ISOLATING METEOROLOGICAL INFLUENCES ON $PM_{2.5}$

IN FUTURE EARTH SYSTEM MODEL SIMULATIONS

by

ALISON BANKS

(Under the Direction of Gabriel Kooperman)

ABSTRACT

Biomass and fossil fuel burning impact air quality by injecting fine particulate matter ($PM_{2.5}$) into the atmosphere, which poses a serious threat to human health. However, the concentration of $PM_{2.5}$ depends not only on emissions, but also removal. As the planet warms with greenhouse gas increases, the meteorological conditions that remove aerosol, primarily through rainfall and wet deposition, shift in pattern, frequency, and intensity. This combination of factors can worsen air quality purely through meteorological forces, even without changes in emissions. By fixing aerosol emissions at present-day levels in the Community Earth System Model, but increasing greenhouse gases through the 21_{st} century, an increase in $PM_{2.5}$ (particularly sulfate) is found to be associated largely with precipitation changes. A decrease in wet day frequency (5% globally) contributes to increases in the concentrations of black carbon, primary organic matter, and sulfate (in addition to changes in aqueous production) at the surface.

INDEX WORDS: Earth System Modeling, Climate, Air Quality

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DEDICATION

For my grandmothers, who were strong and tenacious.

Thank you for showing me the way.

ACKNOWLEDGEMENTS

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CHAPTER 1

INTRODUCTION

Air pollution poses a serious threat to human health, and even mild pollution exposure can cause respiratory infections, heart disease, lung cancer and stroke (*EPA*, 2017; *WHO*, 2016). The World Health Organization estimates that 4.2 million people die every year from exposure to fine particulate matter ($PM_{2.5}$; $\leq$ 2.5 micrometer diameter) aerosol alone, which is capable of penetrating lung membranes (*WHO*, 2016). The United Nations Environment Program names air pollution as the "most important environmental health risk of our time" (*UNEP*, 2019). Fortunately, direct anthropogenic emissions are projected to decrease globally as countries implement air quality policy changes through the 21st century (*O'Neill et. al.,* 2016).

In addition to direct pollution emissions however, atmospheric processes can influence the transport and removal of particle pollutants from the atmosphere. Precipitation has a unique role in removing atmospheric pollutants by wet deposition through the processes of in- and below-cloud scavenging (uptake by rain droplets), which "rains out" aerosol particles from the atmosphere that are deposited at the surface (*Poeschl*, 2006). This is a critical, "natural" process that removes pollutants from the atmosphere. In a recent study based on satellite observations, rainfall frequency and

intensity were shown to have a high impact on aerosol burden (or concentration of aerosol particles in a column of the atmosphere), with an increase in removal of 10% globally over the last 30 years (*Hou,* 2018). Similar studies that incorporate observations can be useful for quantifying how realistic climate and Earth system models (ESMs) are in representing these processes (*Gryspeerdt et al.,* 2015), and thus their effectiveness for informing our understanding of future changes.

As the concentration of greenhouse gases increases, meteorological conditions, namely rainfall, are expected to change in ways that may influence the removal of aerosol particles from the atmosphere. Consequently, the projected efficiency of atmospheric cleansing events is in question, which may affect future air quality, even if anthropogenic aerosol emissions were to remain at constant present-day levels. Aerosol concentration has the potential to increase in some regions due to projected changes in rainfall characteristics alone that may result from climate change (*Xu and Lamarque,* 2018). Recent studies using ESMs project the frequency of intense precipitation events will increase, while the frequency of light-to-moderate events are reduced, leading to an overall decrease in total precipitation frequency on a global scale (*O'Gorman,* 2009; *Pendergrass and Hartmann,* 2014; *Sun,* 2007), and a few studies have begun to looked at the consequences of these changes for aerosol pollution (*Allen et al.,* 2015; *Xu and Lamarque,* 2018).

In this study, I simulate aerosol emissions at a continuous present-day rate, and compare the results to a fully coupled model with projected aerosol emissions changes.

Both will utilize SSP585 (Shared Socioeconomic Pathways), which is a modeled simulation of increasing greenhouse gas emissions through the 21st century. By analyzing these simulations, this work sheds light on the potential benefits of lowering aerosol emissions rates in the future. This work adds to our understanding of how changes in the physical climate system, driven by anthropogenic greenhouse gas emissions, can impact air quality by answering the following research questions.

Research Questions

1. If anthropogenic aerosol emissions were to remain constant at present-day rates but greenhouse gases continue to increase, what regions would experience changes in air quality due to climate and precipitation changes?
2. How do the results from a global earth system model simulation with constant aerosol emissions rates compare to a simulation with a "realistic" projected decrease in emissions rates globally over the 21st century?
3. Where is worsening air quality a result of precipitation and wet deposition change versus changes in transport and/or secondary aerosol production, and which processes impact the different aerosol species?

CHAPTER 2

LITERATURE REVIEW

Health Impacts of Fine Particulate Matter

In 2017, outdoor air pollution contributed to 6% of deaths globally, and is responsible for 3.4 million deaths each year (*WHO*, 2016). However, a global annual average statistic concerning air quality does not fully characterize the regional disparity of this environmental health risk. For industrializing low- and middle-income countries, deaths attributed to poor air quality can be 10% or higher. Additionally, regular exposure to fine particulate matter increases the risk of non-communicable diseases (stroke, heart disease, respiratory disease etc.) which have the strongest effect on older populations (*EPA*, 2017). Both severe air quality events and long-term exposure to $PM_{2.5}$ have negative health effects, and zero exposure is ideal. The effects of chronic (long-term) exposure to $PM_{2.5}$ have been assessed in previous studies which find that every 10 $\mu g/m^3$ increase in fine aerosol is associated with a 6% increased risk of mortality (*Pope*, 2002). Other studies that utilize satellite derived products find that acute (short-term) exposure increases mortality in a population by 2.8% for every 10 $\mu g/m^3$ increase (*Kloog et. al.*, 2013). To address this, the World Health Organization set recommended daily exposure limits on $PM_{2.5}$ concentrations (25 $\mu g/m^3$), as well as annual exposure limits (10 $\mu g/m^3$).

Aerosol Processes in the Atmosphere

The concentration of aerosol at a given location depends on sources, sinks, and transport. These factors vary regionally, and local changes to aerosol concentration can be driven by large-scale and global effects. For example, atmospheric conditions such as changes in horizontal transport of aerosols also impact the spatial distribution of aerosol concentration and resulting wet-deposition changes (*Fang*, 2001; *Allen et al.*, 2015). Likewise, aerosol can have a significant impact on the large-scale climate system. For example, a suite of climate model experiments found that widespread drought in South Asia was directly tied to the slowing of the South Asian monsoon, a result of increasing anthropogenic aerosol emissions in the area (*Wang et. al.*, 2015). However, these coupled interactions can be complicated and depend on the covariations and timing of aerosol and atmospheric conditions. Aerosol magnitude and type can be measured globally through their varied optical properties, primarily through scattering and absorption of solar radiation (*Myhre*, 2009).

A. Aerosol Sources

Primary aerosol particles in the atmosphere can come from natural sources such as sea salt and dust, as well as anthropogenic sources like biomass burning and fossil-fuel combustion (*Poschl*, 2005). The line occasionally gets blurred, for example, as wildfires (a natural source of black carbon emissions from biomass burning) can result from natural (lightning) or human ignition, and longer wildfire seasons may be extended by

global warming (*Abatzoglou and Williams*, 2016). Additionally, particles can be emitted directly as solid/liquid particles, or form from secondary gas-phase processes, which nucleate, condense, and coagulate into larger groups that grow into particle-size diameter range (*Jacob*, 1999). Locally, the aerosol concentrations depend in part on emissions as input, but are also influenced by sinks and horizontal transport, which include column removal through wet/dry deposition and lateral transport to/from the region.

B. Aerosol Sinks

Ultimately, aerosol particles leave the atmosphere through either wet or dry deposition. Wet deposition is a process where precipitation reaches the Earth's surface and brings with it cloud condensation nuclei as well as scavenges aerosol particles which are removed from the atmosphere and deposited at the surface (*Poschl*, 2005). These atmospheric cleansing events keep the lifetime of aerosols relatively short, often lingering in the atmosphere for days or weeks (*Jacob and Winner*, 2008). Another major sink of aerosol particle is through dry deposition, which is a process that deposits particulate matter on the surface through gravitational settling. Both of these forms of deposition have conditional meteorological qualities as well as regional preferences that influence the range of lifetimes aerosols may take in the atmosphere (*Poschl*, 2005). I will focus primarily on the largest aerosol sink for $PM_{2.5}$ aerosol, wet-deposition, while also addressing non-precipitation related sinks (dry deposition) in my analysis.

C. Aerosol Transport

Transportation of aerosols also plays a role in regional sources and sinks. For example, the aerosol emissions from Asia can cross the Pacific Ocean, which is widely studied as a contribution to aerosol concentrations in the western United States (*Heald et. al*., 2006). Thus, large-scale circulation changes, such as the poleward expansion and weakening of the Hadley cell, may have the potential to change pollutant transport pathways and processes. After aerosols are vertically lofted, the stronger upper winds of the free troposphere can dramatically change horizontal distribution. It has been found in observational studies that increased $PM_{2.5}$ aloft may contrast with regional decreases near the surface (*Glotfelty*, 2016), but is uncertain in conventional model representation *(Jacob and Winner*, 2009). The dynamics of aerosol transport are also projected to change as global mean temperature increases by 3°C, a result of increasing greenhouse gas emissions. However, some regional effects of global warming and aerosol patterns are already observed. For example, a strengthening of the African easterly jet and summer heat low contribute to desertification in the Sahara, and naturally emitting mineral dust aerosol decreases globally though rainfall pattern shifts and soil changes (*Cook and Vizy*, 2015). These trends in combination emphasize the importance of studying these complex interactions in a global framework, which accounts for seasonality and efficiency of precipitation, as well as changes in transport.

Precipitation in Response to Greenhouse Gas Emissions

Increasing greenhouse gases, which absorb and remit outgoing longwave radiation, have been a growing source of global warming since the early 20th century (*Ramanathan et. al.*, 2001; *IPCC*, 2013). Changes in the climate system directly and indirectly induced by warming can in turn influence the characteristics of precipitation. There is an expectation of increased global average precipitation resulting from radiative balance shifts from global warming by 1-3% for every °C increase (*Allen and Ingram*, 2002). In examining the variation of precipitation on regional scales, the current literature describes a general trend toward wetting over the tropics and high latitudes, and drying over the subtropics (*Held and Soden*, 2006). Global models are needed to assess future changes in large-scale precipitation in response to further increases in greenhouse gases, and generally show an expansion of the tropics (*Seidel et al.*, 2008), poleward shifts of storm tracks (*Yin*, 2005), and increases in severe thunderstorm days (*Diffenbaugh*, 2013). Along with the average increase, a statistical shift to heavy rainfall events (shown in models and observations) is found increasing in frequency (*O'Gorman*, 2009; *Sun*, 2007), while light- to-moderate events decrease (*Kooperman et al.*, 2016; *Pendergrass and Hartmann*, 2014). Though global average rainfall increasing by 3-7% over the 21st century (*Xu and Lamarque*, 2018), reduction of moderate rainfall in these models is observed as a direct response to energetic constraints on precipitation from global warming (*Pendergrass and Hartman*, 2014). This leads to an overall reduction in the frequency of precipitation on global scales.

Using 3D chemical transport models, it becomes clear that the efficiency of aerosol removal, and therefore the lifetime of pollutants in the atmosphere, is more sensitive to the frequency of precipitation than the intensity, but neither of which are accounted for in analysis of annual averages alone (*Hou*, 2018). Given this more nuanced examination of precipitation, the wet deposition removal of particles in the atmosphere is potentially changing due to changes in precipitation frequency, which may alter aerosol lifetime and concentration in some regions (*Xu and Lamarque*, 2018).

Expectations for Aerosol Changes due to Climate Change

As discussed above, aerosol burden depends both on emissions and removal rates, which may be indirectly affected by greenhouse gas induced warming. In a simulation of the 21st century with no change in aerosol emissions, a recent study conducted by *Xu and Lamarque* [2018] found that sulfate, black carbon, and primary organic carbon increased by 5%-10% due to greenhouse gas driven climate changes alone. The authors showed that a decrease in wet removal flux (1%-2% during the 21st century) was correlated with global $PM_{2.5}$ increase at the surface by 3%, which has a more direct consequence and hazard to health than the total column burden (*Xu and Lamarque*, 2018). Through the increase of aerosol concentrations due to meteorological factors that increase their lifetime in the atmosphere, the projected future of air quality around the globe may be of greater concern than previously understood. However, many competing processes

influence aerosol in the atmosphere and there are many unanswered about aerosol concentrations and air quality with respect to these precipitation changes.

CHAPTER 3

METHODS

In this section, I summarize the model and its representation of relevant aerosol processes, simulations and experiment design, and analysis methods used in the study.

Community Earth System Model

In this study, the National Center for Atmospheric Research's (NCAR) Community Earth System Model (CESM2.1.1), with aerosol treatment through the Community Atmosphere Model (CAM6) component, is used to investigate the connection between rainfall and air quality. CESM is a fully coupled Earth system model (ESM) including atmosphere, ocean, land, and ice components (*Danabasoglu et. al*., 2020). The CAM6 atmospheric component of CESM includes aerosol interactions that are ideal for this project. The two-moment cloud microphysics in CAM6 includes state transitions (evaporation, collision, droplet formation and fall) (*Gettelman and Morrison*, 2008), which are linked to wet-deposition and in-and-below cloud scavenging of aerosol. The atmospheric model interacts with changes in large scale patterns such as ENSO, and can be run with Shared Socioeconomic Pathways that simulate varied levels of greenhouse gas emissions trends as well as aerosol emissions.

Figure 1. Components of the Community Earth System Model (*Hurrell et. al*., 2013).

Aerosol Representation in CAM6

In CAM6, aerosols are represented by the Modal Aerosol Module (MAM4), which uses four modes and six species for modeling atmospheric composition (*Liu et. al.*, 2016). The three standard internally mixed modes (maintaining the chemical and physical average properties of all species within) are Aitken, Accumulation, and Coarse in order of smallest aerosol particle to largest (Figure 2.). MAM4 has an additional primary carbon mode that explicitly treats carbonaceous aerosols, and uses a series of parameterizations to represent tropospheric and stratospheric aerosols in the atmosphere. These parameterizations are linked to atmospheric processes in CAM6 (e.g., cloud microphysics and radiation) and larger climate-system processes in CESM (e.g., dust

emissions from the land-surface). MAM4 is a prognostic modal aerosol scheme (*Liu et. al.,* 2016) that represents six species of aerosol (sulfate, black carbon, primary and secondary organic aerosol, sea salt and dust), for which each mode and species has an associated prediction of mass mixing ratio and total number concentration (*Ghan et al.,* 2012). These predictions are influenced by emissions, aging, and transferring between modes through processes like condensation and coagulation. MAM4 also determines a time mean distribution for aerosol lifetime in the upper atmosphere, which evolves with the changing dynamics of the model (*Gettelman and Morrison*, 2008).

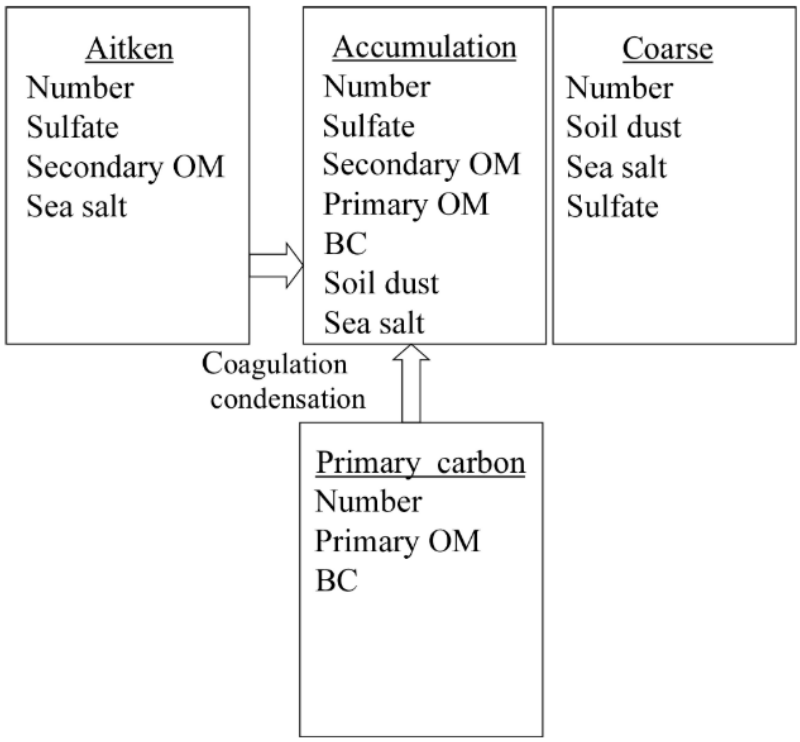

Figure 2. Four modes of classification for aerosol in MAM4 models (*Liu et. al.,* 2016).

Sulfate Representation in CAM6

Of the six species of aerosol represented in the MAM4 classification, SO_4 is unique in its production, which can be influenced by the changing hydrology of the model. The concentration of sulfate in the atmosphere is subject to the same sources, sinks, and transports as other species, but has the additional effect of secondary production by aqueous chemistry and gas-phase reactions. While some SO_4 is directly emitted in the model, SO_2 is also represented as a product of industrial emissions and uses conversion processes (condensation, new particle formation, and thermodynamic partitioning) to form new SO_4 aerosol particles though gas phase oxidation (*Tie et. al*., 2001; *He et. al*., 2015). The set of parameterizations that calculate global distributions of chemical species is called MOZART (Model for Ozone and Related Chemical Tracers), a tropospheric chemistry scheme that converts gas-phase sulfur into sulfate primarily by OH (Hydroxide) via oxidation. CESM2.1.1 uses MOZART-T1, which improves upon secondary aerosol representation and gas-phase reactions (*Emmons et. al*., 2020).

In addition to the gas phase production, sulfate is also produced through aqueous chemistry, a source that is tied to the meteorological conditions examined in this study. Aqueous phase production of sulfate has a strong reliance on the presence of clouds and the pH of cloud droplets. The conversion of SO_2 to SO_4 through the aqueous phase in the presence of cloud water is represented through reactions with ozone and hydrogen peroxide (*Lamarque et. al.*, 2012). Therefore, the availability of atmospheric moisture

and cloud water, which are impacted by climate change, has consequences for the production of this hazardous particulate matter, and is explored further in Chapter 4.

Simulations

A. Shared Socioeconomic Pathways

Climate models are often used to simulate the 21st century for projections of future climate change. In the Coupled Model Intercomparison Project Phase 5 (CMIP5), these simulations were called Representative Concentration Pathway (RCP) scenarios. In this study, I use the updated Coupled Model Intercomparison Project Phase 6 (CMIP6) experiment design to investigate changes in air quality due to greenhouse gas increases that result from socioeconomic and policy change. These CMIP6 simulations are called Shared Socioeconomic Pathway (SSP) scenarios, which differ from RCP scenarios in that they include a narrative element that incorporates urbanization, GDP, and population growth (*O'Neill et. al.,* 2016). In this study, the high emissions (or "business-as-usual") scenario, called SSP5-8.5 (similar to RCP8.5), is used, which sets a baseline for future plans concerning aerosol and greenhouse gas emissions as well as land-use change. In CESM, this is the model component set BSSP585, which includes a fully coupled ocean and sea-ice configuration. In this "realistic" scenario (SSP5-Full), air pollution is managed in many countries such that aerosol emissions generally reduce over the 21st century, yet greenhouse gas emissions continue to rise. SSP5, therefore, examines greenhouse gas driven climate change with a global-scale reduction in aerosol emissions.

B. Investigating Future Changes with Fixed Aerosol Emissions

The results of the standard SSP5 experiment design, described above, are compared to a scenario in which aerosol emissions are held fixed at the present-day rates (Figure 3.) (SSP5-Fixed). This approach follows the methods from Xu and Lamarque (2018) for RCP8.5, and allows for assessment of the climate system under an increased greenhouse gas scenario, which has been predicted to drive a precipitation increase across a global average, but decrease in precipitation frequency. In this scenario, an identical simulation was run with a fixed aerosol emissions signature, cycling aerosol emissions (and related precursor emissions, e.g., SO_2) at 2010 values, with all other variables consistent with SSP5. This included surface emissions, external forcings (3D emissions), and prescribed chemical constituents data. Naturally emitting aerosols such as dust and sea salt in the model are interactive and have the ability to change in the simulation, prompting the need for focused analysis on the anthropogenic species in the model.

Figure 3. Schematic of the two key simulations run in CESM. While both SSP5 simulations include increasing greenhouse gas emissions, (a) SSP5-Full decreases aerosol, and (b) SSP5-Fixed cycles annual aerosol emissions at present-day (2010) rates.

Analysis

A. Present-Day Comparison to Observational Data

To answer the research questions outlined in the introduction, I validated the two CESM simulations described above with observational satellite data. Aerosol validation was provided by Obs4MIP (Observations for Model Comparison Project), a collaborative effort to consolidate datasets for model validation (*Teixeira,* 2018). Most of the datasets that quantify global aerosol properties use Aerosol Optical Depth (AOD), which is measured by the scattering and absorption of radiation at wavelengths reflected to space (*Ramanathan et. al.*, 2001). This can have limitations, as cloudy skies prohibit aerosol detection, and the types of aerosol are inferred by Aerosol Detection Products (ADP) algorithmically. Despite that, satellite data has global and sometimes hourly coverage, and increasingly fine resolution as technology improves.

MERRA-2 (Modern-Era Retrospective analysis for Research and Applications) provides AOD measurements that span a 20-year time period from 2000 to 2020 (*Bosilovich*, 2019), making it comparable to my analysis of simulated AOD from 2015 to 2035 (Figure 4. b). GPCP (Global Precipitation Climatology Project) 1DD (1 degree daily) is a daily output precipitation dataset provided by NOAA (*Pendergrass* 2016; *Huffman*, 2016). By having a robust temporal resolution, and 1-degree grid resolution, this observational data can be compared directly to model results. The long climatology and daily data, allows for a comparison of the annual average of precipitation as well as the frequency of rain days.

B. Future Changes

For the simulations described above, I performed analysis of aerosol column burden as well as surface aerosol concentration (particularly $PM_{2.5}$), focusing on species of aerosol that are represented in MAM4. I assessed aerosol as global averages and regional averages, as well as the 99_{th} percentile of aerosol, and surface concentration of $PM_{2.5}$ above a recommended daily threshold for extreme event analysis. This global understanding of aerosol is followed by analysis of precipitation statistics (average, frequency, and intensity) and its influence on wet deposition rates and deposition efficiency. Additionally, area-weighted averages of climate variables, including aerosol related variables, were calculated throughout. Finally, the species of anthropogenic aerosols are assessed individually and analyzed with respect to their particular characteristics in regards to production and sinks.

CHAPTER 4

RESULTS

The analysis presented below is based on comparisons between 20-year time periods at the beginning and at the end of simulations that represent the 21st century (2016 to 2035 and 2079 to 2098). Unless otherwise stated, the global maps show the annual climatological statistics of aerosol in the atmospheric column (including aerosol burden and surface concentration) and surface precipitation. This includes anthropogenic aerosols such as black carbon (BC), primary organic matter (POM), secondary organic matter (SOA), and sulfate (SO_4), as well as sea salt and dust.

Part 1: Changes in Column Burden and Surface Aerosol Concentrations in SSP5-Full versus SSP5-Fixed Simulations

In this study, I examine changes in air quality through the lens of a mass balance perspective. This includes assessing the sources, sinks, and transport of aerosol. To isolate the dynamic influences on aerosol sinks in a changing climate system, I have employed an experiment design that fixes the sources (emissions of BC, POM, SOA, and SO_4) at present-day (2010) levels, as described above. In order to understand the overall impact of fixing emissions, I first compare simulated aerosol concentrations from this controlled SSP5-Fixed run with the SSP5-Full run, which included changes in aerosol

sources. SSP5-Fixed present-day conditions were validated against satellite observations for accuracy in the representation of aerosol spatial pattern and magnitude (Figure 4.). The Aerosol Optical Depth Analysis (AOD) units are a vertically integrated quantity. This includes extinction coefficients with all six species of aerosol, which are multiplied by the vertical integral. When comparing the AOD satellite product with AOD in the model, "hot-spots" (Central Africa in particular) show in the same regions, as well as global coverage of aerosols such as sea salt and dust. It should be noted that simplifications in conventional ESMs often poorly represent the frequency and intensity of precipitation (*Allen and Soden*, 2008; *Pendergrass and Hartmann*, 2014), leading to uncertainties in aerosol removal, such that Aerosol Optical Depth (AOD) can be overestimated compared to satellite data (*Wang et. al.*, 2011).

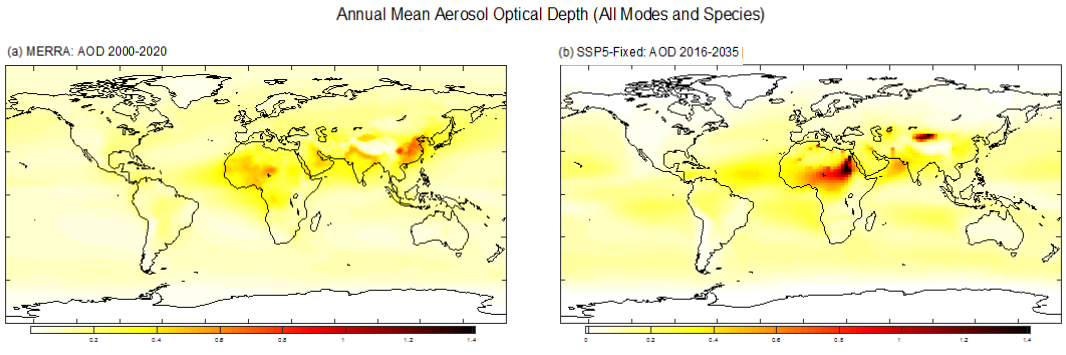

Figure 4. Annual Mean Aerosol Optical Depth for present-day conditions. (a) MERRA-2 AOD (2000-2020) and (b) SSP5-Fixed AOD (2016-2035).

Projecting into the future, the SSP5-Full scenario has an expected decrease in the anthropogenic aerosol burden over most of the world, primarily around India and China (Figure 5 c.). In contrast, despite unchanging emissions in the fixed scenario, there is a general increase over land masses over the 100-year simulation (Figure 5 b.). This difference supports the hypothesis that meteorological changes tend to drive an increase in aerosol burden. However, this figure includes all modes of the aerosol module, including the largest coarse mode aerosol (i.e., larger than $PM_{2.5}$). This analysis also assesses the entire vertical column of aerosol in the atmosphere, not just the surface, which is a more appropriate measure for "air quality."

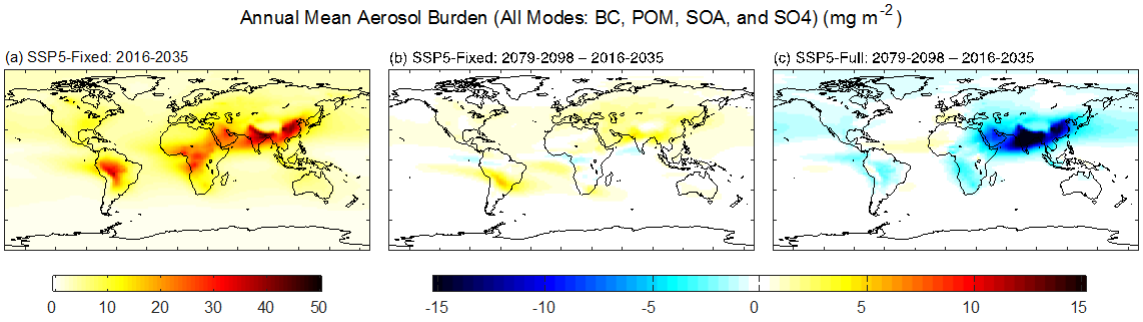

Figure 5. Annual mean aerosol burden for (a) present-day conditions (2016-2035), and changes (2079-2098 minus 2016-2035) from (b) SSP5-Fixed and (c) SSP5-Full simulations. Burden includes all modes for BC, POM, SOA, and SO_4 in units of [mg/m_2].

The fine modes of the modal aerosol module (i.e., Aitken and Accumulation) can be used as a proxy for $PM_{2.5}$ and focus more directly on the fine particles that would

affect human health at the surface. Though there are major decreases in emissions planned in the SSP5-Full simulation through time, regionally some areas of increase emerge. Surface aerosol conditions (Figure 6.) show some distinct difference from the column burden. For example, there is an increase in the column burden of anthropogenic aerosols in South America, yet there is a decrease in fine mode surface aerosol in the fixed emissions simulation (Figure 6 b.). While in other areas there is more agreement, such as a decrease in both the burden and in surface fine particulate matter over Central Africa. Over the middle latitudes there is an increase of surface $PM_{2.5}$, as seen over India, Asia, and the Mediterranean. I further examine these changes below by breaking down the individual species of aerosols, as well as their links to changes in precipitation and secondary production that influence these patterns.

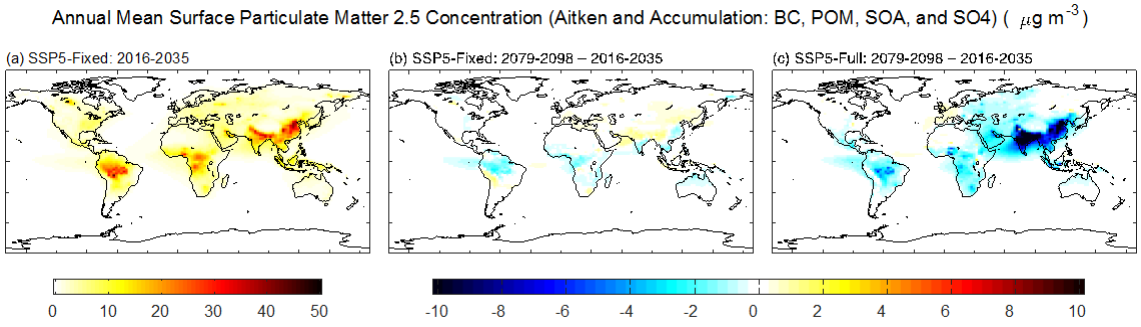

Figure 6. Annual mean $PM_{2.5}$ at the surface for (a) present-day conditions (2016-2035), and changes (2079-2098 minus 2016-2035) from (b) SSP5-Fixed and (c) SSP5-Full simulations. $PM_{2.5}$ includes all models for BC, POM, SOA, and SO_4 in units of [$\mu g/m_3$].

Fine aerosol at the surface in the model with fixed emissions has a varied change spatially through the century. However, averaging globally at the end of the century, there is a 1.3% increase in fine surface aerosol from the four anthropogenic species (Table 1.). In some areas we observe that the surface air quality improves in respect to $PM_{2.5}$, places like Brazil and coastal East Asia, but this is accompanied by areas of increase nearby, such that a dipole pattern emerges. For example, a decrease of $PM_{2.5}$ in the Amazon is accompanied by increases to the South. Similarly, the decrease of $PM_{2.5}$ observed along the coast of South and East Asia is accompanied by increases to the North. As the source of aerosol emissions remains constant, the dipole pattern that emerges will investigated through wet deposition change analysis and transport changes.

Table 1. Area-Weighted global averages for aerosol concentrations through the column of the atmosphere, at the surface, and associated lifetimes of aerosols in the fixed aerosol simulation.

Aerosol Area-Weighted Global Averages	PRESENT	FUTURE	CHANGE
Aerosol Burden [mg/m_2]			
Total (BC+POM+SOA+SO4)	6.109	6.657	0.548
BC Burden	0.277	0.288	0.011
POM Burden	1.554	1.634	0.080
SOA Burden	1.821	1.998	0.177
SO4 Burden	2.457	2.736	0.279
Surface Aerosol [$\mu g/m_3$]			
Total (BC+POM+SOA+SO4)	2.250	2.278	0.029
BC Surface	0.128	0.132	0.004
POM Surface	0.661	0.672	0.010
SOA Surface	0.804	0.757	-0.047
SO4 Surface	0.656	0.717	0.061
Global Average Lifetimes [*Days*]			
Total (BC+POM+SOA+SO4)	4.783	5.163	0.380
BC Lifetime	5.355	5.563	0.207
POM Lifetime	5.963	6.260	0.296
SOA Lifetime	4.208	4.613	0.405
SO4 Lifetime	4.617	5.037	0.420

When assessing global averages of aerosol in the fixed simulation, it becomes apparent that increases in particulate matter are clear in the changes for burden, surface concentration, and lifetime of aerosols. In understanding these aerosols, it also becomes

apparent that the species of aerosols must be addressed individually. Black carbon and primary organic matter have similar functions with sources from only primary emissions in the model, and are directly affected by precipitation frequency and wet deposition (investigated in part 3). Together, they increase in burden and fine surface aerosol by 5% and 2%, respectively. SOA has strong increases in atmospheric column burden (10%) but decreases in surface $PM_{2.5}$ (6%). This is related to secondary reactions of this aerosol in the model, rather than removal processes, and is thus not directly related to the focus of this analysis. The largest increase in both burden and surface concentration belongs to sulfate (11% and 9% respectively), which will be explored in part 4.

Due to the planned decreases in emissions of anthropogenic aerosols, an expected concentration of aerosol declines in the SSP5-Full simulation. However, as we continue on with emissions fixed at a present-day rate, an increase aerosol burden is observed over all continents in the SSP5-Fixed simulation. At the surface, air quality appears to worsen under these fixed emissions conditions, as $PM_{2.5}$ increases over some areas in South America, Central Africa, and South to East Asia.

Part 2: Precipitation Changes in Simulations with Increased Greenhouse Gases

As a global average, precipitation increases in the fixed aerosol simulation (SSP-Fixed), as well as the full aerosol emissions simulation (SSP5-Full), because these changes are driven by greenhouse gas changes, which are represented in both. However, the frequency of daily precipitation events may better explain the aerosol wet deposition

changes (a primary sink for aerosol particles) than the average, and the frequency tends to decrease in response to greenhouse gas increases (Table 2). In this section, I provide an overview of rainfall pattern change and explore its relationship with sinks such as wet deposition.

First, the present-day SSP5-Fixed conditions are validated against observations. In comparing the GPCP 1DD observations to model projections, the general pattern is captured by the model (Figure 7.). Annual precipitation rates are similar in scale, and the largest values are focused around the equator in both the SSP5-Fixed simulation and GPCP. A known issue related with model representation is the presence of a pronounced "double ITCZ", which is seen here. Similarly, when comparing the wet day frequency in the model to observations, there is a stronger chance of rain near the equator, but there are also areas of lesser precipitation frequency nearby in the subtropics and desert regions. The overall pattern of wet day frequency is well represented in CESM, but, like many ESMs, it has a tendency to rain too frequently, particularly over the ocean, which may impact the representation of aerosol removal to some degree.

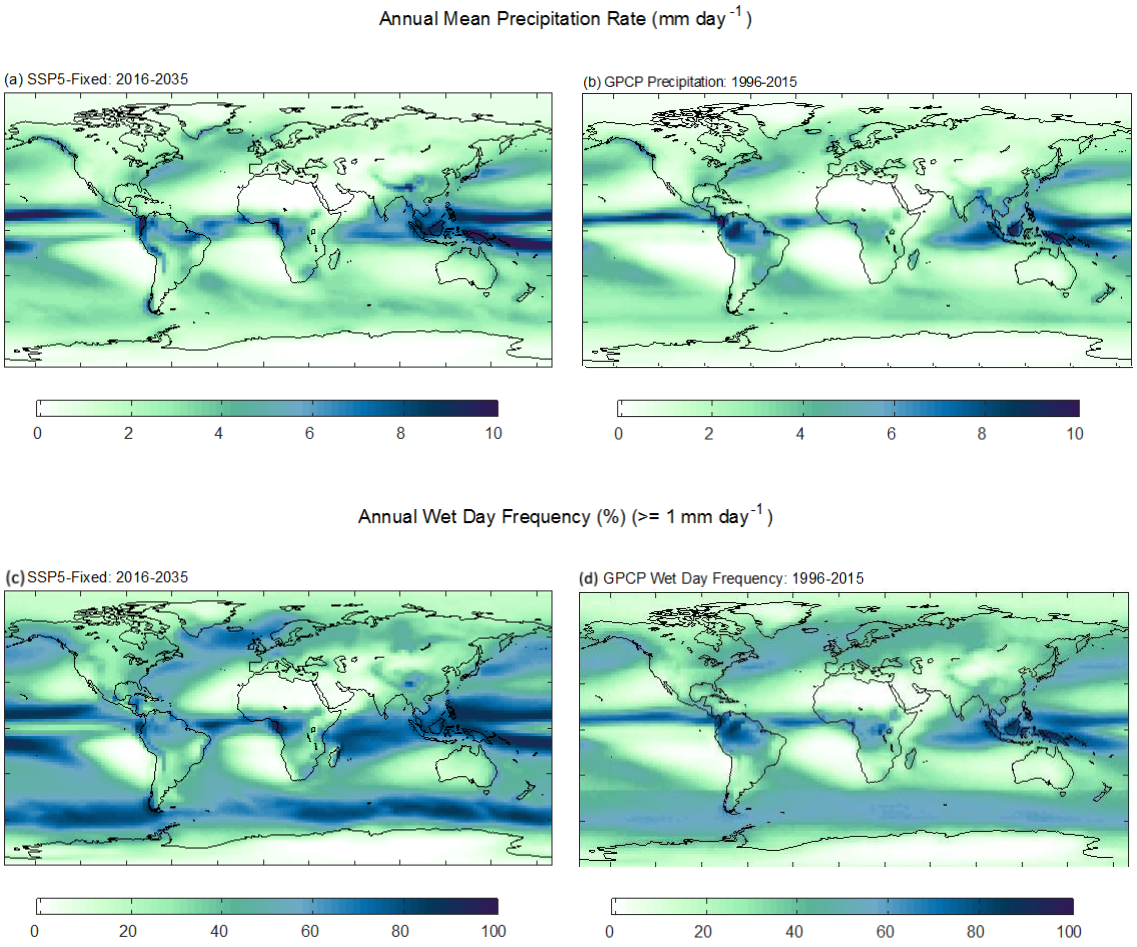

Figure 7. Annual mean precipitation rate for (a) SSP5-Fixed (2016-2035), and (b) GPCP observations (1996-2015). Precipitation rate is in units of [mm/day]. Annual wet day frequency for (c) SSP5-Fixed (2016-2035), and (d) GPCP observations (1996-2015). Wet days are defined as >= 1 mm/day and converted to units of [%].

As precipitation increases globally, one may expect to see more wet deposition, and a decrease in aerosol concentration and improvement of air quality. However, this is not necessarily the case, as the average precipitation does not assess regional patterns or how continuous/frequent the rainfall is over aerosol laden continents. The average precipitation change (Figure 8) shows that the global average increase is strongly concentrated over the equator, and does not represent the amount of rainfall over regions that are more relevant to air quality concerns. As a global average, precipitation increases by 4% (Table 2.), but has notable average decreases in some continental tropical regions. Regions that experience decreasing rainfall include a majority of South America (particularly centered around the Amazon), South Africa, Indonesia, and Australia. The majority of the 4% increase that we see globally is centered around the equatorial pacific, and not likely to contribute to bettering air quality in populated areas.

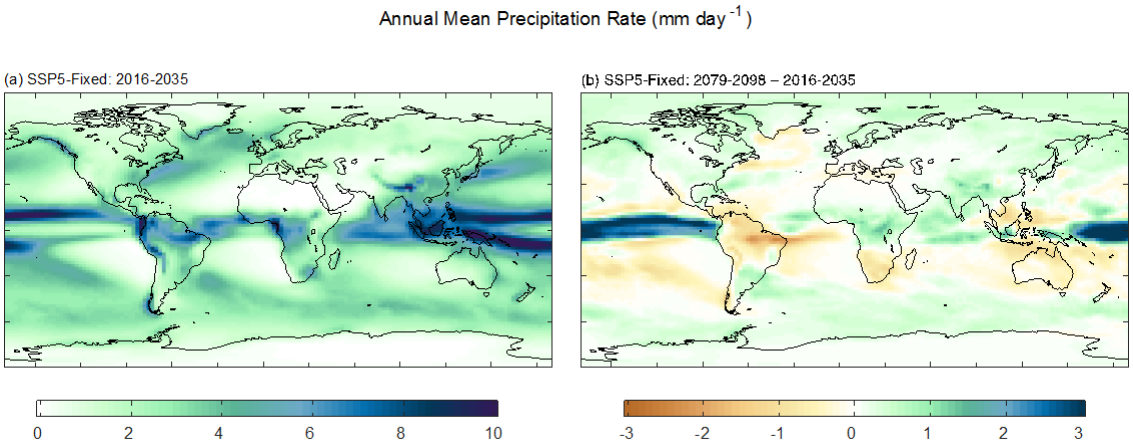

Figure 8. Annual mean precipitation rate for (a) present-day conditions (2016-2035), and (b) changes (2079-2098 minus 2016-2035) in the SSP5-Fixed simulation. Precipitation rate is in units of [mm/day].

The change in wet day frequency (Figure 9) shows a more complex pattern, with more widespread decreases. Wet days decrease in the global average by 5%, but have a strong regional element (Table 2.). For example, the annual average precipitation rate was relatively unchanged in Europe and the Mediterranean, but here we see a much stronger change in the decrease of rain events. Not only are the tropical areas mentioned above (South America, South Africa, and Indonesia/Australia) decreasing in wet day frequency, but parts of the United States and East Asia show that the average of precipitation may increase, but the number of rain days decreases. Overall, while the mean of precipitation is generally increasing globally, the decrease in frequency of precipitation allows for an aerosol to stagnate, and worsen air quality.

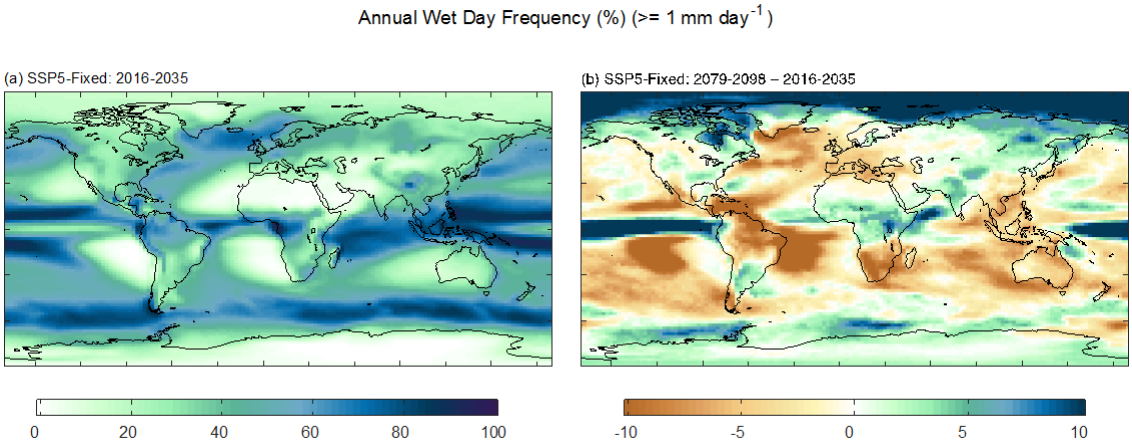

Figure 9. Annual wet day frequency for (a) present-day conditions (2016-2035), and (b) changes (2079-2098 minus 2016-2035) in the SSP5-Fixed simulation. Wet days are defined as >= 1 mm/day and converted to units of [%].

In SSP5-Fixed there are dramatic changes in the climatic conditions by the end of the 21st century, as demonstrated by the magnitude of annual average changes (Table 2), which result from increasing greenhouse gas emissions in the simulation. As greenhouse gasses increase, we see an associated increase in global mean temperature (+3°C), which results in an increase of precipitable water due to the increased evaporation and ability of the atmosphere to uptake moisture. Aside from hydrological changes, these global mean changes also may have varied effects on the transport of aerosol. This applies specifically to the planetary boundary layer (PBL) height changes seen in the simulation, as well as horizontal circulation (not shown). As PBL height lowers by 2.5%, the vertical transport of aerosols is changed. Aerosols meet a lower "cap" and are lofted into the upper

atmosphere less often. This might explain the increases in $PM_{2.5}$ increase at the surface in some regions such as South America, but decrease in the column burden of aerosols.

Table 2. Area-Weighted global averages for climatic variables in SSP5-Fixed. As greenhouse gases increase, global precipitation increases but the frequency of rain events decreases by 5%.

Climate Area-Weighted Global Averages	PRESENT	FUTURE	CHANGE
Specific Humidity [*kg/kg*]	0.011	0.013	0.002
Relative Humidity [%]	77.557	77.434	-0.123
Precipitable Water [kg/m^2]	26.194	32.302	6.108
Column Integrated Liquid [kg/m^2]	0.062	0.064	0.002
Temperature [*K*]	288.361	291.495	3.134
Precipitation [*mm/day*]	**2.964**	**3.089**	**0.126**
Surface Humidity [*kg/kg*]	0.010	0.012	0.002
PBL Height [*m*]	739.298	720.874	-18.424
Wet Day Frequency [%]	**74.687**	**69.763**	**-4.924**

Over land, the wet day frequency changes are mirrored in the annual mean wet deposition flux of the four anthropogenic aerosol species (Figure 10). Wet deposition rates drop dramatically in the Northern part of South America, the Southern tip of Africa, the Mediterranean, Indonesia and Australia, while they increase in other regions, such as over Central Africa, the Southeastern US, and India. Generally, however, where there is a decrease in wet day frequency, we also observe a decrease in wet deposition flux. The pattern seen over the tropics has a strong correlation with the aerosol changes seen there,

and is analyzed in more detail with discussion of primarily emitted aerosols (black carbon and primary organic matter) in Part 3. Decreases in wet deposition are observed in our model where $PM_{2.5}$ (particularly sulfate) is increasing at the surface. Over India however, where wet day frequency increases along with mean wet deposition, the aerosol changes are more complicated. This is likely due to large changes in secondary sulfate production (again, in the simulation with fixed emissions) that also contributes to future changes in the aerosol concentration, which is explored in Part 4.

As greenhouse gases increase in the SSP5-Fixed simulation, annual average precipitation increases 4%. However, the frequency of rainy days decreases by 5% globally, leading to a decrease in wet deposition and aerosol removal.

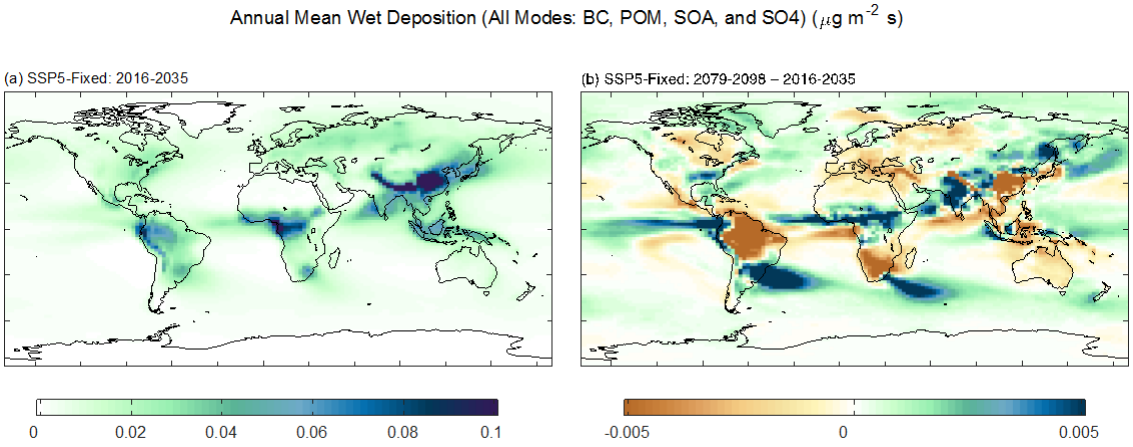

Figure 10. Annual mean wet deposition rate for (a) present-day conditions (2016-2035), and (b) changes (2079-2098 minus 2016-2035) in the SSP5-Fixed simulation for the four species of anthropogenic aerosols in units of [$\mu g/m_2/s$].

Part 3: Primary Emitting Aerosols; Black Carbon and Primary Organic Matter

BC and POM are from the same source of primary production, combustion, and can be assessed similarly in the model. Without changes in aerosol emissions (SSP5-Fixed), a pattern emerges around the tropics. Surface concentration of primary fine particulate matter increases broadly in the Northern Hemisphere, but decreases in the tropics in very specific areas. Over Central Africa, a decrease occurs over the equator with increases to the North and South (Figure 11). This matches very clearly with the deposition rates seen in Figure 12, with a strong increase in wet deposition in Central Africa and strong decrease in $PM_{2.5}$.

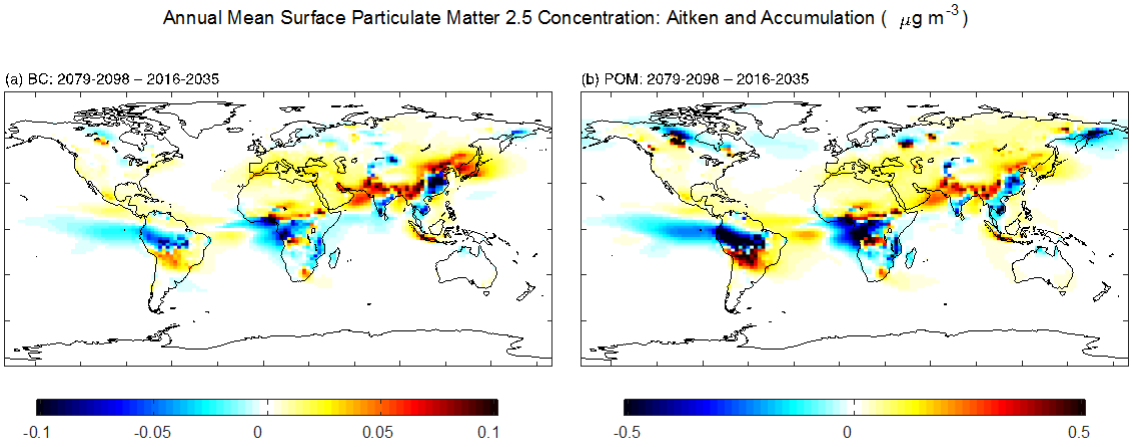

Figure 11. Surface $PM_{2.5}$ concentration of (a) BC and (b) POM for changes (2079-2098 minus 2016-2035) in the SSP5-Fixed simulation in units of [μg/m3].

However, the expected results of increased wet deposition and wet day frequency do not always correlate with decreases in $PM_{2.5}$. Here we see another dipole pattern emerge, where a decrease in aerosol has an associated increase nearby. Since our system is characterized by sources, sinks, and transport, we may rule out emission sources (which are fixed) and only focus on sinks and transport to explain this effect. Using South America as an example, we see a decrease in surface aerosol to the North and an increase in the South. There is also a strong dipole pattern reflected in wet deposition, with strong decreases in the North and increases to the South. We can hypothesize, therefore, that the increase in deposition is due to a horizontal aerosol transport to the South, causing a decrease in both concentration and deposition to the North. A similar pattern is observed in India and much of Asia, where aerosol increases are to the West and decreases in both aerosol and deposition are to the East. India is of particular interest here, where wet deposition increases, but is paired with a decrease of deposition to the North (the Himalayas) and to the East. This dipole effect may also be observed around South Africa, where wet deposition is decreasing on the continent and increasing over the ocean to the South. Under this new hypothesis, the efficiency of removal will be reflected in these areas where aerosols are moved from areas of high concentration to areas of high wet day frequency and wet deposition.

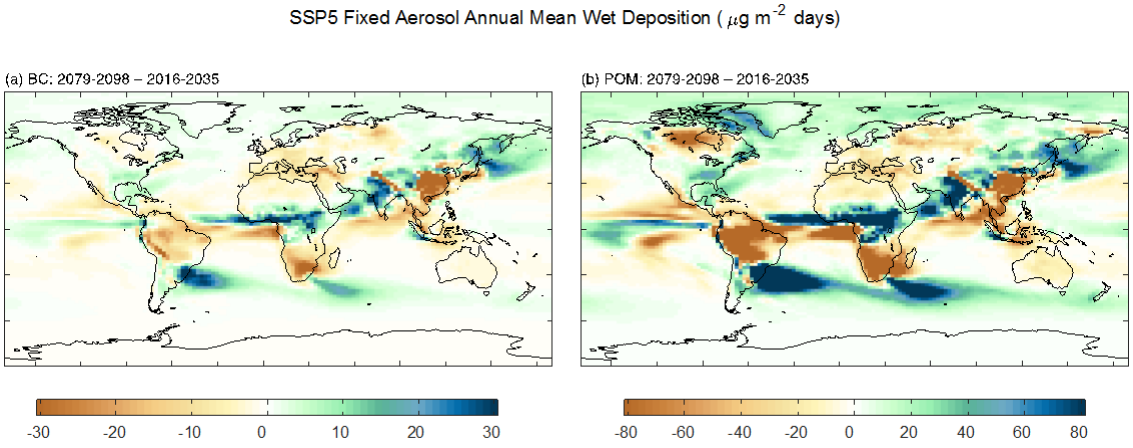

Figure 12. Annual Average of wet deposition rates for (a) BC and (b) POM, for changes (2079-2098 minus 2016-2035) in the SSP5-Fixed simulation in units of [μg/m2/days].

A new metric for analysis, wet deposition efficiency, is used to understand the rate of deposition relative to the burden of aerosol that is available to be removed (Figure 13.). The efficiency is calculated as the rate of deposition relative to the concentration of aerosol available to better understand the precipitation effect relative to the potential of aerosol removal. Again, we observe black carbon and primary organic matter in the atmosphere is very similar and both are less efficiently removed from the atmosphere over continents. In South America, efficiency is somewhat less, suggesting that the decrease in aerosol and deposition rates are generally balanced (there is less burden, and therefore lower efficiency in that region). In this dipole region, concentration decreases in the North due to the removal of aerosol via transport to the South where not only does wet deposition and wet day frequency increase, but the concentration of air pollutants

outweighs these sinks. Wet deposition efficiency in the model shows some increases in other regions of the deep tropics, minimizing poor air quality around India and Central Africa, but otherwise decreasing in the subtropics and worsening air quality.

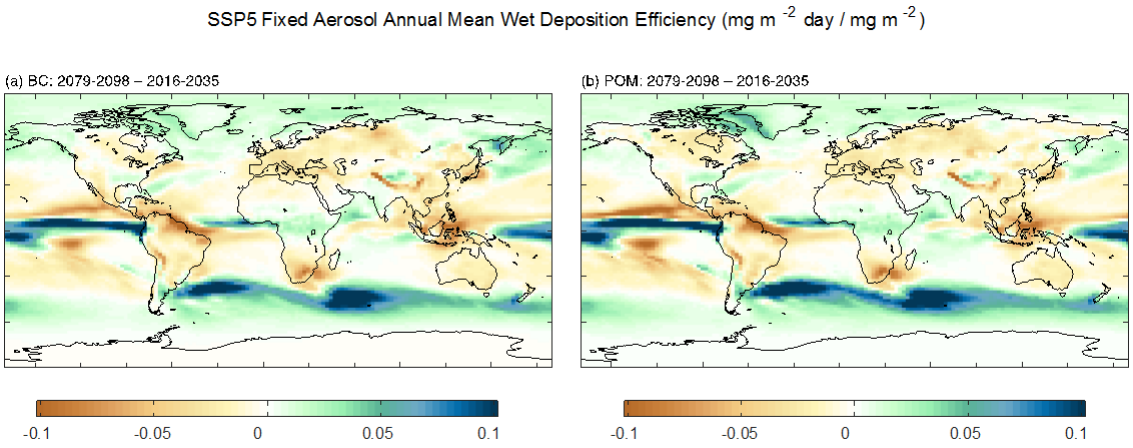

Figure 13. Annual average of wet deposition efficiency, or wet deposition as a rate that is balanced with the aerosol burden available in the atmosphere, for (a) BC and (b) POM, for changes (2079-2098 minus 2016-2035) in the SSP5-Fixed simulation.

As a global average, lifetimes of these aerosols are increasing over the 21st century. The annual average lifetime for the four species of anthropogenic aerosols increases by 8% (Table 1.). The primary emitting aerosols show a smaller change in lifetime (BC at 4% and POM at 5%), suggesting that SOA and SO4 have a more pronounced transport change in the model (10% and 9% respectively).

For the primary species of aerosol, transport and lifetime changes are a major factor in shifting wet deposition spatial patterns. The result is a dipole effect, where weak deposition is followed by a stronger rate of deposition nearby.

Table 3. Area-Weighted global averages of emissions and removal variables for BC and POM in the SSP5-Fixed simulation.

Area-Weighted Global Averages [$\mu g/m^2/day$]	PRESENT	FUTURE	CHANGE
BC Emissions	62.191	62.191	0
BC Wet Deposition	35.735	35.641	-0.140
BC Dry Deposition	15.895	16.130	0.235
POM Emissions	260.161	260.161	0
POM Wet Deposition	179.356	179.760	0.405
POM Dry Deposition	81.214	81.263	0.049
Total Removal (Deposition) BC	51.676	51.771	0.096
Total Removal (Deposition) POM	260.570	261.023	0.453

Part 4: Focusing on Sulfate Production and Removal

Assessing the impacts of climate change on sulfate is key for understanding future air quality at the surface level, with the largest increase in concentration over the course of the fixed aerosol simulation (0.61 $\mu g/m^3$ and 9% globally) (Table 4.). However, the increases in sulfate are not completely characterized by decreases in sinks like wet deposition, and are also affected by secondary production in the model (aqueous chemistry and gas to aerosol exchange factors). The changing hydrology of the climate system goes beyond the rainfall changes discussed above, and includes changing

humidity and available moisture in simulations with increasing greenhouse gases. This prompted a new question about changing air quality from these simulations. Is the production of secondary aerosol such as sulfate enhanced in a warmer climate? In Part 4, I first examine the concentration of SO_4 and the changes in the production variables associated with sulfate before continuing analysis of the removal changes in sulfate as a result of rainfall frequency.

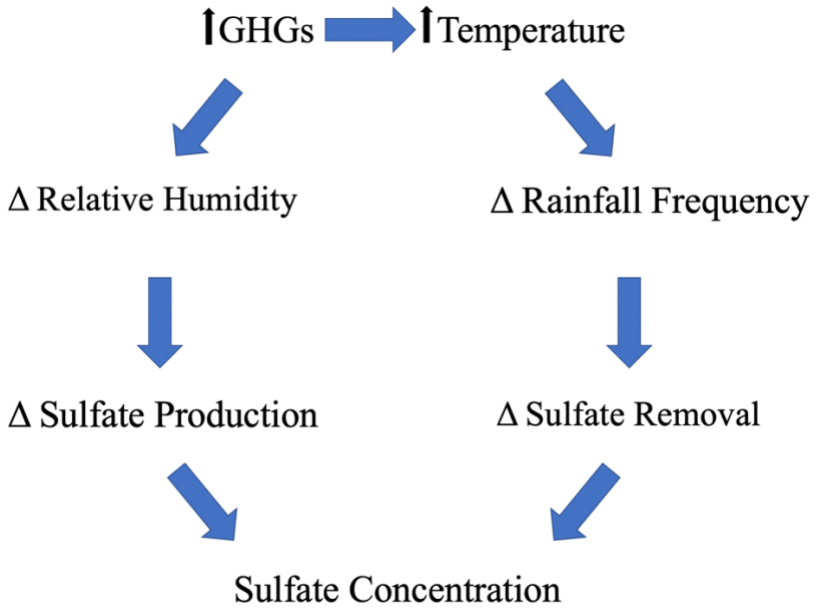

Figure 14. Schematic diagram illustrating the competing effects on SO_4 concentration in the model. As CO_2 and temperature increases globally, the changes in humidity and rainfall frequency lead to competing influences on sulfate production and removal.

As a starting point, the schematic above summarizes the competing processes that can influence sulfate in the atmosphere. Though an increase of sulfate concentration in the simulation is seen in most regions (Figure 15.), the balance of sources and sinks is complex, despite the direct emissions of SO_4, SO_2, and DMS (dimethyl sulfide) remaining fixed at 2010 rates. In SSP5 simulations, increasing greenhouse gases and temperature lead to two competing effects. Those effects are the sulfate removal change from rainfall changes at a given place, and the changes in temperature, cloud water droplets, and surface humidity that contribute to sulfate production. Both contribute to a sulfate concentration change (generally increased, depending on the region) over the course of the 100-year simulation. These factors combine together to explain increases in surface fine sulfate and are separately examined below.

A. SO_4 Concentration

Continuing with the aerosol emissions at the present-day rate, increases in sulfate are observed, particularly affecting surface concentration in populated areas. Sulfate increases are observed across the Northern hemisphere, and particularly over the Southern Asian Continent (with exceptions off the coast of Ecuador and the Arabian Sea) (Figure 15 b.). At the surface level, fine sulfate increases on continents similarly to burden concentration with the exception of East Asia. Global averages in this case are at first glance misleading, as the balance of sulfate generally increases across the globe even with increases in wet deposition which are balanced out by production of sulfate, particularly Gas/Aerosol exchange (Table 4.).

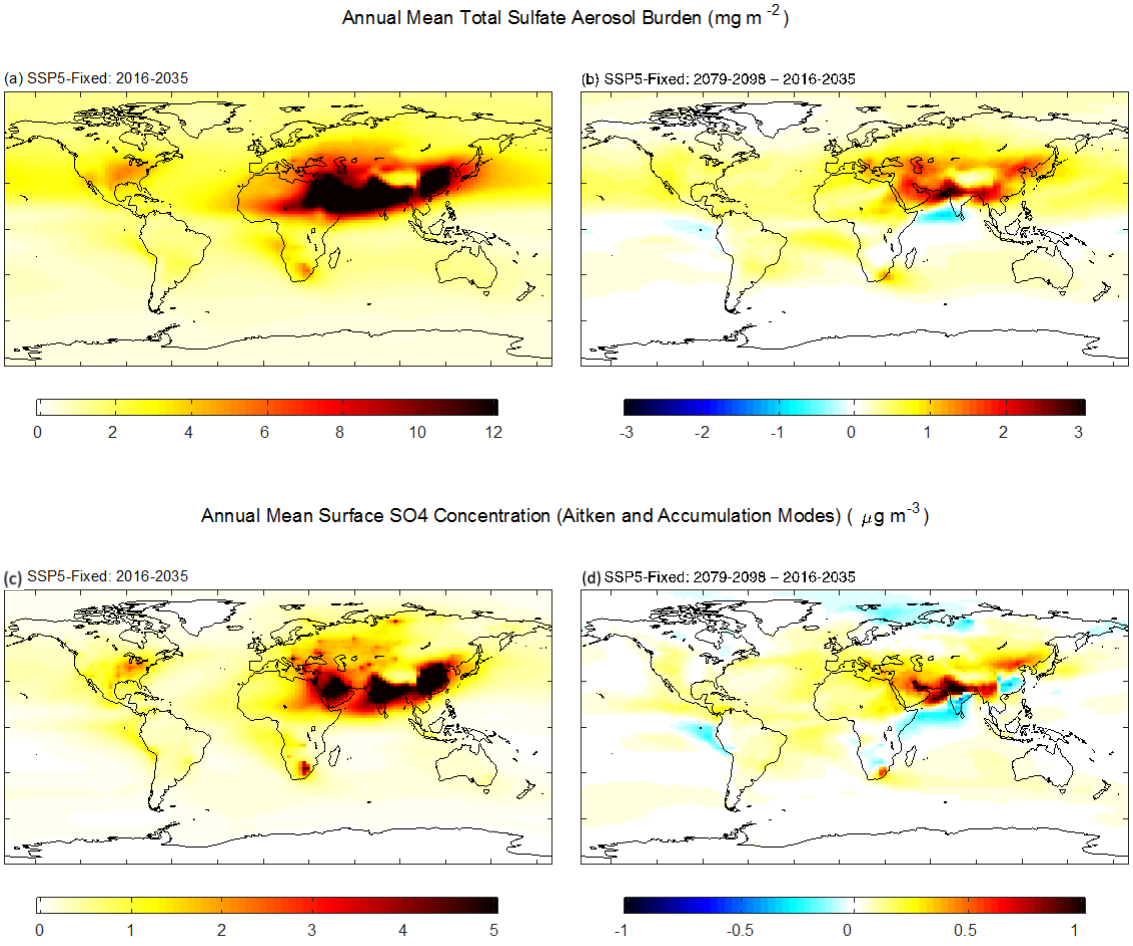

Figure 15. Annual mean of sulfate burden in (a) present-day conditions (2016-2035), and (b) changes (2079-2098 minus 2016-2035) for the SSP5-Fixed simulation. Surface $PM_{2.5}$ concentration of sulfate in (c) present-day conditions (2016-2035), and (d) changes (2079-2098 minus 2016-2035) in units of [μg/m3].

Table 4. Area-weighted global averages for sulfate concentration, deposition, production, and emissions in the SSP5-Fixed simulation.

Area-Weighted Global Averages	PRESENT	FUTURE	CHANGE
SO4 Burden [*mg/m2*]	2.457	2.736	0.279
SO4 Surface Concentration [*μg/m3*]	0.656	0.717	0.061
Wet Deposition [*μg/m^2/day*]	450.024	455.969	5.945
Dry Deposition [*μg/m^2/day*]	82.174	87.260	5.085
Aqueous [*μg/m2/day*]	254.080	246.304	-7.776
Gas [*μg/m2/day*]	263.263	282.377	19.114
Total Secondary [*μg/m2/day*]	517.343	528.681	11.338
Total Deposition [*μg/m^2/day*]	532.198	543.229	11.031
Emissions [*μg/m2/day*]	23.391	23.391	0

B. SO_4 Production

Total sulfate secondary production in the atmosphere results from two types of processes, aqueous chemistry and gas to aerosol exchange. In this simulation, I find that the change in gas to aerosol exchange is the main source of increases in secondary production (with Yemen / Oman and India and as notable exceptions wherein production through aqueous chemistry is increasing, Figure 16). A strong inverse relationship is shown between the changes in these production modes. Particularly in the tropics, where there is a decrease in aqueous chemistry, there is an increase in gas to aerosol exchange. This inverse relationship between production modes is highly dependent on available moisture for aqueous production. Decreases in the availability of moisture over land in the tropics, particularly cloud liquid water, are associated with decreases in aqueous

chemistry, and opposite increases in gas to aerosol exchange production. For example, South America loses available moisture for sulfate production in the form of humidity at the surface (Figure 17.). This results in a decrease in aqueous chemistry SO_4 production, but an increase in the gas to aerosol exchange of sulfate. The result is an increase in surface concentration of sulfate. We see a similar effect over China, where the loss of surface humidity decreases aqueous chemistry and allows gas-to-aerosol exchange to become the primary mode of production. As surface level sulfate $PM_{2.5}$ increases globally, so does total production, primarily driven by gas to aerosol exchange.

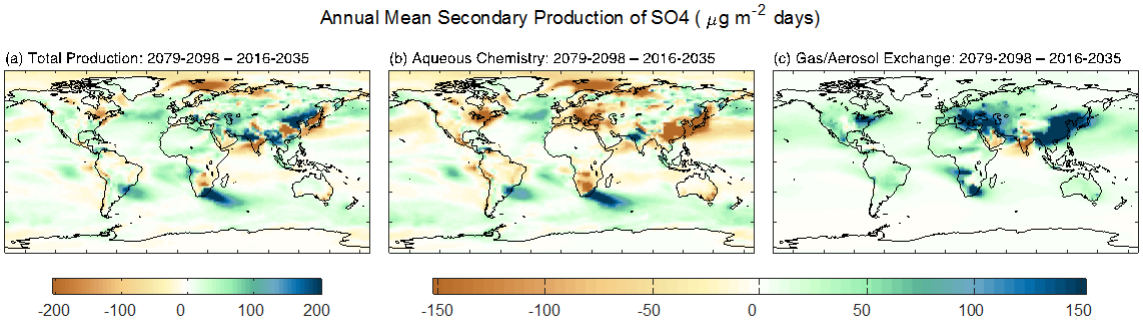

Figure 16. Annual mean secondary production of sulfate changes (2079-2098 minus 2016-2035) for (a) total production, (b) aqueous chemistry, and (c) gas/aerosol exchange in units of changes in units of [μg/m2/days].

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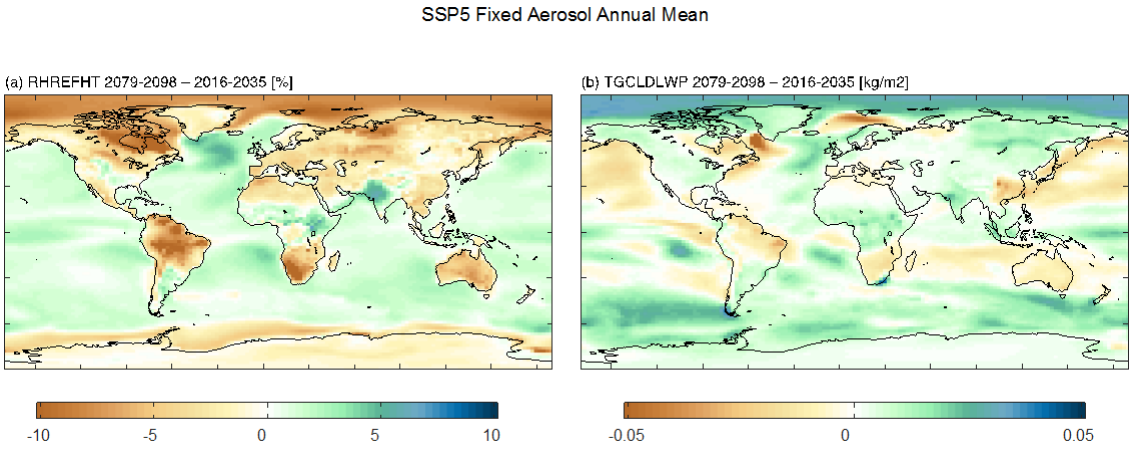

Figure 17. Annual mean (a) relative humidity at the surface [%], and (b) total grid box cloud liquid water path [kg/m2].

There are some notable exceptions to where the gas to aerosol exchange increase is counterbalanced by a reduction in aqueous production. For example, the annual mean surface SO_4 concentration increases in northern India without increases in emissions. Northern India shows increases in both relative humidity and available cloud liquid water. This allows for a stronger aqueous production in the model, and a higher total production and concentration. However, to the South we see the reverse effect, where the humidity hot-spot ends, aqueous chemistry is not dominant. Overall, the total secondary production signature matches well with annual surface concentration of SO_4. This is also observed in Yemen / Oman, where there is an increase in SO_4 concentration to the North, and less so in the South. This is reflected in a similar pattern of relative humidity increase, gas to aerosol decrease, and overall less production.

C. SO_4 Removal

The primary removal mechanism of sulfate is wet deposition, which, as discussed above, is influenced by climate changes in the fixed aerosol simulation. Sulfate deposition increases over areas of concentration increase, particularly around South Asia (Figure 18 b.). However, we find that the increases in wet deposition are typically found over the ocean, and decreases are not the only factor influencing concentration. Nor are increases in wet removal a signal for better air quality. A stronger production of sulfate may outweigh sinks and increase concentration, even while the rate of deposition of decreases due to the availability of aerosols. For this reason, a wet deposition efficiency metric was calculated, based on the inciting study conducted by *Xu and Lamarque* [2018], to understand the rate of deposition and removal of sulfate in respect to the total burden in the atmosphere. This new metric of deposition efficiency correlates with changes in aerosol lifetime, with sulfate showing a 9% increase in global mean lifetime change (Table 1.), and shows near total decreases over continents (Figure 18 c.).

Sulfate behaves differently from the primary emitting aerosols in the model. The methods of production are varied, but it appears to be gas phase production which dominates the increase in sulfate for most regions. This is inversely related to aqueous phase production, which acts as a function of decreased availability of moisture in the model over continents.

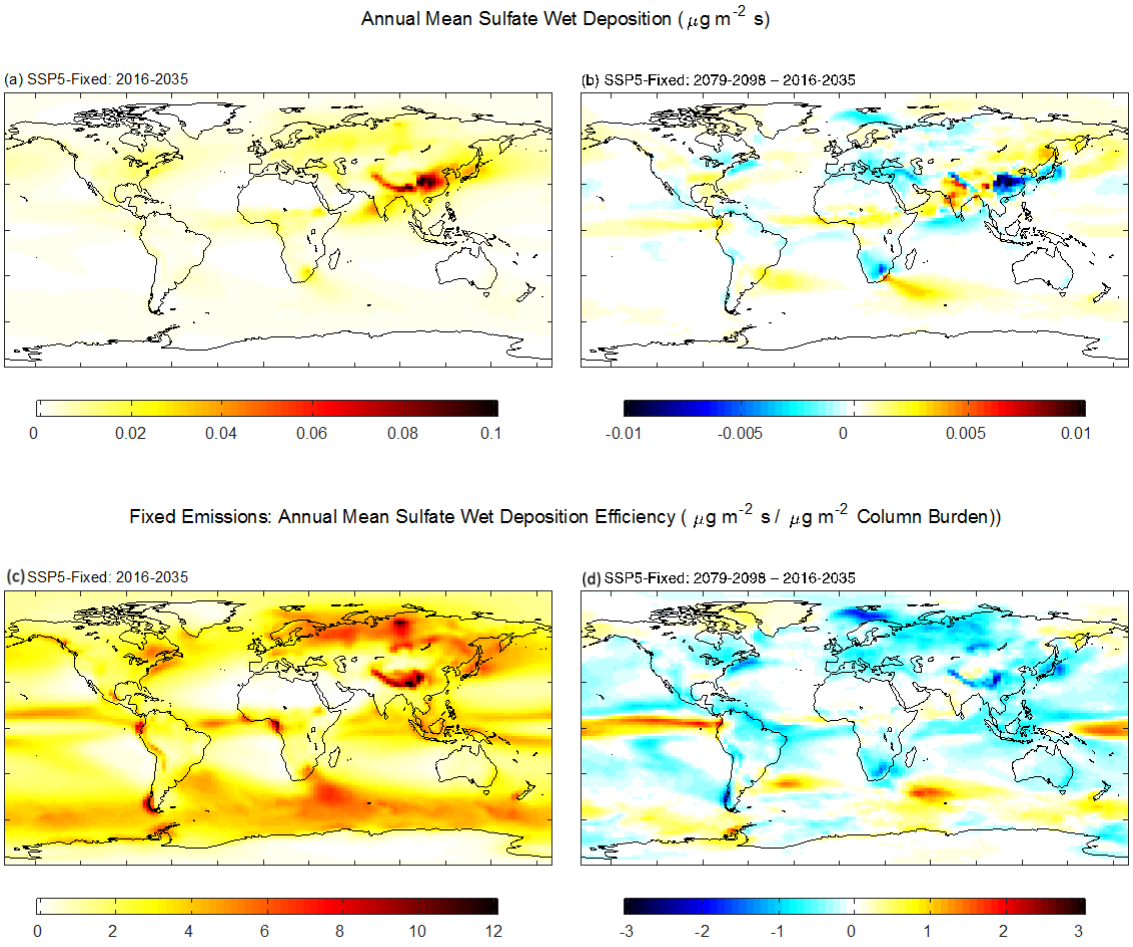

Figure 18. Annual mean of sulfate wet deposition in (a) present-day conditions (2016-2035), and (b) changes (2079-2098 minus 2016-2035) for the SSP5-Fixed simulation in units of [μg/m2s]. Annual mean of sulfate wet deposition efficiency for (c) present-day conditions (2016-2035), and (d) changes (2079-2098 minus 2016-2035) in units of [s].

Part 5: Analysis of Daily Extreme Air Quality Events

In addition to changes in mean aerosol loading, the 99.9th percentile of $PM_{2.5}$ concentration from daily output shows a clear increase of the infrequent but extreme air quality events in the SSP5-Fixed simulation (Figure 19.). Since all forms of $PM_{2.5}$ are hazardous to health, this analysis of air quality events includes sea salt and dust. The Amazon, Central Africa, and East Asia all have hotspots of extreme air quality changes, but there also emerge stronger concentrations of $PM_{2.5}$ in the midlatitudes. While there are increases everywhere, the focus of the 99.9th percentile increases are strongest over continents.

The results from the SSP5-Fixed simulation show 99.9th percentile air quality events (the most extreme values for $PM_{2.5}$ concentration) are consistent with regions of concern addressed in Figure 5. The addition of dust adds a higher concentration in the model, particularly in Central Africa. However, areas of worsening air quality are consistent with the changes of anthropogenic surface PM2.5 in the model (The Amazon, Central Africa, and East Asia).

The World Health Organization sets the guidelines for hazardous air quality sets a cap for $PM_{2.5}$ at 25 $\mu g/m^3$ for a 24-hour period (*WHO*, 2016). It should be noted that zero exposure is ideal, and any daily intake of particulate matter may have adverse health effects. While the 99.9th percentile changes discussed above provide an assessment of how locally defined extreme values change, it is useful to also evaluate changes in the exceedances of a fixed extreme value (i.e., extreme value is defined to be the same

everywhere). In the SSP5-Fixed simulation, the percentage of these hazardous air quality days increases by up to 20% in some regions through meteorological effects alone.

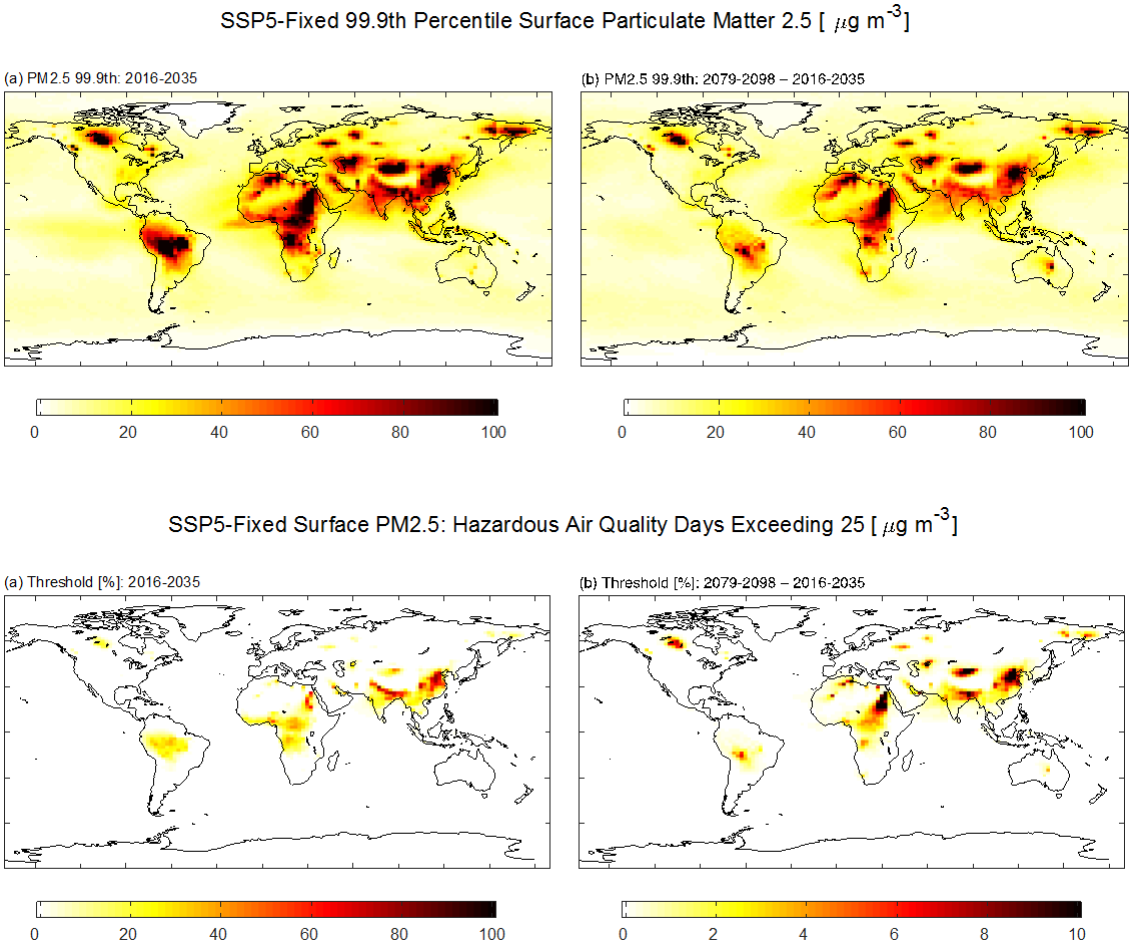

Figure 19. (Above) 99.9th Percentile of surface $PM_{2.5}$ changes (2079-2098 minus 2016-2035) for the SSP5-Fixed simulation in units of [μg/m3]. (Below) Hazardous air quality day frequency. Surface $PM_{2.5}$ changes (2079-2098 minus 2016-2035) for the SSP5-Fixed simulation which have exceeded a daily 25 μg/m3 threshold in units of [%].

Hazardous air quality days are consistent with the regional extremes of $PM_{2.5}$ in the 99.9^{th} percentile analysis. However, the percentage of poor air quality days are increasing in the model less uniformly (not showing major increases over oceans or large portions of continents). Instead, hazardous air quality days are focused on particular points and intensifying in the model through the 21^{st} century (countries like Bolivia, Egypt, and Bangladesh or cities like Beijing).

CHAPTER 5

DISCUSSION

In comparing the two key simulations of this study, I find that concentration of aerosol declines in the SSP5-Full simulation, which is a projection of the future climate under planned air pollution control measures. When aerosol emissions are cycled at present-day rates in the SSP5-Fixed simulation, an increasing burden of aerosol is observed over all continents, with PM2.5 at the surface in parts of South America, Central Africa, and South to East Asia. Due to increasing greenhouse gases in these simulations, global average precipitation increases by 4%. However, wet day frequency of rainfall decreases by 5%. This finding is supported by previous studies of precipitation in ESMs, wherein the frequency of light-to-moderate rainfall events are reduced at a global scale (*O'Gorman*, 2009; *Pendergrass and Hartmann*, 2014; *Sun*, 2007). Findings in this study are also supported by previous research suggesting that there is a decrease in wet removal flux over the course of the 21st century in simulations with increasing greenhouse gases, leading to an increased global surface $PM_{2.5}$ concentration (*Xu and Lamarque*, 2018). "Hazardous Air Quality Days" (i.e. days in which air quality fails to reach EPA daily standards of $PM_{2.5}$, or 25 $\mu g/m^3$) increase in SSP5-Fixed through meteorological effects alone (with climate change), worsening health issues without changing aerosol emissions. Regionally, this increase of hazardous air quality days are a

result of spatial patterns developed though precipitation changes and aerosol interactions that vary by species. For primary species (BC and POM), transport becomes a major factor in wet deposition spatial patterns, setting up a dipole effect of weak wet deposition followed by strong wet deposition nearby. For SO_4, aqueous and gas phase production are inversely related, and decreased availability of moisture in the model encourages gas phase production to dominate most regions. The increase of SO_4 production is observed in the simulation spatially from Egypt to the East China Sea, a region that is also subject to the highest increases in hazardous air quality days.

CHAPTER 6

CONCLUSIONS

In this study, I investigated the influence of climate change on future air quality by comparing the standard SSP5-Full simulation, with aerosol emissions decreasing over the 21st century, to a simulation with a fixed aerosol signature cycled on 2010 rates (i.e., SSP-Fixed). The largest changes between these two future projections are a result of policy change, but some important changes result from greenhouse gas driven changes in the climate system. When aerosol emissions are fixed at 2010 rates, air quality is found to worsen in South America and Central Africa (primarily due to increases in black carbon and primary organic matter), along with higher concentrations of $PM_{2.5}$ in South to East Asia (primarily sulfate). Greenhouse gas related precipitation changes have a strong influence on primary emitting species (i.e., BC and POM) through changes in the frequency of rainfall, while the availability of moisture for production becomes more important to the concentration of sulfate in the model (which is enhanced in simulation). When it comes to primary aerosol species (BC and POM), transport becomes a major factor in wet deposition spatial patterns, setting up a dipole effect of weak wet deposition followed by strong wet deposition nearby. For SO_4, aqueous and gas phase production are inversely related, and decreased availability of moisture in the model encourages gas phase production to dominate most regions.

Here I presented an analysis focused of climatological timescales using simulations from an ESM that has conventional parameterizations of convection. The results of this study motivate future work in several areas, including analysis of air quality changes on shorter timescales (e.g., seasonal) and applications of models that improve the representation of atmospheric processes (e.g., rainfall characteristics). For example, a seasonal analysis for India, a standout region from this study, may further our understanding of the regional changes identified there. India has shown a slowing monsoon season as a result of aerosol interactions in simulations with increased greenhouse gases (*Wang et. al.*, 2015). The seasonality of the monsoon cycle may be more nuanced than the annual wet day frequency analysis would suggest, leading to varied effects on air quality throughout the year. Additionally, CESM has the capability to be run with "resolved convection" using a method called super-parameterization (SP). In SP-CESM (*Randall et al.*, 2003), embedded cloud-resolving models represent sub-grid convection at 3-kilometer resolution explicitly, as opposed to CAM6 approximated convection (*Hurrell et. al*., 2013). Cloud super-parameterization may provide a useful tool to further investigate the decreases in rainfall frequency that contribute to an increase in aerosol concentrations despite an overall average increase in rainfall globally.

The results of this study suggest that without changes in our present-day emissions, we can expect to see an increase in surface level fine aerosols, which are hazardous to vulnerable populations in many regions. According to the World Health Organization, the consequences that fall on low- and middle-income countries account

for 91% of fine particulate matter deaths due to outdoor aerosol pollution (*WHO*, 2016), and many of these countries show changes in rainfall frequency that poorly impact their future air quality. This study provides insight into these impacts of climate change and the air quality of those who are most at risk, which I hope will motivate future work in the area and inform regulatory decisions.

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