

Interactive comment on

“Attribution of Chemistry-Climate Model Initiative (CCMI) ozone radiative flux bias from satellites” by Le Kuai et al.

We would like to thank the Editorial Review Board and the reviewers for their helpful comments. These feedbacks help us to improve the clarity and readability of the manuscript. Please find our replies following the comments.

Anonymous Referee #2

Review of Kuai et al, “Attribution of Chemistry-Climate Model Initiative (CCMI) ozone radiative flux bias from satellites”

In this paper, Kuai et al. quantify and attempt to interpret model biases in outgoing longwave radiation (OLR) in the wavelength band of strong absorption by tropospheric ozone, 9.6 microns. Using assimilated data, they first construct instantaneous radiative kernels (IRKs) to quantify the sensitivity of OLR at 9.6 μ to four driving variables: tropospheric ozone, water vapor, atmospheric temperature, and surface temperature. The IRKs are calculated at a range of altitudes and across the globe. They then apply the observation-based IRKs to the biases in the four variables in an ensemble of climate models; this step yields the contributions of these variables to biases in OLR at 9.6 μ in each models. The subsequent analysis shows the importance of accurate representations of tropospheric ozone and water vapor in calculating OLR in this wavelength region.

The topic of the paper is important. Understanding the contribution of tropospheric ozone to current and future climate change requires models that can accurately simulate OLR in the relevant wavelengths. The calculations in the paper are fairly straightforward and yield some interesting results – e.g., that the overestimate of tropical tropospheric water vapor present in many models leads to an underestimate of OLR in the ozone wavelength band. But the paper feels like an early draft: much of the analysis in the paper is shallow and the paper is not well written.

I recommend acceptance only after major revisions.

Major comments.

1. A key reason to improve OLR at 9.6 μ is to improve estimates of the radiative forcing from the change in tropospheric ozone from the preindustrial era to the present-day or from the present-day to the future. The manuscript leaves the reader wondering how important are the biases uncovered in the manuscript. Do the biases have much of an effect on ozone forcing estimates over time?

We agree these are all very good points. Yes, the biases we studies have effect on the ozone forcing. We have showed in our early study (Kuai et al., 2017) that the tropospheric ozone radiative effect will be weaker under optical thick sky, such as with cold and wet condition through the secondary effect on ozone IRK. Therefore, the incorrect water vapor or temperature

will cause the response in IRK in 9.6 μm band and also effect the other band in OLR. And also the wrong water vapor and temperature will also cause incorrect cloud cover and then ozone loss. Too much or too less ozone and water vapor will have wrong heating rate and effect temperature and cloud. All these will effect the ozone forcing estimates from present day to the past.

In meanwhile don't have the O₃ RF estimation over time from CCMI models to do the similar analysis as Bowman et al., 2013 for ACCMIP. We have raised our request in the most recent CCMI workshop for the RF calculation from the CCMI models. Hope in near future, the data will be available. Then we will do more analysis to understand the uncovered biases and their effect on the ozone forcing over time.

We mentioned in the introduction how 9.6 m band bias would impact the ozone RF over time at Page 3, line 44.

'Aghedo, et al. (2011) applied the TES IRKs to evaluate the O₃ radiative effect of chemistry-climate models' O₃ biases in the Atmospheric Chemistry Climate Model Inter-comparison project (ACCMIP) (Lamarque, et al., 2013). Bowman et al. (2013) found model OLR bias due to O₃ is correlated with RF in the ACCMIP models. This correlation helped to reduce the inter-model divergence in RF by about 30% (Myhre, et al., 2013).'

2. In discussing the sources of biases in the four driving variables, the manuscript provides some detail about how modelers have struggled to improve estimates of tropospheric ozone (e.g., Oman et al., 2011 and 2013; Stenke et al., 2013; Revell et al., 2015). But nothing is said about model biases in the other three variables. Are these biases well-known or new to the community? If known, what are the potential reasons for these biases?

The different biases in the models are known since there are regularly evaluation versus all kinds of observations but the reason are poorly understood. However, the biases in the four driving variables for CCMI study and their radiative biases are new to CCMI community and the reason are not fully understood. To my knowledge, for the CCMI comparison, there are not yet specific analysis about the lower tropospheric water vapor biases and temperature biases. Most of such studies are about past inter-comparison project, like CMIP6 or ACCMIP. For example, Lamarque et al., 2013 did a fairly rough analysis in water vapor biases for ACCMIP (Fig. S4). There is some information on the bias for the GEOSCCM in the technical report by Molod et al., 2012 (see figures 18 and 19). However, this is not the CCMI run specifically. Monks et al., 2015 showed the correlation between the intermodal differences in OH in the POLMIP study with inter-model differences in water vapor. They found GEOSCCM simulated specific humidity has high bias relative to MERRA reanalysis in most troposphere and also in mid troposphere compared to Atmospheric Infrared sounder (AIRS) data. Strode et al., 2015 section 3.4.3 discussed some water vapor biases impact on OH, methane and methyl chloroform lifetime in GESCCM. Again these analyses are not for CCMI study and didn't provide any potential reasons for the biases.

We add some words at Page 15 line 38 as below.

‘We compute the model biases against reanalysis data for four key variables: O₃, H₂O, Ta, and Ts. Especially for O₃ biases, the newly developed TCR-1 O₃ assimilation data (Miyazaki et al., 2015; Miyazaki and Bowman 2017) are, for the first time, used as the state-of-the-art benchmark for tropospheric O₃ in models. These specific bias comparisons for the CCMI study cause the modelers to investigate the reasons for these biases and motivate them to improve their simulations. For example, MRI-ESM1r1 shows the reduced LNO_x emission help to improve their tropical upper tropospheric O₃ and its radiative bias.’

References mentioned above:

Lamarque, J.-F., Shindell, D. T., Josse, B., Young, P. J., Cionni, I., Eyring, V., Bergmann, D., Cameron-Smith, P., Collins, W. J., Doherty, R., Dalsoren, S., Faluvegi, G., Folberth, G., Ghan, S. J., Horowitz, L. W., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Naik, V., Plummer, D., Righi, M., Rumbold, S. T., Schulz, M., Skeie, R. B., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., Voulgarakis, A., and Zeng, G.: The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): overview and description of models, simulations and climate diagnostics, *Geosci. Model Dev.*, 6, 179–206, <https://doi.org/10.5194/gmd-6-179-2013>, 2013.

Molod, A., Takacs, L., Suarez, M., Bacmeister, J., Song, I.-S., and Eichmann, A.: The GEOS-5 atmospheric general circulation model: Mean climate and development from MERRA to Fortuna, NASA, Goddard Space Flight Center, Greenbelt, MD, 2012.

Monks, S. A., Arnold, S. R., Emmons, L. K., Law, K. S., Turquety, S., Duncan, B. N., Flemming, J., Huijnen, V., Tilmes, S., Langner, J., Mao, J., Long, Y., Thomas, J. L., Steenrod, S. D., Raut, J. C., Wilson, C., Chipperfield, M. P., Diskin, G. S., Weinheimer, A., Schlager, H., and Ancellet, G.: Multi-model study of chemical and physical controls on transport of anthropogenic and biomass burning pollution to the Arctic, *Atmos. Chem. Phys.*, 15, 3575–3603, [doi:10.5194/acp-15-3575-2015](https://doi.org/10.5194/acp-15-3575-2015), 2015.

Strode, S. A., Duncan, B. N., Yegorova, E. A., Kouatchou, J., Ziemke, J. R., and Douglass, A. R.: Implications of carbon monoxide bias for methane lifetime and atmospheric composition in chemistry climate models, *Atmos. Chem. Phys.*, 15, 11789–11805, <https://doi.org/10.5194/acp-15-11789-2015>, 2015.

3. The paper focuses on OLR under clear-sky conditions, which is fine. But cloud cover varies significantly among models. Could variation in clouds also affect model estimates of ozone radiative flux – i.e., OLR at 9.6 μ ? The text should include a discussion of how the focus on clear sky conditions affects the conclusions.

We added some discussion based on reviewer’s feedback at Page 3, Line 26:

‘In addition, the presence of clouds is the primary control on atmospheric opacity. Under the cloudy sky conditions, the roles of these variables other than cloud on TOA flux are much weaker. In addition, the variation in clouds could affect model estimates not only of the ozone but also of the flux sensitivity to ozone and other variables. Both ozone and sensitivity will impact the ozone radiative flux but in opposite directions. With cloud cover, the O₃ loss will be reduced. That means too much cloud would lead to more ozone production. The presence of the

cloud would also cause weaker flux sensitivity to O₃ and other variables (IRKs). Therefore, the cloud effect is a battle between the impact on ozone estimation and the radiative sensitivity to ozone (IRK). The differences in cloud variations between the models will complicate the radiative effect. Furthermore, the study of the cloud effect is also currently limited by the global observations of total cloud cover and IRK product under realistic cloud conditions. Without knowing which models have better cloud cover, we benefit from using IRK based on the observed cloud free data by TES. Therefore, here we first try to access the role of O₃, H₂O, T_a and T_s in the variation of the TOA flux without cloud effect.'

4. The paper is poorly written with many lapses in English, about 1-3 per paragraph throughout. Those co-authors for whom English is a first language should read the manuscript much more closely and rewrite as needed. As is, the manuscript gives the impression that the co-authors did not read it carefully.

We have got the professional grammar check after we addressed all comments from two reviewers.

2

Minor comments.

Page 2. Line 10. Abstract should make clear that the overestimate of atmospheric opacity in the models is due to too much tropospheric ozone and/or water vapor. Also, the large number of significant digits looks suspect.

We addressed reviewer's suggestions by adding words for the reason of the opacity in Page 2, line 8 as below and change the number from 132.9 to 133.

"Overall, the multi-model ensemble mean bias is -133 ± 98 mWm⁻², indicating that they are too atmospherically opaque due to trapping too much radiation in the atmosphere by overestimated tropical tropospheric O₃ and H₂O. Having too much O₃ and H₂O in the troposphere would have different impacts on the sensitivity of TOA flux to O₃ and these competing effects add more uncertainties on the ozone radiative forcing."

Page 2. Line 22. The text should make clear whether these estimates of radiative forcing are instantaneous or adjusted.

This is adjusted radiative forcing. Page 2, Line 21 has been updated as

'Tropospheric O₃ adjusted RF ranges widely from +0.2 to +0.6 Wm⁻² computed from chemistry-climate model ensembles (IPCC AR5, 2013) (Bowman, et al., 2013;Stevenson, et al., 2013).'

Page 3. Line 18. Text should clarify why the western Pacific is particularly opaque.

Page3, line 15 has been updated:

“For example, O₃ changes in more opaque regions, e.g., the Western Pacific, a wet region due to convection, result in a much smaller change in TOA flux than in more transparent regions, e.g., the Middle East, a dry region due to downwelling (Kuai, et al., 2017).”

Page 6. Line 38. Reader wonders whether surface characteristics matter to OLR calculations, or are these captures by T_s?

Yes, surface characteristics like emissivity will effect the OLR calculations. For different surface types, like rock or vegetation, the emissivity could vary from 0.9 to 1. The surface type could relate to the differences of T_s but such difference would be much smaller than seasonal variation of T_s. T_s is a dominate factor to determine OLR. Here we are discussing the variability of the IRK for T_s is mainly controlled by T_s.

Page 9. Line 41. “The flux bias due to T_a is found to be negligible in all models, which indicates that the T_a is relatively accurate.” The statement should probably be qualified to say simply that the modeled T_a estimates provide reasonable radiative fluxes at 9.6 μm. The temperatures may not be so accurate for other purposes.

We addressed the comments now at Page 10, line 5.

‘The flux bias due to T_a is found to be negligible in all models, which indicates that the model T_a estimates provide reasonable radiative fluxes.’

Page 10. Lines 20-25. The authors might consider reordering the figures so that the figures showing biases in each driving variable come right after the figures showing the biases in the fluxes attributable to that variable – e.g., Figure 9 right after Figure 5. That way readers can more easily see how the driving variables affect fluxes.

We did reconsider the reviewer’s great suggestion but decided to keep the current structure. Section ‘5.3 Vertically-resolved radiative bias of the O₃, H₂O and T’ introduce the partitioned radiative biases for all four variables and summarized their features briefly. This let the readers to do the cross comparison between the variables. Then, in the section ‘5.4 The Spatial Source of TOA Flux Bias’, we did the discussion in the style as reviewer’s way. For example for O₃, we talk about Fig. 5 and Fig. 9. For H₂O, we discuss Fig. 6 and Fig. 10 together.

Page 11. Lines 20-25. Are these ozone biases, reported in ppb, weighted by air density? If not, perhaps they should be, given that the opacity of the atmosphere is affected by total ozone column, not average ozone mixing ratio in that column.

Yes, the ozone biases are reported in ppb. When we do the vertical integration in free troposphere (Eq. 5), the column density is considered.

Page 12. Line 8. What updates did Oman 2011 and Oman 2013 implement in GEOSCCM to improve the model’s representation of ozone? Readers will want to know.

We added more detailed discussion about how does GEOSCCM improved their ozone representation in Page 12, line 26.

‘The GEOSCCM has been used to study the tropospheric O₃ response to variations in the El Nino-Southern Oscillation (ENSO) where Oman, et al., (2011;2013) compared the model to satellite observations. These regular comparisons may have led to the improved simulation of tropospheric O₃ profiles and consequently lower vertical O₃ bias. The GEOSCCM model in the CCMI study uses the tropospheric/stratospheric chemical package developed within the Global Modeling Initiative (GMI) program (Duncan, et al., 2007) which has more realistic ozone chemistry, an internally generated quasi-biennial oscillation, an improved air/sea roughness parameterization and other improvements (Oman et al., 2014).

Nielsen et al., (2017) showed that GEOSCCM successfully reproduces the changes in the quasi-global (60°S–60°N) annual-mean trend in total O₃ column since 1960s to the present day. For the present-day atmosphere, simulated tropospheric partial column O₃ from GESCCM Ref-C1 for CCMI was compared to satellite observations of OMI and MLS (Ziemke et al., 2011). The differences are mostly a few Browner Dobson (DU) except the Northern Hemisphere subtropics and middle latitudes in autumn and winter with the 4–6 DU bias which are under investigation.’

New References:

Nielsen, J. E., S. Pawson, A. Molod, B. Auer, A. M da Silva, A. R. Douglass, B. Duncan, Q. Liang, M. Manyin, L. D. Oman, W. Putman, S. E. Strahan and K. Wargan, 2017. **Chemical mechanisms and their applications in the Goddard Earth Observing System (GEOS) earth system model**. Journal of Advances in Modeling Earth Systems, 9, 3019-3044, DOI: <https://doi.org/10.1002/2017MS001011>.

Molod, A., Takacs, L., Suarez, M., Bacmeister, J., Song, I.-S., & Eichmann, A. (2012). *The GEOS-5 Atmospheric General Circulation Model: Mean Climate and Development from MERRA to Fortuna* (NASA Tech. Rep. Series on Global Modeling and Data Assimilation, NASA/TM-2012–104606, Vol. 28, 117 pp.). Greenbelt, MD: NASA Goddard Space Flight Center.

Oman, L. D., & Douglass, A. R. (2014). Improvements in total column ozone in GEOSCCM and comparisons with a new ozone-depleting substances scenario. *Journal of Geophysical Research: Atmospheres*, 119, 5613– 5624. <https://doi.org/10.1002/2014JD021590>

Ziemke, J. R., Chandra, S., Labow, G. J., Bhartia, P. K., Froidevaux, L., & Witte, J. C. (2011). A global climatology of tropospheric and stratospheric ozone derived from Aura OMI and MLS measurements. *Atmospheric Chemistry and Physics*, 11, 9237– 9251. <https://doi.org/10.5194/acp-11-9237-2011>

Duncan, B. N., Strahan, S. E., Yoshida, Y., Steenrod, S. D., and Livesey, N.: Model study of the cross-tropopause transport of biomass burning pollution, *Atmos. Chem. Phys.*, 7, 3713-3736, <https://doi.org/10.5194/acp-7-3713-2007>, 2007.

Page 12. Line 25. The text should provide a reference to substantiate the claim that MRI-EMS1r1 overestimates lightning NO_x emissions and underestimates convective updrafts in the tropics.

We revised now Page 13, Line 4 as below

“Meanwhile the strong tropical upper tropospheric O₃ biases in MRI-EMS1r1 are believed to be related to the weak tropical convective updraft and the large lightning NO_x emissions in the model. The model with weak updraft fails to bring enough low O₃ air from the surface to the upper troposphere in the tropics or overestimates the upper tropospheric mixing of stratospheric ozone-rich air. In addition, the global lightning NO_x (LNO_x) emission used in MRI-EMS1r1 is 10 TgN/yr. The best estimate of annual mean LNO_x based on satellite data assimilation is 6.3 TgN/yr (Miyazaki et al, 2014). The LNO_x in GEOSCCM is approximately 5 TgN/yr (Martini et al., 2011), which shows less tropical upper tropospheric O₃ bias compared to MRI-EMS1r1. Thus, the overestimation of the O₃ precursor in the upper troposphere is another reason for too much O₃. Figure. A1 shows the improvement in the radiative biases due to less O₃ bias in the experiment by half the LNO_x emissions in MRI-EMS1r1 (see the appendix).”

Appendix

Here we compare the MRI-EMS1r1 experiment run: RefC1_50%LNO_x with its RefC1 run. The new run’s emission decreases by about 50% compared with the original run. The global lightning NO_x emission annual mean in 2006 simulated in the experiment run is reduced from ~10.79 TgN/yr in RefC1 to ~5.21TgN/yr. The 10-year average changes from ~10.44 TgN/yr to ~5.18 TgN/yr.

We found the total flux bias is much reduced due to the improved O₃ radiative bias (Fig. A1 top two plots). The vertical resolved O₃ radiative bias shows as we expect, the strong overestimation of the tropical upper tropospheric O₃ radiative biases are much weaker in the new run (the middle two panels). This improvement is due to the less O₃ biases in this region due to reduced LNO_x emission (the bottom plots).

We also see some changes in the latitudinal distributions of the H₂O radiative bias. This is because the reduction of the upper tropospheric O₃ will cause by the model responses in the O₃ heating rate, which would have radiative effect on the temperature, atmospheric stabilities, and convective activity (e.g. Newack et al. 2015). All these would impact water vapor and cloud.

New references:

Miyazaki, K., Eskes, H. J., Sudo, K., and Zhang, C.: Global lightning NO_x production estimated by an assimilation of multiple satellite data sets, *Atmos. Chem. Phys.*, 14, 3277-3305, <https://doi.org/10.5194/acp-14-3277-2014>, 2014.

Martini, M., Allen, D. J., Pickering, K. E., Stenchikov, G. L., Richter, A., Hyer, E. J., & Loughner, C. P. (2011). The impact of North American anthropogenic emissions and lightning

on long-range transport of trace gases and their export from the continent during summers 2002 and 2004. *Journal of Geophysical Research*, 116, D07305.
<https://doi.org/10.1029/2010JD014305>

Nowack, P. J., Abraham, N. L., Maycock, A. C., Braesicke, P., Gregory, J. M., Joshi, M. M., Osprey, A., and Pyle, J. A.: A large ozone-circulation feedback and its implications for global warming assessments, *Nat. Clim. Chang.*, 5, 41–45, doi:10.1038/nclimate2451, 2015.
<https://www.nature.com/articles/nclimate2451>

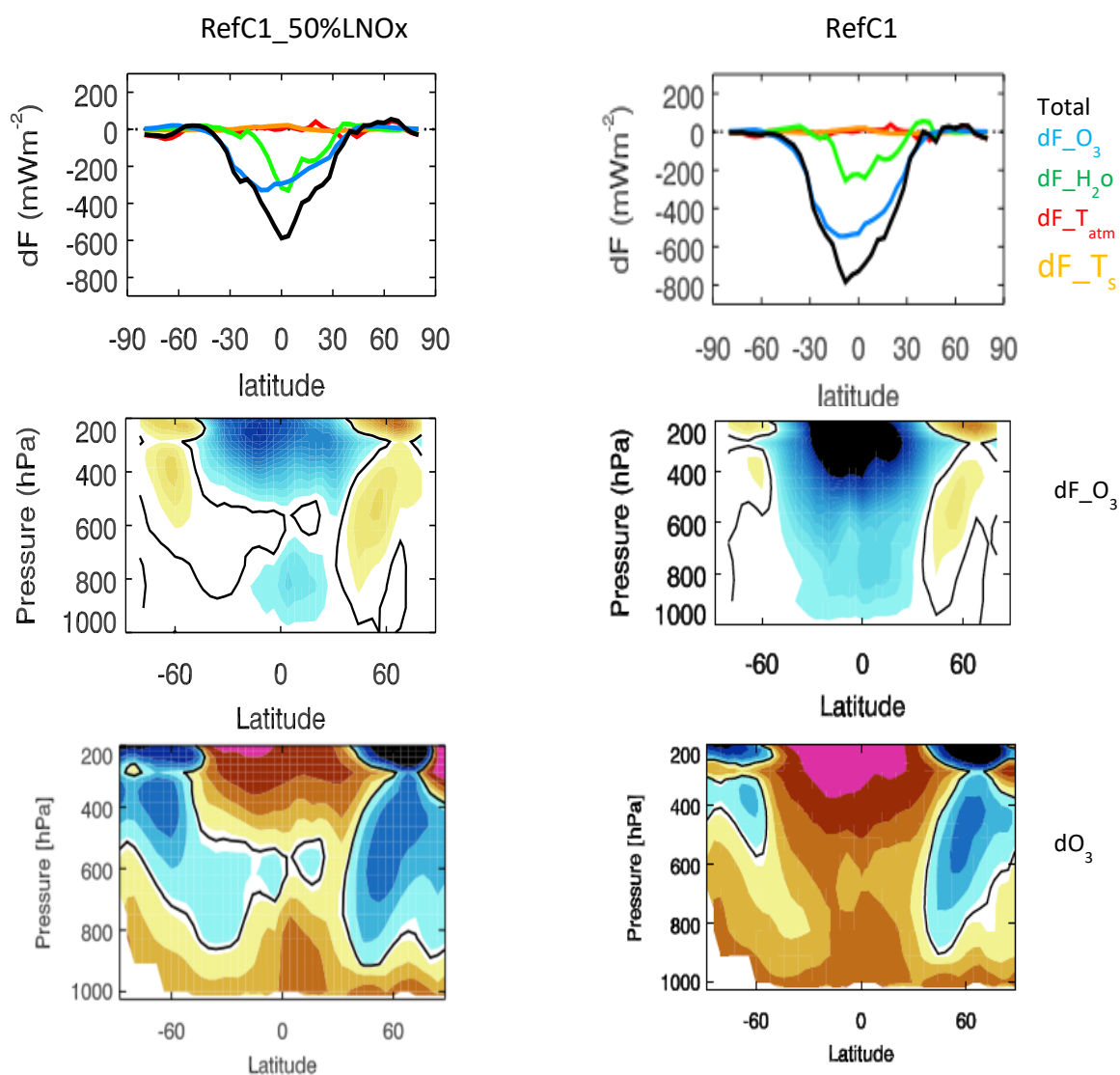


Fig. A1. The comparison of the MRI-ESM1r1 experiment of half LNOx in total flux bias (top row), O_3 radiative bias (middle row), and O_3 bias (bottom row). Left: new run with half LNOx. Right: RefC1 run. The black curves in the contour are the zero lines.

Page 12. Line 35. This reader does not understand “fact” 3, which gives this explanation for the bias in the radiative flux: “the systematic bias throughout the tropical troposphere, when propagate into the TOA flux, causes an accumulated bias in the radiative effect.”

We revised the statement now at Page 13 Line 19 as below

‘3) the simulations with the systematic bias throughout the tropical troposphere, when vertically integrated, accumulated into a larger column bias when compared to the models with vertically random biases.’

Page 13. Line 20. Why do the Ta biases in the tropics “shift between positive and negative vertically” in most models?

That means simulated air temperatures stay around the reanalysis profiles. These models better represent the air temperature than trace gases like H₂O and O₃.

We added the sentences in now Page 14, Line 7:

‘The oscillated T_a biases suggest that simulated air temperatures stay around the reanalyzed profiles. These models better represent the air temperature than the trace gases like H₂O and O₃.’

Page 14. Lines 6-35. The text should clarify what of value is learned in the analysis of how biases in 9.6 μ band affect biases in the entire OLR band.

We added some statements for the value of the understanding about how biases in 9.6 μ band affect biases in the entire OLR band. Now Page 15, line 31:

‘The anti-correlation between the biases in the 9.6-μm band and in the entire OLR band would suggest some bias drivers in the 9.6-μm band must play different roles at the other part of the OLR band. The further investigation of these processes would help to explain the radiative effect of different biases on the OLR estimations from models (Huang et al., 2008; Huang et al., 2014).’

New References:

Huang, X. L., W. Yang, N. G. Loeb, and V. Ramaswamy (2008), Spectrally resolved fluxes derived from collocated AIRS and CERES measurements and their application in model evaluation: Clear sky over the tropical oceans, *J. Geophys. Res.*, 113, *D09110*, doi:[10.1029/2007JD009219](https://doi.org/10.1029/2007JD009219).

Huang, X. L., Chen, X. H., Potter, G. L., Oreopoulos, L., Cole, J. N. S., Lee, D. M., & Loeb, N. G. (2014). A global climatology of outgoing longwave spectral cloud radiative effect and associated effective cloud properties. *Journal of Climate*, 27(19), 7475–7492. <https://doi.org/10.1175/JCLI-D-13-00663.1>

Pages 14-16. The conclusion should emphasize that the paper focuses only on clear-sky OLR.

We added the statement for the clear sky in the following sentence. Now Page 15, line 31

‘We have demonstrated a new method to quantitatively attribute the biases in O₃ band TOA flux from chemistry-climate model ensembles to O₃, H₂O, T_a, and T_s radiative components without cloud effect using observationally-constrained IRKs in the clear sky.’

Figure 4. The caption could say that the black curves are the same as the colored curves in Figure 3.

Added to the caption.

‘Figure 4. The attribution of the total TOA flux bias for each model to four dominant components and their latitudinal distribution. The black curves are the same as the colored curves in Figure 3.’

Figure 5. What do the black curves represent?

We add the words as below

‘Figure 5. Vertical resolved O₃ radiative bias. The black curves are the zero lines.

Figure 6. Vertical resolved H₂O radiative bias. The black curves are the zero lines.

Figure 7. Vertical resolved T_a radiative bias. The black curves are the zero lines.

Figure 9. The zonal averaged vertical-latitudinal distribution of O₃ model biases to the TCR-1 O₃ assimilation data. The black curves are the zero lines.

Figure 10. The zonal averaged vertical-latitudinal distribution of H₂O biases (model to the ERA reanalysis data). The black curves are the zero lines.

Figure 11. The zonal averaged vertical-latitudinal distribution of T_a biases from models to the reanalysis data. The black curves in the contour are the zero lines.’

Figure 13. Equations should be shown only for those correlations and slopes that are statistically significant.

OK. We removed the equations of the fitting lines with weak correlations.

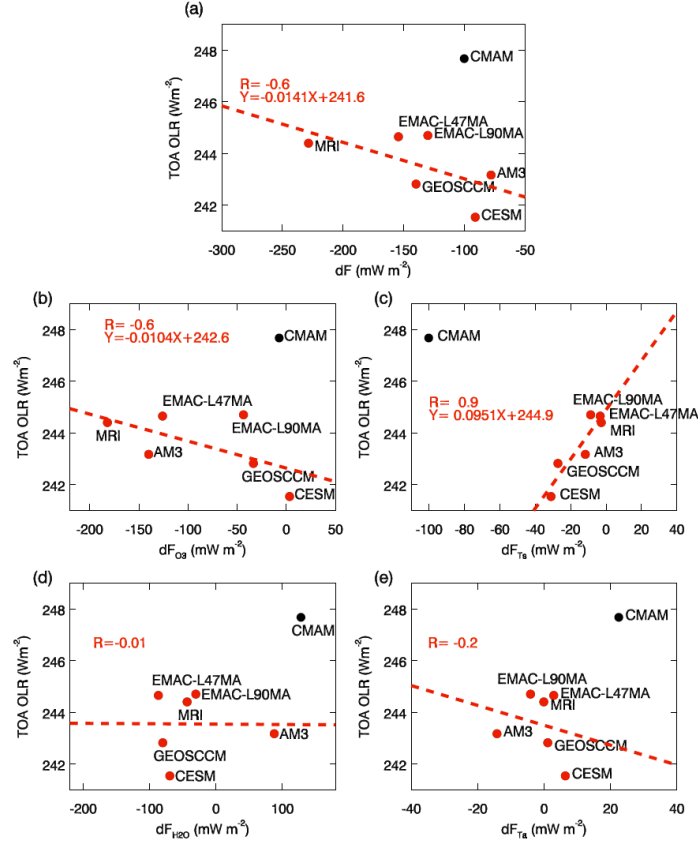


Figure 13. The correlation of the ozone band TOA flux biases to the model calculated broadband OLR (a) and the correlation of the attributed radiative components to the broadband OLR (b – e).

Table 3. Are the mixing ratios for ozone and water weighted by air density in each layer?

No. This is just the simple regional concentration average. We didn't compute the column bias by account for air density in each layer. The vertical weighting is considered while multiply with IRK and integral into tropospheric column.

Attribution of Chemistry-Climate Model Initiative (CCMI) ozone radiative flux bias from satellites

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Abstract

The top-of-atmosphere (TOA) outgoing longwave flux over the 9.6- μm ozone band is a fundamental quantity for understanding chemistry-climate coupling. However, observed TOA fluxes are hard to estimate as they exhibit considerable variability in space and time that depend on the distributions of clouds, ozone (O_3), water vapor (H_2O), air temperature (T_a), and surface temperature (T_s). Benchmarking present day fluxes and quantifying the relative influence of their drivers is the first step for estimating climate feedbacks from ozone radiative forcing and predicting radiative forcing evolution.

To that end, we constructed observational instantaneous radiative kernels (IRKs) under clear-sky conditions, representing the sensitivities of the TOA flux in the 9.6- μm ozone band to the vertical distribution of geophysical variables, including O_3 , H_2O , T_a , and T_s based upon the Aura Tropospheric Emission Spectrometer (TES) measurements. Applying these kernels to present-

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day simulations from the Chemistry-Climate Model Initiative (CCMI) project as compared to a 2006 reanalysis assimilating satellite observations, we show that the models have large differences in TOA flux, attributable to different geophysical variables. In particular, model simulations continue to diverge from observations in the tropics, as reported in previous studies of the Atmospheric Chemistry Climate Model Inter-comparison Project (ACCMIP) simulations. The principal culprits are tropical mid and upper tropospheric ozone followed by tropical lower tropospheric H₂O. Five models out of the eight studied here have TOA flux biases exceeding 100 mWm⁻² attributable to tropospheric ozone bias. Another set of five models *have* flux biases over 50 mWm⁻² due to H₂O. On the other hand, T_a radiative bias is negligible in all models (no more than 30 mWm⁻²). We found that AM3 and CMAM have the lowest TOA flux biases globally but are a result of cancellation of *opposite biases due to* difference processes. Overall, the multi-model ensemble mean bias is -13.3 ± 98 mWm⁻², indicating that they are too atmospherically opaque *due to trapping too much radiation in the atmosphere by overestimated tropical tropospheric O₃ and H₂O. Having too much O₃ and H₂O in the troposphere would have different impacts on the* sensitivity of TOA flux to O₃ and *these competing effects add more uncertainties on the* ozone radiative forcing. We find that the inter-model TOA *outgoing longwave radiation (OLR)* difference is well anti-correlated with their ozone band flux bias. This suggests that there is significant radiative compensation in the calculation of model outgoing longwave radiation.

1. Introduction

Tropospheric ozone (O₃) is the third important anthropogenic greenhouse gas (GHG) in terms of radiative forcing (RF) as a consequence of O₃ precursor and methane (CH₄) emission increases since pre-industrial *times to the* present day. Tropospheric O₃ *adjusted* RF ranges widely from +0.2 to +0.6 Wm⁻² computed from chemistry-climate model ensembles (IPCC AR5, 2013) (Bowman, et al., 2013;Stevenson, et al., 2013). The large uncertainty of the tropospheric O₃ RF is driven in part by the model responses to climate change. Without a good long-term record of the historical O₃ levels (Young, et al., 2017;Gaudel, et al., 2018), such estimates are highly dependent on the model assumptions of past O₃ levels. Differences between models in physical climate, chemical, and radiative processes conspire to complicate the assessment of the accuracy of these RF calculations. Consequently, a method to disentangle the key players caused the model differences to observations as well as the difference between the models is critical to robust estimates of chemistry-climate coupling.

About 80% of tropospheric O₃ RF is due to O₃ longwave absorption with the remaining 20% from the shortwave absorption (IPCC AR5, 2013). In the longwave, 97% of the total longwave absorption is in the 9.6-μm O₃ band (Rothman, et al., 1987). The global outgoing longwave radiation (OLR) spectra were first observed from space for a few months in 1970. Radiance observations were taken during April 1970 and January 1971 by *the* NASA Infrared Interferometric Spectrometer (IRIS) and then from October 1997 for 9 month by the Interferometric Monitor of Greenhouse Gases (IMG) instrument, on board the Japanese Advanced Earth Observing Satellite “Midori” (ADEOS) satellite. Harries, et al. (2001) showed that the changes in the greenhouse gas features between the observed spectra taken 30 years

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apart by these two instruments suggest increases in greenhouse gas forcing. Over the last two decades, a new generation of thermal infrared satellite instruments have provided a unique opportunity to continuously monitor the outgoing radiances covering the 9.6- μm O_3 band globally, such as NASA's Tropospheric Emission Spectrometer (TES) and Atmospheric Infrared Sounder (AIRS), ESA's Infrared Atmospheric Sounding Interferometer (IASI), and NOAA's Cross-track Infrared Sounder (CrIS). These valuable long-term global measurements can be used to derive the top-of-atmosphere (TOA) O_3 band flux and the sensitivity of the flux to the vertical distributions of O_3 , defined as instantaneous radiative kernels (IRKs) (Worden, et al., 2011; Doniki, et al., 2015).

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The TES observed global TOA outgoing fluxes at the 9.6- μm O_3 band in clear sky (Fig. 1) show strong geographic variations as a result of the short life-time of O_3 (Worden et al., 2011; Bowman et al., 2013). Consequently, the global O_3 GHG effect is more unevenly distributed than long-lived GHG, such as CO_2 . In addition, the variations of the TOA fluxes are not only highly dependent on the distributions of O_3 , but are also dependent on water vapor (H_2O), air temperature (T_a), and surface temperature (T_s) (Kuai et al., 2017).

There is an additional factor where the large-scale atmospheric structure sets the overall atmospheric opacity, which describes the fraction of the light that fails to pass through the atmosphere due to the absorption or scattering. For example, O_3 changes in more opaque regions, e.g., the Western Pacific, a wet region due to convection, result in a much smaller change in TOA flux than in more transparent regions, e.g., the Middle East, a dry region due to downwelling (Kuai, et al., 2017). This opacity has a direct impact on radiative forcing calculations.

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Chemistry-climate models diverge significantly in the simulation of these processes, which are difficult to disentangle because it is hard to quantify the response of the TOA flux due to the change in atmospheric opacity. In this study, we introduce a method to use observational based IRKs to quantitatively estimate the contributions of the model biases in O_3 , H_2O , T_a and T_s to the TOA flux biases.

In addition, the presence of clouds is the primary control on atmospheric opacity. Under the cloudy sky conditions, the roles of these variables other than cloud on TOA flux are much weaker. In addition, the variation in clouds could affect model estimates not only of the ozone but also of the flux sensitivity to ozone and other variables. Both ozone and sensitivity will impact the ozone radiative flux but in opposite directions. With cloud cover, the O_3 loss will be reduced. That means too much cloud would lead to more ozone production. The presence of the cloud would also cause weaker flux sensitivity to O_3 and other variables (IRKs). Therefore, the cloud effect is a battle between the impact on ozone estimation and the radiative sensitivity to ozone (IRK). The differences in cloud variations between the models will complicate the radiative effect. Furthermore, the study of the cloud effect is also currently limited by the global observations of total cloud cover and IRK product under realistic cloud conditions. Without knowing which models have better cloud cover, we benefit from using IRK based on the

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observed cloud free data by TES. Therefore, here we first try to access the role of O_3 , H_2O , T_a and T_s in the variation of the TOA flux without cloud effect.

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Worden et al., 2008 first attempted to disentangle these effects from satellites. They subsequently developed the IRK in Worden et al., (2011) for O_3 , which is used in this study as a powerful tool to attribute model variability. IRKs for O_3 represent the sensitivity of TOA fluxes to the vertical distributions of the observed O_3 . Aghedo, et al. (2011) applied the TES IRKs to evaluate the O_3 radiative effect of chemistry-climate models' O_3 biases in the Atmospheric Chemistry Climate Model Inter-comparison project (ACCMIP) (Lamarque, et al., 2013). Bowman et al. (2013) found model OLR bias due to O_3 is correlated with RF in the ACCMIP models. This correlation helped to reduce the inter-model divergence in RF by about 30% (Myhre, et al., 2013). Doniki et al. (2015), updated the IRKs' calculation with a more accurate, but computationally more complicated method, a five Gaussian Integration (GI) method, to replace the anisotropic approximation. They computed the O_3 IRKs with IASI observations and also showed that between the two methods there are about 20% differences in IRKs and about 20-25% differences in the Longwave Radiative Effect (LWRE). They also found that the day and night difference of LWRE is mainly controlled by the T_s change instead of O_3 amount change. Kuai et al., (2017) updated the computational method for the TES O_3 IRK product with the five GI method and revealed the hydrological controls on the global distribution of the O_3 GHG effect. The study showed that H_2O , T_a and T_s affect the O_3 IRK strength through relative humidity.

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Therefore, the TOA flux in the 9.6-micron band depends on more than O_3 . Consequently, in this study we expand the TES observation-based IRKs to other quantities, including H_2O profiles, T_a profiles, and T_s . We apply these IRKs to help understand the reasons for the model divergence in the TOA flux.

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The questions that have never been answered before include 1) How do the model-based flux and the flux sensitivity compare to the observational-based flux and sensitivity? 2) How do they compare between the models? 3) How do the flux biases in models relate to the RF variation? Thus, benchmarking present day O_3 band flux is the first step in answering all these questions, and would help to further understand the correlations between the bias in TOA flux and the bias in O_3 RF, and eventually improve the estimation of the climate feedbacks from O_3 forcing.

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To benchmark the model simulated geophysical quantities, a recently developed multi-species multi-satellite Tropospheric Chemistry Reanalysis (TCR) product (Miyazaki, et al., 2015) is used in this study to compare to the model results. This chemical reanalysis assimilates data from multiple satellites with sensitivity over complementary parts of the atmosphere, which provides better information than single-species chemical data assimilation. Satellite observations have the occasional issue of temporal discontinuity due to instrument performance and irregular spatial coverage, which can be circumvented by chemical data assimilation. Miyazaki et al., (2015) showed statistically the model error against independent aircraft and ozonesonde observations in the assimilated species, e.g. O_3 , NO_2 , and CO , is significantly reduced. The multi-species assimilation improves the Northern/Southern Hemisphere OH ratio and provides the

emission estimates with interannual variation. The comparison of O₃ reanalysis to the ACCMIP ensemble O₃ simulation in Miyazaki and Bowman (2017) quantified the model discrepancies in terms of seasonal amplitude, spatial variability, and inter-hemispheric gradient. For example, the ensemble mean is 6-11 ppb too high in the northern extra tropics, while up to 18 ppb too low in the southern tropics over the Atlantic in the lower troposphere. In this study, we use the same O₃ reanalysis data (Miyazaki and Bowman, 2017) to understand the model bias in the Chemistry-Climate Model Initiative (CCMI) project (Morgenstern et al., 2017), a follow up model inter-comparison study for ACCMIP. The multi-species assimilation also provides the opportunity to optimize the chemical related species of O₃ and the emission sources of the precursors simultaneously. Further work by Miyazaki, et al. (2017) showed that the surface emission of nitrogen oxides (NO_x) over a 10-year period (2005-2014) has a positive trend in regions including India, China, and the Middle East, but a negative trend over the USA, southern Africa, and western Europe. The global total emission stays almost constant between 2005 (47.9 Tg N yr⁻¹) and 2014 (47.5 Tg N yr⁻¹). Therefore, the O₃ reanalysis data from TCR represents the state-of-the-art for the current knowledge of the global distribution of tropospheric O₃ by combining the complementary information from model and satellite observations for O₃ and its precursors.

In this paper, we demonstrate a method to use the IRK products and the model biases relative to the reanalyzed tropospheric composition (O₃ and H₂O) and atmospheric state (T_s and T_a) to quantitatively attribute the radiative biases of the flux in a suite of CCMI models to these dominant components. The method and IRKs are described in section 2. The models and reanalysis data are introduced in the next section. Section 4 discusses the inter-comparison between models' flux biases, the bias attribution to the dominant components, and the geospatial distribution of the biases. Lastly, conclusion and future directions are summarized in section 5.

2. Instantaneous radiative kernels (IRKs) for the climate variables

The TOA flux in the 9.6-μm O₃ band (Fig. 1) is defined as

$$F_{TOA} = \int_v \int_0^{2\pi} \int_0^{\frac{\pi}{2}} L_{TOA}(v, \theta, \phi, q) \sin(\theta) \cos(\theta) d\theta d\phi dv \quad (1)$$

where v is the frequency, integrated over the O₃ band from 980 to 1080 cm⁻¹. $L_{TOA}(v, \theta, \phi, q)$ is the upwelling TOA radiance at frequency v , zenith angle θ and azimuth angle ϕ . We assume here that the radiance is symmetric in the azimuthal direction. The outgoing TOA radiances, L_{TOA} , are also a function of the atmospheric state, which is represented by variable ' q ', e.g. H₂O, O₃, and T_a, that is in turn a function of altitude, z .

The IRKs (Equation 2) represent the sensitivities of the TOA radiative flux in the 9.6-μm O₃ band to the changes in the vertical distribution of an atmospheric variable.

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$$\frac{\partial F_{TOA}}{\partial q(z_l)} = \int_v \int_0^{2\pi} \int_0^{\frac{\pi}{2}} \frac{\partial L(v, \theta, \phi, q)}{\partial q(z_l)} \sin(\theta) \cos(\theta) d\theta d\phi dv \quad (2)$$

Where z_l is altitude in discretized level l . When q represents the T_s , z_l becomes a single surface value at $l = 0$. The partial derivative term is the spectral radiance Jacobians calculated analytically by the TES radiative transfer model.

In this study, we expanded the TES global O_3 IRKs to IRKs with respect to H_2O , T_a , and T_s . The TOA flux sensitivities to H_2O or T_a still refer to the spectral window region in the $9.6\text{-}\mu\text{m}$ O_3 band for the flux. All the kernels are computed with the five-angle Gaussian Integration method (Doniki et al., 2015, Kuai et al., 2017). Figures 2 (a), (c), and (e) show examples of IRK profiles for O_3 , H_2O , and T_a for 2006. The TOA flux is most sensitive to each variable at very different vertical levels. The O_3 IRK peaks in the middle and upper troposphere (600 to 200 hPa), a higher level than the peaks in both H_2O and T_a IRKs. The mid and upper tropospheric O_3 near 500 hPa has the largest impact on the TOA flux change (close to $1 \text{ mWm}^{-2}\text{ppb}^{-1}$ in the tropics). The H_2O IRK peaks near 700 hPa, a little higher than the T_a IRK. The T_a IRK is maximal closest to the surface, suggesting that the O_3 band flux is most sensitive to boundary layer T_a near 900 hPa. The strength of the peaks all decrease with increasing latitude for all the three variables but the peak altitude does not change significantly except for the H_2O IRKs in the polar region, which peaks at a slightly higher level than in lower latitudes.

In addition, the T_s IRK is greater than zero, which means increases in T_s would increase the outgoing TOA flux. However, the IRKs for the GHGs, i.e. H_2O and O_3 , are negative, because the increase in gas concentrations reduces the upwelling flux at TOA due to radiative absorption by the gas.

The global vertical distribution of the zonal averaged kernels for O_3 , H_2O , and T_a are also shown below their profile plots in Fig. 2 (b), (d), and (f). The sensitivities of the TOA flux to these three variables are strongest in the tropics and decrease with latitude. Furthermore, the IRK for T_s is also shown in Fig. 2 (g) and (h). Unlike the other IRKs, the T_s IRK is not a function of altitude so we show the winter (Dec.-Feb.) and summer (Jun.-Aug.) seasonal average of its global distribution. The flux sensitivities are found to be largest over the major deserts, like the Sahara, Middle East, and Australia, corresponding to the regions with the highest values of T_s . We also notice that the values of the T_s IRKs in the Inter Tropical Convergence Zone (ITCZ) are much lower than in the subtropics, which suggests that the atmosphere opacity has an impact on the strength of the T_s IRKs.

3. A method to attribute the flux biases

The flux biases between observations and models under the clear-sky conditions can be described as:

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$$\delta F_{TOA}^i \approx \sum_{l \in L} \frac{\partial F_{TOA}^i}{\partial O_3^{i,l}} \delta O_3^{i,l} + \sum_{l \in L} \frac{\partial F_{TOA}^i}{\partial H_2O^{i,l}} \delta H_2O^{i,l} + \sum_{l \in L} \frac{\partial F_{TOA}^i}{\partial T_a^{i,l}} \delta T_a^{i,l} + \sum_{l \in L} \frac{\partial F_{TOA}^i}{\partial T_s^{i,l}} \delta T_s^{i,l} \quad (3)$$

where δF_{TOA}^i is the total TOA flux bias in the O_3 band at the i^{th} location. The four terms on the right hand side of the equation are the products of the IRKs and the biases in the geophysical quantities (i.e. O_3 , H_2O , T_a , and T_s). These biases are then vertically integrated on index, l , over the domain L , which in our case is the troposphere. The summation is the vertical integral from the surface to the tropopause.

Here we assume that the biases due to other physical processes, e.g., surface emissivity or other atmospheric species, have much less influences on the TOA flux variation. For example, the model bias in global emissivity is not accessible, but is believed to be quite small compared to O_3 , H_2O , T_a , and T_s . We also assume that the nonlinearity terms are much smaller than these four first order terms.

Following Bowman et al., (2013), the delta terms in Equation (3) are the model biases with respect to the reanalysis data, defined as below

$$\delta q^{i,l} = q_{mod}^{i,l} - q_{assim}^{i,l} \quad (4)$$

where $q_{mod}^{i,l}$ and $q_{assim}^{i,l}$ represents the model and reanalysis O_3 , H_2O , T_a , or T_s at the i^{th} location and the l^{th} altitude level respectively.

The mean flux bias or the mean bias components from tropospheric uncertainties are calculated from Equations (3) and (4) as

$$\delta F_q^j = \frac{1}{N_j} \sum_{i \in D_j} \sum_{l \in L} w_i \frac{\partial F_{TOA}^i}{\partial q^{i,l}} (q_{mod}^{i,l} - q_{assim}^{i,l}) \quad (5)$$

where w_i is area-weighted for the latitude bands, D_j is a set of observed locations, N_j is the number of locations in the domain of D_j and tropospheric levels up to the tropopause. We use the chemical tropopause $O_3 = 150$ ppb (Naik, et al., 2005; Hansen, et al., 2007; Bowman, et al., 2013; Kuai, et al., 2017). The domain of D_j can be zonal bands for the zonal mean or global area for the global mean, respectively. The global mean of the flux bias and its components will be denoted as $\overline{\delta F}$ and $\overline{\delta F_q}$, respectively.

4. Chemistry-climate models and the reanalysis data

4.1 Models and simulations

We analyze six models from the CCM1 study (Table 1) (Morgenstern, et al., 2017; Eyring, et al., 2013). It is a combined activity of the International Global Atmospheric Chemistry (IGAC) and

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Stratosphere-troposphere Processes And their Role in Climate (SPARC) (Randel, et al., 2004). The CCMI coordinates a number of model experiments that capture the variability and evolution of air quality, tropospheric chemistry, stratospheric O₃, and global climate. This approach builds on the legacy of previous chemistry-climate model inter-comparisons, such as the Chemistry-Climate Model Validation (CCM-Val; SPARC, 2010) (Eyring, et al., 2010) and the ACCMIP. In this study we use the experiment REF-C1, which is analogous to the REF-B1 experiment of CCMVal-2 (Table S30 in Morgenstern et al., 2017). REF-C1 require to use historic forcing and observed sea surface conditions. The models are free-running and simulate the recent past (1960–2010). We did not choose to use REF-C1SD (specified dynamics) because specified dynamics nudged the wind and temperature of the model to be constrained to the reanalysis data. The long-term climatological biases relative to the reanalysis between the models are minimized. Our study aims to find a correlation between the present day radiative bias and the RF from present day to future by the model predictions. Therefore, we prefer to keep the model differences in simulating longer-term climatology between their free runs.

We note that SOCOL3 and EMAC are both based on different versions of the ECHAM5 climate model. We also added two additional model simulations with AM3 from NOAA and CESM from NCAR. These two simulations are not the specific CCMI experiment run; however, these two models have been used in many studies, and including them in this study provides more useful information on the TOA flux diversity among the most recent models.

4.2 Tropospheric Chemistry Reanalysis (TCR-1) data

We computed the biases in the geophysical variables between the model and the reanalysis data. To compute the O₃ bias in models, we used the satellite-based O₃ reanalysis from multi-constituent multi-satellite data assimilation: Tropospheric Chemistry Reanalysis version 1 (TCR-1) (Miyazaki, et al., 2015; Miyazaki and Bowman, 2017) as the best synthesis of the observations. The reanalysis provides comprehensive spatiotemporal and multi-variable evaluation of model performance that complements direct comparisons against individual measurements, which may suffer from significant sampling bias (Miyazaki and Bowman, 2017).

TCR-1 assimilated multiple species data from multiple satellite products for the period from 2005 to 2017, e.g., combined TES and MLS observations for O₃, integrated OMI, SCIAMACHY and GOME-2 for tropospheric NO₂ column, MOPITT for CO, and MLS for HNO₃. TCR-1 used a global CTM MIROC-Chem (Watanabe, et al., 2011) as a forecast, which includes 92 species and 262 reactions. The model has 2.8° horizontal resolution with 32 vertical layers up to 4 hPa. The data assimilation was based on an ensemble Kalman filter with 32 ensemble members, which was used to simultaneously optimize concentrations and emissions of various species.

As summarized by Miyazaki and Bowman (2017), the mean bias in the reanalysis dataset against the WOUDC ozonesonde observations is from –3.9 to –2.9 ppb at the NH high latitudes (55°N –90°N); –0.9 to –0.1 ppb at the NH mid-latitudes (15°N –55°N); and –1.0 to –0.1 ppb at the SH mid-latitudes (55°S –15°S), between 850 and 500 hPa. On average, the bias is about 0.9

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ppb at the tropics and mid-latitudes between 500 and 200 hPa. These biases are much smaller than biased in the model simulation without data assimilation, demonstrating that the multi-satellite data assimilation provides comprehensive constraints on the entire tropospheric profile of O₃.

For the purpose of consistency, we also use outputs of H₂O, T_a, and T_s from the reanalysis to estimate the model biases. In the reanalysis calculation, meteorological fields simulated by the atmospheric general circulation model MIROC-AGCM (Watanabe et al., 2011) were nudged toward the 6-hourly ERA-Interim meteorological reanalysis (Dee et al., 2011) for zonal wind ($\tau = 1$ day) and temperature ($\tau = 3$ days) to reproduce past meteorological fields while simulating short-term (<six hours) meteorological variations, which were used to drive the CTM, as similarly employed in CCM1 C1SD simulations. Thus, the reanalysis dataset provides realistic and comprehensive estimates for both chemical and meteorological fields required for the TOA flux evaluations.

5. Results

5.1 The latitudinal distribution of the TOA flux bias

Figure 3 shows the latitudinal distribution of the zonal and annual mean of the TOA flux bias from each model relative to the reanalysis. The largest divergence between models is located at the tropics where most models underestimate the flux with the exception of CMAM. The low bias in the model ensemble implies the model atmosphere is more opaque than the chemical reanalysis leading to a 13.3 mWm⁻² outgoing flux reduction on average. The TOA flux in an opaque atmosphere is less sensitive to changes in tropospheric composition than a more transparent one. Under those conditions, the models would underestimate the radiative feedback from composition since the IRKs estimated under an opaque atmosphere will be weaker than those under a realistic (more transparent) atmosphere.

Two models that have larger low biases at the equatorial region than other models are SOCOL3 and MRI-ESM1r1. Their global means of the flux bias are more than -200 mWm⁻² (Table 2). The following analysis will help to clarify the source of the bias in the models.

5.2 Flux bias attribution

The total TOA flux bias is caused by biases from atmospheric composition and temperature. In order to determine the primary drivers of these biases, we apply the IRKs to the differences between model and the chemical reanalysis as described in Equation (3). Figure 4 shows the contribution of O₃ (blue), H₂O (green), T_s (red), and T_a (yellow) for each model to the total TOA flux bias (black). The global mean bias is summarized in Table 2.

In general, O₃ and H₂O are the two dominant drivers for most models where the large biases are concentrated in the tropics and subtropics. There are only three models (GEOSCCM, CMAM, and CESM) whose O₃ radiative biases (δF_{O_3} in Table 2) are less than 50 mWm⁻² and are almost

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negligible zonally. While the flux bias is better represented in these models it does not follow that they represent more accurately tropospheric O_3 as will be shown in the following section. The other five models (AM3, SOCOL3, EMAC-L47MA, EMAC-L90MA, and MRI-ESM1r1) have significant negative peaks at low latitudes (Fig. 3 and 4), actually resulting from their strong O_3 contributed biases (from 80 to 180 mWm^{-2} , numbers are highlighted in Table 2).

The TOA flux bias from H_2O is the second largest component for most models. Similar to O_3 , most models show the fluxes are biased low in the tropics due to the H_2O uncertainties with the exception of CMAM, which has the strongest global mean bias (127.9 mWm^{-2}). Note that, in the reanalysis, no data assimilation (or nudging) was applied for specific humidity. Watanabe et al (2011) demonstrated a dry bias in the lower troposphere and a wet bias in the middle and upper troposphere in MIROC-AGCM, primarily attributable to temperature biases. Nevertheless, the reported H_2O biases can be greatly reduced in the reanalysis, because of the nudging applied for temperature.

The flux bias due to T_a is found to be negligible in all models, which indicates that the model T_a estimates provide reasonable radiative fluxes, T_s radiative bias is also meridionally weak relative to the flux bias in O_3 and H_2O (Fig 4). With the exception of CMAM, the T_s ensemble global mean bias is less than 35 mWm^{-2} (see Table 2). Figure 4 suggests the strong bias from T_s in CMAM ($-100.2 mWm^{-2}$) comes from the two subtropical regions.

Interestingly, the positive flux bias due to H_2O (127.9 mWm^{-2}) is compensated by the negative flux bias due to T_s ($-100.2 mWm^{-2}$) in CMAM leading to the lowest global mean in δF (42.9 mWm^{-2} , calculated with Equation 5). This compensation is also true for AM3 but between a positive H_2O radiative bias (87.7 mWm^{-2}) and negative O_3 radiative bias ($-140 mWm^{-2}$). This analysis reveals that these two models are both right but for wrong—and opposite—reasons.

However, all the other models have a strong negative global mean bias and are mostly driven by the two major components (O_3 and H_2O). SOCOL3 and MRI-ESM1r1 are the two models that have the strongest low bias up to $-200 mWm^{-2}$, which is mainly due to their strong O_3 radiative bias ($-180 mWm^{-2}$). Their O_3 estimates are both biased high in the tropics and subtropics. We will show later that such bias is particularly strong in the upper troposphere.

5.3 Vertically-resolved radiative bias of the O_3 , H_2O and T

The zonal flux biases among the models are both significant and mainly in the tropics. However, those biases are the vertically integrated product of the model profile bias and the IRKs both with their own vertical structures. The vertically-resolved radiative bias can provide more insight into the processes leading to the biases. To further investigate, we examined the vertically-resolved flux bias for O_3 , H_2O and T_a (Fig. 5-7) and the global distribution for T_s (Fig. 8). These are computed from Equation 3 before the vertical summation. These figures show that the maximum contribution to the flux bias is a balance between the peak of the IRKs (Fig 1) and the peak of the geophysical quantities' bias (Fig. 9-12). The positive tropical O_3 radiative bias for GEOSCCM, CMAM, and CESM is commonly centered in the mid-troposphere corresponding to

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the peak of the IRKs (Fig. 5). On the other hand, the primary O₃ flux bias contribution in the tropics for SOCOL3, EMAC-L47MA, EMAC-L90MA, and MRI-ESM1r1 is in the upper troposphere around 200 hPa even though the IRKs are roughly half the peak sensitivity. These strong negative biases exceed 15 mWm⁻².

The strong tropical H₂O radiative bias collapses to shallower tropical regions below 400 hPa and is maximized near 800 hPa for most models exceeding 50 mWm⁻² (Fig. 6). CMAM has the unique and strongest net positive bias of above 50 mWm⁻² centered lower and close to 900 hPa. While most models flux bias is centered near 800 hPa, particularly, GEOSCCM, AM3, and CESM show a more vertically uniform—and opposing—flux bias.

Figure 7 indicates that tropical T_a radiative bias is largely negligible for vertical layers above 600 hPa as a consequence of the rapid decrease in sensitivity of the T_a IRKs. The maximum bias is in the lower troposphere between 900 hPa to surface. CMAM and CESM both show the strongest positive bias exceeding 10 mWm⁻² over most of the tropics. However, CESM has a compensating negative bias from 700-800 hPa that leads to a mean global bias of only 6.4 mWm⁻² (Table 2), whereas CMAM has a positive bias throughout, leading to an atmospheric T_a radiative bias of 22.5 mWm⁻², the largest of the models studied here.

Surprisingly, the model ensemble T_s turns out to be the 2nd largest contributor to the total bias (Table 2) instead of H₂O, driven primarily by 3 models: CMAM, CESM, and GEOSCCM as shown in Fig. 8. CMAM shows a negative bias that covers all of Africa, exceeding 500 mWm⁻², and Asia centered over India. Consequently, CMAM has the largest total bias (−100.2 ± 93.3 mWm⁻²). CESM and GEOSCCM T_s radiative biases, on the other hand, are centered at high latitudes in the Western Hemisphere over the Eastern US and Canada exceeding 300 mWm⁻².

The vertically and spatially concentrated radiative biases provide clues as to what processes are the most important for the total flux bias. These processes drive the distribution of the constituents, which we will discuss in detail in the next sections.

5.4 The spatial source of TOA flux bias

The source of the attributed flux biases can be traced back to their spatial origins, which can provide more insight into the underlying processes and the differences between the models.

5.4.1 O₃ bias

Figure 9 shows a vertically resolved zonal averaged distribution of O₃ biases between the model and the chemical reanalysis similar to that in Fig. 5. Three models (GEOSCCM, CMAM, and CESM) have the weakest globally averaged O₃ radiative bias reported in Table 2 (−33.5 mWm⁻², −7.3 mWm⁻², and 3.3 mWm⁻²). These three models also have the lowest O₃ bias in tropical troposphere on average (−1.1 ppb, −1.3 ppb, and 3.0 ppb, reported in Table 3) and as a consequence have weaker radiative bias in the region with the strongest O₃ IRK globally (0.9

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mWm⁻², 1.4 mWm⁻², and 2.1 mWm⁻², reported in Table 4). On the other hand, the O₃ bias is greater than 7 ppb for all the other models and results in a large O₃ radiative bias in the tropics, especially SOCOL3 (13.4 ppb in the tropical O₃ bias and -9.2 mWm⁻²; see Table 3 and Table 4) and MRI-ESM1r1 (13.7 ppb and -10 mWm⁻²).

GEOSCCM, CMAM, and CESM also commonly have a vertically compensated pattern in the tropics that biased high in the upper troposphere while biased low in the middle and lower troposphere (Fig. 9). Their O₃ low biases in the middle troposphere are approximately 5 to 10 ppb, where the peak of the IRK centered, but the high biases in the upper troposphere are about 5 ppb. Such a high-low pattern leads to compensation during the vertical integration through the troposphere into the radiative effect at the top of the atmosphere. The corresponding vertical resolved O₃ radiative bias for these three models in Fig. 5 shows the consistent tropical vertical distribution but in an opposite sign since the O₃ IRK is negative. In contrast, the other five models have vertically systematic biases high in the tropical O₃, and the biases increase from the middle troposphere to the upper troposphere. Especially SOCOL3 and MRI-ESM1r1 strongly overestimate O₃ by more than 20 ppb in a wide region of tropical upper troposphere. The O₃ radiative biases in this region remain significantly high, stronger than -15 mWm⁻², causing these two models to have the highest O₃ radiative biases in the global and annual mean (both about -183 mWm⁻²).

The systematic bias in the entire tropical tropospheric O₃ and strong overestimation of upper troposphere in SOCOL3 and MRI-ESM1r1 could be caused by several factors. For example, the transport from the lower stratosphere could be too high. Alternatively, precursor emissions of tropospheric O₃ could also be too high. The analysis with the spatially explicit biases provides important clues to implicate the specific processes that individual modeling groups can investigate.

The GEOSCCM has been used to study the tropospheric O₃ response to variations in the El Niño-Southern Oscillation (ENSO) where (Oman, et al., 2011;2013) compared the model to satellite observations. These regular comparisons may have led to the improved simulation of tropospheric O₃ profiles and consequently lower vertical O₃ bias. The GEOSCCM model in the CCMI study uses the tropospheric/stratospheric chemical package developed within the Global Modeling Initiative (GMI) program (Duncan, et al., 2007), which has more realistic ozone chemistry, an internally generated quasi-biennial oscillation, an improved air/sea roughness parameterization and other improvements (Oman and Douglass, 2014).

Nielsen, et al. (2017) showed that GEOSCCM successfully reproduces the changes in the quasi-global (60°S–60°N) annual-mean trend in total O₃ column since 1960s to the present day. For the present-day atmosphere, simulated tropospheric partial column O₃ from GEOSCCM Ref-C1 for CCMI was compared to satellite observations of OMI and MLS (Ziemke, et al., 2011). The differences are mostly a few Browner Dobson (DU) except the Northern Hemisphere subtropics and middle latitudes in autumn and winter with the 4–6 DU bias which are under investigation.

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The finding that SOCOL3 and MRI-EMS1r1 both have strong overestimates in the tropical upper troposphere is also understandable. SOCOL3 is the third generation of the coupled chemistry-climate model (CCM) SOCOL (modeling tools for studies of Solar Climate Ozone Links). Several steps have been taken to improve the SOCOL model simulation of O₃. Stenke et al., 2013 first attempted to reduce the O₃ bias in their middle-atmosphere by updating their middle-atmosphere general circulation with an advanced advection scheme. Revell, et al. (2015) revealed that ozone precursor emissions are the biggest players that control the global-mean change in tropospheric ozone. In a parallel study, Revell, et al., 2018 developed an updated version of "SOCOL3.0", "SOCOL3.1", to reduce the tropospheric ozone bias. By improving the treatment of ozone sink processes, the tropospheric column ozone bias in "SOCOLv3.1" is reduced up to 8 DU, mostly due to the inclusion of N₂O₅ hydrolysis on tropospheric aerosols. We expect that the future similar analysis with the SOCOL3.1 could show a reduced flux bias for this model.

Meanwhile the strong tropical upper tropospheric O₃ biases in MRI-EMS1r1 are believed to be related to the weak tropical convective updraft and the large lightning NO_x emissions in the model. The model with weak updraft fails to bring enough low O₃ air from the surface to the upper troposphere in the tropics or overestimates the upper tropospheric mixing of stratospheric ozone-rich air. In addition, the global lightning NO_x (LNO_x) emission used in MRI-EMS1r1 is 10 TgN/yr. The best estimate of annual mean LNO_x based on satellite data assimilation is 6.3 TgN/yr (Miyazaki, et al., 2014). The LNO_x in GEOSCCM is approximately 5 TgN/yr (Martini, et al., 2011), which shows less tropical upper tropospheric O₃ bias compared to MRI-EMS1r1. Thus, the overestimation of the O₃ precursor in the upper troposphere is another reason for too much O₃. Figure A1 shows the improvement in the radiative biases due to less O₃ bias in the experiment by half the LNO_x emissions in MRI-EMS1r1 (see the appendix).

In summary, the potential reasons for the prevalence of O₃ radiative bias in tropical mid and upper troposphere in the models could be due to following facts: (1) the tropical O₃ IRK is strongest in this region (Fig. 2); (2) the largest O₃ bias in the models also centered in the same place (e.g. SOCOL3 and MRI-EMS1r1, Fig. 9); 3) the simulations with the systematic bias throughout the tropical troposphere, when vertically integrated, accumulated into a larger column bias when compared to the models with vertically random biases.

5.4.2 H₂O bias

H₂O turns out to be the primary contributor for three models (GEOSCCM, CMAM, and CESM) since their O₃ radiative bias is small. It is also the second dominant driver after O₃ in the other five models. Different from O₃, H₂O IRKs in Fig. 2 show the strongest sensitivity to the tropical lower troposphere centered at 800 hPa, where H₂O is most concentrated globally. We found the model biases in H₂O are strongest in the tropical lower troposphere. It explains why the strongest radiative bias from H₂O is also located in the tropical region near 800 hPa in all models as shown in Fig. 6. Figure 10 and Table 3 further help to indicate that H₂O is biased low

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only in two models, AM3 (−586.5 ppm) and CMAM (−506.6 ppm). We note that H₂O IRKs are also negative as O₃. Therefore, these two models have the unique overestimates in H₂O radiative bias at low latitudes (see Fig. 4) while all the other models are predominantly biased high in tropical H₂O concentrations, which result in the negative radiative biases.

5.4.3 T_a bias

We found the T_a radiative biases in these model ensembles are all negligible. There are two reasons. One is that the T_a biases are small overall (less than 2 K) even at the tropical lower troposphere (below 1 K on average in Table 3). The other reason is that the compensation in the vertical integration helps to reduce the radiative bias at the top of atmosphere.

Figure 11 shows that the model biases in T_a range within ± 2 K for all the models because the current chemistry-climate models have been well-developed to simulate the global atmospheric T_a fields relative to reanalysis. The region with strongest sensitivity, identified by the T_a IRKs (Fig. 2), is the tropical lower troposphere (the region within $\pm 30^\circ$ and below 800 hPa). The T_a biases in the tropics shift between positive and negative vertically in most models except CMAM, which is systematically biased (Fig. 11). The oscillated T_a biases suggest that simulated air temperatures stay around the reanalyzed profiles. These models better represent the air temperature than the trace gases like H₂O and O₃. The oscillation around the reanalyzed profile leads to vertical compensation in the air T_a radiative bias. Therefore, the flux bias from T_a is a small component compared to the radiative bias from O₃ and H₂O. Figure 4 suggests the only model that has a small tropical peak in the T_a radiative component is CMAM, which has the strongest T_a radiative bias (22.5 ± 40.5 mWm^{−2} in Table 2) among all the models. Figure 11 shows that this model has a deep region with strong bias of about 2 K at tropical and also persistently overestimated T_a vertically. Figure 7 further suggests the strong radiative bias in CMAM mainly comes from the tropical lower troposphere (>10 mWm^{−2} below 800 hPa). While the other models have vertical compensation in the tropics (less than 0.5 K bias on average, see Table 3) and therefore they are less biased in TOA flux (less than 1 mWm^{−2} in Table 4). The two EMAC models both have strong biases at the southern high latitude but still have weak radiative effect at this region (see Fig. 7) due to much weaker T_a IRK at high latitudes.

5.4.4 T_s bias

The global distribution of the T_a biases indicates that the biases in Sea Surface Temperature (SST) are smaller than the biases in land T_s for all the models (Fig. 12) because the CCMI experiment (REF-C1) selected in this study used the observed SST. CMAM is the model that has the strongest T_s radiative bias (-100.2 ± 93.3 mWm^{−2} in Table 2) which peaks in both sub-tropical regions (Fig. 4). These large negative biases are due to the large underestimates of the T_s over the major deserts, e.g., the Sahara, Middle East and Australia (Fig. 12). In other words, the real deserts' surface is hotter than the model's prediction. At the same time, the T_s IRKs at the subtropical desert region are also strongest globally since the TOA flux is more sensitive to T_a when the atmosphere is transparent, which is due to the downdraft of the Hadley cell control the region (Kuai et al., 2017). The downwelling airflow results in less precipitation and less

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cloud as well as higher T_s during summer over the desert surface. These factors cause the CMAM to have the largest T_s radiative bias compare to all the other models.

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In contrast, two EMAC models and MRI-ESM1r1 also have strong high bias in T_s at Siberia (> 4 K in Fig. 12) but the radiative bias is much less significant compared with the Middle East in CMAM in Fig. 8. The T_s IRKs are weaker during the winter season at high latitudes than the low latitudes if the T_s is low (Fig. 2). However, the IRKs at the subtropical desert region stay strong during winter. Therefore, the annual mean of the T_s radiative bias is much weaker at Siberia in two EMAC models and MRI-ESM1r1 than the Middle East region in CMAM. Consequently, the global annual means of the T_a radiative biases for two EMAC models and MRI-ESM1r1 are small although the large biases in T_s are found in their Siberia region.

6. Correlation to the broadband OLR

The analysis up to this point has been limited to the 9.6 micron band. We posed the question as to whether biases in this band could provide any insight into biases in the entire OLR band. To that end, we found an anti-correlation ($R = -0.6$) between the global mean of the O_3 band flux biases and the clear sky broadband OLR calculated internally by the models as shown in Fig. 13 (a). The CMAM OLR is inconsistent with the ensemble (more than 2 Wm^{-2} higher than all the other models) and therefore it is excluded in the correlation. Interestingly, a very similar regression line and anti-correlation coefficient ($R = -0.6$) are found between the O_3 radiative bias and the broadband OLR (Fig. 13 b). The similar regression line indicates that the O_3 radiative bias dominates the 9.6-micron TOA flux distribution, which is confirmed by the attribution analysis that O_3 radiative bias is the largest term in five of eight models. The anti-correlation suggests a radiative compensation between the 9.6-micron band and the other parts of the OLR assuming a constant globally integrated OLR at TOA. More interestingly, a strong correlation ($R = 0.9$) is found between T_s radiative bias and broadband OLR (Fig. 13 [c]) because the T_s affects the entire baseline of the outgoing radiance and its radiative effect plays the same role in the O_3 band as in the entire OLR. However, there is no significant correlation found between T_a radiative bias and OLR, likely because there is no coherent bias in T_a radiative effect between the O_3 band and in the entire OLR. There is neither a correlation between the H_2O radiative bias and broadband OLR. H_2O absorption is ubiquitous in the OLR. Consequently, biases in the 9.6-micron band do not drive the magnitude of the overall H_2O absorption in spite of the H_2O biases.

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The anti-correlation between the biases in the 9.6- μm band and in the entire OLR band would suggest some bias drivers in the 9.6- μm band must play different roles at the other part of the OLR band. The further investigation of these processes would help to explain the radiative effect of different biases on the OLR estimations from models (Huang, et al., 2008)(Huang, et al., 2014).

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7. Conclusions

We have demonstrated a new method to quantitatively attribute the biases in O₃ band TOA flux from chemistry-climate model ensembles to O₃, H₂O, T_a, and T_s radiative components without cloud effect using observationally-constrained IRKs in the clear sky. The study also provides the first vertically and globally resolved view of the radiative bias for each component. An IRK depicts the sensitivity of TOA fluxes to the vertical distribution of the geophysical quantities, such as O₃, H₂O, T_a, and T_s. While the products of 9.6-μm O₃ band IRK for O₃, H₂O, T_a, and T_s have been developed with the satellite observations by Aura TES, the record could be extended by MetOP-IASI and SNPP-CrIS Fourier Transform Spectrometer (FTS) measurements. We compute the model biases against reanalysis data for four key variables: O₃, H₂O, T_a, and T_s. Especially for O₃ biases, the newly developed TCR-1 O₃ assimilation data (Miyazaki et al., 2015; Miyazaki and Bowman 2017) are, for the first time, used as the state-of-the-art benchmark for tropospheric O₃ in models. These specific bias comparisons for the CCMI study cause the modelers to investigate the reasons for these biases and motivate them to improve their simulations. For example, MRI-ESM1r1 shows the reduced LNOx emission help to improve their tropical upper tropospheric O₃ and its radiative bias.

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O₃ abundance is found to be the dominant driver for the ensemble flux bias. Tropical tropospheric O₃ is too high for most models and accounts for about 70% of the flux bias (Table 2). The second driver in the model ensemble becomes the T_s instead of H₂O because the T_s radiative components are commonly biased low in the model ensemble while the H₂O radiative biases between models are biased randomly in both directions with large diversity. For individual models, however, H₂O is the second most important driver, a larger component than T_s, for many cases, such as AM3, SOCOL3, and MRI-ESM1r1.

In addition to determining that the tropospheric O₃ and H₂O are overestimated, and the surface is too cold, the study also tells us the geolocations, in latitudes and altitudes, of the deviations in these geophysical quantities that propagate into the flux bias.

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The largest spread of the flux bias between the models is found in the tropics. The principal contributors governing each model are different and controlled by different processes over different regions. The flux biases in five of the eight models (AM3, SOCOL3, EMAC-L47MA, EMAC-L90MA, and MRI-ESM1r1) are primary driven by too much O₃ in the tropical middle and upper troposphere. H₂O is a big driver in five models (AM3, SOCOL3, GEOSCCM, CMAM, and CESM). T_s is an important contributor in CMAM in addition to its H₂O.

Although AM3 and CMAM overall have relative lower TOA flux biases globally, we found they are actually right for wrong reasons. In AM3, the dominant positive H₂O radiative bias (87.7 mWm⁻² in Table 2) happens to be cancelled by the dominant negative O₃ component (−140 mWm⁻²). While in CMAM, the large positive H₂O component (127.9 mWm⁻²) is mostly be compensated by T_s radiative bias (−100.2 mWm⁻²). The two relatively young models among the model ensembles, SOCOL3 and MRI-ESM1r1, have a large potential to be improved for their fluxes by reducing their strong negative radiative biases from both tropical upper tropospheric O₃ and tropical lower tropospheric H₂O.

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On average, the model ensemble underestimates the flux by about 133 mWm⁻² due to overestimated tropical tropospheric O₃ and H₂O. The underestimate of the TOA flux implies the model atmosphere is too opaque. In a more opaque atmosphere, the change in flux will be weaker for the same change in tropospheric O₃ because the sensitivity (i.e., IRKs) is weaker.

5 With such feedback, the O₃ RF, the changes in O₃ GHG effect from pre-industrial times to the present day, would likely be underestimated. The opacity of the atmosphere is controlled by climate processes, such as the hydrological cycle, that is showed can indirectly affect the O₃ GHG effect and RF, as discussed in Kuai et al., (2017).

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10 The spatially explicit and process-focused differences could be used as a basis for emergent constraints (Bowman et al, 2013). New techniques such as hierarchical emergent constraints (HEC) can harness this spatial information so that specific processes affecting O₃ RF can be identified (Bowman, et al., 2018). Moreover, if this correlation exists between the TOA flux bias and the O₃ RF, then the similar issue could be found in the RF of other GHGs, such as CO₂ and CH₄. That is a subject for future research.

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Finally, although the chemical reanalysis dataset provides comprehensive information on model radiative biases, we need to understand its performance. For instance, further improvements are still needed for lower tropospheric O₃ (Miyazaki and Bowman 2017). Ingesting more datasets and applying a bias correction procedure would be useful to improve reanalysis accuracy. The lower tropospheric O₃ analysis would benefit from the recently developed satellite retrievals with high sensitivity to the lower troposphere (Fu, et al., 2018) and the optimization of additional precursor emissions.

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25 Author contributions

LK and KB designed the analysis and organized the paper. LK developed the IRK product, performed all the analysis, and drafted the paper. KB helped interpret the results. HW and SK provided the code used to further develop the new IRK products. KM contributed the TCR-1 data. The other authors ran the individual model, contributed the model output, and helped revise the paper.

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providing the HPC and data archiving resources for the consortial project ESCiMo (Earth System Chemistry integrated Modelling). The GEOSCCM is supported by the NASA MAP program and the high-performance computing resources were provided by the NASA Center for Climate Simulation (NCCS). The coauthor, ER, is partially supported by the Swiss National Science Foundation under grant 200020_182239 (POLE) and the information gained will be used to improve next versions of the CCM SOCOL.

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Appendix

Here we compare the MRI-EMS1r1 experiment run: RefC1_50%LNOx with its RefC1 run. The new run's emission decreases by about 50% compared with the original run. The global lightning NOx emission annual mean in 2006 simulated in the experiment run is reduced from ~10.79 TgN/yr in RefC1 to ~5.21 TgN/yr. The 10-year average changes from ~10.44 TgN/yr to ~5.18 TgN/yr.

We found the total flux bias is much reduced due to the improved O₃ radiative bias (Fig. A1 top two plots). As we expected, the vertical resolved O₃ radiative bias shows that the overestimation of the tropical upper tropospheric O₃ radiative bias is much weaker in the new run (the middle two panels). This improvement is due to the lower O₃ biases in this region caused by reduced LNOx emission (the bottom plots).

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We also see some changes in the latitudinal distributions of the H₂O radiative bias. This is because the reduction of the upper tropospheric O₃ will cause the model responses in the O₃ heating rate, which would have radiative effect on the temperature, atmospheric stabilities, and convective activity (Nowack, et al., 2015). All these factors would impact water vapor and cloud formation.

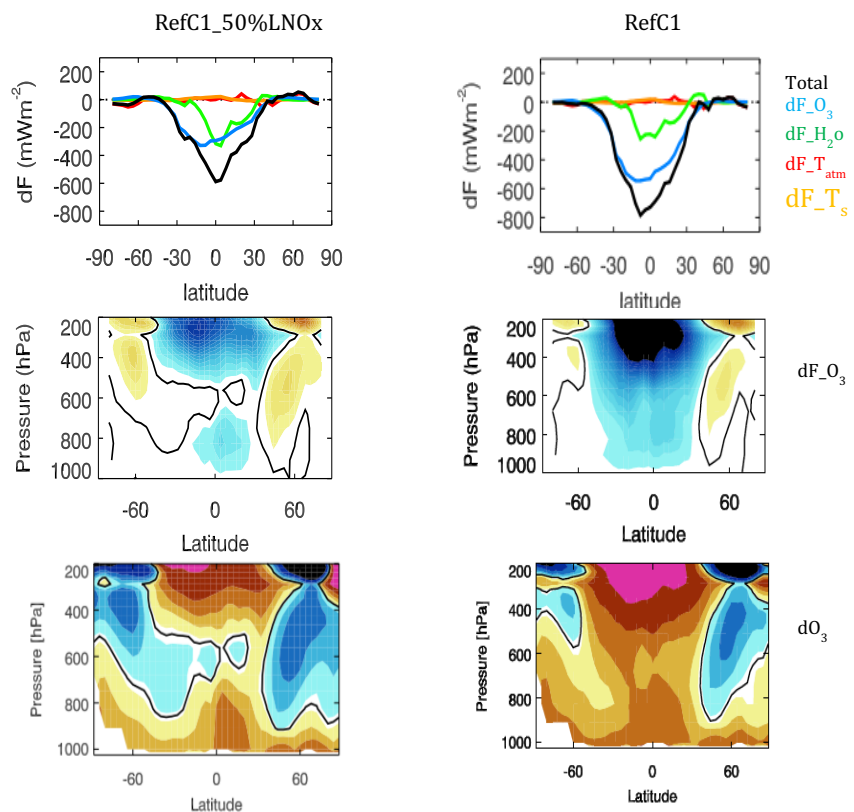


Fig. A1. The comparison of the MRI-ESM1r1 experiment of half LNOx in total flux bias (top row), O₃ radiative bias (middle row), and O₃ bias (bottom row). Left: new run with half LNOx. Right: RefC1 run.

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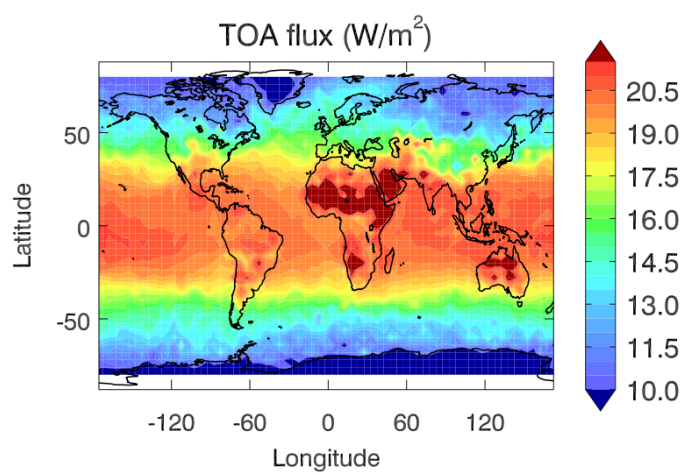


Figure 1. The clear sky TES observed TOA flux at 9.6- μm O_3 band, annually averaged in 2006.

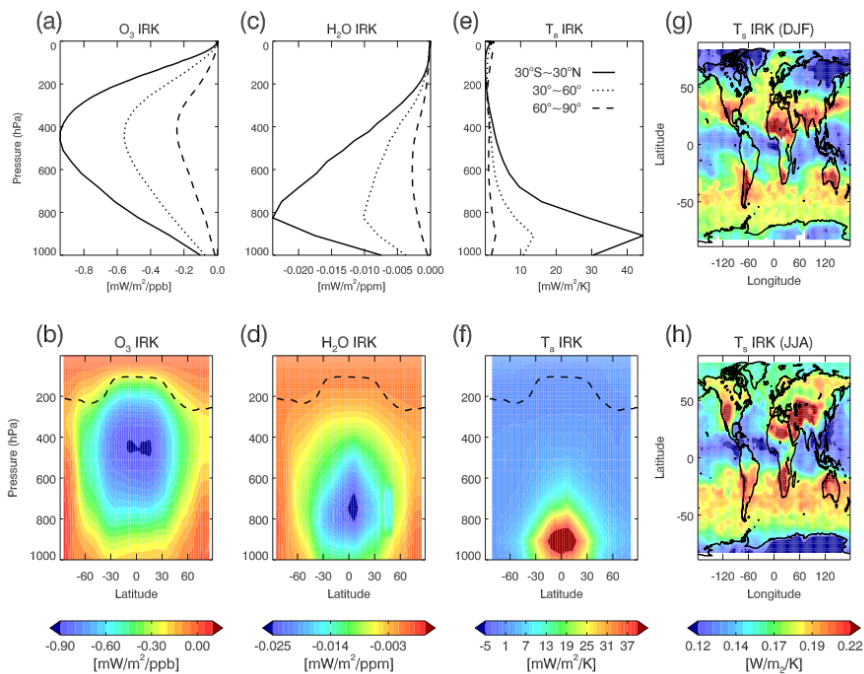


Figure 2. TES 2006 IRK for four primary components (O_3 , H_2O , T_a , and T_s). Figures (a), (c), and (e) are IRK of latitudinal band averages in the tropics ($30^\circ\text{S}\sim 30^\circ\text{N}$), mid-latitudes of both hemispheres ($30^\circ\sim 60^\circ$), and high-latitudes of both hemispheres ($60^\circ\sim 90^\circ$). The figures below them are the pole-to-pole vertical distribution of the zonally averaged IRK. The global distribution of IRK for T_s is plotted for winter season (December to February) in (g) and summer season (June to August) in (h).

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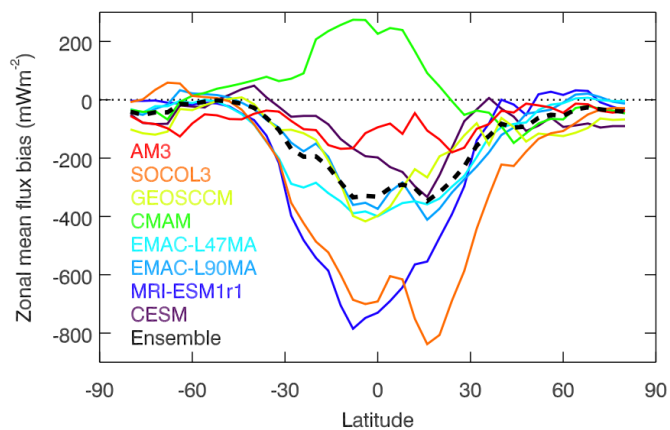


Figure 3. The latitudinal distribution of the zonal flux bias (model – reanalysis) with latitude weight.

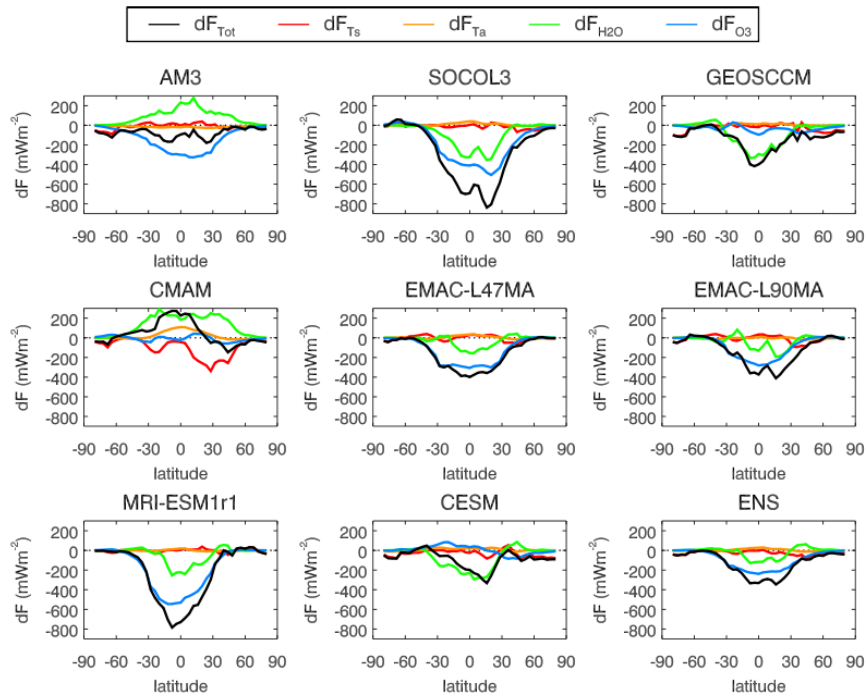


Figure 4. The attribution of the total TOA flux bias for each model to four dominant components and their latitudinal distribution. [The black curves are the same as the colored curves in Figure 3.](#)

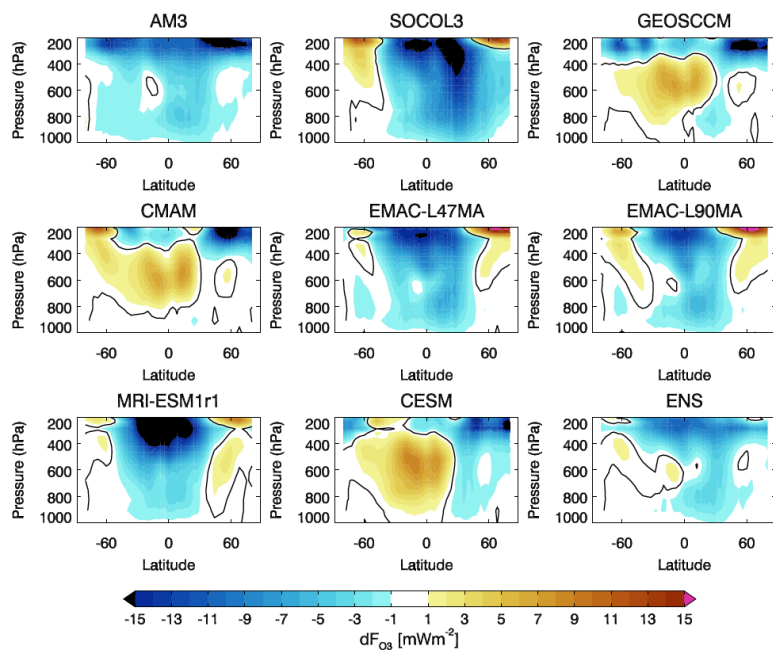


Figure 5. Vertical resolved O_3 radiative bias. The black curves are the zero lines.

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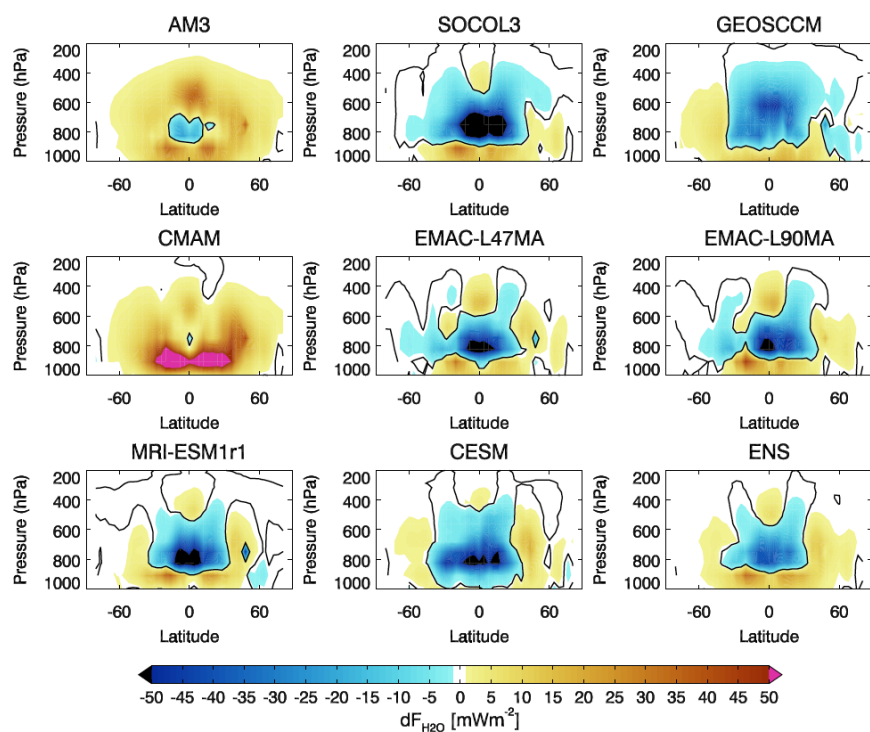


Figure 6. Vertical resolved H_2O radiative bias. The black curves are the zero lines.

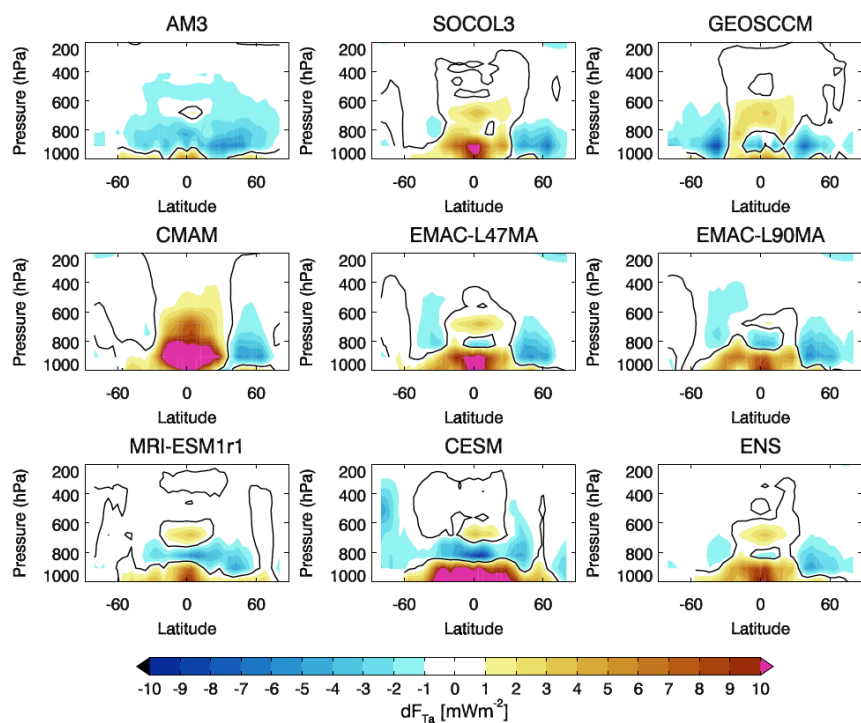


Figure 7. Vertical resolved T_a radiative bias. The black curves are the zero lines.

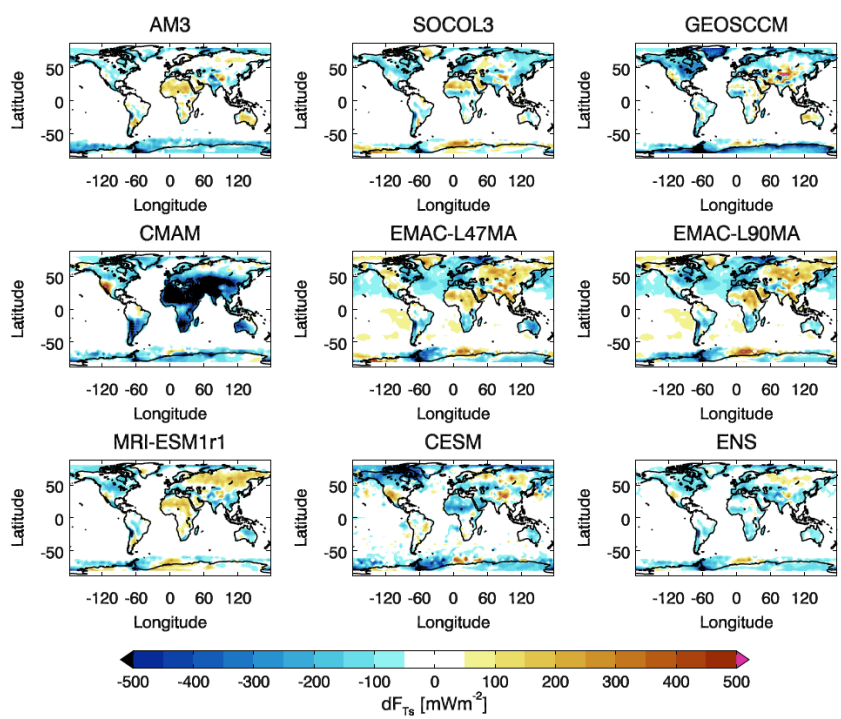


Figure 8. Global distribution of the T_s radiative bias.

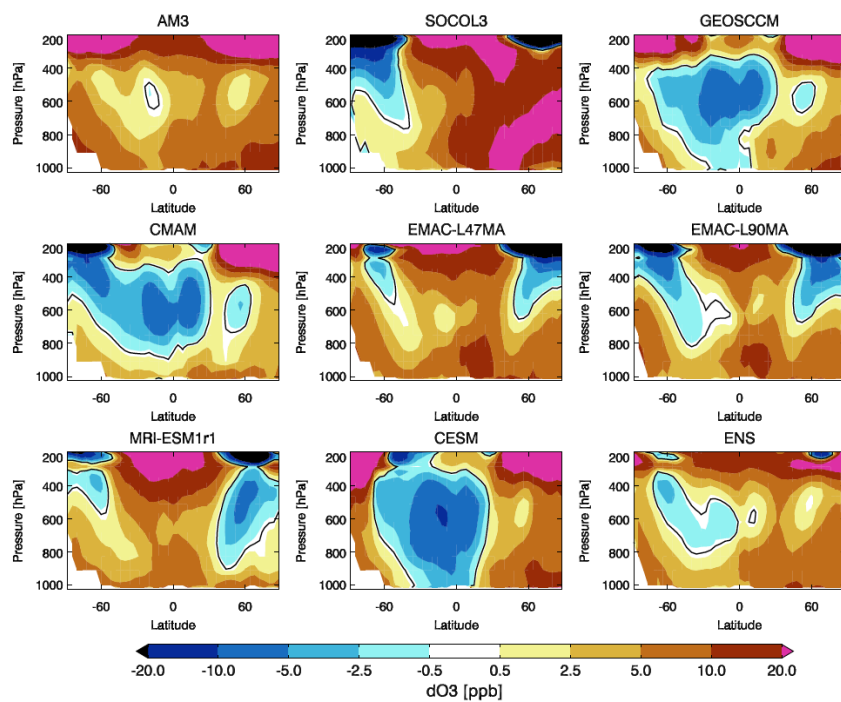


Figure 9. The zonal averaged vertical-latitude distribution of O₃ model biases to the TCR-1 O₃ assimilation data. *The black curves are the zero lines.*

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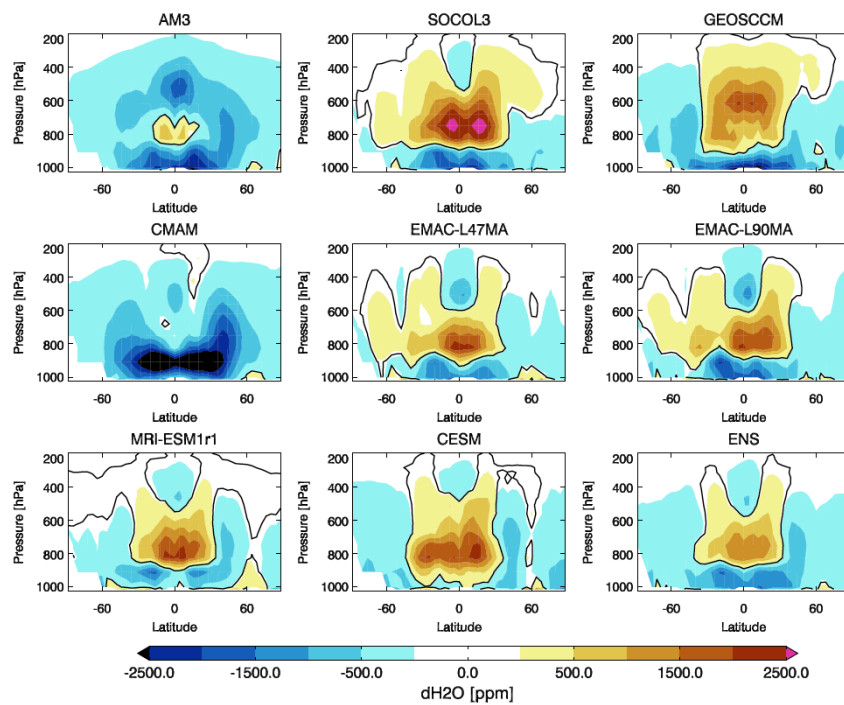


Figure 10. The zonal averaged vertical-latitude distribution of H_2O biases (model to the ERA reanalysis data). The black curves are the zero lines.

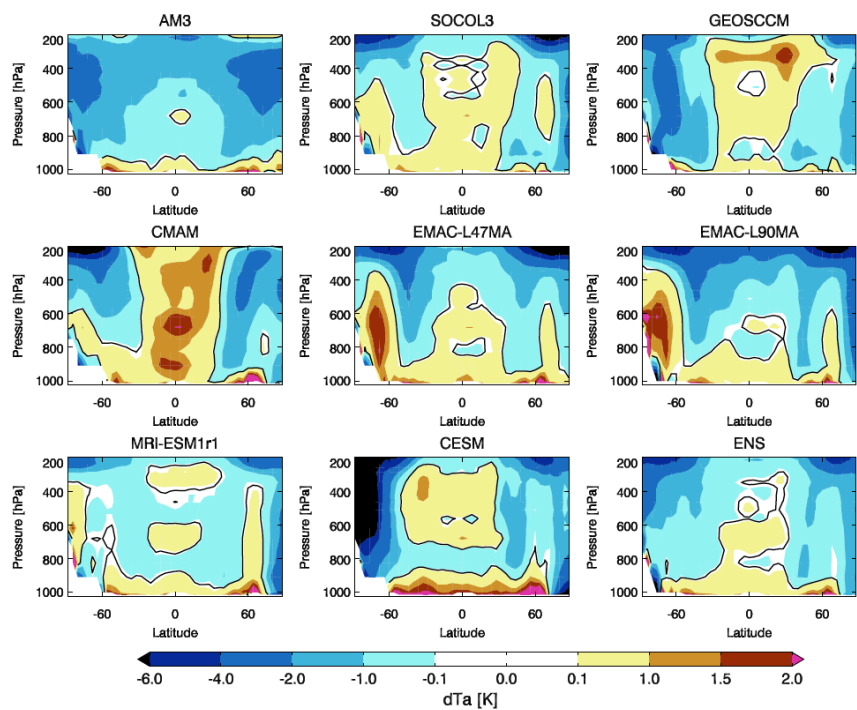


Figure 11. The zonal averaged vertical-latitude distribution of T_a biases from models to the reanalysis data. The black curves are the zero lines.

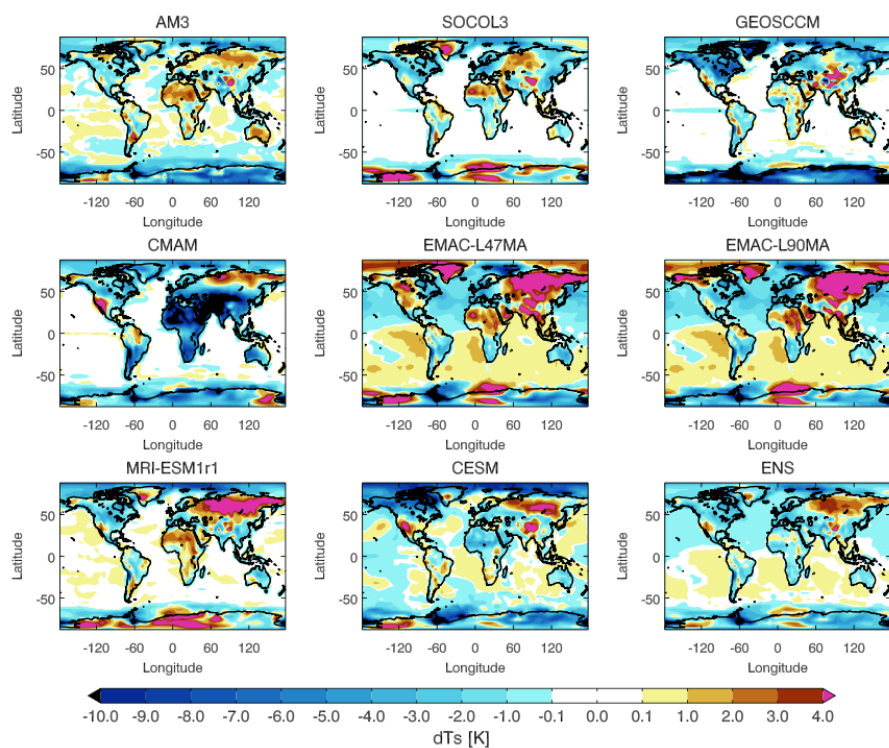


Figure 12. The global distribution of T_a biases from models to the reanalysis data

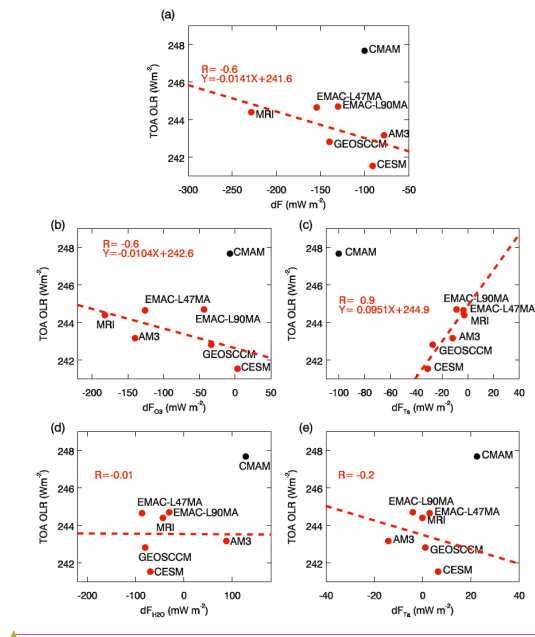


Figure 13. The correlation of the ozone band TOA flux biases to the model calculated broadband OLR (a) and the correlation of the attributed radiative components to the broadband OLR (b – e).

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Table 1. The chemistry-climate models and their experiment simulations

	Model	Institutes	CCMI runs		
		CCCma, Environment and Climate			
1	CMAM	Change Canada	REF-C1	r1i1p1	v1
2	SOCOL3	ETH-Zurich, PMOD/WRC	REF-C1	r1i1p1	v1

3	GEOSCCM	NASA/GSFC	REF-C1	r1i1p1	v1
4	EMAC-L47MA	DLR-IPA,KIT-IMK-ASF,KIT-SCC-SLC,	REF-C1	r1i1p1	v1
5	EMAC-L90MA	FZJ-IEK-7,FUB, UMZ-IPA,MPIC, CYI	REF-C1	r1i1p1	v1
6	MRI-ESM1r1	MRI	REF-C1	r1i1p1	v1
7	AM3	NOAA GFDL	-	-	-
8	CESM	NCAR	-	-	-
	ERA-Interim, TCR-1	Reanalysis	-	-	-

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Table 2 The global mean of the flux bias (mWm^{-2}) and the dominant components due to tropospheric O_3 , H_2O , T_a , and T_s . The numbers in parentheses are the standard deviation of the zonal distribution. For the ensemble, the standard deviation is computed from the variation between the models. The highlighted red numbers are the extreme values for the large biases. Two green numbers highlight the models that have relative small global and annual averaged TOA flux bias.

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Models	$\overline{\delta F}$	$\overline{\delta F_{T_s}}$	$\overline{\delta F_{T_a}}$	$\overline{\delta F_{H_2O}}$	$\overline{\delta F_{O_3}}$
AM3	-78.1 (46.2)	-11.7 (30.0)	-14.2 (7.8)	87.7 (76.4)	-140 (117.1)
SOCOL3	-283.6 (290.0)	-11.6 (22.3)	5.9 (13.9)	-94.5 (124.6)	-183.5 (187.5)
GEOSCCM	-139.9 (112.0)	-27.2(38.5)	1.1 (12.0)	-80.3 (120.9)	-33.5 (28.0)
CMAM	42.9 (118.5)	-100.2 (93.3)	22.5 (40.5)	127.9 (100.7)	-7.3 (28.8)
EMAC-L47MA	-154.4 (150.0)	-3.3 (27.3)	2.9 (14.8)	-28 (56.1)	-125.9 (128.2)
EMAC-L90MA	-130.2 (142.7)	-8.7 (36.0)	-4.1 (6.6)	-30.5 (63.0)	-86.9 (108.7)
MRI-ESM1r1	-228.5 (281.8)	-2.7 (17.9)	-0.1 (6.8)	-43.6(86.6)	-182 (213.7)
CESM	-91.1 (89.0)	-31.1 (33.9)	6.4 (13.9)	-69.7 (113.0)	3.3 (43.8)
Ensemble	-132.9 (98)	-24.6 (32)	2.6 (10)	-16.4 (81)	-94.5 (75)

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Table 3. Models' bias in the tropical troposphere between 25°S and 25°N, and below 200 hPa.

Models	$\overline{\delta O_3}$ (ppb)	$\overline{\delta H_2O}$ (ppm)	$\overline{\delta T_a}$ (K)
AM3	7.4 (5.0)	-586.5 (669.9)	-1.0 (0.8)

SOCOL3	13.4 (6.8)	330.3 (1067.3)	0.1 (0.4)
GEOSCCM	-1.1(4.3)	-506.6 (949.1)	0.5 (0.5)
CMAM	-1.3 (3.9)	18.1 (949.5)	1.0 (0.4)
EMAC-L47MA	10.4 (6.1)	-20.2 (729.1)	-0.2 (0.6)
EMAC-L90MA	8.2 (5.6)	203.1 (845.1)	-0.9 (1.0)
MRI-ESM1r1	13.7 (9.3)	386.9 (679.9)	0.1 (0.4)
CESM	-3.0 (4.4)	22.0 (630.7)	0.3 (0.7)

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Table 4. Models' flux bias (mWm^{-2}) in the tropic troposphere between 25°S and 25°N, and below 200 hPa.

Model	$\overline{\delta F_{O_3}}$	$\overline{\delta F_{H_2O}}$	$\overline{\delta F_{T_a}}$
AM3	-4.8 (3.7)	6.9 (10.5)	-0.7 (1.4)
SOCOL3	-9.2 (5.3)	-9.1 (21.6)	1.0 (2.2)
GEOSCCM	1.0 (3.3)	-8.5 (14.1)	0.7 (1.4)
CMAM	1.4 (2.8)	9.1 (18.5)	3.1 (5.0)
EMAC-L47MA	-7.2 (4.8)	-3.2 (15.3)	1.0 (2.9)
EMAC-L90MA	-5.6 (4.2)	-2.7 (16.4)	0.2 (2.2)
MRI-ESM1r1	-10.0 (7.4)	-5.5 (16.4)	0.3 (2.0)
CESM	2.1 (3.4)	-8.3 (15.0)	1.3 (4.5)
Ensemble	-4.06 (4.90)	-2.66 (7.02)	0.86 (1.1)

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