

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

Comments:

".... Thank you for making the BCC climate model source code and input files available through a repository, including the provision of a doi.

However, I note that my request to precisely cite the model output data from the CMIP6 archive hasn't been addressed. May I please ask that you correct this, so that the revised manuscript fully complies with the GMD data policy?

As mentioned previously, model output on ESGF is very good practice but can be difficult to find. ESGF provides the correct data citation for each piece of data on the corresponding catalogue page. Precise instructions for citing CMIP6 data are here: https://docs.google.com/document/d/1SnwBL9MJQNEU1_nJ661SN3-SSxR0j1mAYan6-WXfaSU/edit

As soon as I'm satisfied that the model output is being cited precisely, I will accept your manuscript for final publication in Geoscientific Model Development. "

Response:

Dear Dr. Fiona O'Connor

Thanks for your helpful comments. We have modified the description in the section of "6. Code and data availability" as: "...Model output data of BCC CMIP6 AerChemMIP simulations described in this paper are available on the Earth System Grid Federation (ESGF) (<https://cera-www.dkrz.de/WDCC/ui/cersearch/cmip6?input=CMIP6.AerChemMIP.BCC.BCC-ESM1>, <https://doi.org/10.22033/ESGF/CMIP6.1733>; Zhang et al., 2019) ..." That modification maybe help readers to easily find the BCC model output data.

Best regards,

Tongwen Wu, and Co-authors.

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

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

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22 Submit to Geosci. Model Dev.

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27

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 (O₃), carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O),
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 O₃ 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 O₃ depletion (e.g., Cariolle et al. 1990; Austin et al. 1992; Solomon, 1999), tropospheric O₃
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 CO₂ from combustion of fossil fuels and
76 land use change, BCC-CSM1.1 successfully reproduced the trends of observed atmospheric
77 CO₂ 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 CO₂ concentration is calculated from a prognostic equation of CO₂ budget
83 taking into account global anthropogenic CO₂ emissions and interactive land-atmosphere and
84 ocean-atmosphere CO₂ 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. O₃, CH₄, N₂O and CO₂) 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 (SO₂) and ammonia (NH₃), 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 O₃, carbon monoxide (CO), CH₄, formaldehyde (CH₂O), acetic acid
132 (CH₃OOH), hydrogen peroxide (H₂O₂), nitrogen dioxide (NO₂), nitric acid (HNO₃),
133 polyacrylonitrile (PAN), acetone (CH₃COCH₃), peroxyacetic acid (CH₃COOOH),
134 acetaldehyde (CH₃CHO), methylglyoxal (CH₃COCHO), nitric oxid (NO), and pernitric acid
135 (HNO₄) 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 (C₂H₅OH), 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, SO₂ and NH₃, 13 prognostic aerosol species including sulfate
182 (SO₄²⁻), 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 SO₄²⁻ which gas-phase and aqueous phase conversion from SO₂ 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 SO₂, DMS, NH₃, and Sulfate**

192 SO₂ is a main sulfuric acid precursor to form aerosol sulfate SO₄²⁻. Conversions of SO₂
193 to SO₄²⁻ occur by gas phase reactions (Table 2) and by aqueous phase reactions in cloud
194 droplets. The dry deposition velocity of SO₂ 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 SO₂ 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 SO₂ mainly come from fuel combustion, industrial activities, and
202 volcanoes. SO₂ 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>). SO₄²⁻ 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 SO₂ (in Table 2) and by aqueous phase
209 oxidation of SO₂ 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 SO₄²⁻
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 SO₂, HNO₃, H₂O₂, NH₃, O₃, HO₂, and
214 SO₄²⁻. NH₃ 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). SO₄²⁻ 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 NH₃. The sulfate in- and below-cloud scavenging follows Neu and Prather (2011). Washout
223 of SO₄²⁻ is set to 20% of the washout rate of HNO₃ following Tie et al. (2005) and Horowitz
224 (2006). Dry deposition velocity of SO₄²⁻ 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 μm) 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 4.05×10^{-15} , 4.52×10^{-14} , 1.15×10^{-13} , 1.20×10^{-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} (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):

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

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 β 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 ρ_w is the liquid water density and LWC the cloud liquid water content (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, ρ_a and ρ_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 μm .

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 20th 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). O₃ in the historical
330 simulation is an interactive prognostic variable and feedbacks on radiation, and the
331 concentrations of other WMOGHG, e.g. CH₄, N₂O, CO₂, CFC11, and CFC12 are prescribed
332 using CMIP6 historical forcing data (Meinshausen et al., 2017). Although CH₄ and N₂O 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 H₂ and N₂O are from monthly climatological dataset provided by the MOZART-2
351 standard package. N₂O 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 (CH₃COCH₃) and anthropogenic emission (CH₃CHO, CH₃OH, and CH₃COCH₃) 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 (CH₃COCH₃), C₂H₄, C₃H₈, and Monoterpenes (C₁₀H₁₆) 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 CO₂ 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 SO₂ emissions from volcanic eruption on the variation of SO₂ in the atmosphere and
380 then on the variation of tropospheric SO₄²⁻ concentration are considered, and the SO₂
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 NO₂, CO,
383 CH₄, NH₃, NO, SO₂, 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⁻¹, which is in agreement with observations within the range of 3 to 6 Tg(N) yr⁻¹
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 (O₃, CH₄, N₂O, NO, NO₂, HNO₃, CO, and N₂O₅) 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, NO₂, HNO₃, CO and
397 N₂O₅ 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 (O₃, CH₄, and N₂O) 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 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 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 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 SO_2 to form SO_4^{2-} (Table 2), keeps at a stable level in the atmosphere. 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 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 SO₂ emissions, the
437 SO₄²⁻ over land are evidently smaller than those over oceans especially over the tropical
438 Pacific and Atlantic Oceans, where DMS can be oxidized to SO₂ and then form SO₄²⁻. 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 O₃ and aerosol simulations in the 20th century**

458 The rate of sulfate formation is dependent on the levels of oxidants in the troposphere.
459 O₃ is an important oxidant. So, the evaluation of simulated tropospheric O₃ 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 O₃ 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 O₃ are higher than those in the

466 Southern Hemisphere.

467 Figure 5 shows the vertical profiles of O₃ 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 SO₂, OC, and BC to the
484 atmosphere from natural and anthropogenic sources. Emissions of SO₂ are largely due to
485 industrial production. From 1850 to 1915, SO₂ emissions increased year by year as the
486 Industrial Revolution intensified and expanded. But from 1915 to 1945, the increase trend of
487 SO₂ emissions became slower as broke out the First and the Second World Wars. After that
488 period, with growing industrial productions, SO₂ emissions increased again and reached a
489 maximum around the end of 1970s. During the 1980s and 2000s, with a substantial decrease
490 of SO₂ emissions in Europe and the United States, the global SO₂ emissions has been
491 decreasing since the 1980s despite the rapid increase of SO₂ 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⁻¹. Anthropogenic black carbon

496 emissions increased from 1 Tg yr⁻¹ in 1850 to nearly 8 Tg yr⁻¹ in 2010.

497 Anthropogenic SO₂, 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 SO₄²⁻, OC, and BC
502 in the whole atmosphere column as simulated by the BCC-ESM1 20th century historical
503 simulation are generally consistent with the values derived from CMIP5-recommended
504 aerosols concentrations. Due to increasing SO₂ emissions from 1850 to present day (Fig. 6),
505 the global SO₂ 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 SO₂ is short. The burden directly followed the emission. DMS in
508 the atmosphere is oxidized by OH and NO₃ to form SO₂ (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 SO₂ 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 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 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%) SO_2 is
540 produced from the DMS oxidation, only $0.1 \text{ Tg(S) yr}^{-1}$ 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 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 SO_2
544 production is balanced by 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 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 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 SO_2 oxidation ($38.73 \text{ Tg(S) yr}^{-1}$)
551 and partly from gaseous phase oxidation of 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 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⁻¹ BC and 13.91 Tg yr⁻¹ OC from fossil and bio-fuel
559 emissions and 18.38 Tg yr⁻¹ 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⁻¹, higher than AeroCom multi-model
570 mean (1840 Tg yr⁻¹, 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⁻¹, slightly lower than
576 the simulated value in Liu et al. (2012), and substantially lower than the AeroCom mean
577 (16600 Tg yr⁻¹, 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 (SO₄²⁻), 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 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., O₃, CH₄, N₂O) 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 CH₄, N₂O, CO₂, CFC11 and CFC12 are prescribed
701 using CMIP6 prescribed concentrations (to replace prognostic values of CH₄ and N₂O 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 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 SO_2 , OC, and BC from 1850 to 2014 and
708 their natural emissions implemented in BCC-ESM1, the model simulated 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 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 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. O_3 is evaluated in this work. Other GHGs such as CH_4 and

734 N₂O 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 input files, and scripts to reproduce the
738 simulations that are presented in the article have been archived and made publicly available
739 for downloading from <https://zenodo.org/record/3609337> (Wu et al., 2020). Model output
740 data of BCC CMIP6 AerChemMIP simulations described in this paper are available on the
741 Earth System Grid Federation (ESGF)
742 ([https://cera-www.dkrz.de/WDCC/ui/cerasearch/cmip6?input=CMIP6.AerChemMIP.BCC.B](https://cera-www.dkrz.de/WDCC/ui/cerasearch/cmip6?input=CMIP6.AerChemMIP.BCC.BCC-ESM1)
743 <https://doi.org/10.22033/ESGF/CMIP6.1733>; refer to Zhang et al., (2019) and is
744 distributed through ESGF and freely accessible through the ESGF data portals after
745 registration. Details about 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

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758

759 **References**

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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 ₃	✓				
N ₂ O			✓		
N					
NO	✓		✓	✓	
NO ₂	✓				
NO ₃					
HNO ₃	✓	✓			
HO ₂ NO ₂	✓	✓			
N ₂ O ₅					
CH ₄	✓		✓	✓	
CH ₃ O ₂					
CH ₃ OOH	✓	✓			
CH ₂ O	✓	✓	✓		
CO	✓		✓	✓	
OH					
HO ₂					
H ₂ O ₂	✓	✓			
C ₃ H ₆			✓		
ISOP			✓		
Gas tracers					
PO ₂					
CH ₃ CHO	✓	✓	✓		
POOH	✓	✓			
CH ₃ CO ₃					
CH ₃ COOOH	✓	✓			
PAN	✓				
ONIT	✓	✓			
C ₂ H ₆			✓		
C ₂ H ₄			✓		
C ₄ H ₁₀			✓		
MPAN	✓				
ISOPO ₂					
MVK		✓			
MACR		✓			
MACRO ₂					
MACROOH	✓	✓			
MCO ₃					
C ₂ H ₅ O ₂					
C ₂ H ₅ OOH	✓	✓			
C ₁₀ H ₁₆			✓		

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

1097 Table 2. Gas-phase chemical reactions for NH₃ and bulk aerosols precursors following
 1098 CAM-Chem (Lamarque et al., 2012). The reaction rates (s⁻¹) 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⁻³, ki and ko in cm³ molecule⁻¹ s⁻¹.
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Chemical reactions	Rate
NH ₃ + OH → H ₂ O	1.70E-12*exp(-710/T)
SO ₂ + OH → SO ₄ ²⁻	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 ₂	9.60E-12*exp(-234./T)
DMS + OH → .5*SO ₂ + .5*HO ₂	1.7E-42*exp(7810/T)*M*0.21/(1+5.5E-31*exp(7460/T)* M* 0.21)
DMS + NO ₃ → SO ₂ + HNO ₃	1.90E-13*exp(520/T)
BC1 → BC2	7.10E-06
OC1 → OC2	7.10E-06

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Table 3. Size and density parameters of bulk aerosols.

Aerosols	Species Name	Mean radius (μm) / bin size (μm)	Geometric standard deviation (μm)	Density (g cm^{-3})
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

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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 ₂ H ₄	CMIP6	CMIP6	On-line computation		MOZART2		
C ₂ H ₅ OH	CMIP6	CMIP6					
C ₂ H ₆	CMIP6	CMIP6	ACCMIP		MOZART2		
C ₃ H ₆	CMIP6	CMIP6	On-line computation		MOZART2		
C ₃ H ₈	CMIP6	CMIP6	ACCMIP		MOZART2		
C ₄ H ₁₀	CMIP6	CMIP5	MOZART2		MOZART2		
CH ₂ O	CMIP6	CMIP6					
CH ₃ CHO	ACCMIP	CMIP6					
CH ₃ COCHO		CMIP6					
CH ₃ OH	ACCMIP	CMIP6	ACCMIP				
CH ₃ COCH ₃	ACCMIP	ACCMIP	On-line computation		MOZART2		
ISOP		CMIP5	On-line computation				
C ₁₀ H ₁₆		CMIP6	On-line computation				
CH ₄	CMIP6	CMIP6	MOZART2		MOZART2	CMIP6	
CO	CMIP6	CMIP6	ACCMIP	MOZART2	ACCMIP	CMIP6	
H ₂	MOZART2	CMIP6		MOZART2	MOZART2		
N ₂ O	MOZART2	CMIP6		MOZART2	MOZART2		
NH ₃	CMIP6	CMIP6		ACCMIP	ACCMIP	CMIP6	
NO	CMIP6	CMIP6		ACCMIP		CMIP6	
SO ₂	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, SO₂, and sulfate in the period of 1991 to 2000. Units are sources and sinks, Tg(S) yr⁻¹; 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 ^a
	Sinks	28.0	
	Gas-phase oxidation	28.0	
	Burden	0.12	0.04-0.29 ^a
	Lifetime	0.78	0.5-3.0 ^a
SO ₂	Sources	76.93	
	Emission at surface	63.63	
	Emission from airplane	0.10	
	DMS oxidation	13.20	10.0-24.7 ^a
	Sinks	76.96	
	Dry deposition	18.53	16.0-55.0 ^a
	Wet deposition	9.36	0.0-19.9 ^a
	Gas-phase oxidation	10.33	6.1-16.8 ^a
	Aqueous-phase oxidation	38.74	24.5-57.8 ^a
Burden	0.48	0.40-1.22 ^a	
Lifetime	1.12	0.6-2.6 ^a	
SO ₄ ²⁻	Sources	49.05	59.67 ± 13.13 ^b
	Emission	0.00	
	SO ₂ aqueous-phase oxidation	38.73	
	SO ₂ gas-phase oxidation	10.32	
	Sinks	49.06	
	Dry deposition	2.20	4.96-5.51 ^d
	Wet deposition	46.86	39.34-40.20 ^d
	Burden	1.89	1.98 ± 0.48 ^b , 1.71 ^c , 1.2 ^g , 2.22-2.43 ^h
	Lifetime	4.69	4.12 ± 0.74 ^b , 3.72-3.77 ^d 3.3 ^g , 3.7-4.0 ^h

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, SO₂, 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⁻¹; 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 ^b , 7.8 ^g
	Sinks	7.24	7.75 ^d , 7.8 ^g
	Dry deposition	0.90	0.27 ^g , 1.30-1.64 ^e
	Wet deposition	6.34	7.5 ^g , 6.10-6.45 ^e
	Burden	0.13	0.114 ^c , 0.24 ± 0.1 ^b , 0.11 ^g , 0.14-0.26 ^h , 0.084-0.123 ^e
	Lifetime	6.60	7.12 ± 2.35 ^b , 3.95-4.80 ^e , 5.0 ^g , 6.3-7.5 ^h
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 ^c , 1.7 ± 0.45 ^b , 1.0-2.2 ^h
Lifetime	5.00	6.54 ± 1.76 ^b , 4.56-4.90 ^d , 6.4 ^g , 5.4-6.6 ^h	
Dust	Sources	2592.0	1840 ^b , 2943.5-3121.9 ^d , 2677 ^g
	Sinks	2592.0	
	Dry deposition	1630.8	1444 ^g
	Wet deposition	961.2	1245 ^g
	Burden	22.93	20.41 ^c , 22.424.7 ^d , 35.9 ^f , 19.2 ± 7.68 ^b , 28.5 ^g , 16.5-17.9 ^h
	Lifetime	3.23	4.14 ± 1.78 ^b , 2.61-3.07 ^d , 3.9 ^g , 5.3-5.7 ^h
Sea Salt	Sources	4667.2	4965.5-5004.1 ^d , 5039 ^g
	Sinks	4667.4	
	Dry deposition	2978.5	2158 ^g
	Wet deposition	1688.9	2918 ^g
	Burden	11.89	7.58-10.37 ^a , 6.4 ± 3.4 ^b , 11.84 ^c , 13.6 ^g , 3.9 ^h
	Lifetime	0.93	0.41 ± 0.24 ^b , 0.55-0.76 ^d , 0.98 ^g , 1.2-1.3 ^h

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).

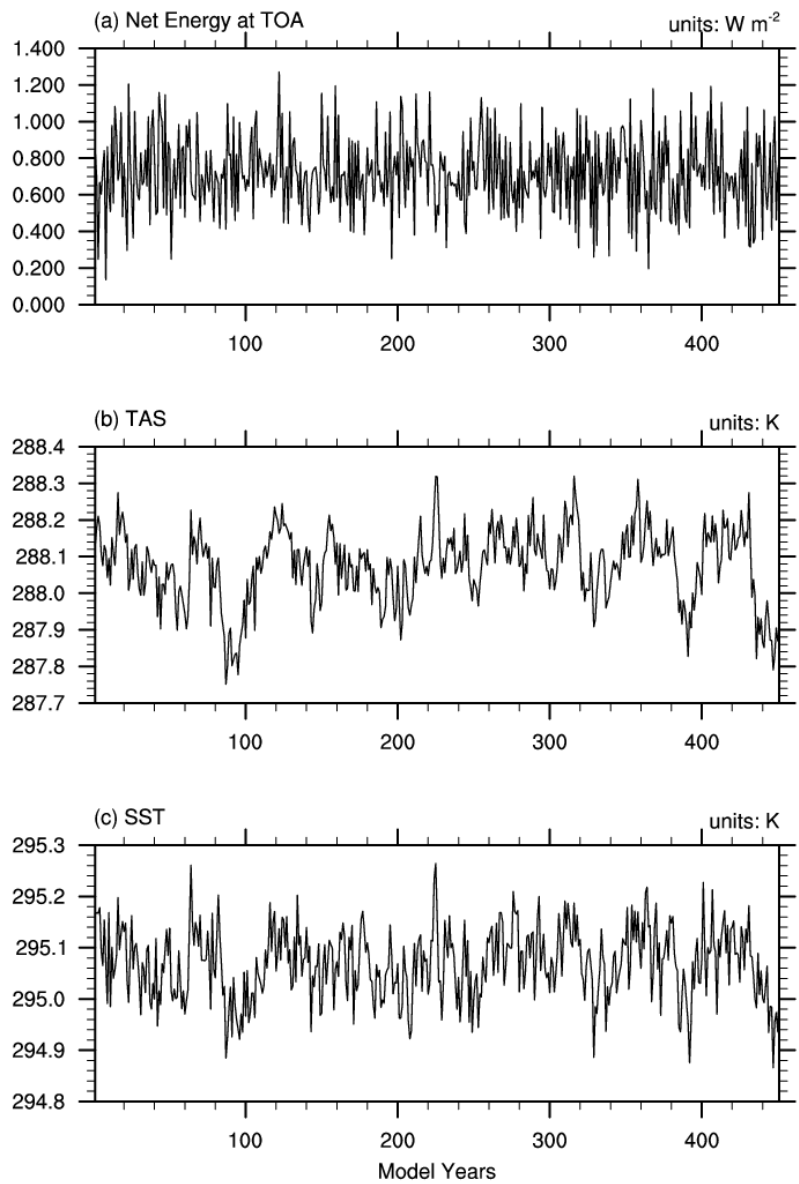


Figure 1. The time series of global and annual mean of (a) net energy budget at top of atmosphere (W m^{-2}), (b) near-surface air temperature (K), and (c) sea surface temperature (K) in the last 450 years of the piControl simulation.

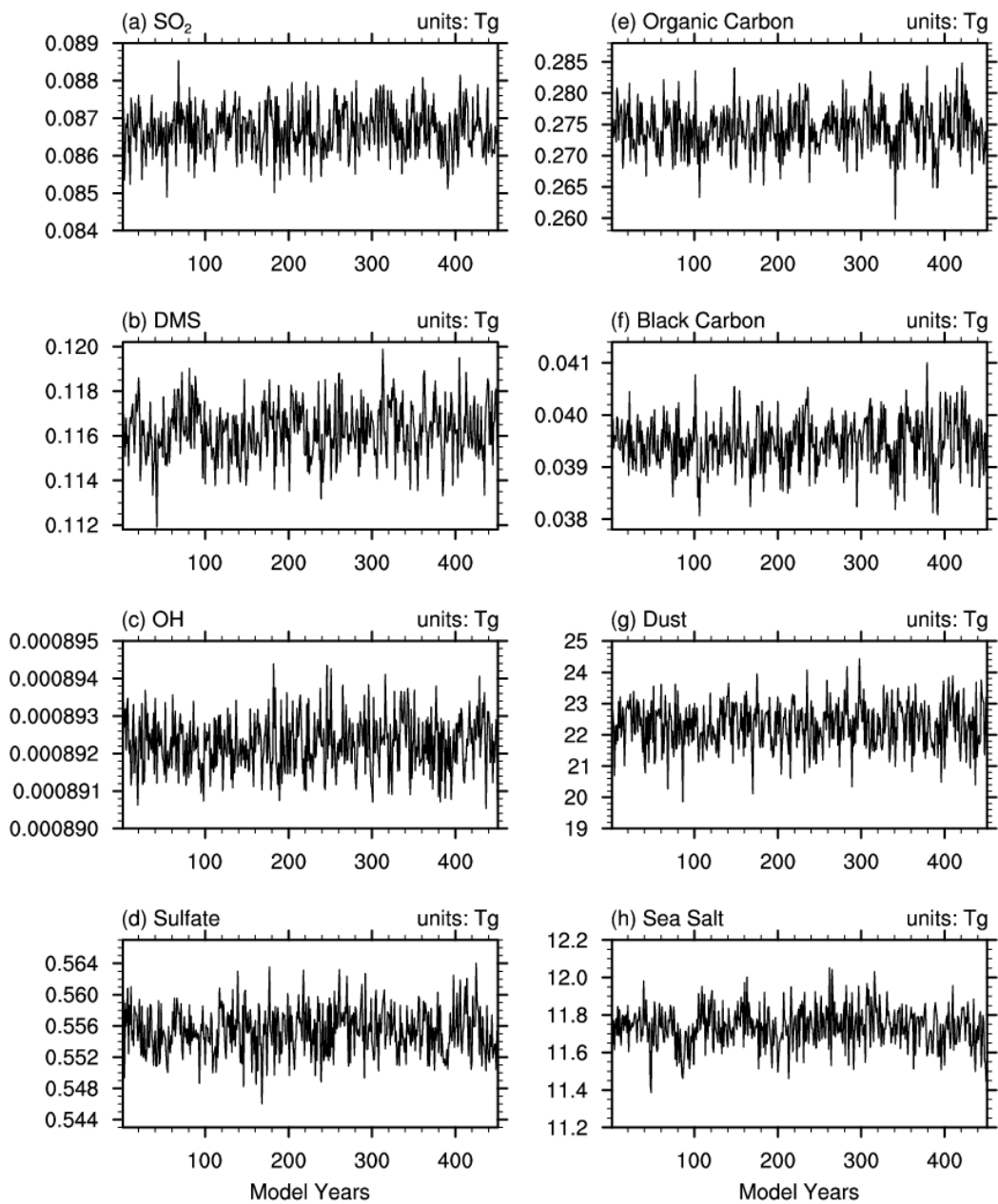


Figure 2. Same as in Figure 1, but for the global burdens of (a) SO_2 , (b) DMS, (c) OH, and (d-h) different aerosols in the troposphere (below 100 hPa). Units are Tg.

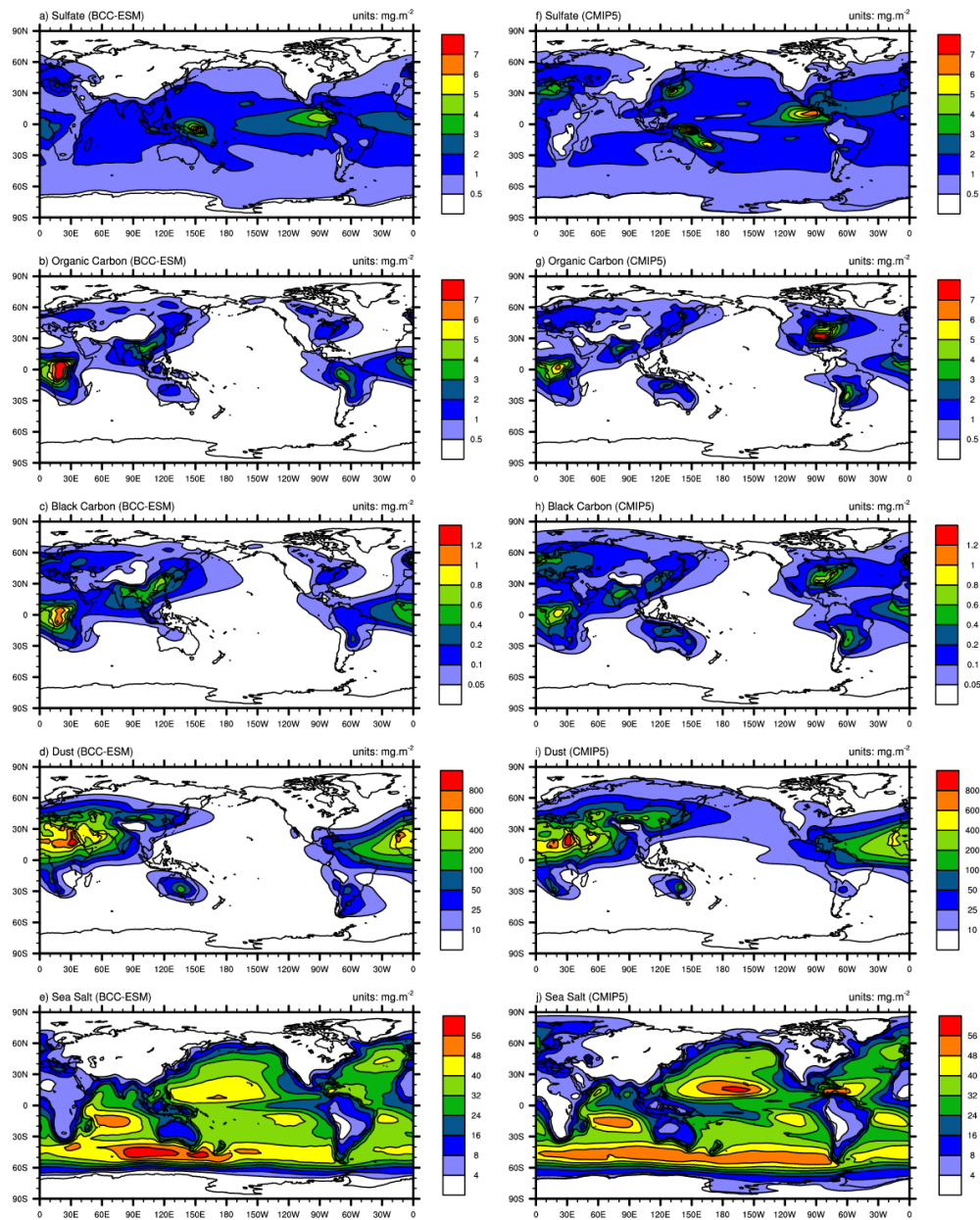


Figure 3. Global distributions of annual mean mass burdens of sulfate (SO_4^{2-} ; first row), organic carbon (OC; second row), black carbon (BC; third row), dust (fourth row), and sea salt (fifth row) aerosols in the whole atmospheric column. The left panels show the mean averaged for the last 100 years 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: mg m^{-2} .

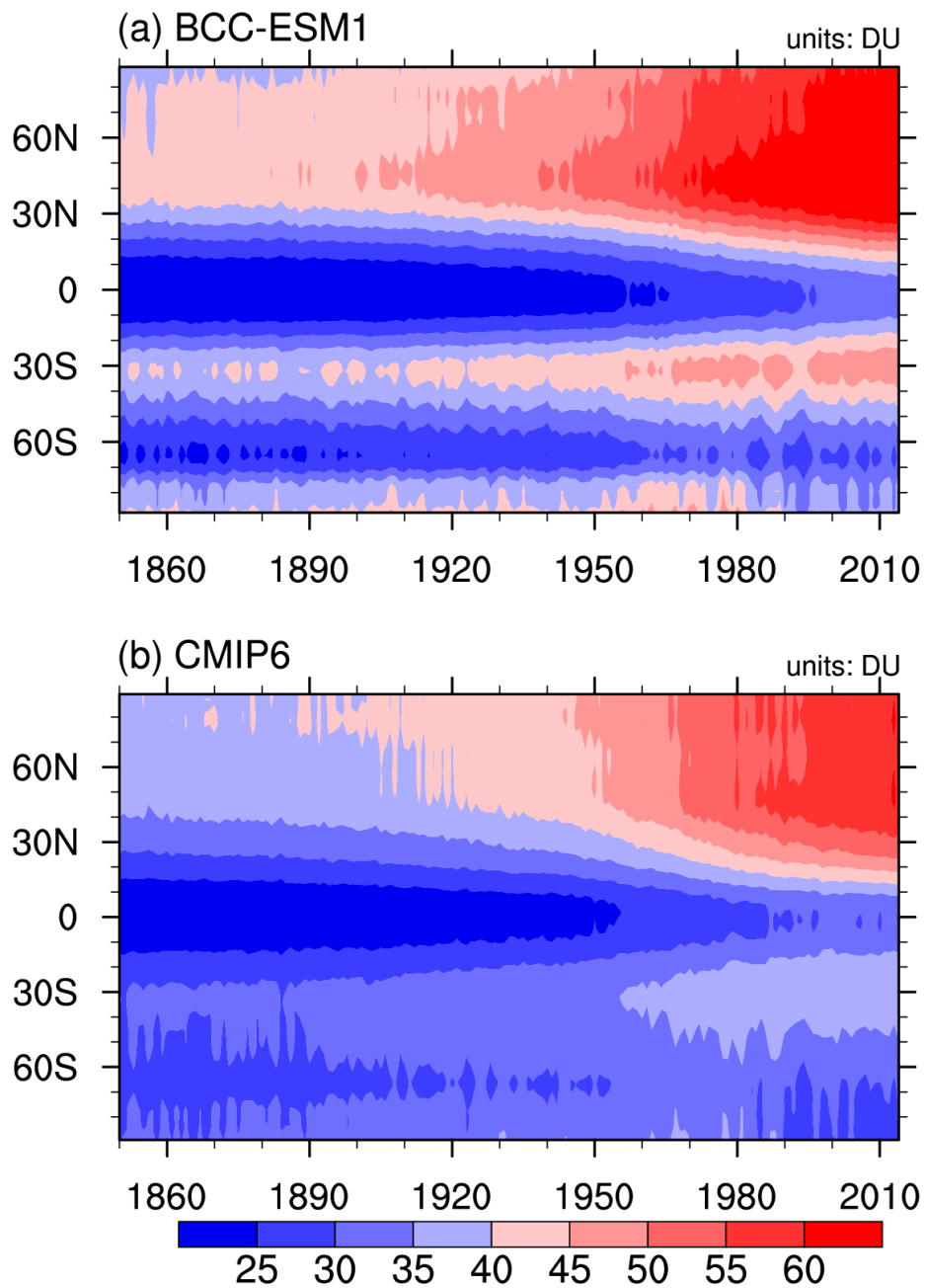


Figure 4. Zonal mean of yearly mean concentration of ozone column in the troposphere below 300 hPa to the ground from 1871 to 1999 for (a) BCC-ESM1 and (b) CMIP6 data. Unit: DU.

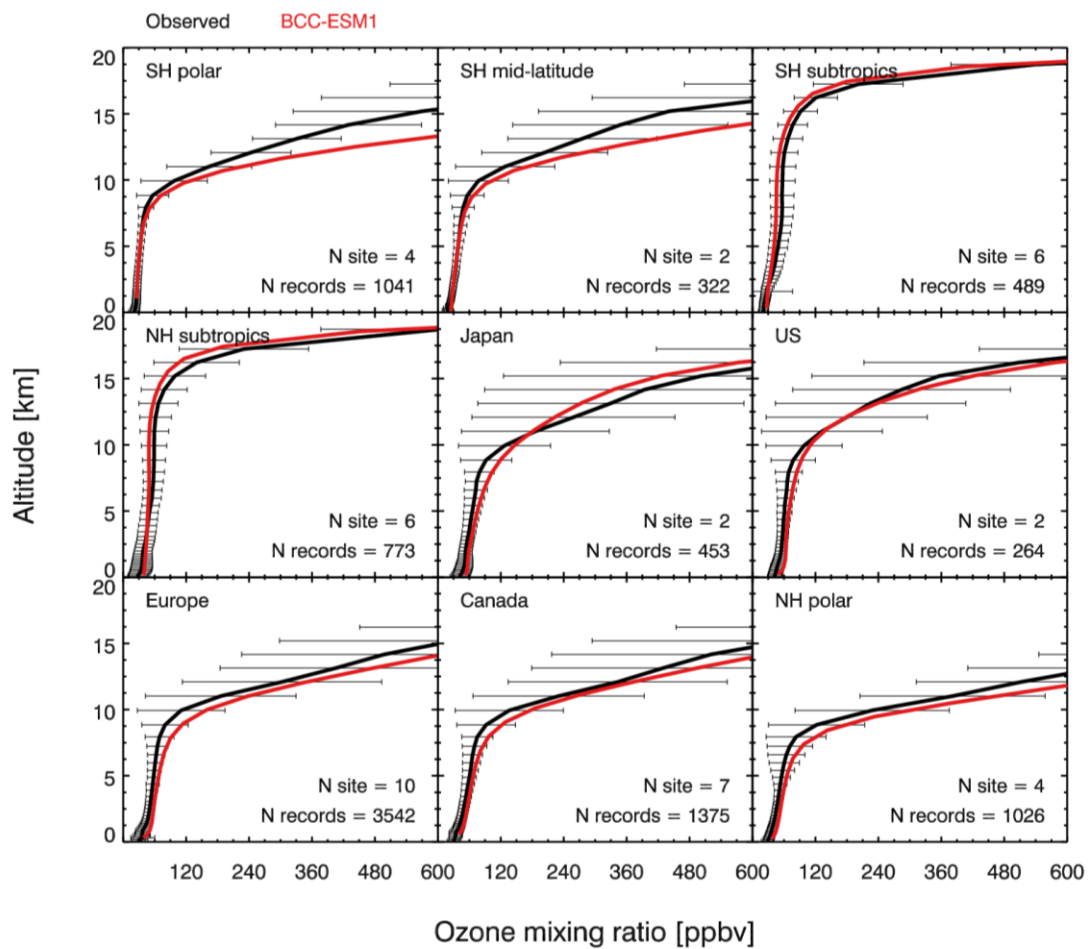


Figure 5. Vertical profiles of annual mean ozone concentrations from observations averaged for 2010-2014 in nine regions (black) and from the BCC-ESM1 simulations (red). The observations are derived from 41 global WOUDC sites.

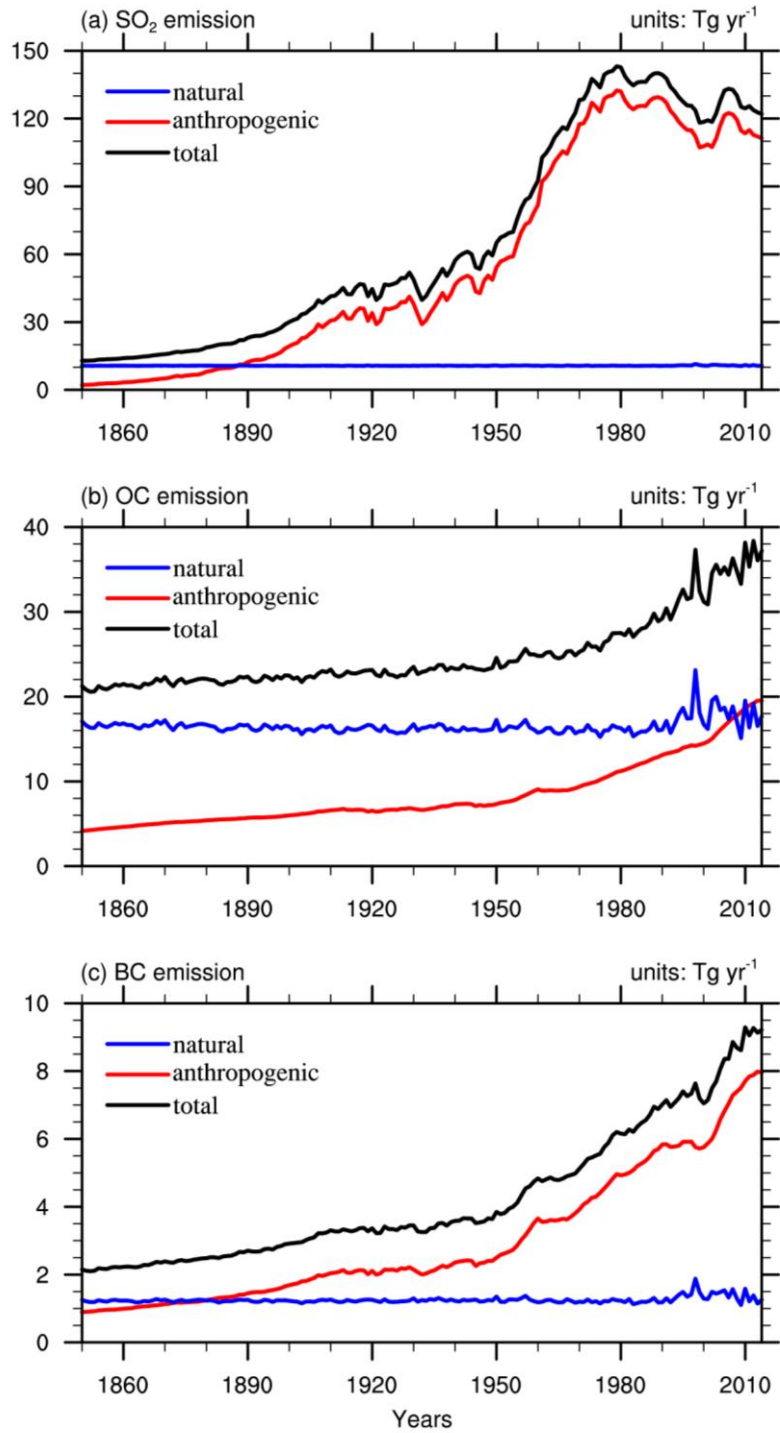


Figure 6. Global annual anthropogenic, natural, and total emissions of SO₂, organic carbon (OC), and black carbon (BC) in the BCC-ESM1 historical simulation. All the biomass burning emissions are included in natural emissions in (a)-(c). Units: Tg yr⁻¹.

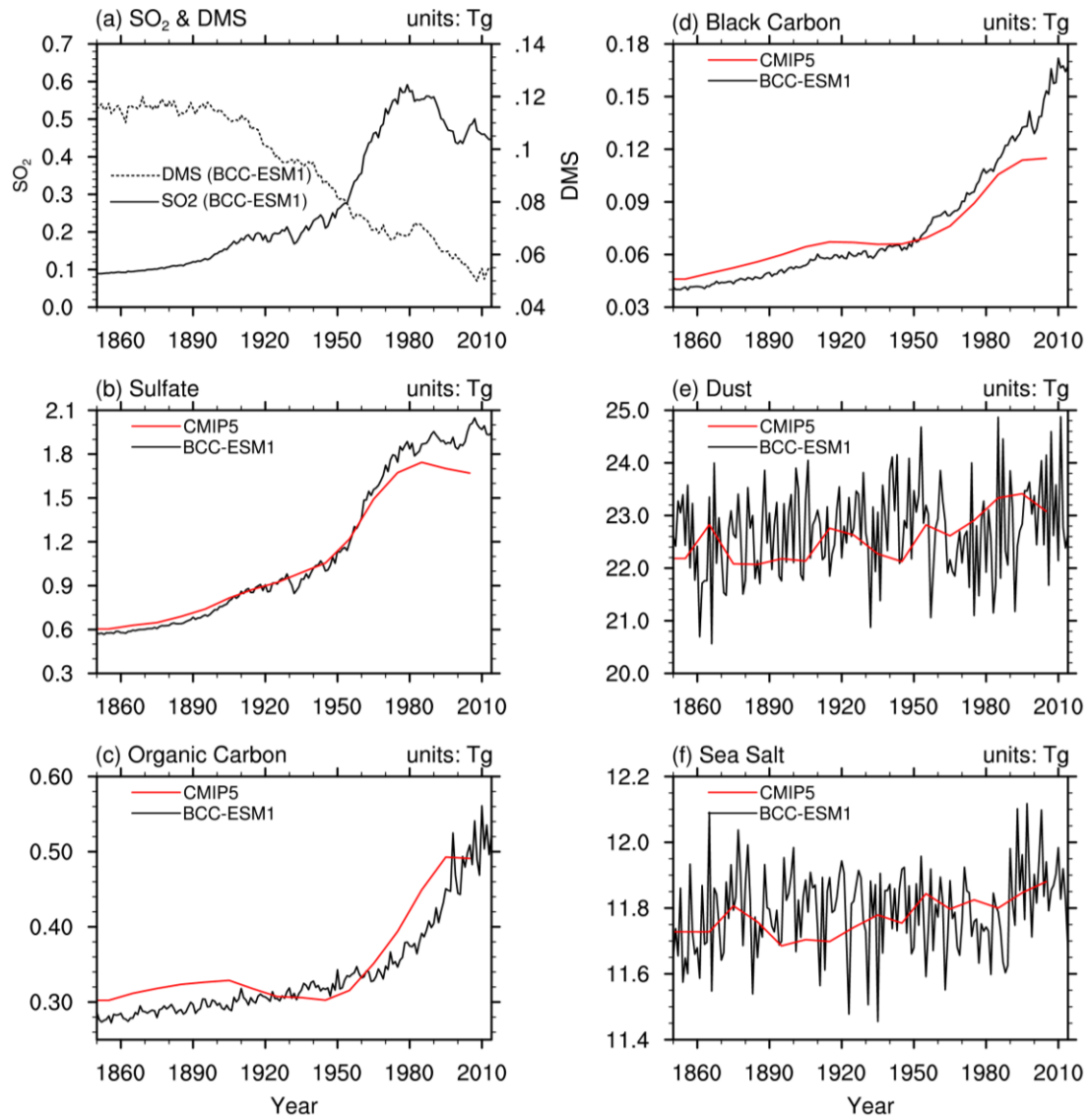


Figure 7. The time series of global yearly amounts of (a) SO₂ and DMS and (b-f) aerosols in the whole atmosphere column from the CMIP6 historical simulations of BCC-ESM1 (black lines) and the CMIP5-recommended aerosols masses (red lines). The yearly CMIP5 data are interpolated from the time series in 10-year interval. Units: Tg.

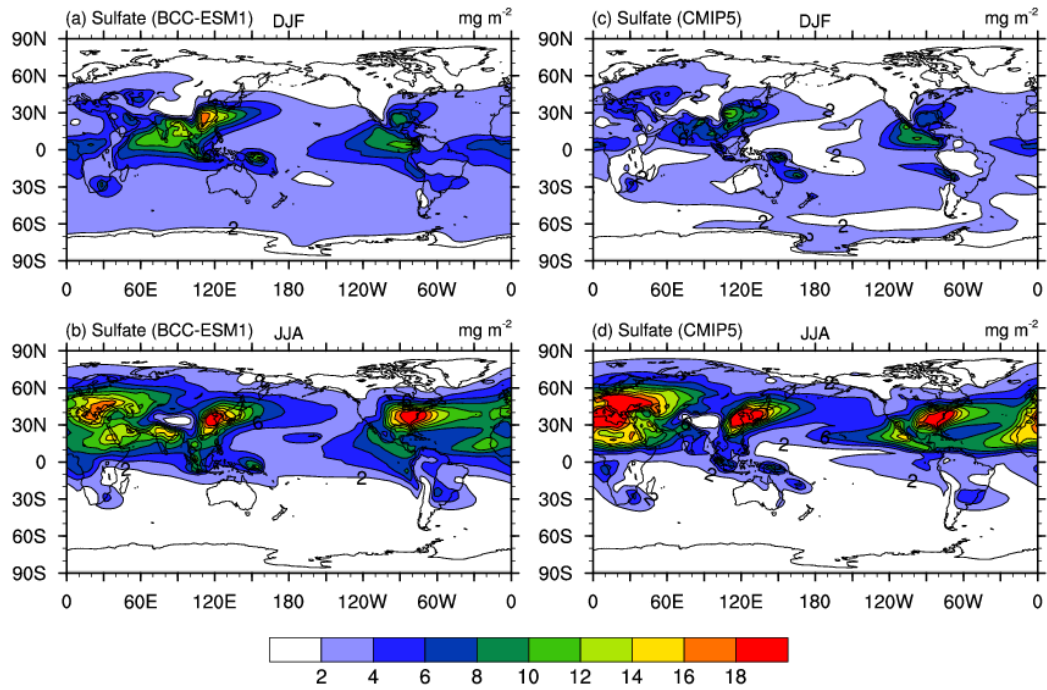


Figure 8. December-January-February (DJF; top panels) and June-July-August (JJA; bottom panels) mean sulfate (SO_4^{2-}) aerosol column mass concentrations averaged for the period of 1971-2000. Left panels show the historical simulations of BCC-ESM1, and right panels the CMIP5-recommended data. Units: mg.m^{-2} .

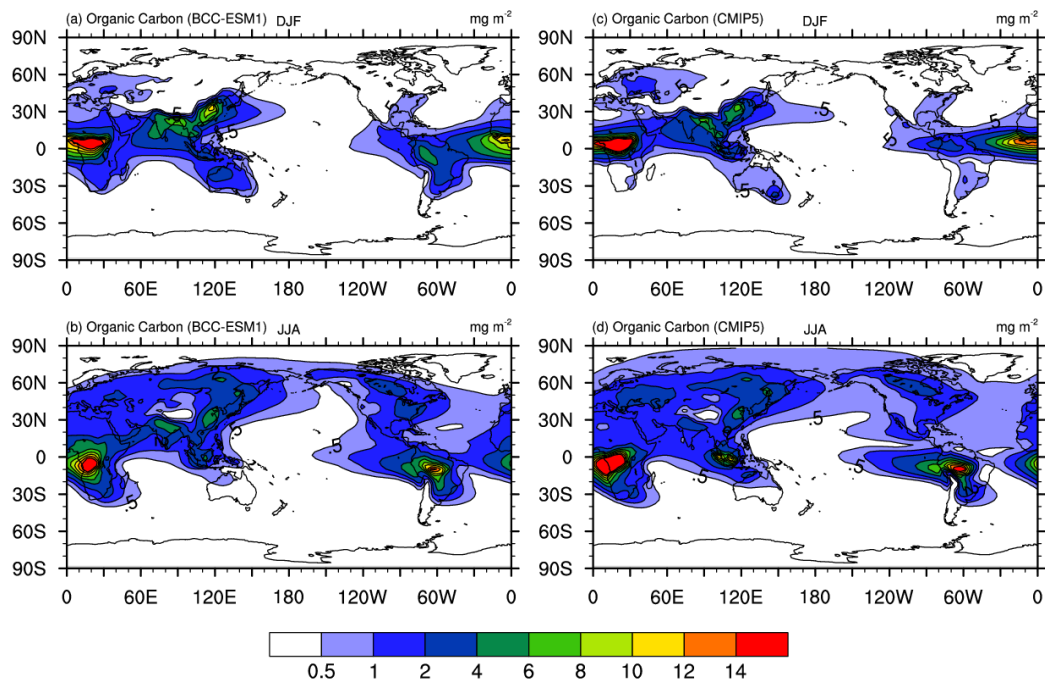


Figure 9. The same as in Figure 8, but for organic carbon (OC) aerosol column mass concentrations. Units: mg m^{-2} .

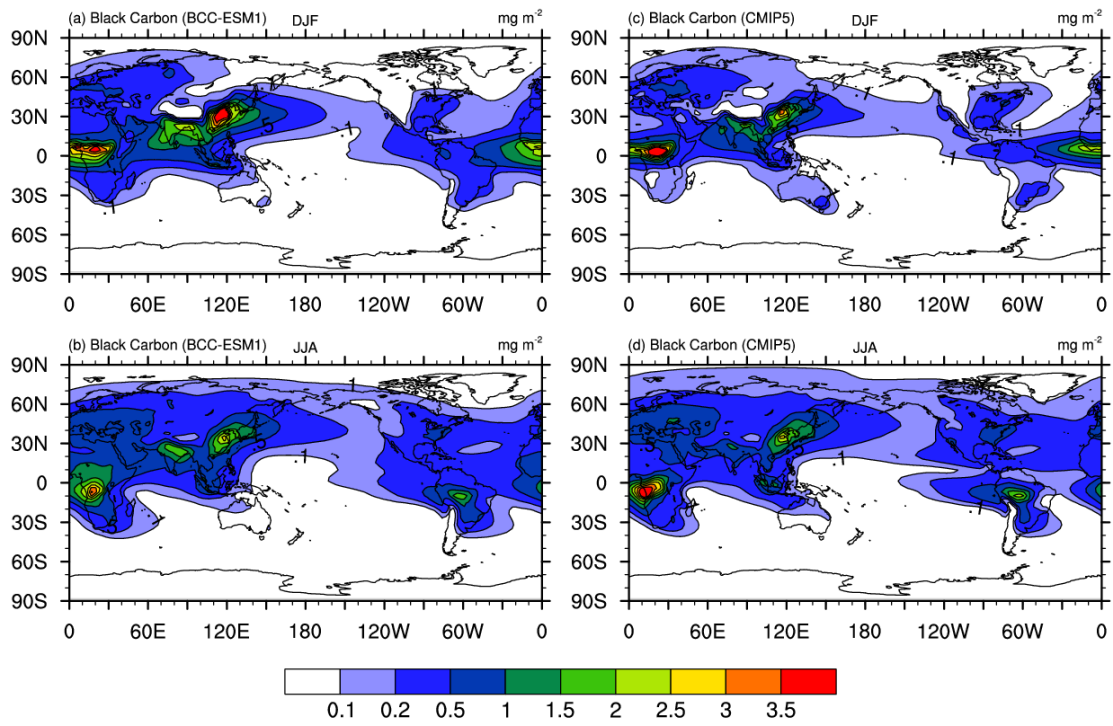


Figure 10. The same as in Figure 8, but for black carbon (BC) aerosol. Units: mg.m^{-2} .

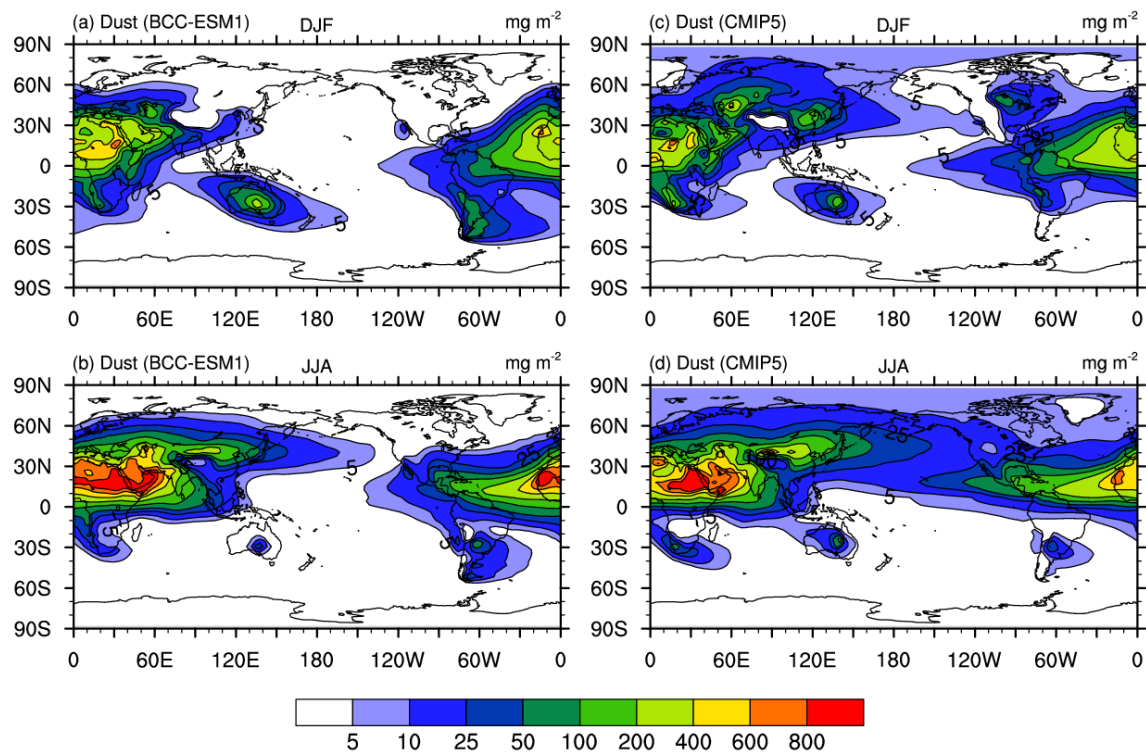


Figure 11. The same as in Figure 8, but for dust aerosol. Units: mg.m^{-2} .

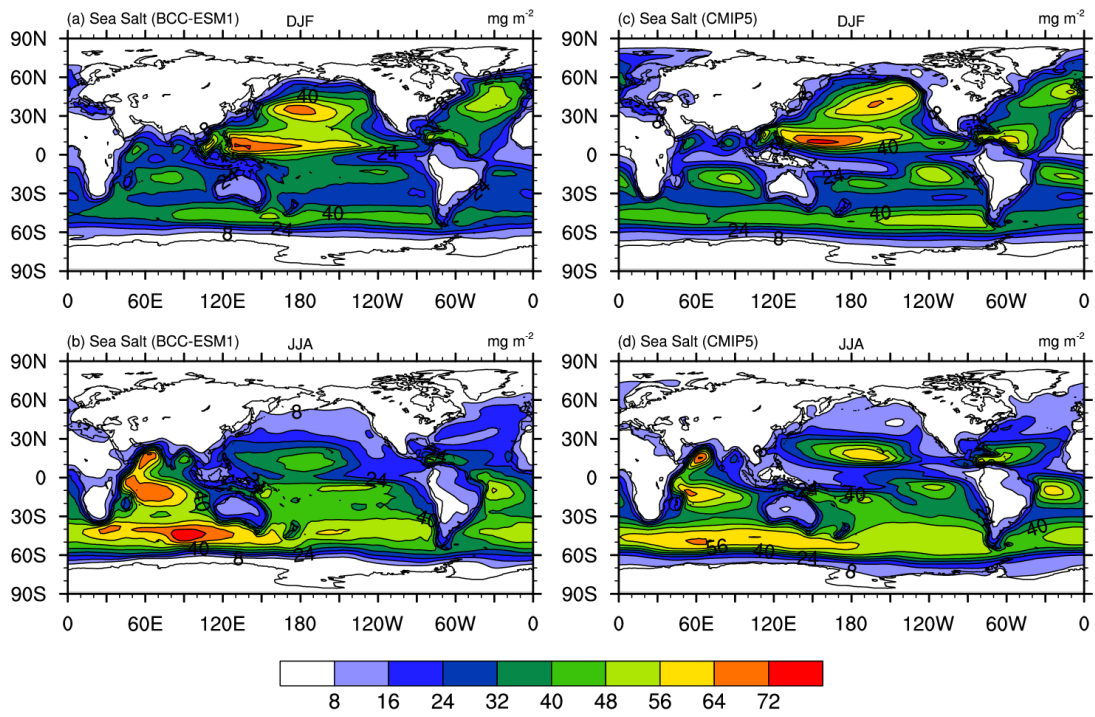


Figure 12. The same as in Figure 8, but for sea salt (SSLT) aerosol. Units: mg.m^{-2} .

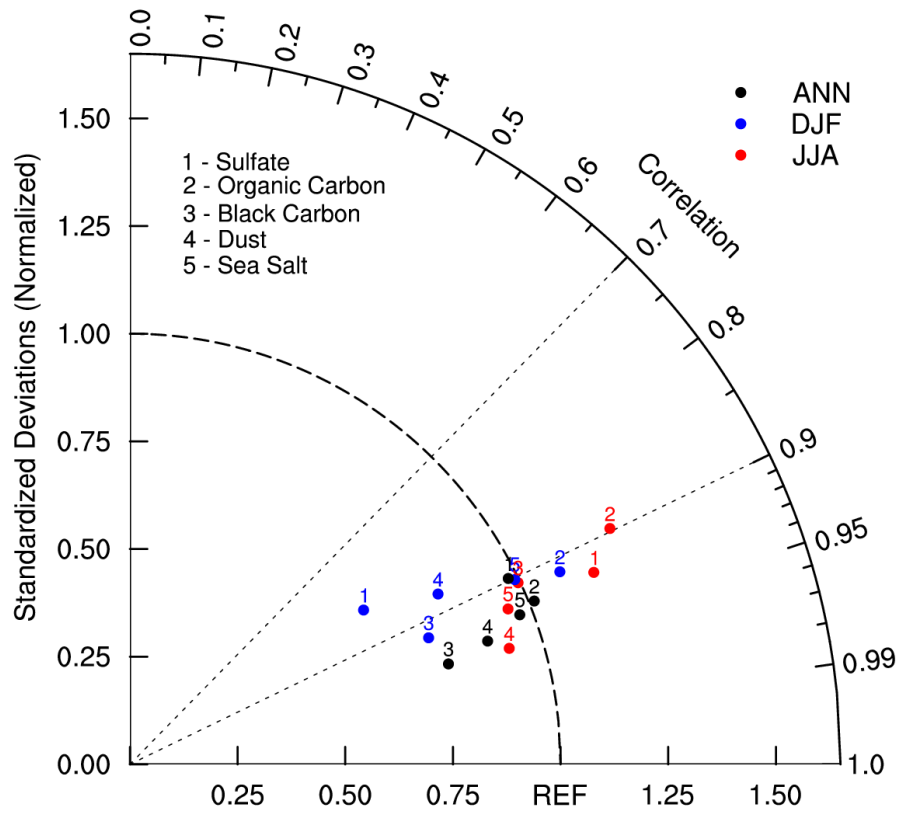


Figure 13. Taylor diagram for the global aerosols climatology (1971–2000) of sulfate, organic carbon, black carbon, dust, and sea salt averaged for December-January-February (DJF), June-July-August (JJA), and annual respectively. The radial coordinate shows the standard deviation of the spatial pattern, 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.

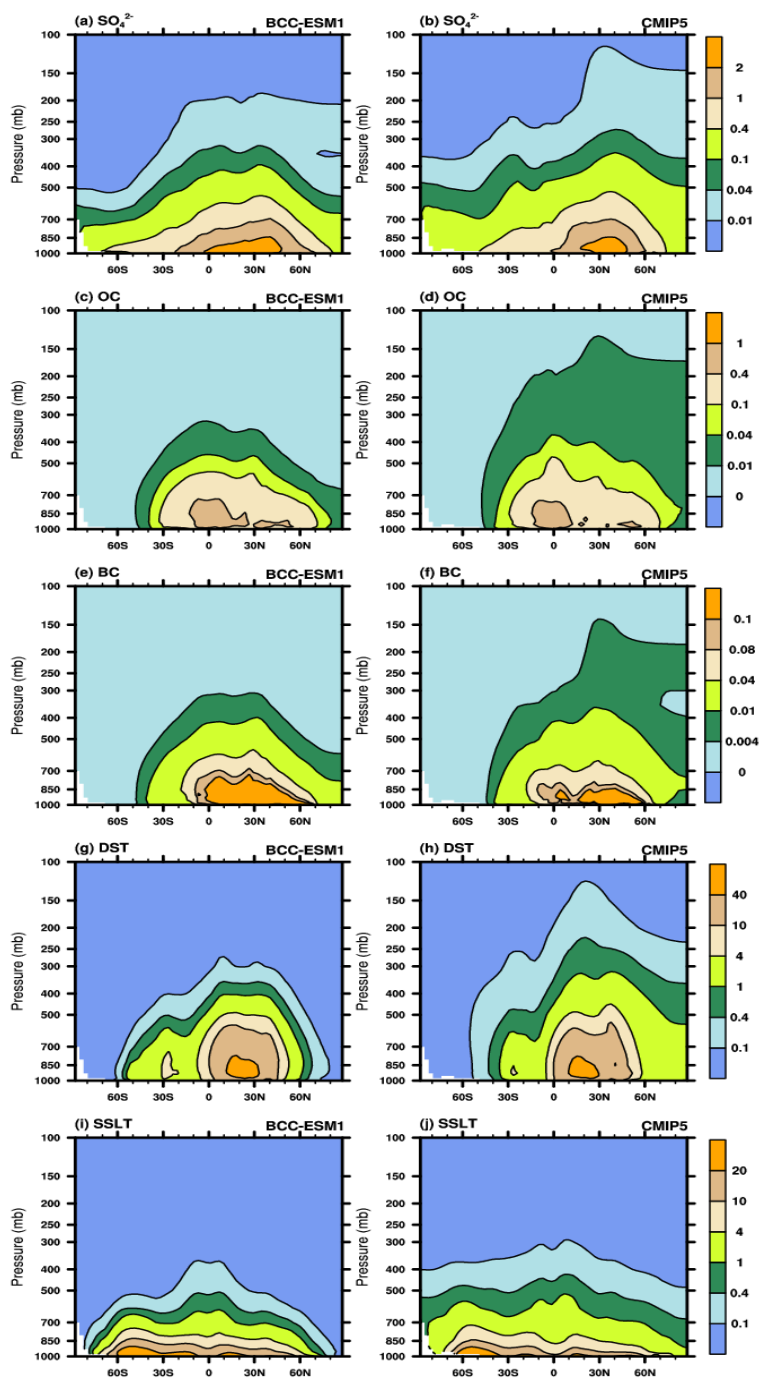


Figure 14. Latitude-pressure distributions of zonally-averaged annual mean sulfate, organic carbon, black carbon, dust, and sea salt aerosol concentrations for the period of 1971-2000. Left panels show the CMIP6 historical simulation of BCC-ESM1, and right panels the CMIP5 recommendation data. Units: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

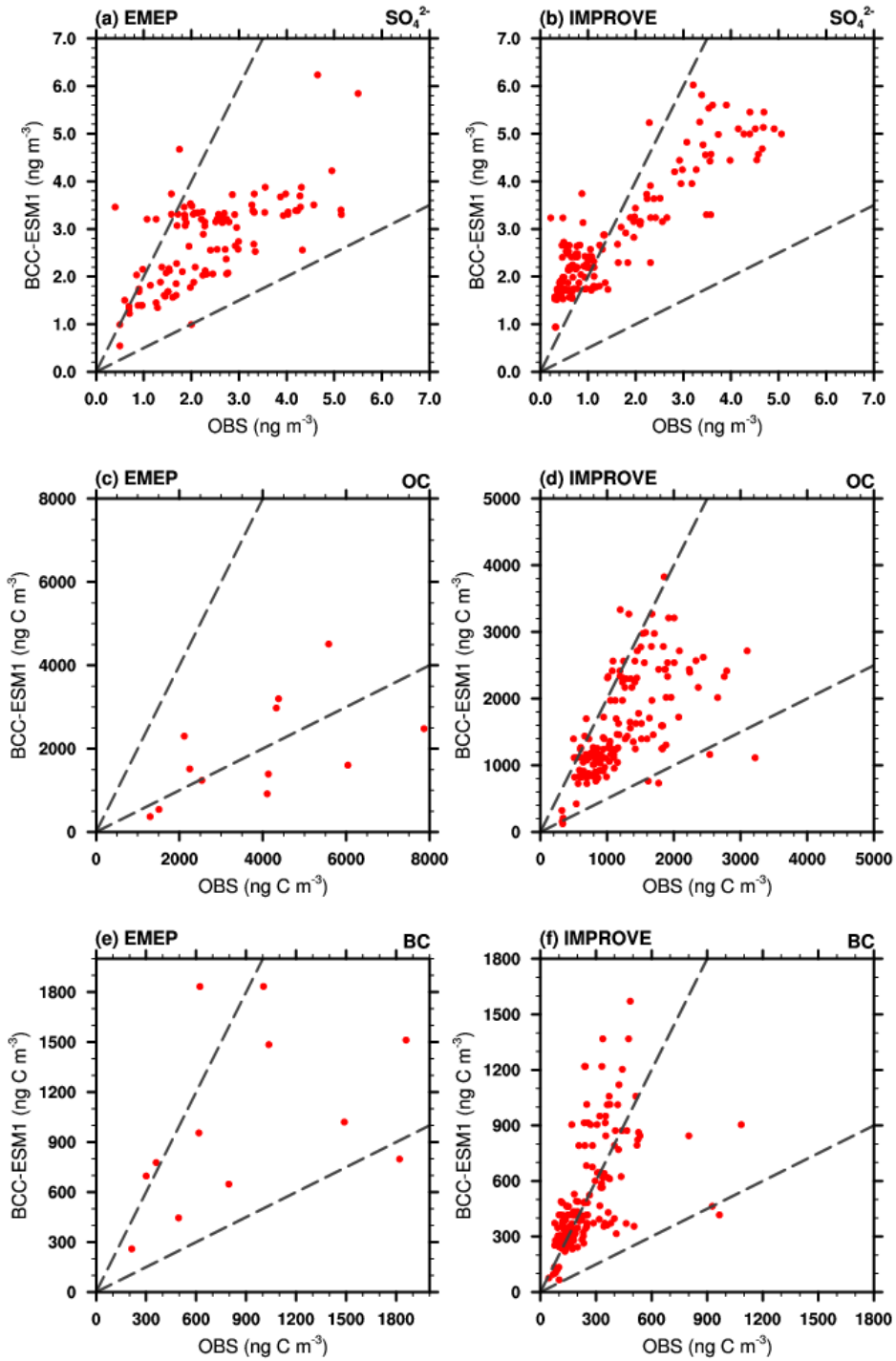


Figure 15. Scatter plots showing observed versus simulated multi-years averaged annual mean sulfate (SO_4^{2-}), organic carbon (OC), black carbon (BC) mixing ratios at IMPROVE and EMEP network sites. Observations are averages over the available years 1990–2005 for IMPROVE sites, and 1995–2005 for EMEP sites. Simulated values are those at the lowest layer of BCC-ESM1.

Table 7. Observed versus simulated concentrations of sulfate (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
	SO_4^{2-} (Obs/Model)	OC (OBS/Model)	BC (OBS/Model)	SO_4^{2-} (OBS/Model)	OC (OBS/Model)	BC (OBS/Model)	BC (OBS/Model)
Mean Values	2.37/2.74	3844/1919	884/1022	1.53/2.79	1215/1565	249/504	8.2/11.1
Std Deviation	1.16/0.93	1997/1215	572/526	1.30/1.20	572/745	164/296	27.9/21.0
Min Values	0.40/0.55	1296/369	214/259	0.22/0.94	322/123	45/66	0.0025/0.066
Max Values	5.50/6.24	7867/4510	1859/1834	5.07/6.02	3219/3827	1084/1570	558.91/267.11
Correlation (Obs and Model)	0.67	0.56	0.40	0.90	0.63	0.55	0.51

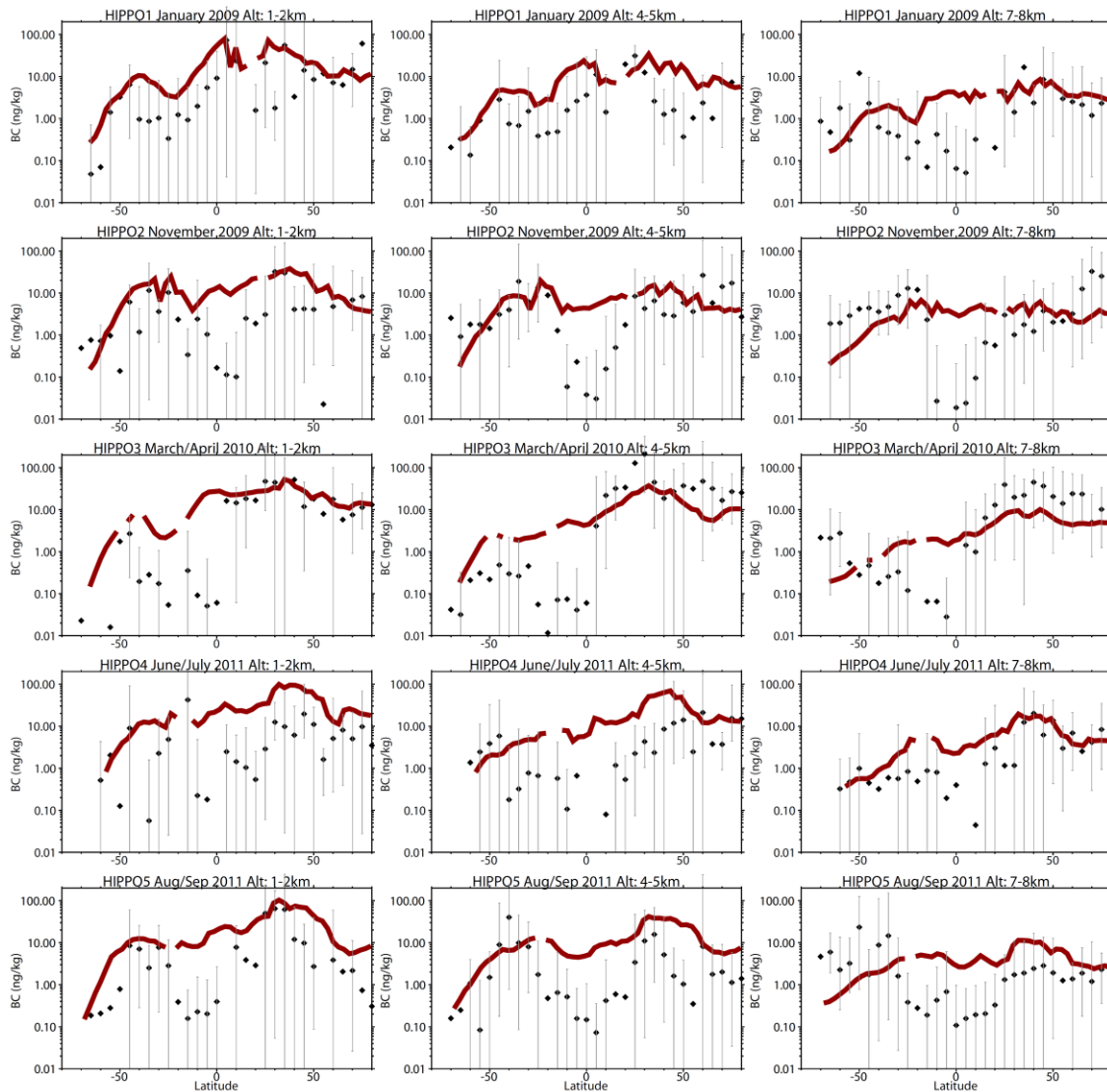


Figure 16. Comparison of modelled black carbon (BC) aerosol (red lines) with observations from HIPPO aircraft campaigns over the Pacific Ocean (black symbols, bars represent the full data range). Observations from different HIPPO campaigns were averaged over 5 °latitude bins and three different altitude 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.

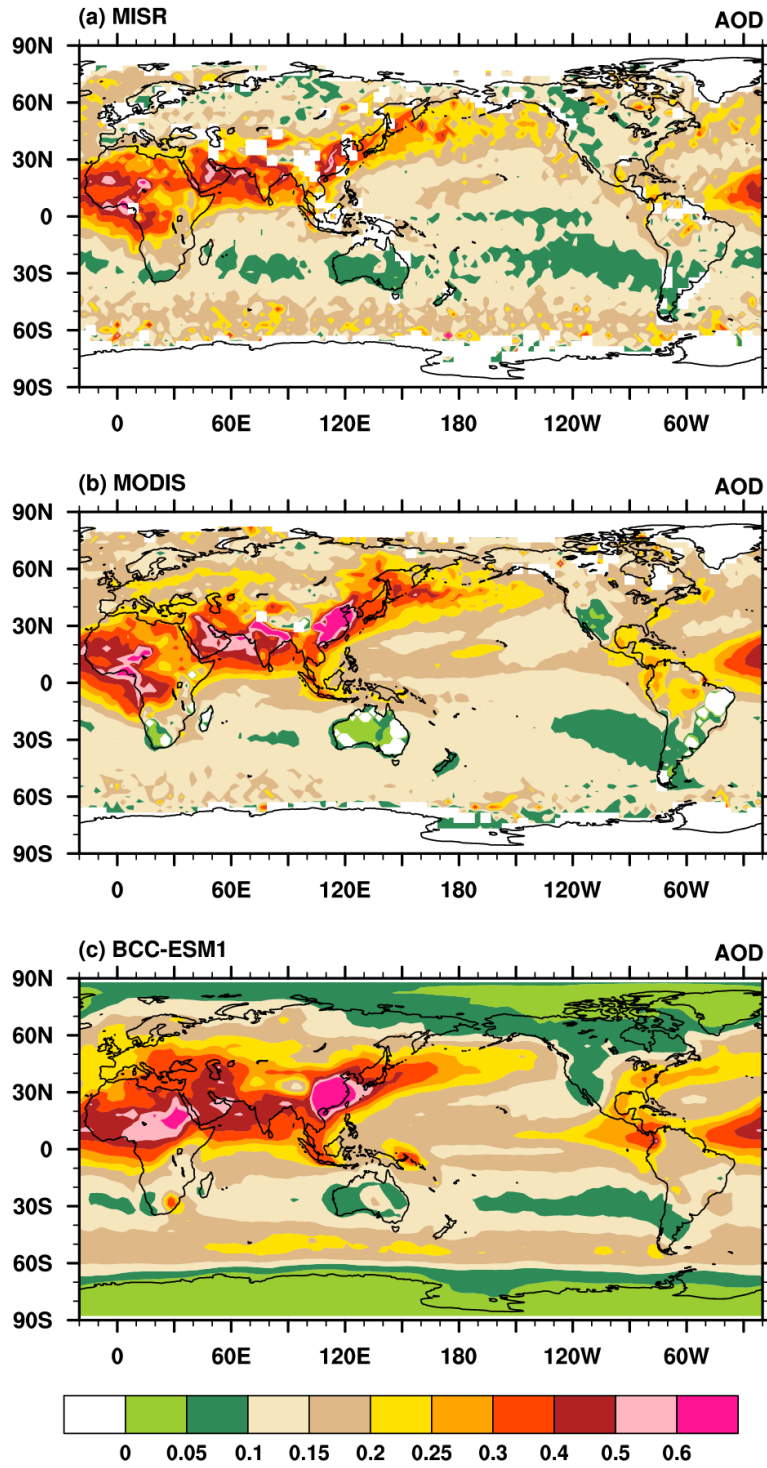


Figure 17. Global distribution of annual mean AOD simulated in BCC-ESM1 compared with the MISR and MODIS data for the year 2008.

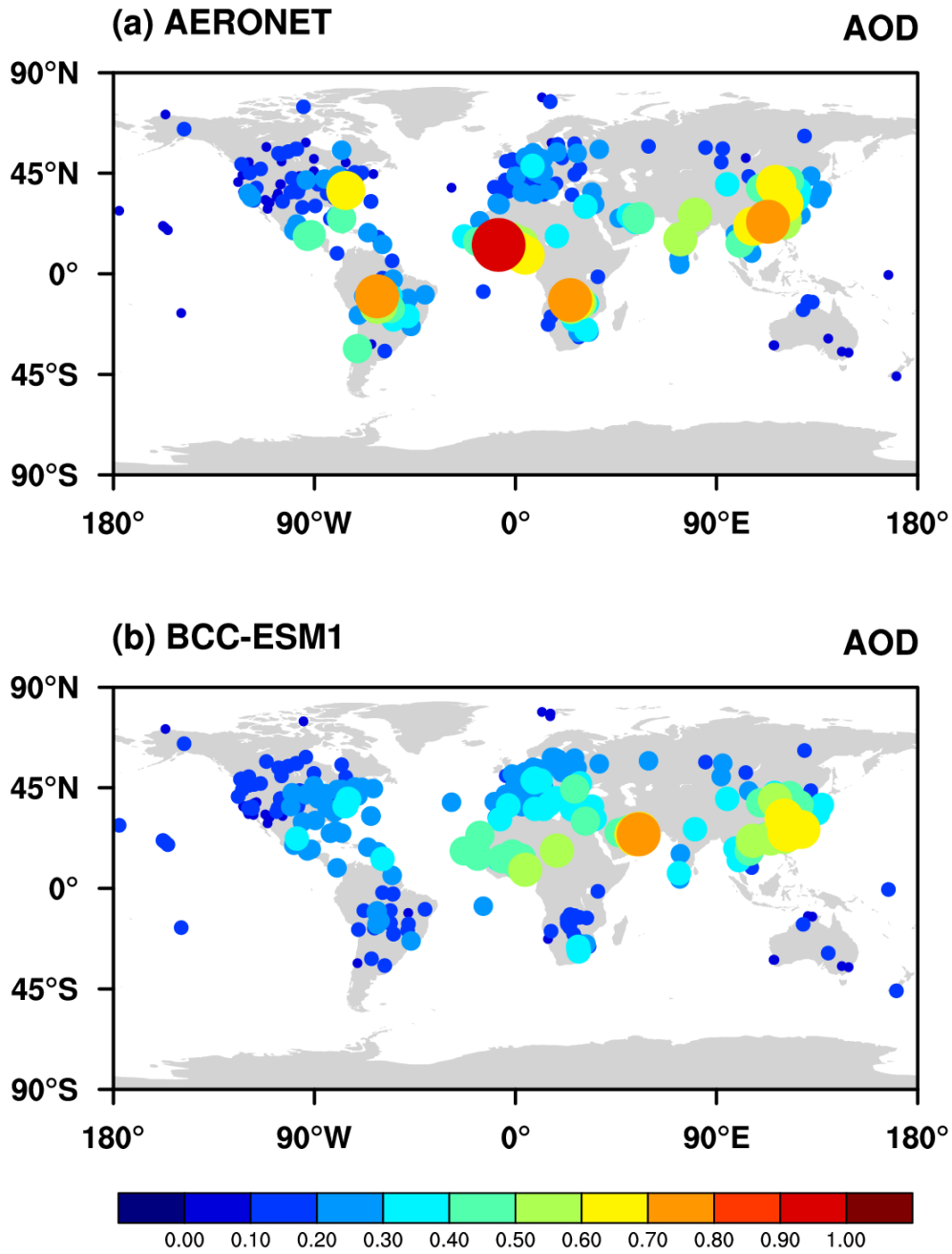


Figure 18. Observed versus simulated annual means of AOD at AERONET sites. Each data point represents the mean averaged for available monthly values of AOD. The dot sizes denote the magnitudes of AOD at sites. The spatial correlation is 0.56.

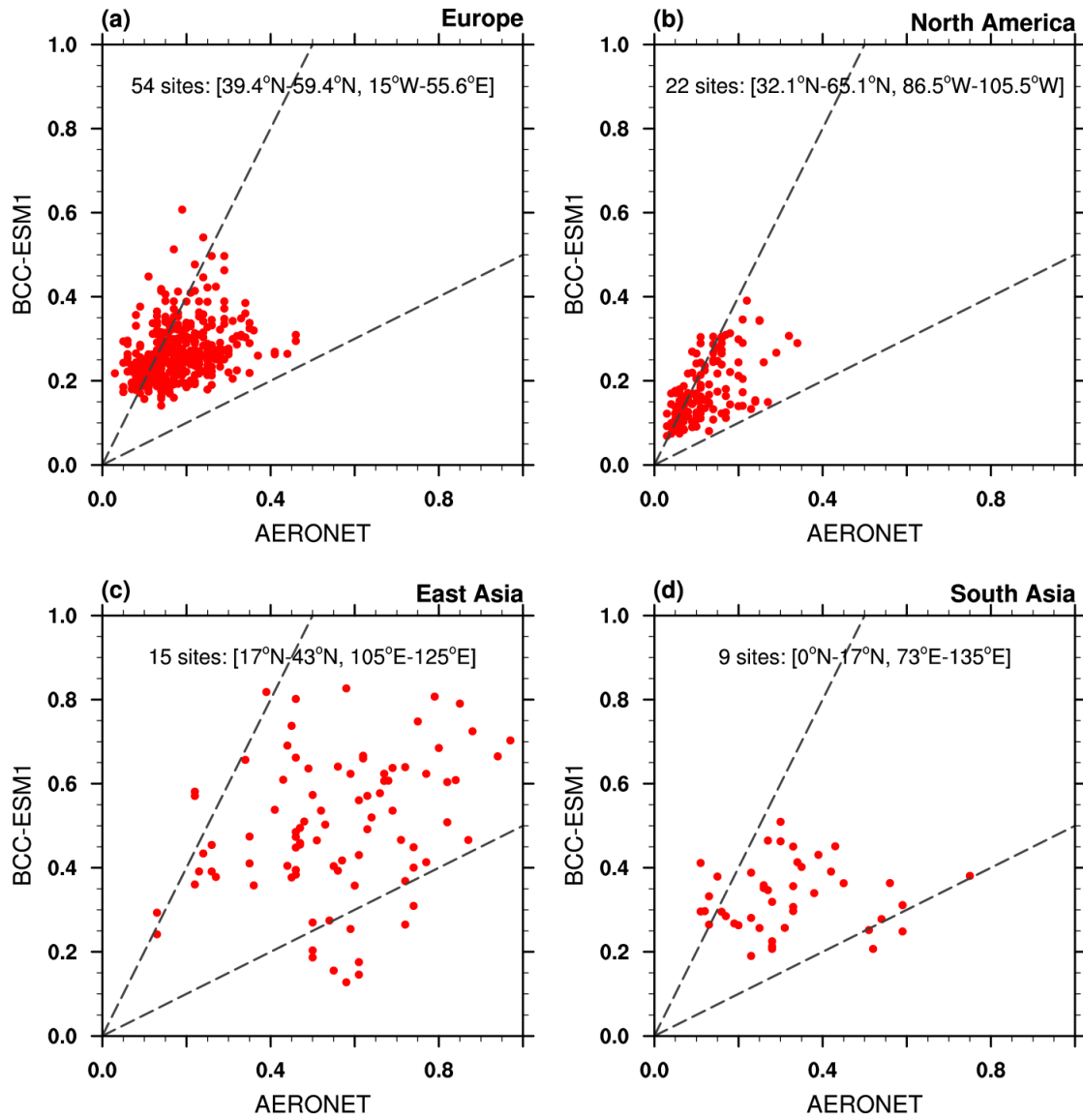


Figure 19. Scatter plots of observed versus simulated monthly mean AOD at AERONET sites in Europe, North America, East Asia, and South Asia over the period of 1998-2005.