

Title: "Short-Lived Halogen Sources and Chemistry in the Community Earth System Model v2 (CESM2-SLH)"

Tracking #: 2024-01975

Authors: Fernandez et al.

Within this response letter, we use the following text format: Reviewer comments appear in regular black font, while our responses are in blue font. The original manuscript text is shown with double quotes and in *"italic-blue format"*, while the changes introduced in the revised draft are in ***"bold-italic blue"***. Please note that the line numbering used here (e.g., **L528-533**) points at the track-changed version of the revised draft.

Reviewer Comments:

Reviewer #1:

The manuscript by Fernandez et al. describes the implementation of short-lived halogen chemistry already available in CESM1 into CESM2, thereby making it accessible to the broader CESM community. It provides a detailed description of the implemented SLH chemistry (incl. offline/online emissions, chemical reactions and deposition), an analysis of the SLH budgets, spatial/vertical distributions and impacts on oxidants. The analysis is very detailed and can act as benchmark evaluation for future studies using CESM2-SLH. It fits well into the scope of GMD and I see no reason why it should not be published after addressing the following minor comments.

GENERAL ANSWER: We thank the reviewer for recognizing the detailed description and analysis of manuscript, and for the very useful and constructive suggestions that helped to improve the quality of our work.

Given the large number of individual comments, and to avoid excessive repetition, in this section we present a short and general answer to the main comments, and provide below detailed response to each of the specific comments and independent questions.

General comments:

1.

R1-C1.0: Parts of the manuscript (especially the beginning of Section 2 and Section 2.5) read a bit like a CESM user manual. The information is without doubt very useful for CESM users. However, some adaptations might help to make it easier to follow for readers outside the CESM community. The manuscript is also relatively long, so the authors might want to consider moving some more technical parts to the supplement.

R1-C1.4: L662-686: Could be moved to the supplement?

GENERAL ANSWER: We agree with the reviewer that the original manuscript had a lot of technical information and accepted his/her suggestions of moving several blocks to the supplement. As detailed below, we have re-structured the manuscript and included a completely new Appendix that contains the main technical information as well as specific user-namelist configurations. We hope these changes help to shorten the manuscript length and ease the reading for the general audience, while keeping fine details of relevance

for the CESM community. In particular, note that original Table 7 has now been moved to the Appendix and renamed to Table A1. Please refer to the last section of the revised manuscript for the complete Appendix text (see also related responses to **R1-C1.3** below).

R1-C1.1: L138-164: The authors might want to consider merging this section and Section 2.5 as some of the information is redundant (e.g. CESM branches, compsets), and it would be beneficial to have the information at the same place. Additionally, a definition/introduction of “compset” would be helpful. I understand that it is the CESM name for “simulation setup” and specifies the used BC and emissions, and settings like resolution etc.. It would be helpful if Table 6 would be referenced here (if not merged) to summarise the mentioned compsets. Additional columns could be introduced in Table 6 to explain the difference between compsets. For example, I assume that FCnudged means nudged meteorology and FCHIST historical free-running, but it is not explained explicitly. Also the difference between FCnudged and FWnudged is not explained explicitly. What is FCASE?

GENERAL ANSWER: We thank the reviewer for this very important suggestion, which is also in line with comment **R2-C1.4** and **R2-C9.6** from the second reviewer. Based on both suggestions, we introduced a whole new section entitled “**3.2 Model Experiments and Sensitivity Simulations**”, where we include the new Table 7 and describe the whole set of experiments performed in this work. In addition, we removed the usage of the term “*compset*” from the abstract and introduction, and is then introduced in Section 3, where we define and explain the different CESM2-SLH model configurations first, followed by an independent description of the whole set of experiments performed.

Based on the above, we have revised the manuscript as follows:

L515-518: “*The implementation of SLH sources and chemistry described in this work was performed over the base FCnudged and FCHIST atmospheric chemistry compsets (<https://docs.cesm.ucar.edu/models/cesm2/config/2.2/compsets.html>). In CESM terminology, compset is an acronym for ‘component setup’ and refers to specific model configuration of the Earth System Model, specifying which components are used and how they are coupled (Danabasoglu et al., 2020).*”

L548-565: “*Two main CESM2-SLH configurations have been developed based on the available CAM6 base atmospheric chemistry compsets: FCHIST_slh and FCnudged_slh (see Table 6), where the former resolves atmospheric dynamics internally (free-running) and the latter is forced by prescribed meteorological fields (new nudging approach for specified dynamics). Both configurations employ the coarse f19_f19_mg17 horizontal resolution (1.9° latitude × 2.5° longitude, hereafter 2°×2°) and 32 vertical levels (low-top) from the surface to approximately 40 km (~4 hPa)” ... “In addition to the low-top configurations, whole atmosphere (high-top) fully-coupled BWHIST_slh (f19_g16) and FWnudged_slh (f19_f19_mg17) compsets were developed, both considering the coarse 2°×2° resolution and 70 vertical levels as their corresponding base atmospheric WACCM compsets. Table 6 summarizes the names and specifications of the main CESM2-SLH compsets.*”

L569-578: “**3.2 Model Experiments and Sensitivity Simulations**

The complete set of model experiments performed in this work is presented in Table 7. Each experiment’s name is composed by a prefix denoting the model used and the inclusion of short-lived halogens (e.g., CC-SLH for CAM6-Chem with SLH turned on) and a suffix indicating the model resolution and meteorology option (e.g., [2×2-ndg] for coarse resolution and nudging approach). Although all model configurations have been validated, initial results presented in Section 4 are mostly based on low-top and coarse CC-SLH [2×2-ndg] and/or CC-SLH [2×2-hst]

experiments. We note that for each of the six SLH experiments listed in Table 6, an additional sensitivity was conducted neglecting the sources and chemistry of short-lived halogens (e.g., the corresponding CC-NOH [2×2-ndg] experiment, see Appendix). For most analysis comparing CC-SLH and CC-NOH distributions, we used the FCnudged_slh compset as this ensures that dynamical transport is consistently represented across experiments, which is crucial for isolating and quantifying chemical changes.

Table 7: CESM2-SLH experiments performed in this work.

Experiment name	SLH compset	Resolution (lat × lon)	Period of time
CC-SLH [2×2-ndg]	FCnudged_slh	(1.9° × 2.5°)	1980 - 2020
CC-SLH [2×2-hst]	FCHIST_slh	(1.9° × 2.5°)	1950 - 2020
CC-SLH [1×1-ndg]	FCnudged_slh	(0.9° × 1.25°)	2000-2005
CC-SLH [1×1-hst]	FCHIST_slh	(0.9° × 1.25°)	2000-2005
FW-SLH [2×2-ndg]	FWnudged_slh	(1.9° × 2.5°)	2000-2005
BW-SLH [2×2-cpl]	BWHIST_slh	(1.9° × 2.5°)	2000-2005

In addition to the CESM2-SLH experiments presented above, and with the intention to validate current model performance compared with previous CESM1 studies, we also analysed model output from the main simulations originally published by Li et al. (2022) and Saiz-Lopez et al. (2023), as well as in Barrera et al. (2023) and Bossolasco et al., (2025). For consistency, these experiments are referred to, respectively, CESM1-SLH [QL22], CESM1-SLH [SL23], CESM1-SLH [JB23] and CESM1-SLH [AB25]. Although these CESM1 configurations are not strictly identical, the experiments were conducted considering analogous spatial resolution and nudging options to those employed here in CESM2-SLH. They are therefore considered as the benchmark simulations for quantitative and qualitative intercomparison between CESM model versions. Further details of the main similarities and differences between SLH results performed with CESM1 and CESM2 are described below.”

R1-C1.2: L606: It is not clear to me what the difference between cesm2.2-asad-branch_slh and cam_cesm2_2_rel09_slh is. Could you clarify why you need two branches?

GENERAL ANSWER: CESM is a modular Community Earth System Model where each independent component is developed and maintained by different working groups. One of these components is CAM (Community Atmospheric Model) which includes the main dynamical core, cloud microphysics, radiative transfer routines and certainly, the atmospheric chemistry package described in this work. Despite most of the SLH developments described in this work (e.g., FORTRAN modules and routines, compset definitions, user-namelist, etc.) the fully-coupled compsets and some other configuration scripts must be updated within the main CESM branch (cesm2.2-asdbranch_slh). To avoid any confusion and make this clear to the reader not familiarized with CESM, we have rephrased the text as follows:

L528-533: “CESM2-SLH porting was performed on top of version 2.2.0 of CESM2 (Danabasoglu et al., 2020), particularly over branch cesm2.2-asdbranch, which includes CAM6 tagged version cam_cesm2_2_rel_09. Based on these versions, we forked and created the new CESM2 and CAM6 tags called cesm2.2-asdbranch_slh and

cam_cesm2_2_rel_09_slh, respectively, which incorporate SLH updates within the main FORTRAN routines as well as modifications to **building scripts** (e.g., *cime* and *cime_config*) and default namelist variables (see **Appendix**). These **open-access community developments allow to download, clone and build all SLH compsets available (see Code and Data Availability below; Fernandez et al., 2025).**

R1-C1.3: L610-619: Could be moved to the supplement? The information could be presented in a table listing the additional species, and if they emit, dry/wet deposit etc.

GENERAL ANSWER: We thank the reviewer for the careful reading and accept his/her suggestion of moving the referred text to the Appendix. The new Appendix includes now a brief introduction of the original CESM modules and routines modified, as well as a detailed description of the main configuration changes and namelist variables that were updated when implementing SLH sources, chemistry and deposition. In particular, we move one of the original tables of the main text to the Appendix, and also compiled in a new table all the relevant photolysis, bimolecular and termolecular reactions of organic and inorganic SLH included in the chemical mechanism.

R1-C1.6: Also Section 4 “Final Remarks is quite technical (e.g. L1602-1606). I think it would be valuable here to discuss if the performance of CESM2-SLH (regarding SLH budgets and impact on atmospheric composition) is similar/better/worse compared to CESM1-SLH/ CAM4-Chem, and other models including SLH chemistry.

GENERAL ANSWER: We thank the reviewer for the suggestion. Following this and another comment from reviewer R2, we have reorganized the manuscript structure and include all technical blocks into the Appendix, while also include a dedicated Discussion section (Section 5) that contains a comprehensive analysis of CESM2-SLH performance. This includes a detailed analysis of the main changes in the representation of halogen distribution between the different experiments and previous studies (Section 5.1) as well as a brief evaluation of the atmospheric impacts estimated with different models. In doing so, we also modified the title of the final Section 6, which is now called “Summary and Conclusions”. Below we provide a couple of examples of sentences included in Sections 5.1 and 5.2, and we kindly ask the reviewer to refer to the revised manuscript for the complete changes introduced in the text:

L1415-1424: *“The current CESM2-SLH configuration applied ~15% enhancement to oceanic VSL bromo- and chloro-carbons to reproduce previous CESM1 distributions. This resulted in SG vertical profiles and trends that lay in the lower margin of the most recent assessment of VSL_{Cl} contribution to chlorine stratospheric injection (Fig. 5a). Therefore the offline emissions and LBCs for anthropogenic chlorinated halocarbons described in Section 2.1.2 could be increased by a factor larger than 15% to reproduce the observed trend in the recent past (Hossaini et al., 2024; WMO, 2022). Due to the large anthropogenic contribution from developed regions over US, Europe and Asia (Claxton et al., 2020; Hossaini et al., 2019), the VSL_{Cl} distribution for the Northern and Southern mid-latitudes shown in Fig. 7 shows a clear hemispheric asymmetry just below the tropopause. This asymmetry on VSL distributions have already been described in the literature for chlorine (Roozitalab et al., 2024) as well as bromine (Jesswein et al., 2022; Keber et al., 2020), although note that anthropogenic VSL_{Br} sources are currently not considered in CESM2-SLH.”*

L1448-1459: *“The tropospheric OH burden of 222.8–226.2 Mg obtained for nudged CC-SLH [2×2-ndg] and free-running CC-SLH [2×2-hst] experiments corresponds to a global mean tropospheric OH concentration of approximately 1.23–1.27×10⁶ molec. cm⁻³, consistent with previous model results neglecting SLH chemistry (Lelieveld et al., 2016) and in the higher edge of the range reported in the literature for the 21st century (11.7±1.0×10⁵ molec. cm⁻³; Voulgarakis et al., 2013). While the relative contribution of SLH to OH production and loss remains consistent between CESM1 and CESM2-SLH (Table 11), the absolute OH burden is substantially higher in CESM2-SLH compared to CESM1 (Table 9). This discrepancy is not attributable to the implementation of SLH sources and*

chemistry, but rather due to structural changes in between CESM versions, including changes in VOCs degradation, pollutant emissions, prescribed LBCs and the overall atmospheric oxidative capacity of CAM6-Chem relative to CAM4-Chem (Emmons et al., 2020). Although the absolute values of P and S differ between CESM1 and CESM2-SLH due to the different background state of the modelled atmosphere, the overall magnitude and relative contribution of the dominant OH production pathways remain consistent with previous studies using CESM1 and other global chemistry models (Bossolasco et al., 2025)."

2.

R1-C2.0: The comparison with observations is somewhat limited. In Section 3.2.2. "Evaluation of SLH Abundance with Observations" Source Gas Injection and Product Gas Injection to the stratosphere is compared to observations. It would be beneficial to extend the evaluation to SLH abundance to the (lower) troposphere, MBL or surface (for example comparison to station data). In Section 2, a comparison of VSL halocarbons to observation is mentioned (e.g. in L273), but no details are given.

R1-C1.5: Page 12: line 16: Again, please provide a map of these sites

ANSWER: We thank the reviewer for the careful reading and for the suggestion of strengthen the model validation. Following the reviewer request, in addition to the comparison of VSL_{Cl}, VSL_{Br} and VSL_I tropical vertical profiles with WMO assessment, the revised validation Section 4.2.1 now includes a comparison of surface reactive halogen abundance (in particular, BrO and IO) obtained during different coastal and ship-born campaigns. The new Figure 6 presents the modeled distribution of BrO and IO extracted at exactly the same locations and periods of time as daily-mean observations performed during 4 different campaigns. The new figure is accompanied by a short description of CESM2-SLH performance and limitations based on model-observations correlations.

L913-931: *"Additional model validation was performed extracting hourly surface reactive halogens mixing ratios for the CC-SLH [2×2-ndg] experiment at the same location and time as previously published observations obtained in multiple measurement campaigns. Three ship-based campaigns measured IO concentrations: TransBrom, conducted in October 2009 from Japan to Australia (Großmann et al., 2013), Poseidon, which surveyed the coasts of Northwest Africa between the Canary Islands and Cape Verde in June 2010 (Bange, 2011) and Malaspina, conducted between January and July 2011, which circumnavigated the globe from Brazil to Spain via Australia and the Panama Canal (Prados-Roman et al., 2015b). Additionally, we compared measurements from a coastal station in Cape Verde between April and June (Mahajan et al., 2010; Read et al., 2008). The ship routes and the location of Cape Verde are shown in the central panel of Fig. 6. The side panels compare daily mean modelled (red) and observed (blue) values, where the shading and error bars represents the modelled spread and the reported observational uncertainty, respectively. A general good agreement is achieved, especially for IO where observed values ranging between 0.5 and 2 pptv with a sharp variability that can exceed 1 pptv between consecutive days is captured in magnitude by the model, although not always for the exact time and location. The maximum values predicted in Malaspina during late May and early June (Fig. 6d) are attributed to highly localized IO bursts due to the large partitioning shift between Iy species driven by SSA recycling (Prados-Roman et al., 2015b). Similarly, near-zero BrO values are modelled in Cape Verde (Fig. 6a) due to the negligible contribution of SSA-debromification predicted by the model during early April. However, during late May the model is capable of reproducing the magnitude of sporadic BrO peaks due to the improved representation of SSA abundance over the east-Atlantic. Despite the remaining model-observation discrepancies, we highlight that CESM2-SLH results in a general underestimation of observed reactive halogen abundance, which implies that the predicted influence of SLH on atmospheric composition is, in any case, a lower limit."*

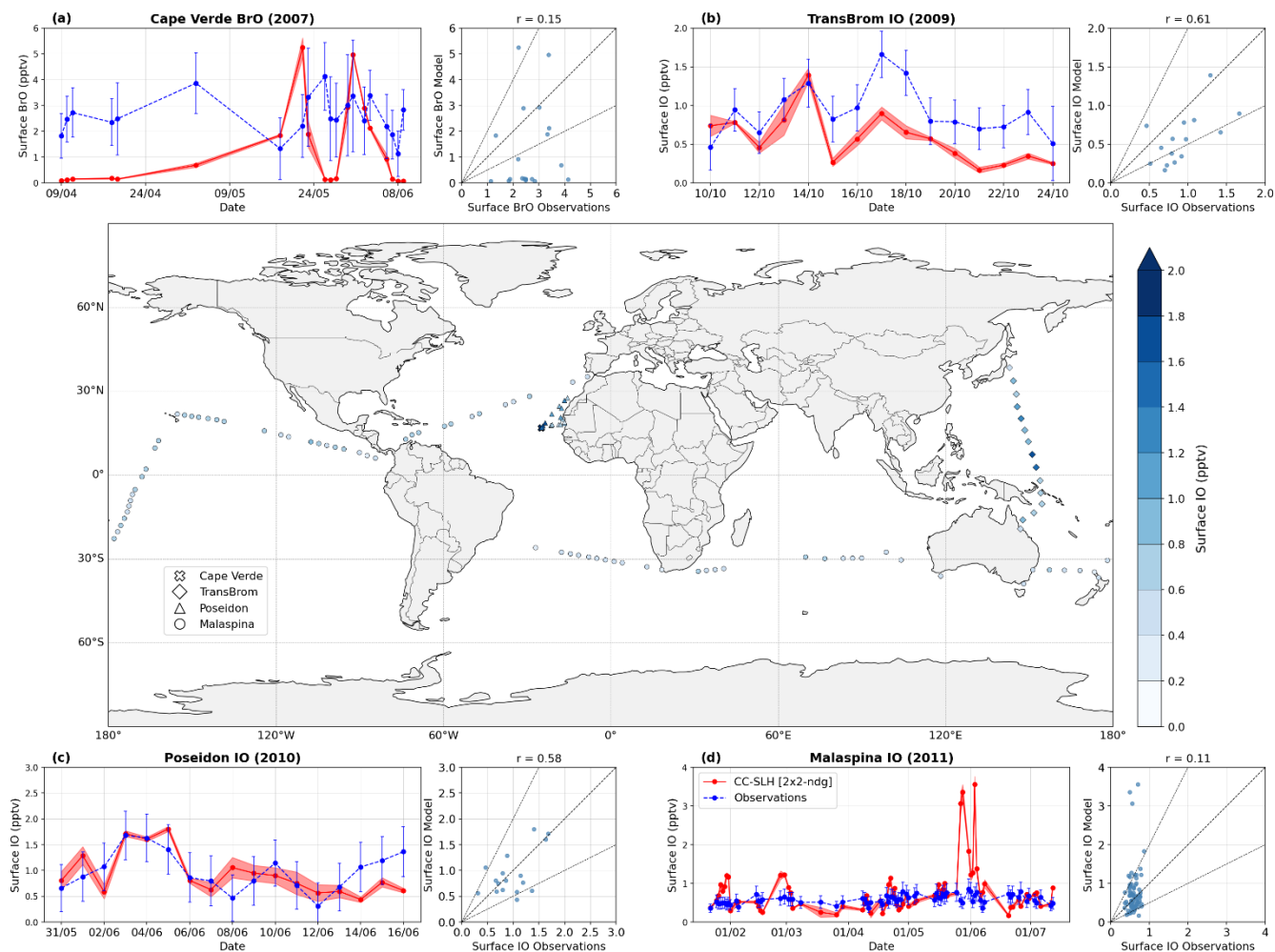


Figure 6: Comparison of nudged CC-SLH [2x2-ndg] surface mixing ratios with IO and BrO observations from multiple campaigns. The central panel shows the location of the Cape Verde station which measured BrO in 2007 (cross) and the routes of three ship-based IO campaigns: TransBrom in 2009 (diamond), Poseidon in 2010 (triangle) and Malaspina in 2011 (circle). Markers colours indicate IO volume mixing ratio in pptv. Side panels show time series of observations (blue dashed line with reported root-mean square error (RMES) bars) and model outputs (red line with ± 1 standard deviation), along with modelled vs. observed scatterplots with correlation coefficients (r). Measurement campaigns are (a) Cape Verde; (b) TransBrom; (c) Poseidon and (d) Malaspina. All data corresponds to daytime conditions.

In addition, we repeated the CC-SLH [2x2-ndg] and CC-NOH [2x2-ndg] experiments for year 2015 and extracted the surface ozone abundance with hourly resolution. This allowed us to compare our modelling results with the same temporal resolution as the TOAR dataset shown in the original Figure 14 (now Figure 15). The revised validation figure, now moved to Section 4.3.1, shows the mean $\pm$ range (computed as the standard deviation of hourly values) for modelled (shading) and observations (black bars).

L1197-1205: “Overall, the model captures the observed seasonal ozone cycles across most stations. This agreement is particularly strong at northern mid-latitude stations ($>30^\circ$ N), where the model reproduces both the magnitude and seasonality of ozone, with peaks in spring-summer driven by enhanced photochemical production. Similarly, good representation of ozone seasonality is observed at station h) located in east Asia as well as in station l) south of New Zealand. Most notably, the inclusion of SLH chemistry results in an overall reduction of ozone that reduces the model bias and in most locations is larger than the spread of modelled values between the CC-SLH and CC-NOH experiments. However, discrepancies emerge at some stations like i) in central Chile, where the model successfully reproduces the observed ozone magnitude but fails to capture the seasonal cycle. This divergence likely stems from

incomplete representation of ozone sources and sinks in this pristine region, as well as due to the coarse resolution used.”

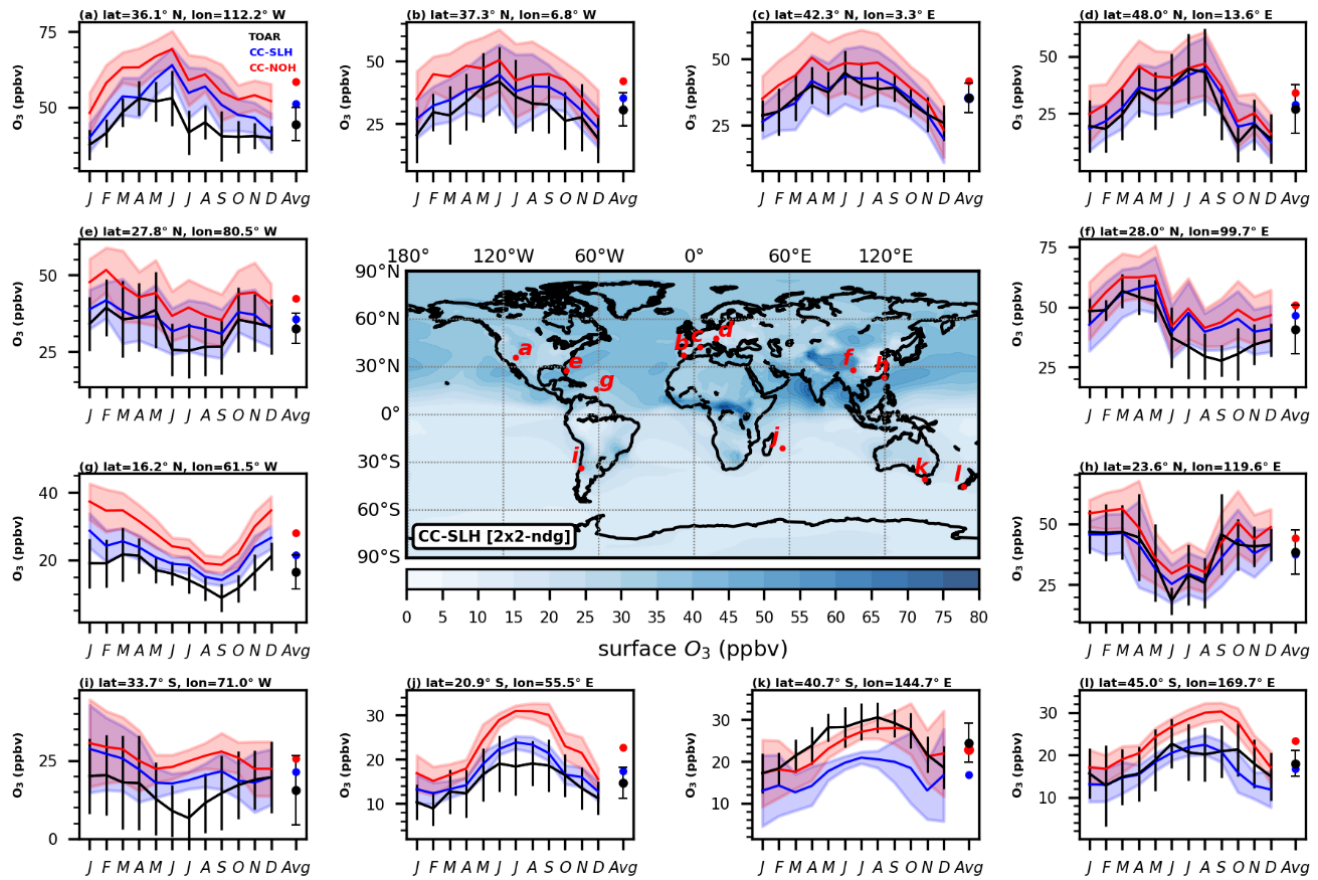


Figure 15: Comparison of CESM2-SLH nudged CC-SLH [2x2-ndg] and CC-NOH [2x2-ndg] simulations with TOAR-I observations. The center panel shows the geographical distribution of surface ozone for the CC-SLH [2x2-ndg] experiment, as well as the location of each of the observational sites shown in the side panels (red points). The side panels (a-l) show the monthly mean $\pm$ range seasonality (solid lines and shading) and the annual average (separate marker) of surface ozone for CC-SLH [2x2-ndg] (blue) and CC-NOH [2x2-ndg] (red) experiments. Black lines and error bars show surface observations reported in the first phase of Tropospheric Ozone Assessment Report (TOAR-I) (Schultz et al., 2017). Both model output and observations ranges have been computed as the standard deviation of hourly data for year 2015.

Finally, we note that we have revised the manuscript to avoid pointing at model evaluation details in other sections of the paper focused on the technical description of the SLH developments implemented.

Specific comments

R1-C2.1: L214: Another model with SLH chemistry would be TOMCAT (Hossaini et al., 2016; <https://doi.org/10.1002/2016JD025756>).

ANSWER:. The reviewer is completely right. We apologize for the unintended omission. We have rephrased the sentence as follows:

L189-191: “These include global and regional models such as *TOMCAT* (Hossaini et al., 2016), *GEOS-Chem* (Sherwen et al., 2016a, b), *CMAQ* (Sarwar et al., 2015), *WRF-Chem* (Badia et al., 2019), *SOCOL* (Karagodin-Doyennel et al., 2021) and *LMDZ-INCA* (Caram et al., 2023).”

R1-C2.1: L241: Could you explain which VSL halocarbons are compared to observations, and add references for the surface and aircraft observations you compare to?

ANSWER: Table 1 in Ordóñez et al. (2012) presents a compilation of the 13 airborne observations used in the development of the emission inventory, while 16 different observational campaigns at different locations in the MBL over the world are compiled in their Table 2. These include: PEM-Tropics (A and B), TRACE-P, INTEX-A, INTEX-B, TC4, ARCTAS, STRAT, POLARIS, SOLVE, Pre-AVE, H-AVE II and CR-AVE, as well as Cape Grim, Christmas Island, Hawai, Hateruma Island, Thomson Farm, Cape Oxhiishi, Roscoff, Mace Head, Norfolk, Dagebull and other locations over Atlantic, Pacific and Southern Ocean. We appreciate the reviewer suggestion, but we believe that including such a long description is not necessary in this manuscript and prefer to point at the original reference for scientific support. We note that most of the aircraft campaigns detailed above are also considered in WMO (2022) report to assess the contribution of VSL bromine to stratospheric ozone depletion, which is used in this work to validate SGI and PGI (see Figures 8 and 9). Therefore, we have modified the text in the revised manuscript as follows:

L211-214: “The methodology follows a top-down approach based on chlorophyll-a monthly climatology from the *SeaWIFS* project for the 1998–2003 period (Melin, 2013), and the iterative adjustment of emission flux strength by comparing modelled distributions and vertical profiles of VSL halocarbons with *a compilation of aircraft campaigns and surface observations* (see Tables 1 and 2 in Ordóñez et al. (2012)).”

R1-C2.2: L292: Did you compare VSL chlorocarbons to observations as well (if yes, please provide more details), or is the rationale being consistent with the scaling of natural emissions?

ANSWER: For the particular case of VSL chlorocarbons, we did not develop our own emission inventory but implemented the anthropogenic emissions and LBC described in the Hossaini et al. (2019) and Claxton et al. (2020). Therefore, no iterative adjustment to observations was performed in this case. Once we noted that 15% enhancement to oceanic VSLS emission was required, we applied the same enhancement to the anthropogenic VSLS inventory and, as expected, we were able to produce previous results.

In order to be completely clear and transparent about the procedure we follow during VSL chlorine implementation, we have rephrased the complete section as follows:

L247-257: “For anthropogenic VSL sources of chlorocarbons, we follow the Hossaini et al. (2019) and Claxton et al. (2020) emission inventories, which include time-dependent emission of two major contributors to the organic chlorine load (CH_2Cl_2 and C_2Cl_4) as well as surface LBCs for two other compounds (CHCl_3 and $\text{C}_2\text{H}_4\text{Cl}_2$). Based on these works, an increasing trend from 2000 to 2020 is applied to the emission and LBC files for the recent past. Beside a fraction of CHCl_3 emissions arise from natural sources (with minor contributions from the other compounds), chlorinated VSL are dominated by anthropogenic emissions that display a pronounced hemispheric asymmetry (WMO, 2022). Therefore, hereafter we assume all of these offline emissions to have only an anthropogenic origin. Consistent with the scaling factor applied to the Ordóñez et al. (2012) inventory, both VSL chlorocarbon surface emissions and LBCs were globally scaled by the same constant factor (1.15 or 15% enhancement). It should be noted that additional anthropogenic halocarbon sources, as those arising from the waste treatment of power-plants as well as seaweed aquaculture are not considered (Carpenter et al., 2000; Jia et al., 2023; Leedham et al., 2013).”

R1-C2.3: L340: What is the reasoning for assuming free-regime approximation, i.e. why is diffusion not considered (Schwartz 1986)? Could it play a role for larger SSA particles?

ANSWER: The representation of heterogeneous recycling of halogens on SSA is a complex process with multiple elemental reactions that can only be implemented in global models through a simplified parameterization. Due to the rapid reaction of oxidized halogen reservoirs with SSA bromide and chloride, as well as the insolubility of the di-halogen species produced (Br_2 , BrCl , IBr , ICl and Cl_2 , see our Table 1), in Eq. 3 we assume that the initial uptake of inorganic halogen species onto aerosols is the rate-limiting step. This is based on the original work of McFiggans et al. (2000) as described in the Supplementary Material of Ordóñez et al. (2012). Given that in CESM2-SLH we do not explicitly treat neither aqueous phase chemistry nor diffusion within the bulk of the sea salt aerosols, we compute the free molecular collisional frequency using FRA and assume this process represents the rate limiting step of the reaction.

In order to make this clear, and to highlight the main limitations of the parameterized implementation, we have rephrased the referred sentence as follows:

L285-292: *“This complex redox process is parameterized to proceed in a single-step heterogeneous reaction dependent on the collisional frequency of the gas-phase species and the substrate, as well as on the degassing efficiency of the halogenated product released to the atmosphere (Ordóñez et al., 2012). Given that CESM2 does not include an explicit treatment of aqueous phase chemistry and diffusion, we assume the rate-limiting step is the uptake of inorganic halogen species onto SSA and computes the first-order reaction rate based on the Free-Regime Approximation (FRA) approach (McFiggans et al., 2000) following Eq. (3)”.*

L304-309: *“The FRA implementation in CESM2-SLH assumes that the bromide and chloride content of SSA is sufficiently large to act as an effectively infinite halide reservoir capable of sustaining the surface heterogeneous-redox reaction. This implies that the bromide and/or chloride content in the SSA bulk is always in excess compared to the abundance of the gas-phase halogen reservoir that deposits over the aerosol surface, which is valid for fresh SSA typically found close to the ocean surface. Therefore, the implementation of SSA-dehalogenation does not account for the slowing down of the halogen recycling when aged sea-salt aerosols become depleted in bromide and chloride (von Glasow et al., 2002).”.*

R1-C2.4: L396: Could you provide the formula to calculate the yield (e.g. add it in Table 1). I could not find the formula in the referenced publication Li et al., 2022 either.

ANSWER: We have now included the ClNO_2 yield formula in the footnote of Table 1 as follows:

L322-324: *“& The ClNO_2 yield is computed as $\text{yield}^{\text{ClNO}_2} = \left(\frac{\text{SSA}_{\text{mass}} - \text{SSA}_{\text{min}}}{\text{SSA}_{\text{max}} - \text{SSA}_{\text{min}}} \right)^3$, where SSA_{mass} is the total sea-salt aerosol mass mixing ratio of all aerosol bins in each model gridbox, while $\text{SSA}_{\text{min}} = 5.0 \times 10^{-10}$ and $\text{SSA}_{\text{max}} = 1.0 \times 10^{-9}$ are the minimum and maximum SSA_{mass} adjusted thresholds for $\text{N}_2\text{O}_5 \rightarrow 2 \cdot \text{HNO}_3$ implemented in Lamarque et al. (2012).”*

R1-C2.5: L520: Could you clarify what is meant by "following the HNO_3 acid displacement developments from Hossaini et al., 2016"? The HNO_3 acid displacement is het_ss_9, isn't it? Do you mean that a similar approach as for this reaction is used? Hossaini et al., 2016 also implemented heterogeneous recycling on non-SSA particles (e.g. their reaction 6).

ANSWER: We apologize by the confusion introduced by pointing at “ HNO_3 acid displacement developments” here. Note that in current Section 2.2.3 (Table 2) we are pointing at “stoichiometric”

heterogeneous reaction implemented in the model (i.e., *usr_N2O5_aer2*) that results in a net change of partitioning. This differs from previous processes described in Section 2.1.3 (Table 1) that represented “non-stoichiometric” reactions that constitute a net source of chlorine (i.e., *het_ss_12*). Given that the halide content of non-SSA is limited, it is assumed that the initial condensation of gas-phase HCl must occur for the heterogeneous reaction to proceed (Osthoff et al., 2008).

In order to be completely transparent about the implementation of stoichiometric recycling reactions in CESM2-SLH, we have rephrased the complete paragraph as follows:

L428-437: *“The heterogeneous recycling reaction of N_2O_5 shown in Table 2 (*usr_N2O5_aer2*) differs from the net source of chlorine occurring over sea-salt aerosols (*het_ss_12* in Table 1). Indeed, tagged reaction *usr_N2O5_aer2* occur over other tropospheric aerosols (e.g., black-carbon, organic-carbon, sulphate and/or nitrate) over continental domains, considering accommodation coefficients and production yields from McDuffie et al. (2018, 2019). The resulting $ClNO_2$ can further photolyze to release NO_2 and consequently result in additional ozone production within polluted environments (i.e., high- NO_x regimes). Given that main aerosol components within MAM4 do not represent a halide reservoir, reaction *usr_N2O5_aer2* assumes that the gas phase HCl is initially condensed at the substrate surface (Osthoff et al., 2008), which further react to produce $ClNO_2$ that is released back to the gas phase (reactive uptake). This stoichiometric recycling of nitrogen oxides is based on Li et al. (2022) following the original implementation of chlorine activation over non-SSA substrates described in Hossaini et al. (2016).”*

R1-C2.6: L708: What about the study by Mirrezaei et al. (<https://essopenarchive.org/doi/full/10.22541/essoar.175288325.58779694/v1>). They seem to use CESM2.2.-SLH in a CH_4 -emission driven setup.

ANSWER: We thank the reviewer for the careful reading and suggestions. We note that the referred preprint, currently accepted and published in JGR-Atmospheres, was currently under evaluation when we submit the first version of the manuscript. We have now updated the manuscript as follows:

L608-609: *“Nevertheless, an initial implementation of emission-driven CH_4 configuration in CESM2-SLH is presented in Mirrezaei et al. (2026).”*

R1-C2.7: Figure 2 and Figure 3: Would it be possible to show the interannual variability (e.g. standard deviation) for CESM1 Cyclical_slh? Or is it negligible?

ANSWER: The inter-annual variability of the referred *cyclical_slh* simulation (whose name has been updated to CESM1-SLH [SL23] following R2 request) is very small and hardly distinguishable from the size of the symbol used in each panel. We thanks the reviewer for the suggestion, but given that Figures 2 and 3 already include a large number of colored lines and symbols, as well as a long list of experiments within the legend (see **R2-C10.3** below), we rather keep the main panels as originally drawn.

R1-C2.8: Figure 2: Could you provide a bit more context on the differences in the trends of O_3 , OH, and NO_2 between CESM2-SLH and CESM1-SLH shown in Figure 2? You explain that these differences drive differences in the trends of halogen budgets. Are the differences of O_3 , OH, and NO_2 mainly caused by differences in prescribed boundary conditions and emissions (e.g. NO_x and VOCs) or by model updates? Are differences between CESM2 and CESM1 (without SLH) similar to the differences shown here? How do the trends compare to other models and observations (e.g. are differences between CESM1 and CESM2 small compared to inter-model differences or biases towards observations)?

ANSWER: We note that the main drivers of the different modeled trends in O₃, OH, NO₂ and VOCs between CESM1 and CESM2 are the changes in the benchmark air-pollutant emissions considered, as well as on the updates on the chemical degradation and speciation of large hydrocarbons. Indeed, that is one of the main reasons why we performed several experiments to assure that the CESM2-SLH release was able to reproduce previously published results performed with CESM1:

To make this clear, we followed the reviewer suggestion and have modified the description of Figure 2 as follows:

L702-713: *“For example, given that the dominant halogen sources in the troposphere are computed online depending on the background concentration of NO₂ and O₃ (Figure 2e,f), the surface abundance of Cl_y, Br_y and I_y (Figure 2a-c) follows the temporal evolution of the former species within each model version. **Indeed, the increasing trend in surface ozone abundance during the 1980–2020 period in CESM2-SLH compared to the more flatten trend observed in CESM1 (Fig. 2f) results in an increasing trend in iodine volume mixing ratios in the former, while I_y abundances in the latter remained approximately constant, particularly after year 2000 (Fig. 2c). A similar behaviour is also observed for Cl_y and Br_y, as their main sources depends mostly on the HNO₃ and N₂O₅ abundance as well as on the extent of partitioning of halogen reservoirs to ClONO₂ and BrONO₂, all of which increase for larger NO_x background conditions (Barrera et al., 2023). Compared with CESM1, the new benchmark tropospheric chemistry implemented in CESM2 includes updates in the oxidation of isoprene and terpenes, organic nitrate speciation, aromatic speciation and oxidation, as well as in the prescribed anthropogenic and biomass burning emission inventories, which in turn improves the representation of ozone and other secondary air-pollutants (Emmons et al., 2020).”***

Importantly, although the relative contribution of SLH to OH production and loss remains broadly consistent between CESM1 and CESM2-SLH, the absolute OH abundance is larger in CESM2. As clarified in the revised manuscript (Section 5.1), this difference mainly reflects structural updates in CAM6-Chem framework (e.g., complexity of VOC oxidation, pollutant’s emissions and NO_x–HO_x partitioning) rather than the implementation of SLH chemistry itself.

L1451-1456: *“While the relative contribution of SLH to OH production and loss remains consistent between CESM1 and CESM2-SLH (Table 11), the absolute OH burden is substantially higher in CESM2-SLH compared to CESM1 (Table 9). This discrepancy is not attributable to the implementation of SLH sources and chemistry, but rather due to structural changes in between CESM versions, including changes in VOCs degradation, pollutant emissions, prescribed LBCs and the overall atmospheric oxidative capacity of CAM6-Chem relative to CAM4-Chem (Emmons et al., 2020).”*

Please note that following reviewer R2 suggestion, we have modified the arrangement of Figure 2 to avoid including VOCs vmr in the second row where otherwise species ratios are presented (see response to **R2-C10.2** below).

R1-C2.9: L914: An even more recent estimate for GEOS-Chem might be given by Wang et al., 2021 (<https://acp.copernicus.org/articles/21/13973/2021/>)

ANSWER: We have now modified the manuscript and include an updated discussion with the most recent GEOS-Chem predictions as follows:

L794-801: *“We note that our present-day global chlorine source from acid displacement is equivalent to CESM1 (21.5 Tg Cl yr⁻¹) and around 4 times smaller than an equivalent implementation of acid displacement dehalogenation in the TOMCAT model (~90 Tg Cl yr⁻¹; Hossaini et al., 2016). **Similarly, the SSA-dehalogenation source in CESM2-SLH is between 1.5 and 3 times lower than the range of values estimated by Graedel and Keene (1995) (~37–73 Tg Cl yr⁻¹) and less than half of the recent predictions from Wang et al. (2019, 2021) (~50–64 Tg Cl yr⁻¹). For the case of bromine, our global annual flux of SSA-dehalogenation (~2.8–4.4 Tg Br yr⁻¹) remains lower than other estimates obtained with GEOS-Chem when the reactivity of bromine with tropospheric aerosols is considered (3.5–6.4 Tg Br***

yr⁻¹; Chen et al., 2018; Zhu et al., 2018), and between 4 to 7 times smaller than their most recent predictions (~20 Tg Br yr⁻¹; Wang et al., 2019, 2021). ”

R1-C2.10: L1227: Cl_y, Br_y, and I_y are usually defined as total inorganic halogens including XO_x as far as I know. Here it represents the reservoir only excluding XO_x, which might be confusing. Maybe better to choose a slightly different abbreviation for the reservoir, and make clear that the striped and empty bars give total inorganic chlorine/bromine/iodine.

ANSWER: We understand the reviewer is pointing at original lines L1237, and greatly appreciate him/her for catching up this important mistake. Throughout the whole manuscript, we made sure to highlight in the discussions the importance of distinguishing between the reactive XO_x and reservoir fractions of total inorganic halogens (X_y) to properly evaluate the halogen impact on other tropospheric components. We apologize for the unintentional error remaining in the original caption of Figure 11. We prefer not to introduce an additional acronym for reservoir species, and consequently we have modified the manuscript for current Figure 12 as follows:

L1111-1115: *“Figure 12: Seasonal distribution of total SLH abundance during present-day (2015-2020) for a-c) Northern Hemisphere (NH = 20°N - 90°N) extratropics; d-f) Tropical (20°N – 20°S) mean; and g-i) Southern Hemisphere (SH = 20°S - 90°S) extratropics. Each coloured stacked bar distinguishes the contribution from organic VSL_x (stippled bars), reactive (XO_x, striped bars) and reservoir (empty bars) halogens (i.e., the sum of reactive XO_x plus reservoir species representing total inorganic halogens, X_y) for the CC-SLH [2×2-ndg] and CC-SLH [2×2-hst] experiments.”*

R1-C2.11: Table 9: This table provides really interesting information. Considering the following question might make it even more valuable:

- How is stratospheric O₃ defined, i.e. everything above the chemical tropopause? The footnote \$ is missing.
- Tropospheric mean OH could better be expressed as number concentration (in molecules cm⁻³) to facilitate comparison with other published estimates (e.g. multi-model comparison in Lee et al., 2021; <https://doi.org/10.1073/pnas.2115204118>).
- How large is the interannual standard deviation compared to the difference?
- How do the results compare to SLH effects in other models?

ANSWER: We note that the tropospheric OH burden has been expressed in total mass (Mg or Tg) throughout this work to facilitate direct comparison with previous studies using CESM1 (e.g., Saiz-Lopez et al., 2023; Bossolasco et al., 2025) as well as with the OH production budgets reported in Lelieveld et al. (2002, 2016). Nevertheless, we accept the reviewer suggestion and have now included an additional column to Table 9 with global mean OH abundance expressed in molecules cm⁻³ to facilitate comparison with other studies. We have added the following sentence to the revised text:

L1448-1451: *“The tropospheric OH burden of 222.8–226.2 Mg obtained for nudged CC-SLH [2×2-ndg] and free-running CC-SLH [2×2-hst] experiments corresponds to a global mean tropospheric OH concentration of approximately 1.23–1.27×10⁶ molec. cm⁻³, consistent with previous model results neglecting SLH chemistry (Lelieveld et al., 2016) and in the higher edge of the range reported in the literature for the 21st century (11.7±1.0×10⁵ molec. cm⁻³; (Voulgarakis et al., 2013).”*

In Addition, we have now introduced a dedicated discussion section where we explicitly compare our CESM2-SLH results with the literature. The following lines have therefore been added in Section 5:

L1439-1447: “Given the general higher ozone abundance in **base CESM2 CAM6-Chem** compared to CESM1 CAM4-Chem (Emmons et al., 2020, **see Fig. 2**) the absolute SLH-driven tropospheric and surface ozone reductions are larger for current CESM2-SLH simulations compared to previous studies. However, we highlight that the corresponding percentage decreases of global mean ozone at the surface (~22%) and the troposphere (~17%) **obtained in CESM2-SLH** are in very good agreement with previous CESM1 studies, ranging from –13% to –24% at the surface and –16% to –20% in the troposphere (Badia et al., 2021; Barrera et al., 2023; Iglesias-Suarez et al., 2020; Saiz-Lopez et al., 2023). Nevertheless, we note that some discrepancies remain due to **several SLH developments not yet implemented in CESM2-SLH (such as continental inorganic halogens, emissions from dust and/or polar halogens, see Section 3.3), as well as due to the continuous improvements implemented in the base CESM development branch, which directly or indirectly impact on SLH abundance and chemistry.**”

Finally, we have updated the table footnote as follows:

L1160: “^sThe stratospheric ozone column is expressed in Dobson Units (DU) and computed for $O_3 \geq 150$ ppbv.”

R1-C2.12: Figure 16: Panels c) and f) of Figure 16 suggests that the total ozone loss is the same in FCnudged_SLH and FCnudged_NOH, and that only the contribution of different families changes. Could you confirm that this is indeed the case? It might be worth mentioning.

ANSWER: The reviewer is correct, the gross tropospheric chemical loss is equivalent regardless of considering (4800–4850 Tg yr⁻¹) or neglecting (4870–4960 Tg yr⁻¹) SLH in the model, which represents a small change of only 1.5–2.3 %. Similarly, the net photochemical production differs only by 1.5–1.7 % between equivalent model sensitivities (see Table 10). However, we highlight that the global tropospheric ozone burden is reduced by 16-17% between the SLH and NOH simulations, in agreement with previous estimates (Barrera et al., 2023) . This is qualitatively shown in original Figure 16c,f (current Figure 17c,f) as pointed out by the reviewer, as well as in the quantitative values compiled in Table 10.

We accept the reviewer suggestion and have modified the manuscript to explicitly clarify this behavior as follows:

L1247-1251: “Indeed, for the **CC-SLH [2×2-ndg] experiment** (Fig. 17b), the total contribution of halogens increases significantly and represents between 10% and 30% globally (Fig. 17c), with a variable vertical profile that is dominated by IO_x cycles in the troposphere, and an increasing contribution mostly from BrO_x close to the tropopause. **This increase in halogen driven ozone loss is compensated** by a proportional decrease in the **OddO_x_{Loss}** destruction by HO_x and O_x in comparison with **CC-NOH [2×2-ndg]** (negative change in Fig. 17c,f).”

L1267-1271: “These values show excellent agreement with previously reported halogen influences obtained with CESM1 (Badia et al., 2021). Table 10 also shows **that the tropospheric ozone photochemical production and the gross chemical loss remains almost unaltered (with minor changes below ~3%) regardless of the consideration or not of SLH sources and chemistry, consistent with previous studies (Barrera et al., 2023).**”

R1-C2.13: L1618: By how much does SLH increase computing time?

ANSWER: The inclusion of SLH chemistry in CESM2 results in an increment of the computational cost of ~20-25%, regardless of the model configuration and resolution considered. Following the suggestion of both

reviewers about moving all technical and user's guide comments into a dedicated section, in the revised manuscript we have now include this information within the Appendix.

Technical corrections

R1-C3.1: L52: "de" --> "the"

ANSWER: Thanks. Done.

R1-C3.2: L53: "SLH have been shown"? SLH refers to short-lived halogens, or "SLH chemistry has been shown"?

ANSWER: Thanks. Done.

R1-C3.3: L57: The acronym VSL refers to "very short-lived" only. Change to "VSL halocarbons"? Or define new acronym VLSL (for very short-lived species); same in L743

ANSWER: Thanks. Note that in the original manuscript we used the acronym VSLs (with lower capital s) when referring to more than one species, and VSL for singular and/or when pointing at a specific species (i.e., VSL chlorocarbons). We have revised the manuscript for a consistent usage of the singular and plural terms.

R1-C3.4: Table 1, Table 2 and Table 5: Rename "Molecular weight" to something more meaningful, e.g. "Molecular weight dependent factor ". And provide units for it.

ANSWER: Thanks for the careful reading. We followed the reviewer suggestion and replaced it by the new variable MW_{factor} (molecular weight factor) throughout all tables and text. We made an explicit clarification in each Table footnote as follows:

L320-321, L440-442, 491-492: *"^{\$} Values for the **molecular weight factor** (MW_{factor}) correspond to the following term from Eq. (3), $\left(\sqrt{\frac{8 \times R}{\pi \times MW_{\text{ox}}}}\right)$, where temperature has been excluded from the square-root and is later multiplied by an independent $(\sqrt{T})$ term."*

R1-C3.5: L606: "respectively" used twice

ANSWER: Thanks. Note we have completely rephrased the sentence to properly use the terms 'fork' and 'tag'.

R1-C3.6: Caption of Figure 3 (L852): "compset_non" --> "compset_noh"

R1-C3.8: Figure 5: "FCNudged_NON" --> "FCNudged_NOH". Might need to check for a consistent naming of the setup without SLH in general.

ANSWER: Thanks for the careful reading. To avoid confusion about the different configurations and terms in CESM2-SLH, in the revised manuscript we have split the original Table 6 into two independent tables: the first one introducing the new "compsets" available, and a second table describing the different "experiments" performed. Therefore, in all figures, tables and throughout the text, we now point at the specific experiment

name (e.g., CC-SLH [2×2-ndg] instead of *FCnudged_slh* and/or CC-SLH [2×2-ndg] instead of *compset_noh*) when presenting and discussing the results.

R1-C3.7: Table 8: How is present-day defined, i.e. average over which years is shown? Please provide the information in the caption. Units are missing for Long-lived halogens

R1-C3.15: Table 12: Please include in the table caption which years were analysed.

ANSWER: We thank the reviewer for the careful reading. Present-day was already “*defined as the mean 2015–2020 period*” in the original manuscript (see **L642**). We have now included units for Long-Lived Halogens, which are the same as for VSLS. To make this clear, we have modified the title of Table 8 and original Table 12 (split into current Tables S4 and S5) as follows:

L803: “*Table 8: Quantitative values for global annual SLH burdens during present-day (2015–2020) conditions.*”

Supplementary Material: “*Table S4: Global mean surface and tropospheric SLH loading for different experiments during present-day (2015–2020).*”

Supplementary Material: “*Table S5: Global mean surface and tropospheric changes in O₃, OH, and NO₂ abundance due to SLH during present-day (2015–2020).*”

R1-C3.9: Figure 7: “BRCL” --> “BrCl” typo in legend of panel b

ANSWER: Thanks. Corrected.

R1-C3.10: l. 1061: Figure 10 could be referenced here as it shows the geographical distribution.

ANSWER: Thanks for the careful reading and for the suggestion. Given that original Figure 10 (current Figure 11) show surface mixing ratios instead of oceanic fluxes, we have clarified the sentence as follows:

L1033-1036: “*As summarized in Table 8, SSA-dehalogenation is the dominant source of bromine and the second largest source for chlorine. However, Fig. 10c highlights that these sources are primarily confined to the **lower troposphere where most of the washout takes place**. For chlorine, the contribution of the acid-displacement HCl release dominates, increasing surface Cl_y particularly in the NH mid-latitudes and coastal locations (Fig. 11d).*”

R1-C3.11: L1097: Figure 8 shows profiles of the absolute VMR of the species I think. The “change in inorganic halogen product gases from the Earth’s surface to the stratosphere” could be misunderstood as “difference between the Earth’s surface and the stratosphere”.

ANSWER: Thanks for the suggestion. We have rephrased the sentence as follows:

L854-857: “*Figure 4 shows the tropical mean (20° N – 20° S) vertical profiles of all VSL source gases (SG_x) along with the associated increment of inorganic product gases (PG_x). Since the simulations neglecting SLH (CC-NOH) have non zero Cl_y and Br_y abundance, the PG_{Cl} and PG_{Br} vertical profiles were computed as ΔCl_y and ΔBr_y, respectively (i.e., the difference between the CC-SLH and CC-NOH experiments).*”

R1-C3.12: L228: Do you mean “summer” instead of “winter” here?

ANSWER: Thanks for the careful reading and for catching up the unintended error. We have corrected in the revised manuscript.

R1-C3.13: Table 10: Units missing for PO₃ – LO₃, drydep, and O₃ STE

ANSWER: We thank the reviewer for the careful reading. We have now including the missing units for ozone photochemical production, chemical loss, and dry deposition terms (which are Tg yr⁻¹) in Table 10.

R1-C3.14: L1422: The estimates in the text (505 and 522 Tg yr⁻¹) refer to IO_x loss according to Table 10.

ANSWER: Thanks for the careful reading and for catching up the unintended error. To avoid confusion, we now point at the total halogen loss followed by the percentage contribution of iodine, bromine and chlorine. We have rephrased the complete sentence as follows:

L1265-1267: “Globally, *the halogen driven tropospheric photochemical ozone destruction reaches 659.4 Tg yr⁻¹ for CC-SLH [2×2-ndg] and 662 Tg yr⁻¹ for CC-SLH [2×2-hst], dominated by IO_x-induced OddOx_{Loss} (accounting for 76–79%) and with minor contributions for bromine (16-18%) and chlorine (5%).*”

R1-C3.16: L1618: “chemitry” --> “chemistry”

ANSWER: Thanks. Done.

Reviewer #2:

General Comments:

Overall quality of the preprint

R2-C1.0: This preprint is clearly targeted towards the Community Earth System Model (CESM) community and presents the implementation of short-lived halogen (SLH) sources and very short-lived (VSL) halocarbons into CESM version 2.2.0. The authors demonstrate substantial expertise in this subject area.

GENERAL ANSWER: We thank the reviewer for the recognition in the field and for the very useful and constructive suggestions that helped to improve the quality of our work.

Given the large number of individual comments, and to avoid excessive repetition, in this section we present a short and general answer to the main comments, and provide in next section below detailed and specific responses to each of the independent questions/comments.

The manuscript is organized as follows:

- Section 2 (2.1–2.4) provides a detailed account of historical developments related to SLH sources and VSL halocarbons, combined with model implementation details and scientific background.
- Section 2.5 describes shared community model configurations and setups for SLH and VSL experiments.
- Section 3 presents results from CESM-SLH experiments, comparing them to previous CESM1 work and observational benchmarks. These include global distributions, vertical species partitioning, and atmospheric burdens.
- A short summary concludes the manuscript.

R2-C1.1: This topic and the results presented are highly relevant to the SLH/VSL research community. However, the current presentation limits accessibility and clarity. Specifically, the manuscript is:

- insufficiently structured,
- repetitive, with numerous filler words and phrases (e.g. "we highlight/note that", "Here, we note/should be noted")
- grammatically inconsistent (e.g., plural nouns combined with singular verbs),
- using lab-specific jargon or unexplained community terms, and
- affected by persistent typesetting issues, particularly with citations (e.g., parentheses instead of in-text citation).

GENERAL ANSWER: We thank the reviewer for highlighting the relevance of our developments and results, and appreciate the very careful reading and constructive comments provided. As detailed below, we have re-structured the manuscript (including a dedicated Discussion section and a completely new Appendix), removed the numerous filler phrases, checked for grammar consistency, avoid using jargon style and formatted the whole text and references following GMD guidelines.

Equations, tables, and figures:

- Several tables and headings do not follow the journal’s formatting standards
- Formulae are incorrectly typeset (see some examples in Section 3).
- Figures are sometimes low-resolution, with frame artifacts and inconsistent labeling
- Colormaps draw disproportionate attention to light blue and yellow regions.
- Empty figure panels are included.
- Exponential expressions are written as E-02 instead of 10^{-2} .

R2-C1.2: Although submitted to *GMD*, the manuscript does not yet meet expectations for a clear and comprehensive model description. For instance, the term compset is never explained for readers outside the CESM community.

GENERAL ANSWER: We appreciate the careful reading and detailed comments to follow the journal guidelines. The revised manuscript has now been checked for format consistency, particularly for table heading and figure captions. We provide high-quality figures, all of which has been re-generated using colorblind-friendly pallets with sequential scales. All equations, physical and chemical variables, range of values and exponential expressions have been re-formatted. We appreciate the reviewer suggestion to explain the term “*compset*” for readers outside the CESM community. Therefore, we have removed it usage from the abstract and introduction, and only defined it within the Model Description section as follows:

L515-518: “*The implementation of SLH sources and chemistry described in this work was performed over the base FCnudedged and FCHIST atmospheric chemistry compsets (<https://docs.cesm.ucar.edu/models/cesm2/config/2.2/compsets.html>) In CESM terminology, compset is an acronym for ‘component setup’ and refers to specific model configuration of the Earth System Model, specifying which components are used and how they are coupled (Danabasoglu et al., 2020).*”

Suggested Improvements

R2-C1.3: To enhance clarity and accessibility, the following structural changes are recommended:

- Split and reorganize Sections 2 and 3.
- Following common conventions, Section 4 should be titled “Summary and Conclusions.”
- Introduce an additional section dedicated to discussion, consolidating currently fragmented discussion points.

GENERAL ANSWER: We thank the reviewer for his/her constructive suggestions. Therefore, we implemented several structural changes to improve the manuscript clarity and accessibility. In particular, we split and reorganize original Section 2 into two independent sections (new Sections 2 and 3), the results are now shown in Section 4, we included a new Section 5 dedicated to Discussions and entitled the final Section 6 as “Summary and Conclusions”. See also the detailed response to **R2-C1.4** below.

More detailed structural advise:

- The new *Section 2* (Model description) should begin with a concise, general overview of *CESM2* and its relevant components. For example, explain:
 - CESM structure (refer to Supplement Chart S1),
 - definition of specified dynamics (SD),
 - what a compset is,

- differences between compsets (e.g., SD vs. free-running).
- Only after this overview should CESM2 be contrasted with CESM1.
- Follow with an overview of CESM2-SLH developments and available compsets (currently Section 2.5). Detailed implementation should then be presented in the next section 3.
- Section 2.1 (historical developments), while interesting, is unnecessary for this manuscript and could be removed, as this is not intended as a review paper.
- The new *Section 3* (Model implementation of SLH and VSL) should combine Sections 2.2–2.4, focusing on implemented schemes, mechanisms, and chemistry.
- Consider including Figure S1 here, as it illustrates key differences in sea salt properties between CESM1 and CESM2.
- Avoid historical context and developments in this section. It is enough to briefly introduce the Ordoñez inventory.
- Move “user guide” like paragraphs into an Appendix section (User guide) or rephrase them for a more general audience.
- The new *Section 4* (Results) could be subdivided into:
 - 4.1 Validation (currently Sections 3.2.2 and 3.3.1),
 - 4.2 Comparison to CESM1 results,
 - 4.3 Sensitivity studies with CESM2-SLH and additional single-experiment studies.
- First describe sensitivity experiments and summarize them in tabular form (in parts Tab. 6).
- Consolidate all discussion into a dedicated *Section 5* (Discussion).
- Rename the final section as *Section 6* (Summary and Conclusions).

R2-C1.4: Implementing these changes will significantly improve readability, accessibility, and compliance with journal standards.

GENERAL ANSWER: While we accepted the reviewer suggestion to split the original Section 2 of the manuscript (Model Description), the intention of this manuscript is not to present an overview of CESM2 (which is comprehensively described in Danabasoglu et al. (2020)), but instead, to only point at the specific developments and configurations that are of importance to SLH. Therefore, the new structure of the manuscript includes first a detailed description of independent SLH and VSL developments (“**2. Implementation of SLH chemistry in CESM**”) followed by an overview of the new model configurations available in the current release (“**3. CESM2-SLH compsets and experiments**”). Here, we have split original Table 6 into two different tables: one describing the new compsets available and another one summarizing the main experiments performed in this work. Note that we only turn-on “track-changes” in the revised version for the subsection titles, but not the whole block of paragraphs that were moved up/down.

We have also rearranged Section 4 (Results) to focus on the distribution and evolution of SLH for the main model configurations (Section 4.2), including as the first subsection the evaluation of SLH abundance in comparison with observations (4.2.1); followed by the influence of SLH over the main atmospheric components (Section 4.3), also initialized by the model validation (4.3.1). In doing so, we split and reorganized all “technical” blocks into the Appendix, while the comprehensive analysis and intercomparison between the different experiments and previous studies is all condensed in the new Section 5 (Discussion).

Finally, we refrain from moving Figure S1 from the supplement to Section 2 because the implementation of sea-salt aerosol (SSA) in CESM2 is not related to the SLH developments and have been performed by other groups. Similarly, we rather keep the original Section 2.1 (Historical Developments) with a double porpoise: *i)* to highlight that individual sources, sinks and chemistry for each of the independent halogen families were implemented and validated in the model throughout a long-lasting period and following a coordinated work

among several groups; and ii) to provide for the non-expert in SLH chemistry, particularly those outside the CESM community, with a comprehensive review of the relevance of considering SLH chemistry in chemistry-climate studies. Note original Fig. S1 has now been shifted to Fig. S2.

R2-C1.5: The title may be expanded along the lines of “– implementations and benchmark results”

ANSWER: We appreciate the reviewer suggestion but prefer to keep the original title of the paper.

Specific comments:

Individual scientific questions/issues

GENERAL ANSWER: We thank the reviewer for the detailed analysis and for rising constructive questions and suggestions, whose implementation had substantially improve the manuscript quality. Below we provide individual answers to all individual issues belonging to **Section 2**. To avoid repetition, in some occasions we have grouped a few independent comments into a single response and/or point at the responses to the related technical comments included below in **Section 3**.

Section 2

R2-C2.1: L259–263: “*Despite [...]*” It seems there is no variable in Eq. (1) that connects emissions to the long- or shortwave incoming solar irradiance of the atmospheric basemodel which would vary with atmospheric state, season, time of the day, and orbit. Or is this hidden behind our “diurnal emission profiles”? From the text, it is not clear how these “profiles” are implemented.

R2-C2.2: L261–263: “*Emissions follow either [...]*” What does this mean? Can the user choose between the two?

ANSWER: Eq. (1) was used to generate monthly-mean VSLs emission files, computing each individual halocarbon flux strength at all latitudes and longitudes offline. Once those emission files are ready, they are read during CESM2-SLH execution, and a temporal profile is applied online to represent the hourly variability. Most species follow the Gaussian profile, with exception of one VSL iodocarbon, that present a top-hat shape. The implementation of all these developments has been described in the Ordóñez et al. (2012) inventory, although the hourly profile is not shown in the original reference. Therefore, we have included an additional figure to the Supplementary Material and rephrased the referred sentence as follows:

L231-235: “*Once the monthly-mean prescribed fields are read, hourly dependent profiles are applied to the flux strength of all halocarbons to represent the photosynthetic dependence on radiation intensity. Therefore, VSLs emissions follow a Gaussian diurnal profile with peak emissions at local solar noon (Ordóñez et al., 2012), with exception of CH₂I₂ that follows a top-hat shape with a uniform distribution during the day and no-null emissions at night (see Fig. S1).*”

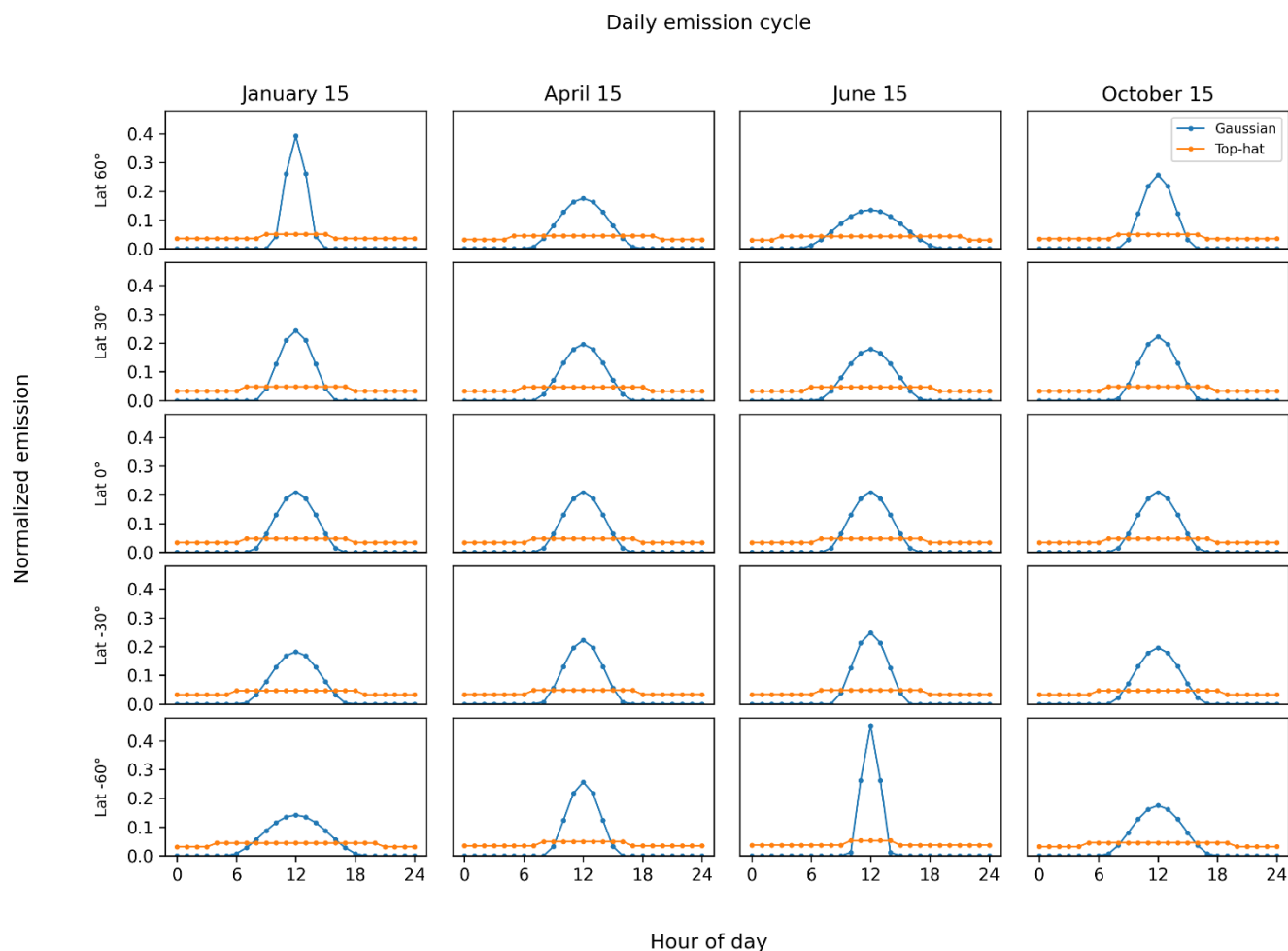


Figure S1: Hourly diurnal variation applied to all oceanic VSL halocarbon emissions considered in the in Ordóñez et al. (2012) inventory. Either the Gaussian or the top-hat temporal profiles are multiplied by the monthly offline emission strength interpolated to each day of the year, and considers the natural daily solar cycle at each specific latitude and longitude.

R2-C2.3: L263–264: “Given that no long-term trend has been established [...]” Has a trend been observed or not? Please include a recent reference (post 2007 and/or 2023) that could support your statement.

ANSWER: We have modified the sentence and provide a citation as follows:

L235-236: “Given that no **global** long-term trend has been **observed or** established (Tinel et al., 2023; WMO, 2022), the emission strength of oceanic halocarbons is assumed to follow a constant climatology.”

R2-C2.4: L264–268: This paragraph is an example of “user guide” like paragraphs. As indicated above, please collect this and similar paragraphs in a dedicated Appendix section. I will not point out each of them individually.

R2-C2.6: L311: “routine [...] iodine_emissions.F90 [...]” Here you refer to a Fortran (module) file as routine. Please be more precise and distinguish between subroutine and module file. What is the actual name of the subroutine? Please also refer my rephrasing suggestion in Section 3.

ANSWER: Following **R1** and **R2** comments, we have reduced the main text length and included a new Appendix with technical details and user-defined configurations. Regarding *iodine_emissions.F90* file, the

reviewer has correctly identified it is a Fortran module. The corresponding subroutine is called *iodine_emissions_set* (#). Note the whole block has now been moved to the Appendix. See also the related responses to **R2.C4.13-15** below.

Please refer to the revised manuscript for the complete Appendix.

R2-C2.5: L286–287: *Based on these works, [...] With respect to the preceding sentence, it is unclear whether this imposed trend is part of the inventories or an alternation in present work. If latter is the case this imposed trend needs more explanation and references. If former is the case this sentence may be dropped as it doesn't provide much additional information.*

ANSWER: Thanks for the suggestion, but we rather keep the sentence pointing at the trend of chlorinated VSLS emissions with the intention to highlight the different behavior with respect to oceanic VSL bromocarbons (cyclical climatology). To avoid being interpreted as a novel alternation of our work, we have rephrased the referred sentence as follows (see response to **R2.C4.9-12** below as the complete paragraph has been changed):

L247-250: *“For anthropogenic VSL sources of chlorocarbons, we follow the Hossaini et al. (2019) and Claxton et al. (2020) emission inventories, which include time-dependent emission of two major contributors to the organic chlorine load (CH_2Cl_2 and C_2Cl_4) as well as surface LBCs for two other compounds (CHCl_3 and $\text{C}_2\text{H}_4\text{Cl}_2$). Based on these works, an increasing trend from 2000 to 2020 is applied to the CESM2-SLH emission and LBC files for the recent past (see Appendix).”*

R2-C2.7: L325–326: *“Note that the fitting [...]”* This sentence is not well written and should be rephrased along the lines: → ‘Equation (2a,b) are only valid for wind speeds $\omega \geq 3 \text{ m s}^{-1}$ (Inamdar et al., 2020). Below this threshold, anonymously high iodine fluxes are predicted

R2-C2.8: L326–328: *“Therefore, a wind speed mask imposing [...] periods.”* This paragraph is not entirely clear from an implementation point of view in the code. Rephrase: → To avoid this overprediction, $\omega = 3 \text{ m s}^{-1}$ is assumed for all grid cells where $\omega < 3 \text{ m s}^{-1}$.

ANSWER: We understand the reviewer meant anomalously instead of anonymously. We have modified the manuscript as follows:

L277-279: *“Equations (2a,b) are only valid for wind speeds $w \geq 3 \text{ m s}^{-1}$. Below this threshold, anomalously high iodine fluxes are obtained. To avoid this overprediction, $w = 3 \text{ m s}^{-1}$ is assumed for all grid cells where $w < 3 \text{ m s}^{-1}$. Not imposing this wind speed mask result in an overestimation of iodine fluxes from the ocean during calm periods (Inamdar et al., 2020).”*

R2-C2.9: L350: *“[...] above a pressure threshold [...]”* Does this mean below or above 300 hPa?

ANSWER: To avoid confusion, we have replaced the text by *“below a pressure threshold ($P < 300 \text{ hPa}$).”*

R2-C2.10: L367–373: *“Given, the large changes [...] (see Section 3.2.2).”* Be more explicit. How large are the uncertainties and how much have you adjusted the reaction rates compared to the most recent JPL catalog?

ANSWER: We apologize for the unspecific description of the changes in accommodation coefficients required to replicate previous estimation of the SSA-dehalogenation flux. We have now modified to explicitly mention the scaling factors implemented in the latest version of CESM2-SLH as follows:

L311-317: *“Given the large **increase** in SSA abundance and **vertical** distribution between CESM1 and CESM2 (see Fig. S2), particularly over the boundary layer where the latter show up to 10 times larger values than the former, the accommodation coefficients (γ_{ox}) for halogenated reservoirs compiled in Table 4 of the Supplementary Material of Ordóñez et al. (2012) was reduced by a factor of 3 for chlorine and iodine and by a factor of 8 for bromine (see Table 1). This scaling factors are within the reported uncertainty and variability of γ^{ox} for the different halogen families and aerosol compositions (Burkholder et al., 2020) and allowed to reproduce previously estimated SSA-dehalogenation fluxes and burdens (see Section 4.2).”*

R2-C2.11: L379–382: *“Careful consideration [...] budget and burden.”* This content of this paragraph is of more general nature and should be formulated that way. → ‘The representation of aerosols in general circulation models, their resulting distribution, abundance, and properties play a key role for prediction of SLH production via SSA dehalogenation in the troposphere’.

R2-C2.12: L384–385: *“[...] the vertical extent of sea-salt in the free-troposphere was largely reduced [...]”* Be more explicit. What does this mean? How was this achieved?

R2-C2.13: L478–482: *“CESM-SLH uses the Modal Aerosol Model with 4 Modes [...]”* This has been implicitly mentioned earlier in that section. Either include this, when you first mention the aerosol model difference between CAM4-chem and CAM6, or in the general CESM model description section as proposed.

ANSWER: We have implemented all of them in the revised version and rephrased the complete paragraph as follows (see also the related response to **R2-C6.23** and **R2-C6.32-33** below):

L534-545: *“The representation of aerosols in general circulation models, including CESM, plays a key role for predicting halogen production via SSA-dehalogenation, which in turn impacts on the SLH tropospheric budget and burden. The major difference between CESM1 (CAM4-Chem) and CESM2 (CAM6-Chem) is that the former uses a continuous bulk representation of aerosols with four size bins, whereas the latter uses a modal representation with three bins (Danabasoglu et al., 2020). Another relevant difference is that MAM4 is the default dust scheme in CESM2 (Liu et al., 2016). Consequently, the vertical extent of SSA in the free-troposphere was largely reduced to improve the model overestimation obtained in previous studies (Lamarque et al., 2012; Tilmes et al., 2023). Therefore, to allow adjusting the online efficiency of the SSA-dehalogenation flux and avoid unrealistic changes in the atmospheric burden of tropospheric halogens due to parallel SSA developments, the released CESM2-SLH configuration now includes a group of SLH scaling factors (&slh_nl,) within the user-defined namelist (user_nl_cam). These tuneable scaling factors allow individual users to adjust the SSA-dehalogenation source in the different model resolutions and configurations in order to assure a consistent halogen atmospheric loading that are consistent with those shown in this work (see Appendix for further details).”*

In order to define MAM4 on its first mention, we have updated the closing paragraph of current Section 2.2.2 as follows:

L393-401: *“Oxidation of DMS to produce sulphur dioxide (SO₂) by BrO, Cl and IO are also included in the updated SLH chemical scheme. These improvements in sulphur chemistry representation in CESM2 with respect to CESM1, including tropospheric sulphate formation and washout, are a major source of uncertainty for radiative forcing estimations (Ge et al., 2022) and are currently under development. Therefore, the current CESM2-SLH mechanism*

has been developed starting from the benchmark sulphur scheme implemented in TS1.2 (Emmons et al., 2020). Similarly, the Modal Aerosol Model with 4 modes (MAM4) scheme is the default aerosol option in CESM2, which includes updates in the formation and growth of sulphate, black carbon and organic matter, secondary organic aerosols, sea salt, and dust (Liu et al., 2016). Although no direct SLH changes to the MAM4 scheme were implemented, we note that indirect perturbations of reactive halogens on OH abundance can influence the global aerosol burden (Saiz-Lopez et al., 2023)."

R2-C2.14: L528–530: *"Regarding stratospheric heterogeneous [...]"* This sentence seems to refer to the stratospheric heterogeneous chemistry in the context of Polar Stratospheric Clouds (PSCs) that had been previously implemented in mo_strato_rates.F90. But it is unclear what "complemented [...]" mapping" means. The expression "historical implementation" here seems inadequate. This sentence should be rephrased.

R2-C2.15: L536–539: *"In doing so, [...]"* This sentence is unclear and should be rephrased. Do you mean that you used reaction yields measured for chlorine and bromine species for the respective iodine species instead of the actually observed iodine yields rates? What is the reason for this?

ANSWER: We have rephrased the whole paragraph describing stratospheric heterogeneous reactions as follows (see also the related response to **R2-C6.35** below):

L445-455: *"Implementation of stratospheric heterogeneous reactions of inorganic halogen reservoirs arising from the photodecomposition of long-lived halogens in WACCM are important for representing the ozone layer (Marsh et al., 2013). This includes temperature and water-vapour dependent re-activation reactions of chlorinated and brominated reservoirs occurring over stratospheric sulphate (SAD_{sulfc}), ice-crystals (SAD_{ice}) and nitric acid trihydrate (SAD_{NAT}) surfaces (Kinnison et al., 2007), including recent updates of individual stratospheric accommodation coefficients (γ^{strat} ; Solomon et al., 2015). Based on Cuevas et al. (2022), in CESM2-SLH we extended WACCM developments by mapping equivalent heterogeneous reactions in the stratosphere for the minor halogenated reservoirs (mainly HOCl and HOBr), as well as the previously neglected iodine species (see Table 3). Here, we assumed identical accommodation coefficients and reaction yields for iodine than those used for the equivalent chlorinated and brominated reservoirs. This assumption represents a lower-limit of iodine impacts, as previous studies suggest that the reactive efficiencies of iodine species are larger than those for bromine and chlorine (Koenig et al., 2020; Solomon et al., 1994)."*

R2-C2.16: L604–608: Putting on my code developer hat... Considering that the VSL/SLH developments in CESM1 were not directly transferable to CESM2, how sustainable are your developments based on CESM2? First, forking implies that the development may never be merged back into the CESM repository and is in jeopardy to end up on a stale branch (again). Second, taking a look at your github repository shows that the last pull request from the upstream repository has been more than one year ago. Third, since submitting this preprint you have added changes to the branches on your CESM2 and CAMS forks without (as far as I can tell) tagging the versions you have based this preprint on. Therefore, it would not be possible to reproduce your results, although your repository is publicly available (which is actually great!).

ANSWER: First, we apologize for the erroneous usage of the terms fork and tag in the original manuscript. We have now corrected and used them appropriately. Most importantly, we have now created two CESM2-SLH tags: one reverted to the exact commit used to perform all simulations described in the manuscript (*cesm2.2-asdbranch_slh-gmdd* and *cam_cesm2_2_rel_09_slh-gmdd*), and another one where we have updated all SLH default namelists variables in the building scripts of CESM with the final user-defined values described in this work (*cesm2.2-asdbranch_slh_tag* and *cam_cesm2_2_rel_09_slh_tag*). To avoid confusion

for new users, and given that FORTRAN changes were not implemented between tagged versions, we only point at the latest one in the revised manuscript.

Second, we appreciate the reviewer comment and understand his/her concerns about the importance of making sure the described SLH developments do not end up on a stale branch. This was indeed the case when performing the initial pull-request of our SLH commits to NCAR. We were suggested to shift all developments into the latest CAM6_dev branch as this would assure SLH developments to be maintained and updated in future versions of CESM. Here we note that once the final CAM6_dev version is frozen, it will become the core CAM7 atmospheric component of CESM v3. Therefore, we have now ported all SLH developments of CESM2-SLH into CAM6_dev, whose pull request had already been approved by NCAR and made available in the official GitHub repository (tag *cam6_4_153*).

Based on the above, we have rephrased the referred sentence, as well as the Code and Data Availability section as follows:

L528-533: *“CESM2-SLH porting was performed on top of version 2.2.0 of CESM2 (Danabasoglu et al., 2020), particularly over branch *cesm2.2-asdbranch*, which includes CAM6 tagged version *cam_cesm2_2_rel_09*. Based on these versions, we forked and created the new CESM2-SLH and CAM6-SLH branches called *cesm2.2-asdbranch_slh* and *cam_cesm2_2_rel_09_slh*, respectively, which incorporate SLH updates within the main FORTRAN routines as well as modifications to building scripts (e.g., *cime* and *cime_config*) and default namelist variables (see Appendix). These open-access community developments allow to download, clone and build all SLH compsets available (see Code and Data Availability below; Fernandez et al., 2025).”*

L1551-1561: *“The Community Earth System Model (CESM) code is maintained by the NSF National Center for Atmospheric Research (NCAR). The benchmark version of CESM2 (Danabasoglu et al., 2020) is distributed via GitHub (<https://github.com/ESCOMP/CESM>) and available at the NCAR’s official site (<https://www.cesm.ucar.edu/models/cesm2>). The final tag of the CESM2-SLH branch used in this work (*cesm2.2-asdbranch_slh_tag*) can be cloned from GitHub (https://github.com/RafaPedroFernandez/CESM/tree/cesm2.2-asdbranch_slh_tag), which was originally forked from the latest CESM2 tag available in the NCAR repository (<https://github.com/ESCOMP/CESM/tree/cesm2.2-asdbranch>). All SLH code and data supporting this work, including the complete set of building scripts, Fortran routines, individual namelist templates and input files for SLH and NOH compsets, as well as the annual mean output used to generate all Figures and Tables, can be obtained from Mendeley Datasets at <https://doi.org/10.17632/f87hvr25v.1> (Fernandez et al., 2025). All SLH developments described here have also been ported into the latest CAM6 development branch maintained at NCAR (https://github.com/ESCOMP/CAM/tree/cam6_4_153) and will be available in the next release of CESM v3.”*

R2-C2.17: L625–631: “[...] factors are maintained identical to the default FCHIST and FCnudged parent’s configuration [...]” It remains unclear how the two compsets differ (without going through additional resources). I suggest elaborating on the difference between the two in the beginning of the results section

R2-C2.18: L643–645: *“Finally, a fully-coupled [...] setups, both of them at [...] resolution [...] were mapped to their corresponding WACCAM compsets.”* This sentence is grammatically not correct, hard to understand, and needs rephrasing. What do you mean by “mapped”? Do you mean that you have created also compsets for WACCAM based on your BWHIST_slh and FWnudged_slh compsets that use CAM6?

ANSWER: To avoid any confusion about the new SLH configurations available, we have removed the term “parent’s” and rephrased the opening and closing sentences of the paragraph referred in both comments as follows:

L548-565: “Two main CESM2-SLH configurations have been developed based on the available CAM6 base atmospheric chemistry compsets: FCHIST_slh and FCnudged_slh (see Table 6), where the former resolves atmospheric dynamics internally (free-running) and the latter is forced by prescribed meteorological fields (new nudging approach for specified dynamics). Both configurations employ the coarse f19_f19_mg17 horizontal resolution (1.9° latitude × 2.5° longitude, hereafter 2°×2°) and 32 vertical levels (low-top) from the surface to approximately 40 km (~4 hPa)” ... “In addition to the low-top configurations, whole atmosphere (high-top) fully-coupled BWHIST_slh (f19_g16) and FWnudged_slh (f19_f19_mg17) compsets were developed, both considering the coarse 2°×2° resolution and 70 vertical levels as their corresponding base atmospheric WACCM compsets. Table 6 summarizes the names and specifications of the main CESM2-SLH compsets.”

R2-C2.19: L664 and following: “stabilization” Expression is unusual. Usually, this is referred to as ‘equilibrium’.

ANSWER: Chemical species in the atmosphere, particularly SLH concentrations, do not reach chemical equilibrium. In any case, we can talk of a steady state. We have rephrased the sentence and included in the Appendix as follows:

L1586-1587: “The initial condition (ncdata) data has been replaced to ensure background SLH species and atmospheric compounds are stabilized.”

Section 3

R2-C3.1: L766–769: “[...] their photodecomposition constitutes the critical first step in releasing inorganic chlorine and bromine [...]” This is a repetition of scientific background presented in Section 2. Here, and elsewhere in this section, you should focus on the results and not on repeating scientific background.

ANSWER: The original sentence was oriented to connect the distinct influence of unreactive VSL halocarbons with that of reactive halogen species for the non-expert. We have now accepted the reviewer suggestion and removed the referred sentence as well as many others equivalent connections throughout Section 4 (Results). In some occasions, we have shifted down and rephrased as part of the SLH discussion in the new Section 5.

A detailed response to the suggestion of including a dedicated Discussion section is provided above (**R2-C1.3**) while individual answers to specific blocks that have been moved to the new Section 5 are detailed below (**R2-C3.8**, **R2-C3.9**, **R2-C3.12** and **R2-C3.14**).

R2-C3.2: L898: “[...] in agreement with previous estimates [...]” You may cite the numbers from the reference.

ANSWER: Thanks for the suggestion, but we do not further need it as we removed the final half-sentence to shorten the manuscript length.

R2-C3.3: L900–901: “model surface” What “surface” does this refer to? Is it the actual land/water surface or the lowermost model level above the surface?

ANSWER: Thanks. We have rephrase the sentence to explicit mention Lower Boundary Conditions:

L784-786: *“Here, it should be noted that the additional contribution from other anthropogenic VSL chlorocarbons (CHCl_3 and $\text{C}_2\text{H}_4\text{Cl}_2$) are also included as **LBCs** instead of considering offline emissions, reaching a total of ~48 pptv at the **lowest model level above the surface.**”*

R2-C3.4: L904–907: *“This is explained by the large non-linear response of the [...]”* Can you further elaborate on this? Where do the non-linearities in Eq. (3) come from? I understand that there are non-linearities in the involved atmospheric feedback processes, e.g. radiation-temperature-photolysis.

ANSWER: The reviewer is correct that Eq. (3) alone cannot explain the non-linearity of the SSA-dehalogenation. Here, we do not point at radiation-temperature atmospheric feedbacks, but instead to the fact that every time a non-stoichiometric heterogeneous reaction occurs, the system contains more and more Br and Cl in the gas phase, which in the next cycle, will uptake once again bromide and chloride from the SSA. To make this clear, we have modified the referred sentence as follows:

L789-792: *“This is explained by the non-linear **release of Cl_2 , Br_2 and BrCl through the non-stoichiometric heterogeneous reactions described in Table 1, which in turn depend on the parameterized fields of SAD_{SSA} (Fig. S2) and the partitioning shift between different inorganic halogen reservoirs (Fig. 10).**”*

R2-C3.5: L952: *“[...] the IO_x burden change [...] remains close to 10%.”* It is not clear, to what these 10% refer to. In Table 8 no “burden change” is found, rather the tropospheric burdens in Gg are displayed. The percentage difference between the two SLH experiments and the given burden of 3.2 Gg IO_x amounts to 9.4% and 12.5%, respectively. Please clarify and rephrase accordingly.

ANSWER: We were not pointing at the burden change for a specific experiment, but to the spread of burden values between different experiments. To clarify the modeled changes in IO_x and I_y abundance between experiments, we have modified the complete paragraph as follows (see also the response to **R2-C7.20** below):

L831-839: *“In contrast to bromine and chlorine, surface I_y abundance in **CC-SLH [2×2-ndg]** is ~40% higher compared to **CESM1-SLH [SL23]** (2.3 pptv I_y), which is mainly due to the different IO_x/I_y and OH/HO_2 ratios between the different nudging approaches among CESM versions (see Fig. 2i,k). However, free-running experiments (**CC-SLH [2×2-hst]** and **CESM1-SLH [QL22]**) show equivalent surface abundances, particularly for the 2015–2020 period. The larger OH abundance and lower HO_2/OH ratio in CESM2-SLH compared to CESM1 increases the contribution of HOI to the total I_y loading, and therefore, there is a major shift on the partitioning from reactive to reservoir species for iodine (see Table 8). Therefore, the reactive IO_x tropospheric burden in CESM2 (3.5-3.6 Gg I) is only ~10% larger than the 3.2 Gg I estimated value found in Saiz-Lopez et al. (2023). Most notably, the spread in global I_y abundance between free-running and nudging CESM2 compsets is smaller than the spread in CESM1.”*

R2-C3.6: L1023–1038: Given the varying height of the tropopause between 90°N - 90°S (~400 - 90 hPa) and that the ratio between ocean to land is roughly 2 to 3, I don't quite understand this paragraph. First, a global average over pressure/height lumps together polar stratospheric and tropical tropospheric air. Therefore, you absolutely cannot draw any conclusion with respect to the UTLS region. Second, you refer to the Marine Boundary Layer (MBL) but average globally including also grid cells over land, I suppose. Therefore, it seems impossible to draw conclusions with respect to the MBL. You would need to convert your data into vertical coordinates relative to the tropopause height and use a land-sea-mask to include ocean grid cells only to be able to draw robust conclusions.

R2-C3.7: L1051–1082: The concerns raised above also apply to these paragraphs and analyses.

ANSWER: The reviewer has a point that given to the latitudinal variation of the tropopause within the UTLS, as well as to the different representation of the boundary layer over the ocean (MBL) and continental domains (PBL), it is hard to draw robust (i.e., quantitative) conclusions. However, we respectfully disagree with R2 that it is not possible to draw any conclusion based on original Figure 6 (current Figure 9). Indeed, the computation of the global mean average was made on purpose to summarize, in a single figure, the general behavior of organic and inorganic chlorine, bromine and iodine from the Earth's surface to the stratosphere. As conceived, we believe current Fig. 9 summarizes the main similarities and differences between halogen families.

We appreciate the careful reading and have rephrased the opening paragraph to avoid pointing at the UTLS and MBL explicitly. In addition, the revised manuscript now includes explicit clarification of the different regions and domains that are considered when computing the global mean,

L994-997: *"We note that these global and annual mean averages lumped together continental and oceanic domains within the boundary layer, as well as tropospheric and stratospheric parcels due to the latitudinal variation of the tropopause (see dashed horizontal lines in Fig. 9). However, three main differences characterizing the vertical distribution for each halogen family are distinguished."*

and remove on the forthcoming paragraphs any reference to the UTLS and MBL, as for example:

L1033-1039: *"As summarized in Table 8, SSA-dehalogenation is the dominant source of bromine and the second largest source for chlorine. However, Figure 7c highlights that these sources are primarily confined to the **lower troposphere**. For chlorine, the contribution of the acid-displacement HCl release dominates, **increasing surface Cl_y** particularly in the NH mid-latitudes and coastal locations (**Fig. 11d**). The photochemical degradation of both VSL_{Cl} and VSL_{Br} dominates the release of Br and Cl atoms throughout the **free troposphere**, although the biogenic ocean flux is the sole source of bromine, while for the case of chlorine the contribution from anthropogenic VSL_{Cl} is at least two orders of magnitude larger than the natural oceanic source."*

Please refer to the related responses to **R2-C2.9** above as well as to **R2-C3.11** and **R2-C6.8** below.

R2-C3.8: L1132–1139: *"Please note that due to [...]"* This paragraph belongs to a discussion section.

ANSWER: The original sentence was oriented to provide insights about the similarities and differences of SGI and PGI for chlorine w.r.t. bromine, and to support our results with the available literature. We agree with the reviewer that the referred sentence better fit in a dedicated Discussion section. Please also refer to the responses to **R2-C1.3** and **R2-C3.1** above.

R2-C3.9: L1116–1165: *"Here we recall [...]"* Beside this odd expression, this paragraph belongs into a discussion section.

ANSWER: The original sentence was oriented to provide insights about the uncertainties and implications associated with the low model bias for SGI_{Cl} w.r.t. the dominant role played by CFCs and HCFCs. We agree with the reviewer that the referred sentence better fit in a dedicated Discussion section. Please also refer to the responses to **R2-C1.3** and **R2-C3.1** above, as well as **R2-C3.10**, **R2-C3.14** and **R2-C7.36-37** below.

R2-C3.10: L1165–1167: *"[...] should not represent a significant bias in our model implications on other atmospheric components."* Quite unclear. Do you mean that atmospheric (stratospheric) chemistry would not be affected (or only to a small degree) by the underestimation of SGI_I ?

ANSWER: We are explicitly pointing at the minor contribution of SGI_{Cl} to total stratospheric chlorine. Based on the reviewer suggestion of including a dedicated Discussion section (**R2-C1.3** and **R2-C3.1**), we have moved the sentence to the new Section 5 and rephrased it as follows (see also the response to **R2-C7.36-37** below):

L1425-1427: *“Given that chlorine influence on stratospheric ozone is dominated by long-lived CFCs and HCFCs, the current model underestimation of SGI_{Cl} does not introduce a significant bias in the total stratospheric chlorine budget and, consequently, does not substantially affect chlorine-driven ozone destruction.”*

R2-C3.11: L1177–1182: *“[...] over the boundary layer (i.e., mean modelled value from the surface up to ~ 850 hPa).”* This should be rephrased, as the boundary layer is highly variable. → ‘average over model levels $P \geq 850$ hPa and the time period 2015 – 2020’

ANSWER: The reviewer is correct, thanks for the suggestion. We have rephrased the sentence as follows (see also response to **R2-C7.21** regarding pointing at individual figure’s panels below):

L1058-1060: *“Figure 11 shows the spatial distribution of carbon-bonded VSLs (Fig. 11a-c), total inorganic halogens (Fig. 11d-f) and the most relevant reactive fraction (Fig. 11g-i) of each halogen family averaged over model levels with $P \geq 850$ hPa and for the time period 2015–2020.”*

R2-C3.12: L1278–1284: *“Given [...] in the troposphere [...].”* This paragraph could be placed in a discussion section.

ANSWER: The original sentence was oriented to provide insights about the equivalent although not exactly identical absolute and percentage changes in ozone abundance in CESM2-SLH compared to CESM1. We agree with the reviewer that the referred sentence better fit in a dedicated Discussion section. Please also refer to the responses to **R2-C1.3** and **R2-C3.1** above.

R2-C3.13: L1434–1438: *“the old nudging over model levels [...] most recent nudging approach [...] the chemical influence of SLH on tropospheric and stratospheric transport are of equivalent magnitude.”* The first part is basically a repetition from the model description section. Albeit, the formulations differ. Please rephrase in more general terms. Regarding the second part, you have previously discussed in this preprint, that dynamics are important for the amount of VSL and SLH in the troposphere and stratosphere and therefore for the ozone depletion potential and ozone abundance. Please clarify, how the model results presented in this section could indicate an impact of SLH chemistry on atmospheric transport, especially in a SD or nudged setup where dynamics are prescribed.

ANSWER: We agree with the reviewer that the first part of the sentence is a repetition of the model description section. Regarding the second part, we believe that including a comprehensive analysis and discussion of the role played by STE on the transport of VSLs and SLH in this section would excessively enlarge an already long paper. Therefore, we decided to leave this detailed discussion for future studies and rephrased the sentence as follows (see also the related response to **R2-C2.18** above):

L1278-1280: *“Differences between model versions are attributed to dynamical changes in the representation of air subsidence and downward ozone transport across the tropopause between the old and new nudging approaches (Davis et al., 2022) and will be evaluated in future studies.”*

R2-C3.14: L1517–1550: Large parts of this paragraph are written as if they were meant for a user guide which should clearly not be the purpose of a scientific paper. Please rephrase with a more general modeling community audience in mind

ANSWER: We agree with R2 that some parts of the referred block belong to the new Appendix. However, we believe that the final couple of sentences belongs to the new Discussion section. Therefore, we have substantially reduced the length of the original Section 3.4 almost by half and adapted the remaining text to other sections of the manuscript. We added the following sentence to the revised manuscript (see also response to **R2-C1.4**, **R2-C2.6** and **R2-C3.1** above):

L1358-1359: *“A brief description of the main routines and user-defined namelist required to setup and modify different model experiments within CESM2-SLH is included in the Appendix.”*

Section 4

R2-C4.1: For your Summary and conclusions section, it would also be important to mention the necessary scaling of emissions depending on compset and model resolution. Potentially, the scaling factors for the nudged case will depend on the nudging datasets (e.g. ERA5, MERRA, CRUJRA, ...) as well. As part of this section, but also in the model development section, you should clearly state, how these scaling factors should be derived

ANSWER: We thank the reviewer for the suggestion, which is an important consideration for studies using other configurations. However, we believe the suggested clarification belongs to the new subsection 5.3 within *“Discussion”* instead of the final Section 6 (*“Summary and Conclusions”*). Therefore, we added the following sentence to the revised manuscript:

L1489-1494: *“. For the particular case of the FCnudged_SLH and FWnudged_SLH compsets, the scaling factors compiled in the Appendix for VSL emissions, SSA-dehalogenation and washout following FRA were adjusted considering the default meteorological MERRA2 dataset (Rienecker et al., 2011). However, we note that all &slh_nl values are affected by changes in the dynamical transport, and must be fine-tuned in case other meteorological fields are imposed, as for example those from the fifth generation ECMWF (European Centre for Medium-Range Weather Forecasts) reanalysis (ERA5, 2023). See the Appendix and the CESM2-SLH Mendeley repository for further details.”*

Finally, we note that the description of the SLH scaling factors (including the original Table 7) as well as the basic guidance about how to adjust and/or extend them is now condensed in the new Appendix.

Technical corrections

GENERAL ANSWER: Thanks a lot for the very detailed evaluation. Below we provide direct answers to all technical corrections. In some cases, we have grouped consecutive comments into a single response. Note that “format/style” modifications have not been highlighted in the track-changes version of the manuscript.

Section 1

R2-C5.1: L82: "X, Y = Cl, Br, I" Maybe write this as mathematical expression? $\rightarrow X, Y \in \{\text{Cl, Br, I}\}$.

ANSWER: No, thanks. We rather use the same style as in previous publications.

R2-C5.2: L99–118: Reactants in chemical formulae are supposed to be set upright, e.g. using `\chem` in LATEX. $\rightarrow$ <https://www.geoscientific-model-development.net/submission.html#math>

ANSWER: Thanks. We now use “normal/upright” format for all chemical species. Note we have not used LATEX but the “MS Word template” instead.

Section 2

R2-C6.1: L151: "(e.g., (Saiz-Lopez et al., 2023))" Missing ')'. $\rightarrow$ e.g. using LATEXcommand `cite[e.g.][]<sl23>`, where `<sl23>` is the reference label $\Rightarrow$ (e.g., Saiz-Lopez et al., 2023).

R2-C6.2: L239 (and many others): "[...] described in the original work of (Ordoñez et al., 2012)." When citing a reference in-text, in-text citation should be used, e.g. in LATEX `\citet{}` $\Rightarrow$ Ordoñez et al. (2012).

ANSWER: Thanks. All references and citations (including in-text citation) now follow GMD style. Note we have not used LATEX but the “MS Word template” instead.

R2-C6.3: L239: "[...] has been extensively described in the original work of [...]" This expression is odd. $\rightarrow$ 'has been described in'.

ANSWER: Thanks. Done.

R2-C6.4: L248: "20°-50°" em-dash should be used according to journal house-style. Please check similar expressions! $\rightarrow 20^{\circ} - 50^{\circ}$

ANSWER: Corrected here and elsewhere.

R2-C6.5: L252: Formula not set correctly. Please refer to the journal house-style, e.g. non-counting indices should be typeset upright. $\rightarrow$ <https://www.geoscientific-model-development.net/submission.html#math>

ANSWER: Done.

R2-C6.6: L256: "Note that, differing from previous [...]" As indicated above, historical context is unnecessary and can be omitted.

ANSWER: The context is not historical, we aimed to highlight the advantages of the Ordoñez inventory in comparison to other approaches. To avoid confusion, we have rephrased the sentence as follows:

L228-231: "In comparison to other VSLS approaches considering only only latitudinal bands (Butler et al., 2007; Jones et al., 2010; Warwick et al., 2006b, a), the Ordoñez et al. (2012) inventory introduces a tropical geographically-

heterogeneous and seasonally dependent variability, which allows for an improved spatio-temporal representation of VSL halocarbon distribution in the global troposphere.”

R2-C6.7: L259–263: *”Despite [...]”* This sentence is hard to understand like most of your sentences that start with ”despite”. Try to rephrase this and similar sentences! → ’The emission flux is predefined and read from file to which we referred to as offline emissions. The FORTRAN routine mo_srf_emissions.F90’ was modified to compute a time-dependent halocarbon emission in hourly resolution accounting for the radiation dependency of photosynthesis. Emissions follow either [...]’

ANSWER: We have carefully checked all sentences starting with *Despite* and apply changes accordingly. We have rephrased the referred sentence as follows (see complete answer to **R2-C2.1**):

L231-233: *“Once the monthly-mean prescribed fields are read, hourly dependent profiles are applied to the flux strength of all halocarbons to represent the photosynthetic dependence on radiation intensity.”*

R2-C6.8: L269–281: *”Finally, [...]”* Please refrain from giving historical context, rather concentrate on the important content! This paragraph should be rewritten along the lines: → ’Due to improvements in MBL dynamics and resulting different oxidative capacity, the Ordoñez emission inventory for brominated and chlorinated species has to be rescaled by a factor of 1.15 (15%) to achieve the observed stratospheric halogen loading.’

ANSWER: The context is not historical, we aimed to highlight the advantages of the Ordoñez inventory in comparison to other approaches. To avoid confusion, we have rephrased the sentence as follows:

L237-244: *“Due to improvements in the representation of large-scale ascent and transport across the boundary layer in CESM2 (Simpson et al., 2020) as well as in the emission strength and speciation of air pollutants altering the oxidative capacity of the troposphere (Emmons et al., 2020) the original Ordóñez et al. (2012) inventory for chlorinated and brominated species was increased by a constant factor of 1.15 (i.e., 15%) to reproduce the stratospheric halogen loading obtained in previous CESM1 studies (Fernandez et al., 2014, 2021). The rationale for increasing the offline emission fluxes is that the reduction of transport across the boundary layer and the increase near-surface OH abundance in CESM2-SLH results in more efficient conversion from SGs to PGs at lower model levels. Given the much shorter photochemical lifetime of iodine species, no scaling was applied to VSL iodocarbons.”*

R2-C6.9: L284–286: *”For the particular case of chlorine, [...]”* In-text citation style instead of parenthesis needed. Rephrasing of this sentence is recommended as it is hard to understand. → ’For anthropogenic SLH sources of chlorine, we follow the Hossaini et al. (2019) and Claxton et al. (2020) emission inventories. These inventories include time-dependent emission of two major contributors to the organic chlorine load, CH₂Cl₂ and C₂Cl₄.’

R2-C6.10: L289–291: *”Given that the dominant VSL chlorine sources [...]”* This sentence is difficult to understand. Rephrase along the lines: → ’Beside significant emissions of CHCl₃ from natural sources, chlorinated VSL are dominated by anthropogenic emissions that display a pronounced hemispheric asymmetry.’

R2-C6.11: L293: *”[...] globally scaled [...] globally.”* Doubling. Remove one ”globally”

R2-C6.12: L297–298: “[...] but not yet implemented [...]” You may remove the part of the sentence as it is not important whether it has not been implemented in any CESM version.

ANSWER: Thanks for the suggestions. We have substantially modified and shorten the text as follows:

L247-257: *“For anthropogenic VSL sources of chlorocarbons, we follow the Hossaini et al. (2019) and Claxton et al. (2020) emission inventories, which include time-dependent emission of two major contributors to the organic chlorine load (CH_2Cl_2 and C_2Cl_4) as well as surface LBCs for two other compounds (CHCl_3 and $\text{C}_2\text{H}_4\text{Cl}_2$). Based on these works, an increasing trend from 2000 to 2020 is applied to the emission and LBC files for the recent past. Beside a fraction of CHCl_3 emissions arise from natural sources (with minor contributions from the other compounds), chlorinated VSL are dominated by anthropogenic emissions that display a pronounced hemispheric asymmetry (WMO, 2022). Therefore, hereafter we assume all of these offline emissions to have only an anthropogenic origin. Consistent with the scaling factor applied to the Ordóñez et al. (2012) inventory, both VSL chlorocarbon surface emissions and LBCs were globally scaled by the same constant factor (1.15 or 15% enhancement). It should be noted that additional anthropogenic halocarbon sources, as those arising from the waste treatment of power-plants as well as seaweed aquaculture are not considered (Carpenter et al., 2000; Jia et al., 2023; Leedham et al., 2013).”*

R2-C6.13: L298–301: “To include [...]” This would be part of the proposed “user guide” Appendix section.

R2-C6.14: L308: “[...] variables HOI and I_2 [...]” HOI and I_2 are not “variables” but ‘trace gases’. Please phrase accordingly.

R2-C6.15: L307–310: “To include [...] and replaced by the online computation.” This would also be part of the “user guide”

ANSWER: We have moved most of the referred text to the new Appendix (see complete response to **R2-C1.4** and **R2-C2.6** above), and rephrased the remaining sentence as follows:

L263-269: *“Following Prados-Roman et al. (2015b), the parameterized expressions for HOI and I_2 fluxes from MacDonald et al. (2014) are computed online by Eq. (2a) and (2b), respectively, ...”*

R2-C6.16: L315–317: Formulae not set correctly, e.g. $\mathbf{F}_{I_2} \rightarrow \mathbf{F}_{I_2}$; $\ln(\omega) \rightarrow \ln(\omega)$; $1.75 \times 10^9 \rightarrow 1.75 \cdot 10^9$; →<https://www.geoscientific-model-development.net/submission.html#math>

ANSWER: Done.

R2-C6.17: L310–313: “The implementation of this source [...]. The online emission fluxes [...]”. Rephrase. → ‘Following Prados-Roman et al. (2015)b; MacDonald et al. (2014) the online emission fluxes of [...] are included in the Fortran module file iodine emissions.F90 and computed in subroutine ??? with Eq. [...].’

ANSWER: These technical descriptions have now been moved to the Appendix as follows:

L1619-1622: *“Online oceanic sources for inorganic iodine are included by adding variables HOI and I_2 to the standard srf_emiss_specifier used for offline VSLS halocarbons, although the imposed offline iodine file represents just a place-holder as the input values are forced to zero after being read within mo_srf_emissions.F90 routine and replaced by the online computation performed in iodine_emissions.F90.”*

R2-C6.18: L326,327: m/s → m s⁻¹

R2-C6.19: L328–329: *"Finally, we highlight [...] with ozone abundance [...]"* This sentence seems to be grammatically not correct and is one example for the extensive use of filler phrases mentioned above. *"Finally, we highlight"* should be removed and the sentence (and other sentences of similar kind, too) rephrased. → 'The first order dependency on ozone abundance of F_{HOI} and F_{I2} has important [...].'

ANSWER: Done.

R2-C6.20: L338: *"[...] following Eq. (3)"* Might be removed as the referred equation is following right after.

ANSWER: Done.

R2-C6.21: L345: *"[...] (R is the universal [...])."* Rephrase and resolve the brackets. → '[...], with R the universal [...].'

R2-C6.22: L346: 3 (*three*) Not clear. According to house standard this should read 'three'.

ANSWER: Done, thanks.

R2-C6.23: L382–385: *"This is indeed the case, [...]"* This sentence should be rephrased and split to make it clearer. → 'The major difference for aerosols between CESM1 (CAM4-chem) and CESM2 (CAM6) is the change from a continuous bulk representation with four size bins to a modal representation with three bins.'

ANSWER: Thanks, we have split the sentence and rephrased the whole text as follows (see complete response to **R2-C2.11-13** above):

L535-540: *"The major difference between CESM1 (CAM4-Chem) and CESM2 (CAM6-Chem) is that the former uses a continuous bulk representation of aerosols with four size bins, whereas the latter uses a modal representation with three bins (Danabasoglu et al., 2020). Another relevant difference is that MAM4 is the default dust scheme in CESM2 (Liu et al., 2016). Consequently, the vertical extent of SSA in the free-troposphere was largely reduced to improve the model overestimation obtained in previous studies (Lamarque et al., 2012; Tilmes et al., 2023)."*

R2-C6.24: L397: Wrong arrow. '→'

ANSWER: Replaced here and elsewhere.

R2-C6.25: L400–403: *"The CESM-SLH implementation [...]"* This sentence should be split after *"Eq. (3)"*. The second part is difficult to understand and should be rephrased along the lines: → 'The emission of chlorine from the bulk sea salt aerosols is driven by the heterogeneous reaction of oxidized nitrogen species that do not contain halogens.'

ANSWER: We have clarified the sentence as follows:

L334-336: *"The implementation of both chlorine sources follows the FRA-approach described above for oxidized halogen reservoirs (Eq. 3), although in this case the uptake of oxidized nitrogen species drives the SSA-dehalogenation."*

R2-C6.26: L412–413: *"[...] (e.g. [...])"*. Missing parenthesis. It might be better to connect the half-sentence by comma. → '[...], e.g., [...].'

ANSWER: Done.

R2-C6.27: L427–432: The 'checksum' of reaction types seems to be unnecessary. You may consider constructing a matrix/table of the following form:

Family	ph	odd	org	sh	het	ss	strat	total
Chlorine	11	6	20	2	7	11	10	62
Bromine	10	9	8	1	6	6	13	53
Iodine	17	39	2	1	10	6	18	93
Total	31	46	24	4	17	16	25	163

ANSWER: We thanks the reviewer for the suggestion. Following the arrangement of other tables, we have reshaped and included the new Table A2 in the Appendix. The revised text reads as follows:

Table A2: Total number of additional gas-phase and heterogeneous-phase halogen reactions included in CESM2-SLH chem_mech.in.

Halogen Family Type of Reaction	Chlorine	Bromine	Iodine	Total ^{&}
Photolysis	11	10	17	31
Odd-Halogen	6	9	39	46
Organic Halogen	20	8	2	24
Sulfur Halogen	2	1	1	4
Het-recycling (Tropo)	7	6	10	17
SSA-Recycling	11	6	6	16
Het-recycling (Strat)	10	13	18	25
Total	67	53	93	163

[&] Some reactions involve inter-halogen interactions (e.g., ClO + BrO) and are double-counted within each halogen family. Therefore, the total reaction sum by halogen families is not additive. The complete list of halogen reactions is provided in main Tables 1 to 5 and Supplementary Tables S1 to S3.

L1637-1642: “Overall, the final SLH chemical mechanism implemented in CAM-Chem introduces 12, 9 and 19 additional **chlorine, bromine and iodine** species that participate in 67, 53 and 93 new reactions. These new reactions are categorized as: photolysis, odd-halogen reactions, organic-halogen reactions, sulfur-halogen reactions, heterogeneous recycling on tropospheric aerosols, sea-salt recycling, and stratospheric mapped reactions (see Table A2), where some of the reactions involve inter-halogen interactions (e.g., ClO + BrO), and therefore are double-counted within each family. The inclusion of SLH chemistry in CESM2 results in an increment of the computational cost of ~18-22%.”

R2-C6.28: L442–443: “JPL [...] IUPAC” Acronyms have not been previously defined.

R2-C6.30: L453: “JPL 19-5 handbook” inconsistent with L442.

ANSWER: Done.

R2-C6.29: L452–456: “[...], some laboratory measurements show [...]. [...] recommends neglecting the photochemical breakdown [...]. Within CESM-SLH [...] forcing to zero [...] was required [...].” These sentences are unclear and overcomplicated. Refrain from using a term like “forcing to zero”. You are basically

trying to say: → 'We are following the JPL recommendation, although some laboratory measurements suggest [...].'

R2-C6.31: L456–457: *"Not doing so [...]."* This sentence is somewhat redundant. References are missing. The sentence could be removed or should be rephrased if kept: → 'Including photolysis rates above [...] resulted in a complete decomposition of C₂Cl₄ down to the surface which is not in line with observations.'

ANSWER: Thanks for the suggestions. We have clarified the whole sentence as follows:

L373-376: *"In addition, we follow the JPL 19-5 recommendation (Burkholder et al., 2020) of neglecting the C₂Cl₄ absorption cross-section beyond $\lambda > 270$ nm (Keller-Rudek et al., 2013), as not doing so resulted in a complete photodecomposition in the lower troposphere which is not in line with observations (Roozitalab et al., 2024)."*

R2-C6.32: L475–477: *"Here, we highlight [...]"* Rephrase and avoid phrases like *"Here, we highlight"*. Furthermore, it is not clear which CESM version this sentence refers to.

R2-C6.33: L477–478: *"Therefore [...]."* Missing comma after *"Therefore"*. Overall, this sentence seems to be in disorder grammatically. Please rephrase.

ANSWER: We have clarified the whole sentence as detailed in **R2-C2.13**:

R2-C6.34: L491: *rate_{ICE}* : Variable typesetting not in accordance to house style. → *rate_{ice}*

ANSWER: Following the house style, we now use upright lower-capital letters for the subindex, but maintained the italic format for the reaction-rate (*rate_{ice}*), as it represent a varying magnitude as the flux (*F₁₂*).

R2-C6.35: L491–493: *"[...] assumed to be uptake [...]"* This sentence is grammatically incorrect. Please correct.

ANSWER: We have clarified the text as follows (see also the related response to **R2-C2.14-15** above):

L447-449: *"This includes temperature and water-vapour dependent re-activation reactions of chlorinated and brominated reservoirs occurring over stratospheric sulphate (SAD_{sulfc}), ice-crystals (SAD_{ice}) and nitric acid trihydrate (SAD_{NAT}) surfaces (Kinnison et al., 2007), including recent updates of individual stratospheric accommodation coefficients (γ^{strat} ; Solomon et al., 2015)."*

R2-C6.36: L533: *"[...] a logical condition of humidity [...]"* This phrase is unclear. What does this mean? Please rephrase and make clearer.

ANSWER: We have clarified the text as follows:

L410-420: *"For heterogeneous reactions occurring over ice-crystals, we use the FRA approach to compute the reaction rate constant (rate_{ice}) considering the total number of gas-phase species that suffer uptake on the aerosol surface, resulting in the following expressions for uni-molecular and bimolecular reactions (see Table 2)."*

R2-C6.37: L540: *"FC"* is not defined and should be explained

ANSWER: We now explicitly point at FCnudged_slh and FCHIST_slh compsets.

R2-C6.38: L575: "*Henry law coefficient*" inconsistent naming. Called "constants" in L576.

ANSWER: We have now modified the text to use always "constants" following Sander (2015).

R2-C6.39: L630: "*maintained*" → 'kept'.

ANSWER: "*are identical to those used in*".

R2-C6.40: L634–635: *Despite none of them have been used in this work, we note [...]* Odd expression. The quoted part of the sentence may be removed.

R2-C6.41: L639: "*[...] many [...] depends [...]*" → 'depend'.

R2-C6.42: L640–642: "*Similarly, we highlight that [...] significantly impact on [...]*" Filler-phrase and grammar. You may remove the filler-phrase. The preposition "on" doesn't belong to "impact" and should be removed.

R2-C6.43: L642–643: "*[...] shifting [...]*" The expression might not to be correct. → 'changing'

ANSWER: Thanks for the suggestions. We have split the referred sentence between new Sections 3.1 ("*Model compsets and Resolutions*") and 5.1 ("*Discussion*") and clarified all of the above as follows:

L558-562: "*We note that additional emission and LBC files for other configurations using different SSP scenarios are available in the published CESM2-SLH repository and can be changed through user-defined namelist options (see below). In addition, equivalent model configurations using a finer resolution f09_f09_mg17 grid (0.9° latitude × 1.25° longitude, hereafter 1°×1°) have also been tested and adjusted to reproduce results obtained with the 2°×2° grids.*"

L1477-1484: "*Despite a high-spatial resolution being available (see Table 7), we recommend new users to select the coarse resolution setup as many of the online photochemical sources and recycling reactions described in Section 2 depend on highly variable resolution-dependent atmospheric fields, such as SAD_{SSA} and SAD_{ice}, which typically show larger variability with increasing resolution (see Tables S4 and S5). Similarly, we found that resolution-dependent scaling factors for lightning-NO_x production typically used in CESM2 (Emmons et al., 2020; Wild et al., 2020) result in significant alterations of the chemical partitioning and washout efficiency of SLH. Therefore, caution should be taken when moving from a coarse to a fine resolution, as the resulting halogen abundances can significantly vary between them.*"

R2-C6.44: L705: "*rationale*" This expression may not be correct in this context. → 'reason'

ANSWER: Done.

R2-C6.45: L719: "*(the so called AANE sensitivity in (Saiz-Lopez et al., 2023))*" Missing parentheses and formatting of the citation. Acronym AANE is not resolved. The insertion should be elaborated on and written out.

ANSWER: We have clarified the Anthropogenically Amplified Natural Emissions (ANNE) concept as follows:

L612-617: "*This inventory is mostly relevant for air quality studies focused on urban and/or continental regions (Chang et al., 2024; Fu et al., 2024), without contribution to the free troposphere and stratospheric halogen loading.*"

Indeed, Saiz-Lopez et al. (2023) found that the SLH influence on global atmospheric composition and radiative balance is dominated by natural SLH emissions, which have been anthropogenically amplified since pre-industrial periods due to the efficient coupling of halogenated species with background air pollutants, e.g. the so called AANE sensitivity in Saiz-Lopez et al. (2023)."

Section 3

R2-C7.1: L738–740: *"Results presented [...] are based on [...] sensitivities [...].* Sentence contains lab-slang. "sensitivities" → 'sensitivity studies' or 'sensitivity experiments'. Please change all occurrence accordingly!

ANSWER: Following reviewer comments **R2-C1.4** and **R2-C9.6**, we have now include a whole new section describing the CESM2-SLH release, where we split original Table 6 into two separate tables: Updated Table 6 which includes the new SLH compsets and configurations, and the new Table 7, that summarizes the full set of experiments performed. To make this clear, the manuscript structure has been modified to include the following subsections **"3.1 Model Compsets and Resolutions"** and **"3.2 Model Experiments and Sensitivity Simulations"**. We have revised the manuscript to make sure to unify the usage of the terms *"experiment"* (for SLH) and *"sensitivity"* (for NOH) throughout the text, while using the general term and *"simulations"* when referring to either of them. We have defined all terms in Section 3.2 as follows:

L570-576: *"The complete set of model experiments performed in this work is presented in Table 7. Each experiment's name is composed by a prefix describing the model used and the inclusion of short-lived halogens (e.g., CC-SLH for CAM6-Chem with SLH turned on) and a suffix indicating the model resolution and meteorology option (e.g., [2×2-ndg] for coarse resolution and nudging approach). Although all model configurations have been validated, initial results presented in Section 4 are based on low-top and coarse CC-SLH [2×2-ndg] and/or CC-SLH [2×2-hst]. We note that for each of the six SLH experiments shown in Table 7, an additional model sensitivity was performed neglecting the sources and chemistry of short-lived halogens (e.g., the corresponding CC-NOH [2×2-ndg] experiment, see Appendix)."*

R2-C7.2: L747–748: *"Finally [...] model inter-comparison between the different CESM2-SLH resolutions and configurations.* Usually the term "model inter-comparison" is reserved for the Coupled Model Intercomparison Project (CMIP) and derivatives – organized comparisons between different models in the Earth System and Climate modeling community with a clear modeling protocol. "model intercomparison" → 'comparison' or 'sensitivity experiments' or 'sensitivity studies'.

R2-C7.3: L754: *"Therefore, we highlight that [...]"* As mentioned previously, these filler-phrases should be removed throughout the text to improve readability.

ANSWER: We made sure not to use "model inter-comparison" here and elsewhere to avoid misleading interpretations. We also removed filler-phrases to improve readability.

R2-C7.4: L780–782: *"Here, we highlight [...]"* This sentence should be split after "iodine".

R2-C7.5: L782: *"on-line"* → 'online'

ANSWER: We have completely rephrased and split the sentence and made sure to correct “online” and “offline” here and elsewhere:

L662-667: *“This online inorganic halogen flux shows an increasing emission trend between 1980 and 2020 (Fig. 1) for the three halogen families, although the main drivers of the parameterized expressions are significantly different for chlorine and bromine with respect to iodine (see Section 2.1.3). Indeed, (Prados-Roman et al., 2015a) suggested that this online ocean–atmosphere coupling represents a negative geochemical feedback loop by which current ocean emissions of iodine ($\text{HOI} + \text{I}_2$) would be 2-3 times higher than in pre-industrial times, constituting a natural buffer for ozone pollution in the global marine environment.”*

R2-C7.6: L809: “allow” → ‘allow for’

R2-C7.7: L809–810: “to provide [...] to show” Correcting the above, these infinitive forms become unnecessary.

R2-C7.8: L816: “[...] presents a larger increment than in CESM1 [...]” This sentence is unclear. Do you mean → ‘Iodine volume mixing ratios continue to increase after year 2000 in the CESM2 experiments, while they flatten out in the CESM1 results’? This could be construction of your CESM1 experiments, e.g. recycling of SSTs and SICs after the end of the hindcast period...

ANSWER: We have corrected the whole text as follows:

L699-707: *“These CESM1 configurations were selected as they allow for a better qualitative visualization and quantitative comparison of the absolute abundances of each halogen family and their reactive to inorganic halogen ratios (XO_x/X_y , with $\text{XO}_x = \text{X} + \text{XO}$ and $\text{X} = \text{Cl}, \text{Br}$ and I), as well as the main atmospheric components driving the trends during the hindcast period.”... “Indeed, the increasing trend in surface ozone abundance during the 1980–2020 period in CESM2-SLH compared to the more flatten trend observed in CESM1 (Fig. 2f) results in an increasing trend in iodine volume mixing ratios in the former, while I_y abundances in the latter remained approximately constant, particularly after year 2000 (Fig. 2c).”*

R2-C7.9: L864–868: “Here we note that [...]” Remove filler-phrase and split the sentence before “highlighting”

R2-C7.10: L870–873: “In addition, [...] in any case [...] in any case” Repetitive expression. At least on “in any case” should be removed.

R2-C7.11: L870–873: “[...] in the lower edge [...]” grammatically not correct. → ‘at the lower margin’

ANSWER: We have corrected the whole text as follows:

L750-758: *“Second, the spread of the global mean tropospheric halogen burden between the different CESM2-SLH experiments is smaller than that obtained with CESM1 in previous studies. Indeed, for the case of bromine, the tropospheric Br_y burden for both CC-SLH [2×2-ndg] and CC-SLH [2×2-hst] are equivalent to those obtained for CESM1-SLH [QL22], although they lie in between CESM1-SLH [SL23] and CESM1-SLH [JB23] (Fig. 3e). This range of tropospheric Br_y burdens arise due to changes in the seasonal and latitudinal sea-salt aerosol masks used when the CESM1 polar module is enabled (Fernandez et al., 2014, 2021), highlighting the large sensitivity of the SSA-dehalogenation source to the modelled changes in SSA abundance and distribution (see Fig. S2). The tropospheric Cl_y burden for both CESM2-SLH nudged and free-running configurations are equivalent to those obtained for CESM1-SLH [JB23] and lower than for CESM1-SLH [QL22] and CESM1-SLH [SL23] (Fig. 3d).”*

R2-C7.12: L874: *"timeline simulation"* This expression is odd. Do you mean 'transient simulation' or 'hindcast' ?

ANSWER: Done.

R2-C7.13: L877–880: *"The larger tropospheric [...]"* This sentence is grammatically not sound and should be rephrased. How is present-day conditions defined? → 'The larger present-day tropospheric I_y burden in *FCHIST_slh* and *FCnudged_slh* ([...]) compared to estimates by Saiz-Lopez et al. (2023) and Barrera et al. (2023) ([...]) are due to substantial changes [...]'

ANSWER: Present-day was already *"defined as the mean 2015–2020 period"* in the original manuscript (see **L642**). Note we corrected to use en-dashes (–) instead of hyphens (-) for concatenating period ranges here and elsewhere. We have rephrased the referred sentence as follows:

L762-764: *"The larger present-day tropospheric I_y burden between current CESM2-SLH experiments (17.2-17.7 Tg I) compared to to CESM1-SLH [SL23] and CESM1-SLH [JB23] (14–14.5 Tg I) are explained by the significant changes in reactive (IO_x) vs. reservoir iodine partitioning (IO_x/I_y) between CESM1 and CESM2."*

R2-C7.14: L885–886: *"Table 8 [...]"* This is a rather unusual way to start a section. You should rather start with a sentence of introduction.

R2-C7.15: L887–888: *"Equivalent results highlighting [...]"* This sentence is unclear and should be rephrased or, as it references a later section, it may be removed

ANSWER: We have modified the opening paragraph of the section as follows (see response to comment **R2-C9.8** below related to the shift in the numbering of tables in the revised manuscript):

L770-774: *"Annual mean tropospheric halogen abundances, LBCs, sources and sinks during present-day (2015–2020) are summarized in Table 8. These global-mean reference values for the CC-SLH [2×2-ndg] and CC-SLH [2×2-hst] experiments should be taken as a guideline for any other CESM2-SLH configuration. The main similarities and differences between the recommended setup with respect to the remaining CESM2-SLH compsets and/or resolutions available (Tables 6 and 7) are described in Section 4.4."*

R2-C7.16: L890–893: *"Here, we recall [...]"* Odd style AND repetition. Should be removed.

ANSWER: Done.

R2-C7.17: L893–896: *"Thus, VSL iodine mean global emissions are the same between [...]"* This half-sentence is grammatically not sound and difficult to understand. It should be rephrased. → 'Global mean iodine emissions are (by definition) the same in CESM1 and CESM2, while [...]'

ANSWER: We have modified the opening paragraph of the section as follows:

L776-780: *"Due to changes in transport and background OH abundance between CESM1 and CESM2 (see Figure 2), we increased the flux strength of the original VSL inventory (Ordóñez et al., 2012) by 15% for all VSL bromo- and chloro-carbon (e.g., CH_2Br_2 and CH_2BrCl) but retained the magnitudes of iodo-carbon species, including CH_2ICl and CH_2IBr . As a result, Cl and Br emissions are ~14% and 6% larger in CESM2 compared to CESM1. As a result, chlorinated (VSL_{Cl}) and brominated (VSL_{Br}) halocarbon emissions are, respectively, ~6% and 14% larger in CESM2 compared to CESM1"*

R2-C7.18: L902: "*online sources*" → 'online emissions'

R2-C7.19: L919: "[...] *inspection of Table 8 shows [...]*" This expression is odd. → 'We show in Table 8 [...]' or 'In Table 8 it is shown [...]'

ANSWER: Done both.

R2-C7.20: L950: "[...] *remains similar compared to [...]*" This is another odd expression and should be rephrased. Please also include some numbers from the cited publication. → 'remains comparable with'

ANSWER: Thanks for the suggestion. We have modified the referred sentence as follows: (see also the response to **R2-C3.5** above):

L836-838: "*Therefore, the reactive IO_x tropospheric burden in CESM2 (3.5-3.6 Gg I) is only ~10% than the 3.2 Gg I estimated value found in Saiz-Lopez et al. (2023).*"

R2-C7.21: L955: "*Fig. 3 bottom row*" Here and elsewhere, please reference the subplots with their respective subcaptions.

ANSWER: We have changed all sentences in the text where pointing at rows or columns of Figures, and replaced by the corresponding panel letters (e.g., Fig. 3a-c for the referred comment). However, we keep pointing at rows and/or columns within some figures' captions and table's footnotes to ease the reading.

R2-C7.22: L958: Wrong arrow in the in-line formula. → '→'

R2-C7.23: L968: "*Here it is evident [...]*" Yet another filler-phrase that can be safely removed.

R2-C7.24: L984: "[...] *the difference between both configurations [...]*" "Configurations" is not the right term. As indicated above, you should clearly distinguish between setups/configurations/compsets and model experiments → 'experiments'.

R2-C7.25: L986–987: "[...] *from organic SGs to inorganic halogen (PGs) [...]*" The brackets around PGs seem to be misplaced.

ANSWER: Done all. See the response to **R2-C1.4** and **R2-C7.1** above for a detailed answer about the experiments.

R2-C7.26: L989–990: "[...] *(see for example Table A-1 in the Annex of (WMO, 2022) and elsewhere) [...]*". In-text citation needed. You should refrain from using "elsewhere", either you would cite other sources or not. The sentence is too complicated and should be split here, removing the parentheses.

ANSWER: We have modified the sentence as follows:

L964-966: "*Given the typically longer photochemical lifetimes of long-lived chlorinated compounds compared to brominated halons (see Table A-1 in the Annex of WMO (2022)), the latter presents a steeper vertical gradient in the lower stratosphere and an almost complete conversion to inorganic bromine in the upper stratosphere (Fig. 9b).*"

R2-C7.27: L1004–1005: “[...] which is driven by [...] (see Fig. 7).” Either the order of figures is wrong here and you are actually referring to Figure 6 or the order of the text does not match the figure order.

ANSWER: The order of the figures is correct. We are pointing at the green lines showing the SSA dehalogenation sources of chlorine and bromine presented in original Figure 7a,c (current Figure 10a,c). We’ve updated the text to avoid confusion.

R2-C7.28: L1010: “*throughout tropical injection*” → ‘through the tropical tropopause region’

ANSWER: Thanks for the suggestion. Done.

R2-C7.29: L1086–1089: “*In doing so, [...]*” This is a repetition of model description and development and does not belong here.

R2-C7.30: L1089–1090: “*For this exercise, [...]*” This expression is odd in the context of scientific paper. Furthermore, you should give references to the indicated “standardized evaluations against observations and international assessments” and describe them briefly.

R2-C7.31: L1086–1090: Given the comments above, you could also remove the whole paragraph and only keep the first sentence.

ANSWER: We completely changed the validation section and therefore removed the complete paragraph.

R2-C7.32: L1099: “*configurations*” Do you mean ‘model experiments’ ?

ANSWER: Yes. We have rephrased the whole sentence. See related response to **R2-C7.1** above.

R2-C7.33: L1105: “[...] the more extreme contributions [...]” Unclear expression. What do you mean by “extreme contributions”?

ANSWER: We removed the referred half-sentence to avoid confusion.

R2-C7.34: L1120: “[...] modelled range indicate the variability between compsets.” Grammatically not correct. “indicate” → ‘indicates’. Furthermore, you may need to rephrase this sentence as it is not quite clear. → ‘The results show that the modelled SG to PG partitioning and total abundance of VSL in the stratosphere depend on the chosen compset.’ You may discuss the reason (difference between the compsets) in a dedicated discussion section.

ANSWER: We have split the sentence in two, and rephrased the second half-sentence as follows:

L863-865: “*Notably, all experiments simulate SGs vertical profiles that are within the accepted range reported in the last Ozone Assessment Report (WMO, 2022) throughout the tropical troposphere regardless of the compset and resolution considered.*”

R2-C7.35: L1130: “*Differing [...]*” → ‘Unlike’

ANSWER: Done.

R2-C7.36: L1165: "Having said this, we acknowledge [...]" This expression is misplaced in the context of a scientific paper.

R2-C7.37: L1169: "online sources" This seems to be lab-slang. → 'online emissions' or 'emission computed during runtime of the model'.

ANSWER: Based on the reviewer suggestion of including a dedicated Discussion section (**R2-C1.3** and **R2-C3.1**), we have moved the sentence to the new Section 5 and rephrased it to avoid using lab-slang as follows (see also response to **R2-C3.10** above):

L1409-1410: *"In particular, VSL_{Cl} photodegradation represents only a minor contribution to atomic Cl release in the troposphere, which is largely dominated by the online acid-displacement emissions."*

R2-C7.38: L1183–1204: This whole paragraph should be revised. Many sentences are grammatically incorrect or use odd expressions: "Several distinct features are observed", "This species presents", "Indeed, despite maximum Br_y peaks in the NH are up to".

ANSWER: We have completely re-structured the whole paragraph, without using an explicit bullet for each halogen family and avoiding the use of odd expressions. The revised paragraph reads as follows:

L1062-1079: *"Driven by the prevalence of anthropogenic sources in the NH, VSL_{Cl} exhibits a pronounced South-North latitudinal gradient between ~60 and ~260 pptv (Fig. 11a), with peak values exceeding 500 pptv over China. In contrast, VSL_{Br} shows a more homogeneous spatial distribution with maximum abundances observed over the equatorial regions due to the dominant source from the tropical oceans (Ordóñez et al., 2012). Minimum, maximum and mean global VSL_{Br} mixing ratios reach 2.9 pptv, 12.9 pptv and 6.8 pptv, respectively. The global mean VSL_I is ~1.0–1.2 pptv (see Table 8). Inorganic chlorine also maximizes over the NH, particularly over coastal locations where the mixing of halogen-rich SSA and HNO_3/N_2O_5 plumes results in an enhanced chlorine source. The highest Cl atom concentrations occur over the coastal regions of Europe, US and East Asia, with maximum values smaller than 0.4×10^{-3} pptv and a global mean average of 4×10^{-5} pptv. These distributions are consistent with maximum and mean Cl_y levels of ~640 pptv and ~47 pptv, respectively (Fig. 11d). Similarly, modelled Br_y also maximizes over coastal regions co-located with high SSA abundance (Fig. S2). However, the inter-hemispheric bromine enhancement is less pronounced than for chlorine because SSA-debromination is not controlled by HNO_3 and N_2O_5 , but instead by shifts in Br_y partitioning between $BrONO_2$, $BrNO_2$ and $HOBr$ (see Table 1). Maximum Br_y peaks in the NH are up to 2 times larger than in the SH. However, the corresponding maximum BrO abundances does not present such inter-hemispheric asymmetry (Fig. 11h), highlighting the strong dependence of reactive bromine partitioning on the background atmospheric composition of NO_x and HO_x . For iodine, global I_y distributions is primarily controlled by oceanic HOI/I_2 emissions that peak within the tropics. The mean modelled I_y and $I+IO$ surface abundances reach ~1.25 pptv and ~0.15 pptv, respectively, with corresponding volume mixing ratios spanning between (0.05 – 21) pptv for I_y and (0.008 – 0.90) pptv for $I+IO$. Notably, the global mean IO_x/I_y ratio is ~0.12, whereas in regions with the highest I_y abundances it remains below ~0.04."*

R2-C7.39: L1276: "significant". If significance is not strictly proven the term should not be used. → 'strong' or 'substantial'

ANSWER: Done.

R2-C7.40: L1293: "Regardless of the compset [...]" This phrase is not correct and should be removed, because the results for the CESM1 experiments do not fall within the given bounds of $-(2.7 - 3.9)\%$

ANSWER: The referred values point at CESM2-SLH results. To avoid confusion, we have rephrased the text as follows:

L1144-1146: *“CESM2-SLH reductions range between -2.7% and -3.9% at the surface and approximately -6% integrated in the troposphere for OH, while for NO₂ differences remain below -1% at the surface and ranging between -7.2% and -8.6% in the troposphere.”*

R2-C7.41: L1301: *“[...] is encouraged for future studies.”* Odd expression. Rephrase. → ‘should be subject to future studies.’

ANSWER: Done.

R2-C7.42: L1303–1304: *“[...] (top) [...] (middle) [...] (bottom row).”* Please reference the subfigures by their labels. → ‘(a–c) [...] (d–f) [...] (g–i)’

ANSWER: We removed the referred sentence. Note that we now point at the specific subfigures labels throughout the text. “”

R2-C7.43: L1306–1308: *“[...], although due to the high [...].”* “although” can be removed.

ANSWER: Done.

R2-C7.44: L1327: *“[...] is similar to the analogous pattern [...].”* It is not clear what “analogous” refers to in this context. It can be removed.

R2-C7.45: L1333: *“Indeed, and despite [...].”* Filler-phrase. Can be removed.

ANSWER: Done both.

R2-C7.46: L1348: *“60N-60S”*. Wrong formatted geographic range. → ‘60° N – 60° S’

ANSWER: Corrected here and elsewhere. See complete answer to **R2-C1.2** above.

R2-C7.47: L1353–1354: *“[...], where the model [...] fails to capture their seasonal cycle.”* “their” probably shall refer to “stations 11 and 12”. The stations do not have a “seasonal cycle”, but the ozone observed at the stations does. “their” → ‘the’

R2-C7.48: L1356: *“[...] impact in the [...].”* Grammatically not sound. → ‘impacts the’

R2-C7.49: L1357–1358: *“[...] predicted by a [...] pixel size in the model.”* Odd expression. A pixel size cannot predict anything. → ‘predicted by the model in a [...] resolution.’

R2-C7.50: L1358–1360: *“[...] demonstrate that the inclusion of SLH chemistry in the model helps to close the gap with observations”* Preposition “with” is not grammatically correct. → ‘on’ or ‘between model and observations’

ANSWER: Thanks for the suggestions. We have rephrased the complete sentence as follows:

L1202-1208: *“However, discrepancies emerge at some stations like 11 and 12, where the model successfully reproduces the observed ozone magnitude but fails to capture **the** seasonal cycle. This divergence likely stems from incomplete representation of ozone sources and sinks in this pristine region. Finally, the coarse resolution of the model is **well-known factor affecting** the comparison with in-situ observations, as the local ozone values in some observational sites do not necessarily represent the mean background levels predicted **over** a $1.9^\circ \times 2.5^\circ$ pixel size. Nevertheless, Fig. 14 clearly shows that the inclusion of SLH chemistry in CESM2 helps to close the gap **between the model and observations.**”*

R2-C7.51: L1399: *“setup”* Please clearly distinguish between ‘model experiments’ (in this section) and ‘model configurations’ in the model description section. Consistently use either ‘configuration’ or ‘compset’ or ‘setup’. It is not clear if those are the same.

ANSWER: Done. See the response to **R2-C1.4** and **R2-C7.1** above for a detailed answer about the experiments.

R2-C7.52: L1421: *“Globally, of the total halogen [...]”* This sentence is grammatically not correct and difficult to understand. Please rephrase.

ANSWER: To avoid confusion, we now point at the total halogen loss followed by the percentage contribution of iodine, bromine and chlorine. We have rephrased the complete sentence as follows:

L1265-1267: *“Globally, **the** halogen driven tropospheric **photochemical** ozone destruction **reaches** 659.4 Tg yr^{-1} for CC-SLH [2×2-ndg] and 662 Tg yr^{-1} for CC-SLH [2×2-hst], dominated by IO_x -induced **OddOx**_{Loss} (accounting for 76–79%) and with minor contributions for bromine (16–18%) and chlorine (5%).”*

R2-C7.53: L1513: *“on-line”* Inconsistency. → ‘online’.

R2-C7.54: L1568: *“ 1×1 ”* → $1^\circ \times 1^\circ$

R2-C7.55: L1570: *“This is associated with to the [...]”* → Remove “to”.

ANSWER: Done all of the above.

Section 4

R2-C8.1: This section accumulates many of the repeated issues with expressions and lab-slang. Please rephrase in accordance to all suggestions made. I refrain from giving a detailed account.

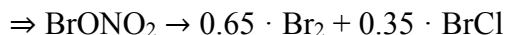
ANSWER: We have now checked the whole manuscript and rephrased to avoid repetition and using lab-slang. In particular, note that the revised manuscript now include a dedicated Section 5 for Discussions and that the original Section 4 has been re-numbered to Section 6. Please see also the general response to main comment **R2-C1.1-4** above and refer to the revised manuscript for the implemented changes.

Tables and Figures

Tables

R2-C9.1: Table 1

- Chemical reactions are not correctly set, e.g. $\text{BrONO}_2 \rightarrow 0.65 \cdot \text{Br}_2 + 0.35 \cdot \text{BrCl}$



- Exponential expressions are not correctly set.
- Empty lines and subheadlines should be removed. The information contained in the subhead-lines should be part of the table caption. $\rightarrow$ 'Table 1: [...]. Reactions het_ss_0...8 include heterogenated-reservoir species and het_ss_9...12 oxidized nitrogen-compounds.'

R2-C9.2: Table 2: same as Table 1.

R2-C9.3: Table 3: same as Table 1. What does "mapped to X" mean?

R2-C9.4: Table 4: same as Table 1. There us a lot of unnecessary whitespace in one column. This could be optimized.

R2-C9.5: Table 5: same as Table 4.

ANSWER: Done. Empty lines and blank spaces have been removed, while we kept a few essential subheadings to facilitate manipulation and formatting with the journal's graphical editors. To avoid an excessive length of the table caption, most information originally provided in subheading has been moved to table footnotes whenever needed. We have also corrected column titles and text format. These changes have been applied to all tables in the main text and supplementary material.

R2-C9.6: Table 6: Mixes the overview over available compsets with performed model experiments. It should be split. The part containing a summary of the available SLH-compsets should remain here. A Table summarizing ALL model experiments (also CESM1) should be place in the next section (Model experiments and results). You should clearly distinguish between compsets and model experiments. It is rather unfortunate that model experiments at different horizontal resolutions are listed under the same name. This ambiguity has to be resolved. Each experiment should have a unique identifier. "Suggested" and "available" do not make sense in the table. The recommendation should be part of the table caption.

ANSWER: We thanks the reviewer for this very important suggestion, that allowed us to clearly distinguish among the different model configurations used for each independent experiment. Based on his/her suggestions, as well as on the main comment **R2-C1.4** oriented to reorganize the manuscript and also on **R2-C7.1** pointing at the importance of unify and/or discern between the usage of setup, configuration, simulation and sensitivity, we introduced a whole new section entitled "**3.2 Model Experiments and Sensitivity Simulations**", were we include the new Table 7 and describe the whole set of experiments performed in this work.

The revised manuscript also clarifies that all CESM1 results shown in this work refer to previously published results and reads as follows:

L569-578: “3.2 Model Experiments and Sensitivity Simulations

The complete set of model experiments performed in this work is presented in Table 7. Each experiment’s name is composed by a prefix denoting the model used and the inclusion of short-lived halogens (e.g., CC-SLH for CAM6-Chem with SLH turned on) and a suffix indicating the model resolution and meteorology option (e.g., [2×2-ndg] for coarse resolution and nudging approach). Although all model configurations have been validated, initial results presented in Section 4 are mostly based on low-top and coarse CC-SLH [2×2-ndg] and/or CC-SLH [2×2-hst] experiments. We note that for each of the six SLH experiments listed in Table 7, an additional sensitivity was conducted neglecting the sources and chemistry of short-lived halogens (e.g., the corresponding CC-NOH [2×2-ndg] experiment, see Appendix). For most analysis comparing CC-SLH and CC-NOH distributions, we used the FCnudged_slh compset as this ensures that dynamical transport is consistently represented across experiments, which is crucial for isolating and quantifying chemical changes.

Table 7: CESM2-SLH experiments performed in this work.

<i>Experiment name</i>	<i>SLH compset</i>	<i>Resolution (lat × lon)</i>	<i>Period of time</i>
<i>CC-SLH [2×2-ndg]</i>	<i>FCnudged_slh</i>	<i>(1.9° × 2.5°)</i>	<i>1980 - 2020</i>
<i>CC-SLH [2×2-hst]</i>	<i>FCHIST_slh</i>	<i>(1.9° × 2.5°)</i>	<i>1950 - 2020</i>
<i>CC-SLH [1×1-ndg]</i>	<i>FCnudged_slh</i>	<i>(0.9° × 1.25°)</i>	<i>2000-2005</i>
<i>CC-SLH [1×1-hst]</i>	<i>FCHIST_slh</i>	<i>(0.9° × 1.25°)</i>	<i>2000-2005</i>
<i>FW-SLH [2×2-ndg]</i>	<i>FWnudged_slh</i>	<i>(1.9° × 2.5°)</i>	<i>2000-2005</i>
<i>BW-SLH [2×2-cpl]</i>	<i>BWHIST_slh</i>	<i>(1.9° × 2.5°)</i>	<i>2000-2005</i>

In addition to the CESM2-SLH experiments presented above, and with the intention to validate current model performance compared with previous CESM1 studies, we also analysed model output from the main simulations originally published by Li et al. (2022) and Saiz-Lopez et al. (2023), as well as in Barrera et al. (2023) and Bossolasco et al., (2025). For consistency, these experiments are referred to, respectively, CESM1-SLH [QL22], CESM1-SLH [SL23], CESM1-SLH [JB23] and CESM1-SLH [AB25]. Although these CESM1 configurations are not strictly identical, the experiments were conducted considering analogous spatial resolution and nudging options to those employed here in CESM2-SLH. They are therefore considered as the benchmark simulations for quantitative and qualitative intercomparison between CESM model versions. Further details of the main similarities and differences between SLH results performed with CESM1 and CESM2 are described below.”

R2-C9.7: Table 7: Empty rows should be removed. The subtitles ”SSA-Sources” and ”FRA-Sinks” should be moved into the Table caption.

ANSWER: Done. See related response to **R2-C9.1-5** above. Due to suggestion of moving the technical details to the Appendix (see response to **R2-C1.4** and **R2-C2.4-6** above), Table 7 has now been moved to Table A1.

R2-C9.8: Table 8: Table is oddly formatted. Horizontal lines appear broken and not all columns are labeled correctly, e.g. "species" seems to refer to 'species families'. The caption is not correct because the table displays different emission fluxes, volume mixing ratios, sinks, and atmospheric abundances. The rows "Abundance–Long-lived halogens" have no unit assigned. You may consider splitting the table into three or four parts which would make it also easier to reference in the text.

ANSWER: We implemented all format suggestions, corrected the table title, point at individual halogen families and included the missing units. However, we regret to split the table in parts as all sources, volume mixing ratios and burdens are of importance to properly evaluate the performance of different experiments. Therefore, we removed most of the horizontal broken lines but kept essential table subheadings to separate the different magnitudes. Please refer to the revised manuscript for the reformatted table.

R2-C9.9: Table 9: As mentioned earlier: "sensitivities" → 'sensitivity experiments'. Here and in other parts of the preprint, it remains unclear whether the CESM1 results refer to previously published results, which would need explicit citations, or new results based on old/repeated model experiments.

ANSWER: The revised text now refers to SLH and NOH experiments, and avoid using the term *compset* (see related response to **R2-C7.1** above). We have also clarified that all CESM1 results shown in this work refer to previously published results and modify the text as follows (see related response to **R2-C10.2** below).

L697-699: *"In order to compare the current CESM2-SLH inorganic halogen abundances with previous studies, Fig. 2 also shows the results obtained in previous studies (i.e., CESM1-SLH [QL22] and CESM1-SLH [SL23] corresponding to Li et al. (2022) and Saiz-Lopez et al. (2023), respectively)."*

R2-C9.10: Table 10: Wrong font. Empty lines should be removed. All tropospheric magnitudes [...] This text should be part of the table caption. What are "tropospheric magnitudes"? What is a "minimum lapse rate pressure"? Do you mean the pressure at which the lapse rate minimum is reached? A dot is missing at the end of the caption.

R2-C9.11: Table 11: A dot is missing at the end of the caption. The line below the table should be part of the caption. Wrong font.

ANSWER: We implemented all of the above format requirements in both tables, correcting font type and size and removing empty lines and blank-spaces. We also corrected definitions provided in the footnotes. To avoid an excessive length of the table caption, we rather keep the information as footnotes of each table.

R2-C9.12: Table 12: This looks like two pictures rather than a continued sideways table with a lot of artifacts ("continue next page", "page", cut in half row "Simulation ..."). Horizontal lines are broken.

ANSWER: We agree with the reviewer that original Table 12 was ugly formatted. We initially submitted inside a landscape page but we were requested to change it to portrait format during GMD quality check, and therefore it didn't fit into a single page. We have now carefully split the table in two and reformatted following the reviewer suggestions in **R2-C9.1-5** and **R2-C9.8**. Note that due to the major manuscript rearrangements applied to include the new Discussion Section 5, we decided to move original Table 12 to the Supplementary Material and split it in two separate tables (Tables S4 and S5).

Figures

R2-C10.1: Figure 1: "NH = 20°N(S) - 90°N(S)" Do not apply to the journal style guide (ranges and em-dash)

ANSWER: Thanks for the careful reading. We have now corrected the latitudinal and time-period ranges in all figures and captions. See also the response to **R2-C6.4** above.

R2-C10.2: Figure 2: As indicated above. The CESM1 experiments are not well characterized in this preprint and difficult to set in context. The arrangement of this figure is a bit unfortunate, putting VOC volume mixing radiations to a (random) spot in second row together with species ratios. It might be possible to split this figure into two (7:5 panels each).

ANSWER: We thanks the reviewer suggestion and have modified the arrangement of the figure to avoid including VOCs vmr in the second row where otherwise species ratios are presented. The new disposition of Figures panels, with the corresponding updated caption, is shown below:

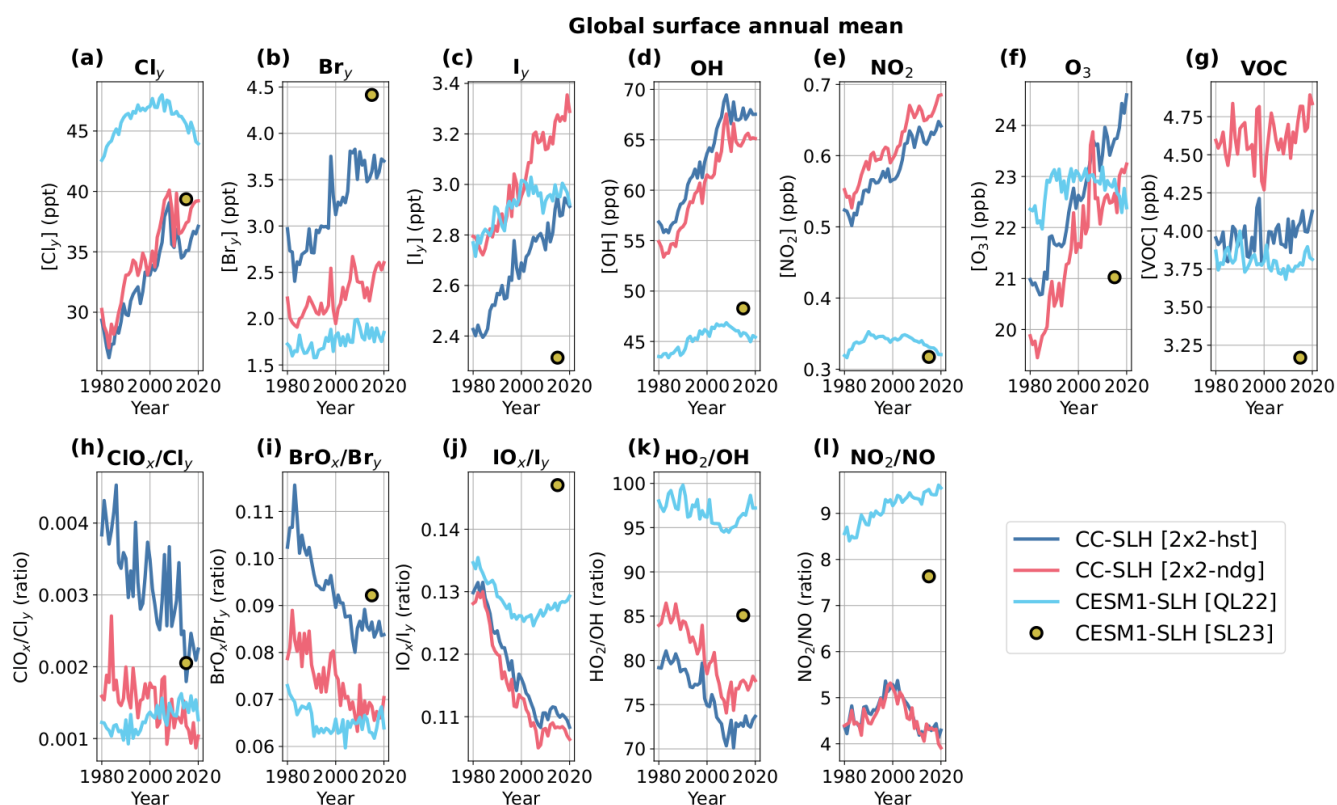


Figure 2: Historical evolution of global annual mean surface abundance and partitioning of main atmospheric components during 1980-2020. Top-row shows the total inorganic halogen surface abundance for a) Cl_y , b) Br_y and c) I_y , as well as the surface mixing ratio for d) OH , e) NO_2 , f) O_3 , and g) VOC (calculated as the sum of species: ISOP, MTERP, CH_3OH , C_2H_5OH , CH_2O , CH_3CHO , CH_3COOH , CH_3COCH_3 , $HCOOH$, C_2H_2 , C_2H_4 , C_2H_6 , C_3H_8 , C_3H_6 , BIGALK, BIGENE, MEK, TOLUENE, BENZENE, XYLENES). Bottom row shows the reactive (XO_x) to reservoir (X_y) surface ratio (i.e., XO_x/X_y) for h) chlorine, i) bromine and j) iodine, as well as the k) HO_2/OH and l) NO_2/NO mean ratio at the model surface. Results for the nudged CC-SLH [2x2-ndg] (pink) and free-running CC-SLH [2x2-hst] (blue) experiments obtained with the CESM2-SLH release are compared with those obtained in previous CESM1 studies (see Section 3.3).

Finally, we have rephrased the text to clarify that the CESM1 results shown here belongs to previously published work (see related response to **R2-C9.9** above).

L697-699: “In order to compare the current CESM2-SLH inorganic halogen abundances with previous studies, Fig. 2 also shows the *results obtained in previous studies (i.e., CESM1-SLH [QL22] and CESM1-SLH [SL23] corresponding to Li et al. (2022) and Saiz-Lopez et al. (2023), respectively).*”

R2-C10.3: Figure 3: The caption is too long and should not repeat the figure legend. Time ranges are not in accordance to journal style. Yellow, open markers are difficult to see. Another color could improve this. `compset_non` was not defined previously. Do you mean 'compset_noh' ?

ANSWER: Thanks for the careful reading. We have now corrected the latitudinal and time-period ranges in all figures and captions. We have now reduced the caption length to avoid repetition with the legend, and also modified all markers are clearly visible. Based on the reviewer suggestion, all figures and tables now point at the independent experiments shown (see related responses to **R2-C7.1** and **R2-C9.5** above).

R2-C10.4: Figure 4:

- The resolution of this figure is rather low. It should be improved.
- Panel (d) and (f) are misaligned compared to panel (a) and (c).
- Panel (c) does not contain any data as stated in the figure caption (“empty panel c”) and should be removed.
- The colormap is not colorblind-friendly and draws the attention towards light blue and yellow. It should be replaced. See also <https://www.geoscientific-model-development.net/submission.html#figurestable>.
- The uppermost tick and every second tick on the pressure-axis are unlabeled. With respect to the space restrictions on the pressure-axis, ticks should be labeled where possible, else removed.
- Subcaptions (a-f) are placed inside the axis frame, which is in contrast to the previous figures. You may consider placing them outside.
- The brackets around units should be round like in the previous figures.
- The colorbar labels indicate that *white* refers to values below zero. From the arrangement of the figure, I guess, *white* is supposed to indicate *invalid* values in panel (c). Usually this would be marked with a ◁, e.g., `extent='left'` in python, matplotlib or *gray* color. Removing panel (c) will probably resolve this issue.
- The zonal mean tropopause (not “mean model tropopause” as stated in the figure caption) displays a strange gap at around 30°S. You may consider plotting the zonal mean tropopause as a line not with markers.

R2-C10.5: Figure 5: In general, the same as Figure 4 applies. In addition:

- Panel (f) does not contain any data and should be removed.
- Panel (i) is identical to panel (c) as it is supposed to show the difference between (c) and (f). It should be removed.

- It might be better to use a sequential colormap for panels (g,h) that would highlight that the difference between the two experiments is shown here and that species concentrations in FCnudged_slh are always higher.
- In figure titles, FCnudged_NON is displayed. Did you mean FCnudged_noh?

ANSWER: Thanks for the careful reading and for the detailed suggestions. We have reproduced the original Figures 4 and 5 (as well as all equivalent figures) following the journal guidelines (i.e., using a color-blind sequential style, removing empty panels, moving sub-caption letters outside each plot frame, etc.). We also thanks for the suggestions regarding the tropopause definition. Note that all figures and tables now point at the independent experiments shown (see related responses to **R2-C7.1** and **R2-C9.5** above). Finally, note that due to the suggested reordering of sections in comment **R2-C1.4**, original Figures 4 and 5 have been shifted down to Figures 7 and 8.

R2-C10.6: Figure 6:

- Captions should not replace figure legends. You may include another legend entry displaying the assignment of each experiment to the open or filled markers.
- The subcaptions (a–c) are inconsistent with previously used placements and style.
- It is not clear, what a "mean height of the global tropopause" is. Do you mean 'global mean tropopause height' ?

R2-C10.8: Figure 8: Probably the most important figure of this manuscript. Although, it can also be improved.

- Observations are missing in the figure legend.
- "vmr" → 'VMR' or use $X_{Cl,Br,I}$. X is preferred by the measurement community.
- Volume mixing ratio (VMR) is not defined in the manuscript and referred to as "mole fractions" in the figure caption which is inconsistent.
- The expression "observed mean $\pm$ range mole fractions" is odd. Do you mean 'Average and range of halogen volume mixing ratios (VMR) compiled from observations in the tropical tropopause region'?
- ΔX_i is not mentioned in the legend. It is called PGs there. Hence, legend and caption are inconsistent.
- "mean $\pm$ range (computed as the standard deviation [...]) is shown by [...] coloured shading [...]". The "coloured shading" is usually referred to as 'error bands'. Why do you call the standard deviation "range"?
- The caption is too long, difficult to understand, and should not replace or repeat the main text.
- The figure caption could read: 'Vertical profiles of the tropical (20°N – 20°S) zonal mean, averaged over the time period 2015 – 2020 for experiments [...] compared to observations for (a) X_{Cl} , (b) X_{Br} , and (c) X_I .

ANSWER: Thanks for the careful reading and for the detailed suggestions. We have now reduced the caption length to avoid repetition with the legend, and moved sub-caption letters outside each plot frame. In addition, we have included additional markers in the legend to distinguish modeled and observational data. We have also rephrased the caption and made sure all acronyms and symbols are mentioned in the legend. Finally, we have corrected the definition of the tropopause to "global mean tropopause height) and explain what the shading represents.

Note that due to the suggested reordering of sections in comment **R2-C1.4**, we have now moved up the halogen validation to be the first subsection 4.2.1, and therefore, the original Figure 6 (global distribution) has been moved down to Figure 9, while original Figure 8 (tropical validation) has been shifted up to Figure 4. The revised figure and caption now reads as follows:

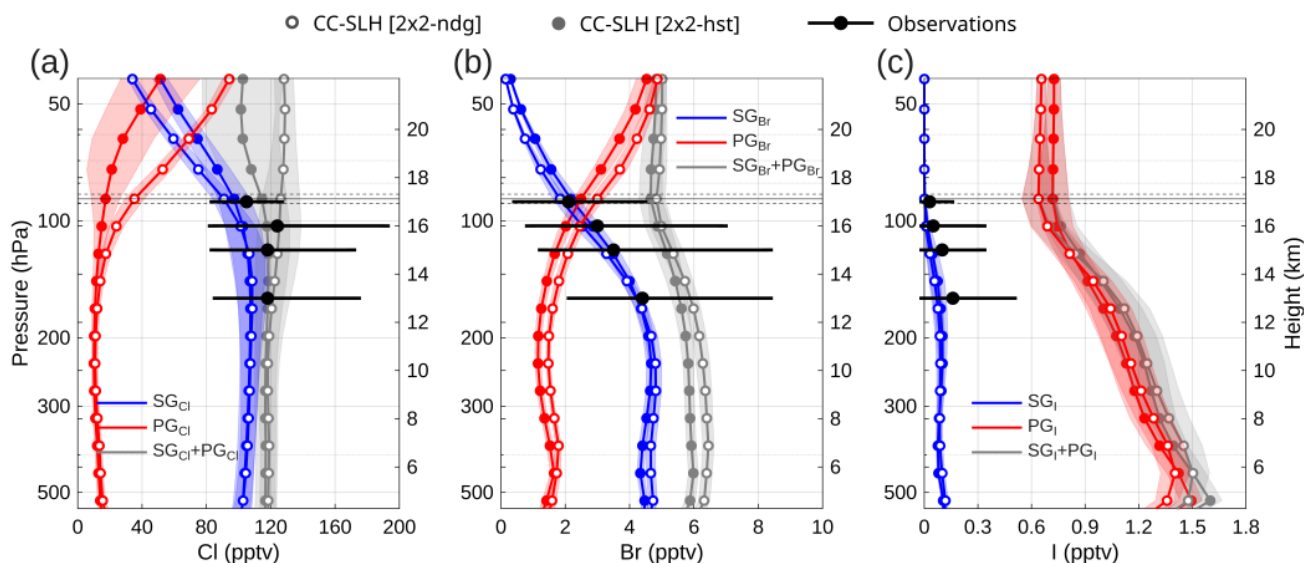


Figure 4: Tropical mean vertical profile of organic SLH source gases (SG_x) and product gases (PG_x) during present-day (2015–2020) for a) chlorine, b) bromine and c) iodine. The SGs profiles (blue lines) correspond to the nudged CC-SLH [2x2-ndg] (empty symbols) and the free-running CC-SLH [2x2-hst] (filled symbols) experiments, while PGs profiles (red lines) are obtained as the difference with respect to the corresponding CC-NOH sensitivity (see text for details). The total additional halogen abundance due to SLH ($SG_x + PG_x$) is shown in grey. The tropical (20°N – 20°S) mean and spread (i.e., $\pm$ the interannual standard deviation for the 2015–2020 period) are shown with solid lines and coloured shading, respectively. Average and range of observed VSL mixing ratios in the tropical tropopause layer compiled in Table 1-5 of WMO (2022) are shown in black symbols and solid-thick horizontal lines at 13, 15, 16 and 17 km, respectively. The solid and dashed horizontal lines indicate the tropical mean tropopause height $\pm$ standard deviation.

R2-C10.7: Figure 7: The line colors used are difficult to distinguish. The legends are overlapping with the data. The x-axis labels of panels (a,c,e) are not correct, as they display a flux. The flux units are rather unusual. Usual flux units are kg s^{-1} or molec s^{-1} is used. You may fix the units. The information contained in this figure may be supplementary material

ANSWER: Thanks for the careful reading and for the detailed suggestions. Flux panels are now shown in volumetric source units ($\text{molec. cm}^{-3} \text{ s}^{-1}$). We rather keep this figure in the main text and moved it to revised Figure 10.

R2-C10.9: Figure 9:

- The caption is too long and should be rephrased.
- Time axis label may be added.
- Units should be placed in round brackets.
- Shouldn't the tropopause height show a climate trend over this period?

ANSWER: Thanks for the careful reading and for the detailed suggestions. We have now reduced the caption length to avoid repetition with the legend, and corrected the temporal axis and units format. Note the solid and horizontal dashed lines do not represent the tropopause, but the mean and range of SGI values as estimated

in the latest WMO (2022) report. Note that due to the suggested reordering of sections in comment **R2-C1.4**, we have now moved up the halogen validation to be the first subsection 4.2.1, and therefore, the original Figure 9 has been shifted up to Figure 5.

R2-C10.10: Figure 10:

- The colormap is not colorblind friendly and highlight the regions early named.
- The colorbars are not labeled.
- Subplot captions are placed inside the axis frames and hardly visible.
- Colorbar ticks in subplot (g) should be formatted in exponential form ($\times 10^{-4}$)

ANSWER: Thanks for the careful reading and for the detailed suggestions. We have reproduced the original Figure 10 (as well as all equivalent figures) following the journal guidelines (i.e., using a color-blind sequential style, including colorbar labels, moving sub-caption letters outside each plot frame, etc.). We also used the exponential format whenever is needed. Following reviewer **R1** suggestion, we added a new validation figure for inorganic halogens (Figure 6), and therefore in the revised manuscript original Figure 10 has been renumbered to Figure 11.

R2-C10.11: Figure 11: The hatches are almost invisible. You may choose different hues or shades of red and blue instead. "Empty" is an odd expression for unhatched bars.

ANSWER: Thanks for the careful reading and for the detailed suggestions. To make sure all hatched and unhatched are clearly distinguished, we have modify the colors and format and shifted to a logarithmic scale highlighting the smaller values. Following reviewer **R1** suggestion, we added a new validation figure for inorganic halogens (Figure 6), and therefore in the revised manuscript original Figure 11 has been renumbered to Figure 12.

R2-C10.12: Figure 12: Some units are not placed in parentheses. Labels are hardly readable due to the low resolution picture. You may consider plotting the zonal mean tropopause height as line not with markers. The color range of subfigure (c,f) is saturated in large parts of the Northern Hemisphere. Upper and lower bounds should be adapted.

R2-C10.13: Figure 13: Same as Figure 12.

ANSWER: Thanks for the careful reading and for the detailed suggestions. We have adapted the ranges of the individual color-scales to avoid saturation and use a solid line for the tropopause. Following reviewer **R1** suggestion, we added a new validation figure for inorganic halogens (Figure 6), and therefore in the revised manuscript original Figures 11 and 12 have been renumbered to Figures 12 and 13, respectively.

R2-C10.14: Figure 14: Albeit the model experiments are only evaluated for one year (2015) a monthly mean should be accompanied by a standard deviation or standard error. It remains unclear what the error bars in the TOAR observations are. If the TOAR data represent climatologies (which years?), aka multi-annual means for each month, then these would represent the inter-annual variability. This should be clarified here and in the main text. Usually the annual averages displayed here would be separated from the monthly means by a vertical line. Is "NON" in the legend the same as model experiment *FCnudged_noh*? Should it read 'NOH' ?

ANSWER: Thanks for highlighting the importance of including the modeled standard deviation of monthly mean values. Following the reviewer suggestions, we repeated the CC-SLH [2×2-ndg] and CC-NOH [2×2-ndg] experiments for year 2015 and extracted the surface ozone abundance with hourly resolution, which allowed to compare our modelling results with the same temporal resolution as the TOAR dataset. The revised validation figure now shows the mean $\pm$ range (computed as the standard deviation of hourly values) for modelled (shading) and observations (black bars). Most notably, the inclusion of SLH chemistry results in an overall reduction of ozone that reduces the model bias and in most locations is larger than the spread of modelled values between the CC-SLH and CC-NOH simulations. Note that all figures and tables now point at the independent experiments shown (see related responses to **R2-C7.1** and **R2-C9.5** above). The revised ozone validation figure and caption reads as follows:

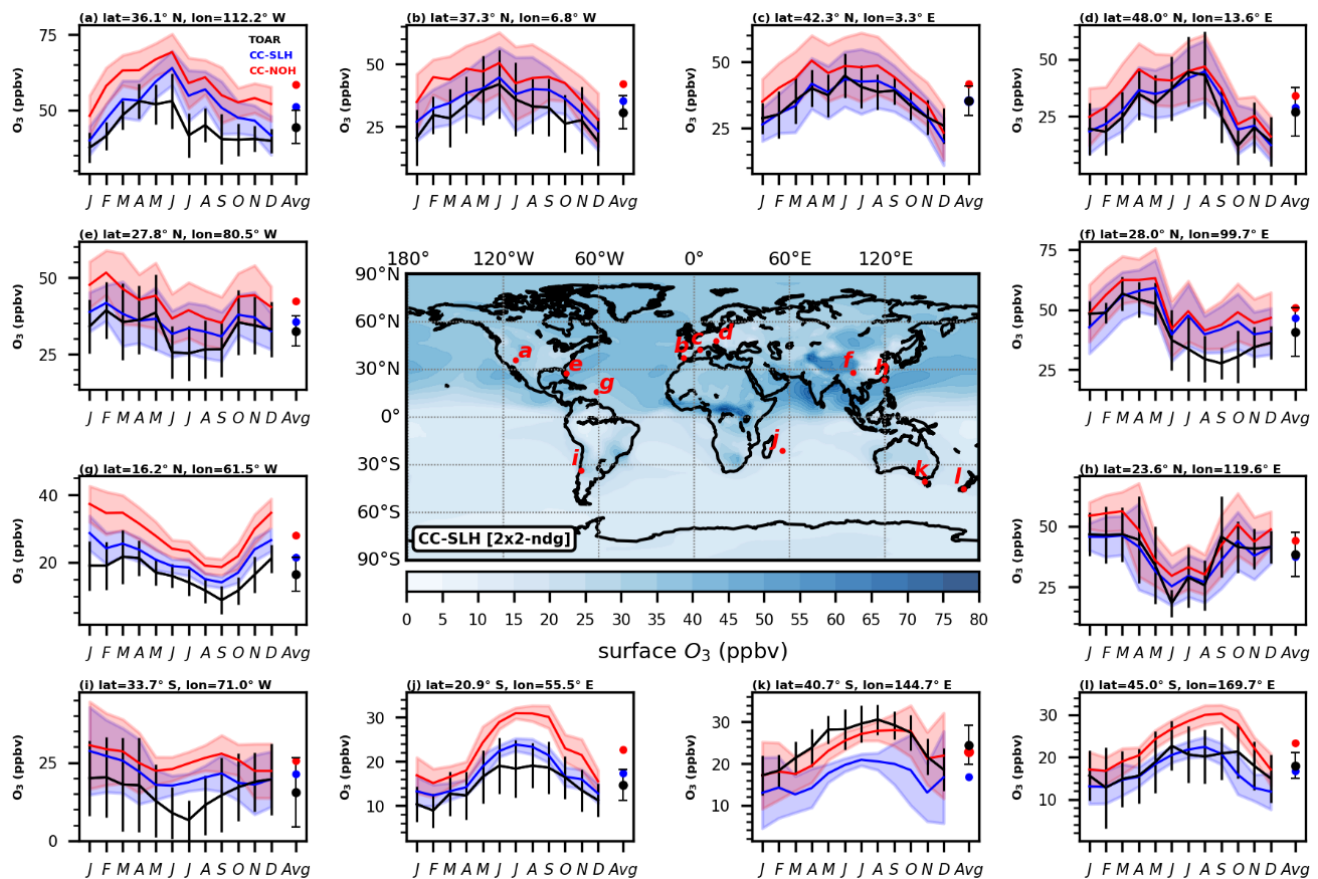


Figure 15: Comparison of CESM2-SLH nudged CC-SLH [2×2-ndg] and CC-NOH [2×2-ndg] simulations with TOAR-I observations. The center panel shows the geographical distribution of surface ozone for the CC-SLH [2×2-ndg] experiment, as well as the location of each of the observational sites shown in the side panels (red points). The side panels (a-l) show the monthly mean $\pm$ range seasonality (solid lines and shading) and the annual average (separate marker) of surface ozone for CC-SLH [2×2-ndg] (blue) and CC-NOH [2×2-ndg] (red) experiments. Black lines and error bars show surface observations reported in the first phase of Tropospheric Ozone Assessment Report (TOAR-I) (Schultz et al., 2017). Both model output and observations ranges have been computed as the standard deviation of hourly data for year 2015.

R2-C10.15: Figure 15: Axis labels and tick labels are too small. The colormap is drawing the attention to the yellow region and hence highlights features that might not be there. A different (sequential) colormap should be chosen. The range of the colormap is not the same for subfigures (a–c) and (d–f).

R2-C10.16: Figure 16: Yet another variant of units not placed on round brackets. Labels and tick labels are too small. Line colors are hardly visible or distinguishable. For subfigure (a–c), the same as for previous

global averages applies – polar stratosphere and tropical troposphere are lumped together. Global averages should be made in vertical coordinate system relative to the tropopause.

ANSWER: Thanks for the careful reading and for the detailed suggestions. We have increase the size of all ticks and labels in all panels, corrected the format in *x*-axis magnitudes and units, and modified the color-palette to a sequential style. Please refer to the response to **R2-C3.6-7** above for a detailed response to the reviewers concerns about showing a global mean vertical profiles. Note we have now added horizontal solid and dashed lines representing the mean $\pm$ range of values for the tropopause, which clear show a much larger spread for the global mean (bottom-panels) than for the tropical average (top-panels). Following reviewer **R1** suggestion, we added a new validation figure for inorganic halogens (Figure 6), and therefore in the revised manuscript original Figures 15 and 16 have been renumbered to Figures 16 and 17, respectively.

R2-C10.17: Figure 17: "°lat $\rightarrow$ Latitude (°)"

ANSWER: Done. Note that this figure has been renumbered to Figure 18.

R2-C10.18: Figure 18: Same as for similar plots.

R2-C10.20: Figure 20: Same as for similar plots: colormap and missing colorbar labeling.

ANSWER: We have applied the same format changes as those described in the response to **R2-C10.4-5** and **R2-C10.10** above. Note that due to the major manuscript rearrangements applied to include the new Discussion Section 5, we decided to move original Figures 18 and 20 to the Supplementary Material Figures S7 and S9.

R2-C10.19: Figure 19: (a–c), same as for similar plots regarding the vertical coordinates and globally averaged tropopause and boundary layer.

ANSWER: Thanks for the careful reading and for the detailed suggestions. Please refer to the response to **R2-C3.6-7** above for a detailed response to the reviewers concerns about showing a global mean vertical profiles. Note we have now moved up/down the bottom/top panels to show global and tropical means in the same order as in the old Figure 16 (current Figure 17, see **R2-C10.16**). In addition, we now added horizontal solid and dashed lines representing the mean $\pm$ range of values for the tropopause, which clear show a much larger spread for the global mean (bottom-panels) than for the tropical average (top-panels).

REFERENCES:

1. Burkholder, J. B., Sander, S. P., Abbatt, J., Barker, J. R., Cappa, C., Crounse, J. D., Dibble, T. S., Huie, R. E., Kolb, C. E., and Kurylo, M. J. (2020). Chemical Kinetics and Photo-chemical Data for Use in Atmospheric Studies, Evaluation No. 19 (JPL Publication 19-5). Jet Propulsion Laboratory, <https://www.google.com/url?sa=t&source=web&rct=j&opi=89978449&url=https://jpldataeval.jpl.nasa.gov/pdf/NASA-JPL%2520Evaluation%252019-5.pdf&ved=2ahUKEwistr4TPsLCRAxV-BdsEHXGNNWYQFnoECBAQAQ&usg=AOvVaw23Z7bVlzdNIUSCUh5CkFcG>.

ANSWER: Thanks for the careful reading. We have corrected the year of JPL 19-5 Publication to 2020.

L1705-1708: “Burkholder, J. B., Sander, S. P., Abbatt, J. P. D., Barker, J. R., Cappa, C., Crounse, J. D., Dibble, T. S., Huie, R. E., Kolb, C. E., Kurylo, M., Orkin, V., Percival, C., Wilmouth, D. and PH, W.: *Chemical Kinetics and Photochemical Data for Use in Atmospheric Studies Evaluation Number 19 - Publication 19-5, JPL_NASA*, 32(15–10), 170 [online] Available from: <https://jpldataeval.jpl.nasa.gov/>, **2020.**”