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 +0.7Wm-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 1.0 Wm-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 (1.0 Wm-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 288.1 K (Fig. 1b) and 295.05 K (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.

   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 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).”

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

The globally-averaged mean of the total NOx 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 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, N2O, HNO3, PAN, CH3COCH3, CH3COOHH, CH3CHO, CH3COOCHO, NO, and HNO4 are not computed interactively and directly interpolated from MOZART2 climatological monthly mean deposition velocities (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, ISOPOOH, XOOH, HYDRA LD, 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 Wesley 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 Wesley (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 Wesley (1989) using the formula, \( W_{SO₂} = \frac{1}{r_a + r_b + r_c} \), 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.”


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 \(4.05 \times 10^{-15}, 4.52 \times 10^{-14}, 1.15 \times 10^{-13}, 1.20 \times 10^{-13}\) 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*
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).”*
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 emissios 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](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 (SO\(_4^{2-}\)), 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 parameter

*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 (SO\(_4^{2-}\)), 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 paper

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

37. L530: you indicate that you used prescribed concentrations for CH\(_4\), and in Table 4 you indicate that you consider CH\(_4\) emissions. Please clarify

*CH\(_4\) is a prognostic variable in the chemistry scheme of BCC-ESM1. So, emission of CH\(_4\) listed in Table 4 is used to simulate CH\(_4\) concentration, but some WMGHGs such as CH\(_4\), N\(_2\)O, CO\(_2\), 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/remark1 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 1º longitude by 1/3º latitude between 30ºS and 30ºN ranged to 1º longitude by 1º latitude from 60ºS and 60ºN 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.


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

Model Description and Evaluation of Aerosol Simulations

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Submit to Geosci. Model Dev.

Revised on Oct. 23, 2019
Abstract. BCC-ESM1 is the first version of a fully-coupled Earth System Model with interactive atmospheric chemistry and aerosols developed by the Beijing Climate Center, China Meteorological Administration. Major aerosol species (including sulfate, organic carbon, black carbon, dust and sea salt) and greenhouse gases are interactively simulated with a whole panoply of processes controlling emission, transport, gas-phase chemical reactions, secondary aerosol formation, gravitational settling, dry deposition, and wet scavenging by clouds and precipitation. Effects of aerosols on radiation, cloud, and precipitation are fully treated. The performance of BCC-ESM1 in simulating aerosols and their optical properties is comprehensively evaluated as required by the Aerosol Chemistry Model Intercomparison Project (AerChemMIP), covering the preindustrial mean state and time evolution from 1850 to 2014. The simulated aerosols from BCC-ESM1 are quite coherent with CMIP5-recommended data, and in-situ measurements from surface networks (such as IMPROVE in the U.S. and EMEP in Europe), and aircraft observations. A comparison of the modeled aerosol optical depth (AOD) at 550 nm for all aerosols with the satellite AOD observations retrieved from Moderate Resolution Imaging Spectroradiometer (MODIS) and Multi-angle Imaging SpectroRadiometer (MISR) and surface AOD observations from AErosol ROBOTic NETwork (AERONET) shows reasonable agreements between simulated and observed AOD. However, BCC-ESM1 seems to show weaker upward transport of aerosols from the surface to the middle and upper troposphere, likely reflecting the deficiency of representing deep convective transport of chemical species in BCC-ESM1. With an overall good agreement between BCC-ESM1 simulated and observed aerosol properties, it demonstrates a success of the implementation of interactive aerosol and atmospheric chemistry in BCC-ESM1.
1. Introduction

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

Since 2013, the Beijing Climate Center (BCC), China Meteorological Administration, has continuously developed and updated its global fully-coupled GCM climate model, the Beijing Climate Center Climate System Model (BCC-CSM) (Wu et al., 2013; Wu et al., 2014; Wu et al., 2019). BCC-CSM version 1.1 was one of the comprehensive carbon-climate models participating in the phase five of the Coupled Model Intercomparison Project (CMIP5, Taylor et al. 2012). When forced by prescribed historical emissions of CO₂ from combustion of fossil fuels and land use change, BCC-CSM1.1 successfully reproduced the trends of observed atmospheric CO₂ concentration and global surface air temperature from 1850 to 2005 (Wu et al., 2013). During recent years, BCC-CSM1.1 has been used in numerous investigations on soil organic carbon changes (e.g. Todd-Brown et al., 2014), ocean
biogeochemistry changes (e.g. Mora et al., 2013), and carbon-climate feedbacks (e.g. Arora et al., 2013; Hoffman et al., 2014). BCC-CSM includes main climate-carbon cycle processes (Wu et al., 2013) and the global mean atmospheric CO₂ concentration is calculated from a prognostic equation of CO₂ budget taking into account global anthropogenic CO₂ emissions and interactive land-atmosphere and ocean-atmosphere CO₂ exchanges.

In recent years, BCC has put much efforts in developing a global climate-chemistry-aerosol fully-coupled Earth System Model (BCC-ESM1) on the basis of BCC-CSM2 (Wu et al., 2019). The objective is to interactively simulate global aerosols (e.g. sulfate, black carbon, etc.) and main greenhouse gases (e.g. O₃, CH₄, NO₂ and CO₂) in the atmosphere and to investigate feedbacks between climate and atmospheric chemistry. BCC-ESM1 is at the point to be publicly released, and it is actively used by BCC for several CMIP6-endorsed research initiatives (Eyring et al. 2016), including the Aerosol Chemistry Model Intercomparison Project (AerChemMIP, Collins et al., 2017) and the Coupled Climate–Carbon Cycle Model Intercomparison Project (C4MIP, Jones et al. 2016).

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

2. Model description

BCC-ESM1 is an Earth System Model with interactive chemistry and aerosol components, in which the atmospheric component is BCC Atmospheric General Model version 3 (Wu et al., 2019) with interactive atmospheric chemistry (hereafter BCC-AGCM3-Chem), land component BCC Atmosphere and Vegetation Interaction Model version 2.0 (hereafter BCC-AVIM2.0), ocean component Modular Ocean Model version 4 (MOM4)-L40, and sea ice component [sea ice simulator (SIS)]. Different components of
BCC-ESM1 are fully coupled and interact with each other through fluxes of a fully-coupled global climate-chemistry-aerosol model. Different components of BCC-ESM1 interact with each other through fluxes of momentum, energy, water, carbon and other tracers at their interfaces. The coupling between the atmosphere and the ocean is done every hour. The atmospheric component BCC-AGCM3-Chem is BCC Atmospheric General Circulation Model version 3 (Wu et al., 2019) with interactive atmospheric chemistry (hereafter BCC-AGCM3-Chem), is able to simulate global atmospheric composition and aerosols from anthropogenic emissions as forcing agents. Its resolution is T42 (approximately 2.8125°x2.8125° transformed spectral grid). The model has 26 levels in a hybrid sigma/pressure vertical coordinate system with the top level at 2.914 hPa. Details of the model physics are described in Wu et al. (2019). The land component is BCC Atmosphere and Vegetation Interaction Model version 2.0 (BCC-AVIM2.0) with terrestrial carbon cycle. The land component BCC-AVIM2.0 is described in details in Li et al. (2019). It includes biophysical, physiological, and soil carbon-nitrogen dynamical processes, and the terrestrial carbon cycle operates through a series of biochemical and physiological processes on photosynthesis and respiration of vegetation. Biogenic emissions from vegetation are computed online in BCC-AVIM2.0 following the algorithm of the Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1, Guenther et al., 2012). The oceanic component is the Modular Ocean Model version 4 with 40 levels (hereafter MOM4.4L40). The land component is BCC Atmosphere and Vegetation Interaction Model version 2.0 (BCC-AVIM2.0) with terrestrial carbon cycle. The sea ice component is Sea Ice Simulator (SIS). Different components of BCC ESM1 interact with each other through fluxes of momentum, energy, water, carbon and other tracers at their interfaces. The coupling between the atmosphere and the ocean is done every hour. BCC-AGCM3-Chem is able to simulate global atmospheric composition and aerosols with anthropogenic emissions as forcing. It is developed on the basis of the recent version 3 of the Beijing Climate Center atmospheric general circulation model (hereafter BCC-AGCM3, Wu et al., 2019). The horizontal resolution of BCC-AGCM3-Chem is T42 (approximately 2.8125°x2.8125° transformed spectral grid). The model has 26 levels in a hybrid sigma/pressure vertical coordinate system with the top level at 2.914 hPa. The land component BCC-AVIM2.0 is described in details in Li et al. (2019). It includes biophysical, physiological, and soil carbon-nitrogen dynamical processes, and the terrestrial carbon cycle
operates through a series of biochemical and physiological processes on photosynthesis and respiration of vegetation. Biogenic emissions from vegetation are computed online in BCC-AVIM2.0 following the algorithm of the Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1, Guenther et al., 2012). Details of oceanic component MOM4-L40 and sea ice component SIS that are used in BCC-ESM1 may be found in Wu et al. (2013) and Wu et al. (2019). MOM4-L40 used a tri-polar grid of horizontal resolution with 1º longitude by 1/3º latitude between 30ºS and 30ºN ranged to 1º latitude at 60ºS and 60ºN and 40 z-levels in the vertical. Carbon exchange between the atmosphere and the ocean are calculated online in MOM4-L40 using a biogeochemistry module that is based on the protocols from the Ocean Carbon Cycle Model Intercomparison Project–Phase 2 (OCMIP2, http://www.ipsl.jussieu.fr/OCMIP/phase2/). SIS has the same horizontal resolution as MOM4-L40 and three layers in the vertical, including one layer of snow cover and two layers of equally sized sea ice.

The BCC-AGCM3-Chem combines 66 gas-phase chemical species and 13 bulk aerosol compounds as listed in Table 1. Apart from 32 gas-phase species of dimethyl sulfide (DMS), and SO2 and NH3, the other 63 gas-phase species are the same as those in the “standard version” of MOZART2 (Model for Ozone and Related chemical Tracers, version 2), a global chemical transport model for the troposphere developed by the National Center for Atmospheric Research (NCAR) driven by meteorological fields from either climate models or assimilations of meteorological observations (Horowitz et al., 2003). Advection of all tracers in BCC-AGCM3-Chem is performed through a semi-Lagrangian scheme (Williamson and Rasch, 1989), and vertical diffusion within the boundary layer follows the parameterization of Holtslag and Boville (1993). The gas-phase chemistry of the 63 MOZART2 gas-phase species as listed in Table 1 is treated in the same way as that in the “standard version” of MOZART2 (Horowitz et al., 2003). There are 33 photolytic reactions and 135 chemical reactions involving 30 dry deposited chemical species and 25 soluble gas-phase species. Their dry depositions are calculated following the resistance-in-series approach originally described in Wesely (1989). 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)) 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 and those for the other 15 species including MPAN, ONIT, ONITR, C\textsubscript{2}H\textsubscript{5}OH, POOH, C\textsubscript{3}H\textsubscript{7}OOH, ROOH, GLYALD, HYAC, CH\textsubscript{3}OH, MACROOH, ISOPOOH, XOOH, HYDRALD, and H\textsubscript{2} are calculated determined using a weighted combination of prescribed deposition velocities of O\textsubscript{3} ozone, CO, or CH\textsubscript{3}CHO or land surface type and surface temperature following the MOZART2 (Horowitz et al., 2003).

Wet removals by in-cloud scavenging for 25 soluble gas-phase species in the “standard version” of MOZART2 use the parameterization of Giorgi and Chameides (1985) based on their temperature dependent effective Henry’s law constants. In-cloud scavenging is proportional to the amount of cloud condensate converted to precipitation, and the loss rate depends on the amount of cloud water, the rate of precipitation formation, and the rate of tracer uptake by the liquid phase water. Other highly soluble species such as HNO\textsubscript{3}, H\textsubscript{2}O\textsubscript{2}, ONIT, ISOPOOH, MACROOH, XOOH, and Pb-210 are also removed by below-cloud washout as calculated using the formulation of Brasseur et al. (1998). Below-cloud scavenging is proportional to the precipitation flux in each model layer and the loss rate depends on the precipitation rate. 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.

The BCC-AVIM2.0 is the land model with terrestrial carbon cycle. It is described in details in Li et al. (2019) and includes biophysical, physiological, and soil carbon-nitrogen dynamical processes. The terrestrial carbon cycle operates through a series of biochemical and physiological processes on photosynthesis and respiration of vegetation. Biogenic emissions from vegetation are computed online in BCC-AVIM2.0 following the algorithm of the Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1, Guenther et al., 2012).
The oceanic component of BCC-ESM1 is the Modular Ocean Model version 4 with 40
levels (hereafter MOM4-L40), and the sea ice component Sea Ice Simulator (SIS).
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. Carbon exchange between the atmosphere and the ocean are
calculated online in MOM4-L40 using a biogeochemistry module that is based on the
protocols from the Ocean Carbon Cycle Model Intercomparison Project-Phase 2 (OCMIP2,
http://www.ipsl.jussieu.fr/OCMIP/phase2/). SIS has the same horizontal resolution as
MOM4-L40 and three layers in the vertical, including one layer of snow cover and two layers
of equally sized sea ice. Details of oceanic component MOM4-L40 and sea-ice component
SIS that are used in BCC-ESM1 may be found in Wu et al. (2013) and Wu et al. (2019).

In the following sub-sections, we will describe the treatments in BCC-ESM1 for 32
gas-phase species of DMS, SO2, and NH3, 13 prognostic aerosol species including
sulfate (SO4^2-), 2 types of organic carbon (hydrophobic OC1, hydrophilic OC2), 2 types of
black carbon (hydrophobic BC1, hydrophilic BC2), 4 categories of soil dust (DST01, DST02,
DST03, DST04), and 4 categories of sea salt (SSLT01, SSLT02, SSLT03, SSLT04).
Concentrations of all aerosols in BCC-ESM1 are mainly determined by advective transport,
emission, dry deposition, gravitational settling, and wet scavenging by clouds and
precipitation, except for SO4^2- which gas-phase chemical reactions and aqueous phase
conversion from SO2 are also considered. The present version of aerosol schemes belongs to a
bulk aerosol aerosol type of model and mainly refers to the scheme of CAM-Chem
(Lamarque et al., 2012), but the nucleation and coagulation of aerosols are still ignored.
in the present version of BCC-AGCM3-Chem of BCC-ESM1.

2.1 SO2, DMS, NH3, and Sulfate
SO2 is a main sulfuric acid precursor to form aerosol sulfate SO4^2-. Conversions of SO2
to SO4^2- occur by gas phase reactions (Table 2) and by aqueous phase reactions in cloud
droplets. 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.
are the aerodynamic resistance and the surface canopy layer resistance, respectively. The loss rate of SO₂ due to wet deposition is computed following the scheme in the global Community Atmosphere Model (CAM) version 4, the atmospheric component of the Community Earth System Model (Lamarque et al., 2012).

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

SO₄²⁻ is one of the prognostic aerosols in BCC-AGCM3-Chem. Its treatment follows CAM4-Chem (Lamarque et al., 2012). It is produced primarily by the gas-phase oxidation of SO₂ (in Table 2) and by aqueous phase oxidation of SO₂ in cloud droplets. The gas phase reactions, rate constants, and gas-aqueous equilibrium constants are given by Tie et al. (2001). The heterogeneous reactions of SO₄²⁻ occur on all aerosol surfaces. Their treatment follows a Bulk Aerosol Model (BAM) used in CAM4 (Neale et al., 2010). The heterogeneous reactions depend strongly on pH values in clouds which are calculated from the concentrations of SO₂, HNO₃, H₂O₂, NH₃, O₃, HO₂, and SO₄²⁻. Only NH₃ is not a prognostic tracer in BCC-AGCM3-Chem and it is estimated using the assumption of an NH₃ to SO₄²⁻ molar ratio of 2.0. NH₃ is a gas tracer apart from MOZART2 (Table 1). Its sources include aircraft and surface emissions due to anthropogenic activity, biomass burning, and biogenic emissions from land soil and ocean surfaces (Table 4). SO₄²⁻ is assumed to be all in aqueous phase due to water uptake, although Wang et al. (2008a) showed that ~34% of sulfate particles are in solid phase globally due to the hysteresis effect of ammonium sulfate phase transition. However, in terms of radiative forcing, consideration of solid sulfate formation process lowers the sulfate forcing by ~8% as compared to consideration of all sulfate particles in aqueous phase (Wang et al., 2008b). Future model development may consider the life cycle of NH₃. The sulfate in- and below-cloud scavenging follows Neu and Prather (2011). Washout
of SO$_4^{2-}$ is set to 20% of the washout rate of HNO$_3$ following Tie et al. (2005) and Horowitz (2006). The dry deposition velocity of SO$_4^{2-}$ is also calculated by the resistance-in-series approach.

2.2 Aerosols of organic carbon and black carbon

BCC-AGCM3-Chem treats two types of organic carbon (OC), i.e. water-insoluble tracer OC1 and water-soluble tracer OC2, and two types of black carbon (BC), i.e. water-insoluble tracer BC1 and water-soluble tracer BC2. As shown in Table 2, hydrophobic BC1 and OC1 can be converted to hydrophilic BC2 and OC2 with a constant rate of 7.1×10$^{-6}$ s$^{-1}$ (Cooke and Wilson, 1996). The 4 tracers of organic carbon and black carbon are mainly from emissions of anthropogenic activities, including both fossil fuel and biomass burning, and are from the CMIP6 data package (https://esgf-node.llnl.gov/search/input4mips/, Hoesly et al., 2018).

Beside anthropogenic and biomass burning emissions, hydrophilic organic carbon OC2 can also come directly from natural biogenic volatile organic compound (VOC) emissions. They are calculated online in the land component model BCC-AVIM2 and assumed to equal to 10% of monoterpenes emission following the algorithm of Guenther et al. (1999).

Dry deposition velocities for all the 4 OC and BC tracers are set to 0.001 m s$^{-1}$. 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), and the transfer of soluble gases into liquid condensate is calculated with Henry’s Law assuming equilibrium between the gas and liquid phases.

2.3 Sea salt aerosols

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

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

(1)

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 for each size bins of sea salt aerosols in BCC-ESM1, respectively.

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

2.4 Dust aerosols

Dust aerosols behave in a similar way as sea salts. Their variations involve three major processes: emission, advective transport, and wet/dry depositions. The dust emission is based on a saltation-sandblasting process, and depends on wind friction velocity, soil moisture, and vegetation/snow cover (Zender et al., 2003). The vertical flux of dust emission is corrected by a surface erodible factor at each model grid cell which has been downloaded from NCAR website [1](https://ncar.ucar.edu/https://svn-ccsm-inputdata.cgd.ucar.edu/trunk/inputdata/atm/cam/dst/).

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

2.5 Effects of aerosols on radiation, clouds, and precipitation

The mass mixing ratios of bulk aerosols are prognostic variables in BCC-ESM1 and directly affect the shortwave radiative transfer in the atmosphere with their treatments following the NCAR Community Atmosphere Model (CAM3, Collins et al., 2004). Indirect effects of aerosols are taken into account in the present version of BCC-AGCM3-Chem (Wu et al., 2019). Aerosol particles act as cloud condensation nuclei and exert influence on cloud properties and precipitation, the hydrological cycle, and ultimately impact the hydrological cycle [2].

Prognostic aerosol masses are used to estimate the liquid cloud droplet number concentration \( N_{\text{cdne}} \) (cm\(^{-3}\)) in BCC-AGCM3-Chem. \( N_{\text{cdne}} \) is explicitly calculated using the empirical function suggested by Boucher and Lohmann (1995) and Quaas et al. (2006):
\[
N_{\text{cdnc}} = \exp\left[5.1 + 0.41\ln \left( m_{\text{aero}} \right)\right]
\]  \hspace{1cm} (2)

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

\[
m_{\text{aero}} = m_{\text{SS}} + m_{\text{OC}} + m_{\text{SO}} + m_{\text{NHNO}_3} \]
\hspace{1cm} (3)

i.e. the first bin of sea salt (m\(_{\text{SS}}\)), hydrophilic organic carbon (m\(_{\text{OC}}\)), sulphate (m\(_{\text{SO}}\)), and

NH\(_4\)NO\(_3\). A dataset of NH\(_4\)NO\(_3\) from NCAR CAM-Chem (Lamarque et al., 2012) is used in our model.

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

\[
r_{\text{el}} = \beta \cdot r_{\text{vol}}
\]
\hspace{1cm} (4)

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

\[
\beta = 0.00084 N_{\text{cdnc}} + 1.22
\]
\hspace{1cm} (5)

\( r_{\text{vol}} \) is the volume-weighted mean cloud droplet radius,

\[
r_{\text{vol}} = \left[ (3LWC) / (4\pi\rho_{\text{H}_{2}O} N_{\text{cdnc}}) \right]^{1/3}
\]
\hspace{1cm} (6)

where \( \rho_{\text{H}_{2}O} \) is the liquid water density and \( LWC \) the cloud liquid water content (g 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),

\[
PWAUT = C_{\text{1,aut}} \rho_{\text{H}_{2}O}^{2} / \rho_{\text{H}_{2}O} \left( q_{\text{i}} / \rho_{\text{H}_{2}O} N_{\text{cdnc}} \right)^{1/2} H \left( r_{\text{vol}} - r_{\text{el}} \right)
\]
\hspace{1cm} (7)
Where $\bar{\rho}_w$ is in-cloud liquid water mixing ratio, $\rho_a$ and $\rho_w$ are the local densities of air and water respectively, and $f_{out}$ is a constant. $H(x)$ is the Heaviside step function with the definition:

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

(8)

$R_{C,vol}$ is the critical value of mean volume radius of liquid cloud droplets $R_{vol}$ and set to 15$\mu$m.

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. As described for BCC-AGCM2 in Wu et al. (2019), it is parameterized as an empirical function of cloud water content and bulk aerosol mass concentration (Boucher and Lohmann, 1995; Quaas et al., 2006).

3. Experiment design for the 20th century climate simulation

There is an Aerosol Chemistry Model Intercomparison Project (AerChemMIP, Collins et al., 2017) endorsed by the Coupled-Model Intercomparison Project 6 (CMIP6) for documenting and understanding past and future changes in the chemical composition of the atmosphere, and estimating the global-to-regional climate response from these changes.

Modelling groups with full chemistry and aerosol models are encouraged to perform all AerChemMIP simulations—climate models that include an interactive representation of tropospheric aerosols and atmospheric chemistry (Collins et al., 2017). To assess the ability of our model to simulate aerosols (mean and variability), we have followed the historical simulation designed by CMIP6 (Eyring et al., 2016) which is named as “historical” experiment in the Earth System Grid Federation (ESGF). The AerChemMIP protocol details of the historical experiment forced with emissions evolving from 1850 to 2014 refer to Collins et al. (2017). $O_3$ in the historical simulation is an interactive prognostic variable and feedbacks on radiation, and the concentrations of other WMGHG, e.g. CH$_4$, N$_2$O, CO$_2$, CFC11, and CFC12 are prescribed using CMIP6 historical forcing data as suggestion in AerChemMIP protocol. Although CH$_4$ and N$_2$O are prognostic variables in the chemistry scheme (Table 1), their prognostic values at each model step in the historical experiment are...
replaced by CMIP6 data. The rest of Other-historical forcing data include: (1) monthly zonally-mean CFC11 and CFC12 concentrations, (2) yearly global gridded land-use forcing data sets, and (23) solar solar forcing. All these datasets were downloaded from https://esgf-node.llnl.gov/search/input4mips/. Climate feedback processes that involve changes to the atmospheric composition of reactive gases and aerosols may affect the temperature response to a given WMGHG concentration level (Collins et al., 2017). Three members of historical experiments are conducted and the first member is analyzed in this work. The AeroChemMIP historical case is a simulation with emissions evolving from 1850 to 2014 and with specified GHG concentrations of CH4, N2O, and CO2 following the protocols defined by CMIP6. Other-historical forcing data include: (1) monthly zonally-mean CFC11 and CFC12 concentrations, (2) yearly global gridded land-use forcing data sets, and (23) solar forcing. All these datasets were downloaded from https://esgf-node.llnl.gov/search/input4mips/. The principal GHGs for radiation calculation in BCC-AGCM-Chem1 include H2O, O3, CH4, N2O, CO2, CFC11 and CFC12. Only one GHG, O3, is a prognostic variable at each model step and interacts with radiations.

3.1 Surface emissions

Surface emissions of chemical species from different sources are summarized in Table 4. They include anthropogenic emissions from fossil fuel burning and other industrial activities, biomass burning (including vegetation fires, fuel wood and agricultural burning), biogenic emissions from vegetation and soils, and oceanic emissions. Most historical emissions from anthropogenic source (surface, aircraft plus ships) and biomass burning in the period of 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 the climatological dataset provided by the standard package of 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 (CH3CHO) and anthropogenic emission (as CH3CHO, CH3OH, and CH3COOH) are from the IPCC ACCMIP emission inventory (http://accent.aero.jussieu.fr/ACCMIP.php) covering the period from 1850 to 2010 with in
10-year time-intervals (see Table 4). Monthly CMIP6-recommended lumped emissions of black carbon and organic carbon aerosols are downloaded from CMIP6-recommended data, but we used assume 80% (for BC) and 50% (for OC) of them in their hydrophobic forms (BC1 and OC1) and the rest in their 20% hydrophilic forms (BC2 and OC2), following the work of Chin et al. (2002).

FiveSeven tracers of ISOP, ACET (CH$_3$COCH$_3$), C$_2$H$_4$, C$_3$H$_8$, and Monoterpenes (C$_{10}$H$_{16}$) in Table 1 belong to biogenic volatile organic carbons (VOCs), i.e. ISOP, ACET (CH$_3$COCH$_3$), C$_2$H$_4$, C$_3$H$_8$, and Monoterpenes (C$_{10}$H$_{16}$), and OC2. As shown in Table 4, those VOCs emissions are directly online calculated in BCC-ESM1 following the modeling framework of the Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1, Guenther et al., 2012) using simple mechanistic algorithms to account for major known processes controlling biogenic emissions. The MEGAN-MEGAN2.1 can provide a flexible scheme for estimating 16 tracers of biogenic emissions from terrestrial ecosystems including five VOCs emissions used in BCC-ESM1 (Table 4). All the VOCs emissions depend on current and past surface air temperature, and solar flux, and the landscape types, and their calculation requires global maps of plant functional type (PFT) and leaf area index (LAI) which is a prognostic variable from the land model BCC-AVIM2. The effect of atmospheric CO$_2$ concentration on isoprene emissions is included, 10% of the biogenic monoterpenes emissions as calculated online with the MEGAN2.1 algorithm in BCC-AVIM2 are converted to hydrophilic organic carbon (OC2) to account for formation of secondary organic aerosols following Chin et al. (2002) in this version of BCC-ESM1.

3.2 Volcanic eruption, lightning and aircraft emissions

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 SO$_2$ emissions from volcanic eruption on the variation of SO$_2$ in the atmosphere and then on the variation of tropospheric SO$_2$ concentration are considered, and the SO$_2$ emissions from 1850 to 2014 are downloaded from the IPCC ACCMIP emission inventory (http://accent.aero.jussieu.fr/ACCMIP.php). Emissions of stratospheric SO$_2$ from volcanic eruption from 1850 to 2014 are prescribed using the CMIP6 recommended data, although
recent studies based on satellite observation of SO$_2$ in 2006-2012 have revealed that this emission data may have a factor of 2-4 high bias in average (Ge et al., 2016). Aircraft emissions are provided for NO$_2$, CO, CH$_4$, NH$_3$, NO, and SO$_2$, and aerosols of OC and BC (Table 1). The emissions of NO from lightning are online calculated in BCC-AGCM3-Chem following the parameterization in MOZART2, and the globally-averaged mean during the period of 1850 to 2014 is 5.19 Tg(N) yr$^{-1}$, which is in agreement with observations within the range of 3 to 6 Tg(N) yr$^{-1}$ (Martin et al., 2002). The lightning frequency depends strongly on the convective cloud top height, and the ratio of cloud-to-cloud versus cloud-to-ground lightning depends on the cold cloud thickness from the level of 0$^\circ$C to the cloud top (Price and Rind, 1992).  

3.3 Upper boundary of the atmosphere  

As no stratospheric chemistry is included in the present version of BCC-AGCM3-Chem, it is necessary to ensure a proper distribution of chemically-active stratospheric species including O$_3$, CH$_4$, N$_2$O, NO, NO$_2$, HNO$_3$, CO, and N$_2$O$_5$. Concentrations of different tracer species (O$_3$, CH$_4$, N$_2$O, NO, NO$_2$, HNO$_3$, CO, and N$_2$O$_5$) 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 with a 10-day time scale down to the tropopause. Climatological values of NO, NO$_2$, HNO$_3$, CO and N$_2$O$_5$ at the top two layers model levels are extracted from the 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 traces (O$_3$, CH$_4$, and N$_2$O) at the top two model layers are the zonally-averaged and monthly values from 1850 to 2014 derived from the CMIP6 data package.  

3.4 The preindustrial model states  

The preindustrial state of BCC-ESM1 is obtained from a piControl simulation of over 600 years in which all forcings including emissions data are fixed at 1850 AD conditions. The initial state of the piControl simulation itself is obtained through individual spin-up runs of
each component of BCC-ESM1 in order for the piControl simulation to run stably and fast to reach its equilibrium. Figures 1(a-cb) show the time series of global yearly means of the net energy budget at top of the atmosphere (TOA), near-surface air temperature (TAS), and sea surface temperature (SST) the net energy budget at top of the model (TOM) from the piControl simulation for the last 450 years. It shows that the surface climate in BCC-ESM1 nearly reaches its equilibrium after 600 years piControl simulation. The whole system in BCC-ESM1 fluctuates around $+0.7 \text{ W m}^{-2}$ net energy flux at TOA without obvious trend in 450 years (Fig. 1ab). This level of TOA energy imbalance is close to the average imbalance ($1.0 \text{ W m}^{-2}$) among CMIP5 models (Wild et al., 2013), and do not cause remarkable climate drift in BCC-ESM1. The global mean surface air temperature TAS and SST keep around 288.1 K (Fig. 1b) and 295.05 K shows only a small warming (Fig. 1ca), respectively. During the last 450 years, there are ($\pm 0.2^\circ \text{K}$ amplitude of TAS and SST) oscillations of centennial scale for the whole globe (Figs. 1b and 1c), which are certainly caused by internal variation of the system.

Figures 2a-2c show the time series of global annual total burdens masses of SO2, DMS, and OH in the troposphere (integrated from the surface to 100 hPa) in the last 450 years of the piControl simulation. Without any anthropogenic source, the SO2 amount in the troposphere nearly keeps the level of varies around $0.0868 \pm 0.002$ Tg in the 450 years of the piControl simulation. Tropospheric DMS varies around the value of $0.116 \pm 0.002$ Tg. Tropospheric OH, as an important gas species oxidizing SO2 to form $\text{SO}_4^{2-}$ (Table 2), keeps at a stable level in the atmosphere. $\text{SO}_4^{2-}$ also remains at a stable level of $0.556 \pm 0.004$ Tg in the atmosphere in the whole period of the piControl simulation (Figure 2d). Without any anthropogenic source, the amounts of BC and OC in the troposphere vary around $0.0395 \pm 0.005$ Tg and $0.275 \pm 0.005$ Tg (Figures 2e-2f), respectively. Dust and sea salt aerosols are at the level of $22 \pm 1$ Tg and $11.7 \pm 1$ Tg (Figures 2g-2h), respectively. All those data are close to the global mean concentrations of 0.604 Tg $\text{SO}_4^{2-}$, 0.046 Tg BC, 0.30 Tg OC, 22.18 Tg dust, and 11.73 Tg sea salts in 1850 which are estimated based on the CMIP5 prescribed data in 1850 (Lamarque et al., 2010) recommended concentrations in year 1850 ($0.604 \text{ Tg } \text{SO}_4^{2-}$, $0.046 \text{ Tg } \text{BC}$, $0.30 \text{ Tg } \text{OC}$, $22.18 \text{ Tg } \text{dust}$, and $11.73 \text{ Tg } \text{sea salts}$).

Figure 3 shows the global spatial distributions of annual mean sulfate, organic carbon,
black carbon, dust, and sea salt aerosols in the whole atmospheric column averaged for the last 100 years of the piControl simulation of BCC-ESM. 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 were fewer anthropogenic/biomass SO_2 emissions, the SO_4^{2-} over land are evidently smaller than those over oceans, mainly distributed especially over the tropical Pacific and Atlantic Oceans, where DMS can be oxidized to SO_2 and then form SO_4^{2-}. There are several centers of high values of black carbon and organic carbon in East and South Asia, Europe, Southeast America, and in the tropical rain forests in Africa and South America. They mainly result from biomass burning including vegetation fires, fuel wood and agricultural burning. Dust aerosols are mainly distributed in North Africa, Central Asia, North China, and Australia, where arid and semi-arid areas locate. Dust emitted from Sahara Desert can be transported to the tropical Atlantic by easterly wind. The sea salt aerosols are mainly distributed over the mid-latitude Southern Oceans, the tropical southern Indian Ocean, and the tropical northern Pacific Ocean, where wind speeds near the sea surface are strong. As shown in Fig. 3, all the spatial distribution patterns of CMIP5-derived sulfate, black carbon, organic carbon, dust, and sea salt aerosols (Lamarque et al., 2010) are well simulated in BCC-ESM1. There are high spatial correlation coefficients, 0.76 for sulfate, 0.77 for black carbon, 0.77 for organic carbon, 0.94 for dust, and 0.94 for sea salts, between CMIP5 data and BCC-ESM1 simulations. 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.

4. Evaluation of O_3 and aerosols simulations in the 20th century

The rate of sulfate formation is dependent on the levels of oxidants in the troposphere. O_3 is an important oxidant. So, the evaluation of simulated tropospheric O_3 is helpful to understand the aerosols simulations. BCC-ESM1 is driven by most of the CMIP6-recommended emission data. As shown in Figure 4, the zonal distributions of the total
amounts of tropospheric O$_3$ below 300 hPa to the ground and their changes with time from 1850 to 2014 from the CMIP6-recommend dataset (Table 4) are well simulated by BCC-ESM1. Evident increasing trends since 1850 almost exist in every latitudes, especially in the Northern Hemisphere where the contents of tropospheric O$_3$ are higher than those in the Southern Hemisphere.

Figure 5 shows the vertical profiles of O$_3$ simulations with comparison to global ozonesonde observations averaged for the monthly data over 2010-2014 from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC: http://woudc.org/data.php, last access: 24 September 2019) in nine regions which are averaged from 41 global WOUDC sites. The details of WOUDC data may refer to Lu et al. (2019). As shown in Figure 5, BCC-ESM1 well captures the observed ozone vertical structure at all regions. At the lower and middle troposphere (i.e. below 6 km), the model typically shows positive bias within 5 ppbv for the Southern Hemisphere and 10 ppbv for the Northern mid-latitudes, similar to those simulated from many other global atmospheric chemical models (Young et al., 2013, 2018). The model has larger ozone overestimation in the upper troposphere and stratosphere at most regions, at least partly due to the use of prescribed stratospheric ozone as upper boundary conditions and/or errors in modeling ozone exchange between the stratosphere and the troposphere. Global tropospheric ozone burden derived from our simulation is 335 Tg averaged over 2010-2014, in consistent with recent assessment from multi chemistry models (Young et al., 2018).

4.1 Global aerosols trends

Figure 64(a)-(c) show the time series of global total emissions of SO$_2$, OC, and BC to the atmosphere from natural and anthropogenic sources. Emissions of SO$_2$ are largely due to industrial production. From 1850 to 1915, SO$_2$ emissions increased year by year as the Industrial Revolution intensified and expanded. But from 1915 to 1945, the increase trend of SO$_2$ emissions evidently became slower as broke out the First and the Second World Wars. After that period, with growing industrial productions, SO$_2$ emissions increased again and reached a maximum around the end of 1970s. During the 1980s and 2000s, with a substantial decrease of SO$_2$ emissions in Europe and the United States, the global SO$_2$ emissions has been decreasing since the 1980s despite the rapid increase of SO$_2$ emissions in South and East Asia.
as well as in developing countries in the Southern Hemisphere in recent years (Liu et al., 2009). The OC and BC emissions substantially increased since 1950s just after the Second World War. The global total OC emission in 2010 was nearly twice as much as that in pre-industrial (year 1850) and increased by 18 Tg yr\(^{-1}\). Anthropogenic black carbon emissions increased from 1 Tg yr\(^{-1}\) in 1850 to nearly 8 Tg yr\(^{-1}\) in 2010.

Anthropogenic SO\(_2\), OC and BC emissions strongly affect the variations of atmospheric concentrations of DMS, SO\(_2\), sulfate, OC, and BC. The global 0.5°x0.5° gridded data of CMIP5-recommended aerosols masses with 10-years interval from 1850 to 2000 (Lamarque et al., 2010) provides an important reference to evaluate the aerosol simulations in BCC-ESM1. As shown in Figure 75b-75f, the annual total aerosol burdens of SO\(_4^2-\), OC, and BC in the whole atmosphere column as simulated by the BCC-ESM1 20th century historical simulation are generally consistent with the values derived from CMIP5-recommended aerosols concentrations. Due to increasing SO\(_2\) emissions from 1850 to present day (Fig. 64), the global SO\(_2\) burden in the atmosphere increased from 100 Tg in 1850s to 200 Tg in 1980s (Fig. 75a), and has a high interannual-correlation coefficient of 0.996 with the anthropogenic emissions (Fig. 64a), as the lifetime of SO\(_2\) is short. The burden directly followed the emission. DMS in the atmosphere is oxidized by OH and NO\(_3\) to form SO\(_2\) (Table 2). Its natural emissions from oceans from 1850 to 2010 in the model are the climatological monthly means from MOZART2 NCAR data package. As shown in Fig 75a, 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 possibly results from the speeded rate of DMS oxidation with global warming, the prescribed emissions have not year-to-year variations and the loss of DMS oxidation to produce SO\(_2\) gradually exceeds the source (Table 5) as the rate of DMS oxidation reaction (Table 2) gets large along with global warming in the 20th century, 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.

Largely driven by SO\(_2\) anthropogenic emissions, the sulfate burden shows three different stages from 1850 to present. In the first period from 1850s to 1900s, the sulfate burden had a weak linear increase. It increased significantly in the second stage from 1910’s to 1940’s, and then exploded since 1950’s, until the middle 1970s and early 1980s. The sulfate burden
then remained nearly stable and even showed slightly decreases as seen from the CMIP5 data. As for the trends of 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 are similar to that of sulfate, but they showed continuous increases from 1950 to present.

The dust and sea salt aerosols in the atmosphere are largely determined by the atmospheric circulations and states of the land and ocean surface. We can see that the global dust burden in the atmosphere showed a large interannual variability and was slightly evident increase enhanced from 19850 to 2000, which could be partly caused by evident global warming since 1980 and increasing soil dryness resulting in more surface dust to be released in the atmosphere. Their details will be explored in the other paper.

4.2 Global aerosols budgets

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

The global DMS emission from the ocean is 27.4 Tg(S) yr⁻¹ in BCC-ESM. It is higher than the value reported in Liu et al (2005), largely due to stronger wind speed near the sea surface. This high emission in BCC-ESM is nearly balanced by the gas-phase oxidation of DMS to form SO₂. The DMS burden is 0.1206 Tg S with a lifetime of 0.78 days, which is within the range of other models reported in the literature. As shown in Table 5, the total SO₂ production averaged for the period of 1991 to 2000 is 76.93 Tg(S) yr⁻¹. A rate of 13.2 Tg(S) yr⁻¹ (about 17%) SO₂ is produced from the DMS oxidation, only 0.1 Tg(S) yr⁻¹ SO₂ from anthropogenic activities and volcanic eruption at surface. Here the emissions of SO₂ from volcanic eruption are not included. The amount of SO₂ produced from the DMS oxidation is in the range of other works (10.0 to 24.7 Tg(S) yr⁻¹) reported in Liu et al (2005). All the SO₂ production is balanced by SO₂ losses by dry and wet deposition, and by gas- and aqueous-phase oxidation. Half of its loss (38.74 Tg(S) yr⁻¹) occurs via its aqueous-phase oxidation.
oxidation to form sulfate. Other losses through dry and wet depositions and gas-phase oxidation to form \( \text{SO}_4^{2-} \) are also important (Table 2). All the sinks are in the range from the literature (Liu et al., 2005). The global burden of \( \text{SO}_2 \) in the atmosphere is 0.4824 Tg S with a lifetime of 1.12 days, consistent with values in literature (Liu et al., 2005).

Sulfate aerosol is mainly produced from aqueous-phase \( \text{SO}_2 \) oxidation (38.73 Tg S yr\(^{-1}\)) and partly from gaseous phase oxidation of \( \text{SO}_2 \) (10.32 Tg S yr\(^{-1}\)), and is largely lost by wet scavenging (49.06 Tg S yr\(^{-1}\)). The total \( \text{SO}_4^{2-} \) production in BCC-ESM is at the lower range of values in other models reported in Textor et al. (2006). Its global burden is 1.89 Tg S and the lifetime is 4.69 days, which are within the range of 1.71 to 2.43 Tg S and 3.72 to 5.4 days in the literatures (Textor et al., 2006; Liu et al., 2012; Liu et al., 2016; Liu et al., 2009; Matsui and Mahowald, 2017; Tegen et al., 2019; the value derived from CMIP5 data).

Sources of BC and OC are mainly from anthropogenic emissions. Based on the CMIP6 data, there are, on average, 7.22 Tg yr\(^{-1}\) BC and 13.9 Tg yr\(^{-1}\) OC from fossil and bio-fuel emissions and 18.3 Tg yr\(^{-1}\) OC from natural emission during the period of 1991 to 2000. Most of them are scavenged through convective and large-scale rainfall processes. The rest returns to the surface by dry deposition. The simulated BC and OC lifetimes are 6.6 and 5.0 days, respectively, and are close to the recent values of 5.0-7.5 days BC and 5.4-6.6 days OC in literatures (Textor et al., 2006; Tegen et al., 2019). The simulated BC and OC lifetimes are 6.6 and 5.0 days, respectively.

The emissions of dust and sea salt are mainly determined by winds near the surface. The annual total dust emission in BCC-ESM1 is 2592 Tg yr\(^{-1}\), higher than AeroCom multi-model mean (1840 Tg yr\(^{-1}\), Textor et al., 2006), but comparable to other studies (Chin et al., 2002; Liu et al., 2012; Emmons et al., 2010; Matsui and Mahowald, 2017). The average dust loading is 22.93 Tg, lower than the value of 35.9 Tg in Ginoux et al. (2001) but slightly higher than the value of 20.4 Tg derived from CMIP5 data. The average lifetime for dust particles is
3.23 days that is shorter than the AeroCom mean (4.14 days) and the value of 3.9 days in a recent study (Matsui and Mahowald, 2017). The simulated sea salt emission is 4667.2 Tg yr\(^{-1}\), slightly lower than the simulated value in Liu et al. (2012), and substantially lower than the AeroCom mean (16600 Tg yr\(^{-1}\), Textor et al., 2006). The simulated sea salt burdens are 11.89 Tg and close to the CMIP5 data. Their averaged lifetimes are 0.93 days and close to the value in the recent of Matsui and Mahowald (2017) but longer than the AeroCom mean (0.41 days, Textor et al., 2006).

4.3 Global aerosol distributions at present day

Figures 86-120 show December-January-February (DJF) and June-July-August (JJA) mean column mass concentrations of sulfate (SO\(_4^{2-}\), OC, BC, Dust, and Sea Salt aerosols averaged for the period of 1991-2000, respectively. Here, BCC-ESM1 simulated results are compared with the CMIP5-recommended data for the same period. Unlike the pre-industrial level of sulfate shown in Fig. 2, sulfate concentrations at present day (Fig. 86) are strongly influenced by anthropogenic emissions, and have maximum concentrations in the industrial regions (e.g., East Asia, Europe, and North America). Their seasonal variations are distinct and are characterized by high concentrations in boreal summer and low concentrations in boreal winter. These spatial distributions simulated by BCC-ESM1 are well consistent with the CMIP5 data, with spatial correlation coefficients in DJF and JJA reaching 0.92 and 0.83 (Figure 13), respectively. The deviation of the spatial pattern in BCC-ESM1 is less from the CMIP5 data in DJF but larger in JJA (Figure 13).

Unlike sulfate whose maximum concentrations are mainly distributed between 60\(^\circ\)N and the equator, peaking concentrations of BC and OC as shown in Figs. 92 and 108 are located near the tropics in the biomass burning regions (e.g., the maritime continent, Central Africa, South America), and their seasonal variations from DJF to JJA are evidently weaker than those of sulfate except in South America. In boreal summer, there are centers of high values in the industrial regions in the Northern Hemisphere mid-latitudes (i.e., East Asia, South Asia, Europe, and North America). These main features of spatial and seasonal variations in CMIP5 data are well captured by BCC-ESM1, and the BCC-ESM1 vs. CMIP5 spatial correlation coefficients (Figure 13) are 0.90 (OC in DJF), 0.91 (BC in DJF), 0.91 (OC
There are less deviations of spatial pattern for OC in DJF and JJA, but larger deviation for BC from CMIP5 data (Figure 13).

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

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

Figure 141 shows vertical profiles distributions of zonally-averaged annual mean concentrations of sulfate, organic carbon, black carbon, dust, and sea salt aerosols in the period of 1991-2000. Both BCC-ESM1 and CMIP5 results show that strong sulfur, OC, and BC emissions in the industrial regions of the Northern Hemisphere mid-latitudes can rise upward and be transported towards the North Pole in the middle to upper troposphere. Most of OC, BC, and dust aerosols are confined below 500 hPa, while sulfate can be transported to higher altitudes. Sea salt aerosols are mostly confined below 700 hPa, as the particles are large in size and favorable for wet removal and gravitational settling towards the surface. It can be seen that BCC-ESM1 tends to simulate less upward transport of aerosols than the
CMIP5 data, likely reflecting the omission of deep convection transport of tracers in
BCC-ESM1.

The CMIP5 data used here are mainly from model simulations. We will need to further
evaluate the BCC-ESM1 model results with ground observations. Annual mean SO$_4$$^{2-}$, BC and
OC aerosol concentration observations from the Interagency Monitoring of Protected Visual
Environments (IMPROVE) sites over 1990-2005 in the United States
(http://vista.cira.colostate.edu/IMPROVE/) and from the European Monitoring and Evaluation
Programme (EMEP) (http://www.emep.int) sites over 1995-2005 are used. As shown in
Figure 15a and 15b, the BCC-ESM simulated sulfate concentrations are in general
comparable to the EMEP observations in Europe, but are systematically by about 1 µg m$^{-3}$
higher than the U.S. IMPROVE observations. As for BC, there are large model biases at both
European and U.S. sites (Figs. 15c and 15d), especially BCC-ESM overestimates BC
concentrations at the IMPROVE sites. The observed simulated OC concentrations are slightly
overestimated for IMPROVE sites but systematically underestimated for EMEP sites. Some
statistical features for simulated concentrations versus EMEP and IMPROVE observations are
listed in Table 7. These comparisons are overall fairly reasonable considering the
uncertainties in emissions and the coarse model resolution.

We then evaluate the simulated BC concentrations from BCC-ESM1 with the HIAPER
(Pole-to-Pole Observations (HIPPO) (Wofsy et al., 2011). The HIPPO campaign provided
observations of black carbon concentration profiles over Pacific Ocean and North America
between 2009 and 2011. Following Tilmes et al. (2016), model results here are sampled along
the HIPPO flight tracks and then averaged to different latitude and altitude bands for
comparison. As shown in Figure 16, BCC-ESM1 and HIPPO aircraft observations shows
reasonable agreement in terms of the spatial distributions and seasonal variations of BC levels.
BCC-ESM1 generally reproduces the observed hemispheric gradients of BC, i.e. the larger
burden in the NH compared to the SH, in consistent with Figures 10 and 14. The model shows
large over estimations of BC observations over the tropics, which is also found in the
CAM4-chem global chemical model (Tilmes et al., 2016).
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 aerosol optical depth (AOD) at 550 nm for all aerosols including sulfate, BC, OC, 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 127 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-19d 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. Comparing 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. 17).
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 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
aerosols in the atmosphere are highly correlated with the CMIP5-recommended data. The
long-term trends of CMIP5 aerosols from 1850 to 2000 are also well simulated by
BCC-ESM1. Global budgets of aerosols were evaluated through comparisons of BCC-ESM1
results for 1990-2000 with reports in various literatures observational data at present day for
sulfate, BC, OC, sea salt, and dust. Their annual total emissions, global atmospheric mass
loading, and mean lifetimes are all within the range of values reported in relevant literatures.

Evaluations of the spatial and vertical distributions of BCC-ESM1 simulated present-day
sulfate (SO$_4^{2-}$), OC, BC, Dust, and Sea Salt aerosol concentrations against the CMIP5
datasets and in-situ measurements of surface networks (IMPROVE in the U.S. and EMEP in
Europe), and HIPPO aircraft observations indicate good agreement among them. The
BCC-ESM1 simulates weaker upward transport of aerosols from the surface to the middle and
upper troposphere (with reference to CMIP5-recommended data), likely reflecting a lack of
deep convection transport of chemical species in the present version of BCC-ESM1. The
aerosol optical depth (AOD) at 550 nm for all aerosols including sulfate, BC, OC, sea salt,
and dust aerosols was further compared with the satellite AOD observations retrieved from
MODIS and MISR and surface AOD observations from AERONET. The BCC-ESM1 model
results are overall in good agreement with these observations within a factor of 2. All these
comparisons demonstrate the success of the implementation of interactive aerosol and
atmospheric chemistry in BCC-ESM1.

This work has only evaluated the ability of BCC-ESM1 to simulate aerosols. The
variations of aerosols especially for sulfate are related to other gaseous tracers such as OH
and NO$_3$ (Table 2), which are determined by the MOZART2 gaseous chemical scheme as
implemented in BCC-ESM1, and require further evaluation. 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. O$_3$ is evaluated in this work. Other GHGs such as CH$_4$ and
N$_2$O concentrations can be simulated when forced with emissions and their simulations also
need to be evaluated in future. The variations of aerosols especially for sulfate are related to
other gaseous tracers such as OH and NO$_3$ (Table 2), which are determined by the MOZART2
gaseous chemical scheme as implemented in BCC-ESM1 and require further evaluation.
about the GHGs simulations in the AeroChemMIP historical run? Can the global warming be reproduced? These questions concerning feedbacks of prognostic aerosols on climate change especially, global warming also need to be explored in the future.

5.6. Code and data availability

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

Author contributions

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

Acknowledgements

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

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**Gas tracers**

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

<table>
<thead>
<tr>
<th>Chemical reactions</th>
<th>Rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH₃ + OH → H₂O</td>
<td>1.70E-12*exp(–710/T)</td>
</tr>
<tr>
<td>SO₂ + OH → SO₄²⁻</td>
<td>ko(1.+ko<em>M/ki)<em>f</em>(1./(1.+log10(ko</em>M/ki)), in which ko=3.0E-31*(300/T)**3.3; ki=1.E-12; f=0.6</td>
</tr>
<tr>
<td>DMS + OH → SO₂</td>
<td>9.60E-12*exp(-234./T)</td>
</tr>
<tr>
<td>DMS + OH → 5 SO₂ + 5 H₂O</td>
<td>1.7E-42<em>exp(7810/T)<em>M</em>0.21/(1+5.5E−31</em>exp(7460/T)* M* 0.21)</td>
</tr>
<tr>
<td>DMS + NO₃ → SO₂ + HNO₃</td>
<td>1.90E-13*exp 520/T</td>
</tr>
<tr>
<td>BC₁ → BC₂</td>
<td>7.10E-06</td>
</tr>
<tr>
<td>OC₁ → OC₂</td>
<td>7.10E-06</td>
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</table>
Table 3. Size and density parameters of bulk aerosols.

<table>
<thead>
<tr>
<th>Aerosols</th>
<th>Species Name</th>
<th>Mean radius (µm) / bin size (µm)</th>
<th>Geometric standard deviation (µm)</th>
<th>Density (g cm(^{-3}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>SO(_{4}^{2-})</td>
<td>Sulfate</td>
<td>0.05</td>
<td>2.03</td>
<td>1.77</td>
</tr>
<tr>
<td>BC1</td>
<td>hydrophobic black carbon</td>
<td>0.02</td>
<td>2.00</td>
<td>1.0</td>
</tr>
<tr>
<td>BC2</td>
<td>hydrophilic black carbon</td>
<td>0.02</td>
<td>2.00</td>
<td>1.0</td>
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<tr>
<td>OC1</td>
<td>hydrophobic organic carbon</td>
<td>0.03</td>
<td>2.24</td>
<td>1.8</td>
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<tr>
<td>OC2</td>
<td>hydrophilic organic carbon</td>
<td>0.03</td>
<td>2.24</td>
<td>1.8</td>
</tr>
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<td>DST01</td>
<td>Dust</td>
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<td>DST02</td>
<td>Dust</td>
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<td>DST03</td>
<td>Dust</td>
<td>3.75 / bin: 2.5-5.0</td>
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<td>DST04</td>
<td>Dust</td>
<td>7.50 / bin: 5.0-10.</td>
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<td>2.5</td>
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<td>15.14 / bin: 10. -20.</td>
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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.

<table>
<thead>
<tr>
<th>Species</th>
<th>Anthropogenic emission</th>
<th>Biomass burning</th>
<th>Biogenic emissions from vegetation</th>
<th>Biogenic emissions from soil</th>
<th>Oceanic emissions</th>
<th>Airplane emission</th>
<th>Volcanic emission</th>
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</table>

**Anthropogenic**

- Industrial and fossil fuel use
- Agriculture
- Ships

**Biomass burning**

- Vegetation fires
- Fuel wood
- Agricultural burning

**Biogenic emissions**

- From vegetation
- From soil
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.

<table>
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<th>BCC-ESM (1991-2000 mean)</th>
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<td>Sources</td>
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<tr>
<td>Emission</td>
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<td>10.7-23.7³</td>
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<tr>
<td>Sinks</td>
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<tr>
<td>Gas-phase oxidation</td>
<td>28.0</td>
<td>10.7-23.7³</td>
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<tr>
<td>Burden</td>
<td>0.1206</td>
<td>0.042-0.2915²</td>
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<tr>
<td>Lifetime</td>
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<tr>
<td>Sources</td>
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<tr>
<td>Emission at surface</td>
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<td>Emission from airplane</td>
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<td>Sinks</td>
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<td>16.0-55.0³</td>
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<td>0.0-19.9⁴</td>
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<tr>
<td>Gas-phase oxidation</td>
<td>10.33</td>
<td>6.1-16.8⁴</td>
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<td>Aqueous-phase oxidation</td>
<td>38.74</td>
<td>24.5-57.8³</td>
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<td>0.4824</td>
<td>0.4020-1.220.61²</td>
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<td>Lifetime</td>
<td>1.12</td>
<td>0.6-2.6º</td>
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<td><strong>SO₄²⁻</strong></td>
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<tr>
<td>Sources</td>
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<td>59.67 ± 13.13¹b</td>
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<tr>
<td>Emission</td>
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<td>SO₂ aqueous-phase oxidation</td>
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<tr>
<td>SO₂ gas-phase oxidation</td>
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<tr>
<td>Sinks</td>
<td>49.06</td>
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</tr>
<tr>
<td>Dry deposition</td>
<td>2.20</td>
<td>4.96-5.51¹d</td>
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<tr>
<td>Wet deposition</td>
<td>46.86</td>
<td>39.34-40.20²d</td>
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<tr>
<td>Burden</td>
<td>1.890.63</td>
<td>1.890.63 ± 0.4846², 1.710.62², 0.64¹, 1.2², 2.22-2.43³</td>
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<tr>
<td>Lifetime</td>
<td>4.69</td>
<td>4.12 ± 0.74², 3.72-3.77³</td>
</tr>
</tbody>
</table>

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 Tg S to Tg (species) for units consistence.
Table 6. Same as Table 5, but for global budgets for black carbon, organic carbon, dust, and sea salts. Units are sources and sinks, Tg yr$^{-1}$; burden, Tg; lifetime, days.

<table>
<thead>
<tr>
<th></th>
<th>BCC-ESM (1991-2000 mean)</th>
<th>Other studies and CMIP5 data</th>
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<tbody>
<tr>
<td><strong>BC</strong></td>
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</tr>
<tr>
<td></td>
<td><strong>Sources</strong></td>
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</tr>
<tr>
<td>Emission</td>
<td>7.22</td>
<td>11.9±2.8, 7.8</td>
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<tr>
<td>Sinks</td>
<td>7.24</td>
<td>7.7±2.8</td>
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<tr>
<td>Dry deposition</td>
<td>0.90</td>
<td>0.27±1.30, 1.64</td>
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<td>Wet deposition</td>
<td>6.34</td>
<td>7.5±6.10-6.45</td>
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<tr>
<td>Burden</td>
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<td>0.114±0.24±0.1</td>
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<td><strong>OC</strong></td>
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</tr>
<tr>
<td></td>
<td><strong>Sources</strong></td>
<td></td>
</tr>
<tr>
<td>Fossil and bio-fuel emission</td>
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<td>Natural emission</td>
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<td>Wet deposition</td>
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<td>Burden</td>
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<td>0.69±1.70±0.45±1.0-2.2</td>
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<td>Lifetime</td>
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<td>6.5±1.76±4.56-4.90, 5.4-6.6, 6.5-1.76</td>
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<tr>
<td><strong>Dust</strong></td>
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<td>20.41, 22.42±2.4, 35.9±4.5</td>
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<tr>
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<td>0.41±0.24, 0.55±0.76, 0.98±1.2, 1.3</td>
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</table>
Notes: References denote a for Liu et al. (2005); b for Textor et al. (2006); c derived from CMIP5 prescribed aerosol masses averaged from 1991 to 2000; d for Liu et al. (2012); e for Liu et al. (2016); f for Ginoux et al. (2001); g for Matsui and Mahowald (2017); and h for Tegen et al. (2019).
Figure 1. The time series of global and annual mean of (a) net energy budget at top of atmosphere (W m\(^{-2}\)), surface air temperature (K) and (b) net energy budget at top of the model (W m\(^{-2}\)) near-surface air temperature (K), and (c) sea surface temperature (K) in the last 450 years of the piControl simulation.
Figure 2. Same as in Figure 1, but for the global burdens of (a) SO$_2$, (b) DMS, (c) OH, and (d-h) different aerosols (in unit of Tg) in the troposphere (below 100 hPa). Units are Tg.
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/RepDb/). Units: mg m$^{-2}$. 
Figure 4. Zonal mean of yearly mean concentration of ozone column in the troposphere below 300 hPa to the ground from 1871 to 1999 for (a) BCC-ESM1 and (b) CMIP6 data. Unit: DU.
Figure 5. Vertical profiles of annual mean ozone concentrations from observations averaged for 2010-2014 in nine regions (black) and from the BCC-ESM1 simulations (red). The observations are derived from 41 global WOUDC sites.
Figure 64. Global annual anthropogenic, natural, and total emissions of SO$_2$, 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$^{-1}$. 

(a) SO$_2$ emission

(b) CC emission

(c) BC emission
Figure 25. The time series of global yearly amounts of (a) SO$_2$ and DMS and (b-f) aerosols in the whole atmosphere column from the CMIP6 historical simulations of BCC-ESM1 (black lines) and the CMIP5-recommended aerosols masses (red lines). The yearly CMIP5 data are interpolated from the time series in 10-year interval. Units: Tg.
Figure 86. December-January-February (DJF; top panels) and June-July-August (JJA; bottom panels) mean sulfate (SO$_2^-$) 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: mg m$^{-2}$. 
Figure 92. The same as in Figure 86, but for organic carbon (OC) aerosol column mass concentrations. Units: mg m\(^{-2}\).
Figure 108. The same as in Figure 86, but for black carbon (BC) aerosol. Units: mg·m$^{-2}$. 
Figure 119. The same as Figure 84, but for dust aerosol. Units: mg.m$^{-2}$.
Figure 120. The same as in Figure 84, but for sea salt (SSLT) aerosol. Units: mg m$^{-2}$. 
Figure 13. Taylor diagram for the global aerosols climatology (1971–2000) of sulfate, organic carbon, black carbon, dust, and sea salt averaged for December-January-February (DJF), June-July-August (JJA), and annual respectively. The radial coordinate shows the standard deviation of the spatial pattern, normalized by the observed standard deviation. The azimuthal variable shows the correlation of the modelled spatial pattern with the observed spatial pattern. Analysis is for the whole globe. The reference dataset is CMIP5-prescribed dataset.
Figure 141. 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: µg m⁻³.
Figure 152. Scatter plots showing observed versus simulated \textit{multi-years averaged} annual mean sulfate (SO$_4^{2-}$), organic carbon (OC), black carbon (BC) mixing ratios at IMPROVE and EMEP network sites. Observations are averages over the available years 1990–2005 for IMPROVE sites, and 1995–2005 for EMEP sites. Simulated values are those at the lowest layer of BCC-ESM1.
Table 7. Observed versus simulated concentrations of sulfate (SO$_4^{2-}$), organic carbon (OC), black carbon (BC) for the regional mean and spatial standard deviation, minimum and maximum values at 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.

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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.
Figure 173. Global distribution of annual mean AOD simulated in BCC-ESM1 compared with the MISR and MODIS data for the year 2008.
Figure 18. Observed versus simulated annual means of AOD at AERONET sites. Each data point represents the mean averaged for available monthly values of AOD. The dot sizes denote the magnitudes of AOD at sites. The spatial correlation is 0.56.
Figure 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.
定义网格后不调整右缩进，相同样式的段落间不添加空格，无孤行控制，不调整西文与中文之间的空格，不调整中文和数字之间的空格，制表位：不在 3.43 字符。