

## Response to Topical Editor (Dr. Fiona O'Connor)

### Comments:

"... Having read your response and your revised manuscript, and after consulting with the executive editors of GMD, I am writing to confirm that I can only recommend publication in GMD once you have adhered to GMD's strict policy on model code availability. ...."

### Response:

Following the GMD's policy, we have modified the section of "6. Code and data availability" as: "The source codes of BCC-ESM1, model input files, and scripts to reproduce the simulations that are presented in the article have been archived and made publicly available for downloading from <http://doi.org/10.5281/zenodo.3609337> (Wu et al., 2020). Model output of BCC CMIP6 AerChemMIP simulations described in this paper refer to Zhang et al. (2019) and is distributed through ESGF and freely accessible through the ESGF data portals after registration. *Details about ESGF are presented on the CMIP Panel website at <http://www.wcrp-climate.org/index.php/wgcm-cmip/about-cmip>.*"

Tongwen Wu, and Co-authors.

1 **Beijing Climate Center Earth System Model version 1 (BCC-ESM1):**

2 **Model Description and Evaluation of Aerosol Simulations**

3  
4 **Tongwen Wu<sup>1\*</sup>, Fang Zhang<sup>1</sup>, Jie Zhang<sup>1</sup>, Weihua Jie<sup>1</sup>, Yanwu Zhang<sup>1</sup>, Fanghua Wu<sup>1</sup>,**  
5 **Laurent Li<sup>1,2</sup>, Jinghui Yan<sup>1</sup>, Xiaohong Liu<sup>3</sup>, Xiao Lu<sup>4</sup>, Haiyue Tan<sup>4</sup>, Lin Zhang<sup>4</sup>,**  
6 **Jun Wang<sup>5</sup>, Aixue Hu<sup>6</sup>**

7  
8 <sup>1</sup>Beijing Climate Center, China Meteorological Administration, Beijing, China

9 <sup>2</sup>Laboratoire de M é t é o r o l o g i e D y n a m i q u e, I P S L, C N R S, S o r b o n n e U n i v e r s i t é E c o l e N o r m a l e  
10 S u p é r i e u r e, E c o l e P o l y t e c h n i q u e, P a r i s, F r a n c e

11 <sup>3</sup>Texas A&M University, College Station, TX, USA

12 <sup>4</sup>Laboratory for Climate and Ocean-Atmosphere Studies, Department of Atmospheric and  
13 Oceanic Sciences, School of Physics, Peking University, Beijing, China

14 <sup>5</sup>University of Iowa, Iowa City, IA 52242, USA

15 <sup>6</sup>National Center for Atmospheric Research, PO Box 3000, Boulder, Colorado 80307-3000,  
16 USA

17  
18  
19  
20 *Correspondence to:* Tongwen Wu ([twwu@cma.gov.cn](mailto:twwu@cma.gov.cn))

21  
22 Submit to Geosci. Model Dev.

23  
24 **Revised on Oct. 23, 2019**

25 **Revised on Dec. 14, 2019**

26 **Revised on Jan. 17, 2020**

28 **Abstract.** BCC-ESM1 is the first version of a fully-coupled Earth System Model with  
29 interactive atmospheric chemistry and aerosols developed by the Beijing Climate Center,  
30 China Meteorological Administration. Major aerosol species (including sulfate, organic  
31 carbon, black carbon, dust and sea salt) and greenhouse gases are interactively simulated with  
32 a whole panoply of processes controlling emission, transport, gas-phase chemical reactions,  
33 secondary aerosol formation, gravitational settling, dry deposition, and wet scavenging by  
34 clouds and precipitation. Effects of aerosols on radiation, cloud, and precipitation are fully  
35 treated. The performance of BCC-ESM1 in simulating aerosols and their optical properties is  
36 comprehensively evaluated as required by the Aerosol Chemistry Model Intercomparison  
37 Project (AerChemMIP), covering the preindustrial mean state and time evolution from 1850  
38 to 2014. The simulated aerosols from BCC-ESM1 are quite coherent with  
39 CMIP5-recommended data, in-situ measurements from surface networks (such as IMPROVE  
40 in the U.S. and EMEP in Europe), and aircraft observations. A comparison of modeled  
41 aerosol optical depth (AOD) at 550 nm with satellite observations retrieved from Moderate  
42 Resolution Imaging Spectroradiometer (MODIS) and Multi-angle Imaging  
43 SpectroRadiometer (MISR) and surface AOD observations from AErosol RObotic NETwork  
44 (AERONET) shows reasonable agreements between simulated and observed AOD. However,  
45 BCC-ESM1 shows weaker upward transport of aerosols from the surface to the middle and  
46 upper troposphere, likely reflecting the deficiency of representing deep convective transport  
47 of chemical species in BCC-ESM1. With an overall good agreement between BCC-ESM1  
48 simulated and observed aerosol properties, it demonstrates a success of the implementation of  
49 interactive aerosol and atmospheric chemistry in BCC-ESM1.

50

51 **1. Introduction**

52 Atmosphere is a thin gaseous layer around the Earth, consisting of nitrogen, oxygen and  
53 a large number of trace gases including important greenhouse gases (GHG) such as water  
54 vapor, tropospheric ozone (O<sub>3</sub>), carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O),  
55 and chloro-fluoro-carbons (CFCs). Besides gaseous components, atmosphere also contains  
56 various aerosols, which are important for cloud formation and radiative transfer. Atmospheric  
57 trace gases and aerosols are actually interactive components of the climate system. Their  
58 inclusion in global climate models (GCMs) is a significant enhancement for most  
59 state-of-the-art climate models (Lamarque et al., 2013; Collins et al., 2017). Early attempts in  
60 coupling global climate dynamics with atmospheric chemistry can be traced back to late  
61 1970s, when 3D transport of ozone and simple stratospheric chemistry were firstly  
62 incorporated into a GCM to simulate global O<sub>3</sub> production and transport (e.g., Cunnold et al.  
63 1975; Schlesinger and Mintz 1979). Since mid-1980s, a large number of on-line global  
64 climate/chemistry models have been developed to address issues of the Antarctic stratospheric  
65 O<sub>3</sub> depletion (e.g., Cariolle et al. 1990; Austin et al. 1992; Solomon, 1999), tropospheric O<sub>3</sub>  
66 and sulfur cycle (e.g., Feichter et al. 1996; Barth et al. 2000), tropospheric aerosol and its  
67 interactions with cloud (e.g., Chuang et al. 1997; Lohmann et al. 2000; Ghan and Easter, 2006;  
68 Jacobson 2012). Aerosols and chemically reactive gases in the atmosphere exert important  
69 influences on global and regional air quality and climate (Collins et al., 2017).

70 Since 2013, the Beijing Climate Center (BCC), China Meteorological Administration,  
71 has continuously developed and updated its fully-coupled GCM, the Beijing Climate Center  
72 Climate System Model (BCC-CSM) (Wu et al., 2013; Wu et al., 2014; Wu et al., 2019).  
73 BCC-CSM version 1.1 was one of the comprehensive carbon-climate models participating in  
74 the phase five of the Coupled Model Intercomparison Project (CMIP5, Taylor et al. 2012).  
75 When forced by prescribed historical emissions of CO<sub>2</sub> from combustion of fossil fuels and  
76 land use change, BCC-CSM1.1 successfully reproduced the trends of observed atmospheric  
77 CO<sub>2</sub> concentration and global surface air temperature from 1850 to 2005 (Wu et al., 2013).  
78 During recent years, BCC-CSM1.1 has been used in numerous investigations on soil organic  
79 carbon changes (e.g. Todd-Brown et al., 2014), ocean biogeochemistry changes (e.g. Mora et  
80 al., 2013), and carbon-climate feedbacks (e.g. Arora et al., 2013; Hoffman et al., 2014).

81 BCC-CSM includes main climate-carbon cycle processes (Wu et al., 2013) and the global  
82 mean atmospheric CO<sub>2</sub> concentration is calculated from a prognostic equation of CO<sub>2</sub> budget  
83 taking into account global anthropogenic CO<sub>2</sub> emissions and interactive land-atmosphere and  
84 ocean-atmosphere CO<sub>2</sub> exchanges.

85 In recent years, BCC has put large efforts in developing a global  
86 climate-chemistry-aerosol fully-coupled Earth System Model (BCC-ESM1) on the basis of  
87 BCC-CSM2 (Wu et al., 2019). The objective is to interactively simulate global aerosols (e.g.  
88 sulfate, black carbon, etc.) and main greenhouse gases (e.g. O<sub>3</sub>, CH<sub>4</sub>, N<sub>2</sub>O and CO<sub>2</sub>) in the  
89 atmosphere and to investigate feedbacks between climate and atmospheric chemistry.  
90 BCC-ESM1 is at the point to be publicly released, and it is actively used by BCC for several  
91 CMIP6-endorsed research initiatives (Eyring et al. 2016), including the Aerosol Chemistry  
92 Model Intercomparison Project (AerChemMIP, Collins et al., 2017) and the Coupled  
93 Climate–Carbon Cycle Model Intercomparison Project (C4MIP, Jones et al. 2016).

94 The purpose of this paper is to evaluate the performance of BCC-ESM1 in simulating  
95 aerosols and their optical properties in the 20th century. The description of BCC-ESM1 is  
96 presented in Section 2. The experimental protocol is given in Section 3. Section 4 presents the  
97 evaluations of aerosol simulations with comparisons to CMIP5-recommended data (Lamarque  
98 et al., 2010) and data obtained from both global surface networks and satellite observations.  
99 The regional and global characteristics compared to observations and estimates from other  
100 studies are analyzed. Simulations of aerosol optical properties in the 20th century are also  
101 analyzed in Section 4. Conclusions and discussions are summarized in Section 5. Information  
102 about code and data availability is given in Section 6.

## 103 **2. Model description**

104 BCC-ESM1 is an Earth System Model with interactive chemistry and aerosol  
105 components, in which the atmospheric component is BCC Atmospheric General Model  
106 version 3 (Wu et al., 2019) with interactive atmospheric chemistry (hereafter  
107 BCC-AGCM3-Chem), land component BCC Atmosphere and Vegetation Interaction Model  
108 version 2.0 (hereafter BCC-AVIM2.0), ocean component Modular Ocean Model version 4  
109 (MOM4)-L40, and sea ice component [sea ice simulator (SIS)]. Different components of  
110 BCC-ESM1 are fully coupled and interact with each other through fluxes of momentum,

111 energy, water, carbon and other tracers at their interfaces. The coupling between the  
112 atmosphere and the ocean is done every hour.

113 The atmospheric component BCC-AGCM3-Chem is able to simulate global atmospheric  
114 composition and aerosols from anthropogenic emissions as forcing agents. Its resolution is T42  
115 (approximately  $2.8125 \times 2.8125^\circ$  transformed spectral grid). The model has 26 levels in a hybrid  
116 sigma/pressure vertical coordinate system with the top level at 2.914 hPa. Details of the model  
117 physics are described in Wu et al. (2019). The BCC-AGCM3-Chem combines 66 gas-phase  
118 chemical species and 13 bulk aerosol compounds as listed in Table 1. Apart from 3 gas-phase  
119 species of dimethyl sulfide (DMS), sulfur dioxide (SO<sub>2</sub>) and ammonia (NH<sub>3</sub>), the other 63  
120 gas-phase species are the same as those in the “standard version” of MOZART2 (Model for  
121 Ozone and Related chemical Tracers, version 2), a global chemical transport model for the  
122 troposphere developed by the National Center for Atmospheric Research (NCAR) driven by  
123 meteorological fields from either climate models or assimilations of meteorological  
124 observations (Horowitz et al., 2003). Advection of all tracers in BCC-AGCM3-Chem is  
125 performed through a semi-Lagrangian scheme (Williamson and Rasch, 1989), and vertical  
126 diffusion within the boundary layer follows the parameterization of Holtslag and Boville  
127 (1993). The gas-phase chemistry of the 63 MOZART2 gas-phase species as listed in Table 1  
128 is treated in the same way as that in the “standard version” of MOZART2 (Horowitz et al.,  
129 2003), and there are 33 photolytic reactions and 135 chemical reactions involving 30 dry  
130 deposited chemical species and 25 soluble gas-phase species. Dry deposition velocities for the  
131 15 trace gases including O<sub>3</sub>, carbon monoxide (CO), CH<sub>4</sub>, formaldehyde (CH<sub>2</sub>O), acetic acid  
132 (CH<sub>3</sub>OOH), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), nitrogen dioxide (NO<sub>2</sub>), nitric acid (HNO<sub>3</sub>),  
133 polyacrylonitrile (PAN), acetone (CH<sub>3</sub>COCH<sub>3</sub>), peroxyacetic acid (CH<sub>3</sub>COOOH),  
134 acetaldehyde (CH<sub>3</sub>CHO), methylglyoxal (CH<sub>3</sub>COCHO), nitric oxid (NO), and pernitric acid  
135 (HNO<sub>4</sub>) are not computed interactively and directly interpolated from MOZART2  
136 climatological monthly mean deposition velocities  
137 ([https://en.wikipedia.org/wiki/MOZART\(model\)](https://en.wikipedia.org/wiki/MOZART(model))) which are calculated offline (Bey et al., 2001;  
138 Shindell et al., 2008) using a resistance-in-series scheme originally described in Wesely  
139 (1989). The dry deposition velocities for the other 15 species including peroxy acetyl nitrate  
140 (PAN), methyl nitroacetate (ONIT), organic nitrates (ONITR), ethyl alcohol (C<sub>2</sub>H<sub>5</sub>OH), organic

141 hydroxiperoxide (POOH), ethyl hydroperoxide ( $C_2H_5OOH$ ), propylhydroperoxide  
142 ( $C_3H_7OOH$ ), methylene glycol mono acetate (ROOH), glycolaldehyde (GLYALD), acetol  
143 (HYAC), methanol ( $CH_3OH$ ), propanoic acid (MACROOH), isoprene hydroxy hydroperoxide  
144 (ISOPOOH), carboxylic acid (XOOH), formaldehyde (HYDRALD), and hydrogen ( $H_2$ ) are  
145 calculated using prescribed deposition velocities of  $O_3$ , CO,  $CH_3CHO$ , or land surface type  
146 and surface temperature following the MOZART2 (Horowitz et al., 2003). Wet removal by  
147 in-cloud scavenging for 25 soluble gas-phase species in the “standard version” of MOZART2  
148 uses the parameterization of Giorgi and Chameides (1985) based on their temperature  
149 dependent effective Henry’s law constants. In-cloud scavenging is proportional to the amount  
150 of cloud condensate converted to precipitation, and the loss rate depends on the amount of  
151 cloud water, the rate of precipitation formation, and the rate of tracer uptake by the liquid  
152 phase water. Other highly soluble species such as  $HNO_3$ ,  $H_2O_2$ , ONIT, ISOPOOH,  
153 MACROOH, XOOH, and lead (Pb-210) are also removed by below-cloud washout as  
154 calculated using the formulation of Brasseur et al. (1998). Below-cloud scavenging is  
155 proportional to the precipitation flux in each model layer and the loss rate depends on the  
156 precipitation rate. Vertical transport of gas tracers and aerosols due to deep convection is not  
157 yet included in the present version of BCC-AGCM3-Chem, which process is considered as a  
158 part of the deep convection and occurs generally in a small spatial region on a GCM-box with  
159 low-resolution ( $2.8^\circ lat. \times 2.8^\circ lon.$ ). Another consideration is that a large uncertainty exists to  
160 treat transport of those water-soluble tracers by deep convection. But this effect will be  
161 involved in the next version of BCC model.

162 The BCC-AVIM2.0 is the land model with terrestrial carbon cycle. It is described in  
163 details in Li et al. (2019) and includes biophysical, physiological, and soil carbon-nitrogen  
164 dynamical processes. The terrestrial carbon cycle operates through a series of biochemical  
165 and physiological processes on photosynthesis and respiration of vegetation. Biogenic  
166 emissions from vegetation are computed online in BCC-AVIM2.0 following the algorithm of  
167 the Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1,  
168 Guenther et al., 2012).

169 The oceanic component of BCC-ESM1 is the Modular Ocean Model version 4 with 40  
170 levels (hereafter MOM4-L40), and the sea ice component Sea Ice Simulator (SIS).

171 MOM4-L40 uses a tripolar grid of horizontal resolution with 1 °longitude by 1/3 °latitude  
172 between 30 S and 30 N ranged to 1 °longitude by 1 °latitude from 60 S and 60 N poleward  
173 and 40 z-levels in the vertical. Carbon exchange between the atmosphere and the ocean are  
174 calculated online in MOM4-L40 using a biogeochemistry module that is based on the  
175 protocols from the Ocean Carbon Cycle Model Intercomparison Project–Phase 2 (OCMIP2,  
176 <http://www.ipsl.jussieu.fr/OCMIP/phase2/>). SIS has the same horizontal resolution as  
177 MOM4-L40 and three layers in the vertical, including one layer of snow cover and two layers  
178 of equally sized sea ice. Details of oceanic component MOM4-L40 and sea-ice component  
179 SIS that are used in BCC-ESM1 may be found in Wu et al. (2013) and Wu et al. (2019).

180 In the following sub-sections, we will describe the treatments in BCC-ESM1 for 3  
181 gas-phase species of DMS, SO<sub>2</sub> and NH<sub>3</sub>, 13 prognostic aerosol species including sulfate  
182 (SO<sub>4</sub><sup>2-</sup>), 2 types of organic carbon (hydrophobic OC1, hydrophilic OC2), 2 types of black  
183 carbon (hydrophobic BC1, hydrophilic BC2), 4 categories of soil dust (DST01, DST02,  
184 DST03, DST04), and 4 categories of sea salt (SSLT01, SSLT02, SSLT03, SSLT04).  
185 Concentrations of all aerosols in BCC-ESM1 are mainly determined by advective transport,  
186 emission, dry deposition, gravitational settling, and wet scavenging by clouds and  
187 precipitation, except for SO<sub>4</sub><sup>2-</sup> which gas-phase and aqueous phase conversion from SO<sub>2</sub> are  
188 also considered. The present version of aerosol scheme belongs to a bulk aerosol model and  
189 mainly refers to the scheme of CAM-Chem (Lamarque et al., 2012), but the nucleation and  
190 coagulation of aerosols are still ignored.

## 191 **2.1 SO<sub>2</sub>, DMS, NH<sub>3</sub>, and Sulfate**

192 SO<sub>2</sub> is a main sulfuric acid precursor to form aerosol sulfate SO<sub>4</sub><sup>2-</sup>. Conversions of SO<sub>2</sub>  
193 to SO<sub>4</sub><sup>2-</sup> occur by gas phase reactions (Table 2) and by aqueous phase reactions in cloud  
194 droplets. The dry deposition velocity of SO<sub>2</sub> follows the resistance-in-series approach of  
195 | Wesely (1989) using the formula,  $W_{\text{SO}_2} = 1/(r_a + r_b + r_c)$ , in which  $r_a$ ,  $r_b$ , and  $r_c$  are the  
196 aerodynamic resistance, the quasi-laminar boundary layer resistance, and the surface  
197 resistance, respectively and they are interactively computed in each model time step. The loss  
198 rate of SO<sub>2</sub> due to wet deposition is computed following the scheme in the global Community  
199 Atmosphere Model (CAM) version 4, the atmospheric component of the Community Earth  
200 System Model (Lamarque et al., 2012).



201 The sources of SO<sub>2</sub> mainly come from fuel combustion, industrial activities, and  
202 volcanoes. SO<sub>2</sub> can also be formed from the oxidation of DMS as listed in Table 2 in which  
203 their reaction rates follow CAM-Chem (Lamarque et al. 2012). The main source of DMS is  
204 from oceanic emissions via biogenic processes. It is prescribed with the climatological  
205 monthly data that are extracted from MOZART2 package  
206 (<https://www2.acom.ucar.edu/gcm/mozart-4>). SO<sub>4</sub><sup>2-</sup> is one of the prognostic aerosols in  
207 BCC-AGCM3-Chem. Its treatment follows CAM4-Chem (Lamarque et al., 2012). It is  
208 produced primarily by the gas-phase oxidation of SO<sub>2</sub> (in Table 2) and by aqueous phase  
209 oxidation of SO<sub>2</sub> in cloud droplets. The gas phase reactions, rate constants, and gas-aqueous  
210 equilibrium constants are given by Tie et al. (2001). The heterogeneous reactions of SO<sub>4</sub><sup>2-</sup>  
211 occur on all aerosol surfaces. Their treatment follows a Bulk Aerosol Model (BAM) used in  
212 CAM4 (Neale et al., 2010). The heterogeneous reactions depend strongly on pH values in  
213 clouds which are calculated from the concentrations of SO<sub>2</sub>, HNO<sub>3</sub>, H<sub>2</sub>O<sub>2</sub>, NH<sub>3</sub>, O<sub>3</sub>, HO<sub>2</sub>, and  
214 SO<sub>4</sub><sup>2-</sup>. NH<sub>3</sub> is a gas tracer apart from MOZART2 (Table 1). Its sources include aircraft and  
215 surface emissions due to anthropogenic activity, biomass burning, and biogenic emissions  
216 from land soil and ocean surfaces (Table 4). SO<sub>4</sub><sup>2-</sup> is assumed to be all in aqueous phase due  
217 to water uptake, although Wang et al. (2008a) showed that ~34% of sulfate particles are in  
218 solid phase globally due to the hysteresis effect of ammonium sulfate phase transition.  
219 However, in terms of radiative forcing, consideration of solid sulfate formation process  
220 lowers the sulfate forcing by ~8% as compared to consideration of all sulfate particles in  
221 aqueous phase (Wang et al., 2008b). Future model development may consider the life cycle of  
222 NH<sub>3</sub>. The sulfate in- and below-cloud scavenging follows Neu and Prather (2011). Washout  
223 of SO<sub>4</sub><sup>2-</sup> is set to 20% of the washout rate of HNO<sub>3</sub> following Tie et al. (2005) and Horowitz  
224 (2006). Dry deposition velocity of SO<sub>4</sub><sup>2-</sup> is also calculated by the resistance-in-series  
225 approach.

## 226 **2.2 Aerosols of organic carbon and black carbon**

227 BCC-AGCM3-Chem treats two types of organic carbon (OC), i.e. water-insoluble tracer  
228 OC1 and water-soluble tracer OC2, and two types of black carbon (BC), i.e. water-insoluble  
229 tracer BC1 and water-soluble tracer BC2. As shown in Table 2, hydrophobic BC1 and OC1  
230 can be converted to hydrophilic BC2 and OC2 with a constant rate of  $7.1 \times 10^{-6} \text{ s}^{-1}$  (Cooke and

231 Wilson, 1996). The 4 tracers of organic carbon and black carbon are mainly from emissions  
232 including both fossil fuel and biomass burning, and are from the CMIP6 data package  
233 (<https://esgf-node.llnl.gov/search/input4mips/>, Hoesly et al., 2018). Beside anthropogenic and  
234 biomass burning emissions, hydrophilic organic carbon OC2 can also come from natural  
235 biogenic volatile organic compound (VOC) emissions. Dry deposition velocities for all the 4  
236 OC and BC tracers are set to  $0.001\text{m}\cdot\text{s}^{-1}$ . OC2 and BC2 are soluble aerosols, and their sinks  
237 are primarily governed by wet deposition. Their in- and below-cloud scavenging follows the  
238 scheme of Neu and Prather (2011).

### 239 **2.3 Sea salt aerosols**

240 As shown in Table 3, sea salt aerosols in the model are classified into four size bins (0.2–  
241 1.0, 1.0–3.0, 3.0–10, and 10–20  $\mu\text{m}$ ) in diameter. They originate from oceans and are  
242 calculated online by BCC-ESM1. The upward flux  $F_{sea-salt}$  of sea salt productions for four  
243 bins is proportional to the 3.41 power of the wind speed  $u_{10m}$  at 10 m height near the sea  
244 surface (Mahowald et al., 2006) and is expressed as

$$245 \quad F_{sea-salt} = S \cdot (u_{10m})^{3.41}, \quad (1)$$

246 where  $S$  is a scaling factor and set to  $4.05 \times 10^{-15}$ ,  $4.52 \times 10^{-14}$ ,  $1.15 \times 10^{-13}$ ,  $1.20 \times 10^{-13}$  for four  
247 size bins of sea salt aerosols in BCC-ESM1, respectively.

248 Dry deposition of sea salts depends on the turbulent deposition velocity in the lowest  
249 atmospheric layer using aerodynamic resistance and the friction velocity, and the settling  
250 velocity through the whole atmospheric column for each bin of sea salts. The turbulent  
251 deposition velocity and settling velocity depend on particle diameter and density (listed in  
252 Table 3). In addition, the fact that the size of sea salts changes with humidity is also  
253 considered. The wet deposition of sea salts follows the scheme for soluble aerosols used in  
254 CAM4, and depends on prescribed solubility and size-independent scavenging coefficients.

### 255 **2.4 Dust aerosols**

256 Dust aerosols behave in a similar way as sea salts. Their variations involve three major  
257 processes: emission, advective transport, and wet/dry depositions. The dust emission is based  
258 on a saltation-sandblasting process, and depends on wind friction velocity, soil moisture, and  
259 vegetation/snow cover (Zender et al., 2003). The vertical flux of dust emission is corrected by  
260 a surface erodible factor at each model grid cell which has been downloaded from NCAR

261 website (<https://svn-ccsm-inputdata.cgd.ucar.edu/trunk/inputdata/atm/cam/dst/>). Soil  
 262 erodibility is prescribed by a physically-based geomorphic index that is proportional to the  
 263 runoff area upstream of each source region (Albani et al., 2014). Like sea salts, dry deposition  
 264 of dust aerosols includes gravitational and turbulent deposition processes, while wet  
 265 deposition results from both convective and large scale precipitation and is dependent on  
 266 prescribed size-independent scavenging coefficients.

## 267 **2.5 Effects of aerosols on radiation, clouds, and precipitation**

268 The mass mixing ratios of bulk aerosols are prognostic variables in BCC-ESM1 and  
 269 directly affect the radiative transfer in the atmosphere with their treatments following the  
 270 NCAR Community Atmosphere Model (CAM3, Collins et al., 2004). Indirect effects of  
 271 aerosols are taken into account in the present version of BCC-AGCM3-Chem (Wu et al.,  
 272 2019). Aerosol particles act as cloud condensation nuclei and exert influence on cloud  
 273 properties and precipitation, and ultimately impact the hydrological cycle. Prognostic aerosol  
 274 masses are used to estimate the liquid cloud droplet number concentration  $N_{cdnc}$  ( $\text{cm}^{-3}$ ) in  
 275 BCC-AGCM3-Chem.  $N_{cdnc}$  is explicitly calculated using the empirical function suggested  
 276 by Boucher and Lohmann (1995) and Quaas et al. (2006):

$$277 \quad N_{cdnc} = \exp[5.1 + 0.41 \ln(m_{aero})] \quad (2)$$

278 where  $m_{aero}$  ( $\mu\text{g}\cdot\text{m}^{-3}$ ) is the total mass of all hydrophilic aerosols,

$$279 \quad m_{aero} = m_{SS} + m_{OC} + m_{SO_4} + m_{NH_4NO_2}, \quad (3)$$

280 i.e. the first bin of sea salt ( $m_{SS}$ ), hydrophilic organic carbon ( $m_{OC}$ ), sulphate ( $m_{SO_4}$ ), and  
 281 Ammonium nitrite ( $NH_4NO_2$ ). A dataset of  $NH_4NO_2$  from NCAR CAM-Chem (Lamarque et  
 282 al., 2012) is used in our model.

283  $N_{cdnc}$  is an important factor in determining the effective radius of cloud droplets for  
 284 radiative calculation. The effective radius of cloud droplets  $r_{el}$  is estimated as

$$285 \quad r_{el} = \beta \cdot r_{l,vol}, \quad (4)$$

286 where  $\beta$  is a parameter dependent on the droplets spectral shape and follows the calculation  
 287 proposed by Peng and Lohmann (2003),

$$288 \quad \beta = 0.00084 N_{cdnc} + 1.22. \quad (5)$$

289  $r_{l,vol}$  is the volume-weighted mean cloud droplet radius,

$$290 \quad r_{l,vol} = \left[ (3LWC) / (4\pi\rho_w N_{cdnc}) \right]^{1/3}, \quad (6)$$

291 where  $\rho_w$  is the liquid water density and  $LWC$  the cloud liquid water content ( $\text{g cm}^{-3}$ ).

292 Aerosols also exert impacts on precipitation efficiency (Albrecht, 1989), which is taken  
 293 into account in the parameterization of non-convective cloud processes. There are five  
 294 processes that convert condensate to precipitate: auto-conversion of liquid water to rain,  
 295 collection of cloud water by rain, auto-conversion of ice to snow, collection of ice by snow,  
 296 and collection of liquid by snow. The auto-conversion of cloud liquid water to rain ( $PWAUT$ )  
 297 is dependent on the cloud droplet number concentration and follows a formula that was  
 298 originally suggested by Chen and Cotton (1987),

$$299 \quad PWAUT = C_{l,aut} \hat{q}_l^2 \rho_a / \rho_w \left( \frac{q_l \rho_a}{\rho_w N_{ncdc}} \right)^{1/3} H(r_{l,vol} - r_{lc,vol}) \quad (7)$$

300 Where  $\hat{q}_l$  is in-cloud liquid water mixing ratio,  $\rho_a$  and  $\rho_w$  are the local densities of air and  
 301 water respectively, and  $C_{l,aut}$  is a constant.  $H(x)$  is the Heaviside step function with the  
 302 definition,

$$303 \quad H(x) = \begin{cases} 0, & x < 0 \\ 1, & x \geq 0 \end{cases}. \quad (8)$$

304  $r_{lc,vol}$  is the critical value of mean volume radius of liquid cloud droplets  $r_{l,vol}$ , and set to 15  
 305  $\mu\text{m}$ .

306 The treatment of aerosol single scattering (optical) properties (such as mass extinction  
 307 efficiency, single scattering albedo, and asymmetric factor) follows the look-up table  
 308 approach in CAM (Collins et al., 2004). The optics for black, organic carbon, sea salt, and sea  
 309 salt particles is assumed to be same as the optics for soot and water-soluble aerosols in the  
 310 Optical Properties of Aerosols and Clouds (OPAC) data set (Hess et al., 1998). The optics for  
 311 dust is derived by Mie calculations for the size distribution represented by each size bin  
 312 (Zender et al., 2003). Similarly, for sulfate and nitrate particles, same set of aerosol optical  
 313 properties for ammonium sulfate are used and are taken from Wang et al. (2008b) with  
 314 treatment of aerosol hygroscopicity. The volcanic stratospheric aerosols are assumed to be  
 315 comprised of 75% sulfuric acid and 25% water, as in Hess et al. (1998). For each model year,

316 different aerosol types are assumed to be externally mixed in the calculation of bulk aerosol  
317 single scattering properties that are in turn used in the radiative transfer calculations.

### 318 **3. Experiment design for the 20<sup>th</sup> century climate simulation**

319 There is an Aerosol Chemistry Model Intercomparison Project (AerChemMIP, Collins et  
320 al., 2017) endorsed by the Coupled-Model Intercomparison Project 6 (CMIP6) for  
321 documenting and understanding past and future changes in the chemical composition of the  
322 atmosphere, and estimating the global-to-regional climate response from these changes.  
323 Modelling groups with full chemistry and aerosol models are encouraged to perform all  
324 AerChemMIP simulations (Collins et al., 2017). To assess the ability of our model to simulate  
325 aerosols (mean and variability), we have followed the historical simulation designed by  
326 CMIP6 (Eyring et al., 2016) which is named as “historical” experiment in the Earth System  
327 Grid Federation (ESGF). The historical experiment is forced with emissions evolving from  
328 1850 to 2014 that include biomass burning emissions (Van Marle et al. 2017), anthropogenic  
329 and open burning emissions (Hoesly et al., 2018; Feng et al., 2019). O<sub>3</sub> in the historical  
330 simulation is an interactive prognostic variable and feedbacks on radiation, and the  
331 concentrations of other WMOGHG, e.g. CH<sub>4</sub>, N<sub>2</sub>O, CO<sub>2</sub>, CFC11, and CFC12 are prescribed  
332 using CMIP6 historical forcing data (Meinshausen et al., 2017). Although CH<sub>4</sub> and N<sub>2</sub>O are  
333 prognostic variables in the chemistry scheme (Table 1), their prognostic values at each model  
334 step in the historical experiment are replaced by CMIP6 data (Meinshausen et al., 2017)  
335 throughout the model domain. The rest of historical forcing data include: (1) yearly global  
336 gridded land-use forcing data sets (Hurtt et al., 2011; Hurtt et al., 2017), and (2) solar forcing  
337 (Matthes et al., 2017). All these datasets were downloaded from  
338 <https://esgf-node.llnl.gov/search/input4mips/>. Climate feedback processes that involve  
339 changes to the atmospheric composition of reactive gases and aerosols may affect the  
340 temperature response to a given WMOGHG concentration level.

#### 341 **3.1 Surface emissions**

342 Surface emissions of chemical species from different sources are summarized in Table  
343 4. They include anthropogenic emissions from fossil fuel burning and other industrial  
344 activities, biomass burning (including vegetation fires, fuel wood and agricultural burning),  
345 biogenic emissions from vegetation and soils, and oceanic emissions. Most historical

346 emissions from anthropogenic source (surface, aircraft plus ship) and biomass burning from  
347 1850 to 2014 are CMIP6-recommended data (available at  
348 <https://esgf-node.llnl.gov/search/input4mips>). Anthropogenic or biomass burning sources of  
349 some tracers which are not included in the CMIP6 dataset (see Table 4), anthropogenic  
350 emission of H<sub>2</sub> and N<sub>2</sub>O are from monthly climatological dataset provided by the MOZART-2  
351 standard package. N<sub>2</sub>O is a prognostic variable in BCC-ESM1 but it is replaced by CMIP6  
352 prescribed concentration in the historical run. Other emissions including biomass burning  
353 (CH<sub>3</sub>COCH<sub>3</sub>) and anthropogenic emission (CH<sub>3</sub>CHO, CH<sub>3</sub>OH, and CH<sub>3</sub>COCH<sub>3</sub>) are from the  
354 Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) emission  
355 inventory (<http://accent.aero.jussieu.fr/ACCMIP.php>) covering the period from 1850 to 2010  
356 with 10-year intervals (see Table 4). Monthly lumped emissions of black carbon and organic  
357 carbon aerosols from 1850 to 2014 are downloaded from CMIP6-recommended data, but we  
358 used 80% (for BC) and 50% (for OC) of them in their hydrophobic forms (BC1 and OC1) and  
359 the rest in their hydrophilic forms (BC2 and OC2), following the work of Chin et al. (2002).

360 Five tracers of ISOP, ACET (CH<sub>3</sub>COCH<sub>3</sub>), C<sub>2</sub>H<sub>4</sub>, C<sub>3</sub>H<sub>8</sub>, and Monoterpenes (C<sub>10</sub>H<sub>16</sub>) in  
361 Table 1 belong to biogenic volatile organic carbons (VOCs). As shown in Table 4, those  
362 VOCs emissions are online calculated in BCC-ESM1 following the modeling framework of  
363 the Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1,  
364 Guenther et al., 2012) using simple mechanistic algorithms to account for major known  
365 processes controlling biogenic emissions. The MEGAN2.1 can provide a flexible scheme for  
366 estimating 16 tracers of biogenic emissions from terrestrial ecosystems including five VOCs  
367 emissions used in BCC-ESM1 (Table 4). All the VOCs emissions depend on current and past  
368 surface air temperature, solar flux, and the landscape types. Their calculation requires global  
369 maps of plant functional type (PFT) and leaf area index (LAI) which is a prognostic variable  
370 from the land model BCC-AVIM2. The effect of atmospheric CO<sub>2</sub> concentration on isoprene  
371 emissions is included. 10% of the biogenic monoterpenes emissions as calculated online with  
372 the MEGAN2.1 algorithm in BCC-AVIM2 are converted to hydrophilic organic carbon (OC2)  
373 to account for formation of secondary organic aerosols following Chin et al. (2002) in this  
374 version of BCC-ESM1.

### 375 **3.2 Volcanic eruptions, lightning and aircraft emissions**

376 As there is no stratospheric aerosol scheme in BCC-ESM1, concentrations of sulfate  
377 aerosol at heights from 5 to 39.5 km, which volcanic origin, are directly prescribed using the  
378 CMIP6-recommended data (Thomasson et al., 2018) from 1850 to 2014. The effects of  
379 surface SO<sub>2</sub> emissions from volcanic eruption on the variation of SO<sub>2</sub> in the atmosphere and  
380 then on the variation of tropospheric SO<sub>4</sub><sup>2-</sup> concentration are considered, and the SO<sub>2</sub>  
381 emissions from 1850 to 2014 are downloaded from the IPCC ACCMIP emission inventory  
382 (<http://accent.aero.jussieu.fr/ACCMIP.php>). Aircraft emissions are provided for NO<sub>2</sub>, CO,  
383 CH<sub>4</sub>, NH<sub>3</sub>, NO, SO<sub>2</sub>, and aerosols of OC and BC (Table 1). The emissions of NO from  
384 lightning are online calculated in BCC-AGCM3-Chem following the parameterization in  
385 MOZART2, and the globally-averaged mean during the period of 1850 to 2014 is 5.19  
386 Tg(N) yr<sup>-1</sup>, which is in agreement with observations within the range of 3 to 6 Tg(N) yr<sup>-1</sup>  
387 (Martin et al., 2002). The lightning frequency depends strongly on the convective cloud top  
388 height, and the ratio of cloud-to-cloud versus cloud-to-ground lightning depends on the cold  
389 cloud thickness from the level of 0°C to the cloud top (Price and Rind, 1992).

### 390 **3.3 Upper boundary of the atmosphere**

391 As no stratospheric chemistry is included in the present version of BCC-AGCM3-Chem,  
392 it is necessary to ensure a proper distribution of chemically-active stratospheric species.  
393 Concentrations of different tracers (O<sub>3</sub>, CH<sub>4</sub>, N<sub>2</sub>O, NO, NO<sub>2</sub>, HNO<sub>3</sub>, CO, and N<sub>2</sub>O<sub>5</sub>) at the top  
394 two layers of the model are set to prescribed monthly climatological values, and  
395 concentrations from below the top two layers to the tropopause are relaxed at a relaxation  
396 time of 10-days towards the climatology. Climatological values of NO, NO<sub>2</sub>, HNO<sub>3</sub>, CO and  
397 N<sub>2</sub>O<sub>5</sub> at the top two layers are extracted from MOZART2 data package available at the  
398 Website (<https://www2.acom.ucar.edu/gcm/mozart-4>), originated from the Study of Transport  
399 and Chemical Reactions in the Stratosphere (STARS, Brasseur et al., 1997). Concentrations  
400 for the other tracers (O<sub>3</sub>, CH<sub>4</sub>, and N<sub>2</sub>O) at the top two model layers are the zonally-averaged  
401 and monthly values from 1850 to 2014 derived from the CMIP6 data package.

### 402 **3.4 The preindustrial model states**

403 The preindustrial state of BCC-ESM1 is obtained from a piControl simulation of over 600  
404 years in which all forcings including emissions data are fixed at 1850 conditions. The initial  
405 state of the piControl simulation itself is obtained through individual spin-up runs of each

406 component of BCC-ESM1 in order for the piControl simulation to run stably and fast to reach  
407 its equilibrium. Figures 1(a-c) show the time series of global yearly means of the net energy  
408 budget at top of the atmosphere (TOA), near-surface air temperature (TAS), and sea surface  
409 temperature (SST) from the piControl simulation for the last 450 years. It shows that the  
410 surface climate in BCC-ESM1 nearly reaches its equilibrium after 600 years piControl  
411 simulation. The whole system in BCC-ESM1 fluctuates around  $+0.7 \text{ Wm}^{-2}$  net energy flux at  
412 TOA without obvious trend in 450 years (Fig. 1a). This level of TOA energy imbalance is  
413 close to the average imbalance ( $1.0 \text{ Wm}^{-2}$ ) among CMIP5 models (Wild et al., 2013). It means  
414 that there exists surplus energy of  $+0.7 \text{ Wm}^{-2}$  obtained by the whole system in BCC-ESM1,  
415 but do not cause remarkable climate drift. The global mean TAS and SST keep around 288.1  
416 K (Fig. 1b) and 295.05 K (Fig. 1c), respectively. During the last 450 years, there are ( $\pm 0.2 \text{ K}$   
417 amplitude of TAS and SST) oscillations of centennial scale for the whole globe (Figs. 1b and  
418 1c), which are certainly caused by internal variation of the system.

419 Figures 2a-2c show the time series of global annual total burdens of  $\text{SO}_2$ , DMS, and OH  
420 in the troposphere (integrated from the surface to 100 hPa) in the last 450 years of the  
421 piControl simulation. Without any anthropogenic source, the  $\text{SO}_2$  amount in the troposphere  
422 nearly keeps the level of 0.0868 Tg in the 450 years of the piControl simulation. Tropospheric  
423 DMS varies around the value of 0.116 Tg. Tropospheric OH, as an important gas species  
424 oxidizing  $\text{SO}_2$  to form  $\text{SO}_4^{2-}$  (Table 2), keeps at a stable level in the atmosphere.  $\text{SO}_4^{2-}$  also  
425 remains at a stable level of 0.556 Tg in the atmosphere in the whole period of the piControl  
426 simulation (Figure 2d). The amounts of BC and OC in the troposphere vary around 0.0395 Tg  
427 and 0.275 Tg (Figures 2e-2f), respectively. Dust and sea salt aerosols are at the level of 22 Tg  
428 and 11.7 Tg (Figures 2g-2h), respectively. All those data are close to the global mean  
429 concentrations of 0.604 Tg  $\text{SO}_4^{2-}$ , 0.046 Tg BC, 0.30 Tg OC, 22.18 Tg dust, and 11.73 Tg sea  
430 salts in 1850 which are estimated based on the CMIP5 prescribed data in 1850 (Lamarque et  
431 al., 2010).

432 Figure 3 shows the global spatial distributions of annual mean sulfate, organic carbon,  
433 black carbon, dust, and sea salt aerosols in the whole atmospheric column averaged for the  
434 last 100 years of the piControl simulation of BCC-ESM. We can compare them with CMIP5  
435 recommended concentrations in year 1850, considered as the reference state in the



436 pre-industrial stage. At that time, there are fewer anthropogenic/biomass SO<sub>2</sub> emissions, the  
437 SO<sub>4</sub><sup>2-</sup> over land are evidently smaller than those over oceans especially over the tropical  
438 Pacific and Atlantic Oceans, where DMS can be oxidized to SO<sub>2</sub> and then form SO<sub>4</sub><sup>2-</sup>. There  
439 are several centers of high values of black carbon and organic carbon in East and South Asia,  
440 Europe, Southeast America, and in the tropical rain forests in Africa and South America.  
441 They mainly result from biomass burning including vegetation fires, fuel wood and  
442 agricultural burning. Dust aerosols are mainly distributed in North Africa, Central Asia, North  
443 China, and Australia, where arid and semi-arid areas locate. Dust emitted from Sahara Desert  
444 can be transported to the tropical Atlantic by easterly wind. The sea salt aerosols are mainly  
445 distributed over the mid-latitude Southern Oceans, the tropical southern Indian Ocean, and the  
446 tropical northern Pacific Ocean, where wind speeds near the sea surface are strong. As shown  
447 in Fig. 3, all the spatial distribution patterns of CMIP5-derived sulfate, black carbon, organic  
448 carbon, dust, and sea salt aerosols (Lamarque et al., 2010) are well simulated in BCC-ESM1.  
449 There are high spatial correlation coefficients, 0.76 for sulfate, 0.77 for black carbon, 0.77 for  
450 organic carbon, 0.94 for dust, and 0.94 for sea salts, between CMIP5 data and BCC-ESM1  
451 simulations. Relative lower relations for sulfate, black carbon and organic carbon are possibly  
452 caused as different anthropogenic emission sources are used in BCC-ESM1 and to create  
453 CMIP5 data. Dust and sea salts belong to natural aerosols and depend on the land and sea  
454 surface conditions, so their spatial distributions are easy to be captured and have relatively  
455 higher correlations between CMIP5 data and BCC-ESM1 simulations.

456

#### 457 **4. Evaluation of O<sub>3</sub> and aerosol simulations in the 20<sup>th</sup> century**

458 The rate of sulfate formation is dependent on the levels of oxidants in the troposphere.  
459 O<sub>3</sub> is an important oxidant. So, the evaluation of simulated tropospheric O<sub>3</sub> is helpful to  
460 understand the aerosols simulations. BCC-ESM1 is driven by most of the  
461 CMIP6-recommended emission data. As shown in Figure 4, the zonal distributions of the total  
462 amounts of tropospheric O<sub>3</sub> below 300 hPa to the ground and their changes with time from  
463 1850 to 2014 from the CMIP6-recommend dataset (Table 4) are well simulated by  
464 BCC-ESM1. Evident increasing trends since 1850 almost exist in every latitudes, especially  
465 in the Northern Hemisphere where the contents of tropospheric O<sub>3</sub> are higher than those in the

466 Southern Hemisphere.

467 Figure 5 shows the vertical profiles of O<sub>3</sub> simulations with comparison to global  
468 ozonesonde observations averaged for the monthly data over 2010-2014 from the World  
469 Ozone and Ultraviolet Radiation Data Centre (WOUDC; <http://woudc.org/data.php>, last  
470 access: 24 September 2019) in nine regions which are averaged from 41 global WOUDC sites.  
471 The details of WOUDC data may refer to Lu et al. (2019). As shown in Figure 5, BCC-ESM1  
472 well captures the observed ozone vertical structure at all regions. At the lower and middle  
473 troposphere (i.e. below 6 km), the model typically shows positive bias within 5 ppbv for the  
474 Southern Hemisphere and 10 ppbv for the Northern mid-latitudes, similar to those simulated  
475 from many other global atmospheric chemical models (Young et al., 2013, 2018). The model  
476 has larger ozone overestimation in the upper troposphere and stratosphere at most regions, at  
477 least partly due to the use of prescribed stratospheric ozone as upper boundary conditions  
478 and/or errors in modeling ozone exchange between the stratosphere and the troposphere.  
479 Global tropospheric ozone burden derived from our simulation is 335 Tg averaged over  
480 2010-2014, in consistent with recent assessment from multi chemistry models (Young et al.,  
481 2018).

#### 482 **4.1 Global aerosols trends**

483 Figure 6(a)-(c) show the time series of global total emissions of SO<sub>2</sub>, OC, and BC to the  
484 atmosphere from natural and anthropogenic sources. Emissions of SO<sub>2</sub> are largely due to  
485 industrial production. From 1850 to 1915, SO<sub>2</sub> emissions increased year by year as the  
486 Industrial Revolution intensified and expanded. But from 1915 to 1945, the increase trend of  
487 SO<sub>2</sub> emissions became slower as broke out the First and the Second World Wars. After that  
488 period, with growing industrial productions, SO<sub>2</sub> emissions increased again and reached a  
489 maximum around the end of 1970s. During the 1980s and 2000s, with a substantial decrease  
490 of SO<sub>2</sub> emissions in Europe and the United States, the global SO<sub>2</sub> emissions has been  
491 decreasing since the 1980s despite the rapid increase of SO<sub>2</sub> emissions in South and East Asia  
492 as well as in developing countries in the Southern Hemisphere in recent years (Liu et al.,  
493 2009). The OC and BC emissions substantially increased since 1950s just after the Second  
494 World War. The global total OC emission in 2010 was nearly twice as much as that in  
495 pre-industrial (year 1850) and increased by 18 Tg • yr<sup>-1</sup>. Anthropogenic black carbon

496 emissions increased from 1 Tg yr<sup>-1</sup> in 1850 to nearly 8 Tg yr<sup>-1</sup> in 2010.

497 Anthropogenic SO<sub>2</sub>, OC and BC emissions strongly affect the variations of atmospheric  
498 concentrations of sulfate, OC, and BC. The global 0.5°x0.5° gridded data of  
499 CMIP5-recommended aerosols masses with 10-years interval from 1850 to 2000 (Lamarque  
500 et al., 2010) provides an important reference to evaluate the aerosol simulations in  
501 BCC-ESM1. As shown in Figure 7b-7f, the annual total aerosol burdens of SO<sub>4</sub><sup>2-</sup>, OC, and BC  
502 in the whole atmosphere column as simulated by the BCC-ESM1 20<sup>th</sup> century historical  
503 simulation are generally consistent with the values derived from CMIP5-recommended  
504 aerosols concentrations. Due to increasing SO<sub>2</sub> emissions from 1850 to present day (Fig. 6),  
505 the global SO<sub>2</sub> burden in the atmosphere increased from 100 Tg in 1850s to 200 Tg in 1980s  
506 (Fig. 7a), and has a high correlation coefficient of 0.996 with the anthropogenic emissions  
507 (Fig. 6a), as the lifetime of SO<sub>2</sub> is short. The burden directly followed the emission. DMS in  
508 the atmosphere is oxidized by OH and NO<sub>3</sub> to form SO<sub>2</sub> (Table 2). Its natural emissions from  
509 oceans from 1850 to 2010 in the model are the climatological monthly means (Dentener et al.,  
510 2006) from MOZART2 data package. As shown in Fig 7a, the global amount of DMS in the  
511 whole atmosphere was about 0.12 Tg during 1850-1900 and decreased to 0.055 Tg in 2010.  
512 This decrease trend maybe partly results from the speeded rate of DMS oxidation with global  
513 warming, and the loss of DMS gradually exceeds the source of ocean DMS emission to cause  
514 a net loss of DMS in the atmosphere since 1910s. Largely driven by SO<sub>2</sub> anthropogenic  
515 emissions, the sulfate burden shows three different stages from 1850 to present. In the first  
516 period from 1850s to 1900s, the sulfate burden had a weak linear increase. It increased  
517 significantly in the second stage from 1910's to 1940's, and then exploded since 1950's, until  
518 the middle 1970s and early 1980s. The sulfate burden then remained nearly stable and even  
519 showed slightly decreases as seen from the CMIP5 data. As for global BC and OC burdens,  
520 BCC-ESM1 results show continuous increases since 1850s, especially from 1950 to present.  
521 From 1910's to 1940's, the CMIP5 data show a slight decrease of BC and OC burdens in the  
522 atmosphere.

523 The dust and sea salt aerosols in the atmosphere are largely determined by the  
524 atmospheric circulations and states of the land and ocean surface. We can see that the global  
525 dust burden in the atmosphere showed evident increase from 1980 to 2000, which could be

526 partly caused by evident global warming since 1980 and increasing soil dryness resulting in  
527 more surface dust to be released in the atmosphere. Their details will be explored in the other  
528 paper.

## 529 **4.2 Global aerosols budgets**

530 We further evaluate global aerosols budgets by comparing a 10-year average of  
531 BCC-ESM results from 1990 to 2000 with various studies for sulfate, BC, OC, sea salt, and  
532 dust. Their annual total emissions, average atmospheric mass loading, and mean lifetimes are  
533 listed in Tables 5 and 6. It is worth emphasizing that the global mean total source and sink for  
534 each type of aerosols in BCC-ESM1 are almost balanced.

535 The global DMS emission from the ocean is  $27.4 \text{ Tg(S) yr}^{-1}$  in BCC-ESM. This  
536 emission in BCC-ESM is nearly balanced by the gas-phase oxidation of DMS to form  $\text{SO}_2$ .  
537 The DMS burden is 0.12 Tg with a lifetime of 0.78 days, which is within the range of other  
538 models reported in the literature. As shown in Table 5, the total  $\text{SO}_2$  production averaged for  
539 the period of 1991 to 2000 is  $76.93 \text{ Tg(S) yr}^{-1}$ . A rate of  $13.2 \text{ Tg(S) yr}^{-1}$  (about 17%)  $\text{SO}_2$  is  
540 produced from the DMS oxidation, only  $0.1 \text{ Tg(S) yr}^{-1}$   $\text{SO}_2$  from airplane emissions to the  
541 atmosphere, and the rest ( $63.63 \text{ Tg(S) yr}^{-1}$ , near 82.7%) from anthropogenic activities and  
542 volcanic eruption at surface. The amount of  $\text{SO}_2$  produced from the DMS oxidation is in the  
543 range of other works ( $10.0$  to  $24.7 \text{ Tg(S) yr}^{-1}$ ) reported in Liu et al (2005). All the  $\text{SO}_2$   
544 production is balanced by  $\text{SO}_2$  losses by dry and wet deposition, and by gas- and  
545 aqueous-phase oxidation. Half of its loss ( $38.74 \text{ Tg(S) yr}^{-1}$ ) occurs via its aqueous-phase  
546 oxidation to form sulfate. Other losses through dry and wet depositions and gas-phase  
547 oxidation to form  $\text{SO}_4^{2-}$  are also important (Table 2). All the sinks are in the range from the  
548 literature (Liu et al., 2005). The global burden of  $\text{SO}_2$  in the atmosphere is 0.48 Tg with a  
549 lifetime of 1.12 days, consistent with values in literature (Liu et al., 2005).

550 Sulfate aerosol is mainly produced from aqueous-phase  $\text{SO}_2$  oxidation ( $38.73 \text{ Tg(S) yr}^{-1}$ )  
551 and partly from gaseous phase oxidation of  $\text{SO}_2$  ( $10.32 \text{ Tg(S) yr}^{-1}$ ), and is largely lost by wet  
552 scavenging ( $49.06 \text{ Tg(S) yr}^{-1}$ ). The total  $\text{SO}_4^{2-}$  production in BCC-ESM is at the lower range  
553 of values in other models reported in Textor et al. (2006). Its global burden is 1.89 Tg and the  
554 lifetime is 4.69 days, which are within the range of 1.71 to 2.43 Tg and 3.3 to 5.4 days in the  
555 literatures (Textor et al., 2006; Liu et al., 2012; Liu et al., 2016; Matsui and Mahowald, 2017;

556 Tegen et al., 2019; the value derived from CMIP5 data).

557 Sources of BC and OC are mainly from anthropogenic emissions. Based on the CMIP6  
558 data, there are, on average, 7.22 Tg yr<sup>-1</sup> BC and 13.91 Tg yr<sup>-1</sup> OC from fossil and bio-fuel  
559 emissions and 18.38 Tg yr<sup>-1</sup> OC from natural emission during the period of 1991 to 2000.  
560 Most of them are scavenged through convective and large-scale rainfall processes. The rest  
561 returns to the surface by dry deposition. The simulated global BC and OC burdens are 0.13  
562 and 0.62 Tg, respectively (Table 6), all close to values of 0.114 Tg BC and 0.69 Tg OC  
563 derived from the CMIP5 data, and within the range of 0.11-0.26 Tg BC (Textor et al., 2006;  
564 Matsui and Mahowald, 2017; Tegen et al., 2019) and less than the values of 1.25-2.2 Tg OC  
565 in other literatures (Textor et al., 2006; Tegen et al., 2019). The simulated BC and OC  
566 lifetimes are 6.6 and 5.0 days respectively, and are close to the recent values of 5.0-7.5 days  
567 BC and 5.4-6.6 days OC in literatures (Matsui and Mahowald, 2017; Tegen et al., 2019).

568 The emissions of dust and sea salt are mainly determined by winds near the surface. The  
569 annual total dust emission in BCC-ESM1 is 2592 Tg yr<sup>-1</sup>, higher than AeroCom multi-model  
570 mean (1840 Tg yr<sup>-1</sup>, Textor et al., 2006), but comparable to other studies (Chin et al., 2002;  
571 Liu et al., 2012; Matsui and Mahowald, 2017). The average dust loading is 22.93 Tg, lower  
572 than the value of 35.9 Tg in Ginoux et al. (2001) but slightly higher than the value of 20.41  
573 Tg derived from CMIP5 data. The average lifetime for dust particles is 3.23 days that is  
574 shorter than the AeroCom mean (4.14 days) and the value of 3.9 days in recent study (Matsui  
575 and Mahowald, 2017). The simulated sea salt emission is 4667.2 Tg yr<sup>-1</sup>, slightly lower than  
576 the simulated value in Liu et al. (2012), and substantially lower than the AeroCom mean  
577 (16600 Tg yr<sup>-1</sup>, Textor et al., 2006). The simulated sea salt burdens are 11.89 Tg and close to  
578 the CMIP5 data. Their averaged lifetimes are 0.93 days and close to the value in the recent of  
579 Matsui and Mahowald (2017) but longer than the AeroCom mean (0.41days, Textor et al.,  
580 2006).

### 581 **4.3 Global aerosol distributions at present day**

582 Figures 8-12 show December-January-February (DJF) and June-July-August (JJA) mean  
583 column mass concentrations of sulfate (SO<sub>4</sub><sup>2-</sup>), OC, BC, Dust, and Sea Salt aerosols averaged  
584 for the period of 1991-2000, respectively. Here, BCC-ESM1 simulated results are compared  
585 with the CMIP5-recommended data for the same period. Unlike the pre-industrial level of

586 sulfate shown in Fig. 2, sulfate concentrations at present day (Fig. 8) are strongly influenced  
587 by anthropogenic emissions, and have maximum concentrations in the industrial regions (e.g.,  
588 East Asia, Europe, and North America). Their seasonal variations are distinct and are  
589 characterized by high concentrations in boreal summer and low concentrations in boreal  
590 winter. These spatial distributions simulated by BCC-ESM1 are well consistent with the  
591 CMIP5 data, with spatial correlation coefficients in DJF and JJA reaching 0.92 and 0.83  
592 (Figure 13), respectively. The deviation of the spatial pattern in BCC-ESM1 is less from the  
593 CMIP5 data in DJF but larger in JJA (Figure 13).

594 Unlike sulfate whose maximum concentrations are mainly distributed between 60°N  
595 and the equator, peaking concentrations of BC and OC as shown in Figs. 9 and 10 are located  
596 near the tropics in the biomass burning regions (e.g., the maritime continent, Central Africa,  
597 South America), and their seasonal variations from DJF to JJA are evidently weaker than  
598 those of sulfate except in South America. In boreal summer, there are centers of high values  
599 in the industrial regions in the Northern Hemisphere mid-latitudes (i.e., East Asia, South Asia,  
600 Europe, and North America). These main features of spatial and seasonal variations in CMIP5  
601 data are well captured by BCC-ESM1, and the BCC-ESM1 vs. CMIP5 spatial correlation  
602 coefficients (Figure 13) are 0.90 (OC in DJF), 0.91 (BC in DJF), 0.91 (OC in JJA) and 0.92  
603 (BC in JJA). There are less deviations of spatial pattern for OC in DJF and JJA, but larger  
604 deviation for BC from CMIP5 data (Figure 13).

605 As shown in Figure 11, dust concentrations in the atmosphere show largest values over  
606 strong source regions such as Northern Africa, Southwest and Central Asia, and Australia,  
607 and over their outflow regions such as the Atlantic and the western Pacific. In DJF, the  
608 CMIP5 data shows centers of high concentrations over East Asia and Central North America,  
609 but both centers are missing in BCC-ESM1. However, these two high-value centers in the  
610 CMIP5 data may not be true, since frozen soils in these areas in winter lead to unfavorable  
611 conditions for soil erosion by winds. The spatial correlation coefficients between CMIP5 and  
612 BCC-ESM1 remain high: 0.95 in JJA and 0.88 in DJF (Figure 13). Small deviations of spatial  
613 pattern for dust simulations in BCC-ESM1 show less magnitude of dust maximums against  
614 with CMIP5 data (Figure 13).

615 As shown in Figure 12, high sea salt concentrations are generally over the storm track

616 regions over the oceans, e.g., mid-latitudes in the Northern Oceans in DJF and the Southern  
617 Ocean in JJA where wind speeds and thus sea salt emissions are higher. In addition, there is a  
618 belt of high sea salt concentrations in the subtropics of both hemispheres where precipitation  
619 scavenging is weak. Their spatial distributions in BCC-ESM1 are consistent with the CMIP5  
620 data with correlation coefficients of 0.92 in JJA and 0.90 in DJF (Figure 13). The spatial  
621 deviations of sea salt are much closer to CMIP5 data than those of sulfate, OC, BC, and dust  
622 distributions (Figure 13).

623 Figure 14 shows vertical distributions of zonally-averaged annual mean concentrations  
624 of sulfate, organic carbon, black carbon, dust, and sea salt aerosols in the period of 1991-2000.  
625 Both BCC-ESM1 and CMIP5 results show that strong sulfur, OC, and BC emissions in the  
626 industrial regions of the Northern Hemisphere mid-latitudes can rise upward and be  
627 transported towards the North Pole in the mid- to upper troposphere. Most of OC, BC, and  
628 dust aerosols are confined below 500 hPa, while sulfate can be transported to higher altitudes.  
629 Sea salt aerosols are mostly confined below 700 hPa, as the particles are large in size and  
630 favorable for wet removal and gravitational settling towards the surface. It can be seen that  
631 BCC-ESM1 tends to simulate less upward transport of aerosols than the CMIP5 data, likely  
632 reflecting the omission of deep convection transport of tracers in BCC-ESM1.

633 The CMIP5 data used here are mainly from model simulations. We will further evaluate  
634 the BCC-ESM1 model results with ground observations. Annual mean  $\text{SO}_4^{2-}$ , BC and OC  
635 aerosol observations from the Interagency Monitoring of Protected Visual Environments  
636 (IMPROVE) sites over 1990-2005 in the United States  
637 (<http://vista.cira.colostate.edu/IMPROVE/>) and from the European Monitoring and Evaluation  
638 Programme (EMEP) (<http://www.emep.int>) sites over 1995-2005 are used. As shown in  
639 Figure 15a and 15b, the BCC-ESM simulated sulfate concentrations are in general  
640 comparable to the EMEP observations in Europe, but are systematically by about  $1 \mu\text{g m}^{-3}$   
641 higher than the U.S. IMPROVE observations. As for BC, there are large model biases at both  
642 European and U.S. sites (Figs. 15c and 15d), especially BCC-ESM overestimates BC  
643 concentrations at the IMPROVE sites. The observed OC concentrations are slightly  
644 overestimated for IMPROVE sites but systematically underestimated for EMEP sites. Some  
645 statistical features for simulated concentrations versus EMEP and IMPROVE observations are

646 listed in Table 7. These comparisons are overall fairly reasonable considering the  
647 uncertainties in emissions and the coarse model resolution.

648 We then evaluate the simulated BC concentrations from BCC-ESM1 with the HIAPER  
649 (High-Performance Instrumented Airborne Platform for Environmental Research)  
650 Pole-to-Pole Observations (HIPPO) (Wofsy et al., 2011). The HIPPO campaign provided  
651 observations of black carbon concentration profiles over Pacific Ocean and North America  
652 between 2009 and 2011. Following Tilmes et al. (2016), model results here are sampled along  
653 the HIPPO flight tracks and then averaged to different latitude and altitude bands for  
654 comparison. As shown in Figure 16, BCC-ESM1 and HIPPO aircraft observations shows  
655 reasonable agreement in terms of the spatial distributions and seasonal variations of BC levels.  
656 BCC-ESM1 generally reproduces the observed hemispheric gradients of BC, i.e. the larger  
657 burden in the NH compared to the SH, in consistent with Figures 10 and 14. The mean value  
658 of modelled results along the flight track is 11.1 ng/kg, comparable to 8.2 ng/kg of the HIPPO  
659 observations. The model shows large overestimations of BC observations over the tropics,  
660 which is also found in the CAM4-chem global chemical model (Tilmes et al., 2016).

#### 661 **4.4 Aerosol Optical Properties**

662 Aerosol optical depth (AOD) is an indicator of the reduction in incoming solar  
663 radiation (at a particular wavelength) due to scattering and absorption of sunlight by aerosols.  
664 In this study, we calculate the AOD at 550 nm for all aerosols including sulfate, BC, organic  
665 carbon, sea salt and dust as the product of aerosol dry mass concentrations, aerosol water  
666 content, and their specific extinction coefficients. The total AOD is calculated by summing  
667 the AOD in each model layer for each aerosol species using the assumption that they are  
668 externally mixed. The AOD observations retrieved from MODIS and MISR over the period of  
669 1997-2003, and from AERONET over the period of 1998–2005 (<http://aeronet.gsfc.nasa.gov>)  
670 are used to evaluate the averaged AOD at 550 nm in BCC-ESM. Figure 17 shows averages of  
671 MISR and MODIS AOD with corresponding averages from BCC-ESM. The BCC-ESM1  
672 simulated AOD generally captures the spatial distribution of MISR and MODIS retrievals.  
673 The model overestimates AOD over East China. It also systematically underestimates the  
674 MODIS observations in the Southern Hemisphere, but is closer to MISR observations. Figure



675 18 shows multi-years annual means of BCC-ESM1 simulated AOD values versus  
676 observations from AERONET over the period of 1998–2005. The basic pattern of modeled  
677 global AOD is similar to that of observations and their spatial correlation reaches 0.56. Large  
678 values of AOD are mainly distributed in land continents such as North African, South Asia,  
679 East Asia, Europe, and eastern part of North America. Figures 19a-19d present scatter plots of  
680 observed versus simulated multi-year monthly mean AOD at those sites of AERONET in  
681 Europe, North America, East Asia, and South Asia over the period of 1998-2005, respectively.  
682 Model simulated monthly AOD generally agrees with observations within a factor of 2 for  
683 most sites. BCC-ESM slightly overestimates the AOD in European and North American sites.  
684 In those regions, BCC-ESM also slightly overestimates MODIS and MISR AOD observations  
685 (Fig. 17).

## 686 **5. Summary and discussions**

687 This paper presents a primary evaluation of aerosols simulated in version 1 of the Beijing  
688 Climate Center Earth System Model (BCC-ESM1) with the implementation of the interactive  
689 atmospheric chemistry and aerosol based on the newly developed BCC-CSM2. Global  
690 aerosols (including sulfate, organic carbon, black carbon, dust and sea salt) and major  
691 greenhouse gases (e.g., O<sub>3</sub>, CH<sub>4</sub>, N<sub>2</sub>O) in the atmosphere can be interactively simulated when  
692 anthropogenic emissions are provided to the model. Concentrations of all aerosols in  
693 BCC-ESM1 are determined by the processes of advective transport, emission, gas-phase  
694 chemical reactions, dry deposition, gravitational settling, and wet scavenging by clouds and  
695 precipitation. The nucleation and coagulation of aerosols are ignored in the present version of  
696 BCC-ESM1. Effects of aerosols on radiation, cloud, and precipitation are fully included.

697 We evaluate the performance of BCC-ESM1 in simulating aerosols and their optical  
698 properties in the 20th century following CMIP6 historical simulation according to the  
699 requirement of the AerChemMIP. It is forced with anthropogenic emissions evolving from  
700 1850 to 2014 but some WMGHGs such as CH<sub>4</sub>, N<sub>2</sub>O, CO<sub>2</sub>, CFC11 and CFC12 are prescribed  
701 using CMIP6 prescribed concentrations (to replace prognostic values of CH<sub>4</sub> and N<sub>2</sub>O from  
702 the chemistry scheme). Both direct and indirect effects of aerosols are considered in  
703 BCC-ESM1. Initial conditions of the CMIP6 historical simulation are obtained from a  
704 600-year piControl simulation in the absence of anthropogenic emissions, which well captures

705 the pre-industrial concentrations of  $\text{SO}_4^{2-}$ , organic carbon (OC), black carbon (BC), dust, and  
706 sea salt aerosols and are consistent with the CMIP5 recommended concentrations for the year  
707 1850. With the CMIP6 anthropogenic emissions of  $\text{SO}_2$ , OC, and BC from 1850 to 2014 and  
708 their natural emissions implemented in BCC-ESM1, the model simulated  $\text{SO}_4^{2-}$ , BC, and OC  
709 aerosols in the atmosphere are highly correlated with the CMIP5-recommended data. The  
710 long-term trends of CMIP5 aerosols from 1850 to 2000 are also well simulated by  
711 BCC-ESM1. Global budgets of aerosols were evaluated through comparisons of BCC-ESM1  
712 results for 1990-2000 with reports in various literatures for sulfate, BC, OC, sea salt, and dust.  
713 Their annual total emissions, atmospheric mass loading, and mean lifetimes are all within the  
714 range of values reported in relevant literature. Evaluations of the spatial and vertical  
715 distributions of BCC-ESM1 simulated present-day  $\text{SO}_4^{2-}$ , OC, BC, Dust, and sea salt aerosol  
716 concentrations against the CMIP5 datasets and in-situ measurements of surface networks  
717 (IMPROVE in the U.S. and EMEP in Europe), and HIPPO aircraft observations indicate good  
718 agreement among them. The BCC-ESM1 simulates weaker upward transport of aerosols from  
719 the surface to the middle and upper troposphere (with reference to CMIP5-recommended  
720 data), likely reflecting a lack of deep convection transport of chemical species in the present  
721 version of BCC-ESM1. The AOD at 550 nm for all aerosols including sulfate, BC, OC, sea  
722 salt, and dust aerosols was further compared with the satellite AOD observations retrieved  
723 from MODIS and MISR and surface AOD observations from AERONET. The BCC-ESM1  
724 model results are overall in good agreement with these observations within a factor of 2. All  
725 these comparisons demonstrate the success of the implementation of interactive aerosol and  
726 atmospheric chemistry in BCC-ESM1.

727 This work has only evaluated the ability of BCC-ESM1 to simulate aerosols. The  
728 variations of aerosols especially for sulfate are related to other gaseous tracers such as OH  
729 and  $\text{NO}_3$  (Table 2), which are determined by the MOZART2 gaseous chemical scheme as  
730 implemented in BCC-ESM1, and require further evaluation. As limited length of the text, the  
731 other optical feature of aerosols such as extinction coefficients, single scattering albedo and  
732 asymmetry parameters, and even their feedbacks on radiation and global temperature change  
733 will be explored in the other paper.  $\text{O}_3$  is evaluated in this work. Other GHGs such as  $\text{CH}_4$  and

734 N<sub>2</sub>O concentrations can be simulated when forced with emissions and their simulations also  
735 need to be evaluated in future.

## 736 **6. Code and data availability**

737 The source codes of BCC-ESM1 ~~model are available for use under licence agreement.,~~  
738 model input files, and scripts to reproduce the simulations that are presented in the article  
739 have been archived and made publicly available for downloading from  
740 <https://zenodo.org/record/3609337> (Wu et al., 2020). Readers interested in BCC-ESM1 codes  
741 and the model input files required to reproduce the simulations may contact Dr. Tongwen Wu  
742 ([twwu@ema.gov.cn](mailto:twwu@ema.gov.cn)) for further details. Model output of BCC CMIP6 AerChemMIP  
743 simulations described in this paper refer to Zhang et al. (2019) and is distributed through  
744 ESGF and freely accessible through the ESGF data portals after registration. Details about  
745 ESGF are presented on the CMIP Panel website at  
746 <http://www.wcrp-climate.org/index.php/wgcm-cmip/about-cmip>.

747

## 748 **Author contributions**

749 Tongwen Wu led the BCC-ESM1 development. All other co-authors have contributions  
750 to it. Fang Zhang and Jie Zhang designed the experiments and carried them out. Tongwen Wu,  
751 Laurent Li, Lin Zhang, Xiaohong Liu, Aixue Hu, and Jun Wang wrote the final document  
752 with contributions from all other authors.

753

## 754 **Acknowledgements**

755 This work was supported by The National Key Research and Development Program of China  
756 (2016YFA0602100). All the figures are created by the NCAR Command Language (Version  
757 6.6.2) [Software].

758

## 759 **References**

760 Albani, S., Mahowald, N. M., Perry, A. T., Scanza, R. A., Zender, C. S., Heavens, N. G.,  
761 Maggi, V., Kok, J. F., and Otto-Bliesner, B. L.: Improved dust representation in the  
762 Community Atmosphere Model, *J. Adv. Model. Earth Syst.*, 6, 541–570,

763 doi:10.1002/2013MS000279, 2014.

764 Albrecht, B.: Aerosols, cloud microphysics, and fractional cloudiness, *Science*, 245, 1227–  
765 1230, 1989.

766 Arora, V., Boer, G., Friedlingstein, P., Eby, M., Jones, C., Christian, J., Bonan, G., Bopp, L.,  
767 Brovkin, V., Cadule, P., Hajima, T., Ilyina, T., Lindsay, K., Tjiputra, J., and Wu, T.:  
768 Carbon-concentration and carbon-climate feedbacks in CMIP5 Earth system models. *J.*  
769 *Climate*, 26, 5289–5314, 2013.

770 Austin, J., Butchart, N., and Shine, K. P.: Possibility of an Arctic ozone hole in a  
771 doubled-CO<sub>2</sub> climate, *Nature*, 360, 221–225, 1992

772 Barth, M.C., Rasch, P.J., Kiehl, J.T., Benkowitz, C.M., and Schwartz, S.E.: Sulfur chemistry  
773 in the National Center for Atmospheric Research Community Climate Model:  
774 Description, evaluation, features, and sensitivity to aqueous chemistry. *J. Geophys. Res.*,  
775 105, D1, 1387-1415, 2000.

776 Bey I., Jacob, D. J., Yantosca, R. M., Logan, J. A., Field, B., Fiore, A. M., Li, Q., Liu, H.,  
777 Mickley, L. J., and Schultz, M.: Global modeling of tropospheric chemistry with  
778 assimilated meteorology: Model description and evaluation, *J. Geophys. Res.*, 106,  
779 23,073-23,096, 2001

780 Boucher, O., Lohmann, U.: The sulphate-CCN-cloud albedo effect – a sensitivity study with  
781 two general circulation models, *Tellus* 47B, 281–300, 1995.

782 Brasseur, G. P., Hauglustaine, D. A., Walters, S., Rasch, P. J., Müller, J.-F., Granier, C., and  
783 Tie, X. X.: MOZART, a global chemical transport model for ozone and related chemical  
784 tracers: 1. Model description, *J. Geophys. Res.*, 103, 28,265– 28,289, 1998.

785 Brasseur, G. P., Tie, X. X., Rasch, P. J., and Lefèvre, F.: A three - dimensional simulation of  
786 the Antarctic ozone hole: Impact of anthropogenic chlorine on the lower stratosphere and  
787 upper troposphere, *J. Geophys. Res.*, 102, 8909–8930, 1997.

788 Cariolle, D., Lasserre-Bigorry, A., and Royer, J.-F.: A general circulation model simulation of  
789 the springtime Antarctic ozone decrease and its impact on midlatitudes, *J. Geophys. Res.*,  
790 95, 1883–1898, 1990.

791 Cess, R. D.: Nuclear war: Illustrative effects of atmospheric smoke and dust upon solar  
792 radiation, *Clim. Change*, 7, 237–251, 1985.

793 Chen, C., and Cotton, W. R.: The physics of the marine stratocumulus-capped mixed layer, *J.*  
794 *Atmos. Sci.*, 44 (50), 2951–2977, 1987.

795 Chin, M., Ginoux, P., Kinne, S., Torres, O., Holben, B.N., Duncan, B.N., Martin, R.V., Logan, J.A.,  
796 Higurashi, A., Naka-jima, T.: Tropospheric aerosol optical thickness from the GOCART  
797 model and comparisons with satellite and Sun photometer measurements. *J. Atmos.*  
798 *Sci.* 59:461–483, 2002.

799 Chuang, C. C., Penner, J. E., Taylor, K. E., Grossman, A. S., and Walton, J. J.: An assessment  
800 of the radiative effects of anthropogenic sulfate, *J. Geophys. Res.*, 102, 3761–3778,  
801 1997.

802 Collins, W. J., Lamarque, J.-F., Schulz, M., Boucher, O., Eyring, V., Hegglin, M. I.,  
803 Maycock, A., Myhre, G., Prather, M., Shindell, D., Smith, S. J.: AerChemMIP:  
804 quantifying the effects of chemistry and aerosols in CMIP6, *Geosci. Model Dev.*, 10,  
805 585–607, 2017.

806 Collins, W. D., Rasch, P. J., Boville, B. A., Hack, J. J., McCaa, J. R., Williamson, D. L.,  
807 Kiehl, J. T., Briegleb, B. P., Bitz, C., Lin, S.-J., Zhang, M., and Dai, Y.: Description of  
808 the NCAR Community Atmosphere Model (CAM3). *Nat. Cent. for Atmos. Res.*,  
809 Boulder, Colo., 2004.

810 Cooke, W.F., Wilson, J.J.N.: A global black carbon aerosol model. *J. Geophys. Res. Atmos.*  
811 101, 19395–19409, 1996.

812 Cunnold, D., Alyea, F., Phillips, N., Prinn, R.: A three-dimensional dynamical-chemical  
813 model of atmospheric ozone, *J. Atmos. Sci.*, 32, 170-194, 1975.

814 Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S.,  
815 Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J.-P., Textor, C., Schulz, M.,  
816 van der Werf, G. R., and Wilson, J.: Emissions of primary aerosol and precursor gases in  
817 the years 2000 and 1750 prescribed data-sets for AeroCom, *Atmos. Chem. Phys.*, 6,  
818 4321–4344, doi:10.5194/acp-6-4321-2006, 2006.

819 Eyring, V., Bony, S., Meehl, G. A., Senior, C. A., Stevens, B., Stouffer, R. J., and Taylor, K.  
820 E.: Overview of the Coupled Model Intercomparison Project Phase 6 (CMIP6)  
821 experimental design and organization, *Geosci. Model Dev.*, 9, 1937–1958,  
822 doi:10.5194/gmd-9-1937-2016, 2016.

823 Feichter, J., Kjellstrom, E., Rodhe, H., Dentener, F., Lelieveld, J., Roelofs, G.-J.: Simulation  
824 of the tropospheric sulfur cycle in a global climate model, 30: 1693-1707, 1996.

825 Feng, L., Smith, S. J., Braun, C., Crippa, M., Gidden, M. J., Hoesly, R., Klimont, Z., van  
826 Marle, M., van den Berg, M., and van der Werf, G. R.: Gridded Emissions for CMIP6,

827 Geosci. Model Dev. Discuss., <https://doi.org/10.5194/gmd-2019-195>, 2019

828 Ghan, S. J. and Easter, R. C.: Impact of cloud-borne aerosol representation on aerosol direct  
829 and indirect effects, *Atmos. Chem. Phys.*, 6, 4163–4174, 2006.

830 Ginoux, P., M. Chin, I. Tegen, J. M. Prospero, B. Holben, O. Dubovik, and S.-J. Lin (2001),  
831 Sources and distributions of dust aerosols simulated with the GOCART model, *J.*  
832 *Geophys. Res.*, 106, 20,255 – 20,274.

833 Giorgi, F., and Chameides, W. L.: The rainout parameterization in a photochemical model, *J.*  
834 *Geophys. Res.*, 90, 7872–7880, 1985.

835 Guenther, A. B., Jiang, X., Heald, C. L., et al.: The Model of Emissions of Gases and  
836 Aerosols from Nature Version 2.1 (MEGAN2.1): An Extended and Updated Framework  
837 for Modeling Biogenic Emissions. *Geoscientific Model Development* 5(6): 1471–1492,  
838 2012.

839 Guenther, A., Baugh, B. Brasseur, G., Greenberg, J., Harley, P., Klinger, L., Serca, D., and  
840 Vierling, L.: Isoprene emission estimates and uncertainties for the Central African  
841 EXPRESSO study domain, *J. Geophys. Res.*, 104(D23), 30, 625–630, 639, 1999.

842 Hess, M., Koepke, P., Schult, I.: Optical properties of aerosols and clouds: the software  
843 package OPAC, *Bull. Am. Meteorol. Soc.*, 79, 831–844, 1998.

844 Hoesly, R. M., Smith, S. ., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T.,  
845 Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N.,  
846 Kurokawa, J., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, R. R., and Zhang Q.:  
847 Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the  
848 Community Emission Data System (CEDS), *Geosci. Model Dev.*, 11, 369-408, 2018

849 Horowitz, L.W., Walters, S., Mauzerall, D. L., Emmons, L. K., Rasch, P. J., Granier, C., Tie,  
850 X., Lamarque, J.-F., Schultz, M. G., Tyndall, G. S., Orlando, J. J., Brasseur, G. P.: A  
851 global simulation of tropospheric ozone and related tracers: Description and evaluation  
852 of MOZART, version 2, *J. Geophys. Res.*, 108(D24), 4784, doi:10.1029/2002JD002853,  
853 2003.

854 Horowitz, L. W.: Past, present, and future concentrations of tropospheric ozone and aerosols:  
855 Methodology, ozone evaluation, and sensitivity to aerosol wet removal, *J. Geophys. Res.*,  
856 111, D22211, doi:10.1029/2005JD006937, 2006.

857 Hoffman, F. M., Randerson, J. T., Arora, V. K., Bao, Q., Cadule, P., Ji, D., Jones, C. D.,  
858 Kawamiya, M., Khatiwala, S., Lindsay, K., Obata, A., Shevliakova, E., Six, K. D.,

859 Tjiputra, J. F., Volodin, E. M., and Wu, T.: Causes and implications of persistent  
860 atmospheric carbon dioxide biases in Earth System Models, *J. Geophys. Res. Biogeosci.*,  
861 119, 141–162, doi:10.1002/2013JG002381, 2014.

862 Holtslag, A. A. M., and Boville, B. A.: Local versus nonlocal boundary-layer diffusion in a  
863 global climate model, *J. Climate*, 6, 1825–1842, 1993.

864 Hurtt, G.C., Chini, L.P., Frolking, S. et al.: Harmonization of land-use scenarios for the period  
865 1500–2100: 600 years of global gridded annual land-use transitions, wood harvest, and  
866 resulting secondary lands, *Climatic Change*, 109, 117-161, 2011.

867 Hurtt, G., et al.: input4MIPs.UofMD.landState.CMIP.UofMD-landState-2-1-h, version  
868 20170126, Earth Syst. Grid Fed., <http://doi.org/10.22033/ESGF/input4MIPs.1127>, 2017.

869 Jacobson, M.Z.: Investigating cloud absorption effects: global absorption properties of black  
870 carbon, tar balls, and soil dust in clouds and aerosols. *J. Geophys. Res.* 117, D06205,  
871 2012.

872 Jones, C.D., Arora, V., Friedlingstein, P., Bopp, L., Brovkin, V., Dunne, J., Graven, H.,  
873 Hoffman, F., Ilyina, T., John, J. G., Jung, M., Kawamiya, M., Koven, C., Pongratz, J.,  
874 Raddatz, T., Randerson, J. T., and Zaehle, S.: C4MIP – The Coupled Climate–Carbon  
875 Cycle Model Intercomparison Project: experimental protocol for CMIP6, *Geosci. Model  
876 Dev.*, 9, 2853–2880, doi:10.5194/gmd-9-2853-2016, 2016.

877 Lamarque, J.-F., Shindell, D. T., Josse, B., Young, P. J., Cionni, I., Eyring, V., Bergmann, D.,  
878 Cameron-Smith, P., Collins, W. J., Doherty, R., Dalsoren, S., Faluvegi, G., Folberth, G.,  
879 Ghan, S. J., Horowitz, L. W., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Naik, V.,  
880 Plummer, D., Righi, M., Rumbold, S. T., Schulz, M., Skeie, R. B., Stevenson, D. S.,  
881 Strode, S., Sudo, K., Szopa, S., Voulgarakis, A., and Zeng, G.: The Atmospheric  
882 Chemistry and Climate Model Intercomparison Project (ACCMIP): overview and  
883 description of models, simulations and climate diagnostics, *Geosci. Model Dev.*, 6, 179–  
884 206, doi:10.5194/gmd-6-179-2013, 2013.

885 Lamarque, J.-F., Emmons, L. K., Hess, P. G., Kinnison, D. E., Tilmes, S., Vitt, F., Heald, C.  
886 L., Holland, E. A., Lauritzen, P. H., Neu, J., Orlando, J. J., Rasch, P. J., and Tyndall, G.  
887 K.: CAM-chem: description and evaluation of interactive atmospheric chemistry in the  
888 Community Earth System Model, *Geosci. Model Dev.*, 5, 369–411, 2012

889 Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse,  
890 C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van

891 Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V.,  
892 Riahi, K., and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and  
893 biomass burning emissions of reactive gases and aerosols: methodology and application,  
894 *Atmos. Chem. Phys.*, 10, 7017-7039, <https://doi.org/10.5194/acp-10-7017-2010>, 2010.

895 Lawrence, D. M., Hurtt, G. C., Arneth, A., Brovkin, V., Calvin, K. V., Jones, A. D., Jones, C.  
896 D., Lawrence, P. J., de Noblet-Ducoudré N., Pongratz, J., Seneviratne, S. I., and  
897 Shevliakova, E.: The Land Use Model Intercomparison Project (LUMIP) contribution to  
898 CMIP6: rationale and experimental design, *Geosci. Model Dev.*, 9, 2973–2998,  
899 <https://doi.org/10.5194/gmd-9-2973-2016>, 2016.

900 Lohmann, U., Feichter, J., Penner, J. E., and Leaitch, W. R.: Indirect effect of sulfate and  
901 carbonaceous aerosols: A mechanistic treatment. *J. Geophys. Res.*, 105, 12193–12206,  
902 2000

903 Li, W., Zhang, Y., Shi, X., Zhou, W., Huang, A., Mu, M., Qiu, B., Ji, J.: Development of the  
904 Land Surface Model BCC\_AVIM2.0 and Its Preliminary Performance in  
905 LS3MIP/CMIP6, *J. Meteor. Res.*, 33, 851-869, doi: 10.1007/s13351-019-9016-y, 2019.

906 Liu, X. H., Penner, J. E., and Herzog, M.: Global modeling of aerosol dynamics: Model  
907 description, evaluation, and interactions between sulfate and nonsulfate aerosols, *J.*  
908 *Geophys. Res.-Atmos.*, 110, D18206, doi:10.1029/2004jd005674, 2005.

909 Liu, X., Easter, R.C. Ghan, S.J., Zaveri, R., Rasch, P., Shi, X., Lamarque, J.-F., Gettelman, A.,  
910 Morrison, H., Vitt, F., Conley, A., Park, S., Neale, R., Hannay, C., Ekman, A.M., Hess,  
911 P., Mahowald, N., Collins, W., Iacono, M.J., Bretherton, C.S., Flanner, M.G., and  
912 Mitchell, D.: Toward a Minimal Representation of Aerosols in Climate Models:  
913 Description and Evaluation in the Community Atmosphere Model CAM5.  
914 *Geos.Model.Dev.* 5(3):709-739. 2012.

915 Liu, X., Ma, P. -L., Wang, H., Tilmes, S., Singh, B., Easter, R. C., Ghan, S. J., and Rasch, P.  
916 J.: Description and evaluation of a new four-mode version of the Modal Aerosol Module  
917 (MAM4) within version 5.3 of the Community Atmosphere Model, *Geosci. Model Dev.*,  
918 9, 505–522, <https://doi.org/10.5194/gmd-9-505-2016>, 2016.

919 Liu, J., Mauzerall, D.L., Horowitz, L.W., Ginoux, P., Fiore, A.M.: Evaluation intercontinental  
920 transport of fine aerosols: (1) methodology, global aerosol distribution and optical depth.  
921 *Atmos Environ* 43:4327–4338, 2009.

922 Lu, X., Zhang, L., Wu, T., Long, M., Wang, J., Jacob, D., Zhang F., Zhang, J., Eastham, S.,



923 Hu, L., Zhu, L., Liu, X., and Wei, M.: Development of the global atmospheric general  
924 circulation-chemistry model BCC-GEOS-Chem v1.0: model description and evaluation,  
925 submitted to Geos.Model.Dev.

926 Mahowald, N., Lamarque, J.-F., Tie, X., and Wolff, E.: Sea salt aerosol response to climate  
927 change: last glacial maximum, preindustrial and doubled carbon dioxide climates, *J.*  
928 *Geophys. Res.*, 111, D05303, doi:10.1029/2005JD006459, 2006.

929 Martin, R. V., et al.: Interpretation of TOMS observations of tropical tropospheric ozone with  
930 a global model and in situ observations, *J. Geophys. Res.*, 107(D18), 4351,  
931 doi:10.1029/2001JD001480, 2002.

932 Matsui, H., and Mahowald, N.: Development of a global aerosol model using a  
933 two-dimensional sectional method: 2. Evaluation and sensitivity simulations, *J. Adv.*  
934 *Model. Earth Syst.*, 9, 1887 – 1920, doi:10.1002/2017MS000937, 2017.

935 Matthes, K., Funke, B., Andersson, M. E., Barnard, L., Beer, J., Charbonneau, P., Clilverd, M.  
936 A., Dudok de Wit, T., Haberleiter, M., Hendry, A., Jackman, C. H., Kretzschmar, M.,  
937 Kruschke, T., Kunze, M., Langematz, U., Marsh, D. R., Maycock, A. C., Misios, S.,  
938 Rodger, C. J., Scaife, A. A., Seppälä, A., Shangguan, M., Sinnhuber, M., Tourpali, K.,  
939 Usoskin, I., van de Kamp, M., Verronen, P. T., and Versick, S.: Solar forcing for CMIP6  
940 (v3.2), *Geosci. Model Dev.*, 10, 2247–2302, <https://doi.org/10.5194/gmd-10-2247-2017>,  
941 2017.

942 Meinshausen, M., Vogel, E., Nauels, A., Lorbacher, K., Meinshausen, N., Etheridge, D. M.,  
943 Fraser, P. J., Montzka, S. A., Rayner, P. J., Trudinger, C. M., Krummel, P. B., Beyerle,  
944 U., Canadell, J. G., Daniel, J. S., Enting, I. G., Law, R. M., Lunder, C. R., O'Doherty, S.,  
945 Prinn, R. G., Reimann, S., Rubino, M., Velders, G. J. M., Vollmer, M. K., Wang, R. H. J.,  
946 and Weiss, R.: Historical greenhouse gas concentrations for climate modelling (CMIP6),  
947 *Geosci. Model Dev.*, 10, 2057–2116, <https://doi.org/10.5194/gmd-10-2057-2017>, 2017.

948 Mora, C., Wei, C.-L., Rollo, A., Amaro, T., Baco, A.R., Billett, D., Bopp, L., Chen, Q.,  
949 Collier, M., Danovaro, R., Gooday, A.J., Grupe, B.M., Halloran, P.R., Ingels, J., Jones,  
950 D.O.B., Levin, L.A., Nakano, H., Norling, K., Ramirez-Llodra, E., Rex, M., Ruh, H.A.,  
951 Smith, C.R., Sweetman, A.K., Thurber, A.R., Tjiputra, J. F., Usseglio, P., Watling, L.,  
952 Wu, T., Yasuhara, M.: Biotic and human vulnerability to projected ocean  
953 biogeochemistry change over the 21st century, *PLoS Biol* 11(10): e1001682.  
954 doi:10.1371/journal.pbio.1001682, 2013.

955 NCAR Command Language (Version 6.6.2) [Software], Boulder, Colorado:  
956 UCAR/NCAR/CISL/TDD. <http://dx.doi.org/10.5065/D6WD3XH5>, 2019.

957 Neale, R. B., et al.: Description of the NCAR Community Atmosphere Model (CAM 4.0),  
958 NCAR Tech. Note, TN-485, pp. 212, Natl. Cent. for Atmos. Res., Boulder, Colo., 2010

959 Neu, J. L. and Prather, M. J.: Toward a more physical representation of precipitation  
960 scavenging in global chemistry models: cloud overlap and ice physics and their impact  
961 on tropospheric ozone, *Atmos. Chem. Phys. Discuss.*, 11, 24413–24466,  
962 doi:10.5194/acpd-11-24413-2011, 2011.

963 Olivier, J.G.J., Bouwman, A.F., Van der Maas, C.W.M., Berdowski, J.J.M., Veldt, C., Bloos,  
964 J.P.J., Visschedijk, A.J.H., Zandveld, P.Y.J., Haverslag, J.L., Description of EDGAR  
965 Version 2.0: A set of global emission inventories of greenhouse gases and ozone  
966 depleting substances for all anthropogenic and most natural sources on a per country  
967 basis and on 1° x 1° grid. RIVM Techn. Report nr. 771060002, TNO-MEP report nr.  
968 R96/119. Nat. Inst. Of Public Health and the Environment/ Netherlands Organisation for  
969 Applied Scientific Research, Bilthoven, 1996.

970 Peng, Y., and Lohmann, U.: Sensitivity study of the spectral dispersion of the cloud droplet  
971 size distribution on the indirect aerosol effect, *Geophys. Res. Lett.*, 30(10), 1507,  
972 doi:10.1029/2003GL017192, 2003.

973 Price, C., and Rind, D.: A simple lightning parameterization for calculating global lightning  
974 distributions, *J. Geophys. Res.*, 97, 9919-9933, 1992.

975 Quaas, J., Boucher, O., and Lohmann, U.: Constraining the total aerosol indirect effect in  
976 the LMDZ and ECHAM4 GCMs using MODIS satellite data. *Atmos Chem Phys* 6,947–  
977 955, 2006.

978 Sander, S., Friedl, R. R., Ravishankara, A. R., et al.: Chemical Kinetics and Photochemical  
979 Data for Use in Atmospheric Studies, Evaluation Number 14, JPL Publication 02-25,  
980 NASA, Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA,  
981 2003.

982 Schlesinger, M. E., Mintz, Y.: Numerical simulation of ozone production, transport and  
983 distribution with a global atmospheric general circulation model, *J.Atmos.Sci.*, 36:  
984 1325-1361, 1979.

985 Shindell, D.T., Horowitz, L.W., Schwarzkopf, M.D.: Composition Models in Climate

986 Projections Based on Emissions Scenarios for Long-Lived and Short-Lived Radiatively  
987 Active Gases and Aerosols. H.Levy II, D.T. Shindell, A.Gilliland, M.D.Schwarzkopf,  
988 L.W.Horowitz, (eds.) .A Report by the U.S.Climate Change Science Program and the  
989 Subcommittee on Global Change Research, Washington, D.C., 2008

990 Solomon, S.: Stratospheric ozone depletion: A review of concepts and history, *Reviews of*  
991 *Geophysics*, 37, 275–316, 1999.

992 Taylor, K.E., Stouffer, R. J., Meehl, G. A.: An overview of CMIP5 and the experiment design,  
993 *Bull. Am. Meteorol. Soc.* 93, 485-498, 2012.

994 Tegen, I., Neubauer, D., Ferrachat, S., Siegenthaler-Le Drian, C., Bey, I., Schutgens, N., Stier,  
995 P., Watson-Parris, D., Stanelle, T., Schmidt, H., Rast, S., Kokkola, H., Schultz, M.,  
996 Schroeder, S., Daskalakis, N., Barthel, S., Heinold, B., and Lohmann, U.: The global  
997 aerosol–climate model ECHAM6.3–HAM2.3 – Part 1: Aerosol evaluation, *Geosci.*  
998 *Model Dev.*, 12, 1643–1677, <https://doi.org/10.5194/gmd-12-1643-2019>, 2019.

999 Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen,  
1000 T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Feichter, H., Fillmore, D.,  
1001 Ghan, S., Ginoux, P., Gong, S., Grini, A., Hendricks, J., Horowitz, L., Huang, P., Isaksen,  
1002 I., Iversen, I., Kloster, S., Koch, D., Kirkevåg, A., Kristjansson, J. E., Krol, M., Lauer, A.,  
1003 Lamarque, J. F., Liu, X., Montanaro, V., Myhre, G., Penner, J., Pitari, G., Reddy, S.,  
1004 Seland, Ø., Stier, P., Takemura, T., and Tie, X.: Analysis and quantification of the  
1005 diversities of aerosol life cycles within AeroCom, *Atmos. Chem. Phys.*, 6, 1777–1813,  
1006 <https://doi.org/10.5194/acp-6-1777-2006>, 2006.

1007 Thomason, L. W., Ernest, N., Millán, L., Rieger, L., Bourassa, A., Vernier, J. P., Manney, G.,  
1008 Luo, B.P., Arfeuille, F., Peter, T.: A global space - based stratospheric aerosol  
1009 climatology: 1979-2016. *Earth System Science Data*, 10(1), 469–492, doi:  
1010 10.5194/essd-10-469-2018, 2018.

1011 Tie, X., Brasseur, G., Emmons, L., Horowitz, L., and Kinnison, D.: Effects of aerosols on  
1012 tropospheric oxidants: A global model study, *J. Geophys. Res.*, 106, 2931– 2964, 2001.

1013 Tie, X., Madronich, S., Walters, S., Edwards, D., Ginoux, P., Mahowald, N., Zhang, R., Luo,  
1014 C., and Brasseur, G.: Assessment of the global impact of aerosols on tropospheric  
1015 oxidants, *J. Geophys. Res.*, 110, D03204, doi:10.1029/2004JD005359, 2005.

1016 Tilmes, S., Lamarque, J.-F., Emmons, L. K., Kinnison, D. E., Marsh, D., Garcia, R. R., Smith,  
1017 A. K., Neely, R. R., Conley, A., Vitt, F., Val Martin, M., Tanimoto, H., Simpson, I.,

1018 Blake, D. R., and Blake, N.: Representation of the Community Earth System Model  
1019 (CESM1) CAM4-chem within the Chemistry-Climate Model Initiative (CCMI),  
1020 Geoscientific Model Development, 9, 1853-1890, 2016.

1021 Todd-Brown, K.E.O., Randerson, J.T., Hopkins, F., Arora, V., Hajima, T., Jones, C.,  
1022 Shevliakova, E., Tjiputra, J., Volodin, E., Wu, T., Zhang, Q., Allison, S.D.: Changes in  
1023 soil organic carbon storage predicted by Earth system models during the 21st century,  
1024 Biogeosciences, 11, 2341-2356, 2014.

1025 Van Marle, M.J.E., S. Kloster, B.I. Magi, J.R. Marlon, A.-L. Daniau, R.D. Field, A. Arneth,  
1026 M. Forrest, S. Hantson, N.M. Kehrwald, W. Knorr, G. Lasslop, F. Li, S. Mangeon, C.  
1027 Yue, J.W. Kaiser, and G.R. van der Werf, 2017: Historic global biomass burning  
1028 emissions for CMIP6 (BB4CMIP) based on merging satellite observations with proxies  
1029 and fire models (1750-2015). Geosci. Model Dev., 10, 3329-3357,  
1030 doi:10.5194/gmd-10-3329-2017.

1031 Wang, J., Hoffmann, A. A., Park, R., Jacob, D. J., and Martin, S. T.: Global distribution of  
1032 solid and aqueous sulfate aerosols: effect of the hysteresis of particle phase transitions, J.  
1033 Geophys. Res., 113, D11206, Doi:11210.11029/12007JD009367, 2008a.

1034 Wang, J., Jacob, D. J., and Martin, S. T.: Sensitivity of sulfate direct climate forcing to the  
1035 hysteresis of particle phase transitions, J. Geophys. Res., 113, D11207,  
1036 doi:11210.11029/12007JD009368, 2008b.

1037 Wesely, M. L.: Parameterization of surface resistance to gaseous dry deposition in  
1038 regional-scale numerical models, Atmos. Environ., 23, 1293–1304, 1989.

1039 Wild, M., Folini, D., Schar, C., Loeb, N., Dutton, E.G., Konig-Langlo, G.: The global energy  
1040 balance from a surface perspective, Climate Dynamics, 40: 3107-3134, 2013.

1041 Williamson, D. L., and Rasch, P. J.: Two-dimensional semi-Lagrangian transport with  
1042 shapepreserving interpolation, Mon. Wea. Rev., 117, 102–129, 1989.

1043 Wofsy, S. C. and the HIPPO team: HIAPER Pole-to-Pole Observations (HIPPO): fine-grained,  
1044 global-scale measurements of climatically important atmospheric gases and aerosols,  
1045 Philo T. R. Soc. A, 369, 2073–86, doi:10.1098/rsta.2010.0313, 2011.

1046 Wu, T., Song, L., Li, W., Wang, Z., Zhang, H., Xin, X., Zhang, Y., Zhang, L., Li, J., Wu, F.,  
1047 Liu, Y., Zhang, F., Shi, X., Chu, M., Zhang, J., Fang, Y., Wang, F., Lu, Y., Liu, X., Wei,  
1048 M., Liu, Q., Zhou, W., Dong, M., Zhao, Q., Ji, J., Li, L., Zhou, M.: An overview of BCC  
1049 climate system model development and application for climate change studies. J. Meteor.

1050 Res., 28(1), 34-56, 2014.

1051 Wu, T., Li, W., Ji, J., Xin, X., Li, L., Wang, Z, Zhang, Y., Li, J., Zhang, F., Wei, M., Shi, X.,  
1052 Wu, F., Zhang, L., Chu, M., Jie, W., Liu, Y., Wang, F., Liu, X., Li, Q., Dong, M., Liang,  
1053 X., Gao, Y., Zhang, J.: Global carbon budgets simulated by the Beijing climate center  
1054 climate system model for the last century. *J Geophys Res Atmos*, 118, 4326-4347. doi:  
1055 10.1002/jgrd.50320, 2013.

1056 Wu, T., Lu, Y., Fang, Y., Xin, X., Li, L., Li, W., Jie, W., Zhang, J., Liu, Y., Zhang, L., Zhang,  
1057 F., Zhang, Y., Wu, F., Li, J., Chu, M., Wang, Z., Shi, X., Liu, X., Wei, M., Huang, A.,  
1058 Zhang, Y., and Liu, X.: The Beijing Climate Center Climate System Model (BCC-CSM):  
1059 the main progress from CMIP5 to CMIP6, *Geos.Model Dev.*, 12, 1573-1600,  
1060 <http://doi.org/10.5194/gmd-12-1573-2019>, 2019.

1061 [Wu, Tongwen, Zhang, Fang, Zhang, Jie, et al.: Model code and data for Wu et al, "Beijing](http://doi.org/10.5281/zenodo.3609337)  
1062 [Climate Center Earth System Model version 1 \(BCC-ESM1\): Model Description and](http://doi.org/10.5281/zenodo.3609337)  
1063 [Evaluation of Aerosol Simulations", GMD publication.](http://doi.org/10.5281/zenodo.3609337)  
1064 [http://doi.org/10.5281/zenodo.3609337, 2020.](http://doi.org/10.5281/zenodo.3609337)

1065 Young, P. J., Archibald, A. T., Bowman, K. W., Lamarque, J. F., Naik, V., Stevenson, D. S.,  
1066 Tilmes, S., Voulgarakis, A., Wild, O., Bergmann, D., Cameron-Smith, P., Cionni, I.,  
1067 Collins, W. J., Dalsøren, S. B., Doherty, R. M., Eyring, V., Faluvegi, G., Horowitz, L.  
1068 W., Josse, B., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Plummer, D. A., Righi, M.,  
1069 Rumbold, S. T., Skeie, R. B., Shindell, D. T., Strode, S. A., Sudo, K., Szopa, S., and  
1070 Zeng, G.: Pre-industrial to end 21st century projections of tropospheric ozone from the  
1071 Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), *Atmos.*  
1072 *Chem. Phys.*, 13, 2063-2090, <http://doi.org/10.5194/acp-13-2063-2013>, 2013.

1073 Young, P. J., Naik, V., Fiore, A. M., Gaudel, A., Guo, J., Lin, M. Y., Neu, J. L., Parrish, D. D.,  
1074 Rieder, H. E., Schnell, J. L., Tilmes, S., Wild, O., Zhang, L., Ziemke, J. R., Brandt, J.,  
1075 Delcloo, A., Doherty, R. M., Geels, C., Hegglin, M. I., Hu, L., Im, U., Kumar, R., Luhar,  
1076 A., Murray, L., Plummer, D., Rodriguez, J., Saiz-Lopez, A., Schultz, M. G., Woodhouse,  
1077 M. T., and Zeng, G.: Tropospheric Ozone Assessment Report: Assessment of  
1078 global-scale model performance for global and regional ozone distributions, variability,  
1079 and trends, *Elem Sci Anth*, 6, 10, <http://doi.org/10.1525/elementa.265>, 2018.

1080 Zender, C., Bian, H., and Newman, D.: Mineral Dust Entrainment and Deposition (DEAD)  
1081 model: Description and 1990s dust climatology, *J. Geophys. Res.*, 108(D14), 4416, doi:

1082 10.1029/2002JD002775, 2003.  
1083 Zhang, J., Wu, T., Shi, X., Zhang F., Li, J., Chu, M., Liu, Q., Yan, J., Ma, Q., Wei, M.: BCC  
1084 BCC-ESM1 model output prepared for CMIP6 AerChemMIP. Version  
1085 YYYYMMDD[1], Earth System Grid Federation,  
1086 <https://doi.org/10.22033/ESGF/CMIP6.1733>, 2019.  
1087

1088 Table 1. Chemical species considered in BCC-AGCM3-Chem. Species marked with star (\*)  
 1089 denote those added in BCC-ESM1 apart from the 63 species used in MOZART2. In the  
 1090 column of surface emission, interactive surface emissions are considered for sea salt and dust.  
 1091

Species	Dry deposition	Wet deposition	Surface emission	Aircraft emission	Volcanic emission
O <sub>3</sub>	✓				
N <sub>2</sub> O			✓		
N					
NO	✓		✓	✓	
NO <sub>2</sub>	✓				
NO <sub>3</sub>					
HNO <sub>3</sub>	✓	✓			
HO <sub>2</sub> NO <sub>2</sub>	✓	✓			
N <sub>2</sub> O <sub>5</sub>					
CH <sub>4</sub>	✓		✓	✓	
CH <sub>3</sub> O <sub>2</sub>					
CH <sub>3</sub> OOH	✓	✓			
CH <sub>2</sub> O	✓	✓	✓		
CO	✓		✓	✓	
OH					
HO <sub>2</sub>					
H <sub>2</sub> O <sub>2</sub>	✓	✓			
C <sub>3</sub> H <sub>6</sub>			✓		
ISOP			✓		
Gas tracers					
PO <sub>2</sub>					
CH <sub>3</sub> CHO	✓	✓	✓		
POOH	✓	✓			
CH <sub>3</sub> CO <sub>3</sub>					
CH <sub>3</sub> COOOH	✓	✓			
PAN	✓				
ONIT	✓	✓			
C <sub>2</sub> H <sub>6</sub>			✓		
C <sub>2</sub> H <sub>4</sub>			✓		
C <sub>4</sub> H <sub>10</sub>			✓		
MPAN	✓				
ISOPO <sub>2</sub>					
MVK		✓			
MACR		✓			
MACRO <sub>2</sub>					
MACROOH	✓	✓			
MCO <sub>3</sub>					
C <sub>2</sub> H <sub>5</sub> O <sub>2</sub>					
C <sub>2</sub> H <sub>5</sub> OOH	✓	✓			
C <sub>10</sub> H <sub>16</sub>			✓		

1092

1093

Species name	Dry deposition	Wet deposition	Surface emission	Aircraft emission	Volcanic emission
C <sub>3</sub> H <sub>8</sub>			✓		
C <sub>3</sub> H <sub>7</sub> O <sub>2</sub>					
C <sub>3</sub> H <sub>7</sub> OOH	✓	✓			
CH <sub>3</sub> COCH <sub>3</sub>	✓		✓		
ROOH		✓			
CH <sub>3</sub> OH	✓	✓	✓		
C <sub>2</sub> H <sub>5</sub> OH	✓	✓	✓		
GLYALD	✓	✓			
HYAC	✓	✓			
EO <sub>2</sub>					
EO					
HYDRALD	✓	✓			
RO <sub>2</sub>					
CH <sub>3</sub> COCHO	✓	✓	✓		
Rn-222					
Pb-210	✓	✓			
ISOPNO <sub>3</sub>		✓			
ONITR	✓	✓			
XO <sub>2</sub>					
XOOH	✓	✓			
ISOPOOH	✓	✓			
H <sub>2</sub>	✓		✓		
Stratospheric O <sub>3</sub>	✓				
Inert O <sub>3</sub>	✓				
SO <sub>2</sub> *	✓	✓	✓	✓	✓
DMS*			✓		
NH <sub>3</sub> *			✓	✓	
SO <sub>4</sub> <sup>2-</sup> *	✓	✓			
OC1*	✓	✓	✓	✓	
OC2*	✓	✓	✓	✓	
BC1*	✓	✓	✓	✓	
BC2*	✓	✓	✓	✓	
SSLT01*	✓	✓			
SSLT02*	✓	✓			
SSLT03*	✓	✓			
SSLT04*	✓	✓			
DST01*	✓	✓			
DST02*	✓	✓			
DST03*	✓	✓			
DST04*	✓	✓			



1097 Table 2. Gas-phase chemical reactions for NH<sub>3</sub> and bulk aerosols precursors following  
 1098 CAM-Chem (Lamarque et al., 2012). The reaction rates (s<sup>-1</sup>) refer to Tie et al. (2001) and  
 1099 Sander et al. (2003), and Cooke and Wilson (1996). Temperature (T) is expressed in K, air  
 1100 density (M) in molecule cm<sup>-3</sup>, ki and ko in cm<sup>3</sup> molecule<sup>-1</sup> s<sup>-1</sup>.  
 1101

Chemical reactions	Rate
NH <sub>3</sub> + OH → H <sub>2</sub> O	1.70E-12*exp(-710/T)
SO <sub>2</sub> + OH → SO <sub>4</sub> <sup>2-</sup>	ko/(1.+ko*M/ki)*f**(1./(1.+log10(ko*M/ki))), in which ko=3.0E-31*(300/T)**3.3; ki=1.E-12; f=0.6
DMS + OH → SO <sub>2</sub>	9.60E-12*exp(-234./T)
DMS + OH → .5*SO <sub>2</sub> + .5*HO <sub>2</sub>	1.7E-42*exp(7810/T)*M*0.21/(1+5.5E-31*exp(7460/T)* M* 0.21)
DMS + NO <sub>3</sub> → SO <sub>2</sub> + HNO <sub>3</sub>	1.90E-13*exp( 520/T)
BC1 → BC2	7.10E-06
OC1 → OC2	7.10E-06

1102  
 1103

1104

1105

1106

Table 3. Size and density parameters of bulk aerosols.

Aerosols	Species Name	Mean radius ( $\mu\text{m}$ ) / bin size ( $\mu\text{m}$ )	Geometric standard deviation ( $\mu\text{m}$ )	Density ( $\text{g cm}^{-3}$ )
$\text{SO}_4^{2-}$	Sulfate	0.05	2.03	1.77
BC1	hydrophobic black carbon	0.02	2.00	1.0
BC2	hydrophilic black carbon	0.02	2.00	1.0
OC1	hydrophobic organic carbon	0.03	2.24	1.8
OC2	hydrophilic organic carbon	0.03	2.24	1.8
DST01	Dust	0.55 / bin: 0.1-1.0	2.00	2.5
DST02	Dust	1.75 / bin: 1.0-2.5	2.00	2.5
DST03	Dust	3.75 / bin: 2.5-5.0	2.00	2.5
DST04	Dust	7.50 / bin: 5.0-10.	2.00	2.5
SSLT01	Sea salt	0.52 / bin: 0.2-1.0	2.00	2.2
SSLT02	Sea salt	2.38 / bin: 1.0-3.0	2.00	2.2
SSLT03	Sea salt	4.86 / bin: 3.0-10.	2.00	2.2
SSLT04	Sea salt	15.14 / bin: 10.-20.	2.00	2.2

1107

Table 4. Source of emission data. MOZART2 data denote the standard tropospheric chemistry package for MOZART contains surface emissions from the EDGAR 2.0 data base (Olivier et al., 1996). ACCMIP data are downloaded from the IPCC ACCMIP emission inventory (<http://accent.aero.jussieu.fr/ACCMIP.php>) and they vary from 1850 to 2000, in 10-year steps (Lamarque et al., 2010). CMIP6 data are from <https://esgf-node.llnl.gov/search/input4mips/>. Anthropogenic emission includes Industrial and fossil fuel use, agriculture, ships, and etc. Biomass burning includes vegetation fires incl. fuel wood and agricultural burning.

Species	Anthropogenic emission	Biomass burning	Biogenic emissions from vegetation	Biogenic emissions from soil	Oceanic emissions	Airplane emission	Volcanic emission
C <sub>2</sub> H <sub>4</sub>	CMIP6	CMIP6	On-line computation		MOZART2		
C <sub>2</sub> H <sub>5</sub> OH	CMIP6	CMIP6					
C <sub>2</sub> H <sub>6</sub>	CMIP6	CMIP6	ACCMIP		MOZART2		
C <sub>3</sub> H <sub>6</sub>	CMIP6	CMIP6	On-line computation		MOZART2		
C <sub>3</sub> H <sub>8</sub>	CMIP6	CMIP6	ACCMIP		MOZART2		
C <sub>4</sub> H <sub>10</sub>	CMIP6	CMIP5	MOZART2		MOZART2		
CH <sub>2</sub> O	CMIP6	CMIP6					
CH <sub>3</sub> CHO	ACCMIP	CMIP6					
CH <sub>3</sub> COCHO		CMIP6					
CH <sub>3</sub> OH	ACCMIP	CMIP6	ACCMIP				
CH <sub>3</sub> COCH <sub>3</sub>	ACCMIP	ACCMIP	On-line computation		MOZART2		
ISOP		CMIP5	On-line computation				
C <sub>10</sub> H <sub>16</sub>		CMIP6	On-line computation				
CH <sub>4</sub>	CMIP6	CMIP6	MOZART2		MOZART2	CMIP6	
CO	CMIP6	CMIP6	ACCMIP	MOZART2	ACCMIP	CMIP6	
H <sub>2</sub>	MOZART2	CMIP6		MOZART2	MOZART2		
N <sub>2</sub> O	MOZART2	CMIP6		MOZART2	MOZART2		
NH <sub>3</sub>	CMIP6	CMIP6		ACCMIP	ACCMIP	CMIP6	
NO	CMIP6	CMIP6		ACCMIP		CMIP6	
SO <sub>2</sub>	CMIP6	CMIP6				CMIP6	ACCMIP
DMS					ACCMIP		
OC1	CMIP6	CMIP6				CMIP6	
OC2	CMIP6	CMIP6	On-line computation			CMIP6	
BC1	CMIP6	CMIP6				CMIP6	
BC2	CMIP6	CMIP6				CMIP6	

Table 5. Global budgets for DMS, SO<sub>2</sub>, and sulfate in the period of 1991 to 2000. Units are sources and sinks, Tg(S) yr<sup>-1</sup>; burden, Tg; lifetime, days.

		BCC-ESM (1991-2000 mean)	Other studies and CMIP5 data
DMS	Sources	27.4	
	Emission	27.4	10.7-23.7 <sup>a</sup>
	Sinks	28.0	
	Gas-phase oxidation	28.0	
	Burden	0.12	0.04-0.29 <sup>a</sup>
	Lifetime	0.78	0.5-3.0 <sup>a</sup>
SO <sub>2</sub>	Sources	76.93	
	Emission at surface	63.63	
	Emission from airplane	0.10	
	DMS oxidation	13.20	10.0-24.7 <sup>a</sup>
	Sinks	76.96	
	Dry deposition	18.53	16.0-55.0 <sup>a</sup>
	Wet deposition	9.36	0.0-19.9 <sup>a</sup>
	Gas-phase oxidation	10.33	6.1-16.8 <sup>a</sup>
	Aqueous-phase oxidation	38.74	24.5-57.8 <sup>a</sup>
Burden	0.48	0.40-1.22 <sup>a</sup>	
Lifetime	1.12	0.6-2.6 <sup>a</sup>	
SO <sub>4</sub> <sup>2-</sup>	Sources	49.05	59.67 ± 13.13 <sup>b</sup>
	Emission	0.00	
	SO <sub>2</sub> aqueous-phase oxidation	38.73	
	SO <sub>2</sub> gas-phase oxidation	10.32	
	Sinks	49.06	
	Dry deposition	2.20	4.96-5.51 <sup>d</sup>
	Wet deposition	46.86	39.34-40.20 <sup>d</sup>
	Burden	1.89	1.98 ± 0.48 <sup>b</sup> , 1.71 <sup>c</sup> , 1.2 <sup>g</sup> , 2.22-2.43 <sup>h</sup>
	Lifetime	4.69	4.12 ± 0.74 <sup>b</sup> , 3.72-3.77 <sup>d</sup> 3.3 <sup>g</sup> , 3.7-4.0 <sup>h</sup>

Notes: References denote a for Liu et al. (2005), b for Textor et al. (2006), c for the values derived from CMIP5 prescribed aerosol masses averaged from 1991 to 2000, d for Liu et al. (2012), g for Matsui and Mahowald (2017), and h for Tegen et al. (2019). Values of DMS, SO<sub>2</sub>, and sulfate burdens in the literature d are transferred from TgS to Tg (species) for units consistence.

Table 6. Same as Table 5, but for global budgets for black carbon, organic carbon, dust, and sea salts. Units are sources and sinks, Tg yr<sup>-1</sup>; burden, Tg; lifetime, days.

		BCC-ESM (1991-2000 mean)	Other studies and CMIP5 data
BC	Sources	7.22	
	Emission	7.22	11.9 ± 2.7 <sup>b</sup> , 7.8 <sup>g</sup>
	Sinks	7.24	7.75 <sup>d</sup> , 7.8 <sup>g</sup>
	Dry deposition	0.90	0.27 <sup>g</sup> , 1.30-1.64 <sup>e</sup>
	Wet deposition	6.34	7.5 <sup>g</sup> , 6.10-6.45 <sup>e</sup>
	Burden	0.13	0.114 <sup>c</sup> , 0.24 ± 0.1 <sup>b</sup> , 0.11 <sup>g</sup> , 0.14-0.26 <sup>h</sup> , 0.084-0.123 <sup>e</sup>
	Lifetime	6.60	7.12 ± 2.35 <sup>b</sup> , 3.95-4.80 <sup>e</sup> , 5.0 <sup>g</sup> , 6.3-7.5 <sup>h</sup>
OC	Sources	32.29	
	Fossil and biofuel emission	13.91	
	Natural emission	18.38	
	Sinks	32.30	
	Dry deposition	2.44	
	Wet deposition	29.86	
	Burden	0.62	0.69 <sup>c</sup> , 1.7 ± 0.45 <sup>b</sup> , 1.0-2.2 <sup>h</sup>
Lifetime	5.00	6.54 ± 1.76 <sup>b</sup> , 4.56-4.90 <sup>d</sup> , 6.4 <sup>g</sup> , 5.4-6.6 <sup>h</sup>	
Dust	Sources	2592.0	1840 <sup>b</sup> , 2943.5-3121.9 <sup>d</sup> , 2677 <sup>g</sup>
	Sinks	2592.0	
	Dry deposition	1630.8	1444 <sup>g</sup>
	Wet deposition	961.2	1245 <sup>g</sup>
	Burden	22.93	20.41 <sup>c</sup> , 22.424.7 <sup>d</sup> , 35.9 <sup>f</sup> , 19.2 ± 7.68 <sup>b</sup> , 28.5 <sup>g</sup> , 16.5-17.9 <sup>h</sup>
	Lifetime	3.23	4.14 ± 1.78 <sup>b</sup> , 2.61-3.07 <sup>d</sup> , 3.9 <sup>g</sup> , 5.3-5.7 <sup>h</sup>
Sea Salt	Sources	4667.2	4965.5-5004.1 <sup>d</sup> , 5039 <sup>g</sup>
	Sinks	4667.4	
	Dry deposition	2978.5	2158 <sup>g</sup>
	Wet deposition	1688.9	2918 <sup>g</sup>
	Burden	11.89	7.58-10.37 <sup>a</sup> , 6.4 ± 3.4 <sup>b</sup> , 11.84 <sup>c</sup> , 13.6 <sup>g</sup> , 3.9 <sup>h</sup>
	Lifetime	0.93	0.41 ± 0.24 <sup>b</sup> , 0.55-0.76 <sup>d</sup> , 0.98 <sup>g</sup> , 1.2-1.3 <sup>h</sup>

Notes: References denote a for Liu et al. (2005), b for Textor et al. (2006), c derived from CMIP5 prescribed aerosol masses averaged from 1991 to 2000, d for Liu et al. (2012), e for Liu et al. (2016), f for Ginoux (2001), g for Matsui and Mahowald (2017), and h for Tegen et al. (2019).

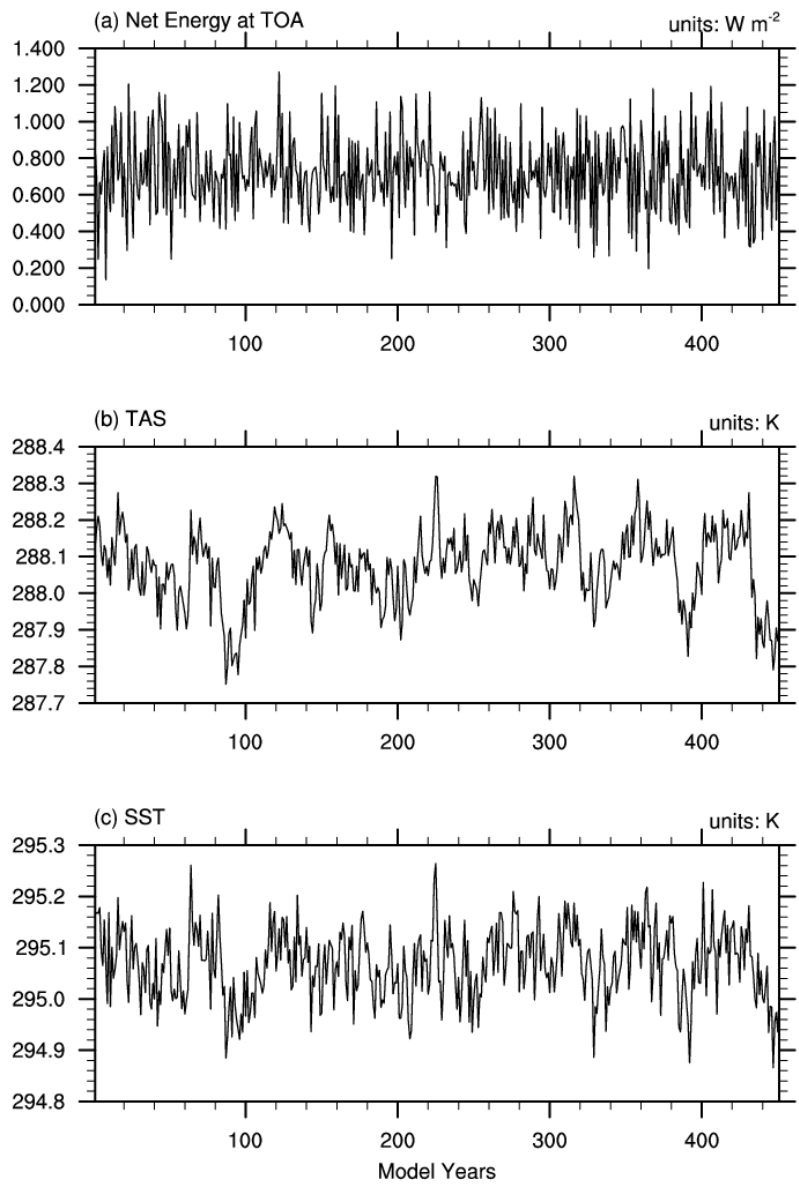


Figure 1. The time series of global and annual mean of (a) net energy budget at top of atmosphere ( $W m^{-2}$ ), (b) near-surface air temperature (K), and (c) sea surface temperature (K) in the last 450 years of the piControl simulation.

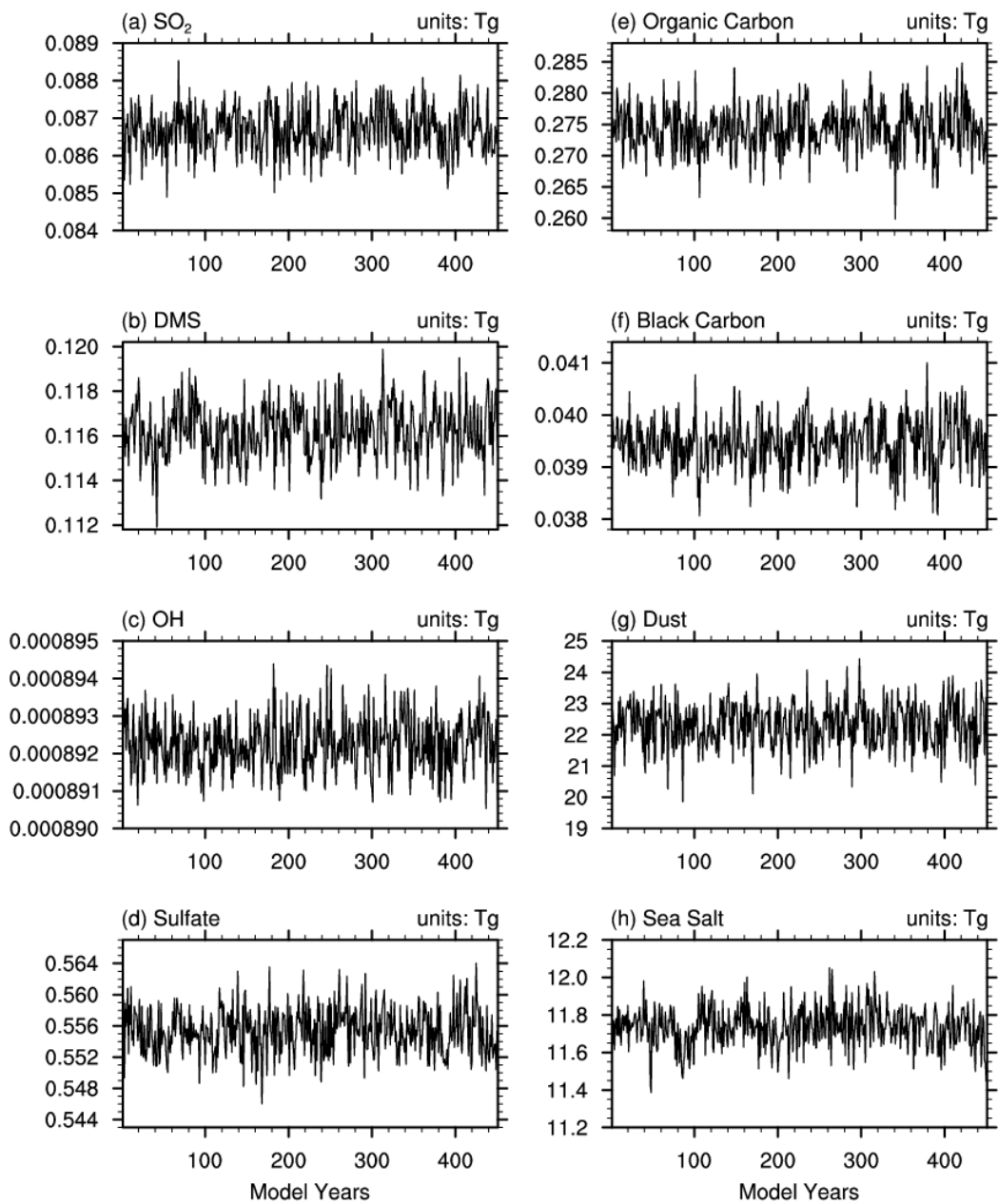


Figure 2. Same as in Figure 1, but for the global burdens of (a)  $\text{SO}_2$ , (b) DMS, (c) OH, and (d-h) different aerosols in the troposphere (below 100 hPa). Units are Tg.

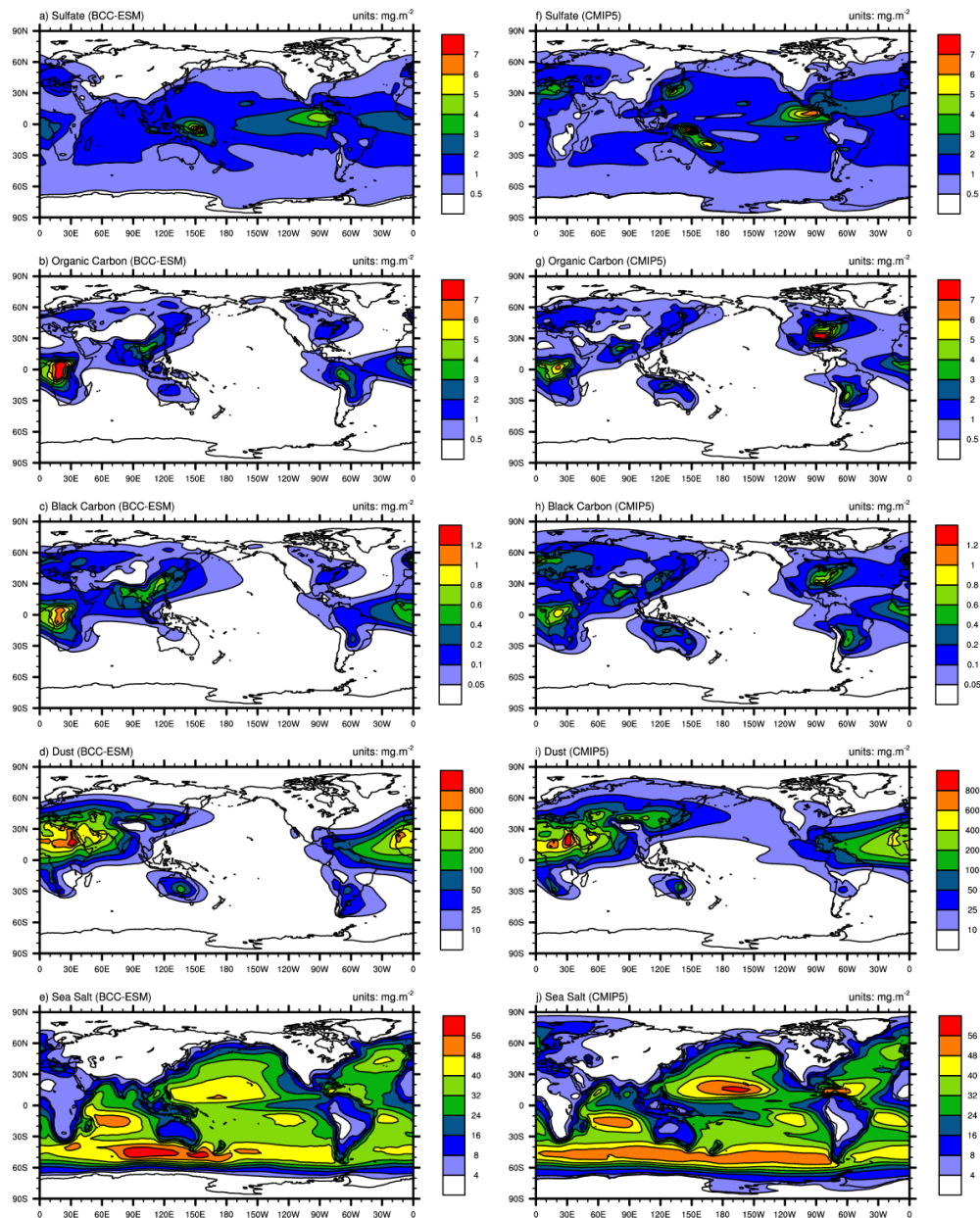


Figure 3. Global distributions of annual mean mass burdens of sulfate ( $\text{SO}_4^{2-}$ ; first row), organic carbon (OC; second row), black carbon (BC; third row), dust (fourth row), and sea salt (fifth row) aerosols in the whole atmospheric column. The left panels show the mean averaged for the last 100 years of BCC-ESM pre-industrial piControl simulations, and the right panels show the CMIP5 recommended aerosol concentrations in year 1850 (the website at IASA <http://tntcat.iiasa.ac.at/RcpDb/>). Units:  $\text{mg m}^{-2}$ .



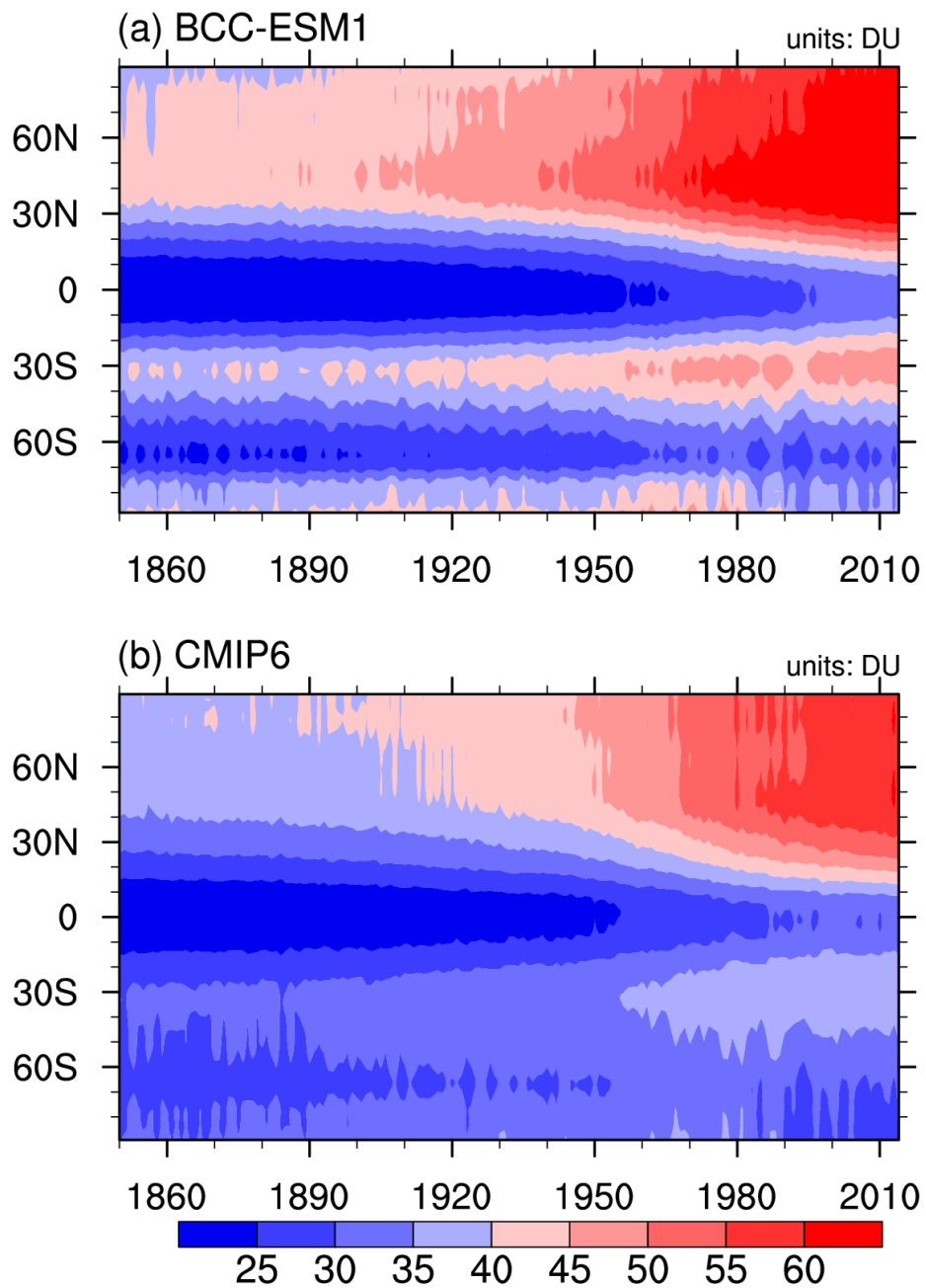


Figure 4. Zonal mean of yearly mean concentration of ozone column in the troposphere below 300 hPa to the ground from 1871 to 1999 for (a) BCC-ESM1 and (b) CMIP6 data. Unit: DU.

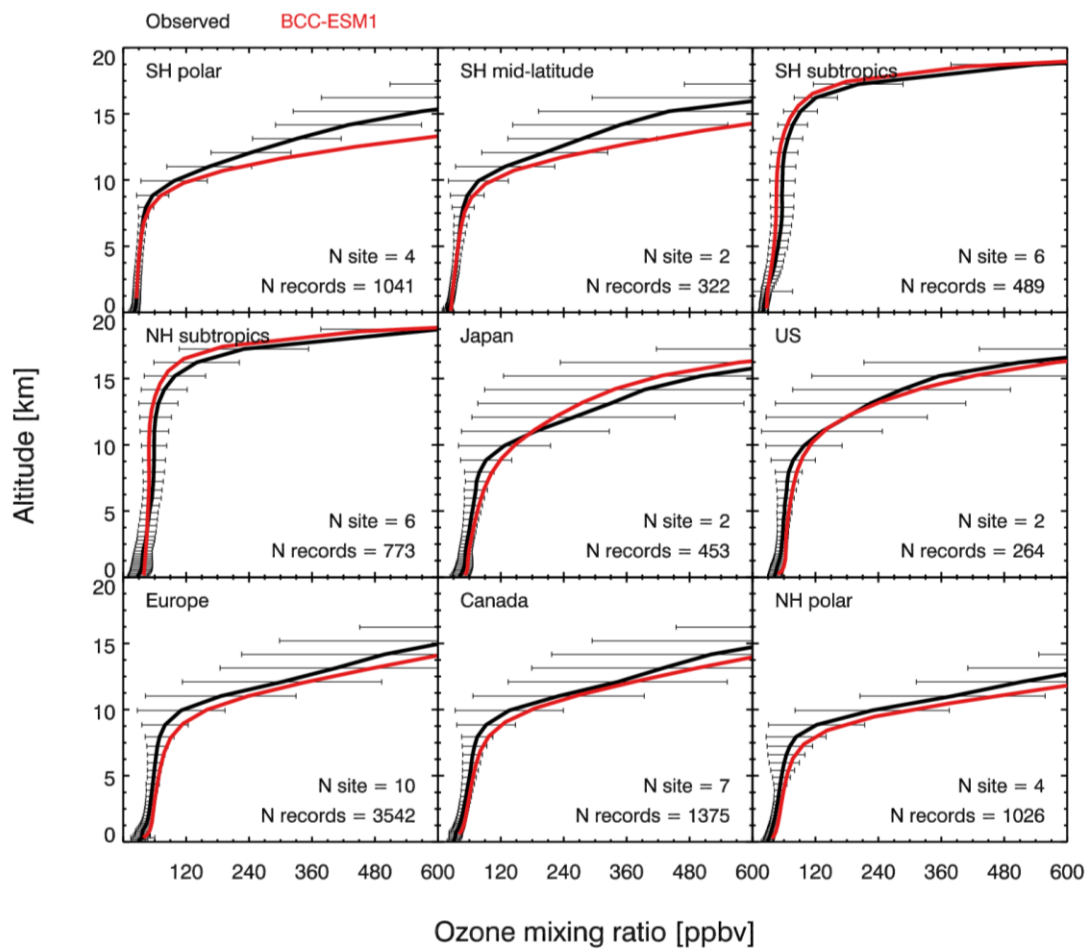


Figure 5. Vertical profiles of annual mean ozone concentrations from observations averaged for 2010-2014 in nine regions (black) and from the BCC-ESM1 simulations (red). The observations are derived from 41 global WOUDC sites.

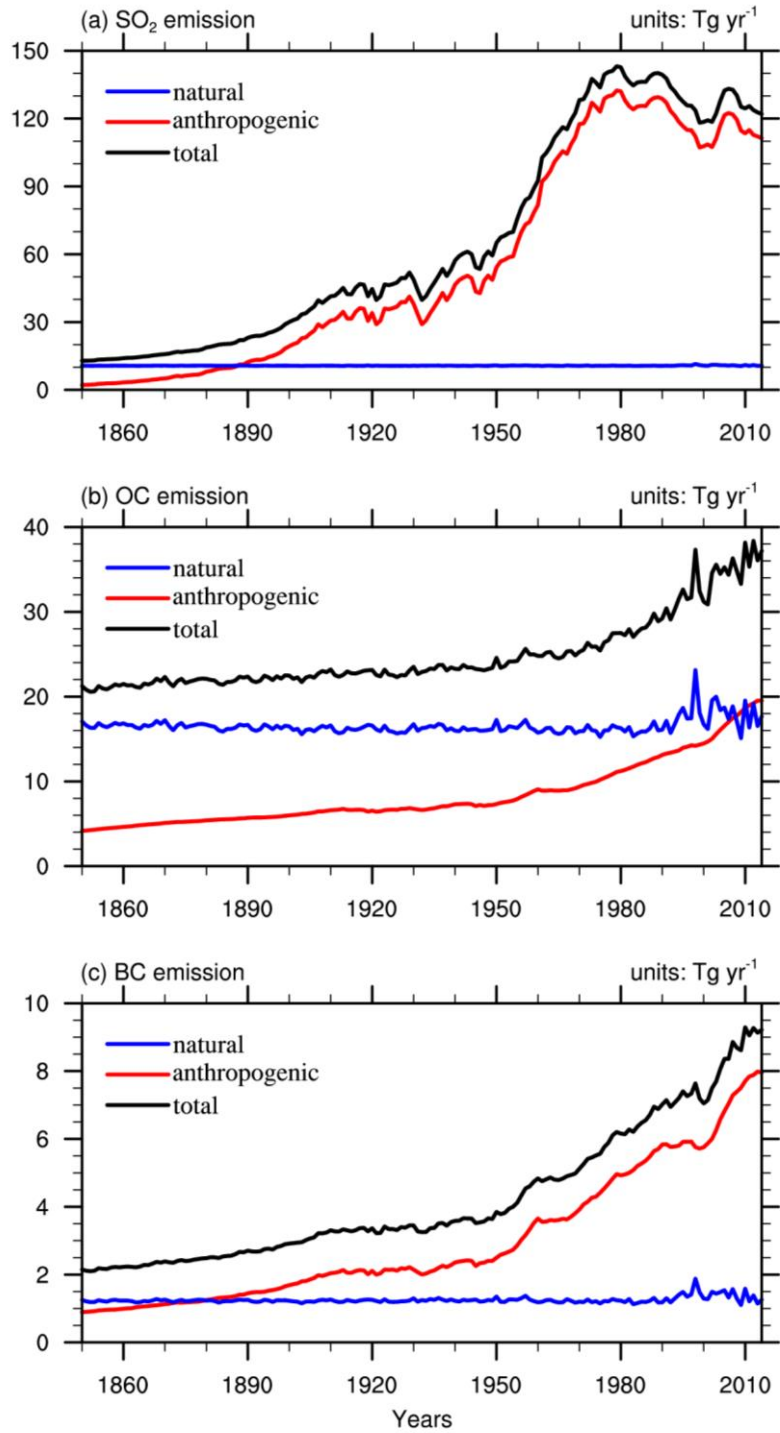


Figure 6. Global annual anthropogenic, natural, and total emissions of SO<sub>2</sub>, organic carbon (OC), and black carbon (BC) in the BCC-ESM1 historical simulation. All the biomass burning emissions are included in natural emissions in (a)-(c). Units: Tg yr<sup>-1</sup>.

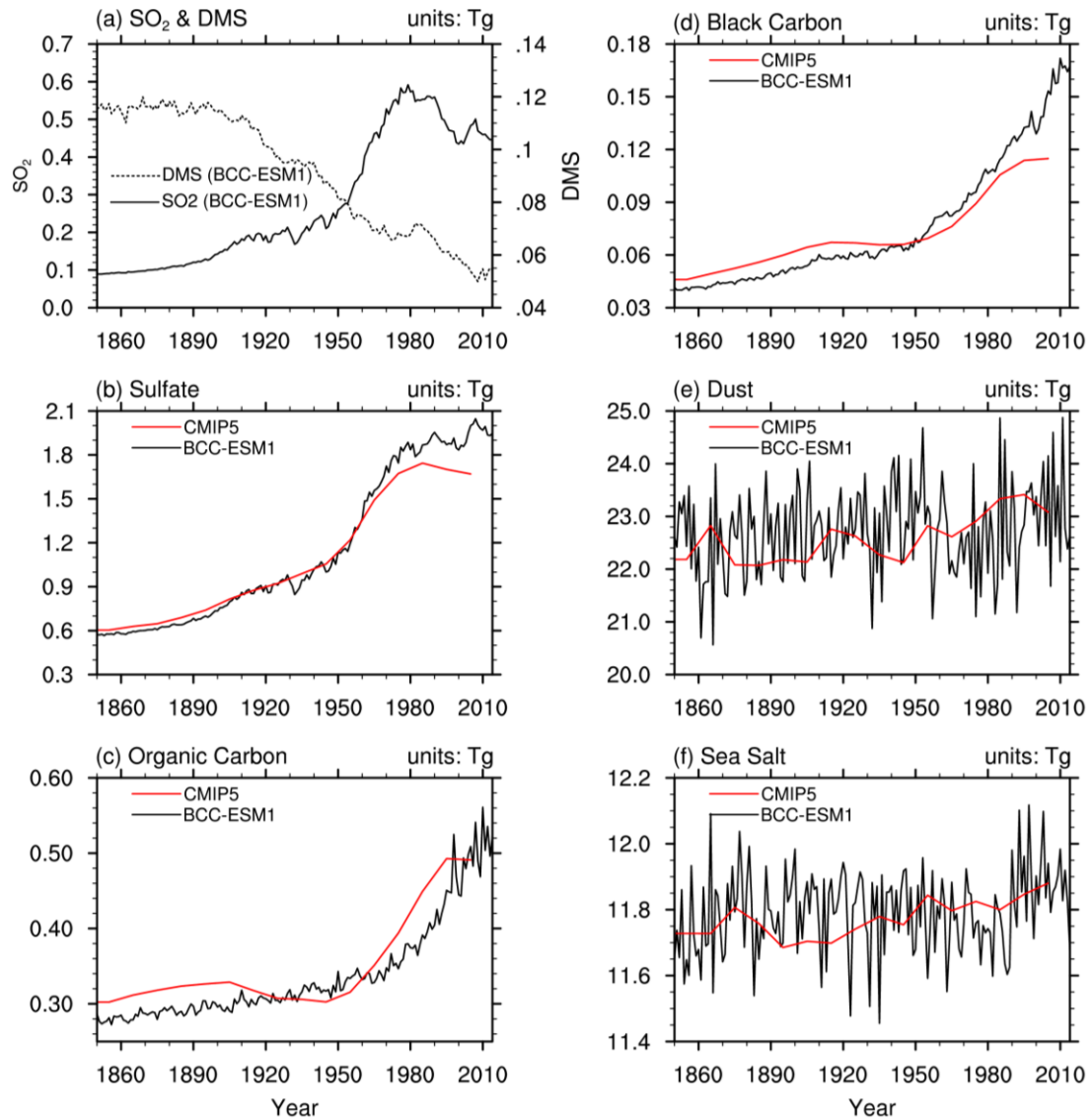


Figure 7. The time series of global yearly amounts of (a) SO<sub>2</sub> and DMS and (b-f) aerosols in the whole atmosphere column from the CMIP6 historical simulations of BCC-ESM1 (black lines) and the CMIP5-recommended aerosols masses (red lines). The yearly CMIP5 data are interpolated from the time series in 10-year interval. Units: Tg.

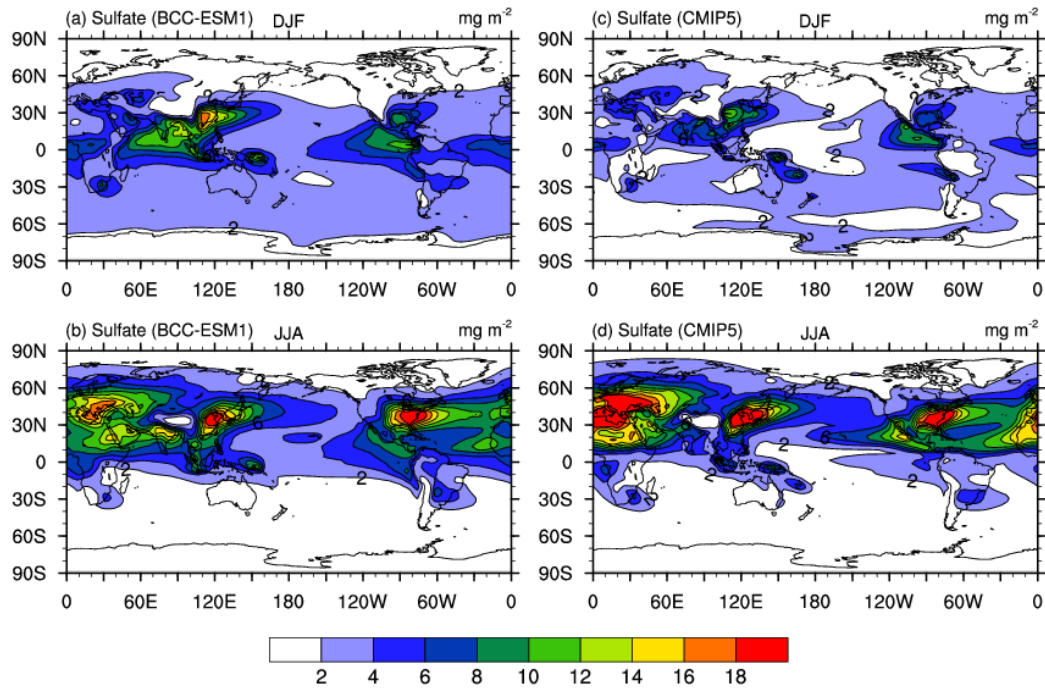


Figure 8. December-January-February (DJF; top panels) and June-July-August (JJA; bottom panels) mean sulfate ( $\text{SO}_4^{2-}$ ) aerosol column mass concentrations averaged for the period of 1971-2000. Left panels show the historical simulations of BCC-ESM1, and right panels the CMIP5-recommended data. Units:  $\text{mg.m}^{-2}$ .

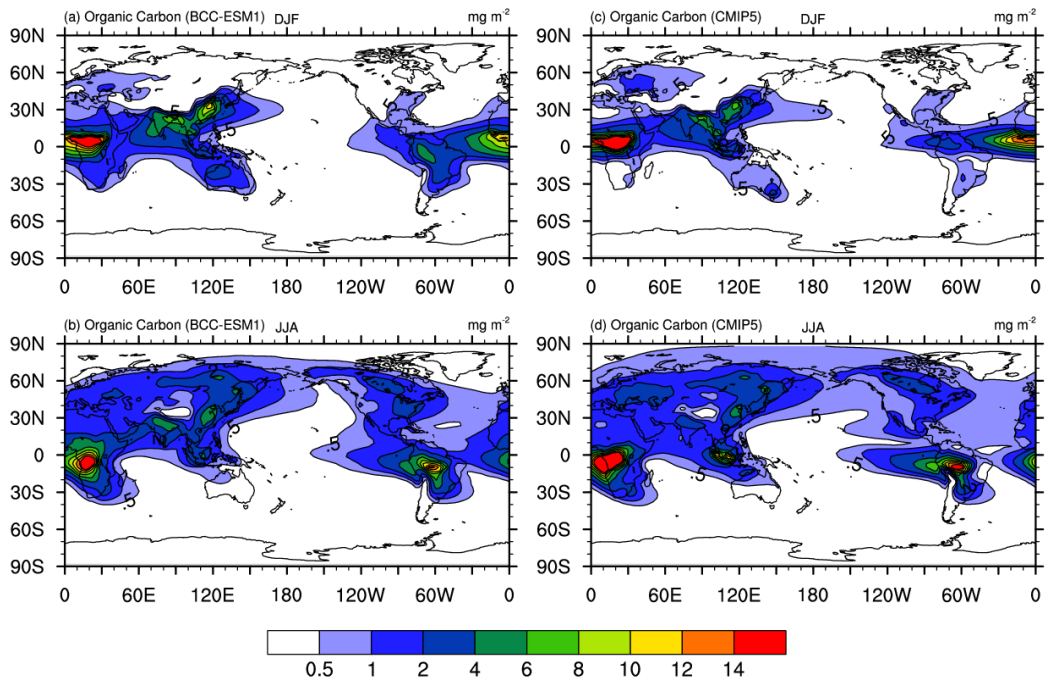


Figure 9. The same as in Figure 8, but for organic carbon (OC) aerosol column mass concentrations. Units:  $\text{mg m}^{-2}$ .

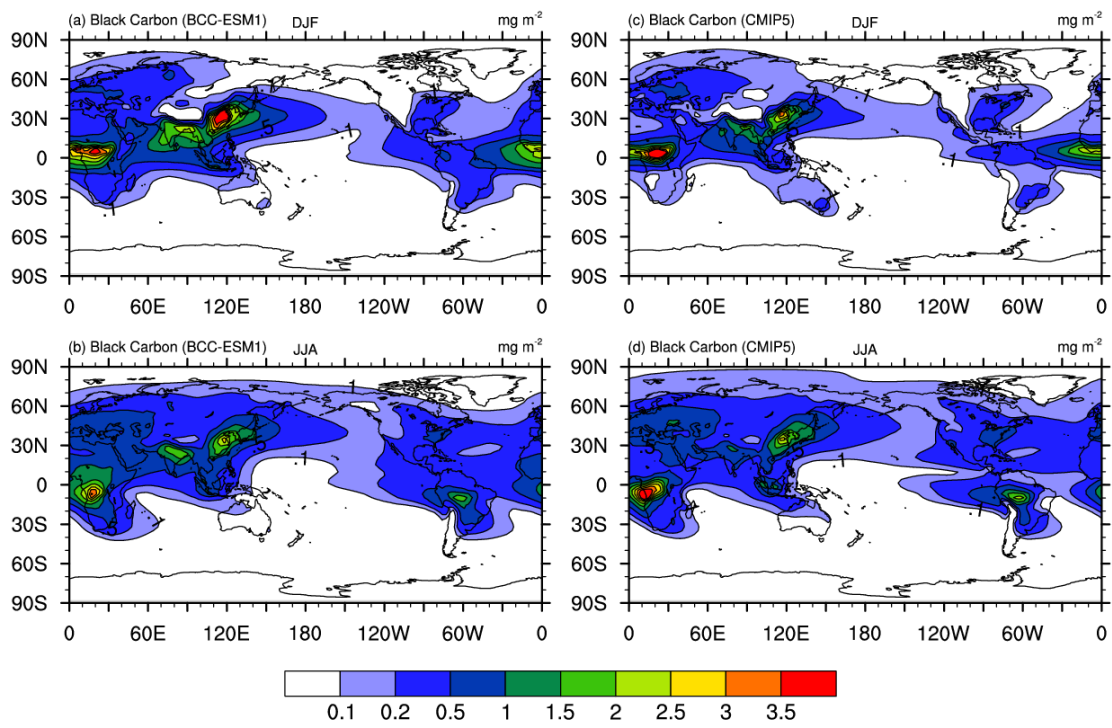


Figure 10. The same as in Figure 8, but for black carbon (BC) aerosol. Units:  $\text{mg.m}^{-2}$ .

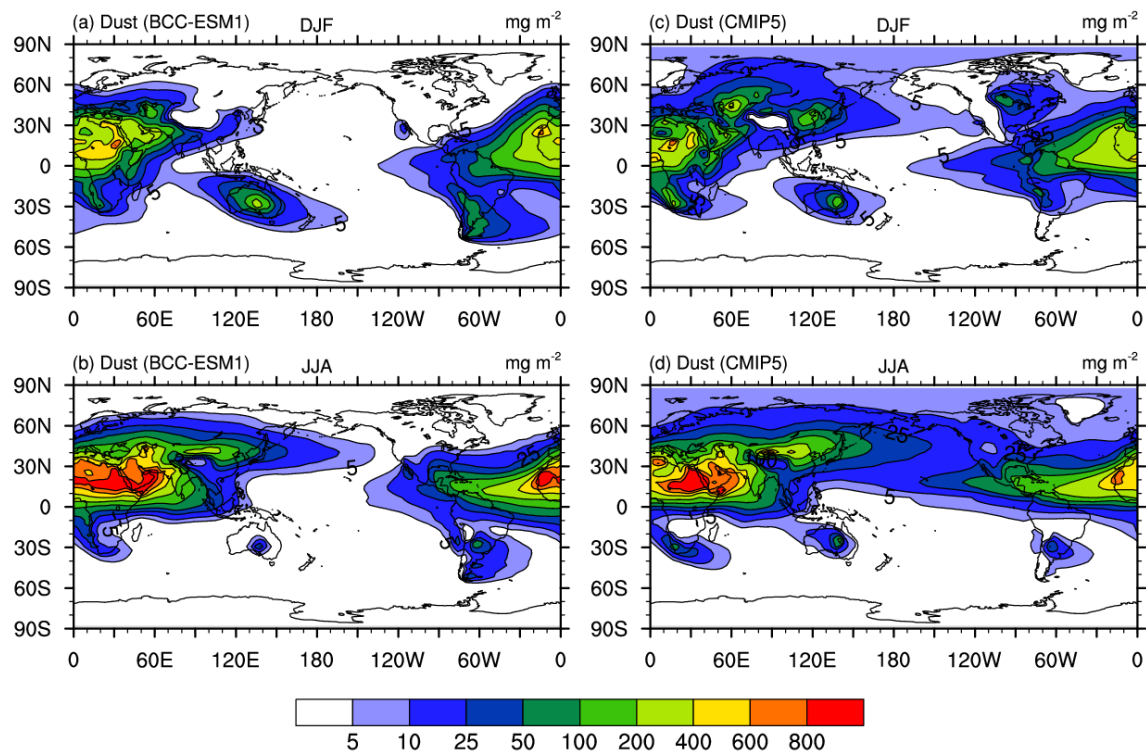


Figure 11. The same as in Figure 8, but for dust aerosol. Units:  $\text{mg.m}^{-2}$ .



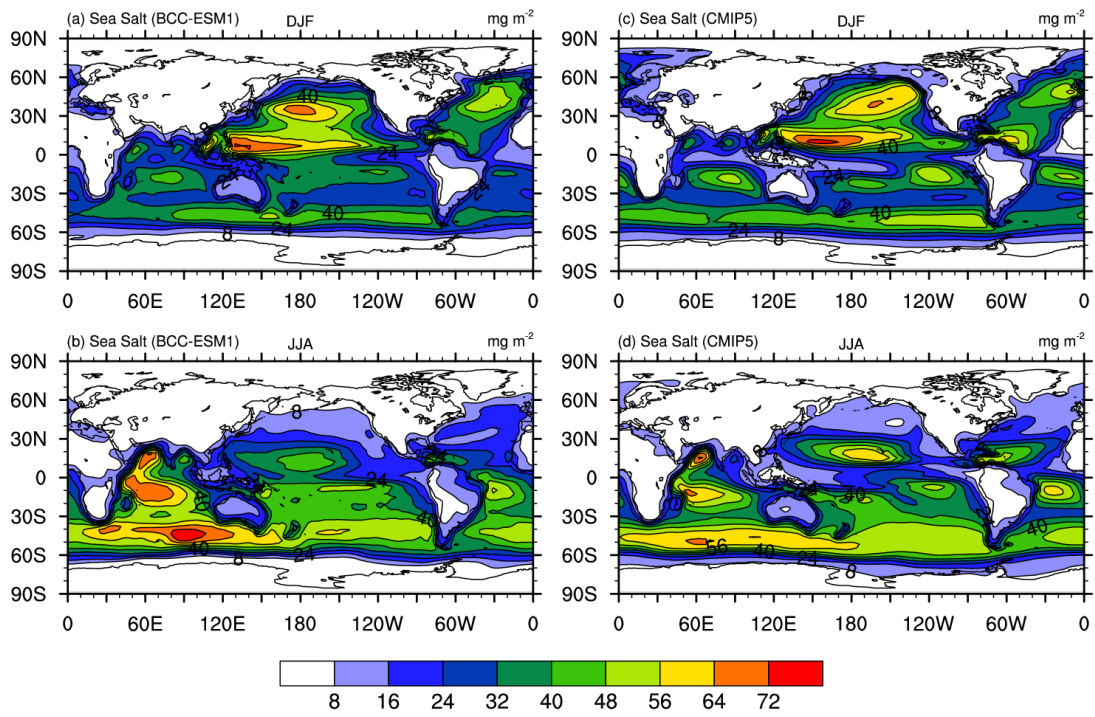


Figure 12. The same as in Figure 8, but for sea salt (SSLT) aerosol. Units:  $\text{mg.m}^{-2}$ .

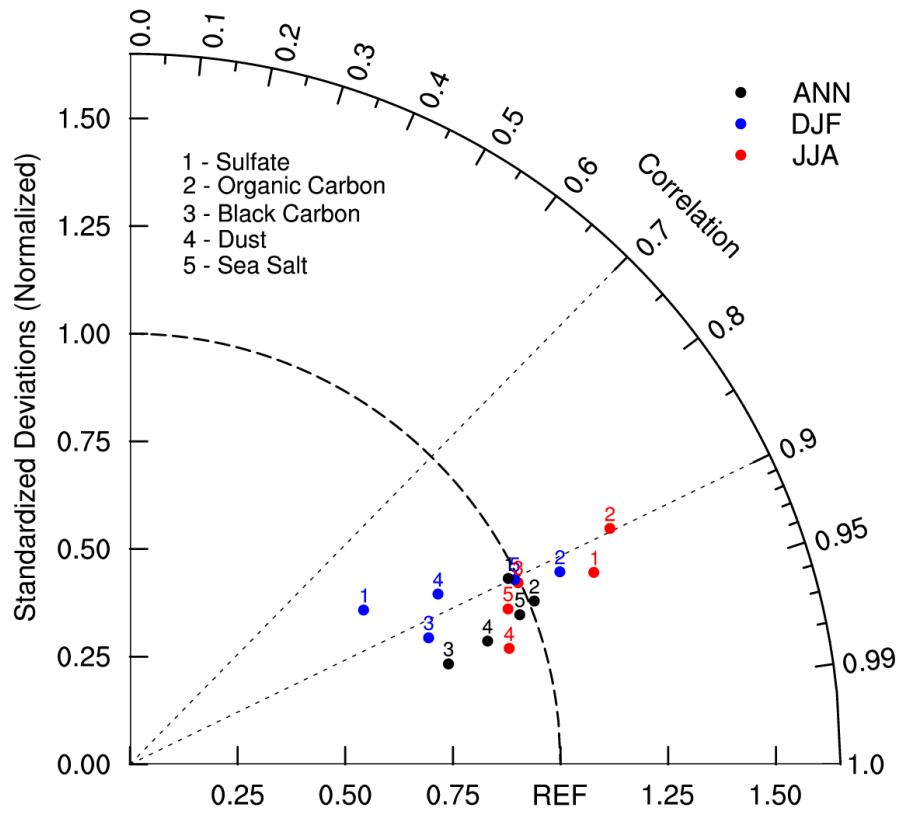


Figure 13. Taylor diagram for the global aerosols climatology (1971–2000) of sulfate, organic carbon, black carbon, dust, and sea salt averaged for December-January-February (DJF), June-July-August (JJA), and annual respectively. The radial coordinate shows the standard deviation of the spatial pattern, normalized by the observed standard deviation. The azimuthal variable shows the correlation of the modelled spatial pattern with the observed spatial pattern. Analysis is for the whole globe. The reference dataset is CMIP5-prescribed dataset.

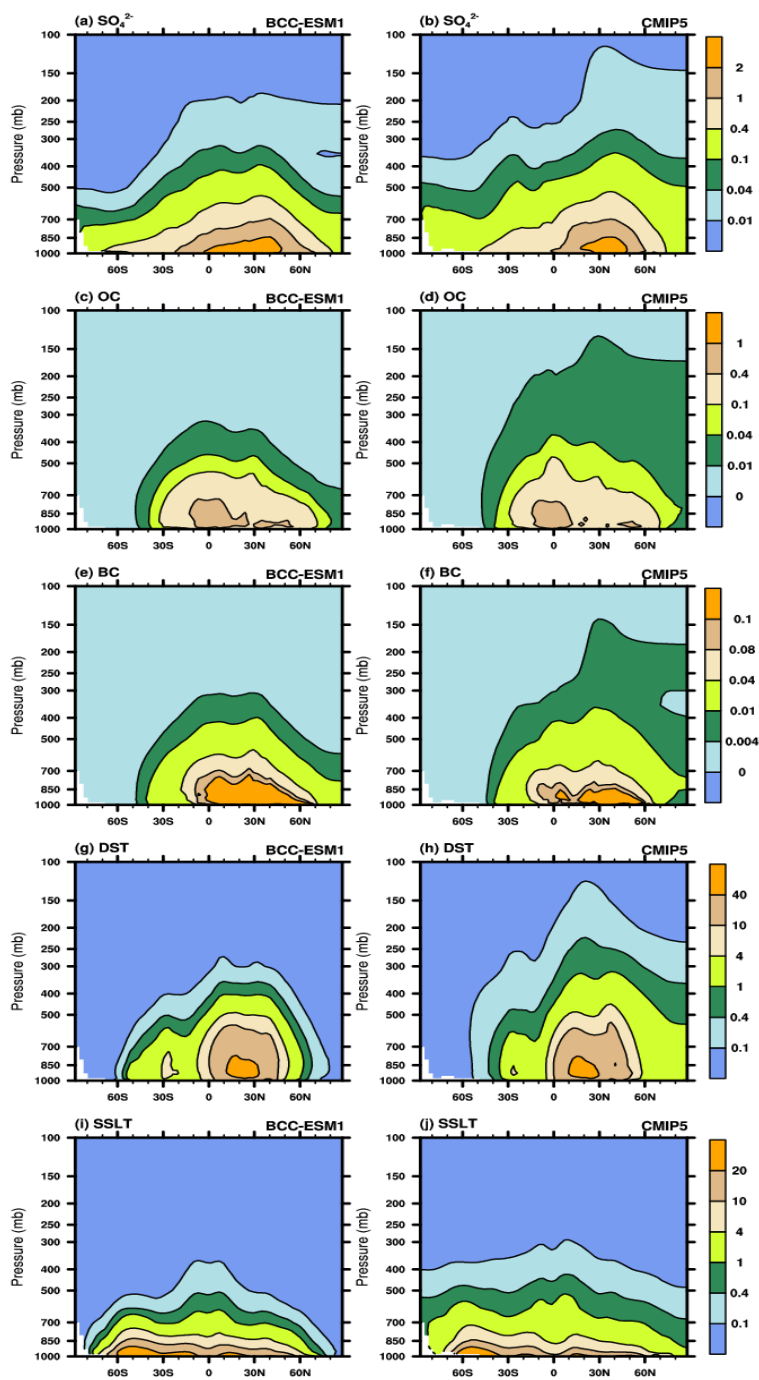


Figure 14. Latitude-pressure distributions of zonally-averaged annual mean sulfate, organic carbon, black carbon, dust, and sea salt aerosol concentrations for the period of 1971-2000. Left panels show the CMIP6 historical simulation of BCC-ESM1, and right panels the CMIP5 recommendation data. Units:  $\mu\text{g m}^{-3}$ .

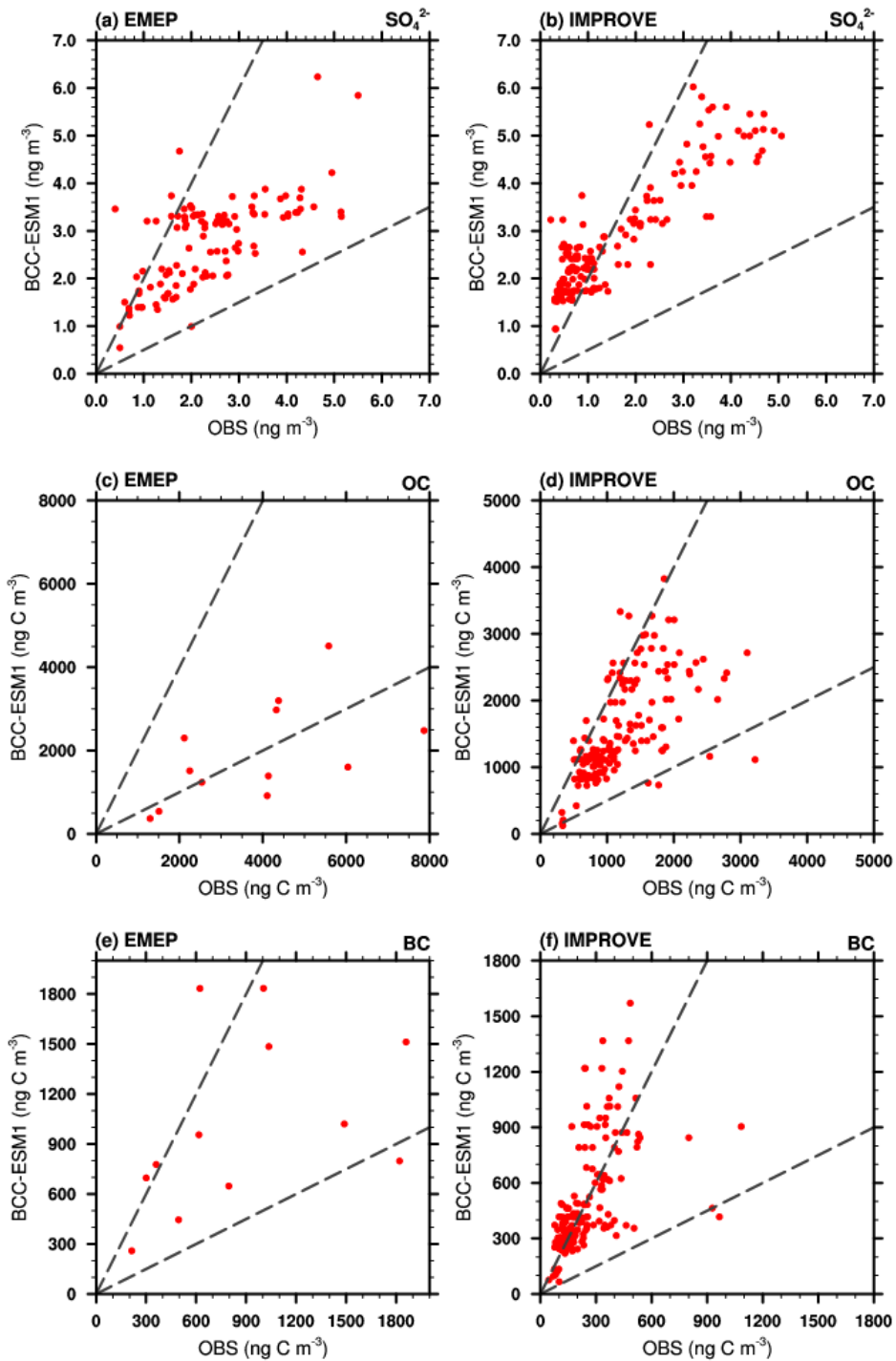


Figure 15. Scatter plots showing observed versus simulated multi-years averaged annual mean sulfate ( $\text{SO}_4^{2-}$ ), organic carbon (OC), black carbon (BC) mixing ratios at IMPROVE and EMEP network sites. Observations are averages over the available years 1990–2005 for IMPROVE sites, and 1995–2005 for EMEP sites. Simulated values are those at the lowest layer of BCC-ESM1.

Table 7. Observed versus simulated concentrations of sulfate ( $\text{SO}_4^{2-}$ ), organic carbon (OC), black carbon (BC) for the regional mean and spatial standard deviation, minimum and maximum values at [HIPPO aircraft observations \(BC only\)](#), IMPROVE and EMEP network sites, and the spatial correlation between observed and simulated multi-years averaged annual means. Simulated values are selected for the same locations and same valid observation time. The data used same as those in Figure 12.

	EMEP			IMPROVE			HIPPO
	$\text{SO}_4^{2-}$ (Obs/Model)	OC (OBS/Model)	BC (OBS/Model)	$\text{SO}_4^{2-}$ (OBS/Model)	OC (OBS/Model)	BC (OBS/Model)	BC (OBS/Model)
<b>Mean Values</b>	2.37/2.74	3844/1919	884/1022	1.53/2.79	1215/1565	249/504	8.2/11.1
<b>Std Deviation</b>	1.16/0.93	1997/1215	572/526	1.30/1.20	572/745	164/296	27.9/21.0
<b>Min Values</b>	0.40/0.55	1296/369	214/259	0.22/0.94	322/123	45/66	0.0025/0.066
<b>Max Values</b>	5.50/6.24	7867/4510	1859/1834	5.07/6.02	3219/3827	1084/1570	558.91/267.11
<b>Correlation (Obs and Model)</b>	0.67	0.56	0.40	0.90	0.63	0.55	0.51

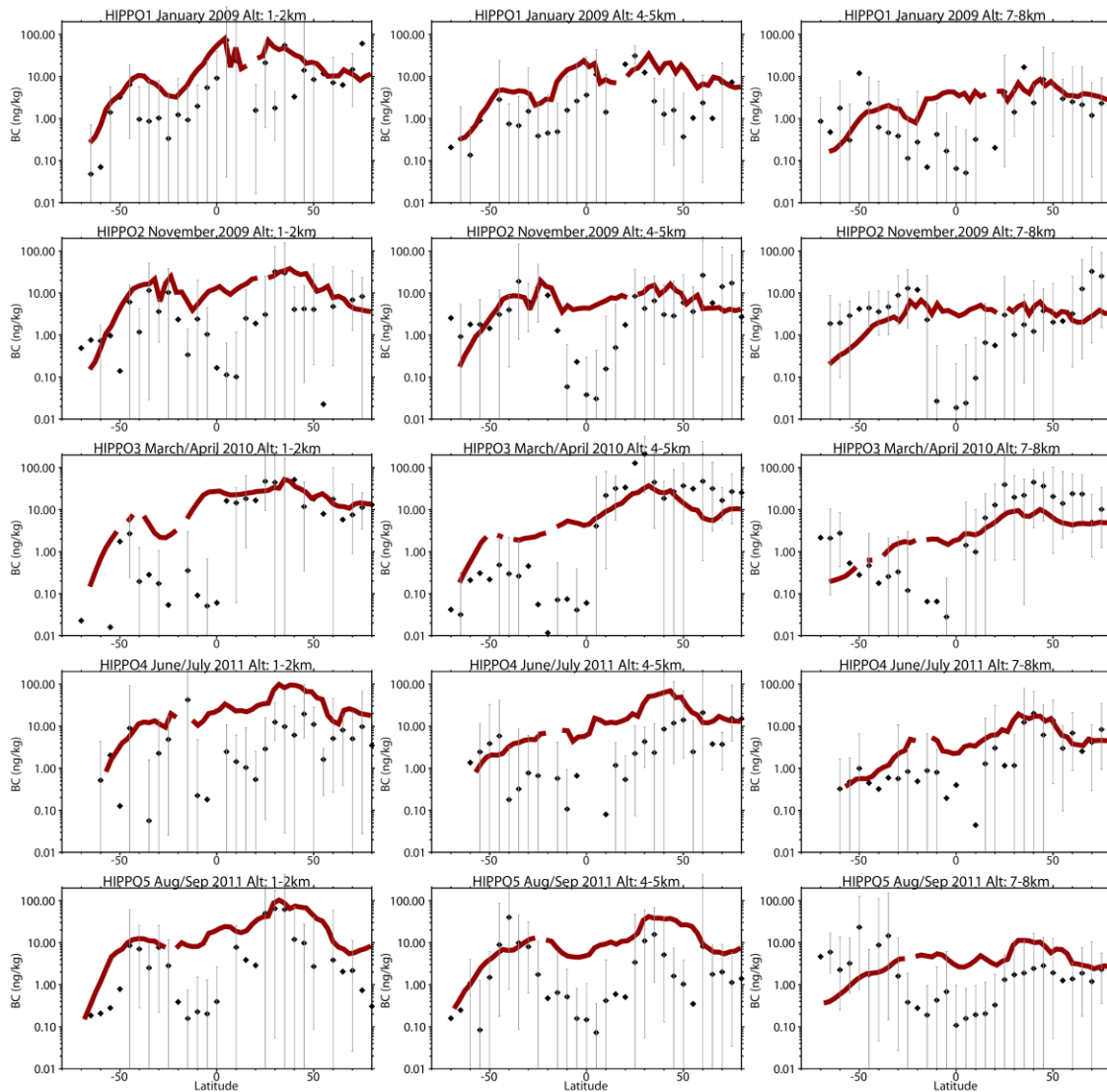


Figure 16. Comparison of modelled black carbon (BC) aerosol (red lines) with observations from HIPPO aircraft campaigns over the Pacific Ocean (black symbols, bars represent the full data range). Observations from different HIPPO campaigns were averaged over 5 °latitude bins and three different altitude bands (left column: 1-2 km, middle column: 4-5 km, and right column: 7-8 km) along the flight track over the Pacific Ocean. Model results were sampled along the flight track and then averaged over the abovementioned regions for comparison.

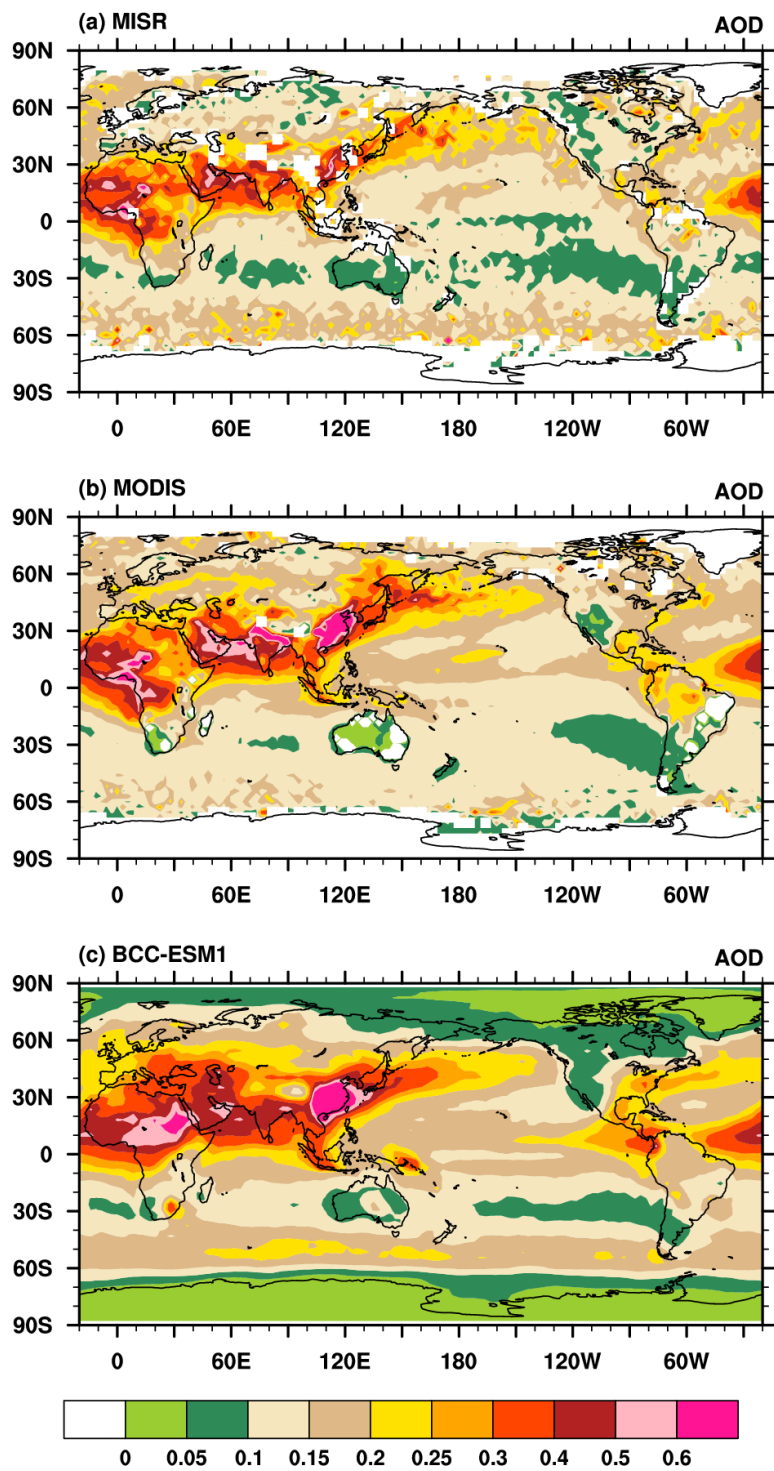


Figure 17. Global distribution of annual mean AOD simulated in BCC-ESM1 compared with the MISR and MODIS data for the year 2008.

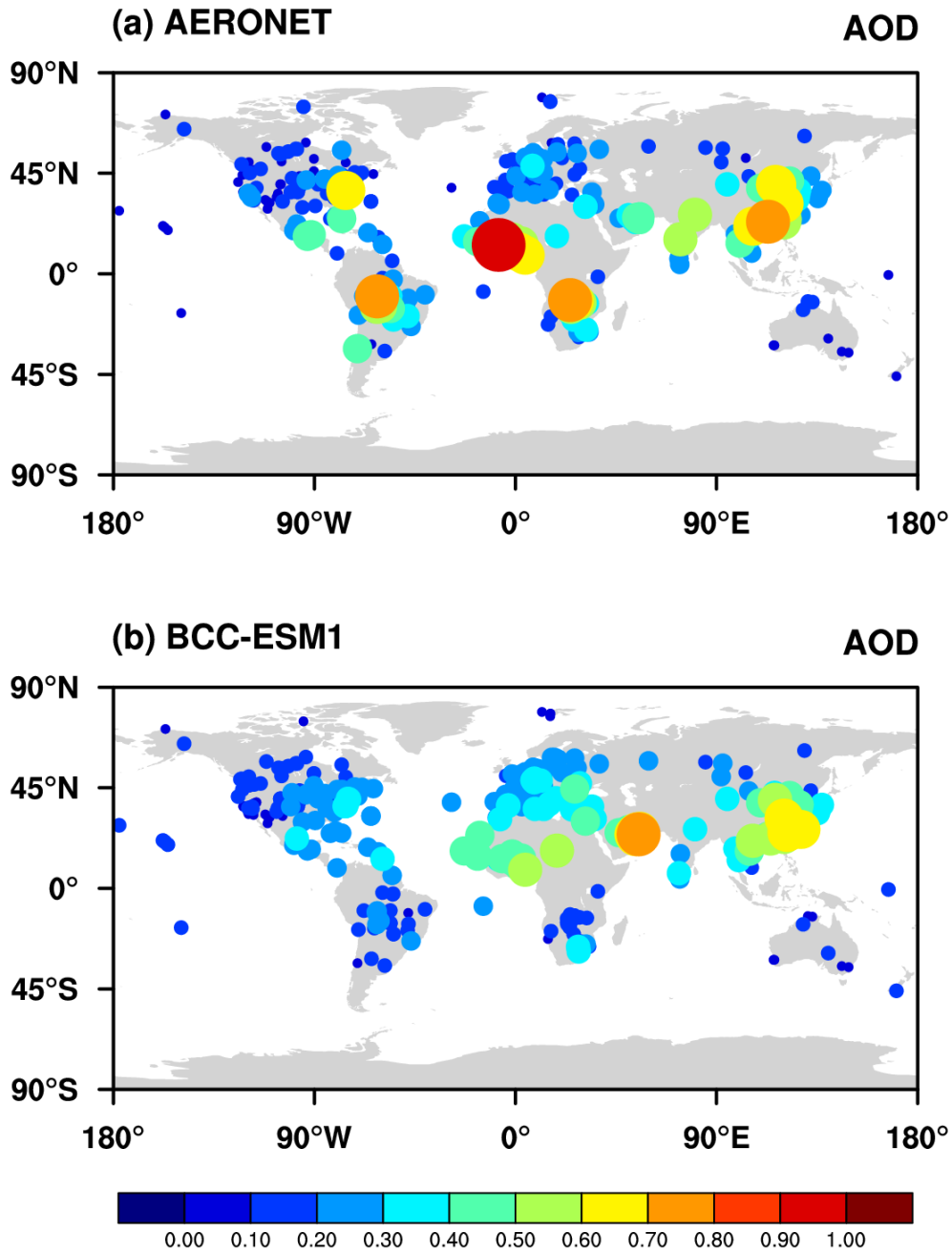


Figure 18. Observed versus simulated annual means of AOD at AERONET sites. Each data point represents the mean averaged for available monthly values of AOD. The dot sizes denote the magnitudes of AOD at sites. The spatial correlation is 0.56.



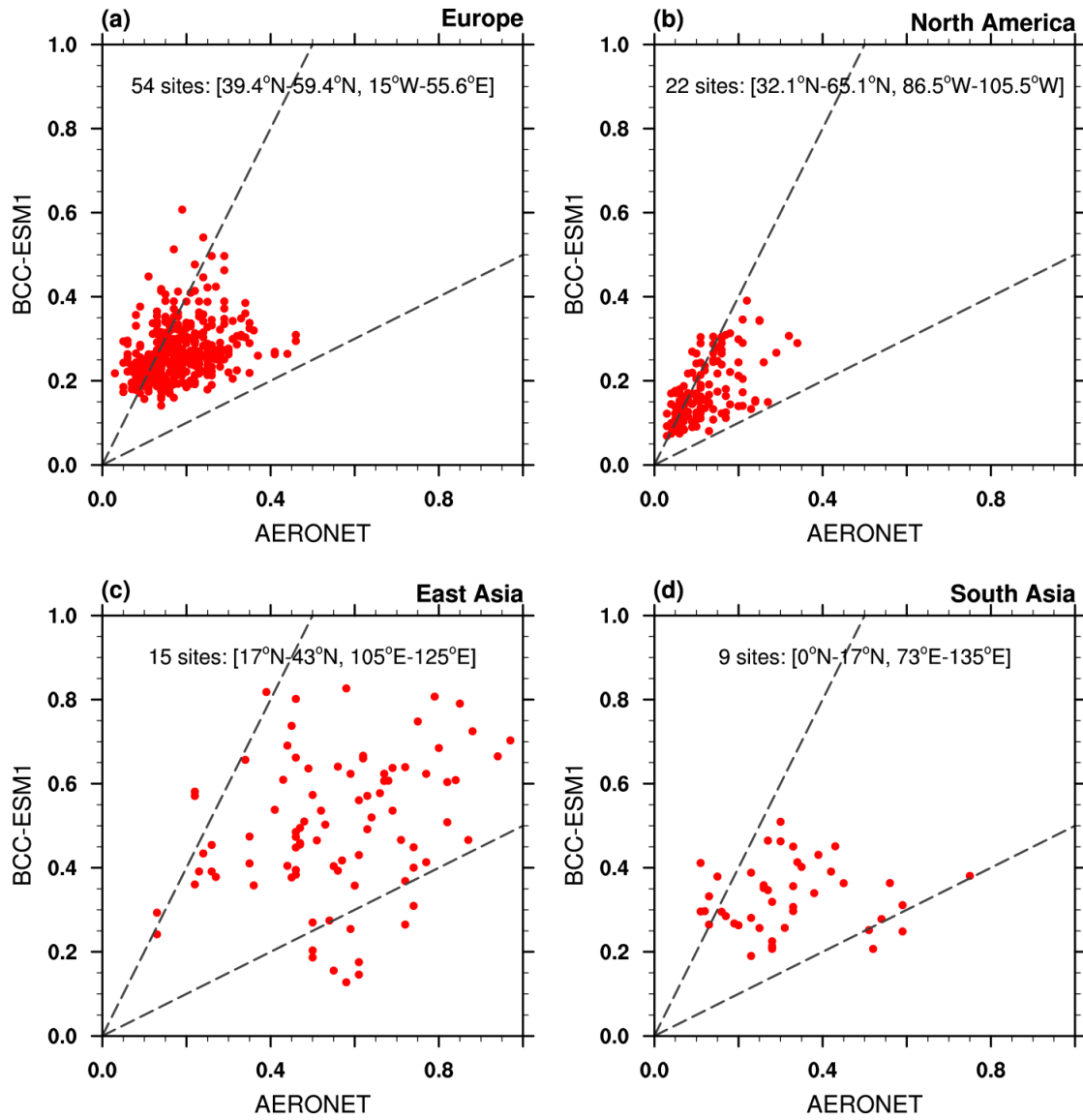


Figure 19. Scatter plots of observed versus simulated monthly mean AOD at AERONET sites in Europe, North America, East Asia, and South Asia over the period of 1998-2005.