Climate Implications of the Heterogeneity of Anthropogenic Aerosol Forcing

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Abstract

Short-lived anthropogenic aerosols are concentrated in regions of high human activity, where they interact with radiation and clouds, causing horizontally heterogeneous radiative forcing between polluted and unpolluted regions. Aerosols can absorb shortwave energy in the atmosphere, but deplete it at the surface, producing opposite radiative perturbations between the surface and atmosphere. This thesis investigates climate and policy implications of this horizontal and vertical heterogeneity of anthropogenic aerosol forcing, employing the Geophysical Fluid Dynamics Laboratory’s AM2.1 and AM3 models, both at a global scale and using East Asia as a regional case study.

The degree of difference between spatial patterns of climate change due to heterogeneous aerosol forcing versus homogeneous greenhouse gas forcing deeply impacts the detection, attribution, and prediction of regional climate change. This dissertation addresses a gap in current understanding of these two forcings’ response pattern development, using AM2.1 historical forcing simulations. The results indicate that fast atmospheric and land-surface processes alone substantially homogenize the global pattern of surface energy flux response to heterogeneous aerosol forcing.

Aerosols’ vertical redistribution of energy significantly impacts regional climate, but is incompletely understood. It is newly identified here, via observations and historical and idealized forcing simulations, that increased aerosol-driven atmospheric absorption may explain half of East Asia’s recent surface insolation decline. Further, aerosols’ surface and atmospheric effects counteract each other regionally—atmospheric heating enhances summer monsoon circulation, while surface dimming suppresses it—but absorbing aerosols’ combined effects reduce summer monsoon rainfall. This thesis constitutes the first vertical decomposition of aerosols’ impacts in this high-emissions region and elucidates the monsoonal response to aerosols’ surface versus atmospheric forcing.

Future aerosol emissions patterns will affect the distribution of regional climate impacts. This dissertation interrogates how international trade affects existing assumptions about
East Asia’s future black carbon aerosol emissions, using integrated assessment modeling, emissions and economic data, and AM3 simulations. Exports emerge as a uniquely large and potentially growing source of Chinese black carbon emissions that could impede projected regional emissions reductions, with substantial climate and health consequences. The findings encourage greater emissions projection sophistication and illustrate how societal decisions may influence future aerosol forcing heterogeneity.
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To my parents and my brother,
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Chapter 1

Introduction

1.1 Background and Motivation

Human emissions of greenhouse gases and aerosol particles are the two most potent anthropogenic forcers of the climate system. Greenhouse gases’ ability to trap longwave radiation within the surface-atmosphere system has resulted in a net positive input to the earth’s radiative balance of $2.8\pm0.6 \text{ W m}^{-2}$ since 1750 [Myhre et al., 2013]. Anthropogenic aerosol particles, on the other hand, are thought to have offset this positive radiative perturbation—by a net negative perturbation of $-0.45\pm0.5 \text{ W m}^{-2}$ since 1750 due to their absorption and scattering of radiation and by an additional $-0.9\pm1.0 \text{ W m}^{-2}$ due to their microphysical interactions with clouds—though these perturbations are composed of a multitude of canceling effects, each of which carries substantial uncertainty [Boucher et al., 2013]. The combined and competing effects of anthropogenic greenhouse gases and aerosols have profound impacts on temperature, precipitation, and other parameters to which human and natural systems are deeply sensitive [IPCC, 2013].

Anthropogenic aerosols, however, have many differences from greenhouse gases beyond the sign of their net radiative perturbation. Although both categories of atmospheric pollutants are often generated by similar industrial, automotive, and residential combustion
processes, anthropogenic aerosols are distinct in the way in which they perturb the Earth’s radiative balance and the timescales and spatial patterns over which they do so. These characteristics can produce complex, intense, and poorly constrained radiative and hydrological effects at the regional scale \[\text{Boucher et al., 2013}\] and make them of particular interest to study both in relation to greenhouse gases and on their own.

Greenhouse gases, such as carbon dioxide, methane, and nitrous oxide, are generally quite long-lived in the atmosphere (i.e. on the order of centuries for carbon dioxide and nitrous oxide and decades for methane) \[\text{Myhre et al., 2013}\]. They are thus relatively uniformly distributed across the globe. They affect the Earth’s energy balance primarily by absorbing longwave radiation emitted by the Earth’s surface and reradiating it into the surface-atmosphere system \[\text{e.g. Arrhenius, 1896; Callendar, 1938; Wang et al., 1976; Hansen et al., 1981}\]. Consequently, they result in warming throughout the troposphere and across the globe, though heterogeneities do exist in the degree to which different regions warm \[\text{e.g. Stouffer et al., 1989; Murphy and Mitchell, 1995}\].

Aerosols, however, come in a range of shapes, sizes, and radiative properties. These solid and liquid airborne particles, ranging in diameter from the size of a virus to the width of a human hair, have both natural and manmade sources. Aerosols can be subcategorized into several key groups: sulfates, nitrates, organic carbon, black carbon, dust and sea salt \[\text{e.g. Boucher et al., 2013}\]. Dust and sea salt aerosols are primarily naturally occurring, via the wind-driven liberation of dust particles from arid surfaces or sea salt particles from ocean waves, respectively \[\text{e.g. Ginoux et al., 2012; O’Dowd et al., 1997}\]. The gaseous precursors of liquid sulfate aerosols can also have natural sources: volcanoes outgas sulfur dioxide that oxidizes into the liquid sulfate aerosol; and ocean microorganisms emit the dimethylsulfide sulfate precursor \[\text{e.g. Graf et al., 1997; Kettle and Andreae, 2000}\]. Forest fires and other natural combustion processes, meanwhile, emit organic carbon and black carbon, as well as sulfate \[\text{e.g. Patterson and McMahon, 1984; Crutzen and Andreae, 1990}\]. Although these naturally occurring aerosols make up the majority of global aerosol concentrations, they
will not be the focus of this dissertation, as they are not generally considered to be an anthropogenic forcing of the climate system.

Approximately 10% by mass of the global aerosol burden, and the majority of black carbon and sulfate aerosols, presently comes from manmade sources [Boucher et al., 2013]. Fossil fuel combustion strongly emits the sulfur dioxide precursor to sulfate aerosol [e.g. Smith et al. 2011a]. Smoke from the burning of biomass as agricultural waste or for land management purposes produces copious organic carbon and black carbon [e.g. Hao and Liu, 1994]. Combustion processes across the transportation, power generation, industrial, and residential sectors are implicated in the generation of sulfate, nitrate, organic carbon, and black carbon aerosols [e.g. Boucher et al., 2013]. Even the predominantly natural aerosols, like dust, can be thought of as having an anthropogenic component, for example as human activities like agriculture and urbanization increase aridity and loosen topsoil [e.g. Ginoux et al., 2012]. This dissertation focuses on the anthropogenic component of aerosol emissions as a key agent of human-induced climate change.

Unlike their gaseous counterparts, aerosols tend to have relatively short atmospheric lifetimes [Myhre et al., 2013]. They are scavenged by clouds droplets and precipitation and settle out of the atmosphere gravitationally [Seinfeld and Pandis, 2012]. In the lower troposphere, all of these processes operate efficiently, resulting in a lifetime of the order of days. If aerosols are lofted into the upper troposphere, for example in deep convective clouds or through intense combustion plumes, removal processes are less efficient. This can result in lifetimes of a week to a few weeks.

Because of this relatively short lifetime, aerosols tend to remain concentrated near their emissions sources, which are primarily located in highly industrialized and developing regions [e.g. Penner et al., 2001]. This creates “hotspots” of aerosol-radiation interactions and their cloud, temperature, and hydrological consequences [e.g. Bollasina et al., 2011; Shindell et al., 2012]. Major regions of industrial aerosol emissions and concentrations include East and South Asia, Eastern North America, and Europe. Major regions of biomass aerosol emissions
and concentrations include Brazil, Sub-Saharan Africa, and Southeast Asia [Lamarque et al., 2010]. The climates of these regions feel the particular effects of their aerosol burdens, but the gradients that are set up between these polluted regions and their unpolluted surrounds can induce large-scale circulation changes that can produce a range of downstream effects [e.g. Charlson et al., 1991; Chou et al., 2005; Wang, 2007; Ming et al., 2011; Bollasina et al., 2011; Allen et al., 2015]. One focus of this dissertation is on how this heterogeneous distribution of anthropogenic aerosols, as compared with homogeneously distributed greenhouse gases, affects the spatial pattern of response to each.

Within this diversity of anthropogenic aerosol types, there are species that counteract the warming from greenhouse gases by scattering shortwave radiation out of the surface-atmosphere system, as well as those that reinforce it by absorbing shortwave radiation within the atmosphere. Sulfate aerosol is the most potent scattering aerosol at visible wavelengths [e.g. Charlson et al., 1991], while black carbon is the most potent absorber [e.g. Sato et al., 2003; Bond et al., 2013]. However, because the different aerosol species often have common emission sources and tend to mix and agglomerate once in the atmosphere, it is increasingly thought that many manmade aerosol populations across the globe are at least partially absorbing [e.g. Andreae et al., 1986; Pósfi et al., 1999; Jacobson et al., 2001; Bond et al., 2013]. This dissertation primarily focuses on sulfate and black carbon aerosols as typifications of scattering and absorbing aerosols, respectively.

The ability of sulfate and black carbon, respectively, to scatter and absorb shortwave radiation has substantial effects on the surface, atmospheric, and top-of-atmosphere shortwave radiative balance. By scattering shortwave radiation back to space, sulfate aerosols reduce the net radiation into the earth system, resulting in a net decrease in global mean temperature [Boucher et al., 2013]. They also deplete the shortwave radiation reaching the earth’s surface, moderating the surface energy balance [Wild, 2012]. In this way, scattering aerosols have a negative influence on net downward energy at both the surface and the top-of-atmosphere.
Aerosols’ ability to absorb shortwave radiation, however, has vertically differentiated impacts on the energy balance [e.g. Ramanathan et al., 2001]. Shortwave radiation is absorbed by the aerosol, transferring energy into the atmospheric column in the layer in which the aerosol resides, which may manifest via responses in atmospheric temperature or motion [e.g. Chung et al., 2002; Ramanathan et al., 2005; Randles and Ramaswamy, 2008; Ming et al., 2010]. Aerosol absorption from species like black carbon thus increases the net energy into the earth system, resulting in a net increase in global mean temperature [Chung and Seinfeld, 2005; Boucher et al., 2013]. However, because aerosol absorption traps radiation within the atmosphere, it still depletes the shortwave radiation reaching the Earth’s surface [e.g. Ramanathan et al., 2005]. This is in crucial contrast to positive radiative forcing from greenhouse gases, which increases net downward radiation at both the top-of-atmosphere and at the surface and warms both the troposphere and surface [e.g. Andrews et al., 2009]. Absorbing aerosols are unique in their ability to produce opposite signs of radiative perturbation at the top-of-atmosphere and surface—a characteristic that is highlighted in this dissertation.

Aerosols also have the capability to influence the structure, optical properties, and persistence of cloud cover, which can produce additional radiative, thermodynamical, and dynamical impacts [e.g. Lohmann and Feichter, 2001; Khain et al., 2005; Ming et al., 2005; Jiang and Feingold, 2006]. Sulfate aerosols can act as nuclei for the condensation of cloud droplets, resulting in clouds composed of more and smaller droplets that reflect more shortwave radiation, precipitate less, and have longer lifetimes than in the absence of aerosols [Twomey, 1977; Albrecht, 1989]. Black carbon aerosol, by heating the atmospheric column, can substantially alter the thermodynamic environment for cloud formation, either invigorating or suppressing cloud growth depending on the particular dynamical environment and the altitude of the heating [e.g. Hansen et al., 1997; Johnson et al., 2004; Koch and Del Genio, 2010; Wang, 2013]. These effects carry substantial uncertainty, however, partially due to our incomplete understanding of cloud physics [Boucher et al., 2013]. They are not the
central focus of this dissertation, but have been investigated by the author elsewhere [Persad et al., 2012].

East Asia emerges as a particular hotspot of absorbing and scattering aerosol emissions and concentrations and, therefore, a particularly appropriate testbed for investigating the impact of the vertical heterogeneity of aerosols’ radiative effects in a regional context. The region composed of China, Japan, and the Koreas has been the locus of globally maximum emissions of both black carbon and sulfate since the 1990s [e.g. Lamarque et al., 2010; Streets et al., 2013]. This dissertation contains a particular focus on quantifying and analyzing the presence of this vertical heterogeneity over East Asia and its impacts on East Asian regional climate.

The implications of aerosols’ climate impacts for societal decision-making are multifold. The many challenges associated with determining aerosols’ net impact, as the primary offset of anthropogenic greenhouse gas warming, are one of the leading sources of uncertainties in predicting future climate change and its impacts [IPCC, 2013]. Further, because aerosols are short-lived and concentrated near emissions sources, the location of aerosol emissions influences the location of aerosol effects, a fact not true of long-lived, dispersed greenhouse gases. A key outcome of this characteristic is that geopolitical processes, like trade, that affect the global distribution of aerosol emissions can result in reallocation of the populations that feel aerosols’ climate impacts [e.g. Lin et al., 2014]. This dissertation also explores such policy dimensions of the heterogeneity in aerosols’ impacts.

1.2 Overview and Research Context

The distinctive ability of aerosols, particularly in their light-absorbing capacity, to redistribute energy both horizontally between polluted and unpolluted regions and vertically between the surface and atmosphere inspires their centrality in this dissertation. Aerosols’ interactions with shortwave radiation have profound influence on the amount of radiation
reaching the Earth’s surface in polluted regions [Wild, 2012]. In the case of absorbing aerosols, this influence is of opposite sign between the surface and atmosphere [Ramanathan et al., 2001]. The regional climate implications of this behavior and the ways in which they propagate at a global scale and in human systems are issues of paramount scientific and societal interest.

In this context, this dissertation seeks to address the following research questions:

• How similar are the patterns of surface energy flux response to heterogeneously-distributed aerosol forcing versus homogeneously-distributed greenhouse gas forcing? (Chapter 2)

• How strongly do aerosols partition radiative energy between the atmosphere and surface through absorption in high-emissions regions like East Asia? (Chapter 3)

• What effect does this have on regional climate when aerosol absorption’s surface and atmospheric effects are considered in isolation and in tandem? (Chapter 4)

• How reliable are assumptions about the manner in which future emissions of absorbing black carbon aerosols will evolve in currently high-emissions regions like East Asia, and what are the societal implications of uncertainties therein? (Chapter 5)

There has been substantial debate in the literature regarding the degree of similarity between the spatial patterns of climate response to positive and homogeneous greenhouse gas forcing versus negative and heterogeneous aerosol forcing, particularly in the context of attributing observed global patterns of temperature and precipitation change to the two modes of anthropogenic emissions [e.g. Ramaswamy and Chen, 1997; Hegerl et al., 2011; Xie et al., 2013; Shindell, 2014]. Previous model studies comparing only sulfate aerosol against carbon dioxide greenhouse gas suggested that the pattern of response to aerosol is substantially different from the pattern of its forcing, and that the eventual temperature change due to a unit change in radiative forcing is larger if that forcing comes from carbon dioxide than if it
comes from sulfate [Taylor and Penner, 1994]. Later model studies comparing all greenhouse gases to all aerosols plus ozone, however, found that the transient change in temperature is larger in response to a unit change in aerosols and ozone than to a unit change in greenhouse gas—a result of aerosols’ concentration in the Northern Hemisphere, where land responses and feedbacks are stronger [Shindell, 2014]. In general, however, it is accepted that the horizontal patterns of temperature response to greenhouse gases and aerosols are sufficiently distinct to allow for identification of their individual signals in observations [Bindoff et al., 2013].

Chapter 2 investigates the spatial pattern of response to aerosols and greenhouse gases in this context. A recent model study has found that the spatial pattern of temperature response over the oceans to present-day greenhouse gases and aerosols have substantial similarity [Xie et al., 2013]. It suggests that common ocean-atmosphere feedbacks excite shared spatial patterns of regional climate change over the oceans in response to either forcing type and recommends that the mechanisms for this pattern formation should be studied in greater detail. Chapter 2 takes up this charge in an analysis of how the initial development of these shared patterns occurs, via investigation of the pattern of ocean surface energy flux produced by the rapid land-and-atmosphere response to present-day aerosols versus greenhouse gases. It is largely composed of a manuscript submitted as Persad et al. [2016].

A key mode by which aerosols affect the surface energy balance, and a distinction in their radiative impact compared to greenhouse gases, is their depletion of shortwave radiation reaching the Earth’s surface, colloquially known as “solar dimming”. Satellite, ground-based measurement, and modeling studies all confirm that aerosols have contributed to detectable, worldwide declining trends in surface solar radiation starting in the 1950s [Wild, 2009, and references therein]. While increased air quality controls in certain regions (e.g. Europe and North America) have allowed their regional trends to recover since the 1980s, other regions (e.g. South and East Asia) have largely continued their decline. Few studies, however, have analyzed the mechanisms via which aerosols achieve this solar dimming in specific regions,
particularly the relative roles that shortwave scattering and shortwave absorption play in contributing to it.

Chapters 3 and 4 fill this critical gap in understanding over East Asia, the region of currently highest black carbon and sulfate emissions [Lamarque et al., 2010]. Chapters 3 mechanistically analyzes the relative contributions of aerosol scattering and aerosol absorption to observed clear-sky solar dimming over East Asia and provides a quantification of the role of different aerosol processes in determining this partitioning. It forms the body of an article published in [Persad et al., 2014]. Chapter 4 builds on this through an investigation of how atmospheric heating and surface dimming by absorbing aerosols operate in isolation and in tandem in affecting East Asian summertime climate.

Future projections of how black carbon emissions will evolve over the next century are highly uncertain, a knowledge gap of considerable concern given the impacts of absorbing aerosol on regional and global climate. While the Representative Concentration Pathways, currently used to capture the range of future emissions trajectories, predict a decline in global emissions that manifests in all regions [Takemura, 2012], the earlier generation of emissions scenarios produced a much broader range of emissions futures [Nakicenovic and Swart, 2000], including those that contained increasing black carbon emissions in already high-emissions regions like East Asia. Assumptions about the economic and sociotechnological transitions that will influence future black carbon emissions are subject to considerable debate [e.g. Dasgupta et al., 2002; Smith et al., 2005; Suri and Chapman, 1998; Malm, 2012], and plausible variations within them can produce substantially different emissions futures [Smith et al., 2011b]. Chapter 5 interrogates the assumptions dictating China’s black carbon emissions trajectory in the RCPs and analyzes the climate and health implications of uncertainties therein in the context of the scientific findings of Chapters 3 and 4.

Chapter 6 concludes the dissertation with a summary of the research findings and their implications, as well as a discussion of the dissertation’s boundaries and the future work inspired by it.
Chapter 2

Spatially Similar Surface Energy Flux Perturbations due to Greenhouse Gases and Aerosols

2.1 Introduction

As discussed in Chapter 1, anthropogenic greenhouse gases and aerosols have substantial differences in their radiative properties, lifetime, and global distribution. However, recent studies suggest that, despite distinct geographic distributions of top-of-the-atmosphere radiative forcing, anthropogenic greenhouse gases and aerosols give rise to similar patterns of climate response (though of opposite sign) in fully atmosphere-and-ocean coupled general circulation model simulations. The surface energy flux perturbation, a crucial pathway by which atmospheric forcing is communicated to the ocean, may be a vital link in explaining the large-scale spatial similarities in the full atmosphere-and-ocean responses to disparate forcings. We here analyze the fast, atmosphere-only change in surface energy flux caused by present-day greenhouse gases versus aerosols to elucidate its role in shaping the subsequent slow, coupled response.
Although it has been demonstrated that the fully coupled response to greenhouse gases (GHGs) and aerosols have significant, though not complete, spatial pattern similarity, the mechanisms for pattern formation remain poorly characterized [e.g. Levy et al., 2008, 2013; Xie et al., 2013]. Xie et al. [2013] suggest that the spatial similarities in the fully coupled response to GHGs and aerosols, analyzed in a subset of the Coupled Model Intercomparison Project Phase 5 (CMIP5) models, are strongly mediated by common patterns of ocean-atmosphere feedbacks that can be separated conceptually into a fast, atmosphere-only component and a slower, ocean-atmosphere coupled component. The degree of spatial similarity in the climate response to GHGs versus aerosols has significant implications for questions in the detection and attribution of anthropogenic climate change [Bindoff et al., 2013, and references therein] and for our understanding of the transient climate response to heterogeneous versus homogeneous forcers [e.g. Shindell, 2014]. It is thus vital to characterize the fast and slow mechanisms via which the spatial patterns of response are produced.

We find that, although the two forcings are largely uncorrelated under clear-sky conditions at the top-of-the-atmosphere, their surface energy flux perturbation patterns are significantly anti-correlated. Our analysis highlights the common modes of atmospheric circulation and surface energy adjustment that are triggered by both greenhouse gas and aerosol forcings. These produce antisymmetric (i.e. symmetric, but of opposite sign) spatial patterns of surface sensible and latent heat flux variations in response to the two forcers, particularly over the winter-hemisphere oceans. Our results suggest that atmosphere and land processes alone are capable of achieving substantial homogenization within a given hemisphere in the climate response to disparate forcers on fast timescales, with implications for detection and attribution and for the understanding and prediction of the regional climate impacts of anthropogenic greenhouse gases and aerosols.
2.2 Methods

2.2.1 Atmospheric General Circulation Model Simulations

All simulations in this study are conducted with a modified version of the Geophysical Fluid Dynamics Laboratory’s AM2.1 Atmospheric General Circulation Model, which prescribes aerosol concentrations and has a prognostic treatment of aerosol indirect effects on liquid clouds [Ming and Ramaswamy, 2009]. We analyze the time-averaged values from a set of three simulations run for 7 years with sea surface temperatures fixed to a repeating climatological annual cycle and different configurations of atmospheric constituents: an all forcing run with all natural and anthropogenic levels varying according to their historical values from 1983–1989 (ALL_F); a run with anthropogenic aerosol concentrations set to 1860 values and all other forcers varying according to their historical values from 1983–1989 (1860_AERO); and a pre-industrial run with all forcers set to 1860 values (PI). Aerosol effects were derived from the ALL_F run minus the 1860_AERO run, and GHG effects were derived from the 1860_AERO run minus the PI run.

2.2.2 Correlation Coefficients and Significance

Pearson correlation coefficients are calculated via linear regression with area-weighting. Scatterplots of all data were analyzed to ensure insensitivity to outliers and qualitative linearity of relationships. All correlation values given are significant at the 95% level.

Results were evaluated for robustness by conducting preliminary analysis on the first 3 and last 3 years of the model simulations separately. The signal is highly consistent between the two 3-year periods in the annual-mean and JJA, but there is greater variability in the DJF midlatitude signal (Table A.1), as expected from the noisy nature of the Northern Hemisphere wintertime weather [e.g., Hurrell and Deser, 2015]. Only features consistent between these two, effectively independent, samples are considered in this analysis.
2.3 Results and Discussion

We here probe the fast, atmosphere-only component of the formation of the spatial patterns of response to present-day GHGs and aerosols in an atmospheric general circulation model—a crucial intermediate step toward understanding the fully coupled response. We find that the top-of-the-atmosphere (TOA), all-sky, effective radiative forcings (ERFs) \cite{Myhre2013} of GHGs and aerosols are only weakly anti-correlated (annual-mean $R = -0.42$) in our simulations, comparable to values found in other studies \cite[e.g.][]{Xie2013}. ERF is calculated as the difference in TOA radiative flux between simulations with and without the forcing agent after the atmospheric and land temperatures have been allowed to re-equilibrate. Aerosols’ TOA ERF pattern (Figs. 2.1b and 2.2b), due to aerosols’ short atmospheric lifetime, is dependent on regional factors like surface albedo and the location of emissions, and is concentrated in the Northern Hemisphere. GHGs’ TOA ERF pattern, meanwhile, is by-and-large hemispherically symmetric and uniformly distributed (Figs. 2.1a and 2.2a).

The clear-sky TOA ERF of GHGs and aerosols is essentially uncorrelated (annual-mean $R = -0.09$), suggesting that the all-sky TOA ERF correlation is strongly influenced by clouds through the so-called cloud masking effect \cite[e.g.][]{Soden2004}. Furthermore, the

| Table 2.1: Spatial correlations of TOA effective radiative forcing and ocean surface energy flux perturbations due to greenhouse gases versus aerosols. |
|---------------------------------|-----------|-----------|-----------|
| TOA Effective Radiative Forcing | Annual Mean | JJA | DJF |
| Clear-sky TOA ERF              | -0.42     | -0.34    | -0.57    |
| Tropics (20°S-20°N)            | -0.32     | -0.45    | -0.53    |
| Surface Energy Flux Perturbation| -0.57     | -0.55    | -0.62    |
| Tropics (20°S-20°N)            | -0.55     | -0.57    | -0.60    |
| NH Extratropics (20°N-90°N)    | -0.67     | -0.22    | -0.64    |
| SH Extratropics (20°S-90°S)    | -0.45     | -0.70    | -0.61    |
| Surface Radiative (SW+LW) Flux | -0.26     | -0.22    | -0.48    |
| Surface Sensible Heat Flux     | -0.58     | -0.57    | -0.61    |
| Surface Latent Heat Flux       | -0.45     | -0.52    | -0.58    |
The spatial anti-correlation of the change in low cloud cover between the two runs (\( R = -0.52 \), not listed) is comparable to that for the all-sky TOA ERF, indicating that the latter can also be explained partially by antisymmetric (i.e. symmetric, but of opposite sign) cloud changes in response to the two forcers.

The spatial differences between the TOA ERF due to GHGs and that due to aerosols, however, are not fully maintained in the surface energy flux perturbation to each. The pattern of surface energy flux perturbation (\( \Delta S \), defined as the change in total surface energy flux, composed of radiative shortwave (SW) and longwave (LW), latent (LH), and sensible (SH) energy) in response to the two forcers is more strongly anti-correlated (annual-mean \( R = -0.57 \)) than the clear-sky TOA ERF (annual-mean \( R = -0.09 \)), indicating a process
of homogenization between the TOA perturbation and the surface perturbation. Because
the surface energy balance over land rapidly re-equilibrates due to the low effective heat
capacity of the land surface, ΔS, is near zero for all land surfaces by necessity. Thus, the
anti-correlation of ΔS manifests almost entirely over the ocean. The oceanic surface energy
balance is not constrained to re-equilibrate on atmosphere-only timescales, and as such will
primarily reflect the atmospheric conditions setting surface fluxes.

ΔS can be construed as an intermediary between the TOA atmospheric perturbation of a
forcing agent and the ocean response thereto, and is thus a telling manifestation of the fast,
atmosphere-and-land-only pathway for fully atmosphere-and-ocean coupled response pat-
tern formation. The annual-mean, global-mean ΔS correlation between GHGs and aerosols
emerges most strongly in the surface latent and sensible heat flux components of the surface
energy balance (Table 2.1). In contrast, the surface radiative (shortwave and longwave) flux change is little correlated (annual-mean $R = -0.26$), indicating that neither climatological cloud masking nor the similar pattern of cloud change can explain the spatial similarities in $\Delta S$ (though the processes that produce the similar patterns of both cloud change and surface energy flux change may be related). The strongest anti-correlation occurs largely over the winter-hemisphere extratropical oceans, as indicated by the seasonal and latitudinal decomposition of $\Delta S$ (Table 2.1), and is driven by antisymmetric wave-like patterns in the surface latent and sensible heat flux change in response to each forcer (Figs. 2.1c,d and 2.2c,d).

The common pattern of surface heat flux change in response to each forcer is a manifestation of a wave perturbation to the atmospheric circulation that is produced by both the forcing due to GHGs and that due to aerosols. Over the extratropical oceans, a barotropic stationary Rossby wave perturbation to the atmospheric flow, consistent with the wave pattern in surface heat fluxes, is evident in alternating positive and negative anomalies collocated in sea level pressure (SLP) and in geopotential height at the 500 mb pressure level ($Z_{500}$) for both forcers (Figs. 2.3a,b and 2.4a,b). The annual-mean patterns of SLP and $Z_{500}$ perturbations due to GHGs and aerosols are correlated at $R = -0.53$ and $R = -0.49$, respectively. Surface winds and temperature and specific humidity gradients between the surface and air, the primary controllers of surface heat fluxes, also exhibit qualitative wave patterns in the extratropics (not shown)—a consequence of the changes in extratropical atmospheric flow demonstrated in the SLP and $Z_{500}$ anomalies.

Rossby wave perturbations to the extratropical atmospheric flow can occur via wave sources located either in the extratropics or in the tropics. Wave excitation within the extratropics can result from changes in extratropical land-sea thermal and diabatic heating contrast caused by land surface temperature adjustments to forcing under the constraint of fixed sea surface temperatures [Held et al., 2002; Ming et al., 2011]. Excitation of extratropical waves from within the tropics, meanwhile, can occur due to changes in tropical deep
Figure 2.3: JJA-mean perturbations due to (a, c, e) GHGs and (b, d, f) aerosols in (a, b) 500mb geopotential height (contour interval, 5 m) and sea level pressure, (c, d) surface temperature, and (e, f) precipitation.
Figure 2.4: DJF-mean perturbations due to (a, c, e) GHGs and (b, d, f) aerosols in (a, b) 500mb geopotential height (contour interval, 5 m) and sea level pressure, (c, d) surface temperature, and (e, f) precipitation.
convection and precipitation (and thus atmospheric latent heating), the signal from which then propagates into the extratropics [Sardeshmukh and Hoskins, 1987; Held and Soden, 2006; Vecchi and Soden, 2007; Ming et al., 2011].

Greenhouse gases and aerosols trigger these wave sources antisymmetrically. In the extratropics, the location of the landmasses serves as a potent fixed wave source in the presence of both forcers. The wintertime land-sea contrast ($T_{L/S}$, quantified as the climatologically negative wintertime hemispheric-mean difference between land and ocean surface temperatures) increases in the presence of aerosols ($\Delta T_{L/S} = -0.15$ K for the Northern Hemisphere, $-0.006$ K for the Southern Hemisphere) and decreases in the presence of GHGs ($\Delta T_{L/S} = 0.31$ K for the Northern Hemisphere, 0.037 for the Southern Hemisphere). This perturbation to the climatological land-sea contrast constitutes a diabatic heating anomaly that acts as an extratropical source of Rossby waves [e.g. Held et al., 2002]. The surface temperature response to GHGs and aerosols (Figs. 2.3c,d and 2.4c,d) is constrained to land in these prescribed sea surface temperature (SST) runs. However, the land surface temperature response to the two forcers is highly anti-correlated (annual-mean $R = -0.71$), likely constrained by local surface energy availability [Andrews et al., 2009] and regional sensitivities and feedbacks that are largely forcing independent [Armour et al., 2012].

In the tropics, changes due to GHGs and aerosols in the atmospheric latent heat release associated with deep tropical ($20^\circ S$–$20^\circ N$) precipitation can also act as a source for extratropical Rossby waves. The fact that the simulated precipitation changes are indeed antisymmetric (annual-mean $R = -0.53$) (Figs. 2.3e,f and 2.4e,f) may contribute to the similar extratropical wave patterns. Interestingly, it is not straightforward why the two forcers would give rise to antisymmetric precipitation changes. In fact, a thermodynamic scaling argument suggests the opposite: under fixed-SST conditions, both GHGs and aerosols increase tropospheric absorption of radiative energy, via increased absorption of longwave radiation in the case of GHGs and of shortwave radiation in the case of aerosols, and thus have suppressing effects on tropical mean precipitation [e.g. Roeckner et al., 1999; Allen and Ingram]...
Figure 2.5: A simple, two-dimensional energy balance between the land and ocean is schematically depicted. Because the land energy balance rapidly reequilibrates ($\Delta R_{\text{surf}}^L + \Delta H_{\text{surf}}^L = 0$), ERF over land ($ERF^L = \Delta R_{\text{surf}}^L + \Delta AA^L$) must be transported over the ocean for longterm storage ($\Delta R_{\text{surf}}^O + \Delta H_{\text{surf}}^O = ERF^O + ERF^L$).

2002; Held and Soden 2006; Ming et al., 2010. As this thermodynamic argument would suggest, in our simulations both forcers decrease tropical mean precipitation (by -0.89% for present-day GHGs and -1.2% for present-day aerosols) and increase atmospheric absorption (Table 2.2) in the annual mean. We suggest that the manifestation of this tropical-mean precipitation decrease in an antisymmetric rather than a symmetric spatial pattern may be the result of the opposite land-sea surface temperature contrast patterns, which in turn may cause opposite dynamically-driven shifts in monsoon precipitation patterns. This is supported by the fact that precipitation in the annual mean decreases over the ocean and increases over land due to GHGs, but decreases over both land and ocean due to aerosols in our simulations.

Our analysis of the mechanisms of surface pattern correlation demonstrates that the atmospheric circulation is an efficient homogenizer of heterogeneous forcings, even under fixed-SST conditions. Indeed, a simple energy balance analysis—depicted schematically in Figure 2.5—allows us to argue this based on energetic constraints alone. Because land surface
Table 2.2: Annual-mean perturbations to energy balance terms due to present-day GHGs and aerosols in $10^{15}$ Watts (globally averaged in W m\(^{-2}\)). Heat flux terms are positive upward, radiative flux terms are positive downward, atmospheric absorption terms are positive into the atmospheric column.

<table>
<thead>
<tr>
<th>Perturbation</th>
<th>Greenhouse Gas</th>
<th>Aerosol</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ocean surf. heat flux ($\Delta H_{\text{surf}}^O$)</td>
<td>0.58 (1.2)</td>
<td>0.22 (0.44)</td>
</tr>
<tr>
<td>Global atmos. abs. ($\Delta AA_{O+L}$)</td>
<td>0.53 (1.0)</td>
<td>0.86 (1.7)</td>
</tr>
<tr>
<td>Land surf. radiative flux ($\Delta R_{\text{surf}}^L$)</td>
<td>0.083 (0.16)</td>
<td>-0.68 (-1.3)</td>
</tr>
<tr>
<td>Ocean surf. radiative flux ($\Delta R_{\text{surf}}^O$)</td>
<td>0.45 (0.89)</td>
<td>-1.3 (-2.5)</td>
</tr>
</tbody>
</table>

and atmospheric energy perturbations rapidly equilibrate to zero, the total ocean surface energy perturbation ($\Delta R_{\text{surf}}^O + \Delta H_{\text{surf}}^O$, with $\Delta R$ and $\Delta H$ denoting the SW+LW radiative and LH+SH heat components, respectively) must equal the TOA ERF over oceans ($ERF^O$) plus that over land ($ERF^L$) in the global mean: $\Delta R_{\text{surf}}^O + \Delta H_{\text{surf}}^O = ERF^O + ERF^L$.

Vertical radiative energy conservation dictates that the TOA ERF must equal the sum of the surface radiative perturbation ($\Delta R_{\text{surf}}$) and any atmospheric SW+LW absorption change ($ERF = \Delta R_{\text{surf}} + \Delta AA$). Thus, the surface heat flux perturbation over ocean will be equal to the total atmospheric SW+LW absorption change over land and ocean plus the land surface radiative perturbation ($\Delta H_{\text{surf}}^O = AA_{O+L} + \Delta R_{\text{surf}}^L$), a balance that is evident in our simulations (Table 2.2). This coupling between the atmospheric and surface energetics over the land and the ocean dictates that most of the spatial heterogeneity in TOA forcing cannot be maintained and must be rapidly transformed (from shortwave and longwave radiation to latent and sensible heat) and redistributed (from the land surface and atmosphere to the ocean surface) by the atmospheric circulation. This circulation adjustment process is fundamentally responsible for the similar ocean heat flux perturbation patterns between GHGs and aerosols.

Notably, the fast homogenization described in this work operates primarily within a single hemisphere. The ratio of Northern Hemisphere to Southern Hemisphere TOA ERF does not differ significantly from that of the surface energy perturbation (1.1 and 1.1, respectively, for GHGs and 2.8 and 3.0, respectively, for aerosols). Past work on the coupled response to
greenhouse gases versus aerosols [e.g. Taylor and Penner 1994, Cox et al. 1995] has highlighted the role that inter-hemispheric homogenization plays in determining the pattern of response to aerosols concentrated in the Northern Hemisphere. Under fixed-SST conditions used here to isolate fast land-and-atmosphere processes, however, the Hadley circulation—the main mode of atmospheric cross-equatorial transport—cannot readily respond to perturbations [e.g. Hill et al. 2014], constraining homogenization to within a given hemisphere. A related key outcome of this fixed-SST constraint is that tropical precipitation shifts that are typically associated with Hadley Circulation changes due to aerosol-driven hemispheric asymmetry [e.g. Broccoli et al. 2006, Ming et al. 2011, Haywood et al. 2013, Hwang et al. 2013, Allen et al. 2015] will not manifest in the fast land-and-atmosphere-only response isolated here.

2.4 Conclusions

The degree of similarity in the spatial pattern of the climate response to GHGs and aerosols has many implications for understanding and predicting the relative climate impacts of these forcers. Detection and attribution studies rely on spatial pattern as one component of the fingerprint of a given forcer, and similarity therein can result in a degradation of the ability to distinguish the signal from GHGs versus aerosols in climate phenomena [Bindoff et al. 2013, and references therein]. Further, our understanding of the regional distribution of present and future climate change relies on constraining the spatial structure of the response to heterogeneous forcers like aerosols versus homogeneous forcers like greenhouse gases [e.g. Shindell and Faluvegi 2009, Shindell et al. 2010]. This issue is of particular importance given that the spatial distribution of aerosol forcing has shifted since preindustrial times and is expected to continue to shift into the future as nations heterogeneously enact emissions mitigation measures for health, air quality, and other reasons [e.g. Takemura 2012].
Our work reveals that, even on the short timescales captured in fixed-SST simulations, GHGs and aerosols can be expected to produce strongly correlated spatial patterns of change across a range of variables. This is enforced by the symmetric perturbation to the extratropical circulation provided by land-sea contrast, the spatial structure of which is fixed by the location of the landmasses and, thus, relatively insensitive to the structure of the initial forcing. Analysis of the spatial correlation of atmosphere-and-land-only change in response to greenhouse gases and aerosols in a range of models will be crucial to improved understanding of this phenomenon. We have here presented a picture of the fast dynamical and thermodynamical mechanisms that drive these similarities in our model, providing a bridge to understanding the fully atmosphere-and-ocean coupled spatial patterns described by Xie et al. [2013] and others, and encourage continued analysis of this phenomenon in additional climate models.

The results of this chapter shed light on the large-scale climate system’s fast processing of aerosols’ regionally heterogeneous radiative perturbations. In the following two chapters, we analyze how this radiative perturbation manifests over the region of largest present-day anthropogenic aerosol loading: East Asia. We demonstrate the regionally intense radiative and climate perturbations that can be induced by localized aerosols, highlighting the need for a robust understanding of the degree to which the climate system can homogenize the signal from these heterogeneous hotspots of aerosol-climate interactions.
Chapter 3

The Role of Aerosol Absorption in Driving Solar Dimming over East Asia

3.1 Introduction

Downward surface solar radiation (SSR) influences the energy available for both sensible and latent heat release, with significant implications for the hydrological cycle and convection \[\text{[Ramanathan et al., 2001; Andrews et al., 2009]}\]. Studies of surface-based observations dating back to the 1950s, however, indicate that there have been decadal variations in the amount of solar radiation reaching the Earth’s surface \[\text{[Wild, 2009; and references therein]}\]. Observation sites world-wide exhibit a decrease in SSR from the 1950s to the 1980s, followed by an increase in the following decades in certain regions, such as Europe and North America. The observed surface trends are an order of magnitude larger than observed variations in top-of-atmosphere insolation \[\text{[Fröhlich and Lean, 1998; Willson and Mordvinov, 2003]}\] and are evident under both all-sky and clear-sky conditions \[\text{[Wild, 2009]}\], indicating that they are the result of changes in atmospheric constituents rather than solar or cloud variability.
In the case of clear-sky SSR, possible trend explanations focus on changes in atmospheric composition. Radiative transfer calculations indicate that changes in water vapor much larger than those observed would be necessary to effect the observed clear-sky SSR changes [Wild, 1997]. This leaves changes in aerosol concentrations as the most plausible explanation for clear-sky SSR variability. Aerosols can attenuate shortwave radiation by either scattering or absorbing it, reducing the amount that reaches the surface. A number of studies have strongly correlated decadal changes in aerosol emissions with decadal changes in SSR, particularly on a regional scale [Streets et al., 2006, 2009]. Modeling studies (including the results of this work) support a causal relationship between aerosol and SSR changes, indicating that increasing aerosol concentrations can drive large regional decreases in SSR [e.g. Nazarenko and Menon, 2005; Ramanathan et al., 2005; Freidenreich and Ramaswamy, 2011].

Over Asia, in particular, trends in SSR have manifested largely as a decrease throughout the observational record. A synthesis of observational studies over East Asia suggests a decreasing trend in all-sky SSR of approximately 7 W m$^{-2}$ decade$^{-1}$ during the 1950s–1980s [Wild, 2012]. This “dimming,” as it is colloquially known, has been strongly correlated with increasing emissions of sulfate and black carbon aerosols regionally [Che et al., 2005; Qian et al., 2006, 2007]. East Asia, therefore, constitutes an ideal location over which to analyze aerosols’ interaction with shortwave radiation and ways in which this interaction may impact SSR values. We, thus, focus our analysis on this region.

Whether an aerosol-driven decrease in SSR comes primarily from increased scattering or from increased absorption can have a significant impact on how the regional climate responds to the SSR perturbation. Absorption traps radiative energy within the atmosphere, while scattering reflects that energy back out of the surface/atmosphere system. As discussed by Ramanathan and Carmichael [2008], surface cooling associated with an SSR reduction, coupled with atmospheric heating from aerosol absorption within the atmospheric column, can weaken the radiative-convective coupling of the atmosphere and decrease evaporation and precipitation. Ming et al. [2010], meanwhile, demonstrate that, although absorbing aerosols
generate surface warming (which is generally associated with increased precipitation in the case of greenhouse gas warming [e.g. Allen and Ingram 2002; Held and Soden 2006]), the increased atmospheric absorption they produce can suppress precipitation on a global mean basis.

Few existing papers, however, analyze the contribution of aerosols to clear-sky SSR variations in particular [Wild, 2009], and fewer yet have analyzed the relative contributions of absorption and scattering in model simulations. Several studies have compared modeled clear-sky SSR with observed clear-sky proxy data [e.g. Norris and Wild 2007, 2009; Ruckstuhl and Norris 2009; Dwyer et al. 2010; Allen et al. 2013], but these studies focus on model trend intercomparison rather than on detailed analysis of the mechanisms behind the modeled trends or the robustness thereof.

In the few studies in which the scattering and absorption contributions to dimming trends have been distinguished, the mechanisms responsible for the modeled scattering and absorption are not elucidated [e.g. Folini and Wild, 2011; Stier et al. 2007] identify that subtle variations in the microphysics of the aerosol representation can significantly affect the modeled amount of absorption and overall shortwave attenuation. Given the many potential climate impacts of aerosol absorption discussed above, an in-depth analysis of how much absorption models produce and of the particular mechanisms via which they do so will be vital to a full picture of how the climate will respond to changes in SSR.

This study seeks to advance the existing literature through a detailed analysis of the relative contributions of aerosol scattering and absorption to modeled clear-sky SSR trends over East Asia, the mechanisms responsible for the simulated absorption, and the sensitivity of that absorption to variations in characteristics of the aerosol parameterization. We achieve this using a model hierarchy that allows us to analyze features from the large-scale trend down to the aerosol microphysics responsible. We use ensemble simulations in the Geophysical Fluid Dynamics Laboratory’s (GFDL) AM2.1 and AM3 atmospheric general circulation models (AGCMs)—included in the CMIP3 and CMIP5 multi-model data archives,
respectively—to isolate the impact of aerosols on the clear-sky dimming trends, and analyze output shortwave radiation variables to characterize the contribution from atmospheric absorption. We then use each model’s standalone radiation module, which allows manipulation of the aerosol parameterizations, to quantify how various aerosol characteristics (including aerosol burden, mixing state, and hygroscopic growth) contribute to the modeled clear-sky dimming and absorption trends. Our goal is both to understand the aerosol mechanisms driving the observed clear-sky trends in SSR over East Asia and to explore its sensitivity to the models’ aerosol parameterizations. Other aerosol characteristics beyond those analyzed here, such as aerosol optical properties and vertical distribution, also contain uncertainties, and the framework developed in this study should be useful in future analysis of additional sensitivities.

We focus our analysis primarily on the effects of sulfate and black carbon aerosols on clear-sky dimming, though other aerosol types are included in all simulations (see Section 3.2). Sulfate aerosol from the oxidation of sulfur dioxide emissions is considered to be the most potent anthropogenic shortwave scatterer [Charlson et al., 1991], while black carbon aerosol from incomplete combustion processes is considered to be the most potent anthropogenic shortwave absorber [e.g. Jacobson, 2000; Sato et al., 2003]. Aerosols can also modify the shortwave radiation budget via their impact on clouds [e.g. Twomey, 1974; Kaufman, 1997; Ackerman, 2000; Lohmann and Feichter, 2001]. However, significant uncertainty is associated with these indirect effects and their representation in models, especially regarding the effect of aerosol absorption on clouds [e.g. Koch and Del Genio, 2010; Persad et al., 2012; Bond et al., 2013]. We, therefore, concentrate solely on issues surrounding the simulation of the clear-sky effects of these two major aerosol species.

Our results demonstrate the importance of aerosol absorption in driving clear-sky solar dimming over East Asia and the contribution to that absorption from different characteristics of the models’ aerosol parameterization. This study constitutes the first time, to our knowledge, that the absorption contribution to regional clear-sky dimming in models has
been mechanistically analyzed. Although our results analyze this behavior for a specific region and suite of models, the outcomes detailed here suggest that such systematic analysis of the absorption contribution to clear-sky dimming is an important diagnostic that models should implement and quantitatively assess the consequences of when evaluating their aerosol formulation.

### 3.2 Methods

#### 3.2.1 Model description

Using experiments conducted with GFDL’s AM2.1 and AM3 AGCMs, we examined the simulated trends in clear-sky SSR over East Asia from 1961-2003 (the period covered by many observational studies). The two models are the atmospheric components of the fully coupled atmosphere-ocean GCMs included in the CMIP3 and CMIP5 model archives (GFDL-CM2.1 and GFDL-CM3, respectively). These two models produce credible simulations highlighting the important role of aerosols in offsetting historic greenhouse gas warming and the global and regional role of aerosols in 20th century temperature evolution [The GFDL Global Atmospheric Model Development Team, 2004; Reichler and Kim, 2008; Donner et al., 2011; Klein et al., 2013]. They thus are excellent tools for studying aerosols’ radiative effects.

The AM2.1 and AM3 aerosol parameterizations contain several differences (summarized in Table 3.1), many of which are typical of improvements made between the CMIP3 and CMIP5 generations of climate models. Generally, advances in computing resources and theoretical understanding have allowed for more complex treatment of aerosols in the newer models [Donner et al., 2011]. Additionally, spatial resolution increases in the newer generation of the model; AM2.1 has a horizontal resolution of 2° latitude × 2.5° longitude (on the order of 200 km on a side) and 24 vertical layers, while AM3’s cubed sphere formulation has a horizontal resolution ranging from 163 km to 231 km, depending on location on the cubed sphere, and 48 vertical layers. Full descriptions of the two models can be found in
Table 3.1: A summary of salient differences in aerosol parameterization between AM2.1 and AM3.

<table>
<thead>
<tr>
<th>Feature</th>
<th>AM2.1</th>
<th>AM3 [Donner et al. 2011]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Emissions</td>
<td>[Olivier et al., 1996] Cooke et al. 1999</td>
<td>[Lamarque et al., 2010]</td>
</tr>
<tr>
<td>Interactivity</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>Prescribed from MOZART</td>
<td>Horowitz et al. 2003</td>
<td></td>
</tr>
<tr>
<td>Mixing State</td>
<td>All external</td>
<td>BC/Sulfate internally mixed</td>
</tr>
<tr>
<td></td>
<td></td>
<td>All else externally mixed</td>
</tr>
<tr>
<td>Hygroscopicity</td>
<td>Sulfate: to 100% RH BC: no</td>
<td>Sulfate: to 97% RH BC: follows sulfate if mixed; else nonhygroscopic</td>
</tr>
</tbody>
</table>
The GFDL Global Atmospheric Model Development Team [2004] and Donner et al. [2011], respectively, but aspects of the models’ aerosol parameterization salient to this investigation are summarized here.

Aerosol concentrations in AM2.1 are prescribed from off-line calculations with the MOZART chemistry transport model [Horowitz et al., 2003] using emissions from Olivier et al. [1996] and Cooke et al. [1999] with optical properties described by Haywood et al. [1999] and Haywood and Ramaswamy [1998]. Once input into AM2.1, these prescribed concentrations can radiatively impact the model meteorology, but are not transported or removed by that meteorology. As such, the meteorological fields used by MOZART to produce the aerosol concentrations seen by AM2.1 are not consistent with the meteorological fields produced by AM2.1 itself. Sulfate, black carbon, organic carbon, sea salt and dust aerosol species are considered. All aerosol types are treated as externally mixed, i.e. though a given aerosol population may contain many different species, any individual aerosol particle is composed purely of one species. Sulfate is treated as hydrophilic, while black carbon is treated as hydrophobic. Hygroscopic growth of sulfate aerosol continues through 100% relative humidity.

Aerosol concentrations in AM3, conversely, are interactive with AM3’s meteorology. Anthropogenic sulfate, black carbon, and organic carbon emissions from Lamarque et al. [2010] are input into AM3 and are transported, aged, and removed according to the meteorology and chemistry within the model itself. Other natural and anthropogenic aerosol species (including sea salt, secondary organic aerosols, and dust) are similarly interactive in the model. Sulfate and black carbon aerosols are assumed to be internally mixed in the model, i.e. coexisting sulfate and black carbon will mix with each other at the individual particle level. The refractive index of the sulfate/black carbon mixture is calculated in the model as a volume-weighted average of the refractive indices of each aerosol species. Black carbon, although largely hydrophobic on its own, will grow hygroscopically when internally mixed with sulfate. Hygroscopic growth is capped at 97% relative humidity in AM3, i.e. aerosol
optical properties are held at those corresponding to 97% relative humidity at all higher relative humidities. Organic carbon contains slight absorption in AM3’s formulation [Donner et al., 2011], but this absorption is minor compared to that of black carbon [Ocko et al., 2012]. The optical properties of other aerosol species, which remain externally mixed, are identical to those used in AM2.1. The simulation and effects of dust also remain identical between the two models and do not contribute to model differences.

3.2.2 Design of experiments

We examine four sets of historical (1861-2003 in AM2.1 and 1870-2005 in AM3) AGCM simulations, forced with observed historical sea surface temperature and sea ice from the HADISST1 dataset [Rayner et al., 2003], in order to isolate the relative contribution of anthropogenic aerosols to the modeled trend in clear-sky SSR over East Asia. These are: (1) A five-member ensemble of experiments that include all forcings (ALL_F), anthropogenic (aerosols, greenhouse gasses, and land-use changes) and natural (solar variations and volcanoes); (2) A three-member ensemble containing only anthropogenic aerosol forcing (AERO); (3) A three-member ensemble containing only anthropogenic well-mixed greenhouse gas and ozone forcings (WMGG); (4) A three-member ensemble containing only natural forcings (NAT). All results shown in this paper are ensemble averages.

3.2.3 Standalone radiative transfer calculation

The radiative transfer modules of AM2.1 and AM3 can be run in a standalone mode, independent of the full models, to produce monthly mean shortwave and longwave fluxes for a set of atmospheric conditions (e.g. temperature, water vapor, clouds, greenhouse gas concentrations, and surface albedo). These conditions are saved from an interactive GCM integration (here, an AM3 all-forcing simulation). Aerosol concentrations are input from an AM2.1 or AM3 aerosol climatology. The standalone code can be modified to substitute one aerosol climatology for another, to change the mixing state of the aerosol population, and to turn
on and off the radiative effects of microphysical processes like hygroscopic growth. The radiative code formulation is otherwise identical across AM2.1 and AM3, ensuring that any differences in output are due to differences in the aerosol characteristics used. Full details of the radiation module used in both models can be found in Freidenreich and Ramaswamy [1999] and Schwarzkopf and Ramaswamy [1999], with modifications described in The GFDL Global Atmospheric Model Development Team [2004].

We utilize this capability to test the influence of various aspects of the aerosol radiative properties on the model-simulated trends in clear-sky SSR and absorption. We perform the following standalone radiative transfer perturbation experiments over one model year: (1) a control case in which the default AM2.1 and AM3 settings are used (AM2_EM and AM3_IM, respectively, with EM denoting external mixing and IM denoting internal mixing), (2) switched mixing state, i.e. AM2.1’s aerosol climatology with internal mixing and AM3’s aerosol climatology with external mixing (AM2_IM and AM3_EM, respectively), (3) hygroscopic growth turned off (..._nohygro), and (4) aerosol radiative effects turned off (..._noaero). These experiments are performed for 1970 and 1990 aerosol burdens derived from AM3 and AM2.1 all-forcing simulations. These years are those closest to the endpoints of the relevant time period for which aerosol concentrations are provided in both models. All other atmospheric and surface constituents are held constant, including surface albedo. The values shown here are for 1990 minus 1970 conditions to provide trend-relevant results.

3.2.4 Observational context

An often-used dataset for comparison of modeled and observed clear-sky dimming over East Asia is that originally published by Norris and Wild [2009]. It is compared with CMIP3 models in Dwyer et al. [2010] and with CMIP5 models in Allen et al. [2013]. We use this clear-sky proxy surface shortwave radiation data as presented in Allen et al. [2013]. They compute monthly mean anomalies in all-sky SSR over East Asia from measurements made at surface pyranometer sites in the Global Energy Balance Archive (GEBA). Clear-sky SSR
values are then derived by subtracting cloud effects from the all-sky values. Allen et al. [2013] used cloud observations from the International Satellite Cloud Climatology Project (ISCCP) and visual cloud observation sources to perform quality control on the surface observation sites and to calculate a shortwave “cloud cover radiative effect anomaly” (CCRE'), which seeks to quantify the shortwave radiative impact of cloud cover anomalies. Allen et al. [2013] then extract time series of clear-sky SSR anomalies from the observed all-sky SSR anomalies by subtracting CCRE' from the all-sky observations using linear regression. The resulting clear-sky SSR proxy anomalies produce a decreasing trend in clear-sky SSR over East Asia of $-0.43 \pm 0.10$ W m$^{-2}$ yr$^{-1}$ over the period from 1961-2003.

There are identified deficiencies in SSR datasets over East Asia [Shi et al., 2008; Tang et al., 2010, 2011]. The clear-sky SSR proxy anomalies can be split into a “dimming” period from 1961-1989 and “brightening” period from 1990-2007, divided by a minimum in the data in 1990 [Allen et al., 2013]. However, Tang et al. [2011] suggest that the minimum in 1990 and following increase in the early 1990s may be a spurious result of instrument retrofits that occurred during that period. We thus choose to focus our observational comparison on the linear trend in clear-sky proxy SSR over the entire time series to minimize possible biases caused by this suspect data.

3.3 Results

3.3.1 AGCM simulations

We analyze the clear-sky SSR and atmospheric absorption anomalies over the period 1961-2003 in the full AGCMs over the eastern portion of East Asia (defined in this study as 22.5°–40° N and 100°–122.5° E), consistent with the spatial and temporal coverage of the Norris and Wild [2009] dataset. The observational proxy for clear-sky SSR is characterized by strong interannual and multidecadal variability, while the model variability is much smaller on both timescales (Figure 3.1). The proxy data may contain higher-than-realistic variability
Figure 3.1: Clear-sky surface solar radiation (SSR) annual-mean anomalies with respect to the 1961-2003 mean in AM3 (black), in AM2.1 (blue), and in the observational proxy estimate from Allen et al. [2013] (red) in W m$^{-2}$.

on interannual timescales due to the cloud removal process used, while the low temporal resolution of the aerosol climatologies used in the models may produce lower-than-realistic model variability on multidecadal timescales, as will be discussed in Section 3.4.4.

Over the entire time period, AM2.1 and AM3 have linear trends (calculated using least squares linear regression) of $-0.47 \pm 0.02$ W m$^{-2}$ yr$^{-1}$ and $-0.30 \pm 0.02$ W m$^{-2}$ yr$^{-1}$, respectively (Figure 3.1). These values are both comparable to the $-0.43 \pm 0.10$ W m$^{-2}$ yr$^{-1}$ trend in the observational proxy; AM2.1 is within the uncertainty of that observational proxy, and AM3’s 95% confidence interval slightly exceeds it. AM2.1’s trend lies on the high end of the observational estimate, however, while AM3’s lies on the lower end. Despite significantly different aerosol characteristics, as mentioned above, both models have been
Figure 3.2: Clear-sky SSR anomaly in W m$^{-2}$ is shown for various attribution experiments in (a) AM3 and (b) AM2.1. The natural forcing (NAT, green) and greenhouse gas only (WMGG, red) experiments show no significant trend, while the aerosol-only (AERO, blue) experiment explains the majority of the trend seen in the all-forcing one (ALL$_F$, black).

This strong clear-sky dimming over East Asia throughout the time period studied is an order of magnitude larger than the global mean clear-sky dimming in both models (AM3: $-0.036$ W m$^{-2}$ yr$^{-1}$, AM2.1: $-0.044$ W m$^{-2}$ yr$^{-1}$). It also stands in stark contrast to Europe ($38^\circ$–$65^\circ$ N and $10^\circ$W–$42^\circ$ E), which exhibits weak clear-sky dimming before 1990 (AM3: $-0.009$ W m$^{-2}$ yr$^{-1}$, AM2.1: $-0.15$ W m$^{-2}$ yr$^{-1}$) and strong brightening thereafter (AM3: $0.35$ W m$^{-2}$ yr$^{-1}$, AM2.1: $0.36$ W m$^{-2}$ yr$^{-1}$) in both the models and in observations [Wild, 2009]. The strength and duration of East Asia’s clear-sky dimming make it an especially important region in which to understand the driving mechanisms of clear-sky SSR reductions.

A pertinent next question is: what is the primary driver of the trends in clear-sky SSR over East Asia produced by the models? Figures 3.2a and 3.2b show the results of the various ensemble simulations (described in Section 3.2.2) for the two models. The natural forcing experiments (NAT) show no significant trend in clear-sky SSR, nor do the WMGG. The AERO experiments, meanwhile, produce a trend in clear-sky SSR over East Asia of $-0.23$ W m$^{-2}$ yr$^{-1}$ in AM3 and $-0.49$ W m$^{-2}$ yr$^{-1}$ in AM2.1, demonstrating that anthropogenic
aerosols are indeed responsible for the majority of the all-forcing clear-sky SSR trends in both models, as previously postulated by many other studies [Wild 2012 and references therein]. The difference between the clear-sky dimming trends in AM3’s AERO and ALL_F runs results from the fact that the trends in sulfate and black carbon column burden are larger in the ALL_F runs than in the AERO runs. This is due to the difference in meteorology produced by the disparate forcings in the two runs, which produces different column burdens from identical emissions when applied to AM3’s fully interactive aerosols.

Aerosol-induced absorption contributes strongly to the reduction in clear-sky SSR (Figures 3.3a and 3.3b). Somewhat surprisingly, the two models produce almost identical increases in absorption (∼0.16 W m⁻² yr⁻¹), despite the many differences in their aerosol formulations. Since the overall reduction in clear-sky SSR is larger in AM2.1 than in AM3, the ratio of absorption change to clear-sky SSR change is smaller in AM2.1 (about one third) than in AM3 (about one half). This indicates that strong absorption is crucial for both models to simulate a clear-sky SSR trend over East Asia that is reasonably close to the range of observations. This can be contrasted with global mean absorption trends of 0.032 W m⁻² yr⁻¹ in AM3 and 0.029 W m⁻² yr⁻¹ in AM2.1, indicating that absorption is much stronger over East Asia than in the global mean.

3.3.2 Standalone radiative transfer calculations

Standalone radiative transfer calculations allow for a process-level analysis of the contribution of various aerosol characteristics to the modeled clear-sky SSR and absorption changes. The key results are shown in Table 3.2 for 1990 conditions minus 1970 conditions. AM3’s baseline configuration (AM3_IM) produces an annual mean surface solar radiation decrease between 1970 and 1990 of 6.9 W m⁻² for AM3’s aerosol concentrations, with an associated increase in atmospheric absorption of 4.3 W m⁻². When external (AM3_EM) rather than internal mixing is used, however, the clear-sky SSR decrease is only 5.6 W m⁻² and the absorption increase is only 2.2 W m⁻². The difference (i.e. between AM3_IM and AM3_EM) in the
Figure 3.3: The clear-sky SSR (left axis, black) and atmospheric absorption (right axis, red) anomalies are shown for (a) AM3 and (b) AM2.1. Increased absorption accounts for approximately one half of the decrease in SSR in both cases.

...
### Table 3.2: A summary of the standalone radiation radiative transfer calculation results.

Clear-sky SSR and absorption flux (W m\(^{-2}\)) values shown are for 1990 conditions minus 1970 conditions. The ‘nohygro’ refers to the versions of each experiment with hygroscopic growth disabled. Normalized absorption (10\(^{6}\) W kg\(^{-1}\)) refers to the 1970 to 1990 change in absorption per unit change in black carbon column burden.

<table>
<thead>
<tr>
<th>Run Description</th>
<th>AM3_IM</th>
<th>AM3_EM</th>
<th>AM2_IM</th>
<th>AM2_EM</th>
</tr>
</thead>
<tbody>
<tr>
<td>SSR (W m(^{-2}))</td>
<td>AM3 aerosol climatology with internal mixing (baseline)</td>
<td>AM3 aerosol climatology with external mixing</td>
<td>AM2.1 aerosol climatology with internal mixing</td>
<td>AM2.1 aerosol climatology with external mixing (baseline)</td>
</tr>
<tr>
<td>_nohygro</td>
<td>-6.9</td>
<td>-5.6</td>
<td>-10.3</td>
<td>-8.5</td>
</tr>
<tr>
<td></td>
<td>_nohygro</td>
<td>-5.2</td>
<td>-4.1</td>
<td>-8.1</td>
</tr>
<tr>
<td>Absorption (W m(^{-2}))</td>
<td>4.3</td>
<td>2.2</td>
<td>6.5</td>
<td>3.1</td>
</tr>
<tr>
<td>_nohygro</td>
<td>3.8</td>
<td>2.3</td>
<td>6.0</td>
<td>3.3</td>
</tr>
<tr>
<td>Scattering (W m(^{-2}))</td>
<td>2.6</td>
<td>3.4</td>
<td>3.8</td>
<td>5.4</td>
</tr>
<tr>
<td>_nohygro</td>
<td>1.4</td>
<td>1.8</td>
<td>2.1</td>
<td>3.0</td>
</tr>
<tr>
<td>Norm. Abs. (MWkg(^{-1}))</td>
<td>8.0</td>
<td>4.1</td>
<td>7.2</td>
<td>3.5</td>
</tr>
<tr>
<td>Column burden (10(^{-6}) kg m(^{-2}))</td>
<td>Black Carbon</td>
<td>0.541</td>
<td>0.541</td>
<td>0.899</td>
</tr>
<tr>
<td></td>
<td>Sulfate</td>
<td>6.37</td>
<td>6.37</td>
<td>12.4</td>
</tr>
</tbody>
</table>
Within the context of the models’ formulation, increased absorption with internal mixing can be explained by the nonlinear relationship between the single scattering albedo of an internally mixed aerosol and its black carbon volume fraction [e.g. Ackerman and Toon 1981, Chylek and Wong 1995, Jacobson et al. 2001, Liao and Seinfeld 2005, Stier et al. 2007].

The radiation code represents internal mixing by calculating refractive indices for the mixed aerosol that are equal to the volume-weighted average of those of black carbon and sulfate. As sulfate volume fraction decreases (i.e. as black carbon volume fraction increases) in the mixed aerosol, single scattering albedo decreases nonlinearly. For the large sulfate to black carbon volume ratios typical of most regions, a 0.1 decrease in sulfate fraction results in a single scattering albedo decrease of more than 0.1. This suggests that the amount of absorption produced by an aerosol population will be strongly sensitive to even small concentrations of black carbon when internal mixing is represented [e.g. Ackerman and Toon 1981], and that absorption will be sensitive to the ratio of BC to sulfate, as discussed in Section 3.4.3.

The absolute change in both clear-sky SSR decrease and absorption increase between mixing states is much larger with AM2.1’s aerosol climatology than with AM3’s. For example, the difference between AM2.IM and AM2.EM absorption is $\sim 3.4 \, \text{W m}^{-2}$, while it is only $\sim 1.9 \, \text{W m}^{-2}$ between AM3.IM and AM3.EM. Strikingly, AM2.1 has almost twice as large of an increase in sulfate and black carbon column burden between 1970 and 1990 as AM3 does ($1.24 \times 10^{-5} \, \text{kg m}^{-2}$ versus $6.37 \times 10^{-6} \, \text{kg m}^{-2}$ of sulfate, respectively, and $8.99 \times 10^{-7} \, \text{kg m}^{-2}$ versus $5.41 \times 10^{-7} \, \text{kg m}^{-2}$ of black carbon, respectively). This suggests that the difference between the two models’ sensitivity to change in mixing state can be explained largely by the difference in aerosol column burden between the two models. Further confirmation of this result and consideration of its implications are presented in Section 3.4.

We also conduct standalone radiative transfer calculations for all previously discussed configurations with hygroscopic growth disabled (i.e. the optical properties of an aerosol is held constant at all relative humidities), the results of which are shown in Table 3.2. Note that the AM3 runs contain the 97% relative humidity cap described in Section 3.2.1 while
there is no capping when AM2.1’s aerosol climatology is used. We do not investigate the impact of the use of relative humidity capping on the modeled radiation, though it has been shown to be nonnegligible [Ginoux et al., 2006].

In both models, with either mixing state implemented, disabling hygroscopic growth decreases the clear-sky SSR reduction between 1970 and 1990. The degree to which this decreased clear-sky dimming comes from decreased absorption versus decreased scattering, though, seems to depend on the mixing state. In the externally mixed case, the modeled absorption increase between 1970 and 1990 seems to be relatively insensitive to whether or not hygroscopic growth is disabled. However, with internal mixing, both models absorb more when hygroscopic growth is enabled. Additionally, the decrease in clear-sky dimming due to disabled hygroscopic growth is larger than the decrease in absorption even in the internally mixed case, indicating that disabling hygroscopic growth also reduces scattering.

Why does hygroscopic growth only result in more absorption if internal mixing is implemented? The implementation of internal mixing makes any mixed black carbon hydrophilic via its inclusion with hydrophilic sulfate, enabling hygroscopic growth of otherwise hydrophobic BC. Hygroscopic growth of the mixed aerosol will then increase the radiation incident on the aerosol due to the focusing effects of the dielectric medium (i.e. the liquid water) [e.g. Danielson et al., 1969]. Because this mixed aerosol is partially absorbing, hygroscopic growth can significantly increase its absorption cross-section [e.g. Chylek et al., 1984; Chylek and Wong, 1995]. Absorbing black carbon only grows hygroscopically in the model when internally mixed with hydrophilic sulfate, and will thus only produce increased absorption from dielectric focusing (represented in the model by an effective refractive index approximation) in the presence of internal mixing.
3.4 Discussion

3.4.1 Compensation between aerosol amount and mixing state

The sensitivity of the models’ clear-sky SSR and absorption to the mixing state of the aerosol, shown in the standalone radiative transfer calculations, provides an explanation for the similarity in the trends in absorption and clear-sky SSR over East Asia that the full models produce, despite significant differences in aerosol characteristics. The change in mixing state, from external to internal, that occurred in the transition from AM2.1 to AM3 tends to increase absorption and decrease clear-sky SSR. This is evinced by the fact that when AM3’s aerosol climatology is run with external mixing, it produces a much weaker signal than AM2.1’s (Table 3.2). However, the change in the aerosol column burdens, from which the models’ number concentrations are proportionally derived according to a lognormal distribution, operates in the opposite direction. Indeed, as discussed in Section 3.3, AM2.1 contains approximately twice as large of an increase in both types of aerosol between 1970 and 1990. This indicates a compensation of effects between aerosol amount and aerosol mixing state that at least partially explains the similarity in the models’ absorption trends.

This compensation can be probed quantitatively by calculating the absorption per unit aerosol produced by each model. If this value, which we term normalized absorption, converges for the two models when they are run in the same mixing state compared to the normalized value in different mixing states, we can argue that there is a compensation between aerosol amount and mixing state in the two models. We choose to normalize by the black carbon burden, since black carbon is relevant to absorption in both the externally and internally mixed case, while sulfate is only relevant in the internally mixed case. However, the ratio of black carbon to sulfate in the two models is very similar, and thus normalization by black carbon can be seen as a proxy for normalization by sulfate as well. The normalized absorption can be calculated as in the schematic equation 3.1 below. The absorption (Abs) with (aero) and without (no_aero) aerosol is calculated by turning aerosol shortwave effects
on and off, respectively, in the standalone radiation code. The ∆ refers to the change over the time period 1970-1990.

\[ \text{Normalized Abs.} = \frac{\Delta \text{Abs}_{\text{aero}} - \Delta \text{Abs}_{\text{no-aero}}}{\Delta \text{BC column burden}} \] (3.1)

The results of this calculation are shown in Table 3.2. From the normalized absorption, it is clear that the absorption values only converge when the effects of aerosol amount and mixing state are both accounted for. In the full AGCMs, the absorption trend is an identical \( \sim 0.16 \text{ W m}^{-2} \text{ yr}^{-1} \) in both AM2.1 and AM3. When the absorption change between 1970 and 1990 in the standalone calculation is normalized by the amount of aerosol in each model, however, the value is significantly different for the two models (8.0 MWkg\(^{-1}\) for AM3.IM versus 3.5 MWkg\(^{-1}\) in AM3.EM). However, when the two models are run in the same mixing state, the normalized absorption values again converge (8.0 MWkg\(^{-1}\) and 7.2 MWkg\(^{-1}\) for AM3.IM and AM2.IM, respectively, and 4.1 MWkg\(^{-1}\) and 3.5 MWkg\(^{-1}\) for AM3.EM and AM2.EM, respectively), indicating that the effects of the aerosol amount and mixing state changes mask each other in the base state of the models.

It should be noted that this cancellation of effects is a byproduct of the shift in aerosol formulation between AM2.1 and AM3 and will not necessarily hold for all regions and model formulations. Although the shift from external to internal mixing does have aerosol lifetime impacts that may contribute to the reduced aerosol burden, this does not account for the entirety of the difference in column burden between the two models, as discussed in Section 3.4.3.

### 3.4.2 Potential effects of seasonality in aerosol amount

The standalone radiative transfer calculations analyzed here provide useful mechanistic insight into the impact of mixing state and hygroscopic growth treatment in aerosol schemes, and the framework developed in this study allows analysis of many other aerosol charac-
Figure 3.4: Seasonal variation of black carbon column burden is shown for AM2.1 (black) and AM3 (blue) normalized by each model’s annual mean value. Also shown is the top-of-atmosphere (TOA) downwelling shortwave radiation (red) over East Asia in W m$^{-2}$ for reference. The two models have significantly different black carbon seasonalities, which lead to different temporal correlations with the downwelling radiation.

Another factor we consider here is the impact of the models’ seasonal cycle of aerosol concentrations on their annual mean shortwave radiative effects. One might expect the annual mean SSR reduction or shortwave absorption induced by an aerosol population to be dependent on how well the seasonal distribution of aerosol correlates with the seasonal distribution of TOA shortwave radiation availability (i.e. insolation). One might also expect this effect to be present in diurnal averaging [e.g. Kassianov et al., 2013].

The models have different seasonal concentrations in aerosol (due to the distinct emissions inventories and interactivity of aerosols), which have different temporal correlation with the seasonal TOA insolation distribution (Figure 3.4). We can hypothesize an expected annual mean sensitivity to this correlation using a simplistic calculation. The ability of a given seasonal distribution of black carbon over East Asia to interact with solar radiation can be calculated by comparing an annual area-averaged BC concentration ($BC'$) that has been
weighted by the area-averaged seasonal insolation ($S$) with the unweighted annual area-averaged concentration ($BC$). This provides a dimensionless, ordinal measure of the clear-sky radiative “potency” ($\Pi$) of a given aerosol seasonality at interacting with shortwave radiation in the annual mean and can be used for first-order comparison of the impact of the seasonal aerosol distribution on the annual mean shortwave values in each model. A larger potency suggests stronger interaction between the BC concentration and insolation. The calculation is as follows for monthly values, $t = \{1, 2, 3, \ldots, 12\}$:

$$BC' = \frac{\sum_{t=1}^{12} BC(t) \times S(t)}{\sum_{t=1}^{12} S(t)} \quad \rightarrow \quad \Pi = \frac{BC'}{BC}$$

(3.2)

Maximum and minimum potency will occur when all black carbon occurs in the month of maximum insolation (July, for East Asia) and minimum insolation (January, for East Asia), respectively. These upper and lower bounds correspond to $\Pi_{\max} = 0.11$ and $\Pi_{\min} = 0.052$. The calculated potency values for AM2.1 and AM3 are 0.086 and 0.084, respectively, indicating moderate potency. The similarity of these values (less than 2.5% difference) suggests that the difference in the models’ seasonal distribution has minimal impact on the annual mean values produced. More analysis via standalone radiation transfer calculations would be needed, however, to confirm this behavior in the context of absorption.

### 3.4.3 Evaluation of aerosol characteristics

The results of this study suggest that both AM2.1 and AM3 are capable of producing trends in clear-sky SSR comparable with observational proxy estimates, but that the aerosol processes responsible are quite different. Given this dichotomy, which model’s aerosol configuration is more physical? Answering this question is vital for improved aerosol modeling, but is made less tractable by persistent uncertainty in many aerosol processes and in aerosol emissions, partially driven by a lack of aerosol observations that are both global and detailed.
We nonetheless attempt to comment on the relative physicality of various relevant aspects of the two models’ aerosol characteristics. Our discussion highlights the sensitivity of models’ absorption to widely varying characteristics, such as the mode of internal mixing used and the ratio of black carbon to sulfate.

Both observational and modeling studies have found that the majority of aerosol populations will be largely internally mixed after aging [e.g. Andreae et al., 1986; Pósfai et al., 1999; Jacobson et al., 2001]. Within modeling studies, however, the representation of the internal mixture can significantly impact the aerosols’ optical properties and radiative perturbation [e.g. Ackerman and Toon, 1981; Jacobson, 2000; Bond et al., 2006]. The uniform mixing scheme used in this study, in which the optical properties of the mixed aerosol are calculated for black carbon and sulfate diffused into a homogeneous aerosol, is one representation. Others include coated core representations, in which optical properties are computed for a black carbon core coated with a sulfate layer [e.g. Stier et al., 2006b; Matsui et al., 2013; Yu et al., 2013], and observationally based parameterizations of internally mixed aerosol properties that do not explicitly assume a physical configuration of the internally mixed particle [e.g. Balkanski et al., 2010]. It is, therefore, useful to place our results in the context of modeling studies that use other internal mixing formulations, albeit for other regions or time periods.

Virtually all manner of internal mixing of black carbon with nonsorbing substances is expected to yield a higher absorption per unit mass of black carbon relative to the aerosols in their externally mixed state [e.g. Chylek et al., 1984].

Matsui et al. [2013] analyze the radiative effects of coated core mixing schemes of varying complexity applied to year 2009 aerosols over East Asia in a standalone radiation calculation, and find lower tropospheric heating rates due to internally mixed aerosols that are comparable to those produced by our 1990 standalone radiation calculations (not shown). Balkanski et al. [2010] analyze the effect of internal vs. external mixing schemes on year 2000 transportation sector aerosols on a global scale using multiple chemical transport models and radiation codes. They represent internal mixing by imposing a 50% absorption increase on
any hydrophilic black carbon in their model, with no dependence on sulfate burden. This formulation makes direct comparison with our results difficult. However, Balkanski et al. [2010] see between a 31% and 47% increase in positive radiative forcing (i.e. absorption) when their internal mixing scheme is used.

Stier et al. [2006b], similarly, conduct idealized experiments using a standalone radiation calculation to determine the change in absorption due to coated core internal mixing versus external mixing in a black carbon and sulfate population. They impose an aerosol population with a sulfate to black carbon mass ratio of 10:1, and find that it produces a 47% increase in absorption when the coated core internal mixing rather than external mixing is utilized. Our results, meanwhile, find an approximately 100% increase in absorption when uniform internal mixing is used on either AM3 or AM2.1’s aerosol burden over East Asia (Table 3.2). This result is quantitatively consistent with that of Stier et al. [2006b], as the mass ratio of sulfate to black carbon in AM3 and AM2.1 is closer to 5.5:1 (Table 3.2). Additionally, Jacobson [2000] found that the use of uniform mixing representations produce a more than 40% increase in global direct radiative forcing from black carbon over coated core representations, so it is unsurprising that our models simulate a stronger increase in absorption due to internal mixing.

The above comparisons highlight the importance, particularly within the context of our volume-weighted uniform mixing scheme (see Section 3.3.2), of the relative burdens of black carbon and sulfate in our region of interest. Matsui et al. [2013] and Stier et al. [2006a] both find that internal mixing increases absorption per unit mass of black carbon, but decreases the black carbon column mass burden due to changes in aerosol lifetime. Thus, the relative abundance of these species may be highly dependent on transport and aging of emissions. Because our mixing perturbation experiments are conducted within a standalone radiative transfer calculation, we do not capture the effects of mixing state on transport or lifetime. However, a comparison between the difference in emissions and the difference in concentrations between AM2.1 and AM3 yields some insight into the role of transport and
lifetime in producing the black carbon and sulfate concentrations to which the standalone radiative transfer calculations are applied.

In our standalone radiative transfer calculations, the change in sulfate column burden between 1970 and 1990 is approximately 2 times larger in AM2.1 than in AM3. However, an analysis of the emissions inventory used in the MOZART simulations that provide AM2.1 with its column burdens and the inventory directly used in AM3 reveals that the change in sulfate emissions between 1970 and 1990 is only 1.3 times higher in AM2.1 than in AM3. Similarly, the change in black carbon column burden between 1970 and 1990 is approximately 1.7 times larger in AM2.1 than in AM3. However, the change in black carbon emissions is, in fact, 1.4 times higher in AM3 than in AM2.1. This indicates that the difference in the black carbon and sulfate column burdens in AM3 and AM2.1 results from a combination of differences in emissions, microphysical or meteorological impacts on lifetime, and transport of emissions. Because differences in input emissions, rather than solely model physics, contribute to the difference in column burden between the two models, it is not guaranteed that other regions will experience the cancellation of effects discussed in section 3.4.1. Even in the absence of emissions differences, the transition in model physics from external to internal mixing may not necessarily result in the burden reduction needed to produce the compensation seen, highlighting the importance of mechanistic analysis such as that conducted in this work.

The models’ aerosol characteristics perform well in relation to other modeling studies, but how do they perform in an observational context? Jacobson [2000] points out that because black carbon is generally insoluble and thus difficult to diffuse into a homogenous aerosol, the uniform mixing representation likely constitutes an overestimation of absorption due to realistic internal mixing. The uniform internal mixing used in AM3 may, therefore, bias that model toward a more pronounced absorption increase than is realistic [Bond et al., 2006; Jacobson et al., 2001]. Comparisons between observations and the CMIP3-generation aerosol models (including AM2.1), however, indicate that those models underestimated BC
absorption [Koch et al., 2009]. Bond et al. [2013] also suggest that many current generation models continue to underestimate black carbon absorption by a factor of three even when emissions biases are accounted for, suggesting that AM3’s strong absorption per unit black carbon may be warranted by observational estimates. Analyzing the relative contributions of diffuse and direct shortwave flux at the surface in models versus observations, as done by [Freidenreich and Ramaswamy, 2011], may provide one means of further constraining realistic absorption.

AM2.1’s surface sulfate and black carbon concentrations were found by [Ginoux et al., 2006] to be lower than observations over East Asia during the period 1996-2000, but within a factor of two. However, it is important to note that surface concentration comparisons may not be transferrable to column burden, which is more relevant for total shortwave attenuation. AM2.1 is known to have an overly vertically diffuse aerosol column over East Asia [Koffi et al., 2012], which would prime the model to underestimate surface concentration while still maintaining a representative or even overestimated column burden. The literature is largely inconclusive on model over- or underestimation of aerosol concentrations over East Asia, partially because of challenges associated with difficult-to-track regional sources. Small but strongly emitting Asian industries, like brick kilns and coking, are often not included in bottom-up emissions inventories, making Asia particularly prone to emissions underestimations. Bond et al. [2013] suggest, nonetheless, that up to a factor of 4 increase in black carbon burdens over those found in current models may be warranted.

### 3.4.4 Limitations of observational comparison

Although the discussion in Section 3.4.3 suggests that AM3’s more complex aerosol parameterization constitutes an improvement over AM2.1’s more simplistic formulation, further advances in aerosol representations in GCMs will require better field measurements against which to validate them [Ginoux et al., 2006; Bond et al., 2013; Allen et al., 2013] and [Ruckstuhl and Norris, 2009], among others, showed that differences in the historical aerosol
emissions used in models cannot by themselves explain divergences in clear-sky dimming trends; significant divergence in aerosol physics remains in the absence of sufficient observations for validation [Koch et al., 2009]. As evinced by the results of this study, observations of aerosol radiative effects alone are not sufficient to constrain aerosol physics, as multiple realizations can produce plausible values. Theoretically, however, multiple, independent observations of different variables—for example, spectral discrimination of aerosols’ radiative effects and diurnal variability of the correspondence between aerosol optical depth and SSR—could prove useful.

While observational datasets provide useful context for model simulation, considerable uncertainty in both models and observations obfuscates direct model/observation comparison. For instance, many studies have analyzed the lack of interdecadal variability in modeled clear-sky SSR, compared to observational proxies [e.g. Ruckstuhl and Norris, 2009; Dwyer et al., 2010; Wild and Schmucki, 2010; Allen et al., 2013]. This should perhaps be unsurprising, however, given the low temporal resolution of models’ aerosol climatologies. In AM2.1, monthly mean aerosol concentrations are only input from MOZART calculations every ten years. Aerosol concentrations between those calculated values are estimated by linear interpolation [Ginoux et al., 2006]. AM3’s aerosol emissions are, likewise, only input directly from the emissions inventory at approximately decadal intervals, with linear interpolation in between. This linear interpolation and coarse temporal resolution will significantly dampen variability of aerosol-driven values in the models.

The large interannual variability seen in the clear-sky proxy data, meanwhile, may be an artifact of the cloud removal process used [Norris and Wild, 2009]. Other studies evaluating clear-sky SSR, but using observations only during clear-sky periods rather than cloud removal, do not find comparable interannual variability, albeit over different regions [e.g. Jing and Cess, 1998; Liepert, 2002]. Furthermore, the interannual aerosol variability necessary to produce the clear-sky SSR variability seen in the dataset of Allen et al. [2013] is not supported by annual mean emissions estimates for this region [Lu et al., 2011].
In addition to the identified observational deficiencies discussed in Section 3.2.4, other issues favor focusing on model simulation rather than model/observation comparison. While Allen et al. [2013] are conscientious in applying the stringent quality standards needed to avoid contamination by possible system deficiencies, this leaves them with only six observation sites. Wild [2009] highlights that small sample sizes are more susceptible to bias from the frequent location of sites near urban centers. Norris and Wild [2009] calculate East Asian clear-sky SSR trends using the same method as Allen et al. [2013], but with a more densely sampled set of observation sites with greater representation of the continental interior, and find different trend values, suggesting a sensitivity to the sampling choices made. We have chosen, therefore, to focus primarily on physical analysis of the model results, using observations primarily for context.

3.5 Conclusions

Our results demonstrate that it is possible to obtain clear-sky SSR reductions over East Asia that are comparable to observed trends via very different combinations of aerosol mechanisms, and that these reductions are strongly driven by increased aerosol absorption. Both the AM2.1 and AM3 AGCMs used in this study capture the decreasing trend in clear-sky SSR over East Asia from 1960 to the mid-2000s, though AM2.1’s trend is larger than AM3’s. The two models contain large, virtually identical increases in absorption over this period, however, despite having significantly different aerosol characteristics, including differences in mixing state, column burden, and aerosol interactivity with model meteorology.

Our analysis using the models’ standalone radiation module reveals that the difference in mixing state and aerosol amount between the two models act on the absorption and clear-sky SSR values in opposing directions, resulting in a compensation of effects that largely explains the similarity in absorption increase between the two models. AM3’s internal mixing increases the absorption produced by its smaller change in black carbon column burden, while
AM2.1 compensates for the smaller normalized absorption induced by its external mixing scheme with a change in black carbon column burden that is a factor of 2 larger than AM3’s. It is important to note that this compensation is not a necessary outcome of the transition from external to internal mixing, making our mechanistic analysis a particularly important undertaking. We find that the hygroscopic growth of internally mixed aerosol in AM3 also acts to enhance the absorption that the model’s aerosol population produces. We also briefly investigate the impact of the seasonality of the aerosol concentrations in the two models on annual mean values, and find that it is largely negligible. The framework developed in this paper can be extended to study the impact of many other aerosol characteristics that may be important for determining the relative contribution of absorption to aerosol-driven clear-sky solar dimming.

Given the climate impacts of aerosol absorption and the sensitivity of that absorption to subtle changes in aerosol characteristics discussed in this study, in-depth mechanistic analyses such as those contained in this paper will be vital to constraining the climate response to aerosol-driven clear-sky solar dimming and for assessing model fidelity. In addition to the single variable dependencies discussed here, cross-correlations between different aerosol characteristics may also exist. For example, seasonal and vertical variations in relative humidity may lead to stronger hygroscopic growth depending on the seasonality and vertical distribution of the aerosols. The standalone radiative transfer calculation framework developed in this paper provides an ideal tool for analyzing these effects in future studies.

This work highlights the important role that aerosol absorption plays in driving clear-sky solar dimming over East Asia, especially in the more recent incarnation of the GFDL model. AM3’s aerosol parameterization contains several advances in the complexity of its aerosol representation, and many of its aerosol characteristics are considered to be more physically realistic than AM2.1’s [Donner et al., 2011]. The particularly strong contribution of absorption to the clear-sky dimming trend produced by AM3, therefore, has many important implications for the climate response that can be expected from clear-sky solar
dimming over East Asia, especially as aerosol emissions evolve in the future. Given the strong regional impacts of the surface-atmosphere radiation dipole that aerosol absorption can impose [Ramanathan et al., 2001], it will be critical to establish greater confidence in the relative contribution of absorption to clear-sky solar dimming values. In the next chapter, we probe the combined and competing effects of this surface-atmosphere radiation dipole on East Asian regional climate to elucidate the implications of uncertain past, present, and future absorption-driven dimming by aerosols.
Chapter 4

Competing Atmospheric and Surface-Driven Impacts of Absorbing Aerosols on the East Asian Summertime Climate

4.1 Introduction

East Asia receives over half of its annual precipitation during the summer months of June, July, and August. The summertime maximum in solar radiation warms the East Asian continent more rapidly than the adjacent ocean due to the land’s lower heat capacity, setting up temperature and pressure gradients that drive on-shore flow of moisture and ascending atmospheric motion over the continent [e.g. Webster 1987]. This land-sea thermal contrast, combined with orographic forcing and seasonal shifts in the subtropical westerly jet, produces a precipitation maximum that peaks in June and July over East Asia, known collectively as the East Asian Summer Monsoon (EASM) [e.g. Murakami 1981 Chen and Bordoni 2014].
Superimposed on this climatological picture are changes in the radiative environment due to anthropogenic emissions over the last several decades. Aerosol concentrations over the East Asian subcontinent decreased the annual mean clear-sky surface solar radiation by 4.3 W m\(^{-2}\) decade\(^{-1}\) from the 1960s to the 2000s \cite{Allen2013}. This “solar dimming” counteracts the climatological land-sea thermal contrast by cooling the land surface more rapidly than the adjacent ocean, both due to the land’s lower heat capacity and because the aerosol is more strongly concentrated over land than over ocean \cite[e.g.][]{Guo2013, Wang2015}. Several modeling studies have suggested that this radiative perturbation from the large increase in anthropogenic aerosol emissions over East Asia could be the primary contributor to a weakening in the EASM since the middle of the twentieth century \cite[e.g.][]{Song2014, Wang2015}.

Recent work, however, has demonstrated that as much as half of the observed clear-sky reduction in surface solar radiation is due to the absorption of solar radiation within the atmospheric column by absorbing aerosols, as opposed to the reflection of that radiation back to space through scattering. \cite{Persad2014} identify in general circulation model (GCM) simulations that aerosol absorption under clear-sky conditions increased over East Asia by \(\sim 1.6\) W m\(^{-2}\) decade\(^{-1}\) from the 1960s to the 2000s, robust across different model aerosol formulations. Consequently, during the 2000s, anthropogenic aerosols (both scattering and absorbing) reduced East Asian summertime clear-sky surface solar radiation by \(\sim 20\) W m\(^{-2}\) and increased atmospheric absorption by \(\sim 10\) W m\(^{-2}\). This vertical radiative dipole of atmospheric heating and surface dimming might be expected to have a substantially different impact on regional summertime circulation and precipitation than surface dimming or atmospheric heating on its own, but the effects of this dipole have not been cleanly decomposed before now.

Attention is often focused on the impact of atmospheric heating from aerosol absorption on regional dynamics due to the extensively discussed “semi-direct effect” of absorbing aerosols \cite[e.g.][]{Hansen1997, Koch2010}, whereby the radiative heat-
ing from direct aerosol absorption changes the thermodynamic and dynamical environment for cloud formation. Studies show that this atmospheric heating can either invigorate or suppress convection, depending on the altitude of the heating relative to the climatological cloud [e.g. Johnson et al., 2004; Feingold, 2005; Persad et al., 2012]. Depending on the convective environment, atmospheric heating on its own has been shown to induce strong rising atmospheric motion regionally that promotes monsoonal moisture flux and convective precipitation [Chung et al., 2002; Wang, 2004; Erlick et al., 2006; Meehl et al., 2008].

However, the same absorption process via which these aerosols heat the atmosphere also reduces the shortwave radiation at the surface. On its own, solar dimming reduces both land surface temperature and the surface energy available for latent and sensible heat fluxes that help drive convection and precipitation [e.g. Roeckner et al., 1999; Huang et al., 2007]. In a global mean sense, solar dimming from purely-scattering aerosols has been shown to spin-down the hydrological cycle [Ramanathan et al., 2001].

It is unclear how these two radiative effects will interact in a given region and what their relative impact on regional climate will be. Depending on the degree of coupling between the surface and atmosphere, atmospheric heating from absorption may or may not be communicated to the surface, affecting whether or not the atmospheric heating is able to counteract the surface temperature effects of the solar dimming [Ramanathan and Carmichael, 2008]. In the case of weak coupling, the combination of surface cooling and atmospheric heating from aerosol absorption can reduce surface-air gradients that drive evaporation, thus limiting the moisture available for rainfall [Ramanathan et al., 2005]. Conversely, with strong coupling, atmospheric heating may be efficiently translated to the surface, counteracting surface cooling from the shortwave attenuation.

Given these uncertainties and the large absorbing aerosol concentrations in East Asia, a robust understanding of the separate, combined, and competing effects of atmospheric heating and surface dimming from aerosol absorption on the East Asian summertime climate is of crucial importance, but until now has been limited. The relative effect of absorbing black
carbon and scattering sulfate aerosols on East Asian climate has been studied previously [e.g. Huang et al., 2007; Randles and Ramaswamy, 2008; Guo et al., 2013; Jiang et al., 2013; Wang et al., 2015], but black carbon representation in climate models is known to be underconstrained [Bond et al., 2013]. Separate analysis in the literature of surface dimming and atmospheric heating in monsoonal systems has shown substantially different impacts on regional climate [e.g. Chung et al., 2002; Roeckner et al., 1999]. However, the coupled effects of the two may be quite different than the sum of the parts. A holistic analysis of the separate and combined impacts of surface dimming and atmospheric heating is thus a crucial and missing element of a full understanding of absorbing aerosol impacts on the EASM and East Asian summertime climate in general.

We here isolate and analyze the surface- vs. atmosphere-driven impacts of aerosol absorption on East Asian summertime climate, using a combination of realistic and idealized forcing simulations in a state-of-the-art climate model, to address the following questions. What are the competing effects on the EASM of increased atmospheric absorption and decreased surface solar radiation due to regional aerosol emissions over the last several decades? Does the atmospheric heating from aerosol absorption enhance or counteract the circulation effects of aerosol-driven solar dimming on the EASM? Which wins out, and why? These are questions of interest both to present day understanding of the forcers of East Asian climate variability and to our predictive ability to understand how future changes in aerosol characteristics over East Asia will affect its projected regional climate.

4.2 Methods

We conduct simulations using both realistic historical aerosol emissions and idealized perturbations in the Geophysical Fluid Dynamics Laboratory’s (GFDL) AM3 Atmospheric General Circulation Model (GCM), which is described in detail in Chapter 3. We here highlight aspects of AM3 of particular relevance to this chapter. Crucially for this analysis, AM3 has
been found to outperform other models in its CMIP generation in the simulation of twentieth century clear-sky solar dimming over East Asia [Allen et al., 2013] via a substantial contribution from aerosol absorption [Persad et al., 2014], making it a reasonable tool with which to investigate the competing effects of aerosol dimming and absorption on the EASM. The GFDL CM3 model suite has been previously used to study East Asian summer climate as part of the CMIP5 model suite [Salzmann et al., 2014; Li et al., 2015]. Over East Asia, AM3 exhibits relatively small biases in comparison with observations in monsoon-critical parameters such as 500 mb geopotential height, surface air temperature, and precipitation [Donner et al., 2011].

We first derive realistic regional aerosol effects from an ensemble of historical single forcing simulations with time-varying sea surface temperature (SST) and sea ice prescribed from the Hadley Centre Sea Ice and Sea Surface Temperature 1 (HADISST1) observational data set [Rayner et al., 2003]. The simulations are: (1) AERO_C, a five-member ensemble of experiments with historically varying anthropogenic aerosol emissions over China (as defined by national boundaries; roughly 20°–50° N and 75°–125° E), but all other anthropogenic emissions (i.e. well-mixed greenhouse gases and ozone, plus all aerosol emissions outside of China) fixed at preindustrial (1860) values; and (2) FIXED, a five-member ensemble with all anthropogenic emissions fixed at preindustrial (1860) values. The realistic regional aerosol signal, hereafter referred to as “Realistic Aerosol”, is calculated as the 1981-2000 average ensemble-mean values from AERO_C minus FIXED.

The realistic regional aerosols absorb and scatter incoming shortwave radiation, resulting in both absorption- and scattering-driven surface dimming and absorption-driven atmospheric heating (Section 4.4). We next use the aerosol radiative effects from the Realistic Aerosol signal to construct a series of idealized forcing simulations that allow us to isolate the effects of these different components of aerosols’ radiative perturbation on East Asian summertime climate. The control simulation for these is a historical simulation (ALL) with time-varying sea surface temperature and sea ice as in FIXED, but with all natural and an-
Table 4.1: Construction and key characteristics of each of the four experiments are summarized. Each experiment name refers to the signal derived from the difference between the given perturbation simulation and control simulation averaged over 1981-2000, and captures the described key characteristics.

<table>
<thead>
<tr>
<th>Experiment Name</th>
<th>Realistic Aerosol</th>
<th>Pure Dimming</th>
<th>Pure Heating</th>
<th>Pure Absorption</th>
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<tr>
<td><strong>Control Simulation</strong></td>
<td>FIXED: -SST and sea ice prescribed from HADISST1 observational dataset -All natural and anthropogenic forcers fixed at 1860 values -Run from 1870–2003 -Five-member ensemble average</td>
<td>ALL: -SST and sea ice prescribed from HADISST1 observational dataset -All natural and anthropogenic forcers varying according to historical values -Run from 1980–2000 -Single member</td>
<td>ALL: -SST and sea ice prescribed from HADISST1 observational dataset -All natural and anthropogenic forcers varying according to historical values -Run from 1980–2000 -Single member</td>
<td>ALL: -SST and sea ice prescribed from HADISST1 observational dataset -All natural and anthropogenic forcers varying according to historical values -Run from 1980–2000 -Single member</td>
</tr>
<tr>
<td><strong>Perturbation Simulation</strong></td>
<td>AERO:C: -SST and sea ice prescribed from HADISST1 observational dataset -Anthropogenic aerosol emissions within China varying historically -All other natural and anthropogenic forcers fixed at 1860 values -Run from 1870–2003 -Five-member ensemble average</td>
<td>ALL: simulation with additional purely scattering optical depth with vertical structure: $\tau_d(P) = \Delta z \times 0.117 \left(\frac{P - 150}{P_0}\right)^2$ for $P \geq 150$ mb, 0 for $P &lt; 150$ mb -Imposed from 1980–2000 -Single member</td>
<td>ALL: simulation with additional shortwave heating rate with vertical structure: $T_{dtsw}(P) = \frac{\bar{S}}{T} e^{-\frac{P}{P_0}}$ for $P \geq 150$ mb, 0 for $P &lt; 150$ mb -Imposed from 1980–2000 -Single member</td>
<td>ALL: simulation with additional purely absorbing optical depth with vertical structure: $\tau_d(P) = \Delta z \times 0.019 \left(\frac{P - 150}{P_0}\right)^2$ for $P \geq 150$ mb, 0 for $P &lt; 150$ mb -Imposed from 1980–2000 -Single member</td>
</tr>
<tr>
<td><strong>Key Characteristics</strong></td>
<td>-Captures effects of all anthropogenic aerosols emitted in China -Contains microphysical aerosol indirect effects in stratiform and shallow cumulus clouds -Used to determine reasonable radiative perturbations for the idealized forcing simulations</td>
<td>-Captures isolated effects of surface dimming from Realistic Aerosol -Allows analysis of how surface dimming from an absorber affects the overall response to absorption; simulates the behavior of dimming from purely scattering aerosols -No microphysical aerosol indirect effects directly induced by perturbation</td>
<td>-Captures isolated effects of atmospheric heating from Realistic Aerosol -With Pure Dimming, allows quantification of how the overall response to absorbing aerosol is influenced by its atmospheric heating versus its surface dimming -No microphysical aerosol indirect effects directly induced by perturbation</td>
<td>-Captures combined effects of surface dimming and atmospheric heating from absorbing aerosols in Realistic Aerosol -No microphysical aerosol indirect effects directly induced by perturbation</td>
</tr>
</tbody>
</table>
thropogenic forcers varying historically (see Donner et al. [2011] for inventories from which emissions are derived). Onto this ALL control simulation, each of the three idealized perturbations described below is imposed in the years 1980–2000 over Southeast China (22.5°–40° N and 100°–122.5° E)—the region of maximum aerosol emissions and radiative forcing in East Asia [Streets et al., 2013], as well as maximum climatological EASM precipitation. This is also the region of greatest observational coverage [Allen et al., 2013; Norris and Wild, 2009; Dwyer et al., 2010] and one in which AM3’s aerosol-driven solar dimming over East Asia has been verified against observational estimates [Persad et al., 2014]. The construction and goal of each idealized forcing simulation is schematically depicted in Figure 4.1, and the construction and characteristics of the realistic aerosol experiment and the three idealized forcing experiments are summarized in Table 4.1.

1. Pure Dimming: In this perturbation, a purely scattering optical depth (i.e. with an effective single scattering albedo of 1), scaled to reduce surface shortwave radiation by a

Figure 4.1: The 3 idealized forcing perturbations are schematically depicted. Each of the depicted perturbations is imposed over Southeast China (22.5°–40° N and 100°–122.5° E) during years 1980–2000 of an ALL control simulation, which contains historically varying anthropogenic and natural emissions and observationally prescribed sea surface temperature and sea ice.
level comparable to that produced by Realistic Aerosol, is imposed onto the ALL control. The scattering optical depth \( \tau_d \) within each model layer of path length or depth \( \Delta z \) is calculated according to the layer average pressure \( P \) via the following equation, which approximates the vertical structure of the Realistic Aerosol perturbation:

\[
\tau_d(P) = \begin{cases} \\
\Delta z \times \alpha_d \left( \frac{P-150}{P_0} \right)^{\beta_d}, & \text{for } P \geq 150 \text{ mb} \\
0, & \text{for } P < 150 \text{ mb}
\end{cases}
\]

where \( \alpha_d = 0.117 \) is a scaling constant designed to achieve a comparable magnitude of dimming as Realistic Aerosol, and \( \beta_d = 2 \) is a decay rate designed to approximate the vertical structure of the realistic aerosol perturbation.

The difference between the above perturbed simulation and the ALL simulation, averaged over 1981–2000, is hereafter referred to as the “Pure Dimming” signal, and isolates the effects of surface dimming from that of atmospheric heating. This allows us to analyze how surface dimming from an absorber affects the overall response to the absorption, or to simulate the behavior of dimming from purely scattering aerosols.

2. Pure Heating: In this perturbation, an idealization of the atmospheric shortwave heating profile from Realistic Aerosol is imposed onto the ALL simulation. The idealized shortwave heating rate \( Tdt_{sw} \) is calculated at each pressure level \( P \) to mimic the bottom-heavy vertical structure and magnitude of that produced by the realistic regional aerosol, and takes the exponential form:

\[
Tdt_{sw}(P) = \alpha_h \frac{S}{\bar{S}} \times e^{\beta_h P_0}
\]

where \( \alpha_h = 1 \) is a scaling constant designed to give a heating rate magnitude comparable to the realistic regional aerosol perturbation, and \( \beta_h = 1 \) is a decay rate designed to approximate the vertical structure of the realistic aerosol perturbation. The variable \( S \) is the time- and grid-varying solar flux in W m\(^{-2} \), \( \bar{S} \) is the regionally and annually averaged solar flux in W
m$^{-2}$, and $P_0 = 1000$ mb is the surface reference pressure. The perturbation heating rate is imposed at every time step, but is scaled according to diurnal and seasonal changes in solar zenith angle and solar flux, as reflected in the $\frac{S}{S}$ term. The resulting regional- and seasonal-mean heating rate are discussed and compared with that produced by Realistic Aerosol in Section 4.4.3.

The difference between the above perturbed simulation and the ALL simulation, averaged over 1981–2000, is hereafter referred to as the “Pure Heating” signal, and isolates the impact of atmospheric heating in the absence of any associated surface dimming. In conjunction with the Pure Dimming signal, this allows us to quantify the degree to which the overall response to an aerosol population that contains absorption is influenced by its atmospheric heating versus its surface dimming.

3. Pure Absorption: In this perturbation, a purely absorbing optical depth (i.e. with an effective single scattering albedo of 0) is scaled to produce atmospheric absorption comparable to that seen in the Realistic Aerosol case. As in the Pure Dimming case, the absorbing optical depth ($\tau_a$) within each model layer of depth ($\Delta z$) is calculated according to the layer average pressure ($P$) via the following equation, which approximates the vertical structure of the realistic regional aerosol perturbation:

$$
\tau_a(P) = \begin{cases} 
\Delta z \times \alpha_a \left(\frac{P-150}{P_0}\right)^{\beta_a}, & \text{for } P \geq 150 \text{ mb} \\
0, & \text{for } P < 150 \text{ mb}
\end{cases}
$$

where $\alpha_a = 0.019$ is the magnitude scaling constant, and $\beta_a = 2$ is the vertical decay rate (identical to the Pure Dimming vertical decay rate, $\beta_d$), both designed to approximate the realistic aerosol perturbation. The resulting shortwave heating rate is compared with that produced by the Realistic Aerosol and Pure Heating cases in Section 4.4.3.

The difference between the above perturbed simulation and the ALL simulation, averaged over 1981–2000, is hereafter referred to as the “Pure Absorption” signal. An absorbing aerosol will both heat the atmosphere by trapping shortwave radiation therein (as in the
Pure Heating simulation) and dim the surface by attenuating shortwave radiation aloft (as in the Pure Dimming simulation). This final simulation allows us to capture both effects acting in combination.

The Realistic Aerosol signal is primarily used as a means of determining reasonable radiative perturbations for the idealized forcing simulations. Due to differences in the formulation of the Realistic Aerosol simulations and the idealized forcing simulations, such as the presence of microphysical aerosol indirect effects in the former case and differences in the control climate, precise correspondence between the Realistic Aerosol case and the 3 idealized forcing signals is not expected. However, it is informative to view the Realistic Aerosol case, which contains both scattering and absorbing aerosols, as some combination of the Pure Absorption and the Pure Dimming signals (with the addition of microphysical aerosol indirect effects). The three idealized forcing simulations, meanwhile, are designed to be more straightforwardly intercompared. Comparison of the Pure Dimming and Pure Absorption signals allows quantification of how the response to absorption-driven and scattering-driven dimming differ. Comparison of all three idealized perturbation simulations allows the full decomposition of how absorption’s atmospheric and surface radiative perturbations operate in isolation and in tandem. Because of the radiative nature of the idealized perturbations’ parameterizations, microphysical indirect effects, such as the Twomey [1977] and Albrecht [1989] effects, are not excited directly by the idealized forcing perturbations. All cloud changes in the idealized forcing simulations will thus be thermodynamically (including semi-direct aerosol effects [e.g. Hansen et al. 1997]) or dynamically driven. All analysis is done on the mean of June, July, and August (JJA), which captures the main Meiyu-Baiu period of EASM rainfall [Sampe and Xie 2010, Chen and Bordoni 2014].
Table 4.2: Regional-mean radiative perturbations over East Asia (22.5°–40° N; 100°–122.5° E) are shown for each of the 4 simulations. The changes in all-sky ($\Delta{SSR_{all}}$) and clear-sky ($\Delta{SSR_{clr}}$) surface shortwave radiation, all-sky ($\Delta{Abs_{all}}$) and clear-sky ($\Delta{Abs_{clr}}$) atmospheric shortwave absorption, and regional all-sky ($\Delta{F_{TOA_{all}}}$) and clear-sky ($\Delta{F_{TOA_{clr}}}$) top-of-atmosphere effective radiative forcing are in units of W m$^{-2}$.

<table>
<thead>
<tr>
<th></th>
<th>Realistic Aerosol</th>
<th>Pure Dimming</th>
<th>Pure Heating</th>
<th>Pure Absorption</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta{SSR_{all}}$</td>
<td>-14.3</td>
<td>-18.6</td>
<td>-0.19</td>
<td>-14.6</td>
</tr>
<tr>
<td>$\Delta{SSR_{clr}}$</td>
<td>-13.5</td>
<td>-20.0</td>
<td>0.04</td>
<td>-16.2</td>
</tr>
<tr>
<td>$\Delta{Abs_{all}}$</td>
<td>6.87</td>
<td>-1.35</td>
<td>-0.49</td>
<td>17.3</td>
</tr>
<tr>
<td>$\Delta{Abs_{clr}}$</td>
<td>7.29</td>
<td>-2.76</td>
<td>0.10</td>
<td>19.1</td>
</tr>
<tr>
<td>$\Delta{F_{TOA_{all}}}$</td>
<td>-6.75</td>
<td>-19.8</td>
<td>2.04</td>
<td>6.57</td>
</tr>
<tr>
<td>$\Delta{F_{TOA_{clr}}}$</td>
<td>-4.35</td>
<td>-27.4</td>
<td>0.09</td>
<td>6.13</td>
</tr>
</tbody>
</table>

4.3 Results

4.3.1 Radiative effects of realistic and idealized aerosols

Table 4.2 summarizes the regional-mean clear-sky and all-sky surface dimming and atmospheric absorption produced by the various model perturbations, as well as their regional-mean top-of-atmosphere effective radiative forcing (defined as the net downward radiative flux at the top-of-the-atmosphere after atmospheric and land surface conditions have been allowed to equilibrate to the perturbation [Myhre et al., 2013]). Clear-sky values are calculated by the model’s radiative transfer code with clouds removed. Microphysical aerosol indirect effects are present in the Realistic Aerosol case and may influence differences between clear-sky and all-sky (i.e. cloud-permitting) values. In the idealized forcing simulations, because microphysical aerosol indirect effects are not in operation, differences between all-sky and clear-sky values are explained by either a) thermodynamically or dynamically driven cloud changes that reinforce or counteract the aerosols’ radiative interactions or b) differences due to cloud masking in the amount of radiation with which the aerosol is interacting in the clear-sky versus all-sky calculation.

The realistic regional aerosols reduce the solar radiation incident at the surface over East Asia by $\Delta{SSR_{all}} = -14.3$ W $m^{-2}$ in the 1981–2000 mean, consistent with the finding of
others in both models and observations [e.g. Norris and Wild 2009; Dwyer et al. 2010; Allen et al. 2013]. Note that, because our simulations only include aerosol emissions within China, they contain less aerosol over East Asia than the models and observations in the above referenced work, which include aerosols transported from both local and remote sources. The surface dimming signal is evident both in the presence and in the absence of cloud cover, and is driven almost equally by scattering and by absorption of shortwave radiation by aerosols within the atmospheric column, consistent with the findings of Persad et al. 2014. The similarity of the clear-sky and all-sky SSR changes should not be interpreted as an absence of aerosol-cloud interactions. Aerosol indirect effects operate in the Realistic Aerosol case and result in a 97% increase in cloud droplet number concentration (not shown) through the activation of aerosols as cloud condensation nuclei, thus increasing the shortwave cloud reflectivity [Donner et al. 2011]. Simultaneously, the presence of cloud above aerosol masks the interaction of the underlying scattering and absorbing aerosol with downwelling shortwave radiation. With the removal of cloud in the clear-sky calculation, these factors tend to compensate for one another, resulting in a net minimal difference in surface shortwave radiation between the clear-sky and all-sky calculations.

The idealized Pure Dimming simulation—designed both to isolate the surface effects of atmospheric absorption and to simulate the response to purely scattering-driven dimming—produces an all-sky SSR reduction of \( \Delta SSR_{all} = -18.6 \text{ W m}^{-2} \). The difference between the clear-sky and all-sky SSR reduction can be largely explained by climatological cloud-masking, as each constitutes a comparable fractional reduction in its respective control SSR budget (10% in the clear-sky case and 9% in the all-sky case). Cloud amount also decreases by approximately 3% (Table 4.4), which appears to be consistent with the smaller all-sky SSR reduction relative to the clear-sky SSR reduction. However, this is the result of a decrease in middle and high cloud and an increase in low cloud, making the overall shortwave effects of the vertically integrated cloud change difficult to determine.
Because the Pure Heating simulation—designed to isolate the atmospheric effects of aerosol absorption—has a heating rate rather than an optical depth imposed, the SSR and absorption perturbations are deliberately minimal in that simulation. As evinced by the difference between the clear-sky and all-sky values (Table 4.2), they are primarily driven by cloud changes that are consistent with the overall circulation response discussed in Section 4.4.3.

Although the absorption optical depth in the Pure Absorption case—designed to probe the combined surface and atmospheric effects of purely absorption-driven dimming—is comparable to that of the Realistic Aerosol case (see Section 4.2), it produces a larger amount of absorption due to the absence of aerosol scattering and aerosol indirect effects, which might otherwise attenuate the shortwave radiation before it reaches an absorber. The increased absorption ($\Delta Abs_{all} = 17.3 \ W \ m^{-2}$) produces a corresponding reduction in SSR ($\Delta SSR_{all} = -14.6 \ W \ m^{-2}$). The atmospheric absorption is larger than the SSR reduction partly due to additional absorption produced by shortwave radiation reflected from the surface, which is not counted in the downward-incident SSR change.

### 4.3.2 Surface energy balance response

On decadal time-scales the land surface energy balance is constrained to maintain equilibrium due to the low effective heat capacity of the land surface. As a result, the reduction in downwelling surface shortwave flux ($\Delta SW_{\downarrow}$) caused by the scattering and/or absorption in the 3 cases that contain dimming (Realistic Aerosol, Pure Dimming, and Pure Absorption) must be compensated for by changes in the other surface energy balance terms: reflected upwelling shortwave flux ($\Delta SW_{\uparrow}$), net longwave flux ($\Delta LW_{\downarrow} + \Delta LW_{\uparrow}$), sensible heat flux ($\Delta SH_{\downarrow}$), and latent heat flux ($\Delta LH_{\downarrow}$), i.e. $\Delta F_s = (\Delta SW_{\downarrow} + \Delta SW_{\uparrow}) + (\Delta LW_{\downarrow} + \Delta LW_{\uparrow}) + \Delta SH_{\downarrow} + \Delta LH_{\downarrow} = 0$ for all values downwelling positive.

These changes are shown in Table 4.3. Slight residuals in the surface energy balance can be attributed to the long adjustment times induced by the deep soil moisture in AM3’s
Table 4.3: Regional-mean surface energy flux changes over East Asia (22.5°–40° N; 100°–122.5° E) are shown for each of the 4 simulations in units of $W \, m^{-2}$. All values are given as downward positive.

<table>
<thead>
<tr>
<th></th>
<th>Realistic Aerosol</th>
<th>Pure Dimming</th>
<th>Pure Heating</th>
<th>Pure Absorption</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta SW_\downarrow$</td>
<td>-14.3</td>
<td>-18.6</td>
<td>-0.19</td>
<td>-14.6</td>
</tr>
<tr>
<td>$\Delta SW_\uparrow$</td>
<td>2.09</td>
<td>2.64</td>
<td>0.03</td>
<td>2.37</td>
</tr>
<tr>
<td>$\Delta LW_\downarrow$</td>
<td>-0.62</td>
<td>-3.22</td>
<td>-0.12</td>
<td>-0.43</td>
</tr>
<tr>
<td>$\Delta LW_\uparrow$</td>
<td>1.16</td>
<td>4.45</td>
<td>-0.09</td>
<td>2.36</td>
</tr>
<tr>
<td>$\Delta SH_\uparrow$</td>
<td>3.23</td>
<td>5.30</td>
<td>2.00</td>
<td>5.71</td>
</tr>
<tr>
<td>$\Delta LH_\uparrow$</td>
<td>7.13</td>
<td>8.95</td>
<td>-1.82</td>
<td>4.14</td>
</tr>
</tbody>
</table>

land model [Donner et al., 2011]. Surface albedo does not change significantly between the simulations, remaining at $\alpha \approx 0.16$. Consequently, the change in upwelling shortwave radiation, $\Delta SW_\uparrow$, is a direct result of the change in downwelling shortwave radiation, $\Delta SW_\downarrow$, and $\Delta SW_\uparrow \approx (0.16)\Delta SW_\downarrow$ in all 4 cases. The surface upwelling longwave flux maintains a quartic relationship with surface temperature, according to the Stefan-Boltzmann law, while sensible and latent heat flux are controlled by temperature and moisture gradients between the surface and near-surface atmosphere, as well as surface wind stress.

The idealized forcing simulations indicate that the response of the surface energy balance to the reduction in SSR is dependent on the source of the dimming. When the dimming is entirely scattering-driven (i.e. the Pure Dimming case), the change in latent heat flux ($\Delta LH_\uparrow$) balances 48% of the dimming ($\Delta SW_\downarrow$), while sensible heat flux ($\Delta SH_\uparrow$) accounts for 28% and net longwave flux($\Delta LW_\downarrow + \Delta LW_\uparrow$) accounts for only 6.6%. Where the dimming is entirely absorption-driven (i.e. the Pure Absorption case), however, the percent contribution from sensible heat flux exceeds that from latent heat flux. Reduced sensible heat flux accounts for the plurality (39%) of the surface energy balance’s reequilibration to the dimming, while reductions in outgoing latent heat and longwave flux account for the remaining 28% and 13%, respectively. Because there is not an explicit radiative perturbation in the Pure Heating simulation, the perturbation to its surface energy balance cannot be construed as a response to $\Delta SW_\downarrow$. Indeed, radiative fluxes remain relatively unperturbed. However, outgoing sensible heat flux decreases, while latent heat flux increases by a similar amount.
The partitioning of the surface energy balance response to dimming from Realistic Aerosol, although it is driven half by atmospheric absorption (Table 4.2), is almost identical to the Pure Dimming case. The reduction in $\Delta LH_{\uparrow}$ balances 49% of $\Delta SW_{\downarrow}$, though $\Delta SH_{\uparrow}$ and $(\Delta LW_{\downarrow} + \Delta LW_{\uparrow})$ also decrease, respectively balancing 23% and 3.8% of $\Delta SW_{\downarrow}$. This suggests that the larger sensible heat flux response seen in the Pure Absorption case requires a dominating absorption contribution to the dimming. We explore the mechanisms behind the relative behavior of the sensible versus latent heat flux response to dimming in Section 4.4.2.

### 4.3.3 Impact on EASM strength

As the primary mechanism for regional rainfall in East Asia, monsoon strength is frequently quantified to the first order by the regional mean precipitation [e.g. Lu et al., 2006; Bollasina et al., 2011]. Because of their role in supporting this monsoonal precipitation, convective activity and the land-sea surface temperature contrast are generally used to evaluate the overall monsoon circulation strength [e.g. Wang et al., 2008; Dai et al., 2013]. Consequently, we use these standard parameters to characterize qualitative changes in the EASM strength: land-sea thermal contrast (i.e. land surface temperature, shown in Figure 4.2); on-shore flow (i.e. 850 mb winds, shown in Figure 4.3); atmospheric ascent (i.e. 500 mb vertical velocity in pressure coordinates, shown in Figure 4.4) over land; and, ultimately, precipitation (Figure 4.5). We also summarize the overall monsoon strength quantitatively using the total precipitation averaged over East Asia, and the 850 mb wind speed averaged over the East Asian monsoon sector ($10^\circ$–$40^\circ$ N and $110^\circ$–$150^\circ$ E) as defined by Li and Zeng [2002] for quantifying EASM variability (Table 4.4).

The Realistic Aerosol signal indicates that historical regional anthropogenic aerosol emissions have reduced the strength of the EASM, consistent with past research [e.g. Song et al., 2014; Wang et al., 2015]. Although atmospheric absorption in the Realistic Aerosol case results in substantial shortwave heating throughout the lower atmosphere (Section 4.4.3),
Figure 4.2: The surface temperature response (shading; in K) to (a) historical regional aerosols, (b) a purely scattering aerosol optical depth, (c) idealized atmospheric heating, and (d) a purely absorbing aerosol optical depth. Climatological values are shown in grey contours, and coastal outlines are shown in black. The red box delineates East Asia (22.5°–40° N; 100°–122.5° E), over which idealized perturbations are imposed and regional-means are calculated.
Figure 4.3: The change in 850 mb wind velocity (vectors; in m s\(^{-1}\)) in response to (a) historical regional aerosols, (b) a purely scattering aerosol optical depth, (c) idealized atmospheric heating, and (d) a purely absorbing aerosol optical depth. Climatological wind speeds (in m s\(^{-1}\)) are shaded, with sign convention following meridional wind direction (southerly positive). Coastal outlines are shown in black. The red box delineates East Asia (22.5\(^\circ\)–40\(^\circ\) N; 100\(^\circ\)–122.5\(^\circ\) E), over which idealized perturbations are imposed and regional-means are calculated.
Table 4.4: Regional-mean responses over East Asia (22.5°–40° N; 100°–122.5° E) are shown for each of the 4 perturbations. The change in column integrated cloud amount (ΔCloud) is given in percent of grid cloud coverage, the change in surface (ΔTs) and near-surface air (ΔTa) temperature is given in degrees Kelvin, the change in precipitation (ΔP/P) is given in percent of climatological values, and the change in vertical pressure velocity at 500 mb (Δω500) is given in 10⁻³ Pa s⁻¹. The change in 850 mb wind speed (ΔV850) is averaged over the East Asian monsoon sector (10°-40° N; 110°-150° E) following Li and Zeng [2002].

<table>
<thead>
<tr>
<th></th>
<th>Realistic Aerosol</th>
<th>Pure Dimming</th>
<th>Pure Heating</th>
<th>Pure Absorption</th>
</tr>
</thead>
<tbody>
<tr>
<td>ΔCloud</td>
<td>-0.28</td>
<td>-0.87</td>
<td>0.41</td>
<td>1.14</td>
</tr>
<tr>
<td>ΔTs</td>
<td>-0.18</td>
<td>-0.74</td>
<td>0.01</td>
<td>-0.38</td>
</tr>
<tr>
<td>ΔTa</td>
<td>-0.08</td>
<td>-0.69</td>
<td>0.04</td>
<td>-0.33</td>
</tr>
<tr>
<td>ΔP/P</td>
<td>-6.0</td>
<td>-8.0</td>
<td>6.3</td>
<td>-1.6</td>
</tr>
<tr>
<td>Δω500</td>
<td>1.0</td>
<td>3.5</td>
<td>-5.2</td>
<td>-3.0</td>
</tr>
<tr>
<td>ΔV850</td>
<td>-0.01</td>
<td>-0.11</td>
<td>0.04</td>
<td>-0.08</td>
</tr>
</tbody>
</table>

the overall radiative effects of the anthropogenic aerosol loading reduces the regional-mean land surface temperature by ΔTs = −0.18 K, which under the fixed-SST conditions in our simulations is equivalent to a reduction in the climatological land-sea thermal contrast (Fig. 4.2a). This counteraction of the land-sea contrast induces a northwesterly perturbation to the 850 mb coastal wind vectors (Fig. 4.3a), as quantified by a small reduction in wind strength over the East Asian monsoon sector (Table 4.4). The land surface cooling also drives strong subsidence over the land surface (Fig. 4.4a), resulting in Δω500 = 1.0 × 10⁻³ Pa s⁻¹ or a 6% reduction in the climatologically ascending motion (i.e. ω500 < 0). This overall counteraction of the climatological EASM circulation contributes to a regional-mean precipitation reduction of ΔP = −0.32 mm day⁻¹ or 6% (Table 4.4 and Fig. 4.5a). An elucidation of how the different aerosol radiative components contribute to this decrease can be achieved via the idealized forcing simulations discussed below.

Solar dimming from a pure scatterer (Pure Dimming) decreases EASM strength by reducing the climatological land-sea thermal contrast (Fig. 4.2b), consistent with previous studies [e.g. Guo et al., 2013; Wang et al., 2015]. The deficit in shortwave energy at the surface produced by the imposed scattering results in a regional-mean land surface cooling of ΔTs = −0.74 K (Table 4.4). This results in a northwesterly perturbation to 850 mb winds
Figure 4.4: The mid-tropospheric (500 mb) vertical pressure velocity ($\omega$) response (shading; in $10^{-3}$ Pa s$^{-1}$) to (a) historical regional aerosols, (b) a purely scattering aerosol optical depth, (c) idealized atmospheric heating, and (d) a purely absorbing aerosol optical depth. Climatological values (Pa s$^{-1}$) are shown in grey contours, and coastal outlines are shown in black. The red box delineates East Asia ($22.5^\circ$–$40^\circ$ N; $100^\circ$–$122.5^\circ$ E), over which idealized perturbations are imposed and regional-means are calculated.
along the coast, counteracting the climatological monsoon southeasterlies (Fig. 4.3b), as quantified by the negative anomaly in wind strength over the East Asian monsoon sector (Table 4.4). The surface cooling coincides with a decrease in atmospheric ascent (Fig. 4.4b), inducing a regionally-averaged reduction of $\Delta \omega_{500} = 3.5 \times 10^{-3}$ Pa s$^{-1}$ or 25% (Table 4.4). Correspondingly, regional-mean precipitation decreases by $\Delta P = -0.39$ mm day$^{-1}$ or 8.0% of climatological levels (Table 4.4 and Fig. 4.5b).

Conversely, pure atmospheric heating in the absence of any surface shortwave perturbation (Pure Heating) enhances EASM circulation in our model. The surface temperature response to the atmospheric heating (Table 4.4 and Fig. 4.2c) is minimal ($\Delta T_s = 0.01$ K). However, the imposed atmospheric shortwave heating induces strong ascending motion over the land surface (Fig. 4.4c); ascending vertical motion at 500 mb increases by a regional mean of $\Delta \omega_{500} = -5.2 \times 10^{-3}$ Pa s$^{-1}$ or 37% (Table 4.4). This atmospheric ascent over land drives lower-level convergence that weakly increases the 850 mb flow of moisture-laden air from the surrounding ocean area (Table 4.4 and Fig. 4.3c). The combined enhancement of atmospheric vertical motion and moisture transport produce a regional-mean precipitation increase of $\Delta P = 0.30$ mm day$^{-1}$ or 6.3% of climatological levels (Table 4.4 and Fig. 4.5c).

Both the solar dimming and the atmospheric heating are active in the Pure Absorption case, and the EASM circulation response is correspondingly a combination of the effects of the two components. While the atmospheric absorption drives an atmospheric shortwave heating rate comparable to the Pure Heating case (Section 4.2), the attenuation of this shortwave radiation within the atmosphere results in a substantial reduction in surface solar radiation (Table 4.2) that drives surface cooling (Fig. 4.2d). The atmospheric heating enhances vertical motion (Fig. 4.4d), resulting in a regional-mean increase in ascending motion of $\Delta \omega_{500} = -3.0 \times 10^{-3}$ Pa s$^{-1}$ or 21% (Table 4.4). However, the solar dimming simultaneously reduces the regional-mean land surface temperature by $\Delta T_s = -0.38$ K (Table 4.4). Although this does reduce the climatological land-sea thermal contrast and induce a northeasterly perturbation to the coastal 850 mb flow that counteracts the climatological onshore transport
Figure 4.5: The change in precipitation (shading; in mm day$^{-1}$) in response to (a) realistic historical aerosols (b) a purely scattering aerosol optical depth from historical aerosol, (c) idealized atmospheric heating, and (d) a purely absorbing aerosol optical depth. Climatological values (in mm day$^{-1}$) are shown in grey contours, and coastal outlines are shown in black. The red box delineates East Asia (22.5°–40° N; 100°–122.5° E), over which idealized perturbations are imposed and regional-means are calculated.
(Fig. 4.3d), it does so by less than the Pure Dimming case (Table 4.4). The precipitation response is small (Table 4.4 and Fig. 4.5d), but net negative ($\Delta P = -0.075$ mm day$^{-1}$ or -1.6%), an outcome that will be further discussed in Section 4.4.

### 4.3.4 Moisture Budget Response

The surface energy balance and monsoon circulation responses combine in determining the overall moisture budget response, as characterized by the response in precipitation ($\Delta P$), evaporation ($\Delta E$, proportional to the change in surface latent heat flux), and the moisture convergence necessary to balance the two ($\Delta(P - E)$). East Asian summertime climate is characterized by climatological moisture convergence ($P > E$).

Table 4.5 shows the moisture budget values for each of the 4 perturbations. In the presence of Realistic Aerosol or idealized Pure Dimming, $P$ decreases by more than $E$ and moisture convergence decreases, consistent with the counteraction of monsoonal flow discussed in Section 4.3.3. In the Pure Heating simulation, conversely, moisture convergence increases via an increase in $P$ that is larger than the increase in $E$. The Pure Absorption case shows a counterbalancing of these two effects; although both $P$ and $E$ decrease as in the Realistic Aerosol and Pure Dimming case, $P$ does so by less than $E$ and moisture convergence consequently increases. The differences in $\Delta P$, $\Delta E$, and $\Delta(P - E)$ between the Pure Absorption and Pure Dimming cases (8.99, 4.81, and 4.18 W m$^{-2}$ respectively) are of the same sign as the values in the Pure Heating case and of comparable magnitude (Table 4.5), suggesting that the Pure Absorption behavior can be interpreted as a superposition of the Pure Dimming and Pure Heating cases. Energy balance constraints provide an investigation of this outcome, discussed in Section 4.4.1.
Table 4.5: The moisture budget averaged over East Asia (22.5°–40° N; 100°–122.5° E) is shown for each of the 4 perturbations. Changes in precipitation ($\Delta P$), evaporation ($\Delta E$, cf. $\Delta$ Latent Heat in Table 4.3), and moisture convergence ($\Delta (P - E)$) values are given in units of W m$^{-2}$.

<table>
<thead>
<tr>
<th></th>
<th>Realistic Aerosol</th>
<th>Pure Dimming</th>
<th>Pure Heating</th>
<th>Pure Absorption</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta P$</td>
<td>-9.35</td>
<td>-11.17</td>
<td>8.77</td>
<td>-2.18</td>
</tr>
<tr>
<td>$\Delta E$</td>
<td>-7.13</td>
<td>-8.95</td>
<td>1.82</td>
<td>-4.14</td>
</tr>
<tr>
<td>$\Delta (P - E)$</td>
<td>-2.22</td>
<td>-2.22</td>
<td>6.95</td>
<td>1.96</td>
</tr>
</tbody>
</table>

4.4 Discussion

4.4.1 An energetic rationalization of absorption’s effect on the EASM

Our results demonstrate that the EASM response to realistic regional aerosol emissions is dominated by the suppressing effect of their surface solar dimming, even in the presence of strong shortwave atmospheric absorption by the aerosols. An interesting first-order question then is whether the EASM response, as summarized in the regional-mean precipitation change, is at all sensitive to the presence of the atmospheric absorption or is simply a response to the surface solar dimming—a question that can be addressed via comparison of the results of our idealized forcing simulations.

Because of variations in how the imposed perturbation translates into a surface radiative effect in each of our simulations (Section 4.2), the surface dimming values across the simulations that contain an explicit radiative perturbation (i.e. Realistic Aerosol, Pure Dimming, and Pure Absorption) are not identical. However, we are concerned with understanding how the response to a given amount of surface dimming depends on the source of that dimming. It is useful, therefore, to normalize out the small differences in the amount of surface dimming across our simulations to allow direct comparison. We refer to this approach as “per unit dimming” in the remainder of the chapter, and calculate it by dividing the response of interest (e.g. $\Delta P$) by the change in surface shortwave radiation ($\Delta SSR_{sw}$) for a given simulation.
The precipitation response does not scale linearly with SSR reduction across the perturbations. The ratio of precipitation decrease to SSR decrease ($\Delta P/\Delta SSR_{\text{all}}$) in the Pure Dimming case is 3-4 times larger than that in the Pure Absorption case, indicating that the atmospheric effects of the aerosol absorption have a damping effect on the precipitation response to the solar dimming. Pure Heating on its own has little to no effect on the surface shortwave radiation or surface temperature, but increases precipitation substantially, hinting at the other processes besides land-sea contrast that can influence EASM strength. The smaller precipitation reduction under Pure Absorption, thus, is not achieved via a modulation of the land-sea surface temperature contrast that drives the circulation decrease under Pure Dimming; another mechanism must be invoked.

We turn to the atmospheric energy and moisture budgets to seek guidance on why absorption-driven dimming (Pure Absorption) would produce a smaller precipitation reduction than the same amount of purely scattering-driven dimming (Pure Dimming). We show that, to the first order, absorption-driven dimming must increase moisture convergence ($P - E$) into the region, while scattering-driven dimming must decrease it due to their opposite top-of-atmosphere radiative perturbations. Then, because surface evaporation ($E$) decreases under either type of dimming and by more (per unit dimming) in the scattering-driven than in the absorption-driven case, the precipitation reduction under absorption-driven dimming must be less than that under scattering-driven dimming. The full argument follows.

The moist static energy (MSE) budget [Neelin and Held, 1987; Chou and Neelin, 2003] dictates that the net energy input into the atmospheric column must be balanced by the divergent moist static energy transport. In monsoonal systems, this balance may be approximated as follows:

$$\langle \frac{\delta h}{\delta t} \rangle \approx 0 = F_{\text{net}} - \langle \vec{v} \cdot \nabla h \rangle - \langle \omega \frac{\delta h}{\delta p} \rangle$$  \hspace{1cm} (4.1)

The seasonal mean vertical integral ($\langle X \rangle$) of the time variation of moist static energy ($h = c_pT + gz + L_vq$, where $c_p$ is specific heat, $T$ is atmospheric temperature, $g$ is gravity, $z$ is
geopotential height, $L_v$ is the latent heat of vaporization, and $q$ is specific humidity) can be approximated as 0, since the atmosphere’s capacity for energy storage is minimal. Thus, the net energy input into the atmospheric column ($F_{\text{net}} = SW_{\uparrow}^{sfc} + SW_{\downarrow}^{sfc} + SW_{\uparrow}^{TOA} + SW_{\downarrow}^{TOA} + OLR + LW_{\downarrow}^{sfc} + LW_{\uparrow}^{sfc} + SH_{\uparrow} + LH_{\uparrow}$, for all values positive into the atmosphere) must be roughly balanced by the horizontal advection ($\langle \vec{v} \cdot \nabla h \rangle$) and vertical advection within the atmospheric column ($\langle \omega \frac{\delta h}{\delta p} \rangle$) of moist static energy.

The horizontal advection term generally can be neglected under summer monsoon conditions, which can be approximated as a quasi radiative-convective equilibrium [Neelin and Held 1987], though it may constitute an appreciable portion of the climatological MSE budget in the EASM [e.g. Chen and Bordoni 2014]. Under this assumption, equation 4.1 can be simplified as $F_{\text{net}} \approx \langle \omega \frac{\delta h}{\delta p} \rangle$. It can be further assumed that $\langle \frac{\delta h}{\delta p} \rangle < 0$ in the troposphere [e.g. Chen and Bordoni 2014], yielding the simplified proportionality: $F_{\text{net}} \propto -\langle \omega \rangle$.

The above relationship can then be used to understand the dynamical response to our imposed perturbations in the adjustment sense: $\Delta F_{\text{net}} \propto -\Delta \langle \omega \rangle$. Further, under a given perturbation, the surface energy balance will rapidly equilibrate to zero over land, as discussed in Section 4.3.2 i.e. $\Delta F_s = (\Delta SW_{\uparrow} + \Delta SW_{\downarrow}) + (\Delta LW_{\downarrow} + \Delta LW_{\uparrow}) + \Delta SH_{\uparrow} + \Delta LH_{\uparrow} = 0$. The change in net energy input into the atmospheric column, therefore, reduces to $\Delta F_{\text{net}} = \Delta F_s + (\Delta SW_{\uparrow}^{TOA} + \Delta SW_{\downarrow}^{TOA} + \Delta OLR) = 0 + \Delta F^{TOA}$, where $\Delta F^{TOA}$ is the change in top-of-atmosphere flux. The proportionality thus can be further approximated as $\Delta F^{TOA} \propto -\Delta \langle \omega \rangle$. Note that, in fixed SST simulations such as ours, the equilibrium change in top-of-atmosphere flux is equivalent to the top-of-atmosphere effective radiative forcing ($\Delta F^{TOA}$ in Table 4.2).

The proportionality, $\Delta F^{TOA} \propto -\Delta \langle \omega \rangle$, indicates that a change in the vertical integral of the vertical pressure velocity ($\omega$) caused by an aerosol perturbation will be proportional to its TOA effective radiative forcing, i.e. a positive (negative) forcing will induce an ascending (subsiding) perturbation to vertical motion in the atmosphere. Comparison of the vertical profiles of $\omega$ (Fig. 4.6a) and $\Delta F^{TOA}$ (Table 4.2) shows that this relationship holds for all
3 cases with a closed energy budget (i.e. Realistic Aerosol, Pure Dimming, and Pure Absorption), indicating that the simplified proportionality is applicable to this regime. Because Pure Absorption results in net positive TOA forcing, even in the presence of strong surface dimming, the absolute value of the vertical integral of $\omega$ is energetically constrained to increase, as fulfilled by the enhanced ascending motion ($\omega < 0$) throughout the atmospheric column (Fig. 4.6a). Conversely, Pure Dimming’s net negative $\Delta F^{TOA}$ implies a positive vertical integral of $\Delta \omega$.

Note that, because the Pure Heating case involves the artificial imposition of a shortwave heating rate, the above framework will not strictly hold, although the relative signs of $\Delta \omega$ and $\Delta F^{TOA}$ are still consistent. In this case, the imposed shortwave heating rate should be considered an additional input of energy into the atmosphere, which would need to be accounted for in the $F_{net}$ term. Because it is artificially imposed rather than induced by an explicit radiative perturbation, unlike the shortwave heating in Pure Absorption, it is otherwise not directly reflected in the $F_s$ or $F^{TOA}$ terms.

The vertically integrated moisture budget then connects this vertical motion to the regional precipitation. To the first order, the moisture budget dictates that any change in moisture convergence will be proportional to the change in the integrated vertical moisture advection, i.e. $\Delta(P - E) \propto \Delta(-\langle \omega \delta q \delta p \rangle)$, again assuming changes in horizontal advection are negligible under summer monsoon conditions [e.g. Chou and Neelin, 2003]. Changes in $\delta q / \delta p$ are small relative to changes in $\omega$ (Fig. 4.7b and Fig. 4.6a, respectively), so the proportionality can be approximated as $\Delta(P - E) \propto -\Delta \langle \omega \rangle$. Therefore, the enhanced ascending motion under Pure Absorption’s positive $\Delta F^{TOA}$ constrains the moisture convergence to increase ($\Delta(P - E) > 0$), and the converse will be true under Pure Dimming ($\Delta(P - E) < 0$), as borne out in Table 4.5.

However, both scattering- and absorption-driven surface dimming have a strongly suppressing effect on evaporation by depleting the radiative energy required to drive it, resulting in $\Delta E < 0$ (shown in terms of the surface energy balance as $\Delta LH$ in Table 4.3). Thus,
\(\Delta(P - E) > 0\) under Pure Absorption implies \(\Delta E < \Delta P < 0\), and \(\Delta(P - E) < 0\) under Pure Dimming implies \(\Delta P < \Delta E < 0\), as seen in Table 4.5. Note that land surface hydrological constraints (i.e. the Budyko relationship) requires the same sign of change in evaporation and precipitation [e.g. Budyko, 1963; Milly, 1994; Koster and Suarez, 1999; Donohue et al., 2011], preventing \(\Delta E < 0 < \Delta P\) under Pure Absorption.

As discussed in Section 4.3.2, the reduction in evaporation per unit dimming is substantially smaller under Pure Absorption than under Pure Dimming, contributing 48% of the surface energy balance adjustment to the SSR reduction in the latter case and just 28% of the adjustment in the former case, i.e. per unit dimming \(\Delta E_{PD} < \Delta E_{PA} < 0\) (where \(PA\) and \(PD\) refer to the Pure Absorption and Pure Dimming cases, respectively). Then, given that \(\Delta P_{PD} < \Delta E_{PD} < 0\) and \(\Delta E_{PA} < \Delta P_{PA} < 0\), the relative changes in precipitation and evaporation per unit dimming are constrained to be \(\Delta P_{PD} < \Delta E_{PD} < \Delta E_{PA} < \Delta P_{PA} < 0\),
Figure 4.7: Vertical profiles of changes in (a) temperature, (b) moisture mixing ratio/specific humidity (q), and (c) relative humidity averaged over East Asia (22.5°–40° N; 100°–122.5° E) are shown for the 4 simulations.

i.e. the reduction in precipitation due to Pure Absorption must be smaller per unit dimming than that due to Pure Dimming.

It is interesting to note that precipitation reduction per unit dimming in the Realistic Aerosol case is comparable to that of the Pure Dimming case. As noted in Section 4.2, the Realistic Aerosol case can be thought of as a combination of the Pure Dimming and Pure Absorption cases, plus aerosol microphysical effects. Aerosols may suppress precipitation via microphysical indirect effects in the model by reducing cloud droplet size, though this process only operates in stratiform and shallow cumulus clouds in AM3’s formulation [Donner et al., 2011]. It is plausible, therefore, that the magnitude of precipitation suppression under Realistic Aerosol should resemble that of Pure Dimming, with the additional suppression from microphysical effects counteracting the reduced suppression from the absorption-driven com-
ponent of the realistic dimming, but direct comparison is difficult for the reasons discussed in Section 4.2.

The above explanation reveals that, under the moisture convergence constraints imposed by their relative top-of-atmosphere radiative forcing, the relationship between the precipitation reduction under purely absorption-driven dimming and that under purely scattering-driven dimming is strongly influenced by their relative reductions in surface evaporation. Fully understanding why absorption-driven dimming produces a smaller precipitation reduction per unit dimming that scattering-driven dimming, therefore, requires an understanding of why absorption-driven dimming produces a smaller evaporation reduction per unit dimming. In the next section, we explore the physical mechanisms behind this difference in the surface energy balance response to absorption- versus scatter-driven dimming.

4.4.2 Absorption’s impact on surface energy flux response partitioning

Over much of South and East Asia, the compensation for total aerosol-driven surface dimming is thought to come largely from a reduction in latent heat release or evaporation [Ramanathan et al., 2001, 2005], consistent with our Realistic Aerosol case. In fully ocean-atmosphere coupled GCM runs conducted by Ramanathan et al. [2005], the evaporation decrease is strongly controlled on a regional basis by a reduction of the temperature and relative humidity gradient between the surface and boundary layer due to dimming-driven surface cooling. This surface latent heat flux reduction can further exacerbate transport-driven moisture deficits due to aerosols, such as those discussed in Section 4.3.3.

Our idealized perturbation results, however, indicate that whether decreased surface latent or sensible heat flux provides the primary balance for the reduction in shortwave radiation depends on the degree to which that dimming is absorption-driven (Table 4.3); in the absence of absorption, dimming is primarily compensated for by a latent heat flux reduction, but in the presence of absorption it is primarily compensated for by a sensible
heat flux reduction (Section 4.3.2). As discussed in Section 4.4.1, this outcome helps to explain the smaller precipitation reduction under absorption-driven dimming than under scattering-driven dimming. In order to understand the relative magnitude of the latent heat flux reduction, we first analyze the factors controlling the behavior of the complementary sensible heat flux reduction.

Sensible heat flux can be thought of as controlled by the local gradient between surface and near-surface atmospheric temperature and the wind speed, as parameterized in GCMs via the bulk formula:

$$SH = \rho c_p c_H v_a (T_s - T_a)$$ \hspace{1cm} (4.2)

where $\rho$ and $c_p$ are the density and specific heat of air, respectively, $c_H$ is a transfer coefficient, $v_a$ is the horizontal wind speed in the near-surface atmosphere, $T_s$ is the surface temperature, and $T_a$ is the near-surface air temperature. The change in horizontal wind speed over land is small under all four of our perturbations (shown at 850 mb in Fig. 4.3), less than 5% of climatological values at the near-surface (10 m), so we focus here on the influence of the change in the gradient between surface and near-surface air temperature ($\Delta T_s - \Delta T_a$) under purely absorption-driven (Pure Absorption) versus purely scattering-driven (Pure Dimming) dimming. Climatologically, the land surface is warmer than the near-surface atmosphere, encouraging the dry convection of turbulent heat flux from the surface to the atmosphere. If this gradient is depressed, either by land surface cooling or atmospheric warming, the surface sensible heat flux can be expected to be proportionally depressed.

Pure Absorption can be thought of as a superposition of the Pure Dimming case and the Pure Heating case, both of which exhibit a reduction in surface-to-air temperature gradient. In the Pure Dimming case, both the surface and the near-surface atmosphere cool due to the depletion of surface shortwave radiation, but the surface cools by more than the atmosphere (Table 4.4), resulting in a reduction in the temperature gradient and a suppression of surface sensible heat flux (Table 4.3). In the Pure Heating case, conversely, both the surface and the near-surface atmosphere warm (Table 4.4) due to the absorption-mimicking imposed
shortwave heating. However, the atmosphere warms by more than the surface, again resulting in a reduction in the temperature gradient between the climatologically cooler atmosphere and warmer surface and, consequently, a reduction in surface sensible heat flux (Table 4.3) even in the absence of an imposed surface shortwave perturbation. In the Pure Absorption case, both the surface cooling from surface shortwave depletion and the atmospheric heating from in situ shortwave absorption are in operation. The combined effects result in a larger reduction in surface-to-air temperature gradient per unit dimming than under the Pure Dimming case (Table 4.4) and, consequently, stronger suppression of sensible heat flux per unit dimming (Table 4.3 and Section 4.3.2). Because of the stronger sensible heat flux suppression induced under Pure Absorption, surface latent heat flux need not decrease as much for a given reduction in surface shortwave input as it does under the Pure Dimming conditions. As a result, latent heat flux (i.e. evaporation) suppression is weaker under Pure Absorption than under Pure Dimming, which (in combination with their relative influence on moisture convergence) helps explain why absorption-driven dimming reduces precipitation less strongly than scattering-driven dimming.

We can also use the relative changes in near-surface atmospheric temperature between Pure Absorption and Pure Dimming to understand the relative reduction in latent heat flux directly, though it is subject to more approximating assumptions. Climatologically, for a given net surface radiation, surface latent heat flux is controlled by the relative moisture content of the two reservoirs it communicates between: the land surface and the atmosphere. Over saturated surfaces (e.g. water or moist land), the surface latent heat flux is primarily determined by the near-surface atmospheric water demand, generally quantified as the potential evapotranspiration (PET) [e.g. Allen et al., 1998]. Conversely, where near-surface atmospheric water demand is greater than surface water availability (e.g. arid land), variability in soil moisture is the primary controller of latent heat flux from the surface to the atmosphere [e.g. Seneviratne et al., 2010]. Because of the climatologically high precipitation rates of the EASM, East Asian summertime latent heat flux is generally considered to be
controlled by atmospheric water demand \cite[e.g.][]{Zhang2011}. Thus, assuming that any changes in precipitation due to the imposed perturbations will not be sufficient to change the region from an atmosphere-controlled to a surface-controlled regime, an understanding of the relative latent heat flux response to absorption-driven versus scattering-driven dimming can be achieved by analysis of the relative impact of each on PET.

PET is most robustly calculated via the Penman-Monteith equation \cite[e.g.][]{Allen1998, Scheff2015, Fu2014}, which is derived from the surface energy balance and the bulk formulae for sensible and latent heat flux under the assumption of a saturated surface:

\[
\text{PET} = \frac{(F_R)(\frac{de^*}{dT})(T_a) + \rho c_p e^* (T_a)(1 - RH_a)c_H v_a}{\frac{c_p}{L_v} + \gamma (1 + r_s c_H v_a)}
\]  

(4.3)

where \( F_R \) is the net radiative flux into the surface, \( \frac{de^*}{dT} \) is the local vertical gradient of the saturation vapor pressure \( (e^*) \), \( \gamma = \frac{c_p p_s}{0.622 L_v} \) given surface pressure \( (p_s) \) and latent heat of vaporization for water \( (L_v) \), \( r_s \) is the bulk stomatal resistance, and all other terms are defined as in equation 4.2. Although eq. 4.3 is most representative when applied to hourly-mean or higher temporal resolution data, it is also applicable in the monthly mean perspective used here \cite{Allen1998, Scheff2014}

The above equation indicates that PET will be directly proportional to \((1 - RH_a)\), i.e. as RH in the near-surface atmosphere increases, PET will decrease, and vice versa. Relative humidity, in turn, is a direct function of specific humidity and an inverse function of atmospheric temperature. The local saturation vapor pressure gradient \( \frac{de^*}{dT} \), meanwhile, is an inverse square function of atmospheric temperature. When all such temperature dependencies are evaluated, PET can be shown to be a direct function of \( T_a \), subject to various simplifying assumptions (see \cite{Scheff2014}).

The smaller decrease in \( T_a \) under Pure Absorption than under Pure Dimming—a result of the balance between the atmospheric absorption-induced heating and surface dimming-
induced cooling—leads to a smaller decrease in PET, according to the first-order scaling of the Penman-Monteith equation. Consequently, the near-surface atmosphere’s demand for moisture, which will be the primary driver of latent heat flux under monsoon-saturated surface conditions, decreases by less per unit dimming under Pure Absorption than under Pure Dimming. Given the same amount of dimming, therefore, the surface shortwave flux reduction will have to be more strongly balanced by reduced sensible heat flux under Pure Absorption (Section 4.3.2), as the latent heat flux reduction will be limited by the atmosphere’s relatively greater demand for moisture under Pure Absorption than under Pure Dimming.

4.4.3 Atmospheric processing of absorption-driven atmospheric heating

The analysis in Sections 4.4.1 and 4.4.2 demonstrates the competing interplay of absorbing aerosols differing surface, atmospheric, and top-of-atmosphere radiative perturbations. It is particularly important to note that the sign of local surface temperature change does not correspond to the sign of local TOA forcing in the case of aerosol absorption (Table 4.2 and 4.4). By depleting surface shortwave radiation locally, absorbing aerosols can reduce regional surface temperatures, especially in regions in which the radiative-convective coupling of the surface and atmosphere is weak [Shindell and Faluvegi, 2009; Ramanathan and Carmichael, 2008; Bond et al., 2013]. They may, however, increase surface temperature elsewhere through tropospheric transport of their atmospheric heating [Menon et al., 2002; Teng et al., 2012]. Although surface-atmosphere coupling is thought to be relatively strong during the EASM [Zhang et al., 2011], our results suggest that it is not sufficient to overcome the cooling effects of the surface dimming.

If the atmospheric heating from atmospheric absorption does not efficiently heat the surface, where does it go? Analysis of the perturbations to various components of the atmospheric heating rates sheds light on this question (Fig. 4.8). In the case of Pure Heating
Figure 4.8: Regional-mean atmospheric heating rates in K day$^{-1}$ are shown for the three idealized forcing simulations: (a) Pure Dimming, (b) Pure Heating, and (c) Pure Absorption. The heating rates due to shortwave radiation (red), longwave radiation (dark blue), latent heat release by convective (light blue) and large-scale (black) cloud formation, vertical diffusion (pink), and dynamical advection of sensible heat (green; computed as residual of other terms) are shown. The shortwave heating rate for the Realistic Aerosol case is also shown (grey) for reference in panels (b) and (c).
(Fig. 4.8b), the imposed shortwave atmospheric heating rate is primarily compensated for by a negative dynamical heating rate. This indicates that the heat is being transported out of the atmospheric column. The absolute value of the dynamical heating rate peaks near 400 mb, indicating that the ventilation is occurring primarily in the upper atmosphere. This completes the circulation pattern indicated by the onshore flow at 850 mb and ascending motion through 500 mb (Section 4.3.3). There is weak cooling in the lower troposphere from vertical diffusion—a signal of the vertical propagation of the surface sensible heat flux suppression discussed in Section 4.4.2.

In the case of the Pure Absorption (Fig. 4.8c), the clear-sky shortwave heating rate is similar by design to that in the Pure Heating simulation, but is compensated for differently. There is some balancing by dynamical cooling aloft, suggesting a similar upper atmospheric ventilation as in the Pure Heating case. However, there is a larger lower atmospheric compensation by cooling from vertical diffusion. This can again be attributed to the vertical propagation of Pure Absorption’s larger sensible heat flux suppression. Pure Dimming exhibits lower tropospheric cooling from vertical diffusion, which balances lower tropospheric dynamical heating from the suppression of vertical motion described in Section 4.4.1 and is consistent with the dimming-driven suppression of sensible heat flux discussed in Section 4.4.2. However, the Pure Absorption case exhibits vertical diffusional cooling that is a superposition of that both from Pure Heating’s atmosphere-driven sensible heat flux suppression and from Pure Dimming’s dimming-driven sensible heat flux suppression, and therefore substantially stronger, as with the surface sensible heat flux suppression itself.

The composition of the heating rate responses to the shortwave heating from aerosol absorption demonstrates how a local positive aerosol radiative forcing can coexist with negative regional surface responses through transport of the heating out of the forcing region. Given the inherently regional nature of aerosol forcing, due to aerosols’ short lifetime and geographically concentrated emissions sources, this result demonstrates that local responses to changes in absorbing aerosol emissions should not be expected to follow global-mean re-
responses and that the surface heating effects from such changes in one region may be primarily felt in other regions.

4.5 Conclusions

The impact of absorbing aerosols on regional climate manifests both through their impact on the atmospheric radiative budget and on surface energy fluxes. This work provides one of the first analyses of the separate and combined effects of aerosol absorption’s atmospheric and surface perturbations on East Asian summertime climate. Our work suggests that the surface energy impacts of aerosol absorption are capable of outweighing its atmospheric impacts in the net response of East Asian Summer Monsoon (EASM) strength, resulting in a net decrease of EASM circulation and precipitation due to the reduced land-sea contrast from dimming-induced land surface cooling (Section 4.3.3). Crucially, however, the precipitation reduction under absorption-driven dimming is smaller per unit dimming than that under purely scattering-driven dimming, due to the moisture convergence constraints imposed by their opposing signs of top-of-atmosphere radiative forcing (Section 4.4.1). This is partially influenced by stronger suppression of surface sensible heat than latent heat under absorption-driven dimming and converse behavior under scattering-driven dimming (Section 4.4.2), which constrains the relative reduction in the evaporative component of moisture convergence and consequently the relative reduction in the precipitation component. Atmospheric heating from aerosol absorption plays a role in this additional suppression of sensible heat, but it is primarily transported out of the region in the upper troposphere. Consequently, absorbing aerosols’ impact on East Asian summertime climate more closely resemble the response to its surface dimming than to its atmospheric heating.

The partitioning of the surface energy balance between sensible and latent heat flux (i.e. the Bowen ratio, \( \frac{SH}{LH} \)) has myriad implications for surface temperature, convection, and boundary layer depth, as well as the atmospheric moisture budget [e.g. Andrews et al.].
Our results indicate that this partitioning is sensitive to the atmospheric forcing associated with a surface forcing: absorption- and scattering-driven dimming perturb the Bowen ratio in opposite directions. We provide a physical explanation here for why sensible heat suppression dominates latent heat suppression under purely absorption-driven dimming, but the partitioning of surface energy fluxes is known to be underconstrained in climate models [e.g. Dirmeyer 2011]. The impact that this partitioning can have on the magnitude of a precipitation response to solar dimming, as discussed in Section 4.4.1, highlights the importance of better constraining this process in models.

This analysis sheds light on the response of East Asian summertime climate to realistic regional aerosol emissions. Surface solar radiation reductions over this region since the 1960s have been driven equally by increased aerosol scattering and increased aerosol absorption. However, the negative EASM response to historical regional aerosol emissions (Realistic Aerosol)—a net reduction of onshore flow, atmospheric ascent, and regional-mean precipitation—scales in proportion to purely scattering-driven dimming (Pure Dimming; Section 4.4.1) and does not exhibit the signal of absorption-driven atmospheric heating (Pure Heating), which on its own invigorates EASM circulation (Section 4.3.3). That even purely absorption-driven dimming reduces EASM strength helps explain why the combined absorption- and scattering-driven dimming of realistic aerosols shows such a strong EASM reduction.

It is important to note that our study does not incorporate the potential additional EASM impacts of aerosol-induced sea surface temperature perturbations. Studies show that the outflow of East Asian aerosol emissions can affect sea surface temperatures in the Western Pacific [e.g. Hsieh et al. 2013; Boo et al. 2015]. The strong modulation of EASM variability by both nearby and remote SSTs [Hu 1997; Lau and Weng 2001; Yang and Lau 2004, e.g.] suggests that this aerosol-driven SST variability could induce additional EASM responses, either reinforcing or counteracting those produced by the aerosols’ in-situ effects. Additionally, the strength of EASM circulation is generally weaker in prescribed
SST simulations, such as ours, than observations suggest [Gao et al., 2010; Kitoh, 2004]. However, our analysis focuses on the land-and-atmosphere driven EASM responses to aerosol absorption, which can be expected to operate on faster timescales than the SST-driven response and warrants analysis separate from the fully-coupled response.

Our idealized forcing simulations, in addition to providing physical insight on the interaction of absorbing aerosols with the EASM, provide test cases at the two limits of possible future absorption/scattering ratios of East Asian aerosol emissions. Since 2000, scattering sulfate emissions have plateaued and declined [Klimont et al., 2013; Li et al., 2013], while absorbing black carbon emissions have continued to rise [Lei et al., 2011] and are expected to continue to do so at least through 2030, though there is considerable uncertainty [Levy, 2009]. The Pure Absorption simulation provides an extreme test case of a situation in which East Asia’s aerosol concentrations are dominated by absorbing rather than scattering aerosols—for example, if China continues to mitigate sulfate aerosol emissions without imposing significant controls on black carbon aerosol emissions. Conversely, the Pure Dimming case provides a test of the extreme of a scenario in which China mitigates its absorbing black carbon emissions without mitigating its scattering sulfate emissions. In combination, these two simulations suggest that increases in either aerosol type will have detrimental effects on EASM strength, but that BC-driven dimming may be less detrimental than sulfate-driven dimming for a given unit of dimming.
Chapter 5

Trade and the Future of China’s Black Carbon Emissions

5.1 Introduction

Over the past several decades, black carbon (BC) aerosol emissions from diesel vehicles, residential cookstoves, agricultural burning, and industrial processes have become a primary concern of a diversity of international institutions due to their combined human health and climate impacts [e.g. United Nations Environment Programme and World Meteorological Organization, 2011; Janssen et al., 2012; Minjares et al., 2014]. Black carbon, the absorbing component of the particulate air pollution produced by incomplete combustion, is a major component of PM$_{2.5}$ concentrations that contribute to over 3 million premature deaths each year due to pulmonary and respiratory disease [World Health Organization et al., 2013]. It is also a potent short-lived climate forcer, with a global warming potential on the order of 1000 times larger than that of carbon dioxide (CO$_2$) on 20 year timescales [e.g. United Nations Environment Programme and World Meteorological Organization, 2011 and references therein] and the potential to substantially alter regional circulation, temperature, and rainfall [e.g. Ramanathan and Carmichael, 2008; Koch and Del Genio, 2010].
China is currently the largest emitter of BC globally. During the 2000s, China emitted approximately 2500 Gg yr\(^{-1}\) of BC [Bond et al., 2013; Wang et al., 2014], accounting for approximately 25% of total global BC emissions [Wang et al., 2014]. Coal and biofuel combustion for residential heating has contributed approximately half of China’s BC emissions over the last two decades, and the remainder of emissions comes from a combination of industrial and mobile sources [Lei et al., 2011; Lu et al., 2011]. Combined with similar BC emissions increases in South Asia and elsewhere, this activity has resulted in increased global BC emissions, from 5.3 Tg yr\(^{-1}\) in 1960 to 9.1 Tg yr\(^{-1}\) in 2007 [Wang et al., 2014].

Projections of future black carbon emissions, however, contain a reversal of this trend after 2030 (Fig. 5.1). The Representative Concentration Pathways (RCPs), used by the Intergovernmental Panel on Climate Change to capture a range of future emissions trajectories, predict that black carbon and other aerosol emissions will decline to 20-40% of current levels.
by 2100 [van Vuuren et al., 2011]. This reduction manifests in all regions and holds across the full suite of RCPs, which range in year 2100 from a target radiative forcing of 8.5 W m$^{-2}$ to 2.6 W m$^{-2}$. This universal aerosol reduction is markedly different than the range of aerosol futures presented by the earlier Special Report Emissions Scenarios (SRES), whose range included both increasing and decreasing 21st century aerosol emissions trajectories [Nakicenovic and Swart, 2000].

The reduction in BC emissions seen across the RCPs, even in the presence of increasing greenhouse gas emissions, arises from the imposition of an empirical relationship between wealth and air quality in the integrated assessment models (IAMs) used to produce the RCPs [Moss et al., 2010; Riahi et al., 2011; Smith et al., 2011b; van Vuuren et al., 2011]. This empirical relationship, known broadly as the Environmental Kuznets Curve (EKC), uses historical economic and emissions data to suggest that there is a level of gross domestic product (GDP) per capita at which nations transition from an inverse relationship to a direct relationship between GDP growth and air quality [e.g. Stern and Common, 2001]. Under this assumption, the relationship between income and air quality takes on an “inverted U-shape”: beyond some critical inflection point of wealth, countries’ air quality tends to improve rather than decline with economic growth. The first-order economic mechanism used to justify the appearance of this relationship is that air quality is a normal good: as individuals’ income increases, so does their demand for improved air quality.

The EKC assumption is subject to challenges, however, that, in some cases, posit that it is too pessimistic regarding the rate at which air quality improvements will occur and, in other cases, posit that it is too optimistic. Some critics argue that the use of historical data underestimates the rate of technology change in developing nations [e.g. Dasgupta et al., 2002; Smith et al., 2005]. The environmental improvements historically achieved by currently high-income countries required the generation of new knowledge of air quality impacts and technologies for mitigation. This occurred in a historical context of weaker regulatory institutions and more limited external funding than now exist. Currently developing countries have
the advantage of access to the extant information and technology that enabled the historical EKC, as well as more sophisticated methods of environmental regulation and more robust instruments for finance. Developing nations, according to these critiques, should therefore be able to reach the critical inflection point toward improving air quality at a lower income level than the historical data suggests.

Other challenges, however, assert that the current downslope of the EKC was achieved via processes that will make it more difficult for countries currently on the upslope to reach the inflection point [e.g. Rothman 1998; Suri and Chapman 1998; Malm 2012]. They suggest that the historical EKC was achieved in developed nations partially via the relocation of their high-emitting industries to less-developed nations. These past trajectories, therefore, cannot be applied to currently developing nations that, in many cases, were the recipients of those outsourced emissions. Suri and Chapman 1998 demonstrate that countries on the upslope of the EKC (i.e. those for whom economic growth and environmental quality are negatively correlated) have energy consumption that is strongly driven by the production of goods for export, while countries on the EKC’s downslope (i.e. those for whom economic growth and environmental quality are positively correlated) have achieved energy use reductions partially by increasing imports. This broad pattern is also evident in global flows of CO\textsubscript{2} through trade: developed nations’ consumption-based CO\textsubscript{2} emissions outweigh their production-based emissions, while the opposite is true of developing nations Davis and Caldeira 2010, implying that a substantial portion of the CO\textsubscript{2} emissions reductions achieved by developed nations have been achieved by shifting to other nations the industrial activity and associated emissions necessary to satisfy domestic consumption.

These trade-relocated emissions, and the challenge they pose to the EKC, are evident in the case of China’s BC emissions. Lin et al. 2014 conduct an input-output analysis on China’s economic activity and trade to determine the proportion of its air pollutant emissions that are the result of foreign consumption. Their work suggests that 17% of China’s black carbon emissions, an average of 170 Gg yr\textsuperscript{-1} from 2000-2007, results from the
production of goods for export; one fifth of those occur from China-to-U.S. export alone. These emissions embodied in trade suggest that, in the U.S. and other of China’s export partners, the outsourcing of those emissions to China enabled some portion of their observed decline in BC emissions with increased income and consumption per capita.

In this paper, we argue that China’s black carbon emissions embodied in trade signify a substantial and potentially increasing source of black carbon-emitting activity that may place additional mitigation pressure on China as it develops compared to currently high income countries. In Section 5.2, we demonstrate that IAM-derived projections of technology change and regulatory stringency in China fall ∼30% short of achieving the EKC-consistent emissions reductions by 2050. China’s future BC-emitting economic activity due to exports provides an explanation for why its techno-regulatory evolution will need to be faster-than-historical in order to achieve EKC-consistent BC emissions reductions. In Section 5.3, we demonstrate in global climate model simulations that unmitigated growth of China’s export-related BC emissions poses a climate and health risk of a magnitude that warrants domestic and international attention. In Section 5.4, we offer an analysis of the export-related sectors in which rapid BC emissions intensity reductions of the type needed to meet the EKC assumption can be achieved using existing technology. Together, these results indicate that China’s BC emissions embodied in export must and can be actively offset if the emissions reductions characterized by the RCPs are to be achieved.
5.2 China’s Export-Related Emissions and Their Implications for the Future

5.2.1 Integrated assessment model projection of China’s future air quality

Integrated assessment models generally combine economic, demographic, political, and technological models with a simplified climate model to analyze the interplay between environmental factors and the human systems that may affect them. They are the key tools used for the development of future emissions scenarios used by the Intergovernmental Panel on Climate Change to assess future climate risk [e.g. Nakicenovic and Swart, 2000; van Vuuren et al., 2011; O’Neill et al., 2015].

Generally, the total emissions of BC ($BC_{tot}$) and other pollutants in an IAM will be computed as the summation over all sectors ($i$) of the product of the activity level ($AL_i$) in a given sector (e.g. in USD or kJ) and the emissions factor (the emissions per unit activity for a given emitting process, $EF_i$) for that sector (e.g. in Gg USD$^{-1}$ or Gg kJ$^{-1}$), i.e.:

$$BC_{tot} = \sum_i (AL_i \times EF_i) \quad (5.1)$$

The activity level in a given sector is generally determined via input-output economic analysis, and the emissions factor is a function of the technologies and emissions control processes assumed to be used. A key result of eq. 5.1 is that increased activity levels ($\Delta AL_i > 0$) can offset a reduced emissions factor ($\Delta EF_i < 0$) so that the total emissions and resulting atmospheric concentrations do not decrease at the same rate as the emissions factor.

Although the atmospheric pollutant concentrations that determine air quality are not explicitly simulated by IAMs, they can be separately determined using atmospheric chemistry transport models initialized with IAM-generated emissions. This information can then be used in the IAMs’ calibration process. IAMs impose the empirical EKC relationship on future
regional air quality in the RCPs by (1) identifying the critical income level of transition, based on the current global distribution of GDP per capita versus air quality, and (2) fixing the stringency of air quality controls in developing countries to achieve the historically seen air quality (as determined via an atmospheric model run with IAM-generated emissions) at that critical income level.

Smith et al. [2011b] (hereafter S2011) detail the development of air pollutant emissions trajectories for the reference scenario in the Global Change Assessment Model (GCAM) IAM on which is based the RCP 4.5 scenario, describing a cost-minimizing socioeconomic and technological pathway for stabilizing global-mean radiative forcing to 4.5 W m$^{-2}$ by 2100. In GCAM, the metric of air quality was chosen to be the population-weighted, national average, seasonal maximum PM$_{2.5}$ concentration, which we refer to here as PM$_{2.5}^*$. The range of values for PM$_{2.5}^*$ seen in 2005 in a subset of countries with current income levels greater than 15,000 USD—in this case, the southern and north-central European Union, the United States, and Japan—was used as the standard air quality range. Determination of PM$_{2.5}^*$ used for calibration in S2011 was conducted using the MOZART chemistry transport model [Horowitz et al., 2003], both for establishing the year 2005 standard range and for calibrating other economies’ projected emissions. Emissions control parameters in different countries in the model were then calibrated such that, as lower-income countries reached 15,000 USD per capita, their PM$_{2.5}^*$ values fell within the standard range [Smith et al., 2011b].

The key outcome of the S2011 results analyzed here is that, compared to high-income countries with current GDP per capita $\geq$ 15,000 USD (HICs), China must have more stringent black carbon emissions controls in place at GDP per capita $\geq$ 15,000 USD in order to achieve PM$_{2.5}^*$ values that are in the range of the HICs. Economy-wide emissions intensity must decrease by an additional 11% in 2050 and by an additional 13% in 2095 beyond those achieved by the HICs at a comparable income level. The additional reduction in China’s
Figure 5.2: Simulated annual-mean surface concentrations of PM$_{2.5}$ from the GCAM model before (a,b) and after (c,d) imposition of the Environmental Kuznets Curve (EKC) assumption in 2050 (a, c) and 2095 (b, d) from Smith et al. [2011b].

BC emissions associated with this faster-than-historical emissions intensity trajectory is substantial.

In initial iterations of GCAM, emissions reductions in the absence of climate policy were achieved by constraining emissions factors ($EF_i$) to converge to currently observed values as a function of GDP per capita. This approach is “semi-Kuznets” in the sense that it creates an income/emissions relationship without explicitly fixing air quality to income levels [Smith, 2005]. Total emissions were then calculated by GCAM as the product of the emissions factor in a given sector and the activity level in that sector, as in eq. 5.1. We refer to this initial formulation as “pre-EKC” in the remainder of this paper. Under the pre-EKC
formulation, PM$_{2.5}$* in 2050 and 2095 in China, Southeast Asia, and the Indian Subcontinent are substantially higher than for current nations at comparable income levels (Fig. 5.2, 5.2b).

In subsequent iterations, S2011 increased the emissions controls for industrial processes in Asia beyond that implied by the income-expected emissions factor improvements to better match EKC-predicted surface concentrations (Fig. 5.2c, 5.2d). We refer to this subsequent formulation as “post-EKC” in the remainder of this paper. These more stringent emissions reduction trajectories were ultimately included in GCAM’s standard reference scenario on which the RCP 4.5 projections are built [Thomson et al., 2011].

Figure 5.3: Emissions of Black Carbon in East Asia before (a, b) and after (c, d) imposition of the Environmental Kuznets Curve (EKC) from the GCAM integrated assessment model.
We use the emissions files input by S2011 into the MOZART model to determine the amount of additional emissions reductions (beyond the pre-EKC estimate) required in China to meet the EKC assumption (Figure 5.3). Analysis of the difference in China’s BC emissions between the pre- and post-EKC reference scenarios indicates that an additional 160 Gg yr$^{-1}$ of black carbon is emitted in China in 2050 and 110 Gg yr$^{-1}$ in 2095 in the pre-EKC compared to the post-EKC scenario (Table 5.1). Consequently, China would have to emit 34% less BC in 2050 and 35% less in 2095 beyond the pre-EKC, income-scaled emissions factor improvements to meet empirically EKC-consistent emissions levels.

5.2.2 Implications of the difference between Pre- and Post-EKC BC emissions

We next decompose the key result of S2011—that China must achieve better-than-HIC emissions factors at GDP per capita $\geq 15,000$ USD in order to achieve comparable PM$_{2.5}^*$—into its possible implications. This decomposition and its outcome is schematically represented in Figure 5.4.

There are two possible explanations for the fact that Chinese BC emissions factors must be better than the HICs at a given income level in order to achieve comparable PM$_{2.5}^*$ values. First, it is possible that there exist region-specific factors that cause a given amount of BC emissions in China to translate into a higher value of PM$_{2.5}^*$ than in the HICs. The translation of a BC emission into a PM$_{2.5}^*$ value involves the meteorological transformation of the BC emission into a component of an atmospheric PM$_{2.5}$ concentration via atmospheric transport and mixing with other pollutant species. The calculation of the PM$_{2.5}^*$ metric from the unprocessed PM$_{2.5}$ concentrations then involves population- and seasonality-weighting and regional averaging. It is possible that some combination of China’s meteorology, seasonality of BC emissions, colocation of population and BC emissions, or other factors could result in disproportionately high PM$_{2.5}^*$ values in China. Additionally, China may be subject to
Figure 5.4: Decomposition of the key result of S2011 is schematically depicted. Analysis suggests that China will require better emissions factors than the HICs’ partially because it will have a higher proportion of BC-emitting economic activity at GDP per capita ≥ 15,000 USD.

Elevated PM$_{2.5}$ concentration due to the meteorological transport of BC emissions from other countries.

We can test this possibility by looking at China’s current ratio of BC emissions to PM$_{2.5}$ compared to a HIC, and find that it is not the case. We compare China’s PM$_{2.5}$ per unit BC emissions with that of the United States in 2005, using total BC emissions data from Lin et al. [2014] and PM$_{2.5}$ values from S2011 and find that they are nearly identical (0.03 $\mu$g m$^{-3}$ of PM$_{2.5}$ per Gg of BC emitted). GCAM uses a fixed spatial distribution of population and emissions sources in its projections, so the covariance of these factors does not change.
over the 21st century in the simulations. This suggests that China’s need for a faster-than-historical emissions reduction trajectory is not a result of an unfavorable sensitivity in China to S2011’s choice of air quality metric.

The above calculation demonstrates that a given amount of BC emissions in China should result in a PM$_{2.5}^*$ increment comparable to the HICs. Thus, China’s need for better-than-HICs emissions factors must be the result of the presence of some higher amount of potentially BC-emitting economic activity at GDP per capita $\geq 15,000$ USD. This additional activity must be offset by more stringent emissions controls to achieve the BC emissions reductions necessary to achieve HIC-consistent PM$_{2.5}^*$.

This information alone is not sufficient to demonstrate that China’s emissions reduction trajectory will be unique. China’s population at GDP per capita $\geq 15,000$ USD will be approximately an order of magnitude larger than the current population of the HICs in the GCAM reference scenario [Clarke et al., 2007]. The economy size (i.e. total GDP) required to support a per capita income of 15,000 USD, therefore, will similarly be an order of magnitude larger. China could thus have more potentially BC-emitting economic activity at GDP per capita $\geq 15,000$ USD than the HICs purely because of its larger economy and not because of any unique sources of potentially BC-emitting economic activity.

However, GCAM projections of China’s per capita BC emissions indicate that potentially BC-emitting activity will be a larger portion of total economic activity at GDP per capita $\geq 15,000$ USD than it was for the HICs. GCAM under both S2011’s pre-EKC and post-EKC conditions (and under the additional climate-related emissions controls of the RCP 4.5 scenario) projects that China’s per capita BC emissions will be larger at GDP per capita $\geq 15,000$ USD than the HICs. This indicates that potentially BC-emitting activity will be a larger portion of total economic activity at GDP per capita $\geq 15,000$ USD in China than in the HICs, in addition to being larger in an absolute sense.

The above decomposition of S2011’s results suggests that China’s requirement of faster-than-historical emissions controls at GDP per capita $\geq 15,000$ USD is at least partially
a result of China having a larger proportion of potentially BC-emitting economic activity at that income level than do the HICs. Disproportionate BC-emitting economic activity contributes to the need for disproportionately aggressive emissions controls, which may affect the magnitude of technological and regulatory transitions that China will need to make relative to the HICs.

### 5.2.3 The role of export growth in China’s emissions future

A crucial next question is: what is the source of this larger proportionality of potentially black carbon emitting activity in China’s economy? We argue that the presence and projected growth of potentially black carbon-emitting activity associated with the production of goods for export offers a compelling explanation for why China’s economy in 2050 and onward is predicted to have a larger proportion of potentially BC-emitting activity than the HICs and why it must achieve better-than-HIC emissions factors to offset that additional activity. As discussed in Section 1, current flows of black carbon emissions embodied in trade demonstrate that the HICs have satisfied current per capita consumption at their income level partially by outsourcing black carbon-emitting activities to other places—mainly, China. Projections of China’s global export share suggest that this pattern will be maintained through a substantial portion of the 21st century. If China is to achieve the same air quality trajectory as the HICs, it will therefore need to either control or outsource both the potentially BC-emitting activity necessary to meet its own domestic consumption with increased income and that necessary to meet continued external demand. The better-than-HIC emissions factors that it must achieve are thus needed in order to offset the additional potentially BC-emitting activity required to support exports.

HICs support some portion of their consumption via BC-emitting economic activity that occurs in other nations, as can be seen via the difference between their production- and consumption-based BC emissions. Consumption-based emissions, i.e. all emissions associated with the production of goods consumed within a region, can be broadly thought of as
representing the amount of emissions required to meet consumption at that region’s income level [e.g. Davis and Caldeira 2010; Peters et al. 2011]. We use the United States (US) as a representative member of the HICs to analyze the consumption-based BC emissions necessary to support consumption at GDP per capita ≥ 15,000 USD. Over the 2000s, the US’s consumption-based black carbon emissions were 15-20% higher than its production-based black carbon emissions [Lin et al. 2014], i.e. those emissions occurring within national borders. This indicates that some portion of the potentially BC-emitting economic activity required to meet the US’s consumption is housed outside of the US. Analysis of black carbon embodied in trade by Lin et al. [2014] demonstrates that a majority of the US’s externally housed BC-emitting activity occurs in China, even when adjusting for the relatively high BC emissions intensity of China’s economy.

Trade-related black carbon flows have not yet been determined at the global scale. However, because black carbon and CO$_2$ are frequently coemitted in industrial processes, we can use global CO$_2$ embodied in trade to provide further support for the assertions (1) that China’s export-related emissions are uniquely large and (2) that HICs’ consumption-based emissions are larger than their production-based emissions. China is the largest net exporter of CO$_2$ emissions by an order of magnitude (1147 Mt CO$_2$ yr$^{-1}$ compared to Russia’s 2nd largest 286 Mt CO$_2$ yr$^{-1}$) [Davis and Caldeira 2010]. That is, the CO$_2$ emitted by the production of goods that China exports outweighs the CO$_2$ emitted by the production of goods that China imports by 1147 Mt CO$_2$ yr$^{-1}$. As with BC (see Section 5.4), this is partially due to the fact that China exports much more goods and services than it imports and partially due to its higher CO$_2$ emissions intensity of production compared to its trade partners [Davis and Caldeira 2010]. Nonetheless, it is clear that export-related economic activity contributes an exceptionally large portion of China’s total emissions. Major countries defining S2011’s HICs (the US and UK, Japan, Germany, France, Italy, and Spain) are also those with the largest gap in consumption-based versus production-based emissions.
of CO₂, demonstrating that a large portion of the CO₂ emissions required to satisfy their consumption is housed elsewhere.

This trade structure can be expected to hold through much of the RCP time horizon. The version of the GCAM model used in the RCP reference scenario does not contain a representation of imports and exports outside of the energy and agricultural sectors, and does not explicitly project changes in trade. Trade evolution in GCAM will, therefore, generally follow current conservative growth estimates. Anderson and Strutt [2016] use the Global Trade Analysis Project (GTAP) economy-wide model [Hertel, 1997] to project global trade through 2030 for a range of growth assumptions for China. Their results indicate that China’s share of global exports remains stable, even under conservative growth assumptions, suggesting China’s future export activity will scale with global GDP in the medium-term.

Based on the expectation in the GCAM projections that China’s export share will remain constant, we construct a simple model combining China’s current BC emissions data from Lin et al. [2014] with GDP growth data from the RCP 4.5 trajectory to provide a first order quantification of potential future export-related BC-emitting activity in China. We use year 2005 BC embodied in export as a proxy for current export-related activity, following the input-output analysis of Lin et al. [2014]. We then project China’s future potentially BC-emitting activity associated with trade into the future by scaling current exports with future global GDP to approximate the GCAM behavior. For consistency with GCAM results, we use the expected global GDP growth from the RCP 4.5 reference scenario [Thomson et al., 2011] to scale China’s year 2005 export-related BC-emitting activity to 2050 and 2095 (Table 5.1).

We first use the simple model to analyze how total BC emissions would be influenced by the projected increase in export-related activity in the absence of any improvements in emissions factor. The production of goods for export in 2005 resulted in approximately 223 Gg of BC out of China’s total 1545 Gg of BC emitted. Scaling this by the ratio of year 2005 global GDP to future global GDP under RCP 4.5 assumptions, this yields an increment in
Table 5.1: Values connecting China’s export-related black carbon emissions to its need for faster-than-historical emissions improvements are shown. Global Gross Domestic Product (GDP) from Thomson et al. [2011] is used to project China’s black carbon (BC) emissions embodied in export (EEE) at fixed year 2005 emissions intensity (EI). Future Chinese pre-EKC emissions intensity (pre-EKC EI) is used to calculate future EEE, which is comparable to the pre- to post-EKC difference in BC emissions (ΔBC).

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<th>2005</th>
<th>2050</th>
<th>2095</th>
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<tbody>
<tr>
<td>Global GDP (10^{12} $2000)</td>
<td>45</td>
<td>120</td>
<td>330</td>
</tr>
<tr>
<td>Chinese BC EI (g/$2000)</td>
<td>0.33</td>
<td>0.087</td>
<td>0.018</td>
</tr>
<tr>
<td>BC EEE (Gg/yr) at year 2005 EI</td>
<td>223</td>
<td>594</td>
<td>1634</td>
</tr>
<tr>
<td>BC EEE (Gg/yr) at pre-EKC EI</td>
<td>–</td>
<td>144</td>
<td>87</td>
</tr>
<tr>
<td>Pre- to Post-EKC ΔBC (Gg/yr)</td>
<td>0</td>
<td>160</td>
<td>110</td>
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Total BC emissions due to increased export-related activity of \( \Delta BC_{2050}^{\text{tot}} \) = 594 Gg yr\(^{-1}\) in 2050 and \( \Delta BC_{2095}^{\text{tot}} \) = 1634 Gg yr\(^{-1}\) in 2095 (Table 5.1).

We next analyze what these emissions would be if subject to the “semi-Kuznets” emissions factor improvements in S2011’s pre-EKC scenario. The pre-EKC scenario contains emissions factor improvements that scale with income at the same rates as they did for the HICs. They can therefore be construed as the emissions factors that would be needed to achieve HICs-consistent PM\(_{2.5}^*\) levels if China’s economic evolution were similar to that of the HICs, i.e. if China were only mitigating those BC emissions associated with its own consumption at a given income level. The more aggressive emissions factor improvements required in the post-EKC scenario are the result of some additional potentially BC-emitted activity (Section 5.2.2), which we argue arises from China supporting both the BC-emitting activity associated with its own consumption and that associated with foreign consumption through exported goods. The difference in emissions factors between the pre-EKC and post-EKC scenarios, therefore, can be construed as that necessary to offset future export-related BC-emitting activity. If this argument holds, the difference in BC emissions between the pre- and post-EKC scenarios should be similar to the future amount of BC emissions embodied in export under pre-EKC emissions factor improvements, since these would be the additional
emissions that would need to be offset by the pre-to-post-EKC increment in emissions factor improvements.

We can translate the future export-related BC emissions from our simple model into the portion of BC emissions they would comprise under the pre-EKC scenario’s emissions factors. We aggregate the sector-specific emissions factors into a Chinese economy-wide BC emissions intensity in grams of BC emitted per unit of economy activity in nonresidential sectors (Table 5.1). This value is 0.33 g BC $2000^{-1}$ in 2005, 0.087 g BC $2000^{-1}$ in pre-EKC 2050 and 0.018 g BC $2000^{-1}$ in pre-EKC 2095 (where $2000$ refers to year 2000 US dollars [Clarke et al., 2007]). Adjusting our future export-related BC emissions for the pre-EKC emissions factor improvements yields export-related BC emissions of $\Delta BC_{2050}^{tot} = 144 \text{ Gg yr}^{-1}$ in 2050 and $\Delta BC_{2095}^{tot} = 87 \text{ Gg yr}^{-1}$ in 2095 (Table 5.1). These projected export-related emissions are comparable with GCAM’s pre- vs. post-EKC emissions difference of 160 Gg yr$^{-1}$ in 2050 and 110 Gg yr$^{-1}$ in 2095, which under our argument should represent the additional potentially BC-emitting activity in China necessary to meet external demand.

The above analysis indicates that export growth can explain the faster-than-historical emissions factor reductions that China must realize in order to achieve EKC-assumed air quality in the middle and late 21st century. In the presence of only “semi-Kuznets” emission factor reductions, excess BC emissions in China contribute to the formation of PM$_{2.5}^*$ values that are beyond those expected for its future income level. In order for China’s mid- and late-21st century air pollutant concentrations to be consistent with those historically found at its corresponding income level, it must attain emissions factor reductions larger than expected at those same income levels, suggesting an additional activity level in China’s case that was not present in the historical patterns. We have shown that increased activity from GDP-scaled export growth will produce BC emissions in China in 2050 and 2095 nearly equivalent to the difference between the pre- and post-EKC emissions. Thus, increasing BC-emitting activity driven by continued foreign demand for goods may help explain why China, and potentially other countries with a similar economic make-up, will need to achieve better emissions factors
with income than has been historically seen in order to achieve EKC-consistent air quality improvements.

5.3 Regional and Global Impacts of Future Export-Related Emissions in China

In the previous section, we demonstrated that emissions embodied in export may help explain China’s need for faster-than-historical BC emissions controls. In order to achieve the projected emissions reductions, this trade-related activity growth must be offset by techno-regulatory improvements more rapid with income than historically seen. However, is the unmitigated growth of these export-related emissions a sufficient concern to warrant offsetting? To address this question, we deploy the future unmitigated export-related emissions from China in 2100, calculated from the simple model in Section 5.2, into a global chemistry-climate model to determine their impact on health-detrimental surface concentrations of black carbon, regional radiative balance, and global climate forcing.

We perturb the Geophysical Fluid Dynamics Laboratory’s AM3 Atmospheric General Circulation Model [Donner et al., 2011] in year 2100 of the RCP 4.5 scenario with China’s GDP-scaled export-related black carbon emissions at the end of the 21st century. This regional total amount is distributed according to the current spatial pattern of BC emissions in China. We run this on a repeating annual cycle for 30 years to ensure that the land and atmosphere equilibrate to the perturbation and to establish a statistically robust signal. Sea surface temperatures and sea ice are derived from a RCP 4.5 simulation in the associated fully coupled CM3 model and repeat the year 2100 annual cycle for the duration of the run.

Table 5.2 summarizes the results of the model simulation. The annual-mean regional maximum surface concentration of black carbon increases by 0.83 µg m\(^{-3}\) or 29% over RCP 4.5 year 2100 background levels due to the export-related activity. This surface concentration increase is equivalent to approximately one third of the BC surface concentration.
Table 5.2: Annual-mean results in year 2100 of simulations comparing the effects of standard RCP 4.5 emissions (RCP 4.5 Baseline) with a scenario including unmitigated growth of China’s export-related BC emissions (Export Growth). The 95% confidence interval on the annual-mean result, produced by model internal variability, are also shown.

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<th></th>
<th>Export Growth</th>
<th>RCP 4.5 Baseline</th>
<th>Growth-Baseline</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>China regional-mean</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Max. sfc. conc. (µg m(^{-3}))</td>
<td>3.68±0.12</td>
<td>2.85±0.16</td>
<td>0.83±0.12</td>
</tr>
<tr>
<td>Atmos. Abs. (W m(^{-2}))</td>
<td>92.9±0.76</td>
<td>85.5±0.68</td>
<td>7.4±0.88</td>
</tr>
<tr>
<td>Surface SW (W m(^{-2}))</td>
<td>190.7±3.8</td>
<td>197.2±4.6</td>
<td>-6.5±3.2</td>
</tr>
<tr>
<td>Global TOA ERF (W m(^{-2}))</td>
<td>–</td>
<td>–</td>
<td>0.16±0.13</td>
</tr>
</tbody>
</table>

decrease achieved in 2030 by the United Nations Environment Programme’s recommended BC mitigation measures [United Nations Environment Programme and World Meteorological Organization, 2011]. Epidemiological studies on the health effects of BC as distinct from PM\(_{2.5}\) are limited [Sasser, 2011]. However, BC contributes up to 15% of total PM\(_{2.5}\) mass, and increases in BC alone are thought to have roughly twice the effect size on mortality per unit mass than increases in PM\(_{2.5}\) overall [Zanobetti and Schwartz, 2009; United Nations Environment Programme and World Meteorological Organization, 2011; Janssen et al., 2012]. Long-term exposure to PM\(_{2.5}\) is estimated to increase mortality due to cardiovascular disease by 6-13% per 10 µg m\(^{-3}\) [World Health Organization et al., 2013]. Thus, the increased surface concentrations of BC due to export-related emissions in 2100 can be expected to have an impact on mortality on the order of a 1% increase. Because the health effects of an incremental change in PM\(_{2.5}\) depend nonlinearly on the baseline PM\(_{2.5}\) levels [Burnett et al., 2014], however, this estimation will potentially vary based on the abundance of other PM\(_{2.5}\) component species.

The regional climate impacts of black carbon manifest in East Asia primarily as a result of its interaction with shortwave radiation. Our simulations indicates that export-related emissions in 2100 decrease surface shortwave radiation over China by 4.3 W m\(^{-2}\) and increase atmospheric absorption of shortwave radiation over China by 5.0 W m\(^{-2}\) compared to the RCP 4.5 baseline. This redistribution of shortwave radiation between the surface and atmosphere has the potential to interfere with East Asian Monsoon circulation that provides
China with approximately half of its annual rainfall through a combination of mechanisms \cite{Menon et al., 2002, Huang et al., 2007}. In Chapter 4 we find that comparable redistribution of shortwave radiation reduces East Asian Monsoon summertime rainfall by 1.5% under present-day conditions by counteracting the climatological land-sea contrast that drives regional monsoon circulation.

Globally, black carbon is acknowledged to be a potent climate warmer. Its current top-of-atmosphere global-mean effective radiative forcing (ERF) is estimated at 0.64 W m\(^{-2}\) \cite{Myhre et al., 2013}, making it the third strongest anthropogenic warming agent behind CO\(_2\) and methane. Our simulation indicates that China’s export-related BC emissions in 2100 would result in an additional 0.16 W m\(^{-2}\) of global mean ERF beyond the RCP 4.5 baseline compared to pre-industrial times. This is comparable to the current ERF of nitrous oxide (0.17 W m\(^{-2}\)), a current focus of UNEP climate mitigation goals \cite{United Nations Environment Programme and World Meteorological Organization, 2011}. For further comparison, the direct, instantaneous radiative forcing (distinguished from ERF by the absence of fast cloud responses) of China’s current total BC emissions has been estimated at 0.09 W m\(^{-2}\) \cite{Li et al., 2016}. The global-mean radiative impact of China’s unmitigated export-related emissions is thus of a magnitude that warrants international interest.

5.4 Strategies for Mitigating Export-Related Emissions in China

In the previous sections, we showed that China’s export-related BC emissions create a need for faster-than-historical future emissions reductions and that their unmitigated growth constitutes a health and climate risk of considerable magnitude. Regional health and climate impacts due to unmitigated growth of these export-related emissions could mobilize domestic interest in their mitigation, and the positive global mean radiative forcing induced by those emissions are of a magnitude that has been shown to elicit international mitigation interest.
Here, we identify China’s export-related sectors in which black carbon emissions factors have
the greatest potential for reduction using existing technologies, and highlight political and
financial instruments that can be used to accelerate emissions factor improvements in these
sectors.

Approximately half of China’s black carbon emissions come from residential sources, with
industry and transport primarily accounting for the remainder [Lei et al., 2011; Lu et al.,
2011; Wang et al., 2012]. We exclude residential sector emissions from consideration of
export-related emissions reductions, as they are not strictly associated with the production
and transport of goods for export [Lin et al., 2014]. These emissions are a major focus of
mitigation efforts, however, due to their acute health effects [United Nations Environment
Programme and World Meteorological Organization, 2011], and have declined in China over
the past two decades [Wang et al., 2012].

Among export-related sectors, coke (17.3%) and brick (7.3%) production are the largest
contributors to total BC emissions. Diesel combustion in industrial (3.4%), agricultural
(2.8%), and vehicular (8.0%) applications is another major contributor [Wang et al., 2012].
BC emissions intensity and total emissions in these sectors show a range of trajectories [Lin
et al., 2014]. The emissions intensity of coking processes showed a 40% reduction between
2002 and 2007, but both brick and coke production total emissions increased during that
period [Lei et al., 2011]. BC emissions intensity in the transportation sector has stayed
constant and relatively high over the same period and is among the highest of export-related
sectors [Lin et al., 2014].

The emissions intensity of China’s production processes is generally higher than that
of its trade partners [Lin et al., 2014]. Its economy-wide emissions intensity is 15-60 times
higher than that of its major trade partners, the United States, European Union, and Japan,
and 5 times higher than the global-mean. Some of this high intensity is indicative of China’s
generally more manufacturing-driven economy [e.g. Guan et al., 2009], but it is also indicative
of the availability of less emissions-intensive technology practices for China’s currently high-emitting sectors.

This availability of lower-emitting technologies can be captured to the first order by differences in the emissions factors in various sectors between China and its trade partners. Table 5.3 shows the percent difference in PM$_{2.5}$ emissions factor in China versus Germany for various export-related, high-emitting sectors from the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) Model [Amann et al., 2011]. The mass ratio of BC in PM$_{2.5}$ emissions varies across sectors [Lei et al., 2011], but the percent difference in emissions factor for a given sector will be the same for PM$_{2.5}$ as for BC, assuming the mass ratio for a given process stays fixed.

Major BC emissions sources, like coke and brick production, exhibit a large percent difference in emissions factor, indicating the availability of lower-emissions technologies than those currently in use in China. The transportation sector also shows the capacity for large improvements in emissions factor, particularly from gasoline-fueled light duty vehicles and natural gas-fueled buses and other heavy-duty vehicles. However, opportunities for lower emissions intensity exist across the industrial, power, and transportation sectors (Table 5.3).

Domestic structures exist that can accelerate the implementation of available emissions reductions. Dasgupta et al. [2002] suggest that economic liberalization has been key in encouraging transitions in sectoral composition that tend to reduce pollutant emissions. This occurs primarily via reduced government subsidization of typically high-emitting sectors and a transition toward larger and, therefore, generally more efficient scales of production [Wheeler, 2001]. However, economic liberalization must be coupled with increased environmental regulation to ensure that total emissions decrease rather than just emissions intensity. Environmental regulation as a structure for enforcing environmental liability can then mobilize other financial instruments, such as bank limitations on credit to high-polluting sectors [Dasgupta et al., 2002]. Economic liberalization also encourages the presence of multina-
Table 5.3: The differential between black carbon emissions factors in Germany versus China is shown for high-emitting, export-involved sectors for which German emissions factors are at least 10% lower than Chinese emissions factors. Ranges are presented where the percent difference is dependent on the fuel grade or control technology. Data is derived from the GAINS model [Amann et al., 2011].

<table>
<thead>
<tr>
<th>Activity</th>
<th>Difference in Emissions Factor (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coal in fuel production</td>
<td>40.4-61.7</td>
</tr>
<tr>
<td>Industrial combustion</td>
<td></td>
</tr>
<tr>
<td>Coal</td>
<td>48.5-80.1</td>
</tr>
<tr>
<td>Gasoline</td>
<td>96.0</td>
</tr>
<tr>
<td>Coke</td>
<td>80.6</td>
</tr>
<tr>
<td>Natural Gas</td>
<td>97.5</td>
</tr>
<tr>
<td>Heavy Fuel Oil</td>
<td>30.4-38.2</td>
</tr>
<tr>
<td>Liquefied Petroleum Gas</td>
<td>98.8</td>
</tr>
<tr>
<td>Diesel</td>
<td>77.5-86.3</td>
</tr>
<tr>
<td>Power Generation</td>
<td></td>
</tr>
<tr>
<td>Coal</td>
<td>42.6-60.5</td>
</tr>
<tr>
<td>Heavy Fuel Oil</td>
<td>44.6-87.5</td>
</tr>
<tr>
<td>Diesel</td>
<td>77.5-86.3</td>
</tr>
<tr>
<td>Light Duty Vehicles</td>
<td></td>
</tr>
<tr>
<td>Gasoline</td>
<td>79.0-97.8</td>
</tr>
<tr>
<td>Liquefied Petroleum Gas</td>
<td>50.0</td>
</tr>
<tr>
<td>Diesel</td>
<td>20.6-34.1</td>
</tr>
<tr>
<td>Buses and Heavy Duty Vehicles</td>
<td></td>
</tr>
<tr>
<td>Natural Gas</td>
<td>71.0</td>
</tr>
<tr>
<td>Diesel</td>
<td>17.4-44.0</td>
</tr>
<tr>
<td>Rail and Other Off-Road</td>
<td></td>
</tr>
<tr>
<td>Diesel</td>
<td>22.9-25.0</td>
</tr>
<tr>
<td>Production</td>
<td></td>
</tr>
<tr>
<td>Brick</td>
<td>60.0</td>
</tr>
<tr>
<td>Cement</td>
<td>97.5</td>
</tr>
<tr>
<td>Coke</td>
<td>58.4</td>
</tr>
<tr>
<td>Other (glass, sinter, pig iron, etc.)</td>
<td>35.1-80.1</td>
</tr>
</tbody>
</table>
tional firms, which may help to flatten technology gradients between the different economies in which they operate [Dowell et al., 2000].

However, it is possible that China could respond to manifest EKC-consistent air quality improvements without decreasing total export-related BC emissions. The emissions/income relationship embodied in the EKC assumption is largely based on increased preference for local air quality improvements as income increases. Three pathways thus exist for China to achieve EKC-consistent population-weighted air quality improvements given continued demand for exported goods: (1) improved technology efficiency; (2) relocation within China of emissions-heavy production away from population centers; (3) outsourcing of emissions-heavy processes to other countries. Only Pathway (1) achieves the global total BC emissions reductions assumed in the RCP scenarios. Pathway (2) is a strategy already being proposed as a solution to China’s urban pollution problems [Stanway, 2014], and global flows of CO$_2$ embodied in trade suggest that Pathway (3) may already be underway [Davis and Caldeira, 2010]. The intra- and inter-national emissions leakage present in Pathway (2) and Pathway (3) are most likely to be prevented by a focus on domestic capacity-building and technological improvement [e.g., Jacoby et al., 1997; Bruvoll and Fæhn, 2006; Peters and Hertwich, 2008].

5.5 Discussion

Our analysis provides an explanation for why China must achieve faster-than-historical emissions factor improvements in order to attain EKC-consistent air quality through the end of the 21st century. However, the use of EKC-determined emissions trajectories in IAMs can also be interrogated from the perspective that developing countries need not hazard the environmentally detrimental upslope of the EKC at all. Zhang et al. [2015] suggest the possibility of developing countries “tunneling through” the EKC and reaching the downslope of the EKC without emissions levels climbing to the critical inflection point. This tunneling
would be made possible by aggressive transfer of efficient technologies and best practices from low to high emissions economies.

Our analysis is not inconsistent with the possibility of such emissions leapfrogging; indeed, from an emissions factor perspective, China’s faster-than-historical emissions factor improvements in the post-EKC iteration of S2011 constitutes precisely this sort of technological leapfrogging. Rather, we highlight the growth of export-related activity as an explanation for why this sort of leapfrogging is necessary in order for China’s future air quality to be EKC-consistent or better. Our work suggests that China’s export activity growth will be an additional impediment to any attempts to tunnel through the EKC.

In general, IAMs’ approach of constraining future air quality to EKC-consistency has been criticized on the grounds that the slope and critical inflection point of the EKC are extremely variable across different pollutants and diverse national economies [Cavlovic et al., 2000; Stern, 2003; Dinda, 2004]. In fact, investigation of whether or not an EKC exists for black carbon emissions has been extremely limited and inconclusive [Cavlovic et al., 2000]. It is generally acknowledged, therefore, that there is not strictly a single EKC in the way that IAMs often treat it [Smith et al., 2011b]. By analyzing one pollutant—black carbon—in one high-emitting nation—China—we are able to construct a mechanistic explanation for how and why China’s emissions future may deviate from the generalized EKC curve and to demonstrate that this deviation has climate and health consequences of a magnitude that must be reckoned with.

Our projection of China’s export-related BC emissions adopts GCAM’s assumption of relatively fixed global export shares and, hence, scales current export-related BC with the RCP 4.5 projected global GDP for consistency. However, there is substantial uncertainty regarding future GDP growth and even currently reported GDP that may result in different outcomes in the real world. The GCAM assumption of fixed global export shares likewise may not be upheld. There is already evidence that China’s export market penetration rate is declining due to increasing production costs [e.g., Mao and Zhang, 2015], with domestic
production turning more toward domestic markets [e.g. Yang, 2014]. Economic data from the World Bank, however, indicates that China’s exports grew at a rate faster than global GDP between 2000 and 2012 [World Bank Group, 2012]. This suggests that a global-GDP scaling of China’s future export activity may be a reasonable median assumption. More precise simulation of potential future changes in the long-term export-driven share of China’s economy would be most robustly conducted in an integrated assessment model or generalized economic model. However, these would be subject to the same complex assumptions that we seek to interrogate here. We opt in our analysis, therefore, for a simplistic, but thoroughly transparent, approach that provides an order-of-magnitude quantification with easily traceable sensitivities.

We offer the growth of export-related potentially BC-emitting activity as an explanation for the discrepancy between the BC emissions in the pre- and post-EKC iterations of the GCAM reference scenario. However, there may be other contributors to the difference between the two iterations. Baseline (i.e. year 2005) emissions were slightly modified in certain regions between the pre- and post-EKC iterations, suggesting that uncertainties in current emissions databases may play a role in explaining the need for China’s perceived faster-than-historical emissions factor improvements. S2011 did not modify baseline emissions over China between the pre- and post-EKC iterations (Table 5.1), however, so this baseline uncertainty is unlikely to offer a complete explanation for the discrepancy in this context.

However, there has been substantial recent debate on the accuracy of China’s reported activity levels, emissions factors, and total emissions that highlights the ongoing challenges associated with national monitoring, reporting, and verification of air pollutant emissions. This may affect the accuracy of the baseline Chinese emissions used in GCAM. For example, gaps in coal use reporting in China were revealed to have resulted in a 17% underestimation in China’s coal use in 2013 and a 4-6% underestimation in carbon dioxide emissions [Buckley, 2015]. Simultaneously, an alternative approach found China’s energy consumption during...
the 2000s to be 10% higher than national statistics, but emissions factors to be 40% lower than commonly used values, resulting in a downward revision of China’s carbon dioxide emissions by 14% compared to existing inventories [Liu et al., 2015]. This baseline emissions uncertainty may affect the accuracy of the current export-related emissions derived from Lin et al. [2014] and used in our emissions projection. Given that this inaccuracy will be present in both the S2011 data used to demonstrate China’s EKC-inconsistency and the Lin et al. [2014] data extrapolated to determine export-related activity growth, it does not substantially detract from our argument that the latter can explain the former, but is uncertainty that should be addressed in future.

GCAM is only one of several integrated assessment models used to develop future emissions scenarios, each of which offer subtly different formulations of the enviro-economic systems they are intended to capture. Different IAMs were used in the development of each of the RCPs, and each RCP involves different assumptions about future GDP, population growth, and other exogenous factors [van Vuuren et al., 2011]. We focus on GCAM and the RCP 4.5 reference scenario here, given the accessibility of data on its implementation of the EKC assumption and the ubiquity of RCP 4.5’s use in analyses of future climate change impacts. The various RCP scenarios, however, are not assigned relative likelihoods, and other model configurations may reveal a different role for China’s export-related emissions. Future research should endeavor to explore this behavior in each of the IAMs and associated RCPs to determine whether it is a pervasive feature of China’s BC emissions evolution. Additionally, the possible importance of China’s export-related economic activity to its future emissions trajectory should be factored into the development of future scenario generations, such as the Shared Socioeconomic Pathways [O’Neill et al., 2015].

The results presented here are based on the assumption in many current IAMs that global export structure remains relatively fixed into the future. A more sophisticated treatment of trade in IAMs may produce projections of China’s future economic make-up in which it phases out export-related activity as income increases. However, the consumption-based
emissions of currently high-income countries demonstrate that the potentially BC-emitting economic activity to satisfy consumption must occur somewhere. A Chinese phase-out of export-related activity would, therefore, require its export-related activity (and potentially some portion of the economic activity to support its own consumption) to be relocated to other regions. Unless there occurs a technological transformation that eliminates the need for potentially BC-emitting activity in the generation of goods and services, the potentially BC-emitting activity required to satisfy high-income countries’ consumption must needs occur somewhere, and the associated greater-than-historical emissions controls will need to be accomplished in that eventual location, even if they do not ultimately need to be achieved in China.

This work focuses on China’s future emissions of black carbon, as both a potent climate warmer and a human health risk. Most combustion processes, however, co-emit black carbon with other aerosol types [e.g. Lin et al. 2014]. The net climate effect of evolving industrial activity may be different when considering all co-emitted pollutants than when considering BC alone, but the exact nature of this difference is uncertain. BC is often co-emitted with less strongly absorbing species, like organic carbon, which may reduce the net climate warming effect of the industrial process [e.g. Kopp and Mauzerall 2010]. However, there is increasing evidence that co-emitted aerosol species rapidly mix and agglomerate in the atmosphere, resulting in hybrid aerosol particles that have optical properties that are a combination of those of their component species [e.g. Bond et al. 2013]. In the presence of black carbon, thus, co-emitted species can ultimately contribute to a larger volume of partially absorbing aerosols, which then increases the overall amount of energy trapped [Persad et al. 2014]. Due to these uncertainties, we focus on a single species here, but a valuable extension of this work would involve analyzing all co-emitted aerosol species from export-related activity in tandem.

Likewise, black carbon is not the only contributor to PM$_{2.5}$ concentrations. Sulfate and nitrate aerosol droplets, aerosolized crustal material, and organic carbon all contribute in
varying proportions to PM$_{2.5}$ concentrations, depending on the emitting activity. The overall health effects of PM$_{2.5}$ are sensitive to the relative abundance of each of these species in the overall concentration \cite{Zanobetti2009}. Further, different health effects are sensitive to whether exposure is acute or extended, and incremental health impacts of PM$_{2.5}$ increases are nonlinearly dependent on the baseline PM$_{2.5}$ concentrations experienced by a population \cite{UNEP2011, Burnett2014}. These factors combine to make the health effects of future export-related BC emissions highly uncertain. We provide in Section 5.3 the health-relevant maximum PM$_{2.5}$ surface concentrations induced by the unmitigated export-related BC emissions growth. Valuable additional insight could be gained, however, by analyzing the health impacts of the BC emissions growth in Section 5.3 within a comprehensive epidemiological concentration-response framework \cite{Saikawa2009, Fang2013}.

## 5.6 Summary and Conclusions

Our results indicate that China’s production of goods for export creates a uniquely large, and potentially growing, source of black carbon-emitting activity that impedes China’s adherence to historical trajectories of air quality improvements with income seen in currently developed nations. The parameterization of emissions controls in China in the integrated assessment models used to project future emissions demonstrates that China would have to improve its emissions factors more rapidly with income than currently high-income nations did in order to meet historically income-proportional air quality levels (Section 5.2). This suggests that China will be subject to additional emitting activities in its future development not present along the development trajectories of currently high-income nations. We show that continued GDP-scaled growth of China’s export-related activity is sufficient to explain the larger emission factor reductions that China must achieve (Section 5.2), that their unmitigated growth poses climate and health risks capable of mobilizing domestic and
international interest in mitigation (Section 5.3), and that pathways exist for rapid emissions intensity improvements in export-related sectors (Section 5.4).

These findings have implications for the emissions trajectories captured in the Representative Concentration Pathways. EKC-consistent air quality may be desirable for the sake of economic consistency [Smith et al., 2011b], particularly when EKC-exceeding emissions do not have a clearly robust source. However, our results identify a concrete mechanism by which the EKC assumption may be too optimistic in the case of China’s BC emissions. We show that the unattainability of EKC-consistent air quality in China through endogenous means may be the function of a very real failure of the EKC to capture potential differences between the past and future BC emissions landscape, particularly the uniquely large portion of China’s emitting activities that are driven by foreign consumption. China’s higher air pollutant concentrations with income could thus become a reality rather than an economic inconsistency, in the absence of active offsetting of export-related emissions growth.

The geopolitical implications of these export-related emissions are multifold. From a climate change perspective, global benefit lies in China reducing its overall BC emissions rather than simply relocating them either away from populated regions in China or to other lower-income nations (Section 5.4), an admittedly difficult task. Domestic and international approaches to mitigating export-related emissions would therefore need to focus on strategies that avoid this potential leakage. Additionally, there is increasing pressure for the accounting systems used in international and mini-lateral environmental agreements to be calculated based on consumption- rather than production-based emissions [e.g. Peters 2008; Peters et al., 2009; Barrett et al., 2013; Bastianoni et al., 2014], making China’s internal export-related emissions increasingly the concern of its trade partners.

We focus our analysis in this work on China, due to the availability of data on BC embodied in export. However, the mechanism described here may be true for other regions that exhibit EKC-inconsistent future air quality. Other regions that show a discrepancy in S2011, such as India and Southeast Asia (Figure 5.2), also have a high export-to-import
ratio in carbon dioxide [Davis and Caldeira, 2010], which suggests that a large proportion of their BC emissions may also be driven by foreign consumption. These regions may therefore also be subject to the future increased export-related activity shown here for China.

Our results indicate that the relocation of emissions-intensive activities from high-income to lower-income countries through trade may restructure the landscape of future emissions trajectories compared to past emissions. As developing nations seek technological and regulatory breakthroughs necessary to improve air quality for their citizens, the role of foreign consumption in domestic air quality crises may become an increasing focus of geopolitical interactions. A comprehensive understanding of the contribution of export-related emissions to the need for steeper emission evolutions in developing countries, which we have provided an initial step toward here, may be an important element in those interactions.
Chapter 6

Summary and Conclusions

6.1 Summary and Implications

Human-caused emissions of aerosols have profound implications for the magnitude and distribution of climate change. Heterogeneous concentrations of these short-lived particles create horizontal gradients in aerosols’ radiative effects and associated thermodynamic and dynamical impacts between polluted and unpolluted regions. When these aerosol concentrations absorb radiation, they also redistribute energy vertically between the surface and atmosphere. This dissertation probes the climate and policy implications of this unique vertical and horizontal heterogeneity of anthropogenic aerosol forcing. It leverages two generations of the Geophysical Fluid Dynamics Laboratory’s atmospheric general circulation model, AM2.1 and AM3, to ask the following questions: How similar are the patterns of surface energy flux response to heterogeneously-distributed aerosol forcing versus homogeneously-distributed greenhouse gas forcing? How strongly do aerosols partition radiative energy between the atmosphere and surface through absorption in high-emissions regions like East Asia? And what effect does this have on regional climate? It also extends these scientific questions into associated policy implications via an analysis of how aerosols relocated to East Asia through trade affect assumptions about China’s emissions future.
Chapter 2 compares the atmosphere-and-land response to greenhouse gases and aerosols in the AM2.1 model. It identifies that, even when only atmosphere and land mechanisms that operate on month-long timescales are considered, the spatial structure of the climate response to greenhouse gases and aerosols (as manifested in the perturbation to the ocean surface energy flux) is substantially similar, though of opposite sign. The landmasses rapidly heat in response to greenhouse gases and cool in response to aerosols. This acts as a geographically fixed perturbation to the extratropical atmospheric circulation, which produces patterns of surface energy change that are relatively insensitive to the structure of the initial forcing. This work reveals that, even on the seasonal to annual timescales on which the land and atmosphere respond, aerosols and greenhouse gases can be expected to produce strongly correlated spatial patterns of change across a range of climate parameters. Our identification of the similar spatial pattern of change in the ocean surface energy flux set by land-and-atmosphere responses provides a valuable bridge to understanding the fully atmosphere-ocean coupled response spatial similarity identified by Xie et al. [2013].

Although patterns of surface energy flux response to greenhouse gases and aerosols show similarity over the oceans, localized aerosols can have profound and complex impacts on surface energy and climate at the regional scale, particularly when both their scattering and absorbing characteristics are appropriately captured. Chapters 3 and 4 highlight this behavior in the particularly high-emissions region of East Asia.

Surface-based observations indicate a significant decreasing trend in clear-sky downward surface solar radiation (SSR) over East Asia since the 1960s. This “dimming” has been explained in the literature by the region’s long-term increase in aerosol emissions [Wild, 2012], but little work has been done to quantify the underlying physical mechanisms or the contribution from aerosol absorption within the atmospheric column. Given the distinct climate impacts that absorption-driven dimming may produce, this constitutes an important, but thus far rather neglected, line of inquiry.
In Chapter 3, we analyze the model-simulated East Asian clear-sky SSR trends in AM2.1 and AM3. We also use the models’ standalone radiation module to examine the contribution from various aerosol characteristics in the two models (such as burden, mixing state, hygroscopicity, and seasonal distribution) to the trends. Both models produce trends in clear-sky SSR that are comparable to that observed, but via disparate mechanisms. Despite their different aerosol characteristics, the models produce nearly identical increases in aerosol absorption since the 1960s, constituting as much as half of the modeled clear-sky dimming. This is due to a compensation between the differences in aerosol column burden and mixing state assumed in the two models, i.e. plausible clear-sky SSR simulations can be achieved via drastically different aerosol parameterizations. Our novel results indicate that trends in aerosol absorption drive a large portion of East Asian clear-sky solar dimming in the models presented here and for the time periods analyzed, and that mechanistic analysis of the factors involved in aerosol absorption is an important diagnostic in evaluating modeled clear-sky solar dimming trends.

In Chapter 4, we build on the findings of Chapter 3 in analyzing how atmospheric absorption and surface solar dimming compete in driving the response of East Asian regional summertime climate to anthropogenic aerosols, which dominates, and why—issues of particular importance for predicting how East Asian climate will respond to projected changes in absorbing and scattering aerosol emissions in the future. We probe these questions using a combination of a realistic aerosol perturbation and idealized decompositions thereof that allow us to analyze the relative influence of absorbing aerosols’ atmospheric and surface-driven impacts on regional circulation and climate. We find that even purely absorption-driven dimming decreases summer monsoon precipitation by cooling the land surface, counteracting climatological land-sea contrast and reducing ascending atmospheric motion and on-shore winds, despite the associated positive top-of-atmosphere regional radiative forcing. Absorption-driven atmospheric heating does partially offset the precipitation and surface evaporation reduction from absorption-driven surface dimming, but the overall response to
aerosol absorption more closely resembles the response to its surface dimming than to its atmospheric heating. Our results provide a novel decomposition of absorbing aerosol’s impacts on regional climate and demonstrate that the response cannot be expected to follow the sign of absorption’s top-of-atmosphere or even atmospheric radiative perturbation.

The future evolution of aerosol emissions is both uncertain and possibly differentiated according to aerosol type. The global distribution of high-emissions regions has evolved over the 21st century [e.g., Takemura, 2012], and there is substantial debate concerning the ways in which this distribution will shift in the future [e.g., Nakicenovic and Swart, 2000; Moss et al., 2010; O’Neill et al., 2015]. Scattering sulfate and absorbing black carbon aerosol trends have been substantially different in the past—sulfate emissions have declined more rapidly than black carbon emissions in many regions—and may continue to evolve differently [Levy et al., 2008]. This dissertation provides insight on both aspects of this uncertain aerosol future. Chapters 3 and 4 illuminate new aspects of the crucial role that shortwave absorption by black carbon aerosol plays in affecting regional climate over East Asia and highlight the influence of the ratio of scattering to absorbing aerosols on regional climate impacts—understanding that will be crucial as the regional ratios of these species evolve into the future. Chapter 2 provides insight on how rapidly the climate system can smooth these regionally intense effects, regardless of their location. In the final chapter of the dissertation, we integrate this scientific information with its policy implications through an analysis of how the relocation of black carbon emissions through trade may affect the future of black carbon emissions in East Asia—providing perspective on the societal factors that contribute to the uncertainty in how future aerosols will be distributed.

Emissions of black carbon aerosols from China have increased by over 200% over the last 50 years, with negative implications both for human health and for regional and global climate. The Representative Concentration Pathway (RCP) emissions scenarios used by the Intergovernmental Panel on Climate Change all project reductions in China’s future black carbon emissions, but this decline is partially dependent on constraining the future
air quality evolution of developing nations to match the empirical historical relationship between air quality and per capita GDP in currently developed nations. Recent research has demonstrated, however, that a substantial portion of China’s black carbon emissions are driven by the production of goods exported for consumption elsewhere—an external demand for emitting activity that is substantially smaller in the currently developed nations on which the empirical relationship is based.

In Chapter 5, we argue using integrated assessment model output, atmospheric general circulation modeling, and emissions and economic data that: (1) the historical empirical air quality/wealth relationship does not emerge from anticipated technology- and regulation-driven changes in black carbon emissions in China (and other currently high-emitting regions); (2) China’s large share of export-related, emitting activities and their potential growth are a possible explanation for this inconsistency; and (3) the climate and health implications of unmitigated future export emissions are non-negligible. Together, these results indicate that the production of goods for export creates a uniquely large source of black carbon emissions in China, for which demand is projected to grow, and which must and can be actively offset to achieve the future emissions reductions empirically assumed in the RCPs.

6.2 Limitations and Future Work

In a general sense, the climate response to a radiative forcer can be thought of as emerging over several different timescales [e.g. Dickinson, 1981; Manabe et al., 1990; Andrews et al., 2010]: the “fast” response that manifests through equilibration of the atmosphere and land to the radiative perturbation; the intermediate response that manifests through the additional equilibration of the shallow ocean mixed layer; and the “slow” or equilibrium response that manifests through the full equilibration of the coupled atmosphere and ocean system, though overlap does exist between these timescales [e.g. Jarvis, 2011]. This dissertation primarily
deals with the fast response of the climate system, which manifests over months to years. However, the mixed-layer and deep ocean are a crucial component of the development of the multidecadal and centennial response to anthropogenic perturbations.

Nevertheless, it is important to understand comprehensively the evolution of the climate response at each of these timescales. Observations of the climate system necessarily capture the transient response of the climate to ongoing human perturbations and the signals therein will be an amalgamation of responses at different stages of evolution. Understanding the nature of the climate response at each of these stages, as a climate model allows, is thus important for unpacking observed climate signals. From a policy perspective, responses that manifests over months to years are especially valuable for validating the impacts of mitigation decisions, particularly with respect to short-lived climate forcers like black carbon. We choose to focus our attention on this facet of the response development in this work, but an interesting extension could involve an exploration of how the processes discussed herein evolve over intermediate and slow timescales.

The research in this dissertation is largely confined to developing understanding within a global climate modeling framework. The extent to which the real world will behave in the way that the models predict will depend on a multitude of factors, including the accuracy of the inputs to the model, parameterizations and approximations required in simulations of the complex and nonlinear earth system, and natural variability of the real climate. Models have many acknowledged deficiencies in simulating the effect of aerosols on climate for a variety of reasons [e.g. Flato et al., 2013 and references therein], as do observational systems [e.g. Hartmann et al., 2013 and references therein], and uncertainties in one often compound uncertainties in the other. A shortage of observational information on aerosol emissions sources, vertical distribution in the atmosphere, and in situ chemical and optical interactions create uncertainties in the aerosol properties and perturbations input into models [Boucher et al., 2013]. Within the models themselves, incomplete representation of the multiple processes by which aerosols are lofted, transported, and removed within the atmosphere creates biases
in model-derived aerosol concentrations \cite{Ginoux2006}. The difficulty of resolving the small scales on which aerosols and clouds interact and limitations in our physical understanding of the mechanisms behind these interactions further compound these uncertainties \cite{Boucher2013}.

However, validation has been done on the models used in this work, as detailed in Chapters 3 and 4, and there are many important aspects of the climate system that they have great fidelity at capturing \cite{Donner2011, Delworth2006}. Further, models are an ideal and powerful tool for studying the sort of hypothetical and mechanistic questions that drive this research. In the real world, it is challenging to separate cleanly the effects of greenhouse gases and aerosols or absorbing and scattering aerosol types, and difficult to probe sensitivities of observed signals to different mechanisms. Models also uniquely allow the examination of different possible futures. The use of models in this work allows us to conduct crucial systematic investigation of complex systems and to look forward into the future in ways that observations alone do not allow. The findings of this work can be greatly complemented, nonetheless, by observational and experimental techniques. For example, future flight campaigns over East Asia could validate the magnitude of regional aerosol absorption, which we have shown regional climate to be highly sensitive to.

A crucial question raised by this work is: how much does the global climate system “care” about where aerosol emissions are located? Hotspots of anthropogenic aerosol emissions have shifted continuously since pre-industrial times: from Western Europe in the late 1800s, to North America and Eastern Europe, to South and East Asia in recent decades. The Representative Concentration Pathways project that portions of the African continent and South America could be the next loci of increased aerosol emissions \cite{Takemura2012}. With this relocation of aerosol-emitting processes comes a relocation of the regional radiative, dynamical, and hydrological impacts of aerosol forcing, which may feed back onto the global climate differently depending on the region in which they occur. Has past relocation affected and
will future relocation affect the global distribution of climate impacts due to anthropogenic aerosols beyond the emissions region?

Chapters 3 and 4 demonstrate the intense regional climate perturbations that can be imposed by aerosols in situ. However, Chapter 2 suggests that the climate system is capable of smoothing out the heterogeneous nature of aerosol forcing, at least insofar as the surface energy budget is concerned. On the other hand, as discussed in Chapter 1, there is evidence that the global response in surface temperature and precipitation is sensitive to the location of an aerosol forcing [Shindell and Faluvegi, 2009; Shindell et al., 2010, 2012; Shindell, 2014]. At a more regional scale, the influence of remote aerosol forcing on local climate has been identified in several distinct regimes, for example in South Asian monsoon circulation [Bollasina et al., 2011], Australian rainfall trends [Rotstayn et al., 2007], and United States temperatures [Teng et al., 2012].

A comprehensive picture of the global pattern of response to aerosol emissions in different regions has not yet been attempted and would be a valuable extension of this work. Chapter 5 demonstrates the substantial impacts that the relocation of aerosols through trade can have on regional emissions trajectories, climate, and health. The establishment of a global map of robust linkages between aerosol concentrations in one region and climate responses in other regions could have profound implications for international trade, air quality control, and other mitigation decisions. Through such work, we may come to better understand how the ever-shifting global distribution of these short-lived and heterogeneous anthropogenic forcers, and the societal factors that determine it, will impact the coupled human-climate system.
Appendix A

Supplement to Chapter 2

Table A.1: Spatial correlations of TOA Effective Radiative Forcing and Surface Energy Flux Perturbations due to greenhouse gases versus aerosols for each of the two 3-year subsets of the data discussed in Chapter 2.

<table>
<thead>
<tr>
<th></th>
<th>Annual Mean</th>
<th>JJA</th>
<th>DJF</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1-3</td>
<td>4-7</td>
<td>1-3</td>
</tr>
<tr>
<td>TOA Effective Radiative Forcing</td>
<td>-0.38</td>
<td>-0.46</td>
<td>-0.33</td>
</tr>
<tr>
<td>Clear-sky TOA ERF</td>
<td>-0.15</td>
<td>-0.19</td>
<td>-0.13</td>
</tr>
<tr>
<td>Tropics (20S-20N)</td>
<td>-0.39</td>
<td>-0.45</td>
<td>-0.44</td>
</tr>
<tr>
<td>Surface Energy Flux Perturbation</td>
<td>-0.43</td>
<td>-0.51</td>
<td>-0.62</td>
</tr>
<tr>
<td>Tropics (20S-20N)</td>
<td>-0.36</td>
<td>-0.52</td>
<td>-0.54</td>
</tr>
<tr>
<td>NH Extratropics (20N-90N)</td>
<td>-0.62</td>
<td>-0.58</td>
<td>-0.35</td>
</tr>
<tr>
<td>SH Extratropics (20S-90S)</td>
<td>-0.29</td>
<td>-0.43</td>
<td>-0.75</td>
</tr>
<tr>
<td>Surface Radiative (SW+LW) Flux</td>
<td>-0.28</td>
<td>-0.31</td>
<td>-0.28</td>
</tr>
<tr>
<td>Surface Sensible Heat Flux</td>
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<td>-0.48</td>
<td>-0.59</td>
</tr>
<tr>
<td>Surface Latent Heat Flux</td>
<td>-0.39</td>
<td>-0.44</td>
<td>-0.56</td>
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</table>
Bibliography


Dai, A., Li, H., Sun, Y., Hong, L.-C., Ho, L., Chou, C., and Zhou, T. The relative roles of upper and lower tropospheric thermal contrasts and tropical influences in driving Asian


Donner, L. J., Wyman, B. L., Hemler, R. S., Horowitz, L. W., Ming, Y., Zhao, M., Golaz, J.-C., Ginoux, P., Lin, S.-J., Schwarzkopf, M. D., Austin, J., Alaka, G., Cooke, W. F.,


Koch, D. and Del Genio, A. D. Black carbon semi-direct effects on cloud cover: review and synthesis. Atmospheric Chemistry and Physics, 10(16):7685–7696, August 2010. ISSN 1680-7324. doi: 10.5194/acp-10-7685-2010. URL http://www.atmos-chem-phys.net/10/7685/2010/


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Randles, C. A. and Ramaswamy, V. Absorbing aerosols over Asia: A Geophysical Fluid Dynamics Laboratory general circulation model sensitivity study of model response to


Saikawa, E., Naik, V., Horowitz, L. W., Liu, J., and Mauzerall, D. L. Present and potential future contributions of sulfate, black and organic carbon aerosols from China to global


Stanway, D. Beijing shuts big coal-fired power plant to ease smog -Xinhua, July 2014. URL http://in.reuters.com/article/china-pollution-beijing-idINL4N0PY3FF20140723


Stouffer, R. J., Manabe, S., and Bryant, K. Interhemispheric asymmetry in climate response to a gradual increase of atmospheric CO2. *Nature*, 342(6250):660–662, December 1989. doi: 10.1038/342660a0. URL http://www.nature.com/nature/journal/v342/n6250/abs/342660a0.html


Takemura, T. Distributions and climate effects of atmospheric aerosols from the preindustrial era to 2100 along Representative Concentration Pathways (RCPs) simulated using the global aerosol model SPRINTARS. *Atmos. Chem. Phys.*, 12(23):11555–11572, December 2012. ISSN 1680-7324. doi: 10.5194/acp-12-11555-2012. URL [http://www.atmos-chem-phys.net/12/11555/2012/](http://www.atmos-chem-phys.net/12/11555/2012/).


