We thank the reviewers for thoughtful and helpful comments. Responses to each of the suggestions are below:

Response to Reviewer 1

Page 9383, line 8: Please provide a brief description of the diurnal steady state approach.

The following text was added:

"In the DSS approach, each input point of in-situ data (generally taken from data merges averaged to a 1-minute common timeline) is integrated by the model to find an internally self-consistent diurnal cycle for all computed species to within a given tolerance (<1%). Predictions are then taken from the computed diurnal cycle at the same time of day as the data for direct comparison of radical predictions and measurements. "

Page 9384, line 14: Please state the tolerance used in this work.

This value (<1%) is now included in the added text described above.

Page 9386, lines 1-4: Please provide further details regarding the TOGA dataset and the correlations of available data with CO/acetone/methanol.

TOGA has been defined *("the NCAR Trace Organic Gas Analyzer (TOGA) Gas Chromatography – Mass Spectrometry instrument")*, and additional details have been added in the text for the correlations, including the percentage of time during each phase that values that were filled using correlations (generally less than 2% of the time). Because of the very small number of affected points, the computed equations for correlations have not been listed for reasons of brevity.

"TOGA data are used to fill values 4% of the time during ARCTAS-A and 8.5% of time during ARCTAS-B. For the few remaining points with missing acetone or methanol data (0.5% and 1.5% during ARCTAS-A and –B), values are filled using observed correlations of PTR-MS acetone or methanol data with CO for each phase of ARCTAS. During ARCTAS-A (spring), the correlations are altitude-independent, while during ARCTAS-B, separate correlations are derived for the lower troposphere where pollution from biomass burning is prevalent (CO>200 pptv) and for cleaner portions of the troposphere (CO<200 pptv).

Methyl-ethyl-ketone (MEK) and ethanol data are taken from the TOGA observations. Where data for MEK are missing (<2% of the time for ARCTAS-A and 8% of the time for ARCTAS-B) values are filled using its observed altitude-dependent correlation to acetone for each phase. Missing ethanol (<1% of the time for both phases)is filled using its observed altitude-dependent correlation to methanol."

Page 9392, line 13: Please correct to 'ozonolysis'

Corrected.

Page 9393, lines 18-26: In the description of the model-measurement comparisons for OH and HO_2 it would be useful to provide a point by point comparison (i.e., modeled mixing ratio vs observed mixing ratio for all points) in addition to the plots shown in Figure 8.

Scatter plot of observed and model-predicted values for OH and HO_2 is now included as new Figure 9.

Page 9394, lines 4-6 and lines 18-20: How are the R-Obs/Calc values impacted by the inclusion of points at or below the LOD which are given R-Obs/Calc = 1? Perhaps the points below the LOD should not be included in R-Obs/Calc, it is not clear how much the R-Obs/Calc below 0.04 is skewed by the inclusion of points below the LOD.

> Because we are looking at the scatter of observations around the model, we do not want to limit the exclusion of points to only observations below LOD because that would unfairly bias the comparison. The assumption that R-Obs/Calc=1 applies when both the model and the observation are at or below the instrument LOD (0.01 pptv) .
>
> In the discussion of Figure 9 (new Figure 10), about 8% of the points below the "threshold" model concentration of 0.04 pptv include paired model and observed values both below LOD. The median R-Obs/Calc ratio below that threshold reported in the text (0.98) has not been adjusted for those cases when both the model and the observations are at or below LOD. The reviewer is correct in pointing out that we should consider the impact of those points. When we adjust the ratio for those 8% of the points to equal a value of 1, the median changes only slightly to a new value of 1.0. This is now corrected and described in the text.
>
> *"Below an arbitrary threshold value of 0.04 pptv (vertical line in Fig. 10), the median R-Obs/Calc is 1.0, while the model over predicts OH at higher concentrations (median R-Obs/Calc of 0.86). Note that the previously described assumption of R-Obs/Calc = 1 for points where both the model and the observation is at or below LOD affects about 8% of the points below the threshold. When that assumption is removed, the median R-Obs/Calc changes only slightly to 0.98."*

Page 9394, line 19 and Figure 8c: 'ppt' is used instead of 'pptv'. Please check that there are not other instances of this in the manuscript.

> Corrected.

Page 9395, lines 14-18: There has been some debate regarding the atmospheric relevance of the reaction of electronically excited NO_2 with water vapour, with subsequent experiments under more appropriate conditions to the atmosphere suggesting that the reaction of NO_2^* with H_2O is unlikely to occur in the atmosphere (S. Carr, D.E. Heard and M.A. Blitz, Science, 324, 5925, 336, 2009). Please include some mention of this work in your discussion.

> Done.
>
> *"Some recent studies have suggested that at high solar zenith angles, an additional source of OH from the reaction of electronically exited NO_2 with H_2O may result in a source of OH and HONO that rivals the magnitude of the primary source from $O(^1D)+H_2O$ (Li et al., 2008), although subsequent studies have suggested that the reaction is unlikely to occur in the atmosphere (Carr et al., 2009)."*

P. 9397, line 7: What was the limit of detection for BrO? Were there any measurements of IO? How would the presence of iodine radicals impact the conclusions in this work?

> The LOD for BrO on the order of 2 pptv. This is now included in the text.
>
> There were no measurements of IO during ARCTAS. A brief discussion has been added at the end of section4.2.3:
>
> *"While there were no measurements of iodine radicals available during ARCTAS, there is recent observational evidence of the presence of IO over isolated regions in the Arctic*

regions of up to a few pptv (Mahajan et al., 2010). That study suggested that at those concentrations, IO will deplete O_3 at rate comparable to BrO, as well as enhance the effect of bromine-catalyzed O_3 depletion. Modeling studies have indicated that iodine chemistry is also expected to have a significant impact on the HO_2/OH ratio, parallel to that from BrO (Bloss et al. 2005, Saiz-Lopez et al., 2011).”

Page 9402, line 26-28: Is there any suggestion as to the source of the remaining model discrepancy? Is the discrepancy consistent with results from other field campaigns in similar regions?

We have done an exhaustive search for the reason for the discrepancy during ARCTAS and have not found an adequate explanation.

Measurements of HO2 were also available from the CIMS instrument during ARCTAS-A, and a detailed comparison is in Ren et al. 2012. In addition, HO_2+RO_2 from CIMS were available during TOPSE, though the number of points with sufficient precursor information is small (<200 points). The following text was added at the end of Section 4.2:

“*Few data sets are available for comparison to these results that include simultaneous measurements of HO_x and its precursors in the free troposphere of the Arctic. A comparison of the ATHOS-LIF HO_x measurements to those from the previously described CIMS instrument which was also on board the DC-8 aircraft during ARCTAS are discussed in detail by Ren et al. (2012). That study shows that CIMS measurements of HO_2 were generally higher than measurements from ATHOS-LIF, with median ratios of $[HO_2]_{CIMS}/[HO_2]_{LIF}$ of 1.65 during ARCTAS-A and1.28 during ARCTAS-B. This results in a median value for R-Obs/Calc (HO_2) within 10% of 1 from the surface through 8 km during ARCTAS-A, compared to values from 0.5 to 0.7 for the ATHOS-LIF (HO_2) measurements described above and in Table 3.*

The same CIMS instrument was used during the TOPSE campaign, including measurements of HO_2+RO_2 (Cantrell et al., 2003), though the number of points available for modeling that include both HO_2+RO_2 observations and the major precursors (H_2O_2 and CH_2O) is limited to less than 200 points. While the CIMS data gave R-Obs/Calc (HO_2) values near 1 during ARCTAS-A, values of R-Obs/Calc (HO_2+RO_2) from TOPSE (using the current version of the LaRC model) range from 0.64 at the surface to 0.38 at 6-8 km, more similar to the values using ATHOS-LIF measurements during ARCTAS-A. Note, however, that there is a considerable amount of scatter in this comparison (r^2=0.35), compared to the highly correlated model-measurements pairs during ARCTAS with r^2=0.75 to 0.85 in the free troposphere.”

P. 9405, lines 15-18: What is the change in the branching ratio for CH_2O production from the surface to the upper troposphere?

During ARCTAS-A, the branching for CH_3O_2+HO_2 to directly produce CH_2O varied from 15% at the surface to 25% in the upper troposphere. During ARCTAS-B, the branching varied from 10% at the surface to 23% in the upper troposphere. Added sentence in text (section 4.2.5):

> *"This branching ratio varies from 10-15% at the surface to 25% in the upper troposphere for ARCTAS conditions."*

Table 1: Please change 'Rate' to 'Rate coefficient' in the table heading, and include appropriate units. Please give the parameters used to calculate the rate coefficient for R13. For reactions 20-23 it looks as though the reactions are with 'hγ', and not 'hν'.

Corrected

Table 7: Please define the term 'Pct'.

Corrected.

Figures 17, 18 and 20: Please see above comment regarding the use of 'ppt'. These figures are rather small in the ACPD format, please check that they will be clearly legible in the ACP format.

Done.

General comment

Recent work by Fuchs et al. (Fuchs et al., AMT, 4, 1209, 2011) has indicated potential interferences from alkene-derived RO_2 radicals in measurements of HO_2 by the FAGE technique. Please provide some indication as to the expected levels of interference for the ATHOS-FAGE instrument used in this work, and whether the interference has been considered in the model comparisons.

The following text was added to section 4.2:

> *Recent work by Fuchs et al. (2011) show that HO_2 measurement techniques using chemical conversion to OH through the addition of NO such as is used for ATHOS are likely to suffer interferences from aromatic-, alkene-, and isoprene-based peroxy radicals (RO_2), resulting in a high bias for the HO_2 measurement. Interferences due to RO_2 from smaller alkanes such as methane and ethane are negligible. During ARCTAS, because the HO_2 observations are persistently lower than the model predictions, any interference from RO_2 radicals would only exacerbate the observed to modeled discrepancies. During the summer phase ARCTAS-B and primarily in the boundary layer, about 17% of the data were comprised of total RO_2 dominated by those from alkene, isoprene, and aromatic chemistry, as predicted by the model. Interestingly, the median R-Obs/Calc for HO_2 for those points is about 25% larger than that for points where RO_2 is dominated by small alkanes, implying an increase in the observation value relative to the model prediction, consistent with the bias identified by Fuchs et al. (2011). Cases where RO_2 is dominated by alkenes, isoprene and aromatic chemistry are rare in the free troposphere during ARCTAS-B, and negligible during ARCTAS-A.*

Response to Reviewer 2

1) It has recently been reported that ground based OH measurements by the Penn State LIF instrument may be subject to an unknown interference related to the presence of ozone and alkenes (Mao et al., ACPD, 12, 6715-6744, 2012). Although the conditions encountered during ARCTAS and TOPSE are likely quite different than the conditions where the interference is observed, the authors should provide a discussion on whether the measurements reported here are subject to any interference.

> This is discussed in Ren et al. 2012. Text has been added at the end of section 4.2, along with a reference to that manuscript.
>
> *"In addition, recent work by Mao et al. (2012) have recently reported that ground based OH measurements by the Penn State LIF instrument may be subject to an interference related to biogenic volatile organic compounds, particularly at higher temperatures. Very few biogenic hydrocarbons were measured during ARCTAS-A, so this interference should be negligible for that data set. There were more biogenic compounds measured during ARCTAS-B, though at relatively low temperatures; Ren et al. (2012) determine that the level of this interference during ARCTAS cannot be quantified at this time."*

2) Similarly, it has recently been found that HO_2 measurements using chemical conversion to OH through the addition of NO may also be subject to interference from alkene-based peroxy radicals (Fuchs et al., Atmos. Meas. Tech., 4, 1209-1225, 2011). The authors should comment on whether the ATHOS instrument is subject to this interference and whether the measurements have been corrected.

> Text added to discussion; See response to Reviewer 1.

3) As discussed in the manuscript, previous modeling of the ARCTAS HO_x measurements found that the measured concentrations of HO_x radicals could be reproduced by including heterogeneous loss of HO_2 onto aerosols (Mao et al. Atmos. Chem. Phys., 10, 5823-5838, 2010). However, including a similar loss process could not account for the model/measurement discrepancy in this manuscript. Unfortunately there is little discussion in the manuscript concerning this discrepancy. The paper would benefit from an expanded discussion regarding the potential reasons for the different results from these two models. If aerosol loss does not explain the model/measurement discrepancy, can the authors suggest other possible reasons?

> The reason for the discrepancy is because many of the HO_x precursor fields that are generated by GEOS-Chem are generally lower than the observed fields; e.g., when the box model is run using the GEOS-Chem predictions as input, the predictions for HO_x by the two models are equivalent. GEOS-Chem predicts lower values for H_2O_2, CH_2O and H_2O than are observed during ARCTAS, as well as lower values of $J(O^1D)$, and this results in the lower predictions of HO_x, which (fortuitously) are at the order of magnitude of its measurements
>
> As stated in the text, global models are useful for assessing relative influences on the chemical system as a whole, and Mao et al. (2010) identified HO_2 loss to aerosol as a potentially important process impacting the Arctic HO_x budget. However, a direct comparison of in-situ HO_x measurements to a model can only be achieved when the observed precursors and physical parameters are used.

While this work confirms that the loss of HO_2 to aerosol as described in Mao et al. (2010) can affect the predictions (up to 23%), we show through direct consideration of the observed precursors that it is not sufficient to fully reconcile the difference between measurements and theory.
This point is now elaborated upon in the text throughout section 4.2.4.

We have done an exhaustive search for the reason for the discrepancy during ARCTAS and have not found an adequate explanation.