

**Reviewer #2:**

*[Comment#2-1] This paper describes and evaluates a new Earth System Model, obtained by combining the GEOS-Chem Chemistry-Transport Model (CTM) with the Beijing Climate Centre Earth System Model (BCCESM). The evaluation comprises tropospheric composition, with a focus on the ozone burden and budget, the impacts of Aerosol-Radiation-Interactions and Aerosol-Cloud-Interactions on radiative fluxes and cloud properties, and a high-resolution simulation of pollution above Asia. Since earlier versions of BCC-ESM participated in CMIP5 and in the CMIP6 Aerosol Chemistry Model Intercomparison Project (AerChemMIP; Zhang et al., 2021), we can expect this new version to contribute to future CMIPs as well. It is thus important to document and evaluate this version properly, preferably by publication in Geoscientific Model Development. Yet the submitted manuscript requires major improvements.*

*[Response#2-1] Thank you very much for your comments. We have revised accordingly. All of them have been implemented in the revised manuscript. Please see our itemized responses below.*

*[Comment#2-2 General Comments] The modelling of stratospheric transport and chemistry can be understood only from two unclear sentences in the description of the setup of model experiments (section 3.1), and seems fundamentally flawed:*

*“We note that as BCC-GEOS-Chem only extends to the height of 2.914 hPa, ozone concentrations at the top two layers are set to prescribed monthly climatological values from CMIP6 data package as upper boundary conditions. [The earlier version] BCC-AGCM-Chem conducted the same treatment but included additional stratospheric species (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>), and [in the current version(?)] their concentrations from below the top two layers to the tropopause are relaxed at a relaxation time of 10 d towards the climatology.”*

*One should first note that this approach precludes any true interactivity between climate change and stratospheric chemistry. This aspect is barely more advanced in BCC-GEOS-Chem v2.0 than in v1.0 and seems actually less developed than in the first version BCC-AGCM-Chem.*

*As I understand it, the 7 listed species are actually included in the current version, but their distributions are not realistic enough to model stratospheric ozone in a usable manner, and this was partly mitigated by the described relaxation to some (undocumented) climatology. I believe that the model top is too low to allow a proper Brewer-Dobson Circulation, hindering a correct stratospheric composition and explaining the (undocumented) statement that “BCC-GEOS-Chem v2.0 shows high ozone bias in the middle stratosphere” (line 575). This would of course explain the very large closure term in the tropospheric ozone budget, i.e. a stratospheric influx residual as large as 977 Tg/year.*

*In my view the authors have a choice between*

- solving this issue by raising the model top, re-running all the experiments, and re-*

*writing all sections related with stratospheric and tropospheric ozone; or...*

- *accepting this flaw, documenting it in a complete and straightforward manner.*

*If the model top is raised, I would recommend using the same 72 layers and top at 0.01 hPa as in the high-resolution configuration described in section 6. If not, the high-resolution configuration provides a means to evaluate the impact of the low top in the low-resolution configuration: see SC7 for more ideas on this route. Note that the vertical grid should be documented in the Supplementary material. This high-top vertical grid seems similar to MERRA-2, which has 72 staggered hybrid-pressure levels, i.e. 73 layers. All modern GCMs use staggered vertical grids. Is this also the case for BCC?*

*In any case, the model evaluation will have to include the vertical distribution of ozone in the stratosphere. Considering the absence of interactive stratospheric chemistry in BCC-GEOS-Chem v1.0 and the “critical role of stratospheric chemistry in modulating global atmospheric dynamics” (lines 108-109), the distribution of ozone in the middle atmosphere must be thoroughly documented by quantitative comparisons with observations or at least a climatology of observations. The evaluation of Total Ozone Columns (Fig. S4) is severely lacking as it is purely qualitative, does not include any observational reference and provides no information on the vertical distribution. These comparisons will also provide a basis to report on future progress with the current overestimation of stratospheric ozone influx (“STE”, section 4.3).*

*Please note that the sections “Specific comments” and “Language improvements” end in section 3.1. This reviewer felt that the text requires too many revisions to warrant such a detailed review of the next sections.*

[Response#2-2] Thank you for pointing it out. We agreed that the modelling of stratospheric chemistry should be clarified in a more explicit way, and the performance of stratospheric chemistry should be carefully evaluated and discussed. As [Comments #2-9, 2-12, 2-13] are closely related to this issue, we organize the responses here.

First, we need to clarify that the capability to simulate interactive stratospheric chemistry in BCC-GEOS-Chem v2.0 is new compared to BCC-GEOS-Chem v1.0 and BCC-AGCM-Chem. We note that there is no stratospheric chemistry in BCC-ACGM-Chem. In BCC-ACGM-Chem, stratospheric chemical species ( $O_3$ ,  $CH_4$ ,  $N_2O$ ,  $NO$ ,  $NO_2$ ,  $HNO_3$ ,  $CO$ , and  $N_2O_5$ ) at the top two layers (2.914 and 4.895 hPa) are set to prescribed monthly climatological values from CMIP6 data package as upper boundary conditions, and concentrations from below the top two layers to the tropopause are relaxed with a relaxation time of 10 d towards the climatological values. BCC-GEOS-Chem v1.0 includes GEOS-Chem v11-02b troposphere-only chemical module from the GEOS-Chem v11-02b, but does not include interactive stratospheric chemistry despite using a simple linearized ozone parameterization

(LINOZ) (McLinden et al., 2000) calculation of stratospheric ozone (Lu et al., 2020). BCC-GEOS-Chem v2.0 implements the standard chemical mechanism in GEOS-Chem v14.0.1, which features a comprehensive O<sub>x</sub>-NO<sub>x</sub>-VOC-halogen-aerosol chemistry in the unified tropospheric–stratospheric chemistry extension (UCX) scheme (Eastham et al., 2014). Unlike BCC-AGCM-Chem, the BCC-GEOS-Chem v2.0 model eliminates the need for stratospheric chemical species relaxation to prescribed fields, since these species can now be calculated directly by the UCX scheme. However, consistent with the treatment in BCC-AGCM-Chem, it still requires upper boundary conditions applied to the top two layers. Besides, BCC-GEOS-Chem v2.0 further includes additional chemical species (e.g. halogens, stratospheric aerosols), which are also simulated interactively. Therefore, BCC-GEOS-Chem v2.0 demonstrates a certain degree of advancement in stratospheric chemistry compared to BCC-GEOS-Chem v1.0 and BCC-AGCM-Chem, and we have modified the following statement to clarify the modelling of stratospheric chemistry:

“The capability to simulate interactive stratospheric chemistry in BCC-GEOS-Chem v2.0 represents a major improvement compared to BCC-GEOS-Chem v1.0 and BCC-AGCM-Chem. BCC-AGCM-Chem does not include stratospheric chemistry, all chemical species in the stratosphere are relaxed to monthly climatological values from CMIP6 data package (Wu et al., 2020). BCC-GEOS-Chem v1.0 incorporates troposphere-only chemical schemes from GEOS-Chem v11-02b. However, it still lacks interactive stratospheric chemistry despite including a simple linearized ozone parameterization (LINOZ) (McLinden et al., 2000) (Lu et al., 2020). Interactive stratospheric chemistry is now available in BCC-GEOS-Chem v2.0, which includes heterogeneous halogen chemistry and the formation, sedimentation and evaporation of polar stratospheric clouds (PSCs) as well as background liquid binary sulfate (LBS) aerosols, thus enabling capture of stratospheric responses and troposphere-stratosphere coupling.” (Section 2.3.2)

“We note that since BCC-AGCM-Chem has no interactive stratospheric chemistry, stratospheric species (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>) concentrations at the top two layers are set to prescribed monthly climatological values from CMIP6 data package as upper boundary conditions, and their concentrations from below the top two layers to the tropopause are relaxed with a relaxation time of 10 d towards the climatological values (Wu et al., 2020). BCC-GEOS-Chem v2.0 model eliminates the need for the relaxation of stratospheric chemical species to prescribed fields, since these species can now be calculated interactively using the UCX chemical scheme. Upper boundary conditions of O<sub>3</sub> and NO<sub>x</sub> are nevertheless still applied to the top two model layers.”

Additional bibliographical references:

McLinden, C. A., Olsen, S. C., Hannegan, B., Wild, O., Prather, M. J., and

Sundet, J.: Stratospheric ozone in 3-D models: A simple chemistry and the cross-tropopause flux, *J. Geophys. Res.*, 105, 14653–14665, <https://doi.org/10.1029/2000jd900124>, 2000.

Second, we enhanced the evaluation of stratospheric chemistry and circulation and discussed the model’s capabilities and limitations explicitly. The vertical discretization in BCC-ESM follows a hybrid sigma-pressure system: upper layers use pressure coordinates, while lower levels transition to sigma coordinates that are purely terrain-following at the surface. We have added this information in the text and documented the reference pressure of each vertical grid under T42L26 and T159L72 resolution in the Supplementary material (Table S5).

We evaluated simulated ozone and other key stratospheric components with the MLS satellite observations and climatological values from the CMIP6 data package. We also evaluate the strength of Brewer-Dobson Circulation through the vertical component of residual circulation with the ERA5 reanalysis data. We acknowledged that the model top is too low to allow a proper representation of Brewer-Dobson Circulation, which may limit the ability to reproduce the spatial distribution of stratospheric compositions. These discussions are shown in Section 4.1:

“However, both BCC-GEOS-Chem v2.0 and v1.0 tend to underestimate ozone levels in the Antarctic UTLS, while BCC-AGCM-Chem shows better agreement with observations as its chemical species are set/relaxed to prescribed monthly climatological values as it does not directly simulate stratospheric chemistry.

Figure 4 compares the zonal mean stratospheric ozone distributions simulated by BCC-GEOS-Chem v2.0 and v1.0 with the MLS satellite observations. Overall, with the incorporation of the UCX scheme for interactive stratospheric chemistry, BCC-GEOS-Chem v2.0 captures the vertical ozone distribution of stratospheric ozone and replicates the ozone maximum between 30-5 hPa seen in the MLS observation. In contrast, BCC-GEOS-Chem v1.0 with a simplified linearized ozone parameterization simulates an anomalously broad ozone maximum and substantially underestimates its magnitude at 30-5 hPa compared to MLS. Furthermore, BCC-GEOS-Chem v2.0 better reproduces Antarctic stratospheric ozone depletion during austral spring than v1.0 (Figure S5), although the depletion remains less pronounced than observed. However, we also note that substantial biases persist in BCC-GEOS-Chem v2.0. We find an underestimation of 5-20% in upper-stratospheric ozone (30-4 hPa) relative to MLS, accompanied by a pronounced cold bias in upper-stratospheric temperature compared to ERA5 reanalysis data. Besides, BCC-GEOS-Chem v2.0 overestimates lower-to-middle stratospheric ozone (100-50 hPa) by over 50% compared to MLS, leading to an overestimation of the total column ozone (Figure S5c). In addition, while the model reproduces the

overall vertical distribution of key stratospheric chemical component (e.g.  $\text{NO}_x$ ,  $\text{NO}_y$ , methane), there are notable discrepancies in both the magnitude and location of concentration hotspots (Figure S6).

These biases may be partly attributable to the model's limited top height in the T42L26 configuration. This constrains its ability to accurately simulate stratospheric circulations, and consequently, to reproduce the distributions and magnitudes of stratospheric chemical components that are significantly modulated by intense climate-chemistry interactions within the stratosphere (such as the interaction between stratospheric ozone and temperature). We find that although BCC-GEOS-Chem v2.0 captures the overall structure of the residual circulation (Figure S7), it underestimates the strength of the Brewer–Dobson circulation in the upper stratosphere over both the equatorial regions and high latitudes. This weaker tropical upwelling may partly account for high biases in lower-stratospheric ozone over the tropics.”

Third, we have also compared the vertical distribution of stratospheric ozone between T159L72 and T42L26 configuration (Figure S8), with special focus on the upper stratosphere. Comparison of the vertical distribution of stratospheric ozone between T159L72 and T42L26 configuration shows that the main features of stratospheric ozone simulated by the two configurations are robust, but T159L72 configuration indeed largely reduces the high ozone bias in the lower to middle stratosphere in the T42L26 configuration. We have added the following discussion: “Our one-month high-resolution simulation using the T159L72 configuration indeed largely reduced the high ozone bias in the lower to middle stratosphere (Figure S8), highlighting the importance of an adequate model top and sufficient vertical resolution for simulating stratospheric ozone.” However, we are currently experiencing some technical difficulties in extending the model vertical levels to 72 layers under T42 resolution. We hope to achieve this capability in the next round of model development.

In summary, BCC-GEOS-Chem v2.0 incorporates an interactive stratospheric chemistry scheme, enabling it to simulate reasonable vertical distributions of key stratospheric constituents. This represents a significant advance over its predecessor, BCC-GEOS-Chem v1.0, and BCC-AGCM-Chem. However, our evaluation indicates that substantial performance improvements are necessary and can be achieved by increasing the model top height. The current configuration also limits a complete representation of stratospheric climate-chemistry interactions. While BCC-GEOS-Chem v2.0 primarily targets improved tropospheric chemistry, resolving these stratospheric limitations is critical and should be prioritized for the model's next generation. We have revised the following statement in the Conclusion section: “In summary, the development of the BCC-GEOS-Chem v2.0 model provides a useful framework for investigating climate–chemistry

interactions and for future projections of global atmospheric chemistry.”

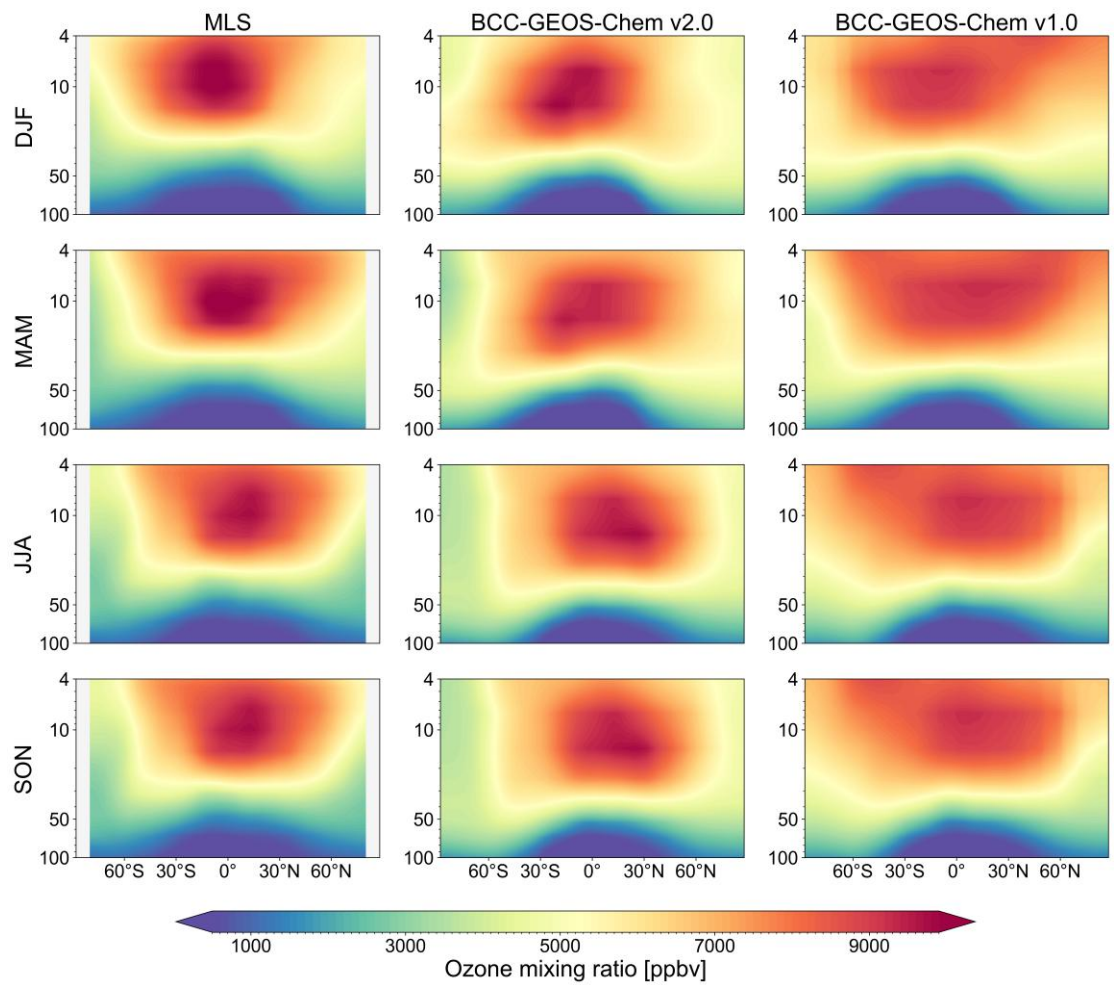


Figure 4. The zonal mean vertical and seasonal distributions of ozone in the stratosphere from BCC-GEOS-Chem v2.0 and BCC-GEOS-Chem v1.0 under T42L26 configuration and MLS. Values are averaged over 2012-2014.

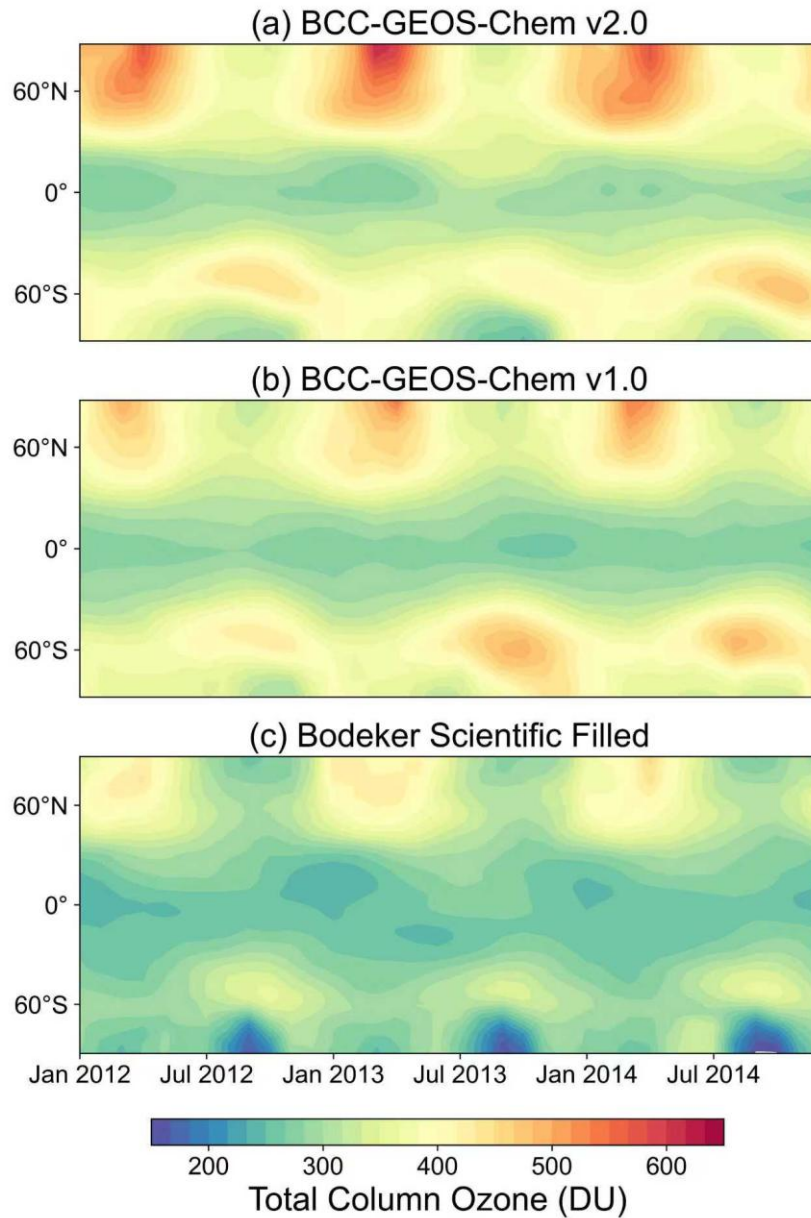


Figure S5. Zonal-mean total column ozone over 2012-2014 from (a) BCC-GEOS-Chem v2.0, (b) BCC-GEOS-Chem v1.0, and (c) Bodeker Scientific Filled data.

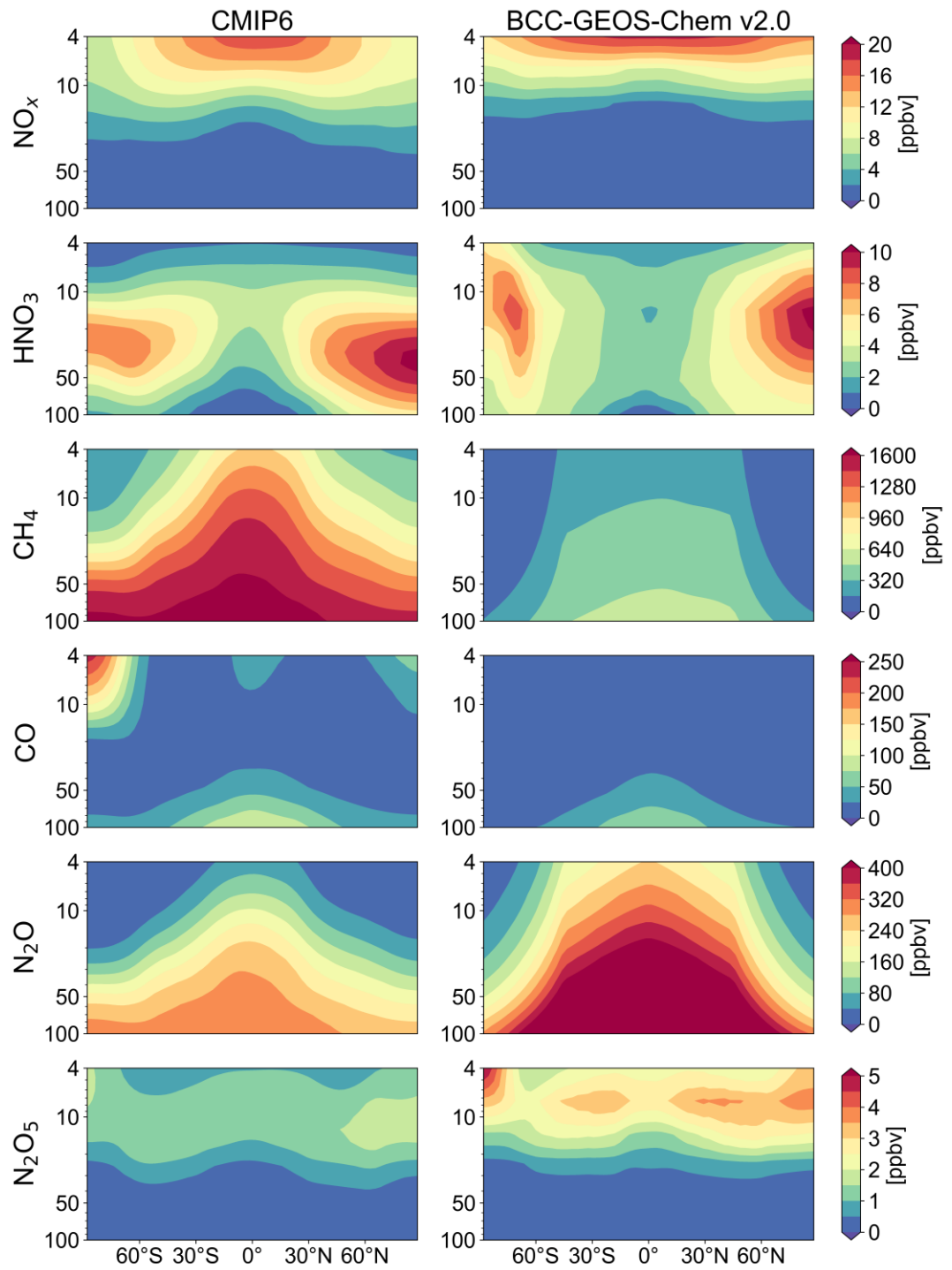


Figure S6. Zonal mean vertical distributions of  $\text{NO}_x$ ,  $\text{HNO}_3$ ,  $\text{CH}_4$ ,  $\text{CO}$ ,  $\text{N}_2\text{O}$ ,  $\text{N}_2\text{O}_5$  in the stratosphere from CMIP6 climatological field and BCC-GEOS-Chem v2.0 under T42L26 configuration.

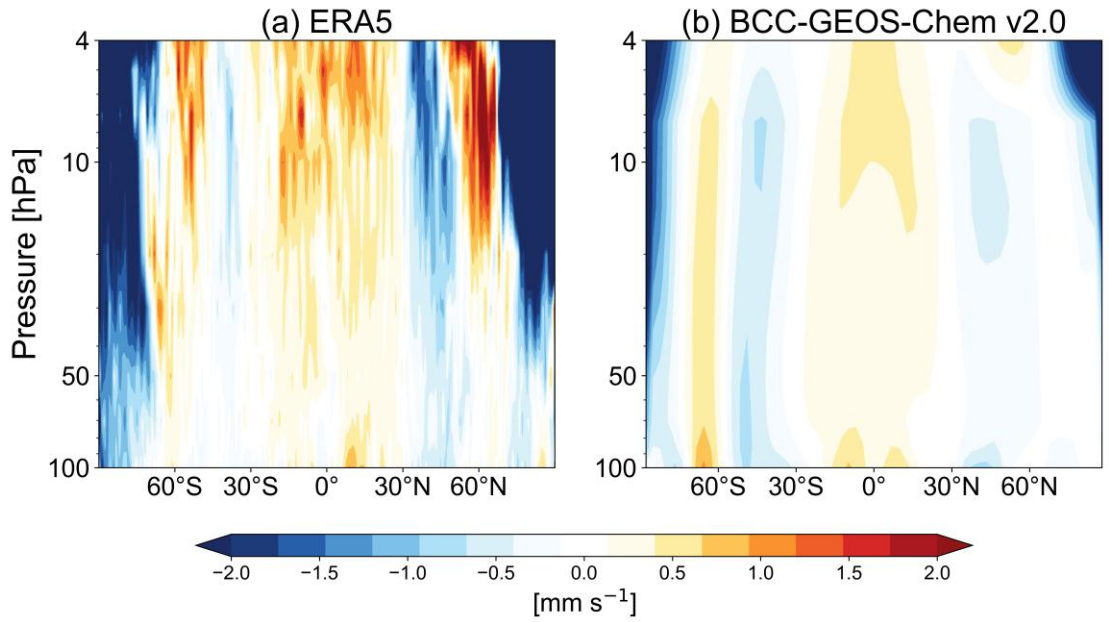


Figure S7. The vertical component of the residual circulation ( $\bar{w}^*$ ) from (a) ERA5 reanalysis mean, and (b) BCC-GEOS-Chem v2.0 under T42L26 configuration. Values are averaged over 2012-2014.

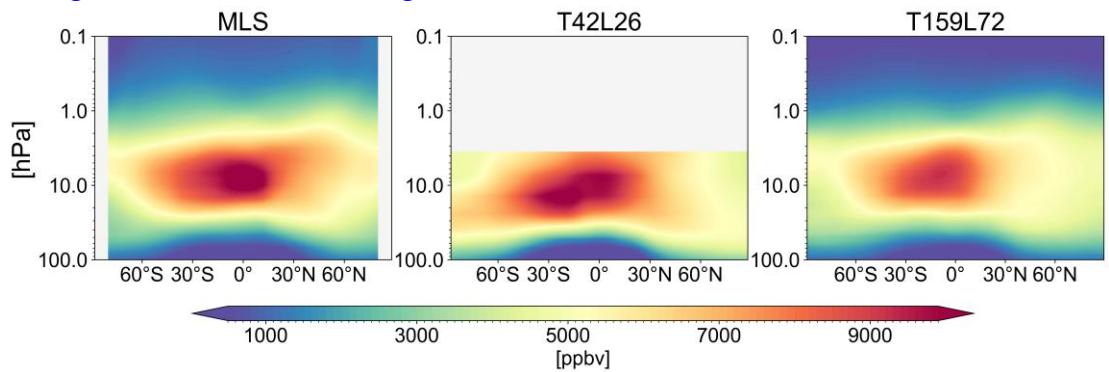


Figure S8. The zonal mean vertical distributions of ozone in the stratosphere from MLS and BCC-GEOS-Chem v2.0 under T42L26/T159L72 configuration. Values are averaged over January 2014.

Table S5 Reference pressures (Pa) under T42L26 and T159L72.

Level edge index	T42L26	T159L72
1	291.4067	0.7983
2	489.5209	1.8412
3	988.2418	3.8939
4	1805.2010	7.5847
5	2983.7240	13.6705
6	4462.3340	23.0105
7	6160.5870	36.4378
8	7851.2430	54.6051

9	9236.5800	77.8864
10	10866.3590	106.7850
11	12783.7080	141.3027
12	15039.3710	180.9154
13	17693.0430	225.1519
14	20814.9440	271.9188
15	24487.7090	320.4182
16	28808.5220	368.0077
17	33891.7310	414.4935
18	39871.8650	457.5551
19	46907.1800	505.4983
20	55183.8710	558.9235
21	64920.9690	618.5109
22	74438.2890	685.0316
23	83102.1230	759.3611
24	90330.0290	842.4946
25	95599.7460	935.4253
26	98511.2200	1038.9650
27	100000.0000	1154.3390
28		1282.9430
29		1426.3410
30		1586.2901
31		1764.7609
32		1963.9671
33		2186.3930
34		2434.8330
35		2712.4252
36		3022.6995
37		3369.6274
38		3757.6757
39		4191.8742
40		4677.8855
41		5222.0914
42		5831.3391
43		6511.8779
44		7271.8302
45		8120.4504
46		9068.0415
47		10126.0499
48		11307.0393
49		12625.0292
50		14095.5424
51		15735.8903

52		17565.4086
53		19605.6228
54		21880.8550
55		24403.9081
56		27157.7947
57		30156.2026
58		33415.3190
59		36952.5895
60		40785.8893
61		44934.0791
62		49416.9101
63		54254.6913
64		59469.0591
65		65082.1298
66		71117.2991
67		77598.9279
68		83529.3613
69		88748.7724
70		93113.8789
71		96503.1638
72		98821.1013
73		100000.0000

*[Comment#2-3 MCI] The section on code availability is very incomplete as it includes only the “coupler” component between GEOS-Chem and BCC-ESM, and a reference to the generic GEOS-Chem code distribution of the Harvard Atmospheric Chemistry Modelling group. Every module of BCCGEOS-Chem v2.0 (see Fig. 1) should be covered in this section. If these modules are distributed through different DOI repositories, then their assembly should build “out of the box”. For example: can GEOS-Chem be linked directly to the coupler code archived for this paper? If the Geos-Chem modules require some tailored modifications, then this forked code should be distributed by the authors of this paper. What is the code availability for the dynamics, physics and HEMCO modules of BCC-AGCM?*

[Response#2-3] We thank the reviewer for raising this important point regarding code availability and reproducibility. The modules in BCC-GEOS-Chem can indeed be assembled and built out of the box. Regarding GEOS-Chem modules, we acknowledge that some tailored modifications are required for their coupling with the BCC model. However, these modifications have already been merged to the GEOS-Chem main code branch through recent updates in version v14.6.0 (<https://github.com/geoschem/geos-chem/releases/tag/14.6.0>). For instance, the BCC-specific implementations are controlled via conditional compilation (e.g., using `#ifdef BCC`), ensuring that the standard GEOS-Chem functionality is

preserved and that no separate forked version needs to be distributed by the authors. As we mentioned in Section 2.2, “Minor modifications to GEOS-Chem source code were made to accommodate the BCC-GC-HEMCO interface, which have been merged to the GEOS-Chem main code branch (<https://github.com/geoschem/geos-chem>, last access: 1 April 2025) to ensure future compatibility between BCC and GEOS-Chem”. We have updated the dynamics, physics, and HEMCO modules in our code availability:

“The GEOS-Chem model is maintained at the Harvard Atmospheric Chemistry Modelling group, which can be obtained at <https://doi.org/10.5281/zenodo.7271960> (last access: 1 April 2025). The HEMCO modules can be obtained at <https://github.com/geoschem/HEMCO> (last access: 1 January 2025). The BCC-GEOS-Chem v2.0 code can be accessed at a DOI repository <https://zenodo.org/records/18448688> (Sun, 2025a), and model outputs for 2012–2014 are available at <https://zenodo.org/records/16734757> (Sun, 2025b). All source code and data can also be accessed by contacting the corresponding authors Lin Zhang ([zhanglg@pku.edu.cn](mailto:zhanglg@pku.edu.cn)).”

*[Comment#2-4 MC2] Climate model evaluations usually rely on mean bias estimates from comparisons between decadal simulations and observational climatologies (for stratospheric composition, see e.g. Froidevaux et al., 2019). When no decadal simulation is available (as seems the case here), the simulations are commonly run in “chemical forecast” mode, with composition running unconstrained but meteorological fields constrained by observations through reanalyses (e.g. MERRA2 or ERA5). Like all CTMs, GEOS-Chem was developed and evaluated in such a configuration. But BCC-GEOS-Chem v2 seemingly lacks this ability, raising the possibility that composition biases are due to unrealistic temperature or circulation patterns. To mitigate this concern, the paper should include, at least in its Supplementary material, all relevant evaluations of temperature and circulation patterns over the evaluated period (2012-2014) and these evaluations should be considered in the discussions of section 4. The introduction should also clarify if BCC-GEOS-Chem v2 can be constrained (or nudged?) by reanalyses, or if there are plans to develop such configurations in the future. This is especially relevant for the Air Quality applications discussed in section 6.*

[Response#2-4] At present, BCC-GEOS-Chem v2.0 operates with a free-running meteorology configuration, and does not support direct nudging or constraint capability to reanalyses like MERRA-2 or ERA5. This capability was not developed because GEOS-Chem already uses assimilated meteorology to drive the identical chemical module. We acknowledge that enabling a “chemical forecast” mode could be valuable in some applications. We also agree that simulated biases in composition identified in this study may partially originate from biases in simulated temperature or large-scale circulation, in addition to chemical process uncertainties.

Beyond the radiation, cloud, and precipitation (Figure 9 and 10), we have added an

evaluation of the simulated temperature and wind fields over the 2012–2014 period in the Supplementary Material (Figure S3). The model accurately reproduces tropospheric temperature, wind fields, radiation, cloud cover, and precipitation. However, it shows limitations in simulating the stratospheric Brewer-Dobson Circulation ([Response#2-2]). Consequently, biases in atmospheric compositions are likely attributable to emission inventory and chemical mechanism uncertainties, though meteorological field biases may contribute. These points are addressed in the text.

In Section 3.2:

“Evaluations with these data shows that BCC-GEOS-Chem v2.0 accurately reproduces tropospheric temperature, wind fields, radiation, cloud cover, and precipitation (Figure S3). However, it has limitations in simulating stratospheric circulations, which may contribute to stratosphere ozone biases, as will be detailed in Section 4.1. Consequently, biases in tropospheric compositions are more likely attributable to uncertainties in emission inventory and chemical mechanisms, which will be the focus of the following discussions. Nevertheless, we acknowledge that the potential contribution from meteorology to biases in tropospheric compositions cannot be ruled out.”

In Section 4.1:

“However, we also note that substantial biases persist in BCC-GEOS-Chem v2.0. We find an underestimation of 5-20% in upper-stratospheric ozone (30–4 hPa) relative to MLS, accompanied by a pronounced cold bias in upper-stratospheric temperature compared to ERA5 reanalysis data. Besides, BCC-GEOS-Chem v2.0 overestimates lower-to-middle stratospheric ozone (100–50 hPa) by over 50% compared to MLS, leading to an overestimation of the total column ozone (Figure S5c). In addition, while the model reproduces the overall vertical distribution of key stratospheric chemical component (e.g. NO<sub>x</sub>, NO<sub>y</sub>, methane), there are notable discrepancies in both the magnitude and location of concentration hotspots (Figure S6).

These biases may be partly attributable to the model’s limited top height in the T42L26 configuration. This constrains its ability to accurately simulate stratospheric circulations, and consequently, to reproduce the distributions and magnitudes of stratospheric chemical components that are significantly modulated by intense climate-chemistry interactions within the stratosphere (such as the interaction between stratospheric ozone and temperature). We find that although BCC-GEOS-Chem v2.0 captures the overall structure of the residual circulation (Figure S7), it underestimates the strength of the Brewer–Dobson circulation in the upper stratosphere over both the equatorial regions and high latitudes. This weaker tropical upwelling may partly account for high biases in lower-stratospheric ozone over the tropics.”

We have also clarified the meteorological fields in model setup:

“These three models are configured with meteorology free-running, and currently does not support nudging or constraining of meteorological fields to reanalysis.”

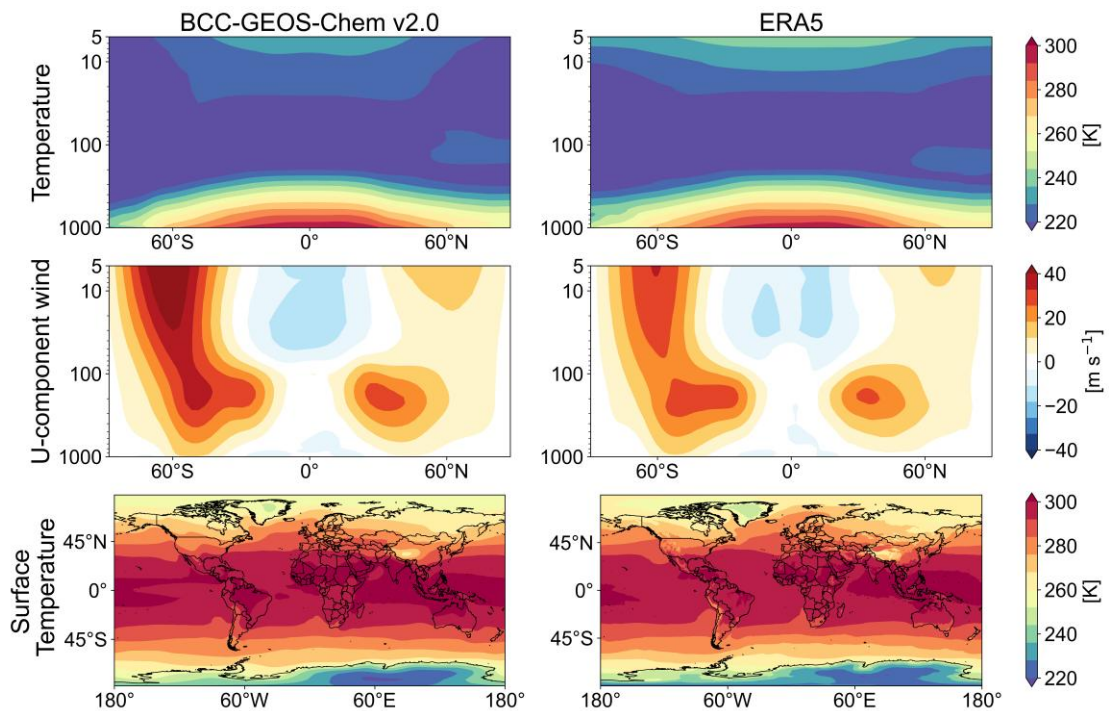


Figure S3. Zonal-mean temperature, U-component wind, surface temperature averaged over 2012-2014 from BCC-GEOS-Chem v2.0 under T42L26 configuration and ERA5 reanalysis.

*[Comment#2-5 MC3] The evaluation and discussion of CH<sub>2</sub>O, NO<sub>2</sub>, CO, and aerosols (section 4.2) is too superficial and requires major improvements. The explanations based on biases emission levels are not credible, because these biases are much too small to explain the reported biases in these three species.*

[Response#2-5] We thank the reviewer for this important comment and agree that the original discussion in Section 4.2 requires improvements. In the revised manuscript, we have substantially expanded Section 4.2 to explicitly distinguish the controlling factors for different species, discuss the relative roles of emissions versus chemistry, and provide a more balanced interpretation of the simulated biases in CH<sub>2</sub>O, NO<sub>2</sub>, CO, and aerosols among the three models:

“Comparison with satellite observations in Figure 6 demonstrate that BCC-GEOS-Chem v2.0 has significant improvements in simulating tropospheric CH<sub>2</sub>O, NO<sub>2</sub>, and CO. BCC-GEOS-Chem v2.0 shows a negative mean bias to OMI tropospheric CH<sub>2</sub>O column of  $-1.2 \pm 1.5 \times 10^{15}$  molecule cm<sup>-2</sup> averaged over 2012-2014, with positive bias at the tropics. In comparison, both BCC-GEOS-Chem v1.0 and BCC-AGCM-Chem show positive bias of over 50% over the Amazon, central Africa, tropical Asia, and the southeastern United States. These two models show 10-50% larger BVOCs emission over the

hotspot regions compared to BCC-GEOS-Chem v2.0 despite comparable global total emissions (410 versus 389 Tg yr<sup>-1</sup>), thereby partly explaining their high biases. In addition, the updated aromatic chemistry in BCC-GEOS-Chem v2.0 (GEOS-Chem version 13.0.0 onwards) further reduces simulated CH<sub>2</sub>O over tropical regions such as central Africa and the Amazon (Bates et al., 2021). BCC-GEOS-Chem v2.0 also shows lower tropospheric CH<sub>2</sub>O columns over the mid-latitudes in both hemispheres compared to OMI retrievals, consistent with the other two models. This pattern may be partly attributed to systematic uncertainties in satellite retrievals, as previous studies have shown that CH<sub>2</sub>O retrievals are biased low on average by 20–51% relative to aircraft observations across mid-latitude regions (Zhu et al., 2016; Zhu et al., 2020).

For tropospheric NO<sub>2</sub>, BCC-GEOS-Chem v2.0 shows no significant global mean bias compared to OMI tropospheric NO<sub>2</sub> column ( $0.0 \pm 1.3 \times 10^{15}$  molecule cm<sup>-2</sup>), but this reflects the compensation of negative bias over emission hotspots such as East Asia, India, Western Europe and central Africa, and positive bias over other regions. In comparison, the simulated tropospheric NO<sub>2</sub> column in BCC-GEOS-Chem v1.0 and BCC-AGCM-Chem show substantially high positive biases of  $4\text{--}7 \times 10^{14}$  molecule cm<sup>-2</sup> (55.4–107.3%) over continental regions, especially over East Asia and India. Given comparable surface NO<sub>x</sub> emissions and consistent lightning NO<sub>x</sub> parameterization across the three models, the improved performance in simulating tropospheric NO<sub>2</sub> columns is likely driven by differences in NO<sub>x</sub> chemistry and nitrate aerosol treatments between the models. Compared to BCC-GEOS-Chem v1.0, BCC-GEOS-Chem v2.0 with up-to-date GEOS-Chem chemical mechanism has incorporated updated aromatic chemistry (GEOS-Chem version 13.0.0 onwards), the photolysis of nitrate (GEOS-Chem version 14.2.0 onwards, not included in this study), as well as the reaction updates in NO<sub>x</sub> chemistry. These updates reduce bias in simulated NO<sub>2</sub> column concentrations, as reported in CESM2-GC (Fritz et al., 2022). Besides, the absence of nitrate aerosol chemistry in BCC-AGCM-Chem (Wu et al., 2020) limits a realistic representation of the gas–particle partitioning of reactive nitrogen, thereby leading to the overestimation of tropospheric NO<sub>2</sub> column.

For CO, we evaluate simulated CO concentrations at 700 hPa, where MOPITT satellite has generally high sensitivity (Emmons et al., 2004; Pfister et al., 2005). BCC-GEOS-Chem v2.0 has also substantially reduced the positive bias relative to observed values, averaged  $4.9 \pm 11.9$  ppbv (5.5%) over the globe, compared with the excessive positive CO bias of BCC-GEOS-Chem v1.0 ( $24.8 \pm 12.9$  ppbv) and BCC-AGCM-Chem ( $50.8 \pm 14.1$  ppbv) with even larger bias over Asia, central Africa, and the East Pacific Ocean. The excessive CO concentrations in BCC-AGCM-Chem are primarily attributable to a severely underestimated atmospheric oxidizing capacity, with the global column-weighted mean OH concentration being nearly 50% lower than that in BCC-

GEOS-Chem v2.0 ( $0.5 \times 10^6$  molecule  $\text{cm}^{-3}$  versus  $1.0 \times 10^6$  molecule  $\text{cm}^{-3}$ ), in addition to higher biomass burning and anthropogenic emissions ( $1140 \text{ Tg yr}^{-1}$  versus  $925 \text{ Tg yr}^{-1}$ ; Table S1).

Figure 7 evaluates the simulated surface fine particulate matter ( $\text{PM}_{2.5}$ ) concentrations with the satellite-derived data as introduced in Section 3.2 (Shen et al., 2024). The satellite-derived data reveal high  $\text{PM}_{2.5}$  concentration over East Asia and India due to intensive anthropogenic emissions, and over northern and central Africa due to mineral dust and biomass burning emissions. BCC-GEOS-Chem v2.0 reproduces the overall spatial distributions and magnitude of observed  $\text{PM}_{2.5}$  concentrations, though it tends to marginally underestimate concentrations in the hotspot regions (e.g., Amazon, central Africa, and India), which is most likely due to the coarse model resolution, and the uncertainties of biomass burning (Reddington et al., 2017). BCC-GEOS-Chem v2.0 demonstrates superior performance in simulating  $\text{PM}_{2.5}$  concentration compared with BCC-GEOS-Chem v1.0, which exhibits pronounced overestimation in East Asia, India, the Arabian Peninsula, and Northern Africa, and BCC-AGCM-Chem, which significantly underestimates  $\text{PM}_{2.5}$  concentrations in major emission hotspot regions. This improvement is likely due to its better performance in simulating the gas-phase precursor as shown above (Figure 6), as well as its more comprehensive representation of the aerosol chemical mechanism. The large negative bias in BCC-AGCM-Chem is mainly due to the absence of nitrate aerosol chemistry and the incomplete representation of secondary organic aerosol formation (Wu et al., 2020).

#### Additional bibliographical references:

Zhu, L., Jacob, D. J., Kim, P. S., Fisher, J. A., Yu, K., Travis, K. R., Mickley, L. J., Yantosca, R. M., Sulprizio, M. P., De Smedt, I., González Abad, G., Chance, K., Li, C., Ferrare, R., Fried, A., Hair, J. W., Hanisco, T. F., Richter, D., Jo Scarino, A., Walega, J., Weibring, P., and Wolfe, G. M.: Observing atmospheric formaldehyde (HCHO) from space: validation and intercomparison of six retrievals from four satellites (OMI, GOME2A, GOME2B, OMPS) with SEAC4 RS aircraft observations over the southeast US, *Atmos. Chem. Phys.*, 16, 13477–13490, <https://doi.org/10.5194/acp-16-13477-2016>, 2016.

Zhu, L., González Abad, G., Nowlan, C. R., Chan Miller, C., Chance, K., Apel, E. C., DiGangi, J. P., Fried, A., Hanisco, T. F., Hornbrook, R. S., Hu, L., Kaiser, J., Keutsch, F. N., Permar, W., St. Clair, J. M., and Wolfe, G. M.: Validation of satellite formaldehyde (HCHO) retrievals using observations from 12 aircraft campaigns, *Atmospheric Chemistry and Physics*, 20, 12329–12345, <https://doi.org/10.5194/acp-20-12329-2020>, 2020.

*[Comment#2-6 MC4] The text is often difficult to read and requires English language improvements beyond the suggestions below.*

[Response#2-6] We thank the reviewer for this comment. In the revised version, we have carefully edited the entire manuscript to improve readability, clarity, and consistency of the language. The text has been thoroughly revised to address grammatical issues, unclear phrasing, and overly complex sentence structures. In addition, we have incorporated all specific language suggestions provided by the reviewer.

*[Comment#2-7 SC1] Lines 50-57: the IPCC reports are not included in the list of references. Please correct this by adding references to the relevant chapters in each WG1 Assessment Report. Please cite additional and specific references to support the statement “most existing CMIP models employ highly simplified representations of atmospheric chemistry” (lines 54-55).*

[Response#2-7] We thank the reviewer for pointing this out. We have corrected the omission of the IPCC references in Lines 50–57 by adding citations to the relevant chapters of the IPCC Working Group I Assessment Reports:

“There is an increasing number of climate system models that incorporate interactive atmospheric chemistry from the Phase 5 to the Phase 6 of the Coupled Model Intercomparison Project (CMIP5&CMIP6) (IPCC AR5, 2013, IPCC AR6, 2021). These models play a pivotal role in supporting the Intergovernmental Panel on Climate Change (IPCC) assessment reports, especially in assessing the contribution of both long-lived and short-lived climate forcers (SLCFs) to global change. However, many existing CMIP models employ highly simplified representations of atmospheric chemistry (IPCC AR5, 2013, Working Group I, Chapter 9, Appendix Table 9. A.1), For example, only 10 of the 39 models participated in CMIP5 simulated atmospheric chemistry interactively, which introduces substantial uncertainties in quantitative assessments, particularly for SLCFs with high spatiotemporal heterogeneities.”

Additional bibliographical references:

IPCC: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Stocker, T. F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P. M., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 1535 pp., <https://doi.org/10.1017/CBO9781107415324>, 2013.

IPCC, 2021: Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change, [Masson-Delmotte, V., P. Zhai, A. Pirani, S.L. Connors, C. Péan, S. Berger, N. Caud, Y. Chen, L. Goldfarb, M.I. Gomis, M.

Huang, K. Leitzell, E. Lonnoy, J.B.R. Matthews, T.K. Maycock, T. Waterfield, O. Yelekçi, R. Yu, and B. Zhou (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, In press, doi:10.1017/9781009157896.

[Comment#2-8 SC2] Line 140: please cite a reference for “the National Centre for Atmospheric Research (NCAR) flux coupler.”

[Response#2-8] We thank the reviewer for pointing this out. We have added an appropriate reference for the National Center for Atmospheric Research (NCAR) flux coupler at Line 140:

“Each component above interacts through bidirectional flux exchanges of momentum, energy, water, and carbon facilitated by the coupler, which is adapted from National Centre for Atmospheric Research (NCAR) flux coupler (Craig et al., 2012, Wu et al., 2019). The coupling under different resolution is achieved using area-weighted or bilinear interpolation schemes that preserve global integrals of energy, momentum, and mass.”

Additional bibliographical references:

Craig, A. P., Vertenstein, M., and Jacob, R.: A new flexible coupler for earth system modeling developed for CCSM4 and CESM1, The International Journal of High Performance Computing Applications, 26, 31–42, <https://doi.org/10.1177/1094342011428141>, 2012.

[Comment#2-9 SC3] Lines 243-244: This sentence is confusing. Please summarize again the aspects of an “interactive troposphere-stratosphere” which were missing in BCC-GEOS-Chem v1.0.

[Response#2-9] We thank the reviewer for pointing out that the original statement was unclear. We have revised Lines 243–244 accordingly to explicitly summarize these missing aspects in v1.0 and to clarify the specific improvements introduced in v2.0, please kindly find [Response #2-2].

[Comment#2-10 SC4] Lines 282-283: please list the modelled SLCF explicitly. It looks like this sentence is not about SLCF in general but aerosols specifically.

[Response#2-10] We thank the reviewer for pointing out this ambiguity. In Lines 282–283, the direct (radiation) and indirect (cloud) climate effects of SLCFs refer to both aerosols and short-lived greenhouse gases (e.g., tropospheric ozone), which are interactively represented in BCC-GEOS-Chem v2.0. We agree that the discussion related to climate feedback mainly concerns aerosol-related processes. To avoid confusion, we have revised the text to clarify that our discussion emphasizes aerosol feedbacks rather than SLCFs in general:

“One significant update in BCC-GEOS-Chem v2.0, relative to BCC-GEOS-Chem v1.0, is the incorporation of the direct (radiation) and indirect (cloud)

climate effects of greenhouse gases and aerosols.”

*[Comment#2-11 SC5] Lines 299-300, this sentence is very unclear: “Finally, aerosols across different size bins are assumed to be externally mixing, which are subsequently used in radiative transfer calculation”. Please re-formulate and clarify!*

[Response#2-11] We thank the reviewer for pointing out that this sentence was unclear. In the original text, our intention was to describe the aerosol mixing state assumed in the model and how it is treated in the radiative transfer calculations. We have reformulated this sentence to clarify both the physical assumption and its implementation in the radiation calculation:

“Aerosols across different size bins are assumed to be externally mixed in the calculation of bulk aerosol single-scattering properties. The derived radiative properties are subsequently used in radiative transfer calculation.”

*[Comment#2-12 SC6] Lines 337-339: “We note that as BCC-GEOS-Chem only extends to the height of 2.914 hPa, ozone concentrations at the top two layers are set to prescribed monthly climatological values from CMIP6 data package as upper boundary conditions.” This is a key issue which needs to be expanded (see General Comment). If these Boundary Conditions are kept, where do they come from? What is the second pressure level? Does this approach introduce vertical discontinuities?*

[Response#2-12] We thank the reviewer for highlighting the importance of the upper boundary condition treatment. Please kindly find [Response#2-2]. In particular, we consider that no sharp vertical discontinuities are introduced. In BCC-GEOS-Chem v2.0, the stratospheric ozone below the top boundary evolves through fully interactive chemistry and transport, ensuring a dynamically and chemically consistent vertical structure throughout the stratosphere. This is supported by Figure 4. Moreover, the use of prescribed upper boundary conditions for ozone is not unique to our model configuration. Even in the “Classical” GEOS-Chem chemical transport model, ozone is influenced by upper boundary constraints provided from the Global Modeling Initiative (GMI), and such treatments have not been shown to introduce noticeable vertical discontinuities. In our implementation, the prescribed ozone fields are vertically interpolated onto the native model levels, further ensuring a smooth transition near the model top.

*[Comment#2-13 SC7] Lines 664-665: “...with 72 vertical hybrid layers extending from the surface to 0.01 hPa.” See the general comment: if the authors decide to keep the current low top at 2.914 hPa, this higher top in the high-resolution configuration allows at least to check the impact of the low top in the low-resolution configuration. The required evaluation of the vertical distribution of ozone should include the results from this high-resolution simulation in the upper stratosphere. The Initial Conditions for this high-resolution simulation should be described because they are very important for stratospheric evaluation. The short duration of this simulation should not be an issue*

*in the upper stratosphere, because the ozone lifetime is shorter than in the middle and lower stratosphere.*

[Response#2-13] We thank the reviewer for this constructive suggestion. Please kindly find [Response#2-2]. In particular, the initial conditions for the simulation are obtained from a long-term GEOS-Chem simulation driven by assimilated meteorological fields over 1995–2017 (Wang et al., 2022), which provides a well spun-up stratospheric chemical state. This information is presented in Section 3.1 “The initial conditions for atmospheric dynamics and physics in 2011 are obtained from the historical simulations (1850-2014) conducted by BCC-ESM1 under CMIP6 framework, and initial states of chemical tracers are obtained from a GEOS-Chem simulation over 1995-2017 (Wang et al., 2022).”

[Comment#2-14] Line 64: *“present-day atmospheric composition and...”*

[Response#2-14] We have revised accordingly.

[Comment#2-15] Line 78: *“BCC-GEOS-Chem v1.0 resulted from the integration of...”*

[Response#2-15] We have revised accordingly.

[Comment#2-16] Line 144: *“Functional advancements that enable feedbacks from aerosol and greenhouse gases to dynamics are introduced separately in Section 2.4.”*

[Response#2-16] We have revised accordingly.

[Comment#2-17] Lines 196-198: *“...future updates of GEOS-Chem and HEMCO in the BCC-GEOS-Chem model would only require downloading and substituting the corresponding source code, avoiding significant modifications to the source code of the other BCC-AGCM modules.”*

[Response#2-17] We have revised accordingly.

[Comment#2-18] Line 210: *“...further modularized HEMCO to enable coupling to the Community Earth System Model (CESM)...”*

[Response#2-18] We have revised accordingly.

[Comment#2-19] Line 217: *“...in the context of rapid updates...”*

[Response#2-19] We have revised accordingly.

[Comment#2-20] Line 222: *“...that are already implemented in HEMCO and driven by...”*

[Response#2-20] We have revised accordingly.

[Comment#2-21] Line 250: *“Details of aerosol chemistry and its interaction with gas-phase chemistry are provided in Lu et al. (2020)”*

[Response#2-21] We have revised accordingly.

[Comment#2-22] Line 252: “... and the ISORROPIA II (Fountoukis and Nenes, 2007) thermodynamic module.”

[Response#2-22] We have revised accordingly.

[Comment#2-23] Lines 258-259: “The scheme requires the input of geography data...”

[Response#2-23] We have revised accordingly.

[Comment#2-24] Lines 276-278: “...instead of using the convection module in BCC-AGCM-Chem described by Wu et al. (2020). This is because the BCC-AGCM-Chem scheme for wet deposition is hardcoded, incompatible with the updated chemical species, and it lacks the scavenging of water-soluble species in convective updrafts...”

[Response#2-24] We have revised accordingly.

[Comment#2-25] Lines 283-284: “This incorporation relies on the radiative transfer module already implemented in BCC-ESM1 (Wu et al., 2020).”

[Response#2-25] We have revised accordingly.

[Comment#2-26] Line 285: “...and incorporates explicit parameterizations for the major absorbers...”

[Response#2-26] We have revised accordingly.

[Comment#2-27] Line 293: “The aerosol direct effect is calculated based on the mass mixing ratios of bulk aerosols, which are prognostic variables in the GEOS-Chem chemical module.”

[Response#2-27] We have revised accordingly.

[Comment#2-28] Line 301: “The indirect effects of aerosols involve their role as cloud condensation nuclei and their influence on...”

[Response#2-28] We have revised accordingly.

[Comment#2-29] Line 306: “...diagnosed from the GEOS-Chem chemical module.”

[Response#2-29] We have revised accordingly.

[Comment#2-30] Lines 310-311 (suggestion to remove words stating the obvious): “...where  $\beta$  is a scaling factor,  $\rho_w$  is the water density, and LWC is the cloud liquid water content. Both  $\rho_w$  and LWC are diagnosed by BCC-AGCM.”

[Response#2-30] We have revised accordingly.

[Comment#2-31] Lines 313-317: “The detailed treatment of the aerosols feedback on precipitation can be found in Wu et al (2020). The current approach for describing the aerosol feedback uses a bulk-mass representation of aerosols in BCC-GEOS-Chem v2.0. This approach is similar to that used in the majority of CMIP5 models, since only two

*of these models include online size-resolved aerosol microphysics (Kodros and Pierce, 2017). This bulk-mass representation of aerosols is computationally efficient, but does not consider...”*

[Response#2-31] We have revised accordingly.

*[Comment#2-32] Lines 320-322: “...microphysics scheme (Kodros and Pierce, 2017) from GEOS-Chem to more accurately simulated size-dependent aerosol chemistry and microphysics. Such developments will be available through BCC-GC-HEMCO interface once these two schemes become compatible with the GEOS-Chem column structure.” Consider a re-formulation starting e.g. with “These two schemes are available in GEOS-Chem and simulate more accurately the size dependency of aerosol chemistry and microphysics, but are not yet integrated in BCC-GEOS-Chem v2.0 because...”*

[Response#2-32] Thank you very much for the comment. We have revised to the following statement:

“For future model development, we hope to explicitly describe aerosol size distributions by integrating size-resolved schemes such as the Advanced Particle Microphysics (APM) scheme (Yu and Luo, 2009) and the Two-Moment Aerosol Sectional (TOMAS) microphysics packages (Kodros and Pierce, 2017). These two schemes are available in GEOS-Chem and simulate more accurately the size dependency of aerosol chemistry and microphysics, but are not yet integrated in BCC-GEOS-Chem v2.0 because these two schemes are currently incompatible with the GEOS-Chem column structure.”

*[Comment#2-33] Line 334: “...under the CMIP6 framework, ...”*

[Response#2-33] We have revised accordingly.

*[Comment#2-34] Lines 335: “The model results in 2012-2014 are used for...”*

[Response#2-34] We have revised accordingly.