Response to Reviewers' Comments on Barth et al. 2012 ACPD

Referee #1

1. P16408L11-14: Because ozone mixing ratios agree well with these same satellite data, it suggests that chemical production of O3 in the model is overpredicted and compensates for the excess convective lofting of BL air. Be more clear with your reasoning. Ignoring biases in STE, if chemical production of ozone is accurately captured, would you expect to see a low-bias in UT ozone due to the lofting of relatively clean BL air? i.e., are you assuming a (relatively) clean BL?

   As suggested below regarding the frequency of high CO being greater in the model than in the satellite data, the abstract was modified to the following: Comparison of UT O_3 and CO frequency distributions between WRF-Chem and satellite data indicates that WRF-Chem is lofting CO too frequently from the boundary layer (BL), but the frequency of O_3 mixing ratios are similar. To compensate for the excess convective lofting of BL air, either the chemical production of O_3 in the model is overpredicted or there is too much stratosphere to troposphere transport in the model.

   Emphasize what is new here. Has WRF-CHEM ever been run at this high resolution (with explicit convection), for this large of a domain, for this long of a time period to study this type of problem?

   The first sentence of the abstract now puts more emphasis on the uniqueness of our simulation.

2. 16409L14: there is concern as to the possibility of substantial –> there are substantial

   The wording of this sentence has been modified as suggested.

3. 16418L6-10: You seem to imply that the use of explicit convection contributes to the poor simulation of unorganized shallow convection? Would you expect to do a bit better with a convective parameterization or are you implying that you need an even finer resolution? Be clear.

   Using either a convective parameterization or finer resolution should give better representation of shallow convection. The numerical weather prediction community suggests that convective parameterizations should be used at grid spacings down to 3 or 4 km (Hong and Dudhia, 2012, BAMS). At these grid spacings, the choice of the convective parameterization is important; one that has subsiding air occurring at neighboring grid cells (instead of the same grid cell) would likely perform better. The following sentence has been added in the 2nd paragraph of section 4.2. It is expected that the small unorganized, scattered convection would be better predicted with either a finer grid spacing (e.g. 1 km) or with inclusion of a convective parameterization that represents upward mass flux at the grid cell and downward mass flux at neighboring grid cells.

4. 16418L20-30: Within the discussion section, you should comment on the impact of this three hour bias on the amount and time evolution of the chemical production of ozone.

   In section 4.3, which discusses ozone production on the geographic regions, we have added the following two sentences pointing out the 3-hour bias in model-predicted

convection and its impact on photochemistry in the UT. Convection occurring later in the day would have less photochemistry from that day. Ozone production is occurring mostly during the next day's daylight hours.

5. 16420: Section 3.2.1 doesn't add much to the paper and could be shortened.

   The discussion of the UT CO and O_3 mixing ratios is helpful in providing a sense of how CO and O_3 are spatially distributed, especially as the anticyclone moves from one part of the U.S. to another. Some extraneous phrases were removed from the description, but the section is not substantially shorter.

6. 16422: L21-25: However, the TES frequency ... This sentence is confusing. CO mixing ratios are not 25% lower. It is the frequency of high-CO amounts that is overestimated. Rephrase.

   Thanks for pointing this out. The sentence has been rewritten.

7. 16423: You do not discuss the Huntsville profile.

   The Huntsville comparison is included in lines 18-19 saying that there is good agreement. On line 23, "Boulder and Huntsville" have been added to specify which profiles have high O3 at 12 km.

8. Section 3.22 & 3.23. Perhaps combine into one shorter section.

   The evaluation with observations has been combined into one sub-section. The discussion of the data and how the model results were analyzed to compare with the data was moved to a separate section (now section3).

9. 16424L20-25: STE caveat probably needs to be added to abstract.

   The STE caveat has been added to the abstract.

10. 16426L23-27: Do you have any thoughts on why ozone production is suppressed when stratospheric air is in the region?

    Examination of one of the stratosphere events (28 July in the Midwest) shows that NOx, HOx, and the HOx precursors H_2O_2 and HCHO all have smaller mixing ratios (10-35% less for NOx, 22% less for HOx, 15-40% less for HOx precursors) when stratospheric air is in the region than the day before. CO is also slightly less (10% less) in the stratospheric air (Fig. 10 of manuscript). It suggests that there is less ozone production because there are less HOx precursors. A phrase explaining why ozone production is less is now included in the paper.

11. 16429L24-26: For point (c), I'd suggest listing which "other VOCs" are important regionally but not necessarily domain-wide.

    The other VOCs are described in the previous paragraph. For the Central Plains and Midwest anthropogenic VOCs e.g. propane are important. For the Gulf Coast, isoprene and its products are important. The last sentence of the section has been clarified.

12. 164321L4-8, These interesting differences between inside and outside of the anticyclone could be highlighted a bit more in the discussion/summary.

The contrast of the BL and stratosphere tracers between the anticyclone region and the region just outside the anticyclone has been added to the Conclusions section.

13. 16433L14-16: The statement that WRF-Chem has a high-bias in UT ozone is inconsistent with what you say at other locations in the text including the abstract.

    Thank you for pointing this out. The sentence has been revised to be consistent with the rest of the text.

14. 16433L24-27: Perhaps state that estimate for organic peroxides may be an upper bound due to model chemistry.

    It is now stated that the contribution estimate for organic peroxides to peroxy radical formation is an upper bound because the lightning-NOx emissions are likely underpredicted due to the underprediction of lightning flash rate.

15. Acknowledgements: A little more information on how much these simulations cost and how much time they took would be useful. Quite an impressive computational feat.

    The following information has been included in the paper. The WRF-Chem simulation performed on the bluefire machine (an IBM clustered Symmetric MultiProcessing (SMP) system based on the Power6 chip) at NCAR used 728 processors. One day of integration took ~20 hours to compute. The hourly output files are 39 Gb in size and are available for scientific investigations upon request.

16. Figures: While (perhaps) obvious make sure to add units to Figures 1 and 2. Figure 3 does not show up well. Make larger.

    Units have been added to the figure captions and with the colorbar of Figures 1 and 2. Figure 3 has been enlarged by splitting it in two.

17. Figure 4. Should we be at all concerned about the overestimation of very weak convection?

    The weak convection is not resolved well by WRF at the 4 km grid spacing. Including a convective parameterization or running at even smaller grid spacing (< 1 km), may help reduce the overestimation.

18. Figure 5. Captions are too small.

    The colorbar labels are now larger.

19. Figure 9 is harder to read than Figure 8.

    Figure 9 has been revised to include thicker lines and larger fonts.

20. Figure 10. This plot is too small. Use entire page.

    Fig. 10 should be using the entire page. The font for the legend is now bigger. We will work with the copy editor to make sure the figure reads well in final format.

Referee #2

Major comments

1. Nowak et al. 2004 show ozone vs CO for several flights from the NOAA P3 during the ITCT experiment off the coast of California in 2002. With this type of plot you can clearly identify measurements from the stratosphere, the marine boundary layer or from Asian pollution plumes. I think if you plotted the MOZAIC ozone vs CO values for the UT you would find some interesting regimes that might help to identify the underlying chemical and transport processes. Then sample the model at the MOZAIC times and location and make a similar plot. Do the WRF data fall out in a similar manner to the observations? This would be a very effective method of evaluating the general model performance.

   Plots of CO versus O_3 for the MOZAIC profiles at Atlanta, Dallas, Philadelphia, and Portland were made (Figure 1). The results from the MOZAIC observations do not show the classic stratosphere-troposphere separation (where stratosphere air has high O_3, low CO, and troposphere air has low O_3, high CO). The WRF-Chem results have more of these characteristics. Most of the WRF-Chem results above 400 hPa pressure level ($p < 400$ hPa) have stratosphere air characteristics, with some points indicating troposphere air. A comparison of CO versus O_3 using the MOZAIC profile locations and times does not seem to be effective for identifying the underlying chemical and transport processes. A more thorough analysis, examining these profiles based on the location of the tropopause and/or on potential temperature regimes, would be the appropriate pathway to follow. However, this is beyond the scope of the current paper.

   The plots of CO versus O_3 were also made for the TES overpass tracks. These plots (Figure 2) show a much more consistent picture between WRF and TES. For the upper troposphere (pressures between 400 and 150 hPa), both TES and WRF are characterized with primarily the stratosphere signature (high O_3, low CO) between CO and O_3, but with some incidents of high CO (> 100 ppbv) and fairly high O_3 (90-120 ppbv). These higher values are often in the same latitude region where convection was occurring that day. In general, the CO-O_3 scatter plot from WRF-Chem results has a more narrow spread of points than the TES data. This is in line with the findings of the frequency distributions shown in the manuscript.

   In summary, it makes sense to compare model results and observations via the CO- O_3 scatter plots, but because of the inconsistencies of the MOZAIC profiles with classic CO-O_3 correlations that would require further investigation we have not included this discussion in the manuscript.

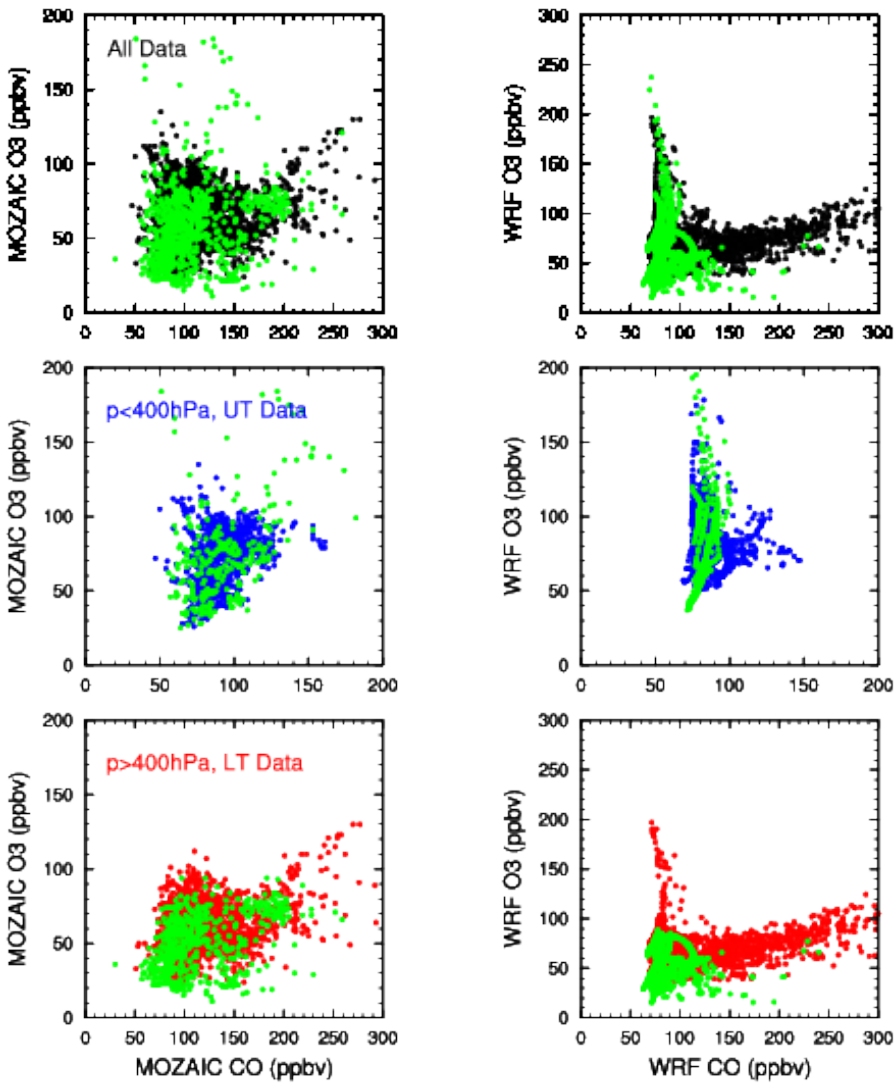

Figure 1. CO versus O_3 mixing ratios from the MOZAIC observations (left panels) and the WRF-Chem simulation (right panels). Top panels are for all altitudes, middle panels for altitudes above the 400 hPa level, and bottom panels for altitudes below the 400 hPa level. The black, blue, and red points are for profiles arriving and departing from Atlanta, Dallas, and Philadelphia. Green points are for profiles arriving and departing from Portland, OR.

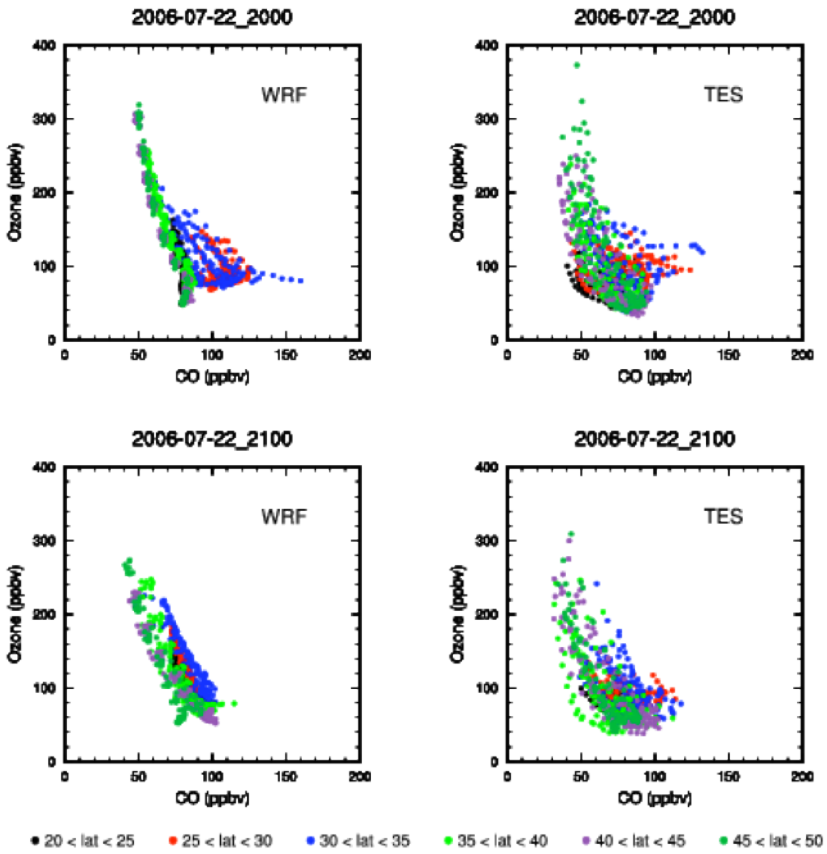

Figure 2. CO versus O_3 mixing ratios from the TES satellite observations (right panels) and the WRF-Chem simulation (left panels) for the two U.S. overpasses on 22 July 2006. TES and WRF-Chem values are for the 400 to 150 hPa region (upper troposphere). The points are segregated by latitude region as denoted in the legend. Plots from the other TES overpasses during the July 16 to August 5, 2006 time period are shown in the appendix.

2. Page 16416 line 16 Why was this particular period chosen for the analysis? While the paper makes good use of the MOZAIC and ozonesonde profiles available during the chosen study period, August 2006 had over 400 ozonesonde profiles from 14 sites to specifically look at ozone produced in the UT from lightning. NASA, NOAA, EPA, Environment Canada and several US universities spent approximately $400,000 (each sonde costs about $1000) so that these data would be freely available for model verification. Why not take advantage of this extensive and unique data set?

   Our original intent was to examine mid-July to the end of August time period. Mid-July was chosen because it coincides more with the start of the North American monsoon allowing a less perturbed upper troposphere with respect to ozone. Further, we had some interest in the heat waves occurring during July and their effects on air quality. Because of the costs of the simulation (both on the computer and in person-hours), we ended the simulation after the first week of August.

3. Page 16424 line 8 How many profiles at each airport? Here it would also be extremely useful to show the MOZAIC O3 and CO profiles from Portland, OR. How much more CO is there above Dallas, Atlanta and Philly, than above Portland? Cooper et al 2006 showed that there was no more CO above Houston and Dallas than above Los Angeles, indicating that the high ozone above Texas didn't have much to do with the lofting of surface pollution, but was more likely from LNOx. How about TES, does it show more CO above the south-central US than above the eastern North Pacific Ocean?

There are 14 profiles for Portland, 11 profiles for Atlanta, 7 for Dallas, and 7 for Philadelphia MOZAIC data. The figure showing the comparison of model results to MOZAIC data now includes the number of data points binned into each altitude level. This is important because the upper altitudes have fewer points because of missing data from the MOZAIC measurements.

Profiles of CO and O_3 were made for the Portland MOZAIC profiles, as the reviewer suggested. There is good agreement between WRF and MOZAIC CO and O_3 mixing ratios (Figure 3). Examination of the individual profiles shows good agreement between model and observations except for two of the CO profiles (out of 14 Portland profiles) that significantly underpredicted observations.

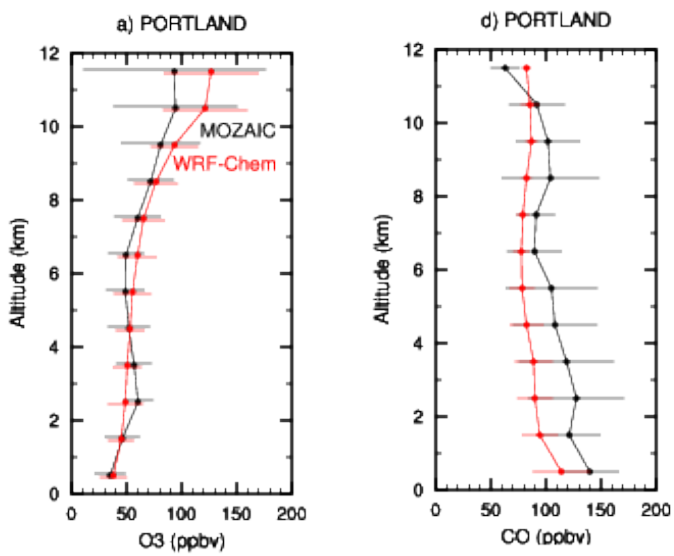

Figure 3. Average vertical profiles for Portland, OR arrivals and departures of O_3 (a) and CO (d) mixing ratios from the MOZAIC data (black lines) and the WRF-Chem simulation (red lines) binned into altitude levels. The profiles are averages over the 15 July to 3 August 2006 when MOZAIC profiles were available. The horizontal lines indicate the standard deviation for each of the datasets.

Specifically, the CO measurements near Portland, which is generally outside the anticyclone (except on 22 July), are of similar magnitude (~100 ppbv) in the UT to the Dallas and Atlanta profiles. In our experience, it has become difficult to discern BL lofting using CO as a tracer because CO has become so well mixed in the troposphere. The CO gradients from the BL to the free troposphere are rather weak. In addition, these high-resolution simulations show that there is an episodic and small-scale nature of convective outflow plumes, stratospheric air filaments, and long-range transport.

Because of the reviewer's points, we have replaced the Philadelphia profiles with the Portland profiles.

4. Page 16434, first paragraph Here the results need to have further discussion and comparison to earlier studies which are listed below. These earlier studies also look at UT ozone over North America using regional scale models and need to be referenced. WRF shows that ozone values within and outside of the anticyclone are similar. But Cooper et al [2007] show that for August 2006 UT ozone above Huntsville is roughly 40 ppbv greater than above the upwind sites (see their Figure 1a). Does the 3 week difference between the 2 studies really make that much difference? How do your results compare if you contrast the center of the anticyclone with the air above the eastern North Pacific or above the Caribbean? Also the WRF finding that stratospheric ozone has a big impact on ozone in the UT anticyclone is different from Cooper et al. 2007, Allen et al 2011, 2012 and from Zhao et al 2009. Why the difference?

   First, thank you for recommending the cited papers for the discussion section of our paper. In comparing the WRF-Chem results with Cooper et al. (2007), there are two things that matter. First, the timing of the events (our study focuses on 15 July to 7 August; while Cooper et al. focus on the monthly average of August 2006) is important. As noted in the manuscript, the anticyclone moved from the central U.S. to the western U.S. and back to the eastern U.S. during July. At the end of July, the anticyclone settled into the southeastern and south-central U.S. and remained there for a few weeks. Because the anticyclone remained mostly stationary over a region rich in VOCs, ozone precursors likely were allowed to accumulate and chemically react to produce ozone. As Jourdain et al. (2010) showed, it is also possible that stratospheric ozone could be accumulating in the anticyclone during August. Second, the resolution of the simulations (both horizontally and vertically) used in this study permits filamentary plumes to be resolved. These plumes are not resolved in the coarse-scale interpolation done by Cooper et al.

   To answer the question of how the results compare between the center of the anticyclone and the air above the eastern North Pacific or above the Caribbean, we compared the O_3 mass per unit area from over a small region of the Caribbean (23-27N, 86-95W) to a region over the south-central U.S. (32-37N, 91-100W) and a region over the southeast U.S. (32-38N, 85-95W). The south-central U.S. and southeast U.S. are not always within the anticyclone, but were more so during the 29 July to 6 August period. The eastern North Pacific is too close to the model's western boundary and was not included in this analysis. Figure 4 shows that the O_3 masses per unit area are similar in magnitude between the three regions. From 23-31 July there is less O_3 over the Caribbean than over the south-central or southeast U.S, but when the anticyclone is over the southeast U.S. in early August, the O_3 over the Caribbean rises to values similar and somewhat higher than those over the south-central and southeast U.S. For the 29 July to 6 August period, all three regions have O_3 masses per unit area between 2.0 and 3.0 kg km^{-2} and average about 2.5 kg km^{-2}.

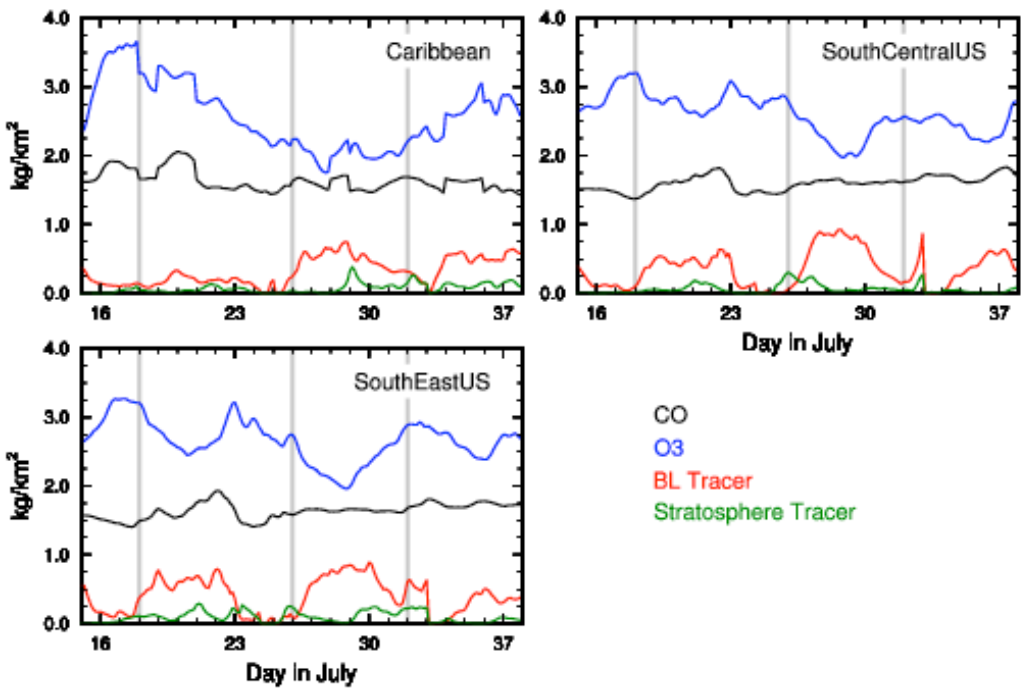

Figure 4. Mass per unit area of CO (black line), O_3 (blue line), decaying boundary layer (red line), and decaying stratosphere tracer (green line) in the upper troposphere (8–12 km altitude range) over the a) Caribbean (23-27N, 86-95W), b) South Central U.S. (32-37N, 91-100W), and c) Southeast U.S. (32-38N, 85-95W). The decaying boundary layer tracer and stratosphere tracer masses per area are the masses of air in units of $2x10^6$ kg km^{-2} and $1x10^6$ kg km^{-2}, respectively. The vertical gray lines delineate the 4 meteorological periods as described in the manuscript.

In comparing our WRF-Chem results at 4 km horizontal grid spacing with the other regional modeling studies (Allen et al 2011, 2012 and from Zhao et al 2009), the main difference between our study and these previous studies is the grid spacing. In all three of these studies, the convection is parameterized requiring the lightning flash rate to depend on the convective parameterization. Figure 15 in the manuscript shows that the stratosphere filaments and chemical production in convective outflow is at a fine resolution. Thus, the finding that stratospheric ozone has a big impact on ozone is due to the high-resolution simulation. However, this result may be an upper bound based on the noted underestimation of lightning flash rate in our simulation. Our finding should motivate further research in improving the representation and understanding of deep convection, lightning-generated NO, and UT ozone by conducting other cloud-scale, large domain, chemistry simulations and through analysis with DC3 field experiment data.

This discussion is now included in the discussion (last paragraph of section 6.1) and conclusions (last 4 sentences of 3rd paragraph of section 7) of the paper.

Minor comments

1. Page 16410 line 14 Here the summer monsoon is described as a seasonal shift in winds that allows low level moisture to flow into the Southwest US. While this is true, the more important feature for deep convection associated with the monsoon is the transport of UT water vapor from the Gulf and Pacific northwards along the Rockies. Without this river of moisture the air above the Rocky Mountains is too dry to sustain deep convection, and the cumulus clouds mix with dry air and are unable to form thunderstorms.

   The following sentence has been added. The low level moisture combined with increased UT water vapor flowing northwards along the Rocky Mountains from the Gulf of Mexico and Pacific Ocean provides ample moisture to sustain deep convection associated with the NAM.

2. Page 16411 line 14 Change to "stratospheric intrusions"

   The wording has been changed.

3. Page 16412 line 7 Please specify if the model is re-initialized across the whole domain or just at the boundaries. Also please describe how WRF-Chem handles stratospheric ozone. Does the model have stratospheric chemistry or is stratospheric ozone just produced at the model boundaries according to ozone profile climatology. Or does CAM-Chem produce the stratospheric ozone at the boundaries? Please provide more information on CAM-Chem. Does it simulate the transport of Asian pollution plumes across the North Pacific Ocean?

   The two outer domains are re-initialized across the entire domains.

   Stratospheric O_3 is obtained primarily from the CAM-Chem model via the initial and boundary conditions, but an upper boundary condition is included for a segment of the simulation. The CO and O_3 values in the troposphere are not affected by the upper boundary condition (compared to tests without the upper boundary condition) for the 13-day segment of the simulation. Between 15-20 km, the WRF-Chem prediction of O_3 remains high (within 20%) compared to ozonesonde observations over the Huntsville site, but is similar in magnitude over the Boulder and Trinidad Head sites. The evaluation for the 15-20 km region has been added to the manuscript.

   The CAM-Chem model is able to simulate plumes from anthropogenic and biomass burning sources originating in Asia and transporting to North America.

   This information has been added to the text in section 2.2.

4. Page 16420 line 11-12 How is the high ozone from across the Pacific simulated? Is this an Asian pollution episode simulated by Cam-Chem?

   This is a confusing question because the statement on page 16420 line 11-12 is about low ozone from the Pacific, which is transported in from the tropics and sub-tropics via the CAM-Chem boundary conditions. The higher ozone in Fig. 5e is from the anti-cyclone circulation and may be a result of in situ chemical production or stratospheric air that has circled around from air above Canada.

5. Page 16423 line 23 Which sites are you referring to, Boulder and Huntsville?

Yes, both of these sites. The text has been modified.

6. Page 16424 line 11-12 Here the model over-predicts CO in the boundary layer. So when the model over-predicts CO in the UT, is this because too much CO is being emitted at the surface, or because the model has too much vertical transport?

   The MOZAIC profiles are representing an urban emissions region as the aircraft land or takeoff from major urban centers. Therefore, the comparison should be interpreted as the model over-predicting CO in the boundary layer in a high emissions region (urban). Unless the storm goes over this specific region, it will be lofting CO concentrations representative of the regional background. The MOZAIC data in the UT represent mixing ratios horizontally advected to the sampling location because commercial aircraft avoid thunderstorm anvils, and therefore are not a result of direct transport from the BL.

7. Page 16424 The comparison between the model and TES and MOZAIC doesn't really make sense. WRF CO is greater than TES, but WRF CO is less than MOZAIC. How can it be both? How does TES compare to MOZAIC? Surely the comparison of WRF to the in situ MOZAIC measurements must be the better comparison?

   One thing to note is that the WRF-Chem comparison to MOZAIC data is good between 9-12 km and worse between 6-9 km. In addition, the comparisons with the MOZAIC data are restricted to aircraft corridors as the planes land or takeoff from the airport. TES data are covering a less specific region, and sampling areas are more representative of the regional background. Figure 6e, as an example, shows the overpass goes through much of the rural Plains states. The convective lofting signature in Fig. 6 is primarily between 26N and 34N. Note also the comment from Referee #1 who correctly pointed out that the frequency of high CO is over-predicted, not the high CO mixing ratios themselves. Again, in Fig. 6c and 6d, what is seen is that the magnitude of the lofted CO is similar between WRF and TES, but the location of that high CO differs. Thus, the conclusion should be that WRF-Chem overpredicts the frequency of convective lofting of high CO values, but underpredicts the CO magnitude in the mid to upper troposphere in aircraft corridors. The text in the manuscript has been modified to reflect this conclusion.

8. Page 16431 line 16 Rather than say the WRF results possibly explain the Cooper et al 2007 results, I would say the WRF results are entirely consistent with the Cooper et al findings.

   The sentence has been changed to say our finding is consistent with Cooper et al, 2007.

References:

Allen, Dale; Pickering, Kenneth; Duncan, Bryan; et al., Impact of lightning NO emis- sions on North American photochemistry as determined using the Global Modeling Initiative (GMI) model, JOURNAL OF GEOPHYSICAL RESEARCH-ATMOSPHERES Volume: 115 Article Number: D22301 DOI: 10.1029/2010JD014062 Published: NOV 19 2010

Allen, D. J.; Pickering, K. E.; Pinder, R. W.; et al., Impact of lightning-NO on eastern United States photochemistry during the summer of 2006 as determined using the CMAQ model,

ATMOSPHERIC CHEMISTRY AND PHYSICS Volume: 12 Issue: 4 Pages: 1737-1758 DOI: 10.5194/acp-12-1737-2012 Published: 2012

Nowak, J. B., et al. (2004), Gas-phase chemical characteristics of Asian emission plumes observed during ITCT 2K2 over the eastern North Pacific Ocean, J. Geophys. Res., 109, D23S19, doi:10.1029/2003JD004488.

Zhao, C.; Wang, Y.; Choi, Y.; et al., Summertime impact of convective transport and lightning NOx production over North America: modeling dependence on meteorological simulations, ATMOSPHERIC CHEMISTRY AND PHYSICS Volume: 9 Issue: 13 Pages: 4315-4327 Published: 2009

Referee #3

General Comments

The authors used the WRF-Chem model with relatively fine grid spacing (4km) to investigate the ozone chemistry during the early stages of the North American monsoon. Since previous studies about convective transport impact on upper tropospheric ozone is mainly estimated based on relatively coarse grid spacing with convection parameterization, the key motivation of this study is to explore the possible substantial uncertainties in those previous studies with coarse spacing. Comparing with meteorological and chemical measurements, WRF-Chem performs reasonable, with biases in simulating convective transport. The ozone budget (chemical production and transport from boundary layer or stratosphere) is analyzed over different geographic regions. The results show that most of ozone chemical production occurs within 24 h of the air being lofted from the boundary layer and ozone mixing ratioes within anticyclone region and outside are similar. Authors also found that ozone in anticyclone is strongly influenced by stratospheric source.

I find this topic about upper troposphere chemistry during the thunderstorms is interesting and important. I would expect this paper's uniqueness in its higher spacing resolution and hence advantages with resolved convective transport and also the budget analysis of ozone. However, this experiment is not well designed. Although, the authors designed some diagnostic parameters to show impact from convection transport, boundary layer, and stratospheric ozone, it's not very clear. I do have some specific comments. I would not suggest this paper for publication in ACP unless the authors address these comments.

We appreciate the reviewer's comments about other valuable ways to design this experiment. Hopefully the reviewer can be convinced that using diagnostic parameters is equally (if not better) as valuable. Using diagnostics allows the analysis of the processes occurring in the simulation without altering the results of the simulation. Emmons et al., (2012, GMDD) point out the deficiencies of perturbing emissions in a region to determine source attribution for ozone and describe a tagged ozone mechanism for a global chemistry transport model. This tagging mechanism was not available in WRF-Chem at the time of our simulation. We think the tracers and diagnostics used in our study provide sufficient information to show the relative importance of in situ chemical production and the contribution of the boundary layer and stratosphere to the upper troposphere ozone. When processes are turned off and on, the resulting non-linear chemistry could significantly change. In our simulation specifically there is the concern that changes in the chemistry could lead to a different meteorological scenario because the simulation allows feedbacks between the chemistry and meteorology through radiation and cloud physics. With these additional changes, it would be difficult to attribute the ozone to the intended change or to the other changes in the simulation. A paragraph has been added to section 2.3 justifying the use of tracers and diagnostics for our study.

Specific Comments

1. The impacts of convective transport, stratosphere ozone, and lightning impact on upper tropospheric ozone are extensively investigated by previous studies, therefore, new insight of this paper is to conduct high spacing resolution simulation. However, authors fail to demonstrate how and to what extent the results from high spacing resolution that explicitly simulating convective transport differ from those from relative low resolution parameterizing convective transport and also fail to point out the uncertainties in the traditional estimate with parameterized convective transport. A sensitivity simulation with a relatively coarse resolution (e.g., 36 km) should be conducted to quantify the difference of impact of convective transport on upper tropospheric chemistry between explicit one and parameterized one.

Presenting a comprehensive comparison between a simulation with parameterized convection and a simulation with convective-permitting grid resolution is one of the goals of our project (but not this paper). We recently implemented a lightning-NOx parameterization for parameterized convection into WRF-Chem and the simulation for the CONUS region during July and August 2006 is just getting underway. So we do not yet have those results to present in this paper.

We do have a simulation conducted without the lightning-NOx parameterization at 36 km grid spacing. Figure 5 shows the July 17-22 results of how the UT mass of CO, O_3, BL tracer, and stratosphere tracer compare for the different grid resolutions. The CO mass has similar values predicted by the two simulations and have similar trends, except for the Gulf Coast region. The BL tracer is similar in magnitude but the 36 km variations do not match those from the 4 km simulation exactly. There are larger differences with the stratosphere tracer mass where the 4 km simulation predicts more stratospheric air entering the UT than the 36 km does, especially for the Midwest region behind the cold front on the 22 July. This comparison is cursory, but indicates that the convective parameterization does fairly well, yet does not exactly match the explicit convection simulation. A paragraph, similar to this one but without the figure, is included at the end of section 5.1.

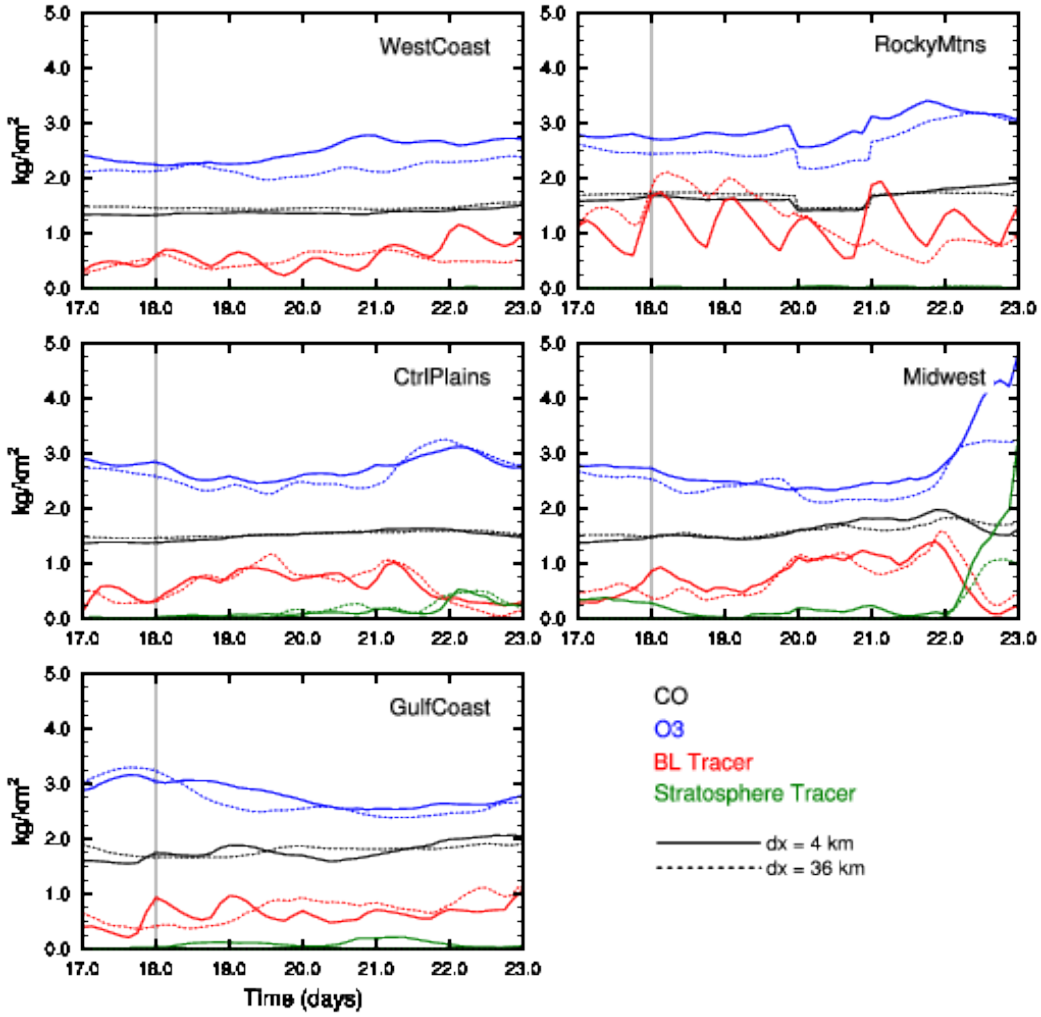

Figure 5. Mass per unit area of CO (black line), O_3 (blue line), decaying boundary layer tracer (red line), and decaying stratosphere tracer (green line) in the upper troposphere (8–12 km altitude range) over the (a) West Coast, (b) Rocky Mountains, (c) Central Plains, (d) Midwest, and (f) Gulf Coast regions. The solid line is from the 4 km grid spacing simulation, the dotted line is from the 36 km simulation. The decaying boundary layer tracer and stratosphere tracer masses per area are the masses of air in units of $2x10^6$ kg km^{-2} and $1x10^6$ kg km^{-2}, respectively. The vertical gray line delineates the 2 meteorological periods as described in the manuscript.

2. This paper tries to understand ozone chemistry during the thunderstorms of North American monsoon. Ozone in the free troposphere/upper troposphere is mainly contributed by chemical production, vertical transport from boundary layer and stratosphere, and lightning. These can be estimated by conducting multiple sensitivity simulations [e.g., Choi et al., 2008; Zhao et al., 2010]:

For the simulation described here, the contributions to the ozone chemistry must be done via diagnostic tools including tracers. The main reason for this is the connection between the chemistry and meteorology via aerosols affecting the radiation and clouds. When emissions, physics, or dynamics are altered for sensitivity tests, this simulation will produce a different meteorology. We acknowledge that the O_3 tracers described in Emmons et al. (2012) would be a better way to analyze the O_3 chemistry. However these tracers were not available for the RACM chemistry mechanism in the version of WRF that was used at the time of the simulation runs.

a. Contribution from transport of boundary layer pollutants can be estimated by subtracting the results of sensitivity simulation without anthropogenic emissions from the results of standard simulation.

The contribution from the boundary layer can be obtained from the BL tracer.

b. Contribution from stratospheric ozone can be estimated by subtracting the results of sensitivity simulation without stratospheric ozone in upper and lateral boundaries from the results of standard simulation.

The contribution from the stratosphere can be obtained from the stratosphere tracer.

c. Contribution from lightning can be estimated by subtracting the results of sensitivity simulation without lightning from the results of standard simulation.

The role of lightning is addressed in an upcoming paper where lightning-NOx emission rates are altered (but not removed).

d. Contribution from convective transport can be estimated by subtracting the results of sensitivity simulation without convective transport from the results of standard simulation. This sensitivity experiment may be difficult for this study since the convective transport is explicitly simulated instead of parameterized. Authors may want to define the convective transport occurs when cloud height or cloud thickness exceed a criteria so that one can artificially turn off convective transport.

Not only would this sensitivity simulation be difficult to do, it would be erroneous. A better comparison would be to compare the chemistry within the convective outflow to the upper troposphere background air surrounding the outflow.

Authors must design these multiple sensitivity experiments to clearly quantify the ozone budget.

The goal of our paper is to identify geographic regions of importance for convective transport and subsequent O_3 production in the upper troposphere and to investigate the role of the anticyclone associated with the NAM on CO and O_3. The goal is not to quantify the ozone budget.

Technical Comments

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1. Line 5 of page 16412, please include the latitude and longitude range of the two outer domains.

The southwest-northeast latitude, longitude values are for the outermost domain (1.7N-55.6N, 129.2W-28.4W) and for the middle domain (13.0N-52.7N, 123W-53W). This information is included in the text.

2. Line 22 of page 16412 and line 2 of page 16414 are not consistent. If aerosol affects cloud microphysics, the Lin scheme should be two-moment.

Chapman et al. (2009, ACP), which is cited in our paper, describe how the aerosol-cloud effects are included with the Lin scheme. From their paper, the following description is given.

"A prognostic treatment of cloud droplet number (Ghan et al., 1997) was added to the Lin microphysics scheme (Lin et al., 1983; Rutledge and Hobbs, 1984; Tao et al., 1989; Chen and Sun, 2002), which treats water vapor and the following classes of hydrometeors: cloud water, rain, cloud ice, snow, and graupel. The parameterizations of Liu et al. (2005) were added to make the autoconversion of cloud droplets to rain droplets dependent on droplet number. Droplet-number nu- cleation and (complete) evaporation rates correspond to the aerosol activation and resuspension rates. A two-moment treatment of cloud water (cloud water mass and cloud droplet number) thus is now available within WRF-Chem."

3. Line 17 of page 16414, provide the reference for your dust emissions.

The reference is Shaw et al. (2008) and is now included in the paper.

Shaw, W., Allwine, K. J., Fritz, B. G., Rutz, F. C., Rishel, J. P., and Chapman, E. G.: An evaluation of the wind erosion module in DUSTRAN, Atmos. Environ., 42, 1907–1921, 2008.

4. Line 20 of page 16414, please provide the reference for lightning NOx production used in the study. The parameterization of lightning NOx production is normally annually dependent (should be scaled by NLDN or other observations for lightning flashes), so please provide the evaluation for the specific period of this study.

The description for the lightning NOx production is given in this paper with specific citations to the previous studies used. There was no scaling to NLDN data (or other observations) done. The placement of the lightning should be fairly accurate since the model predicts convective systems well. The magnitude of the lightning flash rates is low compared to NLDN. A sentence stating that no tuning was performed has been included in the text.

5. Line 24 of page 16424, the high bias of ozone above 12 km may be from stratospheric ozone (i.e., upper boundary)? Did you evaluate the upper boundary ozone?

Stratospheric O_3 is obtained primarily from the CAM-Chem model via the initial and boundary conditions, but an upper boundary condition is included for a segment of the

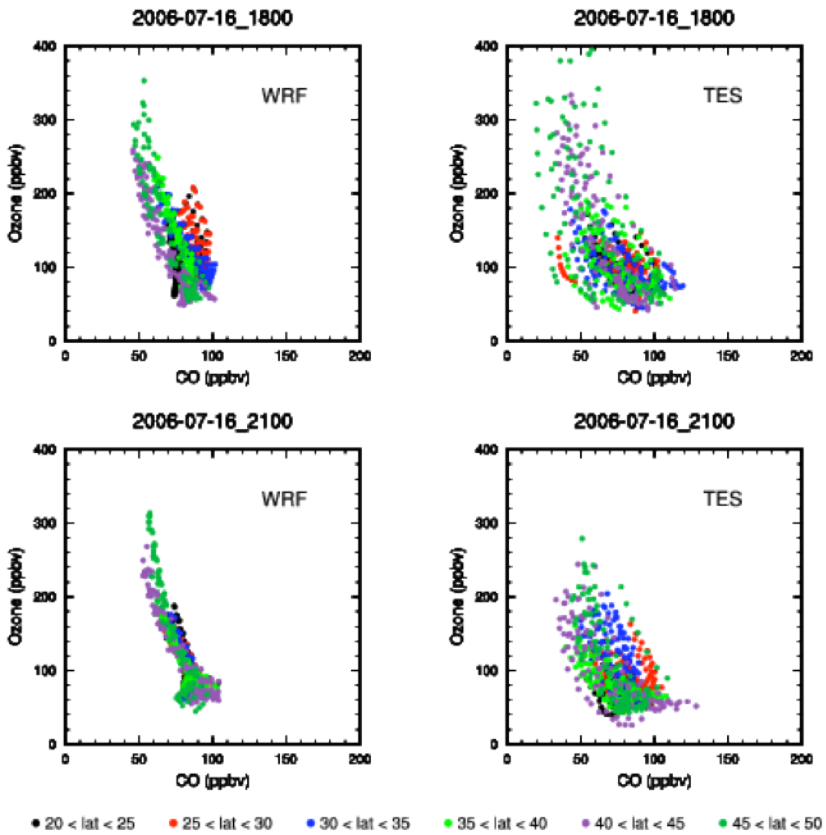
simulation. The CO and O_3 values in the troposphere are not affected by the upper boundary condition (compared to tests without the upper boundary condition) for the 13-day segment of the simulation. Between 15-20 km, the WRF-Chem prediction of O_3 remains high (within 20%) compared to ozonesonde observations over the Huntsville site, but is similar in magnitude over the Boulder and Trinidad Head sites. The evaluation for the 15-20 km region has been added to the manuscript.

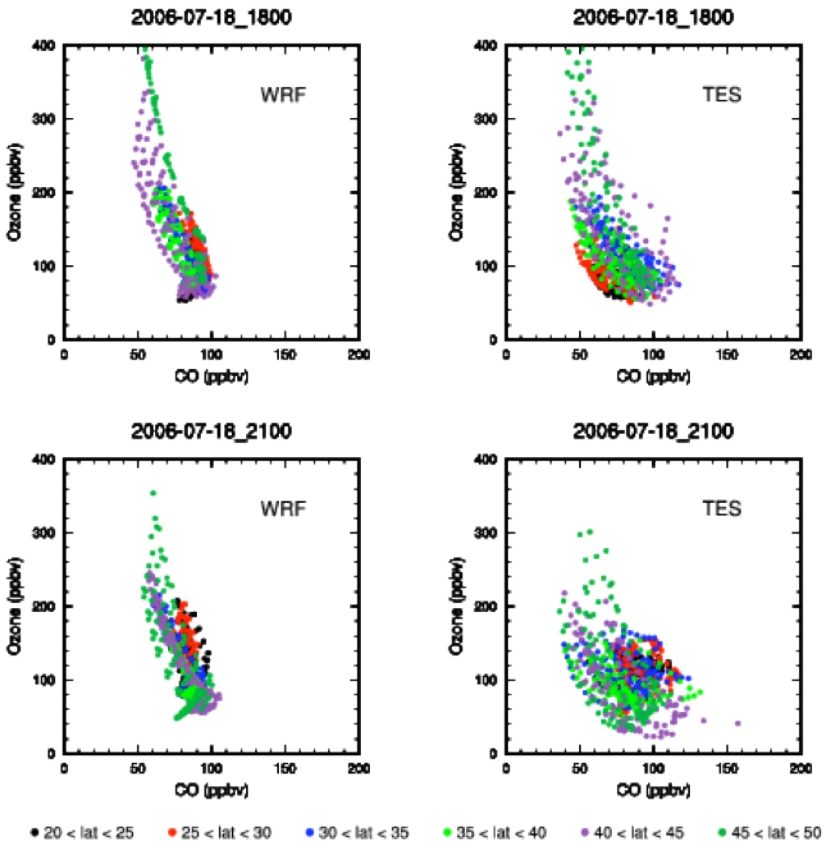
Reference

Choi, Y., Y. Wang, Q. Yang, D. Cunnold, T. Zeng, C. Shim, M. Luo, A. Eldering, E. Bucsela, and J. Gleanson (2008), Spring to summer northward migration of high O3 over the western North Atlantic, Geophys. Res. Lett., 35, L04818, doi:10.1029/2007GL032276.

Zhao, C., Y. Wang, Q. Yang, R. Fu, D. Cunnold, and Y. Choi (2010), Impact of East Asian summer monsoon on air quality over China: The view from space, J. Geophys. Res., 115, D09301, doi:10.1029/2009JD012745.

Appendix: TES observations and WRF-Chem model results showing the correlation between CO and O3 in the upper troposphere (400 > p > 150 hPa) along the TES overpass tracks.

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WRF
2006-07-18_1800
TES
2006-07-18_2100
WRF
2006-07-18_2100
TES
Ozone (ppbv)
CO (ppbv)
0
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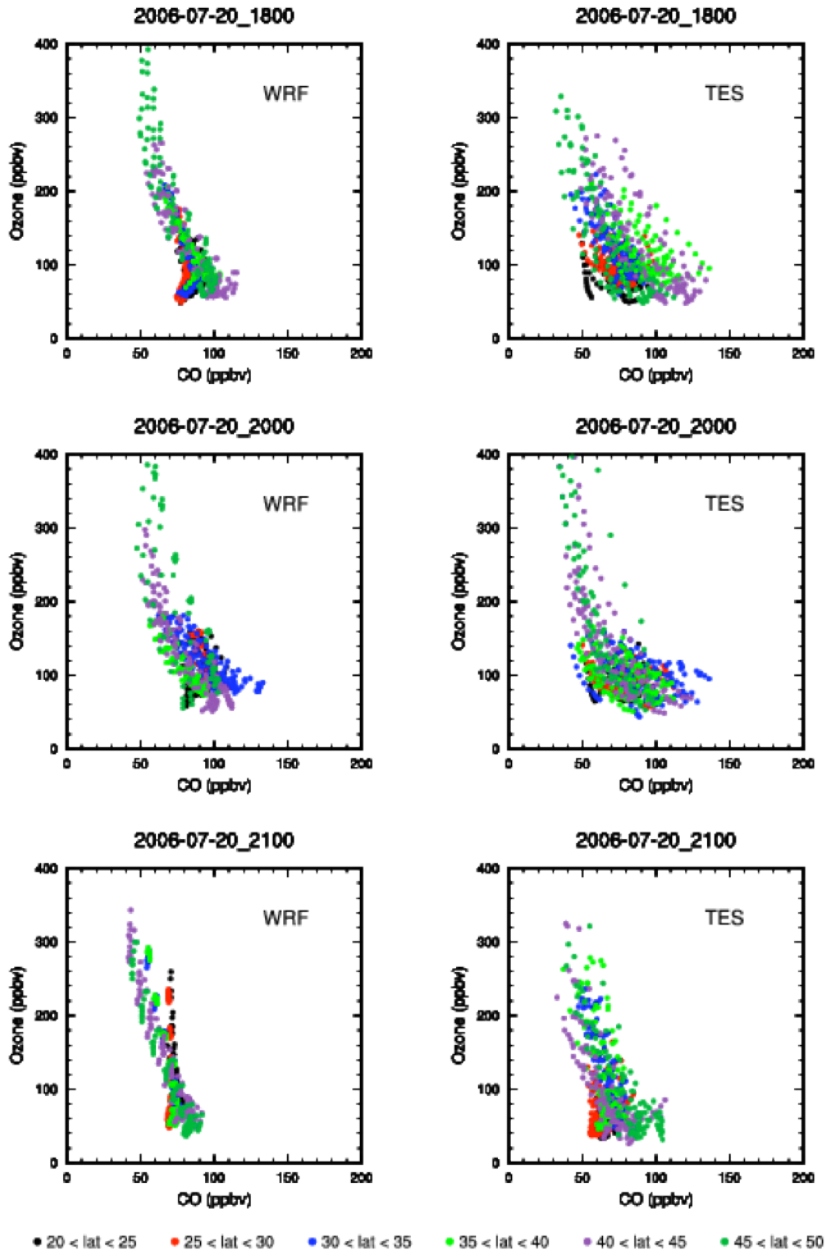
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Ozone (ppbv)
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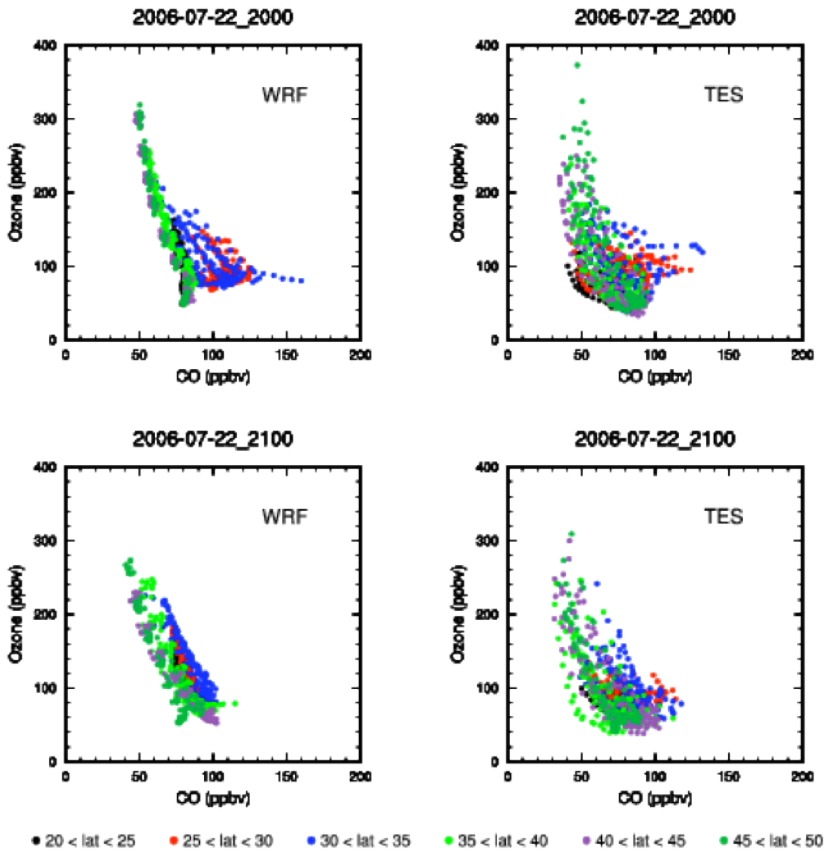
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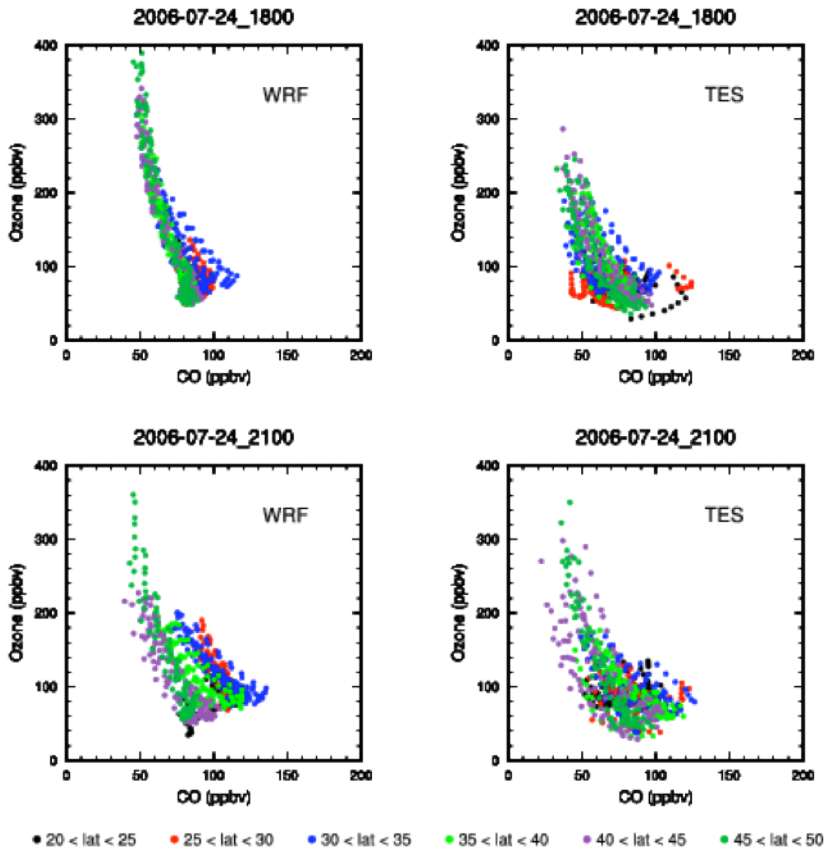
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2006-07-24_1800
TES
2006-07-24_2100
WRF
2006-07-24_2100
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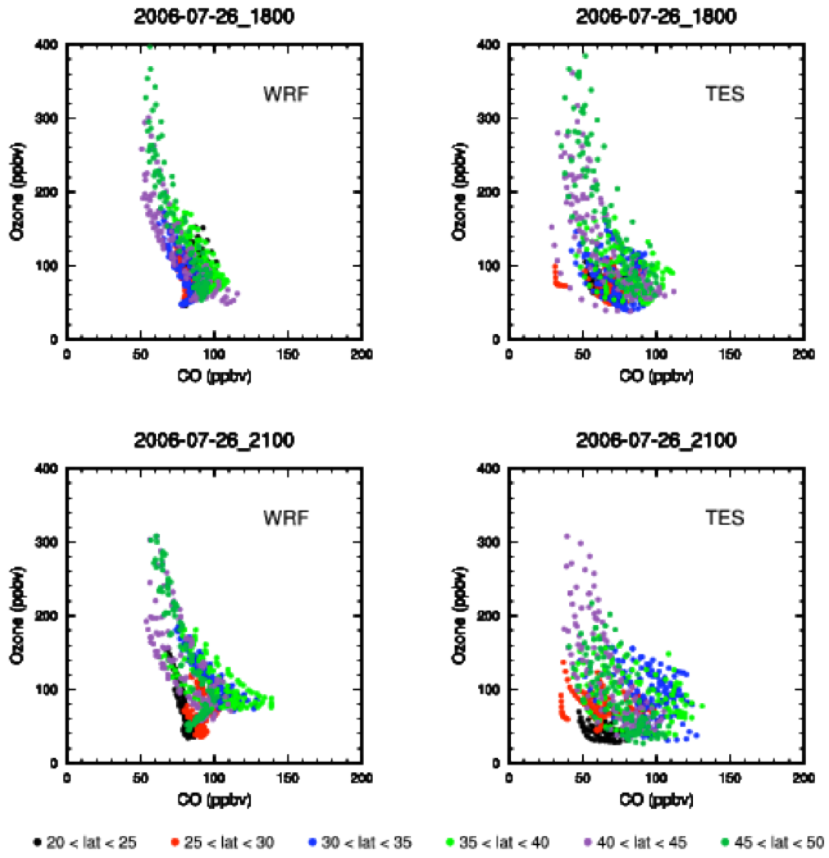
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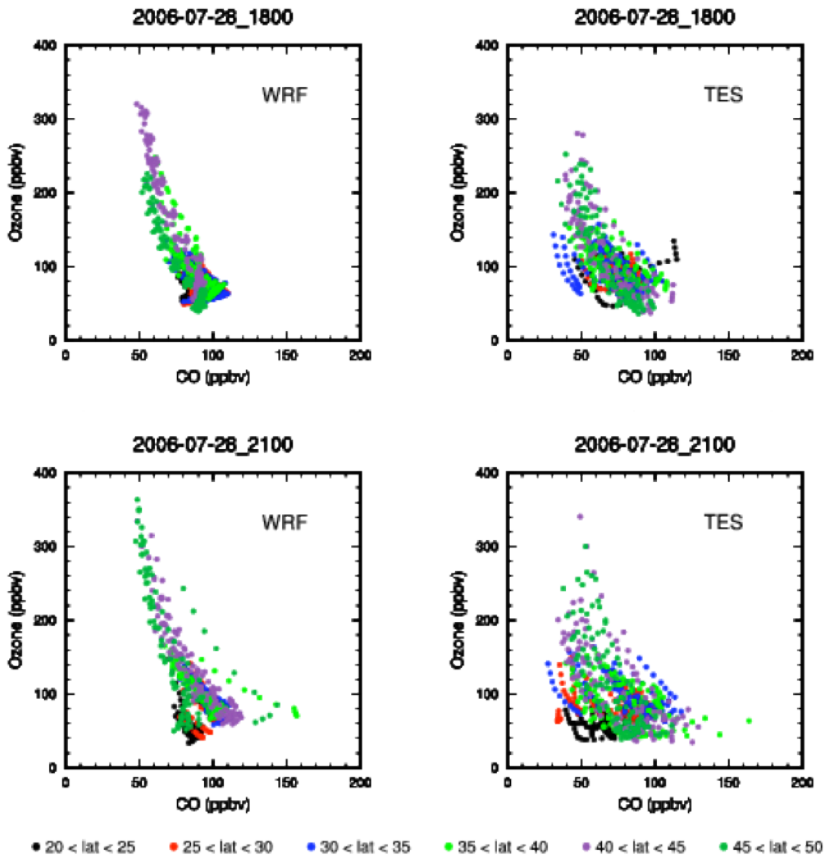
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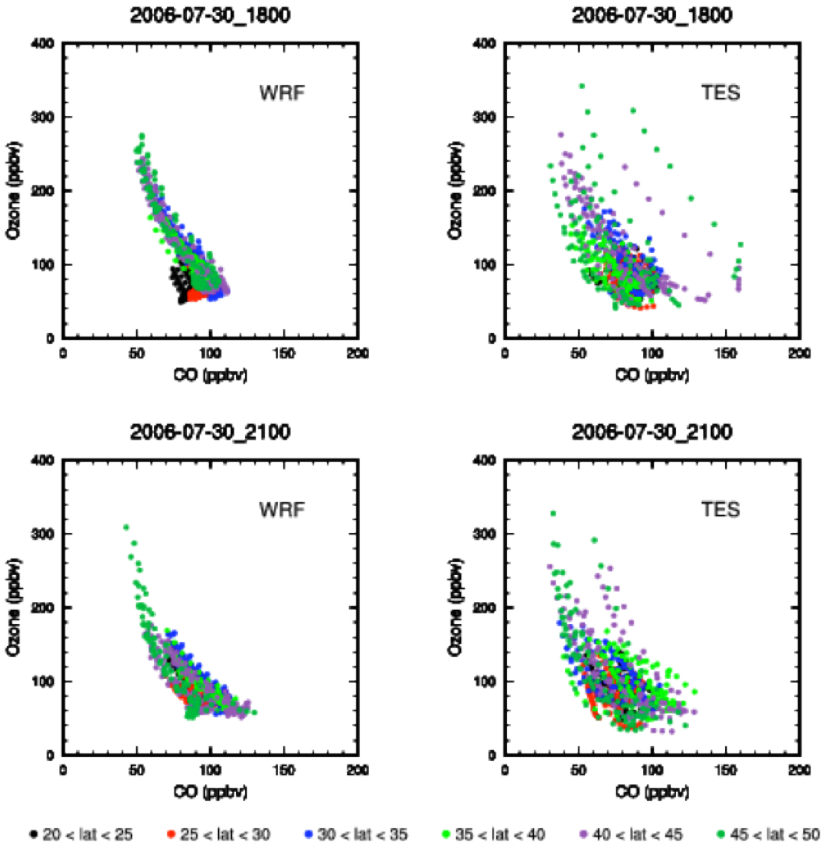
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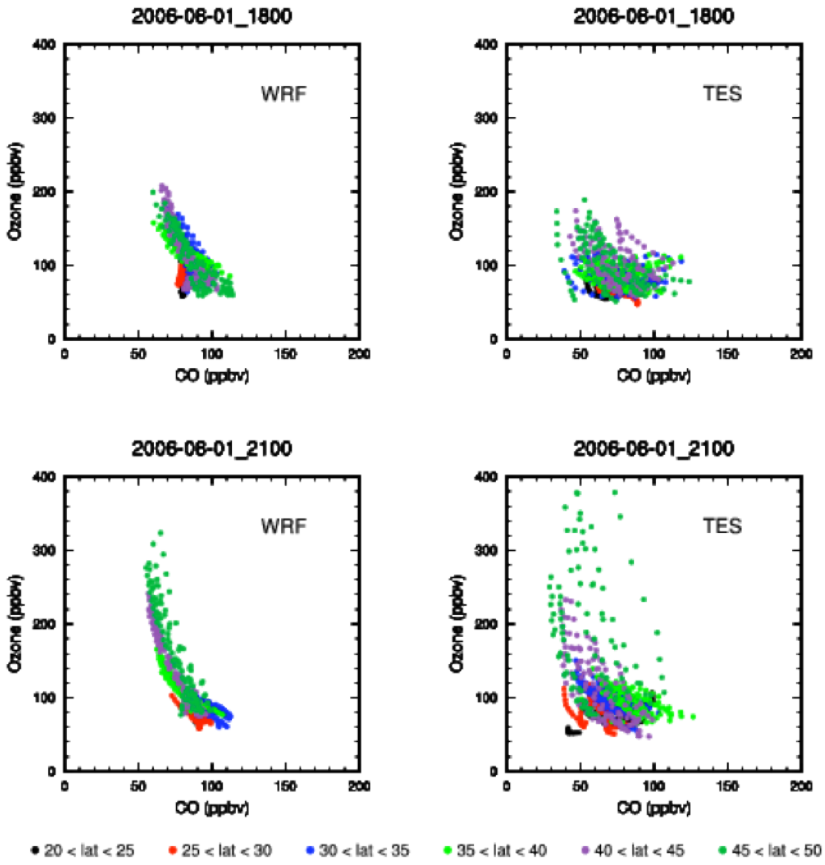
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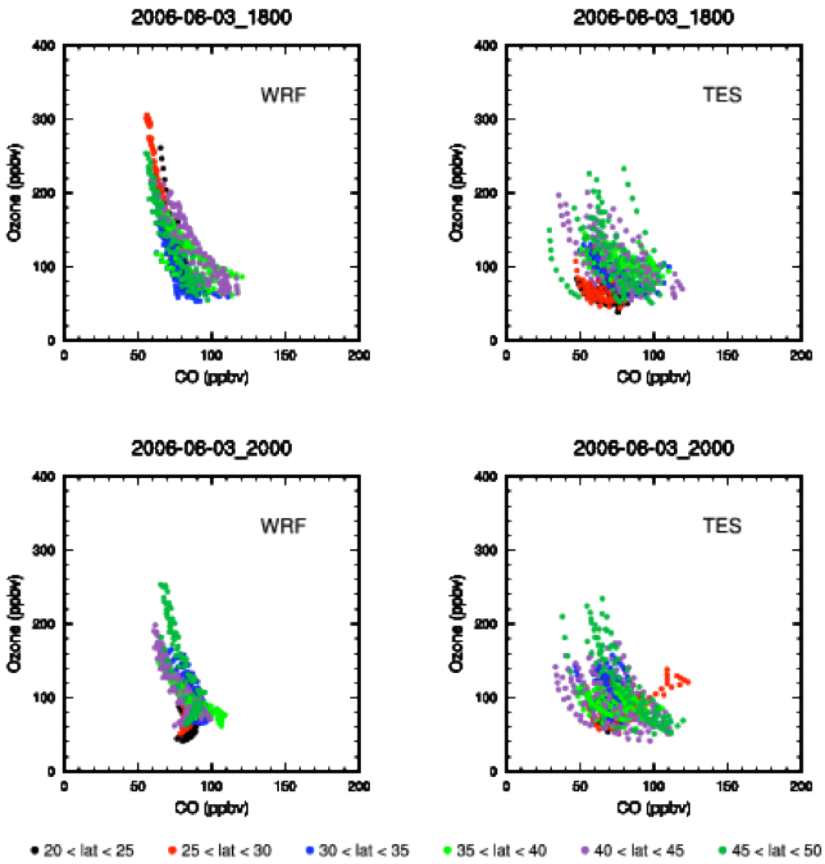
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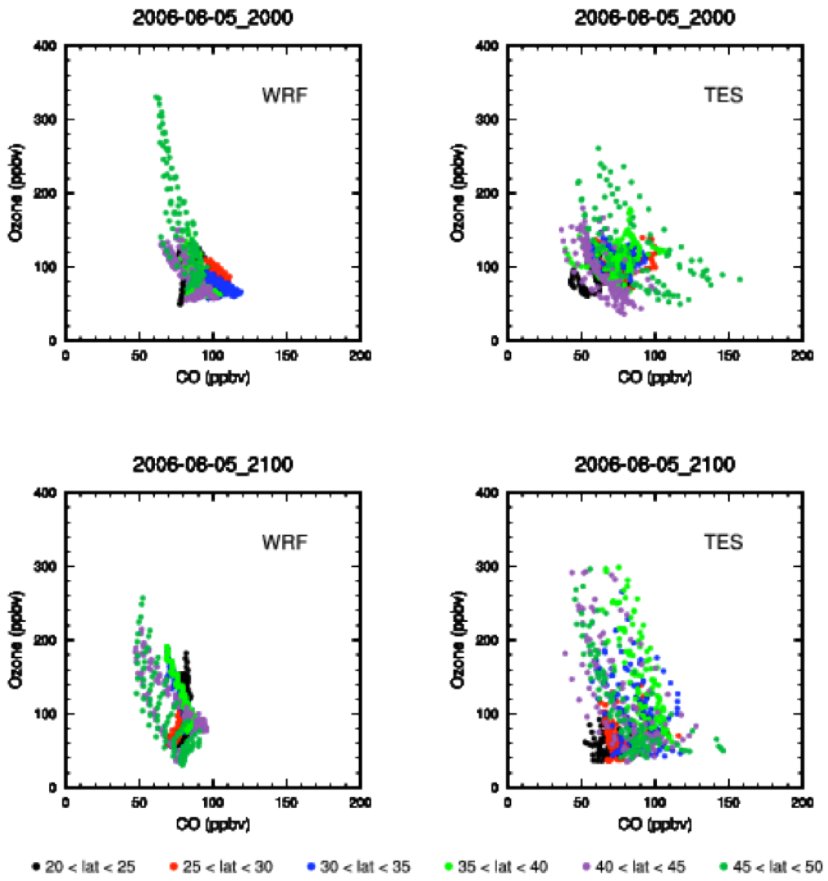
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