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Anthropogenic Direct Radiative Forcing of Tropospheric Ozone and Aerosols from 1850 to 2000 Estimated with IPCC AR5 Emissions Inventories

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Abstract This study estimates direct radiative forcing by tropospheric ozone and all aerosols between the years 1850 and 2000, using the new IPCC AR5 (the Intergovernmental Panel on Climate Change Fifth Assessment Report) emissions inventories and a fully coupled chemistry-aerosol general circulation model. As compared to the previous Global Emissions Inventory Activity (GEIA) data, that have been commonly used for forcing estimates since 1990, the IPCC AR5 emissions inventories report lower anthropogenic emissions of organic carbon and black carbon aerosols and higher sulfur and NO_x emissions. The simulated global and annual mean burdens of sulfate, nitrate, black carbon (BC), primary organic aerosol (POA), secondary organic aerosol (SOA), and ozone were 0.79, 0.35, 0.05, 0.49, 0.34, and 269 Tg, respectively, in the year 1850, and 1.90, 0.90, 0.11, 0.71, 0.32, and 377 Tg, respectively, in the year 2000. The estimated annual mean top of the atmosphere (TOA) direct radiative forcing of all anthropogenic aerosols based on the AR5 emissions inventories is –0.60 W m^{-2} on a global mean basis from 1850 to 2000. However, this is –2.40 W m^{-2} when forcing values are averaged over eastern China (18–45°N and 95–125°E). The value for tropospheric ozone is 0.17 W m^{-2} on a global mean basis and 0.24 W m^{-2} over eastern China. Forcing values indicate that the climatic effect of aerosols over eastern China is much more significant than the globally averaged effect.

Keywords: IPCC AR5 emissions inventories, aerosols, tropospheric ozone, direct radiative forcing

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1 Introduction

Tropospheric ozone and aerosols have been shown to play important roles in regional and global climate change (Huebert, et al., 2003; Mickley et al., 2004; Ramanathan and Feng, 2009). Aerosols perturb the Earth's radiation budget by scattering or absorbing solar radiation (direct effect) and by altering cloud properties (indirect effect) (Ramaswamy et al., 2001). Tropospheric ozone traps the outgoing longwave radiation and induces a warming effect. The Intergovernmental Panel on Climate Change (IPCC) Fourth Assessment Report (AR4) (IPCC, 2007) shows that the global mean top of the atmosphere (TOA) direct radiative forcing by all aerosols is -0.5 ± 0.4 W m^{-2}, with a medium-low level of scientific understanding. The TOA direct radiative forcings of individual aerosol species are found to be –0.4±0.2 W m^{-2} by sulfate, –0.05± 0.05 W m^{-2} by fossil fuel organic carbon, 0.2±0.15 W m^{-2} by fossil fuel black carbon, 0.03±0.12 W m^{-2} by biomass burning, and –0.1±0.1 W m^{-2} by nitrate (IPCC, 2007). The tropospheric O_3 forcing at the tropopause is estimated to be 0.35 W m^{-2} (IPCC, 2007).

The large uncertainties associated with the forcing estimates partly arise from the uncertainties in emissions inventories. A new global emission inventory compiled for the IPCC Fifth Assessment Report (AR5) has recently been released (ftp://ftp-ipcc.fz-juelich.de/). The gridded inventory includes ozone precursors, aerosol precursors, and aerosols from biomass burning and anthropogenic sources. These new AR5 inventories provide emissions data for model intercomparisons that will be presented in the IPCC AR5 report.

China is a rapidly developing country with relatively higher concentrations of tropospheric ozone and aerosols (Chan and Yao, 2008). Studies have shown that climate change in China is closely related to the aerosols (Zhao et al., 2006; Qian et al., 2007; Duan and Mao, 2009). A few studies have examined forcing by limited aerosol species such as sulfate or black carbon (Giorgi et al., 2002; Wang et al., 2003; Qian et al., 2003; Wu et al., 2004, 2008), but no studies, to our knowledge, have estimated anthropogenic direct radiative forcing by all aerosol species in China. Some recent studies estimated forcing based on ground or satellite observations of aerosol optical depth (AOD) and solar irradiance (Li et al., 2007; Xia et al., 2007a, b). These studies accounted for the effects of both natural and anthropogenic aerosols.

In this work, we estimate anthropogenic direct radiative forcing by tropospheric O_3 and aerosol species including sulfate, nitrate, black carbon (BC), primary organic aerosol (POA), and secondary organic aerosol (SOA). To do this, we use a fully coupled chemistry-aerosol general circulation model and the IPCC AR5 emissions inventories. We compare forcing values averaged over eastern China with global mean values to understand the roles of tropospheric ozone and aerosols in regional climate change in China.

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2 Model description and emissions

2.1 The unified model

The model used in this study is a unified tropospheric chemistry-aerosol model in the Goddard Institute for Space Studies (GISS) general circulation model (GCM) II' (Liao et al., 2003, 2004; Liao and Seinfeld, 2005; Liao et al., 2006). The GISS GCM II' has a horizontal resolution of 4° (latitude) by 5° (longitude) and 9 σ vertical layers from the surface to 10 hPa (Rind and Lerner, 1996). The GCM is coupled with a mixed-layer ocean described by Hansen et al. (1984). The model includes a detailed simulation of tropospheric O_3-NO_x-hydrocarbon chemistry, as well as sulfate, nitrate, ammonium, BC, POA, SOA, sea salt, and mineral dust aerosols. The chemical mechanism includes 225 chemical species and 346 reactions for simulating gas-phase species and aerosols. The partitioning of ammonia and nitrate between the gas and aerosol phases is determined by the on-line thermodynamic equilibrium model ISORROPIA (Nenes et al., 1998). SOA forms from oxidation of biogenic monoterpenes and isoprene, which is based on equilibrium partitioning and experimentally determined yield parameters (Griffin et al., 1999a, b; Chung and Seinfeld, 2002). For forcing calculation, all aerosols are assumed to be internally mixed. The refractive index of the mixed aerosols is calculated by volume-weighting the refractive index of each aerosol species and associated water (Sloane, 1984, 1986). The refractive index for each aerosol species follows that listed in Liao et al. (2004). The two-way coupling between gas-phase chemistry and aerosols provides consistent chemical fields for aerosol dynamics. It also gives aerosol mass for heterogeneous processes and calculations of gas-phase photolysis rates.

2.2 The IPCC AR5 emissions inventories

The IPCC AR5 anthropogenic emissions of NO_x, NH_3, BC, OC (Organic Carbon), CO, SO_2, and NMVOC (non-methane volatile organic compounds) include those from agricultural, industrial, energy, and transportation sectors. These anthropogenic emissions for each grid cell are given as annual mean values, which are distributed into each month based on the default monthly profiles in the unified model. Monthly shipping emissions of NO_x, BC, OC, CO, SO_2, and NMVOC are provided. Monthly aircraft emissions of NO_x, BC, and SO_2 have vertical distributions with 25 layers extending from 1 km to 25 km. Monthly biomass burning emissions consider emissions from wild fires of grasslands and forests. All the emissions inventories have a horizontal resolution of 0.5° (latitude) by 0.5° (longitude). Historical variations in emissions over 1850–2000 are given at a 10-yr interval. The IPCC AR5 emissions used in this study are summarized in Table 1.

Natural emissions in the model include dimethyl sulfide (DMS) from the oceans, NO_x from lightning and soil, NH_3 from the oceans and soil, sea salt, and mineral dust, which are calculated based on the meteorological parameters simulated in the model (described by Liao et al., 2006).

Table 1 The IPCC AR5 anthropogenic emissions used in the unified model.

	Global		Eastern China	
	1850	2000	1850	2000
NO_x, Tg N yr^{-1}				
Fossil fuel combustion	0.58	26.5		
Biomass burning	4.8	5.4		
Aircraft *	0	6.1		
Ships	0.04	5.4		
Total	5.42	43.4	0.2	4.9
CO, Tg CO yr^{-1}				
Fossil fuel combustion	63.1	608.3		
Biomass burning	322.5	459.2		
Ships	0.01	1.2		
Total	385.6	1068.7	31.1	150.9
S, Tg S yr^{-1}				
Industrial	2.1	95.6		
Biomass burning	1.2	1.9		
Aircraft *	0	0.06		
Ships	0.07	11.4		
Total	3.4	108.96	0.2	22.0
NH_3, Tg N yr^{-1}				
Agricultural	4.8	30.1		
Biomass burning	5.0	9.2		
Other	0.7	0.7		
Total	10.5	40.0	0.5	6.2
OC, Tg C yr^{-1}				
Fossil fuel combustion	4.6	12.6		
Biomass burning	18.0	23.2		
Ships	0.001	0.1		
Total	22.6	35.9	1.2	4.1
BC, Tg C yr^{-1}				
Fossil fuel combustion	1.1	5.0		
Biomass burning	2.0	2.6		
Ships	0.001	0.1		
Total	3.1	7.7	0.2	1.6

Note that 97% of sulfur emission is assumed to be emitted as SO_2 and the rest as SO_4^{2-}. The domain of eastern China is 18–45°N and 95–125°E.

* Aircraft emission is the default emission in the model.

2.3 Calculation of anthropogenic forcing by tropospheric O_3 and aerosols

Concentrations of tropospheric O_3 and aerosols in 1850 and 2000 are simulated online based on the AR5 emissions for these two years. Both simulations are initialized using present-day climate and integrated for 2 years, with the first year as the model's spin-up. Simulated O_3 and aerosols are not allowed to influence GCM climate, so that the concentrations changes between 1850 and 2000 are the effect of changing emissions alone. Monthly mean concentrations of O_3 and aerosols are saved and then used

for forcing calculations following the approach in Liao and Seinfeld (2005). We compute the direct radiative forcing of a species as the difference in the net flux with and without the species, and then calculate anthropogenic forcing in the present-day by comparison with the preindustrial scenario. In the calculation of forcing by all aerosols, BC aerosol is assumed to be internally mixed with other aerosol species.

3 Model results

Figure 1 shows the geographical distributions of column burdens of aerosols and tropospheric ozone as well as their radiative forcings at the top of the atmosphere. The annual mean global burdens and radiative forcing values of each species are given in Table 2. In 1850, sulfate is the dominant aerosol species with an annual mean burden of 0.79 Tg as a result of the natural DMS emissions over the oceans, followed by POA (0.49 Tg), nitrate (0.35 Tg), SOA (0.34 Tg), and BC (0.05 Tg). The high concentrations of POA in the tropics result from intense biomass burning (Ito and Penner, 2005). SOA has a similar distribution to POA, because the semivolatile gases condense on the preexisting POA particles (Liao et al., 2007). SOA contributes 40% of the total OC burden (POA+SOA) in 1850. Relatively high nitrate concentrations are predicted in the high latitudes in the Northern Hemisphere, because the relatively low temperatures favor nitrate formation.

In the year 2000, burdens of all aerosol species have increased compared to 1850, with the largest changes over populated areas. Sulfate is still the most abundant aerosol species, with a global burden of 1.90 Tg, which is 2.4 times the burden in 1850. Nitrate is the second most abundant aerosol and has a global burden of 0.90 Tg. The POA burden increases to 0.71 Tg, with the highest col-

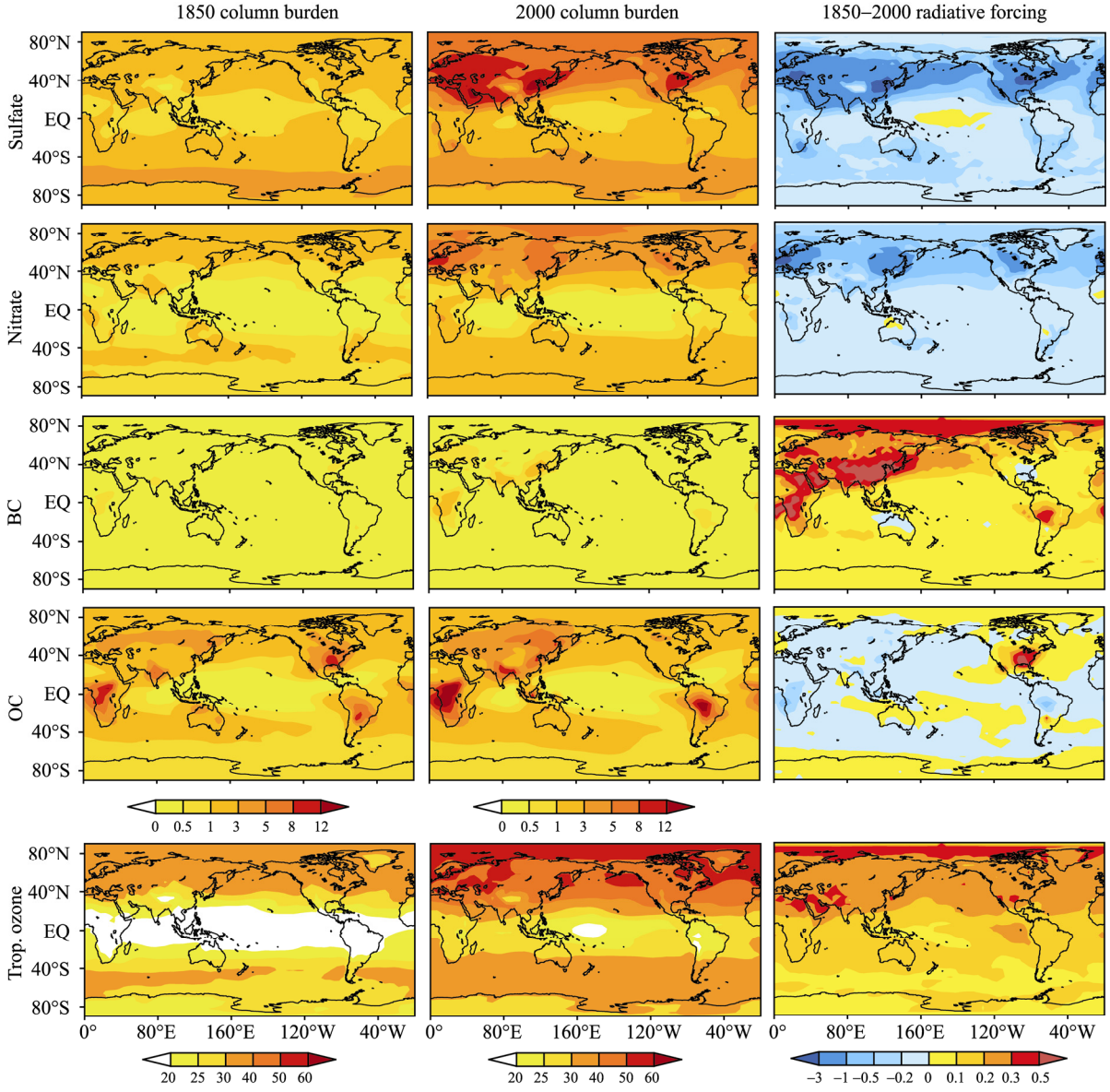

Figure 1 Predicted annual mean burdens of aerosols and tropospheric ozone in 1850 and 2000 (mg m^{-2} for aerosols and DU for ozone) and the estimated corresponding radiative forcings at the top of the atmosphere (W m^{-2}).

Table 2 Predicted annual mean global burdens (Tg yr^{-1}) of aerosols and ozone in 1850 and 2000, as well as the estimated radiative forcings (RF, W m^{-2}) at the top of atmosphere (TOA) and the surface (SRF). Ozone forcing is also given at the tropopause*. Forcing values are averaged over the globe or over eastern China (18–45°N and 95–125°E).

		1850	2000	RF in global		RF in eastern China	
				TOA	SRF	TOA	SRF
SO_4^{2-}		0.79	1.90	–0.56	–0.56	–2.50	–2.55
NO_3^-		0.35	0.90	–0.26	–0.26	–0.75	–0.77
BC		0.05	0.11	0.13	–0.25	0.58	–1.50
OC	POA	0.49	0.71	–0.02	–0.05	–0.13	–0.25
	SOA	0.34	0.32				
All aerosol				–0.60	–1.20	–2.40	–5.40
Tropospheric O_3		269	377	0.17 0.39*	0.05	0.24 0.53*	0.09

umn burdens located over the industrialized areas and biomass burning regions where emissions are high (Ito and Penner, 2005). The burden of BC is doubled relative to 1850 and is 0.11 Tg in 2000. The incomplete combustions in eastern Asia and Europe are the major sources of the predicted BC. The SOA burden has the smallest change among all aerosols. As compared to the estimates by Liao and Seinfeld (2005) with the same model but the GEIA (Global Emissions Inventory Activity) emissions inventories, burdens of BC and POA based on IPCC AR5 are much lower in 2000. The small change in POA burden over 1850–2000 lead to the small change in SOA formation from the preindustrial period to present-day. Tropospheric ozone is predicted to have increased 1.4 times from 1850 to 2000, reaching 377 Tg in the present-day. In populated areas, O_3 increases significantly as a result of the increases in anthropogenic NO_x emissions.

At the top of the atmosphere, sulfate aerosol leads to a global and annual mean cooling of –0.56 W m^{-2}, which is about two times the cooling by nitrate (Table 2). POA, BC, and O_3 lead to annual and global mean forcings of –0.02 W m^{-2}, 0.13 W m^{-2}, and 0.17 W m^{-2} respectively. Figure 2 shows the simulated geographical distributions of the seasonal and annual mean instantaneous anthropogenic direct radiative forcing at the TOA and the surface by all aerosol species. Over the populated and biomass burning areas, the annual mean maximum cooling is predicted to exceed –6 W m^{-2} at the TOA and be up to –13 W m^{-2} at the surface (Fig. 2). Aerosol cooling is stronger at the surface than at the TOA, since both the scattering and the absorption by aerosols reduce solar fluxes that can reach the surface. The global mean for either TOA or surface cooling by all aerosols is stronger in JJA than in DJF because strong photochemical reactions in JJA in the Northern Hemisphere can lead to more sulfate formation. Note that positive TOA forcings exist over places with high surface albedo, such as the Sahara Desert, the Tibetan Plateau, and the North Pole, because the internally mixed aerosols with black carbon absorb both the incoming and the reflected solar radiation.

It is of interest to compare global mean forcing values with anthropogenic radiative forcings over eastern China (18–45°N and 95–125°E). Accounting for all aerosol species, the TOA radiative forcing is –0.60 W m^{-2} on a global

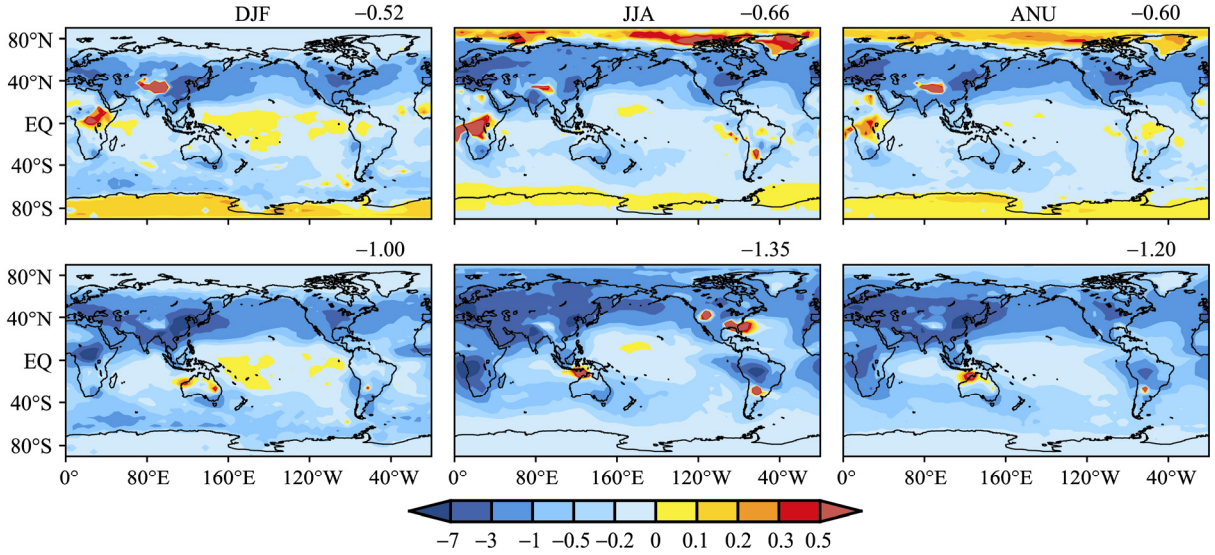

Figure 2 Simulated anthropogenic radiative forcing of all aerosols in winter (December-January-February, DJF), summer (June-July-August, JJA), and the annual mean (ANU) at the top of atmosphere (the above line) and the surface (the below line), because of the changes in concentrations between 1850 and 2000. The global mean value is marked at the top right corner of each panel (W m^{-2}).

mean basis, while it is estimated to be –2.4 W m^{-2} over eastern China. The TOA O_3 forcing is 0.24 W m^{-2} over eastern China, which is higher than the global mean value of 0.17 W m^{-2}. The seasonal variation of aerosol forcing over eastern China is different from that of the global mean forcing; the forcing averaged over eastern China is –2.90 W m^{-2} in DJF and –2.11 W m^{-2} in JJA at the TOA, and is –6.67 W m^{-2} in DJF and –4.00 W m^{-2} in JJA at the surface. The weaker cooling in China in summer than in winter is caused by the abundant precipitation associated with the East Asian Monsoon that cleans out aerosols in the atmosphere.

Our estimated forcing values by aerosols and ozone are compared with previous studies in Table 3. The annual mean aerosol TOA forcing of –0.60 W m^{-2} obtained in this work is close to the estimate of –0.5±0.4 W m^{-2} in the IPCC AR4. Compared to the negligible forcing by the internally mixed aerosols in Liao and Seinfeld (2005), the stronger cooling in this study is a result of the reduced BC emissions while the SO_2 and NO_x emissions increased in the new IPCC AR5 emissions inventories. The tropospheric ozone-induced forcing is 0.17 W m^{-2} at TOA, which is lower than the 0.22 W m^{-2} estimated by Liao and Seinfeld (2005). If we look at ozone forcing at the tropopause, it is estimated to be 0.39 W m^{-2} in this work, close to the 0.35 W m^{-2} in IPCC AR4. Among the previous estimates of radiative forcing over China, the discrepancies in emissions, the consideration of aerosol species, and the model domain make a direct comparison among the studies inappropriate. However, a crude comparison can be made. Our predicted TOA aerosol cooling in eastern China is –2.40 W m^{-2}, which is within the range of previous estimates with multiple aerosol species. TOA ozone forcing from this study is 0.24 W m^{-2} in eastern China, which is about one third of the prediction by Wang et al. (2005) for East Asia. Based on the estimates in our work, the TOA warming by ozone can offset about 10% of the TOA aerosol cooling in eastern China.

4 Summary

We use the new IPCC AR5 emissions inventories to calculate anthropogenic direct radiative forcing of tropospheric ozone and aerosols between 1850 and 2000. Compared to the previously commonly used GEIA emissions inventories, IPCC AR5 emissions have lower anthropogenic emissions of carbonaceous aerosols but higher sulfur and NO_x emissions in year 2000.

Burdens of ozone and aerosols in the years 1850 and 2000 are simulated using a unified chemistry-aerosol general circulation model. The global and annual mean burdens for sulfate, nitrate, BC, POA, and SOA are predicted to be 0.79, 0.35, 0.05, 0.49, 0.34, and 269 Tg, respectively, in the year 1850. As anthropogenic emissions increase in the year 2000, burdens of sulfate, nitrate, and BC increase to 1.90, 0.90, and 0.11 Tg, respectively. The industrialized regions of eastern Asian and Europe are the main contributors to the increased burdens of sulfate and nitrate. The POA burden increases to 0.71 Tg with the

Table 3 A comparison of the estimates of direct radiative forcing at the TOA or tropopause* (W m^{-2}).

	Model	Aerosol/O_3	China	Global
Aerosol				
Wang and Shi, 2001	Euler Transport Model	Sulfate	–0.7	
Wang et al., 2002	RegCM2, offline		–0.6*~ –1.5*	
Girogi et al., 2002	RegCM, online		–1~ –15	
Wang et al., 2003	RegCM2, offline		–4~ –10	
Gu et al., 2006	UCLA/AGCM, offline		–16	
Giorgi et al., 2002	RegCM, online	Black carbon	0.5~2	
Wu et al., 2004	RIEMS, online		4	
Gu et al., 2006	UCLA/AGCM, offline		18.5	
Wu et al., 2008	RegCM3, online		1~1.5	
Zhou et al., 1998	CRCM, offline	AOD data	–5.3~ –13	
Luo et al., 2006	RegCM2, offline	AOD data	–4.1	
Liao and Seinfeld, 2005	The unified model, online	Sulfate, Nitrate, BC, POA, SOA		0
IPCC AR4				–0.5±0.4
This study	The unified model, online	Sulfate, Nitrate, BC, POA, SOA	–2.40	–0.6
Tropospheric ozone				
Wang et al., 2005	RegCM2, online		0.65~0.73	
Mickley et al., 2004				0.49*
Liao and Seinfeld, 2005	The unified model, online			0.22
IPCC AR4				0.35*
This study	The unified model, online		0.24 0.53*	0.17 0.39*

maximum column burdens located over biomass burning regions. SOA has a slight decrease to 0.32 Tg. The global burden of tropospheric ozone in 2000 is simulated to be 377 Tg.

On a global mean basis, the direct radiative forcing of all aerosols is –0.60 W m^{-2} at the TOA and –1.20 W m^{-2} at the surface, and that of tropospheric ozone is 0.17 W m^{-2} at the TOA and 0.05 W m^{-2} at the surface. Forcing values over eastern China are much higher than the global mean values; aerosol forcing over eastern China is –2.40 W m^{-2} at the TOA and –5.40 W m^{-2} at the surface. Tropospheric ozone forcing at TOA is 0.24 W m^{-2} over eastern China, which can offset about 10% of TOA aerosol cooling averaged over the same region. Forcing values indicate that the climatic effect of aerosols over eastern China is much more significant than the globally averaged effect of aerosols.

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