Impact of Future Climate Policy Scenarios on Air Quality and Aerosol-Cloud Interactions using an Advanced Version of CESM/CAM5: Part I. Model Evaluation for the Current Decadal Simulations

Timothy Glotfelty, Jian He, and Yang Zhang

Department of Marine, Earth, and Atmospheric Sciences, North Carolina State University,
Raleigh, North Carolina, USA

Key Points

CESM-NCSU provides a reasonable representation of the current atmosphere.

Aerosol-cloud interactions are well simulated in CESM-NCSU.

Biases in chemical predictions are due to inaccurate emissions, mixing, deposition, and POA volatility.

Abstract

A version of the Community Earth System Model modified at the North Carolina State University (CESM-NCSU) is used to simulate the current and future atmosphere following the representative concentration partway scenarios for stabilization of radiative forcing at 4.5 W m^{-2} (RCP4.5) and radiative forcing of 8.5 W m^{-2} (RCP8.5). Part I describes the results from a comprehensive evaluation of current decadal simulations. Radiation and most meteorological variables are well simulated in CESM-NCSU. Cloud parameters are not as well simulated due in part to the tuning of model radiation and general biases in cloud variables common to all global chemistry-climate models. The concentrations of most inorganic aerosol species (i.e., SO_4^{2-} , NH_4^+ , and NO_3^-) are well simulated with normalized mean biases (NMBs) typically less than 20%. However, some notable exceptions are European NH_4^+ , which is overpredicted by 33.0–42.2% due to high NH_3 emissions and irreversible coarse mode condensation, and Cl^- , that is

negatively impacted by errors in emissions driven by wind speed and overpredicted HNO_3 . Carbonaceous aerosols are largely underpredicted following the RCP scenarios due to low emissions of black carbon, organic carbon, and anthropogenic volatile compounds in the RCP inventory and efficient wet removal. This results in underpredictions of $\text{PM}_{2.5}$ and PM_{10} by 6.4 - 55.7 %. The column mass abundances are reasonably well simulated. Larger biases occur in surface mixing ratios of trace gases in CESM-NCSU, likely due to numerical diffusion from the coarse grid spacing of the CESM-NCSU simulations or errors in the magnitudes and vertical structure of emissions. This is especially true for SO_2 and NO_2 . The mixing ratio of O_3 is overpredicted by 38.9-76.0% due to the limitations in the O_3 deposition scheme used in CESM and insufficient titration resulted from large underpredictions in NO_2 . Despite these limitations, CESM-NCSU reproduces reasonably well the current atmosphere in terms of radiation, clouds, meteorology, trace gases, aerosols, and aerosol-cloud interactions, making it suitable for future climate simulations.

KEYWORDS: CESM/CAM5, Representative Concentration Pathways, Earth System Modeling, Current Air Quality, Model Evaluation, Aerosol Indirect Effects

1. Introduction

The study of the future state of the atmosphere is of crucial importance as it will impact human and ecosystem health [Patz *et al.*, 2005; Haines *et al.*, 2006; Moore *et al.*, 2008; Pereira *et al.*, 2010; Shindell *et al.*, 2012; West *et al.*, 2013]. However, these studies are limited since the true path of future climate and emissions of air pollutants are unknown and as a result the scientific community relies on many different scenarios to explore the range of possible future outcomes. The majority of future air quality studies have used either the older Intergovernmental Panel on Climate Change (IPCC) Fourth Assessment Report (AR4) Special Report on Emissions

Scenarios (SRES) scenarios or the newer IPCC Fifth Assessment Report (AR5) Representative Concentration Pathway (RCP) scenarios [*Fiore et al.*, 2012 and references therein]. IPCC AR4 SRES scenarios are based primarily on socio-economic changes and as such the range in possible futures is described based on assumptions regarding the rate of globalization, population growth, and economic growth, as well as differences in future technology and energy generation [*IPCC*, 2001]. The IPCC AR4 SRES scenarios are a limited projection of the future since none of these scenarios represent the implementation of climate policy initiatives and as a result scenarios with such policies were needed [*IPCC*, 2007]. In a response to this need, the IPCC AR5 RCP scenarios were developed in a parallel process where information is exchanged between disciplines in a rapid manner to develop climate policy scenarios with multiple ways to achieve specific anthropogenic radiative forcing goals [*Moss et al.*, 2008; 2010].

Ultimately, four RCP scenarios were chosen by IPCC AR5 including: a scenario that peaks at approximately 3.0 W m^{-2} and declines to 2.6 W m^{-2} by 2100 (RCP2.6), two stabilization scenarios where radiative forcing stabilizes at target values of 4.5 W m^{-2} (RCP4.5) and 6.0 W m^{-2} (RCP6), respectively, by 2100, and a scenario in which radiative forcing never stabilizes, reaching a value of 8.5 W m^{-2} by 2100 (RCP8.5) [*Moss et al.*, 2010; *van Vuuren et al.*, 2011a]. Some commonalities in all scenarios are the introduction of carbon capture and storage technologies (CCS) to reduce global carbon emissions and greater affluence of developing countries that will lead to greater implementation of emissions control policies for atmospheric pollutants [*Masui et al.*, 2011; *Riahi et al.*, 2011; *Thomson et al.*, 2011; *van Vuuren et al.*, 2011a,b]. The RCP8.5 scenario provides a pseudo baseline for the other scenarios as it is the scenario reflecting the least climate mitigation due in part to slower economic growth and turnover in technology. As a result, this is the worst case scenario in terms of greenhouse gas

(GHG) emissions, as there are minimal controls implemented over energy sector emission of carbon dioxide (CO₂) and methane (CH₄) and increased demand in food supplies from greater populations leads to strong enhancements in nitrous oxide (N₂O) emissions from greater fertilizer use and significant increases in CH₄ emissions from increases in livestock and rice cultivation. Despite representing the worst carbon policy of all RCP scenarios, air pollutants are strongly reduced in RCP8.5 due to assumed successful implementation of current and planned air quality policies and implementation of these policies in developing countries once sufficient affluence is achieved [Riahi *et al.*, 2011]. The RCP2.6 scenario is the most optimistic of the RCP scenarios and achieves reduced radiative forcing through reductions in GHGs, largely from the introductions and large scale implementation of CCS systems on fossil fuel related emissions and introductions of alternative energy sources. Some of these climate policies provide additional air quality benefits as the introduction of CCS systems provides additional reductions in SO₂ emissions from coal and the implementation of hydrogen power sources in the transportation sector helps to reduce total non-methane volatile organic compound (TNMVOC) emissions [van Vuuren *et al.*, 2011b]. The more optimistic stabilization RCP4.5 scenario also relies strongly on CCS systems to achieve the radiative forcing target but includes greater reforestation as a process to control land use emissions and provide a carbon sink [Thomson *et al.*, 2011]. The less optimistic RCP6 scenario is possibly the most interesting pathway from an air quality perspective as Asian emissions in this scenario peak and then decline through the mid-21st century due to rapid expansion in Asian economies, resulting in the largest emissions of some pollutants (e.g., SO₂) under this scenario during the mid-21st century. Another key difference of this scenario compared to RCP4.5 is increases in deforestation and agriculture that drive greater

emissions of CH₄ and N₂O that contribute to the larger radiative forcing target [Masui *et al.*, 2011].

The projection of many air pollutants from these RCP scenarios is significantly different from the IPCC AR4 SRES scenarios where pollutant emissions increase mostly due to the lack of policy considerations [van Vuuren *et al.*, 2011a]. This results in key differences in future air quality, ecosystem health, and impacts from short lived climate forcers. For example, O₃ under the SRES scenarios is increased substantially in many scenarios [Prather *et al.*, 2003], while O₃ decreases by 2100 in a global sense under all RCP scenarios except RCP8.5 [Lamarque *et al.*, 2011; Young *et al.*, 2013; Kim *et al.*, 2015]. Subsequently, radiative forcing from O₃ under the SRES A1B scenario is a projected increase by 2100 [Levy *et al.*, 2008], while all the RCP scenarios except RCP8.5 project decreases in O₃ radiative forcing by 2100 [Stevenson *et al.*, 2013]. In terms of aerosols and radiative forcing the SRES A1B scenario predicted decreases in sulfate (SO₄²⁻) radiative forcing but large increase in black carbon (BC) radiative forcing associated with emissions changes by 2100 [Levy *et al.*, 2008], while under the RCP scenarios the radiative forcing of both SO₄²⁻ and BC decrease by 2100 in response to declining emissions [Lamarque *et al.*, 2011]. Commonalities exist in nitrogen deposition between both types of scenario as the A2 scenario shows enhancements in nitrogen deposition largely from increases in oxidized nitrogen from NO emissions [Dentener *et al.*, 2006]; while the RCP scenarios have enhanced nitrogen deposition in certain regions largely from increases in agricultural emissions of NH₃ [Lamarque *et al.*, 2011; Shindell *et al.*, 2013].

In this work, a version of the Community Earth System Model version 1.2.2 (CESM1.2.2) is employed in order to simulate the impact of future climate policy scenarios on air quality and aerosol-cloud interactions. This work will be presented in a sequence of two parts. Part I

describes model configurations, application, and evaluation. Part II describes the impact of future climate and emission changes on future air quality under the RCP4.5 and RCP8.5 climate policy scenarios. The objective of Part I is to perform decadal simulations of the current atmosphere using emissions from both the RCP4.5 and RCP8.5 scenarios and to compare the state of the atmosphere predicted in these simulations to a current period decadal simulation using more detailed emissions. This will determine how well these scenarios represent the current atmosphere. This work differs from previous studies by providing a comprehensive model evaluation of the current climate as predicted by the RCP scenarios using CESM with advanced treatments for organic aerosol formation and aerosol-cloud interactions.

2. Model Configuration, Evaluation Protocol, and Observational Datasets

2.1 Model Configuration

The CESM1.2.2 model used in this study contains a version of the Community Atmosphere Model version 5.3 (CAM5.3) that has been modified at the North Carolina State University (hereafter CESM-NCSU) [He *et al.*, 2015a, b]. Details of the default treatments within CESM1.2.2 are found on the web at http://www.cesm.ucar.edu/models/cesm1.2/tags/index.html#CESM1_2. CESM-NCSU contains several updated treatments for simulation of gases, aerosols, and aerosol and cloud interactions. Gas-phase chemistry is simulated using the modified version of the 2005 Carbon Bond mechanism with global extension (CB05GE) [Karamchandani *et al.*, 2012; He and Zhang, 2014]. The version of CB05GE in CESM-NCSU also contains updated reactions for isoprene, toluene, xylene and their products, as well as, the inclusion of benzene, ethyne, glycolaldehyde, and glyoxal chemistry from the Carbon Bond Version 6 mechanism (CB6) [Yarwood *et al.*, 2010] for improved simulation of organic aerosol. Inorganic aerosol thermodynamics is

simulated using the computationally-efficient ISORROPIA II module that allows for the explicit simulation of NO_3^- and NH_4^+ in CESM, additionally sea-salt aerosol is split into Na^+ and Cl^- ions and ratios of Mg^{2+} , K^+ , and Ca^{2+} ions are prescribed from dust concentrations that are not considered in the default model to be compatible with ISORROPIA-II [Fountoukis and Nenes, 2007; He and Zhang, 2014]. There are several updates in organic aerosol (OA) treatments. These include (1) a four bin volatility basis set (VBS) approach [Donahue *et al.*, 2006] for simulating secondary organic aerosol (SOA) formation from volatile organic compounds (VOCs) [Ahmadov *et al.*, 2012], (2) a nine-bin VBS approach for simulating the volatility of primary organic aerosol (POA), (3) a nine-bin VBS approach for simulation the organic aerosol formed from semi-volatile organic compounds (SVOCs) and oxidized POA (SVOA) [Shrivastava *et al.*, 2008; Shrivastava *et al.*, 2013], (4) and a simple irreversible uptake approach for simulating SOA formation from glyoxal [Knote *et al.*, 2014]. More details on these OA treatments can be found in Glotfelty *et al.* (2017). CESM-NCSU also contains an updated aerosol nucleation parameterization that selects the largest nucleation rate from the available nucleation parameterizations within the model [He and Zhang, 2014], with two additional nucleation parameterizations included in the selection process. These additional parameterizations are the ion mediated nucleation parameterization of Yu [2010] implemented by He and Zhang, [2014] and the organic gas/ sulfuric acid conucleation parameterization of Fan *et al.* [2006] implemented by Glotfelty *et al.* (2017). Aerosol activation in CESM-NCSU [Gantt *et al.*, 2014] is simulated following the Fountoukis and Nenes [2005] parameterization with additional updates for simulating the impact of giant cloud condensation nuclei (CCN) [Barahona *et al.*, 2010] and adsorptive activation of insoluble aerosols [Kumar *et al.*, 2009].

All CESM-NCSU simulations use a horizontal resolution of $\sim 0.9^\circ \times 1.25^\circ$ and a vertical resolution of 30 layers. The simulation periods are 2001-2010 that represent current climate conditions (hereafter current) and 2046-2055 that represent conditions of the mid-21st century around the year 2050 (hereafter future). Two RCP scenarios: RCP 4.5 and RCP 8.5 are selected. The RCP4.5 scenario is selected for this study since it is a more optimistic stabilization pathway and thus represents most likely future conditions with strict carbon policy implemented, and RCP8.5 is selected as it is the most pessimistic pathway among all RCP scenarios. Two sets of decadal simulations are therefore performed for one current decade and one future decade: one under the RCP4.5 scenario (hereafter RCP4C and RCP4F, respectively) and the other under the RCP8.5 scenario (hereafter RCP8C and RCP8F, respectively). Differences between simulations for the future decade and the current decade provide the impacts of changing emissions and climate on future air quality and its interactions with climate through various feedback mechanisms. An additional sensitivity decadal simulation is performed for the period of 2046-2055 which represents only future greenhouse gas forcing (GHGF) from the RCP4.5 scenario but utilizes RCP4.5 current decade emissions of trace gases and primary aerosols (RCP4F_GHGF). A comparison of RCP4F_GHGF with RCP4F illustrates the impacts of changing emissions on air quality and aerosol-cloud interactions and also their feedback to the climate system. A comparison of RCP4F_GHGF with RCP4C will illustrate the impacts of GHGF from the RCP4.5 scenario on future air quality. RCP4.5 is selected for the GHGF only scenario because it represents stronger emission control and climate mitigation policies than RCP8.5 and thus can better illustrate the impact that these types of policies can have on climate through aerosol-cloud interactions. An additional sensitivity experiment following RCP8.5 is not conducted due to limited computational resources.

The RCP4C and RCP8C simulations are configured based on the CESM1.2.2 B_2000_STRATMAM7_CN component set. This component set represents a fully-coupled CESM configuration including prognostic simulation of the atmosphere, ocean, land, and sea ice from the various component models. The simulation is configured with GHGF comparable to the year 2000 for N₂O, CH₄, CO₂, and Chlorofluorocarbons (CFCs) but with O₃ radiative forcing free to change in response to chemistry. This component set also contains active stratospheric chemistry from CB05GE, aerosols simulated by the seven mode version of the Modal Aerosol Model (MAM7) [Liu *et al.*, 2012], as well as, carbon and nitrogen biogeochemical cycling in the land model. Initial conditions (ICs) for these current decadal simulations are the same as those for the CESM-NCSU current decadal simulations in He *et al.* [2015a], which are generated from a multi-step process. The first set of ICs are obtained from a 10 year (1990-2000) CAM5.1 standalone simulation using chemistry from the Model for Ozone and related chemical Tracers version 4 (MOZART-4) [Emmons *et al.*, 2010] that is provided by the National Center for Atmospheric Research (NCAR). The MOZART-4 ICs are used to drive a one-year simulation of the CESM version 1.0.5 model using the B_1850-2000_CAM5_CN component set to provide initial meteorological conditions and chemical conditions for all species that exist in both MOZART and CB05GE gas-phase mechanism. The ICs from the land model are provided from an NCAR CESM/CAM4 B_1850-2000_CN simulation for the year 2000 and the ocean and sea ice models are initialized from CESM default settings. These ICs are then used to drive a three-month spin up period that provides the final ICs for all meteorological parameters and chemical species; including those in CB05GE that are not available in MOZART-4, that have been spun up from clean conditions.

The ICs for all component models in the RCP4F and RCP4F_GHGF simulations are provided by NCAR from the 2044 restart files of the CESM Phase Five of the Coupled Model Intercomparison Project (CMIP5) b.e11.BRCP45C5CN.f09_g16.004 experiment. The year of 2044 was the closest time period to the starting year of 2046 in this work available from NCAR. These ICs correspond to a simulation using a similar component set as the current decadal simulations but with highly simplified chemistry and RCP4.5 GHGF. Due to the lack of chemical ICs, ICs of GHGs, CH₄, N₂O, CO₂, and O₃, are generated from output of the Whole Atmosphere Community Climate Model (WACCM) RCP4.5 r2i1p1 CMIP5 experiment. ICs of CFCs are generated from default IC files used to drive CESM stratospheric chemistry simulations for the year 2000 that are scaled to the year 2050 based on growth factors derived from the GHG levels used in the b.e11.BRCP45C5CN.f09_g16.004 experiment. These ICs drive a 1-year spin up simulation that generates the ICs for the chemical species used in the future decadal simulations in this work. The ICs for the RCP8F simulation are prepared in the same manner using restart files for the year 2047 from NCAR's CMIP5 b.e11.BRCP85C5CN.f09_g16.001 experiment and WACCM output of CH₄, N₂O, CO₂, and O₃ from the r2i1p1 CMIP5 experiment. The year 2047 was used as this is the closest available time period from NCAR for the starting year of 2046 in this work. All future decadal simulations have GHGF set to 2050 levels for either the RCP4.5 or RCP8.5 scenario.

In addition to the configurations mentioned above, the CESM-NCSU model underwent an additional tuning process for radiation variables. This tuning is necessary since the default tuning parameters in the model are no longer valid once the aerosol activation parameterizations of *Gantt et al.* [2014] were incorporated into the model framework. These aerosol activation treatments generally improved the performance of some cloud variables such as cloud optical

thickness (COT) and cloud liquid water path (LWP) but resulted in an overprediction of shortwave cloud forcing (SWCF) and an associated underprediction in downwelling shortwave radiation at the Earth's surface (FSDS) [Gantt *et al.*, 2014]. These changes were responsible for the strong cold bias reported in He *et al.* [2015a]. Additionally, both the work of Gantt *et al.* [2014] and He *et al.* [2015a] neglected primary number emissions from BC, primary organic carbon (POC), and primary sulfate (SO_4^{2-}). Once the primary number emissions were accounted for in the current decadal simulations, the radiation and temperature biases were further degraded without tuning.

NCAR tunes CESM radiation in climate simulations by adjusting various cloud parameters until top of the atmosphere annual net radiation flux (RESTOA) is equivalent to 0 W m^{-2} for the preindustrial period [Hannay *et al.*, 2014]. Model parameters that are typically adjusted during this process include the threshold relative humidity for formation of low level clouds (rhminl), the critical diameter for ice crystal autoconversion (Dcs), and the cloud fraction for deep convection (personal communication, Cecile Hannay, NCAR, 2014). The method employed to tune CESM-NCSU is slightly different as the time period of the current decadal simulations is not preindustrial. For CESM-NCSU, RESTOA was calibrated to match available estimates of RESTOA from the Cloud and Earth's Radiant Energy System (CERES) / Energy Balanced and Filled (EBAF) dataset, which is roughly 0.8 W m^{-2} as a global area-weighted average for the averaged period of 2001-2010 (personal communication, Cecile Hannay, NCAR, 2014). A series of sensitivity simulations were carried out to find the appropriate tuning parameters. The final result was an adjustment of rhminl from 89.75% in the standard release to 95.5% for CESM-NCSU and an adjustment of Dcs from $400 \mu\text{m}$ in the standard release to $500 \mu\text{m}$ in CESM-NCSU. These changes are consistent with the possible ranges of the tuning

parameter values (personal communication, Cecile Hannay, NCAR, 2014). Given computational constraints and the highly complex nature of the CESM model, the final model global area-weighted value of CESM-NCSU RESTOA was 0.7 W m^{-2} , which was as close to the global area-weighted CERES/EBAF value of 0.8 W m^{-2} as possible without overshooting.

The RCP emission inventories used to drive the simulations are publicly available from version 2 of the RCP Database on the web at <http://tntcat.iiasa.ac.at/RcpDb/dsd?Action=htmlpage&page=welcome>. The current decadal simulations (RCP4C and RCP8C) are broken into three distinct emission periods with the years 2001-2003 utilizing the RCP year 2000 emissions, the years 2004-2006 utilizing the RCP year 2005 emissions, and the years 2007-2010 utilizing the RCP year 2010 emissions. The future decadal periods (RCP4F, RCP4f_GHGF, and RCP8F) utilize emissions based on RCP year 2050 emissions. Emissions of dust, sea-salt, and biogenic VOCs are calculated online in CESM1.2.2 following the parameterizations of *Zender et al.*, [2003], *Martensson et al.* [2003], and the Model of Emissions of Gases and Aerosols from Nature version 2 (MEGAN2) [*Guenther et al.*, 2006], respectively.

2.2 Evaluation and Analysis Protocol

The protocols for evaluation include assessing spatial distributions and statistical measures of model performance following a similar procedure to those used in *Zhang et al.* [2012] and *He et al.* [2015a]. Statistical measures of interest include mean bias (MB), root mean square error (RMSE), normalized mean bias (NMB), normalized mean error (NME), and correlation coefficient (R). Full definitions and details of these metrics can be found in the work of *Yu et al.* [2006] and *Zhang et al.* [2006]. The radiation, cloud, and meteorological variables

evaluated are listed in Table 1 along with the variables' short names used in the remainder of the text, the datasets against which the variables are compared, the spatial coverage and resolution (if applicable) of the datasets, and the temporal coverage during the CESM-NCSU current decadal simulations. Similar information for the chemical species evaluated is listed in Table 2.

All variables are evaluated on an annual basis and some dataset limitations exist. Chemical measurements in East Asia are very limited in terms of both chemical species (i.e., carbon monoxide (CO), nitrogen dioxide (NO₂), sulfur dioxide (SO₂), O₃, and PM₁₀) as well as spatial coverage, with all variables except PM₁₀ only available from South Korea, Japan, and Taiwan. PM₁₀ observations are limited to China and Taiwan for the entire period and available from South Korea beginning in 2006. Traffic and industrial sites are excluded from the Airbase and BDAQ observations because the horizontal grid resolution of these simulations is too coarse to represent these observations [Zhang *et al.*, 2013]. OC observations from EMEP sites have a coarse temporal resolution due to the lack of a routine observational program [Yttri *et al.*, 2007]. As a result, all available observations of OC spanning the time period of 2001-2013 are averaged into an annual climatology to compare with the CESM-NCSU simulation. Similarly, the oxygenated organic aerosol (OOA), hydrocarbon-like organic aerosol (HOA), total organic aerosol (TOA), and SOA measurements compiled in the works of Zhang *et al.* [2007], Jimenez *et al.* [2009], and Lewondowski *et al.* [2013] are based on very limited time periods so these observations are assumed to represent the climatology of the season in which the measurements were taken and are compared against the corresponding seasonal climatology from the CESM-NCSU simulations.

In order to explore how well CESM-NCSU simulates aerosol-cloud interactions and trace gas levels, interannual variations of PM, CCN at supersaturation of 0.5% (CCN5), cloud droplet

number concentration (CDNC), Aerosol Optical Depth (AOD), shortwave cloud forcing (SWCF), tropospheric NO₂ column (TNO₂), tropospheric formaldehyde column (THCHO), and tropospheric ozone residual (TOR) are analyzed over the marine stratocumulus regions identified in the work of *Rausch et al.* [2010]. These regions were selected because the nearly continual presence of clouds in these regions makes them ideal for evaluating aerosol-cloud interactions and the remoteness of these regions provides insights into how well the background levels of trace gases and aerosols are simulated. These stratocumulus regions exist the vicinity of Australia (AUS, 30°S–40°S, 88°E–103°E), North America (NAM, 15°N–35°N, 115°W–140°W), South East Asia (SEA, 10°N–40°N, 105°E–150°E), Southern Africa (SAF, 5°S–25°S, 10°W–15°E), and South America (SAM, 8°S–28°S, 70°W–90°W). However, caution is taken when interpreting the simulated interannual variations in these regions as GHGF for all years is identical, thus year to year variations only result from internal model variability and the changes between the emissions periods. Additionally, the trends in radiation (FSDS and Outgoing Longwave Radiation (OLR)), cloud (CF, cloud water path (CWP)), meteorological (2-m temperature (T2), 2-m specific humidity (Q2), 10-m wind speed (WSP10), and precipitation rate (PR)), and aerosol (AOD) variables between the current and future decadal simulations as predicted by CESM-NCSU will be compared against those predicted by a default version of CESM without complex chemistry from NCAR’s RCP4.5 and RCP8.5 r1i1p1 CMIP5 experiments. This will illustrate the impacts of using the advanced chemical mechanisms to simulate these RCP pathways.

3. Evaluation of Current Climate Periods

3.1 Evaluation of Radiation, Cloud Properties, and Meteorological Parameters

Performance statistics for radiation, cloud, and meteorological parameters from both the RCP4C and RCP8C simulations are listed in Table 3. Zonal mean profiles of radiation and cloud parameters from the RCP4C, RCP8C, and *He et al.* [2015a] (HE15) CESM-NCSU simulations, as well as the CESM CMIP5 data that represent merged historical and RCP4.5 simulations (if available) are compared against satellite estimates in Figure 1. Similar zonal mean comparisons of meteorological and chemical parameters are shown in Figure 2. The performance of cloud, radiation, and meteorological variables are generally similar for both the RCP4C and RCP8C simulations. RESTOA is well simulated after tuning, with a slight underprediction of 3.9% in the RCP4C simulation and slight overprediction of 3.4% in the RCP8C simulation, NME values of ~26%, and R values of 0.99. The sensitivity simulations conducted to find the appropriate tuning parameters utilized the RCP8.5 emissions and as a result, the opposite signs in biases between the two scenarios are caused by their difference in emissions and GHGF. It is important to note that the mean satellite retrieval estimates and mean simulation values of RESTOA in Table 3 are not the same as the RESTOA values mentioned in the model configuration section. This is because the average values mentioned in the model configuration section are area-weighted averages over the globe, while the values in Table 3 are the average grid cell values for model validation. Thus the area-weighted RESTOA average value is positive because grid cells with a positive RESTOA in the tropics are larger in terms of area than higher latitude grid cells where RESTOA is negative. The model performance for RESTOA is the poorest in the tropics with a general underprediction. This is likely caused by the overpredictions of COT in this region that increase outgoing shortwave radiation (OSR) and reduce FSDS compared to satellite estimates, thus reducing the shortwave contribution to RESTOA.

Other shortwave radiation variables are well simulated with NMBs less than $\pm 4\%$, NMEs less than 25%, and R values generally higher than 0.90 with the exception of OSR that has an R value of 0.83. FSDS and FSNS are both slightly underpredicted in response to overpredictions of SWCF and OSR. This indicates that generally clouds within the CESM-NCSU model overestimate reflectance. As shown in Figure 1, the CESM-NCSU model accurately captures the latitudinal variations in the shortwave radiation variables. The overpredictions in SWCF are the most pronounced in the tropics and result from the overpredictions in COT in this region. There is also a smaller overprediction that occurs in the northern hemisphere (NH) mid-latitudes that could potentially be attributed to the large overprediction in CDNC in this region. SWCF is underpredicted in polar regions and this is likely attributed to the underpredictions of cloud and aerosol variables in these environments discussed later in this section. The overpredictions in SWCF in the tropics and NH mid-latitudes are responsible for the overpredicted OSR in these regions. OSR is also overpredicted in the polar regions, likely due to the overpredicted surface reflection from an overpredicted surface albedo. This overprediction of surface albedo could potentially be related to inaccuracies in snow or sea ice coverage or a limitation in the configuration of CESM. The configuration of CESM used in this study does not simulate the interaction between BC and snow albedo, which has been shown to affect the radiative forcing of the snow cover [Flanner *et al.*, 2007]. FSDS is underpredicted in the tropical regions in response to the overpredicted SWCF and overpredicted in polar regions in response to underpredicted SWCF. This pattern is slightly different from FSNS that is also underpredicted in the tropics (due to the SWCF overprediction), but underpredicted in the polar regions. The underprediction in FSNS, despite overpredicted FSDS, in the polar regions provides further evidence of an overpredicted polar surface albedo. In general, the tuning of

RESTOA brings all shortwave radiation variables into better agreement with satellite estimates compared to HE15 and also CMIP5 in the case of FSDS.

Longwave radiation variables are also well simulated in the RCP4C and RCP8C simulations with NMBs within $\pm 11\%$, NMEs less than 21%, and R values above 0.88. FLDS, OLR, and LWCF are all slightly underpredicted, while FSNS is slightly overpredicted. The bias in the longwave radiation variables is in part caused by the underpredictions in FSDS, since the lost energy leads to a cooler surface or less energy emitted as longwave radiation. Similar to the shortwave variables, the longwave variables from CESM-NCSU capture the satellite estimated latitudinal variations. FLDS performs well across the majority of the globe but has the strongest underpredictions in the polar regions and slight overpredictions in the tropics. In the polar regions, this is caused in part by the underprediction in FSNS but may potentially be related to uncertainties in the complex refractive index of water (CRI) that has been shown to result in underestimates of the greenhouse effect in colder regions [Rowe *et al.*, 2013]. The slight overpredictions in the tropics are related to overpredictions in LWCF in this region. LWCF is generally underpredicted in the middle and higher latitudes but overpredicted in the tropics. The underprediction in the mid-latitude regions is likely caused by underpredictions in CF and COT that are maximized in these regions. At the higher latitudes, LWCF is underpredicted due to reduced surface longwave emissions as a result of underpredicted FSNS and the uncertainties in the CRI of water. The overpredicted LWCF in the tropics of the Southern Hemisphere (SH) is likely related to the overpredicted COT in this region. The co-location of the overpredicted COT and LWCF suggests that simulated clouds in this region are physically thick (i.e., the distance from cloud base to cloud top is large), which would explain the strong overpredictions of both SWCF and LWCF. OLR is underpredicted in tropical and subtropical regions of the SH and the

higher latitudes of the NH. The underprediction of OLR in the SH is likely caused by a combination of underpredicted FSDS and the overprediction in LWCF in this region. The underprediction of OLR in the higher latitudes of the NH is likely caused by a combination of the underpredicted FSNS and overpredicted FLNS that leads to an underestimation of T2 as shown in Figure 2, and thus the reduced emission of longwave radiation. FLNS is underpredicted in the tropics similar to all longwave variables in response to underpredicted FSDS; but it is overpredicted in the poles. This is potentially due to the inaccuracies in the simulated land surface or the uncertainties in the CRI of water. In general, the tuning of RESTOA has improved the simulation of longwave radiation variables compared to the HE15 simulation except in tropical regions. Compared to CMIP5, CESM-NCSU has slightly degraded performance for OLR but overall the differences are not substantial.

AOD is moderately well simulated in the RCP4C and RCP8C simulations with relatively low NMBs (-16.2% to -14.5%), moderate error (NMEs of 40.7-41.7%) and moderately high R values (0.61-0.62). The zonal mean profile in Figure 1 shows that the latitudinal variations in AOD from satellite estimates are captured by the model but deviations exist, primarily in the polar regions. Underestimates of aerosol loadings in the polar regions have been reported with the unmodified version of the MAM aerosol module and are believed to reflect either underestimations in global emissions or too efficient wet deposition of aerosols [Liu *et al.*, 2012]. The AOD predicted in the RCP4C and RCP8C simulations is larger and generally performs better than HE15 (e.g., NMBs of -14.5% and -16.2% vs. -35.4% and 49.2%), except in the subtropical regions of the NH and SH where AOD is overpredicted compared to satellite estimates. The larger AOD is the result of the vertically-structured emissions in the RCP inventories that allow for greater lofting of BC, POA, and SO_4^{2-} aerosols into the upper

troposphere where they are more radiatively active. Inaccuracies in these emissions or their vertical structure likely cause the overpredictions in the subtropics as the subtropical regions of the NH contain large primary aerosol emissions from South Asia and Southeast Asia and the subtropical regions of the SH contain large biomass burning emissions from South America and Africa. In general all CESM-NCSU simulations provide an AOD comparable to that of CMIP5 with the RCP simulations providing a slightly higher AOD in most latitudes

CF in CESM-NCSU is well simulated in terms of NMB (-0.9% to -0.7%) and NME (25.9-26.4%) but poor in terms of R value (0.28). This performance is slightly degraded in comparison to the HE15 current climate simulation that slightly overpredicted CF. with an NMB of 0.4%, an NME of 15.3%, and an R value of 0.7. This illustrates the negative aspects of tuning the model as not all variables will benefit. The cloud fraction is underpredicted in the RCP4C and RCP8C simulations because the increased rhminl value reduces low level cloud formation. The loss of low level clouds likely impacts the spatial distribution and thus results in the low R value. Despite this low R value, the RCP8C and RCP8F simulations do capture the latitudinal variations in CF from satellite estimates and are very similar to the values predicted in the CMIP5 experiment as shown in Figure 1. All CESM simulations generally overpredict CF in the polar regions, which is a historical model limitation of CESM and other global climate models [Vavrus and Waliser, 2008; Qian et al., 2012].

CCN5 is underpredicted in the RCP4C and RCP8C simulations with NMBs of -47.3% to -45.7%. The NMEs of 49.4-50.3% for CCN5 indicate that the majority of the simulation errors are a result of the bias. The zonal mean comparison shows that CCN5 values from the RCP4C and RCP8C simulations are generally comparable to MODIS estimates for regions in 30°S-45°N but are largely underpredicted in the middle and higher latitudes. As shown in Table 1, the

MODIS CCN product is only available in marine environments, so the underpredictions in CCN5 are likely from uncertainties in marine aerosol treatments. For example, these underpredictions in CCN5 could potentially be reduced with a treatment for marine organic aerosol [Gantt *et al.*, 2012, 2015]. CCN5 may also be underpredicted in part due to the overpredicted aerosol activation as evidenced by the overprediction of CDNC discussed in detail below. The simulation of CCN5 is greatly improved compared to HE15 due to the addition of particle number emissions of BC, POC, and SO_4^{2-} that were neglected in HE15 and the inclusion of the organic gas/sulfuric acid conucleation treatment in this version of CESM-NCSU (Glotfelty *et al.*, 2017). CDNC is overpredicted in the RCP4C and RCP8C simulations with NMBs of 39.8-45.2%. NMEs are fairly large (58.6-62.6%) and R values are relatively low (0.42-0.43), indicating that the spatial distribution is only moderately captured. The zonal mean profiles show that the RCP4C and RCP8C simulations capture most of the latitudinal variability in CDNC but there is a discrepancy in the middle and higher latitudes of the NH with CDNC levels peaking at $\sim 40^\circ\text{N}$ in the simulation rather than peaking at $\sim 50^\circ\text{N}$ in the satellite estimate. This discrepancy is likely the cause of the moderately large errors and lower R values. The underpredictions at the higher latitudes are likely caused by underpredicted aerosol loadings in these environments, as evidenced by the underpredicted AOD in this region, and uncertainties associated the phase of high latitude clouds in global climate models [Verlinde *et al.*, 2007; Fan *et al.*, 2011]. The systematic overprediction of CDNC by CESM-NCSU can be linked to uncertainties in the model treatment for aerosol activation [Gantt *et al.*, 2014] and also uncertainties in the MODIS derived satellite product [Bennartz, 2007], especially uncertainties in MODIS cloud effective radius [Nakajima *et al.*, 2010; Zhang and Platnick, 2011]. Compared to the HE15 simulation the CDNC

overprediction is larger in the RCP4C and RCP8C simulations in response to higher CCN5 levels.

Precipitable Water Vapor (PWV) is very well simulated in the RCP4C and RCP8C simulations with low NMBs (2.8-3.4%), low NMEs (11.8%), and high R values (0.98). The zonal mean profile of PWV matches that of the MODIS estimates with slight underpredictions in the subtropical and mid-latitudes of the NH and slight overpredictions in the middle and higher latitudes of the SH. PWV is improved from the HE15 simulation due to the enhanced FSDS from model tuning in the tropical region that facilitates greater evapotranspiration. LWP in the RCP4C and RCP8C simulations is underpredicted (NMBs of -30.3% to -29.9%) and only moderately captures the spatial distribution of the satellite estimates (NMEs of 36.4-36.7% and R values of 0.42-0.44). The zonal mean profiles reveal that LWP from CESM-NCSU (RCP4C, RCP8C, and HE15) is generally comparable to satellite estimates in the mid-latitudes but underpredicted in the tropics, subtropics, and polar regions. Despite these underpredictions, the latitudinal variations in the satellite estimates are captured in the simulation in all but the polar regions. The underpredictions in the tropics and subtropics suggest that the model is not accurately representing LWP from convective clouds, as these clouds dominate this region, and may also be related to the underpredicted CF. The poor performance in the polar regions may be related to some of the common issues of CESM and other global climate models in simulating polar aerosols and clouds, as mentioned earlier in this section. It may also be due to inaccuracies in the satellite retrievals at these latitudes due to low solar zenith angles [*Seethala and Horváth, 2010*] and interference from snow cover [*Kay et al., 2012*]. COT is also underpredicted (NMBs of ~ -41.0%) in the RCP4C and RCP8C simulations with even larger errors in spatial distribution compared to LWP (NMEs of 54.9% and R values of -0.12). The zonal mean profiles show that

COT is generally comparable to the MODIS estimates in the middle and lower latitudes but does not capture the high COT values of the polar regions. This is likely due to the same modeling and satellite uncertainties that cause LWP underpredictions in these regions. The general underprediction of COT over the remainder of the globe is likely caused by the underestimates of LWP and CF in these areas. COT and LWP from the RCP4C and RCP8C simulations are comparable to their performance reported by HE15, although some deviations exist, either from a lack of tuning of model radiation in HE15 or from the updates to the microphysics treatment that took place between CAM5.1 and CAM5.3 (http://www.cesm.ucar.edu/models/cesm1.2/tags/cesm1_2/whatsnew_science.html). All CESM-NCSU simulations show improvements in LWP compared to the CMIP5 simulations, illustrating the benefit of the updated aerosol formation and aerosol activation treatments. The moderate to poor performance of cloud related variables is common amongst many global climate models [Lauer and Hamilton, 2013] and regional air quality models [Zhang *et al.*, 2012]. The strong performance of radiation variables compared to these cloud parameters suggests a disconnection between these two fields of study and may reflect offsetting errors in modeled radiation for different assumptions regarding cloud properties [Qian *et al.*, 2012]. The underprediction of LWP and other cloud parameters has been improved using a version of CESM that contains a cloud resolving model (CRM) within each CESM grid cell [Wang *et al.*, 2011], illustrating CRM as a pathway for better representing aerosol-cloud interactions given the increasing availability of computational resources. Also, one important caveat of the results presented here is all simulated values are derived from monthly averaged CESM output files and as a result the annual average simulated results may not necessarily reflect the overpass times of the satellite estimates.

PR is well simulated in the RCP4C and RCP8C simulations with low NMBs (8.3-8.9%) low NMEs (26.3-26.8%) and high R values (0.86-0.87). The zonal mean profiles show that CESM-NCSU captures the latitudinal variability of PR from GPCP but tends to overpredict deep tropical convective precipitation. In general the PR predicted in the current decadal simulation is very similar to that in both the HE15 and CMIP5 simulations. T2 is also simulated well in the RCP4C and RCP8C simulations although a cold bias still persists with MBs ranging from -1.3°C to -1.0°C and RMSEs ranging from 3.0-4.0°C comparing to the NCEP/NCAR and NCDC datasets. The R values are generally high (0.93-0.99). The zonal mean profile shows a very close matching to the T2 from the NCEP/NCAR analysis dataset with slight underpredictions occurring in the polar regions. These underpredictions are related to the underpredictions of FSNS in these regions. In general, the radiation tuning helps to correct the cold bias reported in the HE15 simulation but the T2 values from CESM-NCSU simulations are still slightly cooler than the T2 predicted by the CMIP5 version of the CESM model. The cool bias in T2 leads to reduced evapotranspiration and results in a slight dry bias in Q2 with an MB of -0.5 to -0.3 g kg⁻¹, however, Q2 is generally well simulated with RMSEs of 1.0-1.7 g kg⁻¹ and R values of 0.93-0.99. The zonal mean profile of Q2 shows considerable improvement in Q2 compared to that of HE15 and CMIP5 in the tropics after tuning and similar to T2, the RCP4C and RCP8C simulations have better performance in Q2 than HE15 and weaker performance compared to CMIP5 in the NH. In general these deviations in T2 and Q2 from observations are comparable to those reported across the different models participating in the CMIP5 project [Tian *et al.*, 2013]. WSP10 is moderately well simulated in terms of errors (RMSEs 1.8-3.3 m s⁻¹) and correlation (Rs of 0.47-0.58) comparing to both the NCDC and NCEP/NCAR datasets but there are differences in bias with an underprediction of 0.4 m s⁻¹ against the NCDC observations and an

overprediction of $\sim 2.5 \text{ m s}^{-1}$ against the NCEP/NCAR analysis. This inconsistency between the performance statistics using both observational datasets stems from two possible reasons. The first is that NCDC sites are primarily land based, while NCEP/NCAR has global coverage. This may indicate that CESM-NCSU underpredicts wind speeds over land but overpredicts wind speeds in marine environments. The second possibility is limitation different data processing procedure used for the NCDC observations, which requires values less than 1.5 knots be excluded from evaluation because of uncertainties in anemometer measurements [NOAA, 1998]. This would artificially eliminate or limit overpredictions against WSP10 from NCDC if such biases exist. The zonal mean profile of WSP10 against the NCEP/NCAR analysis shows that despite the biases, the RCP4C and RCP8C simulations adequately capture the latitudinal variations in WSP10 and there is no substantial difference between these simulations and those of CMIP5 or HE15.

3.2 Evaluation of Trace Gases and Aerosols

Table 4 summarizes the performance statistics of trace gas and aerosol variables. CO evaluated against limited measurements from East Asia reveals a substantial underprediction with NMBs of -71.7% to -70.3%. The NMEs are identical to the NMBs indicating most of the error comes from the bias. This is likely caused by inaccuracies in emission levels and simulated vertical mixing in this region. Despite this poor performance in East Asia, TCO is well simulated with NMBs of -11.4% to -9.9%, NMEs of 15.2-16.2%, and R values of 0.83-0.85. The zonal mean profile in Figure 2 reveals that the RCP4C and RCP8C simulations capture the profile from satellite estimates but slightly underpredict TCO in the NH mid-latitudes. This could be related to either underestimates of emissions in the RCP inventory or potentially from overestimated oxidation as evidenced by the O₃ overprediction that will be discussed later in this section. The

overprediction of TCO in HE15 is reduced in the RCP4C and RCP8C simulations but the underprediction in the surface CO concentrations in East Asia is enhanced. This is caused by a correction to the CO reaction rate against OH that underestimates CO destruction by OH in the older version of CESM. This bug was reported in the supplementary material of *Lamarque et al.* [2012]. THCHO is underpredicted (NMBs of $\sim -38.0\%$), resulting in moderate errors (NMEs $\sim 42.0\%$) and a reasonable agreement of the spatial pattern (R values ~ 0.69). The zonal mean profiles from the RCP4.5 and RCP8.5 simulations are qualitatively similar to satellite estimates although the simulations predict less THCHO, especially in SH. These errors could potentially be related to underestimated emissions of formaldehyde or its precursors (e.g., isoprene and terpene) and uncertainties in secondary formation of HCHO. Such uncertainties have been shown to vary by approximately 20% among various commonly-used gas-phase chemical mechanisms including regional versions of CB05 [*Knote et al.*, 2015]. THCHO is improved compared to that in the HE15 simulation due to the inclusion of biogenic emissions that are part of the updated MEGAN2 in the CESM1.2.2 land model (http://www.cesm.ucar.edu/models/cesm1.2/tags/cesm1_2/whatsnew_science.html). Figure S1 of the supplementary material shows absolute differences in the emissions of CO, HCHO, nitric oxide (NO), SO₂, NH₃, anthropogenic VOC (AVOC), BC, and POC between the RCP8.5 and RCP4.5 current emission inventories. In general, CO and HCHO levels are reduced in the RCP8C simulation compared to the RCP4C simulation in response to lower emissions in the RCP8.5 inventory.

NO₂ is strongly underpredicted at surface sites in Europe and East Asia (NMBs of -80.0% to -70%) but TNO₂ is overpredicted (NMBs of 44.9-47.8%). The large errors and poor spatial correlations at the surface sites suggest inaccuracies in the spatial allocation of emissions.

The disparity between the TNO₂ and surface NO₂ levels suggests errors in the vertical structure of NO₂ within the CESM-NCSU model. Such errors could include inaccurate magnitudes and vertical distributions of NO₂ emissions, excess lofting of NO₂ into the higher levels of the troposphere, which is consistent with the overpredicted TNO₂, or excess conversion of NO₂ to HNO₃, which is consistent with the overpredictions of HNO₃ and O₃. Similar errors in the vertical distributions of NO₂ emissions in 2005 over East Asia were reported by Zhang et al. [2015a]. Zhang et al. [2015b] further showed that the adjusted magnitudes and vertical distributions of NO₂ emissions resulted in moderate to large improvement in simulated surface concentrations and column mass abundances of species in terms of domain mean performance statistics at individual sites. The zonal mean profiles show that despite these issues the RCP4C and RCP8C simulations capture the latitudinal changes in TNO₂, which is reflected in the high spatial correlation (R values of 0.82-0.86). The RCP8C and RCP4C simulations overpredict TNO₂ to a greater extent and underpredict surface NO₂ to a lesser extent than the HE15 simulation due to greater global average NO₂ emissions in the RCP emission inventories. The absolute differences in the emissions of CO, HCHO, NO, SO₂, NH₃, AVOC, BC, and POC between the RCP4.5 and RCP8.5 current emission inventories and the emission inventory of HE15 are shown in Figures S2 and S3 in the supplementary material, respectively. HNO₃ is largely overpredicted in the continental U.S. (CONUS) region (NMBs of 82.7-93.1%) but only slightly overpredicted in Europe (NMBs of 0.4-12.3%). However, the small biases in Europe do not indicate excellent performance but are most likely from compensation errors, as evidenced by the high NMEs (63.6-71.2%) and low R values (~0.31). The errors in HNO₃ likely stem from the errors in NO₂ as its poor spatial performance in Europe is similar to that of NO₂ and also likely reflects the inaccuracies in the spatial allocation of emissions. As mentioned previously,

the overpredictions of HNO_3 are consistent with overpredicted oxidation and may be potentially related to the inaccuracies in deposition and overestimated NO emissions, since the strong model performance of NO_3^- does not indicate inaccuracies in gas-to-aerosol partitioning. Poor performance of HNO_3 is also common in regional air quality models due to the strong sensitivity of HNO_3 levels to the concentrations of SO_4^{2-} , NH_3 , and NH_4^+ , emissions, and simulated meteorology [Yu *et al.*, 2005].

O_3 is largely overpredicted in the RCP4C and RCP8C simulations across all NH regions (NMBs of 39.8-76.0%) with greater overpredictions in East Asia and Europe compared to CONUS. The overprediction in O_3 is primarily caused by insufficient O_3 titration and underpredictions in O_3 deposition. The underprediction of O_3 titration by CESM-NCSU has been reported in previous studies on global scale [He and Zhang, 2014; He *et al.*, 2015a] and regional scale [Zhang *et al.*, 2016a, b] and is linked to the underprediction of surface NO_x concentrations mentioned above. The O_3 overprediction is also caused in part by inaccuracies in O_3 deposition within CESM. In particular, the stomatal and leaf resistances in the CESM model are not linked to the leaf area index, which is an oversight that results in positive O_3 biases in both the MOZART-4 and CESM models [Val Martin *et al.*, 2013]. An additional possibility for the overprediction in O_3 is excess mixing of O_3 from higher levels of the troposphere to the surface due to the relative thickness of the CESM first model layer (~99-130 m). Despite the large overpredictions of surface O_3 , TOR is generally well simulated (NMBs of ~ 12.0%, NMEs of ~17.0%, and R values of ~0.8). The zonal mean profile of TOR in the RCP4C and RCP8C simulations is well captured in the SH and lower latitudes of the NH, but largely overpredicted in the NH mid-latitudes. This is consistent with O_3 deposition as the primary cause of the overprediction, since O_3 levels and land mass are both maximized in the NH mid-latitudes. In

general surface O₃ overpredictions are greater than those of HE15 due to the slightly larger NO emissions from the RCP inventories. This emission difference helps to reduce the underpredicted TOR in the SH and lower latitudes of the NH but further enhances the overprediction of NH mid-latitudes.

SO₂ is underpredicted in Europe and East Asia (NMBs of -57.4% to -29.2%). These underpredictions likely reflect either overestimations of deposition or underestimation of emissions as the strong performance of SO₄²⁻ does not indicate that the errors are arising from gas-to-particle partitioning. SO₂ in the CONUS region is largely overpredicted (NMBs of 85.4-112.0%). This large overprediction of SO₂ in CONUS and underpredictions of SO₂ in the other NH locations mirror the performance statistics of HE15, although the underpredictions in Europe and East Asia are larger and the overprediction in CONUS is reduced. These reductions in biases of SO₂ reflect both reduced total column emissions of SO₂ in the RCP inventory compared to the HE15 inventory and reduced emissions of SO₂ in the surface layer in the vertically-structured RCP emissions, which inject more SO₂ at higher altitudes. The large overpredictions of SO₂ in CONUS are attributed to uncertainties in wet deposition, measurements, and emissions in this region and the model performance has been shown to be very sensitive to changes in SO₂ emission levels [*He and Zhang, 2014; He et al., 2015a*]. TSO₂ levels are generally underpredicted against SCIAMACHY satellite estimates (NMBs of -42.9% to -33.7%), with large errors and poor correlations (NMEs of 96.4-98.7% and R values of ~-0.8). This poor performance against the SCIAMACHY estimates is more likely associated with uncertainties in the satellite retrievals than with the model errors as TSO₂ satellite products are relatively new and subject to very large errors and biases especially for the year 2010, which makes them relatively unsuitable for use in data assimilation systems or model validation [*Fioletov et al.,*

2013]. NH_3 concentrations in Europe are overpredicted in the RCP4C and RCP8C simulations (NMBs of 71.2-110.2%). A weaker overprediction was reported in HE15 and was attributed to uncertainties in European emissions of NH_3 . It is difficult to ascertain whether or not the increased overprediction in NH_3 is related to differences in the RCP inventories compared to the HE15 inventory as the spatial allocations of NH_3 appear different in both inventories, reflected in the dipole differences of NH_3 emissions in Figures S2 and S3. Extracting the average emission differences at the monitoring site locations reveals that NH_3 emission fluxes are approximately $1.7 \times 10^7 \text{ molec. cm}^{-2} \text{ s}^{-1}$ less in the RCP4.5 inventory and $5.2 \times 10^9 \text{ molec. cm}^{-2} \text{ s}^{-1}$ greater in the RCP8.5 inventory compared to the HE15 inventory. This difference and the greater NH_3 emissions in the RCP8.5 inventory compared to the RCP4.5 inventory explain why the RCP8C has a much greater overprediction compared to both the RCP45 and HE15 simulations. The exact cause of the overprediction in the RCP4C simulation compared to the HE15 simulation is not apparent and may simply be related to compensation effects from different emission fluxes in grid cells surrounding the monitoring sites. Overall, the simulation of trace gases in the RCP4C and RCP8C reasonably represents the conditions of the current atmosphere within model constraints. In addition to uncertainties in emissions and deposition of these species that seem to be the primary driving force for deviations from observations, the CESM-NCSU simulations use fairly coarse horizontal ($\sim 0.9^\circ \times 1.25^\circ$) and vertical resolution (30 layers). The model first layer with heights of ~ 99 -130 m is also considerably deeper than most regional air quality models that range in height from 25-45m [Zhang *et al.*, 2013]. The coarse grid resolution makes the model more susceptible to numerical diffusion, which in part accounts for underpredictions of many surface gases (e.g., CO , NO_2 , and SO_2) and possibly the overprediction of O_3 .

SO_4^{2-} , NH_4^+ , and NO_3^- concentrations are all well simulated in the RCP4C and RCP8C simulations over the CONUS and European regions with NMBs generally within the range of $\pm 18.0\%$. The exception is NH_4^+ over Europe that is moderately overpredicted (NMBs of 33.0-42.0%) in response to overpredictions of European NH_3 . It is important to note that all particle speciation measurements from Europe used in this study represent a size range consistent with particulate matter with diameter less than and equal to $10\text{ }\mu\text{m}$ (PM_{10}), while chemical speciation measurements from CONUS represent a size range consistent with $\text{PM}_{2.5}$. $\text{PM}_{2.5}$ from the CESM-NCSU MAM7 module is assumed to represent all aerosols within the Aiken, accumulation, primary carbon, fine dust, and fine sea-salt modes. With the exception of primary carbon mode, these are the only modes that treat inorganic aerosol thermodynamics. The coarse dust and coarse sea-salt modes treat only irreversible condensation of H_2SO_4 , NH_3 , HNO_3 , and HCl , which may likely lead to overestimations of these species in PM_{10} measurements [He and Zhang, 2014]. These factors explain the discrepancies between particle speciation model performance in the Europe and CONUS regions found for NH_4^+ and Cl^- which will be discussed later in this section. Errors in the simulation of SO_4^{2-} , NH_4^+ , and NO_3^- are moderate (NMEs $\sim 25\text{-}60\%$) and correlations are reasonably high (0.52-0.87), indicating the spatial distributions of these inorganic aerosol species are captured reasonably well. Compared to the simulations of HE15, SO_4^{2-} levels are enhanced in the RCP4C and RCP8C simulations and generally in better agreement with the observations in terms of bias. This is likely caused greater oxidation related to greater O_3 levels and NO emissions. NH_4^+ levels are comparable to those of HE15 but the bias is reduced from reduced NH_3 emissions in portions of Europe and the eastern U.S. The reduced overprediction of NH_4^+ and reduced underprediction of SO_4^{2-} both serve to reduce the overprediction of NO_3^- compared to the HE15 simulation by reducing the free NH_4^+ available to

from NH_4NO_3 , which considerably improves NO_3^- performance in terms of bias and error.

Coarse NO_3^- concentration in Europe is likely not overpredicted due to the small bias in HNO_3 in this region.

Cl^- is simulated poorly in both the RCP4C and RCP8C simulations. Cl^- is largely overpredicted (NMBs ~450-500%) in Europe, with similar large errors. The R values are high (0.73-0.75) for Cl^- , indicating that the majority of the errors are caused by the overprediction rather than errors in spatial distribution. These overpredictions of Cl^- are likely caused by the overpredicted WSP10 in marine regions that result in excess emissions of sea-salt and also as a result of the irreversible condensation of HCl . In the CONUS region Cl^- is underpredicted (NMBs of -22.0- -25.7%) with large errors (NMEs of 94%), and poor spatial correlation (R values of 0.17-0.20). These underpredictions are likely driven by either underestimates of HCl emissions or the overprediction of HNO_3 , which competes against HCl for partitioning into the particulate phase. The Cl^- performance of the RCP4C and RCP8C simulations is comparable to that of HE15.

BC, OC, and TC concentrations in the CONUS region are all underpredicted in the RCP4C and the RCP8C simulations (NMBs of ~-25- -59%), with moderate errors (NMEs of ~34-60%), and moderate spatial correlations (R values of 0.45-0.58). OC concentrations in Europe are strongly underpredicted (NMBs of ~-82%). This strong underprediction also occurs for HOA, OOA, and TOA, compared against data from the *Zhang et al.* [2007] and *Jimenez et al.* [2009] datasets (NMBs of ~ -93% to -65%). However, despite this large bias, there are moderate correlations for OC in Europe (0.46-0.51) and a strong correlation for OOA, HOA, and TOA (0.85-0.94), indicating that the spatial pattern of these species is reasonably captured. The strong underpredictions of carbonaceous aerosols in the RCP4C and RCP8C simulations are related to

not only the low emissions of BC, POC, and AVOC in the RCP inventories compared to other global inventories [Liu *et al.*, 2012; Glotfelty *et al.*, 2014], but also because the rapid aging of BC and POA within the MAM7 aerosol module leads to very efficient wet removal compared to other global models [Liu *et al.*, 2012]. The large underprediction of HOA is also linked to an overpredicted volatility of POA within the VBS treatment (Glotfelty *et al.*, 2017). The errors and R values in the CONUS region are comparable to those from the HE15 simulation but there is a significant enhancement in the underpredictions. This is partially because the RCP biomass burning emissions are distributed vertically in the RCP4C and RCP8C simulations, which reduces surface OC and BC levels compared to the HE15 simulation where all emissions are injected in the bottom model layer. The levels of BC and OC are also reduced in the RCP4C and RCP8C simulations due to large reductions in the emissions of BC, POC, and AVOC in the RCP emission inventories throughout the CONUS region compared to the HE15 inventory. Although HE15 did not evaluate against HOA, OOA, TOA, and European OC data, the underpredictions in the RCP4C and RCP8C simulations are considerably larger than those shown in the simulations of Glotfelty *et al.* (2017), which utilize the same emission inventory as HE15. This is also attributed to the reduced POC and AVOC emissions in the RCP inventories. SOA in the CONUS region performs well against observations from the Lewondowski *et al.* [2013] dataset (NMBs of -15.6% to -8.1%, NMEs of ~33-34%, and R values of 0.51-0.62). This is similar to the findings reported in Glotfelty *et al.* (2017) and reflects improved SOA performance of the OA updated treatments, however, caution must be taken when comparing SOA from CESM-NCSU to Lewondowski *et al.*, [2013], as the anthropogenic and biogenic precursors used to derive observational SOA are not identical to those that contribute to SOA in CESM-NCSU.

Total PM_{2.5} is well simulated over the CONUS region (NMBs of -6.4- -11.3%, NMEs of ~34%, and R values of 0.67), while PM_{2.5} in Europe is reasonably well simulated in terms of bias (NMBs of -32.1% to -9.9%) and errors (NMEs of 31.9-36.1%) but has a poor correlation (0.03-0.06). This poor correlation is similar to other chemical species in Europe and again indicates possible inaccuracies in the spatial allocation of emissions. The larger underprediction in the RCP8C simulation in Europe is due to reduced emissions of all primary particles and PM_{2.5} precursors, except NH₃, in that region compared to the RCP4.5 inventory. Based on the evaluation of the various PM_{2.5} species, the underpredictions can be attributed to the large underpredictions of primary carbonaceous aerosol. These results are generally comparable to PM_{2.5} in the HE15 current decadal simulation, although the underpredictions are larger in response to greater underpredictions of carbonaceous aerosols and reduced overpredictions of inorganic aerosols. PM₁₀ in the CONUS region is not well simulated (NMBs of -43.5% to -42.7%), NMEs of 51.7-55.6%, and R values of 0.05-0.17). This large inconsistency compared with the simulation of PM_{2.5} suggests that coarse particles are not well simulated in the CONUS region, which is likely linked to inaccuracies in the emissions of dust and also because the inorganic aerosol thermodynamics is not treated for these particles. PM₁₀ in Europe is reasonably well simulated (NMBs of -5.4 to 15.3%, NMEs of 29.5-35.5%, and R values of ~0.22) and consistent with the performance of PM_{2.5}. PM₁₀ in East Asia is underpredicted (NMBs of ~-56%) with the majority of the errors explained by the bias. The high R values (~0.67) suggest that the spatial pattern is reasonably well captured. The underprediction of PM₁₀ in East Asia likely results from either underestimations of emissions in this region or from underpredicted dust emissions from underpredicted WSP10 over land areas. Similar to PM_{2.5}, the simulation of PM₁₀

is comparable to that of the HE15 current decadal simulation accounting for emissions differences.

3.3 Temporal Evaluation of Regional Trace Gases, Aerosol, and Aerosol-Cloud Interactions

Figure 3 shows the time series of monthly averages from the RCP4C and RCP8C simulations for $PM_{2.5}/PM_{10}$, THCHO, TNO₂, and TOR, as well as the monthly average of the trace gas satellite estimates over five stratocumulus regions of *Rausch et al.* [2010]. Figure 4 compares similar time series of simulated and satellite-estimated CCN5, CDNC, AOD, and SWCF. In general, PM_{10} and $PM_{2.5}$ are comparable in both the stratocumulus regions of NAM and SAM. However, the aerosol sources are likely slightly different with the NAM region in the vicinity of greater anthropogenic emissions originating from Mexico and the SAM region in the vicinity of greater SOA from biogenic emissions in the Amazon and greater biomass burning emissions. The stratocumulus regions of SAF and SEA have greater aerosol loadings for both PM_{10} and $PM_{2.5}$ and stronger seasonality than the other regions. The enhanced aerosol loadings originate from greater anthropogenic emissions, greater biomass burning emissions, and greater dust emissions in the vicinity of these regions as evidenced by the spatial distributions of RCP4.5 emissions and natural emissions shown in Figure S4 of the supplementary material. In SEA, aerosol loadings peak in the boreal spring months (i.e., March, April, and May), this corresponds to the seasonality of dust storms in East Asia that occur primarily during this season [*Zhao et al.*, 2006]. In the SAF region, there are bi-modal peaks in aerosol loading with the largest peak in the boreal winter months (i.e., December, January, and February) and a smaller peak in the boreal summer months (i.e., June, July, and August). This bimodal seasonality is related to the movement of the intertropical convergence zone (ITCZ) and its subsequent impact on African

biomass burning emissions. During the boreal winter months (DJF), the ITCZ moves southward, resulting in a dry season and greater fires in the savannas below the Sahara, while during the boreal summer months (JJA) the ITCZ moves northward, resulting in a dry season and greater fire activity in the savannas of southern Africa [Cahoon *et al.*, 1992]. The peak in DJF is larger because the emissions from the savannas below the Sahara are larger in the RCP emission inventory and closer to the stratocumulus region. The AUS region represents relatively clean conditions as evidenced by the large gap between $PM_{2.5}$ and PM_{10} , since natural aerosol sources typically contribute more to the PM_{10} size range [Artinano *et al.*, 2003]. This region also has prevailing winds from the Southern Ocean, thus anthropogenic aerosols can only originate from either shipping and aircraft emissions or limited outflow from Australia. Overall the predictions of $PM_{2.5}$ and PM_{10} between the RCP4C and RCP8C simulations are similar in most regions, except SAF where aerosol loading from the RCP4.5 are greater during the first half of the current climate period (2001-2006) but less during the latter half of the current climate period (2007-2010).

THCHO in all stratocumulus regions is generally underpredicted as shown in the annual average statistics and can be attributed to uncertainties in emissions and secondary formation of HCHO. In the NAM region, the CESM-NCSU model fails to capture the seasonality of the satellite estimates and generally oscillates with peaks during JJA that are comparable to satellite estimates and valleys during DJF that show large deviations from satellite estimates. This pattern is related to the dual ITCZ bias that is persistent in all global models included in the CMIP5 project [Oueslati and Bellon, 2015]. Due to this ITCZ bias, CESM underpredicts precipitation in the NAM region during the boreal spring (MAM) and JJA periods and overestimates precipitation during the boreal autumn (i.e., September, October, and November) and DJF

periods, as shown in Figure S5. The impact of this bias is an artificial seasonality of THCHO within CESM-NCSU due to wet removal errors. In the SAM region, the simulated THCHO is comparable to satellite estimates during the 2001-2005 period but does not capture the observed trend of increasing THCHO in the 2006-2010 period. This indicates that the model is likely failing to capture either increasing trends in biogenic emissions or biomass burning emissions during the current climate period. However, considering the proximity of the SAM stratocumulus region to the Amazon and the lack of a similar trend in AOD and CCN5 from OC and BC in satellite estimates, it is likely that the issue is related to inaccuracies in biogenic emissions. This is consistent with findings [Holm *et al.*, 2014], namely, the MEGAN scheme within the CESM-NCSU land model does not adequately capture the climatic and biophysical process that control biogenic emissions of isoprene in the Amazon and thus may not capture the trends in biogenic emissions of HCHO or isoprene that contribute to secondary formation of HCHO. In the SAF region, CESM-NCSU fails to capture the trend of increasing THCHO but adequately captures the seasonality of THCHO satellite estimates despite underpredicted THCHO levels. The general underprediction is likely linked to overestimated wet removal as PR is consistently overpredicted in this region and the failure to capture the trend is likely linked to errors in biogenic emissions of HCHO and isoprene, similar to the SAM region. In the SEA region, THCHO is generally comparable to satellite estimates in JJA and the boreal autumn (SON) seasons but largely underpredicted during the DJF and MAM seasons, which is related to wet removal errors from the dual ITCZ bias. In the AUS region, CESM-NCSU predicted THCHO has an inverted artificial seasonality compared to SEA and NAM, resulting from overpredicted PR in JJA and underpredicted PR in DJF. The disparity in the seasonal patterns in this region likely also

suggests poor model performance from either neglecting natural marine sources of HCHO emissions from phytoplankton or underestimated shipping emissions.

The seasonal variations of TNO₂ are captured in the NAM, SEA, and AUS regions. TNO₂ is generally overpredicted in the NAM and AUS regions, which is consistent with the global statistics and likely linked to overestimated NO emissions in the vicinity of these regions. In the SEA region, the TNO₂ levels are generally consistent with satellite estimates and generally capture the slight upward trend in TNO₂ in both the RCP4C and RCP8C simulations, however, the RCP8C simulation generally captures the seasonality of TNO₂ better than the RCP4C simulation due to higher NO emissions. In the SAF and SAM regions TNO₂ is well simulated during the JJA months but the reductions in TNO₂ levels shown in satellite estimates during the DJF months are not captured. This suggests errors in photochemistry during this period that could be potentially linked to inaccuracies in biogenic emissions of VOCs, since both regions are in close proximity to the largest sources of biogenic VOCs, and photochemistry would be most active during this period since solar radiation is at its maximum.

TOR abundance and seasonality are generally well simulated against satellite estimates in all regions, although strong deviations exist in SON. The deviations in SON do not correspond well with deviations in TNO₂, which suggests poor model performance during this period is related to model dynamics rather than chemistry. SON is the season during which the exchange of mass between the troposphere and stratosphere (STE) is the largest, with the true maximums occurring late autumn in the NH and early autumn in the SH [Skerlack *et al.*, 2014]. In the NH regions of SEA and NAM, TOR is overestimated during this period, indicating a likely overestimation of STE in the NH, while in the SH regions of SAM, SAF, and AUS, TOR is underpredicted, indicating a likely underestimation of STE. The performance in these regions,

with the exception of SEA and AUS, is generally consistent with other comparisons of global model output against satellite estimates [Ziemke *et al.*, 2006].

AOD in all five regions is well simulated in terms of magnitude and seasonality. This is especially true in the NAM, SAM, and AUS regions, with the small deviations from satellite estimates that likely result from uncertainties in sea-salt emissions. In the SAF region, AOD is mostly well simulated but peak levels in the JJA period and the increasing trend in these peaks are not captured. These peak levels in JJA occur because dust transport from North Africa towards this region occurs at higher altitudes and thus has a greater impact on AOD [Gama *et al.*, 2015]. Therefore, it is likely that the underprediction of the peak AOD and the trend are related to the too efficient wet removal process that impacts aerosols within the MAM7 model [Liu *et al.*, 2012]. AOD is underpredicted in SEA, likely due to underestimates of emissions of anthropogenic PM components such as OC and BC as well as PM precursors SO₂, NO, and NH₃ in the region. This finding is also consistent with the underprediction of PM₁₀ in East Asia in this work and during 2005, 2006, 2011 reported in Chen *et al.* [2015] and Zhang *et al.* [2015, 2016a, b].

CCN5 is well simulated in both the NAM and SAF regions, although the JJA maximum is not fully captured in the SAF region which is similar to AOD. CCN5 is generally underpredicted in both the SEA and AUS regions mirroring the global statistics. The underpredictions in SEA are likely from underestimated anthropogenic emissions of PM components such as OC and BC as well as PM precursors SO₂, NO, and NH₃ in that region, while the underprediction in the AUS region is likely related to neglecting marine organic aerosols. In the SAM region, CCN5 is generally comparable to satellite estimates but the seasonality is not captured. This poor seasonality mirrors the poor seasonality of TNO₂ and could

thus be linked to errors in photochemistry that propagate into SOA formation and subsequently new particle formation due to the inclusion of the organic gas/sulfuric acid conucleation parameterization. CDNC is overpredicted in all regions; consistent with the global statistics, but generally the simulation in all five regions captures the seasonality of CDNC. The variabilities in MODIS-derived and the simulated CDNC are not an exact match, but the CESM-NCSU model does at least capture the increased variability in CDNC compared to other variables. The magnitude of the overprediction is the largest in both the SAF and SEA regions, with the largest deviations from satellite estimates during JJA in SAF and MAM in SEA, when dust levels are maximized. It is further evidence that the aerosol activation parameterization within CESM-NCSU overpredicts aerosol activation of insoluble aerosols. Despite the deviations from satellite estimates of CDNC, SWCF is very well simulated in terms of both magnitude and seasonality in all five stratocumulus regions. However, there is a slight overprediction of SWCF in SEA, likely because CDNC levels in the region are the largest amongst the five regions.

4. Summary and Conclusions

In this work, the CESM-NCSU model is used to simulate the current atmosphere using emissions based on the RCP4.5 and RCP8.5 scenarios. The tuned version of CESM-NCSU simulates radiation variables consistent with satellite estimates from CERES/EBAF. Deviations from CERES-EBAF estimates indicate that CESM-NCSU overpredicts tropical cloudiness, while underpredicting the greenhouse effect and overpredicting surface albedo in the polar regions. Compared to satellite estimates, cloud parameters within CESM-NCSU are not as well simulated as radiation. This is common in many global modeling studies, indicating that more work is

needed to better represent and link these individual processes. AOD is well simulated by CESM-NCSU given model limitations; and it generally performs better or comparable to CMIP5. CCN5 and CDNC in CESM-NCSU are not as well simulated as AOD owing to uncertainties in aerosol treatments and satellite estimates. Meteorological parameters including T2, Q2, WSP10, and PR are all in good agreement with observations or reanalysis estimates, although there are deviations that result from errors in simulated radiation and the dual ITCZ bias.

In terms of atmospheric pollutants, CESM-NCSU generally captures the most features of the spatial distribution and the magnitudes of TNO₂, TOR, TCO, and THCHO. CESM-NCSU shows limited skills in simulating surface O₃ mixing ratios because of possible underestimations in O₃ deposition from the default model as released by NCAR, leading to moderate to large overpredictions. Overall, trace gas performance at surface sites is worse than the performance of column mass abundances. This may be due to inaccuracies in emissions, vertical mixing, and deposition but this evaluation is also limited by the use of a coarse spatial grid resolution in the simulations using the CESM-NCSU and subsequent numerical diffusion. In general, the concentrations of inorganic aerosol SO₄²⁻, NH₄⁺, and NO₃⁻ are all well simulated by CESM-NCSU, while the simulation of Cl⁻ concentrations is poor due to errors in WSP10 and competition of HCl with overpredicted HNO₃ in aerosol partitioning. The concentrations of carbonaceous aerosols are underpredicted due to the small magnitude in the emissions of the BC, OC, and AVOC in the RCP emission inventories, overestimates of POA volatility, and possible overpredictions in wet removal of these species by the MAM7 module. This underprediction in the concentrations of carbonaceous aerosols and the good performance of inorganic aerosols lead to underpredictions of PM_{2.5} and PM₁₀ using the RCP emission inventory in CESM-NCSU.

The evaluation of trace gas columns in the five stratocumulus cloud regions indicate that inaccuracies exist in tropical biogenic emissions within CESM-NCSU that may impact photochemistry in these regions and that the ITCZ bias likely plays an important role in the errors in simulating soluble trace gases in the tropics. Large biases in TOR levels during the SON period also imply biases in the STE of O₃ within the CESM-NCSU model. AOD, CCN₅, and SWCF are all reasonably well simulated in these stratocumulus regions although some errors exist, and they can be attributed to the possible overpredictions of aerosol wet removal, underestimated emissions, or inaccuracies in tropical photochemistry. Compared to satellite estimates, the seasonality of CDNC in these regions is captured despite its overprediction. Given the potential inaccuracies in satellite CDNC estimates and the good performance of other variables, the evaluation of CDNC implies that the CESM-NCSU model represents aerosol-cloud and radiation processes reasonably well.

Overall, the CESM-NCSU model does a reasonably good job in representing the current climate period in terms of radiation, clouds, meteorology, trace gases, aerosols, and aerosol-cloud interactions, making it suitable for use in predicting changes in these properties under future climate policy scenarios. These future climate policy scenario projections will be resented in Part II.

Acknowledgements

This work is supported by the National Sciences Foundation (NSF) Earth System Modeling Program Grant No. AGS-1049200 at NCSU. We would like to acknowledge high-performance computing support from Yellowstone ([ark:/85065/d7wd3xhc](https://www.nsls.org/ark:/85065/d7wd3xhc)) provided by NCAR's Computational and Information Systems Laboratory, sponsored by the National Science

Foundation. The authors would like to thank Brett Gantt and Xin Zhang, former members of Air Quality Forecasting Lab, NCSU, for implementing advanced aerosol activation scheme into CAM5.1. MODIS data and CERES data are provided by NASA via <http://ladsweb.nasa.gov/data/search.html> and http://ceres.larc.nasa.gov/order_data.php, respectively. Other surface network data were downloaded from their respective web sites. For simulation results presented in this paper, please contact the corresponding author.

References

- Ahmadov, R., S. A. McKeen, A. L. Robinson, R. Bahreini, A. M. Middlebrook, J. A. de Gouw, J. Meagher, R.-Y. Hsie, E. Edgerton, S. Shaw, and M. Trainer (2012), A volatility basis set model for summertime secondary organic aerosols over the eastern United States in 2006, *J. Geophys. Res.*, *117*, D06301, doi:10.1029/2011JD016831.
- Artinano, B., P. Salvador, D. G. Alonso, X. Querol, and A. Alastuey (2003), Anthropogenic and natural influence on the PM₁₀ and PM_{2.5} aerosol in Madrid (Spain). Analysis of high concentration episodes, *Environmental Pollution*, *126*(3), 453-465, doi:10.1016/S0269-7491(03)00078-2.
- Barahona, D. and A. Nenes (2007), Parameterization of cloud droplet formation in large-scale models: including effects of entrainment, *J. Geophys. Res.*, *112*, D16206, doi:10.1029/2007JD008473.
- Barahona, D., R. E. I. West, P. Steir, S. Romakkaniemi, H. Kokkola, and A. Nenes (2010), Comprehensively accounting for the effect of giant CCN in cloud activation parameterizations, *Atmos. Chem. Phys.*, *10*, 2467-2473, doi:10.1029/2006JD007547.
- Cahoon, D. R., B. J. Stocks, J. S. Levine, W. R. Cofer, and K. P. O'Neill (1992), Seasonal distributions of African savanna fires, *Nature*, *359*, 812-815, doi:10.1038/359812a0.
- Chen, Y., Y. Zhang, J. Fan, L.R. Leung, Q. Zhang, and K.-B. He (2015), Application of an Online-Coupled Regional Climate Model, WRF-CAM5, over East Asia for Examination of Ice Nucleation Schemes: Part I. Comprehensive Model Evaluation and Trend Analysis for 2006 and 2011, *Climate*, *3*(3), 627-667; doi:10.3390/cli3030627.
- Dentener, F., D. Stevenson, K. Ellingsen, M. Amann, C. Atherton, N. Bell, D. Bergmann, I. Bey, L. Bouwman, T. Butler, J. Cofala, B. Collins, J. Drevet, R. Doherty, B. Eickout, H. Eskes, A. Fiore, M. Gauss, D. Hauglustaine, L. Horowitz, I. S. A. Isaken, B. Josse, M.

- Lawrence, M. Krol, J. F. Lamarque, V. Montanaro, J. F. Muller, V. H. Peuch, G. Pitari, J. Pyle, S. Rast, J. Rodriguez, M. Sanderson, N. H. Savage, D. Shindell, S. Strahan, S. Szopa, K. Sudo, R. Van Dingenen, O. Wild, and G. Zeng (2006), The global atmospheric environment for the next generation, *Environ. Sci. Technol.*, *40*, 3586-3594.
- Donahue, N. M., A. L. Robinson, C. O. Stanier, and S. N. Pandis (2006), Coupled partitioning, dilution, and chemical aging of semivolatile organics, *Environ. Sci. Technol.*, *40*, 2635-2643, doi:10.1021/es052297c.
- Emmons, L. K. S. Walters, P. G. Hess, J.-F. Lamarque, G. G. Pfister, D. Fillmore, C. Granier, A. Guenther, D. Kinnison, T. Laepple, J. Orlando, X. Tie, G. Tyndall, C. Wiedinmyer, S. L. Baughcum, and S. Kloster (2010), Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), *Geosci. Model. Dev.*, *3*, 43-67, doi:10.5194/gmd-3-43-2010.
- Fan, J., R. Zhang, D. Collins, and G. Li (2006), Contribution of secondary condensable organics to new particle formation: A case study in Houston, Texas, *Geophys. Res. Lett.*, *33*, L15802, doi:10.1029/2006GL026295.
- Fan, J., S. Ghan, M. Ovchinnikov, X. Liu, P. J. Rasch, and A. Korolev (2011), Representation of Arctic mixed-phase clouds and the Wegener-Bergeron-Findeisen process in climate models: Perspectives from a cloud-resolving study, *J. Geophys. Res.*, *116*, D00T07, doi:10.1029/2010JD015375.
- Flanner, M. G., C. S. Zender, J. T. Randerson, and P. J. Rasch (2007), Present-day climate forcing and response from black carbon in snow, *J. Geophys. Res.*, *112*, D11202, doi:10.1029/2006JD008003.

- Fioletov, V. E., C. A. McLinden, N. Krotkov, K. Yang, D. G. Loyola, P. Valks, N. Theys, M. Van Roozendaal, C. R. Nowlan, K. Chance, X. Liu, C. Lee, and R. V. Martin (2013), Application of OMI, SCIAMACHY, and GOME-2 satellite SO₂ retrievals for detection of large emission sources, *J. Geophys. Res. Atmos.*, *118*, 11,399–11,418, doi:10.1002/jgrd.50826.
- Fiore, A. M., V. Naik, D. V. Spracklen, A. Steiner, N. Unger, M. Prather, D. Bergmann, P. J. Cameron-Smith, I. Cionni, W. J. Collins, S. Dalsoren, V. Eyring, G. A. Folberth, P. Ginoux, L. W. Horowitz, B. Josse, J.-F. Lamarque, I. A. MacKenzie, T. Nagashima, F. M. O'Connor, M. Righi, S. T. Rumbold, D. T. Shindell, R. B. Skeie, K. Sudo, S. Szopa, T. Takemura, and G. Zeng (2012), Global air quality and climate, *Chem. Soc. Rev.*, *41*(19), doi:10.1039/C2CS35095E.
- Fountoukis, C. and A. Nenes (2005), Continued development of a cloud droplet formation parameterization for global climate models, *J. Geophys. Res.*, *110*, D11212, doi:10.1029/2004jd005591.
- Fountoukis, C. and A. Nenes (2007), ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K⁺-Ca²⁺-Mg²⁺-NH₄⁺-Na⁺-SO₄²⁻-NO₃⁻-Cl⁻-H₂O aerosols, *Atmos. Chem. Phys.*, *7*, 4639–4659, doi:10.5194/acp-7-4639-2007.
- Gama, C., O. Tchepe, J. M. Baldasano, S. Basart, J. Ferreira, C. Pio, J. Cardoso, and C. Borrego (2015), Seasonal patterns of Saharan dust over Cape Verde - a combined approach using observations and modeling, *Tellus*, *67*, 24410, <http://dx.doi.org/10.3402/tellusb.v67.24410>.
- Gantt, B., J. Xu, N. Meskhidze, Y. Zhang, A. Nenes, S. J. Ghan, X. Liu, R. Easter, and R. Zaveri (2012), Global distribution and climate forcing of marine organic aerosol – Part 2: Effects

- on cloud properties and radiative forcing, *Atmos. Chem. Phys.*, *12*, 6555-6563, doi:10.5194/acp-12-6555-2012.
- Gantt, B., J. He, X. Zhang, Y. Zhang, and A. Nenes (2014), Incorporation of advanced aerosol activation treatments into CESM/CAM5: model evaluation and impact on aerosol indirect effects, *Atmos. Chem. Phys.*, *14*, 7485-7497, doi: 10.5194/acp-14-7485-2014.
- Gantt, B., T. Glotfelty, N. Meskhidze, and Y. Zhang (2015), Simulating the impacts of marine organic emissions on global atmospheric chemistry and aerosols using an online-coupled meteorology and chemistry model, *Atmospheric and Climate Sciences*, *5*, 266-274, doi: 10.4236/acs.2015.53020.
- Glotfelty, T., Y. Zhang, P. Karamchandani, and D. G. Streets (2014), Will the role of intercontinental transport change in a changing climate?, *Atmos. Chem. Phys.*, *14*, 9379-9402, doi:10.5194/acp-14-9379-2014.
- Glotfelty, T., J. He, and Y. Zhang (2017), New Organic Aerosol Treatments in CESM/CAM5: Development and Initial Application, *Journal of Advances in Modeling Earth Systems*, in review..
- Guenther, A., T. Karl, P. Harley, C. Wiedinmyer, P. I. Palmer, and C. Geron (2006), Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), *Atmos. Chem. Phys.*, *6*, 3181-3210, doi: 10.5194/acp-6-3181-2006.
- Haines, A., R. S. Kovats, D. Campbell-Lendrum, and C. Corvalen (2006), Climate change and human health: Impacts, vulnerability and public health, *Public Health*, *120*(7), 585–596, doi:10.1016/j.puhe.2006.01.002.

- Hannay, C., C. Jackson, G. Danabasoglu, J.-F. Lamarque, D. Lawrence, K. Lindsay, R. Neale, P. Rasch, and J. Trbbia (2014), Coupled model initialization and tuning, presentation at the 19th Annual CESM Workshop, June 16-19 , 2014, Breckenridge, CO.
- He, J. and Y. Zhang (2014), Improvement and further development in CESM/CAM5: gas-phase chemistry and inorganic aerosol treatments, *Atmos. Chem. Phys.*, *14*, 9171-9200, doi: 0.5194/acp-14-9171-2014.
- He, J., Y. Zhang, T. Glotfelty, R. He, R. Bennartz, J. Rausch, and K. Sartelet (2015a), Decadal simulation and comprehensive evaluation of CESM/CAM5.1 with advanced chemistry, aerosol microphysics, and aerosol-cloud interactions, *J. Adv. Model. Earth Syst.*, *7*, 110–141, doi:10.1002/2014MS000360.
- He, J., Y. Zhang, S. Tilmes, L. Emmons, J.-F. Lamarque, T. Glotfelty, A. Hodzic, and F. Vitt (2015b), CESM/CAM5 improvement and application: comparison and evaluation of updated CB05_GE and MOZART-4 gas-phase mechanisms and associated impacts on global air quality and climate, *Geoscientific Model Development*, *8*, 3999-4025, doi:10.5194/gmd-8-3999-2015.
- Holm, J. A., K. Jardine, A. B. Guenther, J. Q. Chambers, and E. Tribuzy (2014), Evaluation of MEGAN-CLM parameter sensitivity to predictions of isoprene emissions from an Amazonian rainforest, *Atmos. Chem. Phys. Discuss.*, *14*, 23995-24041, doi:10.5194/acpd-14-23995-2014.
- Intergovernmental Panel on Climate Change (IPCC) (2000), *Special Report on Emission Scenarios (SRES)*, A Special Report of Working Group III of the Intergovernmental Panel on Climate Change, edited by N. Nakicenovic and R. Swart, 570 pp., Cambridge Univ. Press, Cambridge, U. K.

Intergovernmental Panel on Climate Change (IPCC) (2007), *Report of the 26th session of the IPCC*. Bangkok. April 30- May 4 2007. Intergovernmental Panel on Climate Change, Geneva, Switzerland.

Jimenez, J. L., M. R. Canagaratna, N. M. Donahue, A. S. H. Prevot, Q. Zhang, J. H. Kroll, P. F. DeCarlo, J. D. Allan, H. Coe, N. L. Ng, A. C. Aiken, K. S. Docherty, I. M. Ulbrich, A. P. Grieshop, A. L. Robinson, J. Duplissy, J. D. Smith, K. R. Wilson, V. A. Lanz, C. Hueglin, Y. L. Sun, J. Tian, A. Laaksonen, T. Raatikainen, J. Rautiainen, P. Vaattovarra, M. Ehn, M. Kulmala, J. M. Tomlinson, D. R. Collins, M. J. Cubison, E. J. Dunlea, J. A. Huffman, T. B. Onasch, M. R. Alfarra, P. I. Williams, K. Bower, Y. Kondo, J. Schneider, F. Drewnick, S. Borrmann, S. Weimer, K. Demerjian, D. Salcedo, L. Cottrell, R. Griffin, A. Takami, T. Miyoshi, S. Hatakeyama, A. Shimono, J. Y. Sun, Y. M. Zhang, K. Dzepina, J. R. Kimmel, D. Sueper, J. T. Jayne, S. C. Herndon, A. M. Trimborn, L. R. Williams, E. C. Wood, A. M. Middlebrook, C. E. Kolb, U. Baltensperger, and D. R. Worsnap (2009), Evolution of organic aerosols in the atmosphere, *Science*, 326, 1525-1529, doi:10.1126/science.1180353.

Karamchandani, P., Y. Zhang, S. Y. Chen, and R. Balmori-Bronson (2012), Development and testing of an extended chemical mechanism for global through-urban applications, *Atmos. Pollut. Res.*, 3(1), 1–24, doi:10.5094/APR.2011.047.

Kay, J. E., B. R. Hillman, S. A. Klein, Y. Zhang, B. Medeiros, R. Pincus, A. Gettelman, B. Eaton, J. Boyle, R. Marchand, and T. P. Ackerman (2012), Exposing global cloud biases in the Community Atmosphere Model (CAM) using satellite observations and their corresponding instrument simulators, *J. Climate*, 25, 5190-5207, doi:10.1175/JCLI-D-11-00469.1.

- Kim, M. J., R. J. Park, C.-H. Ho, J.-H. Woo, K.-C. Choi, C.-K. Song, and J.-B. Lee (2015), Future ozone and oxidants change under the RCP scenarios, *Atmos. Environ.*, 101, 103-115, doi:10.1016/j.atmosenv.2014.11.016.
- Knote, C., A. Hodzic, J. L. Jimenez, R. Volkamer, J. J. Orlando, S. Baidar, J. Brioude, J. Fast, D. R. Gentner, A. H. Goldstein, P. L. Hayes, W. B. Knighton, H. Oetjen, A. Setyan, H. Stark, R. Thanlman, G. Tyndall, R. Washenfelter, E. Waxman, and Q. Zhang (2014), Simulation of semi-explicit mechanisms of SOA formation from glyoxal in aerosol in a 3-D model, *Atmos. Chem. Phys.*, 14, 6213-6239, doi: 10.5194/acp-14-6213-2014.
- Knote, C., P. Tuccella, G. Curci, L. Emmons, J. J. Orlando, S. Madronich, R. Baro, P. Jimenez-Guerrero, D. Luecken, C. Hogrefe, R. Forkel, J. Werhahn, M. Hirtl, J. L. Perez, R. San Jose, L. Giordana, D. Brunner, K. Yahya, and Y. Zhang (2015), Influence of the choice of gas-phase mechanism on predictions of key gaseous pollutants during the AQMEII phase-2 intercomparison, *Atmos. Environ.*, 115, 553-568, doi:10.1016/j.atmosenv.2014.11.066.
- Kumar, P., I. N. Sokolik, and A. Nenes (2009), Parameterization of cloud droplet formation for global and regional models: including adsorption activation from insoluble CCN, *Atmos. Chem. Phys.*, 9, 2517-2532, doi:10.5194/acp-9-2517-2009.
- Lamarque, J.-F., G. P. Kyle, M. Meinshausen, K. Riahi, S. J. Smith, D. P., van Vuuren, A. J. Conley, and F. Vitt (2011), Global and regional evolution of short-lived radiatively-active gases and aerosols in the Representative Concentration Pathways, *Clim. Change*, 109, 191-212, doi:10.1007/s10584-011-0155-0.
- Lamarque, J.-F., K. K. Emmons, P. G. Hess, E. E. Kinnison, S. Tilmes, P. Vitt, C. L. Heald, E. A. Holland, P. H. Lauritzen, J. Neu, J. J. Orlando, P. J. Rasch, and G. K. Tyndall (2012),

- CAM-chem: Description and evaluation of interactive atmospheric chemistry in CESM, *Geosci. Model Dev.*, 5, 369–411, doi:10.5194/gmd-5-369-2012.
- Lauer, A. and K. Hamilton (2013), Simulating clouds with global climate models: A comparison of CMIP5 results with CMIP3 and satellite data, *J. Climate*, 26, 3833-3845, doi: 10.1175/JCLI-D-12-00451.1.
- Lewondowski, M., I. R. Piletic, T. E. Kleindienst, J. H. Offenberg, M. R. Beaver, M. Jaoui, K. S. Docherty, and E. O. Edney (2013), Secondary organic aerosol characterization at field sites across the United States during the spring-summer period, *Int. J. Environ. An. Ch.*, 93(10), 1084-1103, doi:10.1080/03067319.2013.803545.
- Levy, H., II, M. D. Schwarzkopf, L. Horowitz, V. Ramaswamy, and K. L. Findell (2008), Strong sensitivity of late 21st century climate to projected changes in short-lived air pollutants, *J. Geophys. Res.*, 113, D06102, doi:10.1029/2007JD009176.
- Liu, X., R. C. Easter, S. J. Ghan, R. Zaveri, P. Rasch, X. Shi, J.-F. Lamarque, A. Gettelman, H. Morrison, F. Vitt, A. Conley, S. Park, R. Neale, C. Hannay, A. M. L. Ekman, P. Hess, N. Mahowald, W. Collins, M. J. Iacono, C. S. Bretherton, M. G. Flanner, and D. Mitchell (2012), Toward a minimal representation of aerosols in climate models: description and evaluation in the Community Atmosphere Model CAM5, *Geosci. Model Dev.*, 5, 709-739, doi:10.5194/gmd-5-709-2012.
- Martensson, E. M., E. D. Nilsson, G. deLeeuw, L. H. Cohen, and H. C. Hansson (2003), Laboratory simulations and parameterizations of the primary marine aerosol production, *J. Geophys. Res.*, 108(D9), 4297, doi:10.1029/2002JD0022263.

- Masui, T., K. Matsumoto, Y. Hijioka, T. Kinoshita, T. Nozawa, S. Ishiwatari, E. Kato, P.R. Shukla, Y. Yamagata, and M. Kainunma (2011), An emission pathway for stabilization at 6 W m^{-2} radiative forcing, *Clim. Change*, 109, 59-78, doi: 10.1007/s10584-011-0150-5.
- Moore, S. K., V. L. Trainer, N. J. Mantua, M. S. Parker, E. A. Laws, L. C. Backer, and L. E. Fleming (2008), Impacts of climate variability and future climate change on harmful algal blooms and human health, *Environmental Health*, 7, S4, doi:10.1186/1476-069X-7-S2-S4.
- Moss, R., M. Babiker, S. Brinkman, E. Calvo, T. Carter, J. Edmonds, I. Elgizouli, S. Emori, L. Erda, K. Hibbard, R. Jones, M. Kainuma, J. Kelleher, J.-F. Lamarque, M. Manning, B. Matthews, J. Meehl, L. Meyer, J. Mitchell, N. Nakicenovic, B. O'Neill, R. Pichs, K. Riahi, S. Rose, P. Runci, R. Stouffer, D. van Vuuren, J. Weyant, T. Wilbanks, J.-P. van Ypersele, and M. Zurek (2008). *Towards New Scenarios for Analysis of Emissions, Climate Change, Impacts, and Response Strategies*. Technical Summary. Intergovernmental Panel on Climate Change, Geneva, 25 pp.
- Moss, R., J. A. Edmonds, K. A. Hibbard, M. R. Manning, S. K. Rose, D. P. van Vuuren, T. R. Carter, S. Emori, M. Kainuma, T. Kram, G. A. Meehl, J. F. B. Mitchell, N. Nakicenovic, K. Riahi, S. J. Smith, R. J. Stouffer, A. M. Thomson, J. P. Weyant, and T. J. Wilbanks (2010), The next generation of scenarios for climate change research and assessment, *Nature*, 463, 747-756, doi:10.1038/nature08823. .
- Nakajima, T. Y., K. Suzuki, and G. L. Stephens (2010), Droplet Growth in Warm Water Clouds Observed by the A-Train. Part I: Sensitivity Analysis of the MODIS-Derived Cloud Droplet Sizes, *Journal of the Atmospheric Sciences*, 67(6), 1884-1896, doi:10.1175/2009jas3280.1.

- NOAA, National Oceanic and Atmospheric Administration, (1998), Automated Surface Observing System User's Guide. <http://www.nws.noaa.gov/asos/pdfs/aum-toc.pdf>.
- Oueslati, B. and G. Bellon (2015), The double ITCZ bias in CMIP5 models: interaction between SST, large-scale circulation and precipitation, *Clim. Dyn.*, *44*, 585-607, doi: 10.1007/s00382-015-2468-6.
- Patz, J. A., D. Campbell-Lendrum, T. Holloway, and J. A. Foley (2005), Impact of regional climate change on human health, *Nature*, *438*, 310-317, doi:10.1038/nature04188.
- Pereira, H. M., P. W. Leadley, V. Proenca, R. Alkemade, J. P. W. Scharlemann, J. F. Fernandez-Manjarres, M. B. Araujo, P. Balvanera, R. Biggs, W. W. L. Cheung, L. Chini, H. D. Cooper, E. L. Gilman, S. Guenette, G. C. Hurtt, H. P. Huntington, G. M. Mace, T. Oberdorff, C. Revenga, P. Rodrigues, R. J. Scholes, U. R. Sumaila, and M. Walpole, (2010), Scenarios for Global Biodiversity in the 21st Century, *Science*, *330*, 1496-1501, doi:10.1126/science.1196624.
- Qian, Y., C. N. Long, H. Wang, J. M. Comstock, S. A. McFarlane, and S. Xie (2012), Evaluation of cloud fraction and its radiative effect simulated by IPCC AR4 global models against ARM surface observations, *Atmos. Chem. Phys.*, *12*, 1785–1810, doi:10.5194/acp-12-1785-2012.
- Rausch, J., A. Heidinger, and R. Bennartz (2010), Regional assessment of microphysical properties of marine boundary layer cloud using the PATMOS-x dataset, *J. Geophys. Res.*, *115*, D23212, doi:10.1029/2010JD014468.
- Riahi, K., S. Rao, V. Krey, C. Cho, V. Chirkov, G. Fischer, G. Kindermann, N. Nakicenovic, and P. Rafaj (2011), RCP 8.5-A scenario of comparatively high greenhouse gas emissions, *Clim. Change*, *109*, 33-57, doi: 10.1007/s10584-011-0149-y.

- Rowe, P. M., S. Neshyba, and V. P. Walden (2013), Radiative consequences of low-temperature infrared refractive indices for supercooled water clouds, *Atmos. Chem. Phys.*, *13*, 11925-11933, doi:10.5194/acp-13-11925-2013.
- Seethala, C., and Á. Horvath (2010), Global assessment of AMSR-E and MODIS cloud liquid water path retrievals in warm oceanic clouds, *J. Geophys. Res.*, *115*, D13202, doi:10.1029/2009JD012662.
- Shindell, D., J. C. I. Kuylenstierna, E. Vignati, R. van Dingenen, M. Amann, Z. Klimont, S. C. Anenberg, N. Muller, G. Janssens-Maenhout, F. Raes, J. Schwartz, G. Faluvegi, L. Pozzoli, K. Kupiainen, L. Hoglung-Isaksson, L. Emberson, D. Streets, V. Ramanathan, K. Hicks, N. T. K. Oanh, G. Milly, M. Williams, V. Demkine, and D. Fowler (2012), Simultaneously Mitigating Near-Term Climate Change and Improving Human Health and Food Security, *Science*, *335*, 183-189, doi:10.1126/science.1210026.
- Shindell, D. T., J.-F. Lamarque, M. Schulz, M. Flanner, C. Jiao, M. Chin, P. J. Young, Y. H. Lee, L. Rotstayn, N. Mahowald, G. Milly, G. Faluvegi, Y. Balkanski, W. J. Collins, A. J. Conley, S. Dalsoren, R. Easter, S. Ghan, L. Horowitz, X. Liu, G. Myhre, T. Nagashima, V. Naik, S. T. Rumbold, R. Skeie, K. Sudo, S. Szopa, T. Takemura, A. Voulgarakis, J.-H. Yoon, and F. Lo (2013), Radiative forcing in the ACCMIP historical and future climate simulations, *Atmos. Chem. Phys.*, *13*, 2939–2974, doi:10.5194/acp-13-2939-2013.
- Shrivastava, M. K., T. E. Lane, N. M. Donahue, S. N. Pandis, and A. L. Robinson (2008), Effects of gas particle partitioning and aging of primary emissions on urban and regional organic aerosol concentrations, *J. Geophys. Res.*, *113*, D18301, doi:10.1029/2007JD009735.

- Shrivastava, M., A. Zelenyuk, D. Imre, R. Easter, J. Beranek, R. A. Zaveri, and J. Fast (2013), Implications of low volatility SOA and gas-phase fragmentation reactions on SOA loadings and their spatial and temporal evolution in the atmosphere, *J. Geophys. Res. Atmos.*, *118*, 3328–3342, doi:10.1002/jgrd.50160.
- Skerlack, B., Sprenger, and H. Weruli (2014), A global climatology of stratosphere–troposphere exchange using the ERA-Interim data set from 1979 to 2011, *Atmos. Chem. Phys.*, *14*, 913–937, doi:10.5194/acp-14-913-2014.
- Stevenson, D. S., P. J. Young, V. Naik, J.-F. Lamarque, D. T. Shindell, A. Voulgarakis, R. B. Skeie, S. B. Dalsoren, G. Myhre, T. K. Berntsen, G. A. Folberth, S. T. Rumbold, W. J. Collins, I. A. MacKenzie, R. M. Doherty, G. Zeng, T. P. C. van Noije, A. Strunk, D. Bergmann, P. Cameron-Smith, D. A. Plummer, S. A. Strode, L. Horowitz, Y. H. Lee, S. Szopa, K. Sudo, T. Nagashima, B. Josse, I. Cionni, M. Righi, V. Eyring, A. Conley, K. W. Bowman, O. Wild, and A. Archibald (2013), Tropospheric ozone changes, radiative forcing and attribution to emissions in the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), *Atmos. Chem. Phys.*, *13*, 3063–3085, doi:10.5194/acp-13-3063-2013.
- Thomson, A., K. V. Calvin, S. J. Smith, G. P. Kyle, A. Volke, P. Patel, S. Delgado-Arias, B. Bond-Lamberty, M. A. Wise, L. E. Clarke, and J. A. Edmonds (2011), RCP4.5: a pathway for stabilization of radiative forcing by 2100, *Clim. Change*, *109*, 77–94, doi: 10.1007/s10584-011-0151-4.
- Tian, B., E. J. Fetzer, B. H. Kahn, J. Teixeira, E. Manning, and T. Hearty (2013), Evaluating CMIP5 Models using AIRS Tropospheric Air Temperature and Specific Humidity Climatology, *J. Geophys. Res. Atmos.*, *118*, 114–134, doi:10.1029/2012JD018607.

- Val Martin, M., C. L. Heald, and S. R. Arnold (2014), Coupling dry deposition to vegetation phenology in the Community Earth System Model: Implications for the simulation of surface O₃, *Geophys. Res. Lett.*, *41*, 2988–2996, doi:10.1002/2014GL059651.
- van Vuuren, D. P., J. Edmonds, M. Kainuma, K. Riahi, A. Thomson, K. Hibbard, G. C. Hurtt, T. Kram, V. Krey, J.-F. Lamarque, T. Masui, M. Meinshausen, N. Nakicenovic, S. J. Smith, and S. K. Rose (2011a), The representative concentration pathways: an overview, *Clim. Change*, *109*, 5-31, doi: 10.1007/s10584-011-0148-z.
- van Vuuren, D. P., E. Stehfest, M. G. J. den Elzen, T. Kram, J. van Vliet, S. Deetman, M. Isaac, K. K. Glodewijk, A. Hof, A. M. Beltran, R. Oostenrijk, and B. van Ruijven (2011b), RCP2.6: exploring the possibility to keep global mean temperature increase below 2°C, *Clim. Change*, *109*, 95-116, doi: 10.1007/s10584-011-0152-3.
- Vavrus, S. and D. Waliser (2008), An improved parameterization for simulating arctic cloud amount in the CCSM3 climate model, *J. Climate*, *21*, 5673–5687, doi:10.1175/2008JCLI2299.1.
- Verlinde, J., J. Y. Harrington, G. M. McFarquhar, V. T. Yannuzzi, A. Avramov, S. Greenberg, N. Johnson, G. Zhang, M. R. Poellot, J. H. Mather, D. D. Turner, E. W. Eloranta, B. D. Zak, A. J. Prenni, J. S. Daniel, G. L. Kok, D. C. Tobin, R. Holz, K. Sassen, D. Spangenberg, P. Minnis, T. P. Tooman, M. D. Ivey, S. J. Richardson, C. P. Bahrmann, M. Shupe, P. J. DeMott, A. J. Heymsfield, and R. Schofield (2007), the mixed-Phase Arctic Cloud Experiment, *B. Am. Meteorol. Soc.*, *88*, 205–221, doi:10.1175/BAMS-88-2-205.

- Wang, M., S. Ghan, M. Ovchinnikov, X. Liu, R. Easter, E. Kassianov, Y. Qian, and H. Morrison (2011), Aerosol indirect effects in a multi-scale aerosol-climate model PNNL-MMF, *Atmos. Chem. Phys.*, *11*, 5431–5455, doi: 10.5194/acp-11-5431-2011.
- West, J. J., S. J. Smith, R. A. Silva, V. Naik, Y. Zhang, Z. Adelman, M. M. Fry, A. Anenberg, L. W. Horowitz, and J.-F. Lamarque (2013), Co-benefits of mitigating greenhouse gas emissions for future air quality and human health, *Nature Climate Change*, *3*, 885–889, doi:10.1038/nclimate2009.
- Yarwood, G., G. Z. Whitten, J. Jung, G. Heo, and D. T. Allen (2010), Final Report—Development, Evaluation, and Testing of Version 6 of the Carbon Bond Mechanism (CB6) , Rep. 582-7-84005-FY10-26, 119 pp., Center for Energy and Environmental Resources, The University of Texas at Austin, Austin, Texas. [Available at <https://www.tceq.texas.gov/assets/public/implementation/air/am/contracts/reports/pm/5820784005FY1026-20100922-environ-cb6.pdf>].
- Young, P. J., A. T. Archibald, K. W. Bowman, J.-F. Lamarque, V. Naik, D. S. Stevenson, S. Tilmes, A. Voulgarakis, O. Wild, D. Bergmann, P. Cameron-Smith, I. Cionni, W. J. Collins, S. B. Dalsoren, R. M. Doherty, V. Eyring, G. Faluvegi, L. W. Horowitz, B. Josse, Y. H. Lee, I. A. MacKenzie, T. Nagashima, D. A. Plummer, M. Righi, S. T. Rumbold, R. B. Skeie, D. T. Shindell, S. A. Strode, K. Sudo, S. Szopa, and G. Zeng (2013), Pre-industrial to end 21st century projections of tropospheric ozone from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), *Atmos. Chem. Phys.*, *13*, 2063–2090, doi:10.5194/acp-13-2063-2013.
- Yttri, K. E., Q. Aas, A. Bjerke, J. N. Cape, F. Cavalli, D. Ceburnis, C. Dye, L. Emblico, M. C. Facchini, C. Forster, J. E. Hanssen, H. C. Hansson, S. G. Jennings, W. Maenhaut, J. P.

- Putaud, and K. Torseth (2007), Elemental and organic carbon in PM₁₀: a one year measurement campaign within the European Monitoring and Evaluation Programme EMEP, *Atmos. Chem. Phys.*, 7, 5711-5725, doi:10.5194/acp-7-5711-2007.
- Yu. F. (2010), Ion-mediated nucleation in the atmosphere: Key controlling parameters, implications, and look-up table, *J. Geophys. Res.*, 115, D03206, doi:10.1029/2009JD012630.
- Yu, S., R. Dennis, S. Roselle, A. Nenes, J. Walker, B. Eder, K. Schere, J. Swall, and W. Robarge (2005), An assessment of the ability of three-dimensional air quality models with current thermodynamic equilibrium models to predict aerosol NO₃⁻, *J. Geophys. Res.*, 110, D07S13, doi:10.1029/2004JD004718.
- Yu, S., B. Eder, R. Dennis, S.-H. Chu, and S. E. Schwartz (2006), New unbiased symmetric metrics for evaluation of air quality models, *Atmos. Sci. Let.*, 7, 26-34, doi:10.1002/asl.125.
- Zender, C. S., H. Bian, and D. Newman (2003), The mineral dust entrainment and deposition (DEAD) model: Description and 1990s dust climatology, *J. Geophys. Res.*, 108(D14), 4416, doi:10.1029/2002JD002775.
- Zhang, Q., J. L. Jimenez, M. R. Canagaratna, J. D. Allan, H. Coe, I. Ulbrich, M. R. Alfarra, A. Takami, A. M. Middlebrook Y. L. Sun, L. Dzepina, E. Dunlea, K. Docherty, P. F. DeCarlo, D. Salcedo, T. Onasch, J. T. Jayne, T. Miyoshi, A. Shimono, S. Hatakeyama, N. Takegawa, Y. Kondo, J. Schneider, F. Drewnick, S. Borrmann, S. Weimer, K. Demerjian, P. Williams, K. Bower, R. Bahreini, L. Cottrell, R. J. Griffin, J. Rautiainen, J. Y. Sun, Y. M. Zhang, and D. R. Worsnop (2007), Ubiquity and dominance of oxygenated

- species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes, *Geophys. Res. Lett.*, *34*, L13801, doi:10.1029/2007GL029979.
- Zhang, Y., P. Liu, B. Pun, and C. Seigneur (2006), A Comprehensive Performance Evaluation of MM5-CMAQ for the Summer 1999 Southern Oxidants Study Episode, Part-I. Evaluation Protocols, Databases and Meteorological Predictions, *Atmospheric Environment*, *40*, 4825-4838.
- Zhang, Y., P. Karamchandani, T. Glotfelty, D. G. Streets, G. Grell, A. Nenes, F. Yu, and R. Bennartz (2012), Development and initial application of the global-through-urban weather research and forecasting model with chemistry (GU-WRF/Chem), *J. Geophys. Res.*, *117*, D20206, doi:10.1029/2012JD017.
- Zhang, Y., K. Sartelet, S.-Y. Wu, and C. Seigneur (2013), Application of WRF/Chem-MADRID and WRF/Polyphemus in Europe, Part I: Model Description and Evaluation of Meteorological Predictions, *Atmos. Chem. Phys.*, *13*, 6807–6843, doi:10.5194/acp-13-6807-2013.
- Zhang, Y., X. Zhang, K. Wang, J. He, L. R. Leung, J.-W. Fan, and A. Nenes (2015), Incorporating An Advanced Aerosol Activation Parameterization into WRF-CAM5: Model Evaluation and Parameterization Intercomparison, *Journal of Geophysical Research*, *120* (14), DOI: 10.1002/2014JD023051.
- Zhang, Y., X. Zhang, L.-T. Wang, Q. Zhang, F.-K. Duan, and K.-B. He (2016a), Application of WRF/Chem over East Asia: Part I. Model Evaluation and Intercomparison with MM5/CMAQ, *Atmospheric Environment*, *124*, Part B, 285-300, doi:10.1016/j.atmosenv.2015.07.022.

- Zhang, Y., X. Zhang, K. Wang, Q. Zhang, F.-K. Duan, and K.-B. He (2016b), Application of WRF/Chem over East Asia: Part II. Model Improvement and Sensitivity Simulations, *Atmospheric Environment*, 124, Part B, 301-320, doi:10.1016/j.atmosenv.2015.07.023.
- Zhang, Z., and S. Platnick (2011), An assessment of differences between cloud effective particle radius retrievals for marine water clouds from three MODIS spectral bands, *J. Geophys. Res.*, 116(D20)10.1029/2011jd016216.
- Ziemke, J. R., S. Chandra, B. N. Duncan, L. Froidevaux, P. K. Bhartia, P. F. Levelt, and J. W. Waters (2006), Tropospheric ozone determined from Aura OMI and MLS: Evaluation of measurements and comparison with the Global Modeling Initiative's Chemical Transport Model, *J. Geophys. Res.*, 111, D19303, doi:10.1029/2006JD007089.

Tables

Table 1. List of Datasets for Meteorological, Cloud, and Radiation Evaluation

Variable (units)	Short Name	Dataset	Spatial Coverage	Temporal Coverage
Downwelling Longwave Radiation at the Earth's Surface (W m^{-2})	FLDS	Clouds and Earth's Radiant Energy Systems (CERES)/ Energy Balanced and Filled (EBAF)	Global ($1^\circ \times 1^\circ$)	2001-2010
Outgoing Longwave Radiation at the Top of the Atmosphere (W m^{-2})	OLR			
Longwave Cloud Forcing (W m^{-2})	LWCF			
Net Surface Longwave Flux (W m^{-2})	FLNS			
Downwelling Shortwave Radiation at the Earth's Surface (W m^{-2})	FSDS			
Outgoing Shortwave Radiation at the Top of the Atmosphere (W m^{-2})	OSR			
Shortwave Cloud Forcing (W m^{-2})	SWCF			
Net Surface Shortwave Flux (W m^{-2})	FSNS			
Top of the Atmosphere Net Flux (W m^{-2})	RESTOA			
Cloud fraction	CF			
Column CCN (ocean) at $S = 0.5\%$ (cm^{-2})	CCN5			
Cloud Optical Thickness	COT			
Precipitable Water Vapor (cm)	PWV			
Aerosol Optical Depth	AOD			
Cloud Droplet Number Concentration from Warm Clouds (cm^{-3})	CDNC	<i>Bennartz [2007]</i>	Global ($1^\circ \times 1^\circ$)	2001-2010
Liquid Water Path (g m^{-2})	LWP	<i>O'dell et al. [2008]</i>	Oceans ($1^\circ \times 1^\circ$)	2001-2008
Precipitation Rate (mm day^{-1})	PR	Global Precipitation Climatology Project (GPCP)	Global ($2.5^\circ \times 2.5^\circ$)	2001-2010
2m Temperature ($^\circ\text{C}$)	T2	National Climatic Data Center (NCDC) and National Centers for Environmental Prediction (NCEP)/ National Center for Atmospheric Research (NCAR)	Global (Intermittent Coverage) and Global ($2.5^\circ \times 2.5^\circ$)	2001-2010
2m Specific Humidity (g kg^{-1})	Q2			
10m Wind Speed (m s^{-1})	WSP10			

Table 2. List of Datasets for Chemical Evaluation

Variable	Short Name	Dataset	Spatial Coverage	Temporal Coverage
Carbon Monoxide	CO	NIESJ, TAQMN, KMOE	East Asia	2001-2010,

Nitrogen Dioxide	NO ₂	NIESJ, TAQMN, KMOE, Airbase, BDQA, EMEP	East Asia, Europe	2006-2010 (KMOE) 2001-2010 2006-2010 (KMOE)
Ozone	O ₃	TAQMN, KMOE, Airbase, BDQA, EMEP, CASTNET	East Asia, Europe, CONUS	2001-2010 2006-2010 (KMOE)
Sulfur Dioxide	SO ₂	NIESJ, TAQMN, KMOE, Airbase, BDQA, EMEP, CASTNET	East Asia, Europe, CONUS	2001-2010 2006-2010 (KMOE)
Nitric Acid	HNO ₃	EMEP, CASTNET	Europe, CONUS	2001-2010
Ammonia	NH ₃	EMEP	Europe	2001-2010
Particulate Matter with diameter less than and equal to 2.5 µm	PM _{2.5}	Airbase, BDQA, EMEP, IMPROVE, STN	Europe, CONUS	2001-2010
Particulate matter with diameter less than and equal to 10 µm	PM ₁₀	MEPC, NIESJ, TAQMN, KMOE, Airbase, BDQA, EMEP, AQS	East Asia, Europe, CONUS	2001-2010 2006-2010 (KMOE)
Sulfate	SO ₄ ²⁻	EMEP, CASTNET, IMPROVE, STN	Europe, CONUS	2001-2010
Ammonium	NH ₄ ⁺			
Nitrate	NO ₃ ⁻			
Chloride	Cl ⁻			
Black Carbon	BC	IMPROVE, STN	CONUS	2001-2010
Total Carbon	TC			
Organic Carbon	OC	EMEP, IMPROVE	Europe, CONUS	2001-2010 (IMPROVE) Assumed Climatology (EMEP)
Secondary Organic Aerosol	SOA	<i>Lewondowski et al.</i> [2013]	CONUS	Assumed Climatology
Oxygenated Organic Aerosol	OOA	<i>Zhang et al.</i> [2007] and <i>Jimenez et al.</i> [2009]	Northern Hemisphere	Assumed Climatology
Hydrocarbon-like Organic Aerosol	HOA			
Total Organic Aerosol	TOA			
Tropospheric CO	TCO	MOPITT	Global (1°×1°)	2001-2010
Tropospheric Formaldehyde	THCHO	GOME, SCIAMACHY	Global (0.25°×0.25°)	2001-2002 (GOME) 2003-2010 (SCIAMACHY)
Tropospheric NO ₂	TNO ₂			
Tropospheric SO ₂	TSO ₂	SCIAMACHY	Global (0.25°×0.25°)	2005-2010
Tropospheric Ozone Residual	TOR	OMI/MLS	Global (1.5°×1.25°)	2005-2010 (OMI/MLS)

*TOMS/SBUV: the Total Ozone Mapping Spectrometer/the Solar Backscatter UltraViolet; OMI: the Aura Ozone Monitoring Instrument; MOPITT: the Measurements Of Pollution In The Troposphere; the Global Ozone Monitoring Experiment; SCIAMACHY: the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY; GOME: the Global Ozone Monitoring Experiment; CASTNET: Clean Air Status and Trends Network; IMPROVE: Interagency Monitoring of Protected Visual Environments; STN: Speciation Trends Network; AQS: The U.S. Environmental Protection Agency's Air Quality System; EMEP: European Monitoring and Evaluation Program; BDQA: Base de Données sur la Qualité de l'Air; AirBase: European air quality database; MEPC: the Ministry of Environmental Protection of China; TAQMN: the Taiwan Air Quality Monitoring Network; NIESJ: the National Institute for Environmental Studies of Japan; KMOE: the Korean Ministry of Environment.

Table 3. Performance Statistics of Radiation, Cloud, and Meteorological Variables

Variable	Mean Obs.	Mean Sim.		MB [*]		RMSE [*]		NMB [*]		NME [*]		R [*]	
		RCP4.5	RCP8.5	RCP4.5	RCP8.5	RCP4.5	RCP8.5	RCP4.5	RCP8.5	RCP4.5	RCP8.5	RCP4.5	RCP8.5
RESTOA	-29.1	-30.2	-28.1	-1.1	1.0	10.3	9.2	-3.9	3.4	26.1	22.3	0.99	0.99
FSDS	163.5	161.5	161.7	-2.0	-1.8	12.5	12.2	-1.2	-1.1	5.5	5.4	0.98	0.98
OSR	102.7	106.4	106.3	3.7	3.6	11.9	11.5	3.6	3.5	8.7	8.5	0.82	0.83
SWCF	-40.7	-42.3	-42.2	1.6	1.5	12.2	11.9	3.8	3.7	21.6	21.1	0.92	0.92
FSNS	128.5	123.8	124.0	-4.7	-4.5	12.8	12.5	-3.7	-3.5	7.6	7.5	0.99	0.99
FLDS	306.3	300.5	300.7	-5.8	-5.6	13.2	13.5	-1.9	-1.8	3.4	3.5	0.99	0.99
OLR	223.7	217.9	219.6	-6.0	-4.2	9.2	7.9	-2.7	-1.9	3.2	2.7	0.98	0.98
LWCF	22.5	20.2	20.1	-2.4	-2.4	5.8	5.8	-10.6	-10.6	20.3	20.6	0.92	0.91
FLNS	-47.6	-49.8	-49.7	2.2	2.0	8.8	8.8	4.6	4.3	13.9	13.9	0.88	0.88
CF	67.2	66.6	66.7	-0.6	-0.5	22.4	22.0	-0.9	-0.7	26.4	25.9	0.28	0.28
CCN5	2.4×10 ⁸	1.3×10 ⁸	1.3×10 ⁸	-1.1×10 ⁸	-1.2×10 ⁸	4.5×10 ⁸	4.6×10 ⁸	-45.7	-47.3	49.4	50.3	0.11	0.11
COT	17.0	10.1	10.0	-6.9	-7.0	12.8	12.8	-40.7	-41.0	54.9	54.9	-0.12	-0.12
PWV	1.9	2.0	2.0	0.05	0.06	0.3	0.3	2.8	3.4	11.8	11.8	0.98	0.98
AOD	0.16	0.13	0.13	-0.02	-0.03	0.1	0.1	-14.5	-16.2	40.7	41.4	0.62	0.61
CDNC	108.5	151.7	157.6	43.2	49.0	83.7	88.0	39.8	45.2	58.6	62.6	0.43	0.47
LWP	85.7	60.1	59.7	-25.6	-26.0	38.9	40.3	-29.9	-30.3	36.4	36.7	0.44	0.42
PR	2.3	2.4	2.5	0.2	0.2	1.0	1.0	8.3	8.9	26.3	26.8	0.87	0.86
T2 ^a	13.1	11.8	11.8	-1.3	-1.3	3.9	4.0	-9.9	-10.1	21.1	20.9	0.94	0.93
T2 ^b	5.4	4.3	4.3	-1.0	-1.0	3.0	3.1	-19.6	-19.5	39.1	39.0	0.99	0.99
Q2 ^a	8.3	8.0	8.0	-0.3	-0.3	1.7	1.7	-3.6	-3.3	14.0	14.1	0.93	0.93
Q2 ^b	8.2	7.6	7.7	-0.5	-0.5	1.0	1.0	-6.7	-6.2	9.4	9.5	0.99	0.99
WSP10 ^a	3.7	3.3	3.3	-0.4	-0.4	1.8	1.8	-9.7	-10.2	32.8	32.6	0.47	0.48
WSP10 ^b	4.2	6.6	6.6	2.4	2.5	3.2	3.3	58.3	59.0	64.4	65.3	0.59	0.58

^a Evaluated against NCDC dataset, ^b Evaluated against NCEP/NCAR dataset, * MB: mean bias; RMSE: root mean square error; NMB: normalized mean bias; NME: normalized mean error; R: correlation coefficient.

Table 4 Performance Statistics of Trace Gas and Aerosol Variables

Variable	Location	Mean Obs.	Mean Sim.		MB [*]		NMB [*]		NME [*]		R [*]	
			RCP4.5	RCP8.5	RCP4.5	RCP8.5	RCP4.5	RCP8.5	RCP4.5	RCP8.5	RCP4.5	RCP8.5
CO	East Asia ^a	503.9	149.5	142.8	-354.4	-361.2	-70.3	-71.7	70.3	71.7	0.23	0.27

NO ₂	East Asia ^a	12.8	2.6	2.8	-10.2	-9.8	-80.0	-78.1	80.2	78.6	0.56	0.57
	Europe ^b	18.6	5.4	5.3	-13.2	-13.3	-71.1	-71.5	73.2	73.2	0.26	0.25
O ₃	East Asia ^a	28.7	50.3	50.6	21.6	21.8	75.1	76.0	75.1	76.0	0.27	0.28
	Europe ^b	54.9	95.6	95.3	40.7	40.4	74.0	73.5	74.3	73.7	0.37	0.37
	CONUS ^a	35.1	49.2	49.0	14.1	13.9	40.1	39.8	40.7	40.3	0.48	0.49
SO ₂	East Asia ^a	3.2	1.6	1.4	-1.6	-1.8	-48.7	-57.4	56.2	64.0	0.42	0.39
	Europe ^b	5.5	3.9	3.1	-1.6	-2.4	-29.2	-44.4	54.6	58.6	0.51	0.54
	CONUS ^b	2.7	5.8	5.1	3.1	2.3	112.0	85.4	114.6	89.7	0.82	0.85
HNO ₃	Europe ^b	0.8	0.9	0.8	9.5×10 ⁻²	3.0×10 ⁻³	12.3	0.4	71.2	63.6	0.31	0.30
	CONUS ^b	1.2	2.3	2.2	1.1	1.0	93.1	82.7	98.6	88.7	0.66	0.66
NH ₃	Europe ^b	1.0	1.7	2.1	0.7	1.1	71.2	110.2	130.9	156.5	0.25	0.28
PM _{2.5}	Europe ^b	13.6	12.7	9.5	-1.4	-4.5	-9.9	-32.1	31.9	36.1	0.03	0.06
	CONUS ^b	8.3	7.8	7.4	-0.5	-0.9	-6.4	-11.3	33.7	33.6	0.67	0.67
PM ₁₀	East Asia ^b	112.8	50.6	50.5	-63.2	-63.4	-55.5	-55.7	56.5	57.2	0.67	0.66
	Europe ^b	23.2	26.8	22.0	3.6	-1.2	15.3	-5.4	35.5	29.5	0.22	0.21
	CONUS ^b	21.9	12.0	12.6	-9.9	-9.4	-43.5	-42.7	51.7	55.6	0.17	0.05
SO ₄ ²⁻	Europe ^b	2.0	2.4	2.0	0.4	4.2×10 ⁻³	17.8	0.2	54.6	38.0	0.57	0.56
	CONUS ^b	2.3	2.5	2.2	0.2	-0.1	7.8	-4.0	28.4	25.4	0.85	0.87
NH ₄ ⁺	Europe ^b	0.9	1.3	1.2	0.4	0.3	42.4	33.0	54.5	49.7	0.69	0.67
	CONUS ^b	1.1	1.3	1.2	0.1	0.1	12.9	5.1	40.1	38.7	0.70	0.68
NO ₃ ⁻	Europe ^b	1.8	1.8	1.8	-2.1×10 ⁻²	-1.1×10 ⁻³	-1.1	-0.1	50.8	53.3	0.52	0.45
	CONUS ^b	1.0	1.2	1.2	0.1	0.2	11.9	15.5	57.8	59.9	0.58	0.58
Cl ⁻	Europe ^b	0.9	5.2	5.6	4.2	4.7	457.2	503.1	457.2	503.1	0.75	0.73
	CONUS ^b	0.1	0.1	0.1	-2.7×10 ⁻²	-2.3×10 ⁻²	-25.7	-22.0	94.0	94.0	0.17	0.20
BC	CONUS ^b	0.4	0.2	0.2	-0.1	-0.2	-37.3	-45.5	50.1	51.9	0.48	0.51
TC	CONUS ^b	2.6	1.2	1.1	-1.4	-1.6	-53.6	-59.2	55.7	60.1	0.45	0.47
OC	Europe ^b	2.9	0.5	0.5	-2.4	-2.4	-82.0	-82.2	83.0	83.2	0.46	0.51
	CONUS ^b	1.1	0.8	0.7	-0.3	-0.4	-24.9	-33.4	33.7	37.2	0.56	0.58
SOA	CONUS ^b	2.9	2.7	2.5	-0.2	-0.5	-8.1	-15.6	33.2	33.5	0.51	0.62
OOA	NH ^b	4.8	1.7	1.7	-3.1	-3.2	-64.6	-65.5	65.2	66.2	0.89	0.91
HOA	NH ^b	2.1	0.1	0.1	-1.9	-1.9	-92.9	-93.2	92.9	93.2	0.93	0.94
TOA	NH ^b	7.9	1.8	1.8	-6.0	-6.1	-76.6	-77.2	77.0	77.6	0.86	0.85
TCO	Global ^c	1.4×10 ¹⁸	1.3×10 ¹⁸	1.3×10 ¹⁸	-1.4×10 ¹⁷	-1.6×10 ¹⁷	-9.9	-11.4	15.2	16.2	0.85	0.83
THCHO	Global ^c	4.2×10 ¹⁵	2.6×10 ¹⁵	2.7×10 ¹⁵	-1.6×10 ¹⁵	-1.5×10 ¹⁵	-37.4	-37.8	42.0	41.7	0.69	0.69
TNO ₂	Global ^c	5.3×10 ¹⁴	7.7×10 ¹⁴	7.9×10 ¹⁴	2.4×10 ¹⁴	2.6×10 ¹⁴	44.9	47.8	62.0	62.8	0.82	0.86
TSO ₂	Global ^d	0.2	0.1	0.1	-0.1	-0.1	-33.7	-42.9	98.7	96.4	-0.08	-0.08
TOR	Global ^d	28.4	31.8	31.6	3.4	3.2	11.9	11.2	17.2	17.2	0.80	0.79

^a Units of ppb, ^b Units of µg m⁻³, ^c Units of molecules cm⁻², ^d Units of DU, * MB: mean bias; NMB: normalized mean bias; NME: normalized mean error; R: correlation coefficient.

Figure Captions

Figure 1. A comparison of the zonal mean profiles for cloud and radiation variables from the RCP4C, RCP8C, and *He et al.* [2015a] simulations using CESM-NCSU and available CESM1-CMIP5 simulations against satellite estimates.

Figure 2. A comparison of the zonal mean profiles for surface meteorological variables and trace gas column mass abundances from the RCP4C, RCP8C, and *He et al.*, [2015a] simulations using CESM-NCSU and available CESM-CMIP5 simulations against satellites/reanalysis estimates.

Figure 3. The monthly-mean times series of tropospheric $\text{PM}_{2.5}$, PM_{10} , and column mass abundances of HCHO , NO_2 , and O_3 from the RCP4C and RCP8C simulations compared against available satellite estimates in the five stratocumulus cloud regions of North America (NAM), South America (SAM), South East Asia (SEA), South Africa (SAF), and Australia (AUS).

Figure 4. The monthly-mean times series (2001-2010) of AOD, column CCN_5 , column CDNC, and SWCF from the RCP4C and RCP8C simulations compared against available satellite estimates in the five stratocumulus cloud regions of NAM, SAM, SEA, SAF, and AUS.

Figure 1. A comparison of the zonal mean profiles for cloud and radiation variables from the RCP4C, RCP8C, and *He et al.* [2015a] simulations using CESM-NCSU and available CESM1-CMIP5 simulations against satellite estimates.

Figure 2. A comparison of the zonal mean profiles for surface meteorological variables and trace gas column mass abundances from the RCP4C, RCP8C, and *He et al.*, [2015a] simulations using CESM-NCSU and available CESM-CMIP5 simulations against satellites/reanalysis estimates.

Figure 3. The monthly-mean times series of tropospheric PM_{2.5}, PM₁₀, and column mass abundances of HCHO, NO₂, and O₃ from the RCP4C and RCP8C simulations compared against available satellite estimates in the five stratocumulus cloud regions of North America (NAM), South America (SAM), South East Asia (SEA), South Africa (SAF), and Australia (AUS).

Figure 4. The monthly-mean times series (2001-2010) of AOD, column CCN5, column CDNC, and SWCF from the RCP4C and RCP8C simulations compared against available satellite estimates in the five stratocumulus cloud regions of NAM, SAM, SEA, SAF, and AUS.