1 Impact of Future Climate Policy Scenarios on Air Quality and Aerosol-Cloud Interactions using
2 an Advanced Version of CESM/CAM5: Part II. Future Trend Analysis and Impacts of Projected
3 Anthropogenic Emissions

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7 **Key Points**

8 Dust, sea-salt, and BSOA dominate changes in future PM from climate change.

9 Surface concentrations of O₃ and PM₁₀ increase under RCP8.5 and decrease under RCP4.5 by
10 2050.

11 Warming caused by RCP4.5 emission reductions by 2050 weakens the Walker circulation

12 **Abstract**

13 Following a comprehensive evaluation of the Community Earth System Model modified
14 at the North Carolina State University (CESM-NCSU), Part II describes the projected changes in
15 the future state of the atmosphere under the representative concentration partway scenarios
16 (RCP4.5 and 8.5) by 2100 for the 2050 time frame and examine the impact of climate change on
17 future air quality under both scenarios, and the impact of projected emission changes under the
18 RCP4.5 scenario on future climate through aerosol direct and indirect effects. Both the RCP4.5
19 and RCP8.5 simulations predict similar changes in air quality by the 2050 period due to
20 declining emissions under both scenarios. The largest differences occur in O₃, which decreases
21 by global mean of 1.4 ppb under RCP4.5 but increases by global mean of 2.3 ppb under RCP8.5
22 due to differences in methane levels, and PM₁₀, which decreases by global mean of 1.2 µg m⁻³
23 under RCP4.5 and increases by global mean of 0.2 µg m⁻³ under RCP8.5 due to differences in

dust and sea-salt emissions under both scenarios. Enhancements in cloud formation in the Arctic and Southern Ocean and increases of aerosol optical depth (AOD) in central Africa and South Asia dominate the change in surface radiation in both scenarios, leading to global average dimming of 1.1 W m^{-2} and 2.0 W m^{-2} in the RCP4.5 and RCP8.5 scenarios, respectively. Declines in AOD, cloud formation, and cloud optical thickness from reductions of emissions of primary aerosols and aerosol precursors under RCP4.5 result in near surface warming of 0.2°C from a global average increase of 0.7 W m^{-2} in surface downwelling solar radiation. This warming leads to a weakening of the Walker Circulation in the tropics, leading to significant changes in cloud and precipitation that mirror a shift in climate towards the negative phase of the El Nino Southern Oscillation.

KEYWORDS: CESM/CAM5, Representative Concentration Pathways, Global Climate Change, Future Air Quality, Climate Change, Emission Changes

1. Introduction

Much work has been done to study how future global air quality will be impacted by climate change and emissions [Mickley *et al.*, 2004; Murazaki and Hess, 2006; Liao *et al.*, 2006; Stevenson *et al.*, 2006; Ungar *et al.*, 2006; Tagris *et al.*, 2007; Vautard and Hauglaustaine, 2007; Heald *et al.*, 2008; Hedegarad *et al.*, 2008; Wu *et al.*, 2008; Zeng *et al.*, 2008; Jacob and Winner, 2009; Weaver *et al.*, 2009; Lamarque *et al.*, 2011; Fiore *et al.*, 2012; Languer *et al.*, 2012; Tai *et al.*, 2012; Doherty *et al.*, 2013; Stevenson *et al.*, 2013; Voulgarakis *et al.*, 2013; Young *et al.*, 2013; Hauglaustaine *et al.*, 2014; Kim *et al.*, 2015, Rieder *et al.*, 2015] . The changes in future air quality and radiative forcing under the RCP scenarios as well as historical changes have been examined from time slice experiments of many online-coupled atmospheric chemistry and climate models as part of the Atmospheric Chemistry and Climate Model Intercomparison

Project (ACCMIP) [Lamarque *et al.*, 2013]. A comparison of future O₃ burdens across the ACCMIP models reveals decreases under all scenarios except RCP8.5 by 2100 [Young *et al.*, 2013] due to feedbacks from large increases in methane (CH₄) under this scenario, which is consistent the trend in future surface ozone (O₃) levels by 2050 predicted in the work of Kim *et al.* [2015]. Although the models tend to predict similar global mean O₃ trends, the changes in O₃ in all ACCMIP models are not well correlated, indicating that different models are sensitive to climate and emissions changes in different ways [Young *et al.*, 2013]. Changes in O₃ radiative forcing mirror the changes in O₃ burden following these scenarios and while no true signal of climate change on O₃ radiative forcing is seen in the ACCMIP models, there are reductions in tropical O₃ levels from greater increases in humidity and greater increases in tropospheric O₃ at higher latitudes from greater lightning NO_x emissions [Stevenson *et al.*, 2013]. Changes in future oxidation state of the atmosphere via CH₄ lifetime and OH level have been examined in ACCMIP and findings show only minor changes in CH₄ lifetime and OH level under all scenarios except RCP8.5, where CH₄ lifetime increases and OH levels decline largely in response to increases in stratospheric O₃ recovery that limits O₃ photolysis and larger CH₄ levels [Voulgarakis *et al.*, 2013].

In addition to the large body of work on climate change and changing emissions, much effort has been put into understanding and simulating the impacts of aerosols on climate [Ghan *et al.*, 2012; Lim *et al.*, 2014; Yu *et al.*, 2014a, b; Archer-Nicholls *et al.*, 2016]. Despite this effort aerosol impacts on climate continue to be one of the most uncertain aspects when assessing climate change [IPCC, 2013]. Simulated anthropogenic radiative effects of aerosols can be broken down into three components including: absorption and scattering of incoming solar radiation (i.e., the aerosol direct effect), changes in the solar and infrared radiation due to

cloud droplet formation and ice crystal nucleation in clouds (i.e., aerosol indirect effects), and changes in cloud formation from radiation absorption of aerosols (i.e., aerosol semi-direct effects) [Ghan *et al.*, 2012]. Under the RCP scenarios reductions in aerosol levels lead to reductions in aerosol radiative forcing from the aerosol direct and indirect effects, significantly reducing negative anthropogenic radiative forcing, which is reduced under the RCP2.6 scenario to larger extent than increases in GHG radiative forcing by 2100 [Shindell *et al.*, 2013].

The impact of changes in aerosol radiative forgings on future climate under these scenarios needs to be explored as the historical impact of aerosol effects have been shown to alter ocean circulations and heat transfer [Cai *et al.*, 2006] and warming introduced under these scenarios has been shown to enhance sea ice retreat [Gagne *et al.*, 2015]. 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. Part I describes this model, as well as, the ability of the model to represent the current atmosphere following the RCP scenarios. The objectives of Part II are to quantitatively examine the changes in air quality, climate, and aerosol-cloud interactions expected under these scenarios by 2050 to assess the possible range of impacts from differences in climate policy scenarios. Additional objectives for Part II include isolating and quantifying the impact of changes in climate on future air quality and the impact that emission changes under a future climate policy scenario will have on future climate from aerosol-cloud interactions and the subsequent feedbacks from these interactions. This work differs from many previous climate-chemistry studies by using CESM/CAM5 with advanced treatments for organic aerosol formation and aerosol-cloud interactions. This work also differs from the ACCMIP studies in

that it is focused on the 2050 time frame, which was an optional experiment in the ACCIMIP studies that focus primarily on changes by 2030 and 2100.

2. Future Trend Analysis

2.1 Trends in Meteorological, Cloud, and Radiation Parameters

Figure 1 shows the future trends in 2-m temperature (T2), 2-m specific humidity (Q2), precipitable water vapor (PWV), precipitation rate (PR), 10-m wind speed (WSP10), aerosol optical depth (AOD), cloud fraction (CF), cloud water path (CWP = liquid water path (LWP) + cloud ice water path (IWP)), downwelling shortwave radiation at the Earth's surface (FSDS), and outgoing longwave radiation (OLR) between the future and current decade simulations under the RCP4.5 scenario (RCP4F and RCP4C, respectively) using CESM_NCSU compared to similar trends predicted by CESM from the CMIP5 project (CESM1-CMIP5) under RCP4.5 scenario. Figure 2 shows similar trends in these variables between the future and current decade simulations under the RCP8.5 scenario (RCP8F and RCP8C, respectively) compared to CESM1-CMIP5 under RCP8.5 scenario. T2 projections between CESM-NCSU and CESM1-CMIP5 differ in magnitude with CESM-NCSU projections, predicting less warming for both the RCP4.5 and RCP8.5 scenarios (global mean increases of 1.1°C and 1.6°C, respectively) compared to CESM1-CMIP5 (global mean increases of 1.5°C and 2.3°C, respectively). This reduced warming trend is due to simulated cooling by CESM-NCSU that occurs in marine regions of the SH and the North Atlantic. These discrepancies in warming estimates are likely linked to three factors including: discrepancies in ocean and sea ice initial conditions, differences in how future greenhouse gas forcing (GHGF) is prescribed, and slight inconsistencies in the current year periods between CESM1-CMIP5 and CESM-NCSU. As mentioned in Part I, the ocean and sea

ice model components in RCP4C and RCP8C simulations are initialized based on CESM's default settings, consistent with the current decadal simulations of *He et al.* [2015a] (HE15). This differs from the current time period initialization of all component models in CESM1-CMIP5, which are initialized based on a transient 20th century simulation. Thus sea ice coverage and sea surface temperatures could differ between the current periods of CESM-NCSU and CESM1-CMIP5, even though the RCP4F and RCP8F simulations are initialized from the CESM1-CMIP5 simulations. These differences likely account for the excess cooling simulated by CESM-NCSU shown in marine regions and Antarctica in Figures 1-2. GHGF in the CESM1-CMIP5 simulations is based on globally uniform prescribed values of carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O), O₃, and Chlorofluorocarbons (CFCs) volume mixing ratios, while the GHGF in the standard release of CESM with advanced chemical treatments (CAM-Chem) [Lamarque et al., 2012], and subsequently CESM-NCSU, have GHGF based on GHG lower (i.e., the lowest model layer) boundary conditions that vary by latitude and higher altitude prognostic simulations of GHG chemistry. These lower boundary conditions for GHGs give greater GHG volume mixing ratios in the northern hemisphere (NH) compared to the Southern Hemisphere (SH) for CO₂ and CH₄, which is more consistent with observed distributions of GHGs like CH₄ [Frankenberg et al., 2006]. This difference is primarily responsible for the larger latitudinal gradient in temperature changes simulated by CESM-NCSU. For example, maximum cooling of 4.7°C and 3.9°C occurs in the SH for the RCP4.5 and RCP8.5 scenarios, respectively, in the CESM-NCSU simulations compared to maximum cooling of -0.3°C under the RCP4.5 scenario and minimal warming of 0.1°C under the RCP8.5 scenario in the CESM-CMIP5 simulations. CESM-NCSU predicts greater maximum warming of 10.1°C in the NH compared to 6.2°C in the CESM1-CMIP5 simulation under the RCP4.5 scenario and more comparable

maximum warming of 9.2°C compared to 10.0°C in the CESM1-CMIP5 simulation under the RCP8.5 scenario. These differences could also be related to the fact that the current year period in the CESM1-CMIP5 simulations is a subset (2006-2010) of the time period in the CESM-NCSU current year simulations, since the period of 2001-2005 is considered to be part of the historical 20th century period in the CMIP5 project. However, despite these discrepancies in the sign and magnitude of the T2 projections, the spatial patterns in T2 changes predicted by CESM-NCSU and CESM1-CMIP5 are qualitatively similar with greater temperature increases over land compared to marine environments and greater warming at higher latitudes. There is also consistency in terms of cooling as the regions that cool in the CESM-NCSU simulation generally either cool or show minimal warming in the CESM1-CMIP5 simulations. The discrepancy in T2 projections could also be a factor of the chemistry as the aerosol changes under these scenarios can impact large scale circulations, which will be illustrated in Section 4.

The reduced warming in the CESM-NCSU simulation results in less evapotranspiration compared the CESM1-CMIP5 simulations, resulting in smaller increases in Q2 (e.g., global mean increases of 0.4 g kg⁻¹ under RCP4.5 and 0.5 g kg⁻¹ under RCP8.5) and PWV (e.g., global mean increases of 1.2 mm under RCP4.5 and 1.8 mm under RCP8.5) compared to CESM1-CMIP5 (e.g., global mean increases in Q2 of 0.5 g kg⁻¹ under RCP4.5 and 0.8 g kg⁻¹ under RCP8.5 and global mean increases in PWV of 1.5 mm under RCP4.5 and 2.3 mm under RCP8.5). Similar to T2, the spatial patterns of the changes in Q2 and PWV are qualitatively similar to those of CESM1-NCSU despite the differences in sign and magnitude. Overall, in addition to reduced warming and moistening of the atmosphere, the inconsistencies mentioned above also result in smaller differences between the RCP4.5 and RCP8.5 scenarios in the CESM-NCSU simulations compared to CESM1-CMIP5. PR in both scenarios and by both models

increases on global average by $\sim 0.05 \text{ mm day}^{-1}$ in the CESM-NCSU simulations and by $\sim 0.06 \text{ mm day}^{-1}$ under the RCP4.5 scenario and $\sim 0.09 \text{ mm day}^{-1}$ under the RCP8.5 scenario in the CESM1-CMIP5 simulations. Both models under both scenarios generally experience increased PR in the tropics and higher latitudes and decreased PR in the subtropics, this is a consistent pattern of climate change from both CMIP5 models and models using older IPCC AR4 scenarios [Knutti and Sedlacek, 2012]. The inconsistencies between CESM-NCSU and CESM1-CMIP5 occur largely due to reductions of PR in the SH related to the cooling and drying seen in CESM-NCSU.

Changes in WSP10 from both the CESM-NCSU and CESM1-CMIP5 simulations are highly complex under both the RCP4.5 and RCP8.5 scenarios, but some commonalities exist. Under both scenarios, WSP10 changes predicted by both models over land are minimal compared to those in marine environments. These minimal changes over land make the trend in land WSP10 difficult to distinguish, but in a general sense the magnitude and spatial extent of WSP10 reductions appear to dominate. In the marine environments, there is a general trend for increasing WSP10 in the Arctic Ocean and Southern Ocean and reduced WSP10 in the Pacific Ocean. This pattern is consistent in the simulations with both models under both scenarios and is consistent with changes in WSP10 predicted under the RCP4.5 and RCP8.5 scenarios across the different models in the CMIP5 project [Dobrynin *et al.*, 2012].

Changes in AOD simulated by both models are similar across both scenarios in the NH with large decreases in Europe, East Asia, and eastern U.S. in response to decreases in primary aerosol emissions and emissions of most anthropogenic aerosol precursors. The changes in anthropogenic (i.e., anthropogenic VOC (AVOC), nitric oxide (NO), carbon monoxide (CO), sulfur dioxide (SO₂), ammonia (NH₃), black carbon (BC), and organic carbon (OC)) and natural

(i.e., biogenic VOC (BVOC), sea-salt, and dust) emissions between the current and future periods under the RCP4.5 and RCP8.5 scenarios are shown in Figures S1 and S2, respectively, in the supplementary material. Increases are simulated by both models in South Asia under both scenarios due to enhanced emissions of primary aerosols and aerosol precursors. Larger deviations in the AOD trends between CESM-NCSU and CESM1-CMIP5 are found in the SH. In the CESM1-CMIP5 simulations, AOD remains similar to the current period or decreases slightly over most continents in response to declining biomass burning emissions of BC and OC in Africa and South America and reduced WSP10 and greater PR in central Australia. In the CESM-NCSU simulations AOD tends to increase in SH, which is in part related to decreased PR but also likely linked to strong enhancements in secondary organic aerosol (SOA) and related new particle formation in the Congo and Amazon predicted by the updated OA treatments in CESM-NCSU and increased WSP10 leading to greater dust emissions in central Australia and South Africa. Additionally, the magnitudes of changes in AOD are larger in the CESM-NCSU (e.g., maximum differences of $\sim \pm 0.6$) compared to CESM1-CMIP5 (e.g., maximum differences of $\sim \pm 0.2$), which is likely related to more advanced chemistry mentioned in Part I.

Both models simulate decreases in CF on global average across both scenarios. Under the RCP4.5 scenario, the simulated decrease in CF is less by CESM-NCSU ($\sim 0.06\%$) compared to CESM1-CMIP5 ($\sim 28\%$). This discrepancy is related to the cooling in the SH that results in greater saturation and greater warming in the Arctic, leading to greater availability of water vapor to form clouds. Under the RCP8.5 scenario, the decreases in CF between both models are similar ($\sim 33\%$ in CESM-NCSU and $\sim 37\%$ in CESM1-CMIP5). These findings are consistent with other CMIP5 model projections that showed declines in CF due to decreases in low and mid-level clouds resulting from greater vertical ascent related to increasing temperatures, with

this effect maximized over land due to the greater increase in temperatures [Zelinka *et al.*, 2013]. CWP increases in both CESM-NCSU simulations (e.g., global mean increases of 0.1 g m^{-2} under RCP4.5 and 0.6 g m^{-2} under RCP8.5), unlike CESM1-CMIP5 that experiences a general decrease under the RCP4.5 scenario (-0.5 g m^{-2}) and a small increase under the RCP8.5 scenario (0.1 g m^{-2}). The increased trend in CWP in CESM-NCSU is likely related to greater increases in CF in the Arctic and Southern Ocean and generally greater aerosol levels compared to CESM1-CMIP5. However, there are several commonalities between the two models under both scenarios including reduced CWP in East Asia, Europe, eastern U.S., southern Africa, and South America and increases of CWP in South Asia. These changes in CWP are mostly related to changes in CF in these regions but also mirror changes in AOD in NH, indicating that reductions in CWP in Europe, North America, and East Asia are caused by reduced formation of cloud droplets from reduced aerosol levels.

FSDS is reduced on global average by both models under both scenarios. Reductions are larger in CESM-NCSU (e.g., global mean reductions of 1.1 W m^{-2} under the RCP4.5 scenario and 2.1 W m^{-2} under the RCP8.5 scenario) compared to CESM1-CMIP5 (e.g., global mean reductions of 0.3 W m^{-2} under the RCP4.5 scenario and 1.0 W m^{-2} under the RCP8.5 scenario). The greater diming in CESM-NCSU is caused by the greater cloud formation in the Arctic and Southern Ocean and greater increases in AOD levels across the SH compared to CESM1-CMIP5. Despite these differences, the simulated spatial patterns of the trends in FSDS by both models match well with the simulated spatial patterns of the trends in AOD and CWP under both scenarios. Increasing FSDS over many land areas is in response to reductions in the strength of the aerosol direct and indirect effects from reduced emissions and reduced cloud formation. OLR is projected to increase in the future in response to greater warming with greater increases in

CESM1-CMIP5 (e.g., global mean reductions of 1.9 W m^{-2} under the RCP4.5 scenario and 2.3 W m^{-2} under the RCP8.5 scenario) compared to CESM-NCSU (e.g., global mean reductions of 0.6 W m^{-2} under the RCP4.5 scenario and 0.8 W m^{-2} under the RCP8.5 scenario). These differences are largely attributed to the cooling in the SH from differences in GHGF and ocean initial conditions between the simulations and also differences in trends in CF over in the tropics. Changes in OLR are generally consistent across the NH. Overall, the trends in radiation, cloud, aerosol, and meteorological parameters are comparable between CESM-NCSU and CESM1-CMIP5 and consistent with predictions of other CMIP5 models and thus can generally be considered representative of these climate policy scenarios.

2.2 Trends in Trace Gases

Figure 3 shows absolute differences in trace gas levels of total non-methane volatile organic compound (TNMVOC), NO_2 , SO_2 , NH_3 , nitric acid (HNO_3), O_3 , CO, hydroxyl radical (OH), hydroperoxy radical (HO_2), and hydrogen peroxide (H_2O_2) between the RCP4F and RCP4C simulations, as well as between the RCP8F and RCP8C simulations. Changes in TNMVOC, NO_2 , SO_2 , and NH_3 largely follow the trends in their emissions, or the emissions of NO in the case of NO_2 . Figure S1 in the supplementary material shows the absolute difference in emissions between the current and future periods under the RCP4.5 scenario for AVOC, NO, SO_2 , NH_3 , CO, BVOC, BC, OC, sea-salt, and dust. Figure S2 shows the trends in these species under the RCP8.5 scenario. TNMVOC levels increase with global average increases of 0.3 ppb and 0.7 ppb under the RCP4.5 and RCP8.5 scenarios, respectively. The global average TNMVOC level increases under the RCP4.5 scenario because enhanced BVOC emissions from warmer temperatures compensate the global average decline in AVOC emissions. Increases are larger under the RCP8.5 scenario because the magnitude of the decrease in AVOC emissions in

North America, Europe, and East Asia is less under the RCP8.5 scenario compared to the decrease under the RCP4.5 scenario. AVOC emissions in tropical regions and the SH increase under the RCP8.5 scenario but decrease under RCP4.5 scenario, and BVOC emissions are larger under the RCP8.5 scenario compared to the RCP4.5 scenario. The greater enhancement in BVOC emissions is the result of greater warming in the RCP8.5 scenario compared to the RCP4.5 scenario, which is a consistent result for changes in BVOC emissions between both scenarios from the ACCMIP ensemble of models [Young *et al.*, 2013]. In general, the spatial distribution indicates that the enhanced BVOC emissions from warmer temperatures and greater FSDS over the continents will dominate the decreases in AVOC during the future decadal period. This is especially true in North America.

NO₂ levels decline on global average under both scenarios, with greater declines in the RCP4.5 scenario (~0.1 ppb) compared to the RCP8.5 (~0.06 ppb). NO₂ levels increase in South Asia and Africa, decrease in North America, East Asia, and Europe, and remain similar in the many other regions. This is a recurring pattern for the anthropogenic emissions and subsequently the atmospheric concentrations of many species in the RCP emission inventories by 2050. However, it is interesting to note that the increase in NO₂ is larger under the RCP4.5 scenario compared to the RCP8.5 scenario in response to greater increases in emissions in South Asia following RCP4.5. Changes in SO₂ levels follow a similar pattern as NO₂, with greater decreases under the RCP4.5 scenario (0.15 ppb) compared to those under the RCP8.5 scenario (0.08 ppb). This is in response to greater emission reductions across the NH under the RCP4.5 scenario and also greater increases in SO₂ emissions in South Asia under the RCP8.5 scenario. Unlike other inorganic gas species, NH₃ levels increase on global average under both scenarios due to increases in agricultural activities [Riahi *et al.*, 2011; Thomson *et al.*, 2011], with greater

increases under the RCP8.5 scenario (~0.2 ppb) compared to those under the RCP4.5 scenario (~0.1 ppb), resulting from larger emission increases under the RCP8.5 scenario. HNO₃ levels decrease on global average in both scenarios with greater decreases under RCP4.5 (0.04 ppb) compared to RCP8.5 (0.03 ppb). The primary cause of this reduction is the reductions in NO emissions. These decreased NO emissions reduce the levels of most oxidized nitrogen gas species including NO, NO₂, nitrate radical (NO₃g), N₂O₅, and organic nitrates, thus limiting the source species of HNO₃. However, there are also decreases in OH, which further limit HNO₃ formation resulted from the oxidation of NO₂ and organic nitrates, and reductions in aerosol levels that reduce the rates of heterogeneous reactions forming HNO₃ from N₂O₅ and NO₃g.

The O₃ mixing ratio decreases on global average under the RCP4.5 scenario by 1.4 ppb but increases on global average by 2.3 ppb under the RCP8.5 scenario. These differences in O₃ mixing ratio between these scenarios are consistent with future trends in surface O₃ from the ACCMIP model ensemble [Young *et al.*, 2013] as well as the work of Kim *et al.* [2015] that predicted a global average decrease of 3.7 ppb under RCP4.5 and an increase of 2.1 ppb under RCP8.5 by 2050 using a version of GEOS-Chem driven by CESM. In both the RCP4.5 and RCP8.5 scenarios, NO emissions decline making the differences in future O₃ predictions slightly counter intuitive. In order to better understand these differences, Figure S3 shows the absolute difference in O₃, OH, and CO surface layer chemical production and net chemical production (production – loss) between the current and future periods for both the RCP4.5 and RCP8.5 scenarios. The decreases of O₃ mixing ratios under the RCP4.5 scenario in many of the models in the ACCMIP project are primarily attributed to reductions in precursor NO and AVOC emissions [Young *et al.*, 2013], which lead to a global average reduction in O₃ production. This is consistent with the global mean reduction in O₃ production (-1.7×10^{-2} ppb s⁻¹) shown under this

scenario in Figure S3. However, the net chemical production (NCP) of O₃ under the RCP4.5 scenario is positive and increases on global average by 1.2×10^{-4} ppb s⁻¹. In South Asia and Africa the increases in NCP of O₃ are related to the increase in absolute production from greater precursor emissions, while the increases shown in eastern U.S., eastern China, and Europe are linked to strong decreases in O₃ chemical loss. These decreases are caused by reduced titration from reduced NO levels, less loss from the oxidation of AVOC, and less O₃ consumption from heterogeneous oxidation of SO₂ on the surface of aerosol due to reduced SO₂ and aerosol levels. The increased NCP of O₃ indicates that additional causes beyond chemistry result in the declining surface O₃ levels. The most likely cause is the increases in O₃ deposition in the future climate or reductions in production aloft. The enhanced O₃ deposition is not apparent in O₃ deposition flux differences shown in Figure 7 because the flux is a function of the O₃ concentration, except over the Arctic region where the O₃ deposition is enhanced despite a declining O₃ level. Increased O₃ deposition during the future period results from weaker aerodynamic resistances because of reduced wind speeds over land, general increases in planetary boundary layer height (PBLH) over land, and reductions in snow cover that have been shown to increase dry deposition [Andersson and Engardt, 2010].

Under the RCP8.5 scenario, the spatial extent of the reductions in NO emissions is less compared to the RCP4.5 scenario and the increases in background emissions from shipping and aviation are larger. Additionally, TNMVOC levels, temperatures, and CH₄ levels increase substantially in the RCP8F simulation, all of which result in a global mean increase in O₃ production (3.2×10^{-3} ppb s⁻¹) under the RCP8.5 scenario. The increase in CH₄ under the RCP8.5 scenario is especially large (~56.4%) compared to the RCP4.5 scenario (~4.7%). This contributes to the increased O₃ production by both enhancing O₃ production rates through warming (since

CH₄ has a strong greenhouse effect), and by enhancing the conversion of NO to NO₂ which further enhances O₃ levels. This impact from CH₄ has been shown to contribute significantly to increases in O₃ in the RCP8.5 scenario compared to the RCP4.5 scenario on both the global and regional scales [Gao *et al.*, 2013; Young *et al.*, 2013]. Under the RCP8.5 scenario, there is a reduction in surface OH, which may lead to reduced destruction of O₃ despite increases in TNMVOC and HO₂. This in addition to enhanced production leads to greater global average increase in NCP of O₃ (1.6×10^{-4} ppb s⁻¹) under the RCP8.5 scenario. Additionally, previous model simulations have shown that this increase in O₃ under the RCP8.5 scenario may be enhanced by greater troposphere and stratosphere (STE) due to a greater strengthening of the Brewer-Dobson Circulation from greater warming [Young *et al.*, 2013].

In addition to changes in O₃ production and destruction it is also important to examine the impact of changing chemical regimes. Figure S4 in the supplementary material shows the changes in NO_x-VOC limited regimes under both the RCP4.5 and RCP8.5 scenarios using two different photochemical indicators suggested by Zhang *et al.* [2009]. These photochemical indicators include the mixing ratio of NO_y, which indicates a transition to VOC limited conditions when the NO_y level is less than or equal to 5.0 ppb, and the O₃/NO_y ratio which indicates a transition to VOC limited conditions when the value exceeds 15.0. These two indicators were found to be the most robust during a comparison of photochemical indicators from several regional online coupled air quality models as part of the Air Quality Model Evaluation International Initiative phase II [Campbell *et al.*, 2015]. Generally, both the RCP4.5 and RCP8.5 scenarios are dominated by a transition from VOC-limited conditions during the current period to NO_x-limited conditions in the future period over most of the NH industrial zones of the eastern U.S., Europe, and East Asia. While transitions from NO_x-limited conditions

in the current period to VOC-limited conditions in the future period occur in some portions of South Asia and central Africa. Both indicators show similar changes under both RCP scenarios. The largest differences between scenarios occur in Asia. In East Asia there is not as strong a transition to NO_x-limited conditions in the future period under the RCP8.5 scenario. This is a result of smaller decreases in East Asian NO emissions under the RCP8.5 scenario. In South Asia there is a stronger transition to VOC-limited conditions under the RCP4.5 scenario, as a result of greater increases in South Asian NO emissions under the RCP4.5 scenario. Based on this analysis, it would seem the other factors mentioned earlier in this section have a much stronger impact on the global disparity in O₃ projections than chemical regime change, but it may still be important at the regional or smaller scales as indicated by *Yahya et al.* [2016].

Surface OH levels decline under both scenarios with a greater global average decrease under the RCP8.5 scenario (6.3×10^{-3} ppt) compared to the RCP4.5 scenario (1.8×10^{-3} ppt). Under the RCP4.5 scenario, OH production from the O₃ destruction by HO₂ and the reaction of water vapor with singlet oxygen (O¹D) formed through O₃ photolysis is reduced in East Asia, the U.S., South America, and Europe in response to decreased O₃ levels. OH production is enhanced in South Asia and Africa from increased O₃ levels due to the same processes. The change in NCP of OH is a global average increase (2.1×10^{-9} ppb s⁻¹) due to reductions in OH loss from the oxidation of SO₂, O₃, nitrogen species, and AVOCs. However, there are decreases in NCP of OH in regions with large ambient OH levels including the mountainous regions of western North America, the Tibetan Plateau, western South America, and northern and southern Africa, likely resulting from increases in BVOC levels which consume more OH. These decreases in NCP of OH are co-located with the largest decreases in surface OH levels and contribute to the global average surface OH reduction. Changes in OH in the RCP8.5 scenario are more complex. Under

this scenario, there is both greater OH production and greater NCP of OH from greater O₃ levels and reductions in other species oxidized by OH, but surface OH levels decrease more than those under the RCP4.5 scenario. In both the RCP8C and RCP8F simulations, the NCP of OH is negative from the surface to roughly 936 hPa because the ambient trace gas and aerosol levels are relatively large due to surface emissions. In the region of the atmosphere from roughly 936 hPa to 760 hPa, the NCP of OH becomes positive because the levels of most species that destroy OH through oxidation decline and OH precursors (e.g., O and O₃) increase or remain elevated. Thus there is a pool of OH in this region of the atmosphere that can be mixed to the surface, especially in the tropical and subtropical marine environments. Under the RCP8.5 scenario there is greater diming of FSDS and reductions in PBLH in these regions from greater cloud formation that results in weaker vertical motion. This reduces mixing between the elevated region of large OH production and the lower region of OH loss. The net result is a compensation of the increase in the NCP of OH by the reduced mixing that results in decreased surface OH levels. Reduced OH levels under the RCP8.5 scenario are consistent with decreases in OH burden from the ACCMIP ensemble of models [Voulgarakis *et al.*, 2013].

Under both scenarios, the HO₂ and H₂O₂ levels increase on global average with larger increases under RCP8.5 (e.g., 0.46 ppt increase in HO₂ and 0.11 ppb increase in H₂O₂) compared to RCP4.5 (e.g., 0.16 ppt increase in HO₂ and 0.02 ppb increase in H₂O₂). The increase in HO₂ levels are likely from greater oxidation of TNMVOC, especially BVOC from enhanced BVOC emissions, and CH₄ under both scenarios. The increased levels of TNMVOC, CH₄, OH production, and O₃ under the RCP8.5 scenario relative to the RCP4.5 scenario explain the larger increase in HO₂ levels. H₂O₂ levels increase in a similar manner since HO₂ is one of the primary precursors of H₂O₂. Increases in H₂O₂ over land may also be related to reductions in aqueous

chemistry from reduced LWP and SO₂ levels. This may also explain the further continental enhancement of HO₂ since HO₂ and H₂O₂ are connected by cyclical chemical reactions.

Similar to O₃, CO levels increase on global average under the RCP8.5 scenario (18.3 ppb) and decrease under RCP4.5 (-4.6 ppb). Under both scenarios, CO emissions decrease in almost all regions except South Asia and Africa, however, the magnitude and spatial extent of the CO emission reductions over land are reduced under the RCP8.5 scenario. Additionally, shipping emissions in the marine environments are projected to increase by ~30-40% under the RCP8.5 scenario compared to ~10-20% under the RCP4.5 scenario. Chemical production and the NCP of CO increase under both scenarios. However, chemical production of CO is much higher in the RCP8.5 scenario (1.4×10^{-4} ppb s⁻¹) compared to the (3.5×10^{-5} ppb s⁻¹) because of greater formation from the oxidation of TNMVOC and CH₄. The NCP of CO between both scenarios is similar, although the magnitude is larger under the RCP8.5 scenario. The culmination of these factors explain the increase in CO under the RCP8.5 scenario where the enhanced shipping emissions and greater chemical production of CO lead to greater background levels of CO even though CO declines in industrial regions from reduced anthropogenic emissions. Under the RCP4.5 scenario chemical production of CO and increases in shipping emissions are less, allowing decreases in anthropogenic CO emissions to dominate the change in CO levels.

2.3 Trends in Aerosols

Figure 4 shows the trends in surface concentrations of individual aerosol species of sulfate (SO₄²⁻), ammonium (NH₄⁺), nitrate (NO₃⁻), dust, sodium (Na⁺), chloride (Cl⁻), BC, PM, oxidized POA (SVOA), and SOA under the RCP4.5 and RCP8.5 scenarios. Figure 5 shows the total change in PM_{2.5} and PM₁₀ under these scenarios. Changes in SO₄²⁻ levels are dominated by

changes in the emissions of SO₂ with decreases across Europe, East Asia, and North America under both scenarios and increase in South Asia and central Africa, with weaker decreases and stronger enhancements under the RCP8.5 scenario. The result is a global average decrease in SO₄²⁻ in both scenarios (e.g., -0.3 µg m⁻³ under RCP4.5 and -0.2 µg m⁻³ under RCP8.5). NH₄⁺ levels follow a similar pattern to those of SO₄²⁻ despite increases in NH₃ levels in order to reach thermodynamic equilibrium with the reduced sulfuric acid (H₂SO₄) and nitric acid (HNO₃) levels. This leads to global average decreases in the NH₄⁺ level by 0.06 µg m⁻³ under the RCP4.5 scenario and 0.03 µg m⁻³ under the RCP8.5 scenario. Changes in NO₃⁻ concentrations mirror those of SO₄²⁻ and NH₄⁺ with the exception of enhanced NO₃⁻ in the Middle East under both scenarios and southeastern China under the RCP8.5 scenario. The increases in the Middle East are related to increased HNO₃ levels, while the increase over southeastern China is likely a nonlinear effect of the reduced SO₄²⁻ allowing for more NO₃⁻ formation as described in *Zhang et al.* [2000] and *Yu et al.* [2005], since changes in meteorology and HNO₃ do not support this enhancement. The net result is a global average decrease in NO₃⁻ of 0.01 µg m⁻³ under the RCP4.5 scenario and a global average increase in NO₃⁻ of 0.02 under the RCP8.5 scenario.

The global level of dust decreases under both scenarios but with greater decreases under the RCP4.5 scenario (1.4 µg m⁻³) compared to the RCP8.5 scenario (0.3 µg m⁻³). The declining trend of dust levels in both scenarios is driven by decreases in WSP10 over land areas but especially in the dust emission source regions of North Africa, the Middle East, and East Asia. Dust concentrations and emissions in South Africa, Australia, and Patagonia increase under both scenarios in response to enhanced WSP10 in these regions. The weaker reduction in dust levels under the RCP8.5 scenario is due to greater complexity in the changes of WSP10 in the dust source regions of North Africa, the Middle East, and East Asia, leading to smaller reductions in

dust emissions and some compensating increases in dust emissions. Na^+ and Cl^- concentrations both increase similarly on global average under both the RCP4.5 (e.g., $0.21 \mu\text{g m}^{-3}$ and $0.34 \mu\text{g m}^{-3}$, respectively) and RCP8.5 scenarios (e.g., $0.24 \mu\text{g m}^{-3}$ and $0.37 \mu\text{g m}^{-3}$, respectively) in response to greater emissions from the enhancements of marine WSP10. It is also interesting to note that sea-salt levels in the Arctic increase by up to 677% in response to greater emissions from sea ice retreat.

BC and primary organic matter (POM) both experience similar trends as other anthropogenic species with increases in South Asia and Africa and decreases in other regions. The only exception is an increase in POM over northern sections of North America and Asia under the RCP4.5 scenario in response to increases in biomass burning emissions. The global average decrease in BC concentrations is greater under the RCP8.5 scenario ($1.6 \times 10^{-2} \mu\text{g m}^{-3}$) compared to that under the RCP4.5 scenario ($1.2 \times 10^{-2} \mu\text{g m}^{-3}$) in response to greater emission increases in Africa and South Asia and smaller maximum reductions in the NH under the RCP4.5 scenario. The opposing trend exists for POM with a greater global average reduction of $2.2 \times 10^{-2} \mu\text{g m}^{-3}$ under the RCP4.5 scenario compared to a global average reduction of $1.3 \times 10^{-2} \mu\text{g m}^{-3}$ under the RCP8.5 scenario. This is in response to smaller reductions in SH biomass burning emissions and little change in emissions in North America under the RCP8.5 scenario. The changes in POM are also reflected in SVOA with a greater global average reduction under the RCP4.5 scenario ($2.1 \times 10^{-2} \mu\text{g m}^{-3}$) compared to the RCP8.5 scenario ($1.3 \times 10^{-2} \mu\text{g m}^{-3}$). This is related to the differences in OC emissions but may also be related to decreases in OH production in the RCP4.5 scenario that limits the aging of SVOCs.

SOA increases under both scenarios with a greater increase under the RCP8.5 scenario ($7.8 \times 10^{-2} \mu\text{g m}^{-3}$) compared to those under the RCP4.5 scenario ($3.0 \times 10^{-2} \mu\text{g m}^{-3}$). The spatial patterns in SOA changes are quite different between both scenarios. Under the RCP4.5 scenario, SOA enhancements of up to $5.1 \mu\text{g m}^{-3}$ occur in South Asia and central Africa. SOA is increased in these regions in response to increased O_3 levels that lead to greater formation of condensable organic vapors from greater VOC oxidation and greater OH production, the latter enhances not only VOC oxidation but also photochemical aging of these organic vapors, leading to a reduction in volatility. There are also large enhancements in AVOC levels in South Asia and BVOC levels in central Africa that contribute to these enhancements. Minor increases in SOA exist over the Amazon ($0.25\text{--}1.0 \mu\text{g m}^{-3}$) in response to greater BVOC emissions but larger decreases in SOA of greater than $2.5 \mu\text{g m}^{-3}$ occur in central South America. The small increases and decreases in South America result from reductions in AVOC emissions, O_3 levels, and OH production. These factors also contribute to the reductions of up to $11.2 \mu\text{g m}^{-3}$ for SOA in Indonesia and East Asia. Under the RCP8.5 scenario, SOA levels are enhanced up to $6.4 \mu\text{g m}^{-3}$ in Africa, South America, South Asia, and East Asia. The nearly global enhancement in SOA under this scenario is related to the global enhancements of O_3 , OH production, BVOC emissions, and enhanced AVOC emissions in the SH. Overall, the changes in both scenarios indicate that changes in oxidant levels have a stronger impact on SOA changes than changes in TNMVOC levels. This is especially true in East Asia where SOA levels increase under the RCP8.5 scenario despite decreases in TNMVOC levels.

The net impact from changes in all these species is global average decreases in $\text{PM}_{2.5}$ under both scenarios with a greater decrease under the RCP4.5 scenario ($-0.7 \mu\text{g m}^{-3}$) compared to the RCP8.5 scenario ($-0.2 \mu\text{g m}^{-3}$). The smaller decrease under the RCP8.5 scenario results

from smaller decreases in the concentrations of most of the aforementioned species and greater increases in SOA. PM₁₀ levels decrease on global average under the RCP4.5 (-1.2 µg m⁻³) scenario but increase under the RCP8.5 scenario (0.2 µg m⁻³). This is related to the changes in PM_{2.5} but the greater enhancements in PM₁₀ under the RCP8.5 scenario result from greater increases in sea-salt emissions and weaker reductions in dust emissions.

2.4 Trends in Chemical Deposition

Figure 6 shows changes in total deposition of O₃, nitrogen, oxidized sulfur (SO_x) and BC, as well as the percentage of oxidized nitrogen, organic nitrogen, and reduced nitrogen that comprise total nitrogen deposition under the RCP4.5 and RCP8.5 scenarios. The changes in O₃ deposition under both scenarios largely mirror the changes in surface O₃ levels except in the Arctic regions under the RCP4.5 scenario, where O₃ deposition increases despite decreases in surface O₃ levels. The O₃ deposition decreases on global average under the RCP4.5 scenario by 1.1×10^{-12} kg m⁻² s⁻¹ and increases on global average by 3.8×10^{-12} kg m⁻² s⁻¹ under the RCP8.5 scenario. As mentioned in Section 2.1, the enhancements in Arctic O₃ deposition are likely linked to reduced snow cover since greater WSP10 and PBLH in this region would favor reduced deposition. Under both scenarios, O₃ deposition is enhanced in South Asia and central Africa, which indicates regardless of scenario there may be enhanced crop damage in these regions from greater O₃ deposition. The enhanced O₃ deposition over much of the globe under the RCP8.5 scenario may also result in greater ecological stress and thus underscores the importance of carbon control policies as well as emission control policies for climate mitigation and air pollution improvement. This is because the large increases in CH₄ under the RCP8.5 scenario result in greater climate change from greater warming, degradation of air quality from increases in O₃, and greater ecological stress from increases in O₃ deposition.

Nitrogen deposition (ND) from CESM-NCSU is defined as the sum of nitrogen deposited from the dry and wet deposition of NO, NO₂, peroxyacetic acid (HO₂NO₂), HNO₃, NH₃, NO₃⁻, NH₄⁺, Peroxyacetylnitrate, and organic nitrates. ND decreases on global average (-2.1×10^{-13} kg m⁻² s⁻¹) under the RCP4.5 scenario but increases (3.2×10^{-13} kg m⁻² s⁻¹) under the RCP8.5 scenario. ND largely decreases under the RCP4.5 scenario in all regions except central Africa and South Asia in response to decreases in NO emissions that limit the formation of many oxidized nitrogen species. ND deposition is enhanced in almost all regions under the RCP8.5 scenario except the industrial regions of East Asia, northern Europe, and the western and eastern coasts of North America. This large increase in ND results from both smaller reductions in NO emissions that lead to smaller reductions in oxidized nitrogen species and greater increases in NH₃ emissions and concentrations that lead to greater deposition of reduced nitrogen species. This further indicates that the greater emission controls of the RCP4.5 scenario would be beneficial for ecosystem health as the enhancements in ND under the RCP8.5 scenario could lead to greater ecosystem stress, since 10.1% of current period ecosystems are exposed to ND greater the critical load estimated to cause ecological stress [Dentener *et al.*, 2006].

The change in the source of ND is also explored by examining the changes in the percentage of ND from inorganic oxidized nitrogen (INON) species (NO, NO₂, HO₂NO₂, HNO₃, and NO₃⁻), organic nitrogen (ON) species (Peroxyacetylnitrate and organic nitrates), and reduced nitrogen (RN) species (NH₃ + NH₄⁺). Under both the RCP4.5 and RCP8.5 the percentage of INON deposition decreases due to reductions in projected NO emissions, with a greater global average reduction of 9.6% under the RCP8.5 scenario compared to the RCP4.5 scenario reduction of 7.7%. Conversely, the amount of RN in the total ND increases under both scenarios with global average increases of 4.8% and 7.5% under the RCP4.5 and RCP8.5 scenarios,

respectively. The increase in the relative amount of RN occurs largely in the interiors of continents in response to increases in future NH₃ emissions, with increases of ~30-40% occurring in North America, East Asia, and Europe. There are also increases in the relative amount of ON by 2.9% and 2.1% on global average following the RCP4.5 and RCP8.5 scenarios, respectively. The increases in the ON percentage is smaller in magnitude under the RCP8.5 scenario because increases in NH₃ emissions are much larger allowing this change to dominate, especially in regions like South America, where increases in the relative amount of RN lead to decreases in the relative amount of ON. Overall, ON amounts appear to increase more in remote regions of continents and the Arctic compared to continental interiors, indicating that ON deposition will increase and replace INON far from source regions and RN will replace INON more closer to populated areas.

BC deposition decreases in both the RCP4.5 and RCP8.5 scenarios in response to reductions in BC concentrations and emissions. The reduction in global average BC deposition is greater under the RCP8.5 scenario ($-4.1 \times 10^{-13} \text{ kg m}^{-2} \text{ s}^{-1}$) compared to the RCP4.5 scenario ($-2.0 \times 10^{-13} \text{ kg m}^{-2} \text{ s}^{-1}$) mirroring the trend in surface BC concentration in Section 2.3. These global reductions in BC deposition likely indicate a reduction in the snow albedo feedback [Warren and Wiscombe, 1980; Clarke and Noone, 1985] from BC in the future under both RCP scenarios. However, increases in BC deposition in South Asia including minor increases over the Tibetan Plateau may impact the snow melt of this region and thus have an impact on the region's water supply [Qian et al., 2011].

SO_x deposition ($\text{SO}_x = \text{SO}_2 + \text{SO}_4^{2-}$) decreases under both the RCP4.5 and RCP8.5 scenarios by $3.2 \times 10^{-12} \text{ kg m}^{-2} \text{ s}^{-1}$ and $2.1 \times 10^{-12} \text{ kg m}^{-2} \text{ s}^{-1}$, respectively. The largest decreases in both scenarios occur in the eastern U.S., Mexico, East Asia, and Europe. This decrease is in

response to decreasing SO₂ emissions in both scenarios. This implies that many of the industrial regions of the NH will be less susceptible to damage from acid deposition under these climate policy scenarios. There are areas of increasing SO_x deposition in both South Asia and central Africa under both the RCP4.5 and RCP8.5 scenarios in response to increasing SO₂ emissions with larger increases in South Asia under the RCP8.5 scenario (e.g., 1.0×10^{-11} – 1.6×10^{-10} kg m⁻² s⁻¹) compared to the RCP4.5 scenario (general less than 1.0×10^{-11} kg m⁻² s⁻¹). As a result both central Africa and South Asia will likely experience greater negative effects from acid deposition, especially following the RCP8.5 scenario by 2050.

2.5 Trends in Aerosol-Cloud Interactions

Figure 7 shows the differences in CCN5, CDNC, COT, and SWCF between the current and Future periods under the RCP4.5 and RCP8.5 scenarios. Reduced aerosol loadings from reduced emissions in both climate policy scenarios result in global average reductions in CCN5 with a greater reduction under RCP4.5 (-1.8×10^7 cm⁻²) compared to RCP8.5 (-8.4×10^6 cm⁻²). The smaller reductions in CCN5 under the RCP8.5 scenario are related to the smaller reduction in PM_{2.5} and PM₁₀ under this scenario but are also likely linked to smaller reductions in new particle formation, since the organic conucleation treatment in CESM-NCSU could compensate some of the reductions in H₂SO₄ levels due to higher SVOC levels from larger O₃ and OH production. Despite the smaller reductions in CCN5 under the RCP8.5 scenario, the global average reductions in warm cloud CDNC are similar with global average reductions of 11.9 cm⁻³ under the RCP4.5 scenario and 11.6 cm⁻³ under the RCP8.5 scenario. The changes in CDNC are similar under both scenarios because, in addition to reduced aerosol loading, there is reduced cloud formation over the continents under both scenarios as discussed in Section 3.4. This reduces the ability of aerosols to activate, resulting in decreased CDNC regardless of changes in

CCN5 levels. An example of this is South America under the RCP8.5 scenario, where CCN5 levels increase from increasing SOA but CDNC still decreases due to reduced CF. However, there is a possible inconsistency between the current and future simulations since the definition of “warm cloud CDNC” (defined as clouds between 960-850 mb from current decade simulation) may change for future years due to warmer temperatures and greater cloud heights.

COT declines on global average under the RCP4.5 scenario by ~ 0.04 but increases under the RCP8.5 scenario by ~ 0.10 . Under both scenarios, COT is generally reduced over land, especially in Europe, East Asia, North America, and South America. These reductions largely result from reduced CF and CWP. However, the reductions in COT over Europe, East Asia, and eastern North America are enhanced via reduced CDNC from reduced aerosol loadings. Under both scenarios, COT is enhanced over the South Asia, Africa, the Arctic, the Southern Ocean, and the southern Pacific Ocean. Enhancements in South Asia and Africa are related to increases in aerosol loadings from enhanced emissions. Enhancements over the Arctic are largely result from increased cloud formation from the increased warming and evapotranspiration. Enhancements in the Southern Ocean are large related to increases in sea-salt aerosol from enhanced WSP10, while the enhancements in the southern Pacific Ocean result from a combination of enhanced sea-salt and enhanced cloud formation. The global average increase under RCP8.5 occurs because cloud formation in the Arctic, sea-salt increases, and aerosol loading in South Asia and Africa are larger under this scenario and compensate the largely continental decreases.

SWCF is increased under both scenarios with a smaller increase of 0.4 W m^{-2} under the RCP4.5 scenario compared to an increase of 0.5 W m^{-2} under the RCP8.5 scenario. SWCF increases are dominant in both scenarios because the increase in COT over the Arctic and the

oceans results in much stronger scattering compared to the reductions over the continents. This is especially true along the Arctic Ocean coastlines where increases in CF and CWP and relative increases in sea-salt are the largest, resulting in increases of 18-31 W m^{-2} in SWCF. Increases in SWCF over South Asia and Africa from enhanced aerosol loadings are much smaller (less than 9.0 W m^{-2}) and may also be partially related to enhanced cloud formation in these regions. SWCF is enhanced under the RCP8.5 scenario because of greater cloud formation in the Arctic. This enhanced SWCF in both scenarios causes the global average dimming of FSDS discussed in Section 2.1.

3. Impact of Climate Change on Air Quality

In this study the impact of climate change on future air quality is examined following the RCP4.5 scenario. As discussed in Part I, only one sensitivity experiment utilizing current period emissions in the future climate is conducted because of limited computational resources. The RCP4.5 scenario is selected for the following sensitivity analysis because it represents stronger climate mitigation and emission control policies and thus better illustrates the impacts these policies will have on future air quality and climate. Figure 8 shows the absolute changes in CO , O_3 , $\text{PM}_{2.5}$, PM_{10} , $\text{PM}_{2.5}$ (no dust), PM_{10} (no dust), dust, biogenic SOA (BSOA), SO_4^{2-} , and Na^+ between the RCP4F_GHGF and RCP4C simulations. CO increases globally with a 7.5 ppb on global average. This increase in CO is in response to both increases in CO biogenic emissions but also secondary formation from the increase in biogenic VOCs. The CESM-NCSU model predicts an increase in surface O_3 of 0.3 ppb on global average due to climate change under the RCP4.5 scenario. This differs from the findings of both *Dentener et al.* [2006] and *Doherty et al.* [2013], that found decreases in surface O_3 in response to climate change from enhanced removal of excited atomic oxygen (O^1D) due to increases in the future water vapor level.

In the CESM-NCSU simulation O_3 is reduced in the tropics and the Arctic by up 4.7 ppb. While it is possible that these reductions in O_3 are caused by the increased water vapor level, it is more likely that the reductions in FSDS from increased cloud formation in these regions leads to the reduction in O_3 . The increases in O_3 predicted by CESM-NCSU also do not follow similar patterns in previous studies, where O_3 increases from climate change largely occur in regions with high NO_x levels [Doherty *et al.*, 2013]. Instead the increases tend to occur in latitudinal bands ranging from approximately 10°S to 30°S in the SH and 20°N to 50°N in the NH. These areas do include industrialized regions like Asia, Europe, and North America, therefore increases in O_3 formation from increased temperatures in high NO_x conditions cannot be ruled out as a possible contributor to the increases in O_3 . There may also be additional regional enhancements from meteorological changes as regions such as the eastern U.S. and Europe have the stronger increases in O_3 corresponding to increases in FSDS (not shown) and reductions in WSP10 that would both lead to enhancements in surface O_3 . However, the most likely reason for increases in O_3 predicted by CESM-NCSU is increases STE of O_3 over these regions. Figure S5 in the supplementary material shows the absolute difference in the zonal mean vertical cross section of O_3 between the RCP4F_GHGF and RCP4C simulations. This figure shows increases in O_3 descending from the upper atmosphere over the latitude bands that experience increases in surface O_3 . This increase in STE of O_3 is likely linked to the strengthening of the Brewer-Dobson circulation that has been projected to occur due to climate change [Buchart *et al.*, 2010].

The deviation of the CESM-NCSU simulation from previous work may be linked to model biases. For example, in Part I it was shown that CESM-NCSU overpredicts surface O_3 due to errors in model deposition and titration and the evaluation of TOR in five stratocumulus regions revealed possible overpredictions of STE. These two biases combined may contribute to

this predicted increasing trend in O₃ from climate change. The deviation in O₃ predictions may also be a limitation associated with the CB05GE gas phase mechanism. The CB05GE mechanism as part of the Global Weather Research and Forecasting Model with Chemistry (GU-WRF/Chem) [Zhang *et al.*, 2012] also predicts an increasing trend in O₃ due to climate change. These findings are based on future climate simulations using the older IPCC AR4 SRES A1B scenario that are part of a publication currently submitted for peer review. The common trend of increasing O₃ using two different future climate scenarios and two different models that use CB05GE may indicate that the pathway of O₃ destruction due to enhanced water vapor is not well represented in CB05GE.

In response to climate change PM_{2.5} and PM₁₀ decrease on global average by ~0.3 and ~1.0 µg m⁻³, respectively. This decrease in aerosol levels is dominated by decreases in dust concentrations of ~1.3 µg m⁻³ on global average. As mentioned in Section 4.3, this decrease in dust is driven by decreases in WSP10 over land that reduce dust emissions in the large dust source regions of North Africa, the Middle East, and East Asia.

In order to understand the impact of climate change on the remainder of the aerosol species, the differences in PM_{2.5} and PM₁₀ without considering the dust modes are shown in Figure 8. Without the influence of dust PM_{2.5} increases by 0.04 µg m⁻³ on global average. This increase is driven by BSOA that increases by ~0.05 µg m⁻³ on global average in response to increasing biogenic VOCs and dominates non-dust aerosol changes in the tropics. There are also decreases in non-dust PM_{2.5} (e.g., 0.6-3.0 µg m⁻³) that occur over North America, Europe, and Southeast Asia. In order to examine the reasons for the additional changes in aerosol levels the remaining aerosol species including: POM, SVOA, anthropogenic SOA (ASOA), NH₄⁺, NO₃⁻, and Cl⁻, as well as changes in planetary boundary layer height (PBLH), WSP10, and PR between

the RCP4F_GHGF and RCP4C simulations are shown in Figure S6 of the supplementary material. BC is not shown because differences in this aerosol component are negligible. The decreases in non-dust $\text{PM}_{2.5}$ over North America and Europe are primarily the result of decreases in SO_4^{2-} , NH_4^+ , and NO_3^- . These decreases occur in conjunction with increases in PBLH indicating that they are likely the result of enhanced vertical mixing in continental interiors from reduced cloud formation. However, the decreases in NO_3^- are greater over much of central North America, indicating that this reduction is also driven by enhanced volatility of NH_4NO_3 from the warmer climate. The decreases in non-dust $\text{PM}_{2.5}$ in Southeast Asia are largely the result of decreases in SO_4^{2-} , ASOA, NH_4^+ , and SVOA. These decreases occur in conjunction with large increases in PR in this region. The increased PR and the fact that all four of the most impacted species are fairly hydrophilic indicate that this decrease is likely from enhanced wet removal.

Non-dust changes in PM_{10} (e.g., global average increase of $0.4 \mu\text{g m}^{-3}$) are driven by increases in sea-salt ($\text{Na}^+ + \text{Cl}^-$) that occur from increases in marine WSP10. It is important to note that due to limited computational resources a non-forced future climate run was not completed and as a result the impact of internal model variability on the results discussed in this section cannot be ruled out. However, most of the changes in future aerosol levels are driven by: (1) reductions in continental cloud cover, (2) reductions in continental wind speeds, (3) increases in maritime wind speeds, (4) increases in near surface temperatures, and (5) increases in tropical precipitation that have been shown to be both consistent with not only CESM1-CMIP5 but also other models that are part of the CMIP5 comparison project. This indicates that these impacts of climate change are fairly robust and thus the results presented here are likely reliable.

4. Impact of Emission Changes on Aerosol Direct and Indirect Effects

In this section the changes between the RCP4F and RCP4F_GHGF simulations are compared to estimate the impacts of the changes in emissions under the RCP4.5 scenario on the future climate through the aerosol direct and indirect effects. In order to illustrate these impacts, Figure 9 shows the changes in AOD, CCN5, CDNC, CF, LWP, COT, PWV, PR, SWCF, FSDS, PBLH, and T2 between the RCP4F and RCP4F_GHGF. Figure S7 shows additional absolute differences in clear sky downwelling shortwave radiation at the surface (FSDSC), latent heat flux (LHX), sensible heat flux (SHX), average lower troposphere relative humidity (LTRH), average lower troposphere temperature (LTT), sea level pressure (SLP), convective precipitation rate (PRC), and large scale precipitation rate (PRL). The reduction in emissions of many primary aerosol and secondary aerosol precursors results in an AOD reduction of 4.0×10^{-2} on global average. From a regional standpoint the changes are the largest in South Asia, East Asia, and Indonesia, with increases in the range of 0.02-0.14 over South Asia and strong decreases in the range of 0.05 – 0.20 over East Asia and Indonesia. There are more moderate decreases in AOD over Europe, North America, and central South America, with decreases typically in the range of $5.0 \times 10^{-2} - 5.0 \times 10^{-1}$. Trends in Australia and Africa are more difficult to discern due to small magnitudes and localized differences in the direction of changes. The global average reduction in AOD is larger than that between the current and future simulations shown in Figure 1, indicating the enhanced sea-salt from greater WSP10 and increased SOA from warming play an important role in future AOD. Regionally, these climate change differences are the strongest in South America, Africa, and Australia as illustrated by the large differences in AOD changes between Figure 1 and Figure 9, while the strong similarities in North America, East Asia, South Asia, and Europe indicate that these regions are more dominated by emission projections. The decreased AOD results in a global mean decrease of $\sim 0.3 \text{ W m}^{-2}$ in FSDSC, as shown in Figure

S7. Regionally, AOD reductions increase FSDSC in the range of 0.2-3.0 W m⁻² in South America and North America, 0.8-4.0 W m⁻² in Europe, 0.4-7.0 W m⁻² in Indonesia, and 1.3-12.9 W m⁻² in East Asia. The increased AOD in South Asia results in dimming of 1.0-9.4 W m⁻². In Africa and Australia, there is a mixed signal in radiation changes similar to those that occur in AOD, however, there is a pattern of dimming in the range of 0.6-2.0 W m⁻² in central Africa where aerosol loadings increase.

The emission reductions decrease CCN5 on global average by 2.2×10^7 cm⁻². Increasing emissions result in increased CCN5 levels of $3.2 \times 10^7 - 1.2 \times 10^8$ cm⁻² in South Asia and smaller increases of 2.0×10^7 - 8.0×10^7 cm⁻² in portions of central Africa. Reductions in CCN5 are the smallest in Australia (e.g., 9.8×10^6 - 3.2×10^7 cm⁻²) and South America (e.g., 2.3×10^7 - 2.2×10^8 cm⁻²) and the largest in Indonesia (e.g., 2.1×10^7 - 4.3×10^8 cm⁻²) and East Asia (e.g., 1.5×10^8 - 5.7×10^8 cm⁻²). Reductions in CCN5 of 1.6×10^8 - 1.9×10^8 cm⁻² occur in Europe and reductions of 1.5×10^7 - 2.6×10^8 cm⁻² occur in North America. The larger range in reduction over North America reflects greater emission reductions in eastern North America compared to western North America. The global mean reduction in CCN5 leads to similar global mean reductions in “warm cloud” CDNC (-11.2 cm⁻³) with decreases over Europe and much of North America ranging from 50.0-145.0 cm⁻³ and greater decreases of 50.0-267.0 cm⁻³ occurring in East Asia. Smaller decreases are simulated over Australia (1.0-20.0 cm⁻³), Indonesia (6.0-80.0 cm⁻³), South America (1.0-60.0 cm⁻³), with greater complexity in the sign of the changes. The increasing CCN5 in the regions of South Asia and portions of central Africa results in increasing CDNC in the range of 13.0-62.0 cm⁻³. Similar to changes in AOD, a comparison of changes in CCN5 and CDNC between Figure 9 and Figure 7 shows enhanced CCN5 and CDNC levels over the SH between the current and future periods that are not caused by anthropogenic emission changes. This differs from trends in

the NH that mirror the changes caused by anthropogenic emissions. Overall, this indicates that emission changes under the RCP4.5 scenario have a stronger impact on aerosol cloud interactions in the NH than the SH. Despite these differences in the SH, the global mean changes in CCN5 and CDNC are quite similar to those shown in Figure 7, indicating that overall changes in emissions under the RCP4.5 scenario dominate the changes in aerosol-cloud interactions. The greater decrease in CDNC in Figure 7 compared to Figure 9 also lends further evidence to support the reduced cloud formation in the future climate as a reason for reduced aerosol activation and reduced CDNC in the future period.

The impacts of changes in aerosol loadings on CF are more complex than the changes in AOD or CDNC due to a number of complicating factors. CF decreases by 0.2% on global average under the RCP4.5 scenario due to emission projections. In general over the continental regions of Europe, South America, Indonesia, and Australia, CF is reduced by ~0.6-3.0%, with greater reductions of up to 5.5% in East Asia and South Asia. However, there are areas of greater cloud formation over the CONUS region where CF increases by 1.0-2.5% and the central Pacific where CF increases up to 8.7%. The reasons for the continental decrease in CF are difficult to discern as is the reason for the difference in behavior over North America. This is because CF can be impacted by changes in FSDSC from aerosol loadings but also changes in radiation from changes in CDNC. This is a cycle of feedbacks in which it is difficult to determine causes and effects since only the net impacts of the feedback cycle are apparent in the decadal averages. In general the increases in FSDSC from reduced AOD in Europe, North America, South America, and East Asia increase the surface LHX (see Figure S7) in the range of 2.0-6.0 W m⁻² with larger increases in North America and East Asia compared to Europe and South America. Similarly, the decrease in FSDSC decreases the LHX in South Asia and central Africa by up to 22.3 W m⁻².

755 The increased global average LHX increases PWV by 0.2 mm on global average. Increases
756 occurring over Europe, South America, and East Asia are generally smaller (0.2-0.7 mm) than
757 those in North America (0.5-1.0 mm). This somewhat mirrors the magnitude of the changes in
758 LHX and does not correspond well with the magnitudes of changes in FSDSC. This is likely
759 caused by differences in land surface, as the impact of aerosol direct effects on ecosystems has
760 been shown to vary significantly due to plant phenology [*Chen and Zhuang, 2014*]. In addition to
761 the continental changes, there are strong increases (up to 5.5 mm) in PWV in the central Pacific
762 and strong decreases in PWV (up to 2.1 mm) in the Northern Indian Ocean and Indonesia.

763 These changes are likely linked to aerosol effects impacting atmospheric and ocean
764 dynamics, as such aerosol radiative effects have been shown to impact oceanic heat transport and
765 current systems [*Cai et al., 2006*]. One of these dynamical feedbacks is a weakening of the
766 Walker circulation. The Walker circulation is the large scale atmospheric flow pattern that
767 describes meridional momentum flow in the tropics and is known at the surface as the Trade
768 Winds. One indicator of the weakening in the Walker circulation is the reductions in sea level
769 pressure (SLP) in the eastern Pacific and increases in SLP over the western Pacific and Indian
770 Ocean. The weakening of the Walker circulation is consistent with the warming induced impact
771 due to the emission projections under the RCP4.5 scenario, since similar weakening of the
772 Walker circulation has occurred in response to warming from anthropogenic forcing [*Vecchi et*
773 *al., 2006*]. The weakening in the Walker circulation also explains the increase in cloud and
774 convective activity in the central Pacific and central Africa and weakening in areas surrounding
775 Indonesia, Australia, and South America. The decrease in LHX over South Asia also results in a
776 PWV reduction of 0.2-1.4 mm. The loss of PWV in South Asia, Indonesia, and Australia and
777 increases in PWV in North America and the central Pacific are adequate to account for the

changes in CF in these regions from enhanced water vapor availability. However, these changes do not support the reductions in other continental areas. The reductions in CF over the remaining continents are the result of reduced saturation as evidenced by the reductions in the average LTRH, which is defined here as the average RH from the surface to ~500 hPa shown in Figure S11. The average LTRH decreases over East Asia, Europe, South America, South Asia, central Africa, and Australia in the range of ~ 0.5-4.2%, while the average LTRH increases by 0.2-2.5% over the CONUS region. The decreased RH occurs because there is an increase in the average LTT, as shown in Figure S7, from greater warming associated with increases in FSDSC. LTT increases are greater over the East Asia, Europe, and South America regions typically in the range of 0.3-0.9°C, while warming over North America is smaller typically in the range of 0.1-0.7°C. This is related to both the changes in LHX and SHX, since in North America most of the increased radiation increases LHX, that do not directly increase atmospheric temperatures, while the remaining regions experienced stronger increases in SHX that warm the atmosphere. This disparity is likely linked to differences in the underlying land characteristics as mentioned previously. These factors contribute to the reduced warming and greater water vapor availability in North America and are the apparent cause of the enhanced cloud formation in this region compared to the other continents, however, the impact of dynamical changes this region cannot be ruled out as increases in moisture over portions North America is consistent with a weakening of the Walker circulation.

Reduced aerosol loadings from emission projections reduce LWP on global average by 1.3 g m⁻². The decreases of LWP in Europe (e.g., by 2.0-17.0 g m⁻²), East Asia (e.g., by 5.0-51.3 g m⁻³), South America (e.g., by 2.0-17.0 g m⁻²), Indonesia (e.g., by 6.0-30.0 g m⁻²), and eastern North America (e.g., by 1.6-12.0 g m⁻²) are related to decreases in CCN5 that reduce

condensation. This is especially true in North America where LWP declines despite increases in LTRH and cloud formation. The changes in LWP are complex in South Asia but generally there is a dominant trend for decreases typically in the range of 0.7-12.0 g m⁻² due to the reduction in cloud formation despite increases in CCN and CDNC. Additionally, LWP is reduced over Australia by 2.0-11.0 g m⁻² in response to reduced cloud formation from the weakening of the Walker circulation. LWP is increased in central Africa (e.g., 0.2-15.0 g m⁻²) from greater CCN5 levels and the central Pacific (e.g., 1.0-21.1 g m⁻²) from greater cloud formation. COT is reduced on global average by ~0.4 in response to decreases in cloud albedo and cloud formation from reduced CF, CDNC, and LWP. The reductions in COT are smaller in Australia, South Asia, South America, and North America typically in the range of 0.02-0.11 compared to larger reductions of 0.06-0.17 in Europe, 0.05-0.21 in Indonesia, and 0.02-11.6 in East Asia. The largest increases in COT occur in central Africa (0.03-0.07) and the central Pacific (0.3-4.2). Globally the reductions in COT due to emission projections are greater than those shown in Figure 11 because of enhanced cloud formation in the Arctic, South Asia, and central Africa that enhance COT. However, over the continents COT reductions are larger than those that occur from declining emissions, illustrating the enhancement from reduced continental cloud formation.

Reduced COT and cloud formation result in a global mean reduction in SWCF of 0.23 W m⁻². The impact on SWCF is the strongest in the vicinity of Indonesia (decreases of 5.6-12.6 W m⁻²) and the central Pacific (increases of 2.0-18.9 W m⁻²). SWCF is reduced in Australia, Europe, and South America typically in the range of 0.5-7.0 W m⁻², while decreases are generally larger in South Asia (e.g., 1.2-11.5 W m⁻²) and East Asia (e.g., 1.3-8.0 W m⁻²). In central Africa, COT enhancements lead to enhancements in SWCF of 0.5-5.5 W m⁻². The changes in SWCF are more

824 complicated over North America with reductions of $0.2\text{--}4.5\text{ W m}^{-2}$ in the north and west from
825 reductions in aerosol loading and increases of $1.5\text{--}5.0\text{ W m}^{-2}$ in the south and west from
826 enhanced cloud formation. The global mean reductions in SWCF and AOD lead to reductions in
827 FSDS of 0.7 W m^{-2} on global average. The impact of reduced AOD and SWCF enhances FSDS
828 in the range of $1.0\text{--}11.0\text{ W m}^{-2}$ in Europe and South America with a greater enhancement of 2.0--
829 18.7 W m^{-2} in East Asia. Although there is a strong direct impact on radiation in Indonesia from
830 aerosol reductions, the changes in the Walker circulation have a greater impact on FSDS in this
831 region, leading to increases in the range of $3.0\text{--}15.0\text{ W m}^{-2}$. This process dominates over the
832 aerosol impacts in Australia resulting in increases of FSDS in the range of $0.9\text{--}5.5\text{ W m}^{-2}$. In
833 central Africa, the increases in AOD and SWCF from emission projections lead to dimming of 0.3--
834 6.2 W m^{-2} . Changes in FSDS over North America and South Asia from emission projections are
835 the most complex representing compensation of impacts from aerosol direct and indirect effects.
836 In South Asia, FSDS is decreased in the range of $1.0\text{--}9.0\text{ W m}^{-2}$ over northern India in response
837 to a dominance of the aerosol direct effect from enhanced emissions, while FSDS increases in
838 the range of $1.0\text{--}9.0\text{ W m}^{-2}$ over the remainder of South Asia in response to dominant changes in
839 aerosol indirect effects. In North America, FSDS increases in the north and west in the range of
840 $0.2\text{--}7.5\text{ W m}^{-2}$ in response to reduced AOD and SWCF, while enhanced SWCF from greater
841 cloud formation over the southwest North America leads to dimming in the range of $1.0\text{--}5.0\text{ W}$
842 m^{-2} . These changes in radiation from aerosol direct and indirect effects have a strong impact on
843 air pollution meteorology and climate. In terms of air pollution meteorology, the global increase
844 in radiation results in a global average increase in PBLH of 1.0 m . The largest increases occur in
845 Europe, South America, South Asia, East Asia, and Australia mirroring changes in SHX. These
846 increases provide a positive feedback to emission reductions as the increase in PBLH will

enhance vertical mixing, leading to greater reductions in surface pollution levels. The increases in FSDS also result in a general global warming trend of 0.2°C on global average with general warming of 0.2-1.2°C over most continental regions with greater increases in Europe, South America, and East Asia, compared to North America, Indonesia, Africa, South Asia, and Australia.

The impact of the RCP4.5 emission projection on global precipitation is dominated by changes in tropical convective precipitation related to the weakening of the Walker circulation, resulting in increases in PR over the central Pacific of 0.2-2.4 mm day⁻¹. This process also results in decreases of precipitation by 0.1-2.8 mm day⁻¹ in the vicinity of Indonesia, South Asia, Australia, and South Asia. In the NH, PRC is increased in North America (e.g., 0.02-0.3 mm day⁻¹) and East Asia (e.g., 0.01-0.3 mm day⁻¹) in response to greater LHX in these regions and the warming from reduced aerosol loadings. The direction of the impact over Europe is more complicated likely from competing influences from the larger increase in SHX compared to LHX in this region. In North America the increase in large scale cloud formation also leads to increases in PRL of 1.0×10^{-3} -0.7 mm day⁻¹ since the greater water level and reduced aerosol loading lead to bigger droplets as evidenced by the 0.01-0.7 µm increases in average cloud droplet effective radius (Figure not shown). There are mixed changes in PRL in East Asia and Europe with decreases in the range of 0.1-0.5 mm day⁻¹ and increases in the range of 0.01-0.1 mm day⁻¹. These reflect similar complex changes in average cloud drop radius likely from competing influences from reduced CDNC and reduced LWP. The combined effect in total PR is a mixed increase and decrease in East Asia, reflecting the dominant changes in PRL and a slight decrease in Europe.

The changes induced by the RCP4.5 emissions projections are similar to those of *Westervelt et al.* [2015] that reported global average changes of -0.87 g m^{-2} , $\sim 0.08 \text{ mm day}^{-1}$, and 0.81°C in LWP, PR, and T2, respectively. These changes are more extreme than the 0.2°C and $\sim 0.02 \text{ mm day}^{-1}$ increases in T2 and PR, respectively, predicted by CESM-NCSU. This is likely because the work of *Westervelt et al.* [2015] examined the changes caused by emissions reductions by 2100 rather than 2050 when emissions reductions are stronger. However, the changes in LWP predicted by CESM-NCSU are more extreme with reductions of -1.3 g m^{-2} . This is likely due to differences in aerosol activation parameterizations between the models as well as differences in simulated time period.

5. Summary and Conclusions

In this work, the CESM-NCSU model is used to explore changes in the future state of the atmosphere due to climate change and emissions, estimate the impact of future changes in climate on air quality, and estimate the impact of emissions changes on future climate. Changes in meteorology, radiation, clouds, and AOD between the CESM-CAM5.1 simulations from the CMIP5 project and CESM-NCSU are generally comparable under both the RCP4.5 and RCP8.5 scenarios. However, differences exist in many meteorological variables in the SH due to cooling over this region in CESM-NCSU. Both models show the impact of changes in aerosol-climate interactions over the NH industrial areas in response to declining aerosol levels under both RCP4.5 and RCP8.5 climate policy scenarios.

The trends of most trace gases at the surface tend to follow the trends of their emissions which are typically decreasing in both scenarios by 2050. Global average surface O_3 experiences a decrease of 1.4 ppb under RCP4.5 and an increase of 2.4 ppb under RCP8.5, due to increased

891 O₃ production from weaker NO emission reductions, large increases in CH₄, larger increases in
892 anthropogenic and biogenic TNMVOC emissions, and warmer temperatures. This trend is
893 consistent with other modeling studies following these scenarios [Young *et al.*, 2013; Kim *et al.*,
894 2015]. There are also decreases in OH under both scenarios from enhanced destruction by
895 increases TNMVOC and CH₄ consistent with the ACCMIP modeling studies [Voulgarakis *et al.*,
896 2013]. This enhanced oxidation in conjunction with greater increases in shipping emissions
897 increase the background CO level under the RCP8.5 scenario, while smaller increases in
898 oxidation and shipping emissions and greater emissions reduction lead to a decrease in CO under
899 the RCP4.5 scenario. PM_{2.5} is reduced on global average by 0.7 µg m⁻³ under RCP4.5 and 0.2 µg
900 m⁻³ under RCP8.5. These reductions are driven by reductions in anthropogenic emissions of
901 primary aerosol species and many precursor gases. However, natural reductions in dust from
902 reduced WSP10 in high dust emission regions play a role. Reductions in simulated PM_{2.5}
903 concentrations are less under RCP8.5 scenario due to smaller reductions in anthropogenic and
904 dust emissions, larger sea-salt emissions, and larger SOA formation. These differences in sea-salt
905 and dust emissions between the RCP4.5 and RCP8.5 scenarios lead to a global average reduction
906 in PM₁₀ of 1.2 µg m⁻³ under the RCP4.5 scenario but an increase of 0.2 µg m⁻³ under the RCP8.5
907 scenario. In general, trends in air pollutants by the year 2050 under both the RCP4.5 and RCP8.5
908 climate policy scenarios are similar with key differences linked mainly to large feedbacks from
909 large increases in CH₄ from the lack of widespread implementation of carbon capture and storage
910 technologies (CCS) technologies in the RCP8.5 scenario. Thus this illustrates the importance of
911 climate/carbon policies and international cooperation for future climate mitigation and air quality
912 improvement as reductions in typical air pollution precursor emissions are not necessarily
913 sufficient to reduce future air pollution levels.

Decreases in aerosol loading and cloud formation lead to global average decreases in CCN5, CDNC, and CWP under both RCP scenarios. Under the RCP4.5 scenario, COT is reduced on global average in response to the large reductions in CDNC, LWP, and CF over continental regions but under the RCP8.5 scenario this is compensated by stronger increases in cloud formation over the Arctic and Southern Ocean. The increase in cloud formation over the Arctic and the oceans dominates changes in SWCF, leading to a global average increase of 0.4 W m^{-2} and 0.5 W m^{-2} under the RCP4.5 and RCP8.5 scenarios, respectively. The increases in SWCF over the Arctic and oceans and greater AOD over South Asia and Africa compensate increases in FSDS from decreases in AOD and SWCF over the remaining continents, resulting in global dimming of 1.1 W m^{-1} under the RCP4.5 scenario and 2.0 W m^{-2} under the RCP8.5 scenario.

Climate change following the RCP4.5 scenario increases O_3 by ~ 0.3 ppb on global average largely in response to enhanced STE of O_3 . This differs from previous work that report decreases in global O_3 from climate change [Dentener *et al.*, 2006; Doherty *et al.*, 2013]. This indicates that the enhancement of O_3 from climate change is possible either the result of model biases in surface O_3 and STE or a limitation of the CB05GE mechanism. Climate change leads to a reduction in both the global average $\text{PM}_{2.5}$ and PM_{10} level by ~ 0.3 and $\sim 1.0 \mu\text{g m}^{-3}$, respectively. These changes are dominated by reductions in dust concentrations but increases in BSOA, sea-salt, PBLH, and PR also play a role in future aerosol levels.

The reductions in pollutant emissions under the RCP4.5 scenario lead to global average increases of 0.3 W m^{-2} , 0.7 W m^{-2} , 0.2°C in clear sky downwelling solar radiation at the surface, all sky downwelling solar radiation at the surface, and near surface air temperatures, respectively. The higher incoming radiation and warmer temperatures result in increases in

PBLH that would further reduce pollution levels through vertical mixing. However, the global warming signal induced from emission reductions leads to a weakening of the Walker circulation in the tropics, resulting in strong changes in tropical precipitation and cloud patterns and a general shift in global climate towards the negative phase of the El Nino Southern Oscillation. These findings illustrate the importance of utilizing earth system modeling when conducting simulations of global aerosol effects since it allows for feedbacks of chemical species to atmospheric/oceanic circulation systems. These findings also imply a fairly strong “climate penalty” from the reduced emission levels. However, this should not imply that reduced air pollution emissions are poor policy decision as there will be benefits to human health. Rather, this further underscores the importance of climate/carbon policies as the best mechanism to mitigate the negative impacts from climate change.

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Figure Captions

Figure 1. A comparison of the spatial trends in available meteorological, cloud, and radiation variables between CESM-NCSU and CESM1-CMIP5 simulations under the RCP4.5 scenario.

Figure 2. A comparison of the spatial trends in available meteorological, cloud, and radiation variables between CESM-NCSU and CESM1-CMIP5 simulations under the RCP8.5 scenario.

Figure 3. A comparison of trends in surface concentrations of trace gases predicted by CESM-NCSU between future and current decades under the RCP4.5 and RCP8.5 scenarios.

Figure 4. A comparison of trends in surface concentrations of aerosol species predicted by CESM-NCSU between future and current decades under the RCP4.5 and RCP8.5 scenarios.

Figure 5. A comparison of trends in surface concentrations of PM_{2.5} and PM₁₀ predicted by CESM-NCSU between future and current decades under the RCP4.5 and RCP8.5 scenarios.

Figure 6. A comparison of trends in deposition of O₃, nitrogen, oxidized sulfur (SO_x), and black carbon, as well as trends in the percentage of oxidized nitrogen, organic nitrogen, and reduced nitrogen that compose the total nitrogen deposition predicted by CESM-NCSU between future and current decades under the RCP4.5 and RCP8.5 scenarios.

Figure 7. A comparison of trends in CCN5, CDNC, COT, and SWCF predicted by CESM-NCSU between future and current decades under the RCP4.5 and RCP8.5 scenarios.

Figure 8. Absolute difference in O₃, CO, PM_{2.5}, PM₁₀, PM_{2.5} (no dust), PM₁₀ (no dust), dust, BSOA, SO₄²⁻, and Na⁺ between the RCP4F_GHGF and RCP4C simulations.

Figure 9. Absolute differences in meteorological, cloud, and radiation variables between the RCP4F and RCP4F_GHGF CESM-NCSU simulation, illustrating the impacts of changing emissions.

Figure 1. A comparison of the spatial trends in available meteorological, cloud, and radiation variables between CESM-NCSU and CESM1-CMIP5 simulations under the RCP4.5 scenario.

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