Changes in Future Air Quality, Deposition, and Aerosol-Cloud Interactions under Future Climate and Emission Scenarios

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Key Points

Global air quality is projected to degrade by the Mid-21st century following the A1B scenario

Treatment of aerosol-cloud interactions results in more moderate future warming

Aerosol-cloud interaction studies should use Earth system models with an ocean model

Abstract

The prospect of global climate change will have wide scale impacts, such as ecological stress and human health hazards. One aspect of concern is future changes in air quality that will result from changes in both meteorological forcing and air pollutant emissions. In this study, the GU-WRF/Chem model is employed to simulate the impact of changing climate and emissions following the IPCC AR4 SRES A1B scenario. An average of 4 future years (2020, 2030, 2040, and 2050) is compared against an average of 2 current years (2001 and 2010). Under this scenario, by the Mid-21st century global air quality is projected to degrade with a global average increase of 2.5 ppb in the maximum 8-hr O₃ level and of 0.3 $\mu\text{g m}^{-3}$ in 24-hr average PM_{2.5}.

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However, PM_{2.5} changes are more regional due to regional variations in primary aerosol emissions and emissions of gaseous precursor for secondary PM_{2.5}. Increasing NO_x emissions in this scenario combines with a wetter climate elevating levels of OH, HO₂, H₂O₂, and the nitrate radical and increasing the atmosphere's near surface oxidation state. This differs from findings under the RCP scenarios that experience declines in OH from reduced NO_x emissions, stratospheric recovery of O₃, and increases in CH₄ and VOCs. Increasing NO_x and O₃ levels enhances the nitrogen and O₃ deposition, indicating potentially enhanced crop damage and ecosystem stress under this scenario. The enhanced global aerosol level results in enhancements in aerosol optical depth, cloud droplet number concentration, and cloud optical thickness. This leads to dimming at the Earth's surface with a global average reduction in shortwave radiation of 1.2 W m⁻². This enhanced dimming leads to a more moderate warming trend and different trends in radiation than those found in NCAR's CCSM simulation, which does not include the advanced chemistry and aerosol treatment of GU-WRF/Chem and cannot simulate the impacts of changing climate and emissions with the same level of detailed treatments. This study indicates that effective climate mitigation and emission control strategies are needed to prevent future health impact and ecosystem stress. Further, studies that are used to develop these strategies should use fully coupled models with sophisticated chemical and aerosol-interaction treatments that can provide a more realistic representation of the atmosphere.

KEYWORDS: global climate and emissions change, future air quality, aerosol direct effect, aerosol indirect effects, online-coupled model, GU_WRF/Chem

1. Introduction

As the global population increases and industrialization grows in a changing climate, it is critical that governmental agencies account for changes in global air quality when forming

successful climate change and air pollution mitigation strategies. A warmer and more humid climate with an increased abundance of atmospheric CO₂ is generally expected in the future and this has important ramifications for global air quality (Jacob and Winner, 2009; Jacobson and Streets, 2009; Fiore et al., 2012). In addition, several studies have indicated that there is a higher probability of heat waves and air pollution episodes in the future due to increased stagnation. This results from a poleward shift in storm tracks and a general decrease in the frequency of mid-latitude cyclones (Mickley et al., 2004; Jacob and Winner, 2009; Young, 2009; Tai et al., 2012), which is consistent with the decreases in the frequency of mid-latitude cyclones observed during the 1980-2006 period (Leibensperger et al., 2008). The simulated impact of climate change on vertical transport is uncertain due to differences in model parameterizations (Murazaki and Hess, 2006; Jacob and Winner, 2009).

The warmer and wetter climate is expected to reduce the background ozone (O₃) level in regions with low NO_x levels, while increasing O₃ level in more polluted regions (Liao et al., 2006; Murazaki and Hess, 2006; Jacob and Winner 2009; Fiore et al., 2012; Doherty et al., 2013). The decreased background concentrations are the result of increased water vapor which reduces the lifetime of O₃ by removing O¹D to form the hydroxyl radical (OH), which in turn can destroy O₃ through HO_x reactions (Doherty et al., 2013). However, this general reduction in O₃ is compensated by increases in troposphere/stratosphere exchange due to strengthening of the Brewer-Dobson circulation expected in a warmer, CO₂ rich climate (Buchart et al., 2010; Fiore et al., 2012). The O₃ level is increased in polluted regions due to higher temperatures leading to higher O₃ formation rates, increases in emissions of biogenic VOCs, and increased decomposition of peroxyacetyl nitrate (PAN), as well as reduced ventilation from source regions (Liao et al., 2006;). In addition to changes in mean O₃, there is a tendency for increases in the 95th percentile O₃ (Languer et al., 2012), even in cases where the mean O₃ level decreases

(Languer et al., 2012). Similarly, the impact of climate change alone results in over half the eastern U.S. exceeding a maximum 8-hr O₃ level of 75 ppb assuming emissions remain the same (Rieder et al., 2015). This indicates increases in the severity and duration of air pollution episodes which have been estimated to increase in duration from 2 to 3-4 days with increased pollution levels of 5-10% in the U.S. (Mickley et al., 2004).

The impact of a changing climate on aerosols is less certain than that of O₃ because it is comprised of many different species that respond differently to perturbations in climate and emissions. The impact of future wet deposition is uncertain as it has been shown to both increase in the future resulting in decreases in PM_{2.5} (Liao et al., 2006) and also potentially decrease globally due to reductions in annual large scale precipitation over land and seasonal changes in precipitation (Fang et al., 2011). Generally, precipitation changes are variable between models and the impacts are more regional, as wet deposition of soluble aerosols is not strongly dependent on global changes in precipitation (Fiore et al., 2012). PM_{2.5} is also highly correlated with meteorological phenomena, such as the passage of cold fronts, but the projected increased stagnation in the future is not expected to induce a climate change penalty of more than 0.5 µg m⁻³ (Tai et al., 2012).

Changes in climate and emissions are also expected to affect PM_{2.5} speciation. The warmer and wetter climate is generally more oxidant rich which results in greater conversion of SO₂ to sulfate (Ungar et al., 2006; Jacob and Winner, 2009). This increased oxidation capacity along with potential increases in biogenic volatile organic compounds (VOCs) will lead to greater formation of both biogenic and anthropogenic secondary organic aerosol (SOA) (Tagaris et al., 2007; Heald et al., 2008). However, the increases in temperature may lead to reduced nitrate in the particulate phase (Jacob and Winner, 2009). These climate-induced changes in future aerosol levels can be either enhanced or offset by changes in future emissions depending

on the emission projection. Under the Intergovernmental Panel on Climate Change (IPCC) Fourth Assessment Report (AR4) Special Report on Emissions Scenarios (SRES) A1B scenario, the decreases by 2100 in emissions of sulfate, nitrate, and ammonium precursors are larger than those of primary organic matter (POM). This coupled with an increase in SOA may lead to organic matter (OM) as a dominant PM_{2.5} species (Tagaris et al., 2007). It is also possible in certain regions that declining SO₂ emissions and increasing NH₃ emissions may lead to nitrate formation that is equivalent to current PM_{2.5} levels (Fiore et al., 2012). This changing speciation over time may lead to variation in climate forcing. The work of Levy et al. (2008) showed that in 2030 following the SRES A1B scenario the climate is only weakly dependent on short lived climate forcers (sulfate, OM, black carbon (BC), O₃) due to compensating effects, but by 2100 enhanced BC emissions of 100% and a roughly 60% decrease in SO₂ emissions will contribute 1.5-2.0°C to global warming. This is a result of an enhanced warming effect from BC and a reduced cooling effect from sulfate (Levy et al., 2008). There may also be potentially large feedbacks to natural emissions of PM_{2.5} including sea-salt, mineral dust, and wildfires (Jacob and Winner, 2009; Fiore et al., 2012).

Changes in future air quality will also have consequences on ecosystem health. O₃ deposition inhibits photosynthesis, damages plant cells, and has been shown to reduce both net primary productivity and carbon sequestration (Felzer et al., 2004). O₃ deposition is expected to increase due to an increase in the leaf area of broadleaf forests (Wu et al., 2012). The impact of O₃ on vegetation is also dependent upon emission scenario. Large increases in O₃ are to be expected under most of the IPCC AR4 SRES scenarios, making it difficult to achieve standards for crop damage and leading to speculation of possible agricultural concerns in the future (Prather et al., 2003). In contrast, the Representative Concentration Pathway (RCP) scenarios, from the IPCC Fifth Assessment Report (AR5), produce surface level O₃ improvements in all

but the RCP8.5 scenario compared to present day (Lamarque et al., 2011, Young et al., 2013; Kim et al., 2015). However, under both types of scenarios nitrogen deposition is expected to increase (Dentener et al., 2006; Lamarque et al., 2011) due to increases in reactive nitrogen (NO_y) and NH_3 under the SRES scenarios and from increases in NH_3 that compensate losses of NO_y under the RCP scenarios. This may result in ecosystem changes, as 10.1% of natural terrestrial ecosystems are already exposed to nitrogen deposition higher than the critical load estimated to impact sensitive ecosystems (Dentener et al., 2006).

In this work, the Global-through-Urban Weather Research and Forecasting Model with Chemistry (GU-WRF/Chem) is employed to study the impact of climate change and emissions on global air quality under the IPCC AR4 SRES A1B emission scenario. The total impacts, as well as the separate contributions from changing emissions and climate, are investigated. The changes in ambient concentrations, deposition, and the impacts of chemistry on cloud and radiative parameters are discussed. The benefit of using this model for climate change studies is that it can represent the complex feedbacks between chemistry and meteorology and it employs the same level of complexity as regional air quality models, compared to the simplified treatments used by most contemporary global climate and chemical transport models. However, this complexity comes at the cost of higher computational resource requirements.

2. Model Configuration, Evaluation Protocol, and Observational Datasets

2.1 Model Configuration and Inputs

GU-WRF/Chem (Zhang et al., 2012b) is an online-coupled model capable of simulating the interactions and feedbacks between the chemical and physical processes in the atmosphere. It is a hybrid model generated by replacing the WRF model in the National Oceanic and

Atmospheric Association (NOAA)'s mesoscale WRF/Chem with the National Center for Atmospheric Research (NCAR)'s global WRF (GWRf) and then improving several model treatments for their applications on hemispheric to global scales. These improvements include online dust and BVOC emissions, gas-phase chemistry, a photolytic rate scheme, aerosol chemistry and microphysics, and aerosol-cloud interactions. Like GWRf and mesoscale WRF/Chem, GU-WRF/Chem has many different parameterizations for various atmospheric processes occurring on spatial and temporal scales smaller than those that can be resolved explicitly by the model. Table 1 contains a list of the chemistry and physics options used and the configuration of the simulations. A more detailed description of the model aspects relevant to this study can be found in the Supplementary Material and Zhang et al. (2012b) and references therein.

GU-WRF/Chem simulations are conducted for a period of six years, with two current years (2001 and 2010) and four future years (2020, 2030, 2040, and 2050). This limited subset of years is used because each year represents a different stage of climate forcing and emissions changes, thus some of the decade to decade variability can be captured within the computational constraints. The initial meteorology and chemical conditions for O₃, N₂O, and CH₄ come from the Community Climate System Model version 3 (CCSM3) SRESA1B data set. Other chemical initial conditions for gas species with moderate lifetimes not found in the CCSM dataset (e.g., SO₂, PAN, and H₂O₂) are generated from the Goddard Earth Observing System Model with Chemistry v7-04-12-Run0 (http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_v7-04-12). Short-lived radical gas species (e.g., OH and NO₃) are initialized with clean conditions and aerosol initial conditions are set with the default values from mesoscale WRF/Chem. A one-month spin up period is then used to generate realistic ambient values for the short-lived species

and aerosols. An additional two year simulation is conducted for the years 2001 and 2010 using meteorological initial conditions from the National Centers for Environmental Prediction Final Operational Reanalysis (NCEP-FNL), without data assimilation, so that the model can be evaluated to ensure that GU-WRF/Chem can reproduce the current atmosphere. Lastly, a sensitivity simulation of the year 2050 is carried out using the same configuration as the base 2050 simulation but with the 2001 emissions (2050_NPE). This sensitivity experiment is compared against both the 2001 simulation to investigate the impact that climate change alone will have on future air quality and the 2050 simulation with projected emissions to examine the impact of changing emissions on the future climate.

The meteorological initial conditions as well as the sea surface temperatures (SSTs) for the non-evaluation simulations are 10 year averages of the CCSM3 data that are centered about each GU-WRF/Chem simulation year (e.g., the year 2030 is initialized with the average of 2025-2034 from the CCSM3 data), with the exception of 2001 which is initialized with the 5 year period 2000-2004 since the CCSM3 simulation started at the year 2000. The meteorology and the SSTs are reinitialized every month. This is especially important as the model uses static SSTs, which need to be reinitialized in order for the annual variation in SSTs to be represented. The chemical species in the model are reinitialized every 3 months. This allows for a mostly continuous chemistry simulation but prevents excess buildup of pollutants near the top of the model and near the polar singularities that can occur due to the lack of a positive-definite advection scheme in the model. This type of reinitialization technique is standard practice in online-coupled regional air quality models such as those that took part in the Phase 2 of the Air Quality Model Evaluation International Initiative including an updated version of mesoscale WRF/Chem on which GU-WRF/Chem is based that employed reinitialization in 2-day intervals (Yahya et al., 2014; 2015).

The base anthropogenic emission inventory for the year 2001 has been provided in Table 2 of Zhang et al. (2012b). The natural emissions of biogenic volatile organic compounds (BVOCs), sea-salt, and dust are calculated online. The BVOCs are generated from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2 (Guenther et al., 2006), sea-salt is calculated using the formulation of Gong et al. (1997) with a correction for the smaller sea-salt particles from O'Dowd et al. (1997), and dust is generated from the scheme of Shaw (2008) with the modifications of Zhang et al. (2012b). Future emissions projections are based on the IPCC AR4 A1B scenario, which is a scenario of increasing economic and population growth that peaks in the mid-21st century and declines thereafter, with a focus on balanced energy sources (IPCC, 2001). The anthropogenic emissions projections for 2030 and 2050 are generated from region wide growth factors for 17 world regions developed by Argonne National Laboratory (ANL) using the Integrated Model to Assess the Global Environment (IMAGE) following the approach of Nakicenovic et al. (2000) and RIVM (2001), with the exception of the growth factors for primary organic carbon (OC) and black carbon (BC), which were derived following the approach of Streets et al. (2004). Tables containing the growth factors can be found in the supplementary material (Tables S1 and S2). The emissions for 2010 and 2020 are a linear interpolation between the base emissions inventory and the projected 2030 emissions and 2040 emissions are a linear interpolation between the 2030 and 2050 emissions. The total global emissions of all major anthropogenic species for each simulated decade are listed Table S3 in the supplementary material.

2.2 Evaluation Protocol

The model's performance with regards to meteorology and chemistry is evaluated for current years using a combination of satellite, global surface networks, and reanalysis data. The

210 evaluation protocols are similar to those of Zhang et al. (2010, 2012a, 2012b) and include
211 statistical evaluations and trend comparisons with observations for current years and a
212 comparison of the climate trends in the CCSM3 data used to initialize the simulations for future
213 years. The performance statistics include the correlation coefficient (Corr), the mean bias (MB),
214 the normalized mean bias (NMB) and the normalized mean error (NME) (Yu et al., 2006). The
215 satellite data used for evaluation include the column abundance of CO from the Measurements
216 Of Pollution In The Troposphere (MOPITT) , the column abundance of NO₂ from the Global
217 Ozone Monitoring Experiment (GOME), the column abundance of NO₂ and SO₂ from the
218 SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY),
219 the column tropospheric ozone residual (TOR) from the Total Ozone Mapping Spectrometer/ the
220 Solar Backscatter UltraViolet (TOMS/SBUV) and the Ozone Monitoring Instrument/ Microwave
221 Limb Sounder (OMI/MLS), downwelling long-wave radiation flux at the Earth's surface
222 (GLW), downwelling short-wave radiation flux at the Earth's surface (SWDOWN), net
223 downwelling short-wave radiation flux at the Earth's surface (GSW), and outgoing long-wave
224 radiation at the top of the atmosphere (OLR) from the Clouds and the Earth's Radiant Energy
225 System/Energy Balanced and Filled (CERES/EBAF), and the cloud condensation nuclei at a
226 supersaturation of 0.5% (CCN), the aerosol optical depth (AOD), the cloud optical thickness
227 (COT), the cloud fraction (CF), the cloud liquid water path (LWP), and the precipitable water
228 vapor (PWV) from the Moderate Resolution Imaging Spectroradiometer (MODIS). Additional
229 satellite datasets for the evaluation include remote sensing estimates of the cloud droplet number
230 concentration (CDNC) in warm clouds from Bennartz (2007) (BE07) and LWP from O'Dell et
231 al. (2008) (OD08). The data from global surface networks include GLW and SWDOWN from
232 the Baseline Surface Radiation Network (BSRN) and the 2-m temperature (T2), the 2-m water
233 vapor mixing ratio (Q2), the 10-m wind speed (WS10), surface pressure (PSFC), and

precipitation rate (PR) from the National Climatic Data Center (NCDC). Reanalysis data used include PR from the Global Precipitation Climatology Project (GPCP) and OLR from the National Atmospheric and Oceanic Administration/ Climate Diagnostic Center (NOAA/CDC).

3. Model Evaluation

3.1 Operational Evaluation

Table 2 summarizes the model performance statistics for the various global networks listed above. Figure 1 shows a comparison of the zonal mean profiles of GU-WRF/Chem and the satellite/reanalysis datasets for the annual averaged current year period of 2001 and 2010 (AOC), while Supplementary Figure S1 shows spatial distributions of simulated meteorological variables overlaid with observations from the surface networks. OLR and GLW are both well simulated in GU-WRF/Chem with correlations varying between 0.94-0.99 to the observations and NMBs within $\pm 3\%$.

Similar to most global climate/air quality models (Kay et al., 2012; Lauer and Hamilton, 2013), cloud parameters are not as well simulated in GU-WRF/Chem. The cloud fraction in GU-WRF/Chem is overpredicted by ~20% domain wide but the overprediction largely occurs over the oceans and at the higher latitudes while the interior of the continents are either close to the observations or slightly underpredicted. Despite the overprediction in CF; COT and LWP are largely underpredicted (an NMB of -82.6% for COT, and NMBs for LWP are -81.0% against MODIS, and -58.9% against OD08). LWP in GU-WRF/Chem only represents resolved clouds and as a result subgrid scale cloud water is missing compared to the observations, which most likely accounts for the large underprediction. Additionally, the Purdue Lin cloud microphysics scheme is fairly outdated and simplified compared to many of the newer microphysics schemes

such as the Morrison double moment scheme (Morrison et al., 2005) in the WRF model and future work for coupling these microphysics schemes with chemistry and representing subgrid scale cloud water may lead to better agreement with the observations. COT is underpredicted in part because of the underprediction in LWP but also because only cloud water and ice are part of the calculation and additional optical thickness from rain, snow and graupel is not represented in the model. The underprediction is especially large in the polar regions as shown in Figure 1. These underpredictions in the polar regions are likely the result of satellite errors due to low solar zenith angles (Seethala and Horváth, 2010) and interference from snow cover (Kay et al., 2012). However, despite these model weaknesses, the performance of GU-WRF/Chem is comparable to that of mesoscale WRF and MM5/CMAQ (Zhang et al., 2012b).

Both SWDOWN and GSW are reasonably well simulated by GU-WRF/Chem with NMBs of ~20-26% and correlations of 0.89 and 0.92, respectively. The bulk of the overprediction in SWDOWN and GSW occurs in the higher and mid-latitudes. The overpredictions in both SWDOWN and GSW are the result of underpredictions in cloud parameters such as COT, especially over the continents and at higher latitudes. Additionally, based on the zonal mean plots in Figure 1, GSW has a larger overprediction in the Arctic compared to SWDOWN, indicating that the albedo in GU-WRF/Chem may be underpredicted, especially in polar regions.

The GU-WRF/Chem meteorological predictions are also in good or reasonably good agreement with observations. The PSFC is only slightly underpredicted (-0.4%) with a fairly high correlation of 0.65. Both PWV and Q2 are very well simulated with NMBs less than or equal to 2% and correlations of greater than or equal to 0.89. GU-WRF/Chem predicts a wetter atmosphere in the higher latitudes and drier tropics than that is observed. In both cases this may

be the result of the warm bias from the model as higher temperatures in the high latitudes would lead to greater evaporation, while the warmer temperatures in the tropics could result in a depletion of soil moisture and thus a drier atmosphere. The model does overpredict the surface temperature by 1.4°C with the overprediction focused on mostly at higher latitudes and the interiors of continents. These overpredictions are caused by the overpredictions in SWDOWN and GSW. Despite the fairly high bias, the temperature is well correlated with the observations (0.94), indicating that the model can reproduce the spatial pattern in temperature rather well. The WS10 in GU-WRF/Chem tends to be underpredicted over the continents and overpredicted over the oceans with a domain average underprediction of -24.9%. The PRs from NCDC and GPCP are underpredicted by -6.1% and -3.4% respectively. The comparison of zonal mean PR of GU-WRF/Chem against GPCP observations (Figure 1) shows that GU-WRF/Chem generally captures the latitudinal variation in precipitation but overpredicts tropical convective precipitation and underestimates mid-latitude precipitation.

The model AOD and CCN are both overpredicted (NMBs of 96.3% and 18.9%). The overprediction in AOD is the largest in biomass burning regions including South America, Africa, and Indonesia, as shown in Figure S2, indicating that either BC or OM emissions may be overestimated or that the strong convection in this region is leading to excess lofting of fine particles. Underpredictions in precipitation may also be responsible for the overpredicted AOD and CCN as wet removal of particles will be underpredicted. The MODIS CCN data is only available in marine regions so the overprediction is largely caused by excess continental outflows resulting from the underpredictions in precipitation and also potential overpredictions in sea salt from overpredicted marine wind speeds. The model underpredicts warm cloud CDNC by 63.5% most likely due to the underpredicted aerosol activation from lack of cloud formation as evidenced by underpredicted LWP. The aerosol activation may be underpredicted in GU-

WRF/Chem because the Abdul Razzak-Ghan activation scheme does not account for the adsorptive activation of hydrophobic aerosol, which has been shown to increase aerosol activation and CDNC, and generally activates fewer aerosols than other available aerosol activation parameterizations (Gantt et al., 2014).

The column abundances of CO and NO₂ are both overpredicted (NMBs of 54.7% and 74.8%). The overpredictions in both species are tied to overestimations of NO_x and CO emissions. As discussed in Zhang et al. (2012b), the emissions of CO and NO_x in the GU-WRF/Chem inventory are larger than most other available emission inventories and in this study their emissions are not scaled down to match the other inventories. The overestimated CO appears again to come largely from the biomass burning regions in the tropics, indicating that biomass burning emissions may be generally overestimated. The zonal mean profile of column NO₂ in Figure 1 reveals that while the column NO₂ is overestimated it follows the same latitudinal variability as the observations. The TOR also appears to follow the same pattern as the observations but is overestimated by 24.3% most likely due to the overestimated NO₂ column. The model also captures the spatial variability of column SO₂ (Figure 1) but it is largely underestimated (with an NMB of 81.2%). This is most likely the result of a number of factors including the lack of emissions and pathways for the formation of SO₂ from volcanoes and dimethyl sulfide emitted from the oceans. There are also large biases in the satellite estimate of column SO₂ that may explain the large difference between the satellite retrieval and simulated values (Fioletov et al., 2013).

3.2 Climate Trend Comparison

GU-WRF/Chem predictions are compared against available data from climate projections of both CCSM3 that utilizes a similar SRES A1B scenario projection and also the Community

Earth System Model version 1.0 /Community Atmosphere Model version 5.1 (CESM1-CAM5) from the Coupled Model Intercomparison Project Phase 5 (CMIP5) that follows the RCP8.5 scenario. The RCP8.5 scenario was chosen as it is the RCP scenario that is the most comparable to the SRES A1B scenario. Figure 2 shows the trend in T2, Q2, GSW, OLR, PR, and sulfate between the average of the current year period (AOC) and the average of the future year period (AOF) for both CCSM3 and GU-WRF/Chem. Figure S3 also shows trend comparisons of meteorological, radiation, and cloud variables between the AOF and AOC periods for GU-WRF/Chem and CESM1-CAM5. The domain mean increase in temperature between the two SRES A1B models is similar with an increase of $\sim 1.0^{\circ}\text{C}$ for CCSM3 and 0.7°C for GU-WRF/Chem, but the RCP8.5 projection shows greater warming of $\sim 1.4^{\circ}\text{C}$. The SRES A1B models show similar patterns in warming and cooling with the exception of the Arctic where CCSM3 produces the strongest warming and GU-WRF/Chem either produces less warming or even cooling. The disagreement in this region likely contributes the most to the more modest warming trend in GU-WRF/Chem and is caused by increases in Arctic cloudiness that result in a decrease in GSW. The greater warming under the RCP8.5 scenario is the result of large increases in CH_4 emissions under this scenario and reductions in warming aerosol species that tend to increase future temperatures (Chalmers et al., 2012; Westervelt et al., 2015). Changes in Q2 between AOF and AOC predicted by both SRES A1B models are similar with global average increases of 0.4 g kg^{-1} for CCSM3 and 0.3 g kg^{-1} for GU-WRF/Chem. The smaller increase of GU-WRF/Chem is likely driven by the more moderate warming compared to CCSM3. The larger increase of 0.5 g kg^{-1} in the CESM1-CAM5 prediction is also likely linked to greater warming under the RCP8.5 scenario. This is also true for PWV between GU-WRF/Chem (e.g., global average increase of $\sim 1.1 \text{ mm}$) and CESM1-CAM5 (e.g., global average increase of $\sim 1.4 \text{ mm}$), although both models show increases across the almost the entire domain. Changes in PR

are also similar in all projections with increasing PR in the tropics, mid-latitudes and polar regions and drying in the subtropics. The global average changes are an increase of 0.05 mm day⁻¹ for CCSM3, 0.06 mm day⁻¹ for CESM1-CAM5, and 0.02 mm day⁻¹ for GU-WRF/Chem. The PR increase is likely less in GU-WRF/Chem since the atmosphere is slightly drier compared to CCSM3 and CESM1-CAM5.

In general, GU-WRF/Chem predicts a sometimes cloudier and more aerosol-rich future atmosphere, which will be discussed in more detail in subsequent sections. Processed cloud variables from CCSM3 are not available from the Earth System Grid repository for comparison against GU-WRF/Chem so cloud variable comparisons are limited to CESM1-CAM5. The global average change in CF is similar between both models with decreases of ~0.17% for CESM1-CAM5 and ~0.15% for GU-WRF/Chem. However, there is not much agreement in terms of spatial variability of the changes except for increasing cloudiness in the Arctic. This lack of consistency in spatial variability could be related to differences in model parameterizations that affect clouds but it is more likely that the discrepancies over land are related to large differences in radiative forcing due to opposing trends in aerosols and the discrepancies over the oceans are caused the use of static SSTs in GU-WRF/Chem compared to an online-coupled ocean model in CESM1-CAM5. There are large differences in the change in LWP path projected by GU-WRF/Chem (which increases by 1.41 g m⁻² on global average) and condensed water path (CWP = LWP + ice) predicted by CESM1-CAM5 (that increases by 0.04 g m⁻²). The global scale increase of LWP from GU-WRF/Chem reflects an enhanced moisture availability from not only a warmer climate but also higher aerosol levels that can increase LWP by increasing cloud lifetimes. The CWP from CESM1-CAM5 also increases due to enhanced moisture availability but over the continents, especially in industrial regions, it decreases due to

the decreasing aerosol levels under the RCP8.5 scenario that reverse the cloud lifetime effect (Westervelt et al., 2015).

The increases in aerosols in GU-WRF/Chem increase AOD globally by 0.03 on global average. Total AOD was not available from the processed CCSM3 output but a comparison against CESM1-CAM5 shows a contrasting decreasing trend in aerosols under the RCP8.5 scenario which causes AOD to decline on global average by 4.6×10^{-4} . GU-WRF/Chem predicts global mean decreases in GSW of 1.05 W m^{-2} and OLR of 0.72 W m^{-2} . This is because the greater aerosol and cloud water amounts reduce the amount of shortwave radiation reaching the surface, thus reducing GSW. The reduced GSW reduces the surface heating and leads to a smaller emission of longwave radiation and this combined with more longwave absorption by the clouds and greenhouse gases leads to a reduction in OLR. These changes are in contrast to the CCSM3 predicted global mean increases of 0.08 W m^{-2} in GSW and 0.32 W m^{-2} in OLR. SWDOWN is projected to decrease by 1.6 W m^{-2} on global average based on GU-WRF/Chem compared to a 0.6 W m^{-2} decrease from CESM1-CAM5. These changes largely mirror changes in LWP and CWP, indicating that global mean reductions in both models result from increased cloud albedos but the reduction is smaller in CESM1-CAM5 due to declines in aerosol levels under the RCP8.5 scenario, which increase SWDOWN over land through the aerosol direct and indirect effects. OLR increases to a stronger degree in the CESM1-CAM5 projection (e.g., 1.4 W m^{-3} on global average) due to the reduced cloudiness and warmer temperatures following the RCP8.5 scenario. However, despite the differences in the global average trends, there are some similarities in the spatial distributions (e.g., the decreasing OLR in the tropics and increasing OLR in the subtropics). The discrepancy in the trends between both SRES A1B projections could be the result of a number of factors. Examples of such factors include: (1) the simulation of additional aerosol species such as ammonium nitrate and additional aerosol chemistry treated in

GU-WRF/Chem that are not treated by this version of CCSM; (2) changes in carbonaceous aerosols in CCSM3 are based on changes in SO₂ emissions rather than having independent emission changes like those treated in GU-WRF/Chem; (3) sea-salt and dust are static between the current and future CCSM3 simulations but are dynamically evolving with climate in GU-WRF/Chem; (4) differences in cloud microphysics, radiation, and cumulus parameterizations; and (5) the use of a static prescribed O₃ field with a varying radiative forcing for future predictions in CCSM3 whereas GU-WRF/Chem allows O₃ and its subsequent feedback to radiative forcing to evolve with changes in meteorology and precursor emissions. Changes in aerosols including sea-salt and dust, as well as changes in O₃ from GU-WRF/Chem are discussed in detail in Section 4.

These differences in chemistry and also potential differences in emissions and the method for applying emissions changes may possibly explain the discrepancies in sulfate between the two SRES A1B models. Both models predict an increase in the global mean sulfate concentration, with CCSM3 predicting an increase of 0.04 µg m⁻³ compared to a larger increase of 0.19 µg m⁻³ by GU-WRF/Chem. However, there are several regional discrepancies including Africa, where GU-WRF/Chem predicts an increase of 0.5 to 2.0 µg m⁻³ in contrast to the 0.25 to 2.0 µg m⁻³ decreases predicted by CCSM3, and Europe, where GU-WRF/Chem predicts a decrease of 0.25 to 1.0 µg m⁻³ in contrast to the 0.25 to 1.0 µg m⁻³ increase predicted by CCSM3. These discrepancies are caused by differences in emissions since the SRES A1B emissions growth factors for GU-WRF/Chem emissions are derived from the IMAGE integrated assessment model rather than the Asian-Pacific Integrated Model (AIM) used for the CCSM3 projections. This type of divergence despite using the same story line occurs among the integrated assessment models (Nakicenovic et al., 2000).

4. Impact on Air Quality

4.1 Trace Gases

Figure S6 shows the response of trace gases including CO, NH₃, SO₂, NO₂, O₃, peroxyacetyl nitrate (PAN), OH, HO₂, H₂O₂, nitrate radical (NO₃g), N₂O₅, and HNO₃, to changing climate and emissions. The changes in CO, NH₃, SO₂, and NO₂ mirror the changes in their emissions and the global changes are comparable to changes reported in Jacobson and Streets (2009). A more detailed description of these changes is provided in the supplementary material.

Figure 3 shows the absolute changes in maximum 8-hr O₃ (O₃8hr), the average PM_{2.5} level, column cloud condensation nuclei concentration, column cloud droplet number concentration, cloud liquid water path, cloud optical thickness, aerosol optical depth, and incoming shortwave radiation between the AOF and AOC periods. The O₃8hr level increases by 2.5 ppb on global average with increases as large as 20.4 ppb in South Asia and East Asia. Similarly, annual average near surface O₃ increases by 2.2 ppb (11.7%) on global average and as much as 15.0 ppb in South Asia and East Asia. The global increase in O₃ level leads to global increases in O₃ deposition ($1.9 \times 10^{-2} \text{ g m}^{-2} \text{ month}^{-1}$ on average) which is discussed in more detail in the supplementary material. An exact comparison of these changes against previous studies is not possible due to differences in scenario, methodology, and simulated period. However, increase in O₃ during the AOF period are consistent with the 14% increase in near surface O₃ by 2030 following the SRES A1B scenario reported by Jacobson and Streets (2009). The O₃ enhancements in this work are less than the 13.7-18.0 ppb average increase reported by Liao et al., (2006) following the SRES A2 scenario by 2100 which is to be expected as A2 generally has higher pollutant emissions than A1B and warming is more intense by 2100 compare to 2020-2050. The global average change in O₃ from GU-WRF/Chem is also similar to 2.1 ppb increase

in surface O_3 following the RCP8.5 scenario in 2050 reported by Kim et al. (2015), although the reasons for O_3 increases under this scenario are more related to increased temperatures and increased CH_4 levels since NO_x emissions decrease under this scenario. The changes in O_3 in this work are significantly different from all other RCP scenarios where O_3 decreases as a result of stronger controls of pollutants and greenhouse gases (Lamarque et al., 2011; Young et al., 2013; Kim et al., 2015).

In order to better explain the increases in O_3 8hr, the changes in O_3 8hr are correlated with changes in both meteorological and chemical variables, and the correlation coefficients (R) are shown in Table S4. The primary cause of the increased O_3 8hr level is increases in NO_x emissions and consequently, the increases in the surface NO_2 level, which correlates with O_3 8hr by 0.57 and 0.65, respectively. The enhancements of the O_3 8hr level by total non- CH_4 VOCs (TNVOC) are less pronounced with a correlation of 0.26 against TNVOC emissions and 0.44 against the surface anthropogenic VOC level (AVOC). These correlation values do not necessarily indicate a strong influence from TNVOC as the emissions of TNVOC and NO_x are highly correlated with each other (R of 0.60). There is a mild contribution to the O_3 8hr level from increasing T2 (R of 0.20), which increases the reaction rates for O_3 formation. Other meteorological factors including Q2, planetary boundary layer height (PBLH), and the photolysis rate of NO_2 do not appear to contribute substantially to the increase in O_3 8hr with R values that are near zero or negative. The near zero correlation likely indicates that the impact of water vapor destruction on O_3 reported in many studies (Fiore et al., 2012 and references therein) is compensated by the increasing NO_x emissions in this scenario. However, such an impact is found in the 2050_NPE simulation discussed in Section 7.1. The change in the isoprene level also does not appear to be leading to enhanced O_3 formation as it has a negative correlation of -0.32 against the increases in O_3 8hr. However, this strong anti-correlation indicates that O_3 enhancements result in much greater

isoprene oxidation and removal, despite slight increases in biogenic emissions (e.g., ~10% increase in isoprene and total biogenic VOCs). The enhancement of isoprene in GU-WRF/Chem is greater than ~1% increase in biogenic emissions reported by Jacobson and Streets (2009) for 2030 and less than 22% increase reported by 2100 in Heald et al. (2008) following the SRES A1B scenario, indicating the changes are within the range of future possibilities following SRES A1B. The global PAN level increases by ~0.03 ppb on average. This occurs, despite the enhanced PAN decomposition from warmer climate, due mainly to the increases in NO_x emissions but also potentially from the increases in emissions of TNVOC precursors (i.e., acetaldehyde, isoprene, and aromatics such as toluene, xylene, and cresol). This is especially true in South Asia and East Asia where emissions of both NO_x and TNVOC increase, resulting in increasing PAN levels by 0.1 to 0.5 ppb.

The enhanced O₃ level in the future environment leads to greater formation of O¹D, 1.8×10⁻¹¹ ppt (8.6%) on global average, from O₃ photolysis. This increase in O¹D and its subsequent reaction with the greater water vapor level lead to greater formation of near surface OH, which increases on global average by 1.6×10⁻³ ppt (7.3%) with the greatest enhancements in the tropic and subtropic continents of 5.0×10⁻² to 0.4 ppt. The increases in O¹D and OH are small even though the enhanced O₃ level favors formation, because there is a decline in O₃ photolysis due to greater aerosol scattering and cloudiness in the future atmosphere that will be discussed in more detail in subsequent sections. This increase in OH differs from the decreases of 3.8% reported by Jacobson and Streets (2009), and also ensembles results using the RCP scenarios (Voulgarakis et al., 2013; Kim et al., 2015). The decreases following the RCP scenarios are potentially linked to large increases in CH₄ in the RCP8.5 scenario and decreased O₃ production in the remaining scenarios, as well as O₃ recovery in the stratosphere predicted under these scenarios (Voulgarakis et al., 2013; Kim et al., 2015). The discrepancy between GU-

WRF/Chem and the work of Jacobson and Streets (2009) could potentially be due to differences in the chemical mechanisms and parametrizations of physics for many atmospheric processes in both models.

The increase in O_3 and OH also lead to enhancements of the HO_2 level of ~ 0.3 ppt (4.8%) on global average but up to as much as 1.0 to 3.3 ppt in tropical regions. The enhancements of OH and HO_2 lead to a higher H_2O_2 level of 0.08 ppb (13.8%) with enhancements of 0.2 to 1.2 ppb over most continents. The increases in O_3 combined with the increased NO_2 levels lead to greater formation of NO_3g , which increases on global average by 0.6 ppt (33.3%). This shows that the impact of the increased NO_x emissions under the SRES A1B scenario is not only to increase the global O_3 level but to increase the global availability of the major atmospheric oxidants, thus increasing the atmosphere's near surface oxidation state. The increased NO_x emissions also increase the level of other nitrogen containing species such as N_2O_5 which increases by 0.9 ppt (48.7%) on global average. This, combined with the increased levels of NO_2 , NO_3g , and other oxidants, leads to greater formation of HNO_3 (0.3 ppb or 99.8% on global average), creating a bigger reservoir for nitrate aerosol formation. This increased HNO_3 level is comparable to the 76% increase reported by Jacobson and Streets (2009). The increase in NO_x emissions also increases nitrogen total deposition (TND) on global average by $1.4 \times 10^{-3} \text{ g m}^{-2} \text{ month}^{-1}$ due in large part to the increases in the atmospheric oxidized nitrogen species. Changes in nitrogen, BC, and mercury deposition are discussed in the supplementary material.

4.2 $PM_{2.5}$

The global $PM_{2.5}$ level increases on average by $0.3 \mu\text{g m}^{-3}$ (3.6%), slightly larger but comparable to the 1.6% increase predicted by Jacobson and Streets (2009). However, the changes are much more regional in nature compared to the O_3 8hr level. Table S5 shows the correlations of the changes in $PM_{2.5}$ compared to changes in meteorological variables, chemical

precursors, and emissions. Figure S7 also shows the impact of the changing climate and emissions on $PM_{2.5}$ and its components. The impact of changing climate on the overall $PM_{2.5}$ level appears to be minimal except in the cases of the WSP10 (R value of 0.12), which impacts the emissions of sea-salt, and Q2 (R of 0.28) which can lead to greater absorption of water into the aerosol. The impacts from changes in PBLH and precipitation are minimal with R values close to zero. However, this assessment only pertains to simulations in which both climate and emissions are changing as the impact of climate change alone has been shown to impact $PM_{2.5}$ in past studies (Jacob and Winner, 2009; Fiore et al., 2012) and will be demonstrated in the 2050_NPE simulation discussed in Section 6.1.

The global $PM_{2.5}$ level is influenced the most by changes in primary particle emissions of BC, POM, and other unspiciated inorganic $PM_{2.5}$ (OIN) (R values of 0.42, 0.40, and 0.56, respectively). The decreases in these primary particle emissions are the primary cause of the decline of 0.6 to 12.5 $\mu g m^{-3}$ in central and southern Africa and the decline of 0.6 to 1.2 $\mu g m^{-3}$ in East Asia. Increases in these emissions also contribute to the 0.3 to 2.1 $\mu g m^{-3}$ increase in Australia. The changes in $PM_{2.5}$ over the oceanic regions can be either decreases or increases and are partially controlled by the WSP10 driven changes in sea-salt emissions of sodium and chloride over the oceans (R of 0.36 and 0.26, respectively). However, there is a difference in not only the R of both sea-salt components but in the global change of sodium, which increases by 0.02 $\mu g m^{-3}$ on global average, and chloride which decreases by 0.14 $\mu g m^{-3}$. This decline in chloride largely occurs off the coast of most continents and is caused by enhancements in the levels of both SO_2 and HNO_3 in these coastal zones, resulting in much greater chloride displacement by sulfate and nitrate. The enhanced chloride displacement, from the increased HNO_3 level, increases the global average nitrate level by 0.2 $\mu g m^{-3}$ with increases as large as 2.9

$\mu\text{g m}^{-3}$ in these coastal zones. The replacement of chloride by sulfate and nitrate is one of the major contributors to the increasing global $\text{PM}_{2.5}$ level since both nitrate and sulfate have a higher molecular weight than chloride.

The impact from the secondary inorganic aerosol precursors is not as strong as the primary particle emissions. Changes in SO_2 emissions and the surface SO_2 level show the greatest impact (R of 0.20 and 0.31), followed by NO_x emissions and the surface HNO_3 level (R of 0.19 and 0.26), while changes in NH_3 emissions and the surface NH_3 level have the smallest impact (R of 0.14 and 0.12). Changes in SO_2 show a strong impact because increases in sulfate have a strong impact in South and Southeast Asia and declines in sulfate dominate the changes in $\text{PM}_{2.5}$ over Europe. The changes in SOA are negligible in most regions except South America and Indonesia, where SOA increases by as much as $1.2 \mu\text{g m}^{-3}$. This is a result of greater biogenic emissions and oxidant levels leading to greater oxidation of isoprene and terpene species that form SOA in these regions. A more detailed discussion of changes in secondary PM components is provided in the supplementary material.

5. Aerosol-Climate Interactions

As shown in Figure S8, the global average NUM level increases by $1.2 \times 10^8 \text{ cm}^{-2}$. The increases are the largest (3.0×10^8 to $1.1 \times 10^9 \text{ cm}^{-2}$) in South America, South Asia, and South Africa, where SO_2 emissions increase the most, while the NUM level decreases mostly over Europe and northern/western Asia by 2.0×10^8 to $9.8 \times 10^8 \text{ cm}^{-2}$ in response to the largest decreases in SO_2 emissions. The comparable spatial trends between the NUM level and the SO_2 emissions indicate that the changes in NUM are dominated more by changes in the nucleation of new particles from sulfuric acid than by changes in primary aerosol emissions. However, the large decreases in primary particle emissions in central Africa do offset the impact of enhanced new particle formation sufficiently to result in a slight decrease in the NUM level (less than

1.0×10⁸ cm⁻²). The changes in the CCN level mirror the changes in the NUM level with a global average increase in CCN of 6.0×10⁵ cm⁻², and decreases in central Africa, Europe, and northern/western Asia due to declines in the NUM level.

LWP increases by 1.4 g m⁻² on global average due to a combination of both increases in the CCN level and a global average increase in PWV of 1.1 mm (not shown). The impacts of increases in both cloudiness and CCN lead to a global average increase in CDNC of 2.2×10³ cm⁻². The increased CCN tends to increase the CDNC and LWP the most in marine clouds. Over land, changes in CDNC are noisier with the exception of Europe and northern/western Asia, where the CDNC level declines by 5.0×10⁴ cm⁻² to 3.5×10⁵ cm⁻² in response to the declines in CCN, despite increases in LWP. The changes in COT mirror the changes in CDNC with a global average increase of 0.05 largely as a result of the increased CDNC in marine clouds.

Although surface concentrations of PM_{2.5} can increase or decrease in some regions depending on the changes in surface emissions, the PM_{2.5} concentration in the middle and upper troposphere (600-200 mb) generally increases by 0.1 to 1.2 µg m⁻³ (shown in Figure S9). This is a result of enhanced secondary formation of PM_{2.5} from the enhanced oxidant levels and the poleward advection of global precursors and particles from tropical and subtropical regions. This increased PM_{2.5} level in the upper portions of the troposphere leads to a global increase in AOD with an average increase of 0.03, despite regional decreases in the PM_{2.5} level near the surface. AOD enhancements are the largest in the tropical and subtropical regions (0.1 to 0.2) due to both the increased formation of particles and the lofting of these particles due to strong vertical motion in this region. There are also strong enhancements of AOD along the coasts of the continents due to the replacement of chloride by sulfate and nitrate that have higher refractive indices.

The combined enhancement of COT and AOD leads to dimming at the Earth's surface with SWDOWN declining by 1.6 W m^{-2} on global average. This declining in SWDOWN leads to a reduction in photolysis. For example, the NO_2 photolysis rate decreases on global average by $1.2 \times 10^3 \text{ min}^{-1}$. This decline in photolysis could decrease the production of O_3 as mentioned in Section 4.1. However, the increases in NO_x emissions are able to overcome this feedback of the increasing aerosol level on photochemical activity. The dimming induced by aerosol enhancements to the global AOD and COT are also likely the cause for the milder warming of T2 compared to the CCSM3 model as mentioned in Section 3.2.

6. Sensitivity Analysis

6.1 Impact of Climate Change on Air Quality

Figure 4 shows the changes in O_3 8hr, $\text{PM}_{2.5}$, T2, Q2, WSP10, PBLH, PR, and the NO_2 photolysis rate between 2001 and the 2050_NPE simulation. The correlations between the changes in O_3 8hr and $\text{PM}_{2.5}$ with these meteorological variables are listed in Tables S6 and S7, respectively. Under the 2050_NPE case, the O_3 8hr level increases on global average by 0.6 ppb with increases of 1.0 to 3.8 ppb in many mid-latitude continental regions. This is largely in response to increases in temperature (R of 0.33), which increases by 1.2°C on global average but up to 4.2°C in these continental regions. The increasing temperature increases the O_3 8hr level by increasing reaction rates, increasing biogenic VOC emissions ($1.8 \text{ mol km}^{-2} \text{ hr}^{-1}$ on global average), and increasing decomposition of PAN (e.g., PAN decreases by 10.0% on global average). However, in tropical and oceanic regions, the O_3 8hr level is reduced by 0.3 to 1.8 ppb in response to increases in water vapor (0.8 to 2.6 g kg^{-1}) that cause greater O_3 destruction in these low NO_x environments. This explains the fairly substantial anti-correlation with Q2 (R of -0.29). Similar to the base 2050 case, the 2050_NPE case predicts a cloudier and more aerosol-

rich future atmosphere. The increase in aerosols is the result of the increase in SOA from greater biogenic emissions and greater sulfate from increases in the level of H_2O_2 (0.1 ppb on global average) and available cloud water (2.7 g m^{-2} on global average). This leads to a global average decrease in SWDOWN of 2.5 W m^{-2} (not shown). The results are global average declines in PBLH of 27.9 m and the NO_2 photolysis rate of $1.6 \times 10^{-3} \text{ min}^{-1}$. The decreased PBLH should lead to greater O_3 levels near the surface, increasing the O_3 8hr level and thus changes in O_3 8hr level and PBLH should be anti-correlated. However, there is a minor positive correlation (R of 0.16) with PBLH, indicating that the declining PBLH does not significantly impact the O_3 8hr level. The correlations with WSP10 and the NO_2 photolysis rate are negligible, indicating that the O_3 8hr level is not very sensitive to changes in photolysis and horizontal advection. These findings contrast somewhat with the multi-model ensemble predictions of O_3 increases over land and decreases in marine environments caused by climate change reported in Dentener et al. (2006) and Doherty et al. (2013). Since O_3 reductions from climate change are driven by increased water vapor in low NO_x environments, the dominant tendency for increased O_3 from climate change in GU-WRF/Chem is likely driven by smaller increases in water vapor compared to CCSM3 discussed in Section 3.2 and also the overprediction of column NO_2 . The impact of increased temperatures on O_3 may also be exaggerated by the overprediction in T2.

The $\text{PM}_{2.5}$ level in the future is enhanced by $0.25 \text{ } \mu\text{g m}^{-3}$ on global average in the 2050_NPE case. The changes in $\text{PM}_{2.5}$ are largely driven by changes in WSP10 (R of 0.38) especially over the oceans (R of 0.50), where the changes in WSP10 control the changes in emissions of sea-salt. In continental regions there is a smaller magnitude anti-correlation with WSP10 (R of -0.14), which indicates that some of the increasing $\text{PM}_{2.5}$ over land is in response to weaker horizontal advection. The future $\text{PM}_{2.5}$ level is also increased in response to increases of

SOA, which increases as much as 0.6 to $1.0 \mu\text{g m}^{-3}$ in central Africa, South America, and Indonesia in response to increases in biogenic VOC emissions.

It is possible that reductions in PBLH height could contribute to increases in $\text{PM}_{2.5}$ but that contribution is likely minor compared to horizontal advection changes since the correlation of $\text{PM}_{2.5}$ to PBLH is close to zero (R of -0.01 globally and -0.06 over land). There is a small positive relationship between Q2 and the $\text{PM}_{2.5}$ level (R of 0.15), which could indicate enhanced $\text{PM}_{2.5}$ due to water availability and enhanced levels of H_2O_2 , however, it is also likely that this is an artifact of changes in horizontal advection since Q2 would respond to changes in WSP10 in a similar fashion to $\text{PM}_{2.5}$. Changes in precipitation show little impact on $\text{PM}_{2.5}$ (R of 0.01), consistent with findings of other studies reviewed in (Fiore et al., 2012). The findings in this work show a fairly strong influence of climate on $\text{PM}_{2.5}$ and contrast with the influence of climate on $\text{PM}_{2.5}$ discussed in Section 4.2. This likely indicates that changes in emissions dominate over these changes from climate; however, internal model variability from comparing only a single year may also play a role in this discrepancy. Glotfelty et al. (2014) showed that the climate change signal of both T2 and Q2 in the simulations discussed in this work was statistically significant compared to current and predicted interannual variability. While this does not mean that the internal model variability is not present in the simulation, this does indicate the trend of the two climate variables is likely greater than the perturbations caused by internal model variability.

6.2 Impact of Changing Emissions on the Future Climate

To examine the impact that changing emissions have on future climate, Figure 5 shows the absolute differences in AOD, NUM, CCN, CDNC, column average cloud drop effective radius (R_{eff}), COT, SWDOWN, NO_2 photolysis rate, CF, PWV, LWP, PR, GLW, PBLH, T2,

and Q2 between the 2050 simulation with projected emissions and the 2050_NPE case. The general increase in the emissions of some primary aerosol and precursor emissions increases the global AOD by 0.03 on global average. Changes in NUM and CDNC between these simulations are similar to the differences between the AOF and AOC periods. The NUM level increases on global average by $2.3 \times 10^8 \text{ cm}^{-2}$, which results in a global average increase of $1.2 \times 10 \text{ cm}^{-2}$ in CCN. The projected emissions increase the global average CDNC level by $1.3 \times 10^3 \text{ cm}^{-2}$. The increases in CDNC primarily occur in the oceanic regions including the Indian Ocean, the South Atlantic Ocean, and the central Pacific Ocean, due to a relatively high level of cloud formation (e.g., LWP of 50 to 192 g m^{-2}) that favors effective aerosol activation. The enhanced CDNC level leads to a reduction in the cloud effective radius (R_{eff}) by 2.0 to $65.5 \text{ }\mu\text{m}$ in these regions, enhancing the cloud albedo and enhancing the COT by 0.1 to 1.9. In Europe and northern Asia, the NUM level is reduced by 1.0×10^8 to $1.2 \times 10^9 \text{ cm}^{-2}$, in response to declining SO_2 and primary aerosol emissions. The result is a decrease in the CDNC level of 5.0×10^4 to $5.2 \times 10^5 \text{ cm}^{-2}$. This reduction in CDNC allows the cloud droplets to grow larger (with R_{eff} increases of 2 to $12 \text{ }\mu\text{m}$) resulting in decreases in COT of 0.1 to 1.0. The findings in this work for the SRES A1B scenario contrast with those of RCP scenarios discussed in the work of Westervelt et al. (2015). Under the RCP scenarios, declines in primary aerosol and precursor emissions by the year 2100 result in consistent trends for reductions in AOD and also LWP of 0.5-1.0 g m^{-2} due to a reversal of the cloud lifetime effect. This allows for larger growth of existing droplets increasing R_{eff} by 0.54-0.60 μm . Similar reductions in AOD and reductions in CDNC were reported with a subset of the RCP scenarios in the work of Chalmers et al. (2012). A fairer comparison with the work of Jacobson and Streets (2009) shows contrasting declines in NUM and subsequently in CDNC following the SRES A1B scenario by 2030, but it also shows increases in AOD, COT, and LWP that are qualitatively similar to those mentioned above from GU-WRF/Chem. Over the

remaining continental regions, the cloud-aerosol interactions are more complex and these changes are discussed in more detail in the supplementary material.

On a global scale, the enhanced COT and AOD from the projected emissions results in a global average decrease in SWDOWN of 1.2 W m^{-2} and up to 18.5 W m^{-2} in the oceanic regions where COT increases the most. This reduction in SWDOWN reduces the NO_2 photolysis rate by $1.4 \times 10^{-3} \text{ min}^{-1}$ on global average. The changes in emissions do not have a substantial impact on global precipitation with a global average increase of $5.7 \times 10^{-4} \text{ mm day}^{-1}$. However, as mentioned in the supplementary material, there can be regional shifts in precipitation. Despite the global decreases in SWDOWN, T2 increases on global average by 0.03°C . This is a result of the increases in SWDOWN over land from changes in cloud formation or the reduced CDNC in Europe and northern Asia. There is no response in T2 over the oceans since GU-WRF/Chem uses prescribed sea surface temperatures. This illustrates a key limitation of such aerosol-climate interaction studies in atmospheric general circulation models and indicates that such a study is better suited for an Earth system model that has an active ocean model component. Westervelt et al. (2015) also reported increases in global average temperature and precipitation of $0.72\text{--}1.04^\circ\text{C}$ and $0.08\text{--}0.09 \text{ mm day}^{-1}$, respectively, following the RCP scenarios. However, these changes were linked to decreasing aerosol levels rather than the complex interactions discussed here. This increased temperature in all the regions except those mentioned above helps to enhance evapotranspiration on a global scale leading to global average enhancements of Q2 ($3.0 \times 10^{-3} \text{ g kg}^{-1}$), PWV (0.03 mm), and LWP (0.4 g m^{-2}). These global enhancements in atmospheric moisture variables lead to a slight enhancement in the downwelling of longwave radiation at the surface (0.1 W m^{-2} on global average).

7. Conclusions

The impacts of changing emissions and climate following the IPCC AR4 SRES A1B scenario are investigated using the online coupled GU-WRF/Chem model. The model simulates radiation and meteorological variables reasonably well, but not well for cloud parameters. Evaluation of column chemistry variables indicates overpredictions in aerosols from underpredicted precipitation and possible overestimations of biomass burning emissions. Column abundances of CO, NO₂, and TOR are overpredicted due to overestimates of CO and NO_x emissions, while the column SO₂ abundance is underpredicted due to missing sources of natural sulfur species. The comparison of GU-WRF/Chem and CCSM3 future climate predictions shows similar changes in T2, Q2, and PR compared to CCSM3. However, GSW and OLR decrease in GU-WRF/Chem in contrast to increases of GSW and OLR in CCSM3 due to the cloudier and more aerosol-rich atmosphere predicted by GU-WRF/Chem. Sulfate levels increase in both models on global average but the spatial patterns are different due to the differences in chemistry treatments and the emissions projected by different integrated assessment models. CESM1-CAM5 projections under the RCP8.5 scenario predict reduced CWP and AOD due to emission reductions in primary aerosols and aerosol precursors, especially over the continents, reducing the aerosol direct and indirect effects. This leads to greater increases in T2, Q2, and PR compared to either GU-WRF/Chem or CCSM3 and opposing trends in the changes of radiation variables compared to GU-WRF/Chem.

Global air quality is projected to degrade by the mid-21st century following the IPCC AR4 SRES A1B scenario. CO, SO₂, and NO₂ all increase on global average due to increases in emissions in the developing world. NH₃ decreases on global average largely due to decreasing emissions in East Asia. These changes are generally comparable to those reported by Jacobson and Streets (2009). The O₃8hr level will increase by 2.5 ppb on global average with large increases of up to 20.4 ppb in heavily populated areas of South Asia and East Asia. This increase

is primarily caused by increasing NO_x emissions but may also be due to increased emissions of TNVOC and increased reaction rates from a warmer climate. These findings of increasing O₃ are consistent with previous findings from the SRES A1B and A2 scenarios and in contrast to changes in O₃ projected by all the RCP scenarios except RCP8.5 (Liao et al., 2006; Jacobson and Streets, 2009; Young et al., 2013; Kim et al., 2015). Increasing O₃ by 2050 reported under the RCP8.5 scenario by 2050 is driven more by increases in CH₄ and TNVOC as emissions of NO_x are reduced (Young et al., 2013; Kim et al., 2015). The increases in NO_x and TNVOC emissions under the SRES A1B scenario also lead to global average increases in PAN and other nitrogen containing trace gases. The combined effect of enhanced O₃, NO₂, and Q2 enhances the near surface atmospheric oxidation state by increasing the abundance of OH, HO₂, H₂O₂, and NO₃g. The enhancement in OH is in contrast to decreases reported under the RCP scenarios due to recovery of stratospheric O₃, large increases in CH₄ under the RCP8.5 scenario, and decreased O₃ production in the remaining RCP scenarios (Voulgarakis et al., 2013; Kim et al., 2015).

PM_{2.5} increases by 0.3 µg m⁻³ on global average but the changes are regional in nature. Changes in primary particle emissions of BC, OC, and OIN have the greatest impact on the PM_{2.5} level, causing increases in South Asia, Southeast Asia, Indonesia, Australia, and South America, and reductions in Africa and East Asia. PM_{2.5} changes in marine regions are driven by WSP10 changes except in coastal zones where increased levels of SO₂ and HNO₃ lead to greater replacement of chloride by sulfate and nitrate, increasing the mass concentration of PM_{2.5}. Reductions in SO₂ and NH₃ emissions also contribute to the reduced PM_{2.5} level in Europe.

The increased SO₂ emissions and increased primary aerosol emissions in South America and South Asia in this scenario lead to enhancements in new particle formation and an increasing global CCN level. This enhanced CCN level, combined with a moister future atmosphere, leads

to enhanced CDNC and LWP. The increased CDNC and LWP result in an increased COT of 0.05 on global average and the increased aerosol level increases the global AOD by 0.03. Thus, the cloudier and more aerosol-rich atmosphere predicted by GU-WRF/Chem leads to dimming at the Earth's surface of 1.6 W m^{-2} on global average, which reduces photolysis and results in more moderate warming compared to CCSM3. These changes in aerosol effects are in contrast to reduced AOD and aerosol indirect effects following the RCP scenarios due to reductions in aerosol and precursor emissions (Lamarque et al., 2011; Shindell et al., 2013).

Similar to those reported under other SRES scenarios (Prather et al., 2003), deposition of both total nitrogen and O_3 is enhanced under this scenario, especially in the developing world due to enhanced emissions of NO_x . This will enhance the possibility of crop damage and ecosystem system stress in these areas that are already heavily-populated. However, the trend in O_3 deposition is in contrast to most RCP scenarios and the trend in total nitrogen deposition tends to mirror that of the RCP scenarios but is caused primarily by increases in reactive nitrogen rather than increases in reduced nitrogen shown in the RCP scenarios (Lamarque et al., 2011). BC deposition is reduced on global average due to declining emissions but there are enhancements in the Ural Mountains, Andes Mountains, and Tibetan Plateau, which may impact the water supplies of populations that depend on meltwater from these regions. Mercury deposition is enhanced from reductions in aerodynamic resistance and PBLH that can be regionally enhanced by increased precipitation. This may increase the amount of mercury that can become bioavailable and increase health impacts.

With only the influence of climate change, both the O_3 8hr and $\text{PM}_{2.5}$ levels are enhanced on global average by 0.6 ppb and $0.25 \mu\text{g m}^{-3}$, respectively. The increased O_3 8hr level is driven by increasing temperatures. This contrasts slightly with other studies that show larger reductions from enhanced water vapor (Dentener et al., 2006; Doherty et al., 2013), likely due to model

biases in temperature, water vapor, and NO_x . The enhanced $\text{PM}_{2.5}$ level is driven largely by reductions in horizontal advection from reduced WSP10 over land and enhanced sea-salt emissions from enhanced WSP10 in some marine regions. The projected emissions under this scenario result in dimming of 1.25 W m^{-2} at the Earth's surface from an enhanced AOD and COT. The COT increase primarily occurs in marine regions as there is ample moisture for cloud formation and aerosol activation. In continental regions, the impact of projected emissions is more complex due to regional perturbations in the hydrologic cycle that impact atmospheric moisture and ultimately aerosol activation. However, despite this dimming induced by changing emissions, the response in T2 is limited to only continental locations due to the use of prescribed sea surface temperatures. This indicates that studies focusing on global scale aerosol-cloud interactions should use an Earth system model with an active ocean model component to account for changes in marine radiation. These findings differ from those under the RCP scenarios that showed increased warming from reductions in AOD, CDNC, and LWP due to reduced aerosol levels (Chalmers et al., 2012; Westervelt et al., 2015).

This work demonstrates that policy makers, especially those in developing countries, need to implement effective climate mitigation and emission control strategies, such as those that are part of the RCP climate scenarios, in order to avoid potential air quality degradation, crop damage, and ecosystem system stress predicted following the SRES A1B scenario.. However, this study has several limitations discussed in Section S7 of the supplementary material. This work also illustrates that policy decisions based on atmospheric general circulation models need to use information provided by models that simulate more complex chemistry and aerosol-cloud interactions as the inclusion of these process will impact not only the prediction of radiation variables but also T2, Q2, and PR, as evidenced by the differences between the GU-WRF/Chem

and CCSM3 models. Some of those treatments have been included in many of the Earth System Models that have taken part in the CMIP5 comparison and also the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) (Lamarque et al., 2013). The results from the models with advanced chemistry, aerosol, and cloud treatments should provide the basis of future policy decisions. This work further demonstrates the need to conduct aerosol-climate interaction studies using an Earth system model framework, like those in the CMIP5 and ACCMIP projects, in order to represent the impact of changing emissions on not only the atmosphere but also the land, oceans, and sea ice. The advantages of using detailed chemistry and aerosol treatments illustrated in this work will potentially enhance the skill of an Earth system model in simulating atmospheric processes and their interactions with other components in the Earth system. The CB05_GE used in GU-WRF/Chem has indeed been implemented along with an advanced aerosol module with volatility basis set SOA treatments into the Community Earth System Model/Community Atmospheric Model version 5 to advance the Earth system modeling, which will be presented in a future paper.

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Table 1. GU-WRF/Chem Model Configurations

Attribute	Configuration
Time period	2001, 2010, 2020, 2030, 2040, and 2050
Horizontal resolution	$4^{\circ} \times 5^{\circ}$, 45 (latitude) $\times$ 72 (longitude)
Vertical resolution	27 layers from 1000–50 mb, with 17 layers in PBL (< 2.6 km)
Atmospheric Process	Parameterization
Longwave radiation	The rapid radiative transfer model (RRTM) (<i>Mlawer et al.</i> , 1997)
Shortwave radiation	Goddard scheme (<i>Chou et al.</i> , 1998)
Cloud microphysics	Purdue Lin (<i>Lin et al.</i> , 1983; <i>Chen and Sun</i> 2002)
Land surface	NOAH (<i>Chen and Dudhia</i> 2001a,b; <i>Ek et al.</i> , 2003)
Cumulus	Grell-Devenyi (<i>Grell and Devenyi</i> , 2002)
Planetary boundary layer	Yonsei University (YSU) scheme (<i>Hong et al.</i> , 2006)
Surface-layer	Monin-Obukhov scheme (<i>Monin and Obukhov</i> , 1954; <i>Janic</i> , 2002)
Gas phase chemistry	CB05_GE (<i>Karamchandani et al.</i> , 2012)
Aqueous chemistry	CMU mechanism (<i>Fahey and Pandis</i> , 2001)
Aerosol dynamics/thermodynamics	MADRID (<i>Zhang. et al.</i> , 2004, 2010, 2012c)
Photolysis	F-TUV (<i>Tie et al.</i> , 2003)
Aerosol Activation	Abdul Razzak-Ghan (<i>Abdul-Razzak and Ghan</i> , 2002)

NOAH - The National Center for Environmental Prediction (NNR), Oregon State University, Air Force, and Hydrologic Research Lab, CB05_GE – carbon bond mechanism version 2005 with global extension, CMU – Carnegie Mellon University, MADRID – Model of Aerosol, Dynamics, Reaction, Ionization, and Dissolution, F-TUV – Fast Tropospheric Ultraviolet-Visible Model.

Table 2. Model Evaluation Statistics for the Average of the Current Years

Variable	Unit	Dataset	Mean Obs.	Mean Sim.	Corr.	MB	NMB	NME
OLR	W m ⁻²	NOAA/CDC	216.0	217.8	0.97	1.8	0.8	3.1
OLR	W m ⁻²	CERES/EBAF	224.1	217.8	0.97	-6.3	-2.8	3.7
GLW	W m ⁻²	BSRN	325.5	329.6	0.94	4.0	1.2	4.5
GLW	W m ⁻²	CERES/EBAF	307.5	313.6	0.99	6.2	2.0	3.8
SWDOWN	W m ⁻²	BSRN	185.5	211.9	0.89	26.5	14.3	17.2
SWDOWN	W m ⁻²	CERES/EBAF	163.7	184.3	0.89	20.6	12.6	17.2
GSW	W m ⁻²	CERES/EBAF	128.9	149.3	0.92	20.3	15.8	22.8
CF		MODIS	0.66	0.80	0.44	0.13	20.2	31.3
LWP	g m ⁻²	MODIS	144.5	27.4	0.46	-117.0	-81.0	81.1
LWP	g m ⁻²	OD08	84.5	34.7	0.01	-49.6	-58.9	60.9
PWV	cm	MODIS	1.93	1.92	0.99	-4.0×10 ⁻³	0.2	13.6
COT		MODIS	16.9	2.9	-0.42	-14.0	-82.6	82.7
PSFC	hPa	NCDC	969.3	965.2	0.65	-4.1	-0.4	3.2
WS10	m s ⁻¹	NCDC	3.8	2/9	0.24	-0.9	-24.9	42.7
T2	°C	NCDC	13.6	15.0	0.94	1.4	10.6	19.2
Q2	g kg ⁻¹	NCDC	8.3	8.5	0.89	0.2	2.0	16.9
PR	mm day ⁻¹	NCDC	1.9	1.8	0.38	-0.11	-6.1	82.4
PR	mm day ⁻¹	GPCP	2.2	2.1	0.70	-0.08	-3.4	53.9
CCN	cm ⁻²	MODIS	2.5×10 ⁸	3.0×10 ⁸	0.38	4.8×10 ⁷	18.9	57.6
CDNC	cm ⁻³	BE07	107.3	39.1	0.20	-68.2	-63.5	67.1
AOD		MODIS	0.15	0.29	0.40	0.14	96.3	107.1
TOR	DU	TOMS/SBUV	29.6	34.7	0.75	5.1	17.3	21.7
(2001)								
OMI/MLS (2010)								
NO ₂	cm ⁻²	GOME (2001)	4.5×10 ¹⁴	7.8°10 ¹⁴	0.80	3.3×10 ¹⁴	73.7	92.9
SCIAMACHY (2010)								
SO ₂	DU	SCIAMACHY	0.20	0.04	0.40	-0.16	-79.7	81.3
CO	cm ⁻²	MOPITT	1.5×10 ¹⁸	2.1×10 ¹⁸	0.88	6.6×10 ¹⁷	44.6	44.6

T2, 2-m temperature; Q2, 2-m water vapor mixing ratio; WS10, 10-m wind speed only evaluated for observed wind speeds greater than 1.54 m s⁻¹; PSFC, surface pressure; DP, total daily precipitation in mm day⁻¹; PR, average precipitation rate (mm day⁻¹); SWDOWN, incoming shortwave radiation flux at the Earth's surface; GLW, incoming longwave radiation flux at the Earth's surface; OLR, outgoing longwave radiation at the top of the atmosphere; PWV, precipitable water vapor; LWP, vertically integrated cloud water; CF, cloud fraction; CCN_{S=0.5%}, vertically integrated cloud condensation nuclei at a supersaturation (S) of 0.5% ; COT, cloud optical thickness; AOD, aerosol optical depth; TOR, tropospheric ozone residual; NO₂, vertically integrated NO₂ column; CO, vertically integrated CO column; Corr., correlation coefficient; MB, mean bias; NMB, Normalized mean bias

Figure Captions

Figure 1

The zonal mean profiles of CF, LWP, COT, CCN, PWV, precipitation rate, OLR, AOD, column SO₂, column CO, column NO₂, and TOR from GU-WRF/Chem compared against the zonal mean profiles of observations from the MODIS, MOPITT, GOME, TOMS-SBUV, OMI-MLS, SCIAMACHY, NOAA/CDC, and GPCP satellite and reanalysis datasets for the average current year period (2001 and 2010).

Figure 2

A comparison of the variation trends in surface temperature, surface water vapor, net shortwave radiation at the Earth's surface, outgoing longwave radiation at the top of the atmosphere, precipitation rate, and surface sulfate concentration between the average current year period (2001 and 2010) and the average future year period (2020, 2030, 2040, and 2050) for both the CCSM3 and GU-WRF/Chem simulations.

Figure 3

Absolute changes in maximum 8-hr O₃, the average PM_{2.5} level, column cloud condensation nuclei concentration at a supersaturation of 0.5%, column cloud droplet number concentration, cloud liquid water path, cloud optical thickness, aerosol optical depth, and incoming shortwave radiation between the AOF and AOC periods.

Figure 4

Absolute differences in O₃8hr, PM_{2.5}, T2, Q2, PBLH, WSP10, NO₂ photolysis rate, and precipitation between the 2050 climate change only simulation and 2001.

Figure 5

Absolute differences in NUM, CCN, CDNC, Reff, AOD, COT, SWDOWN, NO₂ photolysis rate, CF, PWV, LWP, PR, GLW, PBLH, T2, and Q2 between the 2050 simulation with projected emissions and the 2050 simulation without projected emissions.

Figure 1. The zonal mean profiles of CF, LWP, COT, CCN, PWV, precipitation rate, OLR, AOD, column SO_2 , column CO, column NO_2 , and TOR from GU-WRF/Chem compared against the zonal mean profiles of observations from the MODIS, MOPITT, GOME, TOMS-SBUV, OMI-MLS, SCIAMACHY, NOAA/CDC, and GPCP satellite and reanalysis datasets for the average current year period (2001 and 2010).

Figure 2. A comparison of the variation trends in surface temperature, surface water vapor, net shortwave radiation at the Earth's surface, outgoing longwave radiation at the top of the atmosphere, precipitation rate, and surface sulfate concentration between the average current year period (2001 and 2010) and the average future year period (2020, 2030, 2040, and 2050) for both the CCSM3 and GU-WRF/Chem simulation

Figure 3. Absolute changes in maximum 8-hr O_3 , the average $PM_{2.5}$ level, column cloud condensation nuclei concentration at a supersaturation of 0.5%, column cloud droplet number concentration, cloud liquid water path, cloud optical thickness, aerosol optical depth, and incoming shortwave radiation between the AOF and AOC periods.

Figure 4. Absolute differences in O₃8hr, PM_{2.5}, T2, Q2, PBLH, WSP10, NO₂ photolysis rate, and precipitation between the 2050 climate change only simulation and 2001.

Figure 5. Absolute differences in NUM, CCN, CDNC, Reff, AOD, COT, SWDOWN, NO_2 photolysis rate, CF, PWV, LWP, PR, GLW, PBLH, T2, and Q2 between the 2050 simulation with projected emissions and the 2050 simulation without projected emissions.