

## Reply to Comments from Reviewer 1

Geosci. Model Dev. Discuss., 8, C152–C154, 2015

[www.geosci-model-dev-discuss.net/8/C152/2015/](http://www.geosci-model-dev-discuss.net/8/C152/2015/)

© Author(s) 2015. This work is distributed under the Creative Commons Attribute 3.0 License.

Open Access

Geoscientific  
Model Development  
Discussions

**Interactive comment on “Application of WRF/Chem version 3.4.1 over North America under the AQMEII Phase 2: evaluation of 2010 application and responses of air quality and meteorology–chemistry interactions to changes in emissions and meteorology from 2006 to 2010” by K. Yahya et al.**

**Anonymous Referee #1**

Received and published: 12 March 2015

A comprehensive model evaluation study on the WRF/Chem performance for simulating meteorology and air quality over two years with two different configurations (offline and online), respectively, is presented. Effort has been made in putting all the analysis together and trying to make meaningful presentations of the data. It is very challenging to perform mechanistic evaluation of air quality models over different years with so many uncertainties in meteorology, emissions, and ICONs/BCONS. It often entails more advanced skills and techniques to draw credible conclusions about a model’s responses to some specific changes over the years by eliminating or reducing interference from other uncertain factors.

**Reply:**

**We thank the reviewer for careful review of this manuscript and valuable comments to improve the quality of manuscript. In particular, the reviewer recognized the challenge in performing the mechanistic evaluation for long period simulations and for years having different meteorology, emissions, and ICONs/BCONS, especially for the online-coupled model used in this study.**

**We have carefully addressed all the comments raised by the reviewer to improve the technical and presentation quality of our paper. Please see below our point-by-point replies.**

However, the authors are trying to achieve the goal by simply comparing the model results with observations using the simple statistics (Corr, NMB and NME) and some plots. As the authors pointed out that the main objectives of the Part II paper are to examine whether the model has the ability to consistently reproduce observations for two separate years, as well as to examine whether the trends in air quality and meteorology-chemistry interactions are consistent for both

years. But after reading the manuscript from the beginning to the end, the answers to the above questions are not there.

**In order to achieve our goals, we first compared the model results with observations in 2010 (see Table 1) (similar evaluations for 2006 were performed by Yahya et al. (2014), see Table 1 in Yahya et al. (2014)). The evaluation we performed is very comprehensive and includes all major meteorological, chemical, radiation, and cloud related variables using various available surface network and satellite datasets. We have calculated full sets of statistics (> 16 statistical measures), although for the sake of brevity, our discussions on the statistics only focused on a few of them in this paper. We also evaluate agreement of predictions with observations on various temporal resolutions (i.e., diurnal, seasonal, and annual) and spatial correlations. Such a comprehensive evaluation can assess the model's ability to consistently reproduce observations for two separate years. The examination of model ability to consistently reproduce observations for two separate years has been discussed in Section 3.5.**

**Following a comprehensive evaluation, we then calculated the percentage changes in observed and simulated meteorological and chemical variables between 2010 and 2006 (see Table 2) to assess whether the trends in air quality and meteorology-chemistry interactions are consistent for both years. The trends in air quality and meteorology-chemistry interactions for both 2006 and 2010 are further discussed in Sections 4.1-4.3. Based on collective analyses of all those evaluations and trend analyses (instead of just the simple performance statistics), we found that the model is able to reproduce the observations to a large extent for most meteorological surface variables except for precipitation. The model has significant biases in a few aerosol and cloud variables well, such as for AOD, COT and CCN, however, it is able to reproduce the trends in the aerosol-cloud-radiation variables for 2006 and 2010. The model performs better for O<sub>3</sub> mixing ratios and PM<sub>2.5</sub> concentrations for 2006 compared to 2010 due to more realistic chemical initial and boundary conditions ICONs/BCONs and emissions. For 2010, Im et al. (2014a) found that the MACC model underpredicts surface ozone levels over North America by 22%. Im et al. (2014b) also showed that most models that used the MACC boundary conditions underpredicted PM<sub>2.5</sub> concentrations for 2010.**

**In addition, we conducted several sets of sensitivity simulations as described in Section 4.4 (also see a new table (Table 3), for the simulation setup) to examine the model's responses to specific changes such as meteorology or emissions or chemical ICONs/BCONs only and to estimate the relative impacts of changes in meteorology, emissions, and chemical ICONs/BCONs.**

**With a comprehensive model evaluation, trend analyses, and additional sensitivity simulations, we believe that we have achieved our objectives. To address the reviewer's comments, we have revised the manuscript thoroughly to include more in-depth analyses and better relate the findings of this work to the main objectives of the paper. In addition, we added a new table (Table 4) to evaluate if the sensitivity simulations with different**

**meteorology, emissions, and chemical ICONs/BCONs for Jan. and July 2010 can improve the model's capability in reproducing the trends in both meteorological and chemical variables, as comparing to baseline results in 2006 and 2010.**

**The sections that have been revised include:**

- (i) Section 3.1 to explicitly state the similar trends in terms of meteorological performances from 2006 and 2010 as well as additional explanations for several biases in meteorological performance;**
- (ii) Section 3.2 stating that the chemical performance between 2006 and 2010 is more variable compared to the meteorological performance of surface variables;**
- (iii) Section 3.4 stating that the model is able to reproduce generally similar performances against observations for most of the aerosol-cloud variables for both 2006 and 2010;**
- (iv) Section 3.5 stating that overall, the model is able to predict the trends in all the listed meteorological, chemical and aerosol-cloud-radiation variables between 2006 and 2010 with the exception of WS10 against CASTNET, Precip, CF, maximum 8-hr O<sub>3</sub> against CASTNET and 24-hr EC against IMPROVE as well as additional analysis to explain the reasons**
- (v) Section 4.4 in which we added discussions on the model's capability in reproducing trends between Jan./Jul. 2010 and 2006 with incremental changes in meteorology, emissions, and chemical ICs/BCs.**

Throughout the manuscript, the authors were talking about statistics superficially without in-depth analysis about what caused the agreement/disagreement.

**Reply:**

**The manuscript has been thoroughly revised to include more detailed analyses on model evaluation and likely causes for discrepancies. For example, we added that the cold bias in T2 is attributed to the lack of soil data assimilation in this study in Section 3.1.**

When pairing cell-averaged model predictions with point measurement data in space and time (incommensurability), how much confidence do you have in terms of the good/bad performance of a model for different years with a few percentage differences in NME? I don't oppose using the statistics to perform model evaluations, but it seems too much for me if the analysis is heavily dependent on these numbers and the conclusions were drawn based mainly on these numbers.

**Reply:**

**The U.S. EPA has provided benchmarks (US EPA, 2007) in model evaluation in terms of statistics such as the mean normalized gross error (MNGE) and mean normalized bias (MNB). The performance criteria used in this study follow Zhang et al. (2006), which include model bias (e.g., NMB) and error (e.g., NME) for good or poor model performance. For example, an NMB of  $\leq 15\%$  and an NME of  $\leq 30\%$  indicate satisfactory performances for O<sub>3</sub> and PM<sub>2.5</sub>. We fully agree with the reviewer that the assessment of the model**

**performance should not be simply based on performance statistics. As mentioned before, our conclusions for model performance are based on not only statistical evaluation but also other evaluations temporal (e.g., annual, seasonal, diurnal average) and spatial analysis, as well as several sensitivity studies during Jan and Jul.**

Comparing the diurnal variations (Figure 4) using the whole year and all site data doesn't make sense to me. Considering all the averaging effect through space and time, to relate temperature with O<sub>3</sub> concentrations in this context is very weak.

**Reply:**

**Figure 4 was actually averaged over only the summer period of June to August (O<sub>3</sub> season) at monitoring sites from CASTNET. CASTNET consists primarily rural and remote sites, we think that averaging predictions at the CASTNET sites is technically sound (note that the performance statistics is also calculated separately at sites from each network); their comparisons with averaged observations over the same dataset can provide an assessment of the sources of model biases at the CASTNET sites.**

**In addition, such an evaluation can shed light on whether the underprediction of O<sub>3</sub> mixing ratios is a systematic bias, i.e., day and night, or if it was just a portion of the day.**

**To address the reviewer's comments, we have revised the paper to explain why we averaged model predictions and observations at those sites and the purpose of such diurnal assessments.**

In Section 4, I expected to see some in-depth analysis about the model's response to the changes in emissions and meteorology and this should be the central point the authors are trying to make in this manuscript. But after I read the entire section, I was disappointed, because it simply listed the increase or decrease of the species from one year to another with very basic speculations (and some of them are known facts) and the connection between model response and input changes simply wasn't made. The model's response should be reflected (for example) under the percentage changes in emissions, under the similar weather conditions, does the model respond to the same percentage changes in pollutant levels as it was revealed in the observations.

**Reply:**

**Section 4 has been extensively revised to include more in-depth analyses on the sensitivity simulations, which include the effect on the model response when using different sets of emissions, meteorology and chemical initial and boundary conditions.**

**To address the reviewer's last comment, we calculated the changes in the simulated meteorological and chemical variables due to changes in meteorology and emissions individual and collectively in Jan and July and compared them to the observed changes in the trends in those variables in a new table (Table 4). We also calculated percentage changes in emissions between 2010 and 2006 and added this info in a new table (Table S1 in the supplementary material). Relevant discussions along with percentage changes in emissions and meteorology have been added in Section 4.**

**References cited in this reply:**

- Im U., R. Bianconi, E. Solazzo, I. Kioutsioukis, A. Badia, A. Balzarini, R. Baró, R. Bellasio, D. Brunner, C. Chemel, G. Curci, J. Flemming, R. Forkel, L. Giordano, P. Jiménez-Guerrero, M. Hirtl, A. Hodzic, L. Honzak, O. Jorba, C. Knote, J. J.P. Kuenen, P. A. Makar, A. Manders-Groot, L. Neal, J. L. Pérez, G. Pirovano, G. Pouliot, R. San Jose, N. Savage, W. Schroder, R. S. Sokhi, D. Syrakov, A. Torian, P. Tuccella, J. Werhahn, R. Wolke, K. Yahya, R. Zabkar, Y. Zhang, J.-H. Zhang, C. Hogrefe, S. Galmarini, 2014a, Evaluation of operational online-coupled regional air quality models over Europe and North America in the context of AQMEII phase 2. Part 1: Ozone, Atmos. Environ., doi:10.1016/j.atmos.env.2014.09.042.**
- Im, U., R. Bianconi, E. Solazzo, I. Kioutsioukis, A. Badia, A. Balzarini, R. Baró, R. Bellasio, D. Brunner, C. Chemel, G. Curci, H. D. van der Gon, J. Flemming, R. Forkel, L. Giordano, P. Jiménez-Guerrero, M. Hirtl, A. Hodzic, L. Honzak, O. Jorba, C. Knote, P. Makar, A. Manders-Groot, L. Neal, J. L Pérez, G. Pirovano, G. Pouliot, R. San Jose, N. Savage, W. Schroder, R. S. Sokhi, D. Syrakov, A. Torian, P. Tuccella, K. Wang, J. Werhahn, R. Wolke, R. Zabkar, Y. Zhang, J.-H. Zhang, C. Hogrefe, and S. Galmarini, 2014 b, Evaluation of operational online-coupled regional air quality models over Europe and North America in the context of AQMEII phase 2. Part II: Particulate matter, Atmos. Environ., doi: 10.1016/j.atmos.env.2014.08.072.**
- USEPA, 2007, Guidance on the use of models and other analyses for demonstrating attainment of air quality goals for Ozone, PM<sub>2.5</sub> and Regional Haze, The U.S. Environmental Protection Agency, Research Triangle Park, EPA-454/B-07-002, 2007.**
- Yahya, K., Wang, K., Gudoshava, M., Glotfelty, T., and Zhang, Y., 2014, Application of WRF/Chem over the continental U.S. under the AQMEII Phase II: Comprehensive Evaluation of 2006 Simulation, Atmos. Environ, doi:10.1016/j.atmosenv.2014.08.063.**
- Zhang, Y., Liu, P., Pun, B., Seigneur, C., 2006, A comprehensive performance evaluation of MM5-CMAQ for the summer 1999 southern oxidants study episode, part I. Evaluation protocols, databases and meteorological predictions, Atmos. Environ., 40, 4825 – 4838.**

## Reply to Comments from Reviewer 2

Geosci. Model Dev. Discuss., 8, C171–C172, 2015

[www.geosci-model-dev-discuss.net/8/C171/2015/](http://www.geosci-model-dev-discuss.net/8/C171/2015/)

© Author(s) 2015. This work is distributed under the Creative Commons Attribute 3.0 License.

Open Access

Geoscientific

Model Development

Discussions

**Interactive comment on “Application of WRF/Chem version 3.4.1 over North America under the AQMEII Phase 2: evaluation of 2010 application and responses of air quality and meteorology–chemistry interactions to changes in emissions and meteorology from 2006 to 2010” by K. Yahya et al.**

**Anonymous Referee #2**

Received and published: 17 March 2015

This paper evaluates the WRF/Chem model performance and responses of air quality and meteorology-chemical interactions to the meteorological and emission changes in 2006 and 2010. By comparing the model prediction of WRF/Chem and WRF, the chemical feedbacks to meteorology are assessed. And a series of sensitivity simulations are pursued to distinguish the differences driven by emission changes, meteorological variation, and Chemical ICONs and BCONs. This paper is valuable to understand the WRF/Chem model performance in catching the yearly variations, and reveals the necessity of improving the accuracy of emissions and chemical BCONs, the SOA module, and the chemical-meteorology feedbacks in the online-coupled model.

### **Reply:**

**We thank the reviewer for careful review of this manuscript and recognition of the values of this work. We have carefully addressed all the comments raised by the reviewer to improve the technical and presentation quality of our manuscript. Please see below our point-by-point replies.**

Nevertheless, several important points should be addressed to support the paper conclusions. (1) In section 3.5, "The trends for Precip and CF for simulated variables are not consistent with observed trends from 2006 to 2010. Observed NADP Precip increased slightly from 2006 to 2010 by  $\sim 7\%$ , however both simulated WRF and WRF/Chem show a small decrease from 2006 to 2010...." . Can the authors explain why the model fail to reproduce the trends of precipitation and CF between 2006 and 2010?

### **Reply:**

**Although WRF/Chem is a state-of-science online-coupled meteorology-chemistry model, there still exist large uncertainties in the model treatments of the aerosol-radiation-cloud**

feedbacks, e.g., in the microphysics and cumulus parameterization schemes which will affect precipitation predictions. In addition, as mentioned in the text, model precipitation has large biases against observations. It is also likely that the decrease in precipitation between 2006 and 2010 by the model is due to the smaller decrease in SWDOWN compared to observations between 2006 and 2010. This would result in less convective precipitation during the summer but increased CF for 2010. In addition, PM<sub>2.5</sub> concentrations are more underpredicted in 2010 than 2006 (i.e., simulated PM<sub>2.5</sub> is a better agreement with observations in 2006). Underpredicted PM<sub>2.5</sub> concentrations will affect the formation of clouds and precipitation via various direct and indirect effects.

In Section 4, additional trend analyses for Jan. and Jul, 2010 based on baseline and sensitivity simulations were added in a new Table (Table 4). The new analyses showed that even though some of the sensitivity simulations performed better for individual chemical and meteorological variables (Table S2), the model's capability in reproducing observed trends analyses is not necessarily improved. The analyses showed that using different emissions, chemical ICONs/BCONs, and meteorology can help to improve individual variable performance; however the base 2006 and 2010 simulations performed best for the trend analyses compared to observations.

The above points have been added in Sections 3.5 and 4.

(2) In the conclusion section, " In general, the model performs well in terms of Corr and NMEs for almost all meteorological and chemical variables in 2006 but not as well in 2010 despite lower NMBs for most variables in 2010, due mainly to inaccuracies in emission estimates and chemical BCONs and ICONs used for 2010 simulations". But the inaccuracies of emission estimates in 2010, comparing with 2006, have not been in-depth explained in the manuscripts, e.g., section 3.2. Please revise.

**Reply:**

The above sentence was concluded from the analyses in Section 4.4 where 2006 emissions and chemical ICONs/BCONs were used for the 2010 simulations and the sensitivity simulation showed improved performances for O<sub>3</sub> and PM<sub>2.5</sub> for 2010. To avoid confusion, we have revised the above sentence in the conclusion to be "due mainly to inconsistencies for emission estimate approaches between 2010 and 2006 and inaccuracies in chemical BCONs and ICONs used for 2010 simulations". We also added the detailed explanation about this point and cited the corresponding reference in Section 2.1 as follows:

"The major sources of uncertainties or errors in the U.S. NEI emissions include: (1) the emissions are calculated using a bottom-up approach based on information provided by individual state, local and tribal air agencies; and (2) improvements in emission-estimation methodology over the years may result in inconsistencies between the NEI data compiled and released by the U.S. EPA (Xing et al., 2013)."

(3) Figure S2, S5, S8-10, S12 are not in good shape. Please revise.

**Reply:**

**Figures S2, S5, S8 – 10, S12 have been revised. There were also problems in the alignment of the figures in the Supplementary material when they were converted to PDF by the journal online software. We will make sure they are in sufficient resolution and quality for the final publication.**

(4) Figure 13 and 14, please add the explanation of each column, e.g., the Run 2- Run 3 depicts the differences resulted by the emission changes between 2010 and 2006.

**Reply:**

**The explanations have been added. An additional Table (Table 3) explaining the set-up of the sensitivity simulations has also been included.**

**The reference cited in this reply:**

**Xing, J., J. Pleim, R. Mathur, G. Pouliot, C. Hogrefe, C.-M. Gan, and C. Wei, 2013, Historical gaseous and primary aerosol emissions in the United States from 1990 to 2010, Atmos. Chem. Phys., 13, 7531–7549.**

### Reply to Comments from Reviewer 3

Geosci. Model Dev. Discuss., 8, C190–C191, 2015

www.geosci-model-dev-discuss.net/8/C190/2015/

© Author(s) 2015. This work is distributed under the Creative Commons Attribute 3.0 License.

Open Access

Geoscientific  
Model Development  
Discussions

**Interactive comment on “Application of WRF/Chem version 3.4.1 over North America under the AQMEII Phase 2: evaluation of 2010 application and responses of air quality and meteorology–chemistry interactions to changes in emissions and meteorology from 2006 to 2010” by K. Yahya et al.**

**Anonymous Referee #3**

Received and published: 19 March 2015

This paper evaluates the WRF/Chem model performance on reproducing the air quality and meteorology-chemical interactions in years 2006 and 2010 by comparing the model predictions of WRF/Chem and WRF, model results with each other and with observations. A series of sensitivity simulations have been conducted to evaluate the model response to changes in emission, meteorology and chemical BC/IC. The authors put a lot of effort to evaluate the model performance for various variables. The manuscript is well written and very informative in terms of statistical evaluation of the model against observations.

#### **Reply:**

**We thank the reviewer for careful review of this manuscript and recognition of the values of this work. We have carefully addressed all the comments raised by the reviewer to improve the technical and presentation quality of our manuscript. Please see below our point-by-point replies.**

However i have a number of major comments : 1) I would expect a lot more information on the model development part considering the journal it is submitted to. However, there are just references to other papers regarding the developments. The study as is more than the application of the model version developed in previous studies by these authors.

#### **Reply:**

**Most of the model developments have already been described in great detail in Wang et al. (2014). Note that Wang et al. (2014) only conducted simulations over a specific short episode (i.e., July 2006). This paper extended the simulation periods to two full years, which has rarely been done by the air quality communities for online-coupled air quality models in the past. The model evaluation, in particular, the evaluation over a long-term period, is considered to be part of the model development and improvement efforts as most**

**papers on model development and improvement limit their simulations to be a short time period. This work aims to examine the capability of WRF/Chem with a new chemistry and aerosol option (i.e., CB05-VBS) for long-term simulations and also the capability in reproducing the trend of air quality and meteorology-chemistry interactions under different emission, meteorological and chemical initial and boundary conditions. A number of model limitations have been identified via a comprehensive evaluation and analyses, which would be particularly useful for model improvement. We therefore believe that our work is a valuable contribution to model development and improvement and it is within the scientific scope of the Journal of GMD, which is supported by the fact that our paper passed the initial assessment by the journal Editor before its acceptance for GMDD. Further, to our understanding, GMD has accepted papers that focus purely on model evaluation in the past, e.g., see a paper by Appel et al. (2013) at <http://www.geosci-model-dev.net/6/883/2013/gmd-6-883-2013.pdf> and a paper by Tessum et al. (2015) at <http://www.geosci-model-dev.net/8/957/2015/gmd-8-957-2015.html>.**

**To address the reviewer's concern, we added a brief summary of the model development in this version of WRF/Chem in the Introduction Section. We also added a few statements regarding how the major findings from this work help shape the model further development and improvement in the future in the conclusion. For example, improvements in predictions of precipitation as well as cloud-aerosol interaction treatments are necessary. Large uncertainties exist in one of the important cloud-aerosol processes, i.e., aerosol activation to form CCN, the ability of the existing aerosol activation scheme in WRF/Chem to reproduce the state of the atmosphere and also inter-annual trends should be improved.**

2) i find it difficult draw robust conclusions on feedback mechanisms using two different years with different emissions and meteorology. This is particularly challenging over temporally and regionally averaged variables.

Regarding the feedbacks, i would expect to see some episodic evaluations where these feedbacks really make a difference and see if the model is capable of simulating these effects.

**Reply:**

**We agree with the reviewer that it is difficult to draw robust conclusions on feedback mechanisms using different emissions and meteorology for two different years. The aerosol-cloud-radiation feedbacks from a single year (2006) comparing WRF and WRF/Chem meteorological and chemical surface and column variables have been discussed in great detail in Yahya et al. (2014). In this study, our focus is on long-term simulations, however, our analyses are carried out on a seasonal basis as we think that each season has similar characteristics in terms of emissions and meteorology and seasonal analyses are sufficient to quantify the feedbacks and their seasonal variations. In addition, we had conducted several sensitivity simulations for January and July 2010 in our original paper to estimate the relative impacts of changes in emissions, meteorology and chemical ICONs/BCONs on model predictions in 2010.**

**Regarding "some episodic evaluations where these feedbacks really make a difference and see if the model is capable of simulating these effects", such evaluations require comparison**

of model predictions from two sets of simulations: one with and one without feedbacks, and evaluate both sets of model predictions against observations. To our understanding, the feedback mechanisms in WRF/Chem are hard-coded and there is no public version of WRF/Chem that does not treat feedbacks. So, it is not possible for us to perform WRF/Chem simulations without the feedbacks. Such episodic evaluations have been indeed performed using another model, i.e., GEM-MACH, and presented in Makar et al. (2014 a, b) as part of the 2014 AQMEII-Phase II special issues in Atmospheric Environment. In their work, GEM-MACH was specially designed to include and exclude feedback treatments to enable such episodic evaluations.

To address the reviewer's comments, we have added further analyses in Section 4.4 and also acknowledged a need to perform such episodic evaluations using WRF/Chem that excludes such feedback mechanisms. In addition, we added a new Table (Table 4) to evaluate how changes in emissions and meteorology in Jan. and Jul 2010 on the model's capability in reproducing the observed variation trends of meteorological and chemical variables, including variables that can indicate the magnitudes of chemical feedbacks such as CCN, AOD, COT, CWP, CF, Precip, and SWDOWN. Those results are representative episodic evaluation (i.e., in Jan. and Jul.) and they can provide information regarding if the changes in emissions and meteorology can affect the observed trends in CCN, AOD, COT, CWP, CF, Precip, and SWDOWN, which can show whether these feedbacks through changes in emissions and meteorology really make a difference in the model's capability in reproducing observed variation trends in those variables. We feel that such additional evaluations, though not exactly what the reviewer expected, can complementarily provide further insights into the impact of feedbacks on model predictions during the two representative periods of simulations.

3) I would expect more in depth discussion on the sensitivity section rather than just showing increase or decrease in the species.

**Reply:**

We have added more in-depth discussion in Section 4.4. For example, we discussed the effect of different emissions and meteorology on O<sub>3</sub> mixing ratios as follows:

“As shown in Figures 13 and 14 (column 2), changes in O<sub>3</sub> are influenced by all factors and the overall change of O<sub>3</sub> mixing ratio is a combination of changes in emissions, meteorological and chemical ICONs/BCONs. The O<sub>3</sub> mixing ratios are greatly increased due to the use of 2010 emissions as compared to 2006 emissions (column 2 in Figure 13), indicating that using a different set of emissions can produce an increase of up to a domain mean of 6 ppb domainwide. Conversely, O<sub>3</sub> mixing ratios are greatly decreased (with a reduction of a domain mean of 6ppb) due to the use of the 2010 chemical ICONs/BCONs compared to 2006 ICONs/BCONs (column 3 in Figure 13). The use of different meteorological ICONs/BCONs also results in varying degrees of changes of O<sub>3</sub> mixing ratios domainwide as O<sub>3</sub> mixing ratios are influenced by photolysis and other meteorological parameters including wind and PBLH (column 4 in Figure 13).”

**In addition, we added a new Table (Table 4) along relevant discussions in Section 4.4 to evaluate if the sensitivity simulations with different meteorology, emissions, and chemical ICs/BCs for Jan. and July 2010 can improve the model's capability in reproducing the trends in both meteorological and chemical variables, as comparing to baseline results in 2006 and 2010.**

**References cited in this review:**

**Appel, K.W., G. A. Pouliot, H. Simon, G. Sarwar, H. O. T. Pye, S. L. Napelenok, F. Akhtar, and S. J. Roselle, 2013, Evaluation of dust and trace metal estimates from the Community Multiscale Air Quality (CMAQ) model version 5.0, *Geosci. Model Dev.*, 6, 883–899.**

**Makar P., W.-M. Gong, J. Milbrandt, C. Hogrefe, Y. Zhang, G. Curci, R. Zabkar, U. Im, A. Balzarini, R. Baro, R. bianconi, P. Cheung, R. Forkel, S. Gravel, M. Hirtl, L. Honzak, A. Hou, P. Jimenez-Guerrero , M. Langer, M. D. Moran, B. Pabla, J.L. Perez, G. Pirovano, R. San Jose, P. Tuccella, J. Werhahn, J.-H. Zhang, and S. Galmarini,, 2014a, Feedbacks between air pollution and weather, Part I: Effects on weather, *Atmos. Environ.*, doi:10.1016/j.atmosenv.2014.12.003.**

**Makar, P., W.-M. Gong, C. Hogrefe, Y. Zhang, G. Curci, R. Zabkar, J. Milbrandt, U. Im, A. Balzarini, R. Baro, R. Bianconi, P. Cheung, R. Forkel, S. Gravel, M. Hirtl, L. Honzak, A. Hou, P. Jimenez-Guerrero, M. Langer, M. Moran, B. Pabla, J.L. Perez, G. Pirovano, R. San Jose, P. Tuccella, J. Werhahn, J.-H. Zhang, and S. Galmarini, 2014b, Feedbacks between air pollution and weather, part 2: Effects on chemistry, *Atmos. Environ.*, doi:10.1016/j.atmosenv.2014.10.021.**

**Tessum, C.W., Hill, J.D., Marshall, J.D., 2015, Twelve-month, 12 km resolution North American WRF-Chem v3.4 air quality simulation: performance evaluation, *Geosci. Model. Dev.*, 8, 957 – 973, doi:10.5194/gmd-8-957-2015.**

**Yahya, K., Wang, K., Gudoshava, M., Glotfelty, T., and Zhang, Y., 2014, Application of WRF/Chem over the continental U.S. under the AQMEII Phase II: Comprehensive Evaluation of 2006 Simulation, *Atmos. Environ.*, doi:10.1016/j.atmosenv.2014.08.063.**

1 **Application of WRF/Chem version 3.4.1 over North America under the AQMEII Phase 2:**  
2 **Evaluation of 2010 Application and Responses of Air Quality and Meteorology-Chemistry**  
3 **Interactions to Changes in Emissions and Meteorology from 2006 to 2010**

4 Khairunnisa Yahya, Kai Wang, and Yang Zhang\*

5 Department of Marine, Earth, and Atmospheric Sciences, NCSU, Raleigh, NC 27695

6 Tadeusz E. Kleindienst

7 National Exposure Research Laboratory, U.S. EPA, Research Triangle Park, NC 27711

8

9 **Abstract**

10 The Weather Research and Forecasting model with Chemistry (WRF/Chem) simulation  
11 with the 2005 Carbon Bond gas-phase mechanism coupled to the Modal for Aerosol Dynamics  
12 for Europe and the Volatility Basis Set approach for Secondary Organic Aerosol (SOA) are  
13 conducted over a domain in North America for 2006 and 2010 as part of the Air Quality Model  
14 Evaluation International Initiative (AQMEII) Phase 2 project. This paper focuses on comparison  
15 of model performance in 2006 and 2010 as well as analysis of the responses of air quality and  
16 meteorology-chemistry interactions to changes in emissions and meteorology from 2006 to 2010.  
17 In general, emissions for gaseous and aerosol species decrease from 2006 to 2010, leading to a  
18 reduction in gaseous and aerosol concentrations and associated changes in radiation and cloud  
19 variables due to various feedback mechanisms. WRF/Chem is able to reproduce most  
20 observations and the observed variation trends from 2006 to 2010, despite its slightly worse  
21 performance than WRF that is likely due to inaccurate chemistry feedbacks resulted from less  
22 accurate emissions and chemical boundary conditions (BCONs) in 2010. Compared to 2006, the  
23 performance for most meteorological variables in 2010 gives lower normalized mean biases but

\*Corresponding author. Mailing address: Campus Box 8208, Room 1125, Jordan Hall, 2800 Faucette Drive Raleigh, NC 27695-8208, USA. Tel: 1-991-515-9688. Fax: 1-919-515-7802. E-mail address: [yang\\_zhang@ncsu.edu](mailto:yang_zhang@ncsu.edu)

24 higher normalized mean errors and lower correlation coefficients. The model also shows worse  
25 performance for most chemical variables in 2010. This could be attributed to underestimations  
26 in emissions of some species such as primary organic aerosol in some areas of the U.S. in 2010,  
27 and inaccurate chemical BCONs and meteorological predictions. The inclusion of chemical  
28 feedbacks in WRF/Chem reduces biases in meteorological predictions in 2010; however, it  
29 increases errors and weakens correlations comparing to WRF simulation. Sensitivity simulations  
30 show that the net changes in meteorological variables from 2006 to 2010 are mostly influenced  
31 by changes in meteorology and those of ozone and fine particulate matter are influenced to a  
32 large extent by emissions and/or chemical BCONs and to a lesser extent by changes in  
33 meteorology. Using a different set of emissions and/or chemical BCONs help improve the  
34 performance of individual variables, although it does not improve the degree of agreement with  
35 observed inter-annual trends. These results indicate a need to further improve the accuracy and  
36 consistency of emissions and chemical BCONs, the representations of SOA and chemistry-  
37 meteorology feedbacks in the online-coupled models.

38 **Keywords:** AQMEII, Emission variation, WRF/Chem, Meteorology-chemistry Interactions,  
39 SOA, Air Quality Trends

40

## 41 **1. Introduction**

42 Changes in meteorology, climate, and emissions affect air quality (e.g., Hogrefe et al.,  
43 2004; Leung and Gustafson, 2005; Zhang et al., 2008; Dawson et al., 2009; Gao et al., 2013;  
44 Penrod et al., 2014). As federal, state, and local environmental protection agencies enforce the  
45 anthropogenic emission control programs, ambient air quality is expected to be continuously  
46 improved. However, such an improvement may be compensated by adverse changes in climatic

47 or meteorological conditions (e.g., increases in near surface temperature, solar radiation, and  
48 atmospheric stability, or reductions in precipitation) that are directly conducive to the formation  
49 and accumulation of air pollutants and that may result in higher biogenic emissions. It is  
50 therefore important to examine changes in ~~both meteorology~~meteorology/climate and emissions  
51 as well as their combined impacts on air quality. The Air Quality Model Evaluation International  
52 Initiative (AQMEII) Phase 2 was launched in 2011 to intercompare online-coupled air quality  
53 models (AQMs) in their capabilities in reproducing atmospheric observations and simulating air  
54 quality and climate interactions in North America (NA) and Europe (EU) (Alapaty et al., 2012).  
55 The simulations over NA and EU with multi-models by a number of participants have been  
56 performed for two years (2006 and 2010) that have distinct meteorological conditions.  
57 Compared with 2006, 2010 is characterized by warmer summer conditions in eastern U.S. and  
58 less precipitation over NA (Stoeckenius et al., 2014; Pouliot et al., 2014). In addition, the  
59 emissions of key pollutants are reduced in 2010 relative to 2006, e.g., emissions of oxides of  
60 nitrogen (NO<sub>x</sub>) and sulfur dioxide (SO<sub>2</sub>) are reduced by 10-30% and 40-80% for many regions in  
61 NA (Pouliot et al., 2014). Comparison of 2010 and 2006 simulations will thus provide an  
62 opportunity to examine the success of the emission control programs and the impacts of  
63 meteorological/climatic variables on air quality. Compared to model intercomparison during  
64 AQMEII Phase 1 (Rao et al., 2012) in which offline-coupled models were used, the use of  
65 online-coupled AQMs models during AQMEII Phase 2 allows for study of the interactions  
66 between meteorology and chemistry through various direct and indirect feedbacks among  
67 aerosols, radiation, clouds, and chemistry (Zhang, 2008; Baklanov et al., 2014). The two year  
68 simulations further enable an examination of the responses of air quality and meteorology-

69 chemistry interactions to changes in emissions and meteorology from 2006 to 2010 that was not  
70 possible with offline-coupled models.

71         Similar to offline AQMs, large uncertainties exist in online-coupled AQMs, which will  
72 affect the model predictions and implications. Such uncertainties lie in the meteorological and  
73 chemical inputs such as emissions, initial and boundary conditions (ICONS and BCONs), model  
74 representations of atmospheric processes, and model configurations for applications such as  
75 horizontal/vertical grid resolutions and nesting techniques. Several studies examined the  
76 uncertainties in emissions (e.g., Reid et al., 2005; Zhang et al., 2014) and BCONs (e.g., Hogrefe  
77 et al., 2004; Schere et al., 2012). There are also uncertainties in various chemical mechanisms  
78 and physical parameterizations used in AQMs such as gas-phase mechanisms (Zhang et al.,  
79 2012), aerosol chemistry and microphysical treatments (Zhang et al., 2010), microphysical  
80 parameterizations (van Lier-Walqui et al., 2014), convective parameterizations (Yang et al.,  
81 2013), boundary layer schemes (Edwards et al., 2006), and land surface models (Jin et al., 2010).  
82 Due to the complex relationships in online-coupled AQMs among the emissions, ICONs and  
83 BCONs, and model processes that may be subject to inherent limitations, it is difficult to isolate  
84 the contributions of model inputs or the representations of atmospheric processes to the model  
85 biases. In mechanistic evaluation (also referred to as dynamic evaluation), sensitivity  
86 simulations are performed by changing one or a few model inputs or process treatments, while  
87 holding others constant. This approach can help diagnose the likely sources of biases in the  
88 model predictions.

89         The Weather Research and Forecasting model with Chemistry (WRF/Chem) version  
90 3.4.1 with the 2005 Carbon Bond (CB05) gas-phase mechanism coupled with the Modal for  
91 Aerosol Dynamics for Europe (MADE) and the Volatility Basis Set (VBS) approach for

92 secondary organic aerosol (SOA) (hereafter WRF/Chem-CB05-MADE/VBS) has been recently  
93 developed by Wang et al. (2014). The WRF/Chem-CB05-MADE/VBS ishas been coupled to the  
94 aqueous-phase chemistry scheme (AQChem) based on the AQChem version in CMAQ v4.75.0  
95 of Sarwar et al. (2011) for .This option considers the aqueous chemistry in both large-scale and  
96 convective clouds (Wang et al., 2014). WRF/Chem-CB05-MADE/VBS also contains  
97 Hheterogeneous chemistry involving sulfur dioxide on the surface of aerosols based on Jacob  
98 (2000) has been incorporated into this version of the model. In addition, the modeland treats both  
99 aerosol direct and indirect effects-by affecting CCN formation (Wang et al., 2014). The  
100 applications of WRF/Chem-CB05-MADE/VBS to 2006 and 2010 in this work use the same  
101 model physical and chemical parameterizations as those of Yahya et al. (2014) but with different  
102 emissions, meteorological ICONs and BCONs, and chemical ICONs and BCONs. The  
103 mechanistic evaluation by comparing WRF/Chem-CB05-MADE/VBS predictions for the two  
104 years would help understand the sensitivity of the model predictions and performance to  
105 different model inputs, and that by comparing WRF/Chem-CB05-MADE/VBS and WRF only  
106 predictions would quantify the impacts of chemistry-meteorology feedbacks on the  
107 meteorological predictions. A comprehensive evaluation of the 2006 simulation has been  
108 presented in Yahya et al. (2014). In this paper, the differences in emissions, meteorological and  
109 chemical ICONs/BCONs, and meteorology between 2010 and 2006 are first examined briefly.  
110 The model performance in 2010 is then evaluated and compared with that in 2006. Finally, the  
111 responses of air quality and meteorology-chemistry interactions to changes in emissions,  
112 chemical ICONs/BCONs, and meteorology individually and collectively from 2006 to 2010 are  
113 analyzed. The main objectives of this paper are to examine whether the model has the ability to  
114 consistently reproduce observations for two separate years, as well as to examine whether the

115 trends in air quality and meteorology-chemistry interactions are consistent for both years.  
116 Stoeckenius et al. (2014) carried out an extensive analysis of the trends in emissions and  
117 observations of meteorological variables, O<sub>3</sub>, SO<sub>2</sub>, and PM<sub>2.5</sub> concentrations between 2006 and  
118 2010. This paper complements the work of Stoeckenius et al. (2014) by examining the changes  
119 in WRF/Chem predictions and chemistry-meteorology feedbacks in 2010 relative to 2006.  
120 Similar evaluations of 2010 and 2006 are performed for the coupled Weather Research and  
121 Forecasting – Community Multiscale Air Quality (WRF-CMAQ) system (Hogrefe et al., 2014).  
122 Unlike the coupled WRF-CMAQ system used in AQMEII Phase 2 that only simulates aerosol  
123 direct effects, WRF/Chem used in this work simulates both aerosol direct and indirect effects. In  
124 addition, the work by Hogrefe et al. (2014) involves nudging of temperature, wind speed, water  
125 vapor mixing ratio, soil temperature and soil moisture, while the model used for this study did  
126 not include any nudging.

## 127 **2. Differences in Emissions and ICONs/BCONs between 2006 and 2010**

### 128 **2.1 Emission Trends**

129 The emission variation trends are examined for major precursors for ozone (O<sub>3</sub>) and  
130 secondary particulate matters (PM) (i.e., sulfur dioxide (SO<sub>2</sub>), oxides of nitrogen (NO<sub>x</sub>),  
131 ammonia (NH<sub>3</sub>), volatile organic compounds (VOCs) including both anthropogenic and biogenic  
132 VOCs) and primary PM species (elemental carbon (EC) and primary organic aerosol or carbon  
133 (POA or POC)). [As shown in Table S21, emissions of most species decrease from 2006 to 2010](#)  
134 [with a domainwide averages of -10% to -24%.](#) Comparing to emissions in 2006, the annual  
135 emissions of SO<sub>2</sub> and NO<sub>x</sub> decrease significantly in 2010, especially at the point sources (Figure  
136 [A4S1](#)), with similar variation patterns in all seasons (Figure not shown). The annual emissions of  
137 NH<sub>3</sub> decrease over most areas but increase in some areas in California (CA) and Midwest.

138 Unlike the changes in the emissions of SO<sub>2</sub> and NO<sub>x</sub>, NH<sub>3</sub> and VOCs emissions exhibit strong  
139 seasonal variations in the emission trends, as shown in Figure A2S2. Although anthropogenic  
140 VOC emissions decrease over continental U.S. (CONUS) for all seasons (Figure not shown), the  
141 VOC emissions increase in the southeast, which is dominated by enhanced biogenic emissions  
142 from vegetation as a response to temperature increases (Stoeckenius et al., 2014). The total  
143 annual emissions of EC and POA also decrease but to a smaller extent over most areas of the  
144 continental U.S. The changes in annual and seasonal emissions of those species between 2010  
145 and 2006 will affect simulated air quality and meteorology-chemistry interactions. In addition,  
146 there exist uncertainties in the NEI emissions. The major sources of uncertainties or errors in the  
147 NEI emissions include: ~~the fact that~~(1) the emissions ~~awere~~ calculated using a bottom-up  
148 approach based on information provided by individual state, local, and tribal air agencies; and (2)  
149 improvements in emission-estimation methodology over the years may result in inconsistencies  
150 between different years of NEI data (Xing et al., 2013). These will affect the accuracy of the  
151 model simulations.

## 153 2.2 Differences in Chemical and Meteorological ICONs/BCONs

154 Large differences exist in the chemical and meteorological ICONs/BCONs used in the  
155 simulations. For example, Stoeckenius et al. (2014) reported that the mid-tropospheric seasonal  
156 mean O<sub>3</sub> mixing ratios are generally lower by several ppbs in 2010 as compared to 2006,  
157 especially during spring and summer. Less Asian mid-tropospheric fine dust was also transported  
158 over to the U.S. in the spring of 2010 and less African dust reached the U.S. in the summer of  
159 2010 (Stoeckenius et al., 2014). As shown in Figure A3S3, significant differences exist for  
160 January, February, and December (JFD) and June, July, August (JJA) 2010 – 2006 in averaged

161 meteorological ICONs and BCONs of skin temperature and soil moisture fraction 100 to 200 cm  
162 below ground extracted from the National Center of Environmental Prediction's (NCEP).

### 163 **3. Model Performance in 2010 and Its Comparison with 2006**

164 Model predictions in 2010 respond to changes in emissions, BCONs, and meteorology.  
165 The model performance for both meteorological and chemical predictions in 2010 is evaluated  
166 and compared with that in 2006. The surface observational networks used to evaluate 2010  
167 results include the Clean Air Status and Trends Network - CASTNET (rural sites), the  
168 Southeastern Aerosol Research and Characterization - SEARCH (southeastern U.S. only, rural  
169 and urban sites), the Speciated Trends Network - STN (urban sites), the Interagency Monitoring  
170 for Protected Visual Environments - IMPROVE (rural sites), the Air Quality System - AQS  
171 (rural and urban sites) and the National Atmospheric Deposition Program - NADP (rural and  
172 urban sites). The satellite data used include the Moderate Resolution Imaging Spectroradiometer  
173 (MODIS) and TERRA. The Global Precipitation Climatology Center (GPCC) for precipitation is  
174 a blend of rain gauge data, satellite data and reanalysis data. Major differences in model  
175 performance between the two years and their associations with changes in emissions, BCONs,  
176 and meteorology are discussed below.

#### 177 **3.1 Differences in Meteorological Predictions for 2006 and 2010**

178 Table 1 shows the annual mean observed and simulated values as well as correlation  
179 coefficients (Corr) between the observed and simulated meteorological variables from the 2010  
180 WRF/Chem and WRF simulations. Similar statistics from the 2006 WRF/Chem and WRF  
181 simulations can be found in Table 1 in Yahya et al. (2014). Figure 1 shows normalized mean  
182 bias (NMB) vs. normalized mean error (NME) plots for several meteorological variables by  
183 seasons against several observational networks for 2006 and 2010. In general, [there are a number](#)

184 of similar trends in terms of meteorological model performances in 2006 and 2010. These  
185 systematic biases give insight into the consistency of the model performance in reproducing  
186 observations. Based on Figure 1, (i)First, for T2, the model tends to perform the worst among all  
187 seasons for JFD for both 2006 and 2010; ~~(ii) and~~ and with the exception of JFD 2006 against  
188 CASTNET and JJA 2010 against CASTNET, the T2 performance falls within an NMB of 0 to ~-  
189 10%, which means a slight underprediction of T2 for all other seasons for both years;.  
190 ~~(iii)Second, for SWDOWN, the largest overprediction occurs for JFD against CASTNET for~~  
191 ~~both 2006 and 2010 as compared to other seasons; (iv) the evaluation against CASTNET gives~~  
192 ~~overpredictions for all seasons for SWDOWN for all seasons for both years with the largest~~  
193 ~~overprediction in JFD; (v) and the model performs well evaluation against SEARCH givewiths~~  
194 ~~very lowsmall positive and negative NMBs for SWDOWN for all seasons both years; andThird,~~  
195 ~~(vi) WS10 is overpredicted for all seasons and for both years against CASTNET and SEARCH.~~  
196 Overall, the correlation coefficients (Corr) for 2006 are better than those of 2010, as the  
197 correlations between mean observed and simulated values for all meteorological variables are  
198 higher for 2006 compared to 2010. However, the biases are smaller for temperature at 2-m (T2)  
199 (against CASTNET), downward shortwave radiation (SWDOWN), wind speed at 10-m (WS10),  
200 precipitation (Precip) (against NADP), cloud fraction (CF), and cloud droplet number  
201 concentrations (CDNC) for 2010 compared to 2006. T2 is underpredicted against CASTNET and  
202 SEARCH for both 2006 and 2010. The seasonal mean NMBs for both 2006 and 2010 (except for  
203 JFD 2006) are < 15%, with annual mean NMBs of -7.7% and -4.9%, respectively. With the  
204 exception of JFD 2006 against CASTNET, T2 predictions in the other seasons in 2006 for both  
205 CASTNET and SEARCH have lower NMEs (< 25%) for 2006. All the seasons in 2010 have an  
206 NME of > 25% for T2 predictions. For SWDOWN, for both 2006 and 2010, seasonal NMBs

207 range from -10% to 20% with annual mean NMBs of 21.3% and 7.4%, respectively, against  
208 CASTNET and 3.0% and 12.4%, respectively, against SEARCH; however the seasonal and  
209 annual mean NMEs in 2006 are < 40% while those in 2010 range from 40% to 65%. Although  
210 SWDOWN is overpredicted on an annual basis, T2 is underpredicted in all seasons in 2006 and  
211 all seasons except for JJA in 2010, as T2 is diagnosed from the skin temperature, which depends  
212 on not only SWDOWN but also other variables such as soil properties. The NCEP, Oregon State  
213 University, Air Force, National Weather Service Office of Hydrology (NOAH) land surface  
214 model used in this case calculates the heat fluxes and skin temperatures based on SWDOWN, the  
215 land-use type, and soil properties including soil texture, soil moisture, soil conductivity and  
216 thermal diffusivity which vary for different soil types (Chen, 2007). Pleim and Gilliam (2009)  
217 also reported tThe cold bias for T2 especially for the winter of 2006 is also reported in Pleim and  
218 Gilliam (2009) usingfor their WRF simulations-WRF. However in Pleim and Gilliam (2009),  
219 which was reduced by implementing deep soil temperature and moisture nudging in their worka  
220 soil data assimilation method is introduced to reduce the cold biases due to the deep soil  
221 temperature and moisture. In this study, however, deep soil data assimilationnudging was not  
222 used. Annual mean WS10 is overpredicted for both 2006 and 2010 (with NMBs of 17.4-27.4%  
223 in 2006 and 8-27.5% in 2010) but to a much smaller extent compared to previous studies.  
224 Seasonal WS10 is overpredicted for 2006 but underpredicted for 2010 with better performance in  
225 2010 (i.e., smaller NMBs in 2010 and comparable NMEs between the two years). This is  
226 because In this study, the Mass and Owens (2010) surface roughness parameterization is used in  
227 this work in WRF and WRF/Chem, which helps reduce typical overpredictions in WS10 overall  
228 in both years. However, Mass and Owens (2010) also noted that by using this parameterization,  
229 the high wind speeds are affected and suggested switching off this drag parameterization at

230 ~~higher wind speeds.~~ SWDOWN tends to be overpredicted for CASTNET due to  
231 underpredictions in cloud variables which will be covered in Section 3.4. CF is the only  
232 meteorological variable with a better performance in terms of all three measures including Corr,  
233 NMB, and NME in 2010 than in 2006 against MODIS. The better performance in CF in 2010  
234 may help reduce annual mean NMBs in CDNC, SWDOWN, and T2 in 2010, although their  
235 annual mean NMEs increase and annual mean Corr values decrease.

236 For Precipitation, the model performs consistently well against GPCC for both years with  
237 seasonal NMBs within -11% and -12%, and annual NMBs of 0.3% and 1.3%, respectively, for  
238 2006 and 2010. The evaluation against NADP shows larger differences with NMBs of 22.2%  
239 and 2.5% and Corr values of 0.43 and 0.1 for 2006 and 2010, respectively. As compared to other  
240 meteorological variables such as T2, SWDOWN, and WS10, the meteorological performance for  
241 precipitation do not follow a clear trend for all seasons or years against NADP and GPCC. For  
242 example, JJA-precipitation in JJA is underpredicted against NADP and GPCC for 2010 but this  
243 is not the case for 2006. In general, ~~precipitation-the reported biases in precipitation simulated~~  
244 ~~by~~for WRF from literature are significant. For example, Wang and Kotamarthi (2014) ~~conducted~~  
245 ~~extensive research to study~~ied the precipitation behavior in WRF and showed that even with  
246 ~~nudging, the precipitation biases existed~~remained up to a root mean square error (RMSE) of  
247 ~~62.5%~~ due to inherent weaknesses in the microphysics and cumulus parameterization schemes.  
248 Similarly, ~~the WRF/Chem model is also unable to reproduce~~gives large seasonal mean biases (up  
249 to 44% in 2006 and up to -26% in 2010) for ~~the simulated precipitation results accurately~~ for  
250 most seasons in 2006 or 2010, although the annual mean biases are small to moderate (with  
251 NMBs of -2.2% to -1.3% to against GPCC and 9.7-17.6% to against NADP in both years). CF is  
252 ~~the only meteorological variable with a better performance in terms of all three measures~~

253 ~~including Corr, NMB, and NME in 2010 than in 2006 against MODIS. The better performance~~  
254 ~~in CF in 2010 may help reduce annual mean NMBs in CDNC, SWDOWN, and T2 in 2010,~~  
255 ~~although their annual mean NMEs increase and annual mean Corr values decrease.~~

256       Yahya et al. (2014) compared and evaluated the full-year WRF and WRF/Chem 2006  
257 simulations with the same physical configurations to analyze the effects of feedbacks from  
258 chemistry to meteorology. The results for 2006 show that for the evaluation of SWDOWN, T2,  
259 and WS10 against CASTNET and SEARCH, the Corr is almost identical for both WRF/Chem  
260 and WRF simulations. For evaluation of precipitation against NADP, WRF has a higher Corr  
261 compared to WRF/Chem. Unlike 2006, the 2010 WRF only simulation has higher Corr for all  
262 meteorological variables compared to the 2010 WRF/Chem simulation except for Precip against  
263 GPCC and CF against MODIS. This means that the emissions and chemistry-meteorological  
264 feedbacks play an important role in influencing model performance. Section 4.4 will explore this  
265 in further detail. Another obvious difference is that the NMBs for the meteorological variables  
266 for 2010 are smaller compared to 2006 for all the variables except for Precip against GPCC,  
267 while the NMEs are larger for 2010 compared to 2006 for all variables except for Precip against  
268 GPCC. A smaller overall averaged NMB but a larger NME may indicate compensation of over-  
269 and under-predictions leading to a small bias, but the magnitude of the differences are reflected  
270 in the NME values.

271       The same model physics and dynamics options are used for both years. In addition to  
272 different emissions, there are characteristic climate differences between the two years that lead to  
273 lower Corr and larger NMEs for most meteorological fields in 2010 compared to 2006 for both  
274 WRF and WRF/Chem simulations. 2010 is reported to be the warmest year globally since 1895  
275 according the National Climactic Data Center (NCDC) (<http://www.ncdc.noaa.gov/cag/>). Even

276 though 2010 has high temperatures compared to previous years, a trend analysis of extreme heat  
277 events (EHE) from 1930 to 2010 showed that in 2010, there were more than 35 extreme  
278 minimum heat events (where temperatures are extremely low) over southeastern U.S. compared  
279 to about ~10 events in 2006. In fact, the number of extreme minimum heat events is the highest  
280 overall for CONUS in 2010 compared to all the other years from 1930 onwards (Oswald and  
281 Rood, 2014). The Intergovernmental Panel for Climate Change (IPCC) reported that since 1950,  
282 weather events have become more extreme likely due to climate change (IPCC, 2012).  
283 Grundstein and Dowd (2011) stated that on average, by 2010 there would be 12 more days with  
284 extreme apparent temperatures than those in 1949. These studies imply that increased  
285 temperatures change the weather in unexpected ways with uncertainties in the state of science  
286 (Huber and Gullede, 2011), including models. These high and low temperatures could  
287 contribute to the compensation of over- and under-predictions leading to smaller NMBs in  
288 general for 2010. To better simulate model extreme heat events, Meir et al. (2013) suggested  
289 using a higher spatial resolution with a grid size of 12-km or smaller, better sea surface  
290 temperature estimates, and enhanced urbanization parameterization. Gao et al. (2012) reported  
291 better results in reproducing extreme weather events with WRF over eastern U.S. at a 4-km  $\times$  4-  
292 km resolution. In this study, although the urban canopy model is used for both WRF and  
293 WRF/Chem simulations, a 36-km  $\times$  36-km grid resolution might not be sufficient to reproduce  
294 the extreme temperature events (highs and lows) in 2010.

295 | As shown in ~~Figure A~~[Figure S4](#), the spatial distribution of MB values for T2 for JFD  
296 2010 by WRF/Chem show very large negative MBs over southeastern U.S. compared to JFD  
297 2006. T2 is also generally underpredicted over southeastern U.S. in both years, but with larger  
298 negative biases in 2010 than those in 2006. T2 biases also seem to be more extreme for JFD

299 2010 compared to JFD 2006, with dark red and dark blue colors for the MB markers, indicating  
300 large positive and large negative biases, respectively. This could explain the poorer correlation  
301 for T2 in 2010 compared to 2006 as shown in Table 1. On the other hand, the performances of  
302 T2 for JJA 2010 and 2006 are very similar, with MBs  $\sim -0.1$  to  $0.1$  °C in eastern U.S., large  
303 negative MBs at the sites in Montana and Colorado, and a large positive MB at the site in  
304 Wyoming.

### 305 **3.2 Differences in Chemical Predictions for 2006 and 2010**

306 The chemical performance between 2006 and 2010 is more variable compared to the  
307 meteorological performance of surface variables. The lower Corr for 2010 compared to 2006 for  
308 meteorological variables has a large influence on the model performance for 2010. As shown in  
309 Table 1, all the chemical variables for all networks have lower Corr in 2010 compared to 2006.  
310 As shown in Figures 2 and 3, maximum 8-hr O<sub>3</sub> concentrations are underpredicted to a larger  
311 extent in 2010 compared to 2006, dominating the O<sub>3</sub> annual performance in 2010. These results  
312 are consistent with the results of Hogrefe et al. (2014). The large underpredictions of maximum  
313 8-hr O<sub>3</sub> in JFD 2010 over southeastern U.S. are attributed to larger cold biases in T2 shown in  
314 Figure AFigure S4 and reduced NO<sub>x</sub> and VOC emissions in 2010 relative to their levels in 2006.  
315 While reduced NO<sub>x</sub> levels can result in an increase in nighttime O<sub>3</sub> concentrations due to reduced  
316 NO<sub>x</sub> titration of O<sub>3</sub>, the impact of reduced NO<sub>x</sub> titration on the maximum 8-hr O<sub>3</sub> is small. As  
317 shown in Figure AFigure S4, the temperature biases for both years are relatively similar. Over  
318 northeastern U.S., the T2 bias is generally less than  $-0.1$  °C for JJA in both years. However, as  
319 shown in Figure 2, O<sub>3</sub> concentrations over northeastern U.S. in JJA 2010 have negative biases  
320 whereas those over northeastern U.S. in JJA 2006 have positive biases. In this case, emissions  
321 might play a significant role in the underprediction of O<sub>3</sub> concentrations over northeastern U.S.

322 in JJA 2010. Hourly average surface NO<sub>x</sub> emissions decrease significantly over northeastern  
323 U.S. in JJA from 2006 to 2010. As shown in Figure 3, 2006 model performance for O<sub>3</sub> is  
324 generally good for all seasons and all networks.

325 According to Table 1 and Figure 1, WRF/Chem predicts SWDOWN to a lower extent in  
326 2010 compared to 2006 against CASTNET. Khiem et al. (2010) reported that during the  
327 summer, a large percentage of the variations in peak O<sub>3</sub> concentrations during the summer can be  
328 attributed to changes in seasonally averaged daily maximum temperature and seasonally  
329 averaged WS10. Simulated WS10 is lower for 2010 compared to 2006 in general; therefore,  
330 WS10 does not seem to contribute to reduced O<sub>3</sub> concentrations (through dispersion, increased  
331 dry deposition) in 2010. Figure 4 shows diurnal variations of observed and simulated

332 WRF/Chem T2 and O<sub>3</sub> concentrations from CASTNET in JJA 2006 and 2010. The diurnal  
333 averaging provides insight whether the underpredictions of O<sub>3</sub> mixing ratios is a systematic bias;  
334 i.e. during the daytime and/or nighttime or both. -The diurnally averaged observed temperatures

335 show a similar trend in JJA 2006 to 2010 against T2 measurements from CASTNET. This shows  
336 that the model is able to reproduce T2 for different years. The temperature trends also correlate  
337 strongly with the O<sub>3</sub> trends. At night, where the model has cold bias, O<sub>3</sub> concentrations are  
338 underpredicted to a larger extent. The O<sub>3</sub> concentrations show a larger underprediction for JJA  
339 2010 compared to JJA 2006. The underpredictions in O<sub>3</sub> in both 2006 and 2010 can be explained  
340 by several reasons. For example, Im et al. (2014) showed that MACC underpredicts O<sub>3</sub> mixing  
341 ratios, particularly in winter and spring during both day and night and in summer and fall during  
342 nighttime. As indicated by Wang et al. (2014) and Makar et al. (2014), the inclusion of aerosol  
343 indirect effects also tends to reduce O<sub>3</sub> mixing ratios, comparing to the models that simulate

Formatted: Subscript

344 aerosol direct effect only or do not simulate aerosol direct and indirect effects (i.e., offline-  
345 coupled models).

346 Figure 5 shows spatial distribution of NMBs for  $PM_{2.5}$  concentrations for JFD and JJA  
347 2006 and 2010 against IMPROVE, STN, and SEARCH. Overall, JJA 2006 and JJA 2010 have  
348 similar spatial distribution patterns of NMBs for all sites over CONUS except for several sites in  
349 northwestern U.S. where  $PM_{2.5}$  concentrations are underpredicted for JJA 2010 but overpredicted  
350 for JJA 2006. However, many sites have positive NMBs over eastern and central U.S. for JFD  
351 2006, whereas more sites have negative NMBs over eastern and central U.S. for JFD 2010.  
352 Statistics from Yahya et al. (2014) and Table 1 show that in general, the simulated  
353 concentrations of  $PM_{2.5}$  and all  $PM_{2.5}$  species decrease from 2006 to 2010, however, the Corr  
354 values for  $PM_{2.5}$  and  $PM_{2.5}$  species become worse in 2010 compared to 2006. As shown in Figure  
355 6,  $PM_{2.5}$  concentrations for 2006 can be overpredicted or underpredicted, depending on seasons  
356 and networks, with an equal distribution of positive and negative NMBs. However for 2010,  
357  $PM_{2.5}$  concentrations tend to be underpredicted for all seasons and for all networks except for  
358 JFD against SEARCH. As shown in Figure 7, NMBs for  $PM_{2.5}$  species for 2006 at individual  
359 monitoring sites range from -40% to 60%, while those for 2010 range from -80% to 80%. The  
360 markers are more spread out covering a wider range of NMBs and NMEs for 2010 with more  
361 extremes as compared to the markers for 2006 clustered around the zero NMB line. NMEs for  
362  $PM_{2.5}$  species in 2006 remain below 100%.  $NO_3^-$  concentrations are slightly underpredicted in  
363 2006 against all networks; however,  $NO_3^-$  levels in 2010 are largely underpredicted, likely due to  
364 the large decrease in  $NO_x$  emissions from 2006 to 2010 and the increase in T2. The NMBs for  
365 IMPROVE and SEARCH OC remain low from 2006 to [2010,2010](#); however, the NMEs increase  
366 significantly. For TC against IMPROVE, the NMB and NME in 2010 are larger in magnitudes in

367 2010 than those in 2006.  $\text{SO}_4^{2-}$  has lower NMBs but higher NMEs for all networks in 2010  
368 compared to 2006. EC concentrations are generally overpredicted in 2006 for all networks but  
369 underpredicted against SEARCH and largely overpredicted against IMPROVE in 2010.  $\text{NH}_4^+$   
370 also has higher NMEs in 2010 compared to 2006. Overall, the evaluation in 2010 shows larger  
371 NMEs and ~~poor~~ weaker correlations for all  $\text{PM}_{2.5}$  species compared to 2006.

372 Figure 8 shows the time series plots for 24-hr average concentrations of  $\text{PM}_{2.5}$ ,  $\text{SO}_4^{2-}$  and  
373  $\text{NO}_3^-$  against STN for 2006 and 2010. In 2006, the daily-average PM data were collected on a  
374 daily basis in 2006 but every 3 days in 2010. The model is able to predict most of the observed  
375 peaks and troughs for 2006 even though the observed and simulated magnitudes are significantly  
376 different for several days. For 2010, the model does not show large spikes and can reproduce the  
377 magnitudes well, although it does not predict the peaks and troughs as well as 2006 for some  
378 months (e.g., Jan-March and July-Sept. for  $\text{PM}_{2.5}$ ). This could be attributed in part to the ~~poor~~  
379 weaker correlations of meteorological variables in 2010 compared to 2006. For example,  
380 ~~poor~~ inaccurate predictions of WS10 can influence the transport and dry deposition of aerosols.  
381 ~~An Poor overpredictions~~ of precipitation ~~can impact~~ increases the wet deposition of aerosols.  
382 Poor predictions of T2 can influence the planetary boundary layer height (PBLH) and both can  
383 also affect the distribution of aerosol concentrations.  $\text{NO}_3^-$  concentrations for the winter months  
384 are moderately underpredicted in 2006 but largely underpredicted in 2010, likely due to ~~the~~  
385 underpredictions in nitrogen dioxide ( $\text{NO}_2$ ) concentrations (Yahya et al., 2014). Section 4 will  
386 discuss in further detail the role of emissions, meteorology and chemical ICONs/BCONs on  $\text{O}_3$   
387 and  $\text{PM}_{2.5}$  concentrations.

### 388 3.3 SOA Evaluation for 2006 and 2010

Formatted: Subscript

Formatted: Subscript

389           The VBS framework in WRF/Chem of Ahmadov et al. (2012) provides a more realistic  
390 treatment of SOA compared to previous SOA treatments such as the 2-product model by Odum  
391 et al. (1996) used in the Secondary Organic Aerosol Model (SORGAM) of Schell et al. (2001).  
392 Wang et al. (2014) evaluated SOA and OC concentrations simulated from WRF/Chem-CB05-  
393 MADE/VBS and WRF/Chem-CB05-MADE/SORGAM over NA for July 2006 against field  
394 campaign data from Offenberg et al. (2011) at the Research Triangle Park (RTP), NC for July  
395 2006. They showed significant improvement in simulating SOA and total organic aerosol (TOA)  
396 by VBS than by SORGAM. In this study, SOA and OC predictions are evaluated against  
397 available field campaign data at RTP, NC in eastern U.S. from Offenberg et al. (2011) for 2006  
398 only, and Pasadena, CA and Bakersfield, CA in western U.S. from ~~Klendienst et al. (2012)~~ and  
399 Lewandowski et al. (2013) for 2010 only (note that no observations are available at the same  
400 sites for both years). The RTP site is located in a semi-rural area. Pasadena, CA is located about  
401 11 miles from downtown Los Angeles (LA), and Bakersfield, CA is located about ~100 miles  
402 from downtown LA. Both sites are classified as urban/industrial sites. OC concentrations were  
403 measured using an automated, semicontinuous elemental carbon-organic carbon (EC-OC)  
404 instrument. The observed SOA masses were determined from organic tracers extracted from  
405 filter samples (Lewandowski et al., 2013). Simulated OC concentration is calculated by  
406 summing up SOA and POA, and dividing the total OA by 1.4 (Aitken et al., 2008).

407           As shown in Figures 9 and [A5S5](#), the model overpredicts SOA but underpredicts OC at  
408 RTP in 2006, because (1) the SOA formed from alkanes and alkenes is excluded in the  
409 observations from RTP but simulated in WRF/Chem, and (2) WRF/Chem may have  
410 overestimated the aging rate coefficient for both anthropogenic and biogenic surrogate VOC  
411 precursors (Wang et al. (2014)). The SOA overprediction due to those reasons compensates the

412 underprediction in SOA due to omission of SOA from POA, leading to a net SOA overprediction  
413 at RTP in 2006. By contrast, the VBS underpredicts SOA in 2010 with NMBs of -55.3% and -  
414 75.3% at Bakersfield and Pasadena, respectively, which is mainly due to the omission of SOA  
415 formation from POA in the current VBS-SOA module in this version of WRF/Chem. As shown  
416 in [Figure AFigure S6](#), SOA to OC ratios at RTP in 2006 are in the range of 50-80%, whereas  
417 they are < 20% at Bakersfield, CA and < 40% Pasadena, CA in 2010. This indicates that  
418 neglecting SOA formation from POA would have much larger impact on SOA predictions -at the  
419 two CA sites in 2010 than at RTP in 2006, due to the dominance of POA in TOA at the two CA  
420 sites. As shown in Figure 9, the model underpredicts OC at RTP in 2006 and significantly  
421 underpredicts OC at the two sites in CA in 2010. The differences in OC performance in both  
422 years are caused by different locations (i.e., RTP in 2006 and the two CA sites in 2010) that have  
423 different ratios of POC to OC as mentioned previously. OC performance thus largely depends  
424 on SOA performance at RTP but on POA performance at the two sites in CA. This is why the  
425 OC performance remains poor despite a relatively good performance in SOA at the two sites in  
426 CA. Worse OC performance over the two CA sites in 2010 may also indicate potentially large  
427 underestimation of POA emissions over the western U.S.

### 428 **3.4 Differences in Aerosol-Cloud Predictions for 2006 and 2010**

429 Figure 10 shows NMBs vs. NMEs of several aerosol and cloud variables for JFD and JJA  
430 in 2006 and 2010 against satellite data. Table 1 lists the corresponding annual performance  
431 statistics for 2010. [The model is able to reproduce generally similar performances against](#)  
432 [observations for most of the aerosol-cloud variables for both 2006 and 2010 as the trends of](#)  
433 NMBs and NMEs are quite similar for both seasons in both years. For JJA 2006 and 2010, all  
434 cloud variables are underpredicted [with approximately the same magnitudes of NMBs and](#)

435 | [NMEs](#). For JJA, the model performs better for 2010 for CF, aerosol optical depth (AOD), and  
436 | cloud optical thickness (COT) in terms of seasonal mean spatial distribution. For JFD, the model  
437 | performs better for CF and cloud water path (CWP) in 2010. In terms of annual statistics,  
438 | compared to 2006, 2010 has lower NMBs for CF and COT but larger biases in AOD, CWP, and  
439 | cloud condensation nuclei (CCN), leading to large differences in aerosol-radiation and cloud –  
440 | radiation feedbacks, which in turn affect the performance of meteorological and chemical  
441 | predictions. Despite the ~~general worse~~[differences in model](#) performance of meteorological and  
442 | chemical variables in 2010 compared to 2006, performance of cloud variables do not vary  
443 | significantly. One possible reason is because the evaluation of aerosol-cloud variables is based  
444 | on monthly values that are averaged out on a seasonal basis. The meteorological and chemical  
445 | variables shown earlier are evaluated based on site-specific, and hourly, daily, or weekly data.

### 446 | **3.5 Differences in Observed and Simulated Trends between 2010 and 2006**

447 | Table 2 shows the percentage changes in observed and WRF only and WRF/Chem  
448 | simulated variables between 2010 and 2006. [Overall, the model is able to predict the trends in all](#)  
449 | ~~the listed~~[major meteorological, chemical, and aerosol-cloud-radiation variables between 2006](#)  
450 | [and 2010 with thea few exceptions of \(e.g., WS10 against CASTNET, Precip, CF, maximum 8-hr](#)  
451 | [O<sub>3</sub> against CASTNET, and 24-hr EC against IMPROVE\).](#) The trends in simulated T2,  
452 | SWDOWN, and SEARCH WS10 are generally consistent with the observed trends from 2006 to  
453 | 2010. Both observed and simulated temperatures at 2-m (T2) at the CASTNET sites increase by  
454 | ~4 °C or ~35 to 40% from 2006 to 2010. For downward shortwave radiation (SWDOWN), both  
455 | observed and simulated values at the CASTNET and SEARCH sites increase by ~1 to 3% and by  
456 | ~5 to 7%, respectively, from 2006 to 2010. The observed WS10 remains relatively constant at  
457 | CASTNET in both years. The simulated WS10 by WRF also shows no change but that by

Formatted: Subscript

458 | WRF/Chem shows a small decrease (by -8.3%) for the CASTNET sites. Comparing to a  
459 | SEARCH observed change of ~-4% in WS10, WRF and WRF/Chem predict a larger decrease  
460 | from 2006 to 2010 (~-12 to -13%). The trends for Precip and CF for simulated variables are not  
461 | consistent with observed trends from 2006 to 2010. Observed NADP Precip increased slightly  
462 | from 2006 to 2010 by ~7%, however both simulated WRF and WRF/Chem show a small  
463 | decrease from 2006 to 2010. Observed mean GPCP Precip remained relatively constant from  
464 | 2006 to 2010, however, WRF only shows a slight increase (~4%) while WRF/Chem shows a  
465 | larger decrease (-12%) from 2006 to 2010. MODIS CF decreased by -0.2% from 2006 to 2010  
466 | whereas both WRF and WRF/Chem show small increases ~3-4% from 2006 to 2010. Apart from  
467 | the large biases in the evaluation of precipitation, the decrease in precipitation is likely due to the  
468 | smaller decrease in SWDOWN for WRF/Chem compared to observations between 2006 and  
469 | 2010. This would result in less convective precipitation during the summer but increased CF for  
470 | 2010. In addition, PM<sub>2.5</sub> is underpredicted in 2010 but has a better agreement with  
471 | observed PM<sub>2.5</sub> in 2010 than in 2006. Underpredicted PM<sub>2.5</sub> concentrations will also affect the  
472 | formation of clouds and precipitation via various direct and indirect effects.

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

473 | The simulated decreasing trends between 2006 and 2010 are overall consistent with the  
474 | observed decreasing trend between 2006 and 2010 for all species except for maximum 8-hr O<sub>3</sub>  
475 | concentrations from CASTNET and EC from IMPROVE. CASTNET maximum 1-hr and 8-hr  
476 | O<sub>3</sub> concentrations change very little from 2006 to 2010 whereas WRF/Chem shows a moderate  
477 | decrease of 14-15%. The large decrease in simulated O<sub>3</sub> mixing ratios in 2010 can be attributed  
478 | to a large decrease in O<sub>3</sub> mixing ratios from the ICONs and BCONs (Stoekenius et al., 2014).

Formatted: Subscript

Formatted: Subscript

479 | The IMPROVE observed EC concentrations decreased by ~22% from 2006 to 2010, however,  
480 | WRF/Chem shows a small increase (by ~2%). For PM<sub>2.5</sub> concentrations, the simulated decrease

481 from 2006 to 2010 by WRF/Chem is larger than the observed decrease for both STN and  
482 IMPROVE. Similar steeper decreases by WRF/Chem also occur for  $\text{SO}_4^{2-}$  against STN,  $\text{NO}_3^-$   
483 against IMPROVE, TC against STN, and OC against IMPROVE: [likely due to the influence of](#)  
484 [ICONS/BCONs and emissions.](#)

#### 485 **4. Responses of 2010 Predictions to Changes in Emissions and Meteorology**

486 The changes in emissions, boundary conditions, and meteorology between 2010 and 2006  
487 lead to changes in simulated air quality and the chemistry-meteorology feedbacks, which in turn  
488 change meteorological and air quality predictions during the next time step.

#### 489 **4.1 Air Quality Predictions**

490 Simulated air quality responds nonlinearly to the changes in emissions. Figures 11,  
491 [A7S7- A9-S9](#) show the seasonal changes between 2010 and 2006 in ambient mixing ratios of  
492 gases ( $\text{SO}_2$ ,  $\text{NO}_2$ ,  $\text{NH}_3$ ,  $\text{O}_3$ , and hydroxyl - OH) and concentrations of PM species ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  
493  $\text{NH}_4^+$ , organic matter or OM, EC, POA, anthropogenic SOA or ASOA, biogenic SOA or BSOA,  
494 and  $\text{PM}_{2.5}$ ).  $\text{SO}_2$  and  $\text{NO}_2$  concentrations tend to decrease for all seasons at most locations over  
495 CONUS due to the decrease in their emissions. The increases in  $\text{NO}_2$  concentrations over urban  
496 areas in eastern U.S. in March, April, May (MAM) in 2010 relative to 2006 could be due to a  
497 few reasons including decreased photolytic conversion from  $\text{NO}_2$  to NO due to a decrease in  
498 SWDOWN and less  $\text{NO}_2$  conversion to nitric acid ( $\text{HNO}_3$ ) due to decreased OH concentrations.  
499 The  $\text{NO}_2$  hot spots also correlate to the decrease in hourly  $\text{O}_3$  concentrations in urban areas. This  
500 could indicate an increased titration of nighttime  $\text{O}_3$  by NO. This is an important result for policy  
501 implications, as reducing  $\text{NO}_x$  emissions may reduce  $\text{NO}_2$  concentrations overall for CONUS,  
502 but may not reduce  $\text{NO}_2$  concentrations in several areas, especially in urban areas due to a  
503 combination of titration and complex interplay with local meteorology.  $\text{NH}_3$  mixing ratios

504 generally decrease in the U.S., except over eastern U.S. in MAM and September, October, and  
505 November (SON), where there are increases.  $\text{NH}_3$  emissions decrease, however, over eastern  
506 U.S. in all seasons. The increase in  $\text{NH}_3$  concentrations in MAM and SON could be attributed to  
507 a number of reasons including less  $\text{NH}_3$  conversion to  $\text{NH}_4^+$  to neutralize  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  and less  
508 dispersion of  $\text{NH}_3$  concentrations due to decreased wind speeds over eastern and southeastern  
509 U.S. in MAM and SON, respectively, in 2010 compared to 2006. In JJA and SON, high OM  
510 concentrations in Canada are attributed to the enhanced impacts of BCONs by increasingly  
511 convergent flow in this region. OM is made up of both POA and SOA. An increase in VOC  
512 emissions in eastern U.S. in MAM and SON leads to increases in OM concentrations. Decreases  
513 in VOC emissions in western U.S. for all seasons lead to decreases in OM concentrations. The  
514 OM concentrations in some areas, however, do not follow a linear relationship with VOC  
515 emissions, such as southeastern U.S. in JJA, where VOC emissions increase from 2006 to 2010  
516 but OM concentrations decrease. A decrease in POA concentrations must dominate the overall  
517 decrease in OM concentrations, even under increased temperatures and biogenic VOC emissions  
518 in this area.  $\text{PM}_{2.5}$  concentrations decrease for all seasons and most regions of the CONUS,  
519 which is attributed mainly to decreases in precursor gases, especially the inorganic precursors  
520  $\text{SO}_2$  and  $\text{NO}_x$  in eastern U.S. Increased  $\text{PM}_{2.5}$  concentrations in JFD and MAM in the Midwest  
521 are due to surface temperature decreases, dominating in this region (Stoeckenius et al., 2014).  
522 This in turn leads to increased particle nitrate concentrations (Campbell et al., 2014).

#### 523 **4.2 Meteorological Predictions**

524 | [Figure AFigure S10](#) compares the seasonal changes between 2010 and 2006 in several  
525 meteorological variables that affect air pollution including SWDOWN, T2, WS10, PBLH, and  
526 Precip simulated by WRF only simulations without considering chemistry feedbacks. Large

527 changes occur in those variables between the two years, e.g., 10-50 W m<sup>-2</sup> increases in  
528 SWDOWN in western and Midwest in JJA, generally warmer in JJA and SON over most areas  
529 but cooler by 3-10 °C in eastern U.S. in JFD, and with reduced Precip in eastern or southeastern  
530 U.S. in JJA and SON but increased Precip in northwestern U.S. in MAM and JJA and in western  
531 U.S. in JFD. ICONs and BCONs for skin temperatures shown in ~~Figure A~~[Figure S3](#) greatly  
532 influence T2 shown in ~~Figure A~~[Figure S10](#) for JFD and JJA.

533         Figures 12 and ~~A4-S11~~[A4-S11](#) show the seasonal changes between 2010 and 2006 in several  
534 meteorological and cloud variables SWDOWN, T2, WS10, Precip, PBLH, AOD, COT, CF,  
535 CWP, and CDNC) for WRF/Chem that accounts for meteorology-chemistry feedbacks. The  
536 relationships between various meteorological variables have been discussed in Yahya et al.  
537 (2014). Comparing to the differences in predictions of SWDOWN, T2, WS10, Precip, and PBLH  
538 between 2010 and 2006 WRF only simulation shown in ~~Figure A~~[Figure S10](#) and WRF/Chem  
539 ~~simulations—shows~~[simulations shown](#) in Figures 12 and ~~A4-S11~~[A4-S11](#), the differences in those  
540 meteorological variables ~~except for SWDOWN~~ do not vary significantly [in terms of trends of](#)  
541 [average seasonal spatial distributions](#) between 2010 and 2006 WRF simulations and between  
542 2010 and 2006 WRF/Chem simulations. [However, there are differences in magnitudes,](#)  
543 [especially for SWDOWN. SWDOWN is affected most by the addition of chemistry in](#)  
544 [WRF/Chem as compared to WRF, especially for JFD through indirect feedback of clouds on](#)  
545 [radiation.](#) As shown in Figure 12, the decrease in SWDOWN from 2006 to 2010 is larger over  
546 north-central and north-western U.S. and the increase in SWDOWN is smaller over north-eastern  
547 and southwestern U.S. for MAM (WRF/Chem) compared to MAM (WRF). For SON, the  
548 increase in SWDOWN from 2006 to 2010 simulated by WRF/Chem is larger over eastern U.S.  
549 than that by WRF. The differences between WRF and WRF/Chem are the largest for SWDOWN

550 over northeastern U.S. in JFD with an increase in SWDOWN simulated by WRF but a decrease  
551 simulated by WRF/Chem from 2006 to 2010. The differences in SWDOWN are likely due to the  
552 differences in CF between the two sets of simulation pairs, as the spatial distribution for CF is  
553 consistent with that of SWDOWN. As expected, there are slight differences between T2 and  
554 PBLH between WRF and WRF/Chem (2010 – 2006) due to changes in radiation. There are also  
555 negligible small differences between precipitation between WRF and WRF/Chem. The aerosol-  
556 cloud-radiation feedbacks due to the differences between a single year WRF and WRF/Chem for  
557 2010 will be discussed in Section 4.3.

558 The increase in SWDOWN from 2006 to 2010 does not necessarily translate to an  
559 increase in T2. However, in general, increases in SWDOWN lead to increase in T2, as shown in  
560 SON in Figure 12, where SWDOWN generally increases over most of the continental U.S., T2  
561 also increases over most of CONUS. In general, the largest differences in T2 between 2006 and  
562 2010 occur in SON (increase) and JFD (decrease). The decrease in T2 in JFD in north-central  
563 U.S. and parts of Canada is significant as it results in a decrease in WS10 and PBLH. For JJA,  
564 there is an obvious pattern between SWDOWN and Precip, with an increase in SWDOWN  
565 corresponding to a decrease in Precip and vice versa. According to IPCC (2007), in the warm  
566 seasons over land, strong negative correlations dominate as increased sunshine results in less  
567 evaporative cooling. ~~Figure A~~Figure S12 compares wind vectors superposed with T2 in 2006 and  
568 2010 from WRF/Chem and shows the largest differences are in JJA.

569 As expected, the spatial pattern of SWDOWN changes is anti-correlated with CF changes  
570 for all seasons between 2006 and 2010; however, the changes in the spatial pattern of CF do not  
571 correlate with changes in CDNC. CF in each grid cell is set to either 0 (no clouds), or to 1  
572 (cloudy) if total cloud water + ice mixing ratio  $> 1 \times 10^{-6} \text{ kg kg}^{-1}$  (Wu and Zhang, 2005). In this

573 study, the monthly CF is then normalized over the total number of time steps and vertical layers,  
574 giving a value of CF between 0 and 1 in each grid cell. In contrast, the calculations of CDNC in  
575 the model depend on the supersaturation, aerosol concentrations, aerosol hygroscopicity and  
576 updraft velocity (Abdul-Razzak and Ghan, 2004). The changes in CF are controlled by large  
577 scale state variables including temperature and relative humidity, while CDNC depends on more  
578 complex changes in microphysical variables. The dominant CDNC decrease in MAM, JJA, and  
579 SON, is due to lower  $PM_{2.5}$  concentrations, which in turn lower the effective number of cloud  
580 condensation nuclei. However, exception occurs in southeast U.S. where  $PM_{2.5}$  decreases but  
581 CDNC increases. This is because CDNC also depends on other variables including the amount of  
582 liquid water in the atmosphere. The cloud liquid water path over southeastern U.S. increases,  
583 which may explain the increase in CDNC. The spatial pattern for precipitation correlates to that  
584 of CF. The spatial pattern of CWP also corresponds to a certain extent with CF. PBLH increases  
585 when the ground warms up during the day and decreases when the ground cools so PBLH might  
586 be intuitively related to SWDOWN and T2. However, this consistent trend is now obvious in the  
587 plots, because the simulated growth of the planetary boundary layer (PBL) also depends on the  
588 surface sensible latent and heat fluxes and the entrainment of warmer air from the free  
589 troposphere (Chen, 2007).

#### 590 **4.3 Meteorology-Chemistry Feedback Predictions**

591 As shown in Table 1, similar to 2006, comparison of the performance of most  
592 meteorological variables between WRF/Chem and WRF for 2010 is improved in terms of NMBs  
593 when chemistry-meteorology feedbacks are included. This indicates the importance and benefits  
594 of inclusion of such feedbacks in online-coupled models. However, unlike 2006 for which both  
595 WRF only and WRF/Chem simulations show similar values of Corrs and NMEs, the 2010 WRF

596 | simulations give higher Corr and lower NMEs than the 2010 WRF/Chem simulations. –This  
597 | indicates the impact of worse chemical predictions on chemistry-meteorology feedbacks that can  
598 | in turn affect meteorological predictions. These results indicate the needs of further improvement  
599 | of the online-coupled models in their representations of chemistry-meteorology feedbacks.  
600 | Yahya et al. (2014) analyzed differences in meteorological performance between WRF/Chem  
601 | and WRF for 2006. ~~Figure A~~Figure S13 shows absolute seasonal differences between the  
602 | meteorological predictions from WRF/Chem and WRF for 2010. The differences between  
603 | WRF/Chem and WRF are consistent for both 2006 and 2010. SWDOWN in general is higher for  
604 | WRF/Chem compared to WRF for all seasons, with larger differences over the eastern portion of  
605 | the domain compared to the western portion. Other obvious similarities between 2006 and 2010  
606 | include the increase in T2 over the northern portion of the domain for MAM, SON and JFD;  
607 | increase in PBLH over the ocean in the eastern part of the domain for all seasons; and increases  
608 | over the ocean for CF for all seasons. The reasons for the differences between WRF/Chem and  
609 | WRF in terms of meteorological variables have been discussed in Yahya et al. (2014).

#### 610 **4.4 Sensitivity Simulations**

611 | The aforementioned differences in WRF/Chem predictions between 2006 and 2010 are  
612 | caused by changes in emissions, meteorology, and meteorological and chemical ICONs/BCONs.  
613 | Additional sensitivity simulations for the months of January and July 2010 are carried out to  
614 | estimate the individual contributions of each of those changes to the total net changes in model  
615 | predictions. ~~Table 3 he summarizes for the setup configurations of the sensitivity simulations are~~  
616 | ~~in Table 3.~~ The 2006 baseline simulations are designated as Run 1, the 2010 baseline simulations  
617 | are designated as Run 2, and the two sensitivity simulations are designated as Runs 3 and 4. Run  
618 | 3 is the sensitivity simulation using 2006 emissions but keeping all other inputs (e.g.,

619 | meteorology and chemical ICONs/BCONs) and model [set-up configurations](#) the same as Run 2.  
620 | Run 4 is the sensitivity simulation using 2006 emissions and chemical ICONs/BCONs keeping  
621 | all other inputs and model [set-up configurations](#) the same as Run 2. Figures 13 and 14 show the  
622 | changes due to combined effects of emissions, meteorological and chemical ICONs/BCONs  
623 | (~~column 1~~, Run 2\_ - Run 1 [in column 1](#)), changes due to the changes in emissions (~~column 2~~, Run  
624 | 2\_ - Run 3 [in column 2](#)), changes due to the changes in chemical ICONs/BCONs (~~column 3~~, Run  
625 | 3\_ - Run 4 [in column 3](#)), and changes due to the changes in meteorology including  
626 | ICONs/BCONs (~~column 4~~, Run 4 – Run 1 [in column 4](#)) for January and July, respectively. Since  
627 | the impact of ICONs is only important at the beginning of the simulations whereas the impact of  
628 | BCONs persists throughout the simulations, the changes due to changes in chemical BCONs will  
629 | dominate over those due to changes chemical ICONs/BCONs.

630 | Both Figures [13 and 14](#) show that the differences in the meteorology [due to the impact of](#)  
631 | ~~including meteorological~~-ICONs/BCONs generated by WRF/Chem contribute to ~~most of the~~  
632 | [largest](#) differences in T2 and SWDOWN for both months (~~columns 1 and 4~~). ~~For comparison,~~  
633 | ~~the changes in emissions and chemical ICONs/BCONs lead to less significant differences in T2~~  
634 | ~~and SWDOWN are seen with changes in emissions and chemical ICONs/BCONs~~ (~~columns 2~~  
635 | ~~and 3~~). ~~Column 2 from both Figures 13 and 14 show that~~ [The overall decrease in emissions from](#)  
636 | [2006 to 2010 results in a slight increase in both T2 and SWDOWN in January \(column 2 in](#)  
637 | [Figure 13\), and a more significant larger increase in SWDOWN in July \(column 2 in Figure 14\)](#)  
638 | [due to decreases in aerosol loading. Column 3 from both Figures 13 and 14 shows that](#) ~~There is~~  
639 | ~~a small decrease in T2 and SWDOWN in January (column 3 in Figure 13) due to influences of~~  
640 | ~~different years' chemical ICONs/BCONs used for both years, with but a more significant a larger~~  
641 | ~~decrease occurs in SWDOWN in July (column 3 in Figure 14). As shown in Figures 13 and 14~~

642 ~~(€column 21)~~, ~~€~~changes in O<sub>3</sub> are influenced by all factors and the overall change of O<sub>3</sub> mixing  
643 ratio is a combination of changes in emissions, meteorological and chemical ICONs/BCONs.  
644 ~~From Figure 13, Column 2,~~The O<sub>3</sub> mixing ratios are greatly increased due to the use of 2010  
645 emissions as compared to 2006 emissions (~~column 2 in Figure 13~~), indicating that using a  
646 different set of emissions can produce an increase of up to a domain mean of 6 ppb ~~domainwide~~.  
647 Conversely, O<sub>3</sub> mixing ratios are greatly decreased (~~with a reduction of a domain mean of 6 ppb~~  
648 ~~domainwide~~) due to the use of the 2010 chemical ICONs/BCONs compared to the 2006  
649 chemical ICONs/BCONs (~~Figure 13, column 3 in Figure 13~~). The use of different meteorological  
650 ICONs/BCONs also results in varying degrees of changes of O<sub>3</sub> mixing ratios domainwide as O<sub>3</sub>  
651 mixing ratios are ~~determined~~influenced by photolysis and other meteorological parameters  
652 including wind and PBLH (~~Figure 13, column 4 in Figure 13~~). In addition, T2 and SWDOWN  
653 ~~also~~influence the amount of BVOC emissions produced, which ~~also~~also in turn influences O<sub>3</sub>  
654 mixing ratios. In VOC-limited urban centers over eastern U.S. (Campbell et al., 2014), a small  
655 increase in radiation or T2 will increase BVOC emissions, increasing O<sub>3</sub> mixing ratios, and vice  
656 versa. In July (Figure 14), the decrease in O<sub>3</sub> mixing ratios between 2006 and 2010 (~~€column 1~~)  
657 is largely influenced by chemical ICONs/BCONs (~~€column 3~~) and to a smaller extent by  
658 meteorological ICONs/BCONs (~~€column 4~~). In this case, the difference in emissions (~~€column~~  
659 ~~2~~) does not seem to significantly impact the changes of O<sub>3</sub> mixing ratios between July 2006 and  
660 2010 (~~€column 1~~). For January ~~in~~(Figure 13), ~~decreases in~~PM<sub>2.5</sub> concentrations ~~decrease are~~  
661 ~~seen~~due to decreasing emissions and chemical ICONs/BCONs (~~€columns 2 and 3~~). However,  
662 the use of 2010 meteorological ICONs/BCONs ~~in~~Column 4 results in an increase in PM<sub>2.5</sub>  
663 concentrations over most ~~part~~of the domain except for the northeastern U.S. (~~with a domainwide~~  
664 ~~mean increase of 0.4 μg m<sup>-3</sup>~~) (column 4). The overall differences (~~Figure 13, €column 1 in~~

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

Formatted: Font: Symbol

Formatted: Superscript

665 Figure 13) are mainly due to net effects ~~can be caused by increases in precursor emissions (e.g.,~~  
666 ~~BVOCs) in eastern U.S., decreases in chemical ICONs/BCONs in western U.S., and changes in~~  
667 ~~meteorology in the entire U.S., leading to the dipole pattern in the differences of the spatial~~  
668 ~~distribution of O<sub>3</sub> concentrations from 2006 to 2010 (Figure 13, column 1). The net differences~~  
669 ~~in PM<sub>2.5</sub> concentrations in January from 2006 and 2010 are mainly due to decreases in of~~  
670 ~~emissions (column 2) and changes in meteorology (column 4). For O<sub>3</sub> in July, the net changes~~  
671 ~~from 2006 and 2010 are mainly due to decreases in chemical BCONs that compensate the~~  
672 ~~increases resulted from small increases in precursor emissions (e.g., VOCs) and changes in~~  
673 ~~meteorology. For PM<sub>2.5</sub> in July as shown in (Figure 14), the net changes from 2006 and 2010~~  
674 ~~(Column 1) are dominated entirely by changes in emissions (Column 2) that increase in the~~  
675 ~~southeastern and central U.S. but decrease in the remaining domain., even though~~  
676 ~~meteorological ICONs/BCONs also play a significant role (Column 4).~~

677 Table ~~S2-1A-S1~~ in the supplementary material shows the statistics NMB, NME, and Corr  
678 for a number of variables for the sensitivity simulations for January and July. The statistics in  
679 bold show which of highlights the sensitivity simulations with the best performance the best (i.e.  
680 with the lowest NMB, and NME and the highest Corr). The WRF/Chem performance of T2  
681 against CASTNET T2 improves to a large extent in terms of NME and Corr for Runs 3 and 4  
682 (especially for January when Run 2 performs poorly), which use 2006 emissions, especially for  
683 January when Run 2 performs poorly. This indicates that at least for January (and to a smaller  
684 extent for July), the inaccuracy of emissions may have contributed to the poorer worse  
685 performance of T2 against CASTNET. For CASTNET T2, Run 3 also gives the best  
686 performance of T2 against CASTNET is also for Run 3, which indicates that improvement in  
687 both emissions and chemical ICONs/BCONs can improve meteorological performances for both

688 January and July. For SWDOWN, Runs 3 and 4 improve the performance against CASTNET for  
689 January (with lower NMB, and NME and higher Corr). The cloud-aerosol variables are affected  
690 to a smaller extent by changes in emissions and chemical ICONs/BCONs compared to the  
691 meteorological variables. The performance for CF remains relatively the same for January and  
692 July. The performance for COT and AOD improves slightly for January with a lower NMB and  
693 NME but becomes worse in July with a higher NMB and NME. However, as the performance of  
694 meteorological variables is significantly different, a small change in cloud-aerosol variable can  
695 lead to a large change in meteorological variables. The performances for O<sub>3</sub> and PM<sub>2.5</sub>  
696 concentrations in January and July improve to a large extent when using 2006 emissions and  
697 especially when using 2006 chemical ICONs/BCONs are also used. The higher emissions of  
698 NO<sub>x</sub>, VOCs, and CO for July 2006 compared to 2010 contribute to the better O<sub>3</sub> performance,  
699 and the higher emissions of primary SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, EC and OA for 2006 contribute to the better  
700 PM<sub>2.5</sub> performance for Run 3 in July. However for January, a combination for both 2006  
701 emissions and chemical ICONs/BCONs improve the O<sub>3</sub> performance, while PM<sub>2.5</sub> performance  
702 is the best using 2010 emissions and 2010 ICONs/BCONs. This indicates that inaccuracies in  
703 emissions and chemical ICONs/BCONs in 2010, especially in January could contribute to the  
704 poor performance of WRF/Chem in 2010. These will, in turn affect the meteorological  
705 performance to a large extent. Table S2 shows the differences in emissions of major species from  
706 2006 and 2010. It is likely that the emissions for 2010 are underpredicted. Increasing the  
707 emissions for major species for 2010 might help to improve 2010 predictions.  
708 To evaluate if the sensitivity simulations with different meteorology, emissions, and  
709 chemical ICs/BCs for January and July 2010 can improve the model's capability in reproducing  
710 the trends in both meteorological and chemical variables, compared to baseline results in 2006

Formatted: Font: Not Bold

711 ~~and 2010. Table 4 shows the absolute and percentage differences between the monthly mean of~~  
712 ~~observations of major variables data in 2010 and 2006 versus and between simulation results~~  
713 ~~from three sensitivity simulation pairs: Runs 2 and 1, Runs 3 and 1, and Runs 4 and 1 are~~  
714 ~~calculated and summarized in Table 4. The Run 2 – Run 1 indicates the differences in between~~  
715 ~~2010 baseline simulation and the versus 2006 baseline simulations (Run 2 – Run 1) which show~~  
716 ~~the impact of all the changes (including emissions, meteorology, and chemical ICs/BCs) in the~~  
717 ~~2010 simulation relative to the 2006 simulation on the simulated variation trends and the degree~~  
718 ~~of agreement in the variation trends calculated from the two baseline simulations with the~~  
719 ~~observed changes. Comparisons of differences between Run 3 and Run 1 (Run 3 – Run 1) when~~  
720 ~~compared with those between Run 2 and Run 1 (Run 2 – Run 1) and between Run 4 and Run 1~~  
721 ~~(Run 4 – Run 1) with those between Run 2 and Run 1 (Run 2 – Run 1) indicate the impact of~~  
722 ~~changes in emissions and meteorology, respectively, on the simulated variation trends and their~~  
723 ~~degree of agreement with the observed changes. and Run 4 – Run 1 when compared with Run 2~~  
724 ~~– Run 1 will show the impact of changes in meteorology on variation trends and their degree of~~  
725 ~~agreement with observed change. As shown in Table 4, shows that the simulated database~~  
726 ~~model simulations (Run 2 – Run 1) for January and July are not able to reproduce the trends in~~  
727 ~~terms of either the signs or magnitude or both in the observed data ations for some of the~~  
728 ~~variables, including SWDOWN against CASTNET (January), CF against MODIS (July), COT~~  
729 ~~against MODIS (January), maximum 8-hr O<sub>3</sub> against CASTNET (January), and PM<sub>2.5</sub> against~~  
730 ~~STN (in January) and CF against MODIS in July. Changing the emissions (Run 3 – Run 1) does~~  
731 ~~not improve the variation trends from 2006 to 2010 with the exception of SWDOWN against~~  
732 ~~CASTNET (in January) and maximum 8-hr O<sub>3</sub> against CASTNET in July. Changing the~~  
733 ~~meteorology (Run 4 – Run 1) also does not improve the variation trends to a large extent with the~~

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

734 exception of maximum 8-hr O<sub>3</sub> against CASTNET (in January) and SWDOWN against  
735 CASTNET (in July). In fact, Run 2 – Run 1 (which are the originalbaseline simulations) overall  
736 performs the closest to the observed trends of major variables for January and July 2006 to 2010.

Formatted: Subscript

737

Formatted: Normal, Indent: First line: 0"

## 738 5. Summary and Conclusions

739 This study compares model performance in 2010 and 2006 and examines the changes in  
740 emissions, boundary conditions, and meteorology, as well as the responses of meteorology, air  
741 quality and chemistry-meteorology feedbacks to those changes collectively and ~~individually~~  
742 ~~between~~individually between 2010 and 2006. In general, the emissions of most gaseous and  
743 aerosol species over CONUS decrease from 2006 to 2010 with the exception of NH<sub>3</sub> emissions  
744 over several areas in JFD and biogenic VOCs mainly over eastern U.S. in JJA and SON. The  
745 increases in biogenic VOCs are caused by increases in temperatures in 2010 in eastern U.S.  
746 during these seasons. Overall, T2 increases from 2006 to 2010, however, the changes of T2 and  
747 other meteorological variables including SWDOWN, WS10, PBLH, and Precip vary spatially  
748 over CONUS with the largest differences for SWDOWN. The reduced emissions and changed  
749 meteorology result in decreased concentrations in general for gaseous and aerosol species except  
750 for species influenced by high BCONs, e.g., for OM concentrations over Canada in MAM and  
751 JJA. Due to increases in biogenic emissions, OM concentrations increase over eastern U.S.  
752 CDNC generally decreases over the U.S. due to the decreases in PM<sub>2.5</sub> concentrations and CCN  
753 from 2006 to 2010. The spatial distributions of other meteorological and cloud variables are  
754 consistent with known processes, e.g., SWDOWN is high and precipitation is low where CF is  
755 low. There is no clear spatial correlation between CF and CDNC due to the differences in their  
756 inherent prognostic treatments. COT corresponds relatively well to AOD, especially for JJA in

757 both years. CWP also corresponds well to COT. Sensitivity simulations show that the net  
758 changes in meteorological predictions in 2010 relative to 2006 are influenced mostly by changes  
759 in meteorology. Those of O<sub>3</sub> and PM<sub>2.5</sub> concentrations are influenced to a large extent by  
760 emissions and/or chemical ICONs/BCON, but meteorology may also influence them to some  
761 degrees, particularly in winter.

762 In general, the model performs well in terms of Corr and NMEs for almost all  
763 meteorological and chemical variables in 2006 but not as well in 2010 despite lower NMBs for  
764 most variables in 2010, due mainly to inaccuracies in emission estimates and chemical BCONs  
765 and ICONs used for 2010 simulations. The model is able to reproduce the observations to a large  
766 extent for most meteorological surface variables ~~except for precipitation~~. The model performs  
767 relatively well for PM<sub>2.5</sub> concentrations. However, OC concentrations are significantly  
768 underpredicted against field data for 2010 in Bakersfield and Pasadena, CA, due mainly to  
769 underestimations in emissions of POA that contributes to most OC and also in part to  
770 underestimations in emissions of gaseous precursors of SOA and inaccurate meteorological  
771 predictions in 2010. The model also has significant biases for a few aerosol-cloud-radiation  
772 variables except for CF and QVAPOR, however, the model is able to reproduce the trends in  
773 aerosol-cloud-radiation variables for 2006 and 2010. The variation trends for most  
774 meteorological and chemical variables simulated by WRF and WRF/Chem are overall consistent  
775 with the observed trends from 2006 to 2010 but for 2010, WRF/Chem performs slightly worse  
776 than WRF. Similar to 2006, the inclusion of chemistry-meteorology feedbacks reduces NMBs  
777 for most meteorological variables in 2010, although WRF gives higher Corr and lower NMEs  
778 than WRF/Chem.

Formatted: Subscript

779 A number of sensitivity simulations were also conducted for January and July 2006 and  
780 2010 to compare quantify the relative impact of emissions, chemical ICONs/BCONs, and  
781 meteorology on model performance of major meteorological and chemical species as well as on  
782 the variation trends between 2006 and 2010. Using more accurate emissions and chemical and  
783 meteorological ICONs/BCONs will help improve the performance of some individual chemical  
784 and meteorological surface variables. The sensitivity simulations show that the base simulations  
785 for 2006 and 2010 reproduce the observed trends the best. Although the 2006 emissions may not  
786 represent the true emissions for 2010, the 2010 sensitivity simulations using the 2006 emissions  
787 show improved model performance. however using improved emissions, chemical and  
788 meteorological ICONs/BCONs will help to improve the performance of individual chemical and  
789 meteorological surface variables. However, using 2006 emissions for 2010 simulationsthis will  
790 not necessarydoes not improve the degree of agreement with observed the inter-annual trends as  
791 the consistency between the 2006 and 2010 emissions are affected between the simulations. The  
792 baseline simulations for 2006 and 2010 reproduce the observed trends the best as a consistent set  
793 of 2006 and 2010 emissions are used. The current 2006 and 2010 emissions were developed  
794 taking into account the inter-annual trends, when simulating multi-year cases, the improvement  
795 of emissions need to be carried out consistently for all the individual simulation years when  
796 simulating multi-year cases.

797 WRF/Chem with CB05-MADE/VBS option used in this work has been incorporated into  
798 the WRF/Chem version 3.6.1 to be released in version 3.7 of WRF-Chem (available for  
799 download from <http://www.mmm.ucar.edu/wrf/users/>). These results in this work indicate a  
800 need to further improve the accuracy of emissions and chemical BCONs, and the representations  
801 of organic aerosols and chemistry-meteorology feedbacks in the online-coupled

802 ~~models~~WRF/Chem. In addition, the improvements in aerosol-cloud treatments, such as the  
803 aerosol activation parameterization scheme, and in the treatment of microphysics and cumulus  
804 parameterizations that affect the formation of precipitation are needed to be able to improve the  
805 model's capability in reproducing the state of the atmosphere and also inter-annual trends.  
806 While ~~These work also involves~~ long-term air quality simulations using WRF/Chem with  
807 aerosol-cloud-radiation feedbacks in this work can provide guidance on future model  
808 development and improvement, they do not provide the impact of those feedback mechanisms on  
809 the model performance. Quantifying such impacts requires another set of simulations using a  
810 version of WRF/Chem that does not treat aerosol direct and indirect effects, which is not yet  
811 available to public. The simulations with and without aerosol direct and indirect effects have  
812 indeed been performed by Makar et al. (2014a, b) using a different model that was specially  
813 designed to quantify such impacts. It would be useful to develop a version of WRF/Chem that  
814 does not treat aerosol direct and indirect effects for this impact assessment. In particular, There  
815 is also a need to perform episodic evaluations using WRF/Chem that excludes feedback  
816 mechanisms comparison of the episodic or long-term simulation results using WRF/Chem that  
817 includes and excludes feedback mechanisms against observations of aerosol and cloud variables  
818 to analyze the can provide further insight into whether actual impact of inclusion of those aerosol  
819 direct and indirect effects feedbacks and compare them with can improve the model's capability  
820 in reproducing observations. Those simulations should be considered when the version of  
821 WRF/Chem without aerosol direct and indirect effects and computer resources become available.

822 The developments in the WRF/Chem code used in this work have been incorporated into  
823 WRF/Chem version 3.6.1 to be released in version 3.7 of WRF Chem (available for download  
824 from <http://www.mmm.ucar.edu/wrf/users/>)

825 **Acknowledgements**

826           This study is funded by the National Science Foundation EaSM program (AGS-1049200)  
827 at NCSU. The following agencies have prepared the datasets used in this study: the U.S. EPA  
828 (North American emissions processing), Environment Canada, Mexican Secretariat of the  
829 Environment and Natural Resources (Secretaría del Medio Ambiente y Recursos Naturales,  
830 SEMARNAT) and National Institute of Ecology (Instituto Nacional de EcologíaINE) (North  
831 American national emissions inventories), the European Center for Medium Range Weather  
832 Forecasting Global and Regional Earth-system (Atmosphere) Monitoring using Satellite and in-  
833 situ data (ECMWF/GEMS) project and Meteo France/Centre national de recherches  
834 météorologiques (CNRM-GAME) for the Monitoring Atmospheric Composition and Climate  
835 (MACC) IC/BCs. Meteorological IC/BCs are provided by the National Center for Environmental  
836 Protection. Ambient North American concentration measurements are provided by several U.S.  
837 networks (AQS, CASTNET, IMPROVE, SEARCH, and STN). North American precipitation-  
838 chemistry measurements are provided by several U.S. networks (CASTNET, and NADP). GPCP  
839 Precipitation data is provided by the National Oceanic and Atmospheric Administration's Earth  
840 System Research Laboratory in the Physical Sciences Division (NOAA/OAR/ESRL PSD),  
841 Boulder, Colorado, USA, from their web site at <http://www.esrl.noaa.gov/psd/>. 2006 and 2010  
842 SOA/OC data at RTP, NC, Bakersfield, CA and Pasadena, CA were provided by John Offenberg,  
843 U.S. EPA. Cloud variables were provided by MODIS. We thank Georg Grell, NOAA, Christian  
844 Hogrefe, U.S. EPA, Paul Makar, Environment Canada, Christoph Knote, NCAR, and Patrick  
845 Campbell, NCSU, for helpful discussions on inputs and outputs of AQMEII model  
846 intercomparison. We would also like to acknowledge high-performance computing support from  
847 Yellowstone by NCAR's Computational and Information Systems Laboratory, sponsored by the

848 National Science Foundation. This work also used the Stampede Extreme Science and  
849 Engineering Discovery Environment (XSEDE) high-performance computing support which is  
850 supported by the National Science Foundation grant number ACI-1053575.

851 The US Environmental Protection Agency through its Office of Research and  
852 Development collaborated in the research described here. The manuscript has been subjected to  
853 external peer review and has not been cleared for publication. Mention of trade names or  
854 commercial products does not constitute endorsement or recommendation for use.

## 855 **References**

856 Abdul-Razzak, H., and Ghan, S.J.: A parameterization of aerosol activation, 2. Multiple  
857 aerosol types, *J. Geophys. Res.*, 105, 5, 6837-6844, 2000.

858 Ahmadov, R., McKeen, S.A., Robinson, A.L., Bahreini, R., Middlebrook, A.M., de Gouw, J.A.,  
859 Meagher, J., Hsie, E.-Y., Edgerton, E., Shaw, S., and Trainer, M.: A volatility basis set model  
860 for summertime secondary organic aerosols over the eastern United States in 2006, *J.*  
861 *Geophys. Res.*, 117, D06301, doi:10.1029/2011JD016831, 2012.

862 Aitken, A.C., Decarlo, P.F., Kroll, J.H., Worsnop, D.R., Huffman, J.A., Docherty, K.S., Ulbrich,  
863 I.M., Mohr, C., Kimmel, J.R., Sueper, D., Sun, Y., Zhang, Q., Trimborn, A., Northway, M.,  
864 Ziemann, P.J., Canagaratna, M.R., Onasch, T.B., Alfarra, M.R., Prevot, A.S.H., Dommen, J.,  
865 Duplissy, J., Metzger, A., Baltensperger, U., and Jimenez, J.L.: O/C and OM/OC ratios of  
866 primary, secondary and ambient organic aerosols with high-resolution time-of-flight aerosol  
867 mass spectrometry, *Environ. Sci. Technol.*, 42, 4478-4485, 2008.

868 Alapaty, K.V., Mathur, R., Pleim, J.E., Hogrefe, C., Rao, S.T., Ramaswamy, V., Galmarini, S.,  
869 Schapp, M., Vautard, R., Makar, P., Baklanov, A., Kallos, G., Vogel, B., and Sokhi, R.: New

870 Directions: Understanding interactions of air quality and climate change at regional scales,  
871 Atmos. Environ., 49, 3, doi:10.1016/j.atmos.env.2011.12.016, 2012.

872 Baklanov, A., Schlunzen, K., Suppan, P., Baldasano, J., Brunner, D., Aksoyoglu, S., Carmichael,  
873 G., Douros, J., Flemming, J., Forkel, R., Galmarini, S., Gauss, M., Grell, G., Hirtl, M., Joffre,  
874 S., Jorba, O., Kaas, E., Kaasik, M., Kallos, G., Kong, X., Korsholm, U., Kurganskiy, A.,  
875 Kushta, J., Lohmann, U., Mahura, A., Manders-Groot, A., Maurizi, A., Moussiopoulous, N.,  
876 Rao, S.T., Savage, N., Seigneur, C., Sokhi, R.S., Solazzo, E., Solomos, S., Sorensen, B.,  
877 Tsegas, G., Vignati, E., Vogel, B and Zhang, Y.: Online Coupled Regional Meteorology-  
878 Chemistry Models in Europe: Current Status and Prospects, Atmos. Chem. Phys., 14, 317-  
879 398, doi:10.5194/acp-14-317-2014, 2014.

880 Campbell, P., Zhang, Y., Yahya, K., Wang, K., Hogrefe, C., Pouliot, G., Knote, C., Hodzic, A.,  
881 San Jose, R., Perez, J.L., Guerrero, P.J., Baro, R., and Makar, P.: A Multi-Model Assessment  
882 for the 2006 and 2010 Simulations under the Air Quality Model Evaluation International  
883 Initiative (AQMEII) Phase 2 over North America, Part I. Indicators of the Sensitivity of O<sub>3</sub>  
884 and PM<sub>2.5</sub> Formation to Precursor Gases, Atmos. Environ., in press, 2014.

885 Chen, F.: The Noah Land Surface Model in WRF, A short tutorial, NCAR LSM group meeting,  
886 Boulder, CO, 17 April 2007, 2007.

887 Dawson, J. P., Racherla, P. N., Lynn, B. H., Adams, P. J., and Pandis, S. N.: Impacts of climate  
888 change on regional and urban air quality in the eastern United States: Role of meteorology. J.  
889 Geophys. Res., 114, D05308, doi:10.1029/2008JD00984, 2009.

890 Edwards, J.M., Beare, R.J., and Lapworth, A.J.: Simulation of the observed evening transition  
891 and nocturnal boundary layers: single column modeling, Q.J.R. Meteorol. Soc., 132, 61 – 80,  
892 2006.

893 EPA: National Emission Inventory – Ammonia emissions from animal husbandry operations,  
894 Draft Report, January 30, available at :  
895 [http://www.epa.gov/ttnchie1/ap42/ch09/related/nh3inventorydraft\\_jan2004.pdf](http://www.epa.gov/ttnchie1/ap42/ch09/related/nh3inventorydraft_jan2004.pdf), 2014.

896 Gao, Y., Fu, J.S., Drake, J.B., Liu, Y., and Lamarque, J.-F.: Projected changed of extreme  
897 weather events in the eastern United States based on a high resolution climate modeling  
898 system, *Environ. Res. Lett.*, 7, 044025, 2012.

899 Gao, Y., Fu, J. S., Drake, J. B., Lamarque, J. F., and Liu, Y.: The impact of emission  
900 and climate change on ozone in the United States under representative concentration  
901 pathways (RCPs), *Atmos. Chem. Phys.*, 13, 9607–9621, doi:10.5194/acp-13-9607-2013,  
902 2013.

903 Grundstein, A., and Dowd, J.: Trends in extreme apparent temperatures over the United States,  
904 1949-2010, *J. Appl. Meteor. Climatol.*, 50, 1650-1653, 2011.

905 Hogrefe, C., Lynn, B., Civerolo, K., Ku, J.-Y., Rosenthal, J., Rosenzweig, C., Goldberg, R.,  
906 Gaffin, S., Knowlton, K., and Kinney, P.L.: Simulating changes in regional air pollution over  
907 the eastern United States due to changes in global and regional climate and emissions, *J.*  
908 *Geophys. Res.*, 109, doi: 10.1029/2004JD004690, 2004.

909 Hogrefe, C., Pouliot, G., Wong, D., Torian, A., Roselle, S., Pleim, J., and Mathur, R.: Annual  
910 application and evaluation of the online coupled WRF-CMAQ system over North America  
911 under AQMEII Phase 2, *Atmos. Environ.*, in press, 2014.

912 Hong, S., Lakshmi, V., Small, E.E., Chen, F., Tewari, M., and Manning, K.W.: Effects of  
913 vegetation and soil moisture on the simulated land surface processes from the coupled  
914 WRF/Noah model., *J. Geophys. Res.*, 114, D18, doi:10.1029/2008JD011249, 2009.

915 Huber, D.G., and Gullede.: Extreme weather and climate change: Understanding the link and  
916 managing the risk, Center for Climate and Energy Solutions, available at :  
917 <http://www.c2es.org/publications/extreme-weather-and-climate-change>, 2011.

918 IPCC: Climate change 2007: the physical science basis. In: Solomon, S., Qin, D., Manning, M.  
919 (Eds.), Contribution of Working Group I to the Fourth Assessment Report of the  
920 Intergovernmental Panel on Climate Change, 2007.

921 IPCC: Managing the risks of extreme events and disasters to advance climate change adaptation  
922 (SREX), A special report of Working Groups I and II of the Intergovernmental Panel on  
923 Climate Change, [Field, C.B., V. Barros, T.F. Stocker, D. Qin, D.J. Dokken, K.L. Ebi, M.D.  
924 Mastrandrea, K.J. Mach, G.-K., Plattner, S.K. Allen, M. Tignor, and P.M. Midgley (eds.),  
925 Cambridge University Press, Cambridge, UK, and New York, NY, USA, 582 pp., 2012.

926 Im, et al.: Evaluation of operational on-line-coupled regional air quality models over Europe and  
927 North America in the context of AQMEII phase 2. Part 1: Ozone, *Atmos. Environ.*, in press,  
928 2014.

929 [Jacob, D.: Heterogeneous chemistry and tropospheric ozone, \*Atmos. Environ.\*, 34, 2131 – 2159,](#)  
930 [2000.](#)

931 Jimenez, P., Parra, R., and Baldasano, J.M.: Influence of initial and boundary conditions for  
932 ozone modeling in very complex terrains: A case study in the northeastern Iberian Peninsula,  
933 *Environ. Mod. Software*, 22, 1294-1306, 2007.

934 Jin, J., Miller, N.M., and Schlegel, N.: Sensitivity study of four land surface schemes in the WRF  
935 model, *Adv. Met.*, 2010, doi: 10.1155/2010/167436, 2010.

936 Khiem, M., Ooka, R., Huang, H., Hayami, H., Yoshikado, H., and Kawamoto, Y.: Analysis of  
937 the relationship between changes in meteorological conditions and the variation in summer  
938 ozone levels over the central Kanto area, *Adv. Met.*, 2010, doi:10.1155/2010/349248, 2010.

939 ~~Kleindienst, T.E., Jaoui, M., Lewandowski, M., Offenberg, J.H., and Docherty, K.S.: The~~  
940 ~~—formation of SOA and chemical tracer compounds from the photooxidation of naphthalene~~  
941 ~~and its methyl analogs in the presence and absence of nitrogen oxides, *Atmos. Chem. Phys.*,~~  
942 ~~12, 8711—8726, 2012.~~

943 Leung, L., and Gustafson, W.: Potential regional climate change and implications to US air  
944 quality, *Geophys. Res. Lett.*, 32, 16, L16711, doi:10.1029/2005GL022911, 2005.

945 Lewandowski, M., Piletic, I.R., Kleindienst, T.E., Offenberg, J.H., Beaver, M.R., Jaoui, M.,  
946 Docherty, K.S., and Edney, E.O.: Secondary organic aerosol characterization at field sites  
947 across the United States during the spring-summer period, *Intern. J. Environ. Anal. Chem.*,  
948 2013, 2013.

949 ~~Makar, P. et al., 2014, Feedbacks between air pollution and weather, part 2: Effects on~~  
950 ~~chemistry, *Atmos. Environ.*, doi:10.1016/j.atmosenv.2014.10.021~~~~Makar et al.: Feedbacks~~  
951 ~~between Air Pollution and Weather, Part 1: Effects on Chemistry, *Atmos. Environ.*, in~~  
952 ~~review, 2014.~~

Formatted: Font: Not Bold

Formatted: Font: Not Italic

Formatted: Font: Not Bold

953 Mass, C., and Owens, D.: WRF Model Physics: Progress, problems and perhaps some solutions,  
954 Presented at the 11<sup>th</sup> WRF Users' Workshop, Boulder, CO, 21 – 25 June 2010, 2010.

955 Meir, T., Orton, P.M., Pullen, J., Holt, T., Thompson, W.T., and Arend, M.F.: Forecasting the  
956 New York City urban heat island and sea breeze ~~adding during~~ extreme heat events, *Wea.*  
957 *Forecasting*, 28, 1460-1477, 2013.

958 Odum, J.R., Hoffmann, T., Bowman, F., Collins, D., Flagan, R.C., and Seinfeld, J.H.:  
959 Gas/Particle partitioning and secondary organic aerosol yields, *Environ. Sci. Tech.*, 30, 2580-  
960 2585, 1996.

961 Offenberg, J.H., Lewandowski, M., Jaoui, M., and Kleindienst, T.E.: Contributions of biogenic  
962 and anthropogenic hydrocarbons to secondary organic aerosol during 2006 in Research  
963 Triangle Park, NC, *Aero. Air Qual. Res.*, 11, 99 – 108, 2011.

964 Oswald, E.M., and Rood, R.B.: A trend analysis of the 1930-2010 extreme heat events in the  
965 continental United States, *J. Appl. Meteor. Climatol.*, 53, 565-582, 2014.

966 Penrod, A., Y. Zhang, K. Wang, S.-Y. Wu, and Leung, R.L.: Impacts of Future  
967 Climate and Emission Changes on U.S. Air Quality, *Atmos. Environ.*, 89, 533-547, 2014.

968 [Pleim, J.E., and Gilliam, R.: An Indirect Data Assimilation Scheme for Deep Soil Temperature](#)  
969 [in the Pleim-Xiu Land Surface Model, \*J. Appl. Meteor. Climatol.\*, 48, 1362 – 1376, 2009,](#)  
970 [doi:10.1175/2009JAMC2053.1.](#)

971 Pouliot, G., van der Gon, H.D., Kuenen, J., Makar, P., Zhang, J., and Moran, M.: Analysis of the  
972 Emission Inventories and Model-Ready Emission Datasets of Europe and North America for  
973 Phase 2 of the AQMEII Project, *Atmos. Environ.*, in press, 2014.

974 Rao, S., S. Galmarini, and Steyn, D.G.: AQMEII: An International Initiative for the  
975 Evaluation of Regional-Scale Air Quality Models-Phase 1, *Atmos. Environ.*, Special Issue,  
976 53, 1 – 224, 2012.

977 Reid, J.S., Koppmann, R., Eck, T.F., and Eleuterio, D.P.: A review of biomass burning emissions  
978 part II: intensive physical properties of biomass burning particles, *Atmos. Chem. Phys.*, 5,  
979 799 – 825, 2005.

Formatted: Indent: Hanging: 0.25"

980 Samaali, M., Moran, M.D., Bouchet, V.S., Pavlovic, R., Cousineau, S., and Sassi, M.: On the  
981 influence of chemical initial and boundary conditions on annual regional air quality model  
982 simulations for North America, *Atmos. Environ.*, 43, 32, 4873-4885, 2009.

983 [Sarwar, G., Fahey, K., Napelenok, S., Roselle, S., and Mathur, R.: Examining the impact of](#)  
984 [CMAQ model updates on aerosol sulfate predictions, 10<sup>th</sup> Annual CMAQ Models-3 Users's](#)  
985 [Conference, Chapel Hill, NC, 2011.](#)

986 Schell, B., Ackermann, I.J., Hass, H., Binkowski, F.S., and Ebel, A.: Modeling the formation of  
987 secondary organic aerosol within a comprehensive air quality model system, *J. Geophys.*  
988 *Res.*, 106, 28275 – 28293, 2001.

989 Schere, K., Flemming, J., Vautard, R., Chemel, Colette, A., Hogrefe, C., Bessagnet, B., Meleux,  
990 F., Mathur, R., Roselle, S., Hu, R.-M., Sokhi, R.S., Rao, S.T., Galmarini, S.: Trace  
991 gas/aerosol boundary concentrations and their impacts on continental-scale AQMEII  
992 modeling domains, *Atmos. Environ.*, 53, 38-50, 2012.

993 Stoeckenius, T., Chemel, C., Zagunis, J., and Sakulyanontvittaya, T.: A Comparison between  
994 2010 and 2006 Air Quality and Meteorological Conditions, and Emissions and Boundary  
995 Conditions for the AQMEII-2 North American Domain, *Atmos. Environ.*, in review, 2014.

996 Van Lier-Walqui, M., Vukicevic, T., and Posselt, D.J.: Linearization of microphysical  
997 parameterization uncertainty using multiplicative process perturbation parameters, *Mon.*  
998 *Wea. Rev.*, 142, 401 – 413, 2014.

999 [Wang, J., and Kotamarthi, V.R.: Downscaling with a nested regional climate model in near-](#)  
1000 [surface fields over the contiguous United States, \*J. Geophys. Res. Atmos.\*, 119, 14, 2014,](#)  
1001 [doi:10.1002/2014JD021696.](#)

Formatted: Superscript

1002 Wang, K., K. Yahya, Y. Zhang, S.-Y. Wu, and G. Grell.: Implementation and Initial Application  
1003 of A New Chemistry-Aerosol Option in WRF/Chem for Simulation of Secondary Organic  
1004 Aerosols and Aerosol Indirect Effects, *Atmos. Environ.*, in press,  
1005 doi:10.1016/j.atmosenv.2014.12.007, 2014.

1006 Wu, J., and Zhang, M.: Simulations of clouds and sensitivity study by Weather Research and  
1007 Forecast Model for Atmospheric Radiation Measurement case 4, Fifteenth Arm Science  
1008 Team Meeting Proceedings, 14 – 18 Mar 2005, Daytona Beach, FL, 2005.

1009 [Xing, J., Pleim, J., Mathur, R., Pouliot, G., Hogrefe, C., Gan, C.-M., and Wei, C.: Historical](#)  
1010 [gaseous and primary aerosol emissions in the United States from 1990 to 2010, \*Atmos.\*](#)  
1011 [Chem. Phys., 13, 7531 – 7549, 2013, doi:10.5194/acp-13-7531-2013.](#)

1012 Yahya, K., Wang, K., Gudoshava, M., Glotfelty, T., and Zhang, Y.: Application of WRF/Chem  
1013 over the continental U.S. under the AQMEII Phase II: Comprehensive Evaluation of 2006  
1014 Simulation, *Atmos. Environ.*, in press, doi:10.1016/j.atmosenv.2014.08.063, 2014.

1015 Yang, B., Qian, Y., Lin, G., Leung, L.R., Rasch, P.J., Zhang, G.J., McFarlane, S.A., Zhao, C.,  
1016 Zhang, Y., Wang, H., Wang, M., and Liu, X.: Uncertainty quantification and parameter  
1017 running in the CAM5 Zhang-McFarlane convection scheme and impact of improved  
1018 convection on the global circulation and climate, *J. Geophys. Res.*, 118, 395 – 415, 2013.

1019 Zhang, Y.: Online-coupled meteorology and chemistry models: history, current status, and  
1020 outlook, *Atmos. Chem. Phys.*, 8, 2895-2932, 2008.

1021 Zhang, Y., Hu, X., Leung, L. R., and Gustafson Jr., W.I.: Impacts of regional climate change on  
1022 biogenic emissions and air quality, *J. Geophys. Res.* 113, D18310,  
1023 doi:10.1029/2008JD009965, 2008.

1024 Zhang Y., X.-Y. Wen, and Jang C.J.: Simulating chemistry-aerosol-cloud-radiation-climate  
1025 feedbacks over the continental U.S. using the online-coupled Weather Research Forecasting  
1026 Model with chemistry (WRF/Chem), *Atmos. Environ.*, 44, 3568-3582, 2010.

1027 Zhang, Y., Chen, Y.-C., Sarwar, G., and Schere, K.: Impact of Gas-Phase Mechanisms on  
1028 Weather Research Forecasting Model with Chemistry (WRF/Chem) Predictions:  
1029 Mechanism Implementation and Comparative Evaluation, *J. Geophys. Res.*, 117, D1,  
1030 doi:10.1029/2011JD015775, 2012.

1031 Zhang, Y., Wang, W., Wu, S.-Y., Wang, K., Minoura, H., and Wang, Z.-F.: Impacts of Updated  
1032 Emission Inventories on Source Apportionment of Fine Particle and Ozone over the  
1033 Southeastern U.S., *Atmos. Environ.*, 588, 133-154, 2014.

1034

1035 **Table 1. Annual performance statistics for 2010 Predictions of WRF and WRF/Chem**

Network or Site name	Variable	WRF					WRF/Chem				
		Mean Obs	Mean Sim	Corr	NMB (%)	NME (%)	Mean Obs	Mean Sim	Corr	NMB (%)	NME (%)
CASTNET	T2	15.9	15.0	0.93	-5.0	15.8	15.9	15.1	0.64	-4.9	32.9
SEARCH	T2	19.4	18.4	0.94	-4.3	12.3	19.4	18.4	0.65	-5.1	27.6
CASTNET	SWDOWN	176.1	214.7	0.91	21.8	36.2	176.1	189.2	0.80	7.4	50.4
SEARCH	SWDOWN	217.7	245.0	0.91	11.5	31.6	217.7	211.0	0.78	-3.0	47.2
CASTNET	WS10	2.3	3.0	0.44	28.1	66.4	2.3	3.0	0.17	27.5	80.7
SEARCH	WS10	2.2	2.4	0.47	9.6	50.9	2.2	2.4	0.23	8.0	62.3
NADP	Precip	18.9	20.7	0.54	10.2	71.2	18.9	20.5	0.55	9.7	70.6
GPCC	Precip	2.2	2.3	0.83	1.1	22.6	2.2	2.2	0.83	-1.3	22.0
MODIS	CF	57.6	60.4	0.82	6.2	12.7	57.6	57.8	0.87	0.3	8.9
MODIS	AOD	-	-	-	-	-	0.10	0.05	-0.09	-46.6	54.4
MODIS	COT	-	-	-	-	-	17.2	6.3	0.45	-63.5	63.6
MODIS	CWP	-	-	-	-	-	160.1	97.3	0.54	-39.2	54.9
MODIS	QVAPOR	-	-	-	-	-	1.04	1.13	0.96	9.0	27.7
MODIS	CCN	-	-	-	-	-	0.33	0.09	0.60	-73.2	73.2
TERRA	CDNC	-	-	-	-	-	155.0	123.5	0.10	-20.0	59.2
CASTNET	Max 1-h O <sub>3</sub>	-	-	-	-	-	47.4	33.2	0.40	-30.0	34.8
CASTNET	Max 8-h O <sub>3</sub>	-	-	-	-	-	43.8	32.7	0.40	-25.3	32.0
AQS	Max 1-h O <sub>3</sub>	-	-	-	-	-	48.4	40.7	0.34	-15.8	28.0
AQS	Max 8-h O <sub>3</sub>	-	-	-	-	-	42.3	35.3	0.20	-17.0	29.2
STN	24-h PM <sub>2.5</sub>	-	-	-	-	-	11.0	9.7	0.17	-11.5	54.6
IMPROVE	24-h PM <sub>2.5</sub>	-	-	-	-	-	4.5	4.0	0.44	-11.5	56.0
STN	24-h SO <sub>4</sub>	-	-	-	-	-	2.2	2.6	0.33	19.0	68.5
IMPROVE	24-h SO <sub>4</sub>	-	-	-	-	-	1.0	1.3	0.50	21.1	72.3
STN	24-h NO <sub>3</sub>	-	-	-	-	-	1.4	0.7	0.10	-45.6	89.1
IMPROVE	24-h NO <sub>3</sub>	-	-	-	-	-	0.4	0.2	0.30	-43.3	95.5
STN	24-h NH <sub>4</sub>	-	-	-	-	-	1.0	1.0	0.21	1.5	72.5
STN	24-h EC	-	-	-	-	-	0.4	1.0	0.14	147.1	179.5
IMPROVE	24-h EC	-	-	-	-	-	0.2	0.3	0.29	78.5	123.8
STN	24-h TC	-	-	-	-	-	2.8	2.5	0.10	-11.9	62.0
IMPROVE	24-h OC	-	-	-	-	-	0.9	0.6	0.18	-29.6	74.2
IMPROVE	24-h TC	-	-	-	-	-	1.0	0.9	0.21	-11.8	72.8
Pasadena, CA <sup>2</sup>	SOA	-	-	-	-	-	0.63	0.16	0.1	-75.3	78.3
Bakersfield, CA <sup>2</sup>	SOA	-	-	-	-	-	0.51	0.23	0.3	-55.3	65.9

1036  
 1037 <sup>1</sup>Units are as follows: SWDOWN (W m<sup>-2</sup>), GLW (W m<sup>-2</sup>), OLR (W m<sup>-2</sup>), T2 (°C), RH2 (%), WS10 (m s<sup>-1</sup>),  
 1038 WD10 (°), Precip (mm), CWP (g m<sup>-2</sup>), QVAPOR (cm), CCN (10<sup>9</sup> cm<sup>-3</sup>), CDNC (cm<sup>-2</sup>), O<sub>3</sub> (ppb), PM and  
 1039 PM species (µg m<sup>-3</sup>). CASTNET - the Clean Air Status and Trends Network; AQS – the Aerometric  
 1040 Information Retrieval System Air Quality System; SEARCH - the Southeastern Aerosol Research and  
 1041 Characterization; GPCC - the Global Precipitation Climatology Centre; MODIS - the Moderate Resolution  
 1042 Imaging Spectroradiometer; IMPROVE – the Interagency Monitoring for Protected Visual Environmental;  
 1043 STN – the Speciated Trends Network. Note that IMPROVE did not contain NH<sub>4</sub><sup>+</sup> data for 2010. “-”  
 1044 indicates that the results of those variables not available from the WRF only simulation.  
 1045 <sup>2</sup>The observed SOA data are taken from Klendienst et al. (2012) and Lewandowski et al. (2013).

1046

1047 **Table 2. Percentage changes in observed and simulated variables between 2010 and 2006**

Network or Site name	Variable	Obs	WRF	WRF/Chem
CASTNET	T2	35.7	38.6	40.1
SEARCH	T2	1.3	0.0	0.5
CASTNET	SWDOWN	2.1	2.6	1.4
SEARCH	SWDOWN	7.3	7.4	5.2
CASTNET	WS10	0.0	0.0	-8.3
SEARCH	WS10	-4.3	-13.4	-12.4
NADP	Precip	6.7	-4.3	-1.5
GPCC	Precip	0.0	4.5	-12.0
MODIS	CF	-0.2	3.7	3.0
MODIS	AOD	-28.6	-	-44.4
MODIS	COT	4.2	-	6.8
MODIS	CWP	-10.2	-	-11.1
MODIS	QVAPOR	-47.5	-	-42.1
MODIS	CCN	-2.9	-	-30.8
CASTNET	Max 1-h O <sub>3</sub>	-0.5	-	-15.0
CASTNET	Max 8-h O <sub>3</sub>	0.6	-	-13.9
AQS	Max 1-h O <sub>3</sub>	-3.9	-	-14.6
AQS	Max 8-h O <sub>3</sub>	-4.9	-	-17.4
STN	24-h PM <sub>2.5</sub>	-9.9	-	-20.8
IMPROVE	24-h PM <sub>2.5</sub>	-16.1	-	-27.0
STN	24-h SO <sub>4</sub>	-25.8	-	-33.3
IMPROVE	24-h SO <sub>4</sub>	-23.7	-	-26.3
STN	24-h NO <sub>3</sub>	-11.3	-	-27.8
IMPROVE	24-h NO <sub>3</sub>	-20.0	-	-53.5
STN	24-h NH <sub>4</sub>	-25.3	-	-31.9
STN	24-h EC	-39.5	-	-1.6
IMPROVE	24-h EC	-21.6	-	2.4
STN	24-h TC	-38.1	-	-24.2
IMPROVE	24-h OC	-17.3	-	-45.5
IMPROVE	24-h TC	-25.5	-	-35.7

1048

1049 <sup>1</sup>The percentages are calculated according to this formula: [(2010 value – 2006 value) / 2006 value] \* 100%.  
1050 CASTNET - the Clean Air Status and Trends Network; AQS – the Aerometric Information Retrieval  
1051 System Air Quality System; SEARCH - the Southeastern Aerosol Research and Characterization; GPCC -  
1052 the Global Precipitation Climatology Centre; MODIS - the Moderate Resolution Imaging  
1053 Spectroradiometer; IMPROVE – the Interagency Monitoring for Protected Visual Environment; STN –  
1054 the Speciated Trends Network. Note that IMPROVE did not contain NH<sub>4</sub><sup>+</sup> data for 2010. “-“ indicates that  
1055 the results of those variables not available from the WRF only simulation.

1056



1063 |  
1064 **List of Figures**

1065  
1066  
1067 Figure 1. Comparison of seasonal plots of NMB vs NME of various meteorological variables for  
1068 2006 (left column) and 2010 (right column) – T2 (temperature at 2m), SWDOWN  
1069 (downward shortwave radiation), WS10 (wind speed at 10m) and Precipitation where the  
1070 shapes represent different seasons (diamond – MAM, circle – JJA, triangle – SON and  
1071 square – JFD) and the different colors represent different observational data (red –  
1072 SEARCH, blue – CASTNET, green – NADP, black – GPCC).

1073 Figure 2. Spatial Distribution of NMB plots for JFD and JJA 2006 and 2010 for maximum 8-hr  
1074 O<sub>3</sub> concentrations based on evaluation against CASTNET, AQS and SEARCH.

1075 Figure 3. Comparison of seasonal plots of NMB vs NME for maximum 8-hr O<sub>3</sub> concentrations  
1076 where the different shapes represent different seasons (diamond – MAM, circle – JJA,  
1077 triangle – SON and square – JFD) and the different colors represent different  
1078 observational data (purple – CASTNET, black – AQS and green - SEARCH).

1079 Figure 4. Diurnal variation of T2 (top row) and hourly O<sub>3</sub> concentrations (bottom row) against  
1080 CASTNET for JJA 2006 and 2010.

1081 Figure 5. Spatial Distribution of NMB plots for JFD and JJA 2006 and 2010 for average 24-hr  
1082 PM<sub>2.5</sub> concentrations based on evaluation against the IMPROVE, STN and SEARCH  
1083 sites.

1084 Figure 6. Comparison of seasonal plots of NMB vs NME for average 24-hr PM<sub>2.5</sub> concentrations  
1085 where the different shapes represent different seasons (diamond – MAM, circle – JJA,  
1086 triangle – SON and square – JFD) and the different colors represent different  
1087 observational data (purple – IMPROVE, black – STN and green - SEARCH).

1088 Figure 7. Plots of annual statistics (NMB vs NME) for average 24-hr  $PM_{2.5}$  concentrations and  
1089  $PM_{2.5}$  species against different observational networks.

1090 Figure 8. Time series of Obs vs. Sim  $PM_{2.5}$ ,  $SO_4$  and  $NO_3$  concentrations against STN for 2006  
1091 and 2010.

1092 Figure 9. Scatter plots of SOA (left column) and OC (right column) concentrations at various  
1093 sites.

1094 Figure 10. Comparison of scatter plots for JFD and JJA 2006 and 2010 evaluation of aerosol  
1095 and cloud variables. MISR AOD, and SRB CF obs data was not available for 2010.

1096 Figure 11. Changes in hourly average surface concentrations of  $O_3$  and PM species from 2010 to  
1097 2006 (2010 – 2006).

1098 Figure 12. Changes in hourly average predictions of aerosol-cloud variables at surface from  
1099 WRF/Chem simulations from 2010 to 2006 (2010 – 2006).

1100 Figure 13. Spatial difference plots for January where Run 1: 2006 baseline simulations; Run 2:  
1101 2010 baseline simulations; Run 3: 2010 simulations with 2006 emissions and 2010  
1102 meteorology and chemical IC/BCs; Run 4: 2010 simulations with 2006 emissions and  
1103 2006 chemical IC/BCs and 2010 meteorology.

1104 Figure 14. Spatial difference plots for July where Run 1: 2006 baseline simulations; Run 2: 2010  
1105 baseline simulations; Run 3: 2010 simulations with 2006 emissions and 2010  
1106 meteorology and chemical IC/BCs; Run 4: 2010 simulations with 2006 emissions and  
1107 2006 chemical IC/BCs and 2010 meteorology.

1108