**Reviewer #3 (Formal Review for Authors (shown to authors)):**   
  
 In this manuscript, the authors describe the implementation of interactive atmospheric chemistry in the E3SM model facilitating the model to capture the influence of atmospheric composition changes, particularly of short-lived species, on the simulated Earth System responses and feedbacks. Congratulations! This is a significant advancement in the capabilities of E3SM, opening doors to many scientific explorations that were impossible in the previous version of the model with prescribed chemistry.   
 The paper describes the chemistry updates, evaluates their implementation against observations, and compares simulations against a previous version of the model without these updates plus other similar models. The paper is well-written and adheres to the requirements of JAMES. It should be in a publishable state once the specific comments below have been addressed.

We thank the reviewer for acknowledging the significant enhancement of E3SM’s scientific capabilities through the integration of interactive atmospheric chemistry into its standard release version for production simulations. We anticipate that the forthcoming E3SM release will accelerate follow-up studies that require interactive chemistry. We appreciate the reviewer’s valuable suggestions and constructive comments, which have been incorporated into our revised manuscript as noted below.  
  
 Specific Comments:   
  
 An overall specific comment is that the paper focuses solely on the evaluation of gaseous trace species while the implementation of interactive chemistry (especially including interactive representation of oxidants) has implications for aerosols. The paper would be stronger and more informative if it included an analysis of the impact of interactive chemistry on aerosol metrics such as aerosol burdens and AOD, particularly since the lack of consistent chemistry has been attributed as a reason for excessive aerosol-related forcing in the previous versions of E3SM.

This is a good point, and we have changed the title to reflect this specific study: “Interactive gas chemistry for enhanced science capabilities of the Energy Exascale Earth System Model version 3.”

As E3SM development is a multi-pronged effort, the developments of gas-phase chemistry, aerosols, and related cloud physics were undertaken by different teams, each of which is leading an overview paper documenting the respective capabilities for the upcoming E3SM release. The focus of this gas chemistry overview paper is primarily on the interactive gas-phase chemistry. Related aerosol results are presented in companion papers within the same JAMES special issue: the E3SMv3 atmosphere overview paper that documents the results of integrated chemical and physical new features (Xie et al., 2025, under review, https://doi.org/10.22541/essoar.174456922.21825772/v1) and the aerosol overview paper that documents aerosol features (Wang et al., 2025, to be submitted). We have revised Line 112 to reference these companion papers for aerosol results as follows: “More aerosol results are reported in Wang et al. (2025) and Xie et al. (2025).”

L58: It is not only important to capture the forcing caused by non-CO2 greenhouse gases but also all non-CO2 constituents that directly or indirectly impact the Earth's radiation budget. Suggest changing "non-CO2 greenhouse gases" with "non-CO2 constituents".

Done. Thank you!  
  
 L78-80: The consensus-building, scientific understanding gained from multi-model intercomparisons that helps advance our understanding of the chemistry-climate interactions.

That’s right. We added on Line 80 “The consensus and scientific insights gained from MIPs advance our understanding of chemistry-climate interactions.”  
  
 L92 and L129: References for "decadal monthly mean chemistry from an ESM selected as default standard" and "monthly mean climatology recommended for the MIP from another ESM" would be helpful.

Great suggestion. We have now added the reference to Hegglin et al. (2016) in the revised manuscript.

Hegglin, M., Kinnison, D., Lamarque, J.-F., & Plummer, D. (2016). CCMI ozone in support of CMIP6 - version 1.0. Earth System Grid Federation. https://doi.org/10.22033/ESGF/input4MIPs.1115  
  
 L136-138: Does E3SM-chem also alter the radiative forcing by affecting aerosol chemistry and therefore clouds?

Yes. Since radicals are now simulated by chemUCI rather than being prescribed from external data, aerosol chemistry involving these chemUCI radicals is affected, which in turn influences cloud properties. This gas-aerosol interaction will be documented in Wang et al. (2025) as indicated above.

L139-151: How is deposition of trace species considered? How does gas-phase chemistry interact with heterogeneous chemistry on aerosol surfaces? How is methane specified in the model?

Surface deposition is considered for most species (see Table SX in the revised version), using deposition velocities from the parameterization of Wesley (1989) as adapted in the Community Atmosphere Model (CAM).

The gas phase chemistry is coded with heterogeneous (aerosol) reactions of NO3, N2O5 and HO2 that convert the nitrate species to HNO3 and destroy HO2. In the current implementation they were turned off because the aerosol model was not yet stable. Thus, our study is a one-way gas-aerosol model with no feedback of the aerosols on the gas-phase chemistry. The two-way coupling is to be documented in the Wang et al. (2025) paper.

Methane and nitrous oxide are specified with boundary abundances at the surface based on the CMIP6 historical record.

Wesely, M. L. (1989), Parameterization of surface resistances to gaseous dry deposition in regional scale numerical models, Atmospheric Environment, 23 (6), 1293-1304.

L150: Lee et al (2025) is missing from the reference list.

Good catch. We added the reference to Lee et al. (2025)

Lee, H.-H., Q. Tang, M. J. Prather, and J. Xie (2025), A Comprehensive Chemistry Evaluation and Diagnostics Package for E3SM – ChemDyg Version 1.1.0, Environ. Model. Softw., 191, 106498, doi:10.1016/j.envsoft.2025.106498.  
  
 Section 2.2: Was there any retuning necessary to ensure radiation balance in E3SM-chem? If so, please elaborate.

Tuning was not done and not needed. We found the radiation balance was achieved after 10 years of the atmosphere-only AMIP-style run.  
  
 L177: Insert "emissions" after biomass burning.

Done.  
  
 Section 2.3: How are DMS emissions calculated?

We added “The DMS emission is represented as a monthly sea–air flux climatology, following Wang et al. (2018).”

Wang, S., Maltrud, M., Elliott, S., Cameron-Smith, P., & Jonko, A. (2018). Influence of dimethyl sulfide on the carbon cycle and biological production. Biogeochemistry, 138(1), 49–68. [https://doi.org/10.1007/s10533-018-0430-5](https://urldefense.us/v3/__https://doi.org/10.1007/s10533-018-0430-5__;!!G2kpM7uM-TzIFchu!xxluWsZuFxgdqOkhINfyo7FM1hoA-BE9XyfTvp7IRUBzOpxBbXhNBbP60NowOfCgpaI7495EstQ9chA0-_0QQs2G$)  
  
 L184: Insert a reference for the source of these emissions.

We added the reference to Tilmes et al., 2016. Thanks.

Tilmes, S., Lamarque, J.-F., Emmons, L. K., Kinnison, D. E., Marsh, D., Garcia, R. R., Smith, A. K., Neely, R. R., Conley, A., Vitt, F., Val Martin, M., Tanimoto, H., Simpson, I., Blake, D. R., and Blake, N.: Representation of the Community Earth System Model (CESM1) CAM4-chem within the Chemistry-Climate Model Initiative (CCMI), Geosci. Model Dev., 9, 1853–1890, [https://doi.org/10.5194/gmd-9-1853-2016](https://urldefense.us/v3/__https://doi.org/10.5194/gmd-9-1853-2016__;!!G2kpM7uM-TzIFchu!xxluWsZuFxgdqOkhINfyo7FM1hoA-BE9XyfTvp7IRUBzOpxBbXhNBbP60NowOfCgpaI7495EstQ9chA0--aECd8D$), 2016.

L196-198. The Griffiths et al paper did not provide the CO budget (only the burden). Any other references?

Good suggestion. We added “These numbers are consistent with the CO budget reported in Tables 1 & 2 of Duncan et al. (2007).” on Line 211.

Duncan, B. N., J. A. Logan, I. Bey, I. A. Megretskaia, R. M. Yantosca, P. C. Novelli, N. B. Jones, and C. P. Rinsland (2007), Global budget of CO, 1988–1997: Source estimates and validation with a global model, J. Geophys. Res., 112, D22301, doi:10.1029/2007JD008459.

L208-211: The tropospheric CO burden appears to be an order of magnitude higher than simulated in AerChemMIP models and other modeling studies (e.g., Horowitz et al.,2003; Naik et al 2013). Please check.   
  
We checked our calculation and fixed a typo. The tropospheric burden numbers were corrected to 379 Tg for CO and 4,777 Tg for CH4, which are consistent with literature. Thanks.

L214: AMIP can be deleted from "...system over the AMIP historical period."

Done.  
  
 L214-218: Is this description referring to a simulation of CH4 in AMIP and scenarios or a typical representation of CH4 in such simulations? Please clarify.

Sorry for the confusing writing there. Revised to “The temporal evolution of CH4 in the AMIP run for the historical period (up to 2014) is based on observational data.” and deleted the sentence about SSP scenarios, which our study does not include.

L218-219: Why not show the timeseries of methane lifetime in a plot to facilitate direct comparison with the time series from AerChemMIP simulations (Stevenson et al 2020)?

A table is more efficient in space. There is not enough information for a figure.  
  
 L219-220: The tropospheric CH4 lifetime also responds to changes in temperature, natural emissions (such as lightning NOx) as well as overhead stratospheric ozone via their influence on OH (e.g., Stevenson et al., 2020; Chua et al., 2023). Is there any trend in the simulated lightning NOx emissions?

Lightning NOx exhibits a slight increasing trend (2005-2014 vs. 1980s) in our simulation. Revised to “From 1980s to 2010s, the overall decrease in lifetime due to tropospheric OH is consistent with several driving forces: increase in tropospheric O3, H2O, and NOx emissions (including lightning) and decrease in CO emissions (Stevenson et al., 2020; Skeie et al., 2023).”

L229-230: There seems to be a problem in the figure for the CMIP6 results from Griffiths et al (2021). The model values do not match those depicted in Figure 8 of that paper. Also, what definition of the tropopause was used to determine the E3SM tropospheric ozone?

We have trouble reproducing Fig. 8 in their paper from the CMIP archive due to difficulties in defining tropopause as in their paper. Therefore, we used data in Table 1 of Griffiths et al. (2021) to regenerate our Figure 1 in the revised manuscript. We are using e90 = 80 ppb tropopause for E3SM and we confirmed that it closely matched the T-lapse rate tropopause in the tropospheric air mass.

L241-250: There seems to be some diversity in the distribution of tropospheric ozone as seen by satellites (Figure 10 of Gaudel et al., 2018). It would be informative to compare the model with the other satellite estimates. At the minimum, this diversity in observations should be acknowledged and the implications for model-obs comparison should be elaborated on. Further, the time averaging for model results should be as close to that used for observations for consistent comparison.

Here we aim to evaluate the overall magnitude and geographic pattern of tropospheric column ozone (TrCO) as simulated by E3SM-chem. Although different satellite data products may employ distinct retrieval algorithms tailored to the characteristics of their instruments, resulting in varying sensitivities to ozone at different altitudes within the troposphere, they consistently capture the main spatial patterns of TrCO, such as the minimum over the tropical western Pacific and the maximum over the subtropics (see Figure 10 in Gaudel et al., 2018). Consequently, for the purposes of this evaluation, comparison with a single satellite dataset (OMI + MLS) is sufficient to characterize model performance in reproducing large-scale TrCO features. However, it is important to note that the diversity in regional TrCO magnitudes reported among different satellite products, as documented by Gaudel et al. (2018), should be carefully considered in studies focused on improving the simulation of regional TrCO. Such detailed regional assessments are beyond the scope of the present overview.

On Line 250, we added this sentence as part of the revision: “Different satellite products may diverge in regional TrCO magnitudes depending on the distinct retrieval algorithms (e.g., Figure 10 of Gaudel et al., 2018); however, they generally show consistency in the overall spatial patterns analyzed in this study. Incorporating additional observational datasets in future work would be helpful for further improving the E3SM TrCO simulations, particularly at the regional scale.”

Figure 2 was revised to use model time averaging close to observations per reviewer suggestion. Averaging model results over 2005-2014 only slightly alters the results relative to averaging over 1990-2014 (e.g., global mean TrCO: 29.2 DU vs. 28.5 DU) and does not change the conclusions. Additionally, we revised the text related to Figure 2.

“Globally, E3SM-chem TrCO (29.2 DU) is about 7% less than observed (31.3 DU).”

A. Gaudel, O. R. Cooper, G. Ancellet, B. Barret, A. Boynard, J. P. Burrows, C. Clerbaux, P.-F. Coheur, J. Cuesta, E. Cuevas, S. Doniki, G. Dufour, F. Ebojie, G. Foret, O. Garcia, M. J. Granados-Muñoz, J. W. Hannigan, F. Hase, B. Hassler, G. Huang, D. Hurtmans, D. Jaffe, N. Jones, P. Kalabokas, B. Kerridge, S. Kulawik, B. Latter, T. Leblanc, E. Le Flochmoën, W. Lin, J. Liu, X. Liu, E. Mahieu, A. McClure-Begley, J. L. Neu, M. Osman, M. Palm, H. Petetin, I. Petropavlovskikh, R. Querel, N. Rahpoe, A. Rozanov, M. G. Schultz, J. Schwab, R. Siddans, D. Smale, M. Steinbacher, H. Tanimoto, D. W. Tarasick, V. Thouret, A. M. Thompson, T. Trickl, E. Weatherhead, C. Wespes, H. M. Worden, C. Vigouroux, X. Xu, G. Zeng, J. Ziemke; Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation. *Elementa: Science of the Anthropocene* 1 January 2018; 6 39. doi:<https://doi.org/10.1525/elementa.291>

L258: This value of the STE flux does not match with that given in Table 2.

We corrected the global STE flux to 405 Tg/year as indicated in Table 2 and NH STE to 166 Tg/year and SH STE to 239 Tg/year. Thanks for catching this.  
  
 L262: can you be more specific than this speculative statement - "...driven apparently by QBO..."

The STE O3 flux has a QBO-like variability which is consistent with the findings of Ruiz et al., 2021. We revised the text to “...driven apparently by the quasi-biennial oscillation (QBO), which is supported by the conclusions of Ruiz et al. (2021)...”

L270-280: While valuable at the time, the dataset of Schnell et al (2014) is now over 10 years old. It would be more informative to also add the TOAR surface ozone dataset for evaluation consistent with CMIP6 effort (Turnock et al., 2020)

While we recognize the value of incorporating the TOAR (Schultz et al., 2017) and CMIP6 surface ozone datasets (Turnock et al., 2020), our primary conclusion—that E3SM-chem outperforms most ACCMIP models based on the evaluation metrics of Schnell et al. (2014, 2015)—remains scientifically useful. We object to the inference that the Schnell data set - representing 10 years of hourly surface O3 observations (2000-2009) is outdated compared with the Turnock analysis. It remains a perfectly valid data set to compare with CMIP models that did not go past 2014. Turnock uses 2005-2014 O3 data, which is not that different. The Schnell 2015 comparison is still the only one that addresses the diel variation of O3, which is critical if we are to compare monthly means without bias. The Turnock analysis uses monthly mean (24-hour average O3) and is sadly lacking the MDA8 comparison, since a monthly mean can be problematic with the nighttime boundary treatment of O3. Turnock did what could be done with the limited CMIP6 diagnostics (no hourly surface O3). We have added the integration of TOAR and CMIP6 datasets to the ChemDyg future development roadmap.

L287: Please shed some light on "suggesting emission errors in the NH." How has it been determined that CO and VOC emissions are the cause of this bias?

Evidence is in the following sentence: increasing OH sink for CO will only make the CH4 budget worse. Deleted “suggesting emission errors in the NH.”

L290: Table 4 is not in the main text, however the information is in Table S2 which I recommend bringing in the main text.

We corrected the reference to the Table, which is now cited as Table 4 in accordance with the reviewer’s suggestion.  
  
 L297: "slightly lower" - can you be more quantitative?

Yes, we added “(4%)” after “slightly lower”.  
  
 L314: Clarify how tropospheric O3 ERF is calculated from the model? Is this at the TOA or at the tropopause? How is the contribution of stratospheric ozone separated?

We agree that these details are useful. L314 was changed to “...with an ERF of 0.32 W/m2 at the TOA. It includes the stratospheric adjustment because we do not separate stratospheric changes.”

L322-323: As I understand only the MLS sees stratospheric ozone. Can you provide a reference for this dataset?

We added the reference link (https://acd-ext.gsfc.nasa.gov/anonftp/acd/atmos/ziemke/tco\_omi\_1by1\_oct2004\_to\_dec2024.sav, accessed on June 9, 2025), which is the latest version of this OMI + MLS dataset. We also updated Figure 6 to reflect this observational data version update. The minor changes between the two data versions do not affect our conclusions.

MLS observes stratosphere and upper troposphere, so it requires Ziemke’s OMI + MLS algorithm to decide where the tropopause is and hence the stratospheric column ozone.

L325: Can you qualify the statement "Total column ozone is not a good metric" by a reference?

Ziemke et al. 2019; 2022 include tropospheric O3 2D trends, regionally trying to match modeling these are driven by pollution - stratosphere has no such tends and should not be mixed. Stratospheric O3 has only 1D trends. We added these two references to support this.

Ziemke, J. R., Oman, L. D., Strode, S. A., Douglass, A. R., Olsen, M. A., McPeters, R. D., Bhartia, P. K., Froidevaux, L., Labow, G. J., Witte, J. C., Thompson, A. M., Haffner, D. P., Kramarova, N. A., Frith, S. M., Huang, L.-K., Jaross, G. R., Seftor, C. J., Deland, M. T., and Taylor, S. L.: Trends in global tropospheric ozone inferred from a composite record of TOMS/OMI/MLS/OMPS satellite measurements and the MERRA-2 GMI simulation , Atmos. Chem. Phys., 19, 3257–3269, https://doi.org/10.5194/acp-19-3257-2019, 2019.

Ziemke, J. R., Kramarova, N. A., Frith, S. M., Huang, L.-K., Haffner, D. P., Wargan, K., et al. (2022). NASA satellite measurements show global-scale reductions in free tropospheric ozone in 2020 and again in 2021 during COVID-19. Geophysical Research Letters, 49, e2022GL098712. <https://doi.org/10.1029/2022GL098712>

Section 3.3: How do E3SM-chem stratospheric results compare with those from CMIP6 models as discussed by Keeble et al 2021?

Keeble et al. (2021) (<https://acp.copernicus.org/articles/21/5015/2021/>) includes E3SMv1 results, which suffer from the stratospheric dry biases (also seen in other models, see their Figure 11). This is now corrected in E3SM-chem. Further, E3SM-chem results are similar to those CMIP6 models with the representation of stratospheric H2O production from CH4 oxidation (their Figure 12).

For stratospheric ozone, Keeble et al. (2021) shows that E3SM is in good agreement with CMIP6 models. Here, we demonstrate that E3SM and E3SM-chem exhibit relatively small differences, indicating that E3SM-chem is also well aligned with CMIP6 results.

L328-329: Is this an assumption or a statement of fact?

The excellent agreement in SCO for the tropical region means that E3SM-chem will have little bias in J-O1D in the tropical troposphere, and this drives the primary formation of tropospheric OH (from photolysis of tropospheric O3 and O1D + H2O.).

Figure 7: It would be helpful to average the model and ERA5 results over the same time periods for consistent comparison.

Agreed. Done.  
  
 Section 4: It is nice to see that the addition of chemistry does not adversely affect climate variables in the AMIP mode. Will this hold for the coupled model simulation? It would be good to get some indication of this even if preliminary. Further, it would be helpful to show the impact of chemistry on QBO since it affects the surface climate via teleconnections.

We have verified that the new chemistry does not adversely affect climate in the coupled simulation, which is described in Golaz et al. (to be submitted to the same JAMES E3SM special issue).

Thanks for the suggestion on investigating the impact of chemistry on QBO. There is a parallel effort to improve the representation of QBO in E3SM (Yu et al. 2025). Simulation of QBO has been largely improved in E3SMv3, which is mainly due to an increase of vertical resolution in the stratosphere (Xie et al. 2025). We are currently conducting an in-depth analysis of the QBO in E3SMv3 including the impact from the new chemistry and how changes in QBO simulation affects the surface climate via teleconnections. A dedicated manuscript is expected later this year.

Yu, W., Hannah, W. M., Benedict, J. J., Chen, C.-C., & Richter, J. H. (2025). Improving the QBO forcing by resolved waves with vertical grid refinement in E3SMv2. Journal of Advances in Modeling Earth Systems, 17, e2024MS004473. <https://doi.org/10.1029/2024MS004473>

Xie, S., Terai, C. R., Wang, H., Tang, Q., Fan, J., Burrows, S. M., et al. (2025). The Energy Exascale Earth System Model Version 3. Part I: Overview of the Atmospheric Component. ESS Open Archive. https://doi.org/10.22541/essoar.174456922.21825772/v1