Reply of RC2: Review of Lu et al., "Development of the global atmospheric general circulation-chemistry model BCC-GEOS-Chem v1.0: model description and evaluation"

5 **Reviewer #2**

Comment#2-1: The manuscript presents an overview and assessment of a newly constructed chemistry-climate model that has resulted from the linking of GEOS-Chem with the Beijing Climate Center AGCM. The general features of the model are presented and a fairly extensive comparison against observations for ozone are presented. In addition, other aspects of the model chemical climate are presented,

- 10 presented. In addition, other aspects of the model chemical climate are presented, including the global distribution of OH, ozone budget terms, and some limited comparisons for aerosol quantities against observations including AOD and speciated aerosol concentrations over the US.
- 15 The manuscript is very clearly written and presented, providing a fairly complete overview of the model components and an idea of how the chemical climate of BCC GEOS-Chem compares with GEOS-Chem itself and with other chemistry climate models.

Response#2-1: We thank the reviewer for the valuable comments. All of them have been implemented in the revised manuscript. Please see our itemized responses below.

Comment#2-2: My only significant criticism is focused on Figure 10, comparing column amounts of NO2, CO, SO2 and CH2O. In the discussion of Figure 10, lines 394 – 404, the authors say that the averaging kernels for the satellite observations were not applied to the model concentrations when calculating the column amounts and that the comparisons 'mainly focus ... on spatial variations rather than absolute magnitudes.' In this case the comparison is nothing more than a test to make sure the specified emissions

- 30 are being put into the model in the correct locations. Global models have a longstanding low bias for CO in the northern hemisphere that appears to be related to emissions and the hydroxy radical, but Figure 10 shows that BCC-GEOS-Chem has too high CO in the Northern Hemisphere. Due to the lack of a quantitative comparison with the satellite data by application of the averaging kernel it is impossible to judge whether
- 35 the differences signify anything. While CH2O is not predominantly due to direct emission, the spatial distribution is tightly coupled to the emissions of biogenic hydrocarbons, so the comparison will also be largely driven by having regions of high biogenic emissions in the correct place. The differences in the magnitude of CH2O between the satellite and model is quite large and it would be interesting to have a more
- 40 quantitative comparison with the satellite observations as the qualitative comparison focused on the spatial distribution is not informative at all. I would strongly urge the authors to revise the comparison of the column amounts to be more quantitative by application of the appropriate averaging kernel. My other comments are all minor in nature and are given below.

- 45 Response#2-2: We agree with the reviewer on the need to improve the comparison and discussion for these chemical constituents. For CO, we have now applied the satellite averaging kernels to smooth the model results, and focus on the comparison at 700 hPa where MOPITT has generally high sensitivities. As shown in the new Fig. 10a, the large model high bias has been significantly improved
- 50 though still exists. For other constituents, however, we do not apply corresponding averaging kernel, which requires additional 3-year model simulation to co-sample observations along the satellite tracks. Previous studies (Zhu et al., 2016; 2020) showed CH₂O shape factor (a prior) was not the main driver of the discrepancy between GEOS-Chem modeled and retrieved columns. Their studies also revealed
- that satellite CH₂O retrievals showed significant low bias (up to 50%) compared to aircraft measurements, which may largely explain the model high bias as shown here. For SO₂, we have partly reduced the discrepancies between the observed and modelled SO₂, by removing OMI measurements with slant columns greater than 5 Dobson Units (1.34 × 10¹⁷ molecules cm⁻²) which are affected by strong eruptive volcanoes (Lee et al., 2009, 2011).

We have added much more discussions in Section 3.4 (Evaluation of other atmospheric constituents)

"Figure 10 compares the spatial distributions of annual mean simulated CO, NO₂,

- SO₂, and CH₂O with satellite observations. We evaluate CO at 700 hPa where MOPITT satellite has generally high sensitivity (Emmons et al., 2004; Pfister et al., 2005), and apply averaging kernel to smooth the modelled CO. As shown in Figure 10, BCC-GEOS-Chem v1.0 reproduces the high CO levels over the northern midlatitudes driven by high anthropogenic sources, and over the central Africa driven
- 70 by biomass burning emissions (spatial correlation coefficient r=0.92) with some overestimates. It also captures the observed hotspots of tropospheric NO₂ (r=0.87) and PBL SO₂ columns (r=0.32) over East Asia that generally follow the distribution of anthropogenic sources. The sharp land-ocean gradients for both tracers reflect their short chemical lifetime. We find low biases in the modelled PBL SO₂
- 75 especially over the volcanic eruption regions (e.g., Central Africa) but high biases in the industrialized regions such as East Asia, a pattern consistent with previous comparisons between the OMI and GEOS-Chem PBL SO2 columns, which may reflect inappropriate ship and volcanic emissions in the model (Lee et al., 2009) and/or the model bias in the PBL height. High levels of tropospheric CH₂O column
- are simulated over the Amazon, the central Africa, tropical Asia, and the southeastern US, where CH₂O oxidized from large biogenic emissions of VOCs (r=0.67), but the model shows notable overestimates. Previous studies (Zhu et al., 2016; 2020) showed that satellite CH₂O retrievals are biased low by 20-51% compared to aircraft measurements which would partly explain the model bias.
- 85 Future assessments are required to correct the biases of these gaseous pollutants."



Figure 10. Spatial distributions of satellite observed (top panels) and model simulated (bottom panels) annual mean (a) CO mixing ratio at 700 hPa, (b) tropospheric NO₂ column, (c) SO₂ column in planetary boundary layer, and (d) tropospheric CH₂O column,. Values are 3-year averages for 2012-2014.

References added:

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- Lee, C., Martin, R. V., van Donkelaar, A., O'Byrne, G., Krotkov, N., Richter, A., Huey, L. G., and
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 - Lee, C., Martin, R. V., van Donkelaar, A., Lee, H., Dickerson, R. R., Hains, J. C., Krotkov, N., Richter, A., Vinnikov, K., and Schwab, J. J.: SO2emissions and lifetimes: Estimates from inverse modeling using in situ and global, space-based (SCIAMACHY and OMI) observations, J. Geophys. Res., 116, http://doi.org/10.1029/2010jd014758, 2011.
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- 115 from four satellites (OMI, GOME2A, GOME2B, OMPS) with SEAC(4)RS aircraft observations over the Southeast US, Atmos. Chem. Phys., 16, 13477-13490, http://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
- J. M., and Wolfe, G. M.: Validation of satellite formaldehyde (HCHO) retrievals using observations from 12 aircraft campaigns, Atmospheric Chemistry and Physics Discussions,

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http://doi.org/10.5194/acp-2019-1117, 2020.

- Comment#2-3: Lines 80 83: Here the authors state 'Integration of GEOS-Chem chemical module into CSMs has been enabled by separating the module (which simulates all local processes including chemistry, deposition, and emission) from the simulation of transport, and making it operate on 1-D (vertical) columns in a gridindependent manner (Long et al., 2015; Eastham et al., 2018).' How is the 1-D column version of GEOS-Chem integrated with a 3-D CSM for processes that typically occur
- in the physics of the model such as vertical turbulent diffusion and transport by deep convection? (I do find a description of deep convection and wet deposition around line 190, but no mention of how vertical diffusion is performed.)

Response#2-3: Thanks for pointing it out. The vertical diffusion of the tracers is parameterized using a non-local scheme as described in Holtslag and Boville (1993). We now state in the Section 2.2 (Atmospheric Chemistry): <u>"Tracer</u> <u>advection in BCC-GEOS-Chem v1.0 is performed using a semi-Lagrangian</u> scheme (Williamson and Rasch, 1989) and the vertical diffusion within the

boundary layer follows the parameterization of Holtslag and Boville (1993)"

140 **Reference added:**

Holtslag, A. A. M. and Boville, B. A.: Local versus nonlocal boundary-layer diffusion in a global climate model, J. Climate, 6, 1825–1842, 1993.

Comment#2-4: Lines 181 – 183: The dry deposition uses the general characteristics of
 the land surface as given by the CSM land module BCC-AVIM. Are there also links to
 the land surface scheme for more short-term variables such as stomatal resistance, that
 would allow for effects such as drought on dry deposition?

Response#2-4: The stomatal conductance is simulated but its influences on dry deposition are not considered, therefore the model does not allow drought

- 150 influences the dry deposition through modulating stomatal conductance so far. We now state in Section 2.5 (Dry and wet deposition) <u>"Variables needed for the dry</u> <u>deposition calculation such as the friction velocity, Monin-Obukhov length, and</u> <u>leaf area index (LAI) are obtained from the atmospheric dynamics/physics</u> <u>modules or the land module BCC-AVIM, based on which GEOS-Chem calculates</u>
- 155 the aerodynamic, boundary-layer, and surface resistances. The impacts of some other short-term land variables, such as stomatal conductance, on dry deposition are not included yet."

160 **Comment#2-5:** Line 234: Minor typo in 'The model estimates t global annual ...' **Response#2-5: Corrected.**

Comment#2-6: Lines 311 – 314: Somewhere, either in the discussion of Figure 5 or the caption, there should be mention that the comparison is for annual average ozone.

165 Response#2-6: Thanks for pointing it out. We now state "As shown in Figure 5,

<u>the model well reproduces the observed annual mean ozone vertical structures...</u>". We have also revised the figure caption accordingly.

- Comment#2-7: Lines 314 321: I was a bit curious about why the vertical profile of
 ozone for the Japanese stations shows such a different vertical structure between the
 observations and model in Figure 5. Looking at Figure 6, the 300 hPa doesn't show that
 big of a difference. If 300 hPa is somewhere around 10 11 km, shouldn't the annual
 average in the observations be over 120 ppbv, though it is listed as 90 ppbv on Figure 6?
- 175 **Response#2-7: 300hPa over Japan is around 9 km, and therefore Figures 5 and 6** are consistent. This is a region with frequent stratosphere to troposphere transport and the model may have difficulty in capturing the sharp ozone increases with increasing altitude there.
- 180 **Comment#2-8:** Line 364: Discussing the discrepancy in OH in the tropics between the Spivakovsky climatology the authors state 'This discrepancy appears to be mainly driven by the high bias in ozone levels in this region.' Attempts to understand the reasons for differences in OH between models has shown how many different factors play a role see, for example, Nicely et al. Atmos. Chem. Phys. 20, doi:10.5194/acp-
- 20-1341-2020, 2020. Do the authors have some reason to believe that the ozone and hydroxyl biases are related and, if not I would suggest removing this statement.
 Response#2-8: Thanks for pointing it out. We agree that the OH discrepancies in the model can be driven by more factors than the ozone bias. We now change the statement to "Discrepancies in modeling climate and concentrations of methane, ozone, NOx, and CO can all contribute to the OH bias in climate-chemistry models

<u>(Nicely et al., 2020).</u>"

Reference added:

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E., Stenke, A., Stone, K., Strahan, S., Tilmes, S., Tost, H., Westervelt, D. M., and Zeng, G.: A
machine learning examination of hydroxyl radical differences among model simulations for CCMI1, Atmos. Chem. Phys., 20, 1341-1361, http://doi.org/10.5194/acp-20-1341-2020, 2020.

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