Reply to Comments from Reviewer 1

Geosci. Model Dev. Discuss., 8, C152–C154, 2015
www.geosci-model-dev-discuss.net/8/C152/2015/

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_3 mixing ratios and $PM_{2.5}$ 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 $PM_{2.5}$ 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_3 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 ≤ 15% and an NME of ≤ 30% indicate satisfactory performances for O_3 and $PM_{2.5}$. 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 O3 concentrations in this context is very weak.

Reply:

Figure 4 was actually averaged over only the summer period of June to August (O_3 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_3 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, $PM_{2.5}$ 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/

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 _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, $PM_{2.5}$ concentrations are more underpredicted in 2010 than 2006 (i.e., simulated $PM_{2.5}$ is a better agreement with observations in 2006). Underpredicted $PM_{2.5}$ 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_3 and $PM_{2.5}$ 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/

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_3 mixing ratios as follows:

"As shown in Figures 13 and 14 (column 2), changes in O_3 are influenced by all factors and the overall change of O_3 mixing ratio is a combination of changes in emissions, meteorological and chemical ICONs/BCONs. The O_3 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_3 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_3 mixing ratios domainwide as O_3 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.

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

Khairunnisa Yahya, Kai Wang, and Yang Zhang*

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

Tadeusz E. Kleindienst

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

Abstract

The Weather Research and Forecasting model with Chemistry (WRF/Chem) simulation with the 2005 Carbon Bond gas-phase mechanism coupled to the Modal for Aerosol Dynamics for Europe and the Volatility Basis Set approach for Secondary Organic Aerosol (SOA) are conducted over a domain in North America for 2006 and 2010 as part of the Air Quality Model Evaluation International Initiative (AQMEII) Phase 2 project. Following the Part I paper that focuses on the evaluation of the 2006 simulations, t~~T~~his Part II paper focuses on comparison of model performance in 2006 and 2010 as well as analysis of the responses of air quality and meteorology-chemistry interactions to changes in emissions and meteorology from 2006 to 2010. In general, emissions for gaseous and aerosol species decrease from 2006 to 2010, leading to a reduction in gaseous and aerosol concentrations and associated changes in radiation and cloud variables due to various feedback mechanisms. WRF/Chem is able to reproduce most observations and the observed variation trends from 2006 to 2010, despite its slightly worse performance than WRF that is likely due to inaccurate chemistry feedbacks resulted from less accurate emissions and chemical boundary conditions (BCONs) in 2010. Compared to 2006, the

*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

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

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

1. Introduction

Changes in meteorology, climate, and emissions affect air quality (e.g., Hogrefe et al., 2004; Leung and Gustafson, 2005; Zhang et al., 2008; Dawson et al., 2009; Gao et al., 2013; Penrod et al., 2014). As federal, state, and local environmental protection agencies enforce the anthropogenic emission control programs, ambient air quality is expected to be continuously

improved. However, such an improvement may be compensated by adverse changes in climatic or meteorological conditions (e.g., increases in near surface temperature, solar radiation, and atmospheric stability, or reductions in precipitation) that are directly conducive to the formation and accumulation of air pollutants and that may result in higher biogenic emissions. It is therefore important to examine changes in ~~both meteorology~~meteorology/climate and emissions as well as their combined impacts on air quality. The Air Quality Model Evaluation International Initiative (AQMEII) Phase 2 was launched in 2011 to intercompare online-coupled air quality models (AQMs) in their capabilities in reproducing atmospheric observations and simulating air quality and climate interactions in North America (NA) and Europe (EU) (Alapaty et al., 2012). The simulations over NA and EU with multi-models by a number of participants have been performed for two years (2006 and 2010) that have distinct meteorological conditions. Compared with 2006, 2010 is characterized by warmer summer conditions in eastern U.S. and less precipitation over NA (Stoeckenius et al., 2014; Pouliot et al., 2014). In addition, the emissions of key pollutants are reduced in 2010 relative to 2006, e.g., emissions of oxides of nitrogen (NO_x) and sulfur dioxide (SO_2) are reduced by 10-30% and 40-80% for many regions in NA (Pouliot et al., 2014). Comparison of 2010 and 2006 simulations will thus provide an opportunity to examine the success of the emission control programs and the impacts of meteorological/climatic variables on air quality. Compared to model intercomparison during AQMEII Phase 1 (Rao et al., 2012) in which offline-coupled models were used, the use of online-coupled AQMs models during AQMEII Phase 2 allows for study of the interactions between meteorology and chemistry through various direct and indirect feedbacks among aerosols, radiation, clouds, and chemistry (Zhang, 2008; Baklanov et al., 2014). The two year simulations further enable an examination of the responses of air quality and meteorology-

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

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

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

secondary organic aerosol (SOA) (hereafter WRF/Chem-CB05-MADE/VBS) has been recently developed by Wang et al. (2014). The WRF/Chem-CB05-MADE/VBS ~~is~~has been coupled to the aqueous-phase chemistry scheme (AQChem) based on the AQChem version in CMAQ v~~4.7~~5.0 of Sarwar et al. (2011) for ~~. This option considers the aqueous chemistry in~~ both large-scale and convective clouds (Wang et al., 2014). WRF/Chem-CB05-MADE/VBS also contains ~~H~~heterogeneous chemistry involving sulfur dioxide on the surface of aerosols based on Jacob (2000) ~~has been incorporated into this version of the model. In addition, the model~~and treats both aerosol direct and indirect effects ~~by affecting CCN formation (Wang et al., 2014).~~ The applications of WRF/Chem-CB05-MADE/VBS to 2006 and 2010 in this work use the same model physical and chemical parameterizations as those in the Part I paper of Yahya et al. (2014) but with different emissions, meteorological ICONs and BCONs, and chemical ICONs and BCONs. The mechanistic evaluation by comparing WRF/Chem-CB05-MADE/VBS predictions for the two years would help understand the sensitivity of the model predictions and performance to different model inputs, and that by comparing WRF/Chem-CB05-MADE/VBS and WRF only predictions would quantify the impacts of chemistry-meteorology feedbacks on the meteorological predictions. A comprehensive evaluation of the 2006 simulation has been presented in the Part I paper Yahya et al. (2014). In this Part II paper, the differences in emissions, meteorological and chemical ICONs/BCONs, and meteorology between 2010 and 2006 are first examined briefly. The model performance in 2010 is then evaluated and compared with that in 2006. Finally, the responses of air quality and meteorology-chemistry interactions to changes in emissions, chemical ICONs/BCONs, and meteorology individually and collectively from 2006 to 2010 are analyzed. The main objectives of this 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. Stoeckenius et al. (2014) carried out an extensive analysis of the trends in emissions and observations of meteorological variables, O_3, SO_2, and $PM_{2.5}$ concentrations between 2006 and 2010. This Part I paper complements the work of Stoeckenius et al. (2014) by examining the changes in WRF/Chem predictions and chemistry-meteorology feedbacks in 2010 relative to 2006. Similar evaluations of 2010 and 2006 are performed for the coupled Weather Research and Forecasting – Community Multiscale Air Quality (WRF-CMAQ) system (Hogrefe et al., 2014). Unlike the coupled WRF-CMAQ system used in AQMEII Phase 2 that only simulates aerosol direct effects, WRF/Chem used in this work simulates both aerosol direct and indirect effects. In addition, the work by Hogrefe et al. (2014) involves nudging of temperature, wind speed, water vapor mixing ratio, soil temperature and soil moisture, while the model used for this study did not include any nudging.

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

2.1 Emission Trends

The emission variation trends are examined for major precursors for ozone (O_3) and secondary particulate matters (PM) (i.e., sulfur dioxide (SO_2), oxides of nitrogen (NO_x), ammonia (NH_3), volatile organic compounds (VOCs) including both anthropogenic and biogenic VOCs) and primary PM species (elemental carbon (EC) and primary organic aerosol or carbon (POA or POC)). As shown in Table S~~2~~1, emissions of most species decrease from 2006 to 2010 with ~~a~~ domainwide averages of -10% to -24%. Comparing to emissions in 2006, the annual emissions of SO_2 and NO_x decrease significantly in 2010, especially at the point sources (Figure ~~A1~~S1), with similar variation patterns in all seasons (Figure not shown). The annual emissions of NH_3 decrease over most areas but increase in some areas in California (CA) and Midwest.

Unlike the changes in the emissions of SO_2 and NO_x, NH_3 and VOCs emissions exhibit strong seasonal variations in the emission trends, as shown in Figure ~~A2~~S2. Although anthropogenic VOC emissions decrease over continental U.S. (CONUS) for all seasons (Figure not shown), the VOC emissions increase in the southeast, which is dominated by enhanced biogenic emissions from vegetation as a response to temperature increases (Stoeckenius et al., 2014). The total annual emissions of EC and POA also decrease but to a smaller extent over most areas of the continental U.S. The changes in annual and seasonal emissions of those species between 2010 and 2006 will affect simulated air quality and meteorology-chemistry interactions. In addition, there exist uncertainties in the NEI emissions. The major sources of uncertainties or errors in the NEI emissions include: ~~the fact that~~(1) the emissions ~~a~~were 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 different years of NEI data (Xing et al., 2013). These will affect the accuracy of the model simulations.

2.2 Differences in Chemical and Meteorological ICONs/BCONs

Large differences exist in the chemical and meteorological ICONs/BCONs used in the simulations. For example, Stoeckenius et al. (2014) reported that the mid-tropospheric seasonal mean O_3 mixing ratios are generally lower by several ppbs in 2010 as compared to 2006, especially during spring and summer. Less Asian mid-tropospheric fine dust was also transported over to the U.S. in the spring of 2010 and less African dust reached the U.S. in the summer of 2010 (Stoeckenius et al., 2014). As shown in Figure ~~A3~~S3, significant differences exist for January, February, and December (JFD) and June, July, August (JJA) 2010 – 2006 in averaged

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

3. Model Performance in 2010 and Its Comparison with 2006

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

3.1 Differences in Meteorological Predictions for 2006 and 2010

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

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

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

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

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

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

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

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

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

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

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

3.2 Differences in Chemical Predictions for 2006 and 2010

The chemical performance between 2006 and 2010 is more variable compared to the meteorological performance of surface variables. The lower Corr for 2010 compared to 2006 for meteorological variables has a large influence on the model performance for 2010. As shown in Table 1, all the chemical variables for all networks have lower Corr in 2010 compared to 2006. As shown in Figures 2 and 3, maximum 8-hr O_3 concentrations are underpredicted to a larger extent in 2010 compared to 2006, dominating the O_3 annual performance in 2010. These results are consistent with the results of Hogrefe et al. (2014). The large underpredictions of maximum 8-hr O_3 in JFD 2010 over southeastern U.S. are attributed to larger cold biases in T2 shown in ~~Figure A~~Figure S4 and reduced NO_x and VOC emissions in 2010 relative to their levels in 2006. While reduced NO_x levels can result in an increase in nighttime O_3 concentrations due to reduced NO_x titration of O_3, the impact of reduced NO_x titration on the maximum 8-hr O_3 is small. As shown in ~~Figure A~~Figure S4, the temperature biases for both years are relatively similar. Over northeastern U.S., the T2 bias is generally less than -0.1 ºC for JJA in both years. However, as shown in Figure 2, O_3 concentrations over northeastern U.S. in JJA 2010 have negative biases whereas those over northeastern U.S. in JJA 2006 have positive biases. In this case, emissions might play a significant role in the underprediction of O_3 concentrations over northeastern U.S.

in JJA 2010. Hourly average surface NO_x emissions decrease significantly over northeastern U.S. in JJA from 2006 to 2010. As shown in Figure 3, 2006 model performance for O_3 is generally good for all seasons and all networks.

According to Table 1 and Figure 1, WRF/Chem predicts SWDOWN to a lower extent in 2010 compared to 2006 against CASTNET. Khiem et al. (2010) reported that during the summer, a large percentage of the variations in peak O_3 concentrations during the summer can be attributed to changes in seasonally averaged daily maximum temperature and seasonally averaged WS10. Simulated WS10 is lower for 2010 compared to 2006 in general; therefore, WS10 does not seem to contribute to reduced O_3 concentrations (through dispersion, increased dry deposition) in 2010. Figure 4 shows diurnal variations of observed and simulated WRF/Chem T2 and O_3 concentrations from CASTNET in JJA 2006 and 2010. The diurnal averaging provides insight whether the underpredictions of O_3 mixing ratios is a systematic bias~~,~~ ~~i.e.~~ during the daytime ~~and~~or nighttime or both. ~~-~~The diurnally averaged observed temperatures show a similar trend in JJA 2006 to 2010 against T2 measurements from CASTNET. This shows that the model is able to reproduce T2 for different years. The temperature trends also correlate strongly with the O_3 trends. At night, where the model has cold bias, O_3 concentrations are underpredicted to a larger extent. The O_3 concentrations show a larger underprediction for JJA 2010 compared to JJA 2006. The underpredictions in O_3 in both 2006 and 2010 can be explained by several reasons. For example, Im et al. (2014) showed that MACC underpredicts O_3 mixing ratios, particularly in winter and spring during both day and night and in summer and fall during nighttime. As indicated by Wang et al. (2014) and Makar et al. (2014), the inclusion of aerosol indirect effects also tends to reduce O_3 mixing ratios, comparing to the models that simulate

Formatted: Subscript

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

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

2010 than those in 2006. SO_4^{2-} has lower NMBs but higher NMEs for all networks in 2010 compared to 2006. EC concentrations are generally overpredicted in 2006 for all networks but underpredicted against SEARCH and largely overpredicted against IMPROVE in 2010. NH_4^+ also has higher NMEs in 2010 compared to 2006. Overall, the evaluation in 2010 shows larger NMEs and ~~poor~~ weaker correlations for all $PM_{2.5}$ species compared to 2006.

Figure 8 shows the time series plots for 24-hr average concentrations of $PM_{2.5}$, SO_4^{2-} and NO_3^- against STN for 2006 and 2010. In 2006, the daily-average PM data were collected on a daily basis in 2006 but every 3 days in 2010. The model is able to predict most of the observed peaks and troughs for 2006 even though the observed and simulated magnitudes are significantly different for several days. For 2010, the model does not show large spikes and can reproduce the magnitudes well, although it does not predict the peaks and troughs as well as 2006 for some months (e.g., Jan-March and July-Sept. for $PM_{2.5}$). This could be attributed in part to the ~~poor~~ weaker correlations of meteorological variables in 2010 compared to 2006. For example, ~~poor~~inaccurate predictions of WS10 can influence the transport and dry deposition of aerosols. An ~~Poor~~ overpredictions~~s~~ of precipitation ~~can impact~~increases the wet deposition of aerosols. Poor predictions of T2 can influence the planetary boundary layer height (PBLH) and both can also affect the distribution of aerosol concentrations. NO_3^- concentrations for the winter months are moderately underpredicted in 2006 but largely underpredicted in 2010, likely due to the underpredictions in nitrogen dioxide (NO_2) concentrations (Yahya et al., 2014). Section 4 will discuss in further detail the role of emissions, meteorology and chemical ICONs/BCONs on O_3 and $PM_{2.5}$ concentrations.

3.3 SOA Evaluation for 2006 and 2010

Formatted: Subscript

Formatted: Subscript

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

As shown in Figures 9 and ~~A5~~S5, the model overpredicts SOA but underpredicts OC at RTP in 2006, because (1) the SOA formed from alkanes and alkenes is excluded in the observations from RTP but simulated in WRF/Chem, and (2) WRF/Chem may have overestimated the aging rate coefficient for both anthropogenic and biogenic surrogate VOC precursors (Wang et al. (2014)). The SOA overprediction due to those reasons compensates the

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

3.4 Differences in Aerosol-Cloud Predictions for 2006 and 2010

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

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

3.5 Differences in Observed and Simulated Trends between 2010 and 2006

Table 2 shows the percentage changes in observed and WRF only and WRF/Chem simulated variables between 2010 and 2006. Overall, the model is able to predict the trends in all ~~the listed~~major meteorological, chemical, and aerosol-cloud-radiation variables between 2006 and 2010 with ~~the~~a few exceptions ~~of~~ (e.g.,WS10 against CASTNET, Precip, CF, maximum 8-hr O_3 against CASTNET, and 24-hr EC against IMPROVE). The trends in simulated T2, SWDOWN, and SEARCH WS10 are generally consistent with the observed trends from 2006 to 2010. Both observed and simulated temperatures at 2-m (T2) at the CASTNET sites increase by ~4 ºC or ~35 to 40% from 2006 to 2010. For downward shortwave radiation (SWDOWN), both observed and simulated values at the CASTNET and SEARCH sites increase by ~1 to 3% and by ~5 to 7%, respectively, from 2006 to 2010. The observed WS10 remains relatively constant at CASTNET in both years. The simulated WS10 by WRF also shows no change but that by

Formatted: Subscript

WRF/Chem shows a small decrease (by -8.3%) for the CASTNET sites. . —Comparing to a SEARCH observed change of ~-4% in WS10, WRF and WRF/Chem predict a larger decrease from 2006 to 2010 (~-12 to -13%). 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 ~7%, however both simulated WRF and WRF/Chem show a small decrease from 2006 to 2010. Observed mean GPCC Precip remained relatively constant from 2006 to 2010, however, WRF only shows a slight increase (~4%) while WRF/Chem shows a larger decrease (-12%) from 2006 to 2010. MODIS CF decreased by -0.2% from 2006 to 2010 whereas both WRF and WRF/Chem show small increases ~3-4% from 2006 to 2010. Apart from the large biases in the evaluation of precipitation, the decrease in precipitation is likely due to the smaller decrease in SWDOWN for WRF/Chem compared to observations between 2006 and 2010. This would result in less convective precipitation during the summer but increased CF for 2010. In addition, $PM_{2.5}$ is underpredicted ~~in 2010~~ but ~~has a~~agrees better ~~agreement~~ with observed $PM_{2.5}$ in 2010 than in 2006. Underpredicted $PM_{2.5}$ concentrations will also affect the formation of clouds and precipitation via various direct and indirect effects.

The simulated decreasing trends between 2006 and 2010 are overall consistent with the observed decreasing trend between 2006 and 2010 for all species except for maximum 8-hr O_3 concentrations from CASTNET and EC from IMPROVE. CASTNET maximum 1-hr and 8-hr O_3 concentrations change very little from 2006 to 2010 whereas WRF/Chem shows a moderate decrease of 14-15%. The large decrease in simulated O_3 mixing ratios in 2010 can be attributed to a large decrease in O_3 mixing ratios from the ICONs and BCONs (Stoeckenius et al., 2014). The IMPROVE observed EC concentrations decreased by ~22% from 2006 to 2010, however, WRF/Chem shows a small increase (by ~2%). For $PM_{2.5}$ concentrations, the simulated decrease

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

from 2006 to 2010 by WRF/Chem is larger than the observed decrease for both STN and IMPROVE. Similar steeper decreases by WRF/Chem also occur for SO_4^{2-} against STN, NO_3^- against IMPROVE, TC against STN, and OC against IMPROVE~~.~~ likely due to the influence of ICONs/BCONs and emissions.

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

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

4.1 Air Quality Predictions

Simulated air quality responds nonlinearly to the changes in emissions. Figures 11, ~~A7~~S7- ~~A9~~ S9 show the seasonal changes between 2010 and 2006 in ambient mixing ratios of gases (SO_2, NO_2, NH_3, O_3, and hydroxyl - OH) and concentrations of PM species (SO_4^{2-}, NO_3^-, NH_4^+, organic matter or OM, EC, POA, anthropogenic SOA or ASOA, biogenic SOA or BSOA, and $PM_{2.5}$). SO_2 and NO_2 concentrations tend to decrease for all seasons at most locations over CONUS due to the decrease in their emissions. The increases in NO_2 concentrations over urban areas in eastern U.S. in March, April, May (MAM) in 2010 relative to 2006 could be due to a few reasons including decreased photolytic conversion from NO_2 to NO due to a decrease in SWDOWN and less NO_2 conversion to nitric acid (HNO_3) due to decreased OH concentrations. The NO_2 hot spots also correlate to the decrease in hourly O_3 concentrations in urban areas. This could indicate an increased titration of nighttime O_3 by NO. This is an important result for policy implications, as reducing NO_x emissions may reduce NO_2 concentrations overall for CONUS, but may not reduce NO_2 concentrations in several areas, especially in urban areas due to a combination of titration and complex interplay with local meteorology. NH_3 mixing ratios

505 generally decrease in the U.S., except over eastern U.S. in MAM and September, October, and
506 November (SON), where there are increases. NH_3 emissions decrease, however, over eastern
507 U.S. in all seasons. The increase in NH_3 concentrations in MAM and SON could be attributed to
508 a number of reasons including less NH_3 conversion to NH_4^+ to neutralize SO_4^{2-} and NO_3^- and less
509 dispersion of NH_3 concentrations due to decreased wind speeds over eastern and southeastern
510 U.S. in MAM and SON, respectively, in 2010 compared to 2006. In JJA and SON, high OM
511 concentrations in Canada are attributed to the enhanced impacts of BCONs by increasingly
512 convergent flow in this region. OM is made up of both POA and SOA. An increase in VOC
513 emissions in eastern U.S. in MAM and SON leads to increases in OM concentrations. Decreases
514 in VOC emissions in western U.S. for all seasons lead to decreases in OM concentrations. The
515 OM concentrations in some areas, however, do not follow a linear relationship with VOC
516 emissions, such as southeastern U.S. in JJA, where VOC emissions increase from 2006 to 2010
517 but OM concentrations decrease. A decrease in POA concentrations must dominate the overall
518 decrease in OM concentrations, even under increased temperatures and biogenic VOC emissions
519 in this area. $PM_{2.5}$ concentrations decrease for all seasons and most regions of the CONUS,
520 which is attributed mainly to decreases in precursor gases, especially the inorganic precursors
521 SO_2 and NO_x in eastern U.S. Increased $PM_{2.5}$ concentrations in JFD and MAM in the Midwest
522 are due to surface temperature decreases, dominating in this region (Stoeckenius et al., 2014).
523 This in turn leads to increased particle nitrate concentrations (Campbell et al., 2014).

524 **4.2 Meteorological Predictions**

525 ~~Figure A~~Figure S10 compares the seasonal changes between 2010 and 2006 in several
526 meteorological variables that affect air pollution including SWDOWN, T2, WS10, PBLH, and
527 Precip simulated by WRF only simulations without considering chemistry feedbacks. Large

changes occur in those variables between the two years, e.g., 10-50 W m^{-2} increases in SWDOWN in western and Midwest in JJA, generally warmer in JJA and SON over most areas but cooler by 3-10 ºC in eastern U.S. in JFD, and with reduced Precip in eastern or southeastern U.S. in JJA and SON but increased Precip in northwestern U.S. in MAM and JJA and in western U.S. in JFD. ICONs and BCONs for skin temperatures shown in ~~Figure A~~Figure S3 greatly influence T2 shown in ~~Figure A~~Figure S10 for JFD and JJA.

Figures 12 and ~~A11~~ S11 show the seasonal changes between 2010 and 2006 in several meteorological and cloud variables SWDOWN, T2, WS10, Precip, PBLH, AOD, COT, CF, CWP, and CDNC) for WRF/Chem that accounts for meteorology-chemistry feedbacks. The relationships between various meteorological variables have been discussed in Yahya et al. (2014). Comparing to the differences in predictions of SWDOWN, T2, WS10, Precip, and PBLH between 2010 and 2006 WRF only simulation shown in ~~Figure A~~Figure S10 and WRF/Chem ~~simulations shown~~simulations shown in Figures 12 and ~~A11~~S11, the differences in those meteorological variables ~~except for SWDOWN~~ do not vary significantly in terms of trends of average seasonal spatial distributions between 2010 and 2006 WRF simulations and between 2010 and 2006 WRF/Chem simulations. However, there are differences in magnitudes, especially for SWDOWN. SWDOWN is affected most by the addition of chemistry in WRF/Chem as compared to WRF, especially for JFD through indirect feedback of clouds on radiation. As shown in Figure 12, the decrease in SWDOWN from 2006 to 2010 is larger over north-central and north-western U.S. and the increase in SWDOWN is smaller over north-eastern and southwestern U.S. for MAM (WRF/Chem) compared to MAM (WRF). For SON, the increase in SWDOWN from 2006 to 2010 simulated by WRF/Chem is larger over eastern U.S. than that by WRF. The differences between WRF and WRF/Chem are the largest for SWDOWN

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

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

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

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

591 **4.3 Meteorology-Chemistry Feedback Predictions**

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

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

4.4 Sensitivity Simulations

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

meteorology and chemical ICONs/BCONs) and model ~~set-up~~configurations the same as Run 2. Run 4 is the sensitivity simulation using 2006 emissions and chemical ICONs/BCONs keeping all other inputs and model ~~set-up~~configurations the same as Run 2. Figures 13 and 14 show the changes due to combined effects of emissions, meteorological and chemical ICONs/BCONs (~~column 1,~~ Run 2 - Run 1 in column 1), changes due to the changes in emissions (~~column 2,~~ Run 2 - Run 3 in column 2), changes due to the changes in chemical ICONs/BCONs (~~column 3,~~ Run 3 - Run 4 in column 3), and changes due to the changes in meteorology including ICONs/BCONs (~~column 4,~~ Run 4 – Run 1 in column 4) for January and July, respectively. Since the impact of ICONs is only important at the beginning of the simulations whereas the impact of BCONs persists throughout the simulations, the changes due to changes in chemical BCONs will dominate over those due to changes chemical ICONs/BCONs.

Both Figures 13 and 14 show that the differences in the meteorology due to the impact of ~~including~~meteorological ICONs/BCONs generated by WRF/Chem contribute to ~~most of the~~the largest differences in T2 and SWDOWN for both months (columns 1 and 4)~~.~~. For comparison, the changes in emissions and chemical ICONs/BCONs ~~L~~lead to less significant differences in T2 and SWDOWN ~~are seen with changes in emissions and chemical ICONs/BCONs~~ (columns 2 and 3). ~~Column 2 from both Figures 13 and 14 show that t~~The overall decrease in emissions from 2006 to 2010 results in a slight increase in both T2 and SWDOWN in January (column 2 in Figure 13), and a ~~more significant~~larger increase in SWDOWN in July (column 2 in Figure 14) due to decreases in aerosol loading. ~~Column 3 from both Figures 13 and 14 shows that t~~There is a small decrease in T2 and SWDOWN in January (column 3 in Figure 13) due to influences of ~~different years'~~ chemical ICONs/BCONs used for both years, ~~with~~but ~~a more significant~~a larger decrease occurs in SWDOWN in July (column 3 in Figure 14). As shown in Figures 13 and 14

(~~C~~column ~~2~~1), ~~C~~changes in O_3 are influenced by all factors and the overall change of O_3 mixing ratio is a combination of changes in emissions, meteorological and chemical ICONs/BCONs. ~~From Figure 13, Column 2,~~The O_3 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_3 mixing ratios are greatly decreased (with a reduction of a domain mean of 6 ppb ~~domainwide~~) due to the use of the 2010 chemical ICONs/BCONs compared to the 2006 chemical ICONs/BCONs (~~Figure 13,~~ column 3 in Figure 13). The use of different meteorological ICONs/BCONs also results in varying degrees of changes of O_3 mixing ratios domainwide as O_3 mixing ratios are ~~determined~~influenced by photolysis and other meteorological parameters including wind and PBLH (~~Figure 13,~~ column 4 in Figure 13). In addition, T2 and SWDOWN ~~also~~ influence the amount of BVOC emissions produced, which ~~also~~also in turn influences O_3 mixing ratios. In VOC-limited urban centers over eastern U.S. (Campbell et al., 2014), a small increase in radiation or T2 will increase BVOC emissions, increasing O_3 mixing ratios, and vice versa. In July (Figure 14), the decrease in O_3 mixing ratios between 2006 and 2010 (~~C~~column 1) is largely influenced by chemical ICONs/BCONs (~~C~~column 3) and to a smaller extent by meteorological ICONs/BCONs (~~C~~column 4). In this case, the difference in emissions (~~C~~column 2) does not seem to significantly impact the changes of O_3 mixing ratios between July 2006 and 2010 (~~C~~column 1). For January ~~in~~ (Figure 13), ~~decreases in~~ $PM_{2.5}$ concentrations decrease ~~are seen~~ due to decreasing emissions and chemical ICONs/BCONs (~~C~~columns 2 and 3). However, the use of 2010 meteorological ICONs/BCONs ~~in Column 4~~ results in an increase in $PM_{2.5}$ concentrations over most ~~part~~ of the domain except for the northeastern U.S. (with a domain~~wide~~ mean increase of 0.4 $\mu g\ m^{-3}$) (column 4). The overall differences (~~Figure 13, C~~column 1 in

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

Figure 13) are mainly due to net effects ~~can be caused by increases in precursor emissions (e.g., BVOCs) in eastern U.S., decreases in chemical ICONs/BCONs in western U.S., and changes in meteorology in the entire U.S., leading to the dipole pattern in the differences of the spatial distribution of O_3 concentrations from 2006 to 2010 (Figure 13, column 1). The net differences in $PM_{2.5}$ concentrations in January from 2006 and 2010 are mainly due to decreases in~~of emissions (column 2) and changes in meteorology (column 4). ~~For O_3 in July, the net changes from 2006 and 2010 are mainly due to decreases in chemical BCONs that compensate the increases resulted from small increases in precursor emissions (e.g., VOCs) and changes in meteorology.~~ For $PM_{2.5}$ in July ~~as shown in~~ (Figure 14), the net changes from 2006 and 2010 (~~C~~column 1) are dominated entirely by changes in emissions (~~C~~column 2) that increase in the southeastern and central U.S. but decrease in the remaining domain~~.~~, even though meteorological ICONs/BCONs also play a significant role (~~C~~column 4).

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

January and July. For SWDOWN, Runs 3 and 4 improve the performance against CASTNET for January (with lower NMB~~,~~ and NME and higher Corr). The cloud-aerosol variables are affected to a smaller extent by changes in emissions and chemical ICONs/BCONs compared to the meteorological variables. The performance for CF remains relatively the same for January and July. The performance for COT and AOD improves slightly for January with a lower NMB and NME but becomes worse in July with a higher NMB and NME. However, as the performance of meteorological variables is significantly different, a small change in cloud-aerosol variable can lead to a large change in meteorological variables. The performances for O_3 and $PM_{2.5}$ concentrations in January and July improve to a large extent when using 2006 emissions and especially when ~~using~~ 2006 chemical ICONs/BCONs are also used. The higher emissions of NO_x, VOCs, and CO for July 2006 compared to 2010 contribute to the better O_3 performance, and the higher emissions of primary SO_4^{2-}, NO_3^-, EC and OA for 2006 contribute to the better $PM_{2.5}$ performance for Run 3 in July. However for January, a combination for both 2006 emissions and chemical ICONs/BCONs improve the O_3 performance, while $PM_{2.5}$ performance is the best using 2010 emissions and 2010 ICONs/BCONs. This indicates that inaccuracies in emissions and chemical ICONs/BCONs in 2010, especially in January could contribute to the poor performance of WRF/Chem in 2010. These will, in turn affect the meteorological performance to a large extent. ~~Table S2 shows the differences in emissions of major species from 2006 and 2010. It is likely that the emissions for 2010 are underpredicted. Increasing the emissions for major species for 2010 might help to improve 2010 predictions.~~

To evaluate if the sensitivity simulations with different meteorology, emissions, and chemical ICs/BCs for January and July 2010 can improve the model's capability in reproducing the trends in both meteorological and chemical variables, compared to baseline results in 2006

Formatted: Font: Not Bold

and 2010, ~~Table 4 shows t~~the absolute and percentage differences between the monthly mean of observ~~ed~~ations of major variables~~data~~ in 2010 and 2006 ~~versus~~and between simulate~~d~~ion results from three ~~sensitivity~~ simulation~~s~~ pairs: Runs 2 and 1, Runs 3 and 1, and Runs 4 and 1 are calculated and summarized in Table 4. The ~~Run 2 – Run 1 indicates the~~ differences ~~in~~between 2010 baseline simulation and the~~versus~~ 2006 baseline simulations (Run 2 – Run 1) ~~which~~ show the impact of all the changes (including emissions, meteorology, and chemical ICs/BCs) in the 2010 simulation relative to the 2006 simulation on the simulated variation trends and the degree of agreement in the variation trends calculated from the two baseline simulations with the observed changes~~;~~. Comparisons of differences between Run 3 and Run 1 (Run 3 – Run 1) ~~when compared~~ with those between Run 2 and Run 1 (Run 2 – Run 1) and between Run 4 and Run 1 (Run 4 – Run 1) with those between Run 2 and Run 1 (Run 2 – Run 1) ~~-~~indicate the impact of changes in emissions and meteorology, respectively, on the simulated variation trends and their degree of agreement with the observed changes~~;. and Run 4 – Run 1 when compared with Run 2 – Run 1 will show the impact of changes in meteorology on variation trends and their degree of agreement with observed change~~. As shown in Table 4, ~~shows that~~ the ~~simulated data~~baseline model simulations (Run 2 – Run 1)~~for January and July~~ are not able to reproduce the trends in terms of either the signs or magnitude or both in the observ~~ed data~~ations for some ~~of the~~ variables, including SWDOWN against CASTNET ~~(January)~~, ~~CF against MODIS (July),~~ COT against MODIS ~~(January)~~, maximum 8-hr O_3 against CASTNET ~~(January)~~, and $PM_{2.5}$ against STN ~~(~~in January~~)~~ and CF against MODIS in July. Changing the emissions (Run 3 – Run 1) does not improve the variation trends from 2006 to 2010 with the exception of SWDOWN against CASTNET ~~(~~in January~~)~~ and maximum 8-hr O_3 against CASTNET in July. Changing the meteorology (Run 4 – Run 1) also does not improve the variation trends to a large extent with the

Formatted: Subscript

Formatted: Subscript

Formatted: Subscript

exception of maximum 8-hr O_3 against CASTNET ~~(~~in January~~)~~ and SWDOWN against CASTNET ~~(~~in July~~)~~. In fact, Run 2 – Run 1 (which are the ~~original~~baseline simulations) overall performs the closest to the observed trends of major variables for January and July 2006 to 2010.

Formatted: Subscript

Formatted: Normal, Indent: First line: 0"

5. Summary and Conclusions

This study compares model performance in 2010 and 2006 and examines the changes in emissions, boundary conditions, and meteorology, as well as the responses of meteorology, air quality and chemistry-meteorology feedbacks to those changes collectively and ~~individually between~~individually between 2010 and 2006. In general, the emissions of most gaseous and aerosol species over CONUS decrease from 2006 to 2010 with the exception of NH_3 emissions over several areas in JFD and biogenic VOCs mainly over eastern U.S. in JJA and SON. The increases in biogenic VOCs are caused by increases in temperatures in 2010 in eastern U.S. during these seasons. Overall, T2 increases from 2006 to 2010, however, the changes of T2 and other meteorological variables including SWDOWN, WS10, PBLH, and Precip vary spatially over CONUS with the largest differences for SWDOWN. The reduced emissions and changed meteorology result in decreased concentrations in general for gaseous and aerosol species except for species influenced by high BCONs, e.g., for OM concentrations over Canada in MAM and JJA. Due to increases in biogenic emissions, OM concentrations increase over eastern U.S. CDNC generally decreases over the U.S. due to the decreases in $PM_{2.5}$ concentrations and CCN from 2006 to 2010. The spatial distributions of other meteorological and cloud variables are consistent with known processes, e.g., SWDOWN is high and precipitation is low where CF is low. There is no clear spatial correlation between CF and CDNC due to the differences in their inherent prognostic treatments. COT corresponds relatively well to AOD, especially for JJA in

both years. CWP also corresponds well to COT. Sensitivity simulations show that the net changes in meteorological predictions in 2010 relative to 2006 are influenced mostly by changes in meteorology. Those of O_3 and $PM_{2.5}$ concentrations are influenced to a large extent by emissions and/or chemical ICONs/BCON, but meteorology may also influence them to some degrees, particularly in winter.

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. The model is able to reproduce the observations to a large extent for most meteorological surface variables ~~except for precipitation~~. The model performs relatively well for $PM_{2.5}$ concentrations. However, OC concentrations are significantly underpredicted against field data for 2010 in Bakersfield and Pasadena, CA, due mainly to underestimations in emissions of POA that contributes to most OC and also in part to underestimations in emissions of gaseous precursors of SOA and inaccurate meteorological predictions in 2010. The model also has significant biases for a few aerosol-cloud-radiation variables except for CF and QVAPOR, however, the model is able to reproduce the trends in aerosol-cloud-radiation variables for 2006 and 2010. The variation trends for most meteorological and chemical variables simulated by WRF and WRF/Chem are overall consistent with the observed trends from 2006 to 2010 but for 2010, WRF/Chem performs slightly worse than WRF. Similar to 2006, the inclusion of chemistry-meteorology feedbacks reduces NMBs for most meteorological variables in 2010, although WRF gives higher Corr and lower NMEs than WRF/Chem.

Formatted: Subscript

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

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

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

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

Acknowledgements

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

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

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

References

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

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

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

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

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

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

Campbell, P., Zhang, Y., Yahya, K., Wang, K., Hogrefe, C., Pouliot, G., Knote, C., Hodzic, A., San Jose, R., Perez, J.L., Guerrero, P.J., Baro, R., and Makar, P.: A Multi-Model Assessment for the 2006 and 2010 Simulations under the Air Quality Model Evaluation International Initiative (AQMEII) Phase 2 over North America, Part I. Indicators of the Sensitivity of O_3 and $PM_{2.5}$ Formation to Precursor Gases, Atmos. Environ., in press, 2014.

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

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

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

894 EPA: National Emission Inventory – Ammonia emissions from animal husbandry operations,
895 Draft Report, January 30, available at :
896 http://www.epa.gov/ttnchie1/ap42/ch09/related/nh3inventorydraft_jan2004.pdf, 2014.
897 Gao, Y., Fu, J.S., Drake, J.B., Liu, Y., and Lamarque, J.-F.: Projected changed of extreme
898 weather events in the eastern United States based on a high resolution climate modeling
899 system, Environ. Res. Lett., 7, 044025, 2012.
900 Gao, Y., Fu, J. S., Drake, J. B., Lamarque, J. F., and Liu, Y.: The impact of emission
901 and climate change on ozone in the United States under representative
902 concentration
903 pathways (RCPs), Atmos. Chem. Phys., 13, 9607–9621, doi:10.5194/acp-13-9607-
904 2013, 2013.
905 Grundstein, A., and Dowd, J.: Trends in extreme apparent temperatures over the United States,
906 1949-2010, J. Appl. Meteor. Climatol., 50, 1650-1653, 2011.
907 Hogrefe, C., Lynn, B., Civerolo, K., Ku, J.-Y., Rosenthal, J., Rosenzweig, C., Goldberg, R.,
908 Gaffin, S., Knowlton, K., and Kinney, P.L.: Simulating changes in regional air pollution over
909 the eastern United States due to changes in global and regional climate and emissions, J.
910 Geophys. Res., 109, doi: 10.1029/2004JD004690, 2004.
911 Hogrefe, C., Pouliot, G., Wong, D., Torian, A., Roselle, S., Pleim, J., and Mathur, R.: Annual
912 application and evaluation of the online coupled WRF-CMAQ system over North America
913 under AQMEII Phase 2, Atmos. Environ., in press, 2014.
914 Hong, S., Lakshmi, V., Small, E.E., Chen, F., Tewari, M., and Manning, K.W.: Effects of
915 vegetation and soil moisture on the simulated land surface processes from the coupled
916 WRF/Noah model., J. Geophys. Res., 114, D18, doi:10.1029/2008JD011249, 2009.

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

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

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

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

Jacob, D.: Heterogeneous chemistry and tropospheric ozone, Atmos. Environ., 34, 2131 – 2159, 2000.

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

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

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

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

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

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

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

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

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

Formatted: Font: Not Bold

Formatted: Font: Not Italic

Formatted: Font: Not Bold

960 Odum, J.R., Hoffmann, T., Bowman, F., Collins, D., Flagan, R.C., and Seinfeld, J.H.:
961 Gas/Particle partitioning and secondary organic aerosol yields, Environ. Sci. Tech., 30, 2580-
962 2585, 1996.
963 Offenberg, J.H., Lewandowski, M., Jaoui, M., and Kleindienst, T.E.: Contributions of biogenic
964 and anthropogenic hydrocarbons to secondary organic aerosol during 2006 in Research
965 Triangle Park, NC, Aero. Air Qual. Res., 11, 99 – 108, 2011.
966 Oswald, E.M., and Rood, R.B.: A trend analysis of the 1930-2010 extreme heat events in the
967 continental United States, J. Appl. Meteor. Climatol., 53, 565-582, 2014.
968 Penrod, A., Y. Zhang, K. Wang, S.-Y. Wu, and Leung, R.L.: Impacts of Future
969 Climate and Emission Changes on U.S. Air Quality, Atmos. Environ., 89, 533-547, 2014.
970 Pleim, J.E., and Gilliam, R.: An Indirect Data Assimilation Scheme for Deep Soil Temperature
971 in the Pleim-Xiu Land Surface Model, J. Appl. Meteor. Climatol., 48, 1362 – 1376, 2009,
972 doi:10.1175/2009JAMC2053.1.
973 Pouliot, G., van der Gon, H.D., Kuenen, J., Makar, P., Zhang, J., and Moran, M.: Analysis of the
974 Emission Inventories and Model-Ready Emission Datasets of Europe and North America for
975 Phase 2 of the AQMEII Project, Atmos. Environ., in press, 2014.
976 Rao, S., S. Galmarini, and Steyn, D.G.: AQMEII: An International Initiative for the
977 Evaluation of Regional-Scale Air Quality Models-Phase 1, Atmos. Environ., Special Issue,
978 53, 1 – 224, 2012.
979 Reid, J.S., Koppmann, R., Eck, T.F., and Eleuterio, D.P.: A review of biomass burning emissions
980 part II: intensive physical properties of biomass burning particles, Atmos. Chem. Phys., 5,
981 799 – 825, 2005.

Formatted: Indent: Hanging: 0.25"

982 Samaali, M., Moran, M.D., Bouchet, V.S., Pavlovic, R., Cousineau, S., and Sassi, M.:. On the
983 influence of chemical initial and boundary conditions on annual regional air quality model
984 simulations for North America, Atmos. Environ., 43, 32, 4873-4885, 2009.
985 Sarwar, G., Fahey, K., Napelenok, S., Roselle, S., and Mathur, R.: Examining the impact of
986 CMAQ model updates on aerosol sulfate predictions, 10th Annual CMAQ Models-3 Users's
987 Conference, Chapel Hill, NC, 2011.
988 Schell, B., Ackermann, I.J., Hass, H., Binkowski, F.S., and Ebel, A.: Modeling the formation of
989 secondary organic aerosol within a comprehensive air quality model system, J. Geophys.
990 Res., 106, 28275 – 28293, 2001.
991 Schere, K, Flemming, J., Vautard, R., Chemel, Colette, A., Hogrefe, C., Bessagnet, B., Meleux,
992 F., Mathur, R., Roselle, S., Hu, R.-M., Sokhi, R.S., Rao, S.T., Galmarini, S.: Trace
993 gas/aerosol boundary concentrations and their impacts on continental-scale AQMEII
994 modeling domains, Atmos. Environ., 53, 38-50, 2012.
995 Stoeckenius, T., Chemel, C., Zagunis, J., and Sakulyanontvittaya, T.: A Comparison between
996 2010 and 2006 Air Quality and Meteorological Conditions, and Emissions and Boundary
997 Conditions for the AQMEII-2 North American Domain, Atmos. Environ., in review, 2014.
998 Van Lier-Walqui, M., Vukicevic, T., and Posselt, D.J.: Linearization of microphysical
999 parameterization uncertainty using multiplicative process perturbation parameters, Mon.
1000 Wea. Rev., 142, 401 – 413, 2014.
1001 Wang, J., and Kotamarthi, V.R.: Downscaling with a nested regional climate model in near-
1002 surface fields over the contiguous United States, J. Geophys. Res. Atmos., 119, 14, 2014,
1003 doi:10.1002/2014JD021696.

Formatted: Superscript

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

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

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

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

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

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

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

1026 Zhang Y., X.-Y. Wen, and Jang C.J.:, Simulating chemistry-aerosol-cloud-radiation-climate
1027 feedbacks over the continental U.S. using the online-coupled Weather Research Forecasting
1028 Model with chemistry (WRF/Chem), Atmos. Environ., 44, 3568-3582, 2010.
1029 Zhang, Y., Chen, Y.-C., Sarwar, G., and Schere, K.: Impact of Gas-Phase Mechanisms on
1030 Weather Research Forecasting Model with Chemistry (WRF/Chem) Predictions:
1031 Mechanism Implementation and Comparative Evaluation, J. Geophys. Res., 117, D1,
1032 doi:10.1029/2011JD015775, 2012.
1033 Zhang, Y., Wang, W., Wu, S.-Y., Wang, K., Minoura, H., and Wang, Z.-F.: Impacts of Updated
1034 Emission Inventories on Source Apportionment of Fine Particle and Ozone over the
1035 Southeastern U.S., Atmos. Environ., 588, 133-154, 2014.
1036

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

		WRF					WRF/Chem				
Network or Site name	**Variable**	**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_3**	-	-	-	-	-	47.4	33.2	0.40	-30.0	34.8
CASTNET	**Max 8-h O_3**	-	-	-	-	-	43.8	32.7	0.40	-25.3	32.0
AQS	**Max 1-h O_3**	-	-	-	-	-	48.4	40.7	0.34	-15.8	28.0
AQS	**Max 8-h O_3**	-	-	-	-	-	42.3	35.3	0.20	-17.0	29.2
STN	**24-h $PM_{2.5}$**	-	-	-	-	-	11.0	9.7	0.17	-11.5	54.6
IMPROVE	**24-h $PM_{2.5}$**	-	-	-	-	-	4.5	4.0	0.44	-11.5	56.0
STN	**24-h SO_4**	-	-	-	-	-	2.2	2.6	0.33	19.0	68.5
IMPROVE	**24-h SO_4**	-	-	-	-	-	1.0	1.3	0.50	21.1	72.3
STN	**24-h NO_3**	-	-	-	-	-	1.4	0.7	0.10	-45.6	89.1
IMPROVE	**24-h NO_3**	-	-	-	-	-	0.4	0.2	0.30	-43.3	95.5
STN	**24-h NH_4**	-	-	-	-	-	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[2]	**SOA**	-	-	-	-	-	0.63	0.16	0.1	-75.3	78.3
Bakersfield, CA[2]	**SOA**	-	-	-	-	-	0.51	0.23	0.3	-55.3	65.9

[1] Units are as follows: SWDOWN (W m^{-2}), GLW (W m^{-2}), OLR (W m^{-2}), T2 (ºC), RH2 (%), WS10 (m s^{-1}), WD10 (°), Precip (mm), CWP (g m^{-2}), QVAPOR (cm), CCN (10^9 cm^{-2}), CDNC (cm^{-2}), O_3 (ppb), PM and PM species (μg m^{-3}). CASTNET - the Clean Air Status and Trends Network; AQS – the Aerometric Information Retrieval System Air Quality System; SEARCH - the Southeastern Aerosol Research and Characterization; GPCC - the Global Precipitation Climatology Centre; MODIS - the Moderate Resolution Imaging Spectroradiometer; IMPROVE – the Interagency Monitoring for Protected Visual Environmental; STN – the Speciated Trends Network. Note that IMPROVE did not contain NH4+ data for 2010. "-" indicates that the results of those variables not available from the WRF only simulation.

[2] The observed SOA data are taken from ~~Klendienst et al. (2012) and~~ Lewandowski et al. (2013).

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_3**	-0.5	-	-15.0
CASTNET	**Max 8-h O_3**	0.6	-	-13.9
AQS	**Max 1-h O_3**	-3.9	-	-14.6
AQS	**Max 8-h O_3**	-4.9	-	-17.4
STN	**24-h $PM_{2.5}$**	-9.9	-	-20.8
IMPROVE	**24-h $PM_{2.5}$**	-16.1	-	-27.0
STN	**24-h SO_4**	-25.8	-	-33.3
IMPROVE	**24-h SO_4**	-23.7	-	-26.3
STN	**24-h NO_3**	-11.3	-	-27.8
IMPROVE	**24-h NO_3**	-20.0	-	-53.5
STN	**24-h NH_4**	-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

[1] The percentages are calculated according to this formula: [(2010 value – 2006 value) /2006 value] * 100%. CASTNET - the Clean Air Status and Trends Network; AQS – the Aerometric Information Retrieval System Air Quality System; SEARCH - the Southeastern Aerosol Research and Characterization; GPCC - the Global Precipitation Climatology Centre; MODIS - the Moderate Resolution Imaging Spectroradiometer; IMPROVE – the Interagency Monitoring for Protected Visual Environmental; STN – the Speciated Trends Network. Note that IMPROVE did not contain NH4+ data for 2010. "-" indicates that the results of those variables not available from the WRF only simulation.

Table 3. Summary of set-up of Sensitivity Simulations

	Run 1	Run 2	Run 3	Run 4
Emissions	2006	2010	2006	2006
Meteorological ICONs/BCONs	2006	2010	2010	2010
Chemical ICONs/BCONs	2006	2010	2010	2006

Table 44. Absolute and percentage differences between monthly mean of observed / satellite-retrieved data and sensitivity simulations

		Obs 2010 – Obs 2006	Run 2 – Run 1	Run 3 – Run 1	Run 4 – Run 1
Jan	**CASTNET T2 (K/%)**	-3.5/ -1.3	**-2.0/ -0.7**	-1.9/ -0.7	-1.8/ -0.7
	CASTNET SWDOWN (Wm^{-2}/%)	-6.2/ -7.0	27.6/ 29.1	**-0.8/ -0.9**	-0.6/ -0.6
	MODIS CF (%/%)	2.7/ 4.2	**1.5/ 2.3**	1.4/ 2.1	1.4/ 2.1
	MODIS COT (/%)	-0.2/ -1.2	0.2/ 2.9	0.3/ 5.2	0.3/ 5.5
	MODIS AOD (/%)	-0.008/ -7.9	**-0.002/ -3.9**	0.008/ 15.3	0.01/ 28.0
	CASTNET Max 8-hr O_3 (ppb/%)	4.2/ 12.5	-2.9/ -9.8	-6.1/ -20.8	**0.7/ 2.4**
	STN $PM_{2.5}$ ($\mu g\ m^{-3}$/%)	-0.2/ -1.9	1.6/ 19.1	**1.4/ 16.5**	1.5/ 17.7
Jul	**CASTNET T2 (K/%)**	0.03/ 0.0	0.5/ 0.2	0.5/ 0.2	0.5/ 0.2
	CASTNET SWDOWN (Wm^{-2}/%)	-2.8/ -1.1	-7.4/ -2.6	-8.9/ -3.1	**-5.5/ -1.9**
	MODIS CF (%/%)	1.1/ 2.0	-1.8/ -3.4	-1.8/ -3.3	-1.5/ -2.8
	MODIS COT (/%)	-0.4/ -2.7	**-0.6/ -11.1**	-1.0/ -17.8	-0.9/ -16.5
	MODIS AOD (/%)	-0.06/ -31.0	0.04/ 58.3	0.06/ 79.4	0.04/ 50.9
	CASTNET Max 8-hr O_3 (ppb/%)	-4.8/ -9.2	-7.6/ -15.2	**-5.0/ -10.1**	8.6/ 17.2
	STN $PM_{2.5}$ ($\mu g\ m^{-3}$/%)	-0.5/ -3.7	**-0.5/ -4.5**	1.5/ 14.4	1.0/ 9.8

List of Figures