1 Radiative forcing and climate response to projected 21st

2 century aerosol decreases

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13 Abstract

It is widely expected that global emissions of atmospheric aerosols and their precursors will 14 decrease strongly throughout the remainder of the 21st century, due to emission reduction 15 16 policies enacted to protect human health. For instance, global emissions of aerosols and their precursors are projected to decrease by as much as 80% by the year 2100, according to the 17 18 four Representative Concentration Pathway (RCP) scenarios. The removal of aerosols will 19 cause unintended climate consequences, including an unmasking of global warming from 20 long-lived greenhouse gases. We use the Geophysical Fluid Dynamics Laboratory Couple Climate Model version 3 (GFDL CM3) to simulate future climate over the 21st century with 21 22 and without the aerosol emission changes projected by each of the RCPs in order to isolate the radiative forcing and climate response resulting from the aerosol reductions. We find that the 23 24 projected global radiative forcing and climate response due to aerosol decreases do not vary 25 significantly across the four RCPs by 2100, although there is some mid-century variation, 26 especially in cloud droplet effective radius, that closely follows the RCP emissions and energy consumption projections. Up to 1 W m⁻² of radiative forcing may be unmasked globally from 27 28 2005 to 2100 due to reductions in aerosol and precursor emissions, leading to average global 29 temperature increases up to 1 K and global precipitation rate increases up to 0.09 mm d⁻¹. 30 However, when using a version of CM3 with reduced present-day aerosol radiative forcing (-

1.0 W m⁻²), the global temperature increase for RCP8.5 is about 0.5 K, with similar magnitude 1 2 decreases in other climate response parameters as well. Regionally and locally, climate impacts can be much larger than the global mean, with a 2.1 K warming projected over China, 3 Japan, and Korea due to the reduced aerosol emissions in RCP8.5, as well as nearly a 0.2 mm 4 d⁻¹ precipitation increase, a 7 g m⁻² LWP decrease, and a 2 µm increase in cloud droplet 5 effective radius. Future aerosol decreases could be responsible for 30-40% of total climate 6 7 warming (or 10-20% with weaker aerosol forcing) by 2100 in East Asia, even under the high 8 greenhouse gas emissions scenario (RCP8.5). The expected unmasking of global warming 9 caused by aerosol reductions will require more aggressive greenhouse gas mitigation policies 10 than anticipated in order to meet desired climate targets.

11 **1 Introduction**

12 The climate effects of atmospheric aerosols represent one of the most uncertain aspects of 13 current and future climate forcing and response estimates (Myhre et al., 2013). Whereas the 14 greenhouse gas warming influences on climate are relatively well understood, significant 15 questions remain regarding the magnitude, and in some cases, even the sign (cooling or 16 warming), of aerosol-climate interactions. Aerosol radiative forcing of climate can be split into two categories: the direct effect, in which atmospheric aerosols directly scatter or absorb 17 18 incoming solar radiation; and the indirect effect, in which aerosols modify cloud properties 19 which in turn affect the radiation budget. For a fixed amount of cloud water, a more polluted 20 air mass will have smaller and more numerous cloud droplets, leading to a larger surface area 21 and a brighter cloud (Twomey, 1977). This is known as the cloud albedo effect. In addition, 22 more aerosol pollution may also result in a longer cloud lifetime due to the tendency of 23 smaller droplets to remain suspended in the atmosphere longer (Albrecht, 1989), although this 24 cloud lifetime effect is not as well understood. On a global average basis, both the direct and 25 indirect effects tend to exert a net negative radiative forcing on present-day climate, opposing the positive forcing from greenhouse gases, with the total aerosol effective radiative forcing 26 estimated to be -0.9 W m⁻² (uncertainty range -1.9 to -0.1 W m⁻²) (Myhre et al., 2013). This 27 aerosol forcing has likely offset a significant portion of present-day CO₂ and other greenhouse 28 29 gas-induced climate forcing and subsequent global warming. Likewise, any changes in future 30 anthropogenic aerosols will have implications for the overall net impact on climate. Here we 31 evaluate the changes in global and regional aerosol burden, climate forcing, and climate 32 response due to future decreases in aerosol and precursor emissions as projected by all of the

Representative Concentration Pathways (RCPs). We build upon the work of Levy et al.
 (2013) to contrast the global and regional climate response and quantify the expected
 unmasking of warming due to future aerosol reductions across all of the RCPs.

4 Emissions of aerosols and their precursors have increased dramatically since the preindustrial 5 era, due to increasing industrialization and global population. In 2012, air pollution, mostly in 6 the form of atmospheric aerosols, was responsible for 7 million deaths (3.7 million from 7 ambient air pollution, 3.3 million from indoor) worldwide (WHO, 2014). Due to efforts to 8 reduce this enormous human health impact, aerosol and precursor emissions are expected to 9 decline worldwide over the next several decades as governments enact and enforce stricter 10 emission control policies. Emissions of sulfur dioxide (SO₂, a precursor to sulfate aerosol) 11 have already declined about 50% in North America and Western Europe and passed their peak 12 in 2005 in China (Klimont et al., 2013). In some developing countries, emissions are still rising but are expected to begin to decline within the next few decades, as a consequence of 13 14 increasing affluence and more environmental regulation (van Vuuren et al., 2011a).

15 While clearly beneficial for human health, declining aerosol emissions will result in the 16 unintended consequence of unmasking additional climate warming, due to the reduction of the cooling effects from anthropogenic aerosols such as sulfate and organic carbon (OC). Thus, 17 18 careful policy implementation is necessary in order to maximize reduction of unhealthy air 19 pollution while also minimizing the unmasking of additional global warming. Some studies 20 have pointed to reductions in black carbon as a possible approach to address this dilemma 21 (Bond et al., 2013; Kopp and Mauzerall, 2010; Shindell et al., 2012). Black carbon (BC) is an 22 aerosol species that is a strong absorber of incoming solar radiation in the troposphere and 23 thus a climate-warming agent. It is also a major contributor to PM_{2.5} and has an adverse effect 24 on human health (Bond et al., 2013). Despite a clear human health benefit, questions remain 25 whether its reduction will be an effective strategy for avoiding additional warming, due to its frequent co-emission with two strong cooling species, OC and sulfate (Chen et al., 2010; 26 Reddington et al., 2013). Some sources of BC are not large sources of species that exert a 27 28 negative radiative forcing (e.g. diesel fuel combustion), so it has been suggested that these 29 should be preferentially targeted for control first (Kopp and Mauzerall, 2010; Shindell et al., 30 2012).

31 Modeling studies have suggested that aerosols also have impacts on precipitation, cloud 32 cover, cloud droplet size and number, atmospheric circulation, and other climate parameters

(Lohmann and Feichter, 2005; Ming and Ramaswamy, 2009, 2011; Ming et al., 2011; 1 2 Ramanathan et al., 2001; Rosenfeld et al., 2008; Stevens and Feingold, 2009). An increase in aerosol emissions tends to decrease local to regional precipitation rates (warm rain), through 3 4 macrophysical and microphysical processes: 1) less incoming solar radiation penetrates the 5 troposphere and reaches the surface, resulting in less evaporation (Ramanathan et al., 2001) and 2) smaller and more abundant aerosols lead to smaller cloud droplets, which are less 6 7 likely to convert to rain drops via coalescence on the local and regional scale (Radke et al., 8 1989; Rosenfeld, 2000). An exception to (1) is BC or other absorbing aerosols, which may 9 have opposing effects on precipitation through heating the atmosphere (causing stabilization 10 and reduction in precipitation) and warming the surface via redistributed thermal energy 11 (causing an enhancement of precipitation) (Ming et al., 2010). However, the net effect of 12 increasing aerosol concentrations tends to be suppression of precipitation. Thus, the expected 13 reduction of aerosol concentrations should increase rainfall rates (warm rain) globally. Since 14 aerosols serve as seeds for virtually all liquid cloud formation in the atmosphere, decreases in 15 aerosols may also be expected to affect cloud cover, cloud liquid water path, and effective cloud droplet radius. 16

17 To estimate future aerosol emissions and burden, radiative forcing, and climate response, we 18 must utilize future projections or scenarios. The current state-of-the-art emissions scenarios 19 for global climate modeling are the Representative Concentration Pathways (RCP) (Lamarque 20 et al., 2011; Masui et al., 2011; Riahi et al., 2011; van Ruijven et al., 2008; Thomson et al., 21 2011; van Vuuren et al., 2012; Vuuren et al., 2011a, 2011b). The RCPs are different from previously developed scenarios in that they are initialized with radiative forcing beginning 22 23 and endpoints (2005-2100). A consistent but non-unique pathway based on literature and 24 integrated assessment modeling is then selected to match the prescribed endpoint. There are 25 four scenarios, each named for a radiative forcing endpoint in 2100 of 2.6, 4.5, 6.0, and 8.5 W m⁻² (RCP2.6, RCP4.5, RCP6, and RCP8.5). Each scenario contains emissions and/or 26 27 concentrations for all major greenhouse gases and air pollutants, including emissions of three 28 aerosol/precursor species: SO₂, OC, and BC. All RCPs assume an autonomous change in 29 future air pollution control policies in every world region, resulting in sharp decreases in 30 regional and global emissions of SO₂, OC, and BC.

There have been several previous studies on the effects of diminishing emissions of aerosols and their precursors on aerosol burden, radiative forcing, and climate (Arneth et al., 2009; Bellouin et al., 2011; Chalmers et al., 2012; Gillett and Von Salzen, 2013; Kloster et al., 2009; Lamarque et al., 2011; Leibensperger et al. 2012; Makkonen et al., 2011; Menon et al., 2008; Rotstayn et al., 2013; Shindell et al., 2013; Smith and Bond, 2014; Takemura, 2012; Unger et al., 2009). These studies and their results are summarized in Table 1. In order to have consistent comparisons to the present work, we focus Table 1 on recent studies that used a global climate modeling framework with the RCPs through 2100; hence, studies that may have used other scenarios are not included.

8 Recently, Levy et al. (2013) used GFDL CM3 model simulations of RCP4.5 to analyze 9 changes in radiative forcing, temperature, and precipitation driven by reductions of aerosol 10 emissions. To isolate the effects of decreasing aerosols, Levy et al. (2013) compared the 11 results of the RCP4.5 simulations with those of another set of simulations, in which all aerosol and precursor emissions were held fixed at 2005 levels throughout the remainder of the 21st 12 century. The authors found roughly an additional 1° C warming and a 0.1 mm d⁻¹ increase in 13 14 precipitation due to the decreasing aerosols in RCP4.5. Here we expand on the results from 15 the Levy et al. (2013) study by estimating the changes in global and regional aerosol burden, climate forcing, and climate response due to projected reductions in aerosol emissions for all 16 17 four RCPs using an updated version of the same GFDL CM3 model.

18 We first present the experimental design and the RCP emissions scenarios in Sect. 2. We then 19 go beyond the previous studies described in Table 1 by performing a more exhaustive and 20 robust analysis of the unintended climate consequences of reducing particulate air pollution 21 levels in the future. We also test the robustness of our results using sensitivity simulations 22 with a version of the CM3 model in which aerosol forcing is weakened significantly from its 23 default value (Golaz et al. 2013). We present historical to present day to future results from 24 1860 to 2100, focusing first on global changes (Sect. 3) and then on specific regions that may 25 be most strongly impacted (Sect. 4). We also compare our results with those from previous studies and examine similarities and differences in the projected aerosol-driven changes in 26 climate variables, climate forcing, and aerosol burden across the various RCPs. Finally, we 27 attempt to connect changes in aerosols with changes in forcing and climate parameters (Sect. 28 29 5). Conclusions are presented in Section 6.

1 2 Models and simulations

2 2.1 GFDL Climate Model 3

3 We use the Geophysical Fluid Dynamics Laboratory Climate Model version 3 (GFDL CM3) in this work. CM3 is a fully coupled chemistry-climate model containing atmosphere, ocean, 4 5 land, and sea-ice components. We employ the C48 version of the model, which uses a finitevolume cubed-sphere horizontal grid consisting of 6 faces with roughly a 200-km by 200-km 6 7 spatial resolution. Transport of tracers uses the finite volume algorithm of Lin and Rood 8 (1996) with updates as described by Putnam and Lin (2007) and Donner et al. (2011). The 9 vertical grid consists of 48 vertical levels extending from the surface up to about 0.01 hPa (80 10 km). Additional details on the model configuration and performance can be found in Donner et al. (2011), Naik et al. (2013), and references therein. Details on the ocean and sea ice model 11 12 can be found in Griffies et al. (2011).

13 Anthropogenic emissions of aerosols and their precursors (and emissions or concentrations of 14 all other reactive chemical species) are based on decadal estimates from Lamarque et al. (2010) for the historical period (1860-2000) and from Lamarque et al. (2011) for the RCP 15 projections (2005-2100). Concentrations of long-lived greenhouse gases are based on 16 Meinshausen et al. (2011). Source-specific emissions are provided for anthropogenic sources 17 18 (energy use, industrial processes, and agriculture), biomass burning, shipping, and aircraft 19 emissions. Since natural emission sources are not specified by Lamarque et al. (2010), we 20 follow the methodology described by Naik et al. (2013), including emissions of isoprene, soil 21 NO_x, lightning NO_x, marine primary organic aerosol (POA), DMS, dust, and sea salt. Changes 22 in climate do not feed back on natural emissions except for dust and sea salt, which respond to simulated wind speeds, and lightning NOx, which responds to convective activity. The lack of 23 24 temperature feedback on biogenic VOC emissions may lead to underestimates of future isoprene or other biogenic VOCs (Heald et al., 2008). Volcanic emissions are as described by 25 26 Donner et al. (2011).

The tropospheric chemistry component in CM3 is based on Horowitz et al. (2003) with updates from Horowitz (2006) and solves the reaction rate differential equations using an implicit Euler backward method solver with Newton-Raphson iteration. There are 97 total chemical species including 16 aerosol species. Tropospheric chemical reactions, including the NO_x-O_x-VOC system, are included in the model and fully coupled with the emissions and

atmospheric radiation sections of the model. There are a total of 171 gas-phase reactions, 41 1 2 photolysis reactions, and 16 heterogeneous reactions simulated in the model (Naik et al., 2012). Sulfate aerosols are formed via oxidation of SO₂ by the hydroxyl radical (OH), ozone 3 (O₃), and hydrogen peroxide (H₂O₂). The oxidation of DMS to sulfate aerosols is also 4 5 included. DMS emission from the oceans is parameterized based on 10-m reference height wind-speed but is independent of temperature. Carbonaceous aerosols are modeled in CM3 as 6 7 primary organic aerosols (POA), secondary organic aerosols (SOA), and BC. SOA includes 8 both natural and anthropogenic sources. Biogenic terpene oxidation is estimated to provide a directly emitted source of about 30.4 Tg C yr⁻¹ of SOA and butane oxidation by OH yields 9 roughly another 9.6 Tg C yr⁻¹ (Dentener et al., 2006; Naik et al., 2013; Tie, 2005). 10 11 Hydrophobic OC and BC aerosols are converted to hydrophilic aerosol with an e-folding time 12 of 1.44 days. Sea salt and mineral dust aerosol are treated with a five-section size distribution 13 ranging from 0.1 to 10 µm dry radius.

14 Aerosol optics in CM3 is based on Mie theory. Lognormal size distributions are assumed for 15 OC, BC, and sulfate. Sulfate and hydrophilic BC are assumed to be internally-mixed and to undergo hygroscopic growth, while all other aerosols are externally mixed. New to CM3 was 16 17 the addition of prognostic equations for calculation of cloud droplet number according to the 18 Ming et al. (2006) parameterization, allowing for variable cloud droplet number and radius. 19 The cloud lifetime effect is parameterized as an increase in cloud droplet number (e.g. due to 20 aerosol perturbations) resulting in a decrease in the autoconversion rate, thereby delaying 21 precipitation and increasing cloud lifetime. The derivation of the parameterization is beyond 22 the scope of this paper, but it follows the methodology of Khairoutdinov and Kogan (2000). 23 An autoconversion threshold of 8.2 µm is used in CM3. In a given grid box, if the volume-24 mean cloud droplet radius is less than this threshold, autoconversion is suppressed (Rotstayn, 25 2000). Sensitivity of the indirect effect to different thresholds in GFDL AM3 is explored in 26 Golaz et al. (2011). Despite being internally mixed with sulfate in the radiation calculation, 27 black carbon is assumed to be externally mixed with soluble species (sulfate, sea salt, OC) for 28 the aerosol activation calculation. Fractions of BC and OC are emitted as hydrophobic (80% 29 and 50%, respectively) but undergo aging to hydrophilic BC and OC with e-folding times of 30 1.44 and 2.88 days, respectively. Sulfate (treated as pure ammonium sulfate, internally mixed 31 with BC for optics), BC, and OC are assigned individual lognormal size distributions for both 32 the aerosol optics and activation code (Ming et al., 2007). Size-resolved aerosol microphysics are not included in the model due to computational demand. This lack of prognostic aerosol 33

1 number concentration may bias estimates of the effect of aerosol on clouds. Nonetheless, the 2 aerosol activation scheme has performed well, resulting in reasonable agreement with observations of both droplet size and droplet number concentrations (Donner et al. 2011; 3 Ming et al., 2006, 2007). Donner et al. (2011) showed that CM3 improved upon CM2.1 4 5 model-measurement evaluation metrics for several aerosol-relevant quantities, including aerosol optical depth, co-albedo, and clear-sky shortwave surface radiation flux. In the current 6 7 model configuration, neither the radiation nor the activation code currently include nitrate 8 aerosol. At present nitrate aerosols are estimated to have only contributed marginally to aerosol radiative forcing and climate effects. As nitrate may become a more significant 9 10 contributor to aerosol radiative forcing in the future (Bauer et al., 2007; Bellouin et al., 2011; 11 Hauglustaine et al., 2014), the chemical, radiative, and microphysical properties of nitrate 12 aerosol are being incorporated into a new version of the GFDL atmospheric model.

13 2.2 RCPs

14 The Representative Concentration Pathways (RCPs) contain emissions projections for all long- and short-lived climate forcers, including the aerosol and aerosol precursor species SO₂, 15 16 OC, and BC. The RCPs were featured in the Intergovernmental Panel on Climate Change (IPCC) Fifth Assessment Report (AR5) and are the successors to the Special Report on 17 Emissions Scenarios (SRES). The RCPs include globally gridded projections of emissions of 18 19 important atmospheric constituents from 2005 to 2100 with extensions to 2300. Using 20 literature and integrated assessment modeling (IAM), representative pathways are selected to 21 fit the individual RCP starting and ending points for radiative forcing. Concentrations and 22 emissions of short- and long-lived climate forcers (including aerosols) are then harmonized 23 (made consistent with base years) and emissions are downscaled (gridded on a latitude-24 longitude scale). Each of the chosen final pathways are not unique; however, a consortium of 25 experts from the IAM and IPCC communities have selected pathways with desirable qualities such as coverage of the entire literature range and significant spread in concentrations and 26 27 emissions between the individual pathways. The four pathways include a strong mitigation scenario (RCP2.6), two stabilization scenarios in which radiative forcing stabilizes shortly 28 29 after 2100 (RCP4.5 and RCP6), and one high emissions/low mitigation scenario (RCP8.5), 30 which current emissions most closely track. The radiative forcing pathways in each RCP are 31 internally consistent with concentrations of both short and long-lived climate forcers, with the exception of dust and nitrate aerosol forcing (Masui et al., 2011; Riahi et al., 2007, 2011; 32

1 Thomson et al., 2011; Vuuren et al., 2011a, 2011b). In addition to representing different GHG 2 emission trajectories, the RCPs implicitly assume air pollution reduction policies in which 3 emissions of reactive pollutants decrease as a function of increasing income, but 4 independently of GHG mitigation levels.

Figure 2 shows the global total emissions (anthropogenic and biomass burning) of SO₂, BC, 5 6 and OC for the historical period and for each of the RCPs (see Figs. S1 and S2 of 7 supplemental information for the split between anthropogenic and biomass burning). All four 8 scenarios project strong decreases in globally averaged sulfur dioxide (SO₂), organic carbon 9 (OC), and black carbon (BC) emissions throughout the 21st century. SO₂ emissions, according 10 to the RCPs, have already peaked globally (around 1980) and have been on the decline since, 11 except for a slight uptick in the early 2000s. By 2100, SO₂ emissions will have decreased from a 2005 level of around 120 Tg yr⁻¹ to 13-26 Tg yr⁻¹ (RCP2.6 to RCP8.5), about an 80% 12 decrease. RCP2.6 projects the largest decrease, followed by RCP4.5, RCP6, and RCP8.5. This 13 14 order is consistent with each RCP's embedded climate policy, since RCP2.6 has the most 15 stringent climate policy and RCP8.5 the least stringent, and many climate policies that curb CO₂ emissions can also curb SO₂ (e.g. replacement of coal-fired power plants with renewable 16 sources). However, this order does not hold for all species, on either the global and regional 17 18 scale. Air pollution control also should have an impact on the air pollutant emissions 19 trajectory; however, the assumptions regarding air pollution control were too similar across 20 each of the RCPs to wield noticeable influence (Rogelj et al., 2014). Thus, climate policies 21 (specifically reductions in CO₂ emissions from reduced dependence on coal energy) tend to 22 dominate the trend in not only emissions of aerosols and their precursors, but also in aerosol 23 optical depth, radiative forcing, and climate response, as we will show later. Although there is 24 some variability in global SO₂ emissions over the course of the time series, the RCPs all 25 converge on a fairly narrow range of endpoints. In particular, RCP6 and RCP2.6 stand out, the 26 former due to an increase in the rate of coal consumption around mid-century (2030-2060) 27 and the latter due to stringent climate policy including the nearly complete phase-out of non-28 CCS (carbon capture and storage) coal energy by roughly 2050 (Masui et al., 2011; van 29 Vuuren et al., 2011b). The increase in coal energy projected by RCP6 is a surprising feature 30 that is not present in the other RCPs. As a result, SO₂, BC, and OC emissions in RCP6 are 31 higher relative to the other RCPs over roughly the same time period in (Fig. 2). SO₂ emissions 32 briefly increase in absolute terms over a short period mid-century in RCP6, which drives 33 higher sulfate burdens, larger (negative) aerosol direct and indirect forcings, and noticeable

1 changes in climate response, as we will show in the following sections. The narrow range for 2 emissions of air pollutants and their precursors (e.g. SO₂) has been attributed to similar air 3 pollution policy assumptions in each RCP. New scenarios that strive to span the entire 4 literature range for air pollutants in addition to greenhouse gases are undergoing development 5 (Rogelj et al., 2014).

BC emissions (middle panel in Fig. 2) increased from preindustrial times and continue to increase in RCP2.6, peaking around 2010. RCP4.5, RCP6 and RCP8.5 all peak around 2005 (i.e. the beginning of the RCP projections) and decrease continuously until 2100. Present-day values of BC emissions are 8.5 Tg yr⁻¹. By 2100, BC emissions are projected to range from less than 3.3 Tg yr⁻¹ according to RCP2.6 up to 4.3 Tg yr⁻¹ in RCP6. Here, the expected order seen in the SO₂ emissions reductions is not observed, but BC emissions do also converge on a relatively narrow range of endpoints.

13 Emissions of OC, sometimes co-emitted with BC, have a similar trajectory (bottom panel Fig. 2). The main source of OC emissions in the RCPs is biomass burning, which makes up about 14 60% of the total emissions of OC in present day. RCP2.6 OC emissions continue to increase 15 in the early 21st century, peaking around 2020. Emissions of OC in RCP4.5 drop rapidly, due 16 17 to the strong decrease in cropland area and increase in forested area projected by RCP4.5 (in 18 other words, a decrease in biomass burning as a means to clear cropland), a trend that is mostly unique to RCP4.5 (van Vuuren et al. 2011a, Thomson et al., 2011). OC emissions in 19 RCP6 remain high throughout the 21st century due to the land use assumptions embedded in 20 RCP6, which includes shifts to larger amounts of burning to clear land for crops. CO₂ 21 22 fertilization also plays a role in the increased emissions, as higher CO₂ levels can increase 23 biomass growth and increase the amount available to be burned (Kato et al., 2011). Compared 24 to SO₂ and BC, the variability and range of emissions endpoints across the different RCPs is much wider, ranging from 20 Tg yr⁻¹ in RCP4.5 in 2100 to about 32 Tg yr⁻¹ in RCP6. The 25 26 final order of each RCP's global total OC emissions is different from that of both the BC and 27 SO₂ final order, highlighting the difficulty in comparison between different RCPs. In particular, the lack of climate policy in RCP8.5 has little influence on OC and BC emissions 28 29 in comparison to SO₂.

1 2.3 Simulations

2 We conduct simulations using GFDL CM3 to evaluate the role of changing aerosol emissions 3 on aerosol optical depth (AOD), aerosol radiative forcing, and climate response. Table 2 4 summarizes the simulations performed. A series of RCP simulations (denoted RCPx.x where x.x = 2.6, 4.5, etc.) were run from 2006-2100 in which the RCP emissions scenarios were 5 6 used for future aerosol, greenhouse gas, and other reactive species emissions or 7 concentrations. Another series of simulations were run using the RCPs, but with the 8 anthropogenic and biomass burning emissions of SO₂, OC, and BC held constant at 2005 9 levels. All other inputs, including greenhouse gases, were kept the same as in the full RCP 10 scenarios. We denote these fixed emission simulations as RCPx.x_2005AER, where the 2005AER signifies fixed aerosol and aerosol precursor emissions at 2005 levels. Each of the 11 RCPx.x and RCPx.x_2005AER simulations were run as a 3-member ensemble, each 12 initialized with different initial conditions, provided by another 3-member historical ensemble 13 (1860-2005) of GFDL CM3. The RCP4.5 and RCP4.5_2005AER simulations are not 14 scientifically different from those presented by Levy et al. (2013); however, our simulations 15 were run with a newer model version, which included some minor updates. We calculate the 16 17 aerosol-induced climate response as the difference between the two sets (RCPx.x – 18 RCPx.x 2005AER). Unless otherwise specified, all results presented are ensemble means. 19 Meteorological factors, such as increasing temperatures, changes in atmospheric circulation 20 and stability, and changes in precipitation will also induce changes in aerosol concentrations 21 (Dawson et al., 2007; Jacob and Winner, 2009; Leibensperger et al., 2012a; Pye et al., 2009; 22 Tai et al., 2012, 2010). The influences of greenhouse-gas driven meteorological changes are 23 included in both our RCPx.x and RCPx.x 2005AER simulations. Our methodology of running the RCPx.x_2005AER simulations for the full 21st century allows us to simulate the 24 25 climate-driven effects on aerosol abundance (AOD and concentration). Additionally, taking 26 the difference between RCPx.x and RCPx.x_2005AER isolates the changes in aerosols (and 27 climate response) resulting from emissions reductions alone, separate from the influence of 28 well-mixed GHG-driven climate changes. While these RCPx.x 2005AER simulations (or 29 analogues) were performed in some past studies, to our knowledge this is the first study to 30 present the results from such simulations for all four of the RCPs in a consistent framework.

In order to estimate aerosol effective radiative forcing, we ran additional non-coupled, atmospheric component-only simulations (AM3) in which sea surface temperatures (SST)

1 were held constant at the 1981-2000 climatological values. Simulations were run in which the 2 only climate forcings were aerosol emissions changes from present-day through 2100, allowing us to calculate the aerosol forcing by differencing radiative fluxes with a control 3 simulation using 1860 emissions and the same climatological SSTs. Using the fixed SST 4 5 approach allows us to estimate the radiative response to "fast" adjustments (time scale of a few years or less) to the climate system due to a forcing agent (e.g., aerosols) in addition to 6 7 the full response to the forcing agent itself. This method accounts for the indirect effects of 8 aerosols, including the cloud lifetime effect and the cloud albedo effect. This type of fixed-9 SST calculation has been referred to as the "radiative flux perturbation" (Lohmann et al., 2010) or "effective radiative forcing" (Myhre et al., 2013). This series of simulations are 10 11 denoted RCPx.x_RFP for the aerosol-only simulations and RCP_1860 for the control 12 simulation. This pair of simulations is only used for the radiative forcing calculations, while 13 the coupled runs described above provide AOD and climate response estimates.

14 As reported in Levy et al. (2013), the present-day aerosol effective radiative forcing (ERF) in CM3 of -1.7 - -1.8 W m⁻² falls close to the upper bound of the 90% confidence interval, -1.9 15 W m⁻², set by the IPCC. To test the robustness of our results, we conduct additional 16 17 experiments using an alternate model configuration with weaker present-day aerosol effective 18 radiative forcing. With a lower "starting point", future decreases in the magnitude of aerosol 19 forcing will also be reduced, likely resulting in a reduced climate response. We use the 20 "CM3w" experiment described by Golaz et al. (2013), in which cloud parameters, most 21 notably the autoconversion threshold, are tuned to produce the desired changes in climate 22 forcing. In this version, the present-day aerosol effective radiative forcing is around -1.0 W m⁻ 23 2 . To achieve this effect, we adjust the autoconversion threshold from its default value of 8.2 24 μm to 6.0 μm. Since a lower autoconversion threshold is not consistent with observations 25 (Rotstayn, 2000), we emphasize that this change is employed only as a means to test our 26 results with a lower magnitude present-day aerosol ERF, and is not meant to produce a more 27 scientifically accurate model version. The CM3w run is initialized from a historical run of CM3w, using one ensemble member each for RCP8.5 and RCP8.5 2005AER. These 28 29 simulations are denoted CM3w_RCP8.5 and CM3w_RCP8.5_2005AER.

30 3 Global analysis

We first focus our analysis on area-weighted global means ranging from pre-industrial times to 2100. We analyze aerosol optical depth (sulfate, BC, and OC), aerosol climate forcing

1 (direct and indirect), and climate response (temperature, precipitation rate, liquid water path, 2 and effective cloud droplet radius). In subsequent sections, the analysis is extended to examine spatial distributions and mean changes over specific regions. Past, present, and future 3 trends in global aerosol optical depth can be found in the supplemental information Sect. S2 4 5 as well as Figs. S3 and S4. In short, AOD trends are well correlated with emissions trends, with correlation coefficients ranging from 0.7 to 0.9 for each species and each RCP (not 6 7 shown) globally averaged, and 0.9 or greater over continental source regions. By 2100, sulfate 8 AOD globally is projected to decrease by 50% from 2005 levels in RCP2.6, 40% in RCP4.5 9 and RCP6, and 31% in RCP8.5 (see Supplemental Information). Figure S3 shows the 10 RCPx.x 2005AER fixed emissions simulations (dashed lines) along with the RCPx.x 11 decreasing emissions runs (solid lines) indicating that in the absence of emissions changes, 12 future climate changes cause AOD to increase globally for each of the RCPs. However, this 13 climate driven effect is small compared to the substantial decrease in AOD from emissions 14 reductions (compare dashed and solid lines in Fig. S3).

15 **3.1 Aerosol forcing**

16 3.1.1 Aerosol direct and indirect forcing

17 Future changes (mostly decreases) in aerosol emissions and aerosol amount and optical depth lead to changes in Earth's radiative balance. Figure 3 shows the globally averaged total top-18 19 of-atmosphere (TOA) aerosol effective radiative forcing (direct and indirect) for the historical 20 period and four RCP projections using the standard CM3 runs. From 1860 until present day, 21 the increasing abundance of atmospheric aerosols led to a larger (more negative) aerosol 22 forcing, peaking near present day. Preindustrial to present day aerosol forcing simulated by CM3 is about -1.8 W m⁻². This large negative forcing has offset or "masked" some of the 23 positive forcing from greenhouse gases. Although the net forcing is still positive, without the 24 large increase in the 20th century of aerosol emissions, the net positive forcing would be much 25 26 larger. As we discuss in Sect. 3.2, this masking by aerosols of the positive, greenhouse gas warming has important implications for climate. During the 21st century, the large decreases 27 in global aerosol and aerosol precursor emissions projected by the RCPs cause aerosol forcing 28 29 to decrease in magnitude (become less negative). As discussed in Sect. 2.3, we also use a 30 version of CM3 with a reduced present-day aerosol ERF, in order to test the robustness of our 31 results to the magnitude of aerosol forcing. We present climate response results for these

simulations in subsequent sections but did not run fixed-SST simulations with this adjusted
version of CM3, hence we do not present forcing values associated with these runs.

3 As in many of the other trends shown thus far, there is limited spread in the various RCP projections of global aerosol effective radiative forcing (Fig. 3). For the 2096-2100 five-year 4 average, the effective forcing (relative to 1860) is -0.21 W m⁻² for RCP2.6, -0.32 W m⁻² for 5 RCP4.5, -0.46 W m⁻² for RCP6, and -0.53 W m⁻² for RCP8.5. RCP2.6 has the largest decrease 6 7 in magnitude of aerosol forcing over the century, followed by RCP4.5, RCP6, and RCP8.5, 8 which is the expected order according to each RCP's underlying climate policy. For example, 9 reduction of coal energy usage, a GHG mitigation policy featured in the RCPs, also reduces 10 the amount of SO₂ emissions. As a result, total aerosol forcing trends and the end-of-century 11 rank order for each of the RCPs can be traced back ultimately to the energy and climate 12 mitigation policy. Furthermore, sulfate is the main contributor to the aerosol direct and indirect forcing trend for all RCPs, due to its optical properties as well as large CCN activity 13 14 (hygroscopicity). Therefore, energy policies that affect sulfate will have a magnified effect on 15 aerosol direct and indirect forcing. On the other hand, the direct climate effects of BC and OC 16 have been reported to offset each other (OC being the negative forcing, BC positive) in previous studies with CM3 (Levy et al., 2013), and that is the case in this work as well. RCP6 17 projects the smallest decrease in magnitude of aerosol forcing for much of the middle part of 18 19 the century (2045 - 2075), despite passing RCP8.5 eventually. This is consistent with both the 20 emissions and AOD trajectories for RCP6. RCP6 projects mid-century increases in coal for 21 energy supply globally (Masui et al., 2011), which is visible not only in the emissions and 22 AOD trends as described elsewhere but also the aerosol forcing trends.

23 3.1.2 Comparison to 2005 levels

24 The large decrease in the magnitude of aerosol forcing from present-day to 2100 represents a 25 large *positive* forcing for 2100 relative to present. The globally averaged forcing changes from 26 present-day to year 2100 for each RCP scenario are tabulated in Table 3. In order to have a 27 consistent basis for comparison across the four RCPs and to account for noise in the trends, 28 the beginning and end of century forcing values were taken as 5-year averages, 2000-2004 29 The amount of unmasked aerosol forcing follows the order expected and 2096-2100. 30 according to each RCP's climate policy (as explained above), as is the case with SO₂ emissions (Fig. 2). For RCP2.6, the forcing increases by 1.37 W m⁻² from 2000 to 2100. 31 RCP8.5 represents the lower part of the range at 1.05 W m⁻². An additional positive forcing of 32

1 at least 1 W m⁻² would have major climate implications. For comparison, the 2100 RCP8.5 2 CO_2 forcing is about 5 W m⁻² (Myhre et al., 2013). Thus, the resulting positive forcing from 3 the decrease in aerosol emissions by 2100 is projected to be more than 20% of the forcing of 4 CO_2 in 2100.

As a caveat, our reported aerosol effective radiative forcing values may be an overestimate 5 6 due to the tendency of the GFDL CM3 model to overstate the cloud lifetime effect, thus 7 inflating the magnitude of our positive forcing due to aerosol decline (Levy et al., 2013). We 8 test the robustness of our results with respect to the magnitude of aerosol forcing using 9 CM3w, a version of the model with much weaker present-day ERF (see Sect. 2.3). In a 10 multimodel evaluation of proxies for the aerosol indirect effects (both albedo and lifetime) 11 against satellite observations, a prototype version of AM3 was found to be one of several models overestimating the strength of the relationship between cloud liquid water path (LWP) 12 and aerosol optical depth (defined as sensitivity of LWP to AOD perturbations) (Quaas et al., 13 14 2009). Ratios for AM3 were roughly an order of magnitude larger than observations over land 15 and ocean; however, other models performed just as poorly if not worse (all models overestimate the land ratio by at least a factor of two). Generally, a positive correlation is 16 17 expected between aerosol optical depth and cloud liquid water path, since the increase in 18 cloud droplet number concentration leads to a delay in autoconversion rate, increasing cloud 19 lifetime and cloud liquid water path. However, some studies have found potential for a 20 negative correlation due to a drying effect from increased entrainment of air above the clouds 21 (Ackerman et al., 2004). Thus, the autoconversion parameterization in AM3, which is a 22 simple implementation that does not address the effect of increased entrainment and other 23 confounding issues, could conceivably be driving an overestimate in the cloud lifetime effect 24 and thus the indirect effect as a whole.

25 **3.2 Climate response**

26 3.2.1 Global historical and future trends

The emission reductions and aerosol forcing trends discussed earlier are expected to have significant effects on future climate. Figure 4 shows the 1860-2100 globally averaged time series for temperature, precipitation rate, liquid water path, and effective cloud droplet radius for the historical, RCPx.x ensemble mean (solid lines), and RCPx.x_2005AER ensemble mean (dashed lines). Since the RCPx.x simulations contain all climate-forcing agents

1 (compared to the aerosol differences we will discuss later), greenhouse gases have a large 2 influence. Temperature anomaly (relative to 1881-1920 average) is projected to range from about 2-5 K by 2100, mostly due to anthropogenic greenhouse warming. Likewise, global 3 precipitation rates will increase, which will be shown in the following section to result 4 5 partially from an increase in greenhouse gases and a decrease in aerosol emissions. Precipitation rate in the historical simulation had a significant decrease around 1950-1970; 6 7 this time frame coincides with a doubling in the global anthropogenic SO_2 emissions (Fig. 2). 8 Liquid water path (LWP) (Fig. 4, bottom left) steadily increases from 1860 to present day, 9 then has four very different trajectories in the RCP future projections. The higher 10 temperatures of RCP8.5 and RCP6 most likely lead to LWP increases, and the order of LWP 11 values in each RCP in 2100 is consistent with the amount of temperature increase. Effective cloud droplet radius (calculated over the top two units of optical depth for liquid clouds, 12 13 consistent with the Moderate Resolution Imaging Spectroradiometer (MODIS) algorithm 14 (Donner et al., 2011; Guo et al., 2014; King et al., 2003) and weighted by cloud fraction) decreases steadily from 1950 to present day, also due to the large increase in aerosol 15 emissions. Since many ultrafine particles are anthropogenic in nature (e.g., sulfate), an 16 17 increase in their emission drives down the global average cloud droplet effective radius, since smaller sized aerosols are effective CCN that are likely to activate into cloud droplets. 18 Likewise, with aerosol emissions declining in the 21st century, cloud droplet radius increases 19 20 across all of the RCPs.

21 3.2.2 Climate response to aerosol forcing

As with aerosol optical depth, taking the difference between the time-varying simulations 22 discussed in Sect 3.3.1 and the fixed aerosol emission simulations (RCPx.x -23 RCPx.x 2005AER), we deduce the impacts on climate driven solely by the aerosol emissions 24 25 reductions for each year of each future emission scenario. Fig. 5 presents the globallyaveraged differences for temperature, precipitation rate, LWP, and cloud drop effective radius 26 27 resulting from aerosol emission reductions in the RCPs (also tabulated in Table 3 for a five 28 year average of 2096-2100 only). Additionally, the dashed lines in Fig. 4 represent the fixed 29 emission ensemble means (RCPx.x 2005AER), thus comparing these with the values from 30 the time-varying emissions ensemble (solid lines) gives an estimate of the aerosol unmasking-31 driven contribution to the climate response. The projected global temperature increase due to 32 aerosol emissions reduction alone ranges from 0.72 to 1.04 K in the standard CM3 with RCPs,

compared to 2005 temperature levels. This is a significant fraction of the *total* temperature 1 2 increase by 2100 (including greenhouse gas induced warming) of 1-5 K across the RCPs 3 (compare dashed and solid lines in Fig. 4). However, in the reduced-aerosol-forcing run with RCP8.5 (CM3w_RCP8.5), the globally averaged temperature increase due to aerosol 4 5 decreases is only about 0.5 K, half of that in RCP8.5 with standard CM3. This result indicates that the projected climate response to aerosol decreases throughout the 21st century depends 6 strongly on the magnitude of the present-day aerosol ERF, as a weaker ERF (~-1 W m⁻²) can 7 8 cut the global temperature response in half. This is discussed in more detail in Sect. 4.1.3 and 9 shown in Figs. 7 and 8. The impact of aggressive phase-out of coal as an energy source in 10 RCP2.6 can be seen from about 2020-2050 with a strong increase in aerosol driven 11 temperature change. Likewise, the mid-century rise in coal use in RCP6 shows up as a decline 12 in what is an otherwise consistent temperature increase throughout the century (Fig. 5). 13 RCP4.5 and RCP8.5, on the other hand, have a steadier temperature increase that lacks the 14 same noticeable features. There is a significant amount of overlap in climate response from 15 decreasing aerosols among the RCPs throughout the entire century, especially considering the 16 full range of the ensemble members of each simulation. This suggests that despite large 17 differences in many facets of the individual RCPs (e.g., climate mitigation policies, emissions 18 trajectories, land use), the temperature response to decreasing aerosols is relatively 19 homogenous.

20 Although global precipitation rate (Fig. 5 upper right) is mainly controlled by the tropospheric 21 energy balance (Ming et al., 2010), precipitation is linked to aerosols on the local scale, since aerosols serve as seeds for cloud droplet formation and the number and size of the cloud 22 23 droplets influence the distribution and timing of precipitation. Additionally, aerosols can impact precipitation in other ways, including by changing atmospheric dynamics and 24 25 circulation patterns, changing atmospheric heating rates, and changing the surface energy balance. We find increases of $0.08 - 0.09 \text{ mm d}^{-1}$ (up to 3% of 2005 precipitation levels) are 26 27 projected to result from the aerosol decrease in the four RCPs run with standard CM3, again 28 representing a very narrow range of climate endpoints despite the differences among the 29 scenarios. The estimated increase of precipitation is reduced to only about 0.04 mm d⁻¹ increase by the end of the 21st century in the CM3w_RCP8.5 runs with weaker present-day 30 aerosol ERF. Nonetheless, since the total precipitation rate increase from all forcings ranges 31 from 0.11 - 0.25 mm d⁻¹ (Figure 4), these aerosol driven changes are significant (discussed 32 33 more in Sect. 4.1.3 and shown in Figs. 7 and 8).

1 Liquid water path differences over time are shown for each of the RCPs in the lower left panel 2 of Fig. 5. As aerosol concentrations decrease, LWP also decreases; in other words, aerosols and LWP are positively correlated. This is essentially the cloud lifetime effect acting in the 3 opposite direction: increased aerosols cause cloud droplet concentrations to increase leading 4 5 to a decrease in the autoconversion rate, which hinders precipitation formation and increases cloud lifetime and cloud liquid water path (Albrecht, 1989). The decline in aerosol emissions 6 7 leads to a decrease in LWP in all of the standard CM3 runs with the RCPs, around 0.5-1.0 g m^{-2} or 2% of 2005 levels. Accounting for the ensemble member range (shaded areas), the 8 9 LWP decline in each of the RCPs is remarkably similar. As is the case with radiative forcing, temperature, and precipitation, the annual trends in the LWP values also follow the underlying 10 11 RCP energy use trajectories. In particular, a rebound around 2040 in LWP in RCP6 can be 12 seen in the bottom left of Fig. 5, analogous to the temperature decrease in RCP6 due to an 13 increase in coal energy usage rate and ultimately aerosol and precursor emissions. LWP 14 decreases in CM3w_RCP8.5 are remarkably smaller, only about 20-40% of the decreases 15 standard CM3 RCP runs. With a lower autoconversion threshold, the amount of liquid water 16 held in clouds is decreased, resulting in the smaller change in CM3w_RCP8.5 shown in Fig. 17 5c.

18 With decreasing aerosols, effective cloud drop radius may increase due to the loss of smaller-19 sized and more numerous anthropogenic ultrafine aerosol, leaving natural aerosols (such as 20 sea spray, which in general are fewer in number and much coarser in size) to form cloud 21 droplets. Across the four RCPs, the globally averaged increase in cloud drop effective radius 22 due to decreasing aerosols ranges from 0.54 µm to 0.60 µm (Fig. 5, Table 3). The increase due 23 to all forcings ranges from about $0.60 - 0.80 \mu m$ (Fig. 4). As expected, the increases in cloud 24 droplet effective radius due to aerosol reductions make up a large fraction of the all-forcing 25 increases. Cloud droplet effective radius trends follow energy usage and emissions trends in 26 each of the RCPs very closely, with the results from the individual RCPs being quite distinct 27 from one another through most of the century before slightly converging around 2100. Cloud 28 droplet radius does not appear to be significantly affected by the lower autoconversion 29 parameterization, as the differences between RCP8.5 with standard CM3 and RCP8.5 with 30 CM3w are minimal.

1 **4** Spatial distribution and regional analysis

2 Emissions of aerosols and aerosol precursors are highly heterogeneous in space. As expected, 3 the changes of AOD, radiative forcing, and climate change in response to projected future emission changes also exhibit strong spatial structure. In this section, we first examine the 4 spatial distributions of the changes in AOD and climate response, and then consider the 5 6 average responses over two key source regions, East Asia and North America. Judging by 7 historical emissions over the last few years since the RCPs were developed, concentrations of 8 GHG in the atmosphere are tracking at or even above the trajectory predicted by RCP8.5 (the 9 highest emission scenario), so we will focus mainly on that scenario (Peters et al., 2012; 10 Sanford et al., 2014).

11 **4.1 Spatial distributions**

12 4.1.1 Aerosol optical depth

Figure 6 shows the RCP8.5 aerosol optical depth (550 nm) differences (RCP8.5 -13 14 RCP8.5_2005AER) for the end of the 21st century (2096-2100 average) for sulfate, BC, OC, 15 and total AOD. Corresponding results for the other three RCPs can be found in Figs. S6-S8 of 16 the supplemental information. Strong reductions in AOD are simulated over most continental 17 regions, including North America, Europe, and Asia. This coincides with the strong SO₂ emissions decreases projected by RCP8.5 (and all RCPs) for much of the world. East and 18 19 South Asia have the largest and most widespread decreases in AOD for all species. However, there are a few regions in which emissions changes are projected to *increase* sulfate AOD by 20 21 2100. Africa has noticeable areas of sulfate AOD increase, which can be explained by RCP8.5 22 projected increases in emissions at a few locales in this region (see Fig. S5 of supplementary 23 information). Parts of Africa are expected to industrialize and grow throughout the rest of the 24 century and utilize its natural fossil fuel resources (Masui et al., 2011; Riahi et al., 2007). 25 Similarly, RCP8.5 projects sulfate AOD increases for Indonesia, a region that is also projected 26 to industrialize and experience an increase in biomass burning, resulting in elevated emissions of SO₂, BC, and OC. There are also a few areas of increase over the tropical Pacific and 27 Atlantic oceans for OC and SO₂ which are not caused by emissions increases (see Fig. S5) 28 29 and instead result from meteorological changes, specifically a decrease in effectiveness of wet deposition removal of sulfate and OC. Since precipitation is increasing along with sulfate and 30 31 OC aerosol optical depth, a decrease in the efficacy of wet deposition is implied. Although we

1 do not have wet deposition fluxes archived in the model output, we note that using the same 2 model, Fang et al. (2011) confirmed this relationship between increasing precipitation and 3 decreasing wet deposition removal effectiveness. Using an idealized soluble tracer, the authors found that as climate warms, wet deposition of soluble pollutants decreases due to the 4 5 simulated decreases in large-scale precipitation frequency. In the future, moderate and light precipitation is projected to occur less frequently, whereas heavy precipitation occurs more 6 7 frequently. Since wet deposition does not depend on strength of precipitation events (i.e. same 8 wet deposition flux for heavy or light rain), the decrease in frequency of light precipitation 9 events leads to less wet deposition flux in the future and thus increases in AOD (Fang et al., 10 2011).

11 Asian BC and OC AOD decreases by 2100 are much larger than those in North America or Europe (Fig. 6). Some increases are again projected in Africa. BC and OC biomass burning 12 emissions on average in Africa are generally expected to maintain current levels or decline 13 14 slightly; however, in this particular region of Africa (mostly in Democratic Republic of the 15 Congo and the Central African Republic), biomass burning emissions of OC and BC are 16 projected to rise (not shown, see Fig. S5 of supplementary information). In particular, 17 combustion and biomass burning emissions in Africa are expected to increase rapidly in the 18 near-term (Liousse et al., 2014). The total AOD difference in 2100 for RCP8.5 (lower right of 19 Fig. 6) is dominated by the sulfate AOD reductions over the continents (except for in parts of 20 Africa and other developing regions).

4.1.2 Climate response

22 The climate responses to the aerosol reductions in RCP8.5 are shown in Fig. 7 for the end of 23 the century and in Figs. S9-S11 of the supplemental information for the other three RCPs. As 24 with Fig. 6, Fig. 7 shows the spatial pattern of the difference between the time-varying aerosol simulations and the fixed aerosol simulations (RCP8.5 - RCP8.5_2005AER). Areas hatched 25 26 with dots in each of the figures indicate statistically significant regions at the 95% confidence level according to Student's t test. For temperature (upper left), the impact of aerosol 27 28 reductions is almost entirely a warming effect, as expected, and nearly all of these increases 29 are statistically significant. There are large areas of temperature increase in 2100 for East 30 Asia, which is consistent with the largest decreases in AOD and aerosol emissions in this 31 region. However, much of the strongest temperature increases driven by reduced aerosol 32 emissions are located in or near the Arctic, suggesting that temperature change is non-local

and does not necessarily occur in the areas where emissions are changing. Shindell et al.
 (2010) also have reported strong temperature increases in the Arctic where forcing was small
 in a global multimodel study.

Precipitation rate increases due to decreased aerosol emissions are largest over the tropical 4 oceans, although increases are also observed over continents. In particular, some statistically 5 6 significant precipitation rate increases occur over Europe, Russia, and Southeast Asia. 7 Increases over North America are generally found to be statistically significant. However, 8 much of the areas of increase over the oceans are statistically insignificant. Simulating 9 precipitation, especially regional details, remains challenging for most models, including 10 CM3 (Eden et al., 2012). In our results there are two intercontinental tropical convergence zones (ITCZ) near the equator, a common feature not only identified in CM3 but in other 11 models as well (Lin, 2007). Both of the ITCZ show strong areas of precipitation enhancement 12 despite not coinciding with aerosol decreases. The northward shift in the ITCZ is expected 13 14 when removing a negative forcing in the northern hemisphere (i.e. aerosol decreases), but this 15 does not appear to be statistically significant (Allen and Sherwood, 2010). Precipitation in the tropics is mainly a result of deep convection, and several studies have identified the effect of 16 17 aerosols on deep convective circulation and precipitation (Bell et al., 2008; Lee, 2012; 18 Rosenfeld et al., 2008). Using a cloud-system resolving model of a large-scale deep 19 convective system, Lee (2011) found that perturbations of aerosols in one domain can have 20 teleconnections to other domains, acting to enhance precipitation in one domain while 21 suppressing it in another through deep convection. Since the changes in precipitation are both 22 positive and negative (mostly positive), and regions of precipitation increase that do coincide 23 with aerosol decrease are few, it is likely that aerosols are affecting precipitation through other 24 means than the aerosol indirect effect alone. The aerosols are also likely exerting an influence 25 on atmospheric dynamics and weather patterns, causing large, non-uniform increases and 26 decreases in the tropics. In particular, we postulate that the aerosol decreases in the 27 continental domain are teleconnected to deep convection, resulting in both precipitation 28 increases and decreases, as demonstrated by Lee (2011).

Liquid water path changes (Fig. 7, lower left panel) are largest over East Asia, which again is consistent with the simulated aerosol changes. Much of Europe and eastern North America also have large LWP decreases that coincide with aerosol emission decreases. However, there are large LWP increases in the Arctic region, most of which are statistically significant. These

1 increases could be due to a feedback from the aerosol-driven temperature increase, as well as 2 an increase in melting sea ice, making more water available for evaporation. Cloud droplet 3 radius increases universally across the globe, with statistically significant changes occurring in the Northern Hemisphere, which is also where most of the aerosol reductions occur (Fig. 7, 4 5 lower right). The co-location of the large increases in cloud droplet effective radius with large decreases in aerosol optical depth is a strong signal of the aerosol indirect effect, specifically 6 7 the cloud albedo effect (we will explicitly show this in Sect. 5). Since the impacts of aerosol 8 reductions on both liquid water path and cloud droplet effective radius are significant over the 9 oceans in addition to the continents, we can also conclude that the aerosol reduction impacts 10 are not necessary localized to their area of emission. Additionally, the increases of cloud 11 droplet radius over the oceans may be amplified due to the greater susceptibility of clouds in 12 clean environments compared to more polluted conditions.

13 Figure 8 shows the climate response in 2096-2100 for CM3w_RCP8.5. These responses are 14 plotted on the same scale as Fig. 7 for comparison. For temperature, the pattern is quite similar, with the largest increases over the Arctic and the continents, and statistical 15 16 significance nearly everywhere. However, the magnitude of the temperature increases are 17 noticeably smaller, owing to the reduced aerosol radiative forcing in the CM3w RCP8.5 runs. 18 Precipitation changes show a vastly different spatial pattern and slightly smaller magnitude in 19 CM3w_RCP8.5 compared to RCP8.5 standard CM3, and overall is slightly smaller in 20 magnitude. However, much of this difference is due to the lack of a strong positive 21 precipitation anomaly near the equator in CM3w_RCP8.5, which is not statistically significant 22 in either set of simulations. Over the continents, the precipitation change is more qualitatively 23 similar, for example over East and South Asia, where both sets of simulations show 24 precipitation increases. CM3w_RCP8.5 produces a much larger precipitation increase over 25 South Asia, perhaps due to the lower autoconversion threshold, although without statistical 26 significance and only one ensemble member for CM3w, it is not possible to comment on the 27 robustness of this result. Liquid water path decreases across Eastern North America, Europe, 28 and East Asia, consistent with areas where aerosol decreases are largest. Many of these 29 decreases, however, lack statistical significance, and the magnitude of the change is much 30 smaller than in standard CM3, as discussed previously. Cloud droplet effective radius changes 31 are qualitatively similar, with large decreases over the continents. Compared to LWP, 32 temperature, and precipitation, cloud droplet radius is least by the lowered autoconversion 33 threshold (see also Fig. 9).

4.1.3 Comparison of climate response driven by aerosol decreases and by all forcings

3 Although the projected absolute climate response to decreasing aerosols is similar in all the 4 RCP simulations, the magnitude of relative effects differs. By comparing our climate response 5 to aerosol decreases (RCPx.x – RCPx.x_2005AER) to the climate response from all forcings 6 (RCPx.x), we can roughly assess the relative importance of aerosol decreases to the total 7 changes in climate for each RCP. The nonlinear nature of the global climate system means 8 that these aerosol ratios are not directly additive with other ratios (say, GHG-induced climate 9 changes) and are not the "true contributions" per se. However, comparing the magnitudes of 10 the aerosol-induced climate changes to the total climate changes is a useful framing exercise. In instances where the all-forcing response sign differs from the aerosol-driven response, we 11 12 use the absolute value (i.e. magnitude only) of the all-forcing response for the ratio 13 calculation. We project from 2006 to the end-of-century a 0.97 K warming from aerosol emissions reductions in RCP2.6 (Fig. 5, Table 3) compared with a 1.5 K warming from all 14 climate forcings together (Fig. 4), indicating that, under this scenario, two-thirds of the 15 warming by 2100 would result from decreases in aerosol emissions. The RCP2.6 scenario 16 17 indicates that even with aggressive reductions in the emissions of greenhouse gases, the projected reduction in aerosol emissions is likely to push the climate near the 2 K warming 18 19 frequently cited as constituting "dangerous anthropogenic interference with the climate system" (Meinshausen et al., 2009). Since RCP2.6 projects the least warming from 20 greenhouse gases, we find that this scenario is relatively the most susceptible (i.e. the largest 21 22 percent effect) to unmasked aerosol warming as well as aerosol-driven changes in 23 precipitation, LWP, and cloud droplet effective radius.

24 However, since GHG emissions over the past decade have been well above RCP2.6 and even 25 slightly above RCP8.5 (Peters et al., 2012; Sanford et al., 2014), using RCP2.6 as a 26 benchmark for the aerosol fraction of future climate change may be misleading. Figures 4 and 27 5 show that for RCP8.5, warming from aerosol reductions is roughly 1 K globally of a total 28 warming of nearly 5 K, or around 20%. The RCP8.5 precipitation increase of 0.09 mm d⁻¹ is about 36% of the all-forcing increase of ~ 0.25 mm d^{-1} , while the globally averaged ratios for 29 30 LWP and cloud droplet effective radius are 30% and 75%, respectively. The large aerosol fraction for cloud droplet radius is expected, since cloud droplet size is highly dependent on 31 32 existing aerosols, perhaps to a greater extent than the other three climate parameters presented

here. Thus, even considering the high-emissions, low-mitigation RCP8.5 scenario, aerosol reductions are still a surprisingly important player in future global and regional climate change. However, an important caveat remains that our total present day aerosol effective radiative forcing is on the higher end of the IPCC range and thus may bias our results. We quantify this potential bias and discuss further below.

6 Figure 9 summarizes the 2096-2100 five-year average climate responses to decreasing 7 aerosols (red bars, labeled RCP8.5 - RCP8.5 2005AER) for each region and puts them in the 8 context of the all-forcing (GHG and aerosols) results (white hatched bars, labeled RCP8.5). In 9 the standard CM3 RCP runs, surface temperature increases due to aerosols are a substantial 10 fraction of the all-forcing warming, even in RCP8.5 which features the largest warming from long-lived greenhouse gases. Much of East Asia, Australia, and the Middle East have ratios 11 above 20% (seen in Fig. 9, comparing red bars with white hatched bars), indicating that the 12 large aerosol decreases in these regions will contribute significantly to projected warming. 13 14 The Arctic region (defined as everything above 66° N) actually has the largest temperature increase due to decreasing aerosols, consistent with the polar amplification phenomenon. 15 16 However, because climate warming from greenhouse gases is also amplified in the Arctic, the 17 ratio of aerosol-induced warming to all-forcing warming is smaller than in many other 18 regions. Even over the oceans (not shown), where anthropogenic aerosol abundances are low, 19 we find their global decrease accounts for more than 10% of the all-forcing warming in these 20 locations. Parts of East Asia and Europe, regions for which major aerosol decreases are 21 expected, have high precipitation contributions approaching 40% (red and white bars the 22 upper right panel of Fig 9); however, this is not generalizable to all continental regions. Like 23 precipitation, LWP contributions are small near the poles (see Arctic region), with sporadic 24 regions of large LWP contributions in Asia, Australia, and the Middle East, indicating larger 25 aerosol influence compared with all-forcings in these regions. Finally, contributions of aerosol 26 decreases to total cloud droplet radius changes approach 60-90% for much of the globe 27 between the latitudes 30° S and 60°N. Ratios are particularly low in the Southern Ocean, 28 where natural aerosols such as sea salt likely dominate CCN activation, and thus are less 29 affected by decreasing anthropogenic aerosols.

Figure 9 also shows the comparison of aerosol decrease-driven climate response to all-forcing
climate response for the CM3w_RCP8.5 runs with weaker present-day aerosol ERF (yellow
bars in Fig. 9, labeled CM3w_RCP8.5 - CM3w_RCP8.5_2005AER). With some exceptions

1 for precipitation in Asia, the climate responses to decreasing aerosols are always significantly 2 less in CM3w RCP8.5 than in the standard RCP8.5 runs. This is particularly noticeable in the 3 temperature and LWP panels of Fig. 9. The temperature response in CM3w_RCP8.5 is sometimes half of the standard CM3 response, and the LWP is often an even smaller fraction. 4 5 Thus, whereas RCP8.5 aerosol decreases may contribute up to 30-40% of the total warming in East Asia, this ratio is closer to 10-20% with the weakened aerosol forcing. While both 6 7 versions of the model result in aerosol ERF within IPCC ranges, CM3w_RCP8.5 simulates a 8 present-day ERF much closer to the center of the uncertainty range, suggesting that this 9 estimate of aerosol decrease-driven climate response may be more robust.

10 4.2 Regional climate response

11 Using the regions defined in Fig. 1, we quantify changes in AOD, radiative forcing, and 12 climate responses due to changes in aerosol concentrations on a regional scale in Tables 4 and 13 5. We present the effective radiative forcing (or flux perturbation) as a difference between the 14 2096-2100 value and the 2000-2004 levels (as before), which is why these values are mostly positive. The AOD and climate differences are the difference between the time-varying RCP 15 16 simulations and the 2005 fixed aerosol simulations (RCPx.x – RCPx.x_2005AER) for the end of century (2096-2100 5-year average). Values for RCP8.5 are shown in Table 4; RCP2.6 is 17 18 shown in Table 5. These are chosen as the upper and lower ranges of AOD, radiative forcing, 19 and climate response changes. Identical tables for RCP4.5 and RCP6 can be found in 20 supplemental information (Tables S1 and S2). Bolded values in the tables represent the largest regional change for each quantity (e.g., largest SO₄ AOD decrease). Table S3 shows the 21 22 regional changes in climate response for CM3w_RCP8.5, although the AOD changes are not 23 shown as we expect these to be minimal.

24 4.2.1 East Asia

Several of the largest aerosol-driven climate changes are found in the East Asian region. The region is defined to include all of China, Mongolia, Japan, and Korea (Fig. 1). East Asian emissions, AOD, radiative forcing, and climate response are analyzed below.

Figure S12 shows the anthropogenic SO₂, BC, and OC emissions time series from East Asia for the historical period and for each of the RCPs. For all species and all RCPs except RCP6, aerosol and aerosol precursor emissions are projected to peak in the 2010s. The trend for RCP6 for each species consists of a small increase in the 2010s, followed by a brief decrease and then a sharp increase with emissions peaking in 2050. This shifted trend results from the primary energy supply projections in RCP6, in which coal energy usage in East Asia increases steadily until peaking in 2050-2060. This reliance on coal as a primary fuel source results in a mid-century peak in not only aerosol and precursor emissions, but also CO₂ emissions (Masui et al., 2011).

6 Figure S13 shows the East Asian region aerosol radiative forcing (calculated as a flux 7 perturbation or effective radiative forcing) for all the RCPs and the historical period. This 8 calculation is done by simply averaging over the region considering global aerosols as 9 opposed to isolating the effect of East Asian aerosols alone. Radiative forcing from aerosols 10 (direct + indirect) will continue to become more negative (larger in magnitude) until about 2025, when it reaches nearly -5 W m⁻² over the region. For the rest of the century, the 11 decrease in aerosol emissions (and AOD) results in a less negative radiative forcing (smaller 12 in magnitude). Depending on the RCP, the aerosol forcing decreases in magnitude by about 2 13 to 3 W m⁻² in 2100. RCP6 has the weakest recovery over most of the century. This is likely 14 due to the increase in reliance on coal power, allowing emissions to increase and peak over 15 the first half of the 21st century as mentioned above. 16

Climate response over East Asia is shown in Fig. 10 as differences between the time-varying 17 18 RCP simulations and the fixed 2005 global aerosol emission simulations (RCPx.x -19 RCPx.x_2005AER). Temperature increases driven by aerosol emission changes are much 20 larger than the global average, ranging from 1.1 - 2.2. K of warming in the standard CM3 run. 21 This is consistent with the large decrease in aerosol optical depth (-0.48 for sulfate, see Tables 22 4 and 5). In CM3w RCP8.5, the temperature increase is smaller than all of the standard RCPs 23 by the end of the century. The temperature, precipitation, and especially cloud droplet 24 effective radius all respond differently to the unique energy supply trajectory of RCP6 than to 25 the other RCPs. The warming around the middle of the century from aerosol emissions 26 changes in RCP6 lags that in the other RCPs, and reaches only 1.1 K by 2100 (compared to 27 2.2 K for RCP8.5). Similar trends exist for precipitation rate and cloud droplet effective radius, which lag strongly around 2050, a time period in which the rest of the RCPs steadily 28 29 increase. The annual trends in the indirect forcing related quantities, LWP and cloud droplet 30 effective radius, again bear a striking similarity to the emissions trends (Fig. S12) on the 31 regional scale as well as the global scale, as we previously showed.

1 4.2.2 North America

2 The climate responses over North America to global emission changes are shown in Fig. 11, 3 and North American emissions and effective radiative forcing is shown in supplemental 4 information Figs. S14 and S15. Unlike East Asia, anthropogenic emissions peak much earlier 5 in North America. SO₂ emissions reached a maximum in roughly 1970 and have been declining steadily since then, with each of the RCPs projecting the decline to continue in a 6 7 similar manner. Total aerosol forcing follows a similar qualitative pattern as in East Asia; however, aerosol forcing over North America is projected to range from about 0 to -0.5 W m⁻² 8 9 from 2006 to 2100. Regionally averaged North American temperature increases by 1.5 to 2.0 K across each of the standard CM3 runs with the RCPs, and no particular RCP stands out 10 11 from another at the end of the century, especially considering the ensemble member range. Again, temperature and precipitation response is much more muted in CM3w_RCP8.5. 12 Precipitation increases by about 0.1 mm d⁻¹ over North America due to declining aerosol 13 emissions. There is almost no change in LWP over North America, and a slight increase is 14 15 even projected in RCP8.5 by the end of the century, due to the warming effect on LWP over the extreme northern regions of North America. Despite having a major impact globally and 16 in other regions, LWP changes are not noticeably different in CM3w_RCP8.5 compared to 17 18 the standard CM3 runs. Cloud droplet radius increases nearly identically across the RCPs for the entire century, except for RCP2.6, which stands out as having the greatest effective radius 19 20 increase for the first two-thirds of the 21st century. North American RCP2.6 SO₂ emissions 21 (Fig. S14) and AOD (not shown) decrease substantially over the first half and level off for the remainder of the 21st century, which explains the trend in cloud droplet radius. 22

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24 **5** Implications and discussion

25 **5.1 Comparison to previous studies**

Our estimated range $(1.05 - 1.37 \text{ W m}^{-2})$ for the effective radiative forcing change resulting from projected future aerosol and precursor emission reductions falls within the range estimated in previous literature, as shown in Table 1. In particular, Shindell et al. (2013) estimates a present-day to end-of-century aerosol forcing change of $0.68 - 1.42 \text{ W m}^{-2}$. When considering our CM3w_RCP8.5 simulations with lower present-day aerosol ERF, our lower estimate of 1.05 W m⁻² is actually much closer and in better agreement with the values in

1 Shindell et al. (2013). Our global temperature increases from aerosol decreases across the 2 RCPs of 0.72 - 1.04 K compares well with many previous studies (see Table 1), which have also found that temperatures will increase as much as 1 K due to aerosol decreases. Our 3 global precipitation results also compare well with previous modeling studies. For instance, 4 5 both Levy et al. (2013) and Rotstayn et al. (2013) found global increases in 2100 of 0.1 mm d⁻ ¹, similar to the top of our projected range of 0.078 - 0.093 mm d⁻¹. The effect of future 6 7 aerosol decreases on liquid water path and cloud droplet effective radius were not considered 8 by most of the previous literature, with the exception of Takemura (2012), who reports a change in LWP of around 3 g m⁻², significantly higher than the 0.5-1.0 g m⁻² reported here. 9 However, cloud droplet radius values compare much better. The aerosol-driven increases are 10 11 very similar to those reported by Takemura (2012), who reported a difference in effective radius of about 0.6 µm at the tropopause from 2000 to 2100, compared to 0.5-0.6 µm in our 12 13 study. However, Takemura (2012) did not conduct fixed aerosol simulations 14 (RCPx.x_2005AER) and thus did not include the effect of climate change on future aerosol 15 concentrations.

16 **5.2** Correlations between aerosols and climate response

It remains difficult to attribute specific changes in climate parameters to changes in aerosol 17 18 emissions or optical depth, due to the complexity of global coupled chemistry-climate models 19 and the complexity of the climate system as a whole. In Fig. 12 we plot spatial correlations 20 between RCP8.5 emission-driven changes in aerosol optical depth values and four climate 21 parameters as an attempt to relate an aerosol quantity with corresponding climate response 22 endpoints. Correlation plots for the three other RCPs are in the supplemental information 23 (Figs. S16-S18). We also plot $\Delta C/\Delta AOD$ for RCP8.5 in Figure S19, where ΔC represents the change from the beginning to end of the 21st century for a given climate response variable, 24 25 and $\triangle AOD$ for the change in AOD. For RCP8.5 AOD-temperature correlation, there is a strong relationship (r < -0.75) over virtually all of the continental areas and the northern 26 27 Atlantic and Pacific oceans, which is expected due to aerosols having a net cooling influence on climate via the direct and indirect effects. The weak correlations over the tropical Pacific 28 29 correspond to AOD increases and less extreme temperature increases over these regions, 30 whereas the Southern Ocean positive correlations are caused by increases in temperature and 31 very small increases (due to DMS emissions increases) in sulfate AOD (supported by the weak correlation values). AOD increases are also projected in parts of the Arctic, some of 32

which may be related to the Arctic haze phenomenon. Coupled with strong temperature
 increases in the Arctic, positive AOD-temperature correlations are projected.

3 Precipitation is shown in the upper-right and is correlated both positively and negatively with 4 changes AOD (Fig. 12). Over East Asia and parts of Europe, AOD and precipitation are somewhat strongly anti-correlated (r < 0.5), which is expected according to the aerosol 5 6 indirect effect on clouds. However, the mixed representation between positively and 7 negatively correlated regions, as well as the overall weak correlations, suggests that it is not 8 the aerosol indirect effect alone that is the mechanism for the projected effect of aerosols on 9 precipitation. Instead, this is likely a signal of aerosol impacts on circulation, dynamics, and 10 weather patterns, which can have feedback effects on precipitation. As described in Sect. 11 4.1.2, we believe that aerosol perturbations over the continents may have teleconnections to precipitation enhancements in deep-convective cloud systems, accounting for the positive 12 correlation over the tropics. In Fig. S20 we also correlate AOD perturbations with deep 13 14 convective precipitation and find generally stronger negative correlations (r \sim -0.75) over 15 much of the northern hemisphere but roughly the same positive correlations over much of the 16 oceans, including the tropics.

17 Liquid water path (LWP) is correlated positively (r > 0.5) with AOD over nearly the entire 18 globe, with notable exceptions in Alaska and northern Russia. Additionally, areas with the 19 strongest change in AOD seem to have the strongest LWP changes as well, possibly 20 implicating the aerosol indirect effect as the mechanism behind the correlation. Similarly, 21 cloud droplet radius is very strongly anti-correlated almost throughout the entire globe (r >22 0.75), with the exception of the Arctic, which is also affected by the aerosol-driven increase in 23 temperature.

24 **5.3** Similarity among the RCPs

Our results are of course dependent on the GFDL CM3 model as well as the emissions projections provided by each of the RCPs, which have limitations. First, since recent GHG emissions are tracking at or above RCP8.5 (Peters et al., 2012; Sanford et al., 2014), the high emissions case, the RCPs may be optimistic in their greenhouse gas emissions projections. Second, the individual RCP pathways are often very similar to one another on both the global and regional scales, especially for air pollutant emissions and related quantities (see Fig. 2). This leads to a limited spread in the possible climate responses which likely does not explore

1 the breadth of possible technology and policy driven changes in future aerosol emissions. For 2 example, the spread in precipitation rate response driven by decreased aerosol emissions across scenarios is only 0.015 mm d⁻¹. In almost every example, the trends of AOD, radiative 3 forcing, and climate response in each of the RCPs does not substantially differ (Table 3, 4 5 Figure 5). An exception to this is with the CM3w_RCP8.5 runs, which does produce a significantly different and more unique climate response to decreasing aerosols in the 21st 6 7 century. Despite vastly different energy supply scenarios and climate policies, it appears to be 8 of little consequence which RCP is used to determine the impact of the declining aerosol 9 emissions on climate in the standard CM3 runs. Regardless of the level of projected GHG 10 emissions in the future, decreasing aerosol concentrations will likely result in substantially 11 warmer temperatures (see Tables 3-5) and regionally varied changes in precipitation and other 12 climate parameters. Finally, unlike the IPCC SRES inventories, the RCPs do not begin from 13 narrative storylines of what energy, population, and economic conditions will be like in the 14 future making it difficult to link an RCP with a particular development pathway. However, 15 recent work has attempted to map IPCC SRES illustrative storylines onto each of the RCPs (van Vuuren and Carter, 2013). 16

17 6 Conclusions

18 We present a global climate modeling study of the impact of the projected future decrease of 19 aerosol emissions using the GFDL Climate Model 3 (CM3). Future emissions follow the 20 Representative Concentration Pathway (RCP) projections, which consist of four emissions trajectories that are consistent with global average radiative forcing values in 2100 of 21 approximately 2.6, 4.5, 6.0, and 8.5 W m⁻², respectively. In response to the continuing human 22 health cost of air pollution, each of the RCP scenarios projects that global emissions of 23 24 aerosols and their precursors will decline as the result of policy decisions to improve air 25 quality. However, aerosols also have a net cooling effect on climate that will be reduced (i.e., warming) as the emissions decline. Aerosols also impact clouds and precipitation, the effects 26 27 of which will also change under the projected diminishing emissions. We implement the RCP emissions inventories into CM3 and run two sets of simulations for each of the pathways: one 28 29 in which aerosol and aerosol precursor emissions are allowed to vary as projected by the 30 RCPs, and another in which they are fixed at 2005 levels. The sets of fixed emissions 31 simulations serve as controls to analyze the changes in AOD, radiative forcing, and climate solely due to the projected emission reductions of aerosols and their precursors. 32

1 Changes in aerosol optical depth closely follow the emissions changes spatially and 2 temporally. Regionally, there are large AOD decreases over the continents, especially in East 3 and South Asia. A few localities exhibit increases in aerosol optical depth, in some cases this 4 is due to natural sources of aerosol or feedbacks on precipitation and wet deposition. In other 5 cases, the AOD increases are indicative of local emissions increases, which are expected to 6 occur in parts of Central Africa and Indonesia.

7 The projected decreases in aerosol emissions and optical depth lead to a decrease in the 8 magnitude of the aerosol direct and indirect radiative forcing. CM3 predicts an aerosol direct plus indirect radiative forcing of -1.8 W m⁻² from pre-industrial to present-day, which is close 9 to the upper range of -1.9 W m⁻² used by the IPCC (Myhre et al., 2013). To test robustness of 10 11 our results to the strength of aerosol forcing, we run additional simulations in which the 12 autoconversion threshold is lowered, resulting in a present-day aerosol ERF of about -1.0 W m⁻². By 2100, the aerosol forcing is projected to diminish to between -0.21 W m⁻² in RCP2.6 13 and -0.53 W m⁻² in RCP8.5. Our results therefore indicate that a large positive forcing, up to 1 14 W m⁻² or greater (or 0.5 W m⁻² in CM3w RCP8.5), will result from the projected decrease in 15 aerosol and aerosol precursor emissions. This forcing has a strong impact on climate, 16 17 warming temperatures by as much as 1 K globally and up to 2-3 K regionally in the standard CM3 RCP runs, with a much more modest response in the reduced aerosol forcing runs of 0.5 18 19 K globally. Aerosol reduction-driven surface temperature response generally accounts for a 20 large fraction of the total all-forcing response. Ratios over East Asia can exceed 30% in 21 RCP8.5, and are even higher in the other scenarios that do not feature as much greenhouse gas-induced climate warming. However, these ratios are closer to 10-20% in most regions and 22 23 globally when we consider the reduced aerosol forcing RCP8.5 run. Global precipitation rates are projected to increase by as much as 3% of 2005 values or 0.1 mm d⁻¹, with greater 24 25 regional impacts. Inconsistent with Levy et al. (2013), we found significant local precipitation 26 changes co-located with areas of large aerosol decrease (e.g., East Asia). On the global scale, 27 aerosol reduction-driven changes in AOD and climate response trajectories do not vary 28 significantly among the four RCPs, especially towards the end of the century, despite 29 stemming from nominally different scenarios. Mid-century variation in the climate response and radiative forcing trajectories closely follows the aerosol and precursor emissions 30 trajectories (and thus the energy use trajectories), even for climate parameters such as liquid 31 32 water path and cloud droplet effective radius.

Temperature, liquid water path, and cloud droplet effective radius are strongly correlated 1 2 spatially with aerosol optical depth changes (as would be expected from their parameterizations), confirming the localized effect that aerosols have on climate forcing. 3 Precipitation and AOD changes were only weakly correlated, with even the sign of the 4 5 correlation varying spatially. This suggests that local changes in aerosol quantity are not strongly tied to local precipitation events, but that large-scale changes in atmospheric 6 7 circulation instead play a role in the effect of aerosols on precipitation. Future work is needed 8 to identify the mechanisms behind the precipitation response to inhomogeneous changes in 9 aerosol concentrations.

10 Our results indicate the need for a tighter coupling among climate and air pollution control 11 policies, in order to avoid unintended warming from air pollution reductions. To simultaneously clean the air and avoid exacerbating global warming, more aggressive 12 greenhouse gas mitigation will be necessary to compensate for the unintended aerosol-driven 13 14 warming. Future work on reductions of specific emissions sources (e.g., activities with large 15 emissions of BC) that have beneficial impacts for both human health and climate will be beneficial. Additional work is also needed to understand the connection between changes in 16 17 aerosol quantities and corresponding climate responses. Future scenario development that 18 better replicates explicit policy and technology changes driving emission reductions of air 19 pollutants will provide scenarios that have larger variations among them, which will allow a 20 more thorough analysis of the impacts of various mitigation efforts. Finally, the impact of 21 nitrate aerosols on future climate needs to be explored. Many current studies have omitted 22 nitrate, but the combination of increasing ammonia emissions and decreasing sulfate in the 23 future will likely promote nitrate formation. If nitrate does increase in significance, society 24 might simply replace the lost sulfate forcing and human health impact with a similar impact 25 from nitrate aerosol.

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1 Table 1: Summary of previous global climate modeling studies examining future climate effects of aerosols (only those using RCP

2 projections)

Work	Model	RCPs	Output	Domain	Aerosol	Results
Levy et al. (2013)	GFDL CM3	4.5	Emis., AOD, T, P, RF	Global, 1860-2100	Fixed 2005 emissions	1.26 W m ⁻² , 1° C, 0.1 mm day ⁻¹
Shindell et al. (2013)	Multimodel	All	Emis, AOD, RF	Global, 1850-2100	Fixed for RF calc.	$0.68 - 1.42 \text{ W m}^{-2}$
Rotstayn et al. (2013)	CSIRO-Mk3.6	4.5	Emis, burden, AOD, RF, T, P	Global, 1850-2100	Fixed 2005 emissions	1.46 W m ⁻² , 1.1° C, 0.1 mm day ⁻¹
Gillet et al. (2013)	CanESM2	2.6, 4.5, 8.5	Emis, burden, T	Global, 1850-2100	Fixed 2000 emissions	$0.4 - 0.7^{\circ} \mathrm{C}$
Chalmers et al. (2012)	HadGEM2-ES	2.6, 4.5	Burden, RF, AOD, CDNC, CLD, SW	Global, 2000-2037	Compare RCP2.6 and RCP4.5	See Chalmers et al. (2012)
Takemura (2012)	SPRINTARS	All	Emis, burden, AOD, RF, R _{eff} , LWP, IWP	Global, regional, 1850-2100	Compare to 2000 levels	1.72 – 1.96 W m ⁻²
Lamrque et al. (2011)	CAM5	All	Emis, burden, AOD, RF, O ₃	Global, regional, 1850-2100	Compare to 2000 levels	0.44 - 0.57 W m ⁻²
Bellouin et al. (2011)	HadGEM2-ES	All	Emis, , AOD, RF	Global, 2000-2100	Compare to 2000 levels	~1 W m ⁻²
This work	GFDL CM3	All	Emis, AOD, RF, T, P, Cld, R_{eff}	Global, regional, 1860-2100	Fixed 2005 emissions	See Table 3-5

1 Table 2: List of simulations

	RCP simulations	RCP Fixed emis. simulations	RCP Fixed SST simulations		
Simulation	RCP2.6	RCP2.6_2005AER	RCP2.6_RFP		
name	RCP4.5	RCP4.5_2005AER	RCP4.5_RFP		
	RCP6	RCP6_2005AER	RCP6_RFP		
	RCP8.5	RCP8.5_2005AER	RCP8.5_RFP		
	CM3w_RCP8.5*	CM3w_RCP8.5_2005AER*	RCP_1860		
Aerosol setup	Time-varying	SO ₂ , OC, BC emissions fixed at 2005 levels	Time-varying, aerosol forcing only		
GHG	Time-varying	Time-varying	Time varying		

2 *Simulation uses configuration with weaker aerosol effective radiative forcing (see Sect. 2.3)

- 1 Table 3: Global average difference in aerosol optical depth (AOD), forcing (RF), temperature
- 2 (T), total precipitation rate (P), liquid water path (LWP), and cloud droplet effective radius
- 3 (R_{eff}) due to aerosol emissions reductions at the end of the 21st century (2096-2100 average).
- 4 Values represent differences between RCP simulations and fixed 2005 simulations (base
- 5 cases) (RCPx.x RCPx.x_2005AER). RF values represent the difference between 2100 and
- 6 2006 total aerosol forcing.

	SO ₄ AOD	BC AOD	OC AOD	RF (W m ⁻ ²)	T (K)	P (mm d ⁻¹)	LWP (g m ⁻²)	R _{eff} (μm)
RCP2.6	-0.034	-0.0018	-0.0055	1.37	0.97	0.080	-0.555	0.584
RCP4.5	-0.033	-0.0017	-0.0081	1.25	0.81	0.078	-0.870	0.603
RCP6.0	-0.032	-0.0011	-0.0005	1.12	0.72	0.082	-0.502	0.541
RCP8.5	-0.035	-0.0014	-0.0057	1.05	1.04	0.093	-0.748	0.585
CM3w_RCP8.5*	-	-	-	~0.5	0.51	0.048	-0.218	0.472

7 *Simulation uses configuration with weaker aerosol effective radiative forcing (see Sect. 2.3)

Table 4: Regional average changes in aerosol optical depth (AOD), effective radiative forcing (RF), temperature (T), precipitation (P), liquid water path (LWP), and cloud droplet effective radius (R_{eff}) for RCP8.5 due to aerosol effects only at the end of the 21st century. Values represent differences between RCP simulations and fixed 2005 simulations (RCP8.5– RCP8.5_2005AER. Boldface values represent the largest regional change for each variable. RF values represent the difference between 2100 and 2006 total aerosol forcing.

	SO ₄ AOD	BC AOD	OC AOD	RF (W m ⁻²)	T (K)	P (mm day ⁻¹)	LWP (g m^{-2})	R_{eff} (µm)
NM	-0.13	-0.0029	-0.0039	1.53	1.81	0.125	0.852	0.649
SM	-0.053	-0.0053	-0.030	0.89	0.96	0.044	-0.553	0.571
EA	-0.49	-0.027	-0.050	2.68	2.10	0.192	-6.79	1.84
SA	-0.41	-0.020	-0.102	3.85	1.11	0.175	-4.34	1.35
EU	-0.27	-0.012	-0.025	2.79	1.45	0.123	-3.22	1.27
RU	-0.14	-0.0048	-0.0085	1.80	1.97	0.203	2.04	0.93
AU	-0.060	-0.0012	-0.0079	0.42	1.27	-0.22	-3.18	0.76
AF	-0.12	-0.0013	-0.0105	0.90	1.29	0.039	-1.11	0.87
ME	-0.20	-0.0034	-0.0119	0.42	2.07	-0.031	-2.24	1.50

1 Table 5: Same as Table 4, but for RCP2.6 and RCP2.6_2005AER

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	SO ₄ AOD	BC AOD	OC AOD	RF (W m ⁻²)	T (K)	P (mm day ⁻¹)	LWP (g m ⁻²)	R_{eff} (µm)
NM	-0.122	-0.0029	-0.0010	1.95	1.83	0.115	-0.057	0.756
SM	-0.068	-0.0067	-0.0275	1.16	0.95	0.0442	-1.58	0.508
EA	-0.43	-0.033	-0.061	3.06	1.71	0.33	-3.25	1.85
SA	-0.41	-0.024	-0.053	4.52	0.98	0.36	-2.93	1.35
EU	-0.269	-0.013	-0.017	3.20	1.65	0.087	-2.66	1.24
RU	-0.13	-0.0059	-0.0080	2.76	2.21	0.16	1.84	0.993
AU	-0.045	0.0014	0.0023	0.63	0.73	-0.107	-1.05	0.543
AF	-0.14	-0.0075	-0.070	0.97	1.16	0.016	-0.69	1.06
ME	-0.26	-0.0072	-0.016	0.67	1.75	0.039	-1.36	2.40





3 Figure 1: Region definitions. North America (NM), South and Central America (SM), Europe

4 (EU), Russia (RU), East Asia (EA), South Asia (SA), Australia (AU), Africa (AF), Middle

5 East (ME), and Arctic (AR, everything north of the dashed line)

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Figure 2: Global total emissions (anthropogenic + biomass burning) of sulfur dioxide (SO₂),
black carbon (BC), and organic carbon (OC) from 1860-2100. Historical emissions are
colored in magenta, RCP2.6 in green, RCP4.5 in blue, RCP6 in black, and RCP8.5 in red.



2 Figure 3: Globally averaged historical and future time series of top-of-atmosphere effective

3 radiative forcing (W m⁻²) from emissions of aerosols and their precursors alone.





Figure 4: Globally averaged trend in climate response variables from 1860-2100. Upper left: surface air temperature (K), upper right: total precipitation rate (mm day⁻¹), bottom left: liquid water path (kg m⁻²), bottom right: effective cloud droplet radius (μm). Shaded light colors represent the range of the three ensemble members, solid lines are the ensemble means for the RCPx.x time-varying simulations. Dashed lines are the ensemble means for the RCPx.x_2005AER control simulations (ensemble range not shown).





Figure 5: Globally averaged trend in climate variable anomalies from 2006-2100. Values represent differences between the projected RCP simulations and the fixed 2005 aerosol emissions case (RCPx.x – RCPx.x_2005AER). Upper left: surface air temperature (K), upper right: total precipitation rate (mm day⁻¹), lower left: liquid water path (LWP) (kg m⁻²), lower right: effective cloud droplet radius at cloud top (μ m). Shaded light colors represent the range of the three ensemble members, solid lines are the ensemble means. Yellow lines represent the CM3w_RCP8.5 run.

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Figure 6: Anomalies in aerosol optical depth for 2096-2100 five-year average for RCP8.5.
Values represent differences between the projected RCP aerosol optical depth and the fixed
2005 aerosol emissions case (RCP8.5 – RCP8.5_2005AER). Note the different scale for BC
AOD.



Figure 7: Changes in climate variables for a five-year average over 2096-2100 for RCP8.5.
Values represent differences between the projected RCP simulations and the fixed 2005
aerosol emissions case (RCP8.5 – RCP8.5_2005AER). Hatched areas represent statistically
significant changes at the 95% confidence level. Upper left: surface air temperature (K) ,
upper right: total precipitation rate (mm day⁻¹), lower left: liquid water path (kg m⁻²), lower
right: effective cloud droplet radius at cloud top (µm)





2 Figure 8: Same as Fig. 7, but for the case with reduced present-day effective aerosol radiative

- 3 forcing (CM3w). Values represent differences between the projected RCP simulations and the
- 4 fixed 2005 aerosol emissions case (CM3w_RCP8.5 CM3w_RCP8.5_2005AER).
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Figure 9: Regional comparison of climate response to aerosol decreases (RCP8.5 –
RCP8.5_2005AER, red bars), climate response to aerosol decreases under weaker aerosol
forcing (CM3w_RCP8.5 – CM3w_RCP8.5_2005AER, yellow bars), and climate response to
all-forcings (RCP8.5, white hatched bars) for a five-year average over 2096-2100 for RCP8.5.
Upper left: surface air temperature (K), upper right: total precipitation rate (mm day⁻¹), lower
left: liquid water path (g m⁻²), lower right: effective cloud droplet radius at cloud top (µm).
Regions are defined in Fig. 1.





Figure 10: Trends in climate parameter differences from 2005-2100 for the East Asia Region.
Values represent differences between the projected RCP simulations and the fixed 2005
aerosol emissions case (base case). Shaded light colors represent the range of the three
ensemble members, solid lines are the ensemble means. Yellow lines represent the
CM3w_RCP8.5 run.



Figure 11: Trends in climate parameter differences from 2005-2100 for the North America region. Values represent differences between the projected RCP simulations and the fixed 2005 aerosol emissions case (base case). Shaded light colors represent the range of the three ensemble members, solid lines are the ensemble means. Yellow lines represent the CM3w_RCP8.5 run.



Figure 12: Correlations of aerosol-driven changes in temperature (T), precipitation rate
(Precip), liquid water path (LWP), and cloud droplet radius (R_{eff}) with total anthropogenic
aerosol optical depth (sulfate, black carbon, and organic carbon AOD) for RCP8.5.