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

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Abstract.

Surface-based observations indicate a significant decreasing trend in clear-sky downward surface solar radiation (SSR) over East Asia since the 1960s. This “dimming” is thought to be driven by the region’s long-term increase in aerosol emissions, 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.

We examine experiments conducted in the Geophysical Fluid Dynamics Laboratory’s Atmospheric General Circulation Models, AM2.1 and AM3, in order to analyze the model-simulated East Asian clear-sky SSR trends. 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 por-
tion 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.
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 [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 [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 [Fröhlich and Lean, 1998; Willson and Mordvinov, 2003] and are evident under both all-sky and clear-sky conditions [Wild, 2009].

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 de-
creases 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 Wm$^{-2}$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 \cite{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 \cite[e.g. Norris and Wild, 2007, 2009; Ruckstuhl and Norris, 2009; Dwyer et al., 2010; Allen et al., 2013]{Norris:2007, Norris:2009, Ruckstuhl:2009, Dwyer:2010, Allen: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 \cite[e.g. Folini and Wild, 2011]{Folini:2011}. \citet{Stier: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 myriad climate impacts of aerosol absorption, an in-depth analysis of how much absorption models produce and via what particular mechanisms 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 we hope that the framework developed in this study will 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 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.

2. Methods

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 (GAMDT), 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 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 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 The GFDL Global Atmospheric Model Development Team (GAMDT) [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 [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.

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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.

2.2. Design of experiments

We examine a set of four 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: (1) A five-
member ensemble of experiments that include all forcings (ALL$_F$), i.e. anthropogenic (aerosols, greenhouse gases, 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.

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 radiation 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 (GAMDT) [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 in this paper are for 1990 minus 1970 conditions to provide trend-relevant results.

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 $-0.43 \pm 0.10 \text{ Wm}^{-2}\text{yr}^{-1}$ 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. Results
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 $22.5^\circ-40^\circ \text{ N and } 100^\circ-122.5^\circ \text{ E}$), 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 1). The proxy data may contain higher-than-realistic variability 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 4.4.

Over the entire time period, AM2.1 and AM3 have linear trends (calculated using least squares linear regression) of $-0.47 \pm 0.02 \text{ Wm}^{-2} \text{ yr}^{-1}$ and $-0.30 \pm 0.02 \text{ Wm}^{-2} \text{ yr}^{-1}$, respectively (Figure 1). These values are both comparable to the $-0.43 \pm 0.10 \text{ Wm}^{-2} \text{ yr}^{-1}$ 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 acknowledged to outperform others in their respective model generations in capturing the proxy observed trend over Asia [Dwyer et al., 2010; Allen et al., 2013].

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: $-0.036 \text{ Wm}^{-2} \text{ yr}^{-1}$, AM2.1: $-0.044 \text{ Wm}^{-2} \text{ yr}^{-1}$). 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: $-0.009 \text{ Wm}^{-2} \text{ yr}^{-1}$, AM2.1: $-0.15 \text{ Wm}^{-2} \text{ yr}^{-1}$) and strong brightening thereafter (AM3: 0.35 Wm$^{-2}$yr$^{-1}$, AM2.1: 0.36 Wm$^{-2}$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 2 and 3 show the results of the various
ensemble simulations (described in Section 2.1) for the two models. The natural forcing experiments (NAT) shows no significant trend in clear-sky SSR, nor do the WMGG. The AERO experiments, meanwhile, produce a trend in clear-sky SSR of $-0.23 \, \text{Wm}^{-2}\text{yr}^{-1}$ in AM3 and $-0.49 \, \text{Wm}^{-2}\text{yr}^{-1}$ 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\textsubscript{F} runs results from the fact that the trends in sulfate and black carbon column burden are larger in the ALL\textsubscript{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 4 and 5). Somewhat surprisingly, the two models produce almost identical increases in absorption ($\sim 0.16 \, \text{Wm}^{-2}\text{yr}^{-1}$), 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 $0.032 \, \text{Wm}^{-2}\text{yr}^{-1}$ in AM3 and $0.029 \, \text{Wm}^{-2}\text{yr}^{-1}$ in AM2.1, indicating that absorption is much stronger over East Asia than in the global mean.
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 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 Wm\(^{-2}\) for AM3’s aerosol concentrations, with an associated increase in atmospheric absorption of 4.3 Wm\(^{-2}\). When external (AM3\_EM) rather than internal mixing is used, however, the clear-sky SSR decrease is only 5.6 Wm\(^{-2}\) and the absorption increase is only 2.2 Wm\(^{-2}\). The difference (i.e. between AM3\_IM and AM3\_EM) in the absorption increase (2.1 Wm\(^{-2}\)) is larger than the difference in the clear-sky SSR decrease (1.3 Wm\(^{-2}\)), indicating that the increase in scattering from 1970 to 1990 is 0.8 Wm\(^{-2}\) less with internal mixing than with external mixing. The scattering is calculated simply as the difference between the clear-sky SSR decrease and the absorption increase, since any change in clear-sky SSR must come from either absorption or scattering. These perturbations are imposed on an annual mean clear-sky SSR of \(\sim 273\) Wm\(^{-2}\) in the absence of aerosols.

AM2.1’s baseline configuration (AM2\_EM), meanwhile, produces an annual mean clear-sky SSR decrease between 1970 and 1990 of 8.5 Wm\(^{-2}\) with an associated increase in atmospheric absorption of 3.1 Wm\(^{-2}\). When internal (AM2\_IM) rather than external mixing is used, the clear-sky SSR reduction rises to 10.3 Wm\(^{-2}\) and the absorption increase rises to 6.5 Wm\(^{-2}\). The clear-sky SSR reduction change (1.8 Wm\(^{-2}\)) is smaller than the change in the absorption increase (3.4 Wm\(^{-2}\)), again indicating that the scattering increase between 1970 and 1990 is reduced (by \(\sim 1.6\) Wm\(^{-2}\)) when internal rather than external
mixing is used. This indicates that internal mixing promotes aerosol absorption while suppressing aerosol scattering in the models’ radiative transfer calculations.

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 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 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 ∼3.3 Wm⁻², while it is only ∼1.9 Wm⁻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 ×10⁻⁵ kg m⁻² versus 6.37 ×10⁻⁶ kg m⁻² of sulfate, respectively, and 8.99 ×10⁻⁷ kg m⁻² versus 5.41 ×10⁻⁷ kg m⁻² of black carbon, respectively). This suggests

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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 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 2. Note that the AM3 runs contain the 97% relative humidity cap described in Section 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 may 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 oth-
erwise 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.

4. Discussion

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 a possible explanation for the similarity in the trend in absorptions 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 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, 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 (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.

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

(1)

The results of this calculation are shown in Table 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 ∼ 0.16 Wm^{-2}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

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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 4.3.

### 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 characteristics. 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].

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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 6). 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 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 “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, $t = \{1, 2, 3, \ldots, 12\}$:

$$BC' = \frac{\sum_{t=1}^{12} BC(t) \times S(t)}{\sum_{t=1}^{12} S(t)} \rightarrow \Pi = \frac{BC'}{BC} \quad (2)$$

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 $\Pi_{\text{max}} = 0.11$ and $\Pi_{\text{min}} = 0.052$. 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

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transfer calculations would be needed, however, to confirm this behavior in the context of absorption.

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 [Koch et al., 2009; Bond et al., 2013]. 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.,

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2006a; 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. [2006a], 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

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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 2). This result is quantitatively consistent with that of Stier et al. [2006a], as the mass ratio of sulfate to black carbon in AM3 and AM2.1 is closer to 5.5:1 (Table 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 3.2), of the relative burdens of black carbon and sulfate in our region of interest. Matsui et al. [2013] and Stier et al. [2006b] 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, ©2014 American Geophysical Union. All Rights Reserved.
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 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 homogeneous 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 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.
4.4. Limitations of observational comparison

Although the discussion in Section 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.

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.

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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 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 Asia 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.

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 aerosol interactivity, mixing state, and column burden.

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. 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.

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Donner, L. J., et al. (2011), The dynamical core, physical parameterizations, and basic simulation characteristics of the atmospheric component AM3 of the GFDL global cou-


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Table 1. A summary of salient differences in aerosol parameterization between AM2.1 and AM3.

<table>
<thead>
<tr>
<th>Feature</th>
<th>AM2.1 [GAMDT, 2004]</th>
<th>AM3 [Donner et al., 2011]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Emissions</td>
<td>Olivier [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></td>
<td>Prescribed from MOZART [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</td>
<td>Sulfate: to 97% RH</td>
</tr>
<tr>
<td></td>
<td>BC: no</td>
<td>BC: only when mixed</td>
</tr>
</tbody>
</table>
Table 2. A summary of the standalone radiation radiative transfer calculation results. Clear-sky SSR and absorption flux (Wm$^{-2}$) values shown are for 1990 conditions minus 1970 conditions. The ‘nohygro’ refers to the versions of each experiment with hygroscopic growth disabled. Normalized absorption ($10^6$ Wkg$^{-1}$) 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>
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</thead>
<tbody>
<tr>
<td></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>SSR (Wm$^{-2}$) nohygro</td>
<td>-6.9</td>
<td>-5.6</td>
<td>-10.3</td>
<td>-8.5</td>
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<tr>
<td></td>
<td>-5.2</td>
<td>-4.1</td>
<td>-8.1</td>
<td>-6.3</td>
</tr>
<tr>
<td>Absorption (Wm$^{-2}$) nohygro</td>
<td>4.3</td>
<td>2.2</td>
<td>6.5</td>
<td>3.1</td>
</tr>
<tr>
<td></td>
<td>3.8</td>
<td>2.3</td>
<td>6.0</td>
<td>3.3</td>
</tr>
<tr>
<td>Scattering (Wm$^{-2}$) nohygro</td>
<td>2.6</td>
<td>3.4</td>
<td>3.8</td>
<td>5.4</td>
</tr>
<tr>
<td></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$^{-6}$kgm$^{-2}$)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Black Carbon</td>
<td>0.541</td>
<td>0.541</td>
<td>0.899</td>
<td>0.899</td>
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<tr>
<td>Sulfate</td>
<td>6.37</td>
<td>6.37</td>
<td>12.4</td>
<td>12.4</td>
</tr>
</tbody>
</table>

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Figure 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 Wm^{-2}.
Figure 2. Clear-sky SSR anomaly in Wm$^{-2}$ is shown for various AM3 attribution experiments. The natural forcing (NAT, red) and greenhouse gas only (WMGG, green) 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).
Figure 3. Same as for Figure 2, but for AM2.1.
Figure 4. The clear-sky SSR (left axis, black) and atmospheric absorption (right axis, red) anomalies are shown for AM3. Increased absorption accounts for approximately one half of the decrease in SSR.
Figure 5. Same as for Figure 4, but for AM2.1. Increased absorption accounts for approximately one third of the decrease in SSR.
Figure 6. 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 East Asia in Wm$^{-2}$ for reference. The two models have significantly different black carbon seasonalities, which lead to different temporal correlations with the downwelling radiation.