Responses to Reviewer # 2

We thank reviewer # 2 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.

Responses to specific comments

Abstract: There are several formulations in the abstract which somewhat obscure the scientific content. Examples are "a variety of analyses"; "characteristics of "; "to support"; "various sets of"; "our principle method"; "which is the primary focus of this work"; "simple but key"; "our framework's ability"; "These questions are designed to examine"; "establish the robustness". The clarity of the abstract may be improved by removing several of these phrases.

We have changed the text according to some of the reviewer's recommendations.

Abstract: "complimentary"

We have corrected this mistake. Thanks.

Introduction: In general the introduction is well written. I found the content a bit too focussed on the USA, and the authors may consider to add 1-2 lines to balance this a bit more. A reference to MACC is missing, e.g. the recent GMDD paper by Marecal is relevant.

We have added this reference. Thank you.

p4915, l7: "simplistic". This is a very negative word.

We have removed this word.

Scenarios: why do all scenarios include CO ? A scenario with O3, NO2 and HCHO would make sense to me, given the techniques to measure these compounds with satellites. Would that make any change to the ozone forecasts?

Although ozone is relatively weakly sensitive to CO on the three day timescale of our simulation, large perturbations in CO concentrations can lead to non-negligible perturbations in ozone. Therefore, large unresolved uncertainties in CO emissions can contribute to significant ozone prediction uncertainty (for more detail please refer to the response to reviewer #1's general comment 3). It was therefore interesting to examine CO in the scenarios we chose. We have added text to Sections 3.1.1.1 and 4.1 to discuss this point. Also, the performance of CO observations and the resulting CO emission inversions are close to equal in all three of the CN, OCN, and HCN scenarios. Therefore, its inclusion in each of our scenarios allows us to examine the effects of ozone, NO2, and HCHO to this system without having to simultaneously consider the removal of CO observations in one or more of the other scenarios. Overall, this allows us to use fewer observing scenarios. Finally, the HOCN scenario highlights the value of combining HCHO and ozone observations relative to either the HCN or OCN scenarios.

p4918, l2: "averaging kernel and DFS". Readers may associate "averaging kernels" with satellite retrievals. It is good to make clear that emission averaging kernels are meant here.

We thank the reviewer for identifying this problem. We have changed the text in several places to make this point clearer.

p4918, l14: What is a 1D box model. For me, a box model is 0D. If 1D, how many layers? Or does the 1D refer to time?

It would be better if we referred to our model as pseudo 1D. The model in actual fact contains a single vertical layer, but we use a boundary layer parameterisation with a pre-set diurnal variability to alter the mixing height in the model. We have therefore changed the text to reflect this point more clearly.

"A pseudo 1-Dimensional photochemical box model was built ..."

"The model is not truly 1-Dimensional in the vertical because we use a parameterisation to describe variability in the boundary layer height and mixing volume."

p4919, l1: Isoprene emissions and concentration: please give the reader an impression what this corresponds to (e.g. "typical concentrations for Summertime North-East USA, Summertime Southern California"?). Similar for the anthropogenic VOC emissions: is this typical for urbanised regions? (Is mentioned later, but good to mention it here as well)

We have completely reorganised section 2.2 to state more clearly, and earlier in the text, that the model is set up for conditions in urbanised Southern California. We are then able to refer back to this text when we discuss the emissions in the model.

p4919, l10: Again it is unclear what the "box" in the box model represents. Is it the entire boundary layer?

We have changed the text to reflect that the box vertical height represents the height of the boundary layer.

"In our model, the vertical extent represents the full depth of the boundary layer."

p4919, l22: Can emissions be adjusted with an hourly time step, or longer (e.g. daily)?

This is possible, but the complexity of the data assimilation increases greatly when doing so, and the difficulty in carrying this out would also greatly increase. We think the increases in complexity and difficulty mean that these are issues better explored in future work and are beyond the scope of this paper.

p4922, l1: remove subscript at end of the line.

We have corrected this problem.

p4922, eq 10: S_n^{-1}

We have corrected this problem.

p4923: I do not understand eq 11. Does "xt" mean "true state" ?

Yes, ***xt*** is the true state of the emissions.

p4923: What does "x" mean in this case. Again, is this the "true" state? It seems "x" has a different meaning here as in eq.8 ?

There is actually an error here in several places. ***x*** has been written instead of ***xt***. The ozone true state at time, t, has been written as $q_{O_3}(\boldsymbol{x}, t)$ but should be $q_{O_3}(\boldsymbol{xt}, t)$. We have changed the text to reflect this. This also affects equations 12, 13, and 14. These have been changed accordingly.

p4924, l12: "characterise the uncertainties on x and q". But I thought "x' is the uncertainty. So, the sentence reads like "characterise the uncertainty of the uncertaity". Is this what is meant?

x tilde is actually the error on the emissions. So what we wrote was the uncertainty on the error. The accompany text has been changed to state that this method can be used to estimate the variance on these parameters.

p4925, l8: "z = O3 : : :". Should this be "y = O3 : : :" ?

We have changed this from z to y.

Caption fig 4: q_Z(x,t)/dx_NO is repeated 3 times. What are the three colors?

Thank you for identifying these errors. We have changed the text and have now added an explanation of the colours.

Fig 9: lower is NO and upper is VOC ?!

We apologise for this error. We have corrected this figure. Thank you for identifying the problem.

p 4933, top: For Fig 11 it would be interesting to understand if the error reduction is due to the diurnal sampling, or to the reduction of the noise. More observations (n) effectively implies a 1/sqrt(n) decrease of the error. Would the same reduction be obtained if all observations were taken on the same hour? Figure 10 shown that the time of observation is crucial. How does this relate to fig.11 ? For instance: for a sampling distance of 12h, what are these two hours?

We agree with the reviewer that is a point of interest. Figure 10 does imply that there should be an effect on ozone prediction due to the interaction between observing frequency and how this limits the specific times observations can be made. Figure 10 implies that the decrease in error will not simply follow 1/sqrt(n) because observations made at certain times of day appear to have more value compared to others. In Fig 11 we include results from only a single set of observing times for each of the different observing frequencies, e.g., for an observing frequency of 3 hours we used observations at 0, 3, 6, 9, 12, 15, 18, 21, 0 hours as opposed to 1, 4, 7, 10, 13, 16, 19, 22. All of the other observing frequency scenarios began their observing cycle at time 00:00 of the observing period. We do think it would be interesting to explore the interaction between observing frequency and observing time and we would like to explore this topic in a future article. However, we feel that a study of this interaction would be beyond the scope of this paper, and that it would add extra details and length to an already sizeable manuscript. We have added the following text to discuss these issues at the end of section 4.3:

"It is likely that there is an effect on ozone prediction error due to the interaction between observing frequency and observing time. Figure 10 implies that observing scenarios measuring at the same frequency could yield different prediction errors due to when they actually sampled during the diurnal cycle. However, in each test we made at a particular observing frequency the observations were made at a fixed specific set of times, and so our work does not address this issue. We do think that this is interesting and relevant to evaluating different types of observing scenario, and we would therefore like explore this problem in a future paper. "

Table 3, 4, 5: what is the unit of the numbers presented?

The variables in Tables 3 and 5, and the XNO variable in Table 4 are the unitless emission scaling factors. We have added a note to the captions to explain this.

3.2.2. Table 6 not easy to understand. What does "ozone prediction error – standard true state" mean? Error-minus-state does not make sense.

We agree. This is an error and have therefore corrected it.

p4936, top: I do not understand the message behind the comparison in Fig. 12. Evidently there is a clear weekly cycle. However, on top of that there is the full day-to-day variability of weather-related processes and emission variability which complicate reallife comparisons as compared to the simplified box model approach. In fact, for me Fig 12 is not really useful for this study and may be removed.

We had wanted to use this to show that within urban areas the diurnal variability and inter-diurnal variability of anthropogenic emissions is relatively invariant during the midweek, and that one could therefore assume that it was reasonable to use a consistent profile of emission variability from one day to the next in the simulation. However, following the reviewer's recommendation we have removed this figure and the paragraph that discusses this issue.

p4937, l27: "demonstrated our framework's relevance" I do not understand what is meant by demonstrated here. Clearly many issues, such as the various modelling uncertainties, role of vertical distribution, as well as the ground and satellite observation characteristics (kernels, representativity) are not discussed.

We wanted to explain that we have made the first demonstration that our framework is able to address this kind of technical problem, and this is a minimum requirement for the framework to be "relevant" to air quality forecasting. We recognise that there are many more difficulties and challenges that would have to be overcome in a real-world situation. We have therefore changed the text to reflect this more clearly from:

"This therefore demonstrated our framework's relevance to future air quality forecasting systems that might utilize state of the art assimilation and observations made using either the ground station network or from orbiting satellites."

to:

"This therefore demonstrated our framework's relevance to future air quality forecasting systems that might utilize state of the art assimilation and observations made using either the ground station network or from orbiting satellites. Clearly, more difficulties and challenges remain before such a framework could be used in a real-world setting, such as how to incorporate averaging kernels of satellite measurements into the assimilation system or accounting for representativity errors."

p4942, l 14-15: Apart from future 4D-Var, do the authors think that (ensemble) Kalman filter approaches could deliver similar results?

There are differences in these two data assimilation approaches that limit the type of emission solution each can generate. Specifically, Kalman filter approaches are limited by only being able to arrive at emission inversions in future model timesteps. Besides these differences, the Kalman filter method could still be used to solve emission inversion problems for different observations and targetted emissions and it uses model sensitivities of concentrations to emission changes to do this. Therefore, some of our conclusions regarding the effects of photochemistry on emission inversion will still be relevant. We have therefore added the following text:

"Note too that Kalman filter methods can also be used in this application and we should expect that the performance of this method will be similarly affected by photochemistry."

p4943, l2: "are the first to demonstrate this novel approach" Is this true? For instance, Miyazaki et al. (doi 10.5194/acp-12-9545-2012) assimilate ozone and NO2, and the system adjusts the emissions.

We have now modified the text to reflect this:

"…ozone observations with either NO2 or HCHO observations would be beneficial, consistent with Miyazaki et al. (2012), we have actually shown that it could be highly advantageous."

p4944, l14: "non of the current generation of LEO satellites possesses a reliable means of attaining instrument sensitivity to the boundary layer for these gases." Is this true?

In particular in the UV and SWIR spectral ranges there is sensitivity to the ground, and the signals measured with LEO instruments show a clear signal in NO2 and HCHO (in fact also CO) originating from the BL.

The text describing this point is not precise enough. We meant to say that this sensitivity is not unique to the boundary layer for single instruments (this in itself is not true either, see below). In the case of NO2 and HCHO, the vertical sensitivity is too broad to uniquely resolve the boundary layer. One can assume that these pollutants are concentrated in the boundary layer, but this is not information derived from the satellite instruments themselves. We do, however, recognise that the SWIR channel on MOPITT does give this instrument reliable sensitivity to boundary layer CO over widespread areas of land surface (Worden et al. 2013).

We have therefore changed the relevant text from:

"However, none of the current generation of LEO satellites possesses a reliable means of attaining instrument sensitivity to the boundary layer for these gases."

to:

"However, only one of the current generation of LEO satellite instruments (MOPITT) possesses a reliable means of attaining unique instrument sensitivity to the boundary layer for these gases (Worden et al. 2013)."

p4945, l6: Perhaps good to mention the night-time mixing (of ozone) between the boundary layer and free troposphere.

We thank the reviewer for this suggestion and we have therefore changed the text from:

"Of course, if the effects of transported pollution were to be considered, making observations during the night could offer additional utility by improving the estimated contribution to the pollution made by this process."

to:

"Of course, if the effects of transported pollution were to be considered, such as the night time mixing of ozone between the boundary layer and free troposphere, then making observations during the night could offer additional utility by improving the estimated contribution to the pollution made by this process."