer # 1 for taking the time to make a thorough review and for their constructive remarks. We respond to each of the reviewer's comments by quoting or summarising the reviewer's comments in italics, and by quoting the changed text in the paper (shown in bold text), or by describing the changes in normal text.

General Comment 1*. The reviewer indicated that the presentation quality of the manuscript needs to be improved. They indicated that Section 2 and the descriptions of each experiment in Section 3 were not very clear or well organised. The reviewer gave several specific examples of how this information was discussed in the paper in separate locations without a common link. Consequently, the reviewer found that readers need to construct the full list of experiments and their details themselves. As a result, the reviewer recommended that we add a table describing all of our experiments and that we accompany this table with a paragraph in Section 2.*

We thank the reviewer for their recommendation. We agree that these recommended changes would lead to a substantial improvement to the clarity of the manuscript and that they would resolve the problems identified by the reviewer. We have implemented changes that differ slightly from those suggested by the reviewer though. We have added three new tables (Tables 2 and 3 referenced in Section 2.2 and Table 6 referenced in a new Section 2.5) and added a new Section (2.5) at the end of Section 2 that fulfils the requirements explained by the reviewer. We reduced the amount of information presented on page 4918 and moved this to the paragraph in Section 2.5. We have also reduced the amount of numbers and information presented in the paragraph on page 4920 and have moved much of this information into Tables 2 and 3 but kept this discussion in place in Section 2.2 because these details primarily relate to the photochemical box model. Within Section 2.5 and Table 6 we now list all of the experiments that we carry out with the tools described in Section 2.

The reviewer has recommended that we remove Figures 2-4. A reviewer of the previous version of the manuscript actually asked us to keep Figure 4 in the paper. Also, Figure 4 supports some of our arguments and responses to comment 3 of reviewer #1. We have changed the captions for Figures 2-3 to make it clear that these are not results but for illustrative purposes only. While we accept that these figures are superfluous to the reviewer this may not be true for other readers that perhaps have less background in this field. We therefore wish to keep them in the paper.

General comment 2. *The reviewer explained that a lot of our figures were of a poor quality. Specifically, that the numbers and labels were inconsistent within a single figure, and that consequently these figures were not very homogenous as a result.*

We have now reproduced some of the figures in order to address these concerns. We have now made the numbers in the x axis of Figure 2 larger to be more consistent with the y axis. We have improved the resolution of Figure 3. Unfortunately its resolution was degraded in the typesetting process. We have enlarged the numbers in the Figure 4 x axis to be more consistent with the y axis. In Figures 5-9 the numbers in the x axis are not the same size of the y axis. However, due to the dimensions of these figures, and the requirement from a previous reviewer to enlarge the labels, it is not possible to have the labels on both axes to be the same size.

General comment 3. *The reviewer questioned our inclusion of the CO observations into our experiments. The reviewer explains that we describe "…that CO observations do not influence significantly the ozone forecast", and that our paper focuses on the results from ozone and HCHO. Therefore, to them, it is not clear why we included CO. The reviewer proposes several explanations* *for its inclusion while seeking clarification, and also says that it would be interesting to see results that combine ozone and HCHO measurements.*

Having reflected on the reviewer's comments we can see that our results and experiments regarding CO have not been described precisely enough and that consequently, the justification for including CO was not made very clear. A couple of important details were either not included or not well described. In addition, the reviewer's statement "…that CO observations do not influence significantly the ozone forecast" is not precisely what we said nor is it correct for all cases of observation noise. We therefore now clarify the situation and show the changed text below.

First, in our experiments, working on the timescale of three days, and consistent with prior knowledge, ozone is less sensitive to changes in CO concentrations compared to NOx and VOCs. Thus, ozone is overall less sensitive to changes in CO emissions, and, therefore, ozone predictions are less sensitive to CO emission uncertainties.

However, on the timescale of our air quality forecasting scenario, ozone is still sensitive to very large changes in CO concentrations and consequently to large changes in its emissions and similarly to large uncertainties in their emissions. In each of the CN, OCN, and HCN scenarios, where β = 0.1-1.0 (β is the noise parameter), CO emission uncertainties are sufficiently low (i.e. $E_{X_{CO}} < 0.1$) that the effect on ozone prediction uncertainties would be less than 0.5 ppbv (estimated based purely on the perturbation predicted from the Jacobian). Only in the β = 5.0 scenario, where the uncertainty on X_{CO} is 1.1, are the X_{CO} emission uncertainties large enough to lead to significant ozone forecast uncertainty, i.e. $\sim$ 5 ppbv.

These results in the paragraph above show it is desirable to resolve CO emissions to a sufficient degree in order to improve ozone forecasting. However, the requirements for CO observation noise needed to achieve a sufficient estimate of the CO emissions, and consequently a good ozone forecast, are much lower than for either the observations affecting NOx or VOC emission estimation. Further, and consistent with a point already made in the paper, the estimation of CO emissions is only dependent on observation noise and is independent of photochemical regime.

Given the points above, we would like to change the text in the paragraph at the end of section 3.1.1.1 from:

"Until now, we have not directly discussed the impact of CO observations or of the resolution of CO emission uncertainties within the assimilation framework. We do not show a figure here, but a posteriori CO emission uncertainties are virtually invariant with respect to photochemical regime and to the observing scenario (CN, OCN, or HCN). The a posteriori CO emission uncertainties increase from 1 × 10−5 to 1.1 with increasing observing noise from β = 0.1 to β = 5."

to:

"Until now, we have not directly discussed the impact of CO observations or of the resolution of CO emission uncertainties within the assimilation framework. We do not show a figure here, but a posteriori CO emission uncertainties are virtually invariant with respect to photochemical regime and to the observing scenario (CN, OCN, or HCN). The a posteriori CO emission uncertainties increase from 1×10^{-5} to 0.1 as the observing noise increases from β = 0.1 to β = 1.0, respectively. According to the sensitivity of ozone to X_{CO} in the jacobian **K'**, these relatively low levels of CO emission uncertainty would only lead to perturbations in ozone of 0.5 ppbv at most. For the case with the highest amount of noise, β = 5.0, the a posteriori CO emission uncertainty reaches 1.1. Again, using **K'**, we can estimate that this larger level of CO emission uncertainty could lead to a about a 5 ppbv perturbation in ozone. Therefore, only the β = 5.0 noise scenario leads to large enough a posteriori CO emission uncertainties that can have a significant effect on a posteriori ozone prediction errors."

We also change the text in the paragraph at the end of 4.1 to:

"We have indirectly performed a sensitivity test to see if CO observations affect ozone a posteriori prediction errors. We can address their potential impact within the OCN scenario by examining the jacobian matrix (see Fig. 4). This shows that ozone is relatively insensitive to perturbations in CO emissions and, therefore, also to a posteriori CO emission uncertainties. In fact, it appears that only the β = 5.0 noise scenario has sufficiently large a posteriori CO emission error to cause significant a posteriori ozone prediction error (about 5 ppbv)."

The inclusion of CO observations in the different scenarios is useful and we now include more discussion about CO within the final version. The reviewer also mentions that it would be interesting to examine a scenario using both O3 and HCHO. We did include results from a scenario using O3 and HCHO in the HOCN scenario. We think that a comparison between the HOCN and CN scenarios adequately tests for the sensitivity of the inclusion of O3 and HCHO.

Responses to specific comments

P 4912, line 25: the representativity of the measurement should also be discussed. It can bring some limitations when used for data assimilation.

We agree with the reviewer that it would be interesting to discuss this point. We have added the following text to a separate paragraph immediately after the one highlighted by the reviewer:

"Surface station in-situ data is made at a high spatial resolution, which is typically much higher than most air quality models. As a result, this introduces the problem of having representativity errors between the model, which is unable to represent fine-scale variability, and the observations that can measure this variability. This problem therefore limits the efficacy of data assimilation and systems need to be carefully designed to take this type of error into account."

P4914, lines 3-4: reference to Fu et al., ACP, 2013 and Cuesta et al., ACP, 2013 concerning multispectral retrievals (IR+UV) of ozone should be added.

We have now added these references.

P4923, references to Fig. 3 and eq. 12: the choice of E is not judicious as it is already used for the emissions. I am not sure this figure is very useful. One understands the process by the text.

We have changed *E* to *D*.

P4923, line 10: It is not clear for me why the figure "demonstrates the mechanism by which : : :". It seems quite obvious and well admitted for a secondary pollutant that the improvement of its precursor emissions will improve its concentrations.

We thank the reviewer for identifying this problem. We have removed this sentence.

P 4924, line 11: I do not understand what the authors mean by this sentence and what the interest is. They need the Jacobian to go through the error analysis, so it is not redundant.

We meant that Jacobian is redundant specifically for 4D-var. This statement is true because it plays no role in 4D-var. The uncertainty analysis is a framework external to 4D-var that we can use to characterise the errors. However, to improve the clarity of the manuscript we have changed the statement from:

“The Jacobian matrix is redundant within 4-D-variational data assimilation, but it can help characterize the uncertainties…”

To:

“The Jacobian matrix can be used to help characterize the variance…”

P 4924, line 13-14: I would rephrase the sentence more like this: "Within our framework, each element of K represents the forward: : :.”.

We thank the reviewer for this recommendation and have changed the text accordingly.

P 4934, reference to Tab 5.: For the OCN scenario, 2 very large values are reported in the table for XNO=1.25 and 1.5. Are they correctly reported? IF yes, they should be discussed.

These values are correctly reported. These high values occur because the L-BFGS algorithm is only able to find a solution in a local minimum. The XNO = 1.25 and 1.5 scenarios are neither NOx limited or VOC limited. The low sensitivity of ozone to the XVOC parameter therefore likely explains the difficulty the algorithm has in finding the global minimum. We should point out that this error only has a minimal impact on the ozone prediction error because ozone is not strongly sensitive to XVOC for this XNO range. We have therefore added the following text:

“There are also examples where ozone precursor emissions are poorly resolved, but this has only minimal impact on the ozone prediction error, D. This occurs for the OCN scenario when X_{NO} ranges from 1.25 to 1.5. For these cases the unresolved error on X_{VOC} is larger than for many other situations. Again, this occurs because the L-BFGS algorithm is only able to find a local minima. However, in these instances, the relatively low sensitivity of ozone to XVOC means that the resulting ozone prediction errors are relatively low as well.”

P4935, reference to Tab. 6: What about the ozone concentrations outside the ozone maximum? Is the influence similar?

The influence is very similar outside of the maximum. We have now added this text to the relevant paragraph discussing Table 6:

“Although we only show the differences in the maximum ozone mixing ratios, this behaviour is reproduced in the ozone mixing ratios at other times during the sunlit day. This further confirms our general findings from these tests.”

Technical comments:

P 4915, line 10: change “pre-cursor” to “precursor”

P4922, line 19: Is the notation xˆt within the gradient consistent with the notation use elsewhere in the text?

P4924, line 5: change “emissions” to “emissions estimates”

P4929, line 26: it should be “HCN scenario” and not “HCHO scenario”

P4935, line 1: change “varibility” to “variability”

P4945, line 27: change “may too be insufficient” to “may be too insufficient”.

All of the technical remarks shown above have been addressed.