Simultaneous reductions in emissions of black carbon and co-emitted species will weaken the aerosol net cooling effect

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Abstract

Black carbon (BC), a distinct type of carbonaceous material formed from the incomplete combustion of fossil and biomass based fuels under certain conditions, can interact with solar radiation and clouds through its strong light-absorption ability, thereby warming the Earth's climate system. Some studies have even suggested that global warming could be slowed down in the short term by eliminating BC emission due to its short lifetime. In this study, we estimate the influence of removing some sources of BC and other co-emitted species on the aerosol radiative effect by using an aerosol-climate atmosphere-only model BCC_AGCM2.0.1_CUACE/Aero with

prescribed sea surface temperature and sea ice cover, in combination with the aerosol emissions from the Representative Concentration Pathways (RCPs) scenarios. We find that the global annual mean aerosol net cooling effect at the top of the atmosphere (TOA) will be enhanced by 0.12 W m^{-2} compared with recent past year 2000 levels if the emissions of only BC are reduced to the level projected for 2100 based on the RCP2.6 scenario. This will be beneficial for the mitigation of global warming. However, both aerosol negative direct and indirect radiative effects are weakened when BC and its co-emitted species (sulfur dioxide and organic carbon) are simultaneously reduced. Relative to year 2000 levels, the global annual mean aerosol net cooling effect at the TOA will be weakened by 1.7–2.0 W m^{-2} if the emissions of all these aerosols are decreased to the levels projected for 2100 in different ways based on the RCP2.6, RCP4.5, and RCP8.5 scenarios. Because there are no effective ways to remove the BC exclusively without influencing the other co-emitted components, our results therefore indicate that a reduction in BC emission can lead to an unexpected warming on the Earth's climate system in the future.

1 Introduction

Aerosols in the atmosphere can alter the amount of sunlight reaching the Earth, perturb the temperature structure of the atmosphere, and influence cloud cover by directly scattering sunlight (e.g., sulphate, organic carbon (OC) and nitrate) or absorbing it (e.g., black carbon (BC) and dust) (Boucher et al., 2013). Aerosol particles can also change cloud microphysical and optical properties by acting as

cloud condensation nuclei (CCN) or ice nuclei (Twomey, 1977; Albrecht, 1989; DeMott et al., 1997). These changes due to aerosols will directly or indirectly affect the climate. Since the start of the industrial era, an increase in atmospheric aerosol emissions has likely led to a net cooling of the Earth's climate system (Boucher et al., 2013).

BC has a special role in the climate system, although it accounts for less than 5% of the mass of atmospheric aerosol in most areas of the world (X. Y. Zhang et al., 2012). BC can increase the amount of solar radiation absorbed within the Earth's climate system and heat the atmosphere or surface by directly absorbing sunlight in the visible to infrared wavebands (Hansen et al., 2000; Ramanathan and Carmichael, 2008), changing the cloud amount and its brightness due to embedding into clouds (Chuang et al., 2002; Koch and Del Genio, 2010; Jacobson, 2012; Wang et al., 2013a), or by reducing the surface albedo due to deposition onto snow and ice surfaces (Wang et al., 2011; Lee et al., 2013). BC has even been considered as a potential cause of global warming (Hansen et al., 2000; Jacobson, 2010; Bond et al., 2013). Ramanathan and Carmichael (2008) compared the radiative forcings of greenhouse gases and BC, suggesting that the direct radiative forcing due to BC was larger than that due to any other greenhouse gas except CO_2. The radiative heating effect on the whole atmosphere due to BC was almost double that due to all greenhouse gases. By considering all the known ways that BC affects the climate system, Bond et al. (2013) gave an estimate of industrial-era climate forcing of +1.1 W m^{-2} due to BC with 90% uncertainty limits of +0.17 to +2.1 W m^{-2}. BC can therefore be considered the second

most important anthropogenic positive radiative forcing agent after CO_2 in the present-day atmosphere. Some studies have even suggested that global warming could be slowed down in the short term by eliminating BC emission due to its short atmospheric lifetime. For example, eliminating soot generated from fossil fuels, including BC, primary organic matter, and sulphate, was found to decrease global surface air temperature by 0.3–0.5 ℃ in the short term (about 15 year) (Jacobson, 2010). A simultaneous decrease of short-lived BC and methane through the adoption of control measures could reduce projected global mean warming by about 0.5 ℃ by 2050 (Shindell et al., 2012). However, there is a huge uncertainty and an ongoing debate in climate forcing of BC. Other studies, such as Myhre et al. (2013a), have much lower estimate of the direct radiative forcing from BC, which is reflected in the best estimate in the latest IPCC report (Boucher et al., 2013; Myhre et al., 2013b). Recent literature also suggests that the climate effect of BC may be overestimated due to overestimation of its lifetime (e.g., Hodnebrog et al., 2014; Samset et al., 2014; Q. Wang et al., 2014).

Reducing the emissions of absorptive aerosols (e.g., BC) would decrease the absorption of solar radiation by atmospheric aerosols, thereby enhancing the aerosol net cooling effect. However, BC, OC, sulphate, and some other aerosols have many common emission sources (e.g., in the emission sectors of transportation, industrial, residential, and commercial energy consumption, etc.), and they are generally co-emitted into the atmosphere (Lamarque et al., 2010). A technology-based global emission inventory of BC and OC showed that BC and primary OC particles were

co-emitted from combustion including fossil fuels, biofuels, open biomass burning, and urban waste burning (Bond et al., 2004). An inventory of air pollutant emissions in Asia supporting the Intercontinental Chemical Transport Experiment-Phase B showed that sulfur dioxide (SO_2), BC, and OC all were emitted from power, industry, residential, and transportation sources (Zhang et al., 2009). A spatially resolved biomass burning data set also indicated that BC, OC, and SO_2 were proportionally emitted from biofuel and forest fire sources (Reddy and Venkataraman, 2002). Analyses of aerosol emission trends from some important source regions showed that there were same trends for BC, OC, and SO_2 separately emitted from fossil fuel, biofuel, and biomass burning sources from 1980 to 2009 (Chin et al., 2014), which indirectly suggested the co-emissions of BC with some other aerosols. Moreover, actual operational reduction in BC emission in most of severe polluted countries, like China, is often to cut the usage of coal and other fossil fuels, as well as forbid open burning to reduce biomass burning emissions. All these major measures will result in the emission reductions in BC and its co-emitted components at the same time.

Sulphate, BC, and OC are the main aerosol species in the atmosphere, and the emissions of sulphate and OC will be reduced accordingly if the emission of BC is reduced. Both sulphate and OC are strongly scattering and hygroscopic aerosols, and they can cool the climate system by directly scattering solar radiation and increasing the cloud albedo and lifetime by acting as CCN (Boucher et al., 2013). Therefore, would global warming necessarily be slowed down by reducing BC emission in the future? This is the point of this study.

Focusing on the issue mentioned above, the impact of removing some BC sources and other co-emitted species on the aerosol radiative effects was studied in this paper by using an aerosol-climate atmosphere-only model BCC_AGCM2.0.1_CUACE/Aero (Atmospheric General Circulation Model of Beijing Climate Center, BCC_AGCM2.0.1, coupled with the aerosol model of China Meteorological Administration Unified Atmospheric Chemistry Environment for Aerosols, CUACE/Aero) (Wang et al., 2014) with prescribed sea surface temperature (SST) and sea ice cover (SIC), in combination with the Representative Concentration Pathways (RCPs) emission scenarios (van Vuuren et al., 2011) underpinning the Fifth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC AR5). In Sect. 2, we introduce the aerosol-climate model and simulation details. In Sect. 3, we present the effects of reducing only BC emission and then of the simultaneous reduction of BC and other co-emitted aerosol emissions on aerosol direct, semi-direct and indirect, and net radiative effects. Finally, our discussion and conclusions are presented in Sect. 4.

2 Model and Simulation

2.1 Model description

We use the aerosol-climate atmosphere-only model BCC_AGCM2.0.1_CUACE/Aero developed by Zhang et al. (2012a), and improved by Jing and Zhang (2013), Zhang et al. (2014), and Wang et al. (2014) in this study. The aerosol direct, semi-direct, and indirect effects (albedo and lifetime indirect

effects on stratiform clouds) have been included in BCC_AGCM2.0.1_CUACE/Aero. The model has been used to study the impact of aerosol direct radiative effect on East Asian climate (Zhang et al., 2012a), direct radiative forcing of anthropogenic aerosols (Bond et al., 2013; Myhre et al., 2013a), climate response to the presence of BC in cloud droplets (Wang et al., 2013a), effect of non-spherical dust aerosol on its direct radiative forcing (Wang et al., 2013b), anthropogenic aerosol indirect effect (Wang et al., 2014), and direct effect of dust aerosol on arid and semi-arid regions (Zhao et al., 2014).

A detailed description of BCC_AGCM2.0.1 was given by Wu et al. (2010). The model employs a horizontal resolution of T42 (approximately $2.8^{\circ} \times 2.8^{\circ}$) and a 26 layer hybrid sigma-pressure coordinate system in the vertical direction, with a rigid lid at 2.9 hPa. The time step is 20 min. However, the cloud overlap, radiation, and cloud microphysical schemes were improved in the model. The cloud overlap scheme of the Monte Carlo independent column approximation (McICA) (Pincus et al., 2003) and the Beijing Climate Center RADiation transfer model (BCC_RAD) developed by Zhang et al. (2003, 2006a, b) were used instead of the old schemes in the model (Jing and Zhang et al., 2013). These schemes have improved the accuracy of the subgrid cloud structure and its radiative transfer process (Zhang et al., 2014). A two-moment bulk cloud microphysical scheme to predict both the mass and number concentrations of cloud droplets and ice crystals (Morrison and Gettelman, 2008) was implemented into the model instead of the old one-moment bulk cloud microphysical scheme (Wang et al., 2014). The scheme of Abdul-Razzak and Ghan (2000) has been adopted

for the activation of cloud droplets.

The aerosol model CUACE/Aero is a comprehensive module incorporating emission, gaseous chemistry, transport, removal, and size-segregated multi-component aerosol algorithms based on the Canadian Aerosol Module developed by Gong et al. (2002, 2003). A detailed description of CUACE/Aero was given by Zhou et al. (2012). The mass concentrations of the main five aerosols in troposphere, i.e., sulphate, BC, OC, dust, and sea salt, can be calculated. Each aerosol type is divided into 12 bins as a geometric series for a radius between 0.005 and 20.48 μm. Aerosol optical properties from Wei and Zhang (2011) and Zhang et al. (2012b) were calculated based on the Mie theory. The refractive indices of aerosols were adopted from d'Almeida (1991). Hygroscopic growth was considered for sulphate, OC, and sea salt (Zhang et al., 2012a).

2.2 Simulation details

Six simulations were run in this study. In all simulations, the model settings were the same, whereas aerosol emissions were different. All simulations kept greenhouse gas concentrations fixed at year 2000 levels in order to obtain the effect of change in aerosol emissions exclusively. Table 1 gives the emission setups in all simulations. As a base case, the first simulation (SIM1) used emissions of SO_2, BC, and OC for the year 2000, representing the aerosol effect for recent past. In the second simulation (SIM2), BC emission in 2100 under the RCP2.6 scenario was used, but the emissions of SO_2 and OC were the same as those in SIM1. In the third simulation (SIM3), BC

emission in 2100 under the RCP2.6 scenario was also used, but the emissions of SO_2 and OC used were those for 2100 under the RCP8.5 scenario. In the fourth simulation (SIM4), the emissions of SO_2, BC, and OC for 2100 under the RCP2.6 scenario were used. In the fifth simulation (SIM5), BC emission in 2100 under the RCP2.6 scenario was used, but the emissions of SO_2 and OC used corresponded to the 2100 emission of BC under the RCP2.6 scenario by multiplying them with the ratios of the emissions of SO_2 and OC with BC in 2000. The ratios were calculated and applied for each individual grid box and month. In the sixth simulation (SIM6), the emissions of SO_2, BC, and OC in 2100 under the RCP4.5 scenario were used. Aerosol emission inventories from fossil fuel, biofuel, and biomass burning for the year 2000 given by Lamarque et al. (2010) were used. The emission dataset of RCPs scenarios were described by van Vuuren et al. (2011) and can be obtained from http://tntcat.iiasa.ac.at:8787/RcpDb/dsd?Action=htmlpage&page=about. The biomass burning emissions are also changed when using RCPs scenarios. The National Centers for Environmental Prediction (NCEP) reanalysis climatological data on a Gaussian grid was used as the initial field (downloaded from http://www.cesm.ucar.edu/models/atm-cam/download/). Data for the prescribed annual cycle of monthly mean SST and SIC based on the 21-year (1981–2001) climatology from the Hadley Centre (Hurrell et al., 2008) were used in these simulations. Each simulation was run for 20 years, and the simulation data for the last 10 years were averaged and analyzed.

The difference between SIM2 and SIM1 shows the impact on aerosol radiative

effects (AREs) of reducing only BC emission maximally in the four RCPs scenarios. The difference between SIM3 and SIM1 indicates the effect of maximally reducing the emission of absorbing BC, combined with the least reduction in the emissions of precursor (SO_2) of scattering sulphate and OC on AREs. The differences between SIM4 and SIM1, between SIM5 and SIM1, and between SIM6 and SIM1 show the effects of a simultaneous reduction of SO_2, BC, and OC emissions under the RCP2.6 scenario, a reduction of the BC emission with a simultaneous reduction of the emissions of SO_2 and OC (in terms of their ratios with BC), and a simultaneous reduction in the emissions of SO_2, BC, and OC under the RCP4.5 scenario (representing a medium-low emission pathway), on AREs, respectively.

The aerosol direct effect (ADE) was obtained by calling the radiation routine two times (Ghan et al., 2012):

$$\Delta\mathrm{ADE} = \Delta(F - F_{\mathrm{clean}}), \quad (1)$$

where F is the radiative flux at the top of the atmosphere (TOA) and F_{clean} is the flux calculated as a diagnostic with aerosol scattering and absorption excluded; Δ is the difference between 2000 and 2100. The change in cloud radiative forcing (CRF) was used as an approximate way of quantifying the change in combination of the aerosol semi-direct and indirect effects:

$$\Delta\mathrm{CRF} = \Delta(F - F_{\mathrm{clear}}), \quad (2)$$

where F_{clear} is the flux calculated as a diagnostic with clouds neglected. The change in aerosol net effect was assessed by the change in net radiation flux at the TOA (ΔF) (Ghan et al., 2012). We didn't perform additional simulations in which aerosol

scattering and absorption were neglected to exclusively diagnose the effect of aerosols on CRF according to the method by Ghan et al. (2012) and Ghan (2013). Thus, the difference in aerosol net effect is not equal to the sum of ΔADE and ΔCRF in this study.

3 Results

3.1 Aerosol optical depth for present-day conditions

The simulation performance of BCC_AGCM2.0.1_CUACE/Aero has been given by Wang et al. (2014) in detail. They demonstrated that the model has a good ability to simulate aerosols, cloud properties, and meteorological fields. However, we replace the aerosol emission from AeroCom with those given by Lamarque et al. (2010) for present-day conditions in this work. Thus, a comparison of simulated annual mean aerosol optical depth (AOD) with satellite retrievals is shown in Figure 1. The simulated AODs range from 0.3 to 0.6 over the Sahara Desert and are from 0.15 to 0.3 in nearby Arabian areas due to the large dust loading. The AODs are mainly between 0.2 and 0.4 in eastern China, and exceed 0.15 in eastern North America and West Europe due to the large emissions of anthropogenic aerosols. The AODs are above 0.1 over most subtropical oceans because of the contribution of sea salt and sulphate. The model generally reproduces the geographical distribution of AOD well, but it significantly underestimates the AODs over South Asia, eastern China, and tropical oceans. These errors could be caused by several factors such as uncertainties in the aerosol sources, coarse model resolution, the uncertainties of physical processes, and

243 the absence of nitrate, ammonium and secondary organic aerosols in the model
244 (Zhang et al., 2012a).
245
246 **3.2 The effect of aerosol reductions**
247 **3.2.1 Global mean statistics**
248 Tables 2 and 3 show the global emission amounts and annual mean column
249 burdens of aerosols in all simulations and differences in AREs among them. The
250 global emission amount of BC is reduced from 7.8 Tg yr^{-1} at present to 3.3 Tg yr^{-1} at
251 the end of this century under the RCP2.6 scenario due to the operation of various
252 control measures. The global annual mean of simulated BC burden is decreased from
253 0.17 mg m^{-2} in SIM1 to 0.08 mg m^{-2} in SIM2, assuming that only BC emission is
254 reduced under the RCP2.6 scenario (Table 2). The reduction in the mass concentration
255 of atmospheric BC results in less direct absorption of solar radiation by atmospheric
256 aerosols, thereby causing the global annual mean aerosol direct radiative effect at the
257 TOA to be enhanced by 0.07 W m^{-2}. This indicates that the net cooling effect is
258 enhanced. The multi-model comparison showed that our model had much lower
259 normalized radiative forcing for BC than most of the other models (Myhre et al.,
260 2013a). Thus, the change in aerosol direct radiative effect is quite small despite the
261 strong emission reduction for BC. The reduction in the BC concentration also
262 weakens the aerosol semi-direct effect, resulting in an increase of 0.11 W m^{-2} in the
263 absolute value of the global annual mean net CRF (Table 3). Of which, the shortwave
264 cloud forcing (SWCF) and longwave cloud forcing (LWCF) components are

enhanced by 0.14 and 0.03 W m^{-2}, respectively. It is noted that the change in CRF is a combined effect of decrease in cloud evaporation and increase in cloud cover caused by declining BC, changes in other aerosol concentrations due to adjustment of the atmosphere to BC reduction, and the resulting changes in cloud properties. However, the slight decrease in the sulphate mass concentration due to changes in meteorological fields caused by BC reduction partially offsets the net cooling effect in SIM2 compared with SIM1. Consequently, the global annual mean aerosol net cooling effect at the TOA is enhanced by 0.12 W m^{-2} compared with recent past levels when just BC emission is reduced to the level projected for the end of this century under the RCP2.6 scenario (Table 3).

Many previous studies mentioned in Sect. 1 have indicated that there are several common sources of SO_2, BC, and OC (Reddy and Venkataraman, 2002; Zhang et al., 2009; Lamarque et al., 2010; Chin et al., 2014). SO_2 and OC emissions are likely to be reduced proportionally when BC emission is decreased, as there is no effective way of removing BC exclusively without influencing the other co-emitted components. Therefore, we considered four different ways to simultaneously reduce the emissions of SO_2, BC, and OC to the levels projected for the end of this century under the RCP2.6, RCP4.5, and RCP8.5 scenarios. Then, we calculated the effects of emission reductions of all these aerosols on radiation fluxes in SIM3 to SIM6. It can be seen from Table 2 that the global emissions of SO_2, BC, and OC are decreased to 12.9–25.7 Tg yr^{-1}, 3.3–4.3 Tg yr^{-1}, and 20.0–25.3 Tg yr^{-1} under these three scenarios, respectively. Thus, the global annual mean burdens of sulphate, BC, and OC are

reduced by different levels (63–72, 51–55, and 25–31 %, respectively). The concurrent reductions in scattering sulphate and OC burdens weaken the global annual mean aerosol direct radiative effect at the TOA by 0.25–0.3 W m^{-2}, although the absorbing BC burden is also significantly reduced in SIM3 to SIM6. Additionally, sulphate and OC particles can act as CCN due to their hygroscopicity, so any decrease in their emissions would decrease CCN concentrations, then decreasing cloud lifetime and albedo, thereby weakening the negative SWCF. As can be seen from Table 3, the absolute values of the global annual mean SWCF are weakened by 0.87–1.3 W m^{-2} due to simultaneous reductions in emissions of SO_2, BC, and OC. The quick adjustment of the atmosphere to aerosol effects leads to changes in LWCF, causing the longwave cooling by 0.07–0.2 W m^{-2} in SIM3 to SIM6 compared with SIM1. It partly offsets the shortwave warming. The absolute values of global annual mean net CRF are decreased by 0.8–1.1 W m^{-2} in SIM3 to SIM6 compared with SIM1, which greatly exceed the changes in the aerosol direct radiative effect. This is consistent with results obtained by Chen et al. (2010), who reported that a reduction in BC emission would dampen aerosol indirect forcing. Finally, the global annual mean aerosol net cooling effect at the TOA is weakened by 1.7–2.0 W m^{-2} when the emissions of SO_2, BC, and OC are simultaneously reduced to the levels projected for the end of this century based on three different RCP scenarios (Table 3).

3.2.2 Global distributions

Figure 2 shows the global distributions of simulated annual mean sulphate, BC,

309 and OC burdens under all six simulations. As can be seen from Figure 2b, the BC
310 column burdens are significantly decreased in areas with high BC emission such as
311 East Asia, South Asia, central Africa and South America, eastern North America, and
312 Western Europe compared with recent past levels when the emission of only BC is
313 reduced. Changes in other aerosol burdens are not obvious. The reduction in the BC
314 concentration weakens the direct absorption of solar radiation by atmospheric aerosols,
315 leading to a cooling effect at the TOA in these regions. The largest cooling exceeds 1
316 W m^{-2} in China, Europe, and eastern North America (Fig. 3a). The numbers of
317 activated sulphate, OC, and dust particles are increased over East and South Asia,
318 Mediterranean regions, North America and Africa due to the fast adjustment in
319 meteorological fields caused by declining BC (figure not shown), which leads to the
320 increase in CCN concentration (Fig. 4a). Higher CCN concentrations can produce
321 more cloud droplet numbers, with the maximum increase in annual mean column
322 cloud droplet number concentrations (CDNCs) being up to 0.6 $\times 10^{10}$ m^{-2} (Fig. 5a).
323 This enhances the negative SWCF over those regions (Fig. 6a). In addition, the
324 decrease in the absorption ability of aerosols weakens the cloud evaporation and
325 increases the cloud fraction, which further enhances the negative SWCF over the
326 regions with high BC emission and some oceans (Fig. 6a). However, the LWCF is
327 also increased over most areas with enhanced negative SWCF (Fig. 6a), which can
328 partly offset the shortwave cooling. Finally, only the reduction of BC emission results
329 in an increase of more than 2 W m^{-2} in the annual mean aerosol net cooling effect at
330 the TOA over most regions with large BC emission (Fig. 7a).

Figures 2c–f show that there are different levels of reduction in the annual mean sulphate, BC, and OC burdens in SIM3 to SIM6, with decreases of up to 2.0–5.0 mg S m^{-2}, 0.2–1.0 mg m^{-2} and 2.0–6.0 mg m^{-2} in most areas, respectively, when all aerosol emissions are reduced. The combined reduction in scattering and absorbing aerosols weakens the aerosol direct radiative effect at the TOA by over 1 W m^{-2} for most of the Northern Hemisphere (NH) compared with SIM1 (Fig. 3b–e). The CCN concentrations are greatly decreased over the globe, except for individual regions, mainly due to the emission reductions in hygroscopic sulphate and OC, especially over the middle latitudes of the NH (Fig. 4b–e). Correspondingly, the CDNCs are significantly decreased in SIM3 to SIM6 compared with SIM1. The largest decreases in annual mean column CDNCs exceed 5×10^{10} m^{-2} in Western Europe, North America, and eastern China (Fig. 5b–e). Decreased CDNCs can decrease the cloud albedo and lifetime and weaken the negative SWCF in the regions with high anthropogenic aerosol emissions such as North America and Europe (Fig. 6b–e). The negative SWCFs are enhanced due to the increase in low cloud fraction over most of South and East Asia, though the CCNs are clearly decreased. The shortwave warmings (coolings) are also compensated by the longwave coolings (warmings) over most regions (Fig. 6b–e). Finally, the annual mean aerosol net cooling effect at the TOA is weakened over a range of 2.0–10.0 W m^{-2} due to the changes in emissions of all aerosols over most regions of the NH that have large anthropogenic aerosol emissions (Fig. 7b–e).

4 Discussion and conclusions

It has been argued that eliminating BC emission would be an effective measure to slow down global warming and environmental pollution. In this study, we assess the impact of removing some sources of BC and other co-emitted species on aerosol radiative effects by using an aerosol-climate atmosphere-only model BCC_AGCM2.0.1_CUACE/Aero with prescribed SST and SIC, in combination with the RCP scenarios. Compared with the aerosol effect for recent past, the global annual mean aerosol net cooling effect at the TOA is enhanced by 0.12 W m^{-2} due to a decrease in the direct absorption of solar radiation and aerosol semi-direct effect when BC emission is reduced exclusively to the level projected for the end of this century under the RCP2.6 scenario. The annual mean aerosol net cooling effect at the TOA is enhanced by more than 2.0 W m^{-2} in eastern China, northern India, and Mediterranean regions. Therefore, a reduction of BC emission alone could ideally mitigate global warming.

However, our results suggest that associating with the reduction of net cooling effects directly from aerosols, the aerosol indirect effect is also weakened when emissions of SO_2, BC, and OC are simultaneously reduced in different ways to the levels projected for the end of this century under the RCP2.6, RCP4.5, and RCP8.5 scenarios. Relative to the aerosol effect for recent past, the total global annual mean aerosol net cooling effect at the TOA is weakened by 1.7–2.0 W m^{-2} with the reduction according to potential actual conditions in the emission of all these aerosols (i.e., BC and the major co-emitted species). The main cooling regions are over East

375 Asia, Western Europe, eastern North America, and central Africa, with the largest
376 change exceeding 10.0 W m^{-2}. This is somewhat consistent with the results given by
377 Gillett and Salzen (2013) and Levy et al. (2013), who also reported that the reduction
378 in atmospheric aerosols will weaken the aerosol cooling effect in the future.

379 This study highlights that reducing only BC emission could play a positive role
380 in mitigating global warming and environmental pollution, and would be beneficial to
381 human health. However, the emissions of some co-emitted scattering aerosols and
382 their precursor gases will be inevitably reduced when BC emission is reduced due to
383 their homology. Therefore, reducing BC emission could lead to unexpected warming
384 on the Earth's climate in the future, unless certain technical advances in emission
385 reduction technology are available for removal of the BC exclusively without
386 influencing the other co-emitted components.

387 There exists large uncertainty in BC radiative forcing (Bond et al., 2013;
388 Boucher et al., 2013; Myhre et al., 2013a, 2013b). One reason for the uncertainty is
389 from the biases of current emission inventories of BC, mostly obtained from the
390 so-called bottom-up approach (Cohen and Wang, 2014). Cohen and Wang (2014)
391 provided a global-scale top-down estimation of BC emissions, a factor of more than 2
392 higher than commonly used global BC emissions data sets, by using a Kalman Filter
393 method. If present-day BC emissions have been substantially underestimated, increase
394 in aerosol net cooling effect may be larger due to only reduction in BC emission.
395 Furthermore, co-emissions of other compounds with BC, such as CO_2, might be more
396 important than SO_2 and OC (Rogelj et al., 2014). The reduction in CO_2 can mitigate

global warming when reducing BC. However, it is very difficult to fully obtain the ratios of BC with its co-emitted components due to the complexity of emission sources and diversity of energy structure in different regions. These bring about large uncertainties for the relevant research.

Because of the potential uncertainties mentioned above, we need to ongoingly improve our understanding on emissions of BC and its co-emitted species through a lot of observation and analysis. We also encourage other modeling groups to perform similar simulations to check the robustness of these results.

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661 **Table 1.** Simulation setups.

Simulation	BC emission	OC & SO_2 emissions	Interpretation (compared to SIM1)
SIM1	year-2000	year-2000	Recent past reference emissions.
SIM2	RCP2.6 year-2100	year-2000	Maximal reduction in BC; no reductions in OC & SO2.
SIM3	RCP2.6 year-2100	RCP8.5 year-2100	Maximal reduction in BC; minimal reductions in OC & SO2.
SIM4	RCP2.6 year-2100	RCP2.6 year-2100	Simultaneous maximal reductions in BC, OC & SO2.
SIM5	RCP2.6 year-2100	RCP2.6 year-2100 BC by multiplying the ratios of the emissions of OC & SO2 with BC in 2000	Maximal reduction in BC; simultaneous reductions of OC & SO2 in terms of their ratios with BC in recent past
SIM6	RCP4.5 year-2100	RCP4.5 year-2100	Medium-low reductions in BC, OC & SO2.

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677 **Table 2.** Global amounts of aerosol emissions and annual means of aerosol burdens.

	SIM1	SIM2	SIM3	SIM4	SIM5	SIM6
Emission (Tg yr^{-1})						
SO2	107.4	107.4	25.7	12.9	19.8	22.2
BC	7.8	3.3	3.3	3.3	3.3	4.3
OC	35.8	35.8	23.9	25.3	24.9	20.0
Burden (mg m^{-2})						
Sulphate	3.5	3.4	1.3	0.98	1.1	1.2
BC	0.17	0.079	0.078	0.077	0.078	0.084
OC	1.6	1.6	1.2	1.2	1.2	1.1
Dust	39.9	39.9	39.9	40.6	42.7	42.8
Sea salt	14.2	14.2	14.0	14.0	14.0	14.1

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694 **Table 3.** Global annual mean differences of aerosol direct (DRF), semi-direct and
695 indirect (CRF), and net effect at the TOA (FNT) (Positive values mean incoming,
696 units: W m^{-2}) in different simulations*.

	SIM1	ΔSIM2	ΔSIM3	ΔSIM4	ΔSIM5	ΔSIM6
DRF	–2.01	–0.07 ±0.05	+0.27 ±0.03	+0.28 ±0.05	+0.25 ±0.03	+0.3 ±0.02
SWCF	–49.0	–0.14 ±0.2	+0.87 ±0.3	+1.3 ±0.14	+1.1 ±0.17	+1.02 ±0.2
LWCF	+27.8	+0.03 ±0.09	–0.07 ±0.08	–0.2 ±0.1	–0.19 ±0.08	–0.14 ±0.1
CRF	–21.2	–0.11 ±0.17	+0.8 ±0.3	+1.1 ±0.1	+0.91 ±0.11	+0.88 ±0.18
FNT	–0.66	–0.12 ±0.28	+1.7 ±0.2	+2.0 ±0.19	+1.8 ±0.14	+1.8 ±0.21

697 * DRF, SWCF, LWCF and CRF, and FNT in the SIM1 column are aerosol direct radiative forcing, shortwave, longwave and net
698 cloud radiative forcing, and net radiation flux at the TOA (units: W m^{-2}) in SIM1, respectively. Values in the ΔSIM2 – ΔSIM6
699 columns represent the changes of corresponding variables in these simulations vs. those in SIM1.

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Figure captions:

Figure 1. Global distributions of simulated and observed annual mean AOD at 550 nm. (a) Simulation and (b) MODIS&MISR (van Donkelaar et al., 2010).

Figure 2. Global distributions of simulated annual mean aerosol column burdens (units: mg m^{-2}).

Figure 3. Global distributions of difference in simulated annual mean aerosol direct effect (units: W m^{-2}). (a) SIM2 − SIM1, (b) SIM3 − SIM1, (c) SIM4 − SIM1, (d) SIM5 − SIM1, and (e) SIM6 − SIM1.

Figure 4. Global distributions of difference in simulated annual mean CCN concentration at surface (units: cm^{-3}). (a) SIM2 − SIM1, (b) SIM3 − SIM1, (c) SIM4 − SIM1, (d) SIM5 − SIM1, and (e) SIM6 − SIM1.

Figure 5. Global distributions of difference in simulated annual mean column CDNC (units: 10^{10} m^{-2}). (a) SIM2 − SIM1, (b) SIM3 − SIM1, (c) SIM4 − SIM1, (d) SIM5 − SIM1, and (e) SIM6 − SIM1.

Figure 6. Global distributions of difference in simulated annual mean SWCF and LWCF (units: W m^{-2}). (a) SIM2 − SIM1, (b) SIM3 − SIM1, (c) SIM4 − SIM1, (d) SIM5 − SIM1, and (e) SIM6 − SIM1.

Figure 7. Global distributions of difference in simulated annual mean aerosol net effect (units: W m^{-2}). (a) SIM2 − SIM1, (b) SIM3 − SIM1, (c) SIM4 − SIM1, (d) SIM5 − SIM1, and (e) SIM6 − SIM1.

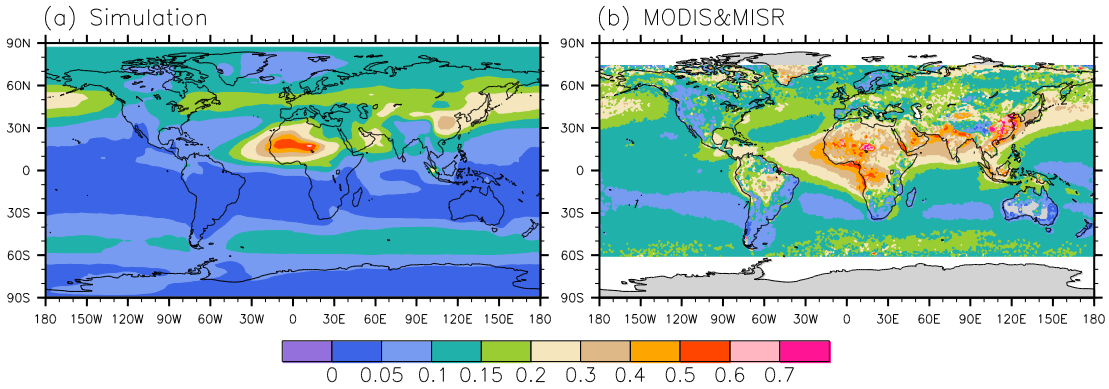

Figure 1. Global distributions of simulated and observed annual mean AOD at 550 nm. (a) Simulation and (b) MODIS&MISR (van Donkelaar et al., 2010).

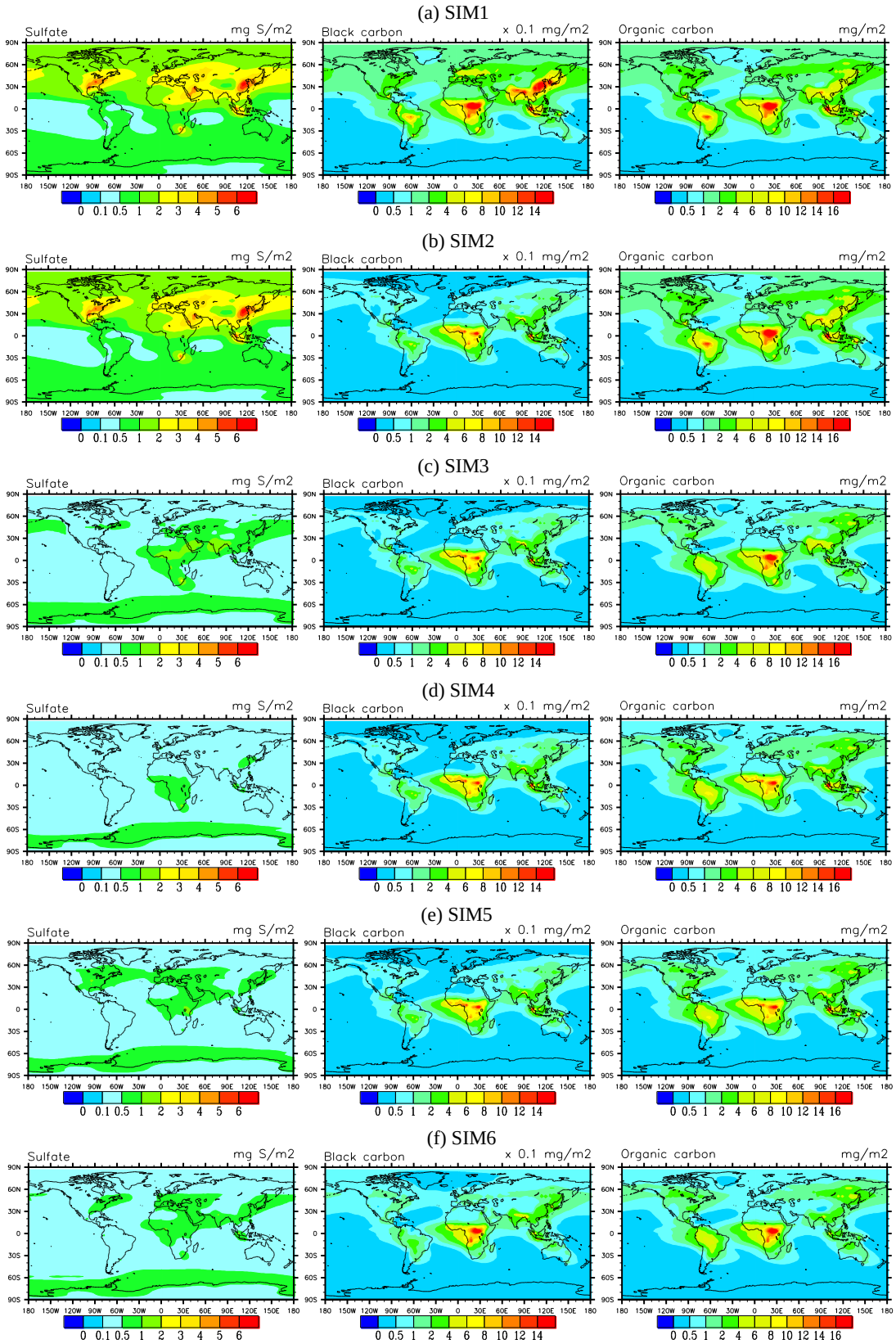

Figure 2. Global distributions of simulated annual mean aerosol column burdens (units: mg m^{-2}).

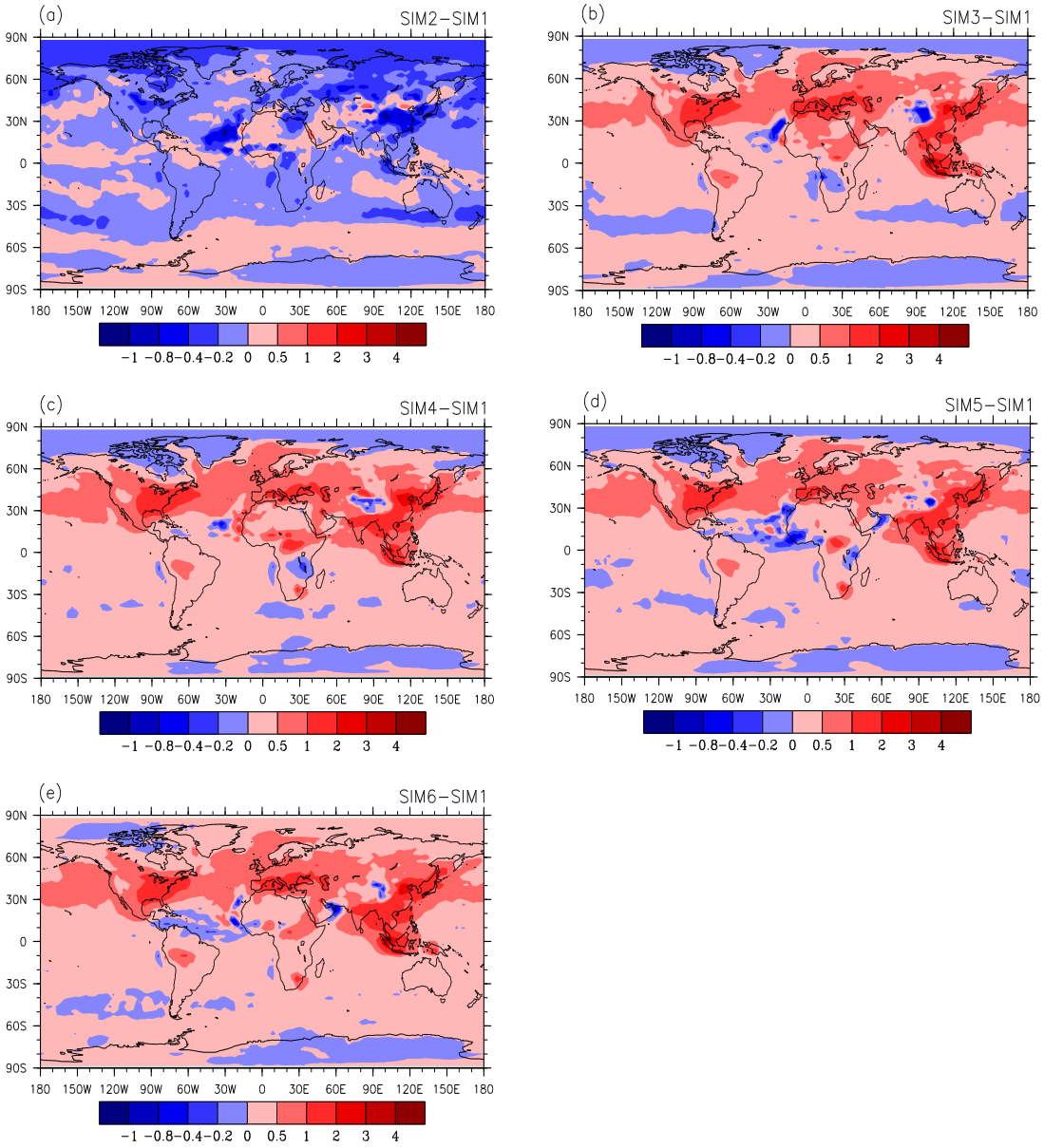

Figure 3. Global distributions of difference in simulated annual mean aerosol direct effect (units: W m^{-2}). (a) SIM2 − SIM1, (b) SIM3 − SIM1, (c) SIM4 − SIM1, (d) SIM5 − SIM1, and (e) SIM6 − SIM1.

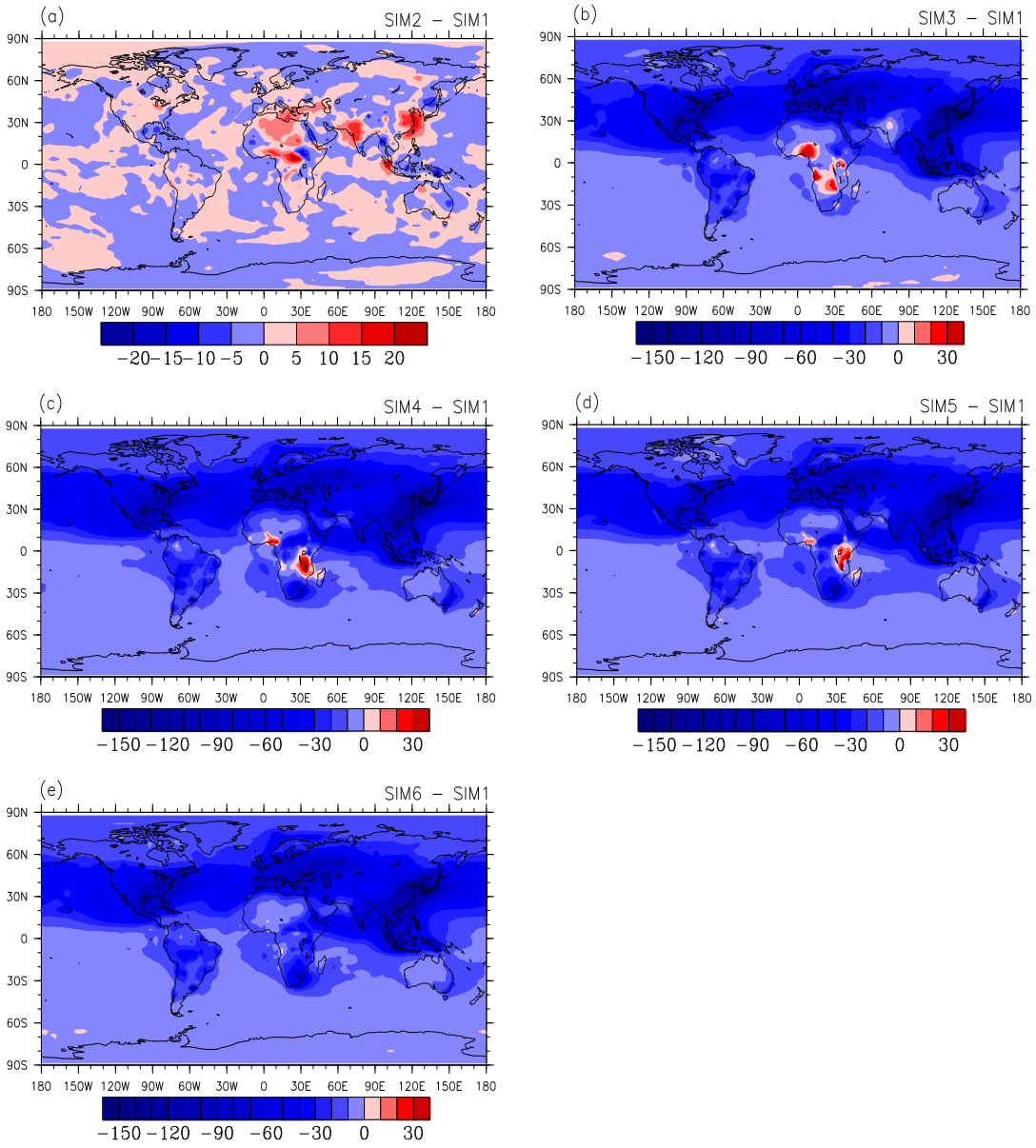

Figure 4. Global distributions of difference in simulated annual mean CCN concentration at surface (units: cm^{-3}). (a) SIM2 − SIM1, (b) SIM3 − SIM1, (c) SIM4 − SIM1, (d) SIM5 − SIM1, and (e) SIM6 − SIM1.

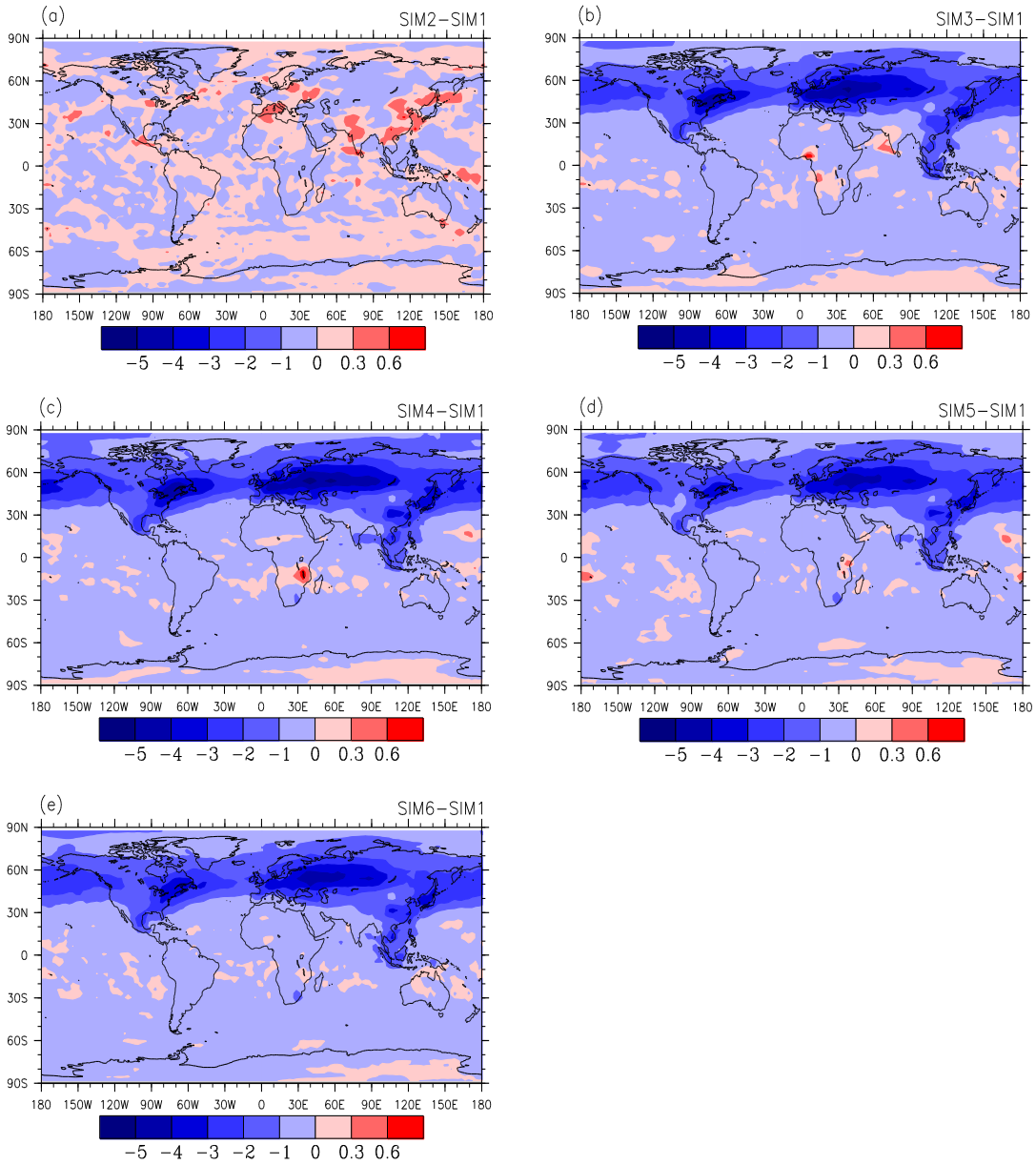

Figure 5. Global distributions of difference in simulated annual mean column CDNC (units: 10^{10} m^{-2}). (a) SIM2 − SIM1, (b) SIM3 − SIM1, (c) SIM4 − SIM1, (d) SIM5 − SIM1, and (e) SIM6 − SIM1.

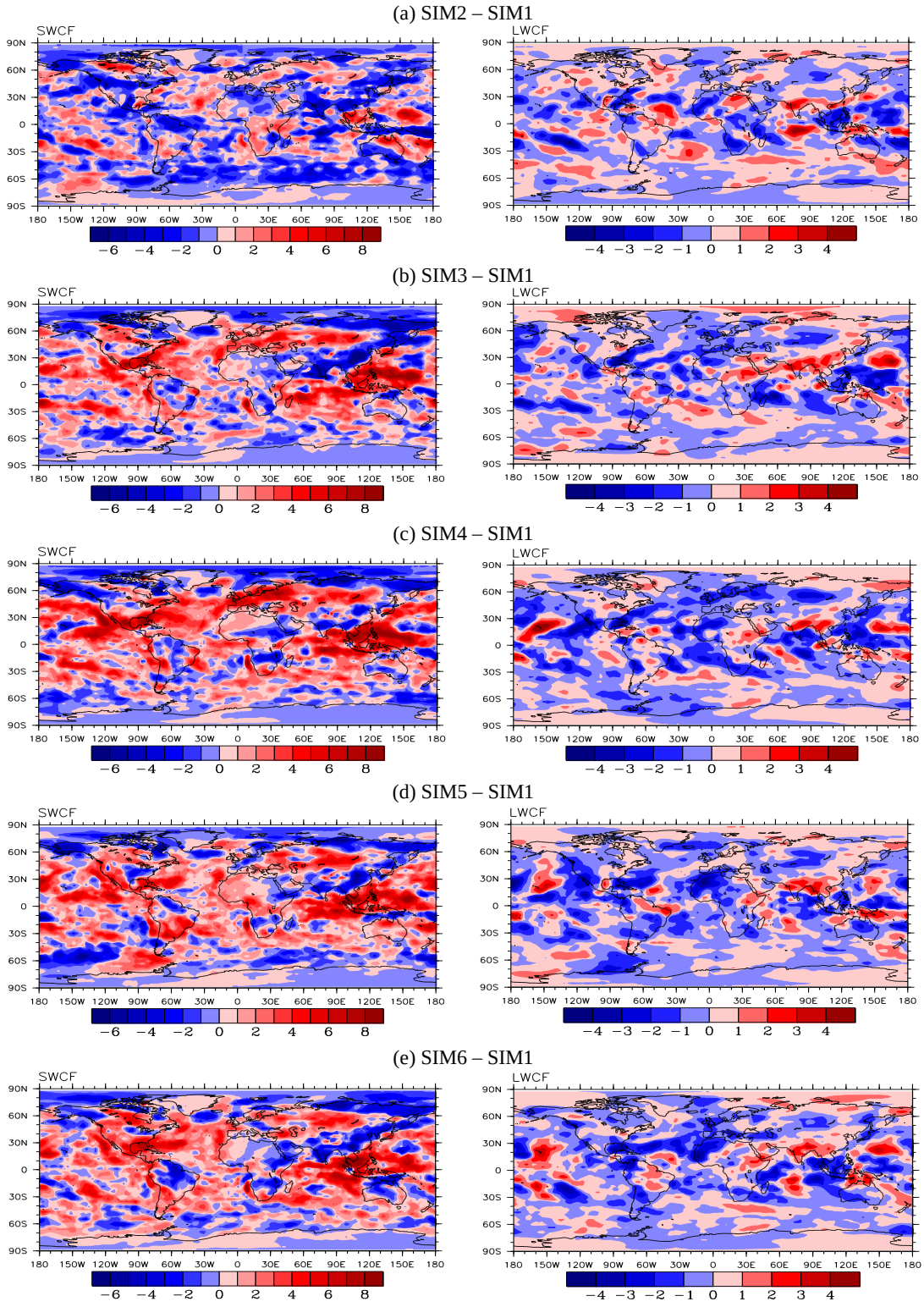

Figure 6. Global distributions of difference in simulated annual mean SWCF and LWCF (units: W m^{-2}). (a) SIM2 − SIM1, (b) SIM3 − SIM1, (c) SIM4 − SIM1, (d) SIM5 − SIM1, and (e) SIM6 − SIM1.

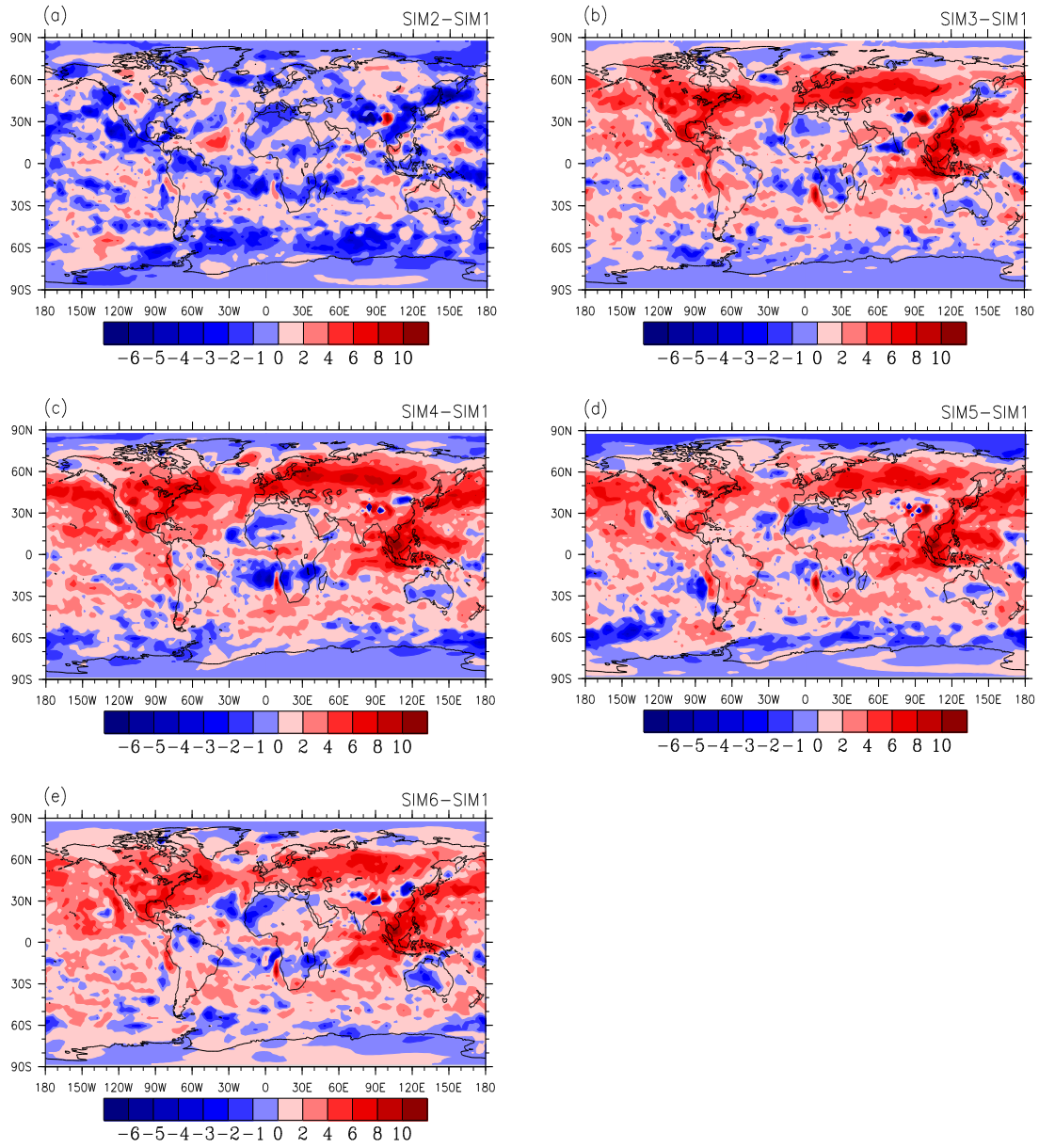

Figure 7. Global distributions of difference in simulated annual mean aerosol net effect (units: W m^{-2}). (a) SIM2 − SIM1, (b) SIM3 − SIM1, (c) SIM4 − SIM1, (d) SIM5 − SIM1, and (e) SIM6 − SIM1.