We thank the reviewer for the many helpful comments, and we believe our responses to these comments have resulted in an improved manuscript that is easier to follow and understand. In particular, we have added a schematic to the Appendix, reproduced at the bottom of these responses, which we believe greatly improves the clarity of the description of the overlap algorithm. We have also improved the discussion of our results to provide better understanding of the role of scavenging in the NO_x and O_3 budgets. Our responses to the detailed comments are below. The original comments are in black, and our responses are in red.

Specific Comments Abstract, line 11: “doubles the lifetime with respect to. . .” It may help if the authors are more concrete here, ie., double with respect of treatment frozen precipitation removal with a Henry’s Law mechanism? Utilization of average cloud cover? (Later in the manuscript the “doubles” applies to the inclusion of improved ice removal). Also, “doubles” is somewhat altitude dependent. Maybe “increases by factors of x to y” is better?

We have changed the statement to “The UCI algorithm doubles the lifetime of HNO_3 in the upper troposphere relative to a scheme with commonly used fractional cloud cover assumptions and ice uptake determined by Henry’s Law and provides better agreement with HNO_3 observations.” While we agree that the change in the lifetime is altitude dependent, we specify that we are referring to the upper troposphere, which is the most important region with respect to impacts on NO_x and O_3. We thank the reviewer for pointing out here and below that on Page 24421, lines 20-21, we state that the doubling of the lifetime was due to the ice treatment alone. That statement was erroneous and we have corrected it as specified below.

Page 24416, line 6: “effectively buried as the crystals continuously evaporate and regrow” this sounds ambiguous; it could be interpreted as the trace gases not being rereleased once they are “buried”, but that does not seem consistent with the last section of the appendix (which in itself is not very clear).

We have changed the wording to “effectively buried as the crystals grow” since the net growth is what matters and the gas is buried as long as the growth rate exceeds the desorption rate, as stated on Page 24420, Line 29-Page 24421, Line 1. We agree that the last section of the appendix was not very clear and have changed Page 24450, Line 20 - Page 24451, Line 2 to read: “For frozen precipitation, we assume that there is minimal diffusion of buried gases within the hydrometeors, i.e. that the gas remains in the layer in which it was initially trapped. Thus, the mass of tracer that evaporates is given by the amount of tracer dissolved in the ice mass that evaporates within the layer, which is proportional to the decrease in precipitation rate:

$$\Delta iwc_{EVAP} = (p'_{MC} - p_{MC}) F_{MC} \frac{\Delta t}{\Delta Z}$$

Following Equation A3.1.1.2, χ_{MCEVAP} is given by

$$\chi_{MCEVAP} = \min(\chi'_{MC},\ \mu(T) \cdot \Delta iwc_{EVAP} \cdot v_{CF} \frac{m_\chi}{m_w})\text{”}$$

Page 24417, line 7: Given the host of different precipitations, grid=averaged vs. local, and at top or bottom level, it may be useful to have a summary table (maybe in the appendix).

As stated above, we have added a schematic to the Appendix, which is reproduced at the bottom of these responses. We believe that this will help alleviate the confusion over the various precipitation rates.

Page 24418, first line: In the equation for Fscav, are we assuming that the parametric rate of conversion to precipitation (k) is the same for liquid water and for ice?

We have clarified the description of k by removing "assumed parametric" from the sentence and adding "For stratiform precipitation, k is usually calculated from the precipitation rate and condensed liquid water content, assuming a minimum value of 1×10^{-4} s^{-1} (Giorgi and Chameides, 1986)."

Page 24418, last paragraph: If I read it correctly, JK 99 assumes maximum-random overlap of clouds, similar to what the authors adopted in their radiation paper. Would their current assumption of maximum overlap only make a difference in the results?

JK99 does indeed assume maximum-random overlap, as stated on Page 24418, Line 21. Based on Figure 1 of JK00, our overlap assumptions are very similar to the ones adopted in that paper, which gave results similar to JK99 as stated on Page 24418, Line 26 – Page 24419, Line2. We have changed Page 24419, Line 5 to read "We assume maximum overlap of clouds in connected precipitating layers, which closely follows JK00 based on their Figure 1."

Page 24421, lines 20-21. See comment re. similar statement in the abstract. However, please note that here the "doubling" of lifetime is due only to the treatment of frozen precipitation, whereas the abstract suggests that it is due to both the treatment of frozen precipitation and cloud overlap.

Thank you for pointing this out. Our statement here was incorrect, as the doubling of the lifetime results from both the ice treatment and the cloud overlap. We have changed the statement to read "We find that this ice treatment increases HNO_3 abundances in the upper troposphere by 100-300% compared to assuming Henry's Law for ice."

Page 24423, section 3, simulations: I found myself constantly searching through these pages to see which case was which (UCI=GMI, UCi-HLAw, etc.). What about a summary table?

We have added the following table, which shows the uptake and overlap assumptions used for the primary set of simulations (excluding the sensitivity runs):

	HNO_3 Ice Uptake			H_2O_2 Ice Uptake			HNO_4 Ice Uptake			Cloud Overlap	
	None	**KV06**	**HLaw**	**None**	**LC98**	**HLaw**	**None**	**KV06**	**HLaw**	**UCI**	**GMI**
UCI		✓		✓			✓			✓	
UCI-HLAW			✓	✓			✓			✓	
UCI-NOICE	✓			✓			✓			✓	
GMI			✓	✓			✓				✓
UCI-H2O2		✓			✓		✓			✓	
GMI-H2O2			✓			✓	✓				✓
UCI-HNO4		✓		✓				✓		✓	
GMI-HNO4			✓	✓					✓		✓

Page 24424, lines 13 and following. The UCI-QUANTIFY scenario seems somewhat irrelevant other scenarios which focus on the impact of different scavenging assumptions. UCI-QUANTIFY simply changes the emission scenario. Given the uncertainties in these scenarios, one could have simply scaled the emissions if one wanted better agreement, or, alternatively, the authors can simply refer to the better agreement without introducing yet another scenario.

While we did not make the case very well in the manuscript, we disagree that the QUANTIFY results could have been achieved by simply scaling the emissions. The key point regarding the change in the emissions scenario is not that the NO_x emissions increase, but that they have a very different spatial distribution, and it is the overlap between this spatial distribution and the spatial variations in scavenging that is interesting. We have changed Page 24424, lines 11-16 to read: "Version UCI-QUANTIFY adopts the new emissions used by the European Union QUANTIFY program (Hoor et al., 2009), which include updated EDGAR emissions and detailed inventories for the road, shipping, and air traffic transport sectors. There is a small net increase (~14%) in surface NO_x emissions, while aircraft NO_x emissions are scaled to be the same as in the standard UCI simulations. However, the spatial distribution of NO_x sources is significantly different than in the UCI simulation. The UCI-QUANTIFY simulation thus allows us to examine the importance of the relationship between the spatial patterns of NO_x sources and the spatial patterns of removal for determining HNO_3 abundances in remote regions." We have also changed Page 24433, Lines 3-4 to read "The results from the UCI-QUANTIFY simulations show that the relationship between the spatial distribution of NO_x sources and the spatial distribution of removal is indeed critical"

Page 24424, line 18. If I am not mistaken, the "GMI" algorithm is really the one originally used in GEOS-CHEM, as explained in Liu et al. and other publications, so proper acknowledgment should be given.

Thank you for pointing out our oversight. We have changed the statement to read "…..we adapt the algorithm used by the GMI model (Considine et al, 2005), which is based on Liu et al. (2001)."

Page 24425, line 8: equation for loss rate. There are two issues here: a) please define F, and b) previously, the units of the precipitation rate have been given as kg m-2 s-1, whereas the units for k seem to assume mm for the precipitation. It will not change the equation, but might as well be consistent in the units.

We have corrected this equation - "F" should have been "F_{scav}", which has already been defined. We have also divided the precipitation rate by the density of water and multiplied by $1x10^3$ mm/m to fix the issue of the units.

Page 24428, first paragraph. This is the first example of results being presented which seem "surprising", yet no explanation or conjecture is given for the model behavior. In this case, HNO3 in UCI and GMI are very similar, despite the more than a factor of 2 difference in lifetimes. Is this due to an averaging effect (i.e., the 250% difference holds in only localized regions), or is it due to the non-linearity of the system, the impact on HOx, etc?

We agree that the phrasing was unclear. The main issue is that the scale of the plots makes it appear as though the differences are very small, when in fact they represent large relative differences. The variations in the difference between UCI and GMI do also reflect variations in the 250% difference in lifetime. We have rephrased Page 24428 Lines 2-5 to read: "The differences in absolute HNO_3 abundance between UCI and GMI are small in many cases, but represent large relative changes (100-500% increases; Section 4.1.3) nearly everywhere. The variations in the differences between UCI and GMI HNO_3 seen in Figure 2 reflect spatial inhomogeneities in the differences in the loss frequency. Given the magnitude of the absolute differences in HNO_3 abundance,…."

Page 24429, line 17: "by the boxes in 3c (not b)".

Fixed, thank you.

Page 24430, lines 5 to 10. First, it seems that the increase in HNO3 in UCI relative to GMI is much larger than the corresponding NOx increase. Why? There is no explanation, and something should be attempted, given the importance of the NOx increase to the ozone increase. For example, is this because there are other important NOx reservoirs, such as PAN? Are we seeing a non-linear behavior in the NOx/HNO3 ratio? Also, the statement that the impact is small in the tropics because of "low concentrations of nitric acid and NOx" is not very convincing: we are looking at percent changes, not absolute differences.

The impact on NO_x is small because HNO_3 recycling into NO_x is a very small source relative to emissions, which we did not explain well in the manuscript. We have improved the discussion of Figure 4 by changing Page 24430, Lines 5-9 to read: "The change in NO_x is small despite large changes in HNO_3 because recycling of HNO_3 into NO_x is a very small source relative to surface pollution, aircraft emissions, and production by lightning, particularly when the HNO_3 removal lifetime is less than a few days as it is in both UCI and GMI. Figure 4 also shows that the upper tropospheric differences in NO_x and O_3 are similar for UCI − GMI and UCI − UCI-HLAW, indicating that NO_x in the upper troposphere is much more sensitive to ice scavenging than to cloud overlap. Ice phase scavenging primarily impacts HNO_3 over the midlatitude oceans (Figure 3b), where other NO_x sources are small, while cloud overlap is important primarily in the tropics (Figure 3a), where lightning NO_x production dominates."

Page 24430, discussion of Figure 5 (and Figure 5): Another unexplained result: the ozone profile seems to increase by quite a bit (20-30%) in the middle-upper troposphere. What is the increase in NOx for the same conditions? Also, why does H2O2 decrease? These questions could be answered if the authors looked at some other diagnostics (such as HOx).

We have expanded the discussion to read: "Differences between UCI and GMI are small, but UCI-NOICE gives much larger O_3 abundances than any of the simulations with ice scavenging. Without ice phase removal, the lifetime of HNO_3 lifetime in the upper troposphere increases to ~2 weeks (Section 4.1.1), allowing sufficient time for significant recycling into NO_x. NO_x increases by 50-100% in UCI-NOICE relative to the ice scavenging simulations in these regions (not shown), resulting in the O_3 increases of 20-30% seen here. The increase in NO_x is in fact large enough that supressing ice scavenging perturbs the entire O_3 chemical cycle. Reaction of HO_2 with NO shifts HO_x toward OH, resulting in a net decrease in HO_x, which is reflected in a decrease in H_2O_2 (not shown)."

Page 24431. H2O2 scavenging: Why is there such a small impact on the ozone budget? Why is the H2O2 change due to scavenging so different relative to that of HNO3 (both are very soluble)? Is it due to ice? As I mentioned earlier, it would be useful to see the impact of these processes on other contributors to the ozone budget, such as HOx.

We have re-written Section 4.2 to provide a better explanation of the H_2O_2 results. It now reads:

"The difference in upper tropospheric H_2O_2 between UCI-H2O2 and GMI-H2O2 (Figure 6a), which both include ice phase scavenging of H_2O_2, exceeds 200% (~40-120 ppt) over large regions and is larger than the difference in HNO_3 between UCI and GMI (Figure 3a). The difference in H_2O_2 is much smaller than that seen in the upper troposphere throughout most of the middle troposphere and boundary layer (Figure 6b), except at Southern high latitudes. The difference between LC98 and Henry's Law H_2O_2 ice uptake is larger than the difference between KV06 and Henry's Law uptake for HNO_3, resulting in a greater sensitivity to the formulation of ice phase scavenging in the upper troposphere. However, the difference in H_2O_2 between UCI-NOICE and the H_2O_2 ice phase scavenging simulations (not shown) is smaller than the difference seen for HNO_3 scavenging primarily due to the lower solubility of H_2O_2, so that even with Henry's Law removal the uptake is not complete.

The difference in O_3 between the UCI-H2O2 and GMI-H2O2 simulations, in which both H_2O_2 and HNO_3 are scavenged on ice surfaces, is only slightly smaller than the difference between UCI and GMI with HNO_3 ice scavenging alone (not shown). Likewise, the difference in O_3 between UCI-NOICE and UCI-H2O2 is very similar to the difference between UCI-NOICE and UCI, indicating that the impact of ice phase H_2O_2 scavenging on O_3 is very small. This is because H_2O_2 affects the O_3 budget only indirectly through reducing HO_2 abundances, and is unimportant except near surface NO_x sources."

Page 24431 lines 21-22: "The reason for this can be seen in Figure 7". The figure does not give a reason for the behavior, it just illustrates it.

We have re-phrased this to read “As seen in Figure 7, …..”

Page 24435 line 7: “likely because H2O2 is not a major source. . .” I believe the statement, but why “likely”? Again, this is something that should be easily diagnosed from the model results.

We have removed the word “likely”.

Appendix A. Granted that the algorithm adopted is rather complex, I found it very difficult to follow this section, and, although I am confident that everything is correct, after a while I found it very time consuming to check all the equations. There are some improvements that the authors may consider:

I. Give a“map” for the logical progression. The problem is that variables are introduced, but it is not until the end that you see everything fitting together.

The schematic included at the bottom of these responses provides an overall picture of how the pieces fit together that the reader can refer to as needed throughout the Appendix.

II. It may be useful to have a sketch of cloud overlap with the different variables denoted.

Done – see the schematic below.

III. What about numbering the equations?

We will number the equations but request guidance from the editor on ACP numbering conventions.

IV. It is stated at the beginning that capitals are used for grid-averaged quantities, whereas small letters are used for “local” (subgrid) quantities. But then, we are introduced to fMC, fNC, etc. Why not FMC, since these denote fractions of the whole grid box?

We agree that these variables should be capitalized and have changed them accordingly.

V. The logic becomes hard (but not impossible) to follow once we start dealing with the f(cap), p, p’, etc. on page 24439 and following. As a function of L, some seem to denote quantities at the bottom of the layer, while others at the top. . . I found myself repeatedly going back to these pages every time I wanted to understand one of the subsequent equations.

We sympathize with the reviewer and hope that the schematic below will help alleviate these problems.

VI. I would encourage the use of “max(1,xxxx)” and “min(0, xxxx”) whenever is appropriate. It is used in some equations later in the Appendix, but why not in the set of equations at the top of page 24439? (I think that it is better than saying that quantities are constrained to be between 0 and 1)

We have removed the statement “All $\hat{f}$ are constrained to be between 0 and 1” and changed it to

$$\hat{f}_{XX\rightarrow YY} = \min(1,\ \max(0,\ \hat{f}_{XX\rightarrow YY}))$$

VII. Page 24439, line 5. This is picky, I know, but the argument of the sum should be something like fXX-,>yy where XX, YY = MC, NC, AM, and the sum should be over YY.

Changed to: $\sum_{YY} \hat{f}_{XX \rightarrow YY} = 1$

Page 24462, Figure 5: You cannot see the blue, black lines on left panel. It they overlap one of the lines, which one is it?

We have added “In both panels, the black, blue, and green lines overlap in part or all of the domain” to the caption.

The schematic on the following page has been added to the Appendix. We will seek guidance from the editor regarding the ACP equation numbering convention.

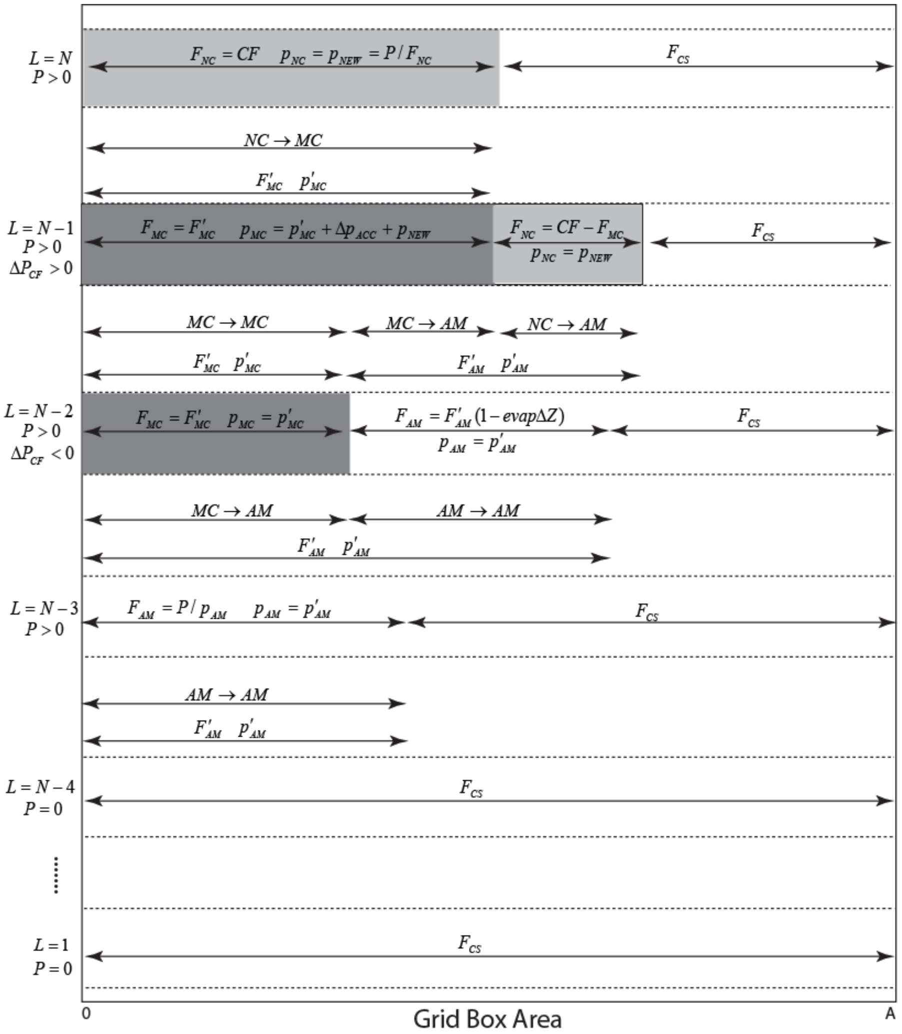

Figure A.1. Schematic of an idealized grid box with N levels, indicated by the horizontal dotted lines. The space between the levels is for illustrative purposes only. Levels N to $N-2$ contain clouds (grey shaded areas) and Levels N to $N-3$ have $P(L) > 0$. All gridbox fractions and precipitation rates are defined in the text and are shown for each level. The terms $XX \to YY$ indicate the gridbox fraction corresponding to $\hat{f}_{XX \to YY}$ as defined in the text (Equations A.x.x to A.x.x). Level N is the first precipitating level, and the precipitating fraction and rate are given by Equations A.x.x and A.x.x. Level $N-1$ provides an example with $CF(L) > CF(L+1)$, $\Delta P_{CF} > 0$ (Equation A.x.x), and $p_{NEW} > 0$ (Equations A.x.x and A.x.x). Level $N-2$ has $CF(L+1) > CF(L)$ and $\Delta P_{CF} < 0$, with the standard evaporation in the ambient region sufficient to account for the decrease in P from level $N-1$ to $N-2$ (Equations A.x.x and A.x.x). Level $N-3$ has $CF(L) = 0$ and the decrease in P from level $N-2$ to $N-3$ exceeds the standard evaporation rate (Equation A.x.x). There is full evaporation of the precipitation in level $N-4$ ($P(L) = 0$).