## Answers to Reviewer 2

We would first like to thank the reviewer for the comments that helped us in improving the manuscript. Detailed point-by-point replies (in blue) are provided below.

# 1. It seems that the approach of aggregating observations into superobservations introduces a resolution dependency. What is the maximum horizontal resolution at which the assimilation approach is practical?

Indeed, the spatial resolution of the superobservations is equal to the resolution of the model. We believe that one way to answer the question concerning the maximum horizontal resolution at which the approach of assimilation is practical is to consider the representativity of the available individual observations on the scale of the model grid. For the model resolution used in this work, using observations helps because the observations help to constrain the average value corresponding to the area covered by each model grid. This is the case because IASI observations cover a scan over more than 2000 km, and because the number of observations in each model grid is generally high (see figure 3a). For IASI we thus believe that the adopted assimilation approach is practical even for grids of the order of several hundred kilometers.

If the number of observations present in very large model grids would be lower (e.g. lower than 4), or if the observations would be representative of only a part of the grid, than one would risk loosing the representativity of the mean concentration by individual observations. For example, having measurements only at nadir would probably limit the representativity of measurements with small footprints at the scale of large model grids (only a part of the model grid would be seen by the instrument).

Our formulation of model error takes into account the error introduced by high variability of individual observations (if the variability is high, we are less likely to capture the mean concentration, and thus the superobservation is given less weight in the assimilation process) and number of measurements falling in each model grid (high number of measurements increases the representativity and thus reduces the error).

# 2. What would be the effect of assimilating each IASI observation separately and to then average the resulting assimilated profiles at each grid box?

We have not tested this approach and instead we have applied the approach of superobservations described in Khattatov et al., 2000. We believe that for the case analyzed in the manuscript, the superobservations are a more meaningful quantity to assimilate since they represent better the grid-average value. The approach with the superobservations also allows us to control the coherency of the error formulation.

If the observations were to be assimilated individually, because of the limiting effect of horizontal resolution of the model, the impact of individual observations would need to be averaged to provide a single innovation value. To show analytically what could be the impact of assimilating the observations individually, we make the simplification that all observations are taken at the grid center that is in the location where  $x_b$ , the model first guess, as well as  $x_a$ , the analysis, are defined. If the averaging kernel (A) and observation error (O) of all of the observations falling in one model grid were the same, than the matrices K and H would be the same for all observations. We could then write the formula for each individual analysis  $x_{a,i}$ :

$$x_{a,i} = x_b + K(y_i - H(x_b))$$

To obtain the final analysis  $x_a$ , one could proceed by averaging the individual  $x_{a,i}$ . This would result in:

$$x_a = x_b + K(\sum_{i=1}^{N} \frac{y_i}{N} - H(x_b))$$
, which would be equivalent to assimilating one superobservation with mean

value  $\sum_{i=1}^{N} \frac{y_i}{N}$ . So the final result would not be too different from assimilating a single super-observation.

Application of Eq 4 (adding the variability term) would still be desirable to have chi2 values near 1. If the errors on the observations are not the same, one would have to take this into account, as more weight would need to be given to an increment calculated for observations with smaller error.

In addition, the approach with superobservations, given the high number of IASI CO retrievals, allows increasing the computational efficiency of the code (assimilating one observation instead of a number that can reach a hundred observation in one model grid).

3. How exactly is the prior formed, is it one global mean profile, or is it variable in time and space? If it is a single profile, how may this affect the quality of the IASI retrievals in terms of temporal/latitudinal biases? Should this uncertainty not be part of the assimilation process as well?

The information on the generation of the prior is given in Section 2.1. It is a single mean profile that was generated based on measurements from aircraft profiles that were completed with ACE-FTS observations in the upper troposphere and above.

This profile is not variable in space and time. The approach of using a single *a priori* profile, with associated (large) *a priori* errors that reflect the global variability, is often used in inversion calculations (see Hurtmans et al., 2012). Using more local *a priori* profiles, with lower errors, could result in lower posterior errors on the retrievals; however, less information would be taken from the measurements and more from the *a priori*. In addition, as the product is global, discontinuities at the limits (temporal or geographical) would appear due to the change in the *a priori*. More detailed discussion of the benefits of using a single prior is beyond the scope of this paper.

The uncertainty of the *a priori* impacts the retrieved CO columns. The value of the *a priori* enters the assimilation as this information is taken into account when the  $H(x_b)$  term in equation 6 is being calculated using an equation similar to the one given in equation 7. The following sentence was added for clarification:

Similar equation in which  $x_r$  is replaced by  $x_b$  is used to construct the corresponding model value (H( $x_b$ ) in equation 6).

4. The presentation of the effect of assimilating CO observations should be presented in more detail and made more quantitative, e.g. by showing difference plots of the assimilated model minus the control run and assimilated model minus the IASI observations. The reason for the discrepancy between the control run and the assimilation run could be further investigated.

As suggested by the reviewer, this section was extended and one additional figure was added (the figure 5c showing the difference between the assimilation and control runs). This discussion now constitutes Section 4. The new figure 5 is shown below.





Figure 5. Monthly averaged CO columns for July 2008 simulated by the model for a run with assimilation (a), model without assimilation (control run) (b) and for the difference between these two simulations (impact of assimilation = simulation with assimilation-control)) (c). The CO columns are calculated by applying the averaging kernels supplied with each observation to the model analysis.

# 5. The combination of a Discussion and Conclusion section seems awkward, it would be much clearer to separate writing this into two separate sections.

As suggested, the Discussion and Conclusion section was split into: Section 5.3- Discussion of the Validation results and Section 6- Summary.

### Specific comments

pg. 31692, I. 13: remove "an" -removed

I. 27: "limited only": certainly, resolution is not the only limitation of the product: do you mean a certain aspect?

This sentence is rewritten as follows: Assimilation techniques offer a powerful tool to propagate the information provided by the satellites in space and time into a global and continuous product whose spatial resolution corresponds to the resolution of the model.

pg. 31693, l. 18: capitalize "Arctic" Corrected

I. 19-26: this section seems out of place, I suggest to place this at the end of the discussion or into the conclusions This section was moved to the end of the summary.

pg. 31697, I. 3-10: This sections seems to explain the same things twice with different formulations.

The following sentence has been removed to avoid repetition. : The error on the superobservation must take into account not only the errors on the individual measurements, but it should also provide information on how representative is the superobservation of individual measurements.

Eq. 3 and 4: The apostrophe at the symbols is not very clearly visible, consider using a different marker symbol, e.g. a tilde.

We agree and the tilde is now used instead of the apostrophe

pg. 31704, I. 7: Do you mean model error parameterisation? Add this to clarify the distinction to other parameterisations (model physics etc.).

Yes, we meant model error parameterization. The text was modified (in Section 3.4 after reorganization). The text now reads:

In order to test the coherence of the parameterisation of the model error (Section 3.3) and of the superobservations (Section 2.3), we apply the chi2 test.

Fig. 5: Include difference plots assimilated-IASI, assimilated-control.

Assimilated-control was added (please see figure 5c above). Assimilated-IASI was not added as the corresponding monthly averaged values are generally weak and no strong features emerge from this plot.

pg. 31705, I. 16: This aspect could be tested by doing the assimilation over Asia only and comparing to the full assimilation.

We do not believe that assimilation over Asia only and comparing with the results with full assimilation would be easy to interpret. Over Asia the results are likely to be similar, however, over the region downwind from Asian emissions, there will be a difference due mostly to the lack of assimilation of the transported plumes (assimilation is important away from source regions) and also somewhat to differences due to lack of assimilation over other source regions.

Fig. 6: Color scale is exceeded in Fig. 6a. A panel should be added showing the IASI observations (not only superobservations) for the same time instance.

The color scale in Fig. 6 was changed. The figure with superobservations was added.



CO column(mol/cm2\*1E18),IASI superobs.

CO column(mol/cm2\*1E18),assimilation run



CO column(mol/cm2\*1E18), control run

CO column(mol/cm2\*1E18), IASI L2

Figure 6. Maps for July 7th 2008 for IASI CO superobservations (upper left), IASI CO individual observations (upper right), model with assimilation (lower left), control run (lower right). For individual observations all available data are plotted (both day and night which leads to overlap) while for the remaining panels the mean values are plotted. The white areas indicate model grid boxes with less than four IASI CO measurements retained during this 24-hour period or regions that were filtered.

### pg. 31709, l. 15: "This plume which mixes. . ." sounds strange, rephrase.

#### The sentence was modified:

This plume which contains contributions from biomass burning and anthropogenic pollution was observed by IASI as shown in Figure 6 and as described in Pommier et al. (2010) and Sodemann et al. (2011).

# I. 16: The presentation in this section would profit from using labels in Fig. 11. Also,

### Fig. 11b is described before Fig. 11a, which is confusing.

The order of figures was reversed in Fig. 11 to correspond to the description in the text. The text was also modified as follows:

We first present a comparison of CO time series obtained during two DC-8 flights that traversed pollution plumes. The first analysed flight took place on July 9th 2008 over Greenland and Canadian Arctic sampling a high altitude plume with Siberian origins. This plume which contains contributions from biomass burning and anthropogenic pollution was observed by IASI as shown in Figure 6 and as described in Pommier et al. (2010) and Sodemann et al. (2011). The analysis of the bottom left panel of Figure 11 shows a significant improvement of the CO level when assimilation is used. The model with assimilation captures relatively well the presence of highly polluted air masses in the upper troposphere during the second half of the flight (bottom left panel of Figure 11). The model values reach 200 ppbv in agreement with observations. The control run with the monthly averaged GFEDv2 emissions is unable to reproduce these high CO signatures

from biomass burning at high latitudes. One day earlier (not shown), on July 8th, the DC-8 aircraft intercepted the outer edge outskirts of the same plume over the Canadian Arctic at about 80°N. Again, the model with assimilation reproduces well the highly elevated values of around 150 ppbv present in the upper troposphere at the time of the flight while the control run predicts mixing ratios below 100 ppbv. The second analysed flight took place four days earlier, on July 5th, 2008. It sampled forest fires plumes over the Saskatchewan region (Canada). For this flight, the model with assimilation reproduces relatively well (and better than the control run) the elevated background values but is unable to reproduce the high CO mixing ratios (bottom right panel of Figure 11). These peaks are likely due to relatively fresh emissions that had less time to be diffused and also that were sampled by fewer IASI assimilation windows than the plumes present at high altitudes over Greenland on July 8th and 9th.



Figure 11. Comparison of model and observed in situ CO values for two flights of DC8: left figures: July 9th over the Arctic and right figures: July 5th over Canada. The top panels show the flight pressure, lower panels show CO mixing ratio from in situ measurements (black) and the simulation with assimilation (green) and without assimilation (red).

# I. 24: "at about 80\_N": How does the fact that data assimilation ends at 75\_N affect this comparison?

The fact that even without assimilation above 75 N there is an improvement in the modeled CO distribution in areas where assimilation is not carried out, signifies that the improvement is due to the better simulations of CO in the source region over Asia. These improved values are then transported across the North Pole.

pg. 31712, I. 17: This discussion seems out of place at this point in the conclusions. I suggest adding a separate Discussion section which also provides the space needed to highlight the important aspects.

We have followed this recommendation. The Summary section was shortened and a separate sub-section was added to the new Section 5 : Section 5.3 : Discussion of the validation results

pg. 31713, I. 7: Similarly, this section seems out of place and better be put in a Discussions section.

This section was taken out of the Summary and transferred to Section 5.3 (Discussion of the validation results)

#### Fig. 6, 7, 10 need to have latitude-longitude coordinates indicated.

The figures were modified to include latitude-longitude coordinates. Figure 6 is already shown above. New figures 7 and 10 are shown below.



Figure 7. CO observations (in ppbv) from MOZAIC measurements taken in May 2008 between 200 and 250 mb.



Figure 10. Flight paths of the five aircraft during the summer campaign (June and July 2008) of POLARCAT.