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**Geoscientific
Model Development
Discussions**

Interactive comment on "Incremental testing of the community multiscale air quality (CMAQ) modeling system version 4.7" *by* K. M. Foley et al.

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We thank the referee for their detailed reading and for the overall positive assessment of the manuscript. We have addressed the questions and suggestions that were provided and we feel that these changes have improved the paper. Below we have listed our response to each of the specific points that were raised, using the page, line and Figure numbers from the paper published in GMDD. Reviewer comments are shown in italics; the author reponses are in regular text.

Major concerns

1. *Status of Carlton et al. and Kelly et al. papers is critical because they describe Increments B and C in more detail.*

The Kelly et al. manuscript has now been published in GMDD and is available online. This reference has been updated in the paper, including Table 1. Additional explanation of the modifications to the SOA module (Increment B) is currently available in the model release notes (available at http://www.cmaq-model.org) and in the documentation of the code itself. The Carlton et al. paper has cleared our internal organizational review and will soon be submitted to Env. Sci. & Technol.

Specific comments

1. *Abstract: Page 1246, line 2: "This paper describes the scientific and structural updates to the latest release of the Community Multiscale Air Quality (CMAQ) modeling system version 4.7 (v4.7)", but it seems that none of the photolysis options reported on pages 1257 and 1258 are available in this release. Not sure if they should be included in this paper.*
All of the photolysis options are included in the model release. User's can select these options when they build the model.

2. *Meteorological input model: Page 1249, lines 19-26: I am assuming nudging and ACM2 were not available in WRF versions prior version 3.0. However, it seems they were used in other models. The authors should name them.*
Nudging, the ACM2 and the PX LSM have been available in MM5 for many years. This information has now been added to the text.

3. *Page 1250, lines 18-21: Why the need to use WRF instead of MM5, when both models are comparable?*
NCAR is no longer releasing new versions of MM5 since WRF is essentially a replacement for MM5. WRF has the same capabilities as MM5 but also includes improvements in the underlying dynamics of the model and updated physics options such as new versions of the land surface model and planetary boundary layer schemes. WRF has already been adopted by many other research groups as the state of the science meteorological model (e.g. it is the current operational model for the National Centers for

Environmental Protection). Because the meteorological inputs are such an important part of the CMAQ modeling system, it was necessary that CMAQ be able to support WRF inputs for future modeling studies.

4. *Page 1250, line 28 – page 1251, line 4: This sentence is long and not easy to read. It should be split into smaller pieces and provide more explanation.*
Following the reviewer's suggestions, in the revised manuscript this discussion now reads: "During winter months, model predictions of particulate nitrate are sensitive to the nighttime hydrolysis of N_2O_5 on particle surfaces (Dentener and Crutzen, 1993). The probability of this heterogeneous reaction (γ N_2O_5) in CMAQv4.6 was parameterized in part using a temperature- and RH-dependent equation published by Evans and Jacob (2005). Lab data indicate that γ N_2O_5 decreases with increased temperature; however this relationship was reversed in the published formula due to a typographical error (Mathew Evans, personal communication)."

5. *Page 1252, line 26: "the CMAQ input file OCEAN_1 has been enhanced" – is this file a part of CMAQ 4.7 release? In other words, are these enhancements available to a reader?*
The OCEAN_1 file is domain specific and as such needs to be created for each new domain application. An example of the OCEAN_1 file is included with the CMAQ release, but this is specific for the limited area tutorial case. As stated in the manuscript discussions, the information in this file was enhanced to better capture the relative fractions of the coastal surf zone, which is needed in the estimation of sea-salt emissions. Tools such as the Spatial Allocator can be used to create the OCEAN_1 file for specific domains. Documentation and code are available from the CMAS website: http://www.cmascenter.org/). Specifically, Section 7.1.6 on http://www.ie.unc.edu/cempd/projects/mims/spatial/alloc.html provides information on the use of the Spatial Allocator tool to compute the grid cell fraction covered by land, open ocean and coastal surf zone.

6. *Page 1253, line 9: "accumulating precipitation" – what is the meaning of that?*

We used the term "accumulating precipitation" to denote the precipitation that impacted the ground. In the meteorological model, the cloud microphysics simulates cloud water and ice, as well as rain, snow, and graupel for all model layers, and the vertical transport of these hydrometeors. The precipitating hydrometeors (rain, snow, and graupel) can undergo evaporation as they descend towards the ground. To avoid confusion, in the revised manuscript, we have removed the word "accumulating" in the associated discussion.

7. *Improvements in atmospheric chemistry: Page 1256, lines 4-6: "the relative impacts of this change were small" – impacts on what? Also, Sarwar et al. (2008) showed that a new HONO treatment improved significantly the model predictions of its mixing ratio. The reason for not performing this increment in this paper should rather be that it was already done in above mentioned paper.*
This sentence has been changed to clarify what changes are being referenced: "Because HONO observations for the time period of this study were not readily available, and the relative impacts of this change on ozone and PM predictions were small, no increment is presented in the paper." This is the reason that we did not present the results of the HONO increment, rather than the fact that Sarwar et al. (2008) also provides an evaluation of these model changes. The impact of many of the other model changes in version 4.7 have also been studied in separate analyses (e.g. coarse mode chemistry is analyzed in Kelly et al., 2009). The value of the incremental testing was to show (when possible) the impact of each model change using a single model simulation for a set time period and domain.

8. *Page 1256, lines 10-12: Has a current version of SMOKE capability of deriving Cl emissions?*
The USEPA maintains a National Emissions Inventory (NEI) for hazardous air pollutant. Since molecular chlorine is a hazardous air pollutant, its anthropogenic emissions are included in the NEI. These emissions as well as emissions for HCl were processed by the SMOKE to generate model-ready emissions and were used in the study. While

sea-salt derived chlorine can affect O_3, these emissions were not included in the study; we intend to incorporate such emissions in the future.

9. *Page 1256, lines 13-18: The authors should describe more thoroughly the motivation for Hg modeling and the improvement of Hg modeling in CMAQ 4.7 compared to previous versions.*
The stand alone mercury model was replaced by the multipollutant version to keep with the original "one atmosphere" approach for air quality modeling in CMAQ. This was done to address an increased interest in modeling multipollutants, including criteria and hazardous air pollutants, in a single modeling framework for air quality management (Scheffe et al., 2007). A manuscript detailing the motivation and evaluation of Hg modeling in CMAQ v4.7 is in preparation.

10. *Research options: Page 1257, lines 10-12: How are the "beta" versions provided to the community? It seems that they are not available with CMAQ 4.7.*
The research options are included in the CVS archive that is downloaded from the CMAS website. The photolysis satellite option and the inline photolysis option can be selected by the user when they build the model. Thus, the inclusion of beta versions of developmental modules enables wider testing across the user community and help in developing more robust algorithms that can be included in future model versions.

11. *Page 1257, line 15 – page 1258, line 2: It seems that the option that utilizes satellite derived cloud information to adjust photolysis rates is not available yet (software problems are currently being addressed). If so, what is the point of reporting unfinished work here.*
The software issues were in the preprocessing software and not in the CMAQ code itself. Therefore, we think that it is important to highlight this capability in CMAQ, since we anticipate that the preprocessing software revisions will be complete in the near future.

12. *Page 1258, lines 26-29: "Bidirectional surface exchange option . . ." It seems that*

this option cannot be used, since even authors could not perform tests. If so, no need to report it in this paper.
It was unfeasible to perform the test at the time of the incremental testing for the test period examined; it should be noted that the option is functional and has been tested for other time-periods. For instance, a revised Hg emissions inventory was developed without a priori estimates of emissions from natural sources and the Hg bidirectional exchange option was described and evaluated in Bash et al. 2010. A revised emissions inventory for NH_3 is in development to evaluate the bidirectional NH_3 air-surface exchange model. The purpose of this sentence was to acknowledge that model options were released as science/beta options, provide references for these options and explain why the analysis of these modules were not included in the incremental testing.

13. *Modelling approach and observational data sets: Page 1259, lines 17-19: Were initial conditions really derived from 36 km simulations? If so, how? A previous statement (line 14) suggest that "a 3-day model spin-up was used".*
The boundary and initial conditions are from a 36km continental U.S. CMAQ application. The 36km simulations used a 10-day spin-up period. We did use a 3-day spin-up for the 12km simulation, initializing the model with the 36km results taken from spin-up period.

14. *Page 1260, lines 1-3: It seems that to derive sesquiterpene fluxes the MEGAN model needs to be used. Previous information on page 1252, lines 1-3, suggests that BEIS itself would derive those emissions.*
Emissions of biogenic species were determined from the BEIS which was updated to incorporate algorithms for estimation of sesquiterpene emissions based on four emission factors in the MEGAN model. Sesquiterpenes were added using available measurements for α-humulene and β–caryophyllene and incorporated as a single species group. Normalized emission factors for sesquiterpene emissions were derived for the four functional plant types in MEGAN based on the work of Sakulyanontvittaya et al. (2008). The broad-leaf, needle-leaf, shrubs, and grassland applied VOC emissions fac-

tors were 0.175,0.1084,0.0552,0.2036 $\mu g g_{dry}^{-1} hr^{-1}$, respectively. Each of the 230 tree species in BEIS was mapped to the four functional plant types to derive an emission factor for each tree species. The only exception was that for Loblolly pine, an emission factor of 0.3 $\mu g g_{dry}^{-1} hr^{-1}$ was used. (Geron, 2009).

15. *Evaluation of major scientific increments: New parameterization for heterogeneous reaction probability: Page 1261, lines 12-16: This part should be moved to section 2.2 and repeated information should be deleted. The paper is already long.*
We thank the reviewer for the suggestion and have modified the text accordingly.

16. *Page 1261, line 23 – page 1262, line 6 and corresponding Figure 1: This discussion needs some more explanation. I understand that column 2 in Figure 1 refers to the corrected typographical error in the CMAQ aerosol model. It is not clear to me what the third and fourth column exactly displays, since my understanding is that Increment A also reflects the impact of corrected typographical error in the CMAQ aerosol model.*
Several sentences have been added to clarify this section. Increment A uses a completely different parameterization of gamma N_2O_5 compared to what was used in CMAQv4.6. Just correcting the typo in the original parameterization led to a large increase in the CMAQ predictions of winter time nitrate. This correction degraded model performance which was the motivation for developing a new parameterization. The third column shows that the new parameterization decreases the unrealistic gamma N_2O_5 values and subsequently the TNO_3 concentrations decrease across the domain. We added this third column because it is more informative than comparing the new model version to the incorrect base model. Simply comparing Increment A to the base model would show an increase in gamma N_2O_5 and an increase in TNO_3. However this is misleading because the base model is getting the "right answer" (in terms of lower model bias compared to observed values) for the wrong reason.

17. *It would be helpful to spell out the CMAQ versions in the headers of the plots.*
We have renamed the increments in the figure headers and throughout the paper:
Old label → New label

Increment A → γN2O5 Increment
Increment B → SOA Increment
Increment C → Coarse Mode Increment
Increment D → Cloud Increment
Increment E → Photolysis Increment

18. *Page 1262, line 13: What is the over prediction in the summer time in quantitative terms?*
The median bias is 0.7 μg/m^3 and the normalized median bias is 43% for the three increments. The following has been added to the text: "TNO_3 concentrations in the summer time are over predicted in all three of the simulations (the normalized median bias for the three increments is 43%), suggesting this bias is not sensitive to the change in the γ parameterization during warmer months."

19. *Page 1263, lines 1-4: It seems in the previous version biogenic SOA were also higher in summer than in winter. So in this respect there is no change using the updated version.*
In the previously released version, biogenic SOA was higher in August in most, but not all of the domain. For example, Figure 3 shows regions in FL and TX where the average biogenic SOA is actually higher in January. This is more clearly seen the difference plot in Figure 1 included here. In contrast, biogenic SOA is now higher in August, compared to January in all grid cells in the updated version. We have updated the text to emphasize that we are referring to an increase across the domain.

20. *SOA model enhancements: Page 1262, line 21 and corresponding Figures 2 and 3: It would be more helpful to spell what you exactly mean with "previous".*
In this case, the "previous increment" is very similar to Increment A. This increment used the gamma N2O5 parameterization given in the body of the Davis et al. paper. In contrast, Increment A (and the released model version, CMAQv4.7) includes the equations in Appendix A of Davis et al. This change was made late in the incremental testing

process in response to reviewer comments for the Davis et al. paper. We felt it would be unnecessarily confusing to explain this small difference in this paper since it has no real impact on the final conclusions of the evaluation of the SOA increment. However, in case other readers would like more information on the "previous increments", we now point them to this the open discussion.

21. *Page 1262, lines 24-25 and corresponding Figure 2: The decrease in anthropogenic SOA in August is not as large as in January. The authors should differentiate their statement.*
This sentence points the reader to Figure 2 which clearly shows the decrease in anthropogenic SOA in August is not as large as in January. We have chosen to leave the text unchanged because this section is lengthy and already highlights other differences between the January and August plots.

22. *Page 1263, line 1: What is the difference between the lower-middle and the lower-center plots in Fig 3?*
This should have said "the lower-center and lower-right plots in Figs. 2 and 3." The correction has been made in the text.

23. *Page 1263, lines 15-16: Were the experimental results in Duke Forest obtained during the entire month of August. Please clarify?*
Yes, the results are from the entire month. The text has been changed to read: "The latter is consistent with the amplitude of 2.6 μgC/m^3 observed throughout August 2006 at the same location."

24. *Page 1263, lines 18-19: Not sure, if you can use experimental data obtained on 17 days of August 2003 to validate the modeling results for the entire month of August in 2006.*
We agree that our conclusions are limited by the availability of semi-continuous TC measurements. We have made an effort to use whatever observations are available at this time to provide an assessment of the changes in model performance that resulted

from our updates to the SOA treatment in CMAQv4.7. As additional datasets become available, we will be able to provide a more in-depth evaluation of the SOA module.

25. *Page 1263, lines 20-21: It sounds like there was a comprehensive update on the aromatic SOA formation treatment, potentially including aromatics like toluene and xylenes. I am not sure, if I have missed something, but section 2.2 the authors are basically referring to benzene. Please clarify.*
Section 2.2 has now been updated to clarify what updates have been made. page 1251, line 25: "The revised model treats the acid-catalyzed enhancement of SOA mass, oligomerization reactions, aqueous-phase SOA formation, and NO_x-dependent SOA yields from aromatic compounds."

26. *Page 1263, lines 20-29: I guess the most important new anthropogenic SOA precursor which was implemented was benzene. The differences in anthropogenic SOA in summer in the new version is really tremendous. Is there a good explanation why anthropogenic SOA is also strongly enhanced over rural areas. Also, it looks like urban areas like Houston do not show though I would expect some benzene related SOA signal. Also the reference of Kleindienst et al is not really helpful, since it refers to "some polluted days" in 2003. So I would expect some bias to higher values. Anyway the Kleindienst et al values of 0.8 mikrogramC/m**3 are actually closer to the previous values, whereas in the new version values up to more than 20 mikrogramC/m**3 are found.*
Most of the differences in anthropogenic SOA arise from the use of NO_x-dependent SOA yields for all aromatic compounds, not from the addition of benzene as an SOA precursor. In low-NO_x environments, aromatic SOA yields are enhanced and the products exhibit very low volatility (Ng et al., 2007). For this reason, anthropogenic SOA is enhanced outside the urban areas. To clarify this point, we have added the words "for all aromatic compounds" to the sentence ending on line 25 of page 1251. Contrary to the reviewer's interpretation, the new model version never produces anthropogenic SOA values in the vicinity of 20 μgC/m^3. The modeled values of 0.2 μg/m^3 (shown in

lower-right plot of Fig. 2) are much closer to the Kleindienst et al. values (0.8 μgC/m^3) than were results from the previous increment (0.05 μg/m^3, shown in lower-left plot of Fig. 2).

27. *Coarse-particle chemistry: Page 1264, line 22: I guess it should be "bottom row of Fig. 6". Page 1264, lines 26-27: I guess it should be "top row of Fig. 6".*
These corrections have been made in the text.

28. *In-line photolysis research option: Page 1266, lines 2-12: This part should be moved to section 2.6 and repeated information should be deleted.*
We agree and have merged this text into section 2.6.

29. *Page 1266, line 19: Higher values of what?*
The sentence is describing differences in photolysis rates. We have modified the sentence to read as follows: "The table interpolation method calculates higher photolysis rates in high elevation areas because vertical interpolation of these rates . . ."

30. *Page 1266, lines 24 – 25: The authors want to describe what the different O3 column values in these models were. Also, the authors want to explain why $O_3 \rightarrow O(^1D)$ photolysis rates at the surface should decrease due to different (supposedly lower) O3 column values. Assuming lower stratospheric O3 values (which make up most part of the O_3 column) would lead to more UV radiation at the surface. Wouldn't this lead to higher $O_3 \rightarrow O(^1D)$ photolysis rates at the surface?*
We have included spatial plots of the O_3 column that compare the values used by the inline module and the table interpolation method (JPROC) (Figure 2 included here). Figure 2c shows that the O_3 column is higher for the inline module (by as much as 14% in the northern part of the modeling domain). This is consistent with our speculation that the lower $O_3 \rightarrow O(^1D)$ photolysis rates in the inline module are the result of higher total O_3 column values used in the inline module. We have also included a plot of the TOMS/OMI O_3 column values to show the data we will be incorporating into the next

release of the model.

31. *Page 1266, line 26 – page 1267, line 1: I agree that there was a stratospheric ozone depletion over the last 30 years, but I am not sure, if this may explain the fact that changes of O3 photolysis depend on the elevation.*
We were not trying to relate the stratospheric O_3 depletion with elevation in the O_3 photolysis discussion. This is not the point we were making. The stratospheric O_3 discussion was used to explain why the $O_3 \rightarrow O(^1D)$ photolysis comparison was different than the NO_2 photolysis comparison. We did look at the layer 34 (top of the model) photolysis rates and noted that the $O_3 \rightarrow O(^1D)$ photolysis rates were lower (15%-20%) with the inline model. This strongly suggests that the differences are related to the O_3 column above. However, the NO_2 photolysis rates were very similar in layer 34 between the two photolysis models. This paragraph has been reworded to clarify this issue in the paper.

32. *Ozone: Page 1269, lines 14-16: According to Appel et al. (2009) cited here the change from MM5 to WRF has a significant impact on ozone. In contrast, on page 1250, lines 16-23, the authors say that WRF and MM5 meteorological fields are comparable and there is a comparable CMAQ performance using either MM5 or WRF meteorology. Is it a different study than Appel et al. (2009)?*
There were differences in the MM5 and WRF model simulations since these are fundamentally different models. The difference in model performance between these two models, based on a comparison to observed data, is different for different time periods, meteorological variables and spatial locations. Similarly, the impact on CMAQ model performance is dependent on the pollutant, time period and location. The statement in section 2.1 was intended to simply summarize the conclusion from Appel et al. (2009) and Gilliam and Pleim (2009) that on the whole, WRF model performance is comparable to MM5-based simulations, i.e. the model performance is not dramatically worse or better. However, we agree that the way this paragraph was originally worded could be misinterpreted. We have expanded this paragraph to

explicitly state that there are differences in the model performance so that this section is consistent with the text in section 5.2.

The paragraph now states: “In two companion studies, multiple MM5 and WRF model simulations were performed for winter and summer months to examine the sensitivity of CMAQ model predictions to the output from each meteorological model. Gilliam and Pleim (2009) compared MM5 and WRF predictions for 2-m temperature, 2-m mixing ratio, 10-m winds and PBL features to observed meteorological data. Appel et al. (2009) evaluated CMAQ output for ozone, PM2.5 species, and wet deposition using MM5-based and WRF-model-based meteorology inputs. While MM5 and WRF error statistics based on observations across on the entire model domain are similar, some regional differences do exist, causing regional differences in the air quality model as noted by Appel et al. (2009). Although MM5 and WRF do not produce the exact same model results, these studies demonstrated that the WRF-model-based simulations generated comparable quality meteorological fields and air quality fields to the MM5-based simulations. The model evaluation provided confidence in the use of WRF model outputs for CMAQ simulations. In addition, the comparable performance of CMAQ when using either MM5-based or WRF-model-based meteorology fields demonstrated the versatility of the CMAQ system.”

33. *Page 1269, lines 25-26: Cannot understand the following sentence: “For higher-level observed O_3 mixing ratios, O_3 predictions tended to decrease.”*
This sentence has been reworded: “Relative to the base model, slightly greater underpredictions at the high mixing ratios (> 70ppb) are noted in CMAQv4.7.”

34. *Evaluation of CMAQ 4.7: - Wet deposition: Page 1270, lines 21- page 1271, line 3 and associated figure 12: It is difficult to decipher any consistent geographic distribution of the parameters. So I am not sure about the value of this figure.*
Boxplots such as Figure 10 were considered for this section but we decided the spatial plots would provide more information in about the same amount of space. In January

wet deposition of sulfate and nitrate are consistently over estimated in the North East and Great Lakes regions. In contrast, the bias in the deposition amounts of these species are very different in the summer time. Although the precipitation tends to be underestimated in these regions in August, sulfate wet deposition is still over estimated at many sites. The cause of the differences in model bias in wet deposition across different regions and in different seasons is currently being investigated in a separate study.

35. *Discussion: Page 1272, line 8: please clarify: is it Mathur et al., 2008a or 2008b?*
This section refers to Mathur et al., 2008b. The correction has been made in the text.

36. *References: I was hoping that the status of these two papers could updated upon a potential acceptance of the manuscript Carlton et al, submitted 2009 Otte and Pleim, submitted 2009.*
Otte and Pleim (2009) has now been published in GMDD and is available online. The Carlton et al. paper has cleared our internal organizational review and will soon be submitted to Env. Sci. & Technol.

37. *Figures: Some of the figures and the legends of figures are not easily readable because they are too small, i.e. figure 1,5,6,7,8,11, and 12.*
We will try to make the figure labels and legends larger, but we are limited somewhat by the available settings in the software being used to create the plots.

38. *Figure 3, figure caption: Were only sesquiterpene emission fluxes added to BEIS 3.14? In chapter 2.2 the authors state that SOA formation from other biogenic VOCs were included as well. Please clarify, if these were also included in the plots of the third column.*
The last sentence in the Figure 3 caption was meant to clarify that the SOA Increment (Increment B) included changes to the SOA module as well as changes in the emissions inputs. Since this caused confusion we have now modified this sentence to simply state: “The right column shows results from the final SOA module described in

section 2.2."

39. *Figure 11: It should be noted somewhere (either in the plots or in the figure caption) what compound is shown.*
The figure caption has been changed to indicate that these plots are based on ozone mixing ratios.

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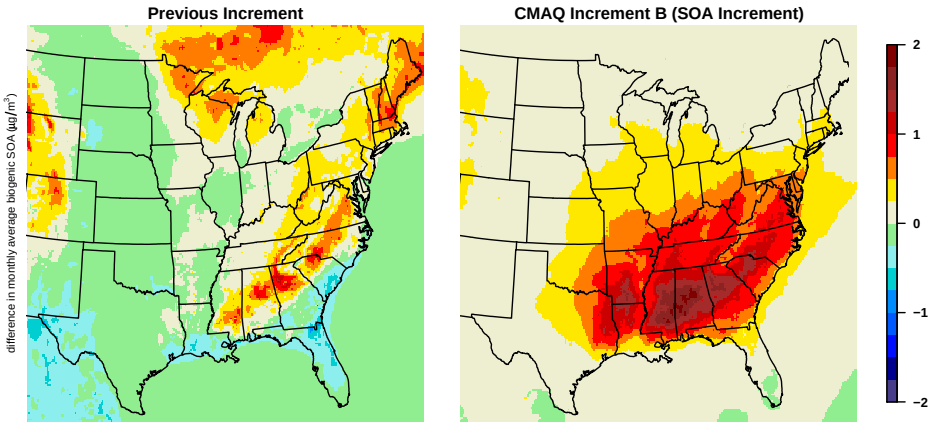

Fig. 1. Difference in monthly average biogenic SOA (μg/m3) (August - January) in the previous increment (left) and Increment B (right).

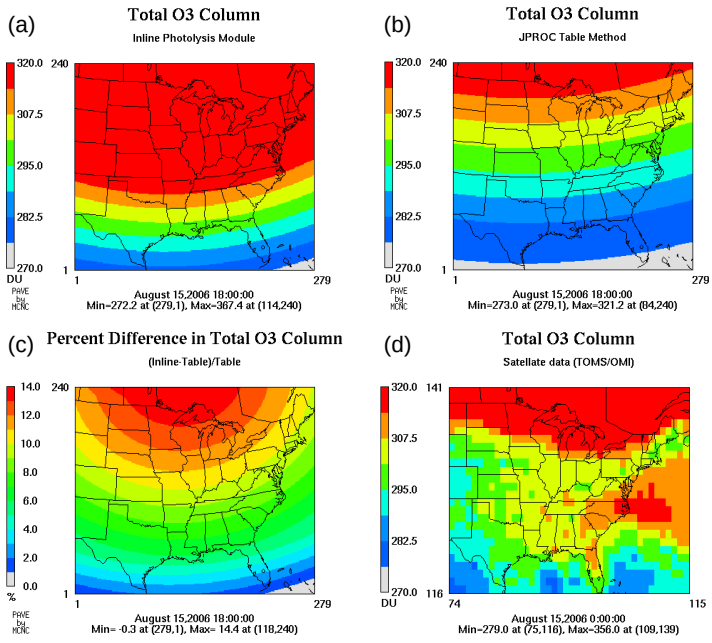

Fig. 2. Spatial plots for August 15, 2006. Total O3 column used by (a) inline photolysis module; (b) JPROC; (d) satellite measurements (TOMS/OMI). Fig. (c) shows percent difference (inline – JPROC values).