Reviewer #1:

This paper describes aircraft observations of concentrations of NO3, N2O5, NO, NO2, O3, biogenic VOCs, CO and other compounds together with a range of aerosol parameter in the lower atmosphere made over a couple of months in the autumn of 2006.
The emphasis is on profiles from the ground through the boundary layer. Three distinct cases are chosen based on the composition of the airmasses and are explored in depth. These cases are then summarized. Finally a box model is used to investigate the impact of NO3 oxidation on biogenic VOCs for aerosol production.

General comments. My major comment about the paper is that it contains a significant amount of work but little summary or synthesis. A summary of sections 3-5 would be useful. What has been learnt by all of this analysis? How has the picture changed from what was expressed in the introduction? What are the features seen in the data interpretation sections that the model is focusing on? What are we trying to analyse with the model? Some form of summary would be useful.

The reviewer makes an excellent comment and suggestion. We have added a section 3.4, following the three case studies, to state the common characteristics of the vertical profiles and their implications for nighttime chemistry, biogenic VOC oxidation, and organic aerosol formation.

"Although the individual profiles for each of the cases described above exhibited substantial variability, they also share a set of common characteristics that indicates both active nighttime chemistry that, in some cases, leads to both rapid BVOC oxidation and secondary aerosol production. First, nitrate radical production rates, $P(NO_3) = k_{NO2+O3}[O_3][NO_2]$, were consistently large at low altitude (0.5–2.7 ppbv hr^{-1}) in urban influenced air and were comparable to or greater than primary OH radical production rates during daytime. Second, NO_2 mixing ratios at low altitude were tightly anticorrelated with O_3, indicating that the chemical transformations of these species were dominated by nighttime chemistry rather than photochemistry. Third, many profiles exhibited distinct increases in BVOC mixing ratios at low altitude, consistent with nighttime BVOC emissions into a shallow boundary layer (also see section 5). BVOC enhancements were apparent in measurements from both the PTRMS (high time resolution) and the canister samples (low time resolution, better speciation) instruments. Fourth, the inverse lifetime of NO_3 , $\tau(NO_3)^{-1} = P(NO_3)/NO_3$, was well correlated with and comparable to the first-order NO_3 loss rate coefficients for NO_3, $k(NO_3) = \Sigma_i k(NO_3+BVOC)_i*[BVOC]_i$, determined from PTRMS measurements of BVOC concentrations, implying a large fraction of NO_3 radical production consumed by NO_3-BVOC reactions. Fifth, organic and nitrate aerosol mass were more consistently enhanced at low altitude than were sulfate or ammonium aerosol mass, with several instances of substantial organic enhancements (> 4 μg m^{-3}). Organic enhancement is consistent with a secondary organic aerosol source from NO_3 + BVOC reactions, although in vertical profiles from the October 12th flight, these effects cannot be separated from organic aerosol emissions from biomass burning. Nitrate enhancement is consistent with a source from either organic nitrate aerosol formation or inorganic nitrate arising from hydrolysis of N_2O_5 and partitioning of HNO_3 to ammonium nitrate. Sixth, for non-biomass burning influenced air, a substantial fraction of organic aerosol enhancements were inferred to arise from secondary sources based on a multivariate regression analysis of the AMS. Finally, organic aerosol enhancements in the NBL were correlated with CO, with slopes of $\Delta OA/\Delta CO$ comparable to or slightly smaller than those inferred from photochemical organic aerosol generation in urban plumes. Some slopes were also likely influenced by biomass burning sources."

Case studies How were the case studies selected? Do they represent the extremes of the campaigns, representative examples of all the data etc. Something here to explain why they have been chosen would be very useful.

A sentence has been added at the end of the introduction to section 3:

"These cases have been chosen subjectively as best illustrations of rapid nighttime chemistry, BVOC emission and oxidation, aerosol production or all three. None of the selected cases is inconsistent with the average characteristics of the vertical profiles (see section 3.4 and 4)."

Box model studies. The final section of the paper is the box modeling study. It's not obvious that the paper requires this study. The interpretation of the field measurements is good and provides insight into the problem. The box modeled feels a little 'tagged on' at the end. The authors should strongly consider the extra insight that the modeling study provides and whether the paper would be cleaner and stronger without the box model.

If the authors would like to keep in the box modeling this section needs to be significantly improved. It is not obvious how much chemistry is included in the model. What reactions are included? Where do the rate constants come from? Where does the mechanism come from? The model seems to include a detailed monoterpene chemistry but then a simplified SOA production where mono-terpene+NO3 or O3 SOA? How does the complex gas phase oxidation chemistry interact with the simplified SOA chemistry? There needs to be significantly more detail here or a reference to a paper that provides such detail.

The authors describe a series of idealized simulations using the box model and discuss the ability of the model to produce SOA for a rural airmass and an urban airmass. The initial conditions used in these simulations are different and the author's don't describe this in any detail. Comments made about P(NO3) are much easier to understand if the initial conditions are described. For example the urban airmass (Fig 18) has different initial conditions as well as different emissions than the rural airmass (Fig 17) however this doesn't seem to be discussed in the text. A clear description of the model, the initial conditions and the objectives of the modeling would be very useful here. However, overall I'm not convinced that the box modeling adds anything significant to the paper. There isn't any really conclusions here, other than the model is in some general sense consistent with the observations based on some generic yield from the

NO3+VOC reactions. Given the lack of detail given about the model simulations themselves I'm not convinced of the usefulness of this section.

We agree with the reviewer that the model is highly simplified, and that the purpose of section 6 was not justified as clearly as it could have been in the discussion paper. The reviewer is also correct that the analysis of the observations could stand on its own without the modeling section. The purpose of the model is to determine if nighttime monoterpene emission and oxidation based on the emissions potential in figure 1, together with laboratory SOA yields for NO_3 and O_3 reactions, are consistent with the observed low altitude, nighttime organic aerosol enhancements. Furthermore, the model simulation illustrates the relative contributions of NO_3 and O_3 to BVOC oxidation and SOA formation. As such, we feel that it does contribute to the overall analysis in demonstrating that the observations are consistent with known BVOC emission and oxidation rates.

Rather than omit the model section, we have made two changes in response to the reviewer's comments. First, we have provided a better justification for the model in the introduction to section 6. Second, we have added a table identifying the model reactions, their rates, and relevant references. If the editor feels that the paper would be stronger without section 6, we could simply omit the section and its associated figures and table.

Minor points.

1) Getting 'NO3' into the title some where would be good.

The analysis includes oxidation by both O_3 and NO_3, even though the latter is more important for the cases studied. Thus the more generally worded title, which we have chosen to retain.

2) The paper refers to primary and secondary aerosol but doesn't give a clear description of what is meant here. Different people do have different meanings for this and it would be useful for the authors to explain early in the text their definition.

The following parenthetical statement has been added following the first reference to primary aerosol:

"(i.e., organic particulate matter directly emitted from a combustion source)"

3) P 11866, Line 23. Primary NO would also react with NO3. I don't think the separation of photochemically produced and by implication non-photochemically produced NO is useful here.

This is always a tricky point to state both clearly and succinctly. The point is not that NO_3 somehow reacts differently with photochemically generated NO than with emitted NO, but that NO is always present during daytime due to photochemistry, and that the daytime sink for NO_3 with such photochemically generated NO is large. Photochemical NO is generally a larger NO_3 sink than photolysis.

The phrase "photochemically generated" has been replaced with the parenthetical statement "(in photochemical steady state with NO_2)"

4) P 11867 Line 2. I think the details of the emissions around Houston are probably best put into the description of the field campaign rather than in the introductory text.

We thank the reviewer for this suggestion. This text has been moved to section 2.

5) P11870 Line 5 is there a missing 'and' between isoprene ___ a factor of two for.

"and" added

6) P11872 Line 11. Tau(NO3)-1 is said to be enhanced but compared to what. Presumably the 'free troposphere' but it would be useful to clarify that.

"relative to the overlying residual layer" has been added

7) P11872 Line 18. The authors use the term 'levels' often in the text. I'm not a big fan of this and perhaps concentration or mixing ratio would be a more precise phraseology.

The word "level" has been replaced with "mixing ratio" in all instances where this term was used in this context.

8) P11873 Line 13. 'Which would undergo rapid and nearly exclusive oxidation with nitrate radicals'. The authors offer little direct evidence for this. Perhaps the language could be softened or more clarity given to how this conclusion was come to.

The two sentences immediately following this statement provide the justification:

"The lifetime of α-pinene with respect to oxidation by O_3 (47±4 ppbv below 280 m) was 2.9 hours, while its lifetime against oxidation by NO_3 (127 ± 17 pptv) was 0.9 min. The lifetime for β-pinene would be 16.5 hours and 2.2 minutes against O_3 and NO_3, respectively (Atkinson and Arey, 2003)."

9) P11880 Line 7 'Well to the north of'. This phrase is used multiple times in the text but it isn't obvious what this means. Could the authors be more quantitative (x km north of etc).

The location of Lone Star airfield has been specified more precisely as being 65 km north of central Houston (rather than "well to the north of"). Additionally, all other qualitative or relative descriptions of locations have been replaced with quantitative ones where possible and / or appropriate.

10) P11882 Line 3. Its not obvious to me why night time oxidation of SO2 is negligible. Cloud processing via H2O2 and O3 would probably continue. Do they mean that gas phase oxidation of SO2 and so the potential to increase H2SO4 on existing aerosol would be slow. The authors should provide a reference here to justify the statement.

The reviewer makes a good point. Our statement is based in part on past experience sampling SO_2 rich power plant plumes from aircraft, which show clear evidence for rapid sulfate production during daytime, but no evidence for sulfate production at night. It was also based in part on the lack of clouds in which heterogeneous reactions of SO_2 could take place. For clarity, the sentence has been revised.

"The presence of sulfate within this residual layer most likely indicates the influence of the previous day's photochemistry, since SO_2 oxidation is likely to have been negligible in the dark under cloud free conditions such as those sampled here."

11) P11883 Line 10. Figure 16 doesn't explain the change of scale clearly. This was confusing until I'd read the text. Can the figure caption be improved?

The figure caption now includes the following:

"Panels B and D show the same data for organics and sulfate as do panels A and C, respectively, but on a scale that is also appropriate for nitrate and ammonium."