Geosci. Model Dev. Discuss., 8, 1639–1686, 2015
www.geosci-model-dev-discuss.net/8/1639/2015/
doi:10.5194/gmdd-8-1639-2015

Geoscientific Model Development Discussions
Open Access

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

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Received: 23 December 2014 – Accepted: 12 January 2015 – Published: 19 February 2015

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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. This 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 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 US 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. These results indicate a need to further

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improve the accuracy of emissions and chemical BCONs, the representations of SOA and chemistry–meteorology feedbacks in the online-coupled models.

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/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 US and less precipitation over NA (Stoeckenius et al., 2015; 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

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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) ap-

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proach for secondary organic aerosol (SOA) (hereafter WRF/Chem-CB05-MADE/VBS) has been recently developed by Wang et al. (2015). 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 of Yahya et al. (2015) 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 Yahya et al. (2015). In this 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 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. (2015) 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 paper complements the work of Stoeckenius et al. (2015) 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

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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)). Comparing to emissions in 2006, the annual emissions of SO_2 and NO_x decrease significantly in 2010, especially at the point sources (Fig. 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 Fig. S2. Although anthropogenic VOC emissions decrease over continental US (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., 2015). The total annual emissions of EC and POA also decrease but to a smaller extent over most areas of the continental US. The changes in annual and seasonal emissions of those species between 2010 and 2006 will affect simulated air quality and meteorology–chemistry interactions.

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. (2015) reported that the mid-tropospheric

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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 US in the spring of 2010 and less African dust reached the US in the summer of 2010 (Stoeckenius et al., 2015). As shown in Fig. 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 US 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.

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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. (2015). 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, 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

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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). Annual mean WS10 is overpredicted for both 2006 and 2010. 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). In this study, the Mass and Owens (2010) surface roughness parameterization is used 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. 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. 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. (2015) 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

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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 US 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 US at a 4 km × 4 km resolution. In this study, although the urban canopy model is used for

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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 Fig. S4, the spatial distribution of MB values for T2 for JFD 2010 by WRF/Chem show very large negative MBs over southeastern US compared to JFD 2006. T2 is also generally underpredicted over southeastern US 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 °C in eastern US, 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 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 Figs. 2 and 3, maximum 8 h 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 h O_3 in JFD 2010 over southeastern US are attributed to larger cold biases in T2 shown in Fig. 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 h O_3 is small. As shown in Fig. S4, the temperature biases for both years are relatively similar. Over northeastern US, the T2 bias is generally less than −0.1 °C for JJA in both years. However, as shown in Fig. 2, O_3 concentrations over northeastern US in JJA 2010 have negative biases whereas those over northeastern US in JJA 2006 have positive biases. In this case, emissions might play a significant

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role in the underprediction of O_3 concentrations over northeastern US in JJA 2010. Hourly average surface NO_x emissions decrease significantly over northeastern US in JJA from 2006 to 2010. As shown in Fig. 3, 2006 model performance for O_3 is generally good for all seasons and all networks.

According to Table 1 and Fig. 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 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, Ulas et al. (2015) 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. (2015) and Makar et al. (2015), the inclusion of aerosol indirect effects also tends to reduce O_3 mixing ratios, comparing to the models that simulate aerosol direct effect only or do not simulate aerosol direct and indirect effects (i.e., offline-coupled models).

Figure 5 shows spatial distribution of NMBs for $PM_{2.5}$ concentrations for JFD and JJA 2006 and 2010 against IMPROVE, STN, and SEARCH. Overall, JJA 2006 and JJA 2010 have similar spatial distribution patterns of NMBs for all sites over CONUS except for several sites in northwestern US where $PM_{2.5}$ concentrations are underpre-

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dicted for JJA 2010 but overpredicted for JJA 2006. However, many sites have positive NMBs over eastern and central US for JFD 2006, whereas more sites have negative NMBs over eastern and central US for JFD 2010. Statistics from Yahya et al. (2015) and Table 1 show that in general, the simulated concentrations of $PM_{2.5}$ and all $PM_{2.5}$ species decrease from 2006 to 2010, however, the Corr values for $PM_{2.5}$ and $PM_{2.5}$ species become worse in 2010 compared to 2006. As shown in Fig. 6, $PM_{2.5}$ concentrations for 2006 can be overpredicted or underpredicted, depending on seasons and networks, with an equal distribution of positive and negative NMBs. However for 2010, $PM_{2.5}$ concentrations tend to be underpredicted for all seasons and for all networks except for JFD against SEARCH. As shown in Fig. 7, NMBs for $PM_{2.5}$ species for 2006 at individual monitoring sites range from −40 to 60 %, while those for 2010 range from −80 to 80 %. The markers are more spread out covering a wider range of NMBs and NMEs for 2010 with more extremes as compared to the markers for 2006 clustered around the zero NMB line. NMEs for $PM_{2.5}$ species in 2006 remain below 100 %. NO_3^- concentrations are slightly underpredicted in 2006 against all networks; however, NO_3^- levels in 2010 are largely underpredicted, likely due to the large decrease in NO_x emissions from 2006 to 2010 and the increase in T2. The NMBs for IMPROVE and SEARCH OC remain low from 2006 to 2010, however, the NMEs increase 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 large NMEs and poor correlations for all $PM_{2.5}$ species compared to 2006.

Figure 8 shows the time series plots for 24 h 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

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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., January–March and July–September for $PM_{2.5}$). This could be attributed in part to the poor correlations of meteorological variables in 2010 compared to 2006. For example, poor predictions of WS10 can influence the transport and dry deposition of aerosols. Poor predictions of precipitation can impact 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 underpredictions in nitrogen dioxide (NO_2) concentrations (Yahya et al., 2015).

3.3 SOA evaluation for 2006 and 2010

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. (2015) 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 US from Offenberg et al. (2011) for 2006 only, and Pasadena, CA and Bakersfield, CA in western US 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 deter-

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mined 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 Figs. 9 and 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., 2015). 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 Fig. 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 Fig. 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 US.

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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 trends of NMBs and NMEs are quite similar for both seasons in both years. For JJA 2006 and 2010, all cloud variables are underpredicted. 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 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. 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 $\sim 4\,^{\circ}$C 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 WRF/Chem shows a small decrease (by

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−8.3 %) for the CASTNET sites. Comparing to 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.

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 h O_3 concentrations from CASTNET and EC from IMPROVE. CASTNET maximum 1 and 8 h O_3 concentrations change very little from 2006 to 2010 whereas WRF/Chem shows a moderate decrease of 14–15 %. 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 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.

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.

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4.1 Air quality predictions

Simulated air quality responds nonlinearly to the changes in emissions. Figures 11, S7–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 US 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 generally decrease in the US, except over eastern US in MAM and September, October, and November (SON), where there are increases. NH_3 emissions decrease, however, over eastern US in all seasons. The increase in NH_3 concentrations in MAM and SON could be attributed to a number of reasons including less NH_3 conversion to NH_4^+ to neutralize SO_4^{2-} and NO_3^- and less dispersion of NH_3 concentrations due to decreased wind speeds over eastern and southeastern US in MAM and SON, respectively, in 2010 compared to 2006. In JJA and SON, high OM concentrations in Canada are attributed to the enhanced impacts of BCONs by increasingly convergent flow in this region. OM is made up of both POA and SOA. An increase in VOC emissions in eastern US in MAM and SON leads to increases in OM concentrations. Decreases in VOC emissions in western US for all seasons lead to decreases in OM concentrations. The OM concentrations in some areas, however, do

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not follow a linear relationship with VOC emissions, such as southeastern US in JJA, where VOC emissions increase from 2006 to 2010 but OM concentrations decrease. A decrease in POA concentrations must dominate the overall decrease in OM concentrations, even under increased temperatures and biogenic VOC emissions in this area. $PM_{2.5}$ concentrations decrease for all seasons and most regions of the CONUS, which is attributed mainly to decreases in precursor gases, especially the inorganic precursors SO_2 and NO_x in eastern US Increased $PM_{2.5}$ concentrations in JFD and MAM in the Midwest are due to surface temperature decreases, dominating in this region (Stoeckenius et al., 2015). This in turn leads to increased particle nitrate concentrations (Campbell et al., 2014).

4.2 Meteorological predictions

Figure S10 compares the seasonal changes between 2010 and 2006 in several meteorological variables that affect air pollution including SWDOWN, T2, WS10, PBLH, and 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 °C in eastern US in JFD, and with reduced Precip in eastern or southeastern US in JJA and SON but increased Precip in northwestern US in MAM and JJA and in western US in JFD. ICONs and BCONs for skin temperatures shown in Fig. S3 greatly influence T2 shown in Fig. S10 for JFD and JJA.

Figures 12 and 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. (2015). Comparing to the differences in predictions of SWDOWN, T2, WS10, Precip, and PBLH between 2010 and 2006 WRF only simulation shown in Fig. S10 and WRF/Chem simulations shown in Figs. 12 and S11, the differences in those meteorological variables except for SWDOWN do not vary significantly between

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2010 and 2006 WRF simulations and between 2010 and 2006 WRF/Chem simulations. As shown in Fig. 12, the decrease in SWDOWN from 2006 to 2010 is larger over north-central and north-western US and the increase in SWDOWN is smaller over north-eastern and southwestern US 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 US than that by WRF. The differences between WRF and WRF/Chem are the largest for SWDOWN over northeastern US 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.

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 Fig. 12, where SWDOWN generally increases over most of the continental US, 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 US 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 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 $> 1 \times 10^{-6}$ kg kg^{-1} (Wu and Zhang, 2005). In this study, the monthly CF is then normalized over the total number of time steps and vertical layers, giving a value of CF between 0 and 1 in each grid cell. In contrast, the calculations of CDNC in the model depend on the

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supersaturation, aerosol concentrations, aerosol hygroscopicity and updraft velocity (Abdul-Razzak and Ghan, 2004). The changes in CF are controlled by large scale state variables including temperature and relative humidity, while CDNC depends on more complex changes in microphysical variables. The dominant CDNC decrease in MAM, JJA, and SON, is due to lower $PM_{2.5}$ concentrations, which in turn lower the effective number of cloud condensation nuclei. However, exception occurs in southeast US where $PM_{2.5}$ decreases but CDNC increases. This is because CDNC also depends on other variables including the amount of liquid water in the atmosphere. The cloud liquid water path over southeastern US increases, which may explain the increase in CDNC. The spatial pattern for precipitation correlates to that of CF. The spatial pattern of CWP also corresponds to a certain extent with CF. PBLH increases when the ground warms up during the day and decreases when the ground cools so PBLH might be intuitively related to SWDOWN and T2. However, this consistent trend is now obvious in the plots, because the simulated growth of the planetary boundary later (PBL) also depends on the surface sensible latent and heat fluxes and the entrainment of warmer air from the free troposphere (Chen, 2007).

4.3 Meteorology–chemistry feedback predictions

As shown in Table 1, similar to 2006, comparison of the performance of most meteorological variables between WRF/Chem and WRF for 2010 is improved in terms of NMBs when chemistry–meteorology feedbacks are included. This indicates the importance and benefits of inclusion of such feedbacks in online-coupled models. However, unlike 2006 for which both 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. (2015) analyzed differences in meteorological performance between WRF/Chem and WRF for

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2006. Figure S13 shows absolute 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. (2015).

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. 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 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 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), changes due to the changes in emissions (column 2, Run 2 – Run 3), changes due to the changes in chemical ICONs/BCONs (column 3, Run 3 – Run 4), and changes due to the changes in meteorology including ICONs/BCONs (column 4, Run 4 – Run 1) for January and July, respectively. Since the impact of ICONs is only important at the beginning of the simulations whereas the impact of BCONs per-

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sists throughout the simulations, the changes due to changes in chemical BCONs will dominate over those due to changes chemical ICONs/BCONs. Both figures show that the differences in the meteorology including ICONs/BCONs generated by WRF/Chem contribute to most of the differences in T2 and SWDOWN for both months. Changes in O_3 can be caused by increases in precursor emissions (e.g., BVOCs) in eastern US, decreases in chemical ICONs/BCONs in western US, and changes in meteorology in the entire US, leading to the dipole pattern in the differences of the spatial distribution of O_3 concentrations from 2006 to 2010 (Fig. 13, column 1). The net differences in $PM_{2.5}$ concentrations in January from 2006 and 2010 are mainly due to decreases in 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, the net changes from 2006 and 2010 are dominated entirely by changes in emissions that increase in the southeastern and central US but decrease in the remaining domain.

Table S1 in the Supplement shows the statistics NMB, NME, and Corr for a number of variables for the sensitivity simulations for January and July. The WRF/Chem performance against CASTNET T2 improves to a large extent in terms of NME and Corr for Runs 3 and 4 which use 2006 emissions, especially for January when Run 2 performs poorly. This indicates that at least for January, the inaccuracy of emissions may have contributed to the poor performance of T2 against CASTNET. For SWDOWN, Runs 3 and 4 improve the performance against CASTNET for January (with lower NMB, 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 perfor-

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mances 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. 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.

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 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 US in JJA and SON. The increases in biogenic VOCs are caused by increases in temperatures in 2010 in eastern US 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 US CDNC generally decreases over the US 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 mete-

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orological 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. 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 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. These results 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.

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/).

The Supplement related to this article is available online at doi:10.5194/gmdd-8-1639-2015-supplement.

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 US EPA (North American emissions processing), Environment Canada,

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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 US networks (AQS, CASTNET, IMPROVE, SEARCH, and STN). North American precipitation-chemistry measurements are provided by several US 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, US EPA. Cloud variables were provided by MODIS. We thank Georg Grell, NOAA, Christian Hogrefe, US 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.

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

Network or Site name	Variable	WRF Mean Obs	WRF Mean Sim	WRF Corr	WRF NMB (%)	WRF NME (%)	WRF/Chem Mean Obs	WRF/Chem Mean Sim	WRF/Chem Corr	WRF/Chem NMB (%)	WRF/Chem 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*	SOA	–	–	–	–	–	0.63	0.16	0.1	−75.3	78.3
Bakersfield, CA*	SOA	–	–	–	–	–	0.51	0.23	0.3	−55.3	65.9

Units are as follows: SWDOWN ($W m^{-2}$), GLW ($W m^{-2}$), OLR ($W m^{-2}$), T2 (°C), 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 ($\mu g m^{-3}$). 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.
*The observed SOA data are taken from Klendienst et al. (2012) and Lewandowski et al. (2013).

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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

The percentages are calculated according to this formula: [(2010value – 2006value)/2006value] × 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.

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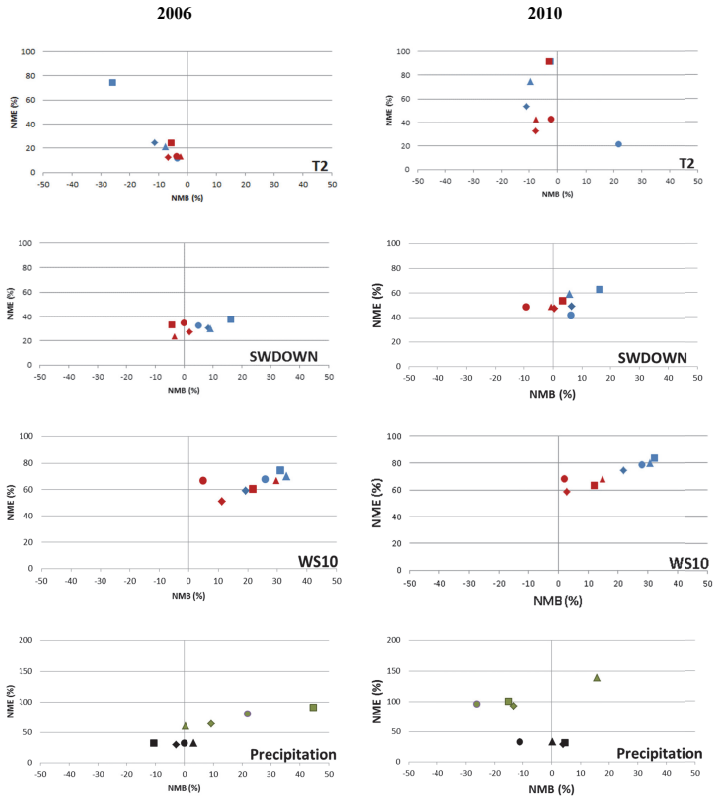

Figure 1. Comparison of seasonal plots of NMB vs. NME of various meteorological variables for 2006 (left column) and 2010 (right column) – T2 (temperature at 2 m), SWDOWN (downward shortwave radiation), WS10 (wind speed at 10 m) and Precipitation where the shapes represent different seasons (diamond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

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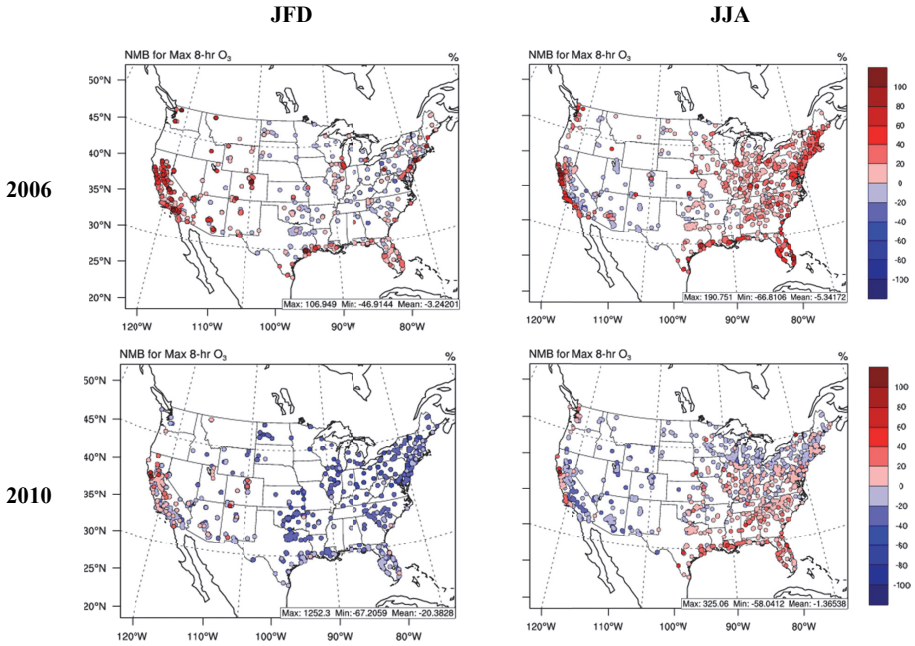

Figure 2. Spatial Distribution of NMB plots for JFD and JJA 2006 and 2010 for maximum 8 h O_3 concentrations based on evaluation against CASTNET, AQS and SEARCH.

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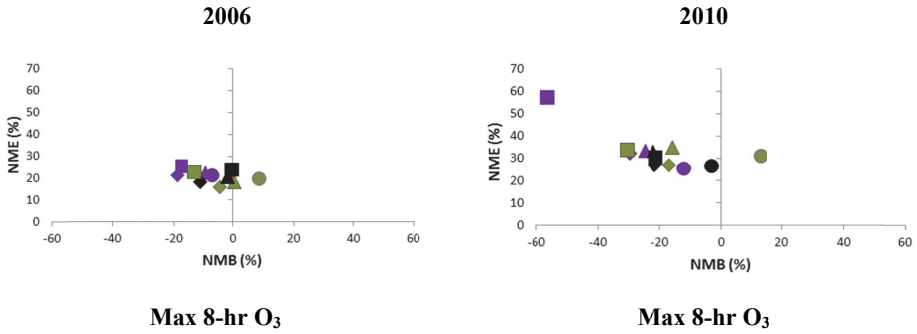

Figure 3. Comparison of seasonal plots of NMB vs. NME for maximum 8 h O_3 concentrations where the different shapes represent different seasons (diamond – MAM, circle – JJA, triangle – SON and square – JFD) and the different colors represent different observational data (purple – CASTNET, black – AQS and green – SEARCH).

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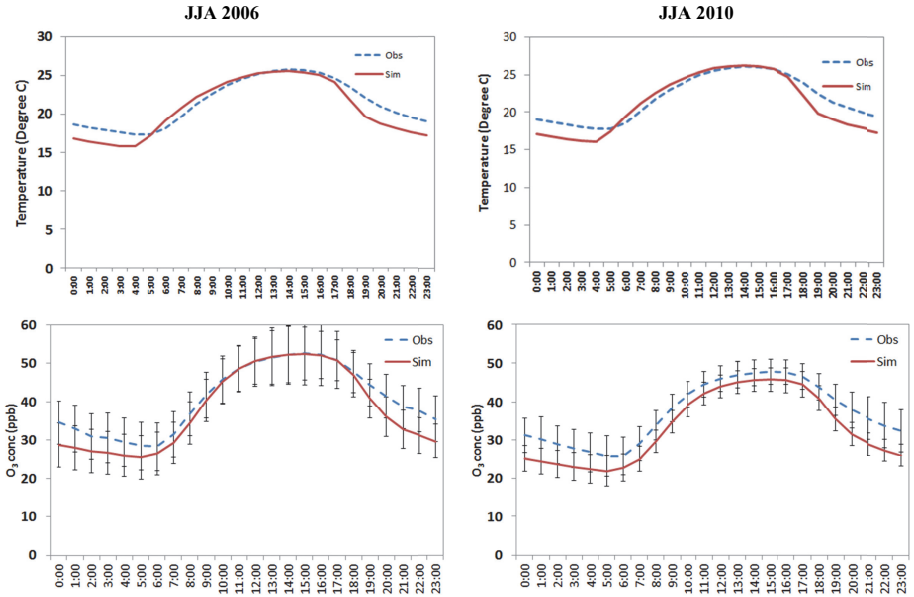

Figure 4. Diurnal variation of T2 (top row) and hourly O_3 concentrations (bottom row) against CASTNET for JJA 2006 and 2010.

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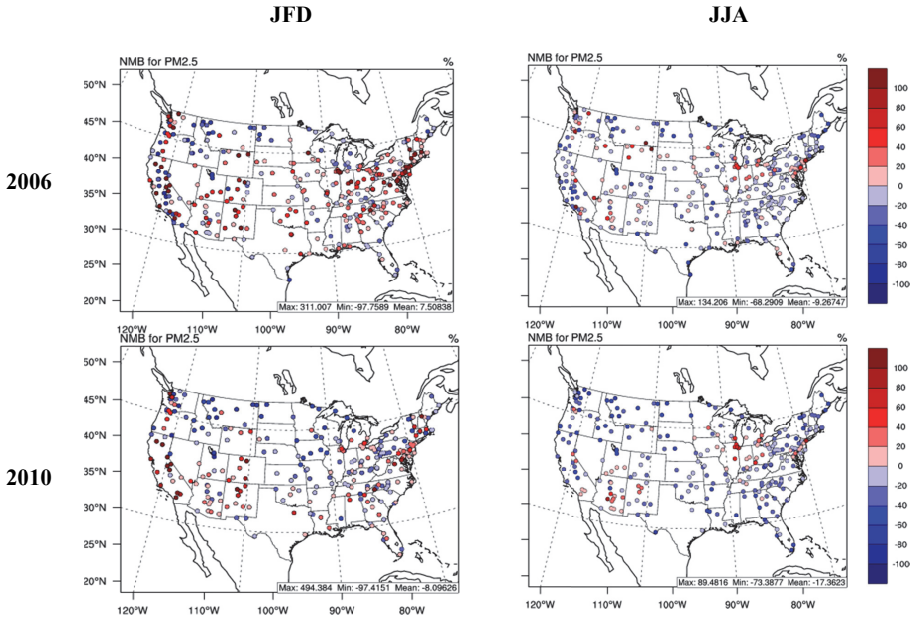

Figure 5. Spatial Distribution of NMB plots for JFD and JJA 2006 and 2010 for average 24 h $PM_{2.5}$ concentrations based on evaluation against the IMPROVE, STN and SEARCH sites.

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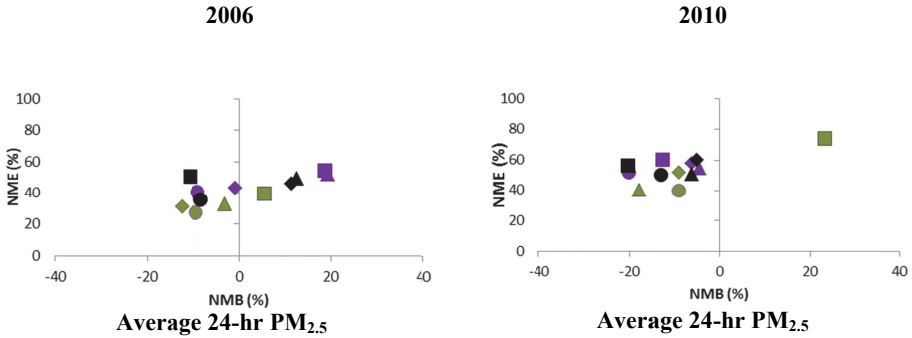

Figure 6. Comparison of seasonal plots of NMB vs. NME for average 24 h $PM_{2.5}$ concentrations where the different shapes represent different seasons (diamond – MAM, circle – JJA, triangle – SON and square – JFD) and the different colors represent different observational data (purple – IMPROVE, black – STN and green – SEARCH).

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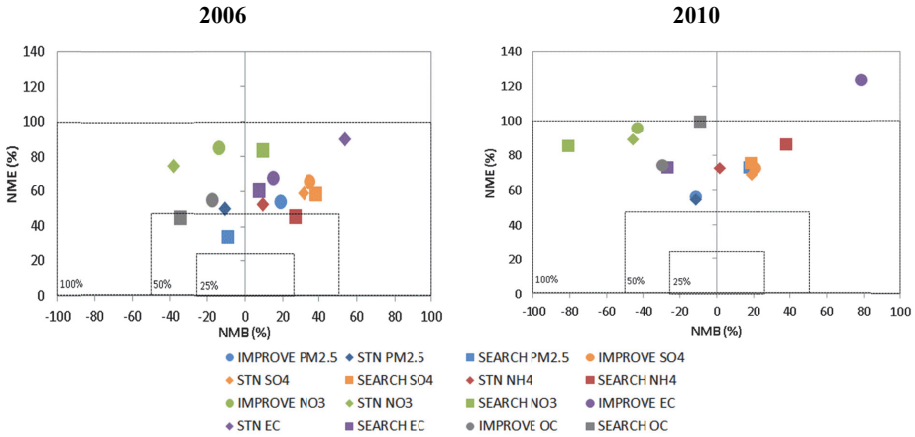

Figure 7. Plots of annual statistics (NMB vs. NME) for average 24 h $PM_{2.5}$ concentrations and $PM_{2.5}$ species against different observational networks.

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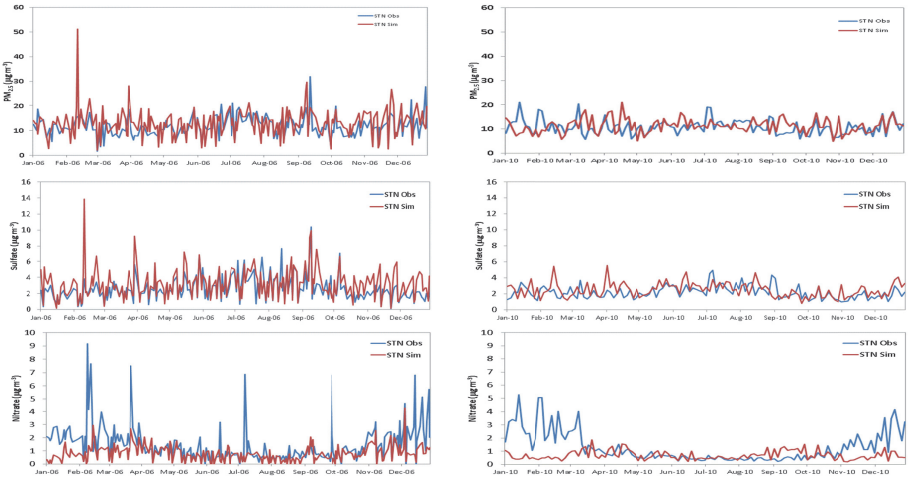

Figure 8. Time series of Obs vs. Sim $PM_{2.5}$, SO_4 and NO_3 concentrations against STN for 2006 and 2010.

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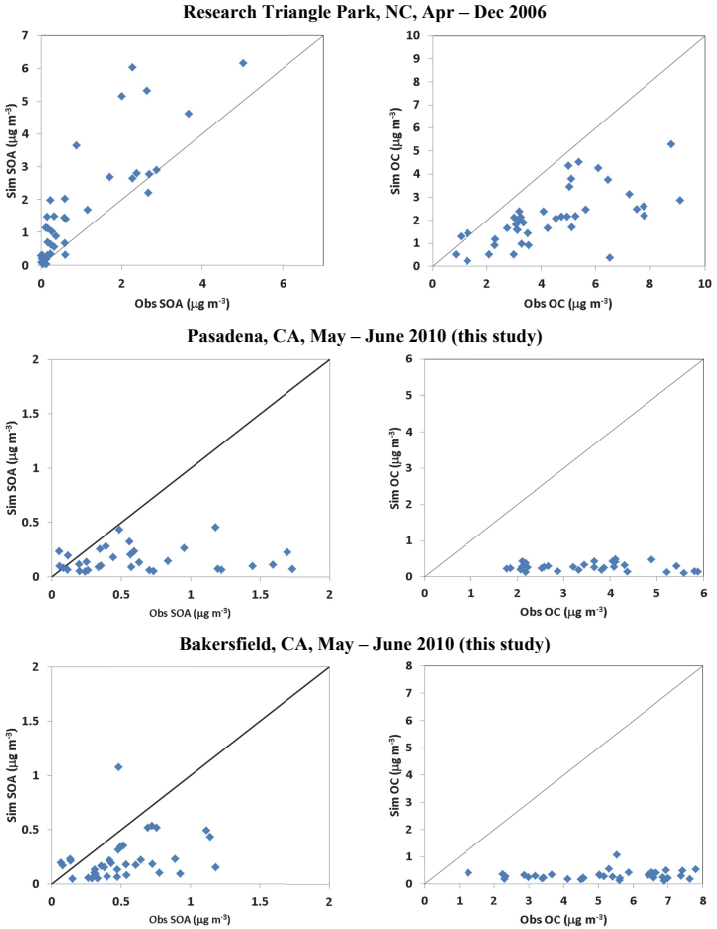

Figure 9. Scatter plots of SOA (left column) and OC (right column) concentrations at various sites.

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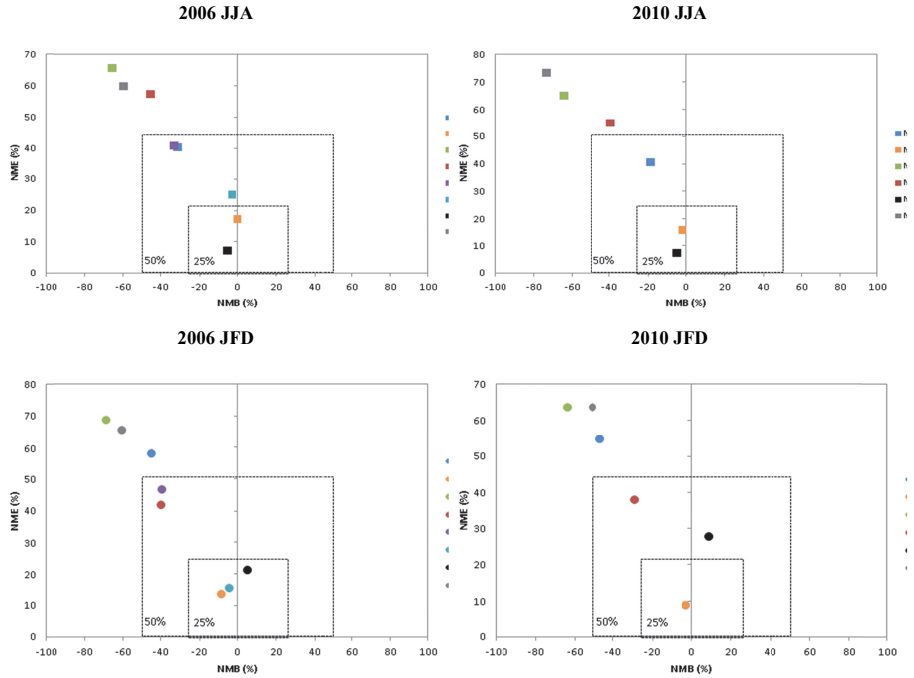

Figure 10. Comparison of soccer plots for JFD and JJA 2006 and 2010 evaluation of aerosol and cloud variables. MISR AOD, and SRB CF obs data was not available for 2010.

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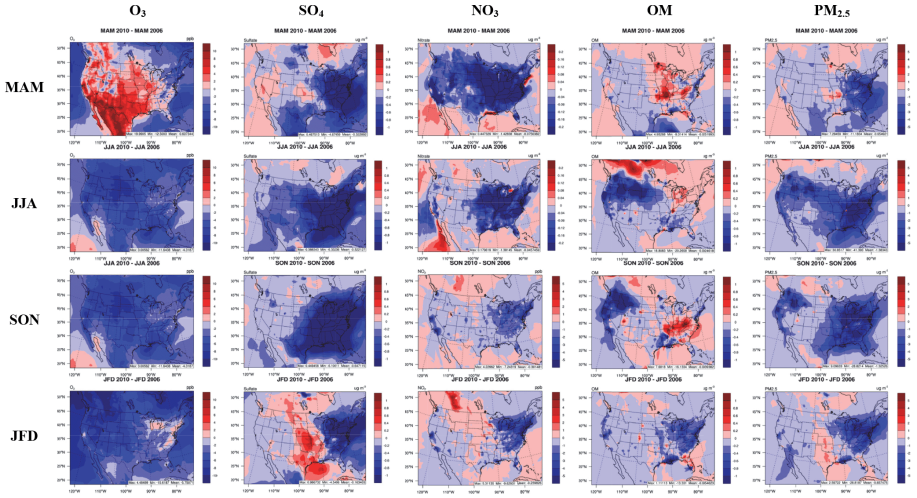

Figure 11. Changes in hourly average surface concentrations of O_3 and PM species from 2010 to 2006 (2010–2006).

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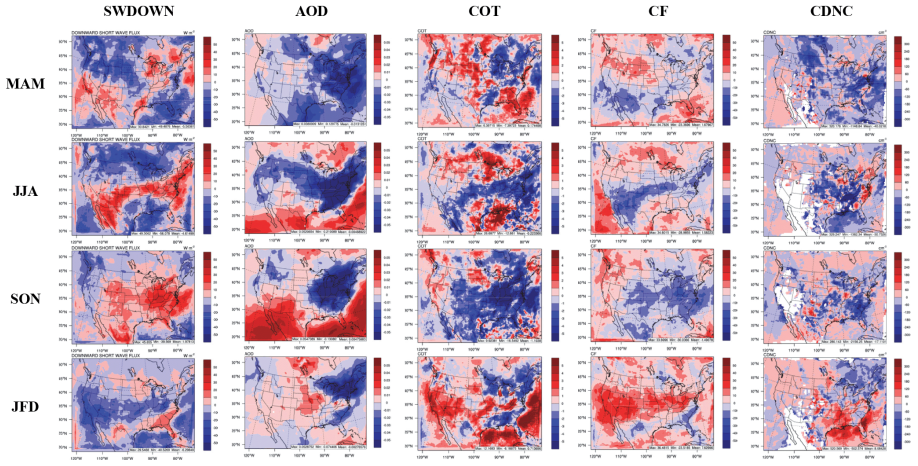

Figure 12. Changes in hourly average predictions of aerosol-cloud variables at surface from WRF/Chem simulations from 2010 to 2006 (2010–2006).

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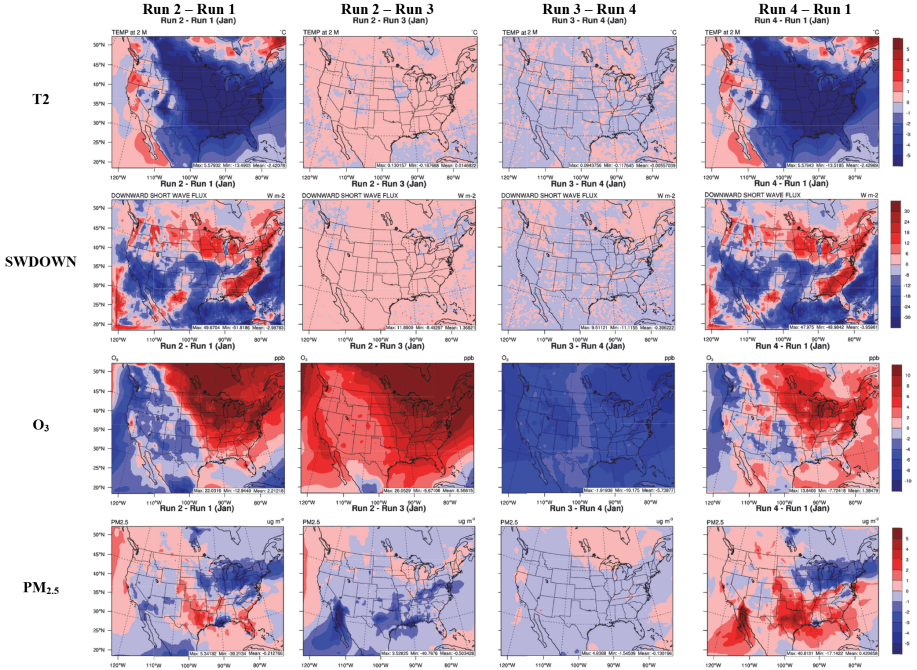

Figure 13. Spatial difference plots for January where Run 1: 2006 baseline simulations; Run 2: 2010 baseline simulations; Run 3: 2010 simulations with 2006 emissions and 2010 meteorology and chemical IC/BCs; Run 4: 2010 simulations with 2006 emissions and 2006 chemical IC/BCs and 2010 meteorology.

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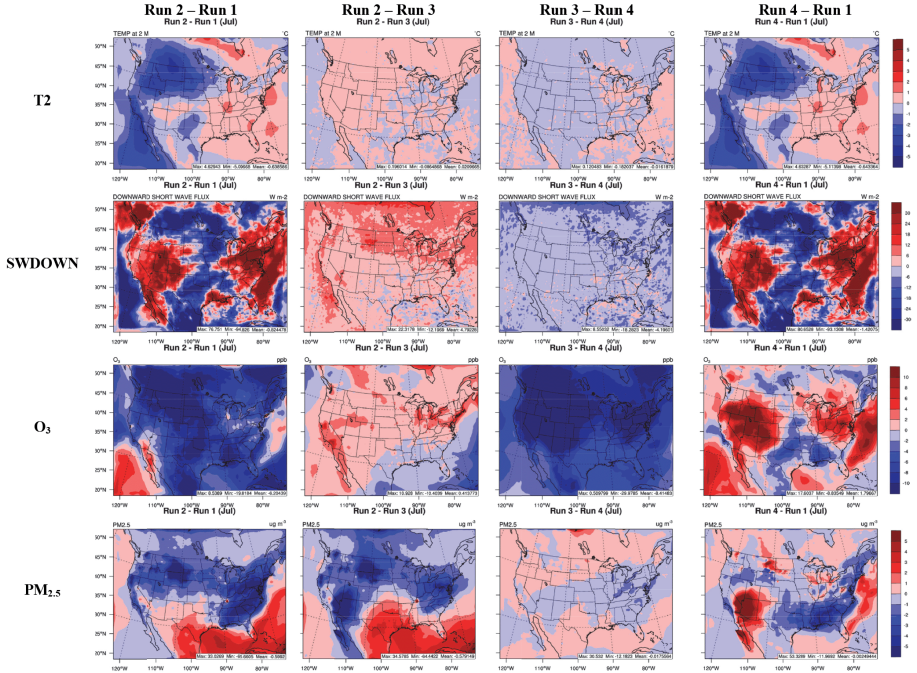

Figure 14. Spatial difference plots for July where Run 1: 2006 baseline simulations; Run 2: 2010 baseline simulations; Run 3: 2010 simulations with 2006 emissions and 2010 meteorology and chemical IC/BCs; Run 4: 2010 simulations with 2006 emissions and 2006 chemical IC/BCs and 2010 meteorology.

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