Authors response to reviewers comments on manuscript acp-2011-723

We would like to thank the three anonymous reviewers for their insightful comments on the manuscript. Responses to their general comments have been collated as follows:

Flow and cohesiveness of the manuscript.

It was noted that the sub-topics presented in each section of the paper did not follow each other very smoothly, with no cohesive section to tie them together. To address this issue, the final paragraph of the introduction has been expanded to clarify the structure of the manuscript to provide a more detailed overview of the work reported in each section. Additional sentences have also been included at the beginning, or end, of each section to provide a clearer link to the subsequent, or following, section.

CO inversion and impact on species emitted from biomass burning.

The justification for using an inversion with CO observations to constrain the biomass burning emissions used in the model was not very clear in the manuscript. The first paragraph of section 5.2 has been rewritten to clarify how the CO inversion relates to the adjoint sensitivity calculations presented in section 5.1. In particular the scaling of the other species emitted from biomass burning through the emission factors is more clearly emphasized, which was also commented on by the reviewers.

Reviewer #1 asked about updated emission factors for biomass burning emissions, and the possible impact. We chose to use the Andreae and Merlet (2001) emission factors as these are already used in the model. Updated emission factor databases are available (e.g. from Simpson et al., 2011, ACP) but, given the uncertainty in the relationship between the emission of different species from biomass burning, it isn't clear what impact this will have on the emissions and chemistry in the model. This is something that is being considered in the analysis of other measurements under the BORTAS project.

Change in ozone due to change in biomass burning emissions, and bias relative to ozonesonde, TES and IASI ozone data.

The change in ozone due to the change in biomass burning emissions is within the variability of the model and observed ozone distribution across the area and time period considered in the analysis. We presented the mean bias of model ozone relative to the ozonesonde, TES, and IASI observations. Figure 13 has been modified to show the standard deviation of the difference for each dataset. Although the change in the mean bias is within the variability, and noise of the measurements, in each case, the variability does not change reflecting that model ozone uncertainty associated with biomass burning emissions are systematic in nature. The mean bias is probably a good enough metric for quantifying the impact of the biomass burning emissions on the model ozone distribution but other metrics should be considered when evaluating the impact of these emissions on the model chemistry, which will be undertaken in subsequent analysis as part of the BORTAS project.

FLAMBE biomass burning emissions in the model.

The text in sections 2.1 and 3 has been modified to clarify some points of how the FLAMBE inventory was derived and implemented in the model. In section 2.1, it has been

made clearer that the inventory used MODIS active fire data from the two MODIS instruments on the NASA EOS Terra and Aqua satellites. In section 3, the text was changed to state that the emissions are aggregated over the resolution of the model simulation which was not clear in the original manuscript.

Response to specific comments from Reviewer #1

How does the mean bias of GEOS-Chem relate to the accuracy of the measurements being compared? I asked this since TES, IASI have different biases. Are these accounted for when reporting the mean bias of GEOS-Chem?

The reported biases in the TES and IASI ozone retrievals are taken into account when the bias between GEOS-Chem and each of these ozone observations is calculated.

P 25116 line 25. The filament described in the text is hard to see.

This has been removed from the sentence.

P 25118 line 5. Is this still consistent with the 8% bias reported in the previous page?

The bias profile defined here is consistent with the 8% bias which Dufour et al. (2011) calculate as a mean bias from the surface to 6km.

P 25122 line 5. What do you mean by "normalized" in this context? Would a simulation of tagged O3 show similar sensitivity?

In this context normalized means that the sensitivity (i.e. the gradient of the cost function with respect to each emission) have been divided by the cost function so that each sensitivity can be directly compared against the others.

Table 1. Please describe the mean (median) bias column in the caption. I understand that this was describe in later section, but at first glance it can be interpreted to be a bias of ozonesonde.

The caption has been modified to clarify that the mean (median) bias is that between the model and ozonesondes.

Figure 9. Where is (a) and (b) in the figure?

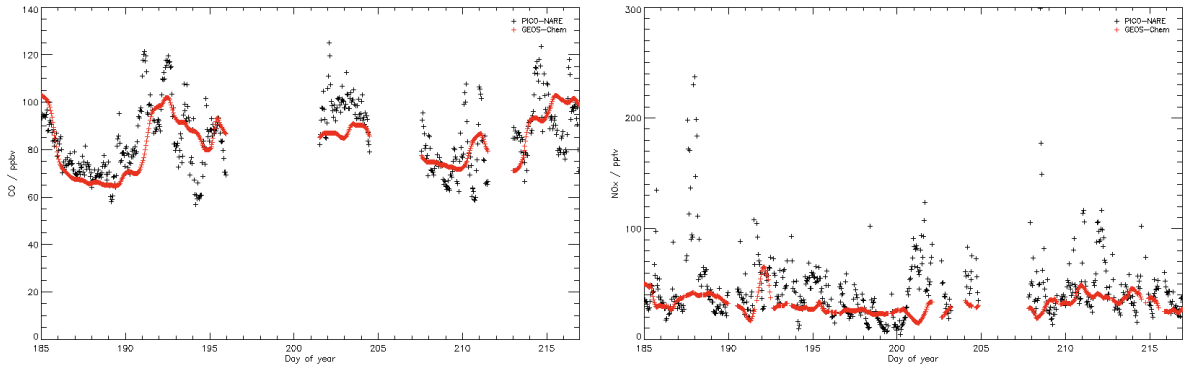
The missing labels have now been included.

Response to specific comments from Reviewer #2

Comparison of the model to ozone precursors would be very valuable. Many models show good agreement with ozone observations while not representing CO, NOx, NOy and VOCs well. An "analysis" of the BORTAS surface measurements at Dalhousie and Pico Mountain Observatory (which the authors say is beyond the scope of this paper) is certainly not required, but using the observations of CO, NO, NOy, etc., to evaluate the model results at those sites would be valuable.

While the main aim of this manuscript has been to evaluate the model ozone distribution, comparisons of other model tracers against some of the surface measurements have also

been made. The figure below shows the comparison of the model CO and NOx to the observations from the Pico Mountain Observatory observatory. The model CO compares reasonably well with the observations, with a mean bias (model minus observation) of -2 ppbv and correlation coefficient of 0.57, and generally follows the observed trend. The model NOx has values comparable to the background NOx mixing ratio observed at the Pico Mountain Observatory with a mean bias of -14 ppbv and correlation coefficient of 0.14.



*Abstract, 1st sentence: Do you mean "We analyse *the sensitivity of* the tropospheric ozone distribution . . . "*

The sentence has been changed.

Section 3: For which species are MEGAN biogenic emissions calculated? Is this online in GEOS-chem, or have you used pre-calculated emissions?

A sentence has been added to clarify that isoprene and non-methane VOCs are calculated online from the MEGAN biogenic emissions.

Section 4.3.2, line 25: The sensitivity of the retrieval does not depend on the vertical grid used in the retrieval. Rewrite this sentence.

The sentence has been rewritten.

p. 25124, line 11-12: Are these all of the CO sources included in the model? What are "monoterpene sources"? These should be explained in section 3. How is the CO produced from oxidation of methane and VOCS treated in the inversion?

All CO sources are included in the model with oxidation of methane and non-methane VOCs modelled following Duncan et al. (2007) - section 3 has been modified with these details.

p. 25128, line 14: You should clarify that PAN is emitted directly by fires in GEOS-chem (I guess that is the case), but there is no evidence of direct emissions in reality.

PAN is not emitted directly by fires in GEOS-Chem. A sentence has been added to section 3 to provide brief details on the species emitted from biomass burning in the model.

Figures 2 and 3: Too many significant figures given. Integers or tenths are sufficient.

The format of the numbers in these figures has been changed.

Fig. 7: Clarify the caption to indicate the model results have been transformed by the satellite retrieval averaging kernel and a priori.

This change has been made.

Fig. 9: "a" and "b" labels are missing.

The labels have been added.

Fig. 13: Do these profiles show the mean of the differences for each observed profile, or is it the difference of the mean? It should be the first, and please show error bars. And clarify in the text what is shown.

The profiles show the mean of the differences for each observed profile. 1-sigma standard deviations are now shown in the plots, and the text has been modified to describe what is being shown.

Response to specific comments from Reviewer #3

*Abstract, line 2. Should this read "We analyse the *sensitivity of* the tropospheric ozone distribution over North America... "?*

The sentence has been changed.

Page 21507, line 20. The text states north of 50○N, but the line in Fig 1a appears to be at 45○N.

50N, as stated in the text, is correct and the figure has been changed to make this clearer.

Page 25116, line 10, rephrase as "... tropospheric distributions (600- 400 hPa, approximately 4-6 km) averaged ...

The sentence has been changed.

line 14, rephrase to "... reported in Table 4 of Boxe et al. (2010), ..."

The sentence has been changed.

Lines 23-24 and Fig.7/Fig.8. I would like to see a little more discussion of why the modelled ozone data have the strong north-south gradient over North America, yet this feature is not seen in either the TES or IASI ozone data. The authors mention this feature in the text of the paper but do not propose any reasons for it.

The north-south gradient in the ozone concentration is due to there being more sources of ozone precursors, from anthropogenic and biogenic as well as biomass burning sources, over the USA compared to Canada. A tagged ozone simulation which differentiates between the boreal (latitudes poleward of 50 degrees N) and non-boreal regions showed that there is a clear partitioning between the influence of precursor emissions from the boreal and non-boreal regions reflecting the gradient seen in Figure 2. The results from the tagged ozone simulation are not shown but are described in section 4.

Page 25123, line 9, and caption of Figure 10, emission units should be molecules cm-2 s-1. Ditto Page 25124, line 11.

The change in the format of these units has been made throughout the manuscript.

Page 25127, lines 5, 8, and 15, and Page 25128, lines 25 and 27. Given all the uncertainties, can the authors really quote ozone level changes to 2 decimal places of accuracy? There are other places in the paper where 2 decimal points are used, and then just 1 decimal place is used elsewhere.

The text has been modified to quote to just 1 decimal place.

Page 25129, lines 22-23. A reference is needed for the Master Chemical Mechanism.

A reference has been added.

Fig 2A, the triangles are hard to see - could they be coloured white and/or made slightly larger so they stand out more? I suggest making the coloured lines of the boxes thicker so they are easier to see (and not on top of each other).

These changes have been made.

Figure 9, (a) and (b) are missing from the figure.

These labels have now been added to the figure.

Figure 10, The figure title uses molec/cm2/s but the text uses molec cm-2 s-1 - Change the figure title to use the second version of the units (with negative powers) to be consistent with the rest of the paper.

These changes have been made.