Decadal Application of WRF/Chem for Regional Air Quality and Climate Modeling over the
U.S. under the Representative Concentration Pathways Scenarios. Part 2: Current vs. Future
Simulations

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Key Points

1. WRF/Chem under RCP4.5 and RCP8.5 scenarios predicts similar domain-mean increases for 2-m temperature but different spatial distributions
2. Future O₃ levels will increase for RCP8.5 but decrease for RCP4.5 due to higher GHGs and BVOCs and dis-benefit of reduced NO_x in some areas
3. Future PM_{2.5} levels will decrease under both RCP4.5 and RCP8.5, leading to lower cloud droplet number concentration and higher radiation

Abstract

Following a comprehensive model evaluation, this Part II paper presents projected changes in future (2046 - 2055) climate, air quality, and their interactions under the RCP4.5 and RCP8.5 scenarios using the Weather, Research and Forecasting model with Chemistry (WRF/Chem). In general, both WRF/Chem RCP4.5 and RCP8.5 simulations predict similar increases on average (~ 2°C) for 2-m temperature (T2) but different spatial distributions of the projected changes in T2, 2-m relative humidity, 10-m wind speed, precipitation, and planetary boundary layer height, due to differences in the spatial distributions of projected emissions, and their feedbacks into climate. Future O₃ mixing ratios will decrease for most parts of the U.S. under the RCP4.5 scenario but increase for all areas under the RCP8.5 scenario due to higher projected

temperature, greenhouse gas concentrations and biogenic volatile organic compounds (VOC) emissions, higher O₃ values for boundary conditions, and disbenefit of NO_x reduction and decreased NO titration over VOC-limited O₃ chemistry regions. Future PM_{2.5} concentrations will decrease for both RCP4.5 and RCP8.5 scenarios with different trends in projected concentrations of individual PM species. Total cloud amounts decrease under both scenarios in the future due to decreases in PM and cloud droplet number concentration thus increased radiation. Those results illustrate the impacts of carbon policies with different degrees of emission reductions on future climate and air quality. The WRF/Chem and WRF simulations show different spatial patterns for projected changes in T2 for future decade, indicating different impacts of prognostic and prescribed gas/aerosol concentrations, respectively, on climate change.

KEYWORDS: WRF/Chem, the representative concentration pathways (RCP) scenarios, Trend analysis, Future air quality, Continental U.S., air quality-climate interactions

1. Introduction

Following a comprehensive model evaluation of the Weather, Research and Forecasting model with Chemistry (WRF/Chem) simulations between RCP4.5 and RCP8.5 simulations for current decade (2001 - 2010) in the Part I paper, this Part II paper examines the impacts of projected emissions on future air quality and climate during 2046-2055 under the Representative Concentration Pathways (RCPs) scenarios with radiative forcing levels of 4.5 and 8.5 W m⁻² by 2100 (hereafter RCP4.5 and RCP8.5) using WRF/Chem. A number of regional climate models (RCMs) and regional air quality model (RAQMs) have been applied to simulate the impacts of projected emissions on future climate and air quality under the RCP scenarios. Table 1 summarizes some of those studies using an RCM or an RAQM to downscale a global climate model (GCM) under RCP scenarios over North America. Three out of the four studies use the

WRF model, which focused on the impact of the RCP emissions on solely climate change without considering changes in air quality. *Trail et al.* [2013] conducted WRF simulations to analyze the changes in meteorological fields which may affect air quality, however, they did not simulate air quality. Most of the studies therefore cannot provide an understanding of the effects of projected emissions on air quality and their trends, which is the motivation for the near-term RCP scenarios described by *Moss et al.* [2008]. The study by *Gao et al.* [2013] is the only one that used the downscaled WRF model to drive the Community Multiscale Air Quality (CMAQ) model to understand the projected climate changes on ozone (O₃). The WRF/CMAQ model used in *Gao et al.* [2013], however, is an offline-coupled model, which does not take into account the feedbacks between meteorology/climate and chemistry. In addition, they only simulated four current and four future years. Table S1 summarizes several studies based on the previous IPCC scenarios, mainly A1B and A2 scenarios. These studies, unlike those in Table 1, focused more on the future climate impacts on O₃ and particulate matter (PM) concentrations, however, they used offline-coupled models for both the GCMs and RCMs.

In this work, an online-coupled meteorology-chemistry model, WRF/Chem, is used to downscale the North Carolina State University's modified Community Earth System Model (CESM_NCSU) to simulate the decadal current and future year air quality and climate over continental U.S. (CONUS) using RCP scenarios. To our best knowledge, this is the first study to use the Weather Research and Forecasting model with Chemistry (WRF/Chem) v3.6.1 model for decadal current and future climate simulations over CONUS, as these simulations are computationally very expensive. The only other similar study is the work of *Molders et al.* [2014], which downscaled CESM using WRF/Chem over Southeast Alaska from 2006 to 2012. However, they did not use the RCP emission scenarios for current and future climate change, nor

they analyzed the air quality impacts from downscaling. The two emission scenarios used in this study, RCP4.5 and RCP8.5, will provide a range of projected levels of greenhouse gases (GHG) and anthropogenic emissions in the U.S. The background mixing ratios of CO₂, CH₄, and N₂O are 379 ppm, 1770 ppb, and 319 ppb, respectively, for the current decade under both RCP4.5 and RCP8.5 scenarios; and 475 and 540 ppm, 1774 and 2743 ppb, and 350 and 367 ppb, respectively, for future decade, under the RCP4.5 and RCP8.5 scenarios.

2. Differences in RCP4.5 and RCP8.5 Future-Year Emissions over the Continental U.S.

RCP4.5 and RCP8.5 emissions are only available for the year 2050 for the future year simulations. Figures S1 and S2 show the absolute and relative differences between future and current emissions of major species under RCP4.5 and RCP8.5 scenarios. The current decade emissions are calculated as a weighted average from the three sets of emissions over 10 years. Overall, for most gas-phase species, the reductions of emissions from current to future scenarios are larger for RCP4.5 compared to RCP8.5. The emissions of some species such as nitrous acid (HONO) increase in 2050 in parts of the U.S. in the RCP8.5 scenario but decrease in the whole CONUS in the RCP4.5 scenario. The emissions of some species such as for aldehydes, ethane, ethanol, formaldehyde, and methanol increase in 2050 over southeastern U.S. under the RCP4.5 scenario. The emissions of oxides of nitrogen (NO_x) and volatile organic compounds (VOCs) are reduced in 2050 to a larger extent under the RCP4.5 scenario compared to the RCP8.5 scenario. NH₃ emissions, however, increase under both scenarios, with larger increases for RCP8.5 compared to RCP4.5. However, as mentioned previously, caution should be exercised when analyzing the difference plots between future and current RCP4.5 and RCP8.5 emission scenarios due to the differences that exist between the current set of emissions between RCP4.5 and RCP8.5. The other obvious difference between the RCP4.5 and RCP8.5 scenarios is the

increase in the emissions of gases and aerosol species over Canada for the future RCP4.5 scenario, but a slight decrease for RCP8.5. This is likely due to the fact that for RCP4.5, there are significant increases in forest burning from deforestation and forest fires from current decade to 2050 under the RCP 4.5 scenario [Van der werf *et al.*, 2006; Clarke *et al.*, 2007; Smith *et al.*, 2006; Wise *et al.*, 2009; Riahi *et al.*, 2007], which is not the case under the RCP8.5 scenario. While the reduction of emissions for some inorganic species such as SO_4^{2-} are larger in 2050 for RCP4.5 compared to RCP8.5, the reverse is true for other species such as EC and unspciated primary $\text{PM}_{2.5}$. RCP8.5 emissions also have a larger percentage reduction of SO_4^{2-} emissions over the ocean compared to RCP4.5. The emissions of NH_3 increase in 2050 under the RCP4.5 and RCP8.5 scenarios with larger increases for RCP8.5. Overall, RCP8.5 has a larger reduction of PM emissions compared to RCP4.5, which will have impacts on simulated $\text{PM}_{2.5}$ concentrations, clouds, and radiation.

3. Comparison between Current and Future WRF/Chem and WRF Simulations

Figures 1 to 4 show the absolute differences between current and future year simulations for major surface meteorological, chemical, and aerosol-cloud-radiation variables for RCP4.5 and RCP8.5 WRF/Chem simulations. For comparison, Figures 1-2 also include the absolute differences between current and future decade WRF only simulations for surface meteorological variables. The figures are based on results averaged over the 10-year period for each current and future decade simulation.

3.1. Comparison for current vs. future WRF/Chem simulations under the RCP4.5 scenario

For the RCP4.5 simulations for WRF/Chem, as shown in Figure 1, domain-average 2-m temperature (T_2) increases by $\sim 2^\circ\text{C}$, while hourly maximum T_2 increases by 3.1°C , mainly over the north-central U.S. The largest relative increases are, however, in the colder areas over

124 Canada. *Trail et al.* [2013] conducted 5-year RCP4.5 current and future year simulations with
125 WRFv3.4 without chemistry. They showed a 1 °C warming in the future; however, the warming
126 mainly occurs in the western U.S. and over Canada. Based on this study, for RCP4.5, the largest
127 warming occurs over north-central U.S., despite significant warming over western U.S. and
128 Canada. Other studies based on the RCP4.5 scenarios have shown similar warming patterns over
129 CONUS. For example, the study of *Nazarenko et al.* [2015] using the global model GISS
130 ModelE2 showed warmer areas in Canada and the central U.S. comparing the results during the
131 late 21st century (2081-2100) to those during 1986-2005; and the study of *Separovic et al.* [2013]
132 using the fifth-generation Canadian regional climate model (CRCM5) showed warmer areas in
133 Canada, the north-central U.S., and the western U.S. comparing T2 during 2071-2100 to that
134 during 1981-2010. The changes in water vapor mixing ratio at 2-m (Q2) between future and
135 current climate follow closely the pattern for changes in T2. In general, average water vapor
136 mixing ratios increase in the future climate. The largest increases in Q2 occur over region where
137 the T2 changes are the smallest near the coastal areas, which gradually decrease inland. WS10 in
138 general increased over most parts of the U.S. due to the increase in T2, with larger increases in
139 Canada and Mexico. The changes in precipitation show a similar trend to the changes in Q2, i.e.,
140 the largest increases in precipitation occur over the ocean and near the coastal areas, while for
141 the central U.S. there is a decrease in precipitation. There are differences in the precipitation
142 projections from different studies. For example, *Trail et al.* (2013) showed a decrease in
143 precipitation over northwestern U.S. and increases in precipitation over all other parts of the
144 U.S., *Separovic et al.* [2013] showed different precipitation trends for December to February,
145 namely, increases over most parts of the U.S. but decreases over Mexico and for June to August
146 and decreases in the north central U.S. and Canada and increases over other parts of the U.S.,

Wang and Kotamarthi [2015] showed decreases in precipitation over southwestern and south-central U.S. and increases in all other parts of the U.S. and Canada with both WRF and CCSM4, and Nazarenko *et al.* [2015] showed the largest increases in precipitation over eastern U.S. and Canada.

As shown in Figure 2, planetary boundary layer height (PBLH) increases over most parts of the U.S. due to the increase in T2, and the combination of the increase in WS10 and PBLH will help to enhance the dispersion of pollutants, reducing their concentrations. The radiation variables such as downward shortwave radiation (SWDOWN) and outgoing longwave radiation (OLR) increase over land and the ocean in the eastern portion of the domain but decrease over the ocean in the western portion of the domain. The net longwave radiation (GLW) is projected to increase over land and ocean due to increases in GHGs in the future scenario by $\sim 10 \text{ W m}^{-2}$. The domain increase for SWDOWN in this study is $\sim 3 \text{ W m}^{-2}$, consistent with the SWDOWN trend projected by Trail *et al.* [2013]. Trail *et al.* [2013] showed increases in SWDOWN of between 0 and 6 W m^{-2} over North America. The increases and decreases in SWDOWN are linked to cloud variables, which will be discussed later in this section. The trend for OLR is similar to that of the WRF simulations, with a general increase over land by $\sim 0.6 \text{ W m}^{-2}$.

As shown in Figure 3, the mixing ratios of SO_2 , NO_x , and CO are projected to decrease and those of NH_3 are projected to increase in responses to reductions and increases, respectively, in their projected emission under the RCP4.5 scenario (see Figure S1). The concentrations of several other gases such as peroxy acetyl nitrate (PAN) are also reduced due to decreases in emissions in those primary species (Figures not shown). As shown in Figure 4, under the RCP4.5 scenario, O_3 mixing ratios are projected to decrease in future decade over most parts of the U.S. with a domain-average decrease of $\sim 2 \text{ ppb}$ with the exception of major urban cities in California

(CA) and over the northeastern U.S. where O₃ mixing ratios are projected to increase with a maximum increase of ~10 ppb. This pattern is also found from the CAM-Chem simulations from *Gao et al.* [2013], especially during the fall and spring seasons. Although both VOCs and NO_x concentrations decrease over major cities in CA and northeastern U.S. in this study, O₃ mixing ratios increase over those regions. According to *Gao et al.* [2013], the increases in O₃ mixing ratios in major urban cities in CA and over northeastern U.S. are due to decreased NO titration resulted from the reduction of significant NO_x emissions, which leads to decreased NO_x concentrations over the whole CONUS as shown in Figure 3. Another reason for the increased O₃ mixing ratios in major cities in CA and northeastern U.S. but decreased O₃ mixing ratios in remaining regions lies in different responses of O₃ chemistry to NO_x emissions reductions. To illustrate this, Figure 5 shows the spatial plots of two O₃ photochemical indicators: total reactive nitrogen (NO_y) and the ratios of the concentrations of O₃ to those of NO_y (O₃/NO_y) under both RCP4.5 and RCP8.5 scenarios. The two indicators are chosen based on the findings of *Campbell et al.* [2015], which showed that these indicators were the most robust, as compared to other O₃ surface indicators including the concentration ratios of hydrogen peroxide to nitric acid (H₂O₂/HNO₃), formaldehyde to NO_y (HCHO/NO_y) and formaldehyde to nitrogen dioxide (HCHO/NO₂). Based on *Zhang et al.* [2009], the NO_x-limited transition values for NO_y and O₃/NO_y are ≤ 5 ppb and ≥ 15 , respectively. Based on these indicators, the current-decade simulations under both RCP4.5 and RCP8.5 scenarios show mainly NO_x-limited O₃ chemistry over western U.S.; and VOC-limited O₃ chemistry over eastern U.S. and major cities and urban areas in western U.S. However, the future-decade simulation under the RCP4.5 scenario shows more NO_x-limited O₃ chemistry in terms of both extent and areas as compared to the current-decade simulation under the RCP4.5 scenario, although O₃ chemistry over major cities in CA

and northeastern U.S. remain VOC-limited. While under the RCP4.5 scenario the significant decrease in NO_x emissions results in the decrease in O₃ mixing ratios over most parts of the U.S. where O₃ chemistry is NO_x-limited, it also results in an increase in O₃ mixing ratios in northeastern U.S. and major cities in CA where O₃ chemistry is VOC-limited.

The decrease in O₃ mixing ratios as well as the increase in surface water vapor results in a decrease in OH mixing ratios in future decade (Figure not shown). As shown in Figure 4, the mixing ratios of total VOCs (TVOCs) are reduced over most areas in the U.S. due to reduced projected anthropogenic VOC emissions. They, however, increase over some areas (e.g., some states in the southeastern U.S.) because of higher mixing ratios of biogenic emissions and therefore biogenic VOCs (BVOCs) such as isoprene, and terpene driven by the increase in T₂ and radiation. The increases in BVOCs and primary OA (POA) (Figure not shown) lead to increases in total SOA (TSOA) over some areas the U.S., due to increases in biogenic SOA (BSOA) from BVOCs with the exception of southern California, western Arizona and southern Nevada where TSOA decreases. The anthropogenic VOC emissions increase over Canada under the RCP4.5 scenario (Figure S2), which leads to an increase in TSOA over Canada. PM_{2.5} concentrations are projected to decrease over CONUS (with an exception in southern Canada) with a domain-average decrease of $\sim 1.5 \mu\text{g m}^{-3}$ ($\sim -30\%$) due to decreases in the concentrations of most PM_{2.5} species including ammonium (NH₄⁺), sulfate (SO₄²⁻), nitrate (NO₃⁻), elemental carbon (EC), and anthropogenic SOA (ASOA) that compensate the small increases in BSOA in some areas.

Due to the decreases in the concentrations of fine particulate matter (PM_{2.5}) under the RCP4.5 scenario, the cloud condensation nuclei (CCN) at supersaturation 0.5% and the cloud droplet number concentration (CDNC) decrease over almost the whole domain (Figure 6). The

spatial distributions of the decrease in CCN and CDNC concentrations are also similar to those in $PM_{2.5}$ concentrations, with the largest decreases occurring over the northeastern U.S. This is to be expected, as CCN is influenced by components of $PM_{2.5}$ even though large uncertainties exist (Yu, 2000, 2014). Due to decreases in CDNC, the cloud optical thickness (COT) also decreases over most parts of the domain except for some areas in western U.S. The cloud fraction (CF) and cloud water path (CWP) decrease over most parts of the land area but increases over the ocean and over a portion of southeastern U.S. The spatial distributions of changes in CWP are very similar to those in precipitation shown in Figure 1. Generally there is an increase in moisture over the ocean, increasing CWP despite decreases in CDNC. The shortwave and longwave cloud forcing (SWCF) generally decrease over the whole domain except for the oceanic areas in the western portion of the domain, which is similar to COT. The projected increases in COT, CWP, and SWCF indicate higher cloud amounts over the ocean in the western part of the domain, which results in a decrease in SWDOWN. By contrast, the projected decreases in COT, CWP, and SWCF over land indicate decreases in cloud amounts, which results in an increase in SWDOWN over land (see Figure 2).

3.2. Comparison of current vs. future WRF/Chem simulations under the RCP8.5 scenario.

As shown in Figure 1, for the RCP8.5 simulations, domain-average T2 also increases by ~ 2 °C, while hourly maximum T2 increases by 2.9 °C, mainly over the western and central U.S. The spatial pattern for T2 for RCP8.5 is different compared to RCP4.5. However, it is more similar to that of *Trail et al.* [2013], and that of differences in summer T2 between the periods of 2045-2055 and 1995-2005 under the IPCC A1B scenario reported by *Leung and Gustafson* [2005]. *Gao et al.* [2012] also showed the largest increases in heat wave duration and frequency based on the daily maximum temperature simulated by WRFv3.2.1 mainly over western U.S.

Similar to the predictions under the RCP4.5 scenario, the changes in water vapor mixing ratio at 2-m (Q2) between future and current climate follow closely the pattern for changes in T2. However, there are smaller changes for Q2 under RCP8.5 compared to RCP4.5 due to the smaller changes in T2. The largest increases in WS10 occur in central U.S. and central Canada with very small decreases in WS10 in eastern and western Canada. Larger changes are simulated for precipitation under the RCP8.5 scenario compared to the RCP4.5 scenario. Precipitation in general increases over the whole domain with the largest increases over southeastern U.S. This trend is similar to the results from *Wang and Kotamarthi* [2015] based on the RCP8.5 scenario using the WRF RCM, which also show the largest increases from 1995-2004 to 2045-2054 over southeastern U.S. *Wang and Kotamarthi* [2015] also showed decreases in precipitation over south-central U.S., which occur over south-central and north-central U.S. in this study. *Nolte et al.* [2008] also simulated a very similar precipitation pattern using the MM5 model based on the A1B scenario when comparing differences between summer 2048-2052 and 1999-2003, with the largest increases in precipitation over southeastern U.S. and very small differences in precipitation over other parts of the U.S. The common findings for this study and other studies from literature are the increases in precipitation over southeastern U.S. for the RCP8.5 scenario.

As shown in Figure 2, PBLH in general increases over most parts of the U.S. The domain-average increase in shortwave radiation is $\sim 3.2 \text{ W m}^{-2}$ with larger increases over the central and eastern U.S. compared to the western U.S. and Canada. The simulated SWDOWN by WRF/Chem, on a domain-average, has a slightly higher increase under the RCP8.5 scenario compared to a domain-average increase of $\sim 3 \text{ W m}^{-2}$ the RCP4.5 scenario. The larger increase in precipitation under the RCP8.5 scenario is likely due to the larger increase in SWDOWN. GLW increases with a domain-average of $\sim 11.0 \text{ W m}^{-2}$. OLR increases over the whole domain with the

exception of some parts of the ocean with a domain-average increase of $\sim 1.0 \text{ W m}^{-2}$. The larger increase in GLW is due to the larger increase in GHG concentrations under the RCP8.5 scenario as compared to the RCP4.5 scenario.

Similar to RCP4.5 scenario, the mixing ratios of SO_2 and NO_x are projected to decrease, and those of NH_3 are projected to increase, respectively, in responses to changes in their emissions. The mixing ratios of CO are reduced over eastern U.S., and some areas in the western U.S., but increase over oceanic areas and some areas in the western U.S., mainly because of larger increases in ship emissions over ocean and higher CH_4 background concentrations under the RCP8.5 scenario than the RCP4.5 scenario. The oxidation of CH_4 increases the mixing ratio of formaldehyde, which in turn produces CO. In contrast to the RCP4.5 scenario, the O_3 mixing ratios are projected to increase over the whole domain under the RCP8.5 scenario with a domain-average increase of ~ 2.6 ppb. Large increases in O_3 mixing ratios of up to ~ 17 ppb are found over major cities mainly in the northeastern U.S. and California. The projected increase and spatial distribution for O_3 mixing ratios under the RCP8.5 scenarios are similar to those of *Gao et al.* [2013] using CAM-Chem for the spring, fall, and winter seasons which showed increases over most parts of the U.S. and the largest increases over major cities in eastern U.S. and California in winter. *Gao et al.* [2013] attributed such O_3 increases to high concentrations of CH_4 , which could contribute to the increases of O_3 mixing ratios as large as 4 to 8 ppb. As shown in Figure 5, similar to the RCP4.5 scenario, the future-decade simulation under the RCP8.5 shows more NO_x -limited O_3 chemistry in terms of both extent and regions compared to the current-decade simulation although most areas in eastern U.S. and major cities throughout the U.S. remain VOC-limited. Compared to the future-decade simulation under the RCP4.5 scenario that shows more NO_x -limited regions, the future-decade

simulation under the RCP8.5 scenario shows more VOC-limited O₃ chemistry in terms of both extent and regions especially over the eastern U.S. and major cities in the rest of U.S. In addition to higher level of GHGs under the RCP8.5 scenario, decreased NO titration and the reduction in NO_x emissions over VOC-limited regions can explain mostly the increase in O₃ mixing ratios over most of U.S. In addition, the decrease in VOC concentrations for RCP8.5 over the western U.S. is small compared to that for RCP4.5. As more locations are VOC-limited under the future RCP8.5 scenario comparing to the future RCP4.5 scenario, a slight increase in VOC concentrations can increase O₃ mixing ratios significantly, leading to increased O₃ mixing ratios domainwide in general for the RCP8.5 scenario. The opposing trends between O₃ mixing ratios for WRF/Chem RCP8.5 and RCP4.5 are explored further in Section 3.4.

Similar to RCP4.5, TSOA concentrations do not vary by much in general over the U.S., with the largest decreases in CA and southern Arizona, but with magnitudes of < 1 µg m⁻³ due to large decreases in VOC concentrations over those areas as shown in Figure 4. TSOA for RCP4.5 shows an obvious increase over Canada, but not so much for RCP8.5, consistent with the increases in the anthropogenic VOC emissions under the RCP4.5 scenario and decrease in them under the RCP8.5 scenario over Canada as shown in Figures S1 and S2. However, as compared to the RCP4.5 scenario which shows increase in TSOA over southeastern U.S., the RCP8.5 scenario shows a decrease, due to the lower VOC mixing ratios over southeastern U.S. as shown in Figure 4. PM_{2.5} concentrations however, decrease over all parts of the U.S. for RCP8.5 with a domain-average decrease of ~1.7 µg m⁻³ (~35%), but with localized decreases of as large as 26 µg m⁻³. The spatial pattern for the decrease in PM_{2.5} concentrations is similar to that under the RCP4.5 scenario with the largest decreases over eastern U.S. but with a larger extent over the whole domain, particularly in urban areas in CA. The larger decrease in PM_{2.5} concentrations for

the RCP8.5 scenario as compared to RCP4.5 are due to larger decreases in EC and ASOA. In addition, POA increases for RCP4.5 but decreases for RCP8.5. The decrease in SO_4^{2-} , NH_4^+ , and NO_3^- are smaller under the RCP8.5 scenario as compared to the RCP4.5 scenario (Figures for PM species are not shown).

Similar to RCP4.5, due to decreases in $\text{PM}_{2.5}$ concentrations, CCN and CDNC also decrease for RCP8.5 over the whole domain as shown in Figure 6. However, the decreases in CCN and CDNC for the RCP8.5 scenario are not as large as that under the RCP4.5 scenario even though the $\text{PM}_{2.5}$ concentrations decrease to a larger extent under the RCP8.5 scenario as compared to RCP4.5 scenario. This is because there are smaller decreases in $\text{PM}_{2.5}$ species (e.g., SO_4^{2-} , NH_4^+ and NO_3^-) that can form cloud droplets under the RCP8.5 scenario as compared to the RCP4.5 scenario. Compared to the RCP4.5 scenario, the changes in CF are similar but COT and SWCF decrease to a larger extent over the whole domain under the RCP8.5 scenario. The increased radiation under the RCP8.5 scenario compared to the RCP4.5 scenario results in increased precipitation and reduced COT. Unlike the RCP4.5 scenario, CWP also increases mainly over land but with some local decreases over the ocean.

3.3. Comparison of RCP4.5 and RCP8.5 current vs. future WRF only simulations

As shown in Figure 1, the maximum difference in T2 between current and future decade WRF only simulations under both RCP4.5 and RCP8.5 scenarios occurs in western U.S. The spatial patterns of projected changes in T2 from both simulations are most similar to those from *Trail et al.* [2013] which showed the largest differences in T2 over western U.S. and western Canada, because *Trail et al.* [2013] also used the WRF model with prescribed gas and aerosol concentrations. Among the four simulations, the WRF RCP8.5 simulation gives the largest domain mean absolute and percentage differences in T2 and Q2 (2.1°C and 16.5%, and 1 g kg⁻¹

and 12% respectively). On the other hand, WS10 differences over land from the WRF only simulations are much smaller compared to those from WRF/Chem simulations due to very small differences in SWDOWN as shown in Figure 2. Precipitation differences from the WRF only simulation are similar to those from the WRF/Chem simulation, with the largest increases occurring over southeastern U.S. However, compared to WRF/Chem, WRF gives much smaller increase in precipitation over southeastern U.S. due to the smaller increase in SWDOWN under both RCP4.5 and RCP8.5 scenarios.

As shown in Figure 2, the spatial patterns of PBLH for WRF/Chem and WRF RCP8.5 are very similar with the largest increases in PBLH over Rocky Mountain Region, and the largest decreases in PBLH occur over the ocean in the western portion of the domain. Increases in PBLH are smaller for RCP8.5 WRF compared to WRF/Chem due in part to smaller differences in SWDOWN. The spatial patterns of PBLH for WRF/Chem and WRF RCP4.5 are also similar, both with maximum increases over the northern and middle portions of the Great plains in the U.S., which correspond to the maximum differences in T2 in the same region. The very small differences (close to zero especially over the land) in SWDOWN between future and current WRF simulations under the RCP8.5 scenario are likely due to the use of the same prescribed aerosol concentrations in current and future decade simulations. Compared to WRF/Chem, WRF gives much larger differences in GLW under both RCP4.5 and RCP8.5 scenarios, with domain mean values higher by $\sim 2 \text{ W m}^{-2}$. The larger increase in GLW by WRF is likely due to larger increases in CF (Figure not shown) and water vapor mixing ratios. For similar reasons, OLR from the WRF simulations are higher than those from the WRF/Chem simulations.

3.4. Influential factors for projected changes in future O₃

There are several differences between the setting of future RCP4.5 and RCP8.5 simulations, including different projected emissions and different background levels of CH₄ (1774 ppb vs. 2743 ppb), CO₂ (475 ppm vs. 540 ppm), and N₂O (350 ppb vs. 367 ppb). One-month sensitivity simulations are carried out to understand the differences in projected O₃ changes under the RCP4.5 and RCP8.5 scenarios and quantify the effects of various influential factors such as chemical BCONs, the NO_x emissions, the background concentration of methane (CH₄) and other GHGs, and the oxidation of CH₄ by OH radicals on the projected changes in O₃ mixing ratios. April 2048 and April 2003 are chosen as representative months; as the differences between projected O₃ mixing ratios for these months (i.e., Base RCP4.5 and Base RCP8.5 in Figure 7) are similar to those based on the 10-year average shown in Figure 4, namely, O₃ will increase in general under the future RCP8.5 scenario and decrease in general over land for under the future RCP4.5 scenario. The sensitivity simulations for future RCP8.5 scenario include (i) the use of the same input as the Base RCP8.5, except that the RCP4.5 background CH₄ levels are used; (ii) the use of the same input as the Base RCP8.5, except that the RCP4.5 background CH₄ levels and NO_x emissions are used; (iii) the use of the same input as the Base RCP8.5, except that RCP4.5 background CH₄ levels and chemical BCONs are used; and (iv) the use of the same input as the Base RCP8.5, except that RCP4.5 background CH₄ levels are used and the reaction of CH₄+OH to produce O₃ is shut off. The above four sets of sensitivity simulations are referred to as Sen (i) to Sen (iv). Figure 7 shows the differences between the results of April 2048 and April 2003. For the Base RCP4.5 and RCP8.5 simulations, domain-average O₃ levels decrease by 1.9 ppb and 2.2 ppb, respectively. The differences between April 2048 Sen (i) and April 2003 simulation show that the domain-average O₃ levels increase by 1.2 ppb, indicating that the elevated CH₄ levels under future RCP8.5 scenario relative to those under the RCP4.5 scenarios

contribute significantly (~1.0 ppb out of 2.2 ppb) to the increase in the future RCP8.5 O₃ levels. These results are consistent with the study by Gao et al. (2013) which attributed the O₃ increases in RCP8.5 to the high concentrations of CH₄. The NO_x emissions also contributed to the differences in O₃ levels between RCP4.5 and RCP8.5. The differences between April 2048 Sen (ii) and April 2003 simulations show a decrease in domain-average O₃ mixing ratios of 0.5 ppb, indicating that approximately ~0.7 ppb decrease of O₃ mixing ratios can be attributed to the differences in NO_x emissions between RCP4.5 and RCP8.5. It is more difficult to carry out a sensitivity simulation with VOC emissions, as the biogenic emissions are calculated online, and they depend on factors such as T2 and SWDOWN. Comparison of differences between Sen (iii) and Base simulations and between Sen (i) and Base simulations shows that the influence of BCONs is significant. Using RCP4.5 chemical BCONs reduce the difference between future and current scenarios by ~2.1 ppb, changing the domain-average difference between future and current April simulations from 1.2 ppb to -0.92 ppb. Switching off CH₄+OH chemistry also reduces the O₃ levels for the future scenario to a large extent. Comparison of the differences between April 2048 Sen (i) and April 2003 simulations and between April 2048 Sen (iv) and April 2003 simulations shows that the domain-average O₃ levels decrease by > 2 ppb (changing from ~1.2 to ~ -1.1 ppb). The sensitivity simulations show that the CH₄ levels and the chemical BCONs contribute the most to the increase in O₃ mixing ratios.

To assess impacts of different GHGs such as CH₄, carbon dioxide (CO₂), and nitrous oxide (N₂O) on projected O₃ changes, additional sensitivity simulations are performed. The sensitivity simulations involve using background RCP4.5 levels for different combination of the greenhouse gases for (v) to (vii), and using background RCP4.5 levels for all greenhouse gases for (viii). Figure 8 shows differences between those additional sensitivity simulations (April

2048 Sen (v) to Sen (viii)) and the April 2003 simulations). Compared to the Base RCP8.5 case in Figure 7, the differences between April 2048 Sen (viii) and April 2003 simulations show that the increase in all GHG levels from RCP4.5 and RCP8.5 contribute to about a 1 ppb increase in O₃ mixing ratios in the future, without considering CH₄+OH chemistry. It is interesting that there are not much differences in future O₃ mixing ratios if only one of the GHGs is set at the RCP8.5 level, while the other two GHGs are set at the RCP4.5 levels. However, the cumulative impact of all two or three GHGs at the RCP8.5 levels contribute to the ~1 ppb increase in O₃ mixing ratios in the future.

3.5. Impact of policy scenarios on future air quality and climate over the continental U.S.

Both RCP4.5 and RCP8.5 WRF/Chem scenarios produce a domain-mean increase of approximately 2°C, however, with varying magnitudes for different locations in the U.S. The differences in spatial distributions of T2 are influenced by the distribution of aerosol particles for the different scenarios. A different spatial pattern for WRF only T2 differences is also found. However, all cases show that the current vs. future differences are smaller for southeastern U.S. compared to western U.S. and Canada. The largest differences in T2 are found for the RCP8.5 WRF only case, followed by the RCP4.5 WRF/Chem case, the RCP8.5 WRF/Chem and lastly the RCP4.5 WRF only case (see Figure 1). The RCP8.5 scenario is supposed to produce the largest radiative forcing level at 2100 at 8.5 W m⁻², as compared to the RCP4.5 scenario, at 4.5 W m⁻². Intuitively, the RCP8.5 scenario at 2050 should project higher T2 levels compared to the RCP4.5 scenario. This is shown in the WRF RCP8.5 simulations but not the WRF/Chem RCP8.5 simulations, indicating the importance of emissions and chemistry and their non-linear effects on T2. In addition, a number of studies used RCMs without simulating aerosol and chemistry [e.g., *Trail et al.*, 2013], or offline-coupled models that do not simulate feedbacks of chemistry into

meteorology/climate [e.g., *Gao et al., 2013; Sun et al., 2015*]. The results in this work and comparison with previous studies show that the meteorological predictions without the inclusion of prognostic aerosol and chemistry can produce different results for important surface meteorological parameters such as T2. In addition, the distribution of emissions over U.S. can influence the magnitude of increase of T2, Q2, average WS10, and precipitation over different locations in the U.S. in the future. The future is also more moist (increases in Q2) for all four cases as compared to the current scenarios. In all cases, precipitation is shown to increase over southeastern U.S. in the future. However, compared to the WRF only simulations, the simulations with WRF/Chem also show a shift in the spatial coverage of increased precipitation over southeastern U.S. under both RCP scenarios, and smaller increases in Florida, and larger increases in Tennessee, Alabama and Mississippi under RCP8.5 scenario.

As discussed in sections 3.2 and 3.4, the predicted changes in O₃ mixing ratios are very different over most areas under the RCP4.5 and RCP8.5 scenarios. Over northeastern U.S. and in major cities in California, even though future NO_x and VOC emissions decrease, WRF/Chem simulations under both RCP4.5 and RCP8.5 scenarios show increases in O₃ mixing ratios, up to a maximum of 10 ppb for RCP4.5 and up to 17 ppb for RCP8.5. Over other parts of the U.S., the simulation under the RCP4.5 scenario shows decreases in future O₃ mixing ratios of up to ~2.2 ppb whereas the simulation under the RCP8.5 scenario shows increases in O₃ mixing ratios. This is an important finding with regards to policy issues in the reduction of O₃ mixing ratios over the U.S. Namely, reducing VOC and NO_x emissions for the whole U.S. might not lead to decreases in O₃ mixing ratios, but might lead to an increase in O₃ mixing ratios in the future as shown in all parts of the U.S. for RCP8.5, and in urban areas in RCP4.5. In addition, regulatory agencies would have to take into consideration the increase in the BVOCs due to increases in T2 in the

future. O₃ mixing ratios in the future are also more likely to increase due to increases in T2, SWDOWN, and GLW at the surface. Each city, county or state, depending whether it is a VOC-limited or NO_x-limited area would have to take into account the possibility of an indicator-regime change in the future to control O₃ mixing ratios.

PM_{2.5} concentrations decrease in general for both cases RCP4.5 and RCP8.5, largely over eastern U.S. (where PM_{2.5} concentrations from the current decade simulations are higher) and over major cities in western U.S. The decrease in PM_{2.5} concentrations in the future will help to improve health effects and visibility overall, however, the changes in PM_{2.5} species concentrations will influence the changes in aerosol-cloud variables which will then affect changes in radiation variables. For example, larger decreases in more soluble PM_{2.5} species such as SO₄²⁻, NO₃⁻, and NH₄⁺ will reduce the CDNC to a larger extent, as compared to reducing insoluble PM species such as EC or dust. The differences in emission scenarios affect radiation at the top of the atmosphere to a larger extent compared to shortwave and longwave radiation at the surface. The increase in OLR from the current to the future scenarios is larger by ~1 W m⁻² under the RCP8.5 scenario as compared to the RCP4.5 scenario.

4. Comparison of Future AQ and Climate Predicted by WRF/Chem and CESM_NCSU

Figures 9 to 12 show the spatial distributions of the changes in several meteorological, chemical, and aerosol-cloud variables projected by CESM_NCSU between future and current decades under the RCP4.5 and RCP8.5 scenarios. Those projected changes are compared with the results from WRF/Chem shown in Figures 1-4 and 6. Figures S3-S7 compare the spatial distributions of selected variables for current and future decades simulated by both models.

As shown in Figure S3, for 2-m temperature (T2), the spatial patterns are similar with the exception of stronger spatial variability for WRF/Chem, which is obvious over western U.S. As shown in Figure 9, Both WRF/Chem and CESM_NCSU project an increase by $\sim 2^{\circ}\text{C}$ for a domain-average T2 and by $\sim 1^{\circ}\text{C}$ for domain maximum temperatures. There are more noticeable differences between WRF/Chem and CESM_NCSU, however, in the future scenarios. For example, higher T2 is predicted over the Gulf of California and northern U.S. by CESM_NCSU for both scenarios compared to WRF/Chem. The higher T2 results in higher 2-m water vapor mixing ratio (Q2) in the future for both WRF/Chem and CESM_NCSU, with higher Q2 for RCP8.5 than for RCP4.5 as shown in Figures 1 and 9. For 10-m wind speed (WS10), in general, both WRF/Chem and CESM_NCSU predict a relatively constant domain-average changes in WS10 for both scenarios despite opposite in signs with slight increases projected by WRF/Chem but slight decreases projected by CESM over most of the domain. For precipitation, as expected, the projected precipitation and its changes by WRF/Chem exhibit much strong spatial variability than CESM over CONUS (Figures S8, 1, and 9). The increases in domain maximum and average precipitation occur for RCP4.5 WRF/Chem and CESM_NCSU simulations and for RCP8.5 WRF/Chem simulations, but a decrease occurs in domain-average precipitation for RCP8.5 CESM_NCSU simulations. For WRF/Chem RCP8.5, however, column water vapor mixing ratios (Figure not shown) are much lower as compared to the WRF/Chem RCP4.5 scenario and the CESM_NCSU RCP8.5 case. Therefore, more of the moisture in the atmosphere in WRF/Chem RCP8.5 is converted to precipitation, rather than remaining as precipitable water vapor.

As shown in Figure S4, PBLH from WRF/Chem and CESM_NCSU shows significant differences over most CONUS and oceanic areas. WRF/Chem uses the YSU PBL scheme, while

CESM_NCSU uses the moisture PBL scheme of *Bretherton and Park* [2009]. For current RCP4.5, over eastern U.S., the differences in PBLH between WRF/Chem and CESM_NCSU are > 60 m. The PBLH for WRF/Chem in general is higher over land (except for some areas in western U.S.) but lower over the ocean as compared to CESM_NCSU for current and future decades under both RCP4.5 and RCP8.5 scenarios. As shown in Figures 2 and 9, all WRF/Chem and CESM_NCSU simulations show increases in PBLH over most areas in the future, due to increases in T2 and SWDOWN. However, the two models predict different areas with large PBLH increases under both RCP scenarios. For example, under the RCP4.5 scenario, the largest increases in PBLH occur over Pacific Northwest by CESM_NCSU but over the northern portion of the Great Plains in the U.S. by WRF/Chem.

As shown in Figures 2, 9, and S5, downward shortwave radiation (SWDOWN) is projected to increase in the future by ~ 4 to 5 W m^{-2} on average by WRF/Chem and $\sim 2 \text{ W m}^{-2}$ on average by CESM_NCSU under both RCP4.5 and RCP8.5 scenarios. In addition, SWDOWN is projected to increase in the future to a slightly larger extent by both WRF/Chem and CESM_NCSU under the RCP8.5 scenario compared to the RCP4.5 scenario. While WRF/Chem projected increases in SWDOWN over most areas under both scenarios, CESM_NCSU projected decreases over western U.S. under the RCP4.5 scenario and over an area in northern U.S. under the RCP8.5 scenario, which results in much smaller domain-average increases in SWDOWN by CESM than by WRF/Chem. Both WRF/Chem and CESM_NCSU projected increases in downward longwave radiation (GLW) over most areas under both scenarios with higher domain average increases by CESM_NCSU. The magnitudes of outgoing longwave radiation (OLR) simulated by CESM_NCSU current and future decade simulations are much higher by $> 5 \text{ W m}^{-2}$ on average compared to those from WRF/Chem simulations. The OLR from WRF/Chem

increases by ~ 1 to 3 W m^{-2} but increases by $\sim 3 \text{ W m}^{-2}$ from CESM_NCSU in the future under both RCP4.5 and RCP8.5 scenarios. This is likely due to total ozone column concentrations which are much higher for CESM_NCSU as compared to WRF/Chem for both current and future decade scenarios. CESM_NCSU and WRF/Chem predict quite different spatial distributions in changes in OLR under both scenarios. Although both WRF/Chem and CESM_NCSU use the Rapid and accurate Radiative Transfer Model for GCM (RRTMG) for both shortwave and longwave radiation, the simulated radiation variables are mainly affected by the tuning of radiation variables for CESM_NCSU, different physical options (e.g., PBL scheme and cumulus parameterizations) used in both models, as well as the horizontal and vertical resolutions used in both simulations.

As shown in Figure S6, there are large differences in SO_2 concentrations for current decade simulations due to the differences in the treatment of SO_2 emissions in WRF/Chem and CESM_NCSU. SO_2 emissions for WRF/Chem are interpolated on an hourly basis from the monthly RCP emissions, and interpolated vertically from the surface RCP emissions, based on the National Emissions Inventory (NEI) emission profile from U.S. EPA as described in *Yahya et al.* [2016]. The hourly temporal profiles of 2001, 2005, and 2010 RCP emissions are based on the 2002 NEI and the Air Quality Modelling Evaluation International Initiative (AQMEII) 2006 and 2010 emissions [Pouliot *et al.*, 2015] derived based on the 2008 NEI, respectively. SO_2 emissions from CESM_NCSU are based on the monthly RCP emissions, and are distributed vertically within the model. SO_2 mixing ratios from WRF/Chem are higher by a domain-average of ~ 0.3 ppb and domain maximum SO_2 mixing ratios of up to ~ 12 ppb than those from CESM_NCSU for both RCP4.5 and RCP8.5 current decade simulations. As shown in Figures 3 and 10, the SO_2 mixing ratios are projected to decrease by 0.84 ppb ($\sim 70\%$) and 0.79 ppb

(~66%) on average by WRF/Chem and 0.62 ppb (~77%) and 0.51 ppb (~72%) on average by CESM_NCSU for the future decade RCP4.5 and RCP8.5 scenarios, respectively. Therefore, both WRF/Chem and CESM_NCSU RCP4.5 scenarios show a larger decrease in SO₂ mixing ratios for RCP4.5 as compared to RCP8.5. Both models project decreases in NO_x mixing ratios with larger decreases by WRF/Chem under both scenarios. The domain-average decreases in NO_x mixing ratios are 1.7 and 1.3 ppb by WRF/Chem and 1.1 ppb and 0.8 ppb by CESM_NCSU under the RCP4.5 and RCP8.5 scenarios, respectively. Both models project increases in NH₃ mixing ratios with larger increases by CESM_NCSU. The changes in NH₃ mixing ratios are affected more by changes PBLH than more reactive species. Under both scenarios, CESM_NCSU predicts smaller domain-average increases in PBLH than WRF/Chem, resulting in larger increases in NH₃ mixing ratios. Both models predict similar changes in CO mixing ratios with decreases over the whole CONUS under the RCP4.5 scenario and over eastern U.S. and a few western states (e.g., California (CA) and Washington (WA)).

As mentioned earlier, O₃ mixing ratios for CESM_NCSU current decade simulations are too high when compared against observational data (Figure S6). The domain-average O₃ mixing ratios are projected to decrease by ~2 ppb by WRF/Chem and ~4 ppb by CESM_NCSU in the future RCP4.5 scenario, but increase by ~3 ppb by both WRF/Chem and CESM_NCSU in the future RCP8.5 scenario. As shown in Figures 4 and 11, both WRF/Chem and CESM_NCSU show similar trends in changes in the domain-average and maximum O₃ mixing ratios under both scenarios with a decrease under the RCP4.5 scenario and an increase under the RCP8.5 scenario. While CESM_NCSU shows larger magnitudes in domain-average changes, WRF/Chem shows larger localized increases in northeastern U.S. under the RCP4.5 scenario and eastern U.S. under the RCP4.5 scenario, and CA under both scenarios.

As shown in Figure S7, the spatial distributions of CCN for current decade simulations are similar for both WRF/Chem and CESM_NCSU simulations, with differences in magnitude. However, current decade WRF/Chem simulations have lower CCN for both RCP4.5 and RCP8.5 cases by $\sim 0.8 \times 10^8 \text{ cm}^{-2}$ and $\sim 0.6 \times 10^8 \text{ cm}^{-2}$ as compared to CESM_NCSU simulations, respectively. As shown in Figures 6 and 12, for RCP4.5, the domain-average decrease in CCN is $1.2 \times 10^8 \text{ cm}^{-2}$ (~63%) and $\sim 1.1 \times 10^8 \text{ cm}^{-2}$ (~58%) for WRF/Chem and $0.9 \times 10^8 \text{ cm}^{-2}$ (~56%) and $0.7 \times 10^8 \text{ cm}^{-2}$ (~28%) by CESM_NCSU under the RCP4.5 and RCP8.5 scenarios, respectively. The relative decrease for WRF/Chem is much higher as compared to CESM_NCSU for CCN, however, both WRF/Chem and CESM_NCSU show that larger decrease in domainwide CCN under the RCP4.5 scenario as compared to the RCP8.5 scenario. Both models predict decreases in cloud droplet number concentration (CDNC), with smaller decreases by WRF/Chem than by CESM_NCSU, which is consistent with the smaller decreases in CCN by WRF/Chem. The spatial distributions for cloud fraction (CF) simulated by WRF/Chem and CESM_NCSU are different for current and future year simulations. As shown in the previous section, CESM_NCSU does not simulate the spatial distribution of CF well (note the difference in scales for WRF/Chem and CESM_NCSU simulations in Figure S7). CESM_NCSU predicts larger domain-average changes and different spatial distributions of such changes in CF than WRF/Chem for both scenarios. Under the RCP8.5 scenario, CESM_NCSU gives mainly increases in CF over southern and eastern U.S. due to the influence of clouds from the ocean from the eastern part of the domain, whereas WRF/Chem predicts increases in CF over smaller areas in southeastern U.S. Both models predict decreases in cloud optical thickness (COT) but with different magnitudes and spatial distributions of such decreases under both scenarios, consistent with the spatial distributions of changes in CDNC. Large differences exist in

predicted changes in cloud water path (CWP) between the two models in terms of magnitudes, signs, and spatial distributions due to the use of different cloud schemes. CESM-NCSU predicts decreases in CWP over most of the domain with domain-average decreases of 5.6 and 5.7 g m⁻² under the RCP4.5 and 8.5 scenarios, respectively. By contrast, WRF/Chem predicts increases in CWP over most of the domain with domain-average decreases of 12.9 and 17.8 g m⁻² under the RCP4.5 and 8.5 scenarios, respectively. Despite differences in spatial distributions of changes in shortwave cloud forcing (SWCF), both models predict decreases in SWCF over most of the domain with similar magnitudes of domain average decreases

5. Summary and Conclusions

WRF/Chem has been applied to project future climate and air quality under RCP4.5 and RCP8.5 scenarios. The future emissions of most gaseous species are projected to decrease to a larger extent under the RCP4.5 scenario as compared to the RCP8.5 scenario. While the SO₄²⁻ emissions will be reduced to a larger extent under the RCP4.5 scenario, however, the emissions of other species such as EC and unspciated primary PM_{2.5} will be reduced to a larger extent under the RCP8.5 scenario. Compared to the RCP4.5 scenario, the RCP8.5 scenario has a larger reduction of total primary PM emissions, with smaller reduction in the SO₄²⁻ emissions but larger reduction in the emissions of other species such as EC and unspciated primary PM_{2.5}. These changes in emissions affect not only air quality but also meteorological and climate trends under both scenarios through feedbacks of chemistry to the climate system. The meteorological and chemical feedbacks are also dependent on the resolution of the RCP emissions. In this case, the RCP emissions have a relatively coarse resolution of 0.5° × 0.5°.

In general, WRF/Chem predicts a domain-average increase of ~ 2°C for T2 and ~0.9 g kg⁻¹ for Q2 under both scenarios with a slightly larger increase in T2 for RCP4.5 than RCP8.5

and a slightly larger increase in Q2 for RCP8.5 than RCP4.5. WRF/Chem predicts slight increases in WS10 and precipitation over most of the domain under both scenarios. It predicts increases in PBLH and radiation variables such as SWDOWN, GLW, and OLR over most of the domain, which result from the decrease in total cloud amounts under both scenarios. Even though the moisture content in the atmosphere generally increases in the future, most cloud variables such as CDNC, CWP, and COT show decreases in the future scenarios over continental U.S. due to the decrease in CCN.

Under both scenarios, while the mixing ratios of SO₂ and NO_x are projected to decrease over the whole domain, those of NH₃ are projected to increase over the whole domain. O₃ mixing ratios decrease over most parts of the U.S. for RCP4.5 with a domain-average decrease of ~ 2 ppb, except for major urban cities in CA and northeastern U.S. where O₃ mixing ratios will increase. However, for RCP8.5, O₃ mixing ratios show a domain-average increase of ~ 3 ppb over all parts of the U.S. This is mainly caused by higher increase in the GHG background concentrations and BVOC emissions for future RCP8.5 scenarios compared to RCP4.5, higher mixing ratios of O₃ in the chemical ICONs/BCONs from CESM_NCSU, as well as the dis-benefit of NO_x emission reductions and reduced NO_x titration over areas with VOC-limited O₃ chemistry.

Sensitivity simulations are carried out to assess the impacts of different factors on the differences in the projected future O₃ mixing ratios between the RCP4.5 and RCP8.5 simulations. The results under the RCP8.5 scenario show that the contributions of the increases in the mixing ratios of individual GHGs to the increase in O₃ mixing ratios are relatively small but their cumulative increases contribute to about 1 ppb increase of O₃ mixing ratios in the future. Compared to RCP4.5, the largest contributor to the increase in O₃ mixing ratios under the

RCP8.5 is CH₄ which is a GHG and also a precursor for O₃ through the CH₄+OH chemistry. Other influential factors include the BCONs from CESM_NCSU under the RCP8.5 scenario, the cumulative impact of the background GHGs, and the differences in NO_x emissions between RCP8.5 and RCP4.5. In this study, the background GHG concentrations used are a single value. For a more accurate representation of the effects of GHGs on O₃ in the future GHG emissions should be vary spatially and temporally.

The mixing ratios of anthropogenic VOCs are projected to decrease under both scenarios with larger domain average decreases under the RCP8.5 scenario. Those of biogenic VOCs will increase resulted from increased T2 and SWDOWN, leading to increases in TVOCs over some areas such as southeastern U.S under the RCP4.5 scenario and southern Canada and northern Mexico under both scenarios. The changes in TSOA correspond to changes in TVOCs. PM_{2.5} concentrations are projected to decrease to a larger extent in the future for RCP8.5 as compared to RCP8.5. However, CDNC decreases to a larger extent in the future for RCP4.5 as compared to RCP8.5, due to the larger decreases in soluble PM_{2.5} species that can form cloud droplets (e.g., SO₄²⁻, NH₄⁺ and NO₃⁻) and smaller decreases in insoluble PM_{2.5} species (e.g., POA and EC) for RCP4.5 as compared to RCP8.5.

The projected future climate and air quality by WRF and WRF/Chem show different spatial patterns under the RCP8.5 scenario. While both simulations predict a domain-average increase of ~ 2°C for T2, WRF, however, predicts higher T2 for RCP8.5 compared to RCP4.5. In addition, the spatial pattern of T2 from the WRF RCP8.5 simulation is more similar to that from literature (e.g., from *Trail et al.* [2013]). The inclusion of precursor emissions, the dynamic gas and aerosol chemistry, and chemistry feedbacks to climate system in WRF/Chem changes the spatial pattern for T2. Precipitation generally increases for WRF RCP8.5, WRF/Chem RCP4.5

and WRF/Chem RCP8.5 simulations with similar spatial patterns but with the largest increases for WRF/Chem RCP8.5, likely due to the effects of the increase in SWDOWN. At the same time, water vapor content from WRF/Chem RCP8.5 simulation decreases in the future. As shown in this study, there are differences in the spatial patterns of surface meteorological predictions from WRF/Chem under different RCP scenarios as well as different models (WRF and WRF/Chem) with prescribed and dynamic gas and aerosol chemistry under the same RCP scenario.

The trends in most variables between future and current decade scenarios for WRF/Chem and CESM_NCSU are generally consistent. For example, both WRF/Chem and CESM_NCSU predict increases in domain-average SWDOWN. However, the magnitudes of increase are larger for CESM_NCSU under both RCP4.5 and RCP8.5 scenarios compared to WRF/Chem, which may be attributed to much higher O₃ column from CESM_NCSU compared to WRF/Chem. In addition, for both WRF/Chem and CESM_NCSU, future O₃ mixing ratios are projected to decrease under the RCP4.5 scenario but increase under the RCP8.5 scenario despite a non-linearity in the O₃ chemistry, and differences in current and future mixing ratios of NO_x and VOCs between WRF/Chem and CESM_NCSU. However, differences exist in the magnitudes and spatial distributions of the projected changes. In particular, WRF/Chem simulations show stronger spatial variability in projected changes for most variables such as precipitation, SWDOWN, CF, CO, O₃, and PM_{2.5}. In addition, both models project changes with different signs for WS10 and CWP under both scenarios, reflecting influences of different physics options used in both models. Further analysis of the differences in the physics, dynamics and chemistry modules between CESM and WRF/Chem is needed to understand the differences in the results.

The above results have important policy implications. The opposite changes in future O₃ mixing ratios predicted under the two RCP scenarios illustrate the benefits of carbon policy with more stringent emission controls under the RCP4.5 scenario than under the RCP8.5 scenarios and the importance of consideration of the O₃ photochemical chemical regimes in designing emission control strategies at urban and local scales. The sensitivity results on the relative importance of several possible contributing factors to such an opposite variation trends in future O₃ predictions can guide the development of emission control strategies and the determination of their priorities. In particular, they support that the controlling of CH₄ emissions is a win-win strategy to reduce O₃ for the U.S. The differences in projected future climate between WRF/Chem and WRF under both scenarios demonstrate the impacts of emissions and chemistry on the climate system through various complex chemistry feedbacks that occur in the real atmosphere, which should be considered for policy-making. While the similarities between projections for most variables by WRF/Chem and CESM support their validity to provide scientific results in support of air pollution control and adverse climate change mitigation, their differences in projected changes in some variables show the sensitivity of the projections to different models at different grid resolutions as well as the benefits in capturing local scale variations in projected changes using high resolution RCMs such as WRF/Chem.

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Table 1. RCP Scenarios modeling studies involving RCMs downscaled from GCMs over North America.

Reference	RCP scenarios	Domain	GCM	RCM	Periods	Focus
Wang and Kotamarthi (2015)	4.5 and 8.5	North America	CCSM4 ¹ (0.9° x 1.25°)	WRF ² v3.3.1 (12km)	Current: 1995 – 2004 Future: 2045 – 2054, and 2085 – 2094	To explore changes in mean and extreme precipitation in the mid and late 21 st century
Gao et al. (2013)	4.5 and 8.5	North America	CESM ³ v1.0/ CAM-Chem ⁴ (0.9° x 1.25°)	WRFv3.2.1 CMAQv5.0 (12km)	Historical: 1850 – 2005 (global) Future: 2005 – 2100 (global) Current: 2001 – 2004 (regional) Future : 2057 – 2059 (regional)	To evaluate the climate impact on ozone concentrations in the 2050s.
Gao et al. (2012)	8.5	Eastern U.S.	CESM v1.0 (0.9° x 1.25°)	WRFv3.2.1 (4km)	Historical: 1850 – 2005 Future: 2005 – 2100	To assess projected changes of extreme weather events
Trail et al. (2013)	4.5	North America	GISS ⁵ ModelE2 (2° x 2.5°)	WRFv3.4 (36km)	Current: 2006 – 2010 Future: 2048 – 2052	To analyze changes in meteorological fields which might affect air quality
Sun et al. (2015)	8.5	Continental U.S.	CESM (0.9° x 1.25°)	WRFv3.2.1 (12km) CMAQv5.0 (12km)	Current: 2002 – 2004 Future: 2057 – 2059	To estimate future PM _{2.5} and ozone-related mortality

¹ Community Climate System Model version 4

² Weather, Research and Forecasting Model

³ Community Earth System Model

⁴ Community Atmosphere Model with chemistry

⁵ Goddard Institute for Space Sciences

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Base RCP8.5

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(ii) RCP8.5 with RCP4.5 CH₄ levels and RCP4.5 NO_x Emissions

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(vi) RCP8.5 with RCP4.5 CH₄ levels and CO₂ levels

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