

Response to Reviewer 1 (Dr. Jean-Francois Lamarque)

We thank Dr. Jean-Francois Lamarque for his insightful and constructive comments. We revised our manuscript accordingly.

This paper provides a description and evaluation of the aerosols in the BCC-ESM. The paper provides a reasonable overview of the model characteristics and sufficient comparisons to be useful. However, it suffers from a certain number of omissions and lack of details that should be fixed before publication moves forward.

My main concern in this paper is the statement at lines 316-318: “The whole system in BCC ESM1 fluctuates around $+0.7\text{Wm}^{-2}$ net energy flux at TOM without obvious trend in 600 years (Fig. 1b), and the global mean surface air temperature shows only a small warming (Fig. 1a)”. If this is the case, then there is a real problem with this model. There cannot be a significant TOA imbalance without a significant trend in surface temperature, unless the ocean is taking up all that excessive forcing. Which would mean huge drifts in the mean ocean temperature. The authors need to clearly identify if this is a mistake, or the difference between TOA and TOM, or whether there is a drift in ocean temperatures. But as stated, this means there is a huge non-conservation of energy in the model.

We apologize for the confusion. TOM (top of model) should be TOA (top of atmosphere). We double checked our data used and there was indeed an imbalance of net energy flux at TOA. In order to verify whether there is a drift in ocean temperatures, Fig.1c representing the variation of global SST is added. It seems that the ocean is stable, at least for its upper layer. If we refer to other models of similar complexity, it seems that a small imbalance commonly exists (Hansen et al., 2005; Wild et al., 2013) and an average of 1.0Wm^{-2} of imbalance is among the CMIP5 models (Wild et al., 2013). In the revised manuscript, we rewrote this paragraph (in lines 390-393 in the revised manuscript) as “This level of TOA energy imbalance is close to the average imbalance (1.0Wm^{-2}) among CMIP5 models (Wild et al., 2013), and does not cause remarkable climate drift in BCC-ESM1. The global mean TAS and SST keep around 288.1K (Fig. 1b) and 295.05K (Fig. 1c) respectively.”

Another concern is that the authors make considerable use of the CMIP5 concentrations (by the way, a correct reference to this data would be Lamarque et al., ACP, 2010), which is a somewhat circular evaluation. Indeed, the CMIP5 data were generated using a chemistry model very similar to the one used in BCC-ESM1. It is true that the emissions are different, but then the main evaluation this analysis provides is on the similarity of emissions. I would therefore encourage the authors to expand their model evaluations to include more observations. For example, the paper <https://www.geosci-model-dev.net/9/1853/2016/gmd-9-1853-2016.pdf> includes analysis against aircraft observations. While I understand that the focus is on aerosols, it cannot be ignored that the rate of formation of sulfate is dependent on the levels of oxidants in the troposphere. It would therefore be very useful if some documentation and evaluation of oxidants (at the very least ozone) is included in the paper.

We appreciate your very relevant comments. The right reference of Lamarque et al. (2010) is now used in the revised manuscript. We also agree entirely that the oxidation capacity should be evaluated, and we followed your suggestion by comparing the simulated O₃ in the 20th century against CMIP6 prescribed data and global ozonesonde observations from WOUDC. We added a new section “4. Evaluation of O₃ and aerosols simulation in the 20th century”. Furthermore, a comparison of BC simulations against HIPPO BC aircraft observations is also added in “4.3 Global aerosol distributions at present day”.

Minor comments

1. Lines 155-157: why is convective transport not considered?

Vertical transport of gas tracers and aerosols due to deep convection is not yet included in the present version of BCC-AGCM3-Chem, which process is considered as a part of the deep convection and occurs generally in a small spatial region on a GCM-box with low-resolution (2.8°lat.×2.8°lon.). Another consideration is that a large uncertainty exists to treat transport of those water-soluble tracers by deep convection. We are working on this issue. This effect will be involved in the next version of BCC model. We feel it is important to mention it since we are aware that the issue can partly matter for the quality of results shown in this manuscript. We added this explanation in lines 146-151 in the revised manuscript.

2. Lines 189-191: Following the work done in CAM4, it would be quite straightforward to include some basic representation of NH₃ chemistry (see Lamarque et al., GMD, 2012, section 5).

We apologize for this mistake about NH₃. In fact, a previous version of BCC-ESM did not include NH₃. But in the frozen version of BCC-ESM1 that is used in this work, NH₃ is indeed a prognostic variable following CAM4 (Lamarque et al., GMD, 2012). So, we added some description about NH₃ in “2.1 SO₂, DMS, NH₃, and Sulfate” and Table 1, Table 2, and Table 4 in the revised manuscript.

3. Line 207: reference Hoesly et al.

In the revision, we have added the reference of Hoesly et al. (2018) in line 223 in the revised manuscript.

4. Lines 251-254: this is an important aspect of the model that needs more discussion. In particular, what is the aerosol indirect effect in this model?

In the revision, we have added a paragraph in “2.5 Effects of aerosols on radiation, cloud, and precipitation” to describe the treatment of aerosol indirect effect in BCC-ESM1.

5. Line 257 (and other places): it is AerChemMIP, not AeroChemMIP

In the revision, we changed “AeroChemMIP” to “AerChemMIP”.

6. Lines 273-276: which emissions are those? The CMIP6 (as the CMIP5) had all emissions

necessary for tropospheric chemistry, as long as some splitting of lumped emissions (like total VOC emissions) were performed.

In the revised manuscript, we added more details for this issue in lines 323-337.

“Most historical emissions from anthropogenic source (surface, aircraft plus ship) and biomass burning from 1850 to 2014 are CMIP6-recommended data (Hoesly et al., 2018; available at <https://esgf-node.llnl.gov/search/input4mips>). Anthropogenic or biomass burning sources of some tracers which are not included in the CMIP6 dataset (see Table 4), anthropogenic emission of H₂ and N₂O are from monthly climatological dataset provided by the MOZART-2 standard package. N₂O is a prognostic variable in BCC-ESM1 but it is replaced by CMIP6 prescribed concentration in the historical run. Other emissions including biomass burning (CH₃COCH₃) and anthropogenic emission (CH₃CHO, CH₃OH, and CH₃COCH₃) are from the IPCC ACCMIP emission inventory (<http://accent.aero.jussieu.fr/ACCMIP.php>) covering the period from 1850 to 2010 with 10-year intervals (see Table 4). Monthly lumped emissions of black carbon and organic carbon aerosols from 1850 to 2014 are downloaded from CMIP6-recommended data, but we used 80% (for BC) and 50% (for OC) of them in their hydrophobic forms (BC1 and OC1) and the rest in their hydrophilic forms (BC2 and OC2), following the work of Chin et al. (2002).”

We check the CMIP6 data website again and cannot find anthropogenic emission data of H₂ and N₂O provided.

7. Line 288: volcanic, not volcano

In the revision, we have corrected to “3.2 Volcanic eruption, lightning and aircraft emissions”

8. Line 290: this is confusing. It is really not clear that stratospheric aerosols are represented in this model. Are those really stratospheric emissions, or tropospheric emissions of the non-eruptive volcanoes?

We apologize for the confusion. We don’t have stratospheric chemistry scheme, and no stratospheric emissions at all. That statement in the initial manuscript indicates surface emissions from non-eruptive volcanos. In the revised manuscript, we rewrote the corresponding paragraph in “3.2 Volcanic eruption, lightning and aircraft emissions”. It reads in lines 354-360 as “As there is no stratospheric aerosol scheme in BCC-ESM1, concentrations of sulfate aerosol at heights from 5 to 39.5 km, which volcanic origin, are directly prescribed using the CMIP6-recommended data (Thomasson et al., 2018) from 1850 to 2014. The effects of surface SO₂ emissions from volcanic eruption on the variation of SO₂ in the atmosphere and then on the variation of tropospheric SO₂- concentration are considered, and the SO₂ emissions from 1850 to 2014 are downloaded from the IPCC ACCMIP emission inventory (<http://accent.aero.jussieu.fr/ACCMIP.php>).”

9. Line 293: what are the total NO_x emissions from lightning (in TgN/year)?

The globally-averaged mean of the total NO_x emissions from lightning during the period of 1850 to 2014 is 5.19 Tg (N)-yr⁻¹. It is in agreement with observations within the range of 3 to

6 Tg(N) yr⁻¹ (Martin et al., 2002). In the revised manuscript, we modified the corresponding description in “3.2 Volcanic eruption, lightning and aircraft emissions”

10. Lines 301-303: this is not clear. Are you describing the relaxation time (of 10-days) of the concentrations towards the climatology? Is the climatology changing over the course of the historical period?

Yes, we are describing the relaxation time (10 days) that we used to relax different chemical variables toward their monthly and zonal mean climatological values, prescribed in the top two layers. During the revision, we rewrote the corresponding paragraph in “3.3 Upper boundary of the atmosphere” in lines 371-379 as

“Concentrations of different tracers (O₃, CH₄, N₂O, NO, NO₂, HNO₃, CO, and N₂O₅) at the top two layers of the model are set to prescribed monthly climatological values, and concentrations from below the top two layers to the tropopause are relaxed at a relaxation time of 10-days towards the climatology. Climatological values of NO, NO₂, HNO₃, CO and N₂O₅ at the top two layers are extracted from MOZART2 data package available at the Website (<https://www2.acom.ucar.edu/gcm/mozart-4>), originated from the Study of Transport and Chemical Reactions in the Stratosphere (STARS, Brasseur et al., 1997). Concentrations for the other tracers (O₃, CH₄, and N₂O) at the top two model layers are the zonally-averaged and monthly values from 1850 to 2014 derived from the CMIP6 data package.”

11. Line 337: there are some anthropogenic/biomass SO₂ emissions in 1850, just small ones. *Yes, that is true, anthropogenic emissions were not entirely negligible, although small in 1850. During the revision, we reformulated the corresponding paragraph in lines 411-415.*

“We can compare them with CMIP5 recommended concentrations in year 1850, considered as the reference state in the pre-industrial stage. At that time, there are fewer anthropogenic/biomass SO₂ emissions, the SO₄ over land are evidently smaller than those over oceans especially over the tropical Pacific and Atlantic Oceans, where DMS can be oxidized to SO₂ and then form SO₄”

12. Line 373: the correlation really only reflects that the lifetime of SO₂ is very short and not changing much, and therefore the burden will directly follow the emissions.

Yes, we agree entirely with this remark. We modified the descriptions in lines 481-484 as “Due to increasing SO₂ emissions from 1850 to present day (Fig. 6), the global SO₂ burden in the atmosphere increased from 100 Tg in 1850s to 200 Tg in 1980s (Fig. 7a), and has a high correlation coefficient of 0.996 with the anthropogenic emissions (Fig. 6a), as the lifetime of SO₂ is short. The burden directly followed the emission”.

13. Line 376: what is the “NCAR data package”?

It is MOZART2 package and corrected in lines 485-487 to “Its natural emissions from oceans from 1850 to 2010 in the model are the climatological monthly means (Dentener et al., 2006) from MOZART2 data package.” in the revised manuscript.

14. Line 400 (and others): a lot of analysis compares to Liu et al (2005). It would be useful to

include more publications, especially more recent ones.

In the revision, we have added more comparison with recent publications such as Liu et al. (2016), Matsui and Mahowald (2017), Tegen et al. (2019) in “4.2 Global aerosols budgets”.

15. Figure 5: why is the BCC ESM1 data also shown as 10-year averages? Also, are those the results of a single ensemble member? More details on the simulation would be useful; in particular I am assuming that this is a fully coupled simulation.

The 10-year averaged from BCC-ESM1 data used in the previous version of manuscript is only based on consideration for intercomparison with the 10-year interval CMIP5 data. In the revised manuscript, we updated those using the yearly mean simulations (Figure 5 is numbered to Figure 7 in the revised version).

Response to Referee #2

Interactive comment on “Beijing Climate Center Earth System Model version 1 (BCC-ESM1): Model Description and Evaluation” by Tongwen Wu et al.

General

The paper presents a description and an evaluation of the tropospheric aerosols included in the Beijing Climate Center Earth System Model. The paper consists of a quite general overview and my main concerns relate firstly to the description of the aerosol scheme which is rather vague in several parts. Furthermore, it is not so clear what is specific or not to this aerosol scheme, what has been developed and/or adjusted, compared to other schemes already in place in other climate models. Secondly, the evaluation is mostly qualitative, and when some quantitative information is provided, it often refers to quite old references.

Both issues needs to be addressed quite thoroughly for the paper to progress in the review process. This requires quite some work. My list of particular points appears below.

We thank reviewer #2 for his/her carefully reading our manuscript. We have revised the manuscript accordingly. We showed more details and descriptions of the aerosol scheme, and added an evaluation for O3 simulation which is helpful to complement aerosols. We also presented more quantitative information, and comparisons with recent observation and references.

Questions/remarks

1. paragraph “Model description”: this paragraph needs some rewriting as some features, eg ACGM3, AVIM2, are presented twice

We have rewritten this paragraph in “2. Model description”. The components of the atmosphere, the land, the ocean and the sea ice in BCC-ESM1 are described in separate paragraph.

2. L146: what is the reference for the “weighted-combination”? Please provide more details.

In the revision, we have reworded this sentence to make our point clearer and rewritten this paragraph in lines 127-137 as “Dry deposition velocities for the 15 trace gases including O3, CO, CH4, CH2O, CH3OOH, H2O2, NO2, HNO3, PAN, CH3COCH3, CH3COOOH, CH3CHO, CH3COCHO, NO, and HNO4 are not computed interactively and directly interpolated from MOZART2 climatological monthly mean deposition velocities ([https://en.wikipedia.org/wiki/MOZART\(model\)](https://en.wikipedia.org/wiki/MOZART(model))) which are calculated offline (Bey et al., 2001; Shindell et al., 2008) using a resistance-in-series scheme originally described in Wesely (1989). The dry deposition velocities for the other 15 species including MPAN, ONIT, ONITR, C2H5OH, POOH, C2H5OOH, C3H7OOH, ROOH, GLYALD, HYAC,

CH3OH, MACROOH, ISOPROOH, XOOH, HYDRALD, and H2 are calculated using prescribed deposition velocities of O3, CO, CH3CHO, or land surface type and surface temperature following the MOZART2 (Horowitz et al., 2003)."

3. L156: is turbulent transport included? if not, then you are missing sub-grid scale transport and the overall distribution of chemical species would be quite different considering this sub-grid scale transport. Please explain what is your rationale for presenting an evaluation without these processes.

Yes, Vertical transport of gas tracers and aerosols due to deep convection is not yet included in the present version of BCC-AGCM3-Chem, which process is considered as a part of the deep convection and occurs generally in a small spatial region on a GCM-box with low-resolution (2.8°lat. ×2.8°lon.). Another consideration is that a large uncertainty exists to treat transport of those water-soluble tracers by deep convection. But this effect will be involved in the next version of BCC model. We have added those expressions in lines 146-151.

4. L173: the Wesely approach has 3 terms. Why did you retain only two terms? Please indicate if you compute the terms interactively or not. This is at the moment not clearly stated.

Yes, the Wesely (1989) approach has 3 terms. We have cleared clarified this in lines 184-187 in the revised manuscript as "The dry deposition velocity of SO2 follows the resistance-in-series approach of Wesely (1989) using the formula, $W_{SO2} = 1/(r_a + r_b + r_c)$, in which r_a , r_b , and r_c are the aerodynamic resistance, the quasi-laminar boundary layer resistance, and the surface resistance, respectively and they are interactively computed in each model time step."

5. L179: it seems to me that the reactions listed in Table 2, and their reaction rates, are the same as the ones that appear in Lamarque et al. 2012. This should be noted in the paper, as therefore both the chemistry and the aerosol modules of the BCC-ESM1 and CAM-Chem used for generating what the authors refer as the "CMIP5 recommended" aerosol concentrations are quite similar. This should be made quite clear in the paper. Possibly a paragraph in the paper could be dedicated to what is specific to this scheme, if this is relevant.

Yes, the reactions listed in Table 2 are referred to CAM-Chem (Lamarque et al., 2012). We have rewritten the description in lines 178-180 in the revised manuscript as "The present version of aerosol scheme belongs to a bulk aerosol model and mainly refers to the scheme of CAM-Chem (Lamarque et al., 2012), but the nucleation and coagulation of aerosols are still ignored."

6. L182: there is no reference to DMS in Benkowitz 1996. Please clarify what you mean.

That is our confusion about the reference. In the revision of the manuscript, we have reworded this sentence in lines 193-196 as "The main source of DMS is from oceanic emissions via biogenic processes. It is prescribed with the climatological monthly data that are extracted from MOZART2 package (<https://www2.acom.ucar.edu/gcm/mozart-4>)."

7. L191: is there a reference for this assumption?

That is related to “NH3”. In the first version of manuscript, we make a mistake about NH3. In fact, the previous version of BCC-ESM did not include NH3 simulation in the chemistry scheme. But in the frozen version of BCC-ESM1 that is used in this work, NH3 is already set as a prognostic variable following CAM4 (Lamarque et al., GMD, 2012). So we added a description about NH3 in “2.1 SO2, DMS, NH3, and Sulfate” and Table 1, Table 2, and Table 4 in the revised manuscript.

8. L215: please clarify why in this paragraph about OC and BC your write about “soluble gases”?

We have rewritten the description in line 226-228 as “OC2 and BC2 are soluble aerosols, and their sinks are primarily governed by wet deposition. Their in- and below-cloud scavenging follows the scheme of Neu and Prather (2011)”.

9. L224: what are the values of this scaling factor?

We have clarified it in lines 236-237 as “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 bins of sea salt aerosols (Table 4), respectively.”

10. L252: Wu et al 2019 is not in the list of references; and what do you mean by “it is parameterized”, what is “it”? Do you refer to the aerosol first indirect effect or to the first and second effects? Please provide further details, in particular if you parameterize the second indirect effect of aerosols that not all climate models consider

We have added Wu et al 2019 in the list of references, and added a paragraph in “2.5 Effects of aerosols on radiation, clouds, and precipitation” to describe the treatment of aerosol indirect effect in BCC-ESM1. In the first version of manuscript “it is parameterized” means “liquid cloud droplet number concentration is parameterized”. Its details are added.

11. L257: “historical” is not an AerChemMIP simulation but rather a CMIP6 simulation that will be a basis for a large number of CMIP6 analyses, including some AerChemMIP analyses, but also other MIPs analyses. Please correct this wording throughout the document. If the simulation you present is an historical CMIP6 simulation, please indicate the baseline name of the corresponding files on the ESGF. Do you present one ensemble member or several members?

We have rewritten description about “historical” experiment in “3. Experiment design for the 20th century climate simulation”. It followed the historical simulation protocol designed by CMIP6 (Eyring et al., 2016) which is named as “historical” in the Earth System Grid Federation (ESGF). The protocol details the historical experiment forced with emissions evolving from 1850 to 2014 refer to Collins et al. (2017). Three members of historical experiments are conducted and the first member is analyzed in this work.

12. L264: “only O₃ is a prognostic variable”: what about CH₄, it is part of the chemistry scheme and therefore it is also a prognostic variable isn't it? what about also CO₂?

CH₄ and N₂O may be selected as prognostic variables. But both are suggested in AerChemMIP to take prescribed values for the historical experiment. CO₂ is also prescribed using CMIP6 historical forcing data. We have clarified this point in lines

307-312 in the revised manuscript.

13. L274: the CMIP6 anthropogenic emissions are meant to cover all that is required for a climate model. Can you explain why this was not the case for your model?

Anthropogenic emissions for most tracers are available in the CMIP6 data. But we cannot find anthropogenic emission data for H2 and N2O that we need. The details about the emission data used are given in the revised manuscript.

14. L276: to my knowledge there is no such CMIP6 recommendation for hydrophobic and hydrophilic forms. Please rephrase your sentence.

Yes, there is no such CMIP6 recommendation for hydrophobic and hydrophilic forms. So, we use monthly lumped emissions of black carbon and organic carbon aerosols and then we divided them separately to 80% of BC and 50% of OC emitted in their hydrophobic forms (BC1 and OC1) and the rest being in their hydrophilic forms (BC2 and OC2) following the work of Chin et al. (2002). This is cleared in lines 333-337.

15. L279 and following: please describe in more details the formation of Secondary Organic Aerosol from vegetation that you consider? what comes out of MEGAN2.1, are they related to OC2 only, and not OC2 and OC1? ...

OC does not belong to biogenic volatile organic carbons (VOCs). The hydrophilic organic carbon (OC2) can be formed from natural biogenic volatile organic compound (VOC) emissions. It is calculated online in the land component model BCC-AVIM2 and assumed to equal to 10% of monoterpenes emission following the algorithm of Chin et al. (2002). Those expressions are added in lines 348-352.

16. L291: factor 2-4 high: this is a strong affirmation! The Ge et al. 2016 study is older than the CMIP6 data. How do they relate? And furthermore, do you have a stratospheric aerosol scheme that uses these data? If yes, please describe the scheme, if not please clarify your sentence.

The work of Ge et al. 2016 is not mentioned, and this statement is now removed in the revised manuscript. As for stratospheric aerosol, we only considered SO4. We have rewritten this paragraph in lines 354-360 as “As there is no stratospheric aerosol scheme in BCC-ESM1, concentrations of sulfate aerosol at heights from 5 to 39.5 km, which volcanic origin, are directly prescribed using the CMIP6-recommended data (Thomasson et al., 2018) from 1850 to 2014. The effects of surface SO2 emissions from volcanic eruption on the variation of SO2 in the atmosphere and then on the variation of tropospheric SO42-concentration are considered, and the SO2 emissions from 1850 to 2014 are downloaded from the IPCC ACCMIP emission inventory (<http://accent.aero.jussieu.fr/ACCMIP.php>).”

17. L304: please clarify what the MOZART2 data package include, data? Chemistry code?...

We have clarified those in lines 374-376 as “Climatological values of NO, NO2, HNO3, CO and N2O5 at the top two layers are extracted from MOZART2 data package available at the Website (<https://www2.acom.ucar.edu/gcm/mozart-4>), originated from the Study of Transport and Chemical Reactions in the Stratosphere (STARS, Brasseur et al., 1997).”

Yes, MOZART2 data package includes data and chemistry code.

18. L307: to my knowledge the CMIP6 data package does not include neither CH₄, nor N₂O: what do you refer here to?

We have checked them. The CMIP6 data package includes zonally and monthly values of CH₄ and N₂O.

19. Table 1 and Table 4: there are incoherences between species listed in both Tables. For example, CH₃COCHO is not emitted in Table 1 and has emissions in Table 4. Please carefully check consistency between these tables.

We have corrected the incoherence between Tables 1 and 4.

20. L318: “only a small warming”: please quantify this

We have rewritten this paragraph in lines 381-395 of the revised manuscript and added the time series of global SST in Figure 1.

21. L324: mean and uncertainty should not be of different orders. Please correct here and in other places in paper.

We have corrected those expressions in “3.4 The preindustrial model states”

22. L331: these are not concentrations but rather loads, and what is the reference for these “CMIP5 recommended concentrations”?

Figures 2a-2c show the time series of global annual total masses in the troposphere (integrated from the surface to 100 hPa) in the last 450 years of the piControl. It is derived from CMIP5 recommended concentrations. The reference of CMIP5 data is Lamarque et al. (2010) and has added in the text.

23. L338: why do you think there is such a distribution?

We added some words about the distribution of SO₄2- in year 1850 in lines 411-415 of the revised manuscript as “We can compare them with CMIP5 recommended concentrations in year 1850, considered as the reference state in the pre-industrial stage. At that time, there are fewer anthropogenic/biomass SO₂ emissions, the SO₄ over land are evidently smaller than those over oceans especially over the tropical Pacific and Atlantic Oceans, where DMS can be oxidized to SO₂ and then form SO₄.”

24. L350: in addition to pointing out similarities, please address differences between CMIP5 and BCC-ESM1 outputs, and why there are such differences/similarities

We have added sentences in lines 428-432 as “Relative lower relations for sulfate, black carbon and organic carbon are possibly caused as different anthropogenic emission sources are used in BCC-ESM1 and to create CMIP5 data. Dust and sea salts belong to natural aerosols and depend on the land and sea surface conditions, so their spatial distributions are easy to be captured and have relatively higher correlations between CMIP5 data and BCC-ESM1 simulations.”

25. L376: what is this particular "NCAR data package"

We have corrected it in lines 485-487. It is MOZART2 data package.

26. L378: sentence "This decrease trend possibly results from the prescribed emissions have not year-to-year variations and ..." is not clear

It is modified in lines 487-491 as "As shown in Fig 7a, the global amount of DMS in the whole atmosphere was about 0.12 Tg during 1850-1900 and decreased to 0.055 Tg in 2010. This decrease trend maybe partly results from the speeded rate of DMS oxidation with global warming, and the loss of DMS gradually exceeds the source of ocean DMS emission to cause a net loss of DMS in the atmosphere since 1910s"

27. L386: the sentence "The trends of global BC and OC burdens are similar to that of sulfate, but they showed continuous increases from 1950 to present." is not clear

This sentence is modified in lines 496-499 of the revised manuscript as "As for global BC and OC burdens, BCC-ESM1 results show continuous increases since 1850s, especially from 1950 to present. From 1910's to 1940's, the CMIP5 data show a slight decrease of BC and OC burdens in the atmosphere."

28. L390: "was slightly enhanced from 1950 to 2000" : I rather see a similar burden in 1950 and in 2000. Please be clearer, and do you have evidence of increasing soil dryness during that period?

We have corrected its description in lines 501-504. Global dust burden in the period from 1980 to 2000, not from 1950 to 2000, shows evident increase. The details about the temperature and soil moisture in drought areas will be explored in other paper.

29. L400: "largely due to stronger wind speed": differences could be due to differences in underlying DMS concentrations in the oceans. What supports your affirmation?

DMS emission from the ocean is computed by wind near the sea surface. We have not compared the wind simulations in BCC-ESM with the data used in Liu et al. (2005). So, we cancelled the original description to account for their difference of DMS emission from oceans between BCC-ESM1 and the values in Liu et al. (2005).

30. L406: air traffic is part of anthropogenic activities; please rephrase your sentence, and what about biomass burning emissions? biomass burning emissions, SO₂ from volcanic eruption?

We have modified descriptions in lines 512-516. There are three parts of SO₂ source listed in Table 5. One is produced from the DMS oxidation, the second is from airplane emissions to the atmosphere, and the rest included emissions from anthropogenic activities and volcanic eruption at surface.

31. L407: you indicate that volcanic emissions are not included. I wonder in Figure 3 what corresponds to the area of large loads of sulfate around Central America?

Corrected it. Volcanic emission of SO₂ at surface is included.

32. L423: it seems that the total of 45.2 Tg/yr for OC is incoherent with what appears in Figure 4b; please correct.

In the Table 6, the units of OC sources and sinks are Tg (OM)/yr in order to compare with the data of Liu et al. (2012), and assumed OC equal to OM/1.4. We have transferred the units of OC sources and sinks to Tg (OC) yr⁻¹ in Table 6 to keep coherence with the data in Figure 4b.

33. L490: please provide some quantitative information with these plots, as for instance appears in the AeroCom web page with scatter plots (https://aerocom.met.no/cgi-bin/surfobs_annualrs.pl)

We have added some statistical values such as Table 7 to list the regional mean and spatial standard deviation, minimum and maximum values at IMPROVE and EMEP network sites versus simulated concentrations of sulfate (SO₄²⁻), organic carbon (OC), black carbon (BC), and the spatial correlation between observed and simulated multi-years averaged annual means.

34. L500: please provide some quantitative elements on the extinction coefficients, also single scattering albedo and asymmetry parameter³

As limited length of the text, the other optical feature of aerosols such as extinction coefficients, single scattering albedo and asymmetry parameters, and even their feedbacks on radiation and global temperature change will be explored in the other paper. It is mentioned in lines 706-709 in “5. Summary and discussions”

35. L502 (and paragraph): do you show a 1997-2003 average or the 2008 year as indicated in the figure; please provide quantitative information (bias, rmse..., or normalized figures as you prefer). This comment is valid for all figures. They should all be accompanied with some quantitative information

We have added Table 7 to list the regional mean and spatial standard deviation, minimum and maximum values at IMPROVE and EMEP network sites versus simulated concentrations of sulfate (SO₄²⁻), organic carbon (OC), black carbon (BC), and the spatial correlation between observed and simulated multi-years averaged annual means.

36. L516: I don't feel the evaluation is “comprehensive” so far. Please review this affirmation as you make some progress in a future version of the paper³

“comprehensive” is changed to “primary” in line 660.

37. L530: you indicate that you used prescribed concentrations for CH₄, and in Table 4 you indicate that you consider CH₄ emissions. Please clarify

CH₄ is a prognostic variable in the chemistry scheme of BCC-ESM1. So, emission of CH₄ listed in Table 4 is used to simulate CH₄ concentration, but some WMGHGs such as CH₄, N₂O, CO₂, CFC11 and CFC12 according to the experimental protocol of AerChemMIP are prescribed using CMIP6 prescribed concentrations (to replace prognostic values of

CH4 and N2O from the chemistry scheme). It is clarified in “3. Experiment design for the 20th century climate simulation” in the revised manuscript.

38. L541: there is no such comparison of all of these aerosols with observations. Please be more precise.

Modified the description in lines 684-685 in the revised manuscript as “Global budgets of aerosols were evaluated through comparisons of BCC-ESM1 results for 1990-2000 with reports in various literatures for sulfate, BC, OC, sea salt, and dust.”.

39. L560: I don't understand “How about the GHGs simulations in the AeroChemMIP historical run?” please be clearer and more precise

O3 is evaluated in this work. Other GHGs such as CH4 and N2O concentrations can be simulated when forced with emissions and their simulations also need to be evaluated in future. Those are added at the end of “5. Summary and discussions”.

40. Figure 4: please add biomass burning emissions, if not done yet, or indicate if they are already part of the figure

Modified the captions of Figure 4, and all the biomass burning emissions are included in natural emissions in (a)-(c).

41. Figure 14: do you compare monthly observations averaged over 1998-2005 with monthly model outputs averaged over 1998-2005? please formulate more precise.

The data plotted in Figure 14 (it is numbered to Figure 15 in the revised manuscript) are multi-years averaged annual means over the available years 1990–2005 for IMPROVE sites and 1995–2005 for EMEP sites and corresponding simulations. The caption of Figure 15 is rewritten as “Scatter plots showing observed versus simulated multi-years averaged annual mean sulfate (SO4), 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.”

Minor questions/remarks at the paper focuses on tropospheric aerosols

1. L1: the title is misleading and should be changed at the paper focuses on tropospheric aerosols

This is a very good suggestion. The title “Beijing Climate Center Earth System Model version 1 (BCC-ESM1): Model Description and Aerosols Simulation Evaluation” is changed to “Beijing Climate Center Earth System Model version 1 (BCC-ESM1): Model Description and Aerosols Simulation Evaluation”

2. L49: “Besides gaseous”

In the revision, we have rewritten this sentence to “Besides gaseous components, atmosphere also contains various aerosols, which are important for cloud formation and radiative transfer.” in lines 52-53 of the revised manuscript.

3. L51: aerosol are particles; so change “aerosol particles” to “aerosols”

It is modified in line 54 of the revised manuscript.

4. L59, and others: homogenise writing of chemical compounds, for instance O3

Expressions for chemical compounds similar to O3 in the whole text are modified to keep homogenies.

5. L99: “BCC-ESM1 is a fully-coupled global climate-chemistry-aerosol model “: it seems to me that BCC-ESM1 is more than that; I would say it is an “Earth System Model with interactive chemistry and aerosol components” if you want to insist on these components

We thank the reviewer for pointing this out. This sentence is rewritten to “BCC-ESM1 is an Earth System Model with interactive chemistry and aerosol components.” in lines 101-102 of the revised manuscript.

6. L120: change “used” to “uses”

It is corrected.

7. L122: please clarify “ranged to”

The sentence is rewritten to “MOM4-L40 uses a tripolar grid of horizontal resolution with 1° longitude by 1/3° latitude between 30°S and 30°N ranged to 1° longitude by 1° latitude from 60°S and 60°N poleward and 40 z-levels in the vertical.” in lines 161-163.

8. L145: it is not clear whether deposition velocities are computed interactively, as in Wesely, or consist of monthly means.

We have clarified those expressions in lines 127-137 of the revised manuscript.

9. L198: remove “Its”

In the revision, we have removed the word “Its”.

10. L238: please be more precise on the Web page

The Web page is <https://svn-ccsm-inputdata.cgd.ucar.edu/trunk/inputdata/atm/cam/dst/>. It is denoted as in line 251 in the revised manuscript.

11. L252: Wu et al 2019 is not in the list of references; and what do you mean by “it is parameterized”, what is “it”?

The reference of Wu et al.(2019) is added in the list of references. The sentence “it is

parameterized” means “liquid cloud droplet number concentration is parameterized”. We have added the description about its parameterization in “2.5 Effects of aerosols on radiation, cloud, and precipitation” in the manuscript.

12. L263: AGCM-Chem1: is this the correct name of the model?

“BCC-AGCM3-Chem” is the name of the atmosphere component model of BCC-ESM1. It is corrected in line 261 of the revised manuscript.

13. L264: please reformulate “at each model step and interacts with radiations”

We have rewritten this expression in lines 307-310 of the revised manuscript.

14. L276: add “see Table 4”

It is modified.

17. L283: MEGAN acronym already introduced

It is modified.

18. L310: change “1850 AD conditions” to “1850 conditions”.

It is modified.

19. L317: change “600” to “450”

It is modified.

20. L385: early 1980s

It is modified.

21. L513: please correct the North American coordinates; and correct also in Figure 14 the European coordinates; and furthermore the coordinates you indicate in the text do not correspond to those of Figure 14

Figure 14 is numbered to Figure 19 in the revised manuscript. We have corrected the legends in Figure 19 and the expressions in the text.

22. L543: in relevant literature

“in relevant literatures” is corrected to “in relevant literature”.

23. Table 1: please indicate that interactive surface emissions are considered for sea salt and dust

We added the expression “In the column of surface emission, interactive surface emissions

are considered for sea salt and dust.” in the caption of Table 1.

24. Table 5: I could not find figures for the sinks of DMS in Liu 2005 Table 4. Where do your figures come from?

It is our mistake as our references confusing and cancelled in the revised manuscript.

25. Table 6: f for Ginoux 2001

Ginoux et al. (2001) is added in the list of references.

26. Figure 1: change SAT into tas official CMIP6 variable

It is modified in Figure 1.

27. Figure 5: what is the "20th historical simulations"? Same question in caption of Figure 11

The expression of “20th historical simulations” is changed to “CMIP6 historical simulations” in Figures 7 and 14 in the revised manuscript.

28. Figure 5: change "blue" to "black"

It is modified. Figure 5 is renumbered to Figure 7 in the revised manuscript.

1 **Beijing Climate Center Earth System Model version 1 (BCC-ESM1):**
2 **Model Description and Evaluation of Aerosol Simulations~~Evaluation~~**

3
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20 *Correspondence to: Tongwen Wu (twwu@cma.gov.cn)*

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22 **Submit to Geosci. Model Dev.**

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

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

49

50 **1. Introduction**

51 Atmosphere is a thin gaseous layer around the Earth, consisting of nitrogen, oxygen and
52 a large number of trace gases including important greenhouse gases (GHG) such as water
53 vapor, tropospheric ozone (O₃), carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O),
54 and chloro-fluoro-carbons (CFCs). Besides ~~the~~ gaseous components, atmosphere also
55 contains various aerosols, which are important for cloud formation and radiative transfer.
56 Atmospheric trace gases and aerosols ~~partieles~~ are actually interactive components of the
57 climate system. Their inclusion in global climate models (GCMs) is a significant
58 enhancement for most state-of-the-art climate models (Lamarque et al., 2013; Collins et al.,
59 2017). Early attempts in coupling global climate dynamics with atmospheric chemistry can be
60 traced back to late 1970s, when 3D transport of ozone and simple stratospheric chemistry
61 were firstly incorporated into a GCM to simulate global ~~ozone (O₃)~~ production and transport
62 (e.g., Cunnold et al. 1975; Schlesinger and Mintz 1979). Since mid-1980s, a large number of
63 on-line global climate/chemistry models have been developed to address issues of the
64 Antarctic stratospheric O₃ depletion (e.g., Cariolle et al. 1990; Austin et al. 1992; Solomon,
65 1999), tropospheric O₃ and sulfur cycle (e.g., Feichter et al. 1996; Barth et al. 2000),
66 tropospheric aerosol and its interactions with cloud (e.g., Chuang et al. 1997; Lohmann et al.
67 2000; Ghan and Easter, 2006; Jacobson 2012). Aerosols and chemically reactive gases in the
68 atmosphere exert important influences on global and regional air quality and climate (Collins
69 et al., 2017).

70 Since 2013, the Beijing Climate Center (BCC), China Meteorological Administration,
71 has continuously developed and updated its ~~global~~ fully-coupled ~~GCMclimate model~~, the
72 Beijing Climate Center Climate System Model (BCC-CSM) (Wu et al., 2013; Wu et al., 2014;
73 Wu et al., 2019). BCC-CSM version 1.1 was one of the comprehensive carbon-climate
74 models participating ~~to~~ in the phase five of the Coupled Model Intercomparison Project
75 (CMIP5, Taylor et al. 2012). When forced by prescribed historical emissions of CO₂ from
76 combustion of fossil fuels and land use change, BCC-CSM1.1 successfully reproduced the
77 trends of observed atmospheric CO₂ concentration and global surface air temperature from
78 1850 to 2005 (Wu et al., 2013). During recent years, BCC-CSM1.1 has been used in
79 numerous investigations on soil organic carbon changes (e.g. Todd-Brown et al., 2014), ocean

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80 biogeochemistry changes (e.g. Mora et al., 2013), and carbon-climate feedbacks (e.g. Arora et
81 al., 2013; Hoffman et al., 2014). BCC-CSM includes main climate-carbon cycle processes
82 (Wu et al., 2013) and the global mean atmospheric CO₂ concentration is calculated from a
83 prognostic equation of CO₂ budget taking into account global anthropogenic CO₂ emissions
84 and interactive land-atmosphere and ocean-atmosphere CO₂ exchanges.

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85 In recent years, BCC has put ~~large much~~ 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, NO₂ 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 ~~into~~ 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 ~~givenshown~~ in Section 3. Section 4
97 presents ~~the~~ evaluations of aerosol simulations with comparisons to CMIP5-recommended
98 data (Lamarque et al., 2010) and data obtained from both global surface networks and satellite
99 observations. The regional and global characteristics compared to observations and estimates
100 from other studies are analyzed. Simulations of aerosol optical properties in the 20th century
101 are also analyzed in Section 4. Conclusions and discussions are summarized in Section 5.
102 Information about code and data availability is ~~givenshown~~ 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

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110 ~~BCC-ESM1 are fully coupled and interact with each other through fluxes of a fully coupled~~
111 ~~global climate chemistry aerosol model. Different components of BCC-ESM1 interact with~~
112 ~~each other through fluxes of momentum, energy, water, carbon and other tracers at their~~
113 ~~interfaces. The coupling between the atmosphere and the ocean is done every hour.~~
114 ~~The atmospheric component BCC-AGCM3-Chem is BCC Atmospheric General~~
115 ~~Circulation Model version 3 (Wu et al., 2019) with interactive atmospheric chemistry (hereafter~~
116 ~~BCC-AGCM3-Chem), is able to simulate global atmospheric composition and aerosols from~~
117 ~~anthropogenic emissions as forcing agents. Its resolution is T42 (approximately 2.8125°x2.8125°~~
118 ~~transformed spectral grid). The model has 26 levels in a hybrid sigma/pressure vertical coordinate~~
119 ~~system with the top level at 2.914 hPa. Details of the model physics are described in Wu et al.~~
120 ~~(2019). The land component is BCC Atmosphere and Vegetation Interaction Model version 2.0~~
121 ~~(BCC AVIM2.0) with terrestrial carbon cycle. The land component BCC AVIM2.0 is described in~~
122 ~~details in Li et al. (2019). It includes biophysical, physiological, and soil carbon-nitrogen~~
123 ~~dynamical processes, and the terrestrial carbon cycle operates through a series of biochemical and~~
124 ~~physiological processes on photosynthesis and respiration of vegetation. Biogenic emissions from~~
125 ~~vegetation are computed online in BCC AVIM2.0 following the algorithm of the Model of~~
126 ~~Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1, Guenther et al.,~~
127 ~~2012). The oceanic component is the Modular Ocean Model version 4 with 40 levels (hereafter~~
128 ~~MOM4 L40). The land component is BCC Atmosphere and Vegetation Interaction Model version~~
129 ~~2.0 (BCC AVIM2.0) with terrestrial carbon cycle. The sea ice component is Sea Ice Simulator~~
130 ~~(SIS). Different components of BCC-ESM1 interact with each other through fluxes of momentum,~~
131 ~~energy, water, carbon and other tracers at their interfaces. The coupling between the atmosphere~~
132 ~~and the ocean is done every hour. BCC-AGCM3-Chem is able to simulate global atmospheric~~
133 ~~composition and aerosols with anthropogenic emissions as forcing. It is developed on the basis of~~
134 ~~the recent version 3 of the Beijing Climate Center atmospheric general circulation model~~
135 ~~(hereafter BCC-AGCM3, Wu et al., 2019). The horizontal resolution of BCC-AGCM3-Chem is~~
136 ~~T42 (approximately 2.8125°x2.8125° transformed spectral grid). The model has 26 levels in a~~
137 ~~hybrid sigma/pressure vertical coordinate system with the top level at 2.914 hPa. The land~~
138 ~~component BCC AVIM2.0 is described in details in Li et al. (2019). It includes biophysical,~~
139 ~~physiological, and soil carbon-nitrogen dynamical processes, and the terrestrial carbon cycle~~

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170 climatological monthly mean deposition velocities
171 ([https://en.wikipedia.org/wiki/MOZART\(model\)](https://en.wikipedia.org/wiki/MOZART(model))) which are calculated offline (Bey et al., 2001;
172 [Shindell et al., 2008](#)) using a resistance-in-series scheme originally described in [Wesely](#)
173 [\(1989\)](#).~~The dry deposition velocities and those~~ for the other 15 species [including MPAN,](#)
174 [ONIT, ONITR, C₂H₅OH, POOH, C₂H₅OOH, C₃H₇OOH, ROOH, GLYALD, HYAC, CH₃OH,](#)
175 [MACROOH, ISOPOOH, XOOH, HYDRALD, and H₂](#) are ~~calculated determined~~ using a
176 ~~weighted combination of prescribed~~ deposition velocities of ~~O₃ozone, CO, or CH₃CHO, or~~
177 [land surface type and surface temperature following the MOZART2 \(Horowitz et al., 2003\).](#)
178 Wet removals by in-cloud scavenging for 25 soluble gas-phase species in the “standard
179 version” of MOZART2 use the parameterization of Giorgi and Chameides (1985) based on
180 their temperature dependent effective Henry’s law constants. In-cloud scavenging is
181 proportional to the amount of cloud condensate converted to precipitation, and the loss rate
182 depends on the amount of cloud water, the rate of precipitation formation, and the rate of
183 tracer uptake by the liquid phase water. Other highly soluble species such as HNO₃, H₂O₂,
184 ONIT, ISOPOOH, MACROOH, XOOH, and Pb-210 are also removed by below-cloud
185 washout as calculated using the formulation of Brasseur et al. (1998). Below-cloud
186 scavenging is proportional to the precipitation flux in each [model](#) layer and the loss rate
187 depends on the precipitation rate. Vertical transport of gas tracers and aerosols due to deep
188 convection is not yet included in the present version of BCC-AGCM3-Chem, [which process](#)
189 [is considered as a part of the deep convection and occurs generally in a small spatial region on](#)
190 [a GCM-box with low-resolution \(2.8°lat.×2.8°lon.\).](#) Another consideration is that a large
191 [uncertainty exists to treat transport of those water-soluble tracers by deep convection. But this](#)
192 [effect will be involved in the next version of BCC model.](#)
193 [The BCC-AVIM2.0 is the land model with terrestrial carbon cycle. It is described in](#)
194 [details in Li et al. \(2019\) and includes biophysical, physiological, and soil carbon-nitrogen](#)
195 [dynamical processes. The terrestrial carbon cycle operates through a series of biochemical](#)
196 [and physiological processes on photosynthesis and respiration of vegetation. Biogenic](#)
197 [emissions from vegetation are computed online in BCC-AVIM2.0 following the algorithm of](#)
198 [the Model of Emissions of Gases and Aerosols from Nature version 2.1 \(MEGAN2.1,](#)
199 [Guenther et al., 2012\).](#)

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200 The oceanic component of BCC-ESM1 is the Modular Ocean Model version 4 with 40
201 levels (hereafter MOM4-L40), and the sea ice component Sea Ice Simulator (SIS).
202 MOM4-L40 uses a tripolar grid of horizontal resolution with 1° longitude by 1/3° latitude
203 between 30°S and 30°N ranged to 1° longitude by 1° latitude from 60°S and 60°N poleward
204 and 40 z-levels in the vertical. Carbon exchange between the atmosphere and the ocean are
205 calculated online in MOM4-L40 using a biogeochemistry module that is based on the
206 protocols from the Ocean Carbon Cycle Model Intercomparison Project-Phase 2 (OCMIP2,
207 http://www.ipsl.jussieu.fr/OCMIP/phase2/). SIS has the same horizontal resolution as
208 MOM4-L40 and three layers in the vertical, including one layer of snow cover and two layers
209 of equally sized sea ice. Details of oceanic component MOM4-L40 and sea-ice component
210 SIS that are used in BCC-ESM1 may be found in Wu et al. (2013) and Wu et al. (2019).

211 In the following sub-sections, we will describe the treatments in BCC-ESM1 for 32
212 gas-phase species of DMS, ~~and~~ SO₂, ~~and~~ NH₃, 13 prognostic aerosol species including
213 sulfate (SO₄²⁻), 2 types of organic carbon (hydrophobic OC1, hydrophilic OC2), 2 types of
214 black carbon (hydrophobic BC1, hydrophilic BC2), 4 categories of soil dust (DST01, DST02,
215 DST03, DST04), and 4 categories of sea salt (SSLT01, SSLT02, SSLT03, SSLT04).
216 Concentrations of all aerosols in BCC-ESM1 are mainly determined by advective transport,
217 emission, dry — deposition, gravitational settling, and wet scavenging by clouds and
218 precipitation, except for SO₄²⁻ which gas-phase ~~chemical reactions~~ and aqueous phase
219 conversion from SO₂ are also considered. The present version of aerosol schemes belongs to a
220 bulk aerosol aerosol type of model and mainly refers to the scheme of CAM-Chem
221 (Lamarque et al., 2012), ~~and but~~ the nucleation and coagulation of aerosols are still ignored.
222 in the present version of BCC-AGCM3-Chem of BCC-ESM1.

223 2.1 SO₂, DMS, NH₃ and Sulfate

224 SO₂ is a main sulfuric acid precursor to form aerosol sulfate SO₄²⁻. Conversions of SO₂
225 to SO₄²⁻ occur by gas phase reactions (Table 2) and by aqueous phase reactions in cloud
226 droplets. The dry deposition velocity of SO₂ follows the resistance-in-series approach of
227 Wesely (1989) using the formula, $W_{SO_2} = 1/(r_a + r_b + r_c)$, in which ~~r_a, r_b, and r_c are the~~
228 aerodynamic resistance, the quasi-laminar boundary layer resistance, and the surface
229 resistance, respectively and they are interactively computed in each model time step ~~r_a and r_c~~

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230 ~~are the aerodynamic resistance and the surface canopy layer resistance, respectively.~~ _The
231 loss rate of SO₂ due to wet deposition is computed following the scheme in the global
232 Community Atmosphere Model (CAM) version 4, the atmospheric component of the
233 Community Earth System Model (Lamarque, et al., 2012).

234 The sources of SO₂ mainly come from fuel combustion, industrial activities, and
235 volcanoes. SO₂ can also be formed from the oxidation of DMS as listed in Table 2 ~~in which~~
236 ~~their reaction rates follow CAM-Chem (Lamarque et al. 2012).~~ The main source of DMS is
237 from oceanic emissions via biogenic processes. It is prescribed ~~with using~~ the ~~climatological~~
238 ~~monthly data that are extracted from~~ MOZART2 ~~data~~ package
239 ~~(https://www2.acom.ucar.edu/gcm/mozart-4).~~ ~~originated from the International Global~~
240 ~~Atmospheric Chemistry/Global Emissions Inventory Activity (IGAC/GEIA, Benkovitz et al.,~~
241 ~~1996).~~

242 SO₄²⁻ is one of the prognostic aerosols in BCC-AGCM3-Chem. ~~Its treatment follows~~
243 ~~CAM4-Chem (Lamarque et al., 2012).~~ It is produced primarily by the gas-phase oxidation of
244 SO₂ (in Table 2) and by aqueous phase oxidation of SO₂ in cloud droplets. The gas phase
245 reactions, rate constants, and gas-aqueous equilibrium constants are given by Tie et al. (2001).
246 The heterogeneous reactions of SO₄²⁻ occur on all aerosol surfaces. Their treatment follows a
247 Bulk Aerosol Model (BAM) used in CAM4 (Neale et al., 2010). The heterogeneous reactions
248 depend strongly on pH values in clouds which are calculated from the concentrations of SO₂,
249 HNO₃, H₂O₂, NH₃, O₃, HO₂, and SO₄²⁻. ~~Only NH₃ is not a prognostic tracer in~~
250 ~~BCC-AGCM3-Chem and it is estimated using the assumption of an NH₃ to SO₄²⁻ molar ratio~~
251 ~~of 2.0. NH₃ is a gas tracer apart from MOZART2 (Table 1). Its sources include aircraft and~~
252 ~~surface emissions due to anthropogenic activity, biomass burning, and biogenic emissions~~
253 ~~from land soil and ocean surfaces (Table 4).~~ SO₄²⁻ is assumed to be all in aqueous phase ~~due~~
254 ~~to water uptake,~~ although Wang et al. (2008a) showed that ~34% of sulfate particles are in
255 solid phase globally due to the hysteresis effect of ammonium sulfate phase transition.
256 However, in terms of radiative forcing, consideration of solid sulfate formation process
257 lowers the sulfate forcing by ~8% as compared to consideration of all sulfate particles in
258 aqueous phase (Wang et al., 2008b). Future model development may consider the life cycle of
259 NH₃. The sulfate in- and below-cloud scavenging follows Neu and Prather (2011). Washout

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260 of SO_4^{2-} is set to 20% of the washout rate of HNO_3 following Tie et al. (2005) and Horowitz
261 (2006). ~~Its d~~Dry deposition velocity of SO_4^{2-} is also calculated by the resistance-in-series
262 approach.

263 2.2 Aerosols of organic carbon and black carbon

264 BCC-AGCM3-Chem treats two types of organic carbon (OC), i.e. water-insoluble tracer
265 OC1 and water-soluble tracer OC2, and two types of black carbon (BC), i.e. water-insoluble
266 tracer BC1 and water-soluble tracer BC2. As shown in Table 2, hydrophobic BC1 and OC1
267 can be converted to hydrophilic BC2 and OC2 with a constant rate of $7.1 \times 10^{-6} \text{ s}^{-1}$ (Cooke and
268 Wilson, 1996). The 4 tracers of organic carbon and black carbon are mainly from emissions
269 ~~of anthropogenic activities~~ including both fossil fuel and biomass burning, and are from the
270 CMIP6 data package (<https://esgf-node.llnl.gov/search/input4mips/>, Hoesly et al., 2018).
271 Beside anthropogenic and biomass burning emissions, hydrophilic organic carbon OC2 can
272 also come ~~directly~~ from natural biogenic volatile organic compound (VOC) emissions. ~~They~~
273 ~~are calculated online in the land component model BCC-AVIM2 and assumed to equal to 10%~~
274 ~~of monoterpenes emission following the algorithm of Guenther et al. (1999).~~

275 Dry deposition velocities for all the 4 OC and BC tracers are set to $0.001 \text{ m} \cdot \text{s}^{-1}$. OC2 and
276 BC2 are soluble aerosols, and their sinks are primarily governed by wet deposition. Their in-
277 and below-cloud scavenging follows the scheme of Neu and Prather (2011) ~~and the transfer~~
278 ~~of soluble gases into liquid condensate is calculated with Henry's Law assuming equilibrium~~
279 ~~between the gas and liquid phases.~~

280 2.3 Sea salt aerosols

281 As shown in Table 3, sea salt aerosols in the model are classified into four size bins (0.2–
282 1.0, 1.0–3.0, 3.0–10, and 10–20 μm) in ~~diameter~~diameter. They originate from oceans and
283 are calculated online by BCC-ESM1. The upward flux $F_{\text{sea-salt}}$ of sea salt productions for
284 four bins is proportional to the 3.41 power of the wind speed u_{10m} at 10 m height near the
285 sea surface (Mahowald et al., 2006) and is expressed as

$$286 F_{\text{sea-salt}} = S \cdot (u_{10m})^{3.41}, \quad (1)$$

287 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} prescribed~~
288 for ~~four~~each size bins of sea salt aerosols in BCC-ESM1, respectively.

289 Dry deposition of sea salts depends on the turbulent deposition velocity in the lowest

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

296 **2.4 Dust aerosols**

297 Dust aerosols behave in a similar way as sea salts. Their variations involve three major
298 processes: emission, advective transport, and wet/dry depositions. The dust emission is based
299 on a saltation-sandblasting process, and depends on wind friction velocity, soil moisture, and
300 vegetation/snow cover (Zender et al., 2003). The vertical flux of dust emission is corrected by
301 a surface erodible factor at each model grid cell which has been downloaded from NCAR
302 website ~~_~~
303 (~~<https://near.ucar.edu>~~~~<https://svn-ccsm-inputdata.cgd.ucar.edu/trunk/inputdata/atm/cam/dst/>~~).

304 Soil erodibility is prescribed by a physically-based geomorphic index that is proportional to
305 the runoff area upstream of each source region (Albani et al., 2014). Like sea salts, dry
306 ~~deposition~~ of dust aerosols includes gravitational and turbulent deposition processes, while
307 wet deposition results from both convective and large scale precipitation and is dependent on
308 prescribed size-independent scavenging coefficients.

309 **2.5 Effects of aerosols on radiation, clouds, and precipitation**

310 The mass mixing ratios of bulk aerosols are prognostic variables in BCC-ESM1 and
311 directly affect the ~~shortwave~~ radiative transfer in the atmosphere with their treatments
312 following the NCAR Community Atmosphere Model (CAM3, Collins et al., 2004). Indirect
313 effects of aerosols are taken into account in the present version of BCC-AGCM3-Chem (Wu
314 et al., 2019). Aerosol particles act as cloud condensation nuclei and exert influence on cloud
315 properties and ~~precipitationthe hydrological cycle~~, and ultimately impact the hydrological
316 cycleprecipitation.~~_~~

317 Prognostic aerosol masses are used to estimate the liquid cloud droplet number
318 concentration N_{cdnc} (cm^{-3}) in BCC-AGCM3-Chem. N_{cdnc} is explicitly calculated using the
319 empirical function suggested by Boucher and Lohmann (1995) and Quaas et al. (2006):

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

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

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

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

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

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

where β is a parameter dependent on the droplets spectral shape and follows the calculation proposed by Peng and Lohmann (2003),

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

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

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

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

The liquid cloud droplet number concentration is an important factor in determining the effective radius of cloud droplets for radiative calculation and in calculating the precipitation efficiency. Aerosols also exert impacts on precipitation efficiency (Albrecht, 1989), which is taken into account in the parameterization of non-convective cloud processes. There are five processes that convert condensate to precipitate: auto-conversion of liquid water to rain, collection of cloud water by rain, auto-conversion of ice to snow, collection of ice by snow, and collection of liquid by snow. The auto-conversion of cloud liquid water to rain (PWAUT) is dependent on the cloud droplet number concentration and follows a formula that was originally suggested by Chen and Cotton (1987).

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

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374 replaced by CMIP6 data. The rest of ~~Other~~ historical forcing data include: (1) ~~monthly~~
375 ~~zonally mean CFC11 and CFC12 concentrations, (2) yearly global gridded land-use forcing~~
376 ~~data sets, and (23) solar forcing. All these datasets were downloaded from~~
377 ~~<https://esgf-node.llnl.gov/search/input4mips/>. Climate feedback processes that involve changes~~
378 ~~to the atmospheric composition of reactive gases and aerosols may affect the temperature~~
379 ~~response to a given WMGHG concentration level (Collins et al., 2017). Three members of~~
380 ~~historical experiments are conducted and the first member is analyzed in this work. The~~
381 ~~AeroChemMIP historical case is a simulation with emissions evolving from 1850 to 2014 and~~
382 ~~with specified GHG concentrations of CH₄, N₂O, and CO₂ following the protocols defined~~
383 ~~by CMIP6. Other historical forcing data include: (1) monthly zonally mean CFC11 and~~
384 ~~CFC12 concentrations, (2) yearly global gridded land use forcing data sets, and (3) solar~~
385 ~~forcing. All these datasets were downloaded from <https://esgf-node.llnl.gov/search/input4mips/>.~~
386 ~~The principal GHGs for radiation calculation in BCC AGCM Chem1 include H₂O, O₃, CH₄,~~
387 ~~N₂O, CO₂, CFC11 and CFC12. Only one GHG, O₃, is a prognostic variable at each model~~
388 ~~step and interacts with radiations.~~

3.1 Surface emissions

390 Surface emissions of chemical species from different sources are summarized in Table
391 4. They include anthropogenic emissions from fossil fuel burning and other industrial
392 activities, biomass burning (including vegetation fires, fuel wood and agricultural burning),
393 biogenic emissions from vegetation and soils, ~~and oand~~ oceanic emissions. Most historical
394 emissions from anthropogenic source (surface, ~~aircraftplane~~ plus ships) and biomass burning
395 ~~in the period offrom~~ 1850 to 2014 are CMIP6-recommended data (Hoesly et al., 2018;
396 available at <https://esgf-node.llnl.gov/search/input4mips/>). Anthropogenic or biomass burning
397 sources of some tracers ~~which are~~ not included in the CMIP6 dataset (see Table 4),
398 ~~anthropogenic emission of H₂ and N₂O are from monthly the climatological dataset provided~~
399 ~~by the standard package of MOZART-2 standard package. N₂O is a prognostic variable in~~
400 ~~BCC-ESM1 but it is replaced by CMIP6 prescribed concentration in the historical run.~~
401 ~~Other emissions including biomass burning (CH₃COCH₃) and anthropogenic emission (or~~
402 ~~CH₃CHO, CH₃OH, and CH₃COCH₃) are from the IPCC ACCMIP emission inventory~~
403 (<http://accent.aero.jussieu.fr/ACCMIP.php>) covering the period from 1850 to 2010 ~~with in~~

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404 10-year ~~time~~ intervals (see Table 4). ~~Monthly CMIP6 recommended lumped~~ emissions of
405 ~~black carbon and organic carbon aerosols from 1850 to 2014 black carbon and organic carbon~~
406 ~~aerosols are downloaded from CMIP6-recommended data, but we used assume~~ 80% (for BC)
407 ~~and 50% (for OC) of them~~ in their hydrophobic forms (BC1 and OC1) and ~~the rest in their~~ 20%
408 ~~in~~ hydrophilic forms (BC2 and OC2), following the work of Chin et al. (2002).

409 ~~Five~~Seven tracers of ISOP, ACET (CH_3COCH_3), C_2H_4 , C_3H_8 , and Monoterpenes ($\text{C}_{10}\text{H}_{16}$)
410 in Table 1 belong to biogenic volatile organic carbons (VOCs), ~~i.e. ISOP, ACET~~
411 ~~(CH_3COCH_3), C_2H_4 , C_2H_6 , C_3H_{10} , Terpenes ($\text{C}_{10}\text{H}_{16}$), and OC2~~. As shown in Table 4, those

412 VOCs emissions are ~~directly~~ online calculated in BCC-ESM1 following the modeling
413 framework of the Model of Emissions of Gases and Aerosols from Nature version 2.1
414 (MEGAN2.1, Guenther et al., 2012) using simple mechanistic algorithms to account for major

415 known processes controlling biogenic emissions. The ~~MEGAN-MEGAN2.1 can provide a~~
416 ~~flexible scheme for estimating 16 tracers of biogenic emissions from terrestrial ecosystems~~
417 ~~including five VOCs emissions used in BCC-ESM1 (Table 4). All the VOCs emissions~~

418 depend on current and past surface air temperature, ~~and solar flux, and the landscape types,~~
419 ~~and~~ their calculation requires global maps of plant functional type (PFT) and leaf area index
420 (LAI) which is a prognostic variable from the land model BCC-AVIM2. ~~The effect of~~

421 ~~atmospheric CO₂ concentration on isoprene emissions is included, 10% of the biogenic~~
422 ~~monoterpenes emissions as calculated online with the MEGAN2.1 algorithm in BCC-AVIM2~~
423 ~~are converted to hydrophilic organic carbon (OC2) to account for formation of secondary~~

424 ~~organic aerosols following Chin et al. (2002) in this version of BCC-ESM1.~~
425 **3.2 Volcanic~~o~~ eruption, lightning and aircraft emissions**

426 ~~As there is no stratospheric aerosol scheme in BCC-ESM1, concentrations of sulfate~~
427 ~~aerosol at heights from 5 to 39.5 km, which volcanic origin, are directly prescribed using the~~
428 ~~CMIP6-recommended data (Thomasson et al., 2018) from 1850 to 2014. The effects of~~
429 ~~surface SO₂ emissions from volcanic eruption on the variation of SO₂ in the atmosphere and~~

430 ~~then on the variation of tropospheric SO₄²⁻ concentration are considered, and the SO₂~~
431 ~~emissions from 1850 to 2014 are downloaded from the IPCC ACCMIP emission inventory~~
432 ~~(<http://accent.aero.jussieu.fr/ACCMIP.php>). Emissions of stratospheric SO₂ from volcanic~~

433 ~~eruption from 1850 to 2014 are prescribed using the CMIP6 recommended data, although~~

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434 ~~recent studies based on satellite observation of SO₂ in 2006-2012 have revealed that this~~
435 ~~emission data may have a factor of 2-4 high bias in average (Ge et al., 2016).~~ Aircraft
436 emissions are provided for NO₂, CO, CH₄, ~~NH₃, NO, and SO₂~~, and aerosols of OC and BC
437 (Table 1). The emissions of NO from lightning are online calculated in BCC-AGCM3-Chem
438 following the parameterization in MOZART2, ~~and the globally-averaged mean during the~~
439 ~~period of 1850 to 2014 is 5.19 Tg(N)·yr⁻¹, which is in agreement with observations within the~~
440 ~~range of 3 to 6 Tg(N)·yr⁻¹ (Martin et al., 2002).~~ The lightning frequency depends strongly on
441 the convective cloud top height, and the ratio of cloud-to-cloud versus cloud-to-ground
442 lightning depends on the cold cloud thickness from the level of 0°C to the cloud top (Price
443 and Rind, 1992).
444

445 3.3 Upper boundary of the atmosphere

446 As no stratospheric chemistry is included in the present version of BCC-AGCM3-Chem,
447 it is necessary to ensure a proper distribution of chemically-active stratospheric species
448 ~~including O₃, CH₄, N₂O, NO, NO₂, HNO₃, CO, and N₂O₅.~~ Concentrations of ~~different these~~
449 tracers (O₃, CH₄, N₂O, NO, NO₂, HNO₃, CO, and N₂O₅) at the top two layers of the model are
450 set to prescribed monthly climatological values, and concentrations from below the top two
451 layers to the tropopause are relaxed at a relaxation time of 10-days towards the
452 ~~climatology with a 10-day time scale down to the tropopause.~~ Climatological values of NO,
453 NO₂, HNO₃, CO and N₂O₅ at the top two layers model levels are extracted from use
454 MOZART2 data package available at the Website
455 (<https://www2.acom.ucar.edu/gcm/mozart-4>), ~~originated from the and are based on~~ Study of
456 Transport and Chemical Reactions in the Stratosphere (STARS, Brasseur et al., 1997).
457 Concentrations for the other tracers of (O₃ ozone, CH₄, and N₂O) at the top two model
458 layer levels are the zonally-averaged and monthly values from 1850 to 2014 derived from the
459 CMIP6 data package.

460 3.4 The preindustrial model states

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

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464 each component of BCC-ESM1 in order for the piControl simulation to run stably and fast to
465 reach its equilibrium. Figures 1(a-cb) show the time series of global yearly means of the net
466 energy budget at top of the atmosphere (TOA), near-surface air temperature (TAS), and sea
467 surface temperature (SST) the net energy budget at top of the model (TOM) from the
468 piControl simulation for the last 450 years. It shows that the surface climate in BCC-ESM1
469 nearly reaches its equilibrium after 600 years piControl simulation. The whole system in
470 BCC-ESM1 fluctuates around $+0.7 \text{ W}\cdot\text{m}^{-2}$ net energy flux at TOA without obvious trend
471 in 450 years (Fig. 1ab). This level of TOA energy imbalance is close to the average
472 imbalance ($1.0 \text{ W}\cdot\text{m}^{-2}$) among CMIP5 models (Wild et al., 2013), and do not cause remarkable
473 climate drift in BCC-ESM1. The global mean surface air temperature TAS and SST
474 keep around 288.1 K (Fig. 1b) and 295.05 K shows only a small warming (Fig. 1ca),
475 respectively. During the last 450 years, there are ($\pm 0.2^\circ\text{K}$ amplitude of TAS and SST)
476 oscillations of centennial scale for the whole globe (Figs. 1b and 1c), which are certainly
477 caused by internal variation of the system.

478 Figures 2a-2c show the time series of global annual total burdens of SO_2 , DMS,
479 and OH in the troposphere (integrated from the surface to 100 hPa) in the last 450 years of the
480 piControl simulation. Without any anthropogenic source, the SO_2 amount in the troposphere
481 nearly keeps the level of $0.0868 \pm 0.002 \text{ Tg}$ in the 450 years of the piControl
482 simulation. Tropospheric DMS varies around the value of $0.116 \pm 0.002 \text{ Tg}$. Tropospheric OH,
483 as an important gas species oxidizing SO_2 to form SO_4^{2-} (Table 2), keeps at a stable level in
484 the atmosphere. SO_4^{2-} also remains at a stable level of $0.556 \pm 0.004 \text{ Tg}$ in the atmosphere in
485 the whole period of the piControl simulation (Figure 2d). Without any anthropogenic source,
486 the amounts of BC and OC in the troposphere vary around $0.0395 \pm 0.005 \text{ Tg}$ and $0.275 \pm$
487 0.005 Tg (Figures 2e-2f), respectively. Dust and sea salt aerosols are at the level of $22 \pm 1 \text{ Tg}$
488 and $11.7 \pm 1 \text{ Tg}$ (Figures 2g-2h), respectively. All those data are close to the global mean
489 concentrations of $0.604 \text{ Tg SO}_4^{2-}$, 0.046 Tg BC , 0.30 Tg OC , 22.18 Tg dust , and 11.73 Tg sea
490 salts in 1850 which are estimated based on the CMIP5 prescribed data in 1850 (Lamarque et
491 al., 2010) recommended concentrations in year 1850 ($0.604 \text{ Tg SO}_4^{2-}$, 0.046 Tg BC , $0.30 \text{ Tg$
492 OC, 22.18 Tg dust , and $11.73 \text{ Tg sea salts}$).

493 Figure 3 shows the global spatial distributions of annual mean sulfate, organic carbon,

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494 black carbon, dust, and sea salt aerosols in the whole atmospheric column averaged for the
495 last 100 years of the piControl simulation of BCC-ESM. We can compare them with CMIP5
496 recommended concentrations in year 1850, considered as the reference state in the
497 pre-industrial stage. At that time, there are fewer anthropogenic/biomass SO₂ emissions, the
498 Without any industrial and fossil fuel anthropogenic emissions of SO₂, the SO₄²⁻ over land are
499 evidently smaller than those over oceans mainly distributed especially over the tropical
500 Pacific and Atlantic Oceans, where DMS can be oxidized to SO₂ and then form SO₄²⁻. There
501 are several centers of high values of black carbon and organic carbon in East and South Asia,
502 Europe, Southeast America, and in the tropical rain forests in Africa and South America.
503 They mainly result from biomass burning including vegetation fires, fuel wood and
504 agricultural burning. Dust aerosols are mainly distributed in North Africa, Central Asia, North
505 China, and Australia, where arid and semi-arid areas locate. Dust emitted from Sahara Desert
506 can be transported to the tropical Atlantic by easterly wind. The sea salt aerosols are mainly
507 distributed over the mid-latitude Southern Oceans, the tropical southern Indian Ocean, and the
508 tropical northern Pacific Ocean, where wind speeds near the sea surface are strong. As shown
509 in Fig. 3, all the spatial distribution patterns of CMIP5-derived sulfate, black carbon, organic
510 carbon, dust, and sea salt aerosols (Lamarque et al., 2010) are well simulated in BCC-ESM1,
511 ~~and~~ there are high spatial correlation coefficients, 0.76 for sulfate, 0.77 for black carbon,
512 0.77 for organic carbon, 0.94 for dust, and 0.94 for sea salts, between CMIP5 data and
513 BCC-ESM1 simulations. Relative lower relations for sulfate, black carbon and organic carbon
514 are possibly caused as different anthropogenic emission sources are used in BCC-ESM1 and
515 to create CMIP5 data. Dust and sea salts belong to natural aerosols and depend on the land
516 and sea surface conditions, so their spatial distributions are easy to be captured and have
517 relatively higher correlations between CMIP5 data and BCC-ESM1 simulations.

519 **4. Evaluation of O₃ and aerosols simulations in the 20th century**

520 The rate of sulfate formation is dependent on the levels of oxidants in the troposphere.
521 O₃ is an important oxidant. So, the evaluation of simulated tropospheric O₃ is helpful to
522 understand the aerosols simulations. BCC-ESM1 is driven by most of the
523 CMIP6-recommended emission data. As shown in Figure 4, the zonal distributions of the total

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554 as well as in developing countries in the Southern Hemisphere in recent years (Liu et al.,
555 2009). The OC and BC emissions substantially increased since 1950s just after the Second
556 World War. The global total OC emission in 2010 was nearly twice as much as that in
557 pre-industrial (year 1850) and increased by 18 Tg_{·yr}⁻¹. Anthropogenic black carbon
558 emissions increased from 1 Tg_{·yr}⁻¹ in 1850 to nearly 8 Tg_{·yr}⁻¹ in 2010.

559 Anthropogenic SO₂, OC and BC emissions strongly affect the variations of atmospheric
560 concentrations of DMS, SO₂, sulfate, OC, and BC. The global 0.5°x0.5° gridded data of
561 CMIP5-recommended aerosols masses with 10-years interval from 1850 to 2000 (Lamarque
562 et al., 2010) provides an important reference to evaluate the aerosol simulations in
563 BCC-ESM1. As shown in Figure 75b-75f, the annual total aerosol burdens of SO₄²⁻,
564 OC, and BC in the whole atmosphere column as simulated by the BCC-ESM1 20th century
565 historical simulation are generally consistent with the values derived from
566 CMIP5-recommended aerosols concentrations. Due to increasing SO₂ emissions from
567 1850 to present day (Fig. 64), the global SO₂ burden in the atmosphere increased from 100 Tg
568 in 1850s to 200 Tg in 1980s (Fig. 75a), and has a high interannual-correlation coefficient of
569 0.996 with the anthropogenic emissions (Fig. 64a), as the lifetime of SO₂ is short. The
570 burden directly followed the emission. DMS in the atmosphere is oxidized by OH and NO₃ to
571 form SO₂ (Table 2). Its natural emissions from oceans from 1850 to 2010 in the model are the
572 climatological monthly means (Dentener et al., 2006) from MOZART2 NCAR data package.

573 As shown in Fig 75a, the global amount of DMS in the whole atmosphere was about 0.12 Tg
574 during 1850-1900 and decreased to 0.055 Tg in 2010. This decrease trend maybe partly
575 possibly results from the speeded rate of DMS oxidation with global warming, the prescribed
576 emissions have not year to year variations and the loss of DMS oxidation to produce SO₂
577 gradually exceeds the source (Table 5) as the rate of DMS oxidation reaction (Table 2) gets
578 large along with global warming in the 20th century and the loss of DMS gradually exceeds
579 the source of ocean DMS emission to cause a net loss of DMS in the atmosphere since 1910s.

580 Largely driven by SO₂ anthropogenic emissions, the sulfate burden shows three different
581 stages from 1850 to present. In the first period from 1850s to 1900s, the sulfate burden had a
582 weak linear increase. It increased significantly in the second stage from 1910's to 1940's, and
583 then exploded since 1950's, until the middle 1970s and earlier 1980s. The sulfate burden

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584 then remained nearly stable and even showed slightly decreases as seen from the CMIP5 data.

585 ~~As for The trends of~~ global BC and OC burdens, BCC-ESM1 results show continuous
586 increases since 1850s, especially from 1950 to present. From 1910's to 1940's, the CMIP5
587 data show a slight decrease of BC and OC burdens in the atmosphere, are similar to that of
588 sulfate, but they showed continuous increases from 1950 to present.

589 The dust and sea salt aerosols in the atmosphere are largely determined by the
590 atmospheric circulations and states of the land and ocean surface. We can see that the global
591 dust burden in the atmosphere showed ~~a large interannual variability and was slightly evident~~
592 increase enhanced from 1985 to 2000, which could be partly caused by evident global
593 warming since 1980 and increasing soil dryness resulting in more surface dust to be released
594 in the atmosphere. Their details will be explored in the other paper.

595 4.2 Global aerosols budgets

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

601 The global DMS emission from the ocean is 27.4 Tg(-S)-yr⁻¹ in BCC-ESM. ~~It is higher~~
602 ~~than the value reported in Liu et al (2005), largely due to stronger wind speed near the sea~~
603 ~~surface.~~ This high emission in BCC-ESM is nearly balanced by the gas-phase oxidation of
604 DMS to form SO₂. The DMS burden is 0.1206 Tg-S with a lifetime of 0.78 days, which is
605 within the range of other models reported in the literature. As shown in Table 5, the total SO₂
606 production averaged for the period of 1991 to 2000 is 76.93 Tg(-S)-yr⁻¹. A rate of 13.2
607 Tg(-S)-yr⁻¹ (about 17%) SO₂ is produced from the DMS oxidation, only 0.1 Tg(-S)-yr⁻¹ SO₂
608 from air ~~planetraffic emissions to the atmosphere~~, and the rest (63.63 Tg(-S)-yr⁻¹, near 82.7%)
609 from anthropogenic activities and volcanic eruption at surface. ~~Here the emissions of SO₂~~
610 ~~from volcanic eruption are not included.~~ The amount of SO₂ produced from the DMS
611 oxidation is in the range of other works (10.0 to 24.7 Tg(-S)-yr⁻¹) reported in Liu et al (2005).
612 All the SO₂ production is balanced by SO₂ losses by dry and wet deposition, and by gas- and
613 aqueous-phase oxidation. Half of its loss (38.74 Tg(-S)-yr⁻¹) occurs via its aqueous-phase

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614 oxidation to form sulfate. Other losses through dry and wet depositions and gas-phase
615 oxidation to form SO_4^{2-} are also important (Table 2). All the sinks are in the range from the
616 literature (Liu et al., 2005). The global burden of SO_2 in the atmosphere is 0.4824 Tg-S with a
617 lifetime of 1.12 days, consistent with values in literature (Liu et al., 2005).

618 Sulfate aerosol is mainly produced from aqueous-phase SO_2 oxidation ($38.73 \text{ Tg}(-\text{S})\text{-yr}^{-1}$)
619 and partly from gaseous phase oxidation of SO_2 ($10.32 \text{ Tg}(-\text{S})\text{-yr}^{-1}$), and is largely lost by wet
620 scavenging ($49.06 \text{ Tg}(-\text{S})\text{-yr}^{-1}$). The total SO_4^{2-} production in BCC-ESM is at the lower range
621 of values in other models reported in Textor et al. (2006). Its global burden is 1.890.63 Tg S
622 and the lifetime is 4.69 days, which are within the range of 1.710.57 to 2.430.66 Tg S-and
623 3.723 to 5.4 days in the literatures (Textor et al., 2006; Liu et al., 2012; Liu et al., 2016; Liu et
624 al., 2009; Matsui and Mahowald, 2017; Tegen et al., 2019; the value derived from CMIP5
625 data).

626 Sources of BC and OC are mainly from anthropogenic emissions. Based on the CMIP6
627 data, there are, on average, $7.22 \text{ Tg}\text{-yr}^{-1}$ BC and $13.9145.2 \text{ Tg}\text{-yr}^{-1}$ OC from fossil and
628 bio-fuel emissions and 18.38 $\text{Tg}\text{-yr}^{-1}$ OC from natural emission during the period of 1991 to
629 2000. Most of them are scavenged through convective and large-scale rainfall processes. The
630 rest returns to the surface by dry deposition. ~~The simulated BC and OC lifetimes are 6.6 and~~
631 ~~5.0 days, respectively.~~ The simulated global BC and OC burdens are 0.13 and 0.62 Tg,
632 respectively (Table 6), all close to values of 0.114 Tg BC and 0.69 Tg OC derived from the
633 CMIP5 data, and within the range of 0.11-0.26 Tg BC (Textor et al., 2006; Matsui and
634 Mahowald, 2017; Tegen et al., 2019) and less than the values of 1.25-2.2 Tg OC in other
635 literatures (Textor et al., 2006; Tegen et al., 2019). ~~The simulated BC and OC lifetimes are~~
636 ~~6.6 and 5.0 days, respectively, and are close to the recent values of 5.0-7.5 days BC and~~
637 ~~5.4-6.6 days OC in literatures (Matsui and Mahowald, 2017; Tegen et al., 2019).~~

638 ~~The emissions of dust and sea salt are mainly determined by winds near the surface.~~ The
639 annual total dust emission in BCC-ESM1 is 2592 Tg yr^{-1} , higher than AeroCom multi-model
640 mean (1840 Tg yr^{-1} , Textor et al., 2006), but comparable to other studies (Chin et al., 2002;
641 Liu et al., 2012; ~~Emmons et al., 2010~~Matsui and Mahowald, 2017). The average dust loading
642 is 22.93 Tg , lower than the value of 35.9 Tg in Ginoux et al. (2001) but slightly higher than
643 the value of 20.41 Tg derived from CMIP5 data. The average lifetime for dust particles is

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644 3.23 days that is shorter than the AeroCom mean (4.14 days) and the value of 3.9 days in
645 recent study (Matsui and Mahowald, 2017). The simulated sea salt emission is $4667.2 \text{ Tg yr}^{-1}$,
646 slightly lower than the simulated value in Liu et al. (2012), and substantially lower than the
647 AeroCom mean (16600 Tg yr^{-1} , Textor et al., 2006). The simulated sea salt burdens are 11.89
648 Tg and close to the CMIP5 data. Their averaged lifetimes are 0.93 days and close to the value
649 in the recent of Matsui and Mahowald (2017) but longer than the AeroCom mean (0.41 days,
650 Textor et al., 2006).

651

652 4.3 Global aerosol distributions at present day

653 Figures 86-120 show December-January-February (DJF) and June-July-August (JJA)
654 mean column mass concentrations of sulfate (SO_4^{2-}), OC, BC, Dust, and Sea Salt aerosols
655 averaged for the period of 1991-2000, respectively. Here, BCC-ESM1 simulated results are
656 compared with the CMIP5-recommended data for the same period. Unlike the pre-industrial
657 level of sulfate shown in Fig. 2, sulfate concentrations at present day (Fig. 86) are strongly
658 influenced by anthropogenic emissions, and have maximum concentrations in the industrial
659 regions (e.g., East Asia, Europe, and North America). Their seasonal variations are distinct
660 and are characterized by high concentrations in boreal summer and low concentrations levels
661 in boreal winter. These spatial distributions simulated by BCC-ESM1 are well consistent with
662 the CMIP5 data, with spatial correlation coefficients in DJF and JJA reaching 0.92 and 0.83
663 (Figure 13), respectively. The deviation of the spatial pattern in BCC-ESM1 is less from the
664 CMIP5 data in DJF but larger in JJA (Figure 13).

665 Unlike sulfate whose maximum concentrations are mainly distributed between 60°N
666 and the equator, peaking concentrations of BC and OC as shown in Figs. 97 and 108 are
667 located near the tropics in the biomass burning regions (e.g., the maritime continent, Central
668 Africa, South America), and their seasonal variations from DJF to JJA are evidently weaker
669 than those of sulfate except in South America. In boreal summer, there are centers of high
670 values in the industrial regions in the Northern Hemisphere mid-latitudes (i.e., East Asia,
671 South Asia, Europe, and North America). These main features of spatial and seasonal
672 variations in CMIP5 data are well captured by BCC-ESM1, and the BCC-ESM1 vs. CMIP5
673 spatial correlation coefficients (Figure 13) are 0.90 (OC in DJF), 0.91 (BC in DJF), 0.91 (OC

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674 in JJA) and 0.92 (BC in JJA). There are less deviations of spatial pattern for OC in DJF and
675 JJA, but larger deviation for BC from CMIP5 data (Figure 13).

676 As show in Figure 119, dust concentrations in the atmosphere show largest values over
677 strong source regions such as Northern Africa, Southwest and Central Asia, and Australia,
678 and over their outflow regions such as the Atlantic and the western Pacific. In DJF, the
679 CMIP5 data shows centers of high concentrations over East Asia and Central North America,
680 but both centers are missing in BCC-ESM1. ~~We think, however, that~~ these two high-value
681 centers in the CMIP5 data may not be true, since frozen soils in these areas in winter lead to
682 unfavorable conditions for soil erosion by winds. The spatial correlation coefficients between
683 CMIP5 and BCC-ESM1 remain high: 0.95 in JJA and 0.88 in DJF (Figure 13). Small
684 deviations of spatial pattern for dust simulations in BCC-ESM1 show less magnitude of dust
685 maximums against with CMIP5 data (Figure 13).

686 As shown in Figure 120, high sea salt concentrations are generally over the storm track
687 regions over the oceans, e.g., middle-latitudes in the Northern Oceans in DJF and the
688 Southern Ocean in JJA where wind speeds and thus sea salt emissions are higher. In addition,
689 there is a belt of high sea salt concentrations in the subtropics of both hemispheres where
690 precipitation scavenging is weak. Their spatial distributions in BCC-ESM1 are consistent with
691 the CMIP5 data with correlation coefficients of 0.92 in JJA and 0.90 in DJF (Figure 13). The
692 spatial deviations of sea salt are much closer to CMIP5 data than those of sulfate, OC, BC,
693 and dust distributions (Figure 13).

694
695 Figure 14 shows vertical ~~profiles-distributions~~ of zonally-averaged annual mean
696 concentrations of sulfate, organic carbon, black carbon, dust, and sea salt aerosols in the
697 period of 1991-2000. Both BCC-ESM1 and CMIP5 results show that strong sulfur, OC, and
698 BC emissions in the industrial regions of the Northern Hemisphere mid-latitudes can rise
699 upward and be transported towards the North Pole in the middle to upper troposphere. Most
700 of OC, BC, and dust aerosols are confined below 500 hPa, while sulfate can be transported to
701 higher altitudes. Sea salt aerosols are mostly confined below 700 hPa, as the particles are
702 large in size and favorable for wet removal and gravitational settling towards the surface. It
703 can be seen that BCC-ESM1 tends to simulate less upward transport of aerosols than the

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704 CMIP5 data, likely reflecting the omission of deep convection transport of tracers in
705 BCC-ESM1.

706 The CMIP5 data used here are mainly from model simulations. We ~~will need to~~ further
707 evaluate the BCC-ESM1 model results with ground observations. Annual mean SO_4^{2-} , BC and
708 OC aerosol ~~concentration~~ observations from the Interagency Monitoring of Protected Visual
709 Environments (IMPROVE) sites over 1990-2005 in the United States
710 (<http://vista.cira.colostate.edu/IMPROVE/>) and from the European Monitoring and Evaluation
711 Programme (EMEP) (<http://www.emep.int>) sites over 1995-2005 are used. As shown in
712 Figure 152a and 152b, the BCC-ESM simulated sulfate concentrations are in general
713 comparable to the EMEP observations in Europe, but are systematically ~~by~~ about $1 \mu\text{g m}^{-3}$
714 higher than the U.S. IMPROVE observations. As for BC, there are large model biases at both
715 European and U.S. sites (Figs. 152c and 152d), especially BCC-ESM overestimates BC
716 concentrations at the IMPROVE sites. The ~~observed simulated~~ OC concentrations are slightly
717 overestimated for IMPROVE sites but systematically underestimated for EMEP sites. Some
718 statistical features for simulated concentrations versus EMEP and IMPROVE observations are
719 listed in Table 7. These comparisons are overall fairly reasonable considering the
720 uncertainties in emissions and the coarse model resolution.

721 We then evaluate the simulated BC concentrations from BCC-ESM1 with the HIAPER
722 (High-Performance Instrumented Airborne Platform for Environmental Research)
723 Pole-to-Pole Observations (HIPPO) (Wofsy et al., 2011). The HIPPO campaign provided
724 observations of black carbon concentration profiles over Pacific Ocean and North America
725 between 2009 and 2011. Following Tilmes et al. (2016), model results here are sampled along
726 the HIPPO flight tracks and then averaged to different latitude and altitude bands for
727 comparison. As shown in Figure 16, BCC-ESM1 and HIPPO aircraft observations shows
728 reasonable agreement in terms of the spatial distributions and seasonal variations of BC levels.
729 BCC-ESM1 generally reproduces the observed hemispheric gradients of BC, i.e. the larger
730 burden in the NH compared to the SH, in consistent with Figures 10 and 14. The model shows
731 large overestimations of BC observations over the tropics, which is also found in the
732 CAM4-chem global chemical model (Tilmes et al., 2016).

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4.4 Aerosol Optical Properties

Aerosol optical depth (AOD) is an indicator of the reduction in incoming solar radiation (at a particular wavelength) due to scattering and absorption of sunlight by aerosols. In this study, we calculate the AOD aerosol optical depth (AOD) at 550 nm for all aerosols including sulfate, BC, organic carbon, sea salt and dust as the product of aerosol dry mass concentrations, aerosol water content, and their specific extinction coefficients. The total AOD is calculated by summing the AOD in each model layer for each aerosol species using the assumption that they are externally mixed.

The aerosol optical depth (AOD) observations retrieved from MODIS and MISR over the period of 1997-2003, and from AERONET over the period of 1998-2005 (<http://aeronet.gsfc.nasa.gov>) are used to evaluate the 1997-2003-averaged AOD at 550 nm in BCC-ESM. Figure 137 shows averages of MISR and MODIS AOD with corresponding averages from BCC-ESM. The BCC-ESM1 simulated AOD generally captures the spatial distribution of MISR and MODIS retrievals. The model overestimates AOD over East China. It also systematically underestimates the MODIS observations in the Southern Hemisphere, but is closer to MISR observations.

Figure 18 shows multi-years annual means of BCC-ESM1 simulated AOD values versus observations from AERONET over the period of 1998-2005. The basic pattern of modeled global AOD is similar to that of observations and their spatial correlation reaches 0.56. Large values of AOD are mainly distributed in land continents such as North African, South Asia, East Asia, Europe, and eastern part of North America. Figures 19a-19d4 present scatter plots of observed versus simulated multi-year monthly mean AOD at those sites of AERONET in Europe, North America, East Asia, and South Asia over the period of 1998-2005, respectively. compares the monthly AOD values at 550 nm from BCC-ESM with 1998-2005-averaged monthly observations from AERONET (<http://aeronet.gsfc.nasa.gov>) at sites in Europe, North America, East Asia, and South Asia. Model simulated monthly AOD generally agrees with observations within a factor of 2 for most sites. BCC-ESM slightly overestimates the AOD in European (43.4-55.4°N and 7.6-27.6°E) and North American (43.4-55.4°N and 7.6-27.6°E) sites. In those regions, BCC-ESM also slightly overestimates MODIS and MISR AOD observations (Fig. 173).

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4.5. Summary and discussions

This paper presents a primary comprehensive evaluation of aerosols simulated in version 1 of the Beijing Climate Center Earth System Model (BCC-ESM1) with the implementation of the interactive atmospheric chemistry and aerosol based on the newly developed BCC-CSM2. Global aerosols (including sulfate, organic carbon, black carbon, dust and sea salt) and major greenhouse gases (e.g., O₃, CH₄, N₂O) in the atmosphere can be are interactively simulated when anthropogenic emissions are provided to the model. Concentrations of all aerosols in BCC-ESM1 are determined by the processes of advective transport, emission, gas-phase chemical reactions, dry deposition, gravitational settling, and wet scavenging by clouds and precipitation. The nucleation and coagulation of aerosols are ignored in the present version of BCC-ESM1. Effects of aerosols on radiation, cloud, and precipitation are fully included.

We evaluated the performance of BCC-ESM1 in simulating aerosols and their optical properties in the 20th century following CMIP6 historical simulation according to the requirement of the ~~Aerosol Chemistry Model Intercomparison Project (AerChemMIP)~~. ~~It is forced with anthropogenic emissions evolving from 1850 to 2014 but some WMGHGs such as CH₄, N₂O, CO₂, CFC11 and CFC12 are prescribed using CMIP6 prescribed concentrations (to replace prognostic values of CH₄ and N₂O from the chemistry scheme).~~ The ~~AeroChemMIP historical simulation uses anthropogenic emissions evolving from 1850 to 2014 and prescribes GHG concentrations (e.g., CH₄, N₂O, CO₂, CFC11 and CFC12) using CMIP6 recommended data.~~ Both direct and indirect effects of aerosols are considered in BCC-ESM1. Initial conditions of the ~~AeroChemMIP-CMIP6~~ historical simulation are obtained from a 600-year piControl simulation in the absence of anthropogenic emissions, which well captures the pre-industrial concentrations of sulfate (SO₄²⁻), organic carbon (OC), black carbon (BC), dust, and sea salt aerosols and are consistent with the CMIP5 recommended concentrations for the year 1850.

With the CMIP6 anthropogenic emissions of SO₂, OC, and BC from 1850 to 2014 and their natural emissions implemented in BCC-ESM1, the model simulated SO₄²⁻, BC, and OC

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793 aerosols in the atmosphere are highly correlated with the CMIP5-recommended data. The
794 long-term trends of CMIP5 aerosols from 1850 to 2000 are also well simulated by
795 BCC-ESM1. Global budgets of aerosols were evaluated through comparisons of BCC-ESM1
796 results for 1990-2000 with reports in various literatures observational data at present day for
797 sulfate, BC, OC, sea salt, and dust. Their annual total emissions, global atmospheric mass
798 loading, and mean lifetimes are all within the range of values reported in relevant literatures.

799 Evaluations of the spatial and vertical distributions of BCC-ESM1 simulated present-day
800 sulfate (SO_4^{2-}), OC, BC, Dust, and sSea sSalt aerosol concentrations against the CMIP5
801 datasets and in-situ measurements of surface networks (IMPROVE in the U.S. and EMEP in
802 Europe), and HIPPO aircraft observations indicate good agreement among them. The
803 BCC-ESM1 simulates weaker upward transport of aerosols from the surface to the middle and
804 upper troposphere (with reference to CMIP5-recommended data), likely reflecting a lack of
805 deep convection transport of chemical species in the present version of BCC-ESM1. The
806 aerosol optical depth (AOD) at 550 nm for all aerosols including sulfate, BC, OC, sea salt,
807 and dust aerosols was further compared with the satellite AOD observations retrieved from
808 MODIS and MISR and surface AOD observations from AERONET. The BCC-ESM1 model
809 results are overall in good agreement with these observations within a factor of 2. All these
810 comparisons demonstrate the success of the implementation of interactive aerosol and
811 atmospheric chemistry in BCC-ESM1.

812 This work has only evaluated the ability of BCC-ESM1 to simulate aerosols. The
813 variations of aerosols especially for sulfate are related to other gaseous tracers such as OH
814 and NO₃ (Table 2), which are determined by the MOZART2 gaseous chemical scheme as
815 implemented in BCC-ESM1, and require further evaluation. As limited length of the text, the
816 other optical feature of aerosols such as extinction coefficients, single scattering albedo and
817 asymmetry parameters, and even their feedbacks on radiation and global temperature change
818 will be explored in the other paper. O₃ is evaluated in this work. Other GHGs such as CH₄ and
819 N₂O concentrations can be simulated when forced with emissions and their simulations also
820 need to be evaluated in future. The variations of aerosols especially for sulfate are related to
821 other gaseous tracers such as OH and NO₃ (Table 2), which are determined by the MOZART2
822 gaseous chemical scheme as implemented in BCC-ESM1 and require further evaluation. How

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823 ~~about the GHGs simulations in the AeroChemMIP historical run? Can the global warming be~~
824 ~~reproduced? These questions concerning feedbacks of prognostic aerosols on climate change~~
825 ~~especially global warming also need to be explored in the future.~~▲

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826 **5.6. Code and data availability**

827 ~~Source codes of BCC-ESM1 model are freely available upon request addressed to~~▲
828 Tongwen Wu (twwu@cma.gov.cn). Model output of BCC CMIP6 AerChemMIP simulations
829 described in this paper is distributed through ~~the Earth System Grid Federation (ESGF)~~ and
830 freely accessible through the ESGF data portals after registration. Details about ESGF are
831 presented on the CMIP Panel website at
832 ~~http://www.wcrp-climate.org/index.php/wgcm-cmip/about-cmip.~~▲

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834 **Author contributions**

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

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842 (2016YFA0602100). All the figures are created by the NCAR Command Language (Version
843 6.6.2) [Software].

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1154 **Table 1. Chemical species considered in BCC-AGCM3-Chem. Species marked with star (*)**
 1155 **denote those added in BCC-ESM1 apart from the 63 species used in MOZART2. In the**
 1156 **column of surface emission, interactive surface emissions are considered for sea salt and dust.**
 1157

Species	Dry deposition	Wet deposition	Surface emission	Aircraftplane 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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Table 1. Continued.

Species name	Dry deposition	Wet deposition	Surface emission	Aircraft lane	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					
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*	✓	✓			
SSLT02*	✓	✓			
SSLT03*	✓	✓			
SSLT04*	✓	✓			
DST01*	✓	✓			
DST02*	✓	✓			
DST03*	✓	✓			
DST04*	✓	✓			

Gas tracers

Aerosols

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1163 Table 2. Gas-phase chemical reactions for NH₃ and bulk aerosols precursors following
 1164 CAM-Chem (Lamarque et al., 2012), The reaction rates (s⁻¹) refer to Tie et al. (2001) and
 1165 Sander et al. (2003), Chin et al. (1996), and Cooke and Wilson (1996). Temperature (T) is
 1166 expressed in K, air density (M) in molecule cm⁻³, ki and ko in cm³ molecule⁻¹ s⁻¹.
 1167

Chemical reactions	Rate
<u>NH₃ + OH → H₂O</u>	<u>1.70E-12*exp(-710/T)</u>
<u>SO₂ + OH ⇌ SO₄²⁻SO₂</u>	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
<u>DMS + OH → SO₂</u>	9.60E-12*exp(-234./T)
<u>DMS + OH → .5*SO₂ + .5*HO₂</u>	1.7E-42*exp(7810/T)*M*0.21/(1+5.5E-31*exp(7460/T)* M* 0.21)
<u>DMS + NO₃ → SO₂ + HNO₃</u>	1.90E-13*exp(520/T)
<u>BC1 → BC2</u>	7.10E-06
<u>OC1 → OC2</u>	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})
$\text{SO}_4^{2-}\text{-SO}_4$	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 surface-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 CH	CMIP6	CMIP6					
C ₂ H ₆	CMIP6	CMIP6	ACCMIP		MOZART2		
C ₃ H ₆	CMIP6	CMIP6	On-line		MOZART2		
C ₃ H ₈	CMIP6	CMIP6	ACCMIP		MOZART2		
C ₄ H ₁₀	CMIP6	CMIP6	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		CMIP6	On-line computation				
C ₁₀ H ₁₆		CMIP6	On-line computation				
CH ₄	CMIP6	CMIP6	MOZART2	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	
ACET	ACCMIP	ACCMIP	On-line computation		MOZART2		
ISOP		ACCMIP	On-line computation				
Terpenes		CMIP6	On-line computation				

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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-S; 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	10.7-23.7 ^a
Burden	0.1206	0.042-0.2915 ^a
Lifetime	0.78	0.5-3.0 ^a
SO₂		
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	0.4824	0.4020-1.2206 ^a
Lifetime	1.12	0.6-2.6 ^a
SO₄²⁻		
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	1.89063	1.98066 ± 0.4816 ^b , 1.71057 ^c , 0.64 ^e , 1.2 ^g , 2.22-2.43 ^h
Lifetime	4.69	4.12 ± 0.74 ^b , 3.72-3.77 ^d , 5.4 ^e , 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); e for Liu et al. (2009); 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.

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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 ^c
Wet deposition	6.34	7.5 ^g , 6.10-6.45 ^c
Burden	0.13	0.114 ^c , 0.24 ± 0.1 ^{ba} , 0.11 ^g , 0.14-0.26 ^h , 0.084-0.123 ^c
Lifetime	6.60	7.12 ± 2.35 ^{bd} , 3.95-4.80 ^g , 5.8 ^c , 5.0 ^g , 6.3-7.5 ^h
OC		
Sources	32.2945.20	
Fossil and bio-fuel emission	13.9145.20	
Natural emission	18.38	
Sinks	32.3045.22	50.4 ^d
Dry deposition	2.443.44	
Wet deposition	29.8641.81	
Burden	0.62	0.69 ^e , 1.70 ± 0.45 ^{ba} , 1.0-2.2 ^h
Lifetime	5.00	6.54 ± 1.76 ^b , 4.56-4.90 ^d , 5.3 ^e , 6.4 ^g , 5.4-6.6 ^h , 6.54 ± 1.76 ^a
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 , 4.14 ± 1.783.9 ^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	11.84 ^e , 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

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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), and e for Liu et al. (2016), f for Ginoux et al. (2001), g for Matsui and Mahowald (2017), and h for Tegen et al. (2019).

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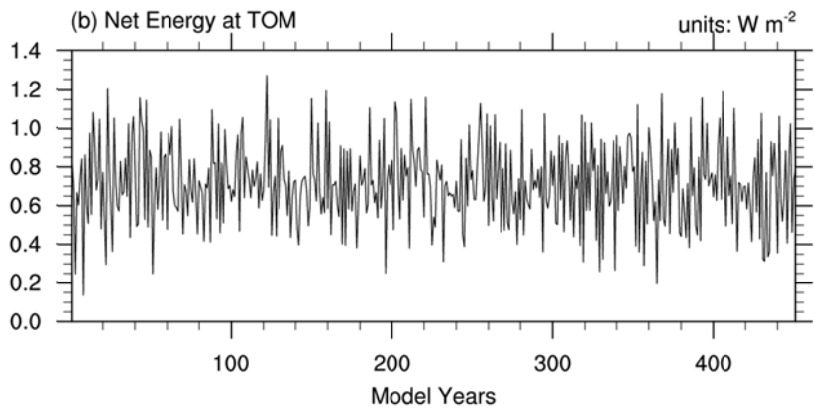
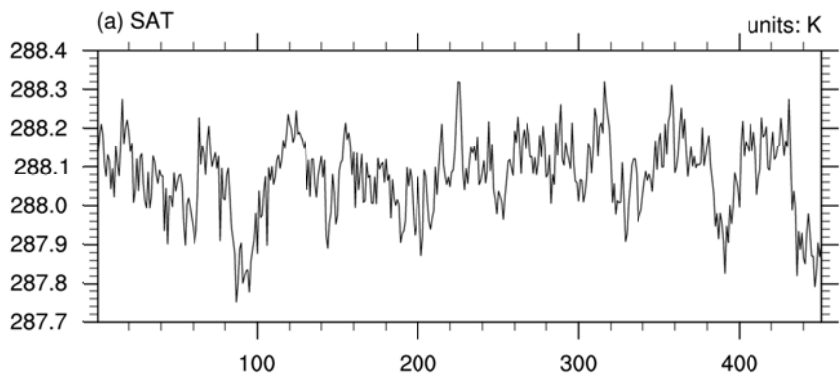
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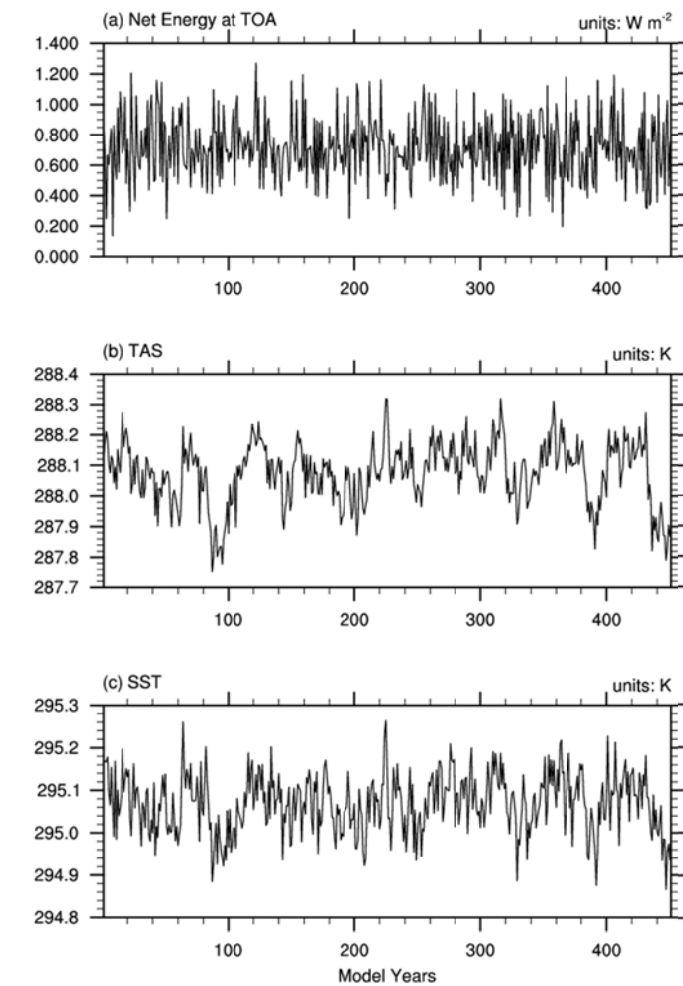


Figure 1. The time series of global and annual mean of (a) net energy budget at top of atmosphere ($W \cdot m^{-2}$), surface air temperature (K) and (b) net energy budget at top of the model ($W \cdot m^{-2}$) near-surface air temperature (K), and (c) sea surface temperature (K) in the last 450 years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

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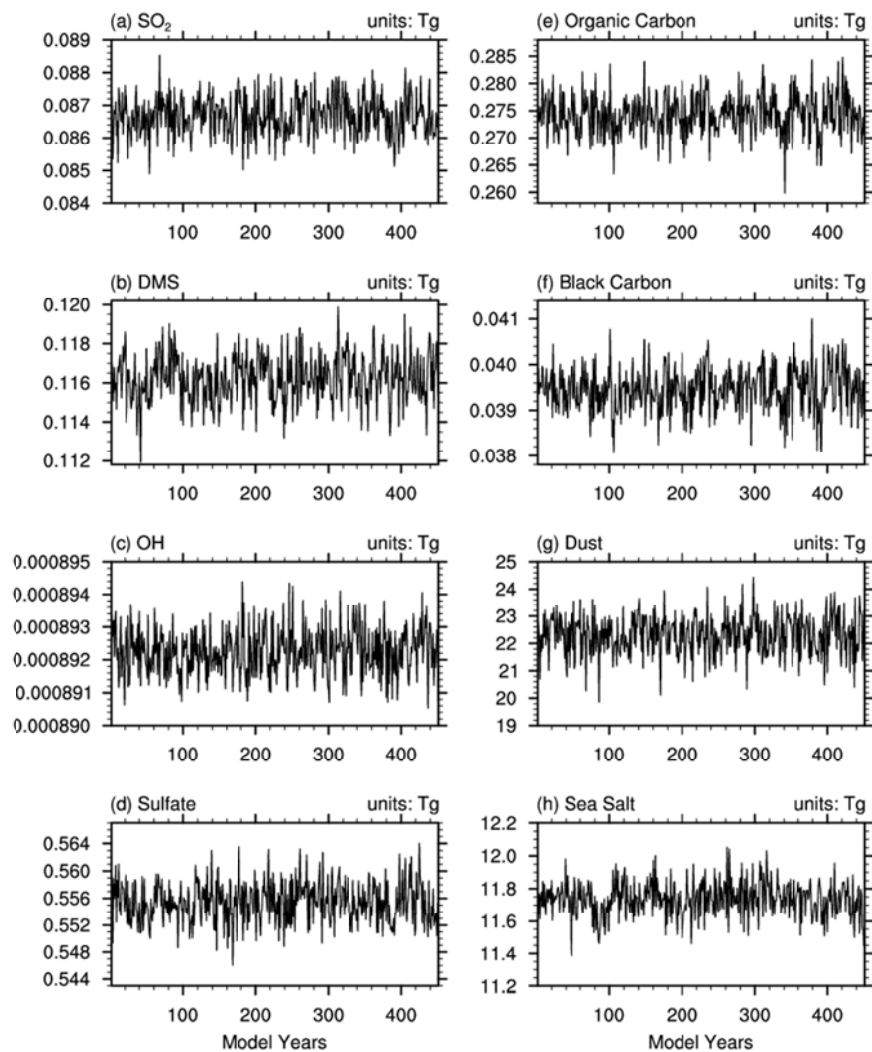


Figure 2. Same as in Figure 1, but for the global burdens of (a) SO₂, (b) DMS, (c) OH, and (d-h) different aerosols (in unit of Tg) in the troposphere (below 100 hPa). Units are Tg.

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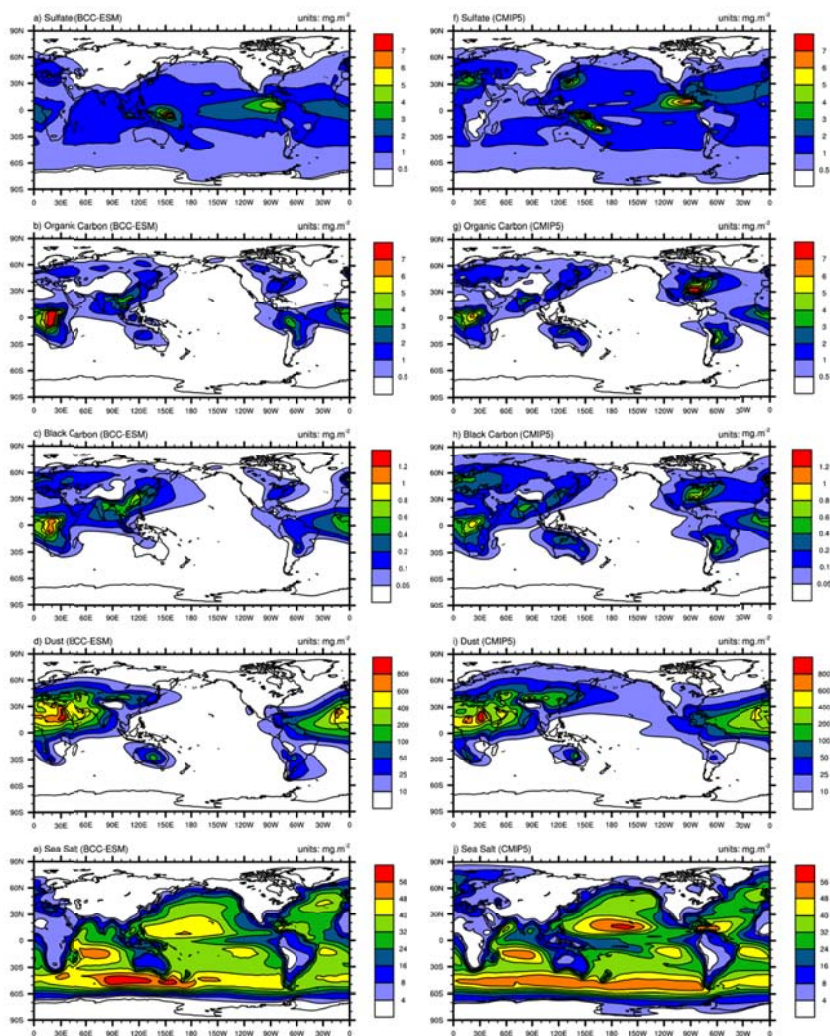


Figure 3. Global distributions of annual mean mass ~~burdens, concentrations~~ of sulfate (SO_4^{2-} ; first row), organic carbon (OC; second row), black carbon (BC; third row), dust (fourth row), and sea salt (fifth row) aerosols in the whole atmospheric column. The left panels show the mean averaged 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 IIASA <http://tntcat.iiasa.ac.at/RcpDb/>). Units: $\text{mg} \cdot \text{m}^{-2}$.

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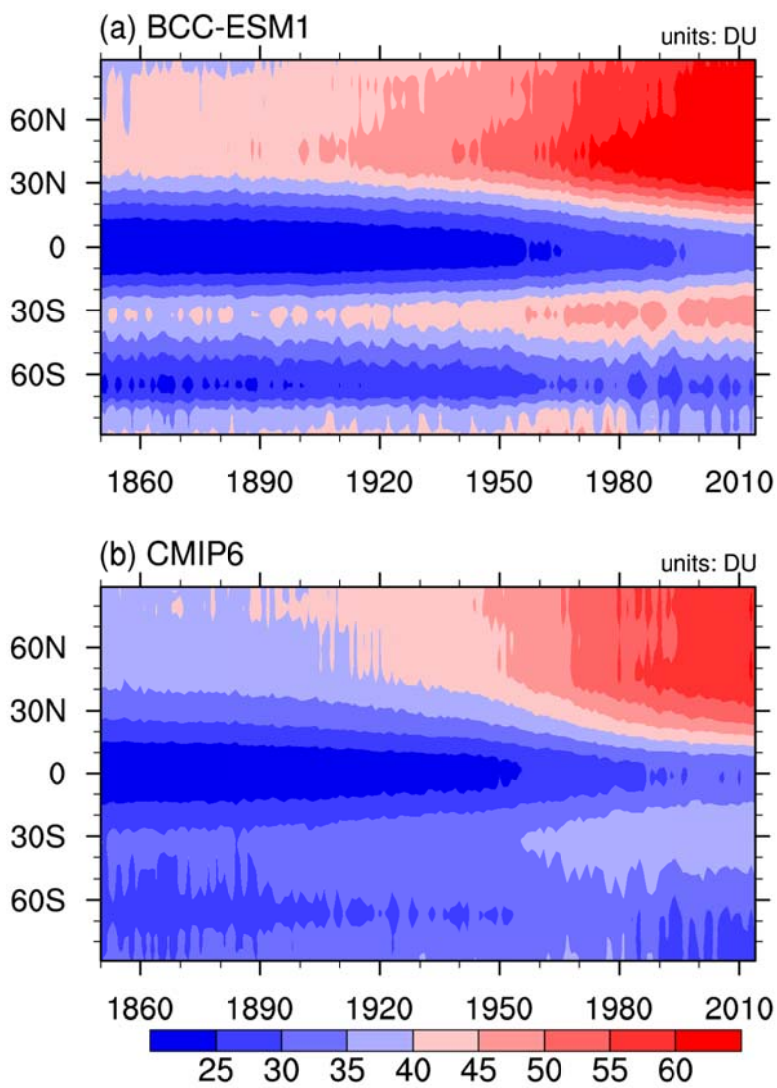


Figure 4. Zonal mean of yearly mean concentration of ozone column in the troposphere below 300 hPa to the ground from 1871 to 1999 for (a) BCC-ESM1 and (b) CMIP6 data. Unit: DU.

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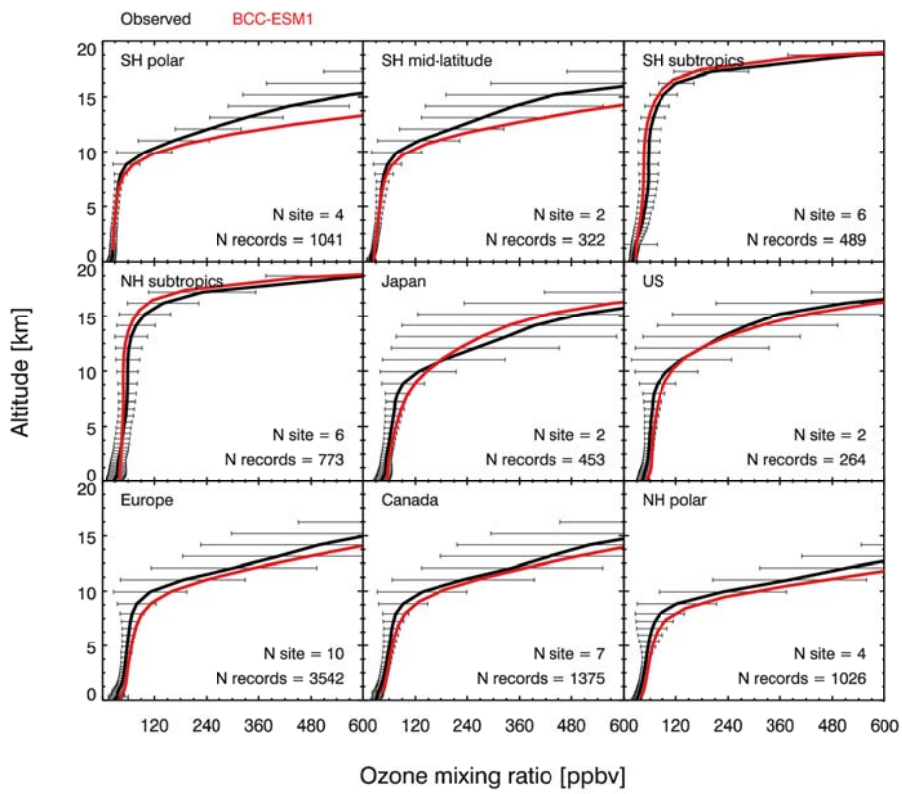


Figure 5. Vertical profiles of annual mean ozone concentrations from observations averaged for 2010-2014 in nine regions (black) and from the BCC-ESM1 simulations (red). The observations are derived from 41 global WUODC sites.

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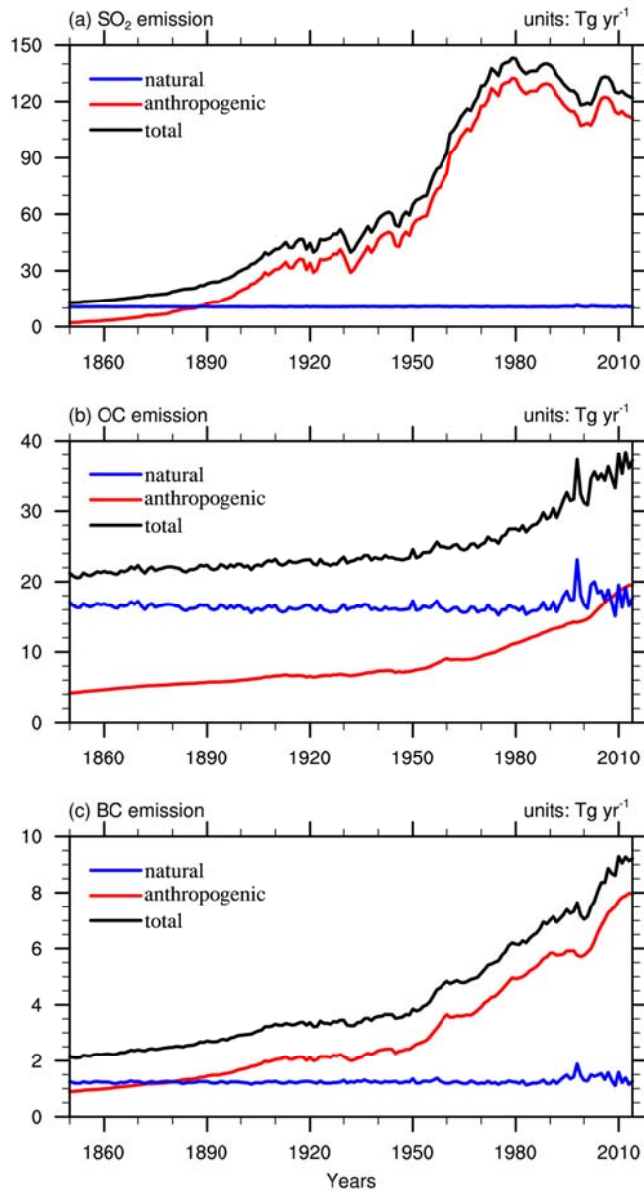
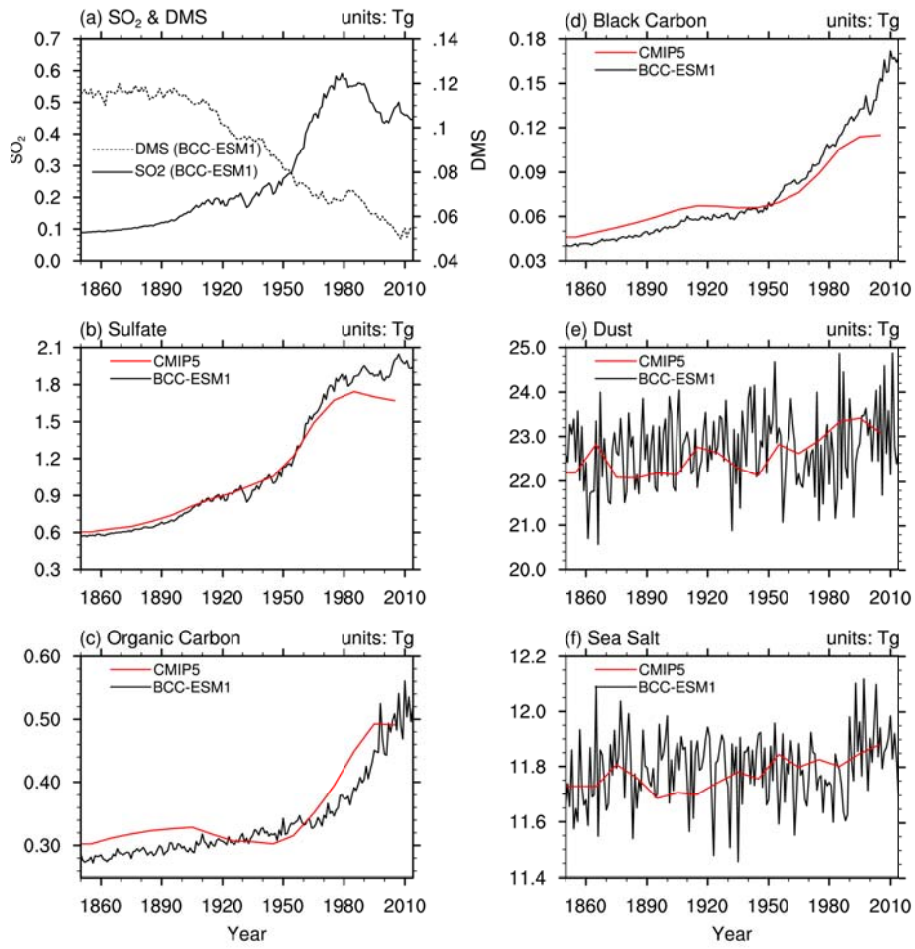


Figure 64. –Global annual anthropogenic, natural, and total emissions of SO₂, organic carbon (OC), and black carbon (BC) in the BCC-ESM1 historical simulation. All the biomass burning emissions are included in natural emissions in (a)-(c). The units are Tg yr⁻¹.

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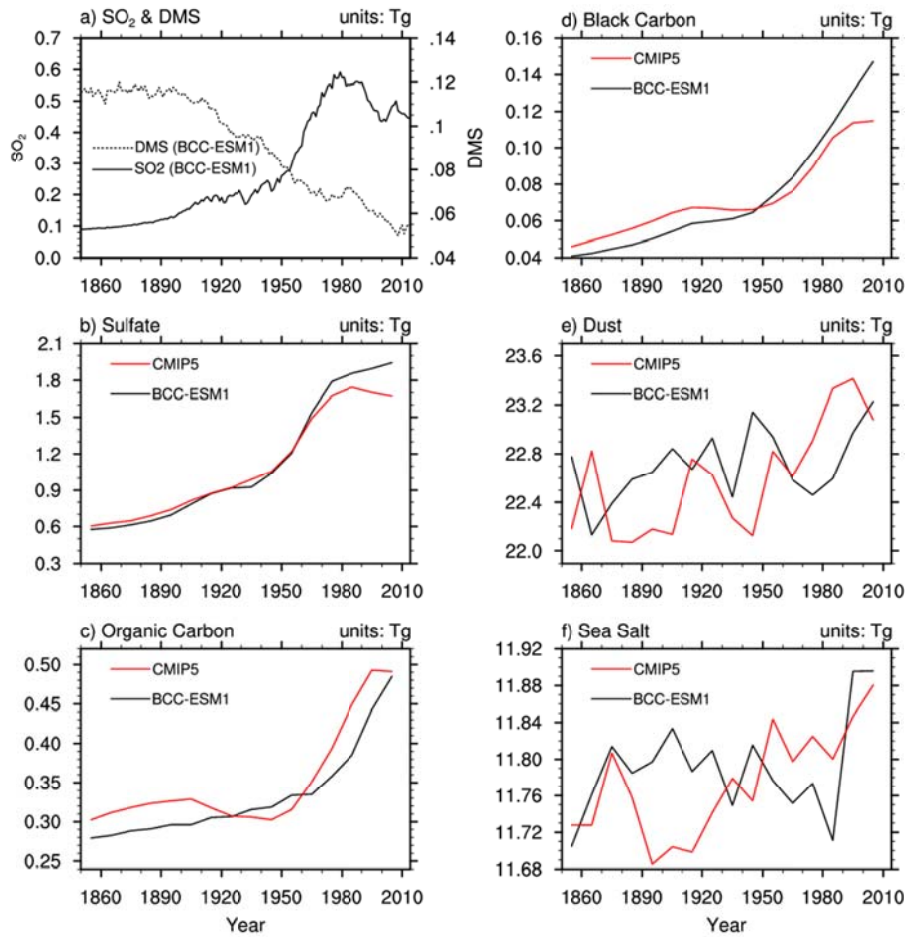


Figure 75. The time series of global yearly annual amounts of (a) SO₂ and DMS and (b-f) aerosols in the whole atmosphere column from the CMIP6 20th historical simulations of BCC-ESM1 (black lines) and the CMIP5-recommended aerosols masses (red lines). The yearly CMIP5 data are interpolated from the time series in 10-year interval. Units: Tg.

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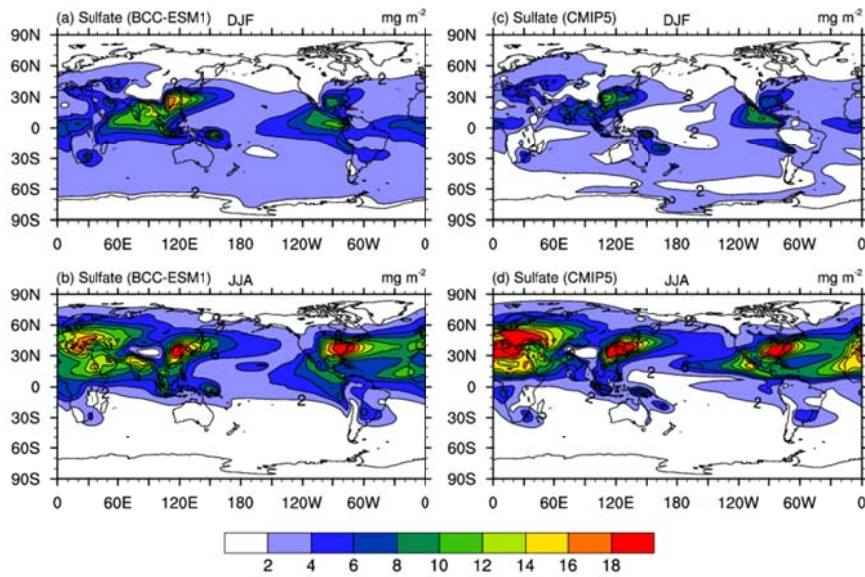


Figure 86. December-January-February (DJF; top panels) and June-July-August (JJA; bottom panels) mean sulfate (SO_4^{2-}) aerosol column mass concentrations averaged for the period of 1979-2000. Left panels show the historical simulations of BCC-ESM1, and right panels the CMIP5-recommended data. Units: $\text{mg}\cdot\text{m}^{-2}$.

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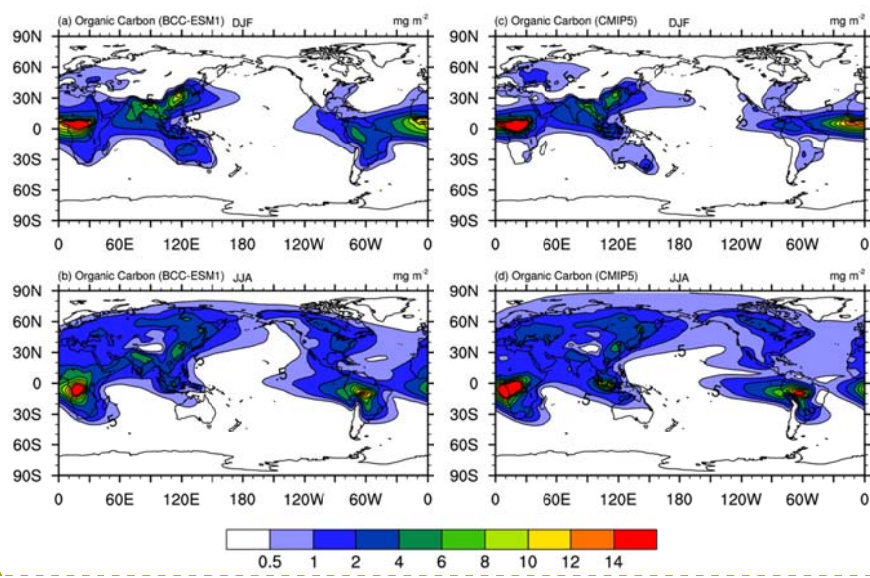
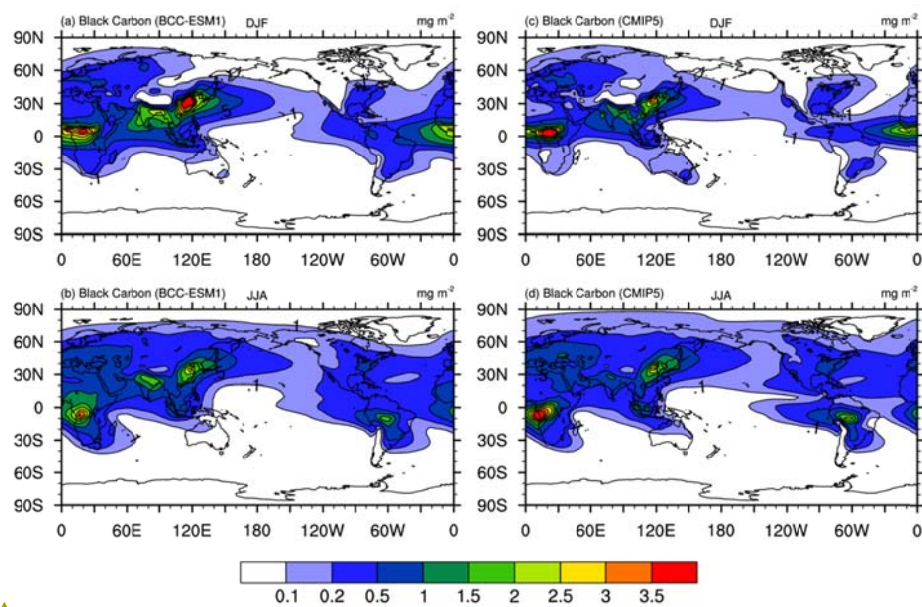


Figure 97. The same as in Figure 86, but for organic carbon (OC) aerosol column mass concentrations. Units: mg m^{-2} .

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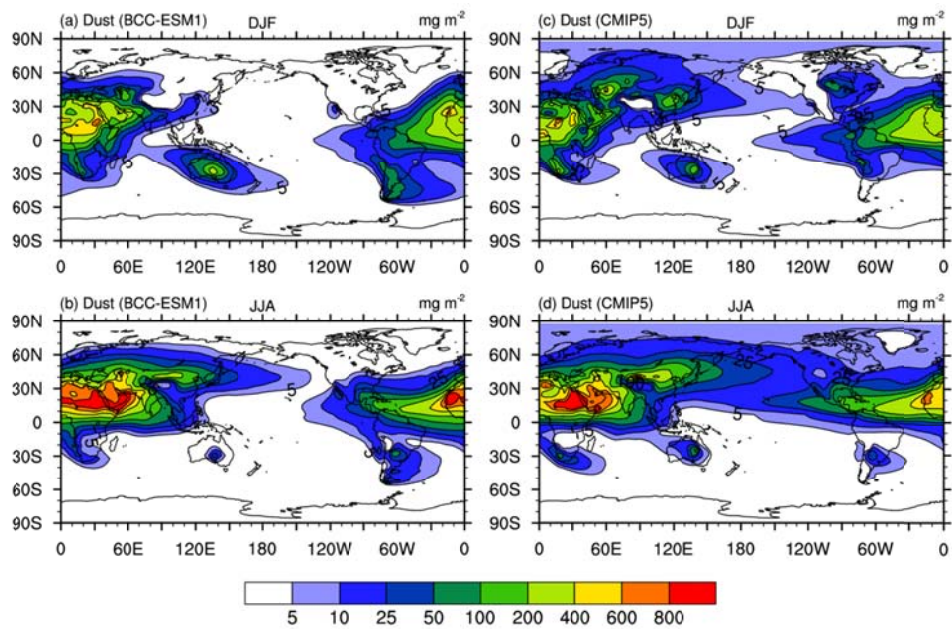
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Figure 108. The same as in Figure 86, but for black carbon (BC) aerosol. Units: mg.m^{-2} .



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Figure 119. The same as in Figure 86, but for dust aerosol. Units: mg.m^{-2} .

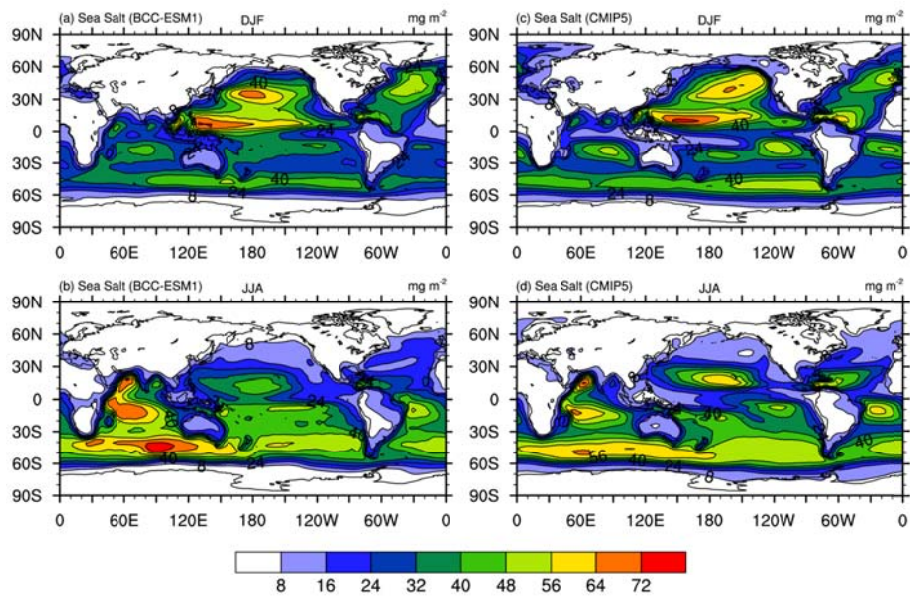


Figure 129. The same as in Figure 86, but for sea salt (SSLT) aerosol. Units: mg.m^{-2} .

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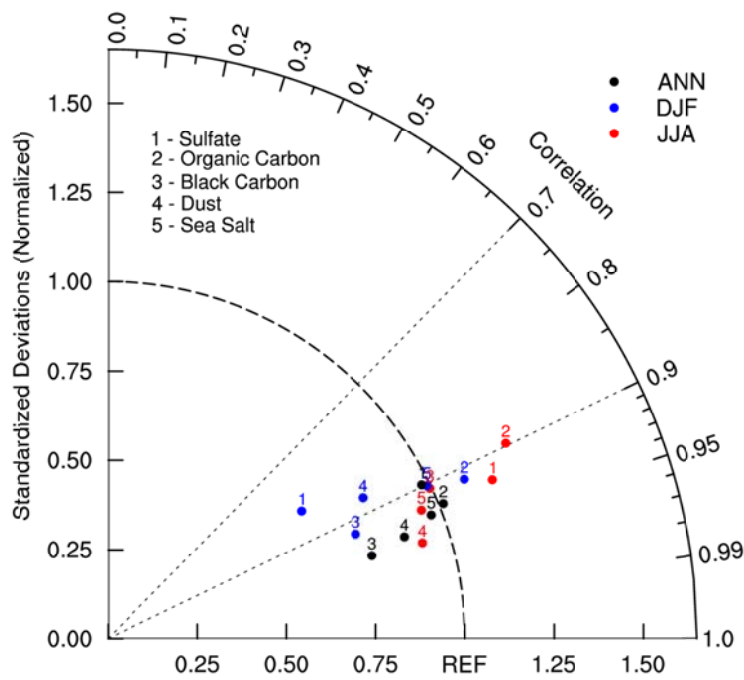


Figure 13. Taylor diagram for the global aerosols climatology (1971–2000) of sulfate, organic carbon, black carbon, dust, and sea salt averaged for December-January-February (DJF), June-July-August (JJA), and annual respectively. The radial coordinate shows the standard deviation of the spatial pattern,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

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带格式的: 两端对齐

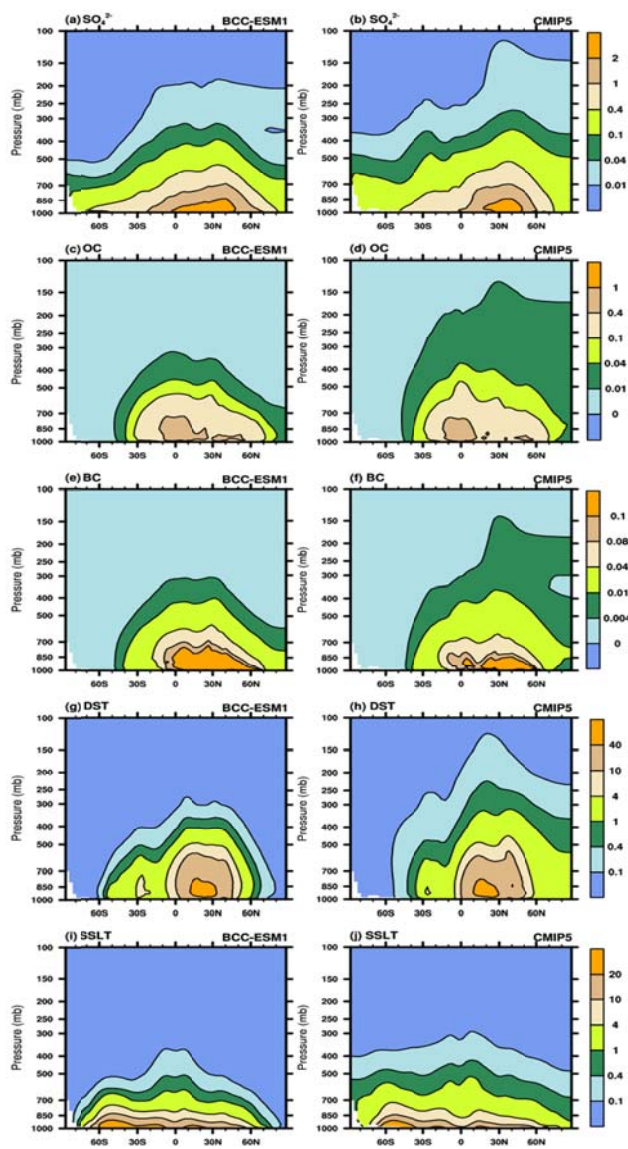


Figure 14. Latitude-pressure distributions of zonally-averaged annual mean sulfate, organic carbon, black carbon, dust, and sea salt aerosol concentrations for the period of 1979-2000. Left panels show the 20th century - CMIP6 historical simulation of BCC-ESM1, and right panels the CMIP5 recommendation data. Units: $\mu\text{g m}^{-3}$.

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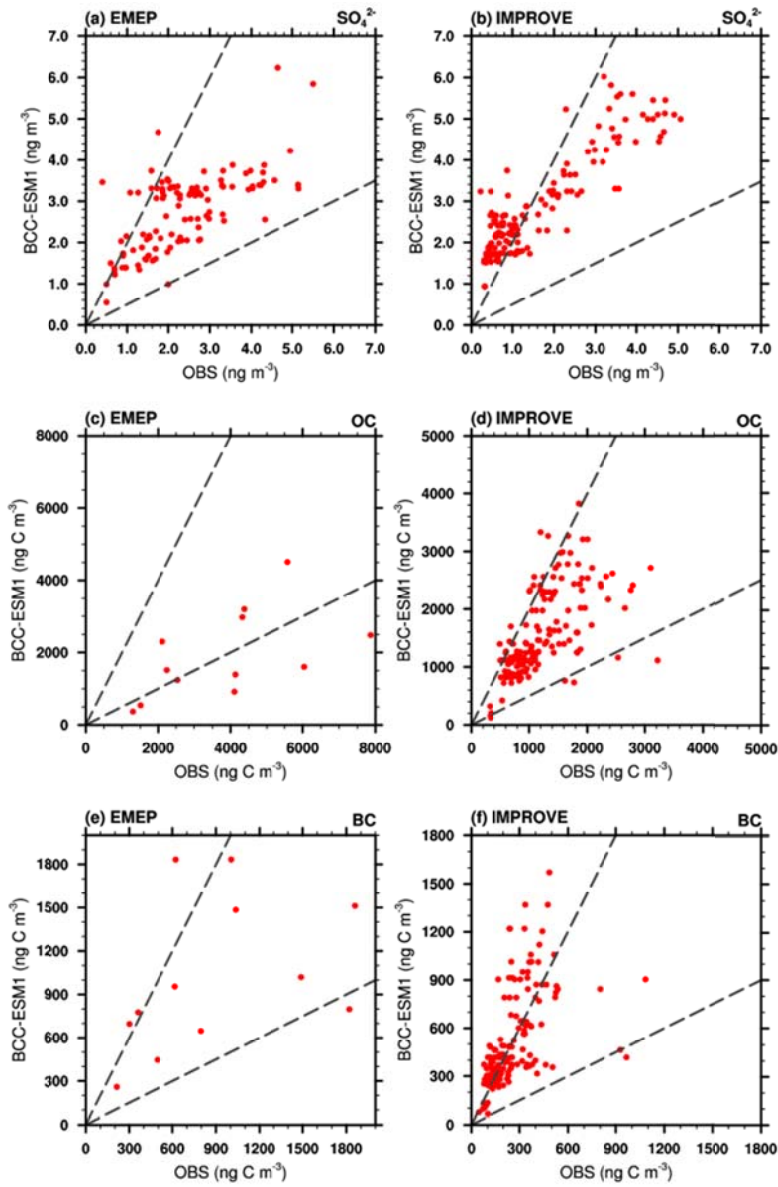


Figure 152. Scatter plots showing observed versus simulated multi-years averaged annual mean sulfate (SO_4^{2-}), organic carbon (OC), black carbon (BC) mixing ratios at IMPROVE and EMEP network sites. Observations are averages over the available years 1990–2005 for IMPROVE sites, and 1995–2005 for EMEP sites. Simulated values are those at the lowest layer of BCC-ESM1.

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Table 7. Observed versus simulated concentrations of sulfate (SO₄²⁻), organic carbon (OC), black carbon (BC) for the regional mean and spatial standard deviation, minimum and maximum values at 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		
	SO ₄ ²⁻ (Obs/Model)	OC (OBS/Model)	BC (OBS/Model)	SO ₄ ²⁻ (OBS/Model)	OC (OBS/Model)	BC (OBS/Model)
▲ Mean Values	2.37/2.74	3844/1919	884/1022	1.53/2.79	1215/1565	249/504
▲ Std Deviation	1.16/0.93	1997/1215	572/526	1.30/1.20	572/745	164/296
▲ Min Values	0.40/0.55	1296/369	214/259	0.22/0.94	322/123	45/66
▲ Max Values	5.50/6.24	7867/4510	1859/1834	5.07/6.02	3219/3827	1084/1570
▲ Correlation (ObsBS and Model)	0.67	0.56	0.40	0.90	0.63	0.55

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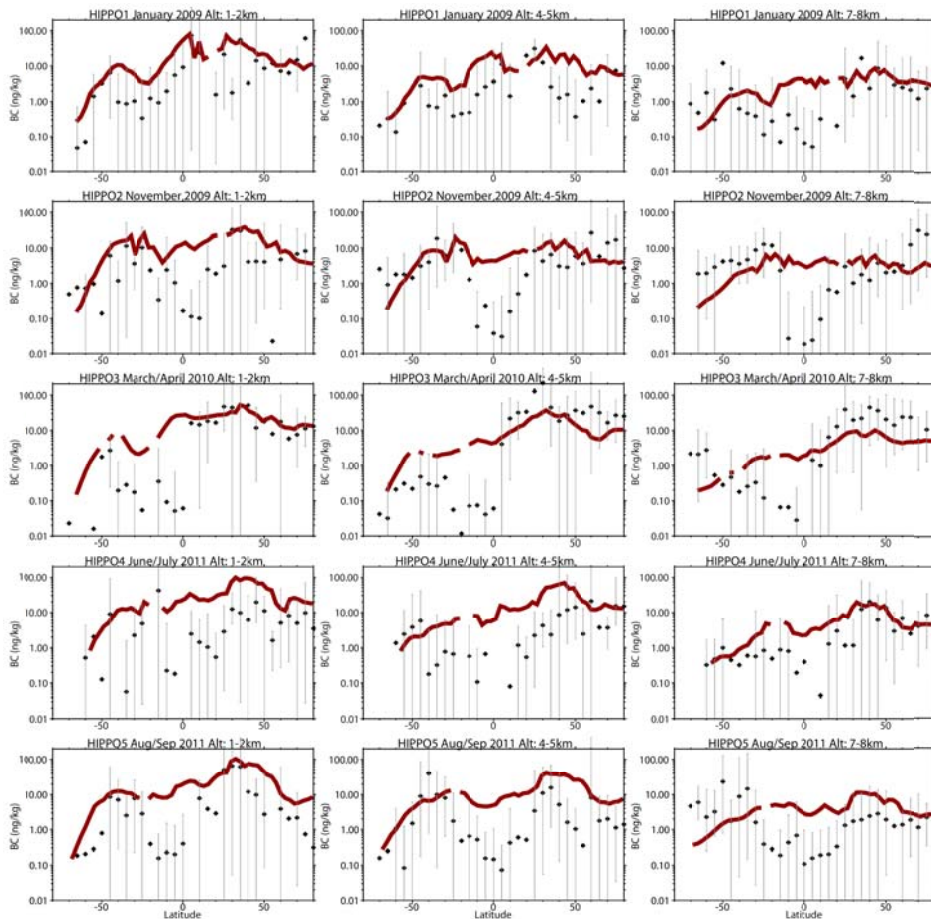
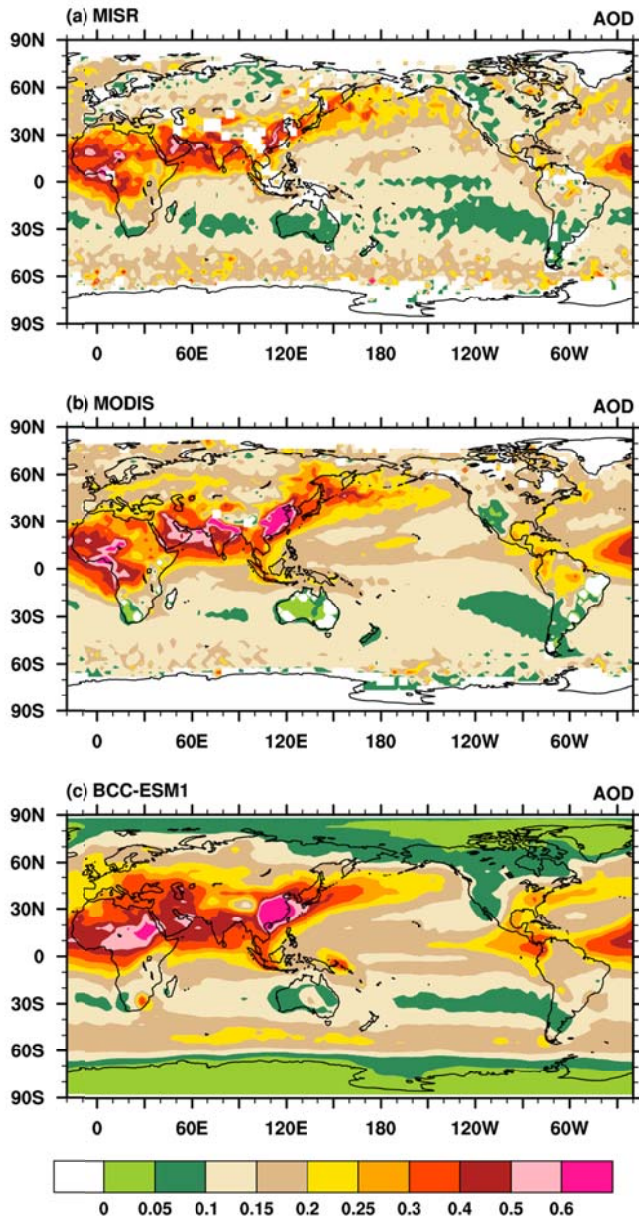


Figure 16. Comparison of modelled black carbon (BC) aerosol (red lines) with observations from HIPPO aircraft campaigns over the Pacific Ocean (black symbols, bars represent the full data range). Observations from different HIPPO campaigns were averaged over 5° latitude bins and three different altitude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

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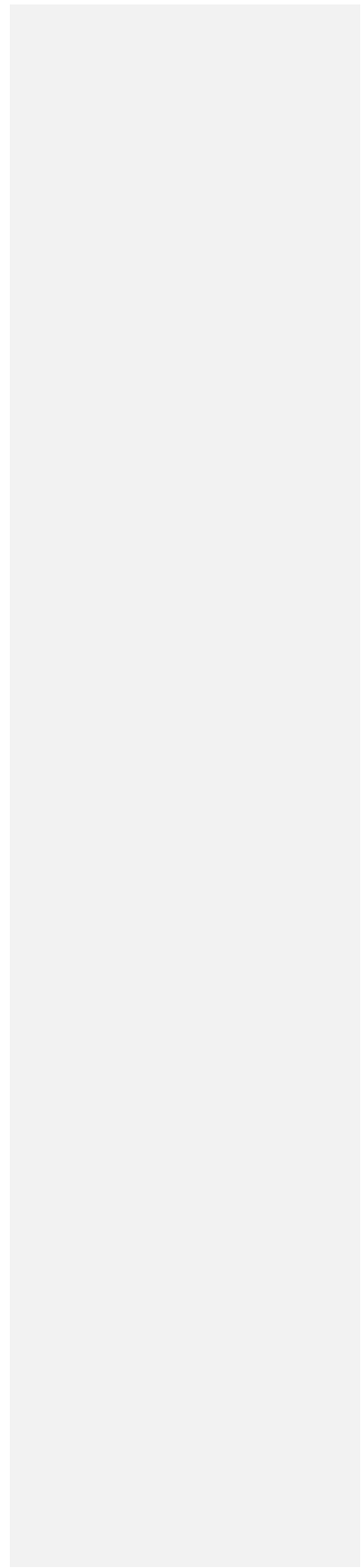


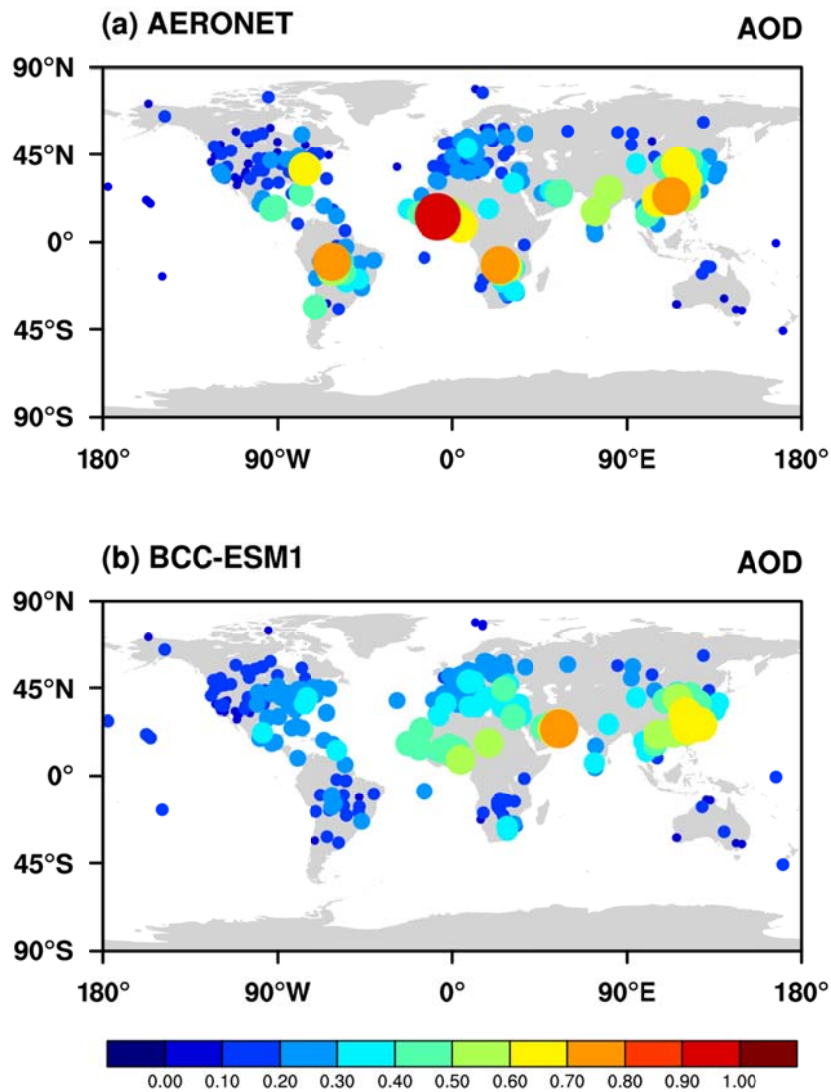
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Figure 173. Global distribution of annual mean AOD simulated in BCC-ESM1 compared with the MISR and MODIS data for the year 2008.

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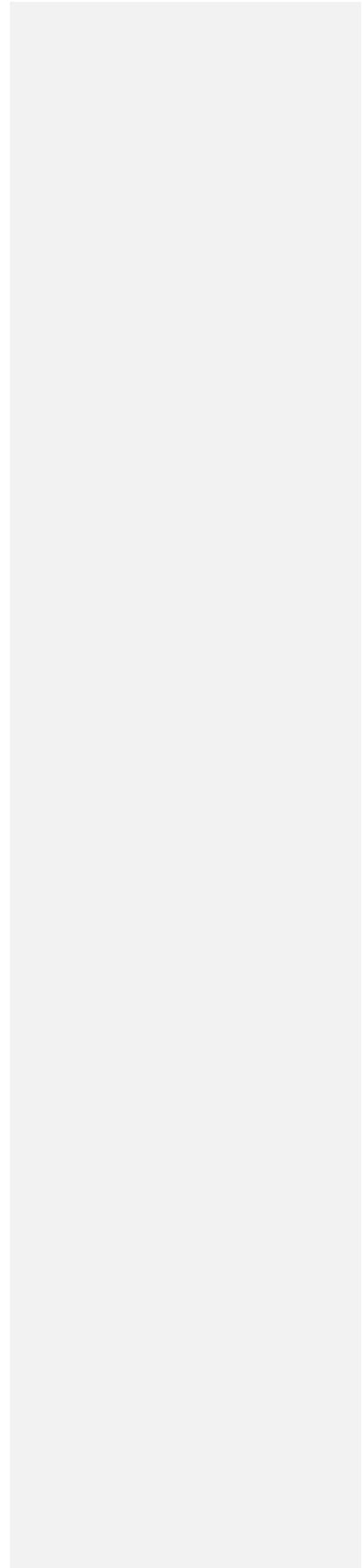


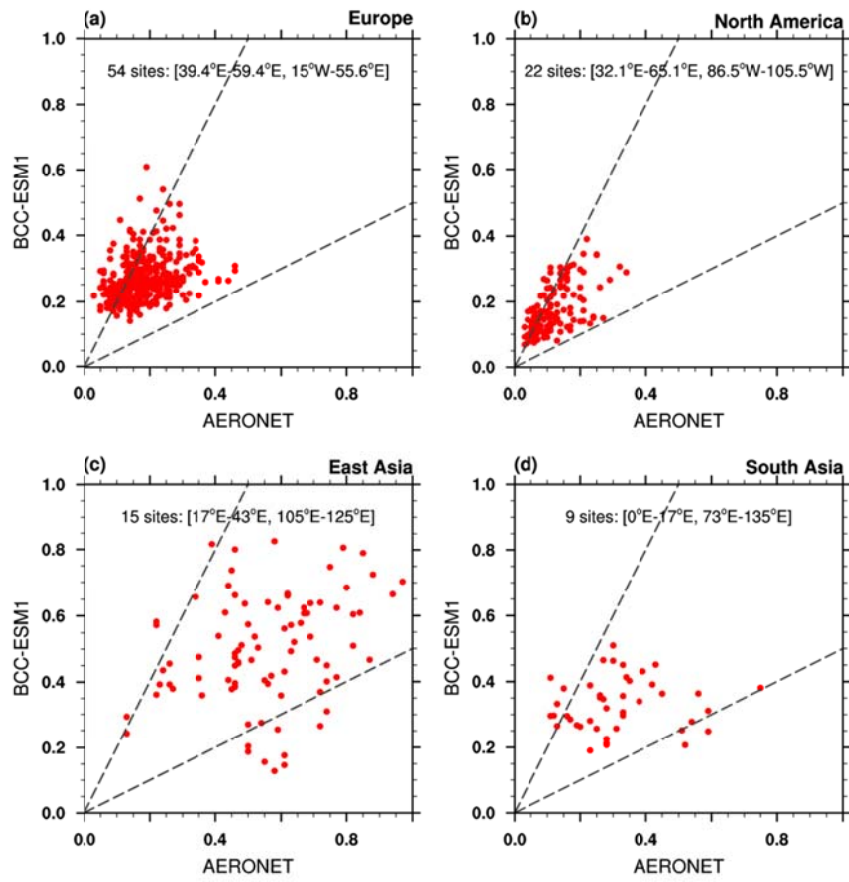
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Figure 18. Observed versus simulated annual means of AOD at AERONET sites. Each data point represents the mean averaged for available monthly values of AOD. The dot sizes denote the magnitudes of AOD at sites. The spatial correlation is 0.56.

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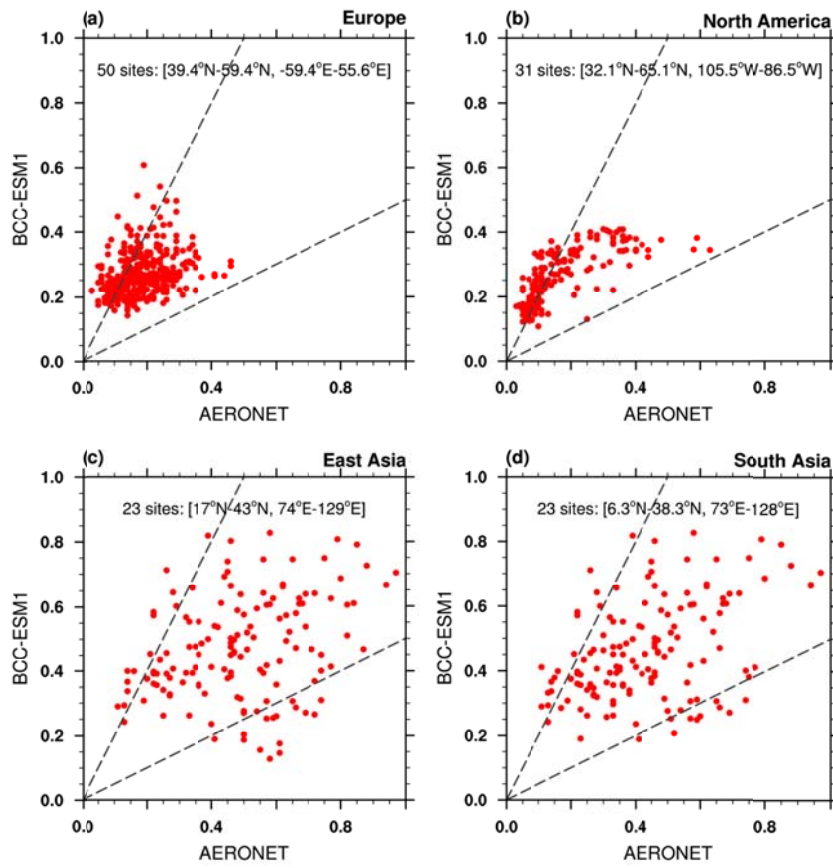


Figure 194. Scatter plots of observed versus simulated monthly mean AOD at AERONET sites in Europe, North America, East Asia, and South Asia over the period of 1998-2005. Each data point represents an available monthly mean AOD at a site and its corresponding model result over 1998-2005.

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页 12: [1] 带格式的 吴统文(拟稿) 2019/10/6 17:14:00

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字体: 11 磅, 字体颜色: 文字 1

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字体: 11 磅, 字体颜色: 文字 1

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字体: 11 磅, 字体颜色: 文字 1

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字体: 11 磅, 字体颜色: 文字 1

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页 13: [18] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

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字体: 小五, 字体颜色: 文字 1

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字体颜色: 文字 1, 下标

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字体颜色: 文字 1

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字体颜色: 文字 1

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字体: 小五, 字体颜色: 文字 1

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字体: 小五, 字体颜色: 文字 1

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字体: 小五, 字体颜色: 文字 1

页 44: [45] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [46] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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页 44: [46] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [46] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [46] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [47] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [47] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

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页 44: [47] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

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字体: 小五, 字体颜色: 文字 1

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字体: 小五, 字体颜色: 文字 1

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字体: 小五, 字体颜色: 文字 1

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字体: 小五, 字体颜色: 文字 1

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字体: 小五, 字体颜色: 文字 1

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字体: 小五, 字体颜色: 文字 1

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字体: 小五, 字体颜色: 文字 1

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字体颜色: 文字 1

页 44: [52] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

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字体: 小五, 字体颜色: 文字 1

页 44: [53] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [53] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [53] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [54] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [54] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [54] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [55] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [55] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [55] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [56] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [56] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [56] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [57] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [58] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [58] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [58] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [59] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [59] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [59] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [60] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [60] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [60] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [61] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体: 小五, 字体颜色: 文字 1

页 44: [62] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [63] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [64] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [65] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [66] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [67] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [68] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [69] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [70] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 44: [71] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: 小五, 字体颜色: 文字 1

页 46: [72] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 上标

页 46: [72] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 上标

页 46: [73] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [73] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [74] 带格式的 吴统文(拟稿) 2019/10/22 16:46:00

非上标/ 下标

页 46: [74] 带格式的 吴统文(拟稿) 2019/10/22 16:46:00

非上标/ 下标

页 46: [74] 带格式的 吴统文(拟稿) 2019/10/22 16:46:00

非上标/ 下标

页 46: [75] 带格式的 吴统文(拟稿) 2019/10/22 16:46:00

上标

页 46: [75] 带格式的 吴统文(拟稿) 2019/10/22 16:46:00

上标

页 46: [76] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [76] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [76] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [76] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [76] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [76] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [77] 带格式的 吴统文(拟稿) 2019/10/22 16:44:00

上标

页 46: [77] 带格式的 吴统文(拟稿) 2019/10/22 16:44:00

上标

页 46: [78] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1

页 46: [78] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1

页 46: [78] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1

页 46: [79] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [79] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [79] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [79] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [80] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 上标

页 46: [80] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 上标

页 46: [81] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [81] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [82] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [82] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [83] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: (默认) Times New Roman, 字体颜色: 文字 1

页 46: [83] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: (默认) Times New Roman, 字体颜色: 文字 1

页 46: [83] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: (默认) Times New Roman, 字体颜色: 文字 1

页 46: [83] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: (默认) Times New Roman, 字体颜色: 文字 1

页 46: [83] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: (默认) Times New Roman, 字体颜色: 文字 1

页 46: [83] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: (默认) Times New Roman, 字体颜色: 文字 1

页 46: [83] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体: (默认) Times New Roman, 字体颜色: 文字 1

页 46: [84] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [84] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [84] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [84] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [85] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [85] 带格式的 吴统文(拟稿) 2019/10/15 19:27:00

字体颜色: 文字 1, 非上标/ 下标

页 46: [85] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [85] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [85] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [85] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [85] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [86] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [86] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [86] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [86] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1, 非上标/ 下标

页 46: [87] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1

页 46: [87] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1

页 46: [87] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1

页 46: [87] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1

页 46: [87] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1

页 46: [87] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1

页 46: [87] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1

页 46: [87] 带格式的	吴统文(拟稿)	2019/10/15 19:27:00
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字体颜色: 文字 1