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A Comparative Study of Two-way and Offline Coupled WRF v3.4 and CMAQ v5.0.2 over the Contiguous U.S.: Performance Evaluation and Impacts of Chemistry-Meteorology Feedbacks on Air Quality

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Abstract

The two-way coupled Weather Research and Forecasting and Community Multiscale Air Quality (WRF-CMAQ) model has been developed to more realistically represent the atmosphere by accounting for complex chemistry-meteorology feedbacks. In this study, we present a comparative analysis of two-way (with consideration of both aerosol direct and indirect effects) and offline coupled WRF v3.4 and CMAQ v5.0.2 over the contiguous U.S. Long-term (five-year of 2008-2012) simulations using WRF-CMAQ with both offline and two-way coupling modes are carried out with anthropogenic emissions based on multiple years of the U.S. National Emission Inventory and chemical initial and boundary conditions derived from an advanced Earth system model (i.e., a modified version of the Community Earth System Model/Community Atmospheric Model). The comprehensive model evaluations show that both two-way WRF-CMAQ and WRF-only simulations perform well for major meteorological variables such as temperature at 2 m, relative humidity at 2 m, wind speed at 10 m, and precipitation (except for against the National Climatic Data Center data) as well as shortwave/longwave radiation. Both two-way and offline CMAQ also show good performance for ozone (O_3) and fine particulate matter ($PM_{2.5}$). Due to the consideration of aerosol direct and indirect effects, two-way WRF-CMAQ shows improved performance over offline-coupled WRF and CMAQ in terms of spatiotemporal distributions and statistics, especially for radiation, cloud forcing, O_3, sulfate, nitrate, ammonium, and elemental carbon as well as tropospheric O_3 residual and column nitrogen dioxide (NO_2). For example, the mean biases have been reduced by more than 10 W m^{-2} for shortwave radiation and cloud radiative forcing and by more than 2 ppb for max 8-h O_3. However, relatively large biases still exist for cloud predictions, some $PM_{2.5}$ species, and PM_{10}, which warrant follow-up studies to better understand those issues. The impacts of chemistry-meteorological feedbacks are found to play important roles in affecting regional air quality in the U.S. by reducing domain-average concentrations of carbon monoxide (CO), O_3, nitrogen oxide (NO_x), volatile organic compounds (VOCs), and $PM_{2.5}$ by 3.1% (up to 27.8%), 4.2% (up to 16.2%), 6.6% (up to 50.9%), 5.8% (up to 46.6%), and 8.6% (up to 49.1%), respectively, mainly due to reduced radiation, temperature, and wind speed. The overall performance of the two-way coupled WRF-CMAQ model achieved in this work is generally good or satisfactory and the improved performance for two-way coupled WRF-CMAQ should be considered along with other factors in developing future model applications to inform policy making.

Keywords: CMAQ, Two-way coupling, Evaluation, Chemistry-meteorology feedback

1. Introduction

The Community Multiscale Air Quality (CMAQ) modeling system developed by the U.S. Environmental Protection Agency (EPA) (Byun and Schere, 2006; Scheffe et al., 2016; San Joaquin Valley APCD, 2018; Pye et al., 2020; U.S. EPA, 2020) has been extensively used by both scientific community and governmental agencies over various geographical regions and under different meteorological and air pollution conditions to address major key air quality issues such as atmospheric ozone (O_3), acid rain, regional haze, and trans-boundary or long-range transport of air pollutants during the past decades over North America (Zhang et al., 2009a,b; Wang and Zhang, 2012; Hogrefe et al., 2015), Asia (Wang et al., 2009, 2012; Liu et al., 2010; Zheng et al., 2015; Li et al., 2017; Xing et al., 2017; Yu et al., 2018; Mehmood et al., 2020), and Europe (Kukkonen et al., 2012; Mathur et al., 2017; Solazzo et al., 2017). The CMAQ model is traditionally driven offline by the three-dimensional meteorology fields generated separately from other meteorological models such as the Weather Research and Forecasting (WRF) model, and the dynamic feedbacks of chemistry predictions on meteorology are neglected. However, more recently (IPCC, 2018), chemistry-meteorology feedbacks have been found to play important roles in affecting the both global and regional climate change and air quality (Jacobson et al., 1996; Mathur et al., 1998; Ghan et al., 2001; Zhang, 2008; Zhang et al., 2010, 2015a,b, 2017; Grell and Baklanov, 2011; Wong et al., 2012; Baklanov et al., 2014; Yu et al., 2014; Gan et al., 2015a; Wang et al., 2015; Xing et al., 2015a,b; Yahya et al., 2015a,b; Hong et al., 2017; Jung et al., 2019). Feedbacks of aerosols on radiative transfer through aerosol-radiation interactions (i.e., aerosol direct forcing) and aerosol-cloud interactions (i.e., aerosol indirect forcing) are especially important (Zhang, 2008; Zhang et al,, 2015a,b; Baklanov et al., 2014; Wang et al., 2015; Yahya et al., 2015a,b). Recognizing this importance, as well as the recent advances in knowledge on chemistry-meteorology interactions and computational resources, the U.S. EPA developed a two-way coupled WRF-CMAQ model that accounts for the aerosol direct effect alone (Wong et al., 2012). This version of CMAQ has been applied for both regional and hemispheric studies (Wang et al., 2014; Hogrefe et al., 2015; Xing et al., 2016, 2017; Hong et al., 2017, 2020; Sekiguchi et al., 2018; Yoo et al., 2019). For example, Xing et al. (2016) showed that aerosol direct feedbacks may further improve air quality resulting from emission controls in the U.S. and also indicated that coupled models are key tools for quantifying such feedbacks. Reduction in atmospheric ventilation resulting from aerosol induced surface cooling can exacerbate ground level air pollution. Hong et al. (2017) estimated an increase by 4.8%-9.5% in concentrations of major air pollutants over China in winter due to incorporation of such effects. Xing et al. (2017) reported that the aerosol direct effects could reduce daily max 1h O_3 by up to 39 μg m^{-3} over China in January through reducing solar radiation and photolysis rates. Hong et al. (2020) found that the benefits of reduced pollutant emissions through weakening aerosol direct effects can largely offset the additional deaths caused by the warming

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effect of greenhouse gases over China. Some of those studies have also found that the missing aerosol indirect effects in WRF-CMAQ may introduce large model biases on their simulations of radiation and thus air quality (Wang et al., 2014; Sekiguchi et al., 2018; Yoo et al., 2019). There has been a growing awareness that both aerosol effects should be considered together to provide greater fidelity in coupling complex atmospheric processes among chemistry, aerosols, cloud, radiation, and precipitation (Grell and Baklanov, 2011). To address this issue and better represent the one-atmosphere modeling capability of CMAQ, Yu et al. (2014) further extended the two-way coupled WRF-CMAQ model by including aerosol indirect effects and improved WRF-CMAQ's capability for predicting cloud and radiation variables.

Different from the traditional online integrated air quality models such as the Gas, Aerosol, Transport, Radiation, General Circulation, and Mesoscale Meteorological (GATOR-GCMM) model (Jacobson, 2001), the WRF model coupled with chemistry (WRF/Chem; Grell et al., 2005) and the WRF model coupled with the Community Atmosphere Model version 5 (WRF-CAM5; Ma et al., 2013; Zhang et al., 2015a,b; 2017), in which atmospheric dynamics and chemistry are integrated and simulated altogether without an interface between meteorology and atmospheric chemistry (Zhang et al., 2013), two-way WRF-CMAQ (also referred to as the online access model) is created by combining existing meteorology (i.e., WRF) and atmospheric chemistry (i.e., CMAQ) models with an interactive interface (Yu et al., 2014). As pointed out by Yu et al. (2014), the main advantage of two-way CMAQ is to allow the existing numerical techniques to be used in both WRF and CMAQ to facilitate future independent development of both models while also maintaining CMAQ as a stand-alone model (the offline capability). In the past, a number of studies have compared and evaluated online vs. offline-coupled model performance (Pleim et al, 2008; Matsui et al., 2009; Wilczak et al., 2009; Lin et al., 2010;

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Herwehe et al., 2011; Yu et al., 2011; Wong et al., 2012; Zhang et al., 2013, 2016a; Choi et al., 2019). However due to the missing offline-coupled mode or component for most online-coupled models, many of those intercomparison studies are subject to some key limitations such as inconsistent model treatments in chemical options (Matsui et al., 2009; Lin et al., 2010; Zhang et al., 2013; Choi et al., 2019) or in both physical and chemical options (Wilczak et al., 2009; Herwehe et al., 2011; Zhang et al., 2016a), different domain projection methods or resolutions (Wilczak et al., 2009; Lin et al., 2010; Zhang et al., 2013), or disunified model inputs (Wilczak et al., 2009; Lin et al., 2010; Zhang et al., 2013). Due to the unique coupling approach, two-way WRF-CMAQ can be used to overcome those limitations and set up ideal intercomparisons between online and offline simulations using consistent model treatments (Pleim et al, 2008; Yu et al., 2011; Wong et al., 2012).

In this study, we provide a robust examination of model improvements by considering chemistry-meteorology feedbacks and their impacts on the U.S. air quality using the two-way WRF-CMAQ model (same version as in Yu et al., 2014) with both aerosol direct and indirect effects. Long-term (five-year of 2008-2012) simulations using both two-way and offline coupled WRF and CMAQ models are carried out and compared to the best of our knowledge for the first time over the contiguous U.S. (CONUS) with anthropogenic emissions based on multiple years of the U.S. National Emission Inventory (NEI) and chemical initial and boundary conditions (ICONs/BCONs) downscaled from the advanced Earth system model, i.e., an updated version of the Community Earth System Model/CAM5 (CESM/CAM5; He and Zhang, 2014; Glotfelty et al., 2017). Our objectives include 1) perform a comprehensive model evaluation for major meteorological variables and chemical species from this long-term application of the two-way

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coupled WRF-CMAQ; and 2) conduct a comparative study of two-way and offline coupled WRF and CMAQ to examine the impacts of chemistry-meteorology interactions on U.S. air quality.

Compared to previous studies in the literature, there are a few key features of this work. First, the intercomparisons between two-way (or online) and offline WRF-CMAQ are performed here using consistent model configurations including both physical/chemical options and inputs. Second, unlike a few previous intercomparison studies (Pleim et al, 2008; Yu et al., 2011; Wong et al., 2012) using two-way WRF-CMAQ with only aerosol direct effects for relatively short episodes, the model version in this work includes both aerosol direct and indirect effects and simulations are conducted for multiple years to provide more robust assessments. Third, compared to other studies (e.g., Yahya et al., 2015a,b; Choi et al., 2019) focusing on the impacts of chemistry-meteorology feedbacks on meteorology only or limited chemical species, this study performs comprehensive and extensive evaluation and comparison to demonstrate importance of chemistry-meteorology feedbacks on regional meteorology and air quality.

2. Model description, simulation setup, and evaluation protocols

Two sets of five-year (i.e., 2008-2012) long-term simulations are conducted using the two-way coupled WRF v3.4-CMAQ v5.0.2 model with both aerosol direct and indirect effects and the sequentially offline-coupled WRF v3.4 and CMAQ v5.0.2 model, respectively, over the CONUS with 36-km horizontal grid spacing. The vertical resolution for these simulations consists of 34 layers from the surface (~38 m) to 100 hPa (~15 km). The two-way coupled WRF-CMAQ includes estimations of aerosol optical properties based on prognostic aerosol size distributions and composition . These aerosol optical properties are then used to modulate the shortwave radiation budget estimated using the Rapid and accurate Radiative Transfer Model for

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General circulation (RRTMG) radiation scheme (Iacono et al., 2008) in WRF. Additionally, aerosol indirect effects, including the first (cloud albedo) and second (cloud lifetime) indirect aerosol forcing and the glaciation (ice and mixed-phase cloud lifetime) indirect aerosol forcing are also modeled. More details on the model development of this version of WRF-CMAQ can be found in Yu et al. (2014). On the other hand, the WRF only model calculates the radiation budgets by using prescribed aerosol optical properties such as aerosol optical depth, single scattering albedo and asymmetry parameters and cloud formation by assuming default droplet number concentration and fixed cloud effective radius, which may not be representative for the large regions with complex air pollution conditions. Both the two-way and offline coupled WRF-CMAQ use the same model configurations as shown in Table S1 in the supplementary material, except that prognostic aerosol impacts on radiation and clouds are fully treated in two-way WRF-CMAQ. The physics options include the RRTMG shortwave and longwave radiation schemes, the Asymmetric Convective Model (ACM2) planetary boundary layer (PBL) scheme (Pleim, 2007), the Pleim-Xiu (PX) land-surface scheme (Xiu and Pleim, 2001), the Morrison two-moment microphysics scheme (Morrison et al., 2009), and version 2 of the Kain–Fritsch (KF2) cumulus scheme (Kain, 2004). The chemical options include the Carbon Bond 2005 (CB05) chemical mechanism (Yarwood et al., 2005) with additional chloride chemistry (Sarwar et al., 2008), the sixth generation CMAQ aerosol module (AERO6) (Appel et al., 2013), and CMAQ's aqueous phase chemistry (AQCHEM). In addition, the time steps of dynamics and radiation for two-way WRF-CMAQ are set as 1 min and 15 mins, respectively, and the call frequency for CMAQ in the two-way coupled model is set to be 5 mins.

The meteorological ICONs/BCONs are generated from the National Centers for Environmental Prediction Final Analysis (NCEP-FNL) datasets and the chemical

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ICONs/BCONs are downscaled from a modified version of CESMv1.2.2/CAM5 (He and Zhang, 2014; Glotfelty et al., 2017). The anthropogenic emissions are based on two versions of NEI. NEI 2008 and NEI 2011 are used to cover the 5-year period, i.e., NEI 2008 for 2008-2010 and NEI 2011 for 2011-2012, respectively. Biogenic emissions are calculated online using the Biogenic Emissions Inventory System (BEIS) v3 (Schwede et al., 2005). The sea-salt and dust emissions are also generated online by CMAQ's inline modules (Zhang et al., 2005; Foroutan et al., 2017). Two-way coupled WRF-CMAQ simulations are reinitialized every 5 days to make meteorology simulations as accurate as possible while preserving the two-way chemistry-meteorology feedbacks. The WRF-only simulations that are used to drive the offline CMAQ simulations apply the same reinitialization method to be consistent with the two-way coupled WRF-CMAQ simulations.

The model evaluation in this work mainly focuses on the long-term climatological type of performance by comparing 5-year average spatially and temporally matched model predictions of major surface meteorological/radiation-cloud variables and surface/column chemical species against various surface/satellite observations and reanalysis data. The surface meteorological data include temperature at 2 m (T2), relative humidity at 2 m (RH2), wind speed at 10 m (WS10), and wind direction at 10 m (WD10) from the National Climatic Data Center (NCDC), and precipitation from the NCDC, the National Acid Deposition Program (NADP), the Global Precipitation Climatology Project (GPCP), the Parameter-elevation Regressions on Independent Slopes Model (PRISM), and the Tropical Rainfall Measuring Mission Multisatellite Precipitation Analysis (TMPA). The radiation and cloud data include downward shortwave radiation at the ground surface (SWDOWN), net shortwave radiation at the ground surface (GSW), downward longwave radiation at the ground surface (GLW), outgoing longwave radiation at the top of the

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atmosphere (OLR), and shortwave and longwave cloud forcing (SWCF and LWCF) from the Clouds and the Earth's Radiant Energy System (CERES); aerosol optical depth (AOD), cloud fraction (CF), cloud water path (CWP), and cloud optical thickness (COT) from the MODerate resolution Imaging Spectroradiometer (MODIS); and cloud droplet number concentration (CDNC) derived based on MODIS data by Bennartz (2007). The chemical data include surface O_3 from the Aerometric Information Retrieval System-Air Quality Subsystem (AIRS-AQS) and the Clean Air Status and Trends Network (CASTNET); surface fine particulate matter ($PM_{2.5}$) and its constituents including sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), elemental carbon (EC), organic carbon (OC), and total carbon (TC = EC + OC) from the Interagency Monitoring of Protected Visual Environments (IMPROVE) and the Chemical Speciation Network (CSN); surface coarse particulate matter (PM_{10}) from the AQS; and column abundance variables such as column carbon monoxide (CO) from the Measurements of Pollution in the Troposphere (MOPITT), tropospheric ozone residual (TOR) from the Ozone Monitoring Instrument (OMI), and column nitrogen dioxide (NO_2) and formaldehyde (HCHO) from the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY).

The satellite datasets used in this study are all level-3 gridded monthly-averaged data with various resolutions (i.e., 0.25° for OMI and PRISM, 0.5° for SCIAMACHY, 1° for CERES, GPCP, MODIS, and MOPITT). For the calculation of model performance statistics, the satellite data with different resolutions are mapped to CMAQ's Lambert conformal conic projection using bi-linear interpolation in the NCAR command language. CMAQ model outputs at approximate time of the satellite overpass are paired with the satellite retrievals to facilitate a consistent comparison. Modeled CDNC is calculated as the average value of the layer of low-level warm clouds between 950 and 850 hPa as suggested by Bennartz (2007). Following the

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approach of Wielicki et al. (1996), the SWCF and LWCF are calculated as the difference between the clear-sky and the all-sky reflected radiation at the top of atmosphere for both simulations and observations.

The statistical performance evaluation follows a protocol similar to that of Zhang et al. (2006, 2009a) and Yahya et al. (2016) and uses well-accepted statistical measures such as correlation coefficient (R), mean bias (MB), root mean square error (RMSE), normalized mean biases (NMB), and normalized mean error (NME) (S. Yu et al., 2006). Because of different sampling protocols among monitoring networks, the evaluation is conducted separately for individual networks for the same simulated variables/species.

3. Comprehensive model evaluation of two-way WRF-CMAQ

3.1 Meteorological evaluation

3.1.1 Surface meteorological variables

Figures 1a-d show the spatial distribution of 5-year average MBs for T2, RH2, WS10, and hourly precipitation from two-way WRF-CMAQ against the NCDC data in 2008-2012 and Table 1 summarizes the statistics for the same variables. All variables except for precipitation show overall good or moderate spatial performance with many sites showing MBs within ±0.6 $^{\circ}$C for T2, ±5 % for RH2, ±1 m s^{-1} for WS10, and ±0.1 mm hr^{-1} for precipitation, respectively. WRF-CMAQ tends to overpredict T2 (i.e., warm bias) over widespread areas of domain especially along the Atlantic coast, the eastern/southeastern U.S., the Central U.S., and Pacific coast. The model also shows cold biases (i.e., underprediction in T2) over the mountainous regions and northeastern U.S. Similar warm biases of T2 have been previously reported by Cohen et al. (2015) and are found to be associated with the relatively deeper PBL depth using the

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non-local ACM2 PBL scheme. The relatively larger warm/cold biases over coastal and mountainous areas are likely caused by the coarse spatial grid spacing of 36-km which cannot resolve the complex topography. Compared to many previous WRF studies (Wang et al., 2012; Brunner et al., 2015; Yahya et al., 2016), which typically show cold T2 biases, the overall small warm biases in this study can be attributed to the soil moisture nudging technique used in the PX land surface scheme (Pleim and Gilliam, 2009). The spatial patterns of MBs for RH2 show a clear anti-correlation compared to T2 (i.e., RH2 is overpredicted where T2 is underpredicted and vice versa). This is consistent with how RH2 is calculated based on T2. The spatial distribution of MBs for WS10 also shows dominant overpredictions especially along coastlines, indicating the prescribed sea-surface temperature might not be sufficient to resolve the air-sea interactions. Systematic overpredictions of hourly precipitation against NCDC data are found to be mainly caused low non-convective precipitation events and should be attributed to the uncertainties associated with the Morrison microphysics scheme (Yahya et al., 2016).

The precipitation performance is further examined by comparing WRF-CMAQ with GPCP and PRISM as shown in Figures 1e-g. The spatial distribution of precipitation is well simulated by WRF-CMAQ especially over the land against both GPCP and PRISM by capturing the hot spots along the Pacific Northwest coast and some areas over eastern U.S. Moderate overpredictions of precipitation against GPCP over the Atlantic Ocean and Gulf of Mexico are also evident, possibly due to overprediction of convective precipitation intensity by the Kain–Fritsch cumulus scheme (Hong et al., 2017) over ocean. As shown in Table 1, the domain-average statistics demonstrate good performance for all variables except for precipitation against NCDC in terms of MBs, NMBs, RMSE, and Rs. For example, the MBs for T2, RH2, WS10, and precipitation are 0.1 °C, 2.2%, 0.44 m s^{-1}, and 0.14-0.28 mm day^{-1}, respectively, and Rs for those

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variables are typically between 0.5-0.98, which are well within the performance benchmark values recommended by Zhang et al. (2013) and Emery et al. (2017).

3.1.2 Radiation and cloud variables

Figure 2 compares the 5-year average spatial distribution of major radiation variables (i.e., SWDOWN, GSW, GLW, OLR, and AOD) based on the satellite retrievals and two-way WRF-CMAQ simulations, and Table 1 summarizes the domain-average model performance statistics. WRF-CMAQ predicts the longwave radiation variables GLW and OLR very well with domain-average of NMBs of -1.9% and 0.8%, respectively, and Rs of 0.99 for both. The shortwave radiation variables SWDOWN and GSW are overpredicted on average with NMBs of 13.0% and 11.1%, respectively, and Rs of 0.97 for both. The simulations also reliably reproduce the spatial distribution of both longwave and shortwave radiation compared to observations. The relatively large overpredictions for shortwave radiation are very likely caused by the underpredictions of aerosol direct radiative forcing reflected from the underpredictions of AOD (Figure 2) as well as underprediction of indirect cloud radiative forcing (see Figure 3). It has been reported that WRF v3.4 does not treat the subgrid cloud feedback to radiation, which could also contribute to the overpredictions in shortwave radiation (Alapaty et al., 2012; Hong et al., 2017). The model largely underpredicts the magnitude of AOD (NMB: -64.8%), while providing a reasonable representation of the spatial distribution of AOD over the U.S., with generally higher values in the east and lower values in the west. The model also underpredicts the elevated AODs over oceans and the northern part of domain. Similar AOD underpredictions have been reported in previous studies over the U.S. using two-way coupled WRF-CMAQ (Gan et al., 2015a; Hogrefe et al., 2015; Xing et al., 2015a). The relatively large underpredictions of AOD may be caused by several factors. First, underprediction of $PM_{2.5}$ concentrations, particularly

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SO_4^{2-} and OC (Table 2), can contribute significantly to the underprediction of AOD, especially over the eastern U.S. Second, the underestimation of dust emissions may contribute to missing hot spots from the model over arid areas in CA and AZ (Foroutan et al., 2017) and underestimates of sea-salt emissions may lead to missing elevated AODs over oceans (Gan et al., 2015b). Third, challenges in adequately representing prescribed and wildfire emissions in the NEI (Kelly et al., 2019) may cause many missing hot spots over large areas of the Pacific Northwest, CA, Canada, and the eastern U.S. Fourth, uncertainties in BCONs of $PM_{2.5}$ concentrations may further contribute to underpredictions of AOD over oceans and the northern part of the domain. For example, Kaufman et al. (2001) found that the background AOD could reach 0.1 over the Pacific Northwest using Aerosol Robotic Network (AERONET) data. The AODs in the current simulation seem to be biased low (between 0.06-0.08) and indicate potential underpredictions of $PM_{2.5}$ BCONs, especially in the free troposphere. Finally, there are uncertainties associated with MODIS retrievals. Remer et al. (2005) found that the uncertainty of level 3 MODIS monthly AODs can be up to ±0.05±0.15AOD over the land due to clouds and surface reflectance. More AOD data from other satellites or AERONET might be considered in the future work to provide more robust ensemble type of evaluation for AOD.

Figures 3 and 4 compare the 5-year average spatial distribution of major cloud and cloud radiative variables for the satellite retrievals and two-way WRF-CMAQ simulations, and Table 1 summarizes the domain-average model performance statistics. As shown in Figure 3, WRF-CMAQ tends to largely underpredict CDNC, COT, and CWP over the whole domain with the domain-average NMBs of -82.1%, -80.1%, and -51.2%, respectively. Despite the large underprediction of those cloud variables, the spatial correlations are generally predicted well, especially for COT and CWP with Rs of 0.84 and 0.79, respectively. Compared to the other cloud variables, CF is much better predicted with an NMB of -12.2% and an R of 0.92, which is consistent with the performance reported in Yu et al. (2014). The model can reproduce the high CFs over northern and northeastern part of domain as well as over oceans while capturing the low CFs over the mountainous and plateau regions in the U.S. and Mexico. In addition to the underprediction of $PM_{2.5}$ (thus underestimating CCN), the large underpredictions of cloud variables (especially CDNC and COT) can be attributed to uncertainties in aerosol microphysics schemes (Yahya et al., 2016) as well as missing aerosol indirect effects on subgrid convective clouds (Yu et al., 2014). Gantt et al. (2014) and Zhang et al. (2015b) also showed the aerosol activation scheme (i.e., Abdul-Razzak and Ghan, 2000) used in the current version of WRF-CMAQ may have underestimated CDNC and thus CWP and COT due to some missing processes such as insoluble aerosol adsorption and giant cloud condensation nuclei. Overall, the relatively poor model performance for cloud variables reflects current limitations in representing aerosol indirect effects and aerosol-cloud interactions in state-of-science online coupled models. Further model improvements that incorporate new knowledge from emerging studies should be conducted in the future.

As shown in Figure 4, WRF-CMAQ predictions of SWCF and LWCF agree well with the satellite-based values. The model partially captures the elevated SWCF and LWCF over the Atlantic Ocean, Pacific Northwest, and widespread areas over the eastern U.S. The domain-average NMBs are -26.0% for SWCF and -22.2% for LWCF, respectively. As discussed earlier, the underpredictions of SWCF may partially contribute the overprediction of SWDOWN (more shortwave radiation reaching the ground) and those of LWCF may further lead to the overpredictions in OLR (more longwave radiation emitted into the space). The performance of SWCF and LWCF is consistent with the 12-km simulation reported in Yu et al. (2014) and even

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slightly better in terms of NMBs, which might be associated with the long-term vs. short-term simulations. It is also worth noting that SWCF (LWCF) is calculated as the difference between the clear-sky and all-sky shortwave (longwave) radiation at the top of atmosphere, and so performance for SWCF and LWCF depends on performance for both radiation and cloud properties. The generally better performance in terms of model bias for SWCF and LWCF compared to the cloud variables seems to be driven by the relatively good performance of shortwave/longwave radiation in the model.

3.2 Chemical evaluation

3.2.1 O_3

Figure 5a shows the spatial distribution of simulated average daily maximum 8-h O_3 from two-way WRF-CMAQ overlaid with observations from both the AIRS-AQS and CASTNET networks. WRF-CMAQ shows good performance by capturing the spatial distribution of max 8-h O_3 over widespread areas of the domain. The model tends to overpredict O_3 along coastlines in the southeastern U.S., Gulf of Mexico, and Pacific coast, which can be attributed to a poor representation of coastal boundary layers (Yu et al., 2007), the warm T2 biases as shown in Figure 1, and lack of O_3 sink via halogen chemistry (Sarwar et al., 2015) and deposition to water (Gantt et al., 2017). The simulation also underpredicts O_3 in widespread areas in the Midwest, eastern, and mountainous regions of the U.S., which is consistent with the results of 36-km simulations from Wang and Zhang (2012) that used an earlier version of CMAQ v4.6 with the same CB05 gas-phase mechanism. In addition to cold T2 biases over those areas (Figure 1), the underpredictions are also believed to be associated with inaccurate representations of precursor emissions and elevated/complex terrain due to the coarse grid spacing of 36-km over those

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regions. Wang and Zhang (2012) found that their 12-km simulation showed improved performance over similar regions.

Figure 5c shows the monthly variation of domain-average 5-year average O_3 mixing ratios between observations from AIRS-AQS and simulations from two-way WRF-CMAQ, and Figure 5d shows the diurnal variation of domain-average 5-year average hourly O_3 mixing ratios between observations from CASTNET and simulations from two-way WRF-CMAQ for representative winter (DJF and blue color) and summer (JJA and red color) seasons. As shown in Figure 5c, the O_3 mixing ratios are overpredicted throughout the year, which is consistent with overprediction of T2 (figure not shown). The largest overprediction occurs in the relatively cold months such as September to December. It is interesting that the observations show the largest monthly O_3 mixing ratios in spring and early summer while the simulation shows the peak during the summer. The difference in timing of peak O_3 between observations and simulations during the year might be associated with uncertainties in the BCONs of O_3 that reflect impacts of the long-range transport and associated stratosphere-troposphere exchange of O_3. As shown in Figure 5d, WRF-CMAQ tends to overpredict O_3 during most hours (i.e., 2:00-18:00) in summer and throughout the whole day in winter partially due to the overprediction of T2, especially in winter (figure not shown). The diurnal pattern of O_3 is captured much better during summer with much less prediction bias, especially during the nighttime, indicating that the model does a better job in predicting the evolution of nocturnal boundary layer and atmospheric chemistry in the warm season than the cold season. The overall overpredictions in this work are also consistent with previous studies (Eder and Yu, 2006; Appel et al., 2007; Wang et al., 2012), although our results show much better nighttime performance owing to the application of the ACM2 scheme that treats both local and non-local closure (Pleim, 2007). As also shown in Table 2, the domain-average NMBs and NMEs for max 8-h O_3 are 12.6% and 13.1% against AIRS-AQS and 1.5% and 8.4% against CASTNET, respectively. The statistics are also consistent with previous studies using the CMAQ model (Zhang et al., 2009a; Appel et al., 2013, 2017; Penrod et al., 2014) and can be considered as good performance according to the criteria suggested by Zhang et al. (2013) and Emery et al. (2017).

3.2.2 Aerosols

Figure 6a shows the spatial distribution of simulated 5-year average $PM_{2.5}$ from two-way WRF-CMAQ overlaid with observations from both the CSN and IMPROVE networks, and Figure S1 shows the spatial distribution of the major $PM_{2.5}$ constituents overlaid with observations from the CSN and IMPROVE network and PM_{10} overlaid with observations from the AQS network. As shown, WRF-CMAQ performs well for $PM_{2.5}$ over widespread areas of the Midwest and northeastern U.S., while $PM_{2.5}$ is underpredicted over the southeastern and western U.S. The model also misses some hot spots of observed concentrations in the western U.S., which are mainly caused by TC underpredictions (Figure S1) that are likely linked to poorly allocated and underestimated wildfire emissions in the NEI (Wiedinmyer et al., 2006; Roy et al., 2007; Kelly et al., 2019). The relatively large underpredictions over the eastern U.S. are mainly caused by the combined effects from SO_4^{2-}, NH_4^+, and TC. As shown in Figure S1, WRF-CMAQ largely underpredicts SO_4^{2-} in the Midwest and southeastern U.S. mainly due to the underprediction of oxidants such as O_3 (see Figure 5a) (which leads to less production from the gaseous oxidation), overprediction of precipitation (see Figure 1d) (which leads to more wet deposition and removal), and large underprediction of cloud fields (see Figure 3) (which leads to less aqueous phase formation), over the same area. On the other hand, NH_4^+ and NO_3^- are either underpredicted or overpredicted, respectively, over the similar areas mainly due to

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underprediction of SO_4^{2-}. According to the aerosol thermodynamics, when SO_4^{2-} is underpredicted, NH_4^+ tends to be underpredicted due to its major role as cation. More gaseous NH_3 will be available to neutralize NO_3^-, thus leading to overprediction of NO_3^- especially over the sulfate poor regions (West et al., 1999). Other potential reasons include the inaccurate assumptions in the thermodynamic module (for example, the internally mixed aerosol state and equilibrium assumption may not be representative over some regions and different time periods, S. Yu et al., 2006), uncertainties in emissions of key species such as NH_3 and non-volatile cations that affect particle acidity (Mebust et al., 2003; Wang and Zhang, 2014; Vasilakos et al., 2018; Pye et al., 2020), and measurement errors especially for NO_3^- and NH_4^+ (X.-Y. Yu et al., 2006; Karydis et al., 2007; Wang and Zhang, 2012). TC underpredictions over most sites of the domain can be attributed to the underprediction of emissions (e.g., wildfire and primary OC) and underestimation of secondary organic aerosol (SOA) formation (Appel et al., 2017; Pye et al., 2017) since EC (a chemically inert species) is overpredicted, which suggest that atmospheric mixing did not drive the TC underpredictions. Figures 6e-6h show the scatter plots of major $PM_{2.5}$ components such as SO_4^{2-}, NH_4^+, and NO_3^-, and TC. The WRF-CMAQ predicts $PM_{2.5}$ constituents well with the majority of data within the 1:2 ratio lines. Systematic underpredictions of SO_4^{2-} and NH_4^+ and overpredictions of NO_3^- are shown, which are consistent with their spatial distributions. Relatively large under- and overpredictions of TC compensate each other and lead to relatively low overall model biases. As also shown in Figure S1, the model fails to reproduce high concentrations of PM_{10} (those > 20 μg m^{-3}) over widespread areas of the domain, especially over dust source areas in CA, AZ, and NM. Hong et al. (2017) found the similar large underprediction of dust using CMAQ v5.0.2 over China and attributed it to a too-high threshold for friction velocity in the current dust module (Dong et al., 2016). Sea-salt also seems to be

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underpredicted by WRF-CMAQ, although sea-salt predictions are better than dust as shown along the coastlines.

Figures 6c and 6d show the monthly variation of 5-year average $PM_{2.5}$ between observations from CSN and IMPROVE, respectively, and simulations from two-way WRF-CMAQ. Both observations and WRF-CMAQ show higher monthly $PM_{2.5}$ concentrations at CSN sites than IMPROVE sites throughout the year because most CSN sites are in more polluted urban areas while IMPROVE sites are in rural areas and national parks. The model tends to underpredict $PM_{2.5}$ over both CSN and IMPROVE sites in the warm months (i.e., April to September) mainly due to the underpredictions of SO_4^{2-} and OC while it overpredicts $PM_{2.5}$ in cold months mainly due to NO_3^-. The model also captures the seasonality of $PM_{2.5}$ better over CSN sites than IMPROVE sites, especially in the summer months. The large underpredictions over IMPROVE sites during summer months are likely due to the underestimation of precursor emissions (such as wildfire emissions).

There are no universally accepted performance criteria for aerosols. As recommended by some previous studies (Zhang et al., 2006; Wang and Zhang, 2012), generally ±15% and ±30% for model biases and 30% and 50% for model errors can be considered as good and acceptable performance. As shown in Table 2, WRF-CMAQ in this work demonstrates an overall good or acceptable performance in predicting aerosols in terms of statistics especially for $PM_{2.5}$, NO_3^-, NH_4^+, and TC. It shows the domain-average NMBs of -7.0% and -13.7% for $PM_{2.5}$ against CSN and IMPROVE, respectively; NMBs of -26.7% and -27.2% for SO_4^{2-} against CSN and IMPROVE, respectively; NMBs of 16.6% and 14.6% for NO_3^- against CSN and IMPROVE, respectively; an NMB of -14.3% for NH_4^+ against CSN; NMBs of 20.6% and 29.4% for EC against CSN and IMPROVE, respectively; an NMB of -28.9% for OC against IMPROVE; and

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NMBs of -9.4% and -9.2% for TC against CSN and IMPROVE, respectively. The relatively large underpredictions of PM_{10}, i.e., an NMB of -45.9% against AQS, indicate further improvements of dust emissions are warranted. Overall, the aerosol performance is also comparable or better than previous CMAQ or WRF-CMAQ applications (Wang and Zhang, 2012; Penrod et al., 2014; Yu et al., 2014). For example, Penrod et al. (2014) showed 5-year (2001-2005) summer mean NMBs of -19.1% to -17.6% for $PM_{2.5}$ against CSN and IMPROVE data over the CONUS using the CMAQ v5.0 and Yu et al. (2014) reported the monthly mean NMBs of -6.2% and -16.8% for $PM_{2.5}$ against CSN and IMPROVE over the eastern U.S. using the same version of WRF-CMAQ as that used in this study.

3.2.3 Column abundance

Figure 7 shows the spatial distribution of 5-year average column abundances between various satellite products and two-way WRF-CMAQ for column CO, TOR, column NO_2, and column HCHO, and Table 2 summarizes the statistics. As shown, WRF-CMAQ can reproduce the spatial distribution of the column abundances of gases quite well with Rs ranging from 0.83 to 0.91. TOR, column NO_2 and column HCHO are also generally well predicted in terms of magnitude with NMBs of 1.6%, -14.5%, and 18.0%, respectively. Systematic underpredictions for column CO occur over the whole domain with an NMB of -26.6% for a few reasons. First, the BCONs of CO may be significantly underestimated from the CESM model. Using WRF/Chem or its variant, Zhang et al. (2016b, 2019) found that the column CO performance could be greatly improved by adjusting the BCON using the satellite observation. A similar approach could be applied in future WRF-CMAQ simulations as well. Second, as pointed by Heald et al. (2003), the regional emissions, especially biomass burning, could be a significant source for elevated CO concentrations and thus underestimation of these emissions could

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contribute to the CO underprediction. A more robust set of fire emissions from FINN generated by NCAR based on satellite retrievals has been applied to the similar time period recently but using the WRF-Chem model (Zhang and Wang, 2019) and were found to improve the CO performance. Last, Emmons et al. (2009) showed positive biases (i.e., 19%) of MOPITT retrievals over the land when compared to in-situ measurements and the biases may have been increasing over time due to the MOPITT bias drift (e.g., 0.5% yr^{-1} for version 7 retrieval). The predicted TOR can capture the observed high values over the eastern U.S. and oceans and the low values in elevated terrain; and it shows the best performance among all gas species. Both satellite observations and simulations can capture the elevated column NO_2 over the industrial and metropolitan areas in the domain where large nitrogen oxide (NO_x) emission sources are located. The model shows moderate underprediction which can be attributed to both uncertainties in the emissions and satellite retrievals. For example, the lightning emissions of NO_x are missing from this study, which have been found by previous studies (Allen et al., 2012) to contribute up to 2.0×10^{15} molecules cm^{-2} over the southern U.S., the Gulf of Mexico, and northern Atlantic Ocean during certain episodes. Boersma et al. (2004) also found that different column NO_2 retrieval approaches may lead to large errors (> 25%) over polluted areas. Column HCHO over the CONUS especially the southeastern U.S. is well predicted in terms of magnitude and spatial distribution and correlates well with the biogenic emission source regions. The underprediction of column HCHO may thus indicate potential underestimation of biogenic emissions from the BEIS. Other reasons including potential low yield of HCHO from isoprene and terpene in the CB05 mechanism and uncertainties in satellite retrievals (Stavrakou et al., 2009; Lorente et al., 2017)

3.2.4 Simulated O_3 and $PM_{2.5}$ exceedances of NAAQS levels

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National Ambient Air Quality Standards (NAAQS) are set for criteria pollutants, including O_3 and $PM_{2.5}$, to provide protection against adverse health and welfare effects (www.epa.gov/criteria-air-pollutants/naaqs-table). In this section, the average number of days per year where the 24-hr $PM_{2.5}$ NAAQS level (35 μg m^{-3}) and the max 8-h O_3 NAAQS level (70 ppb) are exceeded from the WRF-CMAQ predictions is compared with the number of exceedances in the monitoring data (i.e., O_3 from AQS and CASTNET and $PM_{2.5}$ from IMPROVE and CSN). This comparison is intended to better characterize the ability of the model to simulate the high-concentration days that could be especially relevant in regulatory assessments. In Figure 8, the five-year average of the annual number of exceedance days is shown for WRF-CMAQ and the monitoring data at monitor locations. The sizes of circles and shades of color represent the magnitude of exceedances (i.e., larger circles and darker shades indicate a greater number of exceedance days). As shown, the observations indicate a large number of annual exceedance days for max 8-h O_3 over major cities, especially in CA, TX, the Midwest, and northeastern U.S. The spatial distribution of the observed number of exceedance days from the AQS and CASTNET networks aligns well with the nonattainment map reported by the Green Book of U.S. EPA (https://www.epa.gov/green-book). The WRF-CMAQ model also generally captures the distribution of the number of exceedance days very well, especially in CA. The domain-average values of NMB, NME, and R are -3.4%, 14.0%, and 0.98, respectively, also indicating a good performance. For $PM_{2.5}$, the largest number of exceedance days based on the IMPROVE and CSN observations mainly occurs in the northwestern U.S., Midwest, and major cities in the northeastern U.S. The number of exceedance days is generally much lower for $PM_{2.5}$ than O_3. The spatial distribution of the number of exceedance days for observed $PM_{2.5}$ aligns well with nonattainment areas reported by the Green Book from U.S. EPA in CA. However, the

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number of simulated $PM_{2.5}$ exceedance days underpredicts the observation-based values in the western U.S. mainly due to large underpredictions of $PM_{2.5}$ concentrations in the same areas as shown in Figure 6a. The simulation better predicts the distribution of the number of exceedance days in the eastern U.S. where terrain is relatively flat and wildfire less prevalent. The domain-average values of NMB, NME, and R are -29.0%, 80.8%, and 0.21, respectively.

4. Impacts of chemistry-meteorology feedbacks

In this section, the impacts of chemistry-meteorology feedbacks including aerosol direct and indirect effects on regional meteorology and air quality over the U.S. are further examined by comparing results from two-way WRF-CMAQ and offline coupled WRF and CMAQ. Model performance from the two sets of simulations is first compared to demonstrate the potential performance improvements of the two-way model, and the impacts on regional meteorology and air quality are further investigated via the spatial difference plots for selected variables and species.

4.1 Meteorology

Figures 1 and 4 compare observations and simulations from the two-way WRF-CMAQ and WRF-only models for precipitation and SWCF/LWCF, respectively. Table 1 also summarizes the model performance statistics for all major meteorological variables for the two simulations. The statistics of some cloud variables from the WRF-only simulation are not available due to missing model outputs. Overall, good performance is evident for both simulations for surface meteorological variables with slightly better performance (except for RH2) for the two-way WRF-CMAQ simulation than the WRF-only simulation. The MBs for the two-way WRF-CMAQ vs. WRF-only simulation are 0.1°C vs 0.2 °C for T2, 2.2% vs 1.8% for

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RH2, 0.44 m s^{-1} vs 0.46 m s^{-1} for WS10, 32.8 degree vs 33.4 degree for WD10, and 0.14-0.71 mm day^{-1} vs 0.2-0.8 mm day^{-1} for precipitation. The spatial distributions for SWCF and LWCF are slightly better captured especially over the Midwest, Atlantic Ocean, and Pacific Northwest regions. Compared to WRF-only, two-way WRF-CMAQ shows noticeably better performance in terms of both MB and RMSE for radiation and cloud forcing, with MBs of 37.0 vs. 24.2 W m^{-2} for SWDOWN, 28.5 vs 17.6 W m^{-2} for GSW, -10.6 vs. -6.1 W m^{-2} for GLW, 2.8 vs. 2.0 W m^{-2} for OLR, -17.6 vs. -10.7 W m^{-2} for SWCF, and -5.9 vs. -5.3 W m^{-2} for LWCF. These results are consistent with those reported by Yahay et al. (2015a,b) that showed similar improvements in meteorological and radiative variables when comparing predictions from WRF-Chem with those from WRF only. Since identical inputs and physics options are used in both simulations, the differences in performance for meteorological variables is due to the consideration of feedback processes among chemistry, aerosol, cloud, and radiation in the two-way coupled WRF-CMAQ simulation.

Figure 9 shows the difference plots of selected major meteorological variables including SWDOWN, T2, RH2, WS10, PBL height, and precipitation between two-way WRF-CMAQ and WRF-only. As shown, the incoming shortwave radiation is reduced by up to 24.8 W m^{-2} (13.6%) with a domain-average of 13.0 W m^{-2} (6%) due to the combined aerosol direct and indirect radiative effects over the domain. The reduction is predominant over the eastern U.S. where both aerosol loading and cloud cover are high and over the oceans where cloud cover is high. The magnitude of shortwave radiation reduction in this work is consistent with other studies. For example, Wang et al. (2015) found that the combined aerosol direct and indirect effects using the WRF/Chem model, which includes the sub-scale cloud forcing not treated in the current WRF-CMAQ model, may decrease the incoming shortwave radiation by 16.0 W m^{-2} in the summer over the U.S. Hogrefe et al. (2015) reported the reduction of shortwave radiation may reach up to 20 W m^{-2} over the eastern U.S. by only considering the aerosol direct effect using an older version of WRF-CMAQ v5.0.1. Xing et al. (2015b) showed that the aerosol direct forcing may cause the surface shortwave radiation to decrease by up to 10 W m^{-2} over the eastern U.S. over a decadal time period using WRF-CMAQ v5.0. The reduction of shortwave radiation further reduces the surface temperature by up to 0.25 °C over the eastern U.S., which is much larger than the reduction of 0.1 °C reported by Hogrefe et al. (2015), mainly due to the inclusion of aerosol indirect effects. However there are smaller reductions of T2 over the Pacific Ocean and even increases (by up to 0.1 °C) over large areas of Atlantic Ocean and Gulf of Mexico where much larger reductions of shortwave radiation occur. As pointed by Wang et al. (2015), due to the much larger heat capacity of ocean, the response of sea surface temperature is less sensitive to the change of shortwave radiation for ocean compared to the land. The large increase of incoming longwave radiation and latent heat (figures not shown) caused by the aerosol indirect effects and other complex feedback processes over the ocean compensates for the reduction of shortwave radiation, especially over the Atlantic Ocean and Gulf of Mexico, and thus leads to less reduction or even increases of T2. RH2 is found to mostly increase by 3.4% over the land caused by the decrease of temperature while decrease by 2.6% over the ocean caused by either the increase of temperature or large decrease of water vapor. Over the land, the decreases in solar radiation and T2 along with the latent heat (figure not shown) lead to a more stable PBL and thus suppress the wind (by reducing the wind speed as shown). Over the ocean, the changes lead to a more unstable PBL and thus enhance the wind over the ocean. The wind speed and PBL height are reduced by up to 0.05 m s^{-1} and 25 m, respectively, over the U.S. The aerosol feedbacks on precipitation are also mixed with relatively large decreases by up to 0.4 mm day^{-1} over the U.S.

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and increases by up to 0.4 mm day^{-1} over oceans. The suppression of precipitation over the land is mainly due to the formation of more small sized CCNs caused by aerosol indirect effects and align well with areas with high aerosol loadings while the enhancement of precipitation, especially along coastlines and over oceans, might be associated with the larger CCN formation via more activated sea-salt particles as indicated by Zhang et al. (2010) and Wang et al. (2015).

4.2 Air Quality

Figures 5 and 6 compare observations and simulations from two-way WRF-CMAQ and offline CMAQ for O_3, $PM_{2.5}$, and $PM_{2.5}$ constituents. Table 2 summarizes the statistics for all major chemical variables for the two simulations. As shown in Figure 5, two-way WRF-CMAQ shows better performance for both the monthly variation of O_3 (throughout the whole year) over AQS sites and the diurnal pattern of O_3 (especially during winter) over CASTNET sites due to better performance of T2 and radiation compared to offline WRF and CMAQ. As shown in Figure 6, two-way WRF-CMAQ shows similar spatial distribution of $PM_{2.5}$ and better performance for $PM_{2.5}$ for most of months over CSN sites and for cold seasons across IMPROVE sites compared to offline CMAQ. It also shows systematically better performance for SO_4^{2-}, NO_3^-, NH_4^+, and TC with more data within 1:2 and closer to 1:1 ratio lines of scatter plots. Overall, as shown in Table 2, both simulations show generally good performance for all major chemical species except for PM_{10}. For example., the domain-average NMBs are 12.6% (AQS) and 1.5% (CASTNET) vs. 17.7% (AQS) and 7.7% (CASTNET) for O_3 and -7.0% (CSN) and -13.7% (IMPROVE) vs. -3.4% (CSN) and -5.7% (IMPROVE) for $PM_{2.5}$ for two-way WRF-CMAQ and offline-coupled CMAQ, respectively. The two-way WRF-CMAQ shows better domain-wide statistics in terms of both correlation and biases for many variables including O_3, SO_4^{2-}, NO_3^-, NH_4^+, and EC as well as TOR and column NO_2, apparently due to the treatment of

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chemistry-meteorology feedbacks. Offline CMAQ performs better for total $PM_{2.5}$ especially in the western U.S. due to higher dust emissions from higher wind speed and higher SOA due to stronger radiation and higher temperature. However more robust comparisons are needed in the future with improved dust emissions and the use of FINN wildfire emissions.

Figure 10 shows the difference plots of selected chemical variables including CO, O_3, NO_x, volatile organic compounds (VOCs), SO_4^{2-}, SOA, $PM_{2.5}$, and PM_{10} between two-way WRF-CMAQ and offline-coupled CMAQ. As shown, the CO mixing ratios decrease by up to 79.2 ppb (27.8%) especially over the western U.S. with a domain-average reduction of 3.0 ppb (3.1%) due to reduced formation of CO from the oxidation of VOCs caused by reduced solar radiation as indicated by Zhang et al. (2017). Such reductions seem to dominate over the increases caused by reduced PBL height, especially in the western U.S. where PBL height reductions are minimum. The O_3 mixing ratios decrease by up to 5.2 ppb (16.2%) with domain-average of 1.7 ppb (4.2%) mainly due to the reduced solar radiation and T2. The change of O_3 is consistent with other studies such as Makar et al. (2015) and Wang et al. (2015) that also reported lower O_3 mixing ratios caused by aerosol direct and indirect effects. On the other hand, both NO_x and VOC mixing ratios increase over the eastern U.S. while they decrease over the western U.S. The increase should be caused by the combination of the large reduction of PBL mixing and reduced solar radiation which reduces NO_2 photolysis and VOC oxidation to SOA. For aerosol species, SO_4^{2-} concentrations increase by up to 0.38 μg m^{-3} (26.6%) especially over the eastern U.S. In fact, the reduction of O_3 mixing ratios due to aerosol effects is expected to reduce SO_4^{2-} production via the gas-phase oxidation pathway due to the influence of O_3 on OH, but increase SO_4^{2-} production via the aqueous-phase chemistry pathway due to more clouds in the two-way WRF-CMAQ simulation. Thus, the net increase of SO_4^{2-} is more dominate by the

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aqueous-phase chemistry instead of the gas-phase oxidation. This net increase of SO_4^{2-}, in turn, leads to an increase of NH_4^+ and decrease of NO_3^- (figures not shown) through aerosol thermodynamic equilibrium. SOA concentrations decrease by up to 0.34 µg m^{-3} (41.6%) especially over the eastern U.S. due to the large reduction of oxidants. $PM_{2.5}$ concentrations also decrease by up to 5.2 µg m^{-3} (49.1%) with a domain-average of 0.34 µg m^{-3} (8.6%), and PM_{10} concentrations decrease by up to 19.3 µg m^{-3} (64.8%) with a domain-average of 1.1 µg m^{-3} (11.1%). The reductions are more apparent over the western U.S. than the eastern U.S. partially due to the compensation of the increase of SO_4^{2-} and NH_4^+ and decrease of other secondary aerosols over the eastern U.S., as well as the relatively large reduction of dust concentrations over the western U.S. caused by reduced wind speed.

5. Summary and conclusion

In this study, two sets of long-term simulations for 2008-2012 using the two-way coupled WRF-CMAQ and offline coupled WRF and CMAQ, respectively, are conducted, evaluated, and compared to investigate the performance improvements due to chemistry-meteorology feedbacks and impacts of those feedbacks on the reginal air quality in the U.S. First, the two-way coupled WRF-CMAQ simulation with both aerosol direct and indirect radiative forcing is comprehensively evaluated. The results show that WRF-CMAQ performs well for major surface meteorological variables such as temperature at 2 m, relative humidity at 2 m, wind speed at 10 m, and precipitation with domain-average MBs of 0.1 °C, 2.2 %, 0.44 m s^{-1}, and 0.14-0.28 mm day^{-1} (except for 0.71 mm day^{-1} against NCDC), respectively. The overall small warm bias compared to other studies is most likely associated with the soil moisture nudging technique used in the PX land surface scheme. The relatively large positive biases for precipitation are found to be more apparent when observed precipitation is low (dominated more by the non-convective

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precipitation) and are thus believed to be more associated with uncertainties in the Morrison microphysics scheme. The long-term simulation also shows generally good performance for major radiation and cloud radiative variables. Relatively large model biases still exist for cloud variables such as CDNC, COT, and CWP, indicating that the processes associated with aerosol indirect effects are still not well understood and an accurate simulation of those effects is still challenging using state-of-the-science models.

Two-way WRF-CMAQ also shows generally good or acceptable performance for max 8-h O_3, $PM_{2.5}$ and $PM_{2.5}$ constituents, with NMBs generally within ±15% for O_3 and ±30% for $PM_{2.5}$ species. For example, the domain-average NMBs are 12.6 % and 1.5 % for max 8-h O_3 against AQS and CASTNET and -7.0 % and -13.7 % for $PM_{2.5}$ against CSN and IMPROVE, respectively. O_3 mixing ratios are overpredicted for most months, especially in the winter, in part due to the larger overprediction of T2 during the cold season. The overall model biases are small for $PM_{2.5}$ due to the compensation of relatively large underpredictions of SO_4^{2-} and OC, especially in the warm season, and overprediction of NO_3^- in the cold season. In addition to biases inherited from the meteorology, the model performance for chemistry also suffers from uncertainties associated with emissions, the use of a coarse spatial resolution, and representation of aerosol formation pathways in the model. For example, the relatively large biases for EC might be associated with poorly allocated anthropogenic/wildfire emissions and those for OC might be due to underestimation of SOA formation in version 5.0.2 of CMAQ. WRF-CMAQ also predicts the column abundances of chemical species well and the relatively large model biases for CO are found to be associated with an underestimation of BCONs. The model better reproduces the observed number of exceedance days for O_3 than $PM_{2.5}$ mainly due to better performance for O_3 than $PM_{2.5}$ concentrations.

The performance comparison between two-way WRF-CMAQ and WRF-only simulations shows that two-way WRF-CMAQ model performs better for major surface meteorological, radiation, and cloud radiative variables due to the consideration of chemistry-meteorology feedbacks associated with aerosol direct and indirect forcing. The feedbacks are found to reduce the 5-year average SWDOWN by up to 24.8 W m^{-2}, T2 by up to 0.25 °C, PBL height by up to 25 m, wind speed by up to 0.05 m s^{-1}, and precipitation by up to 0.4 mm day^{-1} over the CONUS, which in turn affect the air quality significantly. As a result of feedbacks, two-way WRF-CMAQ outperforms offline CMAQ for O_3, SO_4^{2-}, NO_3^-, NH_4^+, and EC as well as TOR and column NO_2 in terms of both spatiotemporal variations and domain-average statistics due to better meteorology performance for variables such as T2, WS10, radiation, and precipitation. Despite these improvements, the offline CMAQ performs better for total $PM_{2.5}$ in terms of domain-average statistics, which could be partially caused by the compensation of larger under- and over-predictions of $PM_{2.5}$ constituents. More robust comparison for $PM_{2.5}$ should be performed with improved dust and wildfire emissions in future work. Chemistry-meteorology feedbacks are found to play important roles in affecting U.S. air quality by reducing domain-wide 5-year average surface CO by 3.0 ppb (3.1%) and up to 79.2 ppb (27.8%), O_3 by 1.7 ppb (4.1%) and up to 5.2 ppb (16.2%), $PM_{2.5}$ by 0.34 μg m^{-3} (8.6%) and up to 5.2 μg m^{-3} (49.1%), and PM_{10} by 1.1 μg m^{-3} (11.1%) and up to 19.3 μg m^{-3} (64.8%) mainly due to reduction of radiation, temperature, and wind speed.

In summary, the two-way coupled WRF-CMAQ modeling in this study shows generally satisfactory and consistent performance for the long-term prediction of regional meteorology and air quality when compared to other studies in the literature. Possible causes for the meteorological and chemical biases that were identified through this work can provide valuable

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information for future model development to improve the two-way coupled WRF-CMAQ model and those biases should also be considered when making future climate/air quality projections. Non-negligible model improvements for many major meteorological and chemical variables compared to the traditional application of offline coupled WRF and CMAQ suggest the importance of chemistry-meteorology feedbacks, especially aerosol direct and indirect effects. The feedbacks should be considered along with other factors in developing future model applications to inform policy making.

Code Availability

The modeling system used in this study is based on the 2-way coupled WRF-CMAQ model derived from WRF v3.4 and CMAQ v5.0.2. Relevant code for CMAQ v5.0.2, its coupling to WRF and aerosol direct feedbacks are publicly available from: doi:10.5281/zenodo.1079898. WRF v3.4 code can be downloaded from http://www2.mmm.ucar.edu/wrf/users/download/get_source.html. The version of the coupled WRF-CMAQ model with the additional indirect aerosol forcing approach of Yu et al. (2014) can be downloaded from the following website: https://person.zju.edu.cn/shaocaiyu#674502.

Author contribution

YZ and KW designed the study and all the simulations. SY developed the two-way coupled WRF-CMAQ code. KW conducted all the simulations and performed the analyses. KW prepared the manuscript with contributions from all co-authors.

Competing interests

The authors declare that they have no conflict of interest.

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Table 1. The 5-year (2008-2012) average performance statistics for meteorological variables between two-way WRF-CMAQ and WRF-only simulations.

Variables	Datasets	Mean Obs	Two-way WRF-CMAQ					WRF-only				
			Mean Sim	R	MB	NMB (%)	RMSE	Mean Sim	R	MB	NMB (%)	RMSE
T2 (°C)	NCDC	12.9	13.0	0.98	0.1	0.8	1.0	13.1	0.98	0.2	1.8	1.1
RH2 (%)	NCDC	69.1	71.3	0.88	2.2	3.2	5.3	71.0	0.88	1.8	2.6	5.2
WS10 (m s^{-1})	NCDC	3.74	4.18	0.52	0.44	11.7	1.15	4.20	0.52	0.46	12.4	1.16
WD10 (deg)	NCDC	154.4	187.2	0.07	32.8	21.3	47.7	187.8	0.06	33.4	21.6	48.1
Precipitation (mm day^{-1})	NCDC	1.84	2.55	0.62	0.71	38.4	1.27	2.64	0.62	0.8	43.5	1.33
	NADP	2.66	2.81	0.84	0.15	5.8	0.7	2.9	0.84	0.24	9.3	0.73
	GPCP	2.15	2.43	0.79	0.28	13.0	0.9	2.45	0.80	0.30	14.1	0.9
	PRISM	2.16	2.30	0.91	0.14	6.8	0.55	2.36	0.91	0.20	9.5	0.56
	TMPA	2.28	2.50	0.86	0.22	9.9	0.81	2.52	0.86	0.24	10.7	0.82
SWDOWN (W m^{-2})	CERES	185.6	209.8	0.97	24.2	13.0	25.7	222.6	0.96	37.0	19.9	38.3
GSW (W m^{-2})	CERES	158.5	176.0	0.97	17.6	11.1	19.8	187.0	0.95	28.5	18.0	30.6
GLW (W m^{-2})	CERES	322.9	316.8	0.99	-6.1	-1.9	8.1	312.3	0.99	-10.6	-3.3	12.1
OLR (W m^{-2})	CERES	241.2	243.2	0.99	2.0	0.8	3.5	244.0	0.99	2.8	1.2	4.2
SWCF (W m^{-2})	CERES	-41.1	-30.4	0.74	-10.7	-26.0	13.7	-23.5	0.63	-17.6	-42.8	20.1
LWCF (W m^{-2})	CERES	23.7	18.4	0.73	-5.3	-22.2	6.5	17.8	0.74	-5.9	-24.9	6.9
AOD	MODIS	0.15	0.05	0.60	-0.1	-64.8	0.11	N/A	N/A	N/A	N/A	N/A
CF	MODIS	0.57	0.50	0.92	-0.07	-12.2	0.09	N/A	N/A	N/A	N/A	N/A
CDNC (cm^{-3})	MODIS	163.3	29.3	0.35	-134.0	-82.1	138.8	N/A	N/A	N/A	N/A	N/A
CWP (g m^{-2})	MODIS	167.4	81.6	0.79	-85.8	-51.2	90.4	N/A	N/A	N/A	N/A	N/A
COT	MODIS	15.3	3.0	0.84	-12.3	-80.1	12.6	N/A	N/A	N/A	N/A	N/A

*outputs of AOD, CF, CDNC, CWP, and COT are not available from WRF-only simulations

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Table 2. The 5-year (2008-2012) average performance statistics for chemical variables between two-way WRF-CMAQ and offline CMAQ simulations.

Variables	Datasets	Mean Obs	Two-way WRF-CMAQ					Offline CMAQ				
			Mean Sim	R	MB	NMB (%)	NME (%)	Mean Sim	R	MB	NMB (%)	NME (%)
Max 8-hr O_3 (ppb)	**AQS**	43.5	49.0	0.66	5.5	12.6	13.1	51.2	0.66	7.7	17.7	17.9
	CASTNET	42.2	42.8	0.65	0.6	1.5	8.4	45.1	0.65	3.0	7.0	10.5
$PM_{2.5}$ (μg m^{-3})	**CSN**	10.7	9.9	0.50	-0.75	-7.0	21.9	10.3	0.46	-0.36	-3.4	21.7
	IMPROVE	4.78	4.13	0.88	-0.65	-13.7	26.6	4.51	0.87	-0.27	-5.7	23.2
PM_{10} (μg m^{-3})	**AQS**	24.0	13.0	0.02	-11.0	-45.9	49.6	15.4	0.14	-8.6	-35.6	45.0
SO_4^{2-} (μg m^{-3})	**CSN**	2.32	1.70	0.88	-0.62	-26.7	27.1	1.57	0.89	-0.75	-32.3	32.3
	IMPROVE	1.08	0.78	0.98	-0.29	-27.2	27.2	0.76	0.98	-0.32	-29.4	29.4
NO_3^- (μg m^{-3})	**CSN**	1.29	1.51	0.85	0.22	16.6	32.8	1.73	0.85	0.43	33.5	44.9
	IMPROVE	0.41	0.47	0.85	0.06	14.6	42.9	0.57	0.87	0.16	39.0	51.7
NH_4^+ (μg m^{-3})	**CSN**	1.03	0.88	0.86	-0.15	-14.3	18.6	0.87	0.85	-0.16	-15.7	18.7
EC (μg m^{-3})	**CSN**	0.63	0.76	0.34	0.13	20.6	52.4	0.77	0.39	0.14	22.4	50.5
	IMPROVE	0.18	0.23	0.80	0.05	29.4	50.8	0.25	0.79	0.07	37.7	55.6
OC (μg m^{-3})	**IMPROVE**	0.97	0.69	0.59	-0.28	-28.9	44.8	0.74	0.58	-0.23	-23.8	43.4
TC (μg m^{-3})	**CSN**	2.87	2.60	0.10	-0.27	-9.4	29.7	2.71	0.07	-0.16	-5.7	28.8
	IMPROVE	0.68	0.62	0.79	-0.06	-9.2	37.2	0.80	0.72	-0.08	-9.2	39.0
Col. CO (10^{18} mole. cm^{-3})	**MOPITT**	1.96	1.44	0.89	-0.52	-26.6	26.7	1.45	0.89	-0.51	-26.2	26.2
TOR (DU)	**OMI**	30.3	30.8	0.83	0.47	1.6	4.7	31.1	0.82	0.77	2.5	5.1
Col. NO_2 (10^{15} mole. cm^{-3})	**SCIAMACHY**	1.27	1.09	0.91	-0.18	-14.5	27.1	1.08	0.91	-0.19	-14.9	27.3
Col. HCHO (10^{15} mole. cm^{-3})	**SCIAMACHY**	5.13	4.21	0.83	-0.92	-18.0	20.6	4.28	0.83	-0.85	-16.6	19.8

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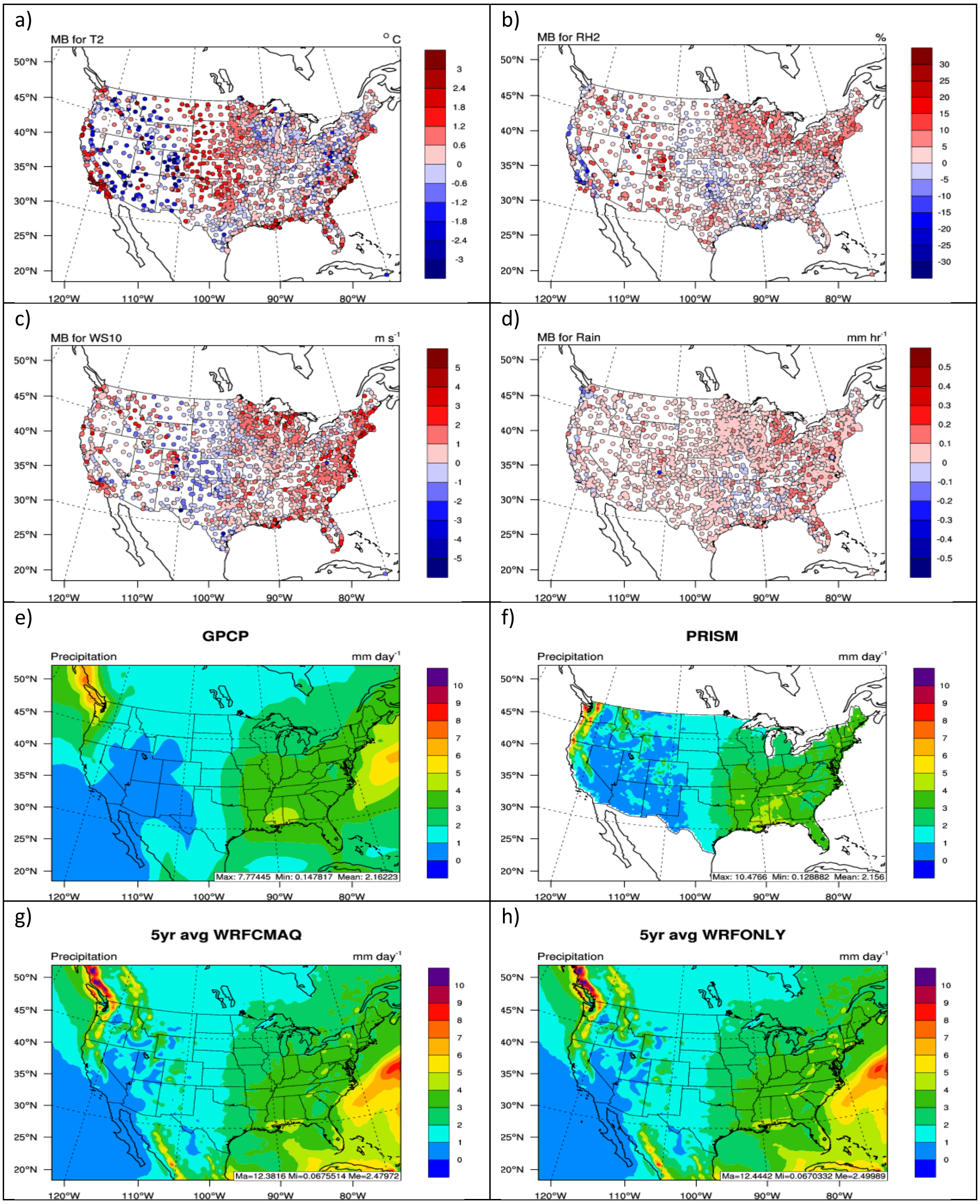

Figure 1. Spatial distributions of 5-year average MBs for a) 2-m temperature (T2), b) 2-m relative humidity (RH2), c) 10-m wind speed (WS10), and d) hourly precipitation from NCDC for two-way WRF-CMAQ in 2008-2012 and 5-year average of daily precipitation for e) GPCP, f) PRISM, g) two-way WRF-CMAQ, and h) WRF-only.

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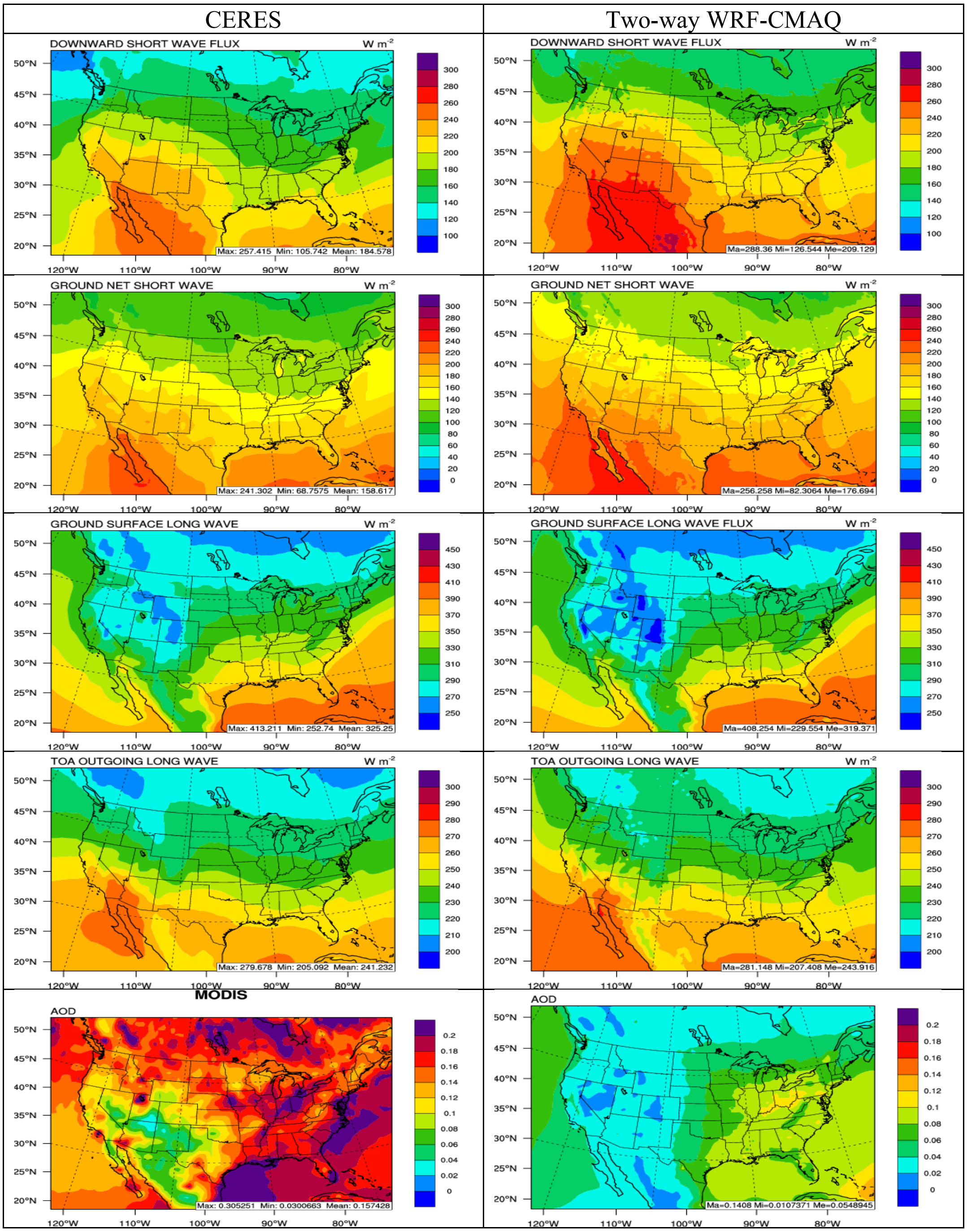

Figure 2. Spatial distribution of 5-year average major radiation variables (from top to bottom: SWDOWN, GSW, GLW, OLR, and AOD) between CERES observations (left panel) vs. two-way WRF-CMAQ (right panel) for 2008-2012.

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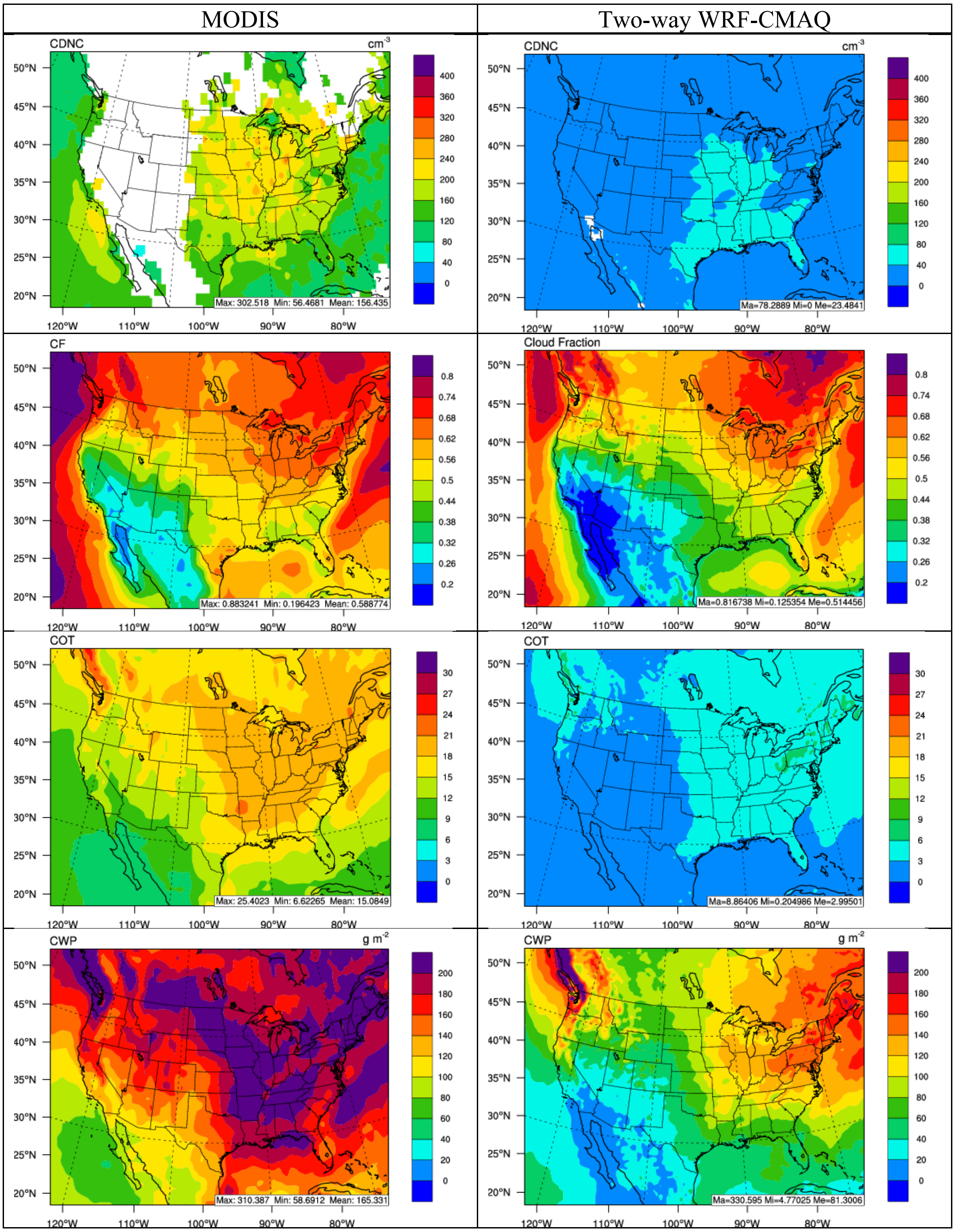

Figure 3. Spatial distribution of 5-year average major cloud variables (from top to bottom: CDNC, CF, COT, and CWP) between MODIS observations (left panel) vs. two-way WRF-CMAQ (right panel) for 2008-2012.

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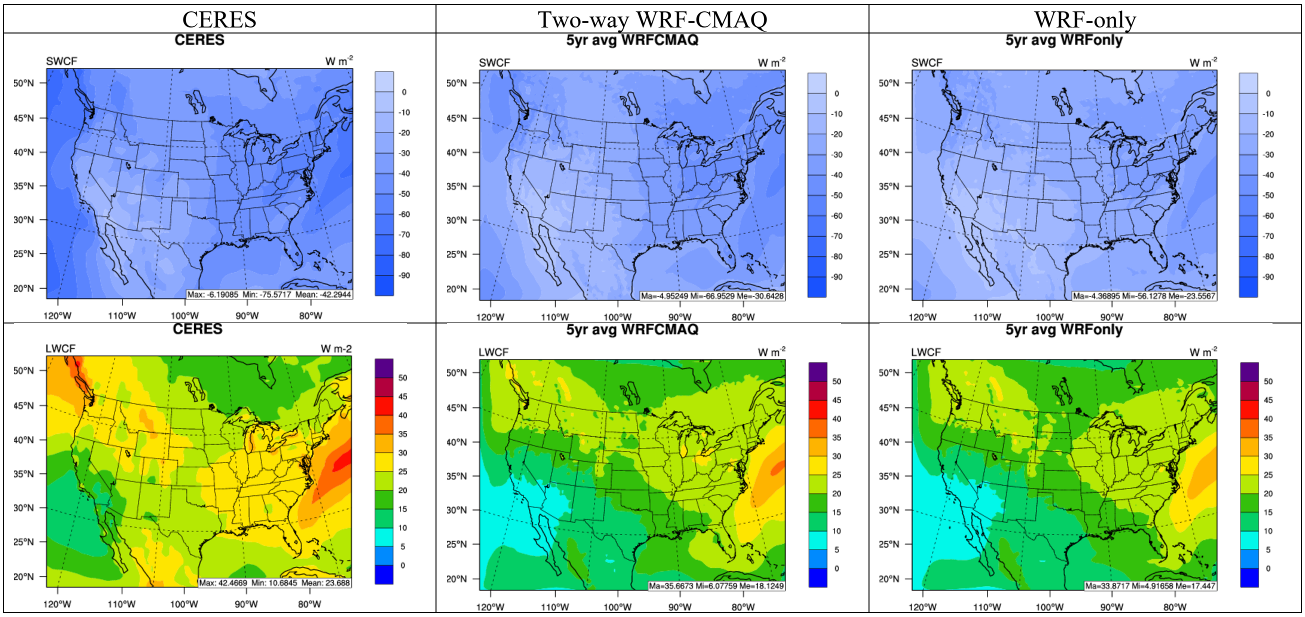

Figure 4. Spatial distribution of 5-year average SWCF (top panel) and LWCF (bottom panel) between SERES observations (left panel) vs. two-way WRF-CMAQ (center panel) and WRF-only (right panel) for 2008-2012.

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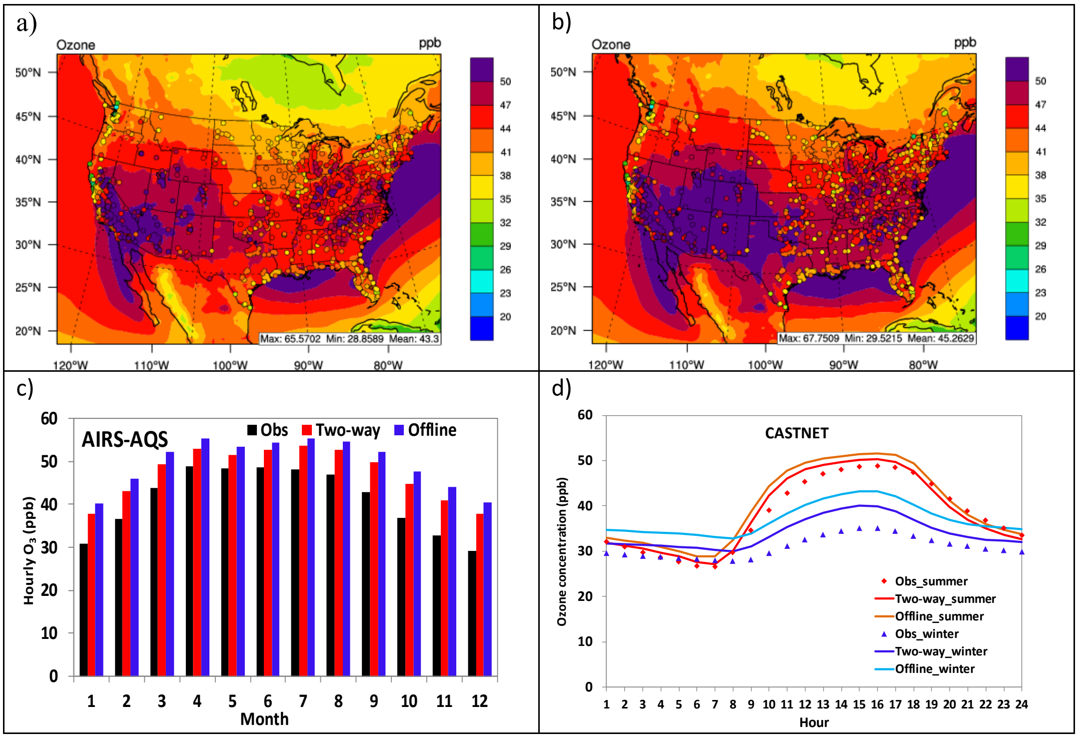

Figure 5. Spatial distributions of 5-year averaged max 8-h O_3 overlaid with observations from AIRS-AQS and CASTNET for a) two-way WRF-CMAQ and b) offline CMAQ; c) bar chart for 5-year average monthly O_3 between observations (black bar), two-way WRF-CMAQ (red bar), and offline CMAQ (blue bar); and d) diurnal plots of observed (dots) vs. simulated (lines) hourly O_3 concentrations against CASTNET for winter (cold colors) and summer (warm colors) in 2008-2012.

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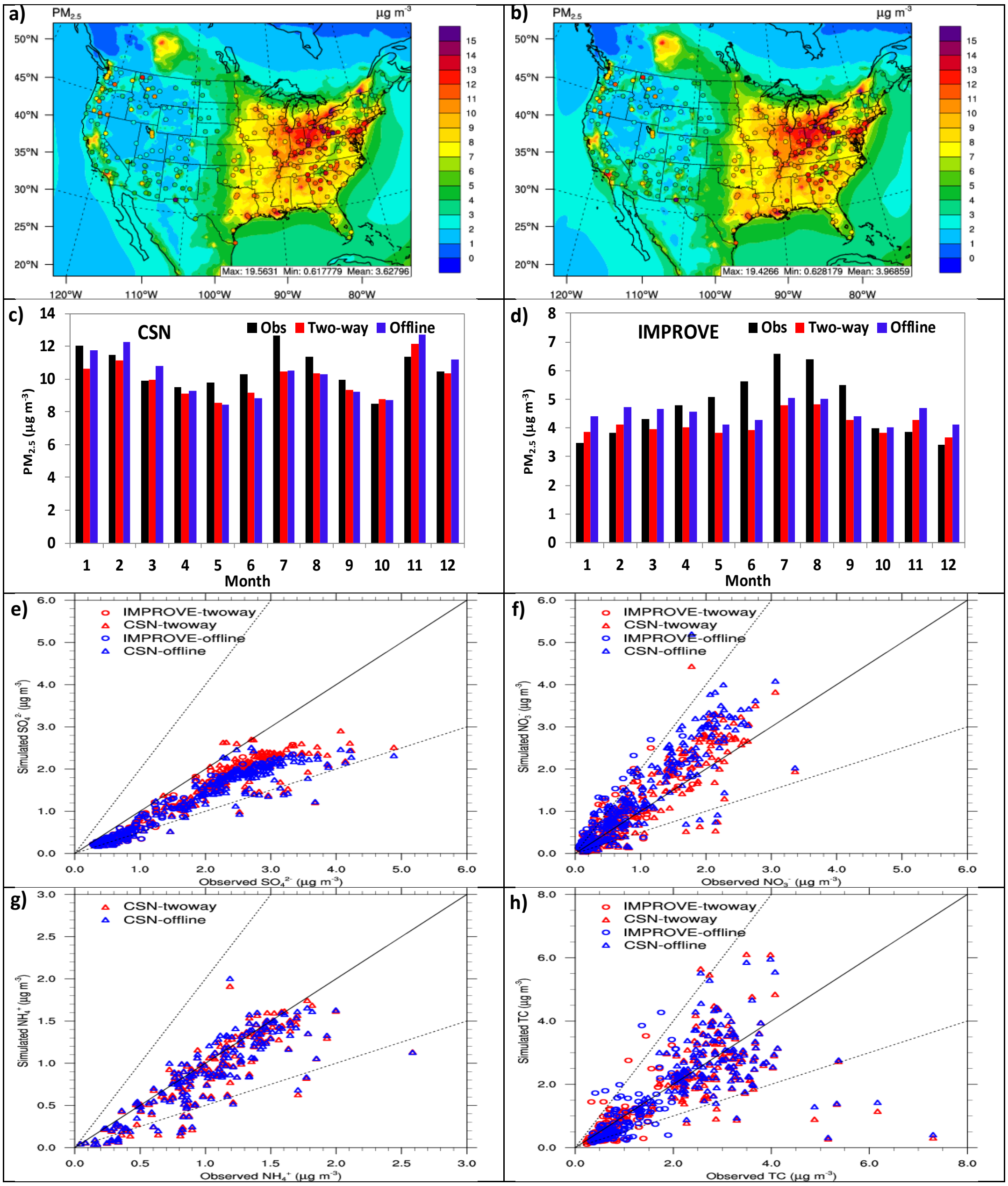

Figure 6. Spatial distributions of 5-year averaged daily $PM_{2.5}$ overlaid with observations from CSN and IMPROVE for a) two-way WRF-CMAQ and b) offline CMAQ; bar charts for 5-year average monthly $PM_{2.5}$ between observations (black bar), two-way WRF-CMAQ (red bar), and offline CMAQ (blue bar) over c) CSN and d) IMPROVE; and scatter plots of $PM_{2.5}$ constituents e) SO_4^{2-}, f) NH_4^+, g) NO_3^-, and h) TC) between observations and simulations of two-way WRF-CMAQ (red color) and offline CMAQ (blue) for 2008-2012.

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Preprint. Discussion started: 3 September 2020

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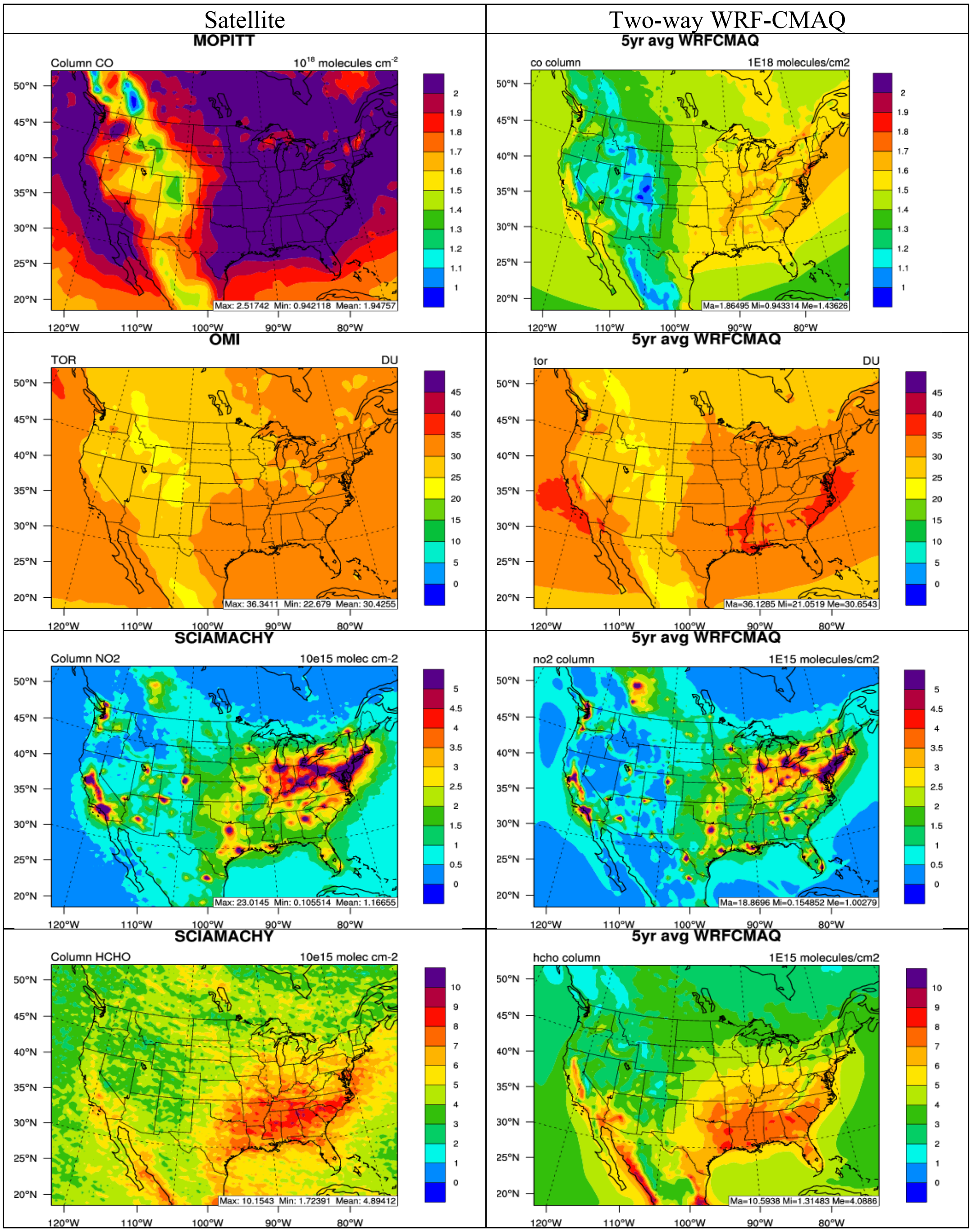

Figure 7. Spatial distribution of 5-year average column abundances (from top to bottom: column CO, TOR, column NO_2, and column HCHO) between various satellite observations (left panel) vs. two-way WRF-CMAQ (right panel) for 2008-2012.

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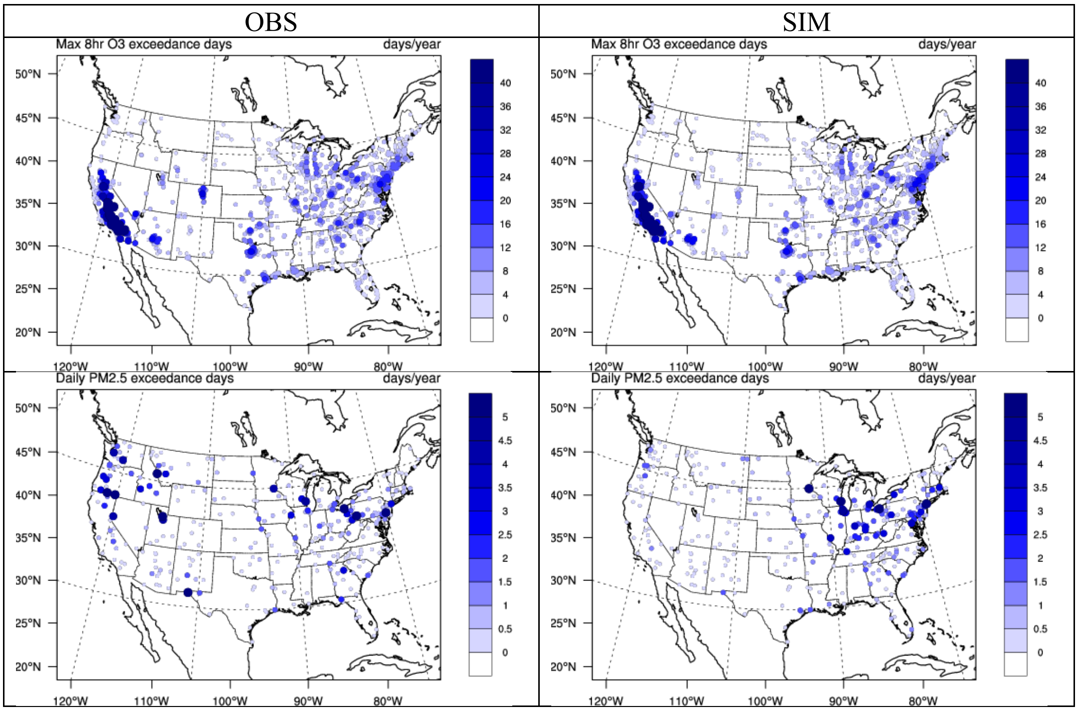

Figure 8. The spatial distribution of 5-year average annual exceedance days of max 8-h O_3 and daily $PM_{2.5}$ between observations (O_3 over the AIRS-AQS/CASTNET network and $PM_{2.5}$ over the IMPROVE/CSN network) and two-way WRF-CMAQ.

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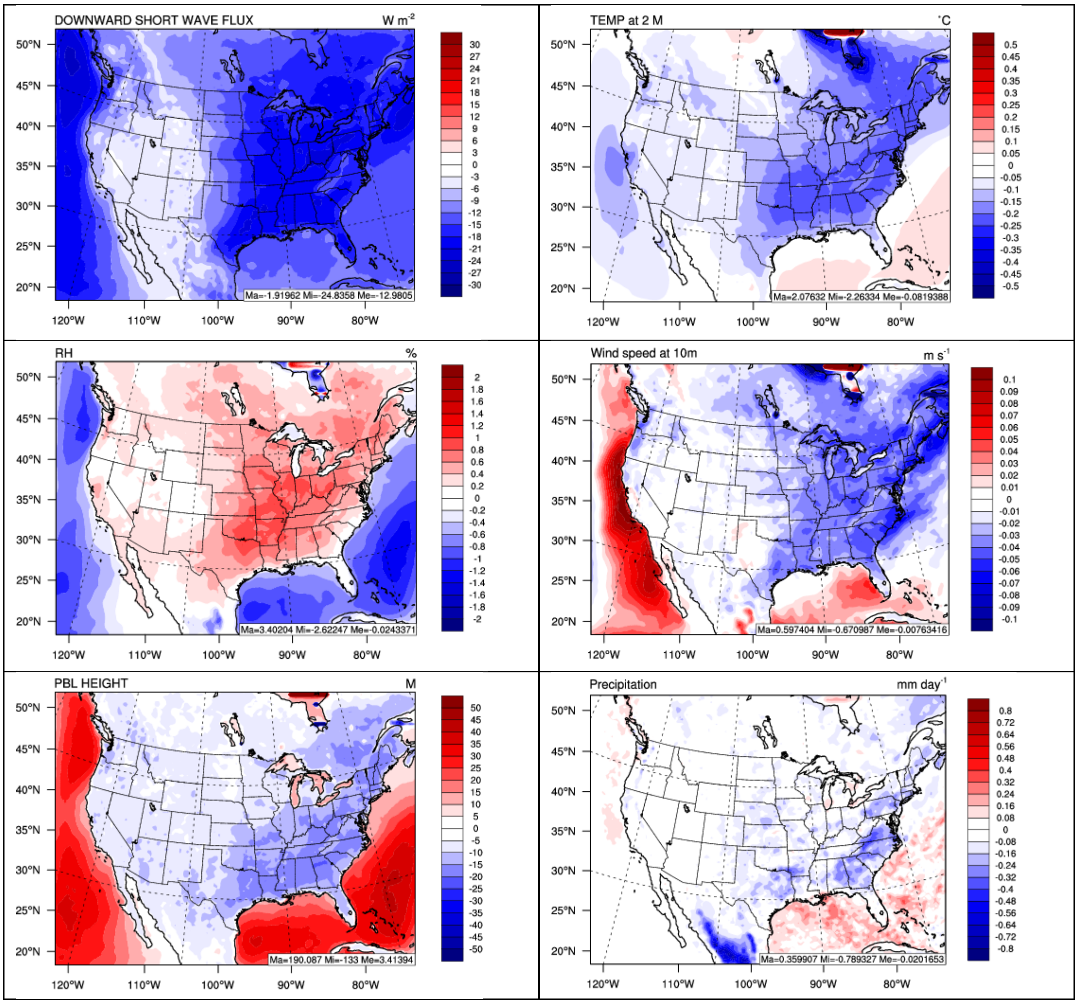

Figure 9. Spatial difference plots (two-way WRF-CMAQ - WRF-only) for major meteorological variables between two-way WRF-CMAQ and WRF-only.

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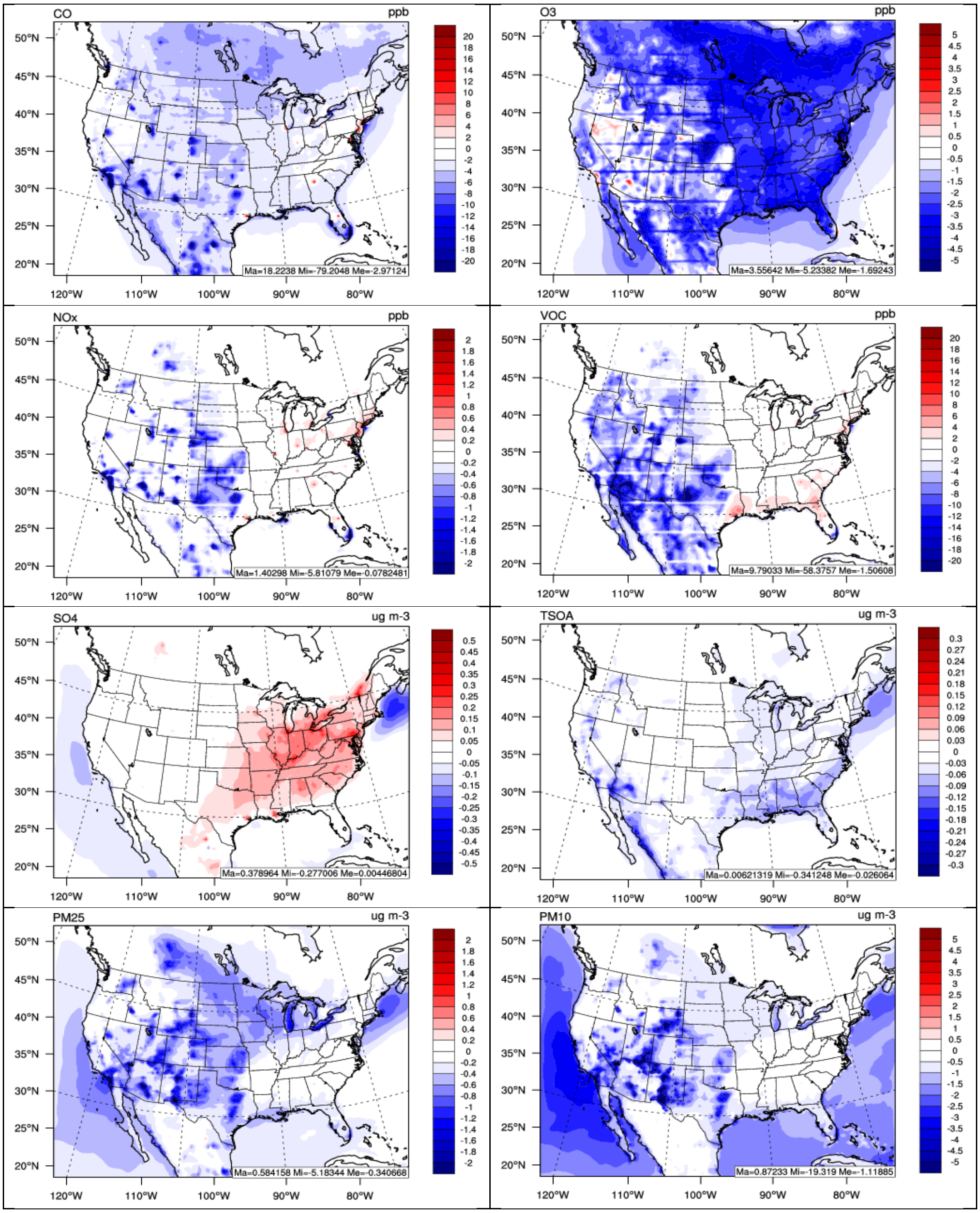

Figure 10. Spatial difference plots (two-way WRF-CMAQ - offline CMAQ) for major chemical species between two-way WRF-CMAQ and offline CMAQ.