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 It fits well into the scope of GMD and I see no reason why it should not be published after addressing the following minor comments.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 adaptions 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, bit 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"***, were 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*	*(1.9º × 2.5º)*	*1980 - 2020*
CC-SLH [2×2-hst]	*FCHIST_slh*	*(1.9º × 2.5º)*	*1950 - 2020*
CC-SLH [1×1-ndg]	*FCnudged_slh*	*(0.9º × 1.25º)*	*2000-2005*
CC-SLH [1×1-hst]	*FCHIST_slh*	*(0.9º × 1.25º)*	*2000-2005*
FW-SLH [2×2-ndg]	*FWnudged_slh*	*(1.9º × 2.5º)*	*2000-2005*
BW-SLH [2×2-cpl]	*BWHIST_slh*	*(1.9º × 2.5º)*	*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-namelists, 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 1.23–1.27×10^6 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±1.0×10^5 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.”***

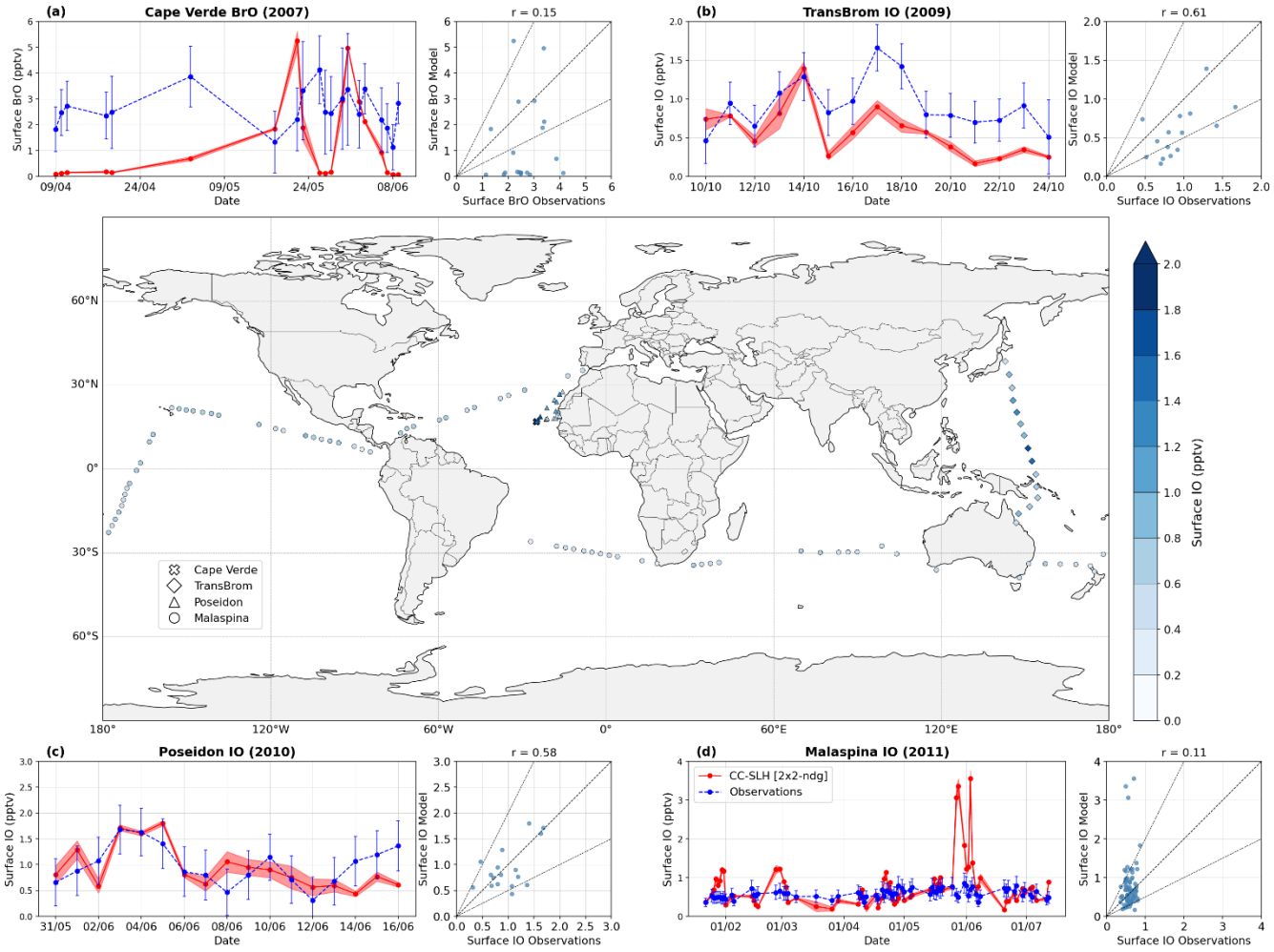

Figure 6: Comparison of nudged CC-SLH [2×2-ndg] surface mixing ratios with IO and BrO observations from multiple campaigns. The central panel shows the location of the Cape Verde station which measured BrO in 2007 (cross) and the routes of three ship-based IO campaigns: TransBrom in 2009 (diamond), Poseidon 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) Caper Verde; (b) TransBrom; (c) Poseidon and (d) Malaspina. All data corresponds to daytime conditions.

In addition, 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. 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 ± 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° 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 Zeeland. 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.”***

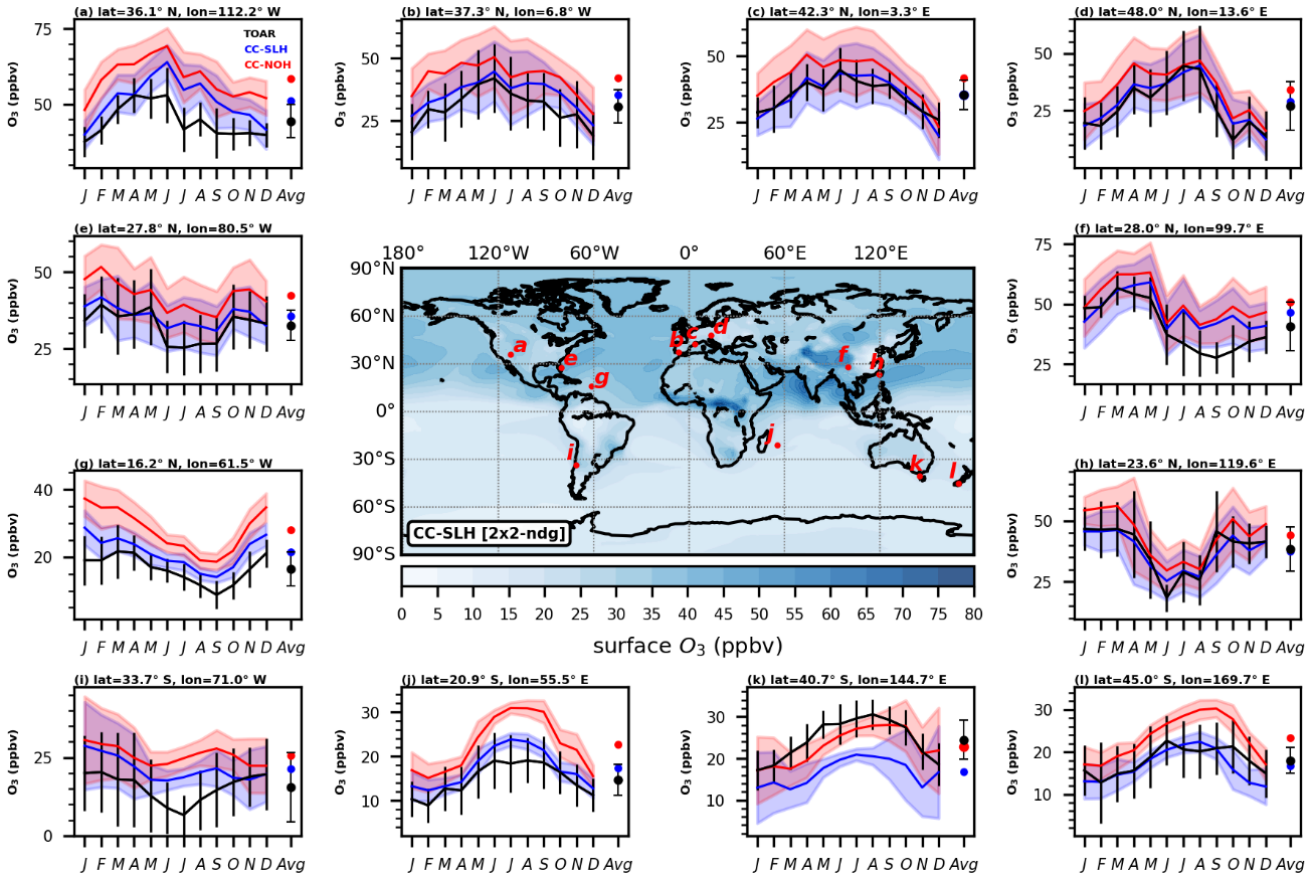

Figure 15: Comparison of CESM2-SLH nudged CC-SLH [2×2-ndg] and CC-NOH [2×2-ndg] simulations with TOAR-I observations. The center panel shows the geographical distribution of surface ozone for the CC-SLH [2×2-ndg] experiment, as well as the location of each of the observational sites shown in the side panels (red points). The side panels (a-l) show the monthly mean ± 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.

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 $C_2H_4Cl_2$).*** *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 $yield^{ClNO2} = \left(\frac{SSA_{mass}-SSA_{min}}{SSA_{max}-SSA_{min}}\right)^3$, where SSA_{mass} is the total sea-salt aerosol mass mixing ratio of all aerosol bins in each model gridbox, while $SSA_{min} = 5.0\times10^{-10}$ and $SSA_{max} = 1.0\times10^{-9}$ are the minimum and maximum SSA_{mass} adjusted thresholds for $N_2O_5 \rightarrow 2\cdot HNO_3$ 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 that the initial condensation of gas-phase HCl must occurs 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 O_3, OH, NO_2 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* ***NO_2 and*** *O_3 (Figure 2****e****,****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 HNO_3 and N_2O_5 abundance as well as on the extent of partitioning of halogen reservoirs to $ClONO_2$ and $BrONO_2$, 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 NOx–HOx 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 O_3 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 $1.23–1.27\times10^6$ 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 ($11.7\pm1.0\times10^5$ 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: *"[$]**The 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. **17**b), the total contribution of halogens increases significantly and represents between 10% and 30% globally (Fig. **17**c), 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 **$OddOx_{Loss}$** 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 VSLS (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), $\left(\sqrt{\frac{8\times R}{\pi\times MW_{ox}}}\right)$, where temperature has been excluded from the square-root and is later multiplied by an independent $\left(\sqrt{T}\right)$ 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 O_3, OH, and NO_2 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 PO_3 – LO_3, drydep, and O_3 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.