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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The effects of global change upon United States air quality" *by* R. Gonzalez-Abraham et al.

R. Gonzalez-Abraham et al.

rodrigoga@mce2.org

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We appreciate the referee for his/her constructive and thorough review. We have revised the manuscript according to the referee's comments. During the revision, we learned there were bugs in CMAQv4.7 in processing emissions, as described here: www.cmascenter.org/cmaq/documentation/4.7.1/RELEASE_NOTES.txt. Accordingly, we have re-run all simulations using CMAQv4.7.1, resulting in substantial changes to the PM2.5 section. Changes in ozone also occurred as a result of upgrading to CMAQ 4.7.1 due a combination of updates in emission, advection and plume rise emission modeling. Most notably, a larger increase in ozone in coastal urban areas is projected as a result of changes in US anthropogenic emissions. We believe the manuscript

has been much improved in addressing comments by both referees and updating the manuscript to reflect the new simulations. Our detailed responses to Referee #2's comments (in italics) appear below.

After the general comments, every comment is numerated. The response is immediately below the comment The revised manuscript is attached.

General Comments

The authors present findings related to the application of the WRF and CMAQ models to downscale future climate predictions estimated with a global model simulation. They find that improvements in ozone and PM air quality due to decreases in anthropogenic emissions are somewhat offset due to climate change and increases in global emissions. The manuscript is generally well written and the approach is thoroughly described. However, certain aspects of the study need further explanation. The scope of this manuscript in its current form does not present particularly novel work in this area, especially for ACP.

Response

While the manuscript does not present a novel methodology, the set of simulations do present different elements than previous publications. The novelty is stated below.

a) The paper separates the effect of climate change on air quality from the effect of changes in biogenic emissions due to climate change. This approach, while unrealistic, situates the largely documented effect of climate change (with changes in biogenic emissions) on air quality on a different perspective. The approach followed in this manuscript shows that climate change alone has a dominant effect on PM2.5 and O3 concentrations over the increase in biogenic emissions in some regions of the US. Also, the regional changes in land use and the resulting effect in the emissions of BVOC with respect to the emissions of NO offer a wider set of sensitivities of the model to changes in emissions.

b) Unlike previous research, the paper shows the sensitivity of Ozone and the inorganic fraction of PM2.5 to different regimes of VOC and NOx concentrations. The approach is followed in the form of a sensitivity analysis.

c) The manuscript presents a wider range in the levels of atmospheric oxidants and their effect on Ozone and PM2.5 due to climate, biogenic and anthropogenic emissions.

d) In the case of DM8O, our approach presents a diverse perspective of the reactions that involve the gas and the organic and inorganic particle phase. One example is the importance of sequestration versus recycling of isoprene nitrates in the SAPRC99 mechanism presented in section 3.3.3.

1) The current year model performance is concerning. If these results are consistent with other downscaling efforts then maybe that is a notable addition to the literature and could be focused on in more detail as a potential confounding factor in interpreting these projected future year scenarios.

As noted above, due to a bug in the emission processing in CMAQv4.7 we have re-ran all simulations using CMAQv4.7.1. As a result, the model performance for PM2.5 has improved (Figure 7 of the revised manuscript). Also, The model performance has to be understood in terms of the climatological perspective of the investigation. Both current and future meteorological conditions are derived from downscaled climate simulations rather than meteorological reanalysis. In our particular study, ECHAM5 global climate model was used. Biases in the drivers of ozone and PM formation and removal are in both the current and future decade simulations. Previous studies that used downscaled climate realizations to drive air quality models also show overestimations of DM8O (Tagaris et al., 2007; Nolte et al., 2008) and underestimations of total PM2.5 mass (Tagaris et al., 2007).

2) RCP scenarios are not always good indicators of future changes in air quality since they are focused on greenhouse gases rather than O3 and PM precursors. The conclusion that ozone reductions due to lower anthropogenic emissions being totally off-

set by increases in global ozone and climate change impacts seems inconsistent with other studies. Even more importantly, observed ozone concentrations continue to trend downward in many places in the United States suggesting US emissions reduction impacts are outpacing climate and international emissions increases. We are 10 years past the baseline for this model simulation so that reality should be recognized in some way.

This study is based on the SRES (used in IPCC's Fourth Assessment Report (AR4) rather than the RCPs (used in AR5). The Greenhouse gas emissions from the SRES scenarios are used to drive the global climate models. We agree that they are not always good indicators of future changes in air quality; but in many countries it is the only proxy for future scenarios. As presented in the results, we explicitly recognize that the scenarios of global and regional anthropogenic emissions are the reason for the inconsistencies between studies. We also explicitly acknowledge in the conclusions that updated emission inventories and future emission scenarios are needed to improve the assessment of the impacts of global change upon the US. While the emission inventory, the assumptions for the regional and global emission projections and the meteorological simulations may be outdated, the results presented in this manuscript are still relevant evidence of the dependence of the US air quality of global emissions and the need of rigorous emission controls. Nevertheless, additional information was added to the manuscript for clarity. A figure with the emissions from the semi-hemispheric domain was added. The figure shows the dramatic increase in NOx emissions across the domain and aims at the reason behind the increase in DM8O. A line addressing a possible higher reduction in ozone as a result of updated future regional emission projections was also added to the conclusion.

3) How closely did the projections using MARKAL match those of the IPCC A1B scenario for areas where both are applied? Are the ozone changes aggregated and post-processed similarly to other similar studies? More clarity is needed regarding methane. The authors state they do not consider methane but may have indirectly as impacts of

increasing methane emissions globally may be part of the global 2050 simulation

Emissions from the IPCC A1B scenario and results of the MARKAL methodology were never overlapped on the same simulation. However, CO2 is underestimated in the A1B scenario in comparison to the US emission inventory and MARKAL projection. The metric used (daily maximum 8 hour average ozone) is common to the studies based on US air quality. Methane emissions are not used in the chemical mechanism used in CMAQ. The impact of methane is in the global climate represented in the ECHAM5 simulations and passed onto the downscaled meteorological fields that drive CMAQ. However, impact of increased methane on chemistry is not considered in this study; in all our CMAQ simulations, methane concentration is fixed at 1.85 ppm.

Author's Change in the manuscript. Two changes have been made:

a) Changes in the text of the introduction to highlight the direct effect of methane. "Furthermore, despite the observed sensitivity of tropospheric ozone to regional emissions and global burden of methane (Zhang et al., 2011; Fiore et al., 2008; Wu et al., 2008a; Nolte et al., 2008; Fiore et al., 2006), in this work, we do not address the direct effect of emissions of methane in the air quality simulations"

b) Section 2.5. Added the line "Methane concentration is fixed at 1.85 ppm for all CMAQ simulations."

Introduction section:

4) First paragraph; the authors have 2 references for negative health effects associated with O3 and PM. Typically journal articles focus on either O3 or PM so it is not clear that both of these references are relevant for both O3 and PM. End of second paragraph; it would be appropriate to also reference Cooper et al 2010 and Cooper et al 2012 here.

For clarity, we added "and, in the case of PM," to the sentence. We also added Cooper et al. 2010 and Cooper et al. 2012, as requested.

5) Page 31847 lines 1-5; It would be helpful for the reader if the authors add what time

scale these increased ozone concentrations are seen. Annual average? Average of the highest days? Just the highest days or are low ozone days part of the average?

The text was modified for clarity: "In summary, despite the differences in modeling elements, all studies found an increase in the summer average of the daily maximum 8-hour average ozone concentrations over large regions of the simulated CONUS domain on the order of 2 to 8 ppb (Weaver et al., 2009)"

6) Page 31847 lines 6-20; this section should mention the time period over which the climate changed and whether the climate impact was isolated or US emission changes were also considered.

The manuscript was modified for clarity and the time periods were also added to the paragraphs: "This downscaling approach has been used in a variety of studies in Europe, Canada, and Asia at different time scales of climate change (e.g., Liao et al., 2006 [2000 to 2100]; Langner et al., 2005 [2000 to 2060]; Forkel and Knoche, 2006 [2990 to 2030]; Meleux et al., 2007 [1975 to 1985]; Kunkel et al., 2007 [1990 to 2090]; Lin et al., 2008 [2000 to 2100]; Spracklen et al., 2009 [2000 to 2050]; Kelly et al., 2012 [2000's to 2050's])." Later in the fifth paragraph of the introduction, we also added "projected to the 2050s" for the US studies funded by the EPA.

7) The A1B scenario has aggressive methane emissions growth through 2040. Is that impact included in the semi-hemispheric model? That is not clear but important in interpretation of the global impacts offsetting decreases in US anthropogenic emissions.

The air quality simulations in this study do not take into account future changes in methane. This caveat was added in the introduction and the first paragraph of section 2.5: "Furthermore, despite the observed sensitivity of tropospheric ozone to regional emissions and global burden of methane (Zhang et al., 2011; Fiore et al., 2008; Wu et al., 2008a; Nolte et al., 2008; Fiore et al., 2006), in this work, we do not address the direct effect of emissions of methane on the air quality simulations" " Section 2.5. Added the line "Methane concentration is fixed at 1.85 ppm for all CMAQ simulations."

Methods section:

8) Sections 2.3 and 2.4: A summary Table similar to Figure 3 showing the non-U.S. emissions would be extremely helpful with interpretation of the results. This is especially critical given the main conclusion that global emissions increases are offsetting reductions in US anthropogenic emissions. It is unclear what is driving the large increases in VOC, primary PM2.5, and ammonia. Primarily emitted PM2.5 is fairly cost effective for point sources and VOC would seemingly be decreasing due to vehicle fleet turnover and mobile source sector regulations. It is not clear why emissions from confined animal operations and/or fertilizer application would just continue to increase in the future. There is only so much land to farm and so much fertilizer than can be put in the soil. One of the reasons the authors see global emission increases compensating for decreases in US anthropogenic emissions is related to the choices made here for these species so the reasoning behind these VOC/PM/ammonia increases are important.

A new figure was added that shows the changes in global emissions (Figure 3 in the revised manuscript). For energy systems, CO2 growth factors were used as a surrogate for CO, NH3, and VOC. For mobile sources, NOx growth factors were used for CO, VOC, and NH3. Non-combustion industrial emission growth factors were developed from projections of economic growth. Growth factors for non-combustion emissions from the residential and commercial sectors are linked to population growth. More text were added in paragraph 4 of Section 2.4 to make this clearer.

We agree that more discussion is required about the effect of increasing CO and VOCs. In the last paragraph of Section 2.4, we have added the following sentence: "The use of CO2 as a surrogate for growth factors as described above means the projected CO and VOC emissions are likely too high." Text was also added in the last paragraph of Section 3.3.1 to address the effect of growing emissions of CO and VOC: "The smaller reduction in ozone concentrations between the future and the current decade in comparison to Nolte et al., (2008) is likely to be a consequence of the increase

in VOC and CO emissions from business-as-usual scenario of MARKAL, which uses surrogates as growth factors for CO and VOC (Loughlin et al. 2011)."

9) Evaluation of model performance (2.6): A tighter scale for Figure 4 would make this Figure more useful for interepretation. More explanation and discussion is needed regarding the overestimated temperatures in the central and eastern U.S. These are rather large overpredictions and would seem to result in overestimated biogenic VOC. These ozone overestimates are extremely large. Mean MDAO3 in the Midwest in current conditions is 50 ppb but the modeling suggests 70 ppb! I appreciate that the authors have not tried to generate Tables and Figures to minimize our focus on the current conditions model performance but it is important to consider these issues when thinking about the results.

The original Figure 4 is now Figure 5 and has been modified as suggested by the referee. We agree with the referee that that overprediction of temperatures and its impact on ozone should be noted. The revised manuscript includes the following statement: DM8O is be over-estimated in regions where temperature maxima are also over predicted, most noticeably in the Midwest, the South, and the Southeast but also in the Northeast."

10) Given the large overestimation bias it would be good for the authors to show that rather than refer to a different manuscript. Also, it would be helpful to know why it is important to represent the correlation between ozone and temperature. If they are both grossly overpredicted is model performance really ok just because these variables correlate well?

Evaluation of model performance was carried out comparing modeled values vs observations (Figure 5 which is 6 in the revised version of the manuscript). The correlation between ozone and temperature across different years is evidence that bias in temperature is the main cause of the bias in DM8O and that the air quality model is responding to the meteorological driver of ozone production and thus can predict the impact of cli-

mate change on DM8O. The text has been edited to have: “Despite the bias, results from the modeling framework presented here have been shown to accurately represent the correlation between ozone and temperature at rural CASTNET sites throughout the US (Avise et al., 2012), suggesting that the bias in temperature is a main cause of the bias in DM8O and that the chemical transport model is responding to the meteorological driver of ozone production and thus can predict the impact of climate change on DM8O.”

11) These underpredictions for PM2.5 are very large and puzzling since ozone is grossly overestimated. Generally when a modeling system can predict a lot of ozone there would be a lot of sulfate, but the spatial plot of average current condition PM2.5 does not show the typical eastern US regional sulfate signature typical for model runs representing the early 2000s. The PM2.5 performance looks like there is something fundamentally wrong with either the CMAQ simulations or the way they were post processed.

As mentioned previously, in the process of addressing this comment, we discovered a bug in CMAQv4.7 that resulted in NH3 emission being read into the model incorrectly. We have re-run all the simulations using CMAQv4.7.1. Modeled to observed PM2.5 comparison is show in Figure 7 in the revised manuscript. The PM2.5 results are significantly improved, though PM2.5 is still underestimated in the western half of the US. We note in the revised manuscript that the causes of underprediction include the lack of wind blown dust and fire smoke emissions, underestimation of SOA formation, and overprediction of precipitation. Note that due to the lack of speciated PM data for our period of simulation, we have removed the section on model performance for speciated PM (the observed data in the ACPD version were from 2003-2008 and we believed is not representative of our period of simulation).

12) Comment from Referee. Despite what the authors suggest in 375-377 this type of underestimate is not typical of other regional CMAQ simulations for this time period and performance here is far worse than Foley et al and Appel et al (and probably the

others but I don’t have time to go back and check them all).

Thanks to the referees’ comment, we encountered a bug on CMAQ version 4.7. We re-ran the simulations with CMAQv4.7.1 and the results changed significantly. Please see comments above.

13) There is really no value to discussing the fractional composition of PM2.5. How is that important for providing confidence in the model? If the model gets the composition right how does that make the large underestimates of all the major species ok? The fractional evaluation should be removed from the manuscript. Please just provide a comparison of speciated mass for the major species.

We agree with the Referee. This section has been removed (see response to comment #11 above)

14) It is very debatable the model is performing well for PM2.5. A five year summer average of 5.6 ug/m3 for total PM2.5 mass during the early 2000s is extremely small. Typically models do quite well capturing regional sulfate and since these simulations are under predicting so much it raises some questions about the model inputs, in particular the emissions.

The text in this section has been changed after we re-ran the simulations using CMAQv4.7.1. After the simulations, PM concentrations did increase across the simulation domain with slight overprediction in the eastern half of the US. For the western US, however, PM2.5 remain heavily underpredicted. The underprediction can be explained by the overprediction of precipitation. The last paragraph of Section 2.6 has been edited to reflex the new simulation results.

Results section:

15) The A1B scenario is a not a great projection of what will happen to global methane and NOX in the future so that caveat would be useful here and in other places where this is mentioned. Does the Hogrefe 2004 paper detail how much of this increase in

related to methane emissions?

In this study, methane concentration is fixed at 1.85 ppm. We have added this caveat to the manuscript; see response to comments #3 and #7. Hogrefe does not detail how much increase is due to methane.

It has been published that the emission projections considered from the SRES have been underestimated (Garnaut et al., 2008; Pozzer et al., 2012; Williams et al., 2014). But the level of uncertainty of the socio economic drivers behind each scenario was the motivation behind the SRES developers to consider each of the scenarios equally plausible. We would be thankful if the referee expands on the evidence behind his/her assertions on future emissions of NOx from the A1B scenario.

16) Comment from Referee. Please provide more explanation about how biogenic VOCs are sequestering ozone precursors and which ozone precursors are being sequestered. NOX? Toluene? How do the authors separate the impacts of this sequestration and recycling of isoprene nitrates? It sounds like this information is available and since this might be a more novel aspect of this research I suggest the authors provide spatial plots or some way to present the relative contributions of these processes here.

We have added Figure 14 and text in Section 3.3.3 regarding the sequestration of NOx: "This [DM8O] decrease is associated with a decrease in NOx concentrations (Fig. 14a). This decrease in NOx suggests that the effect of sequestration of NOx by the biogenic VOCs as organic nitrates (RNO3) is predominant over the effect of recycling of NOx considered in SAPRC-99, which lumps all non-PAN organic nitrates as one compound that has a NOx recycling efficiency of about 30%. This reduction in ozone is consistent with the results of Xie et al. (2013), who reported increases in NOx and ozone in the Southeast when sequestration by isoprene nitrates was reduced relative to the base SAPRC-07T mechanism that has the same RNO3 treatment as SAPRC-99. Evidence of the predominant effect of sequestration over the recycling of NOx in the eastern

US is seen in Figure 14, which shows an increase in RNO3 and reduction in the NOx concentrations in most of the eastern US for Simulation 2 relative to Simulation 1

17) Probably worth noting these increases are driven by global emissions changes.

Reviewer didn't link the comment to any particular paragraph, line, table, figure or section in the manuscript.

Conclusions section:

18) Are Asian and Mexican emissions impacts tracked separately from other countries? It would be nice to see the breakdown by country if that is available.

We did not run additional sensitivity simulations to separate out the effects of Asian and Mexican emissions. It is something to consider for future work

19) The biases in the model current conditions period should be mentioned here again.

The text was added to reflect the referee's suggestion

The next line was added to the conclusion. "The evaluation of the model performance for the base case simulation shows an overestimation of DM8O and an underestimation of PM2.5 across the projected domain"

20) Suggest changing the word "will" to "may" since this assessment is not singularly conclusive.

Referee is correct. The word "will" was changed to "may" throughout the conclusion section.

Figures:

21) Figure 1. The US only map is impossible to read with the current color scale.

We have changed the color scale of Figure 1 so that the US map is more visible.

22) Figure 4. Please show number of stations similar to Figure 5.

We have edited the figure according the referee's suggestion.

23) Figure 6. Please show absolute speciated PM2.5 predictions and observations. The fractional plot does not terribly useful. Since the authors are referencing EPA guidance I suggest showing prediction-observation pairs with shorter averaging times like either the 24-hr or monthly averages to be more consistent with that guidance and to provide more useful information.

The plot has been added to the manuscript. See response to comment #11.

24) Figure 11. Its not clear that any of these panels isolate the biogenic impacts. Is that on here already?

Note that the original Figures 11 and 12 are now Figures 12 and 13. In Figures 12 and 13, panels c and d show the effects of biogenic emissions due to climate change and of biogenic emissions due to climate plus land use changes, respectively. We have edited the figure captions to be this clearer.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/14/C13410/2015/acpd-14-C13410-2015-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 31843, 2014.