Comprehensive Evaluation of Multi-Year Real-Time Air Quality Forecasting Using an Online-Coupled Meteorology-Chemistry Model over Southeastern United States

Yang Zhang^{1,*}, Chaopeng Hong^{1,2}, Khairunnisa Yahya¹, Qi Li¹, Qiang Zhang², and Kebin He^{3,2}

¹Air Quality Forecasting Laboratory, North Carolina State University, Raleigh, NC, USA

²Center for Earth System Science, Tsinghua University, Beijing, P.R. China, 100084

³The School of Environment, Tsinghua University, Beijing, P.R. China, 100084

Abstract

An online-coupled meteorology-chemistry model, WRF/Chem-MADRID, has been deployed for real time air quality forecast (RT-AQF) in southeastern U.S. since 2009. A comprehensive evaluation of multi-year RT-AQF shows overall good performance for temperature and relative humidity at 2-m (T2, RH2), downward surface shortwave radiation (SWDOWN) and longwave radiation (LWDOWN), and cloud fraction (CF), ozone (O₃) and fine particles (PM_{2.5}) at surface, tropospheric ozone residuals (TOR) in O₃ seasons (May-September), and column NO₂ in winters (December-February). Moderate-to-large biases exist in wind speed at 10-m (WS10), precipitation (Precip), cloud optical depth (COT), ammonium (NH₄⁺), sulfate (SO₄²⁻), and nitrate (NO₃⁻) at the IMPROVE and SEARCH networks, organic carbon (OC) at IMPROVE, and elemental carbon (EC) and OC at SEARCH, aerosol optical depth (AOD) and column carbon monoxide (CO), sulfur dioxide (SO₂), and formaldehyde (HCHO) in both O₃ and winter seasons, column nitrogen dioxide (NO₂) in O₃ seasons, and TOR in winter. These biases indicate uncertainties in the boundary layer and cloud process treatments (e.g., surface roughness, microphysics cumulus parameterization), emissions (e.g., O₃ and PM precursors, biogenic, mobile, and wildfire emissions), upper boundary conditions for all major gases and PM_{2.5} species, and chemistry and aerosol treatments (e.g., winter photochemistry, aerosol thermodynamics). The model shows overall good skills in reproducing the observed multi-year

trends and inter-seasonal variability in meteorological and radiative variables such as T2, WS10, Precip, SWDOWN, and LWDOWN, and relatively well the observed trends in surface O₃ and PM_{2.5}, but relatively poor for column abundances of CO, NO₂, SO₂, HCHO, TOR, and AOD. The sensitivity simulation using satellite-constrained boundary conditions for O₃ and CO shows substantial improvement for both spatial distribution and domain-mean performance statistics. The model's forecasting skills for air quality can be further enhanced through improving model inputs (e.g., anthropogenic emissions for urban areas and upper boundary conditions of chemical species), meteorological forecasts (e.g., WS10, Precip) and meteorologically-dependent emissions (e.g., biogenic and wildfire emissions), and model physics and chemical treatments (e.g., gas-phase chemistry in winter conditions, cloud processes and its interactions with radiation and aerosol).

* *Corresponding author:* Yang Zhang, Department of Marine, Earth, and Atmospheric Sciences, Campus Box 8208, NCSU, Raleigh, NC 27695; e-mail: yang_zhang@ncsu.edu

Keywords

WRF/Chem-MADRID, southeastern U.S., discrete evaluation, categorical evaluation, satellite data, multi-year trend analysis

1. Introduction

Real-time air-quality forecasting (RT-AQF) of the concentrations of pollutants of special health concerns such as ozone (O₃) and fine particulate matter (PM_{2.5}) provides a basis for early air quality alerts and preventative actions that reduce air pollution and protect human health.

Increasing public awareness of adverse health impacts of ambient air pollution in both developed and developing countries and the availability of complex, deterministic three-dimensional (3-D) numerical models for RT-AQF have provided driving forces for the establishment and

advancement of RT-AQF. Despite substantial improvements of ambient air quality in major cities in many countries, the frequent occurrences of severe regional hazes in recent years in a number of countries such as China (e.g., Wang et al., 2014), India, and Singapore necessitate the continuous development and application of techniques for RT-AQF worldwide. A number of 3-D air quality models have been deployed for RT-AQF since the mid-1990s on global (e.g., Takigawa et al., 2007; Mangold et al., 2011) and regional scales (e.g., Carmichael et al., 2003; McHenry et al., 2004; McKeen et al., 2005; 2010; Yu et al., 2007, 2008; Eder et al., 2010). Kukkonen et al. (2011) reviewed 18 regional scale RT-AQF models that are currently used in Europe, among which, 3 out of 18 are online-coupled models. Zhang et al. (2012a, b) provided a comprehensive review of history, techniques, current status, and future research needs along with 9 global and 36 regional RT-AQF models that are currently used in Australia, North America, South America, Europe, and Asia, among which, 4 out of 9 global models and 5 out of 36 regional models are online-coupled models. Among those models, the 3-D RT-AQF models with coupled meteorology and chemistry such as the online-coupled Weather Research and Forecasting model with Chemistry (WRF/Chem) (Grell et al., 2005) are advanced tools for RT-AQF that can realistically represent the feedback mechanisms between meteorology and chemistry in the atmosphere. They, however, may not always outperform offline RT-AQF models, as there remain larger uncertainties in RT-AQF models than those originating from the feedback mechanisms, and not all RT-AQF models represent all feedback mechanisms that occur in the real atmosphere. The strengths and limitations of online-coupled models have been reviewed in several papers (e.g., Zhang 2008; Baklanov et al., 2014).

Since May 2009, WRF/Chem with the Model of Aerosol Dynamics, Reaction, Ionization, and Dissolution (MADRID) (WRF/Chem-MADRID) (Zhang et al., 2010a, 2012c) has been deployed by the lead author's group for RT-AQF in southeastern U.S. for ozone (O_3) season

(May-September) and winter season (December-February) (Chuang et al., 2011; Yahya et al., 2014a). The multi-year RT-AQF enables the assessment of the model's capability and robustness in forecasting major pollutants as well as their inter-annual and inter-season variability, and multi-year trends with the long-term forecasting data. In this work, multi-year forecasts of air quality and meteorology during 2009-2015 using WRF/Chem-MADRID are evaluated against surface and satellite-derived observations. The objectives are to evaluate the model's skill in forecasting the observed air quality and meteorology and their variation trends during 2009-2015 and to identify areas of model improvements for more accurate meteorological and chemical forecasts.

2. Model Description and Evaluation Protocol

2.1 Model Description

WRF/Chem-MADRID is an online-coupled meteorology and chemistry model. It was developed based on WRF/Chem version 3.0 (Grell et al., 2005) and CMAQ-MADRID (Zhang et al., 2004) with updates in gas-phase chemistry and aerosol treatments by Zhang et al. (2010a, b, 2012c). WRF/Chem-MADRID treats all major aerosol processes such as the thermodynamic equilibrium for both inorganic and organic species, new particle formation, condensation/evaporation, coagulation, gas/particle mass transfer, dry and wet deposition. Unlike offline-coupled air quality models, WRF/Chem-MADRID simulates aerosol direct and semi-direct feedbacks to photolysis, radiation, and planetary boundary layer (PBL) meteorology, as well as aerosol indirect effects on cloud and precipitation formation via many aerosol-cloud interaction processes. The physics and chemistry options used in this study follow those of Chuang et al. (2011) and Yahya et al. (2014a); they are kept the same for all forecasting periods since 2009. The physics options include the cloud microphysics of Lin et al. (1983); the Rapid

Radiative Transfer Model (RRTM) of Mlawer et al. (1997) for longwave radiation; the Goddard scheme of Chou et al. (1998) for shortwave radiation; the Yonsei University (YSU) PBL scheme of (Hong et al. 2006); the National Center for environmental Prediction, Oregon State University, Air Force, Hydrologic Research Lab (NOAH) LSM (Chen and Dudhia, 2001); and the Grell-Devenyi ensemble cumulus parameterization (Grell and Devenyi, 2002). The chemistry and aerosol-related options chosen include the 2005 Carbon Bond gas-phase chemical mechanism (CB05) (Yarwood et al., 2005); the Carnegie-Mellon (CMU) bulk aqueous-phase chemical kinetic mechanism (Fahey and Pandis, 2001), the MADRID1 aerosol module with 8 size sections over the PM aerodynamic diameter range of 0.025-11.630 μm of Zhang et al. (2004, 2010a, b, 2012c), and the aerosol activation of Abdul Razzak and Ghan (2002). A more detailed description of the model can be found in Chuang et al. (2011) and Yahya et al. (2014a).

2.2 RT-AQF Deployment and Inputs

The forecasting simulations are performed during the O_3 and winter seasons at a horizontal grid resolution of 12 km over an area in southeastern U.S. including the states of Mississippi (MI), Alabama (AL), Georgia (GA), Florida (FL), South Carolina (SC), North Carolina (NC), Tennessee (TN), Kentucky (KY), Virginia (VA), West Virginia (WV), and Delaware (DE), as well as small portions of Louisiana (LA), Arkansas (AR), Missouri (MS), Illinois (IL), Indiana (IN), Ohio (OH), and Maryland (MD). The hourly and daily forecast products are provided at http://www.meas.ncsu.edu/aqforecasting/Real_Time.html. This study analyzes forecast products during six O_3 and winter seasons between May 1, 2009 and February 28, 2015. The National Center for Environmental Prediction's (NCEP) meteorological forecast is downloaded at 7 p.m. (Local Standard Time) to initialize a 60-hr forecasting cycle using WRF/Chem-MADRID with 12-hr spin-up and 48-hr forecasting. The anthropogenic emissions are based on the projected 2009 emissions by the Visibility Improvement State and Tribal Association of the Southeast's

(VISTAS) from the 1999 National Emission Inventories (NEI) version 2 based on historical growth factors and assumed control strategies (Barnard and Sabo, 2008). Those emissions vary hourly and account for seasonal variations. For biogenic emissions, offline biogenic emissions available from the VISTAS emissions were originally used for the RT-AQF during May 1, 2009 and February 28, 2011. The online biogenic emissions from the Model for Gases and Aerosols from Nature (MEGAN) version 2 have been used since December 2011. Mineral dust emissions are simulated using online dust emission of Shaw (2008).

The VISTAS 2009 36-km CMAQ simulation results and those from the previous day's simulation are used to provide daily chemical boundary and initial conditions (BCONs and ICONs), respectively. One-week spin up simulation is performed for the first day of the first 60-hr forecasting cycle for each forecasting season.

2.3 Evaluation Datasets and Protocols

Zhang et al. (2012a) recommended both discrete and categorical evaluation for RT-AQF models, which are carried out for meteorological and chemical forecasts in this work. The PBL meteorological variables evaluated include temperature at 2-m (T2), relative humidity at 2-m (RH2), wind speed and direction at 10-m (WS10 and WD10), and daily precipitation (Precip). The chemical species evaluated include maximum 1-hr and 8-hr O₃, carbon monoxide (CO), sulfur dioxide (SO₂), nitric oxide (NO), nitrogen dioxide (NO₂), nitric acid (HNO₃), 24-hr average PM_{2.5} and PM_{2.5} species such as ammonium (NH₄⁺), sulfate (SO₄²⁻), nitrate (NO₃⁻), elemental carbon (EC), organic carbon (OC) and total carbon (TC = EC+OC). Given the low accuracy of anemometers at low wind speed conditions, the observed and simulated data pairs with the observed value below 0.771 m s⁻¹ are excluded in the statistical calculation following Olerud et al. (2005). A number of surface networks are used for model evaluation, as summarized in Table S1 in the supplementary material. These include the National Climatic

Data Center (NCDC), the AIRNow database, the Air Quality System (AQS), the Clean Air Status and Trends Network (CASTNET), the Interagency Monitoring of Protected Visual Environments (IMPROVE), the Speciated Trends Network (STN), and the Southern Aerosol Research and Characterization (SEARCH). While AIRNow, AQS, and STN include primarily urban and suburban sites, and NCDC, CASTNET and IMPROVE include mainly rural and remote sites. NCDC and SEARCH includes both urban and rural sites in southeastern U.S.

While 14 statistics defined in Zhang et al. (2006, 2012a) and Yu et al. (2006) are calculated against all surface network datasets in the discrete evaluation, the analysis in this study focuses on several commonly-used metrics including the mean bias (MB), normalized mean bias (NMB), the normalized mean error (NME), mean absolute gross error (MAGE), Root mean square error (RMSE), and correlation coefficient (R). The discrete performance statistical criteria for chemical forecasts are based on Zhang et al. (2006) which recommended the use of $NMBs \leq 15\%$ and $NMEs \leq 30\%$ to indicate a satisfactory performance for O_3 and $PM_{2.5}$. For meteorological variables, Tesche and Tremback (2002) suggested a good performance with $MB \leq 0.5 \text{ m s}^{-1}$ for WS10, $MB \leq 10$ degrees and $MAGE \leq 30$ degrees for WD10, and $MB \leq 0.5 \text{ K}$ and $MAGE \leq 2 \text{ K}$ for T2. However, such criteria were developed for meteorological simulations with data assimilation. Data assimilation is not used in this work because it masks the feedbacks between chemistry and meteorology. The model performance may not be as well as those with data assimilation. Brunner et al. (2014) evaluated meteorological simulations for the year of 2010 from eight simulations of WRF version 3.4 with different combinations of physics options and found that the monthly MBs of T2 are within 2 K and MBs of WS10 are within 1.7 m s^{-1} . The reported NMBs of Precip simulated by WRF range from -88% to 66% (e.g., Zhang et al., 2010c; Yahya et al., 2014a, b, 2015a; Penrod et al., 2014). NMBs within $\pm 30\%$ are considered to be acceptable performance for Precip. Categorical statistics are calculated for the maximum 1-hr

and 8-hr O₃ and 24-hr average PM_{2.5} against near-real time observations from AIRNow in terms of accuracy (A), critical success index (CSI), probability of detection (POD), bias (B), and false alarm ratio (FAR), as defined in Kang et al. (2005) and Zhang et al. (2012a). The threshold values are 80 ppb for the maximum 1-hr O₃; 60 ppb for maximum 8-hr O₃; and 15 µg m⁻³ for 24-hr average PM_{2.5} following Chuang et al. (2011). For categorical evaluation, satisfactory performance would yield values close to 1 for A, CSI, and POD and a value close to 0 for FAR. For B, a value of 1 would indicate no bias, and a number greater than 1 means that the model forecasts more exceedances than observed, and vice versa. In addition to domain-mean discrete and categorical statistics, the forecasted meteorological variables and chemical concentrations are evaluated using available observations in terms of domain-mean spatial distributions and site-specific hourly variations. The representative urban and rural sites selected include Atlanta, Georgia; Charlotte and Raleigh in North Carolina; Louisville, Kentucky; Birmingham, Alabama; and Jacksonville, Florida.

In addition to surface evaluation, satellite data are used to assess the model's capability in forecasting column values of meteorological, radiative, and chemical variables, as summarized in Table S1. Such evaluations have not been previously performed for RT-AQF models. These include Precip from the Global Precipitation Climatology Project (GPCP), downward surface shortwave radiation (SWDOWN) and longwave radiation (LWDOWN) from the Cloud's and the Earth's Radiant Energy System (CERES), cloud fraction (CF), aerosol optical depth (AOD), and cloud optical depth (COT) from the Moderate Resolution Imaging Spectroradiometer (MODIS), tropospheric CO column abundances from the Measurements of Pollution in the Troposphere (MOPITT), tropospheric column abundances of NO₂, formaldehyde (HCHO), and sulfur dioxide (SO₂), as well as tropospheric ozone residuals (TOR) from the Ozone Monitoring Instrument (OMI)/ Microwave Limb Sounder (MLS). All satellite data used are level-3 monthly average

(except for column SO₂, which is daily average because monthly average is not available) retrieval data that have been validated and quality assured by data providers (Martin, 2008). Following Zhang et al. (2009), the model outputs for all column variables except for TORs are vertically integrated up to the tropopause and averaged at the same satellite crossing time to generate the tropospheric amounts in order to match the satellite data. Column variables are evaluated in terms of domain-mean discrete statistics and spatial distributions.

3. Evaluation of Model Performance

3.1 Evaluation of Meteorological Variables

Meteorological forecasts are evaluated to understand their influence on chemical forecasts. The meteorological performance for three O₃ seasons and three winter seasons during May 1, 2009-February 28, 2012 has been evaluated in Yahya et al. (2014a). This study focuses on the evaluation of three O₃ seasons and three winter seasons during May 1, 2012-February 28, 2015. Table 1 summarizes domain-mean performance statistics for T2, RH2, WS10, and WD10 against data from CASTNET, NCDC, and SEARCH, Precip against data from CASTNET, NCDC, and GPCP, SWDOWN and LWDOWN against CERES, and CF and COT against MODIS during these three O₃ seasons and three winter seasons.

3.1.1 Ozone Seasons

MBs for T2 range from 0.5-2.1 °C, 0.6-1.8 °C, and 0.9-2.6 °C and MAGEs range from 4.0-4.2°C, 3.7-4.0°C, and 3.9-4.7°C against data from CASTNET, NCDC, and SEARCH, respectively. The values of R for T2 range from 0.6-0.7 at CASTNET and NCDC, and 0.3-0.5 at SEARCH. Low R values at the SEARCH sites indicate possible compensation of large positive and negative biases at different sites. While the MBs and MAGEs of T2 are larger than 0.5 K and 2 K, respectively, suggested by Tesche and Tremback (2002), they fall into the typical

ranges of MBs ($< 2\text{ }^{\circ}\text{C}$) reported for this and newer versions of WRF and WRF/Chem in the literature (e.g., Brunner et al., 2014). Moderate warm biases in T2 are mainly caused by moderate overpredictions in SWDOWN with NMBs of 14%, 17.6%, and 27.6%, and moderate underpredictions in CF with NMBs of -8.3%, -12.7%, and -14.5% at the CASTNET, NCDC, and SEARCH sites, respectively. In this version of WRF, sub-grid cloud feedbacks to radiation are neglected in the cumulus parameterization, contributing in part to the overpredictions in SWDOWN (Alapaty et al., 2012). Limitations in the surface layer and shortwave radiation schemes also contribute to the overpredictions in SWDOWN. The large underpredictions of COT with NMBs of -65.8% to -60.3% reflect the poor ability of the model in simulating cloud variables, due to the limitations in the parameterizations of cloud dynamics, thermodynamics, and microphysics, and interactions with aerosols (Zhang et al., 2012c, d, 2015). The model simulates LWDOWN well, with NMBs within 2%.

The warm biases in T2 directly affect RH2 forecasts. Moderate underpredictions occur in RH2 with MBs of -16.4% to -9.7%, -14.6% to -6.9%, and -20.5% to -10.4%, at the CASTNET, NCDC, and SEARCH sites, respectively. The values of R of RH2 are lower, ranging from 0.2-0.4 at all sites. The model simulates WS10 at the NCDC sites well with MBs of 0.2-0.4 m s^{-1} , MGAEs of 1.8 m s^{-1} , and NMBs of 4.8-5.2%. However, the model moderately or significantly overpredicts WS10 at the SEARCH and CASTNET sites with MBs of 0.3-0.9 m s^{-1} and 1.6-1.8 m s^{-1} , MGAEs of 1.2-1.3 m s^{-1} and 1.9-2.1 m s^{-1} , and NMBs of 15.1-40.9% and 65.6-100.5%, respectively. The MBs at all sites are generally within 1.7 m s^{-1} reported by Brunner et al. (2014) for simulations with WRF version 3.4 and at the NCDC sites they are even smaller than a performance indicator value of 0.5 m s^{-1} suggested by Tesche and Tremback (2002) for simulations with data assimilation. Similar large overpredictions of WS10 by WRF have been reported by a number of studies (e.g., Penrod et al., 2014; Yahya et al., 2014a; Brunner et al.,

2014). The WS10 overpredictions are due in part to unresolved surface roughness and topographical features by the surface drag parameterization used in WRF and in part to the use of coarse horizontal and vertical resolutions in the forecasting simulations (Cheng and Steenburgh, 2005; Mass and Ovens, 2011). Comparing to the NCDC sites that were carefully selected for meteorological measurements, the SEARCH and CASTNET sites were selected for air quality measurements, and many sites are difficult to be resolved at a spatial grid resolution of 12-km because of complex topography and surfaces. MBs for WD10 range from 16.3-29.0°, 42.4-47.9°, and 1.7-24.4° and MAGEs range from 79.1-86.2 °C, 85.1-86.1 °C, and 76.2-94.1°C against data from CASTNET, NCDC, and SEARCH, respectively. The values of MBs and MAGEs are much higher than 10 and 30 degrees, respectively, suggested by Tesche and Tremback (2002), indicating a poor performance for WD10 that is partly because the data assimilation is not used and partly because the surface roughness and topographic features cannot be resolved. The values of R for WD10 range from 0.6-0.7 at CASTNET and NCDC, and 0.3-0.6 at SEARCH. These results indicate certain limitations in the YSU PBL and the Monin-Obukhov surface layer schemes used in resolving main features of the PBL meteorology, particularly over complex terrain with uneven surface topography and mountainous regions (e.g., the Appalachian mountains).

Precip is moderately to significantly overpredicted with NMBs of 52.0-56.2%, 34.4-49.7%, and 29.8-54.6% against data from CASTNET, NCDC, and GPCP, respectively, they are mostly beyond the acceptable performance range of $\pm 30\%$. Similar large overpredictions of Precip by WRF or WRF/Chem have been reported in many studies (e.g., Caldwell et al., 2009; Zhang et al., 2010c, 2012c, d). R values during the O₃ seasons are low, ranging from ~0.0 to 0.4. Figure S1 in the supplementary material compares the spatial distributions of forecasted Precip with GPCP Precip in the O₃ seasons. The forecasted Precip is largely overpredicted over most

areas in the simulation domain. Such large biases and poor correlation can be attributed to three main reasons. First, as reported by Zhang et al. (2010c), the Grell-Devenyi ensemble cumulus parameterization has a tendency to overpredict frequency and the intensity of afternoon convective rainfall. Second, the Purdue Lin microphysics also has a tendency to overpredict cloud ice, graupel, and surface rainfall (Zhang et al., 2012d). Third, as reported in Alapaty et al. (2012), neglecting sub-grid cloud feedbacks to radiation in the cumulus parameterization can overpredict SWDOWN, resulting in unrealistically-large surface forcing for convection thus overpredictions in Precip. Those limitations explain the predicted excessive convection and non-convection rain. While the warm biases in T2 and SWDOWN can lead to higher O₃ and PM_{2.5}, the positive biases in WS10 and Precip and the negative biases in CF and COT can lead to lower O₃ and PM_{2.5}. These effects may compensate each other in chemical forecasts.

3.1.2 Winter Seasons

The MBs for T2 in winter are larger than those in the O₃ season in 2012 but smaller than those in the O₃ seasons in 2013-2014 at the CASTNET and NCDC sites, with a range of 0.7-1.0 °C and 0.8-1.2 °C, respectively. The MB at the SEARCH sites is 1.1 °C during 2014-2015 winter, but -5°C and -3 °C, respectively, in winters during 2012-2013 and 2013-2014. During those winters, heavy snowfall occurred over a large areas in southeastern U.S., particularly during the record-cold winter in Jan-Feb., 2014. The cold biases at the SEARCH sites indicate that the model tends to underestimate the snow melting rates in southeastern U.S. and the effects of urban heat island during winters. RH2 are better forecasted in winters than in O₃ season at all sites except for SEARCH during 2012-2013 and 2013-2014 winters during which large cold biases in T2 occur. Similar to the O₃ season, WS10 in winters is simulated well at the NCDC sites with MBs of 0.2-0.8 m s⁻¹ but largely overpredicted at the CASTNET and SEARCH sites with MBs of 1.6-2.5 and 0.2-1.5 m s⁻¹ because of the model's limited capability in resolving

surface roughness and topographical features. WD10 forecasts are similar to those in the O₃ season at the CASTNET and NCDC sites but worse at the SEARCH sites with MBs of 15.9-30.8°, 38.7-46.7°, and 22.2-46.6° and MAGEs of 89.8-98.3 °C, 92.8-97.5 °C, and 86.7-97.6°C against data from CASTNET, NCDC, and SEARCH, respectively. Comparing to the O₃ seasons, MBs of Precip during winters are smaller at the NCDC sites but similar or slightly worse at other sites, with NMBs of 56.1-60.1%, 19.4-42.2%, 36.9-67.3% against data from CASTNET, NCDC, and GPCP, respectively. As shown in Figure S1, the forecasted Precip is overpredicted in winters over most areas in the simulation domain. The spatial distributions of forecasted Precip with GPCP Precip correlate each other better in winter than in the O₃ seasons, with higher R values of 0.2-0.7. Similar to the O₃ seasons, the model simulates well LWDOWN but moderately overpredicts SWDOWN in winters. Relatively larger underpredictions occur in CF, with NMBs of -23.9% to -18%, leading to slightly larger underpredictions in COT than those during the O₃ seasons. Comparing to the O₃ seasons, the R values are generally higher for all meteorological variables except for CF and COT during winters, indicating that the model can better simulate the spatial/temporal variations of most meteorological variables during winters than in warm seasons.

3.2 Discrete, Spatial, and Temporal Evaluation of Surface Chemical Forecasts

The chemical performance during May 1, 2009-February 28, 2012 has been evaluated in Yahya et al. (2014a). Table 2 summarizes domain-mean performance statistics for chemical species at surface and chemical column abundances during three O₃ seasons and three winter seasons during May 1, 2012-February 28, 2015.

3.2.1 Ozone Seasons

During the three O₃ seasons in 2012-2014, as shown in Table 2a, the maximum 1-hr O₃ mixing ratios are well forecasted with NMBs within $\pm 15\%$ against data at AIRNow, AQS,

CASTNET, and SEARCH (except for SEARCH in 2013 where the NMB is 17%). Maximum 8-hr O₃ mixing ratios are also well forecasted with NMBs within $\pm 15\%$ in 2012 and 2014 but slightly higher NMBs (15-22%) in 2013 at all sites. Larger overpredictions in maximum 1-hr and 8-hr O₃ mixing ratios in 2013 comparing to 2012 and 2014 may be caused by higher warm biases in T2 and greater overpredictions in NO_x (indicated by NMBs of 36% for NO and 56% for NO_x at the SEARCH sites). Higher T2 cause higher emissions of biogenic volatile organic compounds (BVOCs), which also contribute to higher O₃ formation. The high positive biases in NO₂ and other trace gases such as CO and SO₂ at the SAERCH sites may be caused by overestimation of their emissions and also the use of 12-km that cannot represent emissions at those sites. Pan et al. (2014) showed that the use of lower NO_x emissions projected for 2012 than those in 2005 can reduce the positive bias in O₃ forecast during July 2011. Although NO_x mixing ratios are also significantly overpredicted in 2014, smaller warm biases in T2 in 2014 than in 2013, resulting in lower BVOCs emissions, and thus smaller O₃ overpredictions. Although there are no observed BVOCs emissions and mixing ratios for evaluation, the NMBs of OCs are 15% in 2013 but 1% in 2014 and secondary organic aerosol (SOA) dominates OC in southeastern U.S., supporting higher BVOCs emissions and mixing ratios in 2013 than in 2014. Figure 1 (a) compares several discrete statistics of O₃ against data from AIRNow for the six O₃ seasons during 2009-2015. The MBs range from -2.8 to 6.9 ppb and -1.8 to 6.9 ppb for maximum 1-hr and 8-hr O₃ mixing ratios, respectively. The highest and the second highest NMBs for the maximum 1-hr and 8-hr O₃ mixing ratios occur in the O₃ seasons in 2013 and 2009, respectively, with NMBs of 15% and 17.0% in 2013 and 9.6% and 8.5% in 2009. The model's skill in terms of NMEs, RMSEs, and R values is overall similar among all six O₃ seasons. NMEs, RMSEs, and R values are 19.9-26.7%, 13.1-17.0 ppb, 40-60% for maximum 1-hr O₃, and 19.6 to 27.5%, 11.4-14.2 ppb, and 37.5-60% for maximum 8-hr O₃. At sites from

other O₃ measurement networks such as CASTNET, AQS, and SEARCH, the performance statistics for O₃ in O₃ seasons during 2012-2015 in this work are overall similar to those in 2009-2012 shown in Yahya et al. (2014).

Figure 2 shows forecasted maximum 1-hr and 8-hr O₃ mixing ratios overlaid with all available observations during the three O₃ seasons in 2012-2014. Figure S2 shows the corresponding spatial distributions of MBs. In 2013, the model overpredicts at many sites in NC, GA, KY, and AL when the observed maximum O₃ mixing ratios were below 45 ppb, leading to the largest overpredictions among three O₃ seasons and relatively low R values of 0.4-0.5. In 2012, the model captures well the high O₃ mixing ratios in MD, northern GA, eastern TN, western OH, northwestern WV, and regions along the border of IN and KY, although it tends to overpredict at some sites in NC, GA, and KY and underpredicts at some sites in IL, IN, and OH. The overpredictions and underpredictions of O₃ at different sites over different time periods compensate, leading to relatively good R values of 0.5-0.6. In 2014, the observed O₃ mixing ratios are slightly lower than 2012 and 2013, partially because only forecasted results from May-July (MJJ) are averaged (Note that the results in August-September were lost due to the failure of backup drives containing such data). The model captures well the high O₃ mixing ratios in NC (including the hot dots in western NC), GA, IN, KY, VA, although it underpredicts a few hot spots in MD and the border regions between OH and IN. Similar spatial distributions and correlation are found for maximum 8-hr mixing ratios, despite slightly larger overpredictions at some sites in NC, VA, WV, GA, and AL in 2012, and at most sites in 2013. Figure 3 compares forecasted and observed hourly O₃ mixing ratios at the selected six urban sites. The model reproduces well their observed diurnal and daily variations at all six sites in 2012 and MJJ 2014. Larger discrepancies are found at all cities, in particular, Birmingham, Atlanta, and Louisville.

As shown in Table 2a, forecasted PM_{2.5} concentrations agree very well with the observations from AIRNow with NMBs of -4% to 15% and from STN with NMBs of 9-12%, but moderately overpredicted at the IMPROVE and SEARCH sites, with NMBs of 8-25% and 39-53%, respectively. The PM_{2.5} overpredictions are the results of overpredictions of SO₄²⁻ and NO₃⁻ (no observations of NH₄⁺ are available) at the IMPROVE sites, and SO₄²⁻, NO₃⁻, and NH₄⁺ at the SEARCH sites. The overpredicted inorganic PM_{2.5} may be caused by overestimates in the emissions of precursors such as SO₂, NO_x, and NH₃. As shown in Table 2a, the SO₂ and NO₂ mixing ratios at the SEARCH sites are significantly overpredicted with NMBs of 99-725% and 49-56%, respectively. The NO mixing ratios are also overpredicted by 36% and 222% in 2013 and 2014, respectively. The large biases in those precursor gases indicate uncertainties in projected 2009 emissions that are used for RT-AQF during 2009-2015, in particular, such emissions do not reflect the continuous reductions in SO₂ and NO₂ emissions since 2009 as reported in several studies (e.g., Pan et al., 2014). Warm biases in T2 at all sites also contribute to higher inorganic PM_{2.5} because of higher photochemical oxidation rates during the O₃ seasons. Despite overpredictions in WS10 and Precip which tend to reduce PM_{2.5} concentrations, the impacts of overestimated precursor emissions and warm biases on PM_{2.5} formation dominate, leading to a net moderate PM_{2.5} overprediction at all sites. Unlike IMPROVE, STN, and SEARCH, inorganic PM_{2.5} concentrations at the CASTNET sites are mostly underpredicted, likely due in part to the underestimates of anthropogenic of SO₂, NO_x, and NH₃ at remote sites and national parks or the impact of their long-range transport from emissions at nearby urban/rural sites, and in part to the larger wet biases in Precip than at other sites, which scavenges more inorganic PM_{2.5} from the atmosphere at the CASTNET sites. While the model simulates well EC, OC, and TC concentrations at the IMPROVE sites, it underpredicts EC, OC, and thus TC at the SEARCH sites. Such differences are related to different site characteristics

(rural/remote sites in the IMPROVE network vs. urban/rural sites in southeastern U.S. in the SEARCH network) as well as possible underestimates of EC and OC emissions at the SEARCH sites during O₃ seasons.

Figure 1 (a) compares several discrete statistics of PM_{2.5} against data from AIRNow for the six O₃ seasons during 2009-2015. The MBs range from -1.3 to 1.4 $\mu\text{g m}^{-3}$ and NMBs range from -10.1% to 14.7%, indicating a very good performance for PM_{2.5} for all six O₃ seasons. The ranges of NMEs, RMSEs, and R values are 35.8-40.4%, 5.1-8.7 $\mu\text{g m}^{-3}$, and 0.3-0.4, respectively. The model's skill in terms of NMEs, RMSEs, and R values is overall similar among all six O₃ seasons at sites from AirNow, with slightly higher NMEs but lower RMSEs and R values than forecasted O₃ during all six O₃ seasons. Compared to the performance statistics for PM_{2.5} at sites from IMPROVE, STN, and SEARCH in O₃ seasons during 2009-2011 shown in Yahya et al. (2014), those in O₃ seasons during 2012-2014 are worse (particularly at SEARCH sites). Several reasons may contribute to the worse performance of PM_{2.5} during 2012-2014 than during 2009-2011. First, primary PM emissions and the emissions of PM_{2.5} precursors used in the simulations may be higher than actual emissions during those years (resulted from the use of the same emissions as 2009-2011). This leads to higher overpredictions for inorganic PM concentrations during 2012-2014 than during 2009-2011. Second, uncertainties may exist in the spatial allocations of these emissions in both seasons, leading to heterogeneity in model performance at sites from different networks. This uncertainty may explain larger biases in EC and OC predictions during 2012-2014 than during 2009-2011 at SEARCH sites in O₃ season. Third, T2 predictions show larger warm biases in O₃ seasons during 2012-2014 than 2009-2011 at SEARCH sites, which favor the formation of (NH₄)₂SO₄ and thus contribute to higher overpredictions in PM_{2.5} concentrations. As shown in Figures 2 and S2, forecasted PM_{2.5} concentrations agree well spatially with observations in all three O₃ seasons, indicating that the relatively low R values may be mainly

due to mismatching between forecasted and observed hourly $\text{PM}_{2.5}$ concentrations. Such mismatching can be illustrated in Figure 4. For example, in 2012, the model overpredicts $\text{PM}_{2.5}$ concentrations at Atlanta when observed concentrations were relatively low (e.g., July 9-September 30, 2012), but underpredicts $\text{PM}_{2.5}$ concentrations at Louisville when observed concentrations were relatively high (e.g., June 27-July 10, 2012). In 2013, hourly $\text{PM}_{2.5}$ concentrations at Birmingham and Atlanta are largely overpredicted, contributing to large overpredictions and low R values of $\text{PM}_{2.5}$ against data from SEARCH.

3.2.2 Winter Seasons

As shown in Table 2b and Figure 1(b), unlike the O_3 seasons during which O_3 mixing ratios are overpredicted in some years, the maximum 1-hr and 8-hr O_3 mixing ratios are underpredicted in all winters during 2009-2015. The highest and the second highest NMBs for the maximum 1-hr and 8-hr O_3 mixing ratio occur in the winter seasons in 2014-2015 and 2010-2011, respectively, with NMBs of -18.1% and -17.7% in 2014-2015 and -11.9% and -13.5% in 2010-2011. The model's skill in terms of NMEs, RMSEs, and R values is overall similar among all six winter seasons at sites from AirNow, with lower NMEs and RMSEs for both maximum 1-hr and 8-hr O_3 but lower R values for maximum 8-hr O_3 than the O_3 seasons. At sites from other O_3 measurement networks such as CASTNET, AQS, and SEARCH, the performance statistics for O_3 in winter seasons during 2012-2015 in this work are also overall similar to those in 2009-2012 shown in Yahya et al. (2014). Since T2 is moderately overpredicted at most sites during 2009-2015, the O_3 underpredictions are caused in part by large NO_x underpredictions (e.g., an NMB of -67.2% for NO_2 in 2014-2015). Cai et al. (2008) evaluated the forecasting skills of an RT-AQF model that uses the CB4 gas-phase mechanism (which is an older version of CB05) and reported much significant underpredictions of OH and HO_2 radicals at two sites in New York city during January 2004 compared to July 2004. They attributed such underpredictions to greater

uncertainties associated with the CB4 mechanism under low light and low temperature conditions. Their analysis of the predicted and observed CO and NO_x regression slopes also showed a much larger discrepancies between the two slopes in winter than in summer, indicating significant uncertainties associated with the 1999 NEI mobile emission inventories during winter time. In this work, the average observed and forecasted ratios of CO/NO_x at the SEARCH sites for O₃ seasons during 2012-2014 are 28.1 and 29.3, respectively. Those for winters during 2012-2015 are 17.1 and 25.0, respectively. The larger differences in the observed and forecasted ratios of CO/NO_x indicate possibly larger uncertainties in mobile emissions in wintertime than warm seasons. As an example, Figure S3 shows the correlation plots for forecasted and observed CO and NO_x at the SEARCH sites in the 2012 O₃ season. The forecasted ratios of CO and NO_x are higher than their observed ratios in both the 2012 O₃ season and the 2012-2013 winter, with slightly larger differences between the two ratios in winter than in the O₃ season. Those uncertainties associated with winter gas-phase chemistry of HO_x radicals and emissions may also contribute to moderate underpredictions in O₃ at all sites, and large biases in CO, SO₂, and NO_x at the SEARCH sites during winters.

As shown in Table 2a, similar to the O₃ seasons, forecasted PM_{2.5} concentrations during winters agree very well with the observations from AIRNow with NMBs of 0.8 to 8.3% and from STN with NMBs of 4.9-8.3%, but moderately overpredicted at the IMPROVE and SEARCH sites, with NMBs of 57.4-59.3% and 59.7-68.4%, respectively. Unlike the O₃ seasons, the PM_{2.5} overpredictions are the results of overpredictions of OC with NMBs of 80.8-88.7% and EC with NMBs of 24.4-37.3% at the IMPROVE sites, and OC with NMBs of 24-33% and SO₄²⁻ with NMBs of 16.3-24.7% at the SEARCH sites. The concentrations of NO₃⁻ are also moderately overpredicted with an NMB of 16.4% in 2013 at the SEARCH sites, contributing to PM_{2.5} overpredictions. Overpredictions in both OC and EC lead to large overpredictions in TC

concentrations at the IMPROVE sites. Moderate overpredictions in OC dominate over moderate underpredictions in EC, leading to moderate overpredictions in TC at the SEARCH sites. Those results indicate possible overestimates of primary OC emissions at all types of sites and underestimates of EC emissions at urban/rural sites in southeastern U.S. in the SEARCH network during winter seasons.

Figure 1(b) compares several discrete statistics of $PM_{2.5}$ against data from AIRNow for the six winters during 2009-2015. Similar to the O_3 season, the model performs very well for $PM_{2.5}$ for all six winter seasons with the NMBs ranging from -10.2% during 2010-2011 winter to 8.3% during the 2012-2013 winter. As discussed in Yahya et al. (2014a), the underpredictions in 2010-2011 winter are the results of underpredictions in inorganic $PM_{2.5}$, due possibly to underestimates in the emissions of precursors such as SO_2 , NH_3 , and NO_x during winters. Other possible reasons for underpredictions of $PM_{2.5}$ during 2010-2011 include positive biases in both Precip and WS10. Different from underpredictions in $PM_{2.5}$ during 2009-2011 winter seasons at AirNow shown in Figure 1 (b) and STN shown in Yahya et al. (2014), the model overpredicts $PM_{2.5}$ during 2012-2015 winter seasons at all sites from AirNow, STN, IMPROVE, and SEARCH, with larger absolute biases at IMPROVE and SEARCH sites than those during 2009-2011 winter seasons. As discussed in Section 3.2.1, the inaccurate primary PM emissions and the emissions of $PM_{2.5}$ precursors, as well as uncertainties in the spatial allocations of those emissions used in the simulations contribute to the worse performance of $PM_{2.5}$ during winter seasons during 2012-2015 than during 2009-2011. Comparing to $PM_{2.5}$ forecasts during the O_3 seasons, the $PM_{2.5}$ forecasts during winters show slightly higher NMEs and R values and similar RMSEs. Comparing to O_3 forecasts during the winter seasons, $PM_{2.5}$ forecasts during winters show higher NMEs and R values but lower RMSEs. As shown in Figure 2, the model captures well the seasonal variations of $PM_{2.5}$, with higher $PM_{2.5}$ concentrations during O_3 seasons than during

winters. The model shows better spatial correlations with higher R values during winters than O₃ seasons. In particular, the model reproduces several observed hot spots in GA, FL, MO, IN, MD, and LA during the 2012-2013, 2013-2014, and 2014-2015 winters (see Figure 2). As shown in Figure 4, the model reproduces well the observed hourly concentrations of PM_{2.5} at all sites except for Birmingham and Atlanta where overpredictions occur during all three winters.

3.3 Categorical Evaluation of Surface Chemical Forecasts

Figure 5 shows categorical evaluation of O₃ and PM_{2.5} during all six O₃ and winter seasons. The accuracy is high for O₃ forecasts during all six O₃ and winter seasons, with A values of 94-97.7% during O₃ seasons and 98.7-100% during winters. High A values indicate higher percentage of forecasts that correctly predict an exceedance or a non-exceedance, with the number of non-exceedance dominating for both maximum 1-hr and 8-hr O₃ mixing ratios. Because the observed and forecasted maximum 1-hr and 8-hr O₃ mixing ratios during winters are below the threshold values of 80 ppb and 60 ppb, respectively, no values of CSI, POD, B, and FAR can be calculated. During O₃ seasons, the ranges of CSI values are 5.2-15.6 and 9.9-25.3 for maximum 1-hr and 8-hr O₃ mixing ratios, respectively. The relatively low CSI values are caused by relatively high false alarm forecasts. Higher CSI values for maximum 8-hr O₃ than maximum 1-hr O₃ indicate a better skill in forecasting medium range of O₃ mixing ratios during the daytime than the daily peak O₃ mixing ratios. For the same reason, the model gives higher POD values for maximum 8-hr O₃ than for maximum 1-hr O₃, with a range of 26.6-46.7 and 17-31.3, respectively. The model gives similarly low B values for both maximum 1-hr and 8-hr O₃ mixing ratios. The ranges of B values are 0.6-7.9 and 0.6-4.2 for maximum 1-hr and 8-hr O₃, respectively; they are greater than 1 in 2009, 2013, and 2014, indicating overpredictions in those years that are consistent with NMBs shown in Figure 1 (a). The FAR values are high, ranging

from 67-96.1% and 48.6-88.9% for maximum 1-hr and 8-hr O₃ mixing ratios, respectively. High FAR values indicate that a frequent occurrence of forecasted exceedance that did not occur.

Comparing to O₃ forecasts, the A values for PM_{2.5} forecasts are lower, ranging from 70.7-83.2% for O₃ seasons and 83.5-85.9% for winters, indicating that accurately forecasting PM_{2.5} is more challenging than forecasting O₃. The ranges of CSI values are 10.3-27.9% in O₃ seasons and 14.8-22.2% in winters, which are slightly higher than those for O₃ forecasts during most seasons. The POD values range from 15.3-40.1% in O₃ seasons and 28.5-38.3% in winters, which are similar to those for O₃ forecasts during O₃ seasons. B values for PM_{2.5} forecasts are smaller than those for O₃ forecasts, ranging from 0.6-1.3 in O₃ seasons and 0.7-1.2 in winters. FAR values for PM_{2.5} forecasts range from 44.6-75.9% in O₃ seasons and 61.3-76.6% in winters. They are lower than FAR values of O₃ forecasts during O₃ seasons.

3.4 Comparisons of Surface O₃ and PM_{2.5} Forecasting Skill with Other RT-AQF Models

Tables 3 and 4 compare the discrete and categorical performance evaluation for surface O₃ and PM_{2.5} forecasting in this work with those reported over U.S. or a region in the U.S. in the literature. Note that those evaluations did not use the same threshold values and observational data for evaluation nor that they were performed over the same domain and forecasting period. The statistics against AIRNow only and against all datasets are provided for Yahya et al. (2014) and this work because all other evaluations were based on AIRNow. The two sets of performance statistics of max 1-hr and 8-hr O₃ from WRF/Chem-MADRID in this work are within the range reported, with better performance based on AIRNow than most other models. For example, NMBs and NMEs of max 8-hr O₃ from WRF/Chem-MADRID are -17.7% to 17% and 17.8-33.8%, compared to -2.1% to 25.2% and 18.6-30.4%, respectively, reported in the literature. The performance against AIRNow is better than those against all datasets in this work because the model performs worse when the data from the SEARCH sites are included. For 24-h

PM_{2.5} evaluation using all datasets, while the MBs from this work fall into the reported range, the NMBs for the O₃ season and the NMEs for the winter seasons slightly exceed the upper range of report values because of inclusion of all datasets in this work rather than AIRNow only as did in most other work. Using AIRNow only for evaluation, the MBs, RMSEs, NMBs, NMEs for 24-h PM_{2.5} simulated by WRF/Chem-MADRID are -0.5 to 1.4, 5.1-5.7, -4 to 15%, and 36-40%, respectively, during the O₃ seasons, and 0.2 to 0.8, 5.5-6.1, 0.8 to 8.3%, and 42.6-47.4%, respectively, during the winter seasons, which are smaller than corresponding values from most other models, namely, -3.2 to 6.2, 5.5-15.9, -21 to 32%, and 41.2-80%, respectively. As shown in Table 4, the model's categorical performance for PM_{2.5} forecasts is comparable to or better than those reported in the literature. The FAR values for max 8-h O₃ during the O₃ season are slightly beyond the reported range, because of a moderate overprediction in the 2013 O₃ season.

3.5 Discrete and Spatial Evaluation of Column Chemical Forecasts

Table 2 also shows discrete statistics for column mass abundances of CO, NO₂, SO₂, and HCHO, TOR, and AOD during O₃ seasons and winters during 2012-2015. Column CO and SO₂ are moderately underpredicted with NMBs of -42.2% to -36.5% and -55.3% to -54.9%, respectively, in O₃ seasons during 2012-2014. The underpredictions are even larger in winter for both species, with NMBs of -50.7% to -48.2% and -77.2% to -73.2%, respectively. As shown in Table 2a, the surface CO and SO₂ mixing ratios are overpredicted at the SEARCH sites. The overpredictions at surface but underpredictions in their column masses indicate inaccurate vertical profiles used in their boundary conditions. For example, the BCONs of CO used in the forecasts vary from 72.5-96.4 ppb at the surface layer to 50-65 ppb in upper troposphere during July, and from 125-168 ppb at the surface layer to 50-65 ppb in upper troposphere during January. The vertical profiles of CO derived from MOPPIT over the continental U.S. show a value of 105 ppb at surface and 65 ppb at the tropopause during summer

and a value of 125 ppb at surface and 68 ppb at the tropopause during winter (Zhang et al., 2009). While the vertical profiles of CO used reflect the observed seasonal variations, the upper CO mixing ratios used are too low comparing to the MOPITT-derived CO levels in both seasons, and the surface CO mixing ratios are also low in O₃ seasons, leading to moderate to significant underpredictions in column CO in O₃ seasons and winters. The BCONs of SO₂ used in the forecasts vary from 0.04-1.35 ppb at the surface layer to 0.01-0.067 ppb in upper troposphere during July, and from 0.103-1.70 ppb at the surface layer to 0.01-0.067 ppb in upper troposphere during January. Those values are also too low to represent BCONs over southeastern U.S. While column NO₂ is moderately underpredicted with NMBs of -35.3% to -33.4% in the O₃ seasons, NMBs during winters are much smaller, ranging from -7.9% to 26.2%, indicating a more realistic vertical profile used in winters comparing that in O₃ seasons. The BCONs of NO₂ used in the forecasts vary from 0.082-0.181 ppb at the surface layer to 0 ppb in upper troposphere during July, and from 0.316-4.23 ppb at the surface layer to 0-0.0057 ppb in upper troposphere during January. Figure 6 (a) shows spatial distributions of column NO₂, with overall good spatial correlation and R values of 0.7 and 0.9, in the 2012 O₃ season and 2012-2013 winter, respectively. In addition to uncertainties in BCONs, inaccurate/missing emissions and inaccurate vertical allocations of emissions may contribute to the moderate to large underpredictions in column CO, SO₂, and NO₂. For example, while wildfire and lightening NO_x emissions are included, large uncertainties exist in their magnitudes and spatial distributions. Volcanic eruption and/or degassing may make important contribution to column SO₂.

Unlike column CO, SO₂, and NO₂, Column HCHO is moderately overpredicted with NMBs of 13.1-39.9% in O₃ seasons but largely underpredicted with NMBs of -59% to -51.5% in winters. The BCONs of HCHO used in the forecasts vary from 0.599-2.47 ppb at the surface layer to 0 ppb in upper troposphere during July, and from 0.292-0.404 ppb at the surface layer to

0 ppb in upper troposphere during January. The performance statistics show that the BCONs of HCHO are too high in O₃ seasons but too low in winters. Another possible source of errors in simulated column HCHO may come from inaccurate biogenic emissions of isoprene, which can produce secondary HCHO through its photochemical oxidation reactions.

TOR is slightly-to-moderately underpredicted with NMBs of -15.4 to -4.5% in O₃ seasons but moderately overpredicted with NMBs of 29.9-45.1%. The BCONs of O₃ used in the forecasts vary from 26.3-44 ppb in July and from 22.8-39.1 ppb in January at the surface layer to 100.5 ppb in upper troposphere during both months. Although O₃ can be formed through photochemical oxidations of precursor gases such as NO_x, HCHO, and CO above the surface layer, the mixing ratios of those gases are generally low, particularly in mid-to-upper troposphere. Therefore, the column concentrations of O₃ are regulated primarily by BCONs. The performance statistics show that the BCONs of O₃ are more realistic in O₃ seasons than in winters during which the BCON values are too high to represent O₃ vertical profile, leading to moderately overpredicted TOR. AOD is moderately overpredicted with NMBs of 14.4% to 47.6% in O₃ seasons, and significantly overpredicted with NMBs of 59.4% to 95.7% in winters. The overpredictions of AOD are the results of overpredictions of PM_{2.5} at surface and also possible overpredictions of PM_{2.5} in upper layers, indicating that the BCONs used for PM_{2.5} composition may be too high in both O₃ and winter seasons. Figures 6 (a) and (b) show spatial distributions of TOR and AOD. While forecasted TORs correlate well with OMI-derived TORs with an R value of 0.7 during the 2012 O₃ season, they do not correlate in the 2012-2013 winter, indicating a need to adjust the vertical profile of O₃ in winter. The forecasted and MODIS-derived AOD agree better spatially in the 2012-2013 winter than in the 2012 O₃ seasons.

Two sensitivity simulations are performed to further study the importance of BCONs on column forecasts including a sensitive simulation during August 2012 using satellite-constrained

BCONs for CO and a sensitive simulation during December 2012 using satellite-constrained BCONs for O₃. Those sensitivity simulations show large improvement in simulated column CO and TOR. Figures 7 and 8 compare the spatial distributions of satellite-derived CO and TOR and the two simulations in August 2012 and December 2012, respectively. The use of satellite-constrained BCONs for CO and TOR improves the simulated CO and TOR substantially. The MB, NMB, and NME of CO from the sensitivity simulation are -0.2, -10.6%, and 18.2%, respectively, comparing to -0.8, -40.6%, and 40.8% from the baseline simulation. The MB, NMB, and NME of TOR from the sensitivity simulation are -0.2, -0.01%, and 0.1%, respectively, comparing to 11.7, 44.8%, and 44.8% from the baseline simulation. Similar improvements are expected for other column variables including column NO₂ in O₃ season and column SO₂ and HCHO in both season.

3.6 Trend analysis for multiple years

Given interannual variability in climate and emissions, it is useful to assess the robustness of the model in forecasting the relative changes in terms of magnitudes and signs under different climate conditions, as well as the interannual variability from the year of reference.

3.6.1 Meteorological Variables

Figure 9 compares observed and simulated variation trends for T2, Precip, WS10, SWDOWN, LWDOWN, and CF. Note that the trends for SWDOWN, LWDOWN, and CF are only plotted for the 2011-2014 O₃ seasons and 2011-2015 winters because the upper layer model outputs during 2009-2010 were not available due to failures of backup drives containing such data. The model forecasts well the observed changes in terms of both magnitudes and signs, as well as the interannual variability of T2 and WS10 in both O₃ and winter seasons relative to their values in 2009. It simulates reasonably well for the observed interannual variability of Precip at the CASTNET sites, but not well for the observed changes in magnitudes of Precip. The changes

in terms of magnitudes and signs as well as interannual variability relative to their values in 2011-2012 for SWDOWN, LODOWN, and CF are well captured in winters, but in O₃ seasons, while the model reproduces well both the magnitudes of the changes and the interannual variability of LWDOWN, and the interannual variability of SWDOWN and CF, it overpredicts the increases in SWDOWN but underpredicts the increases in CF.

3.6.2 Chemical Variables

Figure 10 compares observed and simulated variation trends for surface O₃ mixing ratios, surface PM_{2.5} concentrations, column mass abundances of CO, SO₂, NO₂, and HCHO, TOR, and AOD. Note that the trends for column variables are only plotted for the O₃ seasons during 2011-2014 and winters during 2011-2015 for the aforementioned reason. Relative to the 2009 O₃ season, the observed O₃ mixing ratios from AIRNow are higher during O₃ seasons in 2010-2012 and 2014 but are lower in O₃ season in 2013, this trend is not well captured by the model, as it forecasts slightly lower O₃ in 2010 and 2014 O₃ seasons, and slightly higher O₃ in other O₃ seasons. While PM_{2.5} forecasts during O₃ seasons generally follow the observed trends, large differences occur in the magnitudes of the changes, with greater increases in 2010-2011 but greater decreases during 2012-2014. Although the differences in the magnitude of the changes for forecasted O₃ are smaller in winters than in O₃ seasons, the observed and forecasted O₃ mixing ratios change in different directions, i.e., the observed O₃ mixing ratios either increase or decrease slightly, the forecasted O₃ mixing ratios continue to decline during 2010-2014 winters. The large differences in magnitudes and signs remain in forecasted and observed trends of PM_{2.5} concentrations during winters comparing to O₃ seasons, however, the forecasted and observed changes of PM_{2.5} concentrations are within $\pm 10\%$. The forecasted and observed changes in column CO are small, within 3% in O₃ seasons and within 1% in winters but they have different signs. The forecasted column SO₂ captures well the observed trends in magnitudes and signs for

column SO_2 in the O_3 seasons, larger differences exist in both magnitude and sign in winters. Although the forecasted and observed changes for column NO_2 are generally within 10%, larger differences exist in magnitude and sign comparing to those for column CO and SO_2 . The OMI-derived TORs decrease in O_3 seasons during 2012-2014, and increase during winters of 2012-2013, 2013-2014, and 2014-2015. Among all column gases, the forecasted column HCHO shows the largest differences in the O_3 seasons in terms of both magnitude and sign. Uncertainties in satellite retrievals of column HCHO may contribute in part to such large discrepancies between forecasts and satellite-derived observations. For example, De Smedt et al. (2008) reported errors in HCHO retrievals of $(0.5\text{-}2.0) \times 10^{15} \text{ molecules cm}^{-2}$, which are on the same order of magnitudes or even larger than the MBs in the forecasted HCHO column for all seasons. The differences in magnitude of the changes in column HCHO are smaller in winters but the signs are opposite in 2012-2013. The forecasted TORs show a slight decrease in 2012 O_3 season and slight increases in 2013 and 2014 O_3 seasons, and slight decreases in winters of 2012-2013 and 2013-2014 as well as a slight increase in winter of 2014-2015.

The forecasted AOD captures the decreasing trend during O_3 seasons of 2012-2014, but with much smaller magnitudes of the changes (by up to 18% versus 40%, respectively). While MODIS-derived AOD shows a large decrease (by up to 22%) from 2011-2012 winter, the forecasted AOD shows a large increase (by up to 23%) in winter. The relatively large discrepancies between satellite-derived and forecasted column variables such as column NO_2 and HCHO, TOR, and AOD in both O_3 and winter seasons, and column SO_2 in winters indicate a need to adjust the vertical profiles of these gases and PM composition in the BCONs.

Summary and Conclusion

An online-coupled meteorology-chemistry model, WRF/Chem-MADRID, has been deployed for RT-AQF in southeastern U.S. since 2009 for six O₃ seasons and six winters. A comprehensive evaluation of meteorological and chemical forecasts is performed using surface and satellite-derived observations in terms of spatial distribution, temporal variation, and discrete and categorical performance statistics. The meteorological evaluation shows moderate to large biases for T2, RH2, WS10, WD10, Precip, SWDOWN, CF, and COT, indicating some limitations in the YSU PBL scheme, the Monin-Obukhov surface layer scheme, the Purdue Lin cloud microphysics module, and the Grell-Devenyi ensemble scheme. In particular, uncertainties exist in the model treatments of PBL processes (e.g., inaccurate representations of surface drag), the dynamics, thermodynamics, and microphysics of clouds, as well as aerosol-radiation-cloud-precipitation interactions (e.g., the missing treatments of sub-grid cloud feedbacks to radiation). Since the forecasts do not use data assimilation, the agreement between meteorological forecasts and observations is not expected to be comparable with the simulations that use data assimilation. The meteorological forecasts for most variables except for WS10, Precip, and COT in this work are therefore deemed to be acceptable. While updating WRF/Chem-MADRID based on the latest WRF/Chem version should help reduce some of those uncertainties with updated schemes and treatments, continuous development and improvement of PBL schemes and cloud parameterizations are important future work to improve meteorological forecasts, which will in turn improve chemical forecasts.

WRF/Chem-MADRID shows consistently good skills for O₃ and PM_{2.5} forecast in terms of both categorical and discrete statistics during 2009-2015. It performs well in both O₃ and winter seasons with most NMBs within $\pm 15\%$ for O₃ forecasts against observations from AIRNow, AQS, CASTNET, and SEARCH. The NMBs for PM_{2.5} forecasts are within $\pm 15\%$ against observations from AIRNow and STN, but larger (up to $\pm 68\%$) against observations from

IMPROVE and SEARCH. Larger biases are also found for secondary PM_{2.5} against surface observations at IMPROVE and SEARCH, and also for some column variables (e.g., column NO₂ in O₃ seasons, TOR, column HCHO, and AOD in winters, and COT and column CO and SO₂ in both O₃ and winter seasons) against satellite data. These biases are due possibly to uncertainties in simulated meteorology (e.g., T2, Precip, and WS10), emissions (e.g., biogenic/wildfire emissions and winter mobile emissions), and BCONs (e.g., inaccurate BCONs for seasonal and inter-annual variations for CO, NO₂, SO₂, O₃, HCHO, and PM_{2.5} composition), as well as limitations in chemical and aerosol treatments (e.g., the production of OH radicals from CB05 in winter, aerosol thermodynamic partitioning, and SOA formation). Comparison of model performance during 2012-2015 with that during 2009-2012 shows that the inaccurate primary PM emissions and the emissions of PM_{2.5} precursors, as well as uncertainties in the spatial allocations of those emissions used in the simulations contribute to the worse performance of PM_{2.5} during both O₃ and winter seasons during 2012-2015 than during 2009-2012.

Despite those biases, the model's performance in terms of surface O₃ and PM_{2.5} forecasts is overall consistent with or better than the performance of other RT-AQF models reported in the literature for different periods over different domains. Although the model shows overall good skills for meteorological and chemical forecasts at the surface, inaccurate representations of species vertical profiles can potentially affect both meteorological and chemical forecasts at the surface because of turbulent mixing and convective cloud updraft and downdraft movements and because of the feedbacks of radiative species (e.g., O₃, NO₂, HCHO, and PM composition) to the radiation calculation in the model. The impacts of chemical BCONs on air quality simulations have been shown in several studies (e.g., Giordano et al., 2015; Yahya et al., 2015b) and in this work. Therefore, the vertical profiles of BCONs of those species should be constrained with satellite-derived observations to more realistically represent vertical and seasonal variations.

Forecasted changes in most meteorological variables except for CF generally reproduce well the observed trends in terms of magnitude and sign and interannual variability. While small changes occur in observed seasonal-mean maximum 1-hr and 8-hr O₃ concentrations from AIRNow since 2009, those for PM_{2.5} show greater decreases and stronger inter-annual variabilities than O₃, reflecting the effects of emission reductions since 2009. Forecasted O₃ levels show weaker inter-annual variabilities than observed O₃ levels during all O₃ and winter seasons. Forecasted PM_{2.5} levels resemble their observed increasing trends from 2009 to 2011 and declining trend from 2011 to 2014 during O₃ seasons and remain nearly constant during winter. Such variabilities are mainly attributed to changes in meteorology and meteorology-dependent biogenic and wildfire emissions. Largest discrepancies are found in the forecasted and observed changes in AOD and column gases including CO, NO₂, SO₂, HCHO, and O₃, due mainly to inaccurate representations of the vertical profiles of the BCONs of those gases and PM composition. More accurate meteorological forecasts, anthropogenic emissions, and meteorology-dependent emissions (e.g., biogenic, wildfire, and volcanic), upper BCONs for chemical species, and model treatments of chemical and aerosol processes should improve the model's ability in reproducing not only the observations but also the interannual and inter-seasonal variation trends in terms of magnitude and sign for major chemical species of concerns.

When resources become available, several limitations in this work should be addressed. These may include the code migration of WRF/Chem-MADRID into the latest version of WRF/Chem, the refinement of configurations using available latest physics and chemistry options (e.g., the use of urban canopy model, updated surface roughness treatments, and the multi-scale cumulus parameterization), and updates in emissions and lateral BCONs including using real-time forecasted emissions, and more realistic BCONs derived from satellite retrievals or dynamic BCONs from a validated global RT-AQF model.

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Table 1a. Discrete statistics of meteorological variables for O₃ seasons.

Variable	Network	O ₃ Season (May – September)														
		2012					2013					2014 ¹				
		Mean Obs ²	Mean Sim	Corr	MB	MAGE	Mean Obs	Mean Sim	Corr	MB	MAGE	Mean Obs	Mean Sim	Corr	MB	MAGE
T2 (°C)	CASTNET	22.2	22.8	0.6	0.5	4.2	21.2	23.3	0.7	2.1	4.0	21.2	23.3	0.7	2.1	4.0
	NCDC	23.7	24.3	0.6	0.6	4.0	23.0	24.8	0.7	1.8	3.7	22.9	24.7	0.7	1.7	3.9
	SEARCH	24.9	25.8	0.4	0.9	4.0	24.1	26.7	0.5	2.6	3.9	23.5	25.6	0.3	2.1	4.7
RH2 (%)	CASTNET	76.3	66.5	0.3	-9.8	17.5	78.2	61.8	0.3	-16.4	20.3	74.3	60.5	0.4	-13.8	18.6
	NCDC	74.4	67.5	0.3	-6.8	17.8	78.5	64.0	0.3	-14.4	19.6	74.1	60.6	0.4	-13.5	20.0
	SEARCH	74.0	63.5	0.3	-10.4	18.5	80.1	59.6	0.2	-20.5	23.2	74.5	54.7	0.3	-19.7	23.1
WS10 (m s ⁻¹)	CASTNET	2.0	3.7	0.4	1.7	1.9	1.8	3.7	0.2	1.8	2.1	2.5	4.1	0.4	1.6	2.1
	NCDC	3.6	3.8	0.2	0.2	1.8	3.6	3.9	0.2	0.4	1.8	3.7	3.9	0.2	0.2	1.8
	SEARCH	2.2	3.1	0.3	0.9	1.3	2.1	2.7	0.3	0.6	1.2	2.2	2.5	0.2	0.3	1.2
WDR10 (°)	CASTNET	201.8	224.8	0.7	23.0	79.1	207.2	223.5	0.7	16.3	86.2	195.6	224.7	0.6	29.0	85.7
	NCDC	186.5	232.1	0.7	45.6	85.2	187.3	235.2	0.7	47.9	85.1	199.4	241.9	0.6	42.4	86.1
	SEARCH	200.7	219.5	0.6	18.9	83.0	206.0	230.5	0.4	24.4	94.3	224.4	226.1	0.3	1.7	76.2
Precip (mm hr ⁻¹)	CASTNET	0.2	0.3	0.0	0.1	0.4	0.2	0.3	0.0	0.1	0.5	0.2	0.3	0.0	0.1	0.4
	NCDC	3.2	4.7	0.0	1.5	5.3	2.8	4.1	0.0	1.4	4.6	3.2	4.3	0.1	1.1	4.9
	GPCP	0.2	0.3	0.4	0.1	0.1	0.2	0.3	0.4	0.1	0.1	0.2	0.2	0.1	0.0	0.1
SWDOWN (W m ⁻²)	CERES	245.2	279.7	0.6	34.4	34.5	239.7	281.9	0.1	42.2	42.2	244.2	311.5	0.4	67.3	67.3
LWDOWN (W m ⁻²)	CERES	399.9	391.0	1.0	-8.9	11.9	397.8	398.5	1.0	0.7	4.4	398.6	390.5	1.0	-8.1	8.1
CF (W m ⁻²)	MODIS	57.8	53.0	0.9	-4.8	6.4	64.4	56.2	0.8	-8.2	9.1	66.6	57.0	0.6	-9.6	10.8
COT	MODIS	14.2	5.4	0.3	-8.8	8.8	14.9	5.9	-0.1	-9.0	9.0	13.5	4.6	0.1	-8.9	8.9

Table 1b. Discrete statistics of meteorological variables for winter seasons.

Variable	Network	Winter Season (December-February)														
		2012-2013					2013-2014					2014-2015				
		Mean Obs ²	Mean Sim	Corr	MB	MAGE	Mean Obs	Mean Sim	Corr	MB	MAGE	Mean Obs	Mean Sim	Corr	MB	MAGE
T2 (°C)	CASTNET	5.7	6.7	0.9	1.0	3.5	3.3	4.3	0.9	0.9	3.6	2.8	3.5	0.9	0.7	3.7
	NCDC	8.1	9.3	0.8	1.2	3.7	6.2	7.4	0.9	1.2	3.7	5.6	6.4	0.8	0.8	3.7
	SEARCH	10.1	5.2	0.4	-5.0	8.7	7.9	5.0	0.1	-3.0	7.6	10.0	11.1	0.6	1.1	4.0
RH2 (%)	CASTNET	75.0	69.0	0.4	-6.0	15.9	72.0	70.1	0.4	-1.9	15.9	74.2	70.2	0.4	-4.1	15.1
	NCDC	76.3	70.4	0.4	-6.0	16.0	74.1	69.9	0.4	-4.2	16.0	74.0	69.7	0.5	-4.3	15.1
	SEARCH	73.3	59.5	0.4	-13.9	21.5	73.7	61.6	0.4	-12.1	21.4	79.2	72.9	0.2	-6.3	16.2
WS10 (m s ⁻¹)	CASTNET	2.4	5.0	0.3	2.5	2.8	2.9	4.9	0.2	1.9	2.5	2.9	4.5	0.2	1.6	2.4
	NCDC	4.3	5.1	0.3	0.8	2.4	4.2	4.8	0.2	0.6	2.3	4.2	4.4	0.2	0.2	2.2
	SEARCH	2.5	4.1	0.6	1.5	1.7	2.4	3.9	0.5	1.4	1.7	2.3	2.5	0.3	0.2	1.1
WDR10 (°)	CASTNET	201.2	217.1	0.5	15.9	91.5	208.4	237.2	0.7	28.8	89.3	212.3	243.1	0.6	30.8	99.8
	NCDC	206.8	245.6	0.6	38.7	96.9	207.8	253.3	0.7	45.5	92.8	209.7	256.4	0.7	46.7	97.5
	SEARCH	212.7	247.9	0.8	35.2	94.1	206.7	228.9	0.8	22.2	97.6	212.2	258.8	0.9	46.6	86.7
Precip (mm hr ⁻¹)	CASTNET	0.1	0.2	0.1	0.1	0.3	0.1	0.2	0.2	0.1	0.2	0.1	0.1	0.3	0.1	0.2
	NCDC	1.9	2.7	0.1	0.8	2.7	1.9	2.5	0.1	0.6	2.4	1.8	2.2	0.1	0.4	2.2
	GPCP	0.1	0.2	0.7	0.1	0.1	0.1	0.2	0.4	0.1	0.1	0.1	0.2	0.5	0.0	0.1
SWDOWN (W m ⁻²)	CERES	118.8	142.4	0.9	23.5	23.7	119.5	141.7	0.9	22.2	22.2	118.4	146.7	0.9	28.4	28.4
LWDOWN (W m ⁻²)	CERES	323.4	320.3	1.0	-3.2	4.1	314.2	318.3	1.0	4.1	5.5	314.3	312.1	1.0	-2.1	3.8
CF (W m ⁻²)	MODIS	64.4	50.0	0.7	-14.4	14.4	64.4	52.8	0.5	-11.6	11.7	65.8	50.0	0.8	-15.7	15.7
COT	MODIS	17.8	5.4	0.2	-12.3	12.3	18.8	5.6	0.2	-13.2	13.2	18.2	4.9	0.1	-13.3	13.3

¹ Data pairs only include simulated and observed data during May, June, and July in 2014 because of loss of simulated data in August and September, 2014 due to failure of backup external hard drives containing such data.

² Mean Obs: Mean observed data; Mean Sim: Mean simulated data; Corr: Correlation coefficient; MB: Mean bias; MAGE: Mean Absolute Gross Error; N/A: Data not available.

Table 2a. Discrete statistics of chemical variables for O₃ seasons.

Variable	Network	O ₃ Season (May – September)														
		2012					2013					2014 ¹				
		Mean Obs ²	Mean Sim	Corr	NMB (%)	NME (%)	Mean Obs	Mean Sim	Corr	NMB (%)	NME (%)	Mean Obs	Mean Sim	Corr	NMB (%)	NME (%)
CO (ppb)	SEARCH	161.6	267.1	0.4	65.3	97.1	177.9	264.2	0.3	48.5	75.5	178.5	322.7	0.4	80.8	99.8
SO ₂ (ppb)	SEARCH	1.0	1.9	0.0	99.0	195.0	0.5	1.9	0.1	308.0	362.0	0.3	2.8	0.3	725.0	736.0
NO (ppb)	SEARCH	1.9	1.4	0.1	-26.0	135.0	1.2	1.6	0.2	36.0	176.0	1.5	4.9	0.1	222.0	339.0
NO ₂ (ppb)	SEARCH	4.6	6.8	0.4	49.0	119.0	4.1	6.4	0.4	56.0	118.0	5.1	8.0	0.5	56.0	120.0
HNO ₃ (ppb)	SEARCH	0.3	0.5	0.1	50.5	124.3	0.3	0.4	0.1	57.0	119.0	0.3	0.4	0.1	23.0	95.0
Max 1-hr O ₃ (ppb)	AIRNow	52.5	52.4	0.5	0.0	22.0	45.6	52.4	0.5	15.0	26.0	49.1	49.1	0.4	0.0	23.0
	AQS	52.4	52.1	0.6	-1.0	22.0	45.6	51.1	0.5	12.0	26.0	N/A	N/A	N/A	N/A	N/A
	CASTNET	51.3	49.8	0.5	-3.0	22.0	46.3	52.0	0.4	12.0	25.0	50.5	51.0	0.4	1.0	19.0
	SEARCH	53.3	55.8	0.6	5.0	21.0	46.3	54.4	0.5	17.0	27.0	48.3	50.2	0.5	4.0	24.0
Max 8-hr O ₃ (ppb)	AIRNow	47.4	47.5	0.5	0.0	22.0	41.0	47.9	0.4	17.0	27.0	44.5	45.0	0.4	1.0	23.0
	AQS	46.8	46.5	0.5	-1.0	22.0	40.3	46.4	0.5	15.0	27.0	N/A	N/A	N/A	N/A	N/A
	CASTNET	45.8	45.1	0.5	-2.0	23.0	37.1	43.6	0.3	18.0	28.0	40.3	41.7	0.3	4.0	20.0
	SEARCH	47.3	49.7	0.6	5.0	21.0	40.7	49.5	0.6	22.0	29.0	43.0	45.7	0.5	6.0	26.0
24-hr Avg PM _{2.5} (µg m ⁻³)	AIRNow	10.8	11.0	0.3	2.0	38.0	9.8	11.2	0.4	15.0	40.0	10.2	9.7	0.3	-4.0	36.0
	IMPROVE	7.7	9.6	0.3	25.0	44.0	7.6	9.5	0.4	25.0	44.0	8.0	8.6	0.4	8.0	33.0
	STN	11.1	12.0	0.3	9.0	38.0	10.4	11.7	0.4	12.0	39.0	N/A	N/A	N/A	N/A	N/A
	SEARCH	9.8	13.6	0.2	39.0	66.0	9.1	13.9	0.3	53.0	74.0	N/A	N/A	N/A	N/A	N/A
NH ₄ ⁺ (µg m ⁻³)	CASTNET	0.8	0.8	0.6	-7.0	29.0	0.8	0.8	0.8	-4.0	25.0	0.7	0.6	0.5	-12.0	29.0
	IMPROVE	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
	STN	0.5	0.9	0.4	64.0	88.0	0.5	0.8	0.4	58.0	87.0	N/A	N/A	N/A	N/A	N/A
	SEARCH	0.8	0.9	0.2	6.0	62.0	0.7	0.8	0.3	14.0	65.0	N/A	N/A	N/A	N/A	N/A
SO ₄ ²⁻ (µg m ⁻³)	CASTNET	2.6	2.5	0.4	-3.0	29.0	2.4	2.6	0.5	6.0	27.0	2.3	2.1	0.2	-9.0	25.0
	IMPROVE	2.1	2.4	0.3	18.0	47.0	2.0	2.4	0.4	16.0	49.0	1.9	2.1	0.4	10.0	40.0
	STN	2.2	2.6	0.3	16.0	46.0	2.1	2.6	0.4	20.0	51.0	N/A	N/A	N/A	N/A	N/A
	SEARCH	2.3	2.9	0.2	29.0	66.0	2.1	3.0	0.3	41.0	72.0	N/A	N/A	N/A	N/A	N/A
NO ₃ ⁻ (µg m ⁻³)	CASTNET	0.4	0.3	0.5	-18.0	62.0	0.3	0.2	0.6	-32.0	60.0	0.4	0.2	0.6	-48.0	67.0
	IMPROVE	0.2	0.3	0.3	54.0	130.0	0.2	0.3	0.3	47.0	136.0	0.2	0.3	0.3	25.0	135.0
	STN	0.4	0.4	0.3	-8.0	89.0	0.4	0.3	0.3	-24.0	83.0	N/A	N/A	N/A	N/A	N/A
	SEARCH	0.2	0.3	0.0	48.0	207.0	0.2	0.3	0.0	83.0	233.0	N/A	N/A	N/A	N/A	N/A
EC (µg m ⁻³)	IMPROVE	0.2	0.2	0.3	0.0	54.0	0.2	0.2	0.5	5.0	54.0	0.2	0.2	0.2	1.0	54.0
	SEARCH	1.5	0.5	-0.1	-67.0	88.0	0.9	0.5	0.0	-40.0	82.0	N/A	N/A	N/A	N/A	N/A
OC (µg m ⁻³)	IMPROVE	1.3	1.9	0.3	50.0	73.0	1.2	1.3	0.1	15.0	60.0	1.2	1.2	0.1	1.0	56.0
	SEARCH	3.0	2.9	0.2	-6.0	57.0	2.2	1.6	0.2	-28.0	61.0	N/A	N/A	N/A	N/A	N/A
TC (µg m ⁻³)	IMPROVE	1.5	2.2	0.3	42.0	66.0	1.4	1.6	0.2	14.0	57.0	1.4	1.5	0.1	1.0	54.0
	STN	2.8	2.7	0.5	-3.0	37.0	2.7	2.0	0.1	-27.0	46.0	N/A	N/A	N/A	N/A	N/A
	SEARCH	2.3	3.0	0.3	30.0	89.0	3.0	2.1	0.3	-29.0	53.0	N/A	N/A	N/A	N/A	N/A
Column CO (10 ¹⁸ molec. cm ⁻²)	MOPITT	2.1	1.2	0.5	-42.2	42.2	2.0	1.3	0.5	-36.5	36.5	2.0	1.3	0.3	-37.2	37.2
Column NO ₂ (10 ¹⁵ molec. cm ⁻²)	OMI	1.7	1.1	0.7	-35.3	45.9	1.6	1.1	0.7	-33.4	42.9	1.7	1.1	0.7	-33.5	40.8
Column SO ₂ (DU)	OMI	0.25	0.11	0.5	-54.9	59.2	0.25	0.11	0.5	-55.1	58.8	0.25	0.11	0.4	-55.3	58.7
Column HCHO (10 ¹⁵ molec. cm ⁻²)	OMI	8.6	9.8	0.8	13.1	31.8	7.6	10.7	0.8	39.9	52.4	8.2	9.2	0.7	13.2	35.3
Column O ₃ (DU)	OMI	39.4	33.3	0.7	-15.4	16.8	38.0	36.0	0.6	-5.3	9.0	37.6	35.9	0.7	-4.5	8.1
AOD	MODIS	0.2	0.2	0.0	14.4	23.6	0.1	0.2	-0.4	47.6	48.4	0.1	0.2	-0.2	37.2	43.8

Table 2b. Discrete statistics of chemical variables for winter seasons

Variable	Network	Winter Season (December-February)														
		2012-2013					2013-2014					2014-2015				
		Mean Obs ²	Mean Sim	Corr	NMB (%)	NME (%)	Mean Obs	Mean Sim	Corr	NMB (%)	NME (%)	Mean Obs	Mean Sim	Corr	NMB (%)	NME (%)
CO (ppb)	SEARCH	200.9	279.1	0.3	38.9	72.4	203.7	345.9	0.3	69.8	89.3	244.6	514.1	0.5	110.2	122.5
SO ₂ (ppb)	SEARCH	0.8	1.9	0.1	123.3	203.1	0.8	2.6	0.1	242.2	298.3	0.5	3.6	0.3	642.6	657.9
NO (ppb)	SEARCH	4.5	2.8	0.3	-37.0	112.8	5.4	4.9	0.3	-8.6	129.0	8.6	25.9	0.4	199.8	263.8
NO ₂ (ppb)	SEARCH	6.4	7.9	0.5	24.3	81.8	7.5	9.4	0.5	24.9	79.0	9.0	3.0	0.5	-67.2	71.0
HNO ₃ (ppb)	SEARCH	0.2	0.4	0.2	111.3	149.1	0.2	0.4	0.2	114.7	155.9	0.2	0.0	0.0	-90.7	91.4
Max 1-hr O ₃ (ppb)	AIRNow	38.3	33.8	0.5	-11.6	19.5	36.8	33.8	0.4	-8.1	17.5	36.7	30.1	0.2	-18.1	23.4
	AQS	38.0	33.1	0.5	-12.8	21.7	33.9	31.7	0.5	-6.5	20.5	N/A	N/A	N/A	N/A	N/A
	CASTNET	38.4	33.9	0.6	-11.7	17.8	38.6	33.7	0.5	-12.8	16.8	38.0	32.5	0.5	-14.3	19.2
	SEARCH	37.6	32.5	0.6	-13.3	20.1	36.0	31.2	0.6	-13.2	20.3	30.5	27.8	0.3	-9.0	22.8
Max 8-hr O ₃ (ppb)	AIRNow	35.6	30.8	0.2	-13.5	22.6	33.8	30.8	0.2	-9.1	19.6	35.0	28.8	0.0	-17.7	24.6
	AQS	33.8	29.4	0.5	-13.0	24.3	29.8	27.8	0.4	-6.5	23.9	N/A	N/A	N/A	N/A	N/A
	CASTNET	32.5	29.7	0.5	-8.7	18.2	32.6	29.2	0.6	-10.2	16.7	29.6	28.6	0.6	-3.2	16.5
	SEARCH	33.6	29.6	0.6	-12.0	21.3	32.2	28.1	0.6	-13.0	21.6	26.4	25.9	0.2	-1.7	25.4
24-hr Avg PM _{2.5} (µg m ⁻³)	AIRNow	9.3	10.0	0.4	8.3	42.6	8.9	9.5	0.3	6.7	47.2	9.2	9.2	0.3	0.8	44.0
	IMPROVE	5.6	8.9	0.4	57.4	72.2	5.2	8.4	0.4	59.3	83.2	N/A	N/A	N/A	N/A	N/A
	STN	9.7	10.2	0.5	4.9	37.7	10.2	11.1	0.7	8.3	45.9	N/A	N/A	N/A	N/A	N/A
	SEARCH	7.8	12.4	0.2	59.7	85.3	7.6	12.9	0.3	68.4	89.0	N/A	N/A	N/A	N/A	N/A
NH ₄ ⁺ (µg m ⁻³)	CASTNET	0.9	0.5	0.6	-42.7	48.2	0.8	0.5	0.6	-32.0	45.2	1.0	0.4	0.6	-55.7	58.8
	IMPROVE	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
	STN	0.8	0.6	0.4	-17.9	63.5	0.7	0.7	0.7	-9.6	63.6	N/A	N/A	N/A	N/A	N/A
	SEARCH	0.8	0.6	0.2	-24.9	62.8	0.7	0.6	0.2	-19.1	61.4	N/A	N/A	N/A	N/A	N/A
SO ₄ ²⁻ (µg m ⁻³)	CASTNET	1.8	1.5	0.2	-21.1	34.1	1.7	1.5	-0.2	-14.6	39.2	1.9	1.3	0.3	-31.0	40.9
	IMPROVE	1.5	1.5	0.2	-3.3	51.3	1.5	1.4	0.1	-7.5	55.6	N/A	N/A	N/A	N/A	N/A
	STN	1.7	1.6	0.1	-5.8	53.7	1.7	1.6	0.5	-4.0	47.9	N/A	N/A	N/A	N/A	N/A
	SEARCH	1.5	1.8	0.1	24.7	68.8	1.5	1.7	0.1	16.3	62.4	N/A	N/A	N/A	N/A	N/A
NO ₃ ⁻ (µg m ⁻³)	CASTNET	1.5	0.6	0.5	-59.8	66.8	1.1	0.6	0.6	-40.8	58.9	1.5	0.2	0.6	-90.2	90.2
	IMPROVE	0.9	0.6	0.2	-33.4	86.4	0.8	0.6	0.3	-23.7	91.9	N/A	N/A	N/A	N/A	N/A
	STN	1.5	0.7	0.4	-54.3	73.9	1.6	1.1	0.5	-32.4	71.8	N/A	N/A	N/A	N/A	N/A
	SEARCH	0.6	0.5	0.0	-12.8	119.2	0.6	0.7	0.3	16.4	121.1	N/A	N/A	N/A	N/A	N/A
EC (µg m ⁻³)	IMPROVE	0.3	0.4	0.5	37.3	74.9	0.3	0.3	0.4	24.4	74.0	N/A	N/A	N/A	N/A	N/A
	SEARCH	1.0	0.6	0.1	-37.5	78.1	1.0	0.7	0.1	-29.5	87.3	N/A	N/A	N/A	N/A	N/A
OC (µg m ⁻³)	IMPROVE	1.1	2.0	0.4	80.8	102.6	1.0	1.9	0.3	88.7	118.3	N/A	N/A	N/A	N/A	N/A
	SEARCH	2.1	2.8	0.2	33.0	85.7	2.2	2.8	0.2	24.0	79.5	N/A	N/A	N/A	N/A	N/A
TC (µg m ⁻³)	IMPROVE	1.3	2.3	0.4	72.5	95.0	1.3	2.3	0.3	75.0	106.4	N/A	N/A	N/A	N/A	N/A
	STN	2.6	2.6	0.4	1.5	44.5	2.5	2.6	0.6	5.4	55.9	N/A	N/A	N/A	N/A	N/A
	SEARCH	2.2	3.2	0.3	41.2	87.2	2.8	3.4	0.2	20.5	73.6	N/A	N/A	N/A	N/A	N/A
Column CO (10 ¹⁸ molec. cm ⁻²)	MOPITT	2.3	1.2	0.3	-50.7	50.7	2.3	1.2	0.1	-48.4	48.4	2.3	1.2	0.1	-48.2	48.2
Column NO ₂ (10 ¹⁵ molec. cm ⁻²)	OMI	2.7	2.7	0.9	1.0	20.1	2.3	2.9	0.9	26.2	32.8	2.5	2.3	0.7	-7.9	32.7
Column SO ₂ (DU)	OMI	0.39	0.09	0.5	-77.2	77.5	0.41	0.09	0.5	-77.1	77.3	0.40	0.11	0.5	-73.2	73.7
Column HCHO (10 ¹⁵ molec. cm ⁻²)	OMI	5.4	2.6	0.0	-51.5	51.6	6.3	2.6	0.1	-59.0	59.0	N/A	N/A	N/A	N/A	N/A
Column O ₃ (DU)	OMI	25.7	35.8	-0.2	39.3	42.4	27.1	35.2	-0.2	29.9	36.6	25.1	36.4	-0.4	45.1	47.9
AOD	MODIS	0.1	0.1	0.8	59.4	62.2	0.1	0.1	0.7	95.7	95.8	0.1	0.1	0.7	75.0	76.5

¹ Data pairs only include simulated and observed data during May, June, and July in 2014 because of loss of simulated data in August and September, 2014.

² Mean Obs: Mean observed data; Mean Sim: Mean simulated data; Corr: Correlation coefficient; NMB: Normalized mean bias; NME: Normalized mean error; N/A: Data not available.

Table 3. Discrete evaluation of RT-AQF results for O₃ and PM_{2.5} predictions

Area	Period	MB	RMSE	NMB (%)	NME (%)	Model	Reference
Maximum 1-hr average O₃							
NE US	8/5-29,2002	1.4	14.6	2.2	18.0	MAQSIP-RT	KA05
NE US	8/5-29,2002	9.5	21.3	15.0	25.8	MM5/Chem	KA05
NE US	8/5-29,2002	3.2	19.1	5.1	23.4	Hysplit/CheM	KA05
E US	7/1-8/15,2004	4.3–8.5	14.8–16.9	7.0–16.4	25.3	Eta/CMAQ	YU07
SE US	5/1-9/30,2009	4.5	16.8	9.5	26.7	WRF/Chem-MADRID	MT11
SE US	5/1-9/30,2009-2011	-3.0 – 4.6	13.4 – 17.0	-5.5 – 9.6	19.9–26.7	WRF/Chem-MADRID	YA14 ^b
SE US	5/1-9/30,2009-2011	-3.0 – 7.3	11.6–17.0	-5.5 – 15.5	17.6–27.4	WRF/Chem-MADRID	YA14 ^c
SE US	12/1-02/28,2009-2012	-4.6 – -2.2	8.0 – 9.7	-11.9 – -6.0	16.1–19.0	WRF/Chem-MADRID	YA14 ^b
SE US	12/1-02/28,2009-2012	-5.6 – 3.7	7.6–10.6	-13.8 – 10.8	15.0–20.3	WRF/Chem-MADRID	YA14 ^c
SE US	5/1-9/30,2012-2014	-0.1 – 6.9	14.0 – 15.1	0 – 15.0	22.0–26.0	WRF/Chem-MADRID	This work ^b
SE US	5/1-9/30,2012-2014	-1.5 – 8.0	12.1–15.9	-3.0 – 17.0	19.0–27.0	WRF/Chem-MADRID	This work ^c
SE US	12/1-02/28,2012-2015	-6.6 – -3.0	9.7–11.1	-18.1 – -8.1	17.5–23.4	WRF/Chem-MADRID	This work ^b
SE US	12/1-02/28,2012-2015	-6.2 – -4.2	8.0–11.1	-18.1 – -6.5	16.8–23.4	WRF/Chem-MADRID	This work ^c
Maximum 8-hr average O₃							
NE US	8/5-29,2002	8.3	18.2	15.1	25.4	MAQSIP-RT	KA05
NE US	8/5-29,2002	2.8	13.0	5.0	18.6	MM5/Chem	KA05
NE US	8/5-29,2002	-1.2	15.8	-2.1	22.5	Hysplit/Chem	KA05
NE US	6/1-9/30,2004	10.2	15.7	22.8	28.1	Eta/CMAQ	ED06
E US	7/1-8/15,2004	6.5–10.4	13.9–16.6	11.9–22.6	19.7–28.8	Eta/CMAQ	YU07
NY	7/1-9/30,2004	6.5	12.8	—	—	Eta/CMAQ	HO07
NY	1/1-3/31,2005	1.4	8.7	—	—	Eta/CMAQ	HO07
NY	6/1-9/30,2005	4.7	13.0	—	—	Eta/CMAQ	HO07
NE US	7/14-8/17,2004	3.4–14.3	11.6–20.9	—	—	WRF/chem	MK07
and		17.0	23.2	—	—	CHRONOS	MK07
SE CA		5.9	16.2	—	—	AURAMS	MK07
		26.4	31.0	—	—	STEM-2K3	MK07
		13.4	17.9	—	—	ET/CMAQ	MK07
E US	6/1-9/30,2005	10.9	16.3	22.4	27.1	WRF-NMM/CMAQ	ED09
E US	6/1-9/30,2006	10.5	15.6	25.2	30.4	WRF-NMM/CMAQ	ED09
E US	6/1-9/30,2007	7.9	14.5	16.5	24.1	WRF-NMM/CMAQ	ED09
SE US	5/1-9/30, 2009	3.5	13.6	8.3	25.0	WRF/Chem-MADRID	MT11
SE US	5/1-9/30,2009-2011	-1.8 – 3.6	11.7 – 13.7	-3.7 – 8.5	19.6–25.0	WRF/Chem-MADRID	YA14 ^b
SE US	5/1-9/30,2009-2011	-2.2 – 6.1	10.5–13.9	-4.5 – 14.6	17.8–26.1	WRF/Chem-MADRID	YA14 ^c
SE US	12/1-02/28,2009-2012	-4.9 – 2.0	8.1 – 12.2	-13.5 – -5.8	16.9–33.8	WRF/Chem-MADRID	YA14 ^b
SE US	12/1-02/28,2009-2012	-4.9 – 2.0	6.7–12.2	-13.5 – -0.3	16.9–21.5	WRF/Chem-MADRID	YA14 ^c
SE US	5/1-9/30,2012-2014	-0.2 – 6.9	13.2–14.2	0.0 – 17.0	22.0–27.0	WRF/Chem-MADRID	This work ^b
SE US	5/1-9/30,2012-2014	-0.8 – 8.8	10.3–15.1	-2.0 – 22.0	21.0–29.0	WRF/Chem-MADRID	This work ^c
SE US	12/1-02/28,2012-2015	-6.2 – -3.1	11.0–17.3	-17.7 – -9.1	19.6–24.6	WRF/Chem-MADRID	This work ^b
SE US	12/1-02/28,2012-2015	-6.2 – -0.5	6.1–17.3	-17.7 – -1.7	18.2–25.4	WRF/Chem-MADRID	This work ^c
24-hr average PM_{2.5}							
NY	7/1-9/30,2004	5.4	13.2	—	—	Eta/CMAQ	HO07
NY	1/1-3/31,2005	6.2	14.5	—	—	Eta/CMAQ	HO07
NY	6/1-7/31,2005	4.4	13.6	—	—	Eta/CMAQ	HO07
PN	8/1-11/30,2004	2.1–2.2	—	17–32	70–81	MM5/CMAQ	CH08
E US	7/14-8/18,2004	-3.2	8.8	-21.0	41.2	Eta/CMAQ	YU08
E Texas	8/31-10/12,2006	-1.3	5.5	—	—	7-model ensemble ^a	DJ10
NA	Summer 2008	-2.08	12.8	—	—	GEM-CHRONOS	MA09
NA	Winter 2008	0.86	14.1	—	—	GEM-CHRONOS	MA09
NA	Summer 2009	-0.70	12.9	—	—	GEM-CHRONOS	MA09
NA	Summer 2008	0.69	13.5	—	—	GEM-MACH15	MA09
NA	Winter 2008	-0.18	15.9	—	—	GEM-MACH15	MA09
NA	Summer 2009	2.08	13.6	—	—	GEM-MACH15	MA09
SE US	5/1-9/30,2009	-0.6	5.9	-5.6	37.0	WRF/Chem-MADRID	MT11
SE US	5/1-9/30,2009-2011	-1.3 – -0.6	5.9 – 8.7	-10.1 – -5.2	36.7 – 38.9	WRF/Chem-MADRID	YA14 ^b
SE US	5/1-9/30,2009-2011	-1.3 – 3.6	4.8 – 20.1	-10.1 – 34.3	35.2 – 65.5	WRF/Chem-MADRID	YA14 ^c
SE US	12/1-02/28,2009-2012	-1.1 – 0.2	5.4 – 6.8	-10.2 – 1.4	39.9 – 41.6	WRF/Chem-MADRID	YA14 ^b
SE US	12/1-02/28,2009-2012	-2.9 – 3.1	4.9 – 9.3	-20.6 – 36.6	0.6 – 65.5	WRF/Chem-MADRID	YA14 ^c
SE US	5/1-9/30,2012-2014	-0.5 – 1.4	5.1–5.7	-4.0 – 15.0	36.0 – 40.0	WRF/Chem-MADRID	This work ^b
SE US	5/1-9/30,2012-2014	-0.5 – 4.8	3.8–10.8	2.0 – 53.0	33.0 – 74.0	WRF/Chem-MADRID	This work ^c
SE US	12/1-02/28,2012-2015	0.2 – 0.8	5.5–6.1	0.8 – 8.3	42.6 – 47.4	WRF/Chem-MADRID	This work ^b
SE US	12/1-02/28,2012-2015	0.1 – 5.2	4.9–10.5	4.9 – 68.4	37.3 – 89.0	WRF/Chem-MADRID	This work ^c

1. MB: Mean Bias; RMSE: Root Mean Square Error; NMB: Normalized Mean Bias; NME: Normalized Mean Error. SE US: Southeastern U.S.; E US: Eastern U.S.; NE US: Northeastern U.S.; SE CA: southeastern Canada; PN: Pacific Northwest; NY: New York State; E Texas: eastern Texas; NA: North America. The unit for MB and RSME are ppb for O₃ and $\mu\text{g m}^{-3}$ for PM_{2.5}.
2. Superscript a: the 7 models include: WRF/Chem-2 (27-km), WRF/Chem-2 (12-km), CHRONOS, AURAMS, STEM-2K3, BAMS (15-km), and NMM/CMAQ; b: statistics based on evaluation against AirNow; c: statistics based on evaluation against all datasets.
3. MT11: Chuang et al. (2011); KA05: Kang et al. (2005); ED06: Eder et al. (2006); HO07: Hogrefe et al. (2007); MK07: McKeen et al. (2007); YU07: Yu et al. (2007); CH08: Chen et al. (2008); MA09: Makar et al., 2009; ED09: Eder et al. (2009); YU08: Yu et al. (2008); DJ10: Djalalova et al. (2010); YA14: Yahya et al. (2014).

Table 4. Categorical evaluation of RT-AQF results against AirNow for O₃ and PM_{2.5} predictions.

Area	Period	A (%)	CSI (%)	POD (%)	B	FAR (%)	Model	Reference
Maximum 1-hr average O₃								
NE US	8/5-29,2002	99.2	9.7	14.0	0.6	76	MAQSIP-RT	KA05
NE US	8/5-29,2002	97.0	9.8	29.8	2.3	87.2	MM5/Chem	KA05
NE US	8/5-29,2002	99.0	8.3	18.2	1.4	86.7	Hysplit/CheM	KA05
SE US	5/1-9/30,2009	94.0	5.2	31.3	5.3	94.1	WRF/Chem-MADRID	MT11
SE US	5/1-9/30,2009-2011	94.6-96	5.2-13.8	17-31.3	0.6-5.3	67-94.1	WRF/Chem-MADRID	YA14
SE US	12/1-02/28,2009-2012	100	0	0	0	0	WRF/Chem-MADRID	YA14
SE US	5/1-9/30,2012-2014	94.2-97.7	3.6-15.5	18.8-30.5	0.9-7.9	71.5-96.1	WRF/Chem-MADRID	This work
SE US	12/1-02/28,2012-2015	100	0	0	0	0	WRF/Chem-MADRID	This work
Maximum 8-hr average O₃								
NE US	8/1-10,2001	80.0	34.0	49.0	1.1	13.0	MM5/MAQSIP_RT	MC04
NE US	8/5-29,2002	85.8	18.1	26.7	0.7	64.0	MAQSIP-RT	KA05
NE US	8/5-29,2002	76.2	17.6	36.4	1.4	74.6	MM5/Chem	KA05
NE US	8/5-29,2002	89.5	5.8	7.1	0.3	76.3	Hysplit/Chem	KA05
NE US	6/1-9/30,2004	98.9	14.2	41.0	2.3	82.1	Eta/CMAQ	ED06
NY	7/1-9/30,2004	84.0-95.2	31.4-53.2	46.5-84.8	—	32.9-55.2	Eta/CMAQ	HO07
	1/1-3/31, 6/1-9/30,2005	96.1-99.8	0.0-29.0	0.0-58.3	—	36.7-82.5	Eta/CMAQ	HO07
NE US	8/12,2005	91.6	23.4	31.3	0.7	51.6	Eta/CMAQ	LE08
E US	8/12,2005	90.4	24.3	37.5	0.9	59.1	Eta/CMAQ	LE08
CONUS	8/12,2005	87.4	26.0	54.2	1.6	66.7	Eta/CMAQ	LE08
SE US	5/1-9/30, 2009	85.6	14.0	33.3	1.7	80.6	WRF/Chem-MADRID	MT11
CONUS	6/1-8/31,2010	86-91	0.17-0.21	0.71-0.76		0.77-0.82	WRF-NMM/CMAQ	CH13
CONUS	01/01-12/31,2010	93-96	0.17-0.21	0.64-0.67		0.76-0.81	WRF-NMM/CMAQ	CH13
SE US	5/1-9/30,2009-2011	81.4-85.7	14-24.9	29.1-33.3	0.6-1.7	48.6-80.6	WRF/Chem-MADRID	YA14
SE US	12/1-02/28,2009-2012	98.7-100	0	0	0	N/A ^a	WRF/Chem-MADRID	YA14
SE US	5/1-9/30,2012-2014	80.2-85.3	9.9-25.3	26.6-46.7	0.8-4.2	54.9-88.9	WRF/Chem-MADRID	This work
SE US	12/1-02/28,2012-2015	98.7-99.2	0	0	0	N/A ^a	WRF/Chem-MADRID	This work
24-hr average PM_{2.5}								
NY	7/1-9/30,2004	60.8-89.7	22.5-53.7	24.3-90.9	—	25.0-55.0	Eta/CMAQ	HO07
	1/1-3/31, 6/1-7/31,2005	91.4-99.7	0-3.6	0-44.7	—	N/A ^a , 96.2-100	Eta/CMAQ	HO07
E Texas	8/31-10/12,2006	—	0.0-8.0	0.0-14	—	80-100	7-model ensemble ^b	DJ10
SE US	5/1-9/30,2009	76.2	22.3	31.5	0.7	56.6	WRF/Chem-MADRID	MT11
SE US	5/1-9/30,2009-2011	70.7-76.2	22.3-27.9	31.5-36	0.6-0.7	44.6-56.7	WRF/Chem-MADRID	YA14
SE US	12/1-02/28,2009-2012	82.2-85.9	14.8-22.2	27.7-38.3	0.7-1.2	61.3-76.6	WRF/Chem-MADRID	YA14
SE US	5/1-9/30,2012-2014	77.5-83.2	10.3-21.3	15.3-40.1	0.6-1.3	68.3-75.9	WRF/Chem-MADRID	This work
SE US	12/1-02/28,2012-2015	83.5-85.3	14.7-17.1	25.5-31.8	1.0-1.2	72.1-74.1	WRF/Chem-MADRID	This work

1. A: Accuracy; CSI: Critical Success index; POD: Probability Of Detection; B: Bias; FAR: False Alarm Ratio. SE US: Southeastern U.S.; NE US: Northeastern U.S.; E US: eastern U.S.; E Texas: eastern Texas; NY: New York State, CONUS: continental U.S.
2. Superscript a: An FAR of N/A indicates that no exceedances were predicted by the AQF model; b: the seven models include: WRF/chem-2 (27-km), WRF/chem-2 (12-km), CHRONOS, AURAMS, STEM-2K3, BAMS (15-km), and NMM/CMAQ;
3. MT11: Chuang et al. (2011); MC04: McHenry et al. (2004); KA05: Kang et al. (2005); ED06: Eder et al. (2006); HO07: Hogrefe et al. (2007); LE08: Lee et al. (2008); DJ10: Djalalova et al. (2010); CH13: Chai et al. (2013); YA14: Yahya et al. (2014).

List of Figure Captions

Figure 1. Discrete evaluation of the maximum 1-hr and 8-hr O₃ and 24-hr average PM_{2.5} for (a) O₃ seasons and (b) winters during 2009-2015.

Figure 2. Spatial distributions of maximum 1-hr O₃ and 8-hr O₃ during the O₃ seasons and average 24-hr PM_{2.5} concentrations during the O₃ and winter seasons during 2012-2015. The observations are symbolled as circles, they are taken from AIRNow, AIRS-AQS, CASTNET, and SEARCH for O₃ and from AIRNow, IMPROVE, STN, and SEARCH for PM_{2.5}.

Figure 3. Time series of the observed and forecasted maximum 1-hr O₃ and 8-hr O₃ concentrations for O₃ seasons during 2012-2015. The observations are taken from AIRNow.

Figure 4. Time series of the average 24-hr average PM_{2.5} concentrations for (a) O₃ seasons and (b) winters during 2012-2015. The observations are taken from AIRNow.

Figure 5. Categorical evaluation of the maximum 1-hr and 8-hr O₃ and 24-hr average PM_{2.5} for (a) O₃ seasons and (b) winters during 2009-2015.

Figure 6. Spatial distributions of satellite-derived and simulated column NO₂, TOR, and AOD during (a) the 2012 O₃ season (rows 1 and 2), and (b) the winter of 2012-2013 (rows 3 and 4).

Figure 7. Comparison of CO spatial distributions in Aug. 2012: (a) satellite observation from MOPPIT, (b) baseline simulation, and (c) sensitivity simulation.

Figure 8. Comparison of TOR spatial distributions in Dec. 2012: (a) satellite observation from OMI, (b) baseline simulation, and (c) sensitivity simulation.

Figure 9. Changes in observed and forecasted T2, Precip, and WS10 (relative to 2009) and SWDOWN, LWDOWN, and CF (relative to 2011) during 2010-2015.

Figure 10. Changes in observed and forecasted surface O_3 and $PM_{2.5}$ concentrations (relative to 2009) and column CO, NO_2 , SO_2 , TOR, AOD (relative to 2011) during 2010-2015.

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(b) Sim_base

(c) Sim_sen

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