August 22nd, 2022

Dear Editor and Reviewers,

Submission of "Implementation and evaluation of the GEOS-Chem chemistry module version 13.1.2 within the Community Earth System Model v2.1" to *Geoscientific Model Development*

Thank you for arranging this review of our work, and for your patience as we have worked to revise the manuscript. Since both reviewers noted the length of the paper, we have worked to both streamline it (by moving less critical figures to the SI) and to make it more readable by breaking specific analyses out into separate sections. We have also worked to make the paper more accessible to non-specialists through the addition of brief descriptions of key chemistry where relevant as requested by Reviewer #1, while aiming to make the paper more relevant to specialists through the addition of more technical data (in particular emissions) and diagnostic data (correlation coefficient tables). Although we recognize that the paper cannot cover all bases, we believe that these changes have helped to improve the relevance and breadth of impact of the manuscript.

As Reviewer #1's mentioned, several of their suggestions and questions took the form of comments which were placed directly on the manuscript itself. Most such comments are listed below with a response; however, for some minor comments, we have simply made the requested change without an explicit response below. These changes are reflected in the updated manuscript.

Please find below our point-by-point responses (in **bold**) to the review comments (in *italics*) and a markedup version of the revised manuscript. A "clean" revised manuscript will be provided through *GMD*'s submission system.

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Reviewer #1:

Summary:

This manuscript documents the new capability of running the Community Earth System Model (CESM) with the GEOS-Chem chemistry online. Comparisons are made to the current coupled model configuration of CESM with CAM-chem chemistry module and the GEOS-Chem chemistry transport model when implemented for high performance computing ("GCHP"). This is an exciting development for both the GEOS-Chem community and the CESM community. The authors provided lengthy inter-model and observation comparisons. This work is highly relevant, and I support publication after my overall comments below are addressed as well as my minor and technical edits within the marked-up PDF are considered.

Comments:

This manuscript is very long and it is a lot to ask of your readers to commit to nearly 50 pages of figures and text. There is a lot of overlapping information in Section 2 (Coupling GEOS-Chem and CESM) and the start of Section 3 & 3.1 (Simulation setup) and I recommend the authors consider synthesizing the details, possibly restructuring these sections. The authors could then make Sections 3.2 and 3.3 their own sections. Given the authors referenced later sections when trying to explain the differences, the authors should reconsider if keeping the model intercomparisons separate from the observations is the best flow for this paper. There were times when reading the model intercomparisons I kept asking myself "which model configuration is closer to observations" and I had to wait to find out if even the model-to-observations comparison was provided.

As discussed in our opening comments, we have attempted to both shorten the paper somewhat and to make it easier to navigate. Any overlapping information between Section 2 and Section 3 has been removed from the latter. Section 3 is now purely about model setup as both Sections 3.2 and 3.3 have been moved respectively to Section 4 and Section 5. This aims to streamline the results section and make it clearer for the reader what is performed in each section. Section 4 is purely about model intercomparison, while Section 5 evaluates the model results against observations.

For the profiles of NOx, NOy, and some of the halogen species, there are satellite observations which could be used for validating at least the stratospheric portions (e.g., MLS, ACE-FTS). There are also groundbased and balloon-based observations of water vapor. The choice of climatologies for the sonde and satellite observation comparisons was not clear to me when observations for 2016 should be available to the authors (also no references were provided for these data sets). Be clear as to the reasoning behind the observations used for the validation section.

The missing references were an oversight, and we are grateful to the reviewer for pointing this out. The relevant references have now been added to the manuscript (lines 948 and 963). With regards to the use of climatologies, we chose this approach as they are part of the standard CESM evaluation package and this comparison is therefore likely to be familiar to users from the CESM community. We felt that this approach was reasonable when the focus was on evaluating differences between model configurations rather than the overall accuracy of the model. This justification is now provided on lines 922-924.

By the end, I was also trying to find ways to reduce the figures, and suggest the authors consider if all panels and figures are necessary or could be included in supplemental information.

We agree that some material is not critical to the main text. We have attempted to shorten the manuscript by moving less critical figures (including the figures showing wet deposition tendencies or surface NO_x concentrations, formerly Figures 12, 13, 14, and 20) to the Supplemental Information.

Especially in the model intercomparison, there is often an assumption of the reader's knowledge of atmospheric chemistry. Provide the chemistry background and references to support statements as to why different chemistry leads to differences between models.

We now provide some detail regarding the chemistry (including references) relevant to specific differences, with the goal of aiding the reader and supporting our statements. This includes referencing DMS chemistry in the context of oceanic sulfates (lines 626-628), how to interpret lower-stratospheric water and ozone differences (lines 552-555), the relationship between NOx and PAN (lines 715), and the importance of halogen chemistry to ozone (lines 747-749).

Be careful quoting figures. I strongly suggest adding panel labels and referencing figure panels whenever possible in the main text. Often there are numbers quoted in a paragraph that I would have expected came from the figure currently being discussed but I do not find these numbers in the figure. In some places, this may have to do with number of significant digits used in the text vs the figure; but it is unclear. When results are discussed including a lot of numbers, I suggest the authors consider tables to make it easier to digest and compare the numbers between the different models (and regions). If numbers are provided but not from the figure being discussed in that paragraph state "not shown" so the reader does not spend time trying to find it.

We have worked to make it clearer when we are not citing specifically from figures (e.g. stating "not shown" as suggested). We have also increased the number of tables in the manuscript (e.g. the new Tables 2 and 4), while trying to balance against the need to avoid further lengthening the manuscript.

The acronyms C-GC, C-CC, and S-GC are so similar it makes reading the comparisons hard to follow. The acronyms were only used in Section 3, not in Section 2 nor in the final Discussion and conclusion Section 4. I struggle to think of alternatives that may be better. Maybe using a lower-case c for CAM-chem will help (e.g., C-GC, C-Cc) or include CESM instead of simply C (e.g., CESM-GC, CESM-Cc). Also, the standalone GEOS-Chem uses GCHP, so maybe simply using GCHP instead of S-GC would help it stand out from the CESM acronym. I had to keep reminding myself if it was using GCHP, not the Classic CTM, while reading the manuscript.

Unfortunately we too were unable to think of acronyms which would work better. Our concern with using GCHP instead of S-GC is that the specific implementation of GEOS-Chem is not significant (GCHP and GC-Classic are essentially identical apart from their handling of transport), so highlighting it in such a way might be distracting. However, we have emphasized in the introduction that S-GC is the GCHP CTM, and not GC-Classic (see next comment).

Specific comments:

Is this referring to the GEOS-Chem Classic CTM or simply a free-running version of C-GC?

We now specify in the abstract that the acronym S-GC refers to the GEOS-Chem High Performance CTM (e.g. on line 393).

globally due to bromine or is this regional?

This statement (regarding the possible causes of differences in ozone) was incomplete. We have now clarified that there are other factors which may contribute, with variable roles in different regions (lines 32-34).

lower altitudes, but still in the stratosphere or in the troposphere?

We have now replaced "lower altitudes" with "in the troposphere" to clarify this sentence (line 32).

Could be rewritten, its a bit difficult to read.

We have clarified this sentence. The sentence now reads: "This difference in tropospheric ozone is not uniform, with tropospheric ozone in C-GC being 30% lower in the southern hemisphere when compared to S-GC but within 10% in the northern hemisphere. This suggests differences in the effects of anthropogenic emissions." (lines 35-35)

So now this sentence is referring to tropospheric ozone?

We now state that this sentence refers specifically to tropospheric ozone (lines 32-35).

i don't understand how the set up of the two versions is different from simply reading the abstract.

We now clarify that the MERRA-2 meteorology is used directly in the GEOS-Chem CTM (lines 39-40).

Change to NASA MERRA-2 Reanalysis meteorology, if that fits in the word limit for the abstract.

This change has been made (lines 39-40).

I'd say reference here but i see you have the url to GEOS-Chem on the following line. What about the Bey et al. 2001 reference?

Indeed, the URL has changed since we first submitted the manuscript. The manuscript now points to <u>http://geos-chem.org</u> (line 58) and we have added the relevant references for both GEOS-Chem (Bey et al. 2001 for GEOS-Chem and Eastham et al. 2018 for GCHP) and CESM (see lines 53-55).

This is true for GEOS FP but MERRA-2 resolution is coarser (0.5deg x 0.625deg).

We now state both meteorological reanalysis datasets with their respective horizontal resolution (lines 81-84).

Does it also have a world-wide network of research groups? Can you give it similar accolades as you did for GEOS-Chem? Is there a good url?

CAM-chem is widely used in the research community as it offers the capability to simulate tropospheric and stratospheric composition within CESM. In particular, users can choose to run CAM-chem with specified meteorology or as "free-running" (with climate feedback). We have now added URLs for both CAM-chem (line 116) and GEOS-Chem (line 58).

I usually recommend only using acronyms after they are defined but are these here so common to the community that they do not need definitions?

The acronyms in question (WACCM, ACCMIP, CCMI, POLMIP, HTAP2, GeoMIP, and CMIP6) admittedly vary in terms of how well they are known. However, all are better known by their acronym than by their full name, and none are used again in the manuscript. We have therefore opted to keep them as acronyms (lines 110-111) rather than potentially compromise readability. However, we would be happy to revisit this decision if necessary.

Is there a url which lists CAM-chem participating in all of these activities?

We have added the following URL (line 116): <u>https://www2.acom.ucar.edu/gcm/cam-chem</u>

I suggest you move sentence to start of next paragraph to better connect the ideas.

We agree with the reviewer's comment. This sentence has now been moved to the beginning of the next paragraph (line 118).

"...the only implementations of GEOS-Chem which are currently publicly available are either designed to run "offline"... or operate at regional scale and do not extend to global simulation": What does this mean? A researcher outside of GMAO can only run GEOS-Chem Classic CTM or GCHP, or WRF-GC?

This is correct. The only other relevant implementations are a frozen (and now outdated) version of GEOS-Chem in the Beijing Climate Center's model, and the implementation in GEOS which is (to our knowledge) only used (or usable) by researchers at NASA.

But you're going to try [to disentangle the root causes of these differences] here, right?

Throughout this study, we have tried to disentangle the source of the differences between the C-GC and C-CC model results.

but you just said you were comparing 2016. is there a spin-up?

We perform a spin up for each simulation setup but only compare the results for the 2016. We now state explicitly the spin up periods used (lines 402-406).

This is a good description of the schematic, but there are some aspects that are not clear. Why are there dashed boxes and arrows in the Chemistry section on the right, versus the solid arrows on the left. Why is Driver/Mediator in a dashed box? Why is the GEOS-Chem source code outside the box with the chemistry options? Are CAM-Chem and GEOS-Chem headers in a table? Dynamics is not connected to anything.

Figure 1 has been regenerated with the same kind of boxes and arrows to avoid any confusion. The large blue box on the right represents a blow-up version of the data exchange between the atmosphere state and the chemistry module (whether CAM-chem or GEOS-Chem). The GEOS-Chem source code stands outside of that blue box because it is not part of the CAM source code but rather downloaded for the GEOS-Chem Github repository. Dynamics are not directly applied through the control layer but rather acts on a "dynamics container" which are then translated to tendencies.

This is not labelled as such in Figure 1.

We now state in the manuscript that this "coupling interface" is labeled as "CESM2-GC interface" in Figure 1.

I assume this is a subfolder within the top level directory of the CESM2 code. Can this be written out in more layman's terms?

We now clearly state that the interface code is kept in a subfolder of the chemistry source code, and we have decided to keep the relative path to this subfolder within parentheses.

Using ESMF or NUOPC was supposed to make sure you could do clean testing within a modular system. How will this be accomplished otherwise?

At this time we are relying on the principle that the GEOS-Chem code base is common between all implementations, relying on interface codes which are now supplied as part of the GEOS-Chem base. We did this because we knew that CESM development is moving towards NUOPC, at which point we intend to update our interfaces to leverage that capability.

define SOA since the acronym is used later.

The acronym is now defined on first occurrence (line 199).

is this a general Modal Aerosol Model acronym or should it be MAM4? My brain defaults to spring March-April-May MAM.

The official acronym of the Modal Aerosol Model is MAM, with MAM3, MAM4 and MAM7 describing the number of modes used in the simulation. The CESM default is to run with MAM4.

Not sure if there is a journal preference but I recommend changing this to m-3

All occurrences of these in the manuscript have been modified to "m⁻³" (instances highlighted – e.g. line 229).

Is there a word missing here? "When implemented as the standalone model,"

Indeed, a word was missing. The sentence now reads: "When implemented as the standalone model, wet deposition is calculated for large-scale precipitation using separate approaches for water-soluble aerosols (Liu et al., 2001) and gases (Amos et al., 2012) with calculation of convective scavenging performed inline with convective transport."

Define

HNO₃ is now defined as nitric acid.

This link did not work for me.

The new URL is now https://geos-chem.seas.harvard.edu/.

add (H2SO4) afterwards to correspond with use in Line 252

We have now added (H₂SO₄) after gas-phase sulfuric acid.

not yet defined.

H₂SO₄ is now defined (see previous comment).

what does this mean here? Are you comparing C-CC and C-GC in this table? Try to be consistent with acronyms.

The table lists the mapping in place between the GEOS-Chem and CAM-chem chemical representations. C-GC and C-CC refer to the fully-coupled CESM alongside its atmospheric chemistry module (GEOS-Chem or CAM-Chem).

Neither of these products have been introduced before. Change to "using the meteorological fields from NASA Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2; Gelaro et al., 2017) and GEOS Forward Processing (FP; Lucchesi, 2018).

Since the MERRA-2 and GEOS-FP acronyms are now defined previously in the manuscript, the sentence now reads "using the NASA MERRA-2 and GEOS-FP meteorological fields" (line 309).

Trying to keep track of all the different module names versus the ESM name is not easy. CAM-Chem here doesn't have its own versioning, it is linked to the version of the CESM?

The Community Atmospheric Model (CAM) does have its own versioning system (C-GC and C-CC both use CAM6), but we are not aware of any subversioning system for CAM-chem. Each release of CESM is tied to its version of CAM. We have attempted to be more explicit in the revised manuscript (e.g. line 331).

Isn't this already stated on Lines 185?

Indeed, we have removed the double occurrence.

what does this mean? C-GC or C-CC? (this refers to "Whichever scenario is chosen")

The expression "Whichever scenario is chosen" refers to the choice between CMIP6 scenarios (e.g. historical or one of the SSPs). This is now clarified.

What does this stand for?

CIME stands for the Common Infrastructure for Modeling the Earth. The sentence now reads: "Additionally, we modify CAM, CLM and the Common Infrastructure for Modeling the Earth (CIME) such that the land model can pass land type information and leaf area indices to the atmosphere model to compute dry deposition velocities."

when is this expected to happen? And on who's authority/guidance?

We now say that this is currently being discussed with the CESM team. Although we interact on a monthly basis with the team, there are no hard guarantees regarding an implementation timeline (lines 370-371).

Explain why [a complete copy of the source code is downloaded]

To make sure that the most recent version of the GEOS-Chem chemical representation is used, a full copy of the code is downloaded when first setting up C-GC. Later updates of the GEOS-Chem code can be brought in using the Git version control tool. The sentence now is: "Although a complete copy of the GEOS-Chem source code is downloaded from the version-controlled remote of GEOS-Chem repository (to ensure that the most-recent release of GEOS-Chem is used), not all files present in the GEOS-Chem source code directory are compiled" (lines 377-379).

There's a fair bit of repetition in Section 3 opening paragraph and Section 3.1 to was already provided in previous Section 2. Given how long this paper is, I recommend revisiting these sections to reduce any redundant information/model descriptions. You may find you can change Section 3.1 to be a subset of Section 2 and then Sections 3.2 and 3.3 can be more stand-alone "result" sections.

Section 3.1 (which is now just "Section 3") only describes the model setup and we made sure that no overlap exists with Section 2, which focuses on the description of each module and their interface with the whole code. Additionally, Sections 3.2 and 3.3 have been moved to Section 4 and Section 5 respectively.

Should you be using C-GC and C-CC throughout Section 2?

We now use the C-GC and C-CC acronyms through all the sections in the manuscript for consistency.

this is odd wording. Did you preform a two-year simulation which you will evaluate against two other model simulations?

We understand that this sentence caused confusion, so we have removed the last few words. A two-year C-GC simulation was performed and evaluated against the other models and observations. The evaluations are described in the same paragraph.

It's odd to reference Section 3.2 and 3.3 without referencing Section 3.1. I advise rewriting this opening paragraph to introduce the models and then describe first a model intercomparison to establish the differences of C-GC to C-CC and then the C-GC evaluation against observations

Sections 3.2 and 3.3 have now been moved to their own Section. Section 3 now just consists of the old "Section 3.1".

How much does the choice of initial conditions impact the difference between the model simulations? How close are the initial conditions between CESM and S-GC? If the CESM initial conditions are used where S-GC doesn't have the fields, could they not have been used entirely to start the C-GC run to be consistent?

The initial conditions used for C-GC have been obtained using the existing C-CC restart file (which contains initial conditions for MAM aerosols, CAM-chem species, and some other meteorological data). However, any species involved in the GEOS-Chem chemistry scheme has been added to this C-GC initial file, using the S-GC restart file. This S-GC restart file is obtained after a decade-long S-

GC simulation. The simulation we performed with C-GC used a one year spin up period, using the same "default" restart file which is expected to provide a reasonable representation of the early 21st century (private communication). A brief description is now provided on lines 402-411.

Our approach allows for the troposphere to be reasonably well spun-up in all three configurations. However, given the model differences between S-GC and C-GC, we realize that stratospheric concentrations might need longer timescales to reach a quasi-equilibrium. Further work would be needed to quantify the impact of the initial spin up time.

Change to 0.01 hPa to be in the same units as the CESM v2.1.1 description in line 393.

The corresponding change has been made in the manuscript.

This is first mentioned at line 299, so it should be defined at first use.

We now define the MERRA-2 acronym on its first use in the main text (lines 84-85). Although we use the term MERRA-2 in the abstract, we do not define it there due to length concerns.

Add references for these schemes

These citations (Bogenschutz et al., 2013; Zhang and McFarlane, 1995) are now included.

not defined yet

The acronym CFC has now been defined. The sentence now reads as "Finally, for long-lived species such as chlorofluorocarbons (CFCs) we use the shared socio-economic pathway 2-4.5 (SSP2-4.5) set of surface boundary conditions in both C-GC and C-CC" (lines 478-479).

have these been defined as H2O and OH?

The sentence now states "water vapor (H₂O) and the hydroxyl radical (OH)".

Based on this comparison being the third panel, I was expecting the middle panel to be discussed first.

We now generally evaluate the differences between C-GC and S-GC first, and then between C-GC and C-CC, thus following the order of the panels.

This result statement should come later where you discuss bromine, or possibly at Line 506, where you discuss reasons for all these differences.

This sentence has been moved to the corresponding paragraph and we now refer to Section 4.4 as a link to the NO_x and bromine comparison Sections.

I encourage the authors to label the panels a, b, c.

We have regenerated each figure with a panel label as requested, and where possible refer to specific figure panels in the main text.

Are you trying to say [the absence of a specific pattern in the ozone delta] is likely related to transport in the online vs offline GEOS-Chem?

Not exactly – we meant more that differences might be related to different treatments of meteorological variables, rather than differences in transport (since all three model configurations use the same prescribed wind fields). We now specify that we are referring to, for example, the differing treatment of water vapor (lines 511-512).

Again, comparing first to C-CC but it is the third panel. Is there a reason to put S-GC in the middle?

We have aimed for consistency in panel ordering throughout the manuscript. However, in this case, we felt that the comparison between C-CC and C-GC resulted in a more logical flow for the comparison to S-GC, and thus began with that rather than a comparison to S-GC.

the next section specifically evaluates the surface ozone. I suggest changing this instead of "at the surface" but "from the surface"

We agree with the reviewer's comment. This change has been implemented in the manuscript.

this figure doesn't show an average. I suggest changing this to "across"

We have made this change in the manuscript (line 30).

The KORUS-AQ campaign is one location, not at all latitudes. Is there a better comparison, such as in Keller et al. (2021) evaulation, where we looked at ozonesondes across all latitudes. If you are specifically trying to highlight the GC vs CC, at least state the caveat that this is regional comparison and not global. [AND] Is this related to the KORUS-AQ study or the current study?

To further clarify that these observations are all made in one geographical location and to more clearly communicate the source of the observed differences, we have added the following sentence: "This suggests that discrepancies observed in KORUS-AQ may be related to chemistry rather than the treatment of meteorology, but a more focused regional analysis would be needed to confirm this" (lines 533-534).

I assume later you'll tell us why this might be. If not, can you provide here a reason?

The sections later in the paper go into detail as to why we observe this difference. We now state on line 544 that this will be discussed in Section 5.2.

Explain why this would impact ozone. Is this seasonal signal that is dominating the annual differences?

To further explain the role of stratospheric water vapor in ozone change, we have added the following sentence at the end of the paragraph: "This is unlikely to be due to HO_x catalytic cycles depleting ozone, as OH in this region is lower in C-GC than in S-GC (panel e) and HO_x cycles are in any case a minor contributor to ozone depletion in the lower stratosphere (Brasseur and Solomon, 2006). The greater water vapor (and therefore humidity) may instead result in faster heterogeneous chemistry, including the liberation of NO_x from HNO_3 " (lines 552-555).

use "approximately" or "about" instead of a tildas.

We have replaced all tildes with the word "approximately".

[Figure 3 middle panel] Is this in reference to the red plume that goes up from the surface to 600 hPa? From the colorbar it is hard to know which color is the 10% to know if this is limited to 900 hPa to the surface.

Yes, this increase of 10% in northern mid-tropospheric OH mixing ratio is a reference to the plumelike feature in the figure. The percentage represents the relative difference between the two model results.

add "generally", as there are some red hot spots in both difference plots

We have made the corresponding change.

I don't see this. It looks darker over the southern ocean.

This sentence was incorrect and has now been removed.

What color is zero? It looks like a slight negative bias to me, with a slight positive bias over the eastern pacific near central america.

This comment refers to Figure 4, middle panel. This panel compares surface ozone as simulated by C-GC and S-GC. We agree that there is a small difference, rather than no difference. The sentence now reads as "In the Northern Hemisphere, we observe a small difference in surface ozone mixing ratio over the oceans (less than 1 ppbv), while a difference of approximately 3 ppbv can be found over North America, Europe and East Asia."

Is there such an asymmetry in bromine from sea salt between southern and northern hemispheres? What is the rational behind this statement?

Given that C-CC does not model sea salt bromine and that ozone is mostly lower over the oceans in C-GC compared to C-CC, we suggest that this difference may be induced by sea salt bromine. On the other hand, the difference between C-GC and S-GC mostly occurs in the Southern Hemisphere, which wouldn't be just explained by differences in sea salt bromine. We therefore believe that this difference may be driven by other factors, such as the larger anthropogenic influence in the Northern hemisphere.

the hemispheric differences seem to be on the order of 2.5 ppb, just in different directions depending on the model comparison. Are you arguing that the C-GC and C-CC difference is not asymmetric?

We agree that there is an asymmetry, but that it is weaker. We now clarify (line 590) that this is specifically in absolute terms, since the asymmetry should also be considered against the baseline hemispheric asymmetry in ozone calculated by C-GC.

Is this suppose to quote the top left panel? The r2 value is 0.91 in Figure 5. i don't see this number in Figure 5. Is it supposed to quote the r2 value from top right?

We indeed had a typo in this sentence. The new sentences are: "In January, we find a correlation coefficient of 0.91 and slope of 0.91 between C-GC and S-GC. In July this agreement is worsened, with a correlation coefficient of 0.80 but a slope of 0.93" (lines 599-600).

Is this an eye ball estimate from the figure or did you output this range? It looks like it starts before 15 ppbv in C-GC to me.

The range provided in the manuscript indeed started approximately around 11 ppbv. The sentence has been modified to the following: "There is also a distinctive "hot spot" in the July parity plot, with a large cluster of grid cells showing mixing ratios in the range 20-25 ppbv in S-GC but 10-20 ppbv in C-GC" (lines 602-603).

DMS has not been defined. Explain to the readers the chemistry behind this connection of DMS to Sulphate aerosols.

We have now properly defined the acronym DMS. We now provide a brief description of the relevant chemistry (lines 626-628).

These large saturated differences start at 700 hPa so I would say at least from the mid-troposphere. This would look lower down in the atmosphere if you used a log-scale for the pressure.

We agree with the reviewer's comment. We have replaced "upper troposphere" with "tropical midto-upper troposphere and extratropical lower stratosphere" (line 631).

Is there a reason that surface ozone got its own subsection but surface aerosols are lumped with the zonal means?

We chose to analyze surface ozone because of its importance to oxidation as well as its importance in air quality. We had originally planned to perform a similar analysis for PM_{2.5}, but chose not to do so in order to save space. Such an analysis could however be included if desired.

Not all of your readers are likely meteorologists and will know where the ITCZ is located. I suggest describing this instead as "off the west coast of southern hemisphere continents" or something like that.

We have made the recommended change on lines 639-640.

Do you think this is because of the emissions in the southern latitudes as you state in line 587? Why do you think this may be? In Line 588 you claim "despite emissions of DMS from oceans".

We are not certain that these differences in surface sulfate concentrations in the ITCZ are caused by the same factors as the difference in sulfate concentrations between C-GC and C-CC in the Southern Hemisphere, in part because DMS emissions are calculated in identical fashion in all three simulations. The differences in surface sulfate concentrations could be explained by differences in a combination of factors such as deposition tendencies, aerosol representation, and OH abundance, but we have not been able to draw a firm conclusion thus far. Further work would be needed to identify the underlying factors causing these differences in surface sulfate concentrations. We now state that this issue is unresolved in Section 4.3.

I suggest this is changed to "mid- and upper tropical troposphere and throughout the lower stratosphere"

We agree with the reviewer and have implemented the corresponding change (lines 653-654).

NOx was first used at line 307 but never defined. It should be written out as NO and NO2.

 NO_x is now first defined on first use (line 312) as NO and NO_2 . In the rest of the manuscript, we refer to nitrogen oxides as NO_x .

Define which species you are adding up here to be NOy

We agree that this can lead to confusion given that GEOS-Chem and CAM-chem have different species included in NO_y . We have added the following sentence in the manuscript: "A full list of the species included in the lumped NO_y reservoir species can be found in the legend of Figure 10 for each model configuration" (lines 663-664).

For those readers who may be red/green color blind, I suggest replotting with different color choices or using dashed styled lines. Also could use the same colors as in the tailor diagrams.

We agree. Where possible, we have modified the plots so that red and green are not used in the same Figure (e.g. Figure 9).

How does the vertical resolution of the model configurations differ and impact the zonal mean comparisons here (and in previous sections).

All three models use the same vertical resolution. The only difference is that S-GC has a higher model top. Indeed, S-GC uses a 72-layer grid where C-GC and C-CC both use a 56-layer grid. In any case, the first 56 layers have identical pressure edges. This is stated in Section 3 (lines 419-423).

I tend to read from left to right so it does throw me off a bit that you usually discuss the right panel first.

We have rearranged our discussion such that the comparison is now made in the same order as the panels (see e.g. Section 4.4.1).

The units on the y-axis are hPa and I think it stops between 2 and 1 hPa.

There was indeed a typo in the manuscript. Figure 10 displays the speciation of NO_y as a function of altitude up to approximately 1 hPa.

I cannot see this subtle difference from Figure 10.

This comment refers to the following sentence: "At 10 hPa, HNO_3 constitutes 20% of total NO_y in C-GC but 23% in both C-CC and S-GC.". These percentages were not read from Figure 10 but rather calculated from the simulation output. We now state in the manuscript (line 689) that these values are not shown explicitly in the figure, although they can in theory be read from the figure data.

Why are there two different legends? Does this mean your NOy estimates have different make up between the GEOS-Chem and CAM-Chem models? What impact does that have on your profile comparisons in the previous figure?

The list of species that make up NO_y differs between GEOS-Chem and CAM-chem. Thus, S-GC and C-GC have the same list of species included in NOy, while it is a different one for C-CC. We now specify this when first mentioning Figure 10 (lines 663-664). The most significant difference is the lack of nitrate aerosols in CAM-chem, which affects the treatment of HNO₃ (see e.g. lines 688 to 695) and is discussed throughout the manuscript. Otherwise the speciation of NO_y does not appear to vary significantly between C-CC and C-GC (see e.g. lines 681-684).

I don't see this purple band on Figure 11.

This was a miscommunication; although N_2O_5 is an important component at altitudes above (pressures less than) 200 hPa, at altitudes below (pressures greater than) this point, PAN is the key component. The discussion has been simplified (lines 700-702) to no longer discuss N_2O_5 and instead focus on PAN.

Sometimes the authors use NOx and sometimes NO and NO2 but I don't think the authors have yet to define NOx as NO and NO2.

 NO_x is now first defined early in the manuscript (line 312) as NO and NO_2 . In the rest of the manuscript, we refer to nitrogen oxides as NO_x .

Can the C-CC legend be moved down slightly or stacked vertically so it doesn't overlap with the NOy speciation

We have regenerated this figure such that the legend does not overlap with the speciation.

For example, this seems redundant with what is in line 657. Can you better connect these two paragraphs as there is overlap in the message.

We have merged this paragraph into the previous one and attempted to reduce redundancy (lines 700 – 718).

Connect this paragraph with the previous paragraph. You give the reasoning for the result before you show/discuss the result. Alternatively, could Figure 12 and 13 be in a supplemental to support the reasoning you provide in Lines 680-686?

In order to reduce the length of the main manuscript, we have decided to move Figures 12, 13 and 14 to the Supplementary Information. The two paragraphs describing the results have been merged into the main text.

How does the difference in species between GC and CC impact these profiles? e.g., Br2

This is an important question given that GEOS-Chem and CAM-chem have different representation of bromine chemistry. The most critical difference is the lack of short-lived bromine source species in this version of CAM-chem, including sea salt, which means that bromine-related tropospheric ozone depletion is largely not captured. We now refer to this difference in the manuscript (lines 761-765).

I got tripped up reading this sentence because of the similar acronyms. Can you change this to "the C-GC total Bry concentration exceeds C-CC by 1000 % at the surface".

We agree with the reviewer's comment and have made the corresponding change.

What do you mean here? Is the slope flattening or steepening?

We agree that this sentence was confusing. We have now rewritten it as "Above 100 hPa, the averaged Br_y mixing ratio levels off, with values between 20 hPa and 2 hPa remaining roughly constant in the range of 16-20 pptv" (lines 767-768).

above 80 hPa for C-CC comparison.

We have added the following statement in the manuscript: "(and even above 80 hPa when compared to C-CC)" (lines 770-771).

in GEOS-Chem?

We now mention that this rapid washout occurs in C-GC and S-GC.

The BrNO3 also looks bigger, less BrCl.

We have added a sentence comparing BrNO₃ and BrCl in the mid stratosphere. This sentence reads as "Larger mixing ratios of BrNO₃ are also present in C-CC (approximately 10 ppbv at 30 hPa) compared to C-GC and S-GC (approximately 7.4 and 7.0 ppbv respectively at 30 hPa). Smaller mixing ratios of BrCl are observed in C-CC, with a mean value of 1.8 ppbv at 30 hPa, while they reach 3.1 ppbv in C-GC and S-GC at 30 hPa. The base causes of these differences are not clear, but may be related to the presence of more complex tropospheric and stratospheric halogen chemistry in the GEOS-Chem chemical mechanism (Wang et al., 20210" (lines 786-790).

"vertical" is redundant as profiles are not in the horizontal.

We agree with the reviewer and we have removed the word "vertical".

Why?

We decided to exclude long-lived chlorocarbons from the analysis of the profiles of reactive chlorine because they have longer lifetimes. However, they are of course included in the simulation.

How do the differences in species representing Cly impact the comparisons in Figure 17?

We find that above 500 hPa, all three simulations have similar chlorine speciation and species not included in CAM-chem do not have major contributions above 500 hPa. Below 500 hPa, the main differences are due to chlorine from sea salt, which is not represented in CAM-chem (~90% of the surface chlorine content, excluding chlorocarbons) (line 829-830).

I do not follow this connection.

We clarified the sentence. It now reads as "As with total Br_y , total Cl_y follows the same vertical distribution as S-GC up to 10 hPa. Above this pressure, the vertical distribution in C-GC is closer to that of C-CC" (lines 815-816).

I read the figures to have negative differences in the middle panel above 10 hPa. **and** Isn't this a negative difference in the right panel of Figure 17? and starting at 200 hPa for C-GC minus C-CC

These are indeed negative differences. The sentence now reads as "Above 10 hPa, the relative difference in Cl_y between C-GC and S-GC increases slowly from -2% at 10 hPa to -5% at 2 hPa, while the difference relative to C-CC remains at approximately -20% above 200 hPa" (lines 817-819).

How can you say this [a statement on production] when looking at a global annual mean? **and** The HCl and CINO3 look similar to me when comparing the left and right plots below 200 hPa (upper troposphere). What are you considering for the region to be lower stratosphere?

We find higher mixing ratios of HCI in C-CC between 200 hPa and 50 hPa. For instance, at 50 hPa, we find a mean HCI mixing ratio of 0.8 ppbv in both C-GC and S-GC, but 1.1 ppbv in C-CC. By the expression "appears to occur faster", we meant to say that there's greater mixing ratios of HCI at these altitudes in C-CC. We now state that "larger mixing ratios of upper tropospheric and lower stratospheric HCI from chlorine source compounds are observed in C-CC" (lines 831-832).

Is there a reason to not use CO? Has it been defined yet?

Carbon monoxide (CO) had not been defined previously. This sentence now reads as "Section 5.3 evaluates the level of agreement of simulated ozone and carbon monoxide (CO) columns to measurements from the OMI/MLS and MOPITT satellite instruments".

I missed the fact at first that each row was a different region. I highly recommend adding a detail to this sentence like "for North America (top row), Europe (middle row), and South-east Asia (bottom row)." Are the bounding boxes for these regions provided else where in the paper? If so remind the reader, if not add these details.

We now state that each row corresponds to a specific region. The sentence now reads "Figure 17 compares surface mass concentrations of NO_2 as estimated by C-GC, S-GC, and C-CC for 2016 against ground station measurements for North America (top row), Europe (middle row), and South-East Asia (bottom row)".

Have you tried comparing it to a "NOy" from the model to see if you get a better comparison?

This is an interesting point and worth studying. However, we have not considered this possibility for the present study. Evaluating NOy model results against NO₂ observations would maybe bypass the consistent overestimation that in-situ monitors generate due to HNO₃. In Section 5.1, we have added the following sentence: "Comparing in-situ NO₂ measurements against NO_y model results could potentially remove the effect of interferants in the observations of surface NO₂ concentrations, but is not considered here" (lines 870-872).

While Figure 19 clearly shows the models underestimate observations across the board, this regional discussion could be provided as a Table and I would then recommend Figure 19 to a supplemental. Other statistics like mean bias, RMSE to go with the correlation values should be considered (see Figure 3 of Keller et al., 2021 JAMES 10.1029/2020MS002413). Also keep in mind that comparing model grid boxes to point source measurements should always come with a caveat in the text that we do not expect perfect matches. I think you are saying something like that in line 828 but not specifically highlighting the grid-box representation errors.

I see later on you have the mean biases. I highly recommend a table for when you have this many numbers that you are quoting in the main text.

We agree with the reviewer's comment and now state in Section 5.1 "By comparing model results at an approximately 2° horizontal resolution to point observations, we expect some differences due to grid-box representation errors.". Additionally, we provide new tables (Tables 8 and 9) listing correlation coefficients of surface-level NO₂ and ozone mass concentrations.

Are the NO and NO2 panels discussed? If not, can they be moved to a supplemental?

We now only present the panels corresponding to the NO:NO₂ ratio, while the previous figure has been moved to the SI (Figure S3).

What about referencing Figure 4 instead?

We now reference Figure 4 as suggested.

Should we be able to see these in the NO2 or NO plots? Would a better color bar show this (e.g., log scale or less saturation below 5 ppbv)?

The new Figure in the SI (Figure S3) plots NO_2 and NO_x on a log scale. Ship tracks can be observed for all three models.

Again, maybe a table would be more useful? or at least put the r values as insets on the panels?

We have added a new table listing the correlation coefficients between surface-level simulation results for ozone and measurements (Table 9).

The authors have already presented a lot of comparisons between the two models. Can the authors be more specific as to what further work needs to be done.

We now state throughout the paper different points that could be envisioned for future work (e.g. NUOPC-based interface, bromine sea salt emissions scaled with sea salt emissions, source of the differences in surface sulfate concentrations, role of heterogeneous chemistry on nitrogen speciation).

Why not use available ozonesonde data for 2016?

Since our goal was to show that CESM-GC produces reasonable results compared to CESM and GEOS-Chem, we used the analysis and processing codes (and datasets) which are typically used in evaluations of CESM and CAM-Chem. Several of the figures shown in the manuscript (including the ozonesonde comparison) have been generated using the CESM post-processor, which directly compares ozone profiles to a climatology of ozone sonde observations from 1995 to 2010. Even though a new climatology came out in 2016, the CESM post-processor still uses the previous dataset. To clarify, the manuscript now includes the time range of the ozone sonde measurements (line 922). We also clarify in the same section the reason for using climatology rather than a specific year.

We found that the choice of restart really made a difference on our stratospheric ozone in the GEOS-CF (GEOS ESM with GEOS-Chem chemistry) (see Section 4, Knowland et al., 2022 JAMES 10.1029/2021MS002852). How well spun up was the stratosphere in the restart file provided from version 13.1.2 GEOS-Chem (line 390)? What is the stratosphere like in the CESM initial conditions (line 389)?

The GEOS-Chem restart file was obtained from a 10-year simulation using GEOS-Chem Classic on a 4°x5° horizontal grid, which was then regridded to the horizontal grid used for this study. We agree that performance of the model stratosphere will be influenced by the restart file chosen, but we do not evaluate that influence in this work. In Section 3, we now state "For C-CC, the standard restart file provided with CESM is used to provide initial conditions. For S-GC, we use a restart file provided with version 13.1.2 of the GEOS-Chem chemistry module, which was obtained from a 10-year simulation. The CESM restart file is intended to represent the early 21st century, so we have followed the lead of previous studies which have used a 1-2 year spin up period (Schwantes et al., 2022; He et al., 2015)" (lines 402-406).

Figure 23 caption states that the observations are for a different period than the model but i do not read that here. State that these comparisons are to climatologies, like in the ozonesondes. My question again though is why not use the actual observations for this year as they are available?

We now address this issue directly in the text (lines 922-924 and 947-956). We agree however that an additional evaluation looking at the performance for a specific year would be a useful supplementary analysis.

Is this shown in Figure 23? If not, state so.

It is not, and we now state this explicitly in the text.

not shown

We now state that these data are not shown in Figure 23 (now Figure 20).

not shown. Again, would these numbers make for a good table?

We now state that this number is not shown in Figure 23 (now Figure 20). Although we agree that a table would be helpful, we have chosen not to create one so as to avoid further lengthening the manuscript.

This should have been defined at first use and then CO can be used throughout. The authors do not make a confusing reference to Colorado which would make using CO for carbon monoxide confusing.

Carbon monoxide (CO) is now defined on first use in Section 5. All other references to carbon monoxide have been replaced with CO.

of CAM-Chem

The sentence has been modified accordingly and now reads as "The CO model estimates using C-CC are characterized by a bias of -9×10¹⁷ molec/cm² in the Northern Hemisphere, consistent with previous evaluations of CAM-chem" (lines 962-970).

Capitalize [Southern Hemisphere]

We have now capitalized Southern and Northern Hemisphere throughout.

I don't follow this statement.

We have clarified the sentence. It now reads as "Across all three model configurations a northsouth gradient is observed in the model bias, with the bias in the southern hemisphere being approximately 10¹⁸ molec/cm², which is of greater magnitude than the (negative) bias in the northern hemisphere."

Are these numbers on Figure 25?

The numbers are included in Figure 25 (now Figure 22), but respectively rounded to 0.6, 0.7 and 0.7.

Is this in a figure?

We have decided to not show the dry deposition fluxes of nitrogen for all three simulations, but rather state the results in the manuscript. The dataset we used has few measurements of dry deposition fluxes and we decided to not include this figure, as the results can be best described in a few sentences. This is now stated in the text in Section 5.4.

Are these numbers from the Figure 26?

These numbers represent the global mean for each of the simulations and they represent thus the mean of the panels on the left in Figure 26 (now Figure 23). This is now stated in the text in Section 5.4.

Are these numbers in the Figure?

The correlation coefficients between model results are not displayed in the figure.

Did the authors expect these numbers to match?

Wet deposition of non-sea salt sulfur is handled by MAM in C-CC and C-GC. Since the same representation of aerosol microphysics and wet deposition is used for sulfates in both C-CC and C-GC, we expected these numbers to be similar.

Am I reading this correctly. Right hand panels, mean of the red dots? The maximum for the model seems less for S-GC than the other two models.

The reviewer is correct. The maximum for S-GC is lower than for other models. We have modified the sentence to "For instance, over North America, measurements indicate a mean sulfur wet deposition flux of approximately 5 kg S/ha/year (for the year 2005), while the results at the same stations are lower with the slope of the linear fit equal to 0.2, 0.1, and 0.2 for C-GC, S-GC and C-CC respectively" (lines 1015-1017).

not only university groups but also government agencies for research

The sentence has been modified accordingly and now reads as "GEOS-Chem is presently used and developed worldwide for research by over 100 university groups and government agencies" (line 1058).

A Data Availability Statement is missing. In the main text, there is no reference to the source of the observation datasets for the ozonesondes, OMI, MLS or MOPITT. Web address and DOIs should be provided.

A Data Availability Statement is now included below the Code availability statement. Web addresses and DOIs are now provided both in said statement, and we provide references (e.g. Deeter et al., 2014 for MOPITT data and Ziemke et al., 2011 for OMI/MLS data) in the main text.

Incomplete reference [Fast-JX]

The reference in question has been removed, as there is no journal paper to cite.

Reviewer #2:

Fritz et al present a landmark overview of a well-designed and implemented configuration of the Community Earth System Model incorporating the GEOS-Chem module. This constitutes a significant technical achievement and represents a very impressive step forward in model capability. This paper fits well within the scope of GMD and I believe is suitable for publication after consideration of the manuscript structure and some further thought is given to the level of detail of the discussion.

As the authors note on L377, the use of the same host ESM allows the differences in results to be attributed to the two chemistry modules. The new configuration offers the possibility to perform interesting chemistry module intercomparisons and offers the possibility to work towards a better understanding of the role of the chemistry scheme, and other processes connected to chemistry (aerosol processes, wet and dry deposition), in determining model performance and intermodel differences.

At this stage, that goal is still someway off, which is understandable given that this is the first paper from this project. There remains a number of differences in the implementation of key processes that inevitably lead to intermodule differences, and it will require further work to unpick the role of, say, the different dry deposition or aerosol schemes in driving differences between the two modules. The CESM-GEOS-Chem framework does allow this work to begin, but the authors might wish to say more about what possibility exists to harmonise further these key processes between modules and to further increase the modularity of the chemistry schemes. This would better facilitate being able to swap between chemistry module process-level treatments to improve attribution which is an important goal, and I would say is the most important potential outcome of this work.

This is a big paper that is doing the work of two or three: it is a description of the technical changes required, a description of the model configurations, a model/module intercomparison paper and a model evaluation paper. This is not to criticize, but it does serve to illustrate the rather huge task of the essential role of model description and evaluation. However, I do wonder if the paper has become rather overlong.

We have worked to reduce the length of the paper by moving several figures to the Supplementary Information, while also aiming to improve the paper's clarity and structure. Section 3 now only discusses the model setup, while Section 4 (previously 3.2) and Section 5 (previously 3.3) go into detail about the model intercomparison and evaluation against observations respectively. Our hope is that this will help the reader to navigate and reduce fatigue.

The evaluation itself is often rather cursory and little is gone into in detail. This puts the success of the paper in some jeopardy - the scope is impressive but the level of detail occasionally leaves the reader hanging, and for specialists it does run the risk of being rather unsatisfactory.

We understand the reviewer's concern, and addressing this has been a significant focus of our revisions. As mentioned above, we have aimed to improve the structure and have moved material which is not critical to the paper into the SI, with the goal of streamlining the reader's experience. However, we have also aimed to improve the level of detail, as will hopefully be evident from our response below. This includes providing more information on total emissions; providing additional context on the likely chemistry causing differences observed between models; and breaking out diagnostic data such as correlation coefficients into separate tables so that they can be more readily accessed and interpreted by specialists. We hope that this has helped both to make the work more accessible to a broad audience and to improve its interest to specialist readers.

The paper performs a comparison between model configurations using zonal mean O3, surface O3, aerosol mass concentration, NOy, Bry and Cly, as well as an evaluation agains observations of surface NOx,O3, ozone profiles (2016 model year vs climatology), satellite O3 (2004-2010 period for troposphere, stratosphere and total column), total CO column (2016 vs 2003-2013 climatology) and wet/dry deposition

(fluxes at various stations 2005-2007). The use of different observational periods for the intercomparison could presumably be addressed with a longer transient, but the text is reasonably caveated on this point.

We understand the reviewer's concern on this point. We have tried to, wherever possible, clarify which observations and which time period are used to compare against model results. In our study, we have used some tools provided by NCAR to post-process results from C-GC and C-CC. Since our goal was to show that CESM-GC produces reasonable results compared to CESM and GEOS-Chem, we used the analysis and processing codes (and datasets) which are typically used in evaluations of CESM and CAM-Chem. Several of the figures shown in the manuscript (including the ozonesonde comparison) have been generated using the CESM post-processor, which directly compares ozone profiles to a climatology of ozone sonde observations from 1995 to 2010.

The intermodule/model comparison is really interesting. The paper describes a whole atmosphere chemistry scheme, and so some whole-atmosphere evaluation is performed, particularly for O3/NOy/Bry/Cly. I think the impact of the structural differences in the model is probably the main result in this paper - wet deposition and Cly/Bry sources are frequently mentioned - so breaking discussion down into C-GC vs C-CC for most of the evaluation and considering in a separate section the offline S-GC runs might make things a bit simpler to follow, not least as there are huge differences arising from the different meteorologies that frequently dominate the S-GC runs, making the comparison not one between modules but more between models at a high level, i.e. between CTM-style offline meteorology and GCM-style free-running experiments, which is interesting but perhaps muddies the waters.

In light of this comment, we experimented with multiple different possibilities for the document structure. Although we agree that the proposed structure (C-GC v C-CC first, with S-GC comparisons separated out) would likely make parts of the analysis much cleaner, we found that it was difficult to avoid repetition in diagnosing model differences. We have therefore opted instead to split the intermodel and intermodule comparison into its own Section (now Section 4), with the comparison of model results to observations now separated into Section 5. We have also worked to improve the degree to which the sections are "signposted", so that interested readers can more easily find the component of the analysis which is most relevant to them.

Better understanding the drivers of inter-module differences would be welcome. I think the manuscript would be improved significantly by examining not just the levels of key species but also the factors controlling the level of their reservoirs in more detail. The manuscript would be improved significantly if this would go further and address the species' budgets, quantifying the inputs and outputs between the modules. While biogenic emissions are compared, it would help - from an ozone evaluation point of view - to add data on other ozone precursors such anthropogenic, soil and LNOx to this table. Similarly, sink terms in the ozone budget would also be beneficial. A table similar to Table 1 in Tilmes et al. Geosci. Model Dev., 9, 1853–1890, 2016 would be ideal for the purposes of comparison. Putting more results into such tables would be helpful for the specialist reader.

We agree and think that this is a valuable addition to the paper. We have thus sought to extract more information from our model configuration data which might inform at least the source magnitudes. Unfortunately, we have only limited output data available from the original simulations (which we do not have the resources to re-run), and are thus not able to extract much additional data such as loss rates and lifetimes (in particular, we do not have this data consistently across all three). However, we were able to provide more information on total emissions. The new Table 2 provides total annual NO_x emissions from anthropogenic, soil, and lightning sources; the new Table 4, shows total annual surface emissions of key aerosols (sulfates, primary organic matter, and black carbon). The values from the model results are consistent with the ones provided in the study from Tilmes et al. (2016). These complement the existing data and tables showing biogenic VOC emissions (Table 3), sea salt emissions (Table 5), dust emissions (Section 3 text), and the burden data shown throughout the results sections.

Similarly, ideally, where key parameters or processes are identified, it may be useful to add references that indicate how the model configurations/chemistry was tuned/optimised when that model configuration was produced (e.g. is it possible to say how the sulfate dry deposition was evaluated originally in CESM2 and GEOS-Chem that means the deposition rates are so different?). This would give some traceability of the model configuration to the evaluation paper.

We have added throughout the manuscript references to the key model papers which describe the technical basis and implementation of specific processes. This includes references to the relevant implementations of stratospheric chemistry (lines 487-488) and halogen chemistry (lines 493-495), and specific references to key differences where appropriate (e.g. lines 788-790).

Specific points

The level of detail is rather variable in section 2.1

We have modified Section 2.1 to try and maintain a more consistent level of detail. In addition, we have attempted to streamline the manuscript by avoiding duplication of information in Sections 2 and 3.

Figure 1 and L161-165 - I am not sure of the timing of the various calls to dynamics, physics and chemistry - can the authors expand on why dynamics does not modify the atmospheric state in the diagram? What order are the routines (physics/chemistry) called in?

The result of dynamics is not directly applied through the control layer but rather acts on a "dynamics container" which are then translated to tendencies. This is now stated in the caption of Figure 1. We have now added a brief but more detailed description of the order of calls in the Supplemental Information.

Section 2 would benefit from a summary table that lists configurations side by side, e.g. aerosol scheme, dry deposition, as in e.g. the supplementary to Turnock et al., Atmos. Chem. Phys., 20, 14547–14579, 2020.

We have added a table summarizing the model configuration (Table 6).

Section 2.3.4 repeats some of the detail in L328 and L185.

We agree that this was somewhat redundant. We have now removed the second occurrence of this statement.

L366 missing words after to ensure

The sentence now reads as "Although a complete copy of the GEOS-Chem source code is downloaded from the version-controlled remote of GEOS-Chem repository (to ensure that the most-recent release of GEOS-Chem is used), not all files present in the GEOS-Chem source code directory are compiled" (lines 377-379)

L470-481 if the authors prefer to keep the three-panel structure (see comment above) it would be helpful to describe the figures in the same order that they are presented (L-R)

We agree with the reviewer. We have kept the same order for all Figures (C-GC, S-GC, C-CC) and we have thus modified the text to describe the results in that order. We also now label all figure panels.

L587 'emission' regions?

We have made the corresponding modification (line 626).

L617 what understanding of the ozone and aerosol do the subsequent analyses aim to improve? What beyond assessment of model skill does the comparison with observation aim to do?

The subsequent analysis (i.e. Section 5) aims to evaluate the model results against both surface ozone concentration measurements and vertical profiles so that we can understand what the dominant factors are in surface ozone when simulated by C-GC, and whether C-GC broadly moves us away from or towards the observations relative to either model. In this sense the reviewer is correct that this paper is concerned more with model skill than with a deeper understanding of atmospheric chemistry, which we hope will be gained through future applications of the model.

L648 are the aerosol reactive tendencies stored? Can this be further assessed?

The aerosol reactive tendencies were unfortunately not stored when the simulations were performed. Further assessment of the role of heterogeneous chemistry on NO_y partitioning in C-GC would be an interesting follow-up to the present study. We now state this in Section 4.4.1.

L657 reads strangely

The corresponding sentence has been split in two and now reads as : "However, between 200 and 900 hPa the dominant contributors are HNO_3 and PAN [peroxyacetyl nitrate]. In this pressure range, the C-GC and S-GC simulations also show a significant contribution from nitrate aerosol (NIT) and $BrNO_3$."

We would again like to thank the reviewers for their time and insight, and believe that their input during this review process has improved the paper substantially. Thank you again for considering our manuscript for publication in *Geoscientific Model Development*.

Best wishes,

Sebastian Eastham

1 Implementation and evaluation of the GEOS-Chem chemistry module

2 version 13.1.2 within the Community Earth System Model v2.1

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Short summary. We bring the state-of-the-science chemistry module GEOS-Chem into the Community Earth System Model (CESM). We show that some known differences between results from GEOS-Chem and CESM's CAM-chem chemistry module may be due to the configuration of model meteorology rather than inherent differences in the model chemistry. This is a significant step towards a truly modular ESM and allows two strong but currently separate research communities to benefit from each other's advances.

18 Abstract. We implement the GEOS-Chem chemistry module as a chemical mechanism in the Community Earth System Model 19 version 2 (CESM). Our implementation allows the state-of-the-science GEOS-Chem chemistry module to be used with 20 identical emissions, meteorology, and climate feedbacks as the CAM-chem chemistry module within CESM. We use coupling 21 interfaces to allow GEOS-Chem to operate almost unchanged within CESM. Aerosols are converted at each time step between 22 the GEOS-Chem bulk representation and the size-resolved representation of CESM's Modal Aerosol Model (MAM4). Land 23 type information needed for dry deposition calculations in GEOS-Chem is communicated through a coupler, allowing online land-atmosphere interactions. Wet scavenging in GEOS-Chem is replaced with the Neu and Prather scheme, and a common 24 25 emissions approach is developed for both CAM-chem and GEOS-Chem in CESM.

26

We compare how GEOS-Chem embedded in CESM (C-GC) compares to the existing CAM-chem chemistry option (C-CC) when used to simulate atmospheric chemistry in 2016, with identical meteorology and emissions. We compare atmospheric composition and deposition tendencies between the two simulations and evaluate the residual differences between C-GC compared to its use as a standalone chemistry transport model in the GEOS-Chem High Performance configuration (S-GC). We find that stratospheric ozone agrees well between the three models with differences of less than 10% in the core of the ozone layer, but that ozone in the troposphere is generally lower in C-GC than in either C-CC or S-GC. This is likely due to 33 greater tropospheric concentrations of bromine, although other factors such as water vapor may contribute to lesser or greater 34 extents depending on the region. This difference in tropospheric ozone is not uniform, with tropospheric ozone in C-GC being 35 30% lower in the Southern Hemisphere when compared to S-GC but within 10% in the Northern Hemisphere. This suggests 36 differences in the effects of anthropogenic emissions. Aerosol concentrations in C-GC agree with those in S-GC at low altitudes 37 in the tropics but are over 100% greater in the upper troposphere due to differences in the representation of convective 38 scavenging. We also find that water vapor concentrations vary substantially between the standalone and CESM-implemented 39 version of GEOS-Chem, as the simulated hydrological cycle in CESM diverges from that represented in the source NASA 40 MERRA-2 reanalysis meteorology which is used directly in the GEOS-Chem CTM.

41

42 Our implementation of GEOS-Chem as a chemistry option in CESM (including full chemistry-climate feedback) is publicly 43 available and is being considered for inclusion in the CESM main code repository. This work is a significant step in the MUlti-44 Scale Infrastructure for Chemistry and Aerosols (MUSICA) project, enabling two communities of atmospheric researchers 45 (CESM and GEOS-Chem) to share expertise through a common modeling framework and thereby accelerate progress in 46 atmospheric science.

47 1. Introduction

48 Accurate representation and understanding of atmospheric chemistry in global Earth System Models (ESMs) has been 49 recognized as an urgent priority in geoscientific model development. The National Research Council (NRC) report on a 50 National Strategy for Advancing Climate Modeling (Bretherton et al., 2012) stresses the need for including comprehensive 51 atmospheric chemistry in the next generation of ESMs. The NRC report on the Future of Atmospheric Chemistry (NRC, 2016) 52 identifies the integration of atmospheric chemistry into weather and climate models as one of its five priority science areas. 53 This work responds to those needs, presenting the implementation of the state-of-science model GEOS-Chem (Bey et al., 2001; 54 Eastham et al., 2018) as an atmospheric chemistry module within the Community Earth System Model (CESM) (Hurrell et al., 55 2013; Tilmes et al., 2016; Lamarque et al., 2012; Emmons et al., 2020).

56

57 GEOS-Chem is a state-of-the-science global atmospheric chemistry model developed and used by over 150 research groups 58 worldwide (http://geos-chem.org). It has wide appeal among atmospheric chemists because it is a comprehensive, state-of-59 science, open-access, well-documented modeling resource that is easy to use and modify but also has strong central 60 management, version control, and user support. The model is managed at Harvard by a GEOS-Chem Support Team with oversight from an international GEOS-Chem Steering Committee. Documentation and communication with users is done 61 62 through extensive web and wiki pages, email lists, newsletters, and benchmarking. Grass-roots model development is done by 63 users, and inclusion into the standard model is prioritized by Working Groups reporting to the Steering Committee. The model 64 can simulate tropospheric and stratospheric oxidant-aerosol chemistry, aerosol microphysics, and budgets of various gases . Simulations can be conducted on a wide range of computing platforms with either shared-memory (OpenMP) or distributed
 memory (MPI) parallelization – with this latter implementation referred to as GEOS-Chem High Performance, or GCHP
 (Eastham et al., 2018).

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For the general atmospheric chemistry problem involving *K* atmospheric species coupled by chemistry and/or aerosol
 microphysics, GEOS-Chem solves the system of *K* coupled continuity equations

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$$\frac{\partial n_i}{\partial t} = -\nabla \cdot (n_i \mathbf{U}) + P_i(\mathbf{n}) - L_i(\mathbf{n})$$
(1)

where $\mathbf{n} = (n_1, ..., n_K)^T$ is the number density vector representing the concentrations of the *K* species, **U** is the 3-D wind vector, and P_i and L_i are local production and loss terms for species *i* including emissions, deposition, chemistry, and aerosol physics. The transport term $-\nabla \cdot (n_i \mathbf{U})$ includes advection by grid-resolved winds as well as parameterized subgrid turbulent motions (boundary layer mixing, convection). The local term $P_i(\mathbf{n}) - L_i(\mathbf{n})$ couples the continuity equations across species through chemical kinetics and aerosol physics.

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80 Standard application of the GEOS-Chem model as originally described by Bey et al. (2001) is off-line, meaning that the model 81 does not simulate its own atmospheric dynamics. Instead, it uses analyzed winds and other meteorological variables produced 82 by Goddard Earth Observation System (GEOS) simulations of the NASA Global Modeling and Assimilation Office (GMAO) 83 with assimilated meteorological observations. The near-real time GEOS Forward Processing (GEOS-FP) output provides data 84 globally at a horizontal resolution of $0.25^{\circ} \times 0.3125^{\circ}$, and the Modern-Era Retrospective analysis for Research and Applications, 85 Version 2 (MERRA-2) provides data at 0.5°×0.625°. GEOS-Chem simulations can be conducted at that native resolution or 86 at coarser resolution (by conservative re-gridding of meteorological fields). Long et al. (2015) developed an on-line capability 87 for GEOS-Chem to be used as a chemical module in ESMs, with initial application to the GEOS ESM. In that configuration, 88 GEOS-Chem only solves the local terms of the continuity equation

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$$\frac{\partial n_i}{\partial t} = P_i(\mathbf{n}) - L_i(\mathbf{n})$$
(2)

and delivers the updated concentrations to the ESM for computation of transport through its atmospheric dynamics. On-line
 simulation avoids the need for a meteorological data archive and the associated model transport errors (Jöckel et al., 2001; Yu
 et al., 2018). It also enables fast coupling between chemistry and dynamics.

95

96 Transformation of GEOS-Chem to a grid-independent structure was performed transparently, such that the standard GEOS-97 Chem model uses the exact same code for on-line and off-line applications. This includes a mature implementation within the

- GEOS ESM. It was applied recently to a year-long tropospheric chemistry simulation with ≈12 km (cubed-sphere c720) global
 resolution (Hu et al., 2018), and is now being used for global atmospheric composition forecasting (Keller et al., 2017, 2021).
 However, the only implementations of GEOS-Chem which are currently publicly available are either designed to run "offline",
 driven by archived meteorological data from the NASA Goddard Earth Observing System (GEOS) (Bey et al., 2001; Eastham
 et al., 2018), or operate at regional scale and do not extend to global simulation (Lin et al., 2020; Feng et al., 2021).
- 103

104 Integration of GEOS-Chem as a chemistry option within an open-access, global ESM responds to the aforementioned calls 105 from the NRC. One of the most widely used open-access ESM is the Community Earth System Model (CESM) (Hurrell et al., 106 2013). CESM is fully coupled and state-of-science. It produces its own meteorology based on fixed sea surface temperatures 107 or with a fully interactive ocean model. It can also be nudged to observed meteorology including from GEOS. The CESM 108 configuration with chemistry covering the troposphere and stratosphere is referred to as CAM-chem (Community Atmosphere 109 Model with chemistry) (Tilmes et al., 2016; Lamarque et al., 2012), CAM-chem is a state-of-science model of atmospheric 110 chemistry; it has participated (along with CESM's WACCM model which extends to the lower thermosphere) in many 111 international model intercomparison activities such as ACCMIP, CCMI, POLMIP, HTAP2, GeoMIP and CMIP6, and has a 112 large international user community. CAM-chem also has a very different development heritage from GEOS-Chem, with each 113 model providing better performance in comparison to observations in different areas (Park et al., 2021; Emmons et al., 2015; 114 Nicely et al., 2017; Jonson et al., 2018). It is widely used for simulations of global tropospheric and stratospheric atmospheric 115 composition, in part because it is able to run either with specified meteorological datasets or with fully-coupled physics 116 (https://www2.acom.ucar.edu/gcm/cam-chem).

117

118 The fundamental differences in implementation of almost every atmospheric process between GEOS-Chem and CAM-chem 119 mean that it is difficult to disentangle the root causes of these differences. Modular Earth system models can resolve this issue. 120 Allowing individual scientific components to be swapped freely allows researchers to evaluate exactly what effect that 121 component has in isolation, while also giving a single user base access to a larger portfolio of options. If two different models 122 each implement five processes in different ways, a researcher must learn to use both in order to compare their results and 123 cannot isolate the effect of any one process with confidence. If process options are implemented in the same framework, this 124 problem is avoided. Such modularity is becoming increasingly possible with the availability of Earth system infrastructure 125 such as the Earth System Modeling Framework (ESMF) and the National Unified Operational Prediction Capability (NUOPC), 126 which describe common interfaces for Earth system modeling components (Hill et al., 2004; Sandgathe et al., 2011). The 127 Multi-Scale Infrastructure for Chemistry and Aerosols (MUSICA) builds upon this trend with process-level modularization, 128 with the goal of allowing researchers to select from a range of community-developed options when performing atmospheric 129 simulations.

130

131 This work integrates the GEOS-Chem chemistry module into CESM as an alternative option to CAM-chem. Our 132 implementation allows researchers to select either model to simulate gas-phase and aerosol chemistry throughout the 133 troposphere and stratosphere, while other processes such as advection, broadband radiative transfer, convective transport, and 134 emissions are handled nearly identically. We demonstrate this capability by comparing simulations of the year 2016 as 135 generated by GEOS-Chem and CAM-chem operating within CESM, with the chemical module being the only difference. 136 Estimates of atmospheric composition are compared between the two models and against a simulation in the standalone GCHP 137 chemistry transport model (CTM). Finally, we evaluate the accuracy of the three approaches against observations of 138 atmospheric composition and deposition.

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Section 2 provides a technical description of the implementation of GEOS-Chem into CESM. Section 3 then describes the model setup. Sections 4 and 5 present a one-year simulation (not including spin up) performed in CESM with GEOS-Chem; CESM with CAM-chem; and the standalone GEOS-Chem CTM. This includes model intercomparison (Section 4), and evaluation against surface and satellite measurements (Section 5).

144 **2.** Coupling between GEOS-Chem and CESM

145 We first describe the interface used within CESM when using either the CAM-chem or GEOS-Chem options (Section 2.1). 146 Unless otherwise stated, "GEOS-Chem" refers to the grid independent chemistry module which is common to all 147 implementations, including standalone GEOS-Chem with OpenMP (Classic) or MPI (GCHP) parallelization, NASA GMAO's 148 GEOS ESM, and WRF coupled with GEOS-Chem (WRF-GC). We then briefly summarize the chemistry and processes 149 represented by the CAM-chem and GEOS-Chem options within CESM (Section 2.2). This is followed by a description of 150 differences between the implementation of GEOS-Chem in CESM and its stand-alone code (Section 2.3), differences in the 151 data flow through CESM when using GEOS-Chem as opposed to CAM-chem (Section 2.4), and finally the installation and 152 compilation process (Section 2.5).

153 **2.1. Interface**

Our approach embeds a full copy of the GEOS-Chem chemistry module source code (version 13.1.2) within CESM (version 2.1.1). All modifications made to the GEOS-Chem source code have been propagated to the GEOS-Chem main code branch (<u>https://github.com/geoschem/geos-chem</u>) to ensure future compatibility between CESM and GEOS-Chem. Information is passed between the CESM Community Atmosphere Model (CAM) version 6 (CAM6) and the GEOS-Chem routines through an interface layer developed as part of this work. A schematic representation of the implementation is provided in Figure 1.

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162 Figure 1. Architectural overview of CESM when running with either the GEOS-Chem or CAM-chem chemistry options. The left section 163 shows the architecture of CESM, where the five major Earth system components are connected through the driver/mediator. The work 164 presented here changes only the contents of the atmosphere component (CAM). Regardless of the chemistry option used, dynamics. 165 physics, and emissions (HEMCO) are handled identically. Each component modifies the "Atmosphere State" while communication occurs 166 through the control layer. The choice of chemistry module is confined to the "Chemistry" subcomponent, where either CAM-chem or 167 GEOS-Chem can be chosen. In each case, data are transmitted between the "Atmosphere State" and the chemistry module, which 168 interacts in turn with the Modal Aerosol Model. Dynamics are shown separately as they act on a "dynamics container" rather than 169 directly on the atmospheric state. Further detail regarding timing of calls is provided in the Supplementary Information.

At each time step, CESM calls the coupling interface (referred to as CESM2-CG interface in Figure 1) which fills in the meteorological variables required by either CAM-chem or GEOS-Chem. Atmospheric transport and physics are identical whether using CAM-chem or GEOS-Chem to simulate atmospheric chemistry. The interface passes species concentrations from CAM to GEOS-Chem, which are then modified by GEOS-Chem and passed back to CAM. Meteorological data and land data are also passed to GEOS-Chem through the same interface. The routine calls in CAM when using either GEOS-Chem or CAM-chem are identical, with the appropriate chemistry module defined at compilation time such that the calls are routed to the appropriate routines.

177

The interface handles the conversion of meteorological variables and concentrations of atmospheric constituents between the state variables in CAM and those used in GEOS-Chem. Since GEOS-Chem operates in a "grid-independent" fashion, changes in the grid specification and other upstream modifications to CESM do not necessitate any changes to this interface (Long et al., 2015). Our version of CESM 2.1.1 is modified such that emissions are handled by the Harmonized Emissions Component (HEMCO), which operates independently of the chemistry module and can provide emissions data to either CAM-chem or
 GEOS-Chem equally (Lin et al., 2021).

184

The interface code is kept in a subfolder of chemistry source code (src/chemistry/geoschem subfolder), which also contains a copy of the source code for GEOS-Chem. Unlike the implementation of GEOS-Chem within GEOS, we do not use ESMF. However, we plan to develop a NUOPC-based interface as part of future work.

188 2.2. Processes represented by CAM-chem and GEOS-Chem

189 CAM-chem uses the Model for OZone And Related chemical Tracers (MOZART) family of chemical mechanisms to simulate 190 atmospheric chemistry (Emmons et al., 2020). The tropospheric-stratospheric MOZART-TS1 scheme which we demonstrate 191 in our intercomparison involves 186 gas-phase chemical species and includes stratospheric bromine and chlorine chemistry. 192 MOZART-TS1 does not include detailed tropospheric halogen chemistry or short-lived halogen sources such as sea salt 193 bromine, although these will be available in a future release (Badia et al., 2021; Fernandez et al., 2021). Photolysis rates are 194 calculated using a lookup table, based on calculations with the Tropospheric Ultraviolet and Visible (TUV) radiation model 195 (Kinnison et al., 2007). Wet deposition is calculated using the Neu and Prather (2012) scheme for both convective and large-196 scale precipitation. Dry deposition velocities over land are calculated for each land type by the Community Land Model (CLM) 197 in CESM using the Wesely (1989) resistance scheme with updates described by Emmons et al. (2020). Deposition velocities 198 over the ocean are calculated separately in CAM-chem. Aerosols are represented using the 4-mode Modal Aerosol Model 199 (MAM4), which includes sulfate, black carbon, primary, and secondary organic aerosols (SOAs) (Mills et al., 2016). 200 Ammonium and ammonium nitrate aerosols are calculated with a parameterization using the bulk aerosol scheme (Tilmes et 201 al., 2016). SOAs are simulated using a 5-bin volatility basis set (VBS) scheme, formed from terpenes, isoprene, specific 202 aromatics and lumped alkanes through reaction with OH, ozone and NO₃, with unique yields for each for each combination of 203 volatility and size bin (Tilmes et al., 2019). This more detailed scheme differs from the default MAM SOA scheme that is 204 used in CAM6 (without interactive chemistry). Aerosol deposition, including dry and wet deposition, and gravitational settling 205 (throughout the atmosphere) are calculated in the MAM code of CESM. CAM-chem also uses a volatility basis set (VBS) 206 approach for SOA with five volatility bins, covering saturation concentrations with logarithmic spacing from 0.01 to 100 µg.m⁻ 207 ³. CAM-chem explicitly represents Aitken and accumulation mode SOA using two separate tracers for each volatility bin but 208 does not include an explicit representation of non-volatile aerosol.

209

GEOS-Chem uses a set of chemical mechanisms implemented with the Kinetic PreProcessor (KPP) (Damian et al., 2002). The standard chemical mechanism has evolved continuously from the tropospheric gas-phase scheme described by Bey et al. (2001) and now includes aerosol chemistry (Park, 2004), stratospheric chemistry (Eastham et al., 2014), and a sophisticated tropospheric-stratospheric halogen chemistry scheme (Wang et al., 2019). The scheme present in GEOS-Chem 13.1.2 includes 299 chemical species. Additional "specialty simulations" such as an aerosol-only option and a simulation of the global mercury

215 cycle are present in GEOS-Chem but are not implemented into CESM in this work. Photolysis rates are calculated using the 216 Fast-JX v7 model (Wild et al., 2000). When implemented as the standalone model, wet deposition is calculated for large-scale 217 precipitation using separate approaches for water-soluble aerosols (Liu et al., 2001) and gases (Amos et al., 2012) with 218 calculation of convective scavenging performed inline with convective transport. A different approach is used to simulate wet 219 scavenging for the implementation of GEOS-Chem in CESM (see Section 2.3.4). Dry deposition is calculated using the Wesely 220 (1989) scheme (Wang et al., 1998), but with updates for nitric acid (HNO₃) (Jaeglé et al., 2018), aerosols (Jaeglé et al., 2011; 221 Alexander et al., 2005; Fairlie et al., 2007; Zhang et al., 2001), and over ocean (Pound et al., 2020). The representation of 222 aerosols in GEOS-Chem varies by species. Sulfate-ammonium-nitrate aerosol is represented using a bulk scheme (Park, 2004), 223 with gas-particle partitioning determined using ISORROPIA II (Fountoukis and Nenes, 2007). Modal and sectional size-224 resolved aerosol schemes are available for GEOS-Chem (Kodros and Pierce, 2017; Yu and Luo, 2009), but are disabled by 225 default and not used in this work. Sea salt aerosol is represented using two (fine and coarse) modes (Jaeglé et al., 2011), while 226 dust is represented using four size bins (Fairlie et al., 2007). We use the "complex SOA" chemistry mechanism in GEOS-227 Chem when running in CESM, as this uses a volatility basis set (VBS) representation of secondary organic aerosol which is 228 broadly compatible with that used in CAM--chem (Pye and Seinfeld, 2010; Marais et al., 2016; Pye et al., 2010). The complex 229 SOA VBS scheme uses four volatility bins covering saturation concentrations on a logarithmic scale from 0.1 to 100 μ g.m⁻³. 230 Two classes of SOA are represented in this fashion: those derived from terpenes (TSOA) and those derived from aromatics 231 (ASOA). For each "class" of SOA, two tracers are used to represent each volatility bin (one holding the gas phase mass, the 232 other holding the condensed phase mass). The only exception is the lowest-volatility aromatic aerosol, which is considered to 233 be non-volatile and therefore has no gas-phase tracer. Two additional SOA tracers, representing isoprene-derived and glyoxal-234 derived SOA, are not represented using a VBS approach.

235

Additional differences between the two chemistry modules include the use of different Henry's law coefficients, gravitational settling schemes, representation of polar stratospheric clouds, and heterogeneous chemistry. Full descriptions of the two models are available at <u>https://geos-chem.seas.harvard.edu/</u> and in Emmons et al. (2020).

239 2.3. Representation of atmospheric processes in GEOS-Chem when running in CESM

Some processes cannot be easily transferred from standalone GEOS-Chem to its implementation in CESM, due to factors such as the different splitting of convective transport in the two models. Processes which vary in their implementation between the standalone and CESM implementations of GEOS-Chem are described below.

243 **2.3.1.** Aerosol coupling in CESM with GEOS-Chem

Since GEOS-Chem and CESM use different approaches to represent aerosols, there is no straight-forward translation between
 the GEOS-Chem representation and that used elsewhere in CESM. We implement an interface between the CESM and GEOS-

- 246 Chem representations, so that GEOS-Chem's processing of aerosols is most accurately represented without compromising the 247 microphysical simulations and radiative interactions of aerosol calculated elsewhere in CESM.
- 248

249 CESM uses the 4-mode version of the Modal Aerosol Model (MAM4) to represent the aerosol size distribution and perform 250 aerosol microphysics (Liu et al., 2016). This represents the mass of sulfate aerosols, secondary organic matter (in five volatility 251 basis set bins), primary organic matter, black carbon, soil dust, and sea salt with advected tracers for each mode (accumulation, 252 Aitken, coarse, and primary carbon), although some species are considered only in a subset of the four modes. A tracer is also 253 implemented for the number of aerosol particles in each mode, resulting in a total of 18 tracers. As discussed above, GEOS-254 Chem instead represents sulfate, nitrate, and ammonium aerosol constituents with three tracers; fresh and aged black and 255 organic carbon with four tracers; fine and coarse sea salt as two tracers; and different sizes of dust with four tracers. Six 256 additional tracers are used to track the bromine, iodine, and chlorine content of each mode of sea salt aerosol, with two more 257 used to track overall alkalinity. Gas-phase sulfuric acid (H_2SO_4) is assumed to be negligible in the troposphere and is estimated 258 using an equilibrium calculation in the stratosphere (Eastham et al., 2014). The GEOS-Chem mechanism therefore represents 259 greater chemical complexity but reduced size resolution compared to the aerosol representation in MAM4.

260

Accordingly, when receiving species concentrations from CESM, the interface to GEOS-Chem lumps all modes of the MAM aerosol into the corresponding GEOS-Chem tracer. This includes gas-phase H_2SO_4 , in the case of the GEOS-Chem sulfate (SO₄) tracer. Aerosol constituents which are not represented explicitly by MAM (e.g. nitrates) are not included in this calculation. The relative contribution of each mode is stored during this "lumping" process for each grid cell. Once calculations with GEOS-Chem are complete, the updated concentration of the lumped aerosol is repartitioned into the MAM tracers based on the stored relative contributions in each grid cell.

267

For SOAs, additional steps are needed. For the bins covering saturation concentrations of $1 \mu g.m^{-3}$ and greater, we assume that 268 269 the relevant volatility bin in MAM4 is equal to the sum of the two classes in GEOS-Chem covering the same saturation 270 concentrations. For example, the tracers TSOA1 and ASOA1 in GEOS-Chem are combined to estimate the total quantity of 271 the Aitken and accumulation modes for species "soa3" in MAM4. Partitioning between the two modes (when transferring from 272 GEOS-Chem to MAM4) is calculated based on the relative contribution of each constituent to the total prior to processing by 273 GEOS-Chem. Partitioning between the two classes (when transferring from MAM4 to GEOS-Chem) is calculated based on 274 the relative contribution of each constituent to the total at the end of the previous time step. For the lowest-volatility species, 275 we split the lowest volatility bin concentrations (and non-volatile species) from GEOS-Chem between the two lowest 276 volatilities in MAM4. A full mapping for all species is provided in Table 1.

277

Table 1. Mapping between tracers used to represent SOA in GEOS-Chem and CAM-chem. Translation between GEOS-Chem and MAM4 is
 performed by preserving the relative contributions provided during the previous transfer.

_				
	GEOS-Chem	Mapping to	Saturation concentration	
	species	CAM-chem species	range (µg.m ⁻³)	Phase
	TSOA0 + ASOAN	$soa1_a1 + soa1_a2 + soa2_a1 + soa2_a2$	0 - 0.1	Aerosol
	TSOA1 + ASOA1	soa3_a1 + soa3_a2	0.1 - 1.0	Aerosol
	TSOA2 + ASOA2	$soa4_a1 + soa4_a2$	1.0 - 10	Aerosol
	TSOA3 + ASOA3	$soa5_a1 + soa5_a2$	10 - 100	Aerosol
	TSOG0	SOAG0 + SOAG1	0 - 0.1	Gas
	TSOG1 + ASOG1	SOAG2	0.1 - 1.0	Gas
	TSOG2 + ASOG2	SOAG3	1.0 - 10	Gas
	TSOG3 + ASOG3	SOAG4	10 - 100	Gas

280

Finally, MAM simulates some chemical processing on and in the aerosol. This includes the reaction of sulfur dioxide with hydrogen peroxide and ozone in clouds, which is already included in the GEOS-Chem chemistry mechanism. We therefore disable in-cloud sulfur oxidation in MAM4 when using the GEOS-Chem chemistry component in CESM, consistent with the GEOS-Chem CTM. A comparison of the effect of each approach is provided in the Supplementary Information.

285 **2.3.2.** Dry deposition

Dry deposition velocities over land are calculated in CESM for each atmospheric constituent by the Community Land Model (CLM) using a species database stored by the coupler. GEOS-Chem is also able to calculate its own dry deposition velocities (see Section 2.2), in situations where a land model is not available such as when running as a CTM. We thus implement different options to compute dry deposition velocities when running CESM with the GEOS-Chem chemistry option:

- Dry deposition velocities over land are computed by CLM and are passed to CAM through the coupler. They are then
 merged with dry deposition velocities computed over ocean and ice by GEOS-Chem, identical to the procedure used
 in CAM-chem. Each of these are scaled by the land and ocean/ice fraction respectively.
- 2. GEOS-Chem computes dry deposition at any location using the land types and leaf area indices from CLM, which
 are passed through the coupler.
- GEOS-Chem obtains "offline" land types and leaf area indices and computes the dry deposition velocities similarly
 to GEOS-Chem Classic.
- This allows researchers to experiment with different dry deposition options, ranging from that most consistent with the approach used in CAM-chem (option 1) to that most consistent with stand-alone GEOS-Chem (option 3). For this work we use option 2, but option 1 will be brought as standard into the CESM main code to reduce data transfer requirements.

300 **2.3.3.** Emissions

The Harmonized Emissions Component (HEMCO) is used to calculate emissions in standalone GEOS-Chem (Keller et al., 2014), and HEMCO v3.0 was recently implemented as an option for CAM-chem (Lin et al., 2021). HEMCO offers the possibility for the user to read, regrid, overlay, and scale emission fluxes from different archived emissions inventories at runtime. Emissions extensions allow for the computation of emissions that depend on meteorology or surface characteristics (e.g. lightning, dust emissions). Some extensions have also been designed to calculate subgrid-scale chemical processes, such as non-linear chemistry in ship plumes (Vinken et al., 2011).

307

308 The GEOS-Chem CTM implementations use archived ("offline") inventories of natural emissions, calculated at native 309 resolution using the NASA GEOS MERRA-2 and GEOS-FP meteorological fields. This ensures that the emissions are 310 calculated consistently regardless of grid resolution. These archived emissions fields can be used within CESM but we also 311 preserve the option for users to employ "online" emissions inventories where relevant. This enables feedback between climate 312 and emissions to be calculated. For instance, lightning nitrogen oxides (NO_x = NO + NO₂) emissions, dust and sea salt emissions, and biogenic emissions are all computed online using parameterizations from CAM and CLM. CAM computes 313 314 lightning NO_x emissions based on the lightning flash frequency, which is estimated following the model cloud height, with 315 different parameterizations over ocean and land. The NO lightning production rate in CAM is assumed proportional to the 316 discharged energy, with 10^{17} atoms of nitrogen released per Joule (Price et al., 1997). The lightning NO_x emissions are then 317 allocated vertically from the surface to the local cloud top based on the distribution described by Pickering et al. (1998). For 318 biogenic emissions, we use the online Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGANv2.1), as 319 established in CLM (Guenther et al., 2012). Aerosol mass and number emissions are passed directly to MAM constituents. 320 Global anthropogenic emissions can be specified from any of the standard GEOS-Chem inventories, but default to the 321 Community Emissions Data System (CEDS) inventory (Hoesly et al., 2018). Sulfur emissions from the CEDS inventory are 322 partitioned into size-resolved aerosol (mass and number) and SO₂ (Emmons et al., 2020). In CAM, volcanic out-gassing of 323 SO_2 is provided from the GEIA inventory with 2.5% emitted as sulfate aerosol (Andres and Kasgnoc, 1998), while eruptive 324 emissions are provided from the VolcanEESM database (Neely and Schmidt, 2016). The option is also available through 325 HEMCO to use the "AeroCom" volcanic emissions, which are derived from OMI observations of SO₂ (Ge et al., 2016; Carn 326 et al., 2015).

327

Although we use HEMCO with both model configurations, there remain differences between the representation of emissions in CAM-chem and in GEOS-Chem when run within CESM. This is because of differences in the species present in their respective mechanisms. For instance, emissions of iodocarbons (CH₃I, CH₂I₂, CH₂ICl, CH₂IBr) and inorganic iodine (HOI, I₂) are not available in CAM-chem since iodine is not explicitly modeled in the versions of CAM-chem available in CESM v2.1.1.

332 VOC lumping is also performed differently (see the Supplemental Information for more detail).

333

Where the emitted species are present in both chemical mechanisms, the emissions calculated by HEMCO in CESM are identical whether running with GEOS-Chem or CAM-chem. If the HEMCO implementations of lightning, dust, sea salt, and biogenic emissions are used, emissions will be identical between CESM and the standalone GEOS-Chem CTM.

337 2.3.4. Wet deposition and convection

For both GEOS-Chem and CAM-chem within CESM, convective scavenging and transport are handled separately. Unlike in the Liu et al. (2001) approach implemented in the GEOS-Chem standalone code, removal of soluble gases within convective updrafts is not explicitly simulated in either CAM-chem or GEOS-Chem when embedded in CESM. When using the CAMchem mechanism within CESM, the Neu scheme is used to perform washout of soluble gaseous species, while wet deposition of MAM aerosols is handled by MAM. When running CESM with the GEOS-Chem chemistry mechanism, the Neu scheme also performs wet scavenging for aerosols which are not represented by MAM4 (e.g. nitrate). For all such aerosols we assume a Henry's law coefficient equal to that for HNO₃.

345 2.3.5. Surface boundary conditions

In CESM, surface boundary mixing ratios of long-lived greenhouse gases (methane, N₂O, and chlorofluorocarbons) are set to the fields specified for CMIP6 historical conditions and future scenarios (Meinshausen et al. 2017). For whichever CMIP6 scenario is chosen, the boundary conditions overwrite those set by the GEOS-Chem chemistry module or by the HEMCO emissions component.

350 2.4. Changes to the data flow in CESM when running with GEOS-Chem

351 In CESM, data such as the Henry's law coefficients required to calculate dry deposition velocities and wet scavenging rates 352 for each species are defined at compile time. For species that are common to GEOS-Chem and CAM-chem but where these 353 factors differ, the GEOS-chem values are used by default. The CAM-Chem values are listed alongside them in the source code 354 to allow users to switch if desired. Additionally, we modify CAM, CLM and the Common Infrastructure for Modeling the 355 Earth (CIME) such that the land model can pass land type information and leaf area indices to the atmosphere model to compute 356 dry deposition velocities. This could be a potential solution for dry deposition of aerosols in MAM, which currently uses fixed 357 land types independent of the ones used in CLM (Liu et al., 2012). However, this comes at the cost of passing land information 358 through the coupler at every time step.

359 **2.5.** Installation and compilation process

The interface between CESM and GEOS-Chem, as well as the GEOS-Chem source code, is automatically downloaded when CAM checks out its external repositories. The versions of GEOS-Chem and of the coupling interface can be changed by modifying the `Externals_CAM.cfg` and by running the `checkout_externals` command. 363

364 When creating a new case, the user chooses the atmospheric chemistry mechanism (GEOS-Chem or CAM-chem). The 365 chemistry option is defined by the name of the CESM configuration (component set, or "compset"), making the process of 366 creating a run directory almost identical when choosing either GEOS-Chem or CAM-chem. Whereas chemistry options in 367 CAM-chem are set explicitly using namelist files, certain options in GEOS-Chem are set using ASCII text input files which 368 are read during the initialization sequence. The installation and build infrastructure of CIME will therefore copy any GEOS-369 Chem specific text input files to the case directory when setting up a simulation which includes GEOS-Chem. This currently 370 includes emissions specifications read by HEMCO, although this is expected to change as HEMCO becomes the standard 371 emissions option for both CAM-chem and GEOS-Chem in CESM (currently being discussed with the CESM team).

372

Although CESM supports both shared-memory parallelization (OpenMP) and distributed memory parallelization (MPI), GEOS-Chem implemented in CESM does not currently support OpenMP. When running CESM with the GEOS-Chem chemistry model, the number of OpenMP threads per MPI task is therefore set to one.

376

377 Although a complete copy of the GEOS-Chem source code is downloaded from the version-controlled remote of GEOS-Chem 378 repository (to ensure that the most-recent release of GEOS-Chem is used), not all files present in the GEOS-Chem source code 379 directory are compiled. For instance, the files pertaining to the GEOS-Chem advection scheme are not needed as advection is 380 performed by CAM, and therefore the GEOS-Chem advection routines are not compiled. To do this we implement a new 381 feature in CIME to use `.exclude` files which list files not needed during compilation. CIME reads each `.exclude` file at 382 compile time and searches subdirectories recursively from the location of the exclude file, preventing any named file from 383 being included in compilation. For example, an `.exclude` file is provided in the chemistry coupling interface folder for 384 GEOS-Chem that lists the files to exclude in the GEOS-Chem source code directories.

385 **3.** Setup used for model evaluation

386 We simulate a two-year period with GEOS-Chem embedded in CESM (hereafter C-GC). The simulation setup is described in 387 the present Section. Then, we perform a comparison of its output to that generated by two other model configurations (Section 388 4). By comparing the results to those produced for the same period by CESM with CAM-chem (hereafter C-CC), we can 389 perform the first comparison of GEOS-Chem and CAM-chem when run as chemistry modules within the same ESM. Any 390 differences between these two simulations can only be the result of differences between the two chemical modules and their 391 implementations in CESM. This includes not only differences in the gas-phase chemical mechanism, but also in the 392 implementation of photolysis calculations, heterogeneous chemistry, aerosol microphysics, and the chemical kinetics 393 integrator itself. We also compare output to that produced by the standalone GEOS-Chem High Performance model (hereafter S-GC). This enables us to evaluate the effect of using CESM's grid discretization, advection, aerosols, and representation of
 meteorology compared to that used in the GEOS-Chem CTM.

396

Lastly, we evaluate the performance of C-GC by comparing output to observational data (Section 5). We also include comparisons of data from the C-CC and S-GC configurations, to provide insight into the relative performance of the model and the root cause of disagreements with observations. This section describes the model configurations in detail, but a brief summary is provided in Table 6.

401

402 All simulations cover January 1st to December 31st 2016, with an additional 6 months (S-GC) or 1 year (C-GC/C-CC) of spin 403 up. For C-CC, the standard restart file provided with CESM is used to provide initial conditions. For S-GC, we use a restart 404 file provided with version 13.1.2 of the GEOS-Chem chemistry module, which was obtained from a 10-year simulation. The 405 CESM restart file is intended to represent the early 21st century, so we have followed the lead of previous studies which have 406 used a 1-2 year spin up period (Schwantes et al., 2022; He et al., 2015). For C-GC, we use initial conditions which are taken 407 from the S-GC restart file where possible, but fill missing species (e.g. MAM4 aerosol tracers) using data from the C-CC 408 restart file. Both simulations performed with CESM v2.1.1 (C-GC and C-CC) use a horizontal resolution of 1.9°×2.5° on 56 409 hybrid pressure levels, extending from the surface to 1.65 hPa. Aerosols are represented in CESM using the 4-mode version 410 of the modal aerosol model, MAM4 (Liu et al., 2012). In C-GC, we use the complex SOA chemistry scheme (Pve and Seinfeld, 411 2010; Pye et al., 2010; Marais et al., 2016). In C-CC, we use the MOZART-TS1 chemistry scheme (Emmons et al., 2020).

412

413 Standalone GEOS-Chem (S-GC) simulations are performed using the GEOS-Chem High Performance (GCHP) configuration, 414 using a C48 cubed-sphere grid (approximately equivalent to a 2°×2.5° horizontal grid) on 72 hybrid pressure levels extending 415 up to 0.01 hPa. In GCHP, chemistry is performed up to 1 hPa (approximately 50 km) with simplified parameterizations used 416 above that point. Aerosols are represented using GEOS-Chem's "native" scheme, without translation to or from MAM4. As 417 in C-GC, we use the complex SOA scheme.

418

419 All three model configurations are driven using meteorological data from MERRA-2. In S-GC all meteorological fields are 420 explicitly specified by MERRA-2, using the same 72-layer vertical grid. The only exception is the specific humidity in the 421 stratosphere, which is computed online. In C-CC and C-GC, we use the "specified dynamics" (SD) configuration of CAM6 in 422 which 3-D temperature, 3-D wind velocities, surface pressure, surface temperature, surface sensible heat flux, surface latent 423 heat flux, surface water flux, and surface stresses are provided by MERRA-2 on a truncated 56-layer vertical grid. These 424 variables are nudged with a relaxation time of 50 hours, resulting in a relatively "loose" nudging strength. All other fields (e.g. 425 cloud fraction) are computed using the CAM physics routines. This includes convection. Whereas S-GC computes convective 426 transport from archived convective mass fluxes and calculates scavenging within the updraft (Wu et al., 2007), convective 427 transport in both C-CC and C-GC is calculated in CAM6 using the CLUBB-SGS scheme for shallow convection (Bogenschutz

428 et al., 2013) and the Zhang-McFarlane scheme (Zhang and McFarlane, 1995) for deep convection. Scavenging within the 429 convective updraft is not simulated explicitly.

430

Water vapor in C-GC is initialized from the specific humidity "Q" restart variable, which is identical to the one used for C-CC; after this point humidity is calculated based on the moist processes represented explicitly in CAM's physics package. The GEOS-Chem CTM does not calculate water vapor in the troposphere, instead prescribing specific humidity directly from MERRA-2 output. Mixing ratios of water vapor in C-CC and C-GC are therefore identical to that in S-GC at initialization time, but from that point onwards may diverge.

436

Emissions are harmonized between the three models, with all three configurations using HEMCO to calculate emissions fluxes. Surface anthropogenic emissions are provided from CEDS and are identical between all three models, apart from small differences in effective emissions from ships due to parameterized plume processing (Vinken et al., 2011). Simulated anthropogenic and biomass burning surface emissions of nitrogen oxides are 128-132 Tg(N) in each of the three models. Aviation emissions are calculated in all three models based on the AEIC 2005 emission inventory, contributing a further 0.8 Tg(N) in addition to other species (Simone et al., 2013).

443

Lightning emissions are calculated in C-CC and C-GC using the online parameterization described in Section 2.3.3, while lightning emissions in S-GC are calculated using archived flash densities and cloud top heights (Murray et al., 2012). Total lightning NO_x emissions are 5.7-6.1 Tg(N) in all three models. A summary of the breakdown of NO_x emissions is provided in Table 2.

448

449 Table 2. Annual global anthropogenic, soil, and lightning NO_x emissions expressed in Tg(N)/year.

	C-GC	S-GC	C-CC
Anthropogenic + biomass burning	40.1	39.1	39.6
Soil	7.23	7.23	7.23
Lightning	6.05	5.82	5.71

450

Biogenic emissions are calculated in C-CC and C-GC using the embedded MEGAN emissions module in CESM, which differs
slightly from the implementation in S-GC and will produce different emissions due to different vegetation distributions. Total
biogenic emissions in S-GC and C-GC are shown in Table 3. In all three simulations we use the "AeroCom" volcano emissions
implemented in HEMCO.

455
456 Table 3. Annual global biogenic emission totals in GEOS-Chem implemented in CESM (C-GC) compared to in standalone GEOS-Chem (S 457 GC).

	Name in	C-GC	S-GC
Species	GEOS-Chem	(Tg/year)	(Tg/year)
Acetone	ACET	42.7	48.2
Acetic acid	ACTA	3.86	-
Acetaldehyde	ALD2	20.8	17.9
Lumped alkanes >= C ₄	ALK4	0.16	-
Ethylene	C2H4	30.4	-
Ethane	C2H6	0.34	0.21
Propane	C3H8	0.03	-
Formaldehyde	CH2O	5.14	-
Carbon monoxide	CO	88.8	-
Ethanol	EOH	20.8	17.9
Limonene	LIMO	11.0	<mark>9</mark> .11
α/β -pinene, sabinene, carene	MTPA	98.6	81.5
Other monoterpenes	MTPO	40.8	38.6
Isoprene	ISOP	502	397.6
Methanol	MOH	119	-
Toluene	TOLU	1.57	-
Lumped alkenes $\geq C_3$	PRPE	22.3	24.2

458

Emissions of aerosols (primary organic matter, and black carbon) are listed in Table 4. These emissions are consistent with thevalues provided in Tilmes et al. (2016).

461

462 Table 4. Annual global emissions of sulfates, primary organic matter and black carbon in all three model configurations.

	C-GC	S-GC	C-CC
SO ₄ in Tg(S)/year	0.22	0.22	0.22
POM in Tg(C)/year	45.86	34.57	57.19
BC in Tg(C)/year	8.14	7.86	6.24

464 Mobilization of mineral dust is calculated in all three models using the DEAD scheme (Zender, 2003). In C-CC and C-GC, 465 the online implementation in CESM is employed, resulting in total natural mineral dust emissions of 5984 Tg/year. A brief 466 discussion of dust emissions in CESM is provided in the Supplementary Information. In S-GC, natural mineral dust emissions

467 are calculated online using the same scheme but with a different scaling and at a slightly different grid resolution, resulting in

total emissions of 1390 Tg/year.

469

Emissions of sea salt are calculated online in CESM for C-GC and C-CC, while S-GC uses a pre-calculated (offline) inventory of sea-salt emissions, as well as sea-salt bromine and chloride. Emissions of sea-salt bromine in C-GC are calculated based on the offline inventory rather than the calculated emissions of sea salt, and therefore do not scale correctly with the estimated sea-salt emissions from CESM (see Table 5). This will be resolved as part of future work.

474

475 Table 5. Annual global emissions of sea salt aerosols (fine and coarse) and bromine in sea salt for C-GC and S-GC. The names of the tracers
 476 used to represent these species in GEOS-Chem are provided in brackets.

Species	C-GC (Tg/year)	S-GC (Tg/year)
Fine sea-salt (SALA)	93.0	59.1
Coarse sea-salt (SALC)	2780	3576
Bromine in fine sea-salt (BrSALA)	0.166	0.126
Bromine in coarse sea-salt (BrSALC)	10.1	7.54

477

Finally, for long-lived species such as chlorofluorocarbons (CFCs) we use the shared socio-economic pathway 2-4.5 (SSP24.5) (Riahi et al., 2017) set of surface boundary conditions in both C-GC and C-CC. In comparisons against S-GC we use
historical emissions from the World Meteorological Organization's 2018 assessment of ozone depletion (Fahey et al., 2018).
However, this difference is unlikely to significantly affect simulation output given the short duration of the simulations.

483 Table 6. Brief summary of the model configuration used for C-GC, S-GC, and C-CC.

	C-GC	S-GC	C-CC
Horizontal resolution	1.9°×2.5°	C48 (~2°×2.5°)	1.9°×2.5°
Vertical levels	L56 (up to 1.65 hPa)	L72 (up to 0.01 hPa)	L56 (up to 1.65 hPa)
Aerosol microphysics	MAM4 with VBS-SOA	Bulk with VBS-SOA	MAM4 with VBS-SOA
Aerosol tracers	Mixed (S-GC for aerosol chemistry; C-CC for microphysics)	SO4, BC, OM, sea salt, dust, AERI, DMS, INDIOL, IONITA, MONITA, MOPI, MOPO, MSA, NH4, NIT, pFe, SOAGX, SOAIE, TSOA, ASOA	SO4, BC, OM (both primary and secondary), sea salt, dust
Treatment of SOA	Explicit calculation of SOA using VBS (five bins)	Explicit calculation of SOA using VBS (four bins)	Explicit calculation of SOA using VBS (five bins)
Chemistry	GEOS-Chem 13.1.2	GEOS-Chem 13.1.2	MOZART-TS1
Biogenic emissions	Online from dynamically evolving vegetation computed in CLM using MEGAN2.1	Offline from archived vegetations using MEGAN2.1	Online from dynamically evolving vegetation computed in CLM using MEGAN2.1

Original model references

This work

484 4. Model intercomparison

We first compare the global distribution of ozone and aerosols between C-GC, S-GC, and C-CC. Section 4.1 evaluates the vertical and latitudinal distribution of ozone and two related species (water vapor (H₂O) and the hydroxyl radical (OH)), followed by the global distribution of ozone at the surface in each model configuration (Section 4.2). Stratospheric chemistry in GEOS-Chem is described by Eastham et al. (2014) and by Emmons et al. (2020) for CAM-chem. A similar evaluation of differences in zonal mean and surface aerosol concentrations follows (Section 4.3).

490

To understand the causes of these differences, we compare the global distribution of reactive nitrogen and halogen species in each model configuration (Section 4.4). When comparing halogen distributions we consider only bromine and chlorine distributions, as iodine is not simulated in this version of CAM-chem. The latest implementation of halogen chemistry in GEOS-Chem and its role in atmospheric chemistry are described by Wang et al. (2021), while its representation in CAM-chem is described by Emmons et al. (2020). Differences in the total atmospheric burden and vertical distribution of these families provides information regarding differences in removal processes. Differences in their internal partitioning (e.g. between NO_x and HNO₃) provide information regarding the representation of atmospheric chemistry.

498 **4.1. Ozone**

Figure 2 shows the annual mean mixing ratio of stratospheric ozone simulated by each of the three model configurations. At 10 hPa in the tropics, where ozone mixing ratios reach their peak, the three configurations agree to within 10% suggesting a reasonable representation of stratospheric ozone. However, near the tropopause the three configurations diverge. Comparison of C-GC to S-GC (panel b) shows mixing ratios 20% lower near the tropical tropopause but more than 50% greater in the extratropical lower stratosphere. However, C-GC simulates mixing ratios of ozone around the tropopause which are 20% lower than C-CC (panel c) at all latitudes.



Figure 2. Comparison of stratospheric ozone simulated with CESM running GEOS-Chem (C-GC) to standalone GEOS-Chem (S-GC) and
CESM running CAM-chem (C-CC). Left column (a): absolute values estimated with C-GC. Center column (b): relative difference between
C-GC and S-GC. Right column (c): relative difference between C-GC and C-CC. Red (blue) shading means that C-GC estimated a higher
(lower) value than the other model.

510 The difference in pattern in the comparison between C-GC, S-GC, and C-CC implies that the cause is likely to be related to 511 factors which are common between C-GC and C-CC, such as the representation of meteorology. One such factor may be water 512 vapor, which is treated differently between the "online" (C-GC, C-CC) and "offline" (S-GC) configurations.

513

To quantify and understand these differences in stratospheric ozone, we analyze concentrations of three different related compounds from the surface to the stratosphere: ozone, the hydroxyl radical OH, and water vapor. OH reacts with most trace species in the atmosphere and its high reactivity makes it the primary oxidizing species in the troposphere, such that differences in abundance between models will affect the simulated abundances of many atmospheric pollutants (Seinfeld and Pandis, 2006). Since OH is produced from water vapor and (indirectly) ozone, these three compounds can collectively be used to understand some of the differences between C-GC, S-GC, and C-CC. Later analyses will focus on NO_x , bromine, and chlorine, each of which also strongly affect tropospheric and stratospheric concentrations of ozone.

- 521
- 522



Figure 3. Comparison of atmospheric composition simulated with CESM running GEOS-Chem (C-GC) to standalone GEOS-Chem (S-GC)
 and CESM running CAM-chem (C-CC). Different rows show different constituents, while different columns show different model results.
 Top row (a-c): ozone. Middle row (d-f): OH radical. Bottom row (g-i): water vapor. Left column (a, d, g): absolute values estimated with

526 *C-GC.* Center column (*b*, *e*, *h*): relative difference between C-GC and S-GC. Right column (*c*, *f*, *i*): relative difference between C-GC and 527 *C-CC.* Red (blue) shading means that C-GC estimated a higher (lower) value than the other model.

The upper row of Figure 3 shows the distribution of ozone as represented by C-GC (panel a), and the difference when compared to S-GC (panel b) or C-CC (panel c). Comparing to C-CC, C-GC estimates mixing ratios of ozone which are 30% lower from the surface (across all latitudes) and throughout the extratropical troposphere. This is consistent with previous work which showed that ozone simulated by GEOS-Chem to match the KORUS-AQ campaign had a normalized mean bias of -26%, compared to -9% in CAM-chem (Park et al., 2021). In the present study, we find that ozone mixing ratios around the tropopause are also lower in C-GC than in C-CC by 15-20%. This suggests that discrepancies observed in KORUS-AQ may be related to chemistry rather than the treatment of meteorology, but a more focused regional analysis would be needed to confirm this.

535

536 Comparing to the differences between C-GC and S-GC provides some insight into possible causes for these discrepancies. 537 Near-surface ozone in C-GC in the Southern Hemisphere is also 30-40% lower than in S-GC, suggesting a potential common 538 cause for the differences with C-CC. However, in the Northern extratropical troposphere below 400 hPa, zonal mean 539 differences between C-GC and S-GC are consistently less than 10%. Ozone concentrations are also lower in the tropical mid-540 troposphere in C-GC than in S-GC by 15-25%, whereas concentrations were well matched in this region between C-GC and 541 C-CC. In the lower stratosphere, ozone concentrations in C-GC are instead greater than in S-GC, with the difference in the 542 Northern extratropical lower stratosphere exceeding 50%. The global ozone burden in C-GC is within 1.5% of that estimated 543 by S-GC, while C-CC has a total atmospheric ozone burden 15% greater than C-GC. These model differences are evaluated 544 against observations in Section 5.2.

545

546 Differences in tropospheric NO_v and halogens, in particular the higher loading of BrO in C-GC, may explain some of these 547 differences (see Section 4.4). However, another possible factor in these differences in ozone is differences in water vapor 548 distribution. The bottom row of Figure 3 shows the annual average simulated distribution of water vapor in C-GC, and the 549 difference relative to S-GC and C-CC. Water vapor concentrations are approximately equal between C-GC and C-CC, since 550 the representation of moist physics in the two models is identical. However, differences of up to 20% arise around the 551 tropopause, possibly due in part to the different representation of stratospheric water chemistry and settling of stratospheric 552 aerosol (including ice). This is unlikely to be due to HO_x catalytic cycles depleting ozone, as OH in this region is lower in C-553 GC than in S-GC (panel e) and HO_x cycles are in any case a minor contributor to ozone depletion in the lower stratosphere 554 (Brasseur and Solomon, 2006). The greater water vapor (and therefore humidity) may instead result in faster heterogeneous 555 chemistry, including the liberation of NO_x from HNO₃.

556

The differences between C-GC and S-GC are larger. Outside of the tropics and below the tropopause, water vapor concentrations are up to 30% greater in C-GC than in S-GC. Differences are smaller in the tropics, but in the tropical upper troposphere water vapor concentrations are instead 15% lower in C-GC than in S-GC. This may be part of the reason that 560 water vapor concentrations in the extratropical lower stratosphere are more than 50% lower in C-GC than in S-GC, since the 561 tropical upper troposphere is the source of water vapor to the stratosphere. This is the same region in which C-GC calculates 562 ozone mixing ratios which are more than 50% greater than in S-GC, potentially due to the lower concentration of water vapor 563 (an indirect sink for ozone).

564

These differences arise due to the different representation of moist processes between CAM's physics package (used in both C-GC and C-CC), and GEOS, which produces MERRA-2 and therefore is represented in S-GC. For example, although total annual average precipitation agrees to within 10% between the models, the mean volumetric cloud fraction in C-GC and C-CC is 15%, compared to 8% in S-GC. Meanwhile the area-averaged cloud water content and cloud ice content are 57% and 38% greater in S-GC than in C-GC (or C-CC).

570

571 Differences in ozone and water vapor result in differences in concentrations of OH, as shown in the middle row of Figure 3. 572 The global OH atmospheric burden is approximately 10% lower in C-GC than in S-GC, but this difference is not evenly 573 distributed. Differences in OH concentrations can be roughly considered to be the product of differences in ozone and 574 differences in water vapor, since both are needed to create OH (along with UV radiation). In the tropical troposphere, OH 575 concentrations are more than 50% lower in C-GC than in S-GC, likely due to a relative lack of both ozone and water vapor. 576 However, in the Northern mid- and upper latitudes below 900 hPa, OH concentrations are 10-20% greater in C-GC than in S-577 GC. This reflects the greater water vapor concentrations and roughly equal ozone concentrations between the two models.

578 **4.2.** Surface ozone

Figure 4 compares the simulated, annually-averaged surface ozone mixing ratios as estimated by C-GC, S-GC, and C-CC. We find that, when globally averaged, C-GC predicts a lower surface ozone mixing ratio than either C-CC or S-GC. Averaged over each Hemisphere, C-GC estimates a lower surface ozone mixing ratio than S-GC (panel b) by 4.9 ppbv and 2.2 ppbv in the Southern Hemisphere and Northern Hemisphere respectively. This varies between the land and oceans. In the Northern Hemisphere, we observe a small difference in surface ozone mixing ratio over the oceans (less than 1 ppbv), while a difference of approximately 3 ppbv can be found over North America, Europe and East Asia.



Figure 4. Comparison of the annually averaged surface ozone mixing ratios simulated with CESM running GEOS-Chem (C-GC, panel a)
 to standalone GEOS-Chem (S-GC, panel b) and CESM running CAM-chem (C-CC, panel c). Red (blue) shading means that C-GC
 estimated a higher (lower) value than the other model.

The difference between C-GC and C-CC (panel c) does not show the same hemispheric asymmetry in absolute terms, and a larger difference over oceans than over land. We find that C-GC estimates 5.4 and 7.9 ppbv less ozone than C-CC in the Southern and Northern Hemispheres respectively. The pattern indicated in Figure 4c suggests that bromine from sea salt may be the principal cause of the differences in surface ozone between C-GC and C-CC, whereas differences between C-GC and S-GC are likely to be related to anthropogenic emissions given the hemispheric asymmetry. The 20-30% increase in ozone over the Amazon in C-GC related to C-CC may instead be related to differences in biogenic emissions.

596

586

597 In addition to annual averages, we also consider seasonal variations of surface ozone. Figure 5 presents parity plots of monthly-598 averaged surface ozone mixing ratios for January and July comparing C-GC to S-GC and C-CC, after outputs from all three 599 model configurations were remapped to a common $2^{\circ} \times 2.5^{\circ}$ grid. In January, we find a correlation coefficient of 0.91 and slope 600 of 0.91 between C-GC and S-GC. In July this agreement is worsened, with a correlation coefficient of 0.80 but a slope of 0.93. 601 This indicates that the sources of differences in surface ozone mixing ratios between C-GC and S-GC are magnified during 602 boreal summer. There is also a distinctive "hot spot" in the July parity plot, with a large cluster of grid cells showing mixing 603 ratios in the range 20-25 ppbv in S-GC but 10-20 ppbv in C-GC. Further research is needed to establish the origin of this 604 cluster, which does not occur during boreal winter.



Figure 5. Parity plots of surface ozone mixing ratios, expressed in ppbv, for January (left) and July (right) comparing C-GC on the X axis
 to S-GC (top) and C-CC (bottom) on the Y axis. Fitting parameters are shown in the top left corner for both months. All panels share the same color scale.

611 Comparison between C-GC and C-CC shows a different pattern. The line of best fit between C-CC and C-GC indicates 30% 612 greater ozone in C-CC in January than in C-GC ($y \sim 1.3x$), but no such normalized mean bias is present in July ($y \sim 1.0x$). As 613 with the comparison of C-GC to S-GC, the absolute bias is greater in July than in January, but the correlation between C-CC 614 and C-GC does not worsen between the two months ($r^2 = 0.87$). This may indicate the strength of the effect of meteorology

615 and non-chemistry processes in the seasonality of simulated surface ozone.

616 4.3. Aerosols

- 617 Figure 6 shows the zonal mean mass concentration of sulfate aerosol as simulated in each of the three model configurations.
- 618 In C-GC and C-CC, this is calculated as the sum across all aerosol size bins, whereas S-GC uses a bulk representation.







623 value than the other model. Differences are restricted to $\pm 100\%$ for clarity.

624 Between 45°S and 45°N, and below 800 hPa, C-GC more closely follows S-GC (comparison in panel b) with regards to sulfate 625 aerosol mass. Compared to C-CC (panel c), sulfate aerosol mass is approximately 50% greater in southern latitudes with 626 differences being greatest over the oceans. Sulfate concentrations in this region are dominated by oxidation of naturally-emitted 627 dimethyl sulfide (DMS) (Seinfeld and Pandis, 2006). Since DMS concentrations are identical between the three configurations, 628 the greater sulfate concentration in C-GC compared to C-CC may instead reflect differences in OH (Figure 3). Elsewhere the 629 concentration of sulfate in C-GC more closely follows that in C-CC, likely due to the common representation of sulfate aerosol 630 in MAM4 and differences in the representation of convective scavenging between CESM and standalone GEOS-Chem. 631 Concentrations of sulfate in the tropical mid-to-upper troposphere and extratropical lower stratosphere in C-GC exceed those 632 in S-GC by over 100%, whereas comparison to C-CC show differences of $\pm 25\%$.



634

Figure 7. Comparison of the annually averaged surface mass concentration of sulfate aerosol simulated with CESM running GEOS-Chem
 (C-GC) to standalone GEOS-Chem (S-GC) and CESM running CAM-chem (C-CC). Red (blue) shading means that C-GC estimated a higher
 (lower) value than the other model.

This is further illustrated in Figure 7, which shows the surface concentration of sulfate aerosol in each model configuration. C-GC simulated greater concentrations in the intertropical convergence zone (off the west coast of Southern Hemisphere continents) than in S-GC (panel b), but in these regions agrees more closely with C-CC (panel c). Elsewhere in the tropics the agreement between C-GC and S-GC is stronger, whereas surface concentrations of sulfate aerosol over (e.g.) the Southern Pacific exceed those in C-CC by over 100%. At high latitudes and over land the agreement between C-GC and C-CC is again stronger than in S-GC, although this varies by location. Further work would be needed to identify the underlying causes leading

to differences in surface sulfate concentrations between all three models.





Figure 8. Comparison of primary organic matter aerosol mass concentration as simulated with CESM running GEOS-Chem (C-GC) to
standalone GEOS-Chem (S-GC) and CESM running CAM-chem (C-CC). Left: absolute values estimated with C-GC. Center: relative
difference between C-GC and S-GC. Right: relative difference between C-GC and C-CC. Red (blue) shading means that C-GC estimated a
higher (lower) value than the other model.

650 We also show the zonal mean concentrations of primary organic matter (POM) aerosol in each configuration (Figure 8). POM

651 in C-GC and C-CC is calculated as the sum of the POM aerosol size bins, whereas in S-GC it is the sum of the hydrophobic

and hydrophilic organic carbon species. As with sulfate aerosol, C-GC and S-GC agree to within 25-50% in the tropics below

- 653 800 hPa, but C-GC simulates concentrations of POM which are over 100% greater than S-GC in the mid- and upper tropical
- troposphere and throughout the lower stratosphere. This is again likely due to differences in the representation of convective
- 655 scavenging. C-GC also simulates concentrations of POM which are lower than C-CC throughout the entire troposphere. This
- is likely due to differences in the implementation of POM emissions between C-CC and C-GC, where emissions of POM in
- 657 C-CC are 29% lower and occur as accumulation-mode rather than primary organic mode aerosol.

658 4.4. Reactive nitrogen (NO_y), bromine (Br_y), and chlorine (Cl_y)

To better understand the source of differences in ozone and aerosols described above, we now investigate differences in reactive nitrogen (NO_y) and halogen families (Br_y and Cl_y).

661 **4.4.1. Reactive nitrogen** (NO_y)

We compare the total concentration and partitioning of reactive nitrogen species in each model configuration, including NO_x and its reservoir species (collectively NO_y). A full list of the species included in the lumped NO_y reservoir species can be found in the legend of Figure 10 for each model configuration. We first compare results in the stratosphere, followed by an evaluation of concentrations and partitioning below 100 hPa. Concentrations of nitrate aerosol concentrations are estimated in CAMchem using a simplified approximation (Lamarque et al., 2012), and particulate nitrate is typically not considered to be simulated by CAM-chem (e.g. Park et al. (2021)). We therefore do not include it in this analysis.



Figure 9. Global annual mean mixing ratio of total reactive nitrogen (NO_y) as a function of altitude. Left (panel a): Profile of NO_y mixing ratio for C-GC (red), C-CC (blue), and S-GC (orange). Middle (panel b): Relative difference in NO_y mixing ratio between C-GC and S-GC. Right (panel c): Relative difference in NO_y mixing ratio between C-GC and C-CC.

Figure 9 shows global mean NO_y at each altitude for C-GC, C-CC, and S-GC. Comparing C-GC to S-GC (panel b), differences in total NO_y are less than \pm 50% at all altitudes. Between 100 and 10 hPa, C-GC differs from S-GC by less than 20%, compared to less than 10% with respect to C-CC (panel c). The difference between C-GC and C-CC increases from -2% at 10 hPa to +20% at the top of the model, compared to an increase from 10% to 25% when comparing C-GC to S-GC. At lower altitudes C-GC more closely follows C-CC than S-GC, with differences between C-GC and S-GC exceeding 50% between 200 and 300 hPa. The global NO_y burden in C-GC (2.74 TgN) is closer to that in S-GC (2.84 TgN) than C-CC (3.01 Tg), likely due to the stronger influence of the troposphere on this quantity.

669

Figure 10 shows the speciation of NO_y as a function of altitude in each model from the surface to 1 hPa. The list of species defined collectively as NO_y differs between C-GC and S-GC on one side and C-CC on the other side. At altitudes above 100 hPa, the dominant contributors to NO_y in all three model configurations are NO, NO_2 , HNO_3 , and N_2O_5 , although ClNO₃ contributes significantly between approximately 80 and 5 hPa. Between 10 and 200 hPa ratios of NO to NO_2 are approximately consistent between the models, lying in the range 0.35 to 0.50. This suggests broad consistency in actinic flux and ozone concentrations, given their role in controlling NO:NO₂ ratios in the stratosphere (Cohen and Murphy, 2003).

- 687
- 688 By contrast, partitioning between NO_x and HNO₃ differs significantly between the three models. At 10 hPa, HNO₃
- 689 constitutes 20% of total NO_y in C-GC but 23% in both C-CC and S-GC (values not shown explicitly). This fraction increases
- 690 with decreasing altitude at differing rates. At 200 hPa, HNO₃ constitutes 60 and 63% of NO_y in C-GC and S-GC
- respectively, but 78% of NO_y in C-CC. One possible cause of these discrepancies is heterogeneous chemistry. GEOS-Chem
- (in both S-GC and C-GC) uses a different representation of N_2O_5 hydrolysis than CAM-chem, but the CESM-driven
- 693 simulation includes a more detailed representation of the sulfate aerosol size distribution through MAM4. The present study
- did not include the analysis of aerosol reactive tendencies. This would be a valuable line of inquiry for future comparisons of
- 695 CAM-chem and GEOS-Chem, given the lack of nitrate aerosol in the former.



696

697 Figure 10. Global annual mean speciation of NO_y as a function of altitude. Results are shown from C-GC (left, a), S-GC (middle, b), and 698 C-CC (right, c) from the surface up to the model top (approximately 2 hPa). Values correspond to the number of N atoms present, such 699 that (e.g.) the mixing ratio of N₂O₅ is multiplied by 2.

- Figure 11 provides a closer look at the speciation of NO_y at altitudes below 100 hPa. NO_y at altitudes below 200 hPa is
- predominantly NO_x , HNO_3 , and peroxyacetyl nitrate (PAN). At 200 hPa, the combination of NO_x , HNO_3 , and PAN make up 86% of total NO_y in C-GC (panel a) and 84% in S-GC (panel b), but 96% in C-CC (panel c). However, between 200 and 900
- hPa the dominant contributors are HNO₃ and PAN. In this pressure range, the C-GC and S-GC simulations also show a
- significant contribution from nitrate aerosol (NIT) and BrNO₃. At 500 hPa, the contributions of NO_x, HNO₃, and PAN are

705 78%, 85%, and 97% respectively for C-GC, S-GC, and C-CC. Below 900 hPa, NO and NO₂ once again become significant
706 contributors to total NO_y. At these lower altitudes C-GC more closely follows C-CC than S-GC, with differences in total NO_y
707 between C-GC and S-GC exceeding 50% between 200 and 300 hPa. As such, we find that the speciation in C-GC more closely
708 follows that in S-GC at lower altitudes.

709

710 Since surface emissions of NO_x are nearly identical between the three configurations (see Table 2) and lightning NO_x emissions 711 are identical between C-GC and C-CC, differences below 100 hPa may instead be related to NO_x chemistry, and nitrate aerosol. 712 However, concentrations of PAN in C-GC more closely follow C-CC than S-GC, suggesting that the representation of 713 meteorology (including wet deposition rates) is also an important factor. At 500 hPa, total PAN in C-GC is 3% lower than the 714 value in C-CC, but exceeds the value in S-GC by 38%. This may be due to the greater emissions of biogenic VOCs in CESM than in the standalone GEOS-Chem (see Table 3Table 3), resulting in more NOx being bound into PAN for long-range 715 716 transport. We also find that HNO₃ concentrations in the mid-troposphere are lower in C-GC than in either C-CC or S-GC. At 717 500 hPa, HNO₃ mixing ratios in C-GC are 43% lower than in S-GC and 52% lower than in C-CC. This does not account for 718 the conversion of HNO₃ in C-GC and S-GC to nitrate aerosol (NIT), which is not represented in C-CC.

719





722 Differences in mid-tropospheric HNO₃ between the models are most likely due to differences in the representation of wet 723 scavenging. In C-CC and C-GC, scavenging of gaseous species is handled by the Neu scheme, while scavenging of modal 724 aerosols is performed by MAM (Neu and Prather, 2012). Any aerosol species not handled by MAM, such as nitrate in C-GC, 725 are also scavenged using the Neu scheme. In C-GC and C-CC, the Neu scheme calculations are performed at the same time as 726 the chemistry and after convective transport, while scavenging of MAM aerosols is performed before. Thus, all species that 727 undergo wet deposition in the Neu scheme are not removed during convective transport. This leads to soluble species and 728 aerosols being carried to higher altitudes without being convectively scavenged. We also find that the Neu scavenging scheme 729 in C-GC and C-CC results in an HNO₃ wet removal rate which is four times higher in C-GC than in S-GC (Figure S1 of the 730 Supplementary Information). This likely explains the greater depletion of HNO_3 in the mid-troposphere calculated by C-GC 731 compared to S-GC. Wet scavenging in C-CC is faster yet, with HNO₃ wet removal rates approximately six times greater than 732 in S-GC, and 50% greater than in C-GC. This is in part because the mixing ratio (or fraction of total NO_v) of HNO₃ in the mid-733 and upper-troposphere as modeled in C-CC is greater than in either C-GC or S-GC, but also because C-GC and S-GC simulate 734 nitrate aerosol explicitly. The application of the Neu scheme to remove nitrate aerosol also affects removal of total NO_{ν} in C-735 GC (Figure S2 in the Supplementary Information). We find that the Neu scheme removes aerosol more rapidly than the 736 scheme used in S-GC, and at lower altitudes.

737

Total HNO₃ removal tendencies in each model configuration are shown in Table 7. The total removal rate of NO_3^- is lowest in S-GC and highest in C-CC, consistent with the finding that total NO_y burdens are lower in S-GC than C-GC or C-CC. However, the removal rate of nitrate aerosol is lower in C-GC than in S-GC despite the greater wet removal rates for C-GC. A possible explanation is that washout rates of nitrate aerosol are sufficiently high in both C-GC and S-GC that all nitrate aerosol is effectively removed, but that the faster washout of HNO₃ in C-GC results in less nitrate aerosol being available for removal.

743

744 Table 7. Total wet removal tendency of HNO₃ and nitrate aerosol in each model configuration. All values are given in units of Tg NO₃/yr.

	0.00	9.00	0.00
	C-GC	S-GC	
HNO ₃	82.0	71.3	119.6
Nitrate aerosol	20.4	22.7	-
Total NO ₃ -	102.4	94.0	119.6

745

746 **4.4.2.** Reactive bromine (Br_y)

Halogens are involved in multiple catalytic ozone-depleting chemical cycles, and are critical to an accurate description of both tropospheric and stratospheric chemistry (Solomon et al., 2015). We therefore analyze the abundance and speciation of the two key halogens – bromine and chlorine – in each configuration. Figure 12 shows the annual average mixing ratio of total reactive bromine as a function of altitude in each of the three models. This does not include long-lived species such as halons or CH₃Br. A full listing is included in the legend of Figure 13.



Figure 12. Global annual mean mixing ratio of reactive bromine as a function of altitude. Left (panel a): Profile of total gaseous inorganic
and organic bromine mixing ratio for C-GC (red), C-CC (blue), and S-GC (orange). Middle (panel b): Relative difference in brominecontaining species mixing ratio between C-GC and S-GC. Right (panel c): Relative difference in bromine-containing species mixing ratio
between C-GC and C-CC. Although relative differences between C-GC and C-CC exceed 1000% near the surface, the limits on the
rightmost panel are clipped to allow comparison to the center panel.

- Globally averaged total Br_y in C-GC is maximized at the surface, exceeding that from S-GC by 100%. This is partially explained by the greater emissions of sea salt bromine, although C-GC's annual emission of sea salt bromine is only 36% greater than that in S-GC (see Table 4). Since C-CC does not include short-lived bromine sources such as sea salt bromine, the C-GC total Br_y concentration exceeds C-CC by 1000 % at the surface.
- 763

From Figure 13, we find that, in all three models, the mixing ratio increases monotonically with altitude above 800 hPa due to the reaction of CH₃Br with OH. Br_y falls sharply from 12 pptv at the surface in C-GC to 3 pptv at 900 hPa, but then increases again to 10 pptv at 100 hPa. This pattern is similar to that displayed by S-GC, although the decrease from the surface is less sharp and the absolute value lower in S-GC. Above 100 hPa, the averaged Br_y mixing ratio levels off, with values between 20 hPa and 2 hPa remaining roughly constant in the range of 16-20 pptv. This is similar to the behavior shown by C-CC but differs from S-GC, in which Br_y continues to rise with altitude – albeit more slowly. The net effect is that total Br_y in C-GC

- exceeds both C-CC and S-GC below 100 hPa, but is lower than the value in either model above 10 hPa (above 80 hPa when
 compared to C-CC).
- 772

In addition to differences in total Br_y , the partitioning of Br_y also varies between the three models (Figure 13). The additional near-surface bromine present in C-GC and S-GC is due to the presence of Br_2 and sea salt bromine (BrSALA and BrSALC, representing bromine in fine and coarse-mode sea salt respectively). This provides a source of active bromine in the planetary boundary layer which is not represented in C-CC, but in forms which are rapidly washed out in C-GC and S-GC. The greater concentrations of Br_y near the surface as calculated by C-GC compared to S-GC are likely due to the greater emissions of sea salt bromine, as shown in Table 4.

779



780

Figure 13. Global annual mean speciation of total organic and inorganic bromine as a function of altitude. Results are shown from C-GC
(left, a), S-GC (middle, b), and C-CC (right, c), from the surface up to the model top (approximately 2 hPa). Values correspond to the number of Br atoms present, such that (e.g.) the mixing ratio of Br₂ is multiplied by 2.

784 Br_y in the model stratosphere is dominated by the same species in all three configurations: BrO_x (Br + BrO), BrCl, BrNO₃,

HBr, and HOBr. The most significant difference is the greater proportion of HOBr in C-CC (approximately 15%) than in S-

GC or C-GC (8-10%). Larger mixing ratios of BrNO₃ are also present in C-CC (approximately 10 ppbv at 30 hPa) compared
 to C-GC and S-GC (approximately 7.4 and 7.0 ppbv respectively at 30 hPa). Smaller mixing ratios of BrCl are present in C-

788 CC, with a mean value of 1.8 ppbv at 30 hPa, while they reach 3.1 ppbv in C-GC and S-GC at 30 hPa. The base causes of

- these differences are not clear, but may be related to the presence of more complex tropospheric and stratospheric halogenchemistry in the GEOS-Chem chemical mechanism (Wang et al., 2021).
- 791

Between 30 hPa and the top of the boundary layer, the three models show greater divergence. The only significant sources of atmospheric Br_y in C-CC are CH₃Br, CH₂Br₂, and very long-lived bromine species such as halons which are insoluble. As a result, tropospheric Br_y concentrations increase only slowly from the surface up to 300 hPa, at which point HOBr, BrO, and BrNO₃ begin to form in significant quantities. In C-GC and S-GC, these sources of bromine are supplemented by bromine from sea salt and surface Br_2 emissions. Mid-tropospheric Br_y concentrations are therefore largely set by the quantity of sea salt bromine emitted, and by the fraction of that bromine which can be released to an insoluble form (e.g. Br_2) before the sea salt is washed out of the atmosphere.

799

The greater concentration of mid-tropospheric Br_y in C-GC than in S-GC is likely due to differences in wet scavenging. Wet removal tendencies of bromine in fine sea salt (BrSALA) from large-scale and convective precipitation as calculated by C-GC and S-GC are shown in the Supplementary Information (Figure S3). We find that there is greater wet deposition of fine sea salt bromine in S-GC than in C-GC, despite removal rates below 900 hPa being greater in C-GC. Since total emissions of BrSALA are also 26% lower in S-GC than in C-GC (Table 4), the slower mid-tropospheric mid-tropospheric removal of bromine in C-GC explains the greater simulated concentration of Br_y in the mid troposphere.

806

807 C-GC also calculates wet deposition of non-MAM aerosols from both convective and large-scale precipitation independent of
 808 convective transport, whereas S-GC calculates convective scavenging as part of convective transport. This means that soluble
 809 species can be transported in convective updrafts in C-GC, unlike in S-GC.

810 4.4.3. Reactive chlorine (Cl_y)

We now focus on atmospheric chlorine by comparing its profile and partitioning in all three models. Annually-averaged vertical profiles of reactive chlorine (Cl_y) are displayed in Figure 14, excluding source species such as chlorocarbons. A full list of the species used to define Cl_y in each configuration is provided in Figure 15.

814

As with total Br_y , total Cl_y follows the same vertical distribution as S-GC up to 10 hPa. Above this pressure, the vertical distribution in C-GC is closer to that of C-CC. The dominant factor in differences below 100 hPa is the lack of short-lived chlorine species such as sea salt in C-CC, which are the dominant source of chlorine to the lower troposphere. Above 10 hPa, the relative difference in Cl_y between C-GC and S-GC increases slowly from -2% at 10 hPa to -5% at 2 hPa, while the difference relative to C-CC remains at approximately -20% above 200 hPa.



821

Figure 14. Comparison of annual average vertical profiles of chlorine-containing compounds in the three models. Left (panel a): Vertical
profile of total gaseous chlorine mixing ratio for C-GC (red), S-GC (orange), and C-CC (blue). Middle (panel b): Relative difference in
Cl_y mixing ratio between C-GC and S-GC. Right (panel c): Relative difference in Cl_y mixing ratio between C-GC and C-CC exceed 1000% near the surface, the limits on the rightmost panel are clipped to allow
comparison to the center panel.

Figure 15 shows the speciation of Cl_y as a function of altitude in each model. The greater near-surface chlorine simulated by

828 C-GC (panel a) and S-GC (panel b) relative to C-CC (panel c) is mostly made up of HCl and chlorine in sea salt (SALACL

and SALCCL). However, the short lifetime of sea salt aerosol and of chlorine in the lower troposphere means that this source

830 is likely not significantly affecting stratospheric chlorine burdens or partitioning. In the stratosphere there is no clear difference

831 between partitioning in C-GC and S-GC, but larger mixing ratios of upper tropospheric and lower stratospheric HCl from

- 832 chlorine source compounds are found in C-CC. At 50 hPa, total Cl_y in C-CC is 15% greater than in C-GC and S-GC, but the
- 833 mean mixing ratio of HCl in C-CC is 45% greater. Differences in ClNO₃ reach their peak at higher altitudes, around 20-30
- 834 hPa.



Figure 15. Global annual mean vertical speciation of total organic and inorganic bromine in C-GC (left, a), S-GC (middle, b), and C-CC
(right, c) from the surface up to the model top (approximately 2 hPa). Values correspond to the number of Cl atoms present, such that
(e.g.) the mixing ratio of Cl₂ is multiplied by 2. SALACL and SALCCL correspond to chlorine in fine and coarse sea salt, respectively.

The global mean tropospheric concentration of Cl atoms is 590 cm⁻³, roughly consistent with a recent evaluation from Wang et al. (2021) which found a value of 630 cm⁻³. This is 24% greater than the value from S-GC (477 cm⁻³) and 160% greater than that from C-CC (224 cm⁻³), likely due to the greater emissions of sea salt and indicating that chlorine will play a greater role in tropospheric oxidation in C-GC.

844 **5.** Comparison of model results to observations

We now compare the results from C-GC to observational data, with results from S-GC and C-CC also provided as context. Section 5.1 evaluates model performance at the surface, comparing to ground measurements of surface NO₂ and ozone. Section 5.2 compares model results to a climatology of vertical profiles of ozone, based on ozone sonde data. Section 5.3 evaluates the level of agreement of simulated ozone and carbon monoxide (CO) columns to measurements from the Aura Ozone Monitoring Instrument (OMI) and Microwave Limb Sounder (MLS), and MOPITT satellite instruments. Finally, Section 5.4 evaluates the model against measurements of dry deposition fluxes and rainwater composition measurements. The CESM wet deposition scheme is presented in Neu and Prather (2012), while the wet deposition scheme implemented in GEOS-Chem uses different
schemes for gases and aerosols (Liu et al., 2001; Amos et al., 2012). Dry deposition in GEOS-Chem is described in several
publications (Wesely, 1989; Wang et al., 1998; Zhang et al., 2001), while the coupling between CAM and CLM is described
in Val Martin et al. (2014).

855 5.1. Surface NO₂ and ozone

Figure 16 compares surface mass concentrations of NO₂ as estimated by C-GC, S-GC, and C-CC for 2016 against ground station measurements for North America (top row), Europe (middle row), and South-East Asia (bottom row) (AirNow API, 2021; Environmental Numerical Database, 2021; China Air Quality Historical Data, 2021; European Air Quality Portal, 2021). All ground station measurements are the average value over 2016. By comparing model results at an approximately 2° horizontal resolution to point observations, we expect some differences in this evaluation due to grid-box representation errors.

861

862 All three model configurations calculate lower mixing ratios than are reported by the ground observations. This is likely to be 863 in part due to the presence of interferants such as HNO₃, which cause in-situ monitors to overestimate the concentration of 864 NO₂ (Dunlea et al., 2007). However, S-GC is consistently biased lower than C-GC or C-CC. We also find that the surface NO₂ 865 concentrations display variable agreement depending on the geographical location. The correlation coefficients for North 866 America, Europe and South-East Asia are provided in Table 8. All three models give similar correlation coefficients for each 867 region. This is expected given that the three model configurations all use the same input wind fields and NO_x emissions 868 datasets. Nonetheless, both C-GC and C-CC estimate higher concentrations of NO₂ in Northern China, Northern Europe, and 869 the northeast US than S-GC. This suggests that the representation of meteorology, photolysis, and NO_y removal processes 870 have a greater impact on simulated NO₂ than the chemistry module alone. Comparing in-situ NO₂ measurements against NO_y 871 model results could potentially remove the effect of interferants in the observations of surface NO₂ concentrations, but is not 872 considered here.

873

874 Table 8. Correlation coefficients of surface-level NO₂ mass concentrations for C-GC, S-GC and C-CC against measurements.

	C-GC	S-GC	C-CC
North America	0.39	0.36	0.38
Europe	0.21	0.21	0.21
South-East Asia	0.42	0.41	0.41



Figure 16. Annual average surface NO₂ mass concentrations simulated by C-GC (left: a, e, h), S-GC (middle: b, f, i), and C-CC (right: c, g, j) for 2016 compared against monitor measurements in North America (top: a-c), Europe (middle: d-e), and South East Asia (bottom: h-880 j).

881 Figure 17 shows the ratio of annual mean NO to annual mean NO_2 , and thus provides some insight into possible causes of 882 these disagreements. The global distribution of surface-level NO2 and NOx is presented in the Supplementary Information. 883 All three configurations show enhanced NO:NO₂ ratios in polluted regions such as eastern China and over icy regions such as 884 Greenland and Antarctica. However, S-GC shows reduced NO:NO2 ratios over land compared to either C-CC or C-GC. For 885 example, ratios over North America in S-GC range from 0.1 to 0.2, compared to a range of 0.01 to 0.1 in C-GC and C-CC. 886 Surface NO:NO₂ ratios are typically dictated by surface ozone and the NO₂ photolysis rate (Seinfeld and Pandis, 2006). Given 887 that surface ozone concentrations in S-GC are typically between those calculated in C-GC and C-CC (see Figure 4) and that 888 S-GC and C-GC share the same photolysis treatment, this discrepancy may instead be caused by the differences in cloudiness 889 calculated by CESM compared to the MERRA-2 fields read in by S-GC.



Figure 17. Surface-level NO:NO2 estimated by C-GC (left, a), S-GC (middle, b), and C-CC (right, c) for 2016. Annual average of NO:NO2
 is calculated as annual mean NO divided by annual mean NO2.

Differences in NO:NO₂ may also be related to differences in emissions and treatment of oxidants such as VOCs and bromine. C-GC and C-CC show a reduction in NO:NO₂ over the Amazon and in the Congo river basin, but this pattern is not reproduced in S-GC. Similarly, topographical features including the Andes and Himalayas are visible in the C-CC and C-GC NO:NO₂ ratios, but not in the S-GC data – whereas a large reduction in NO:NO₂ over the Arctic Ocean is more pronounced in S-GC and C-GC than in C-CC. This latter feature may be related to differences in the response of the simulated atmosphere to anthropogenic emissions, as ship tracks are more visible in the C-GC and S-GC NO:NO₂ ratios (see e.g. Cape Horn and the Cape of Good Hope) than in C-CC.

900

890

Figure 18 compares surface ozone against monitor measurements. The correlation coefficients are shown in Table 9. The geographical pattern is also consistent, with high surface ozone concentrations over the Mediterranean Sea and lower concentrations over Northern Europe.

904

905 Table 9. Correlation coefficients of surface-level ozone mass concentrations for C-GC, S-GC and C-CC against measurements.

	C-GC	S-GC	C-CC
North America	0.37	0.28	0.24
Europe	0.44	0.43	0.44
South-East Asia	-0.24	-0.07	-0.22

906

However, the results from all models appear to be biased low. As discussed in Section 4.2, C-GC estimates surface ozone
mixing ratios lower than either S-GC and C-CC, and therefore exhibits the greatest mean bias. C-GC, C-CC, and S-GC show
mean biases of -15, -9, and -10 ppbv for over Europe; -10, -3, and -5 ppbv over North America; and -20, -11, and -12 ppbv
over South-East Asia.



912

Figure 18. Annual average surface ozone mass concentrations simulated by C-GC (left: a, e, h), S-GC (middle: b, f, i), and C-CC (right: c, g, j) for 2016 compared against monitor measurements in North America (top: a-c), Europe (middle: d-e), and South East Asia (bottom: h-915 j).

The greater negative bias in simulated ozone shown by C-GC is likely related to both the different representation of meteorology compared to S-GC and the greater bromine emissions compared to both S-GC and C-CC. However, further work is needed to disentangle the root causes of discrepancies between the three models, and the common biases relative to observations.

920 **5.2.** Vertical profiles of ozone

We now focus on the evaluation of the profile of ozone mixing ratios by comparing C-GC, C-CC, and S-GC to a climatology of ozone sonde observations from 1995-2010 (Tilmes et al., 2012). This climatology (in addition to those shown in Section 5.3) has been used repeatedly in evaluations of CAM-chem, and is therefore chosen here to provide a familiar point of comparison which also allows the three configurations to be compared to a realistic baseline. Over the past decades, observations from ozone sondes in different locations provide a valuable dataset of the evolution of ozone mixing ratios in the troposphere and stratosphere. Figure 19 provides a Taylor diagram comparison between the C-GC, C-CC, and S-GC simulations of 2016 to the climatology.

- In general, C-GC does not perform significantly better or worse than C-CC or S-GC, producing mean biases and correlations in each region/altitude combination which are within the same range. The clearest exception is at low altitudes (900 or 500 hPa) and mid- to high latitudes. In these regions, C-GC results frequently show a smaller normalized difference from the mean (radius) than either S-GC or C-CC, but also a weaker correlation with the observed seasonal cycle. The C-GC simulation of tropical ozone also shows the smallest mean bias at all altitudes at or below 250 hPa (panels a, d, and g), although again showing a weaker correlation.
- 935

At high altitude (50 hPa, panels j-l), all three models appear to perform similarly. This may however simply reflect the lack of spin up time. Since the three models only simulated 1.5-2 years in total, the simulated stratosphere will not have had time to fully respond to the new model configuration. Longer simulations are needed to fully evaluate the performance and capability of the C-GC stratosphere.



Figure 19. Taylor diagrams of the comparison of C-GC (red), C-CC (blue), and S-GC (black) simulations to a present-day (1995-2010)
ozone sonde climatology. Top row to bottom row: comparisons at 900 (a-c), 500 (d-f), 250 (g-i) and 50 hPa (j-l). Left column to right
column: tropics (a, d, g, j), mid latitudes (b, e, h, j), and high latitudes (c, f, i, l). The normalized mean difference between simulations and
observations for each region is shown on the radius, and the correlation of the seasonal cycle is shown as the angle from the vertical.

946 5.3. Total column ozone and CO

- 947 Figure 20 shows total ozone column climatologies in Dobson Units as measured by OMI/MLS for the 2004-2010 time period
- 948 (Ziemke et al., 2011). The results from the satellite observations (panel a) are compared to results from C-GC, C-CC, and S-
- GC (panels b, d, and f respectively,). We find that on average the results from C-GC are 7.8 DU lower than the observations
- 950 (panel c), mostly driven by an overestimation of stratospheric ozone depletion during the Antarctic spring of up to 16 DU
- 951 (value not shown in figure). C-CC predicts a total ozone column that is 6.6 DU larger than the global mean ozone column
- (panel e). When broken down by tropospheric and stratospheric ozone column, we find that the bias in the stratospheric and
- 953 tropospheric ozone columns for C-GC is -2 and -6 DU respectively, compared to +9.5 and -2.5 DU for C-CC (value not shown
- 954 in figure). Additionally, we find that the bias in seasonal variations of total column ozone as predicted by C-GC range between
- 955 -16 and -6 DU, while the variations range from -3 to +7 DU for C-CC (value not shown in figure). The model results from S-
- 956 GC predict similar geographical biases in total ozone column as C-GC, although with a smaller net bias of -3.3 DU (panel g).



Figure 20. Total ozone column in DU as observed by OMI/MLS for the 2004-2010 time period (panel a), compared to the results from C-GC (2nd row, panels b and c), C-CC (3rd row, panels d and e), and S-GC (4th row, panels f and g) for the year 2016. The measurements and model results are presented on the left (a, b, d, f), while the model biases are shown on the right (c, e, g).

957

962 Figure 21 compares the simulated total columns of CO (panels b, d, and f) to retrievals from the MOPITT satellite instrument 963 (panel a), averaged for each April in the period 2003 to 2012 (Deeter et al., 2014). The model results as well as the model biases are shown for April 2016. The CO model estimates using C-CC (panel d) are characterized by a bias of -9×10^{17} 964 965 molec/cm² in the Northern Hemisphere (panel e), consistent with previous evaluations of CAM-chem (Emmons et al., 2020). In C-GC (panel b), a negative bias still exists in the Northern Hemisphere (panel c), but is smaller at -5×10^{17} molec/cm². Across 966 967 all three model configurations a north-south gradient is observed in the model bias, with the bias in the Southern Hemisphere being approximately 10¹⁸ molec/cm², which is of greater magnitude than the (negative) bias in the Northern Hemisphere. The 968 969 results from S-GC (panel f) are nearly identical to those in C-GC, with a smaller negative bias in the Northern Hemisphere 970 (panel g) than C-CC, but a larger positive bias in the Southern Hemisphere.



973 *Figure 21.* Total CO column in molec/cm² as observed by MOPITT during April for the 2003-2012 time period. (panel a), compared to the
974 results from C-GC (2nd row, panels b and c), C-CC (3rd row, panels d and e), and S-GC (4th row, panels f and g) for April 2016. The
975 measurements and model results are presented on the left (a, b, d, f), while the model biases are shown on the right (c, e, g).

976 5.4. Wet and dry deposition tendencies

Finally, we compare simulated and observed surface deposition. Since deposition is the primary removal mechanism for atmospheric reactive nitrogen and sulfur species, the ability of a model to reproduce observed patterns of deposition provides an aggregate diagnostic for its representation of emissions, atmospheric chemistry, and the physical deposition processes.

980

972

981 Recent measurements have provided wet deposition rates in numerous geographical locations for the years 2005 to 2007 (Vet

982 et al., 2014). Dry deposition fluxes are available from the same study but are limited to sulfur and nitrogen species. They are

also limited to fewer geographical locations. Nonetheless we compare results from all three model configurations to the results

984 from Vet et al. (2014) below.

Figure 22 compares the model-evaluated wet deposition rates of nitrogen at the surface for C-GC, C-CC, and S-GC. The total
nitrogen flux is calculated by adding surface fluxes from each individual nitrogen compound undergoing wet deposition.
Rainwater composition measurements are also displayed where available for comparison. We find correlation coefficients of
0.65, 0.66, and 0.67 for C-GC, C-CC and S-GC respectively with these observations. On average, the results from C-GC (panel
b) are closest to parity with a slope of 0.6, compared to 0.5 and 0.49 for C-CC (panel d) and S-GC (panel f). We do not find
any clear trends by location between the three models.





999 Comparing the dry deposition flux of nitrogen species at the surface from C-GC, C-CC and S-GC to in-situ measurements 1000 over North America (the only region present in this data set) from 2005 to 2007 shows that all models have positive biases. 1001 Relative to an observational mean of 1.57 kgN/ha/yr, C-GC has the best performance with a mean bias of +0.94 kgN/ha/yr, 1002 compared to +1.76 and +2.32 kgN/ha/yr from S-GC and C-CC respectively. These biases from all three models can be 1003 explained by either larger concentrations of nitrogen compounds or enhanced dry deposition velocities. However, we do not 1004 compensate for changes in nitrogen emissions between the time of the observations (2005-2007) and the simulated period, 1005 during which NO_x emissions are estimated to have increased (Emmons et al., 2020).

1006

Figure 23 displays the evaluated wet deposition rates of non-sea salt sulfur from C-GC, C-CC, and S-GC alongside measurements of sulfur in rainwater for 2005. When comparing across model results, we find a global mean deposition rate of 0.58, 0.38, and 0.50 kg S/ha/year in C-GC, S-GC, and C-CC respectively (computed as the global surface average of the left panels in Figure 23). The results from C-GC and C-CC show a correlation coefficient greater than 0.95, whereas C-GC and S-0C results show a correlation coefficient of 0.88.

1012

1013 Comparing to observational data, we find a mean bias of -2.40 kg S/ha/year (C-CC and C-GC) and -2.76 kg S/ha/year (S-GC) 1014 between the simulation results and rainwater composition measurements. This bias is location-dependent, with simulated data 1015 for Asia showing a lower bias than North America or Europe. For instance, over North America, measurements indicate a 1016 mean sulfur wet deposition flux of approximately 5 kg S/ha/year (for the year 2005), while the results at the same stations are 1017 lower with the slope of the linear fit equal to 0.2, 0.1, and 0.2 for C-GC, S-GC and C-CC respectively. This can be explained 1018 by the reduction in the sulfur wet deposition surface flux over the past decades. Previous literature has found that the deposition 1019 rate of sulfur over the Eastern U.S. has been decreasing at a rate of 1 kg S/ha/year² since 1990, with 60% of the reduction being 1020 in wet deposition rates and 40% in dry deposition rates (Zhang et al., 2018). Similar findings have been suggested for wet 1021 deposition rates over Europe (Theobald et al., 2019). A similar, but more recent, decrease over Asia has also been observed 1022 (Aas et al., 2019).



1025 Figure 23. As in Figure 22, but now for non-sea salt sulfur. Rows: C-GC (top), S-GC (middle), and C-CC (bottom).

1026 It is difficult to say with confidence that the calculated bias is purely due to lack of recent data without new measurements to 1027 support this conclusion. However, our results do show that the simulation of sulfur deposition in C-GC more closely follows 1028 that in C-CC than that in S-GC. This could be due to either the simulated distribution of precipitation, the representation of 1029 aerosol, or the representation of scavenging processes, all of which differ between C-GC (or C-CC) and S-GC.

1030 6. Discussion and conclusion

We present the first implementation of the GEOS-Chem chemistry mechanism as an option in the Community Earth System Model (CESM). In addition to allowing users of CESM to take advantage of advancements in atmospheric modeling implemented in the GEOS-Chem model, this also allows the community to better understand why models disagree and how progress might be made to improve model performance and accuracy.

1035

1036 Our results suggest that differences in the representation of tropospheric halogen chemistry – in particular the representation 1037 and magnitude of emissions of short-lived bromine and chlorine sources – may be responsible for differences in simulated 1038 ozone between these model configurations. However, in addition to the recognized differences in chemical mechanisms, subtle 1039 structural differences in atmospheric models may have a significant role. Our evaluation of tropospheric ozone concentrations 1040 suggests that one of the key drivers in differences between CAM-chem and GEOS-Chem ozone fields differences may be 1041 differences in free tropospheric water vapor. Similarly, we show that sulfur deposition rates are approximately twice as great 1042 when running GEOS-Chem in a standalone model as opposed to running GEOS-Chem embedded in CESM, despite the use 1043 of identical emissions.

1044

We also find that differences in the representation of wet scavenging are a significant contributor to differences in reactive nitrogen and halogen species distributions between GEOS-Chem and CAM-chem. The unification of convective transport and scavenging in GEOS-Chem helps to prevent movement of soluble species to the upper troposphere through convective updrafts, and therefore limits the effect of near surface halogen emissions from sea salt on ozone at higher altitudes.

1049

Our implementation of GEOS-Chem in CESM is now publicly available for use. We envision that this model can become a powerful tool for research, forecast, and regulatory applications of global atmospheric chemistry, air quality, and climate research. Furthermore, this is also an important step towards the Multiscale Infrastructure for Chemistry and Aerosols (MUSICA), and thereof a truly modular Earth system model. By enabling us to fairly compare models down to individual processes, we can begin to understand precisely why different models perform better or worse in reproducing different measurements and accelerate our efforts to improve atmospheric modeling fidelity as a whole.

1056

Finally, this work will foster collaboration between the GEOS-Chem and CESM-CAM-chem communities. GEOS-Chem is presently used and developed worldwide for research by over 100 university groups and government agencies, and CAMchem similarly has numerous users and developers. The availability of GEOS-Chem as an option in CESM will stimulate broader interest in the GEOS-Chem community to use CESM, and in the CESM community to use GEOS-Chem. Indeed, we expect that on-line simulation of atmospheric chemistry will become increasingly attractive to GEOS-Chem users as the resolution of dynamical models increase, and that CESM will provide the principal vehicle for this because of its public 1063 availability and support. By enabling improvements developed for GEOS-Chem to percolate into CESM without the need for

1064 re-implementation, this work will accelerate progress in atmospheric chemistry and Earth system modeling.

1065 Author contributions

1066 TMF, SDE, HL, and EWL were responsible for the software development. TMF performed the investigation, formal analysis, 1067 and validation. SDE, LKE, SRHB, and DJJ conceived of the project and acquired funding. SDE, LKE, SG, SRHB, and DJJ 1068 supervised the work. TMF performed all visualization and prepared the original draft. Review and editing were performed by 1069 all co-authors. All contributions are defined according to the CRediT taxonomy (https://casrai.org/credit).

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1074

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 secondary organic aerosols.

1078 Code availability statement

GEOS-Chem as an option within CESM is currently being implemented into the CESM main branch, such that no additional download will be needed to use it. However, a standalone copy of the specific implementation of CESM including GEOS-Chem which was used to generate the results in this manuscript is permanently archived at https://github.com/CESM-083 GC/CESM-GC-Standalone/releases/tag/v1.0.0_review (permanent DOI: https://github.com/CESM-1083 reproduce the results of this work, the repository should be used as-is without using features such as checkout_externals to acquire any additional code.

1085 Data availability statement

Output data from the model are available from the authors upon request. Satellite data retrievals including from OMI/MLS
and of carbon monoxide from the Terra/MOPITT instrument are available from the NASA Earth data archive
(https://earthdata.nasa.gov; ftp://15ftl01.larc.nasa.gov/MOPITT/). Surface observations of carbon monoxide are available from

- 1089 the NOAA Earth System Research Laboratory Global Monitoring Division data archive
- 1090 (https://www.esrl.noaa.gov/gmd/dv/ftpdata.html).

1091 **Competing interests**

1092 The authors declare that they have no conflict of interest.

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