Reply to anonymous referee #2

General Comments:

The manuscript "Inline Coupling of Simple and Complex Chemistry Modules within the Global Weather Forecast model FIM (FIM-Chem v1)" discusses global chemical weather forecast (aerosol and gas phase) using three chemistry schemes from simple to complex and evaluates its result against aircraft measurement over parts of the globe. The study is valuable to understand the merits of these schemes as well as to understand some of the biases/issues that are present in inline couple chemistry transport model. The manuscript needs major revision to improve from its present status and Authors need to address the following comments outlined here:

Reply: We really appreciate the reviewer's very helpful comments and suggestions. The paper has been revised throughout based on all of the specific comments listed by the reviewers, including the text, figures, section number, unit and references, etc.

1. 3 difference chemistry schemes discussed in the manuscript, GOCART, GOCART-RACM and advanced RACM_SOA_VBS here. However it is not very clear how these three differs in terms of number of species advected, chemical reactions and computational expense from each other (both aerosol and gas phase). Authors should include that in the page 6 after describing each of the these schemes.

Reply: Revised. We have added a table to show the comparisons of the 3 chemical schemes, also added corresponding descriptions for each of these schemes. See table 1 and Section 2.

2. Include full form of HTAP in Page 6, line 23.

Reply: Revised. See P7 L3.

3. Is biomass burning emission (3BEM) used in the study have any day lag input in the model, like model forecast for today uses day -1 or -2 emission etc. Also whether same settings for all these 3 schemes were used in PRER-Chem step? It should be included in the text.

Reply: Thanks for asking. This is the retrospective forecast, we have prepared all of the biomass burning emission before the forecast, so there is no day lag input for fire emission in the model. Also, we are using the same setting for these 3 schemes in PREP-CHEM-SRC, expect for different schemes, so only the emission species are not the same for these 3 schemes. We have included that into the text. See P7 L11.

4. Include with some references ow much error is associated with PALMS for different species, Page 8.Anticipating it to be small, but Authors should include it.

Reply: Included. See P8 L25.

5. Comparing Figure 2 for total pm2.5, dust and seasalt sources are identical between GOCART and GOCART-RACM, which is expected to be the case. Looking into sulfate, particularly in the difference plot, it seems GOCART-RACM reduced sulfate emission quite a bit over land (with explicit oxidants simulations in its scheme). But why there is so much overprediction over the ocean all over the globe for sulfate in the different plot ? In Fig 2b, GOCART-RACM, over the coast of Africa and Europe and over East Asia there is reduction in sulfate but in the immediate part of the ocean there is a big surge in sulfate is simulated. What is the cause of this surge ? Chemical transport ? Authors should explain this in the text.

Reply: Sulfate is not coming the emission directly, however, it is produced by the simple sulfur chemical reaction from the precursors of SO_2 and background fields of H_2O_2, OH and NO_3. The major difference for sulfate production between GOCART and GOCART-RACM is the background fields of H_2O_2, OH and NO_3. GOCART is using the climatological backgrounds fields of H_2O_2, OH and NO_3 while GOCART-RACM is using the online simulated backgrounds fields of H_2O_2, OH and NO_3 from the RACM schemes. The differences in the background fields of H_2O_2, OH and NO_3 caused the differences in sulfate productions. We have explained that in the text. See P9 L22.

6. Also is sulfate conc in Fig 2 is at the surface or total column ? Include that in the caption and text

Reply: Revised. See Section 3 and the figure captions.

7. Include units in Figure 3.

Reply: Revised.

8. As we are investigating the reason for decrease in GOCART-RACM simulated oxidants, Authors should consider adding 12z figure for OH in figure 3, it will show the reduction over Africa, India and Asia between the two schemes.

Reply: We thank the reviewer's very helpful suggestions. The 12z figures have been added in Fig. 3d in the revised manuscript, which shows the reduction over Africa, India and Asia. See P10 L4.

9. Line 15-19, Page 9, difference in terms of H2O2 and NO3 impacted over land areas only in Figure 3, GOCART-RACM largely reduced near biomass burning regions. However, NO3 conc is less and impact is smaller. OH difference with valid values over the globe will help to understand the issues.

Reply: We thank the reviewer's comments. Yes, the OH difference is the major factor causing the differences in sulfate production. As answering in last question, we have added the 12z figures to show that. See P10 L4.

10. Include whether OH, H2O2 and NO3 are at the surface conc or total column in the figure caption and test.

Reply: Revised. P9 Line 26.

11. Add unit in the figure 4 caption.

Reply: Added.

12. As there is no verification with any satellite data provided, Line 26, page 9 it is hard to compare GOCART-RACM simulation. I suspect much underprediction in surface O3 predicted over East China. Over middle-east and Mediterranean much of surface O3 present in the summer, but it is very less over the corresponding Ocean position. Authors can comment whether that is related to transport or due to intensity of surface O3.

Reply: Fig.4 is a 5-days (120-hours) forecast of O_3 at 00 UTC and 12 UTC, it is early morning and dark evening over East China. The relative low surface O_3 is likely due to the O3 titration during the early morning and nighttime periods. Over the ocean, there are less O3 precursors from the emissions over the ocean compared to the land with anthropogenic activities. See P10 L16.

13. Again add unit in Figure 5 caption.

Reply: Revised.

14. Add SOA simulation by GOCART-RACM for the same time as in the Figure 5 for VOC to understand the difference between GOCART-RACM and RACM_SOA_VBS to understand the impact of the later. Add a description in the text, line 12, page 10.

Reply: We thank the reviewer's comments. Though the GOCART-RACM scheme including gas-phase chemistry, but for the aerosol, it is still using the GOCART aerosol scheme and the aerosols species are the same as GOCART scheme. So there is no SOA in GOCART-RACM scheme.

15. EC is not defined before in the text, how its relation to OC and BC aerosol of GOCART needs to be explained.

Reply: Defined. See P11 L18.

16. It is confusing to understand what flight data used for Figure 6 and 7, in the test it is mentioned 15th and 17th August, but caption within Figure 6 mentioned only 15th Based on Figure 1, it is aircraft data from S. America to Europe combining 15th and 17th August of ATOM dataset. Remove all captions within the figure in Fig 6.

Reply: Revised.

17. As mentioned in line 30, Page 10, is hydrophilic BC and OC only has biomass burning origin?

Reply: Hydrophilic BC and OC origins from both biomass burning and anthropogenic emission.

18. In the figure 6, CO near the surface over the tropics simulated by both GOCART-RACM and RACM_SOA_VBS is very high compare to ATOM. However O3 profiles matches closely with the observation and in the figure 7 OH shows underestimation by the model near the tropics. Something not adding up. Is it possible to include VOC and NOx comparisons in this figure if they are measured by the aircraft to understand a possible pathway. Add them in the text as well.

Reply: We thank the reviewer's suggestions. We have avoided comparing VOC in this study, since sampling times for the VOC instrumentation are either intermittent or very long compared to the 1-second data used in the rest of the analysis. We have looked at NO_x (see Figure A below in the biomass burning event as in Fig.9a), with NO_x profiles from the RACM models being comparable to the observations except some overestimate at the upper levels.

The CO, OH and O_3 inconsistencies noted by the reviewer are related to the biomass burning signature in that region. To address this concern, we have added analysis and discussion based on Fig.17 in the revised manuscript, showing scatter plots of observed and modeled NO_y versus CO and O_3 versus NO_y for the tropical biomass burning signature seen between 22°S and 12°N below 6km altitude. The text in Section 5 has been modified to include a brief discussion of what the reviewer has noted. We use NO_y in the discussion, rather than NOx, but NO_y is largely NO_x, especially in fresh fire plumes. See P15 L15-25.

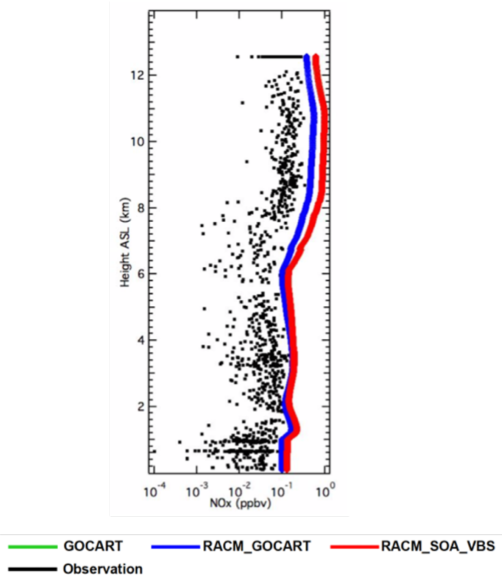

Figure A. ATom-1 observations and model vertical profiles of NOx in the biomass burning events. The biomass burning plume is from August 15, 2016, profile #16 near 20°S.

19. In the figure 10, surprised to see GOCART with prescribed climatological oxidants simulating sulfate conc above 4km closely with ATOM rather than two other schemes which simulates explicit oxidant fields. Is it possible to add H2O2, and NO3 comparison for Continental USA case to understand difference between the schemes that gives such result.

Reply: We do have the comparisons of OH and H_2O_2 in GOCART, RACM_GOCART and RACM_SOA_VBS with ATom-1 observation (see Figure B). We can not compare NO3 because we don't have the observation data. We can see that the prescribed OH is close to the ATom-1 observation, which may be the major factor contributing to get better sulfate production in GOCART. Also, from Fig. 2 and Fig. 3 in the manuscript, OH looks like the major factor impact on the sulfate concentration.

Considering the sulfur chemical reaction mechanism and the aerosol scheme in RACM_SOA_VBS is completely different to that in GOCART and RACM_GOCART, the comparison of oxidants may not be the only reason causing the differences, which still needs more investigations. Though the sulfur chemical reaction mechanism is consistent in GOCART and RACM_GOCART, but the fields of OH, H_2O_2 and NO_3 are not from the prescribed oxidants used in GOCART. Different to the GOCART, the SO_2 and sulfate in RACM_GOCART are also impacted by the gas phase chemistry of RACM scheme, which is also difficult to quantity the impact from the OH, H_2O_2 and NO_3 and needs further study.

We have included all of these descriptions and figures into the revised manuscript. See P16 L6 and L20.

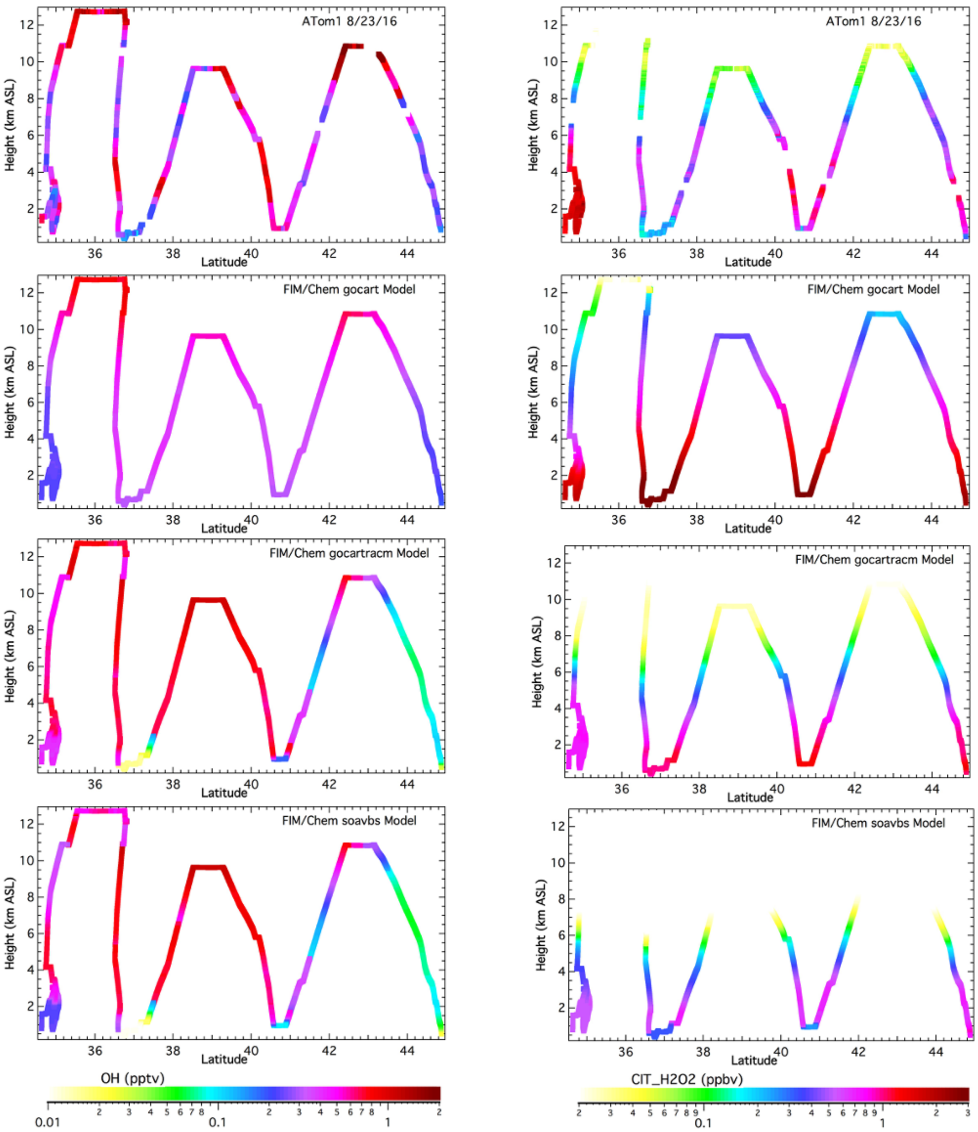

Figure B. Height-latitude profiles of OH and H2O2 over United States on August 23rd, 2016 for (a) ATom-1; (b) GOCART; (c) RACM_GOCART; and (d) RACM_SOA_VBS.