

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

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

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

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

*We apologize for the confusion. TOM (top of model) should be TOA (top of atmosphere). We double checked our data used and there was indeed an imbalance of net energy flux at TOA. In order to verify whether there is a drift in ocean temperatures, Fig.1c representing the variation of global SST is added. It seems that the ocean is stable, at least for its upper layer. If we refer to other models of similar complexity, it seems that a small imbalance commonly exists (Hansen et al., 2005; Wild et al., 2013) and an average of  $1.0\text{Wm}^{-2}$  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 ( $1.0\text{Wm}^{-2}$ ) 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  $288.1\text{K}$  (Fig. 1b) and  $295.05\text{K}$  (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.*

3. Line 207: reference Hoesly et al.

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

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

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

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

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

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

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

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

*“Most historical emissions from anthropogenic source (surface, aircraft plus ship) and biomass burning from 1850 to 2014 are CMIP6-recommended data (Hoesly et al., 2018; available at <https://esgf-node.llnl.gov/search/input4mips>). Anthropogenic or biomass burning sources of some tracers which are not included in the CMIP6 dataset (see Table 4), anthropogenic emission of H<sub>2</sub> and N<sub>2</sub>O are from monthly climatological dataset provided by the MOZART-2 standard package. N<sub>2</sub>O is a prognostic variable in BCC-ESM1 but it is replaced by CMIP6 prescribed concentration in the historical run. Other emissions including biomass burning (CH<sub>3</sub>COCH<sub>3</sub>) and anthropogenic emission (CH<sub>3</sub>CHO, CH<sub>3</sub>OH, and CH<sub>3</sub>COCH<sub>3</sub>) are from the 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 H<sub>2</sub> and N<sub>2</sub>O 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 SO<sub>2</sub> emissions from volcanic eruption on the variation of SO<sub>2</sub> in the atmosphere and then on the variation of tropospheric SO<sub>2</sub>- concentration are considered, and the SO<sub>2</sub> emissions from 1850 to 2014 are downloaded from the IPCC ACCMIP emission inventory (<http://accent.aero.jussieu.fr/ACCMIP.php>).”*

9. Line 293: what are the total NO<sub>x</sub> emissions from lightning (in TgN/year)?

*The globally-averaged mean of the total NO<sub>x</sub> emissions from lightning during the period of 1850 to 2014 is 5.19 Tg (N) yr<sup>-1</sup>. It is in agreement with observations within the range of 3 to*

*6 Tg(N) yr<sup>-1</sup> (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 (O<sub>3</sub>, CH<sub>4</sub>, N<sub>2</sub>O, NO, NO<sub>2</sub>, HNO<sub>3</sub>, CO, and N<sub>2</sub>O<sub>5</sub>) at the top 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, NO<sub>2</sub>, HNO<sub>3</sub>, CO and N<sub>2</sub>O<sub>5</sub> 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 (O<sub>3</sub>, CH<sub>4</sub>, and N<sub>2</sub>O) 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 SO<sub>2</sub> 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 SO<sub>2</sub> emissions, the SO<sub>4</sub> over land are evidently smaller than those over oceans especially over the tropical Pacific and Atlantic Oceans, where DMS can be oxidized to SO<sub>2</sub> and then form SO<sub>4</sub>”*

12. Line 373: the correlation really only reflects that the lifetime of SO<sub>2</sub> 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 SO<sub>2</sub> emissions from 1850 to present day (Fig. 6), the global SO<sub>2</sub> 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 SO<sub>2</sub> 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).*