Supplementary Information:

# Reactive halogens increase the global methane lifetime and radiative forcing in the 21st century

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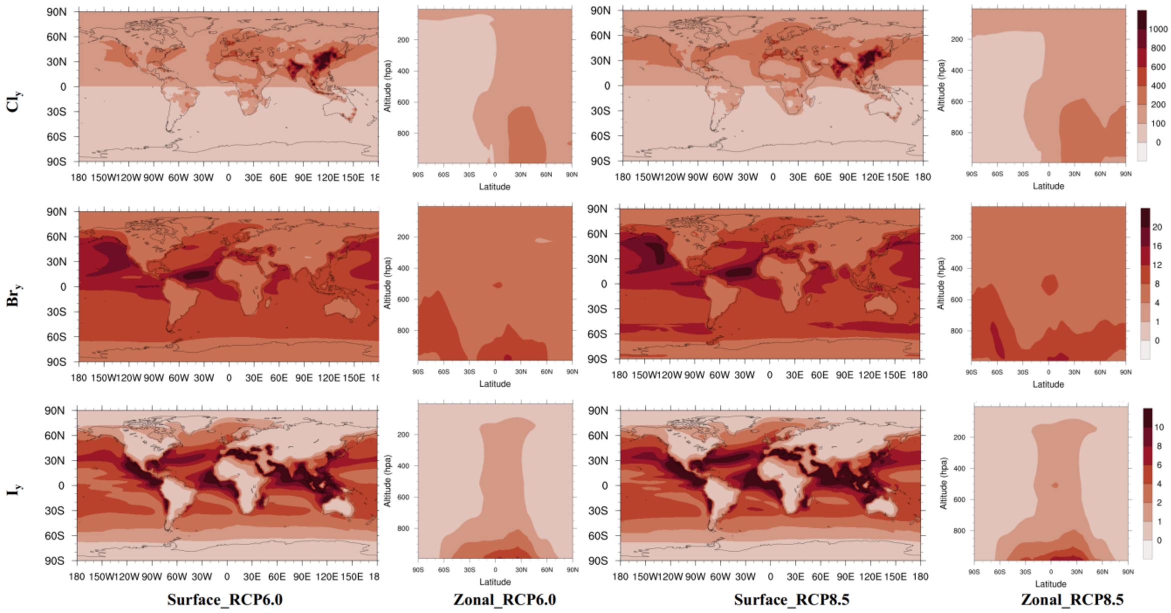
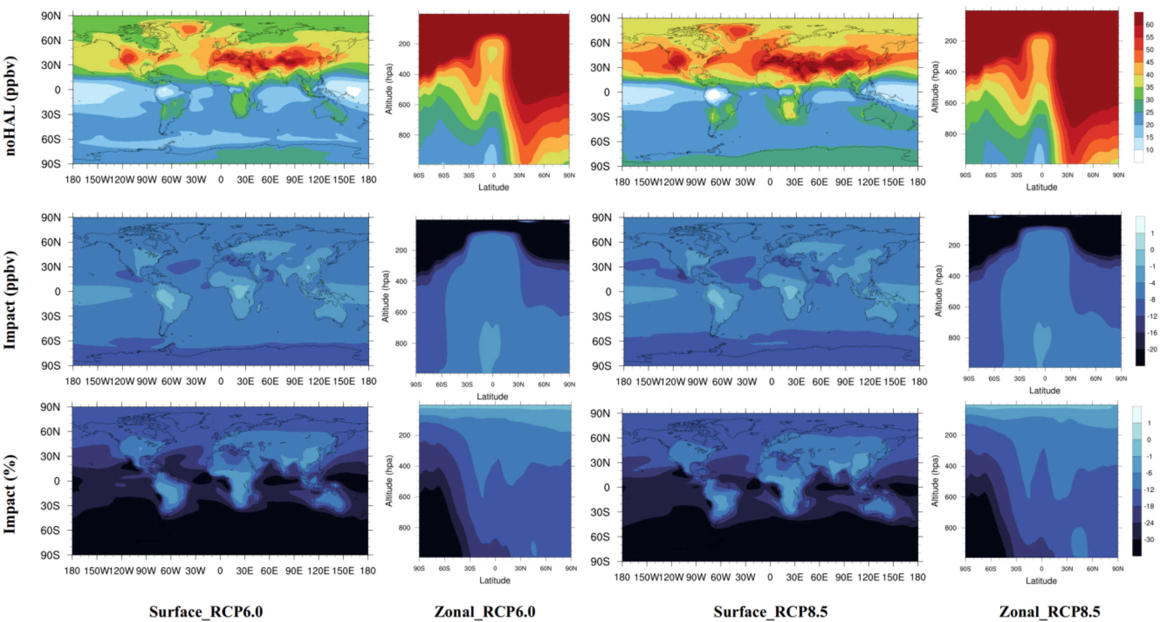
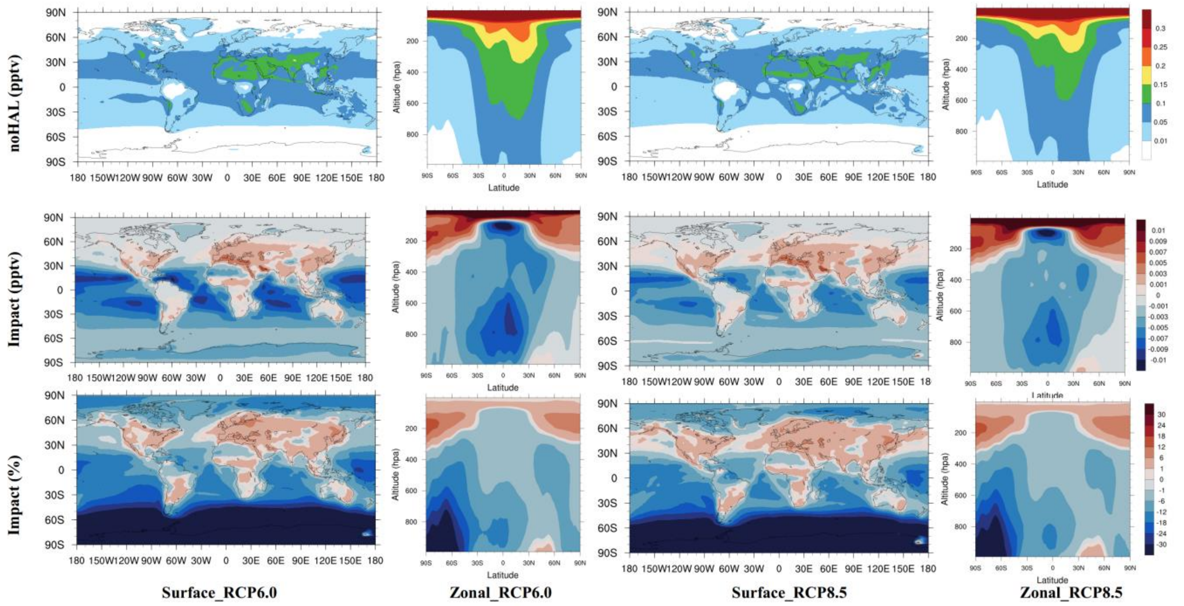
Contents:



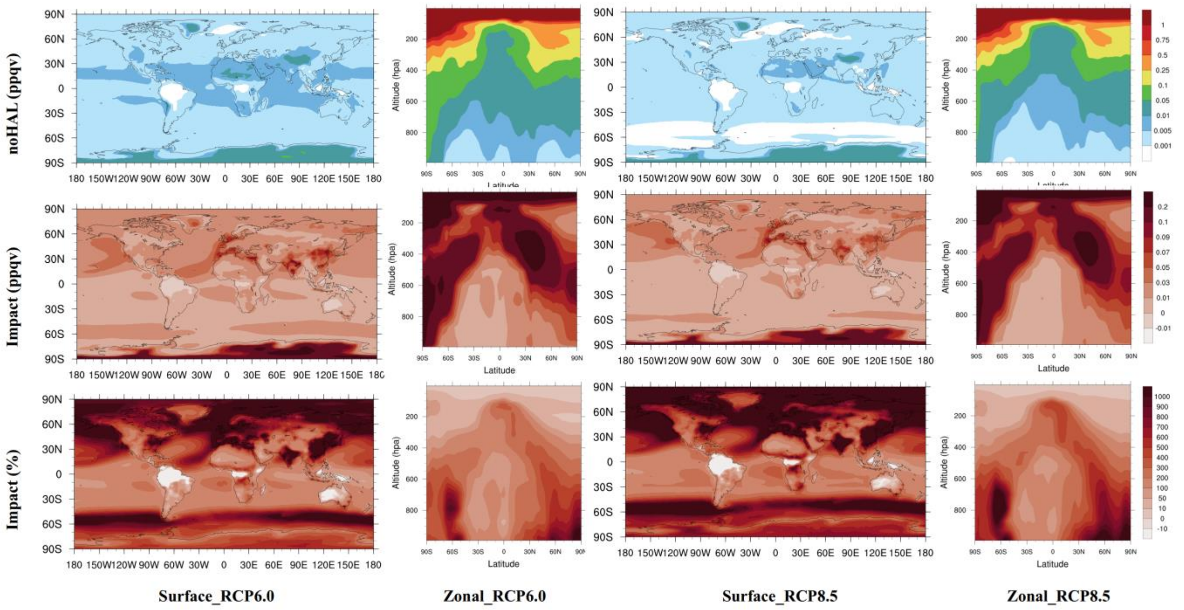

**Supplementary Figure 1 | Simulated changes (difference between HAL and noHAL scenarios) in RHS ($Cl_y$, $Br_y$, and $I_y$) mixing ratios (pptv) for RCP6.0 (left) and RCP8.5 (right) scenarios in the 21st Century.** Both surface and zonal average results are shown. The marked difference in $Cl_y$ between the northern and southern hemispheres is because the source of $Cl_y$ is dominated by anthropogenic emissions which are much higher in the northern hemisphere (particularly in the northern mid-latitude land) than in the southern hemisphere. The increase of bromine and iodine species is confined particularly in the tropical lower troposphere.



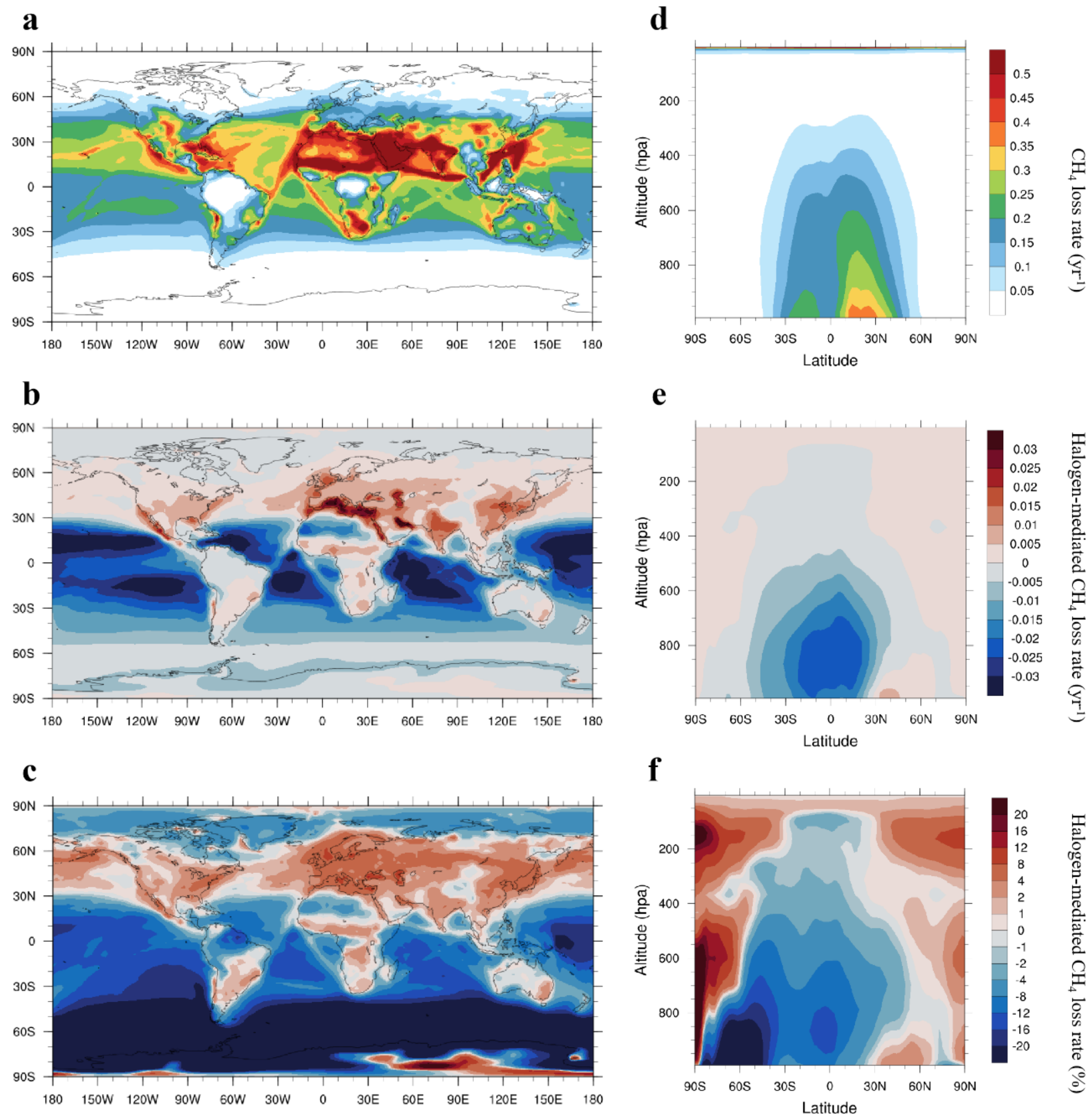

**Supplementary Figure 2 | Predicted $O_3$ mixing ratios (ppbv) in noHAL case and their changes (ppbv and %) due to halogens for RCP6.0 (left) and RCP8.5 (right) scenarios in the 21$^{st}$ Century.** Both surface and zonal average results are shown.



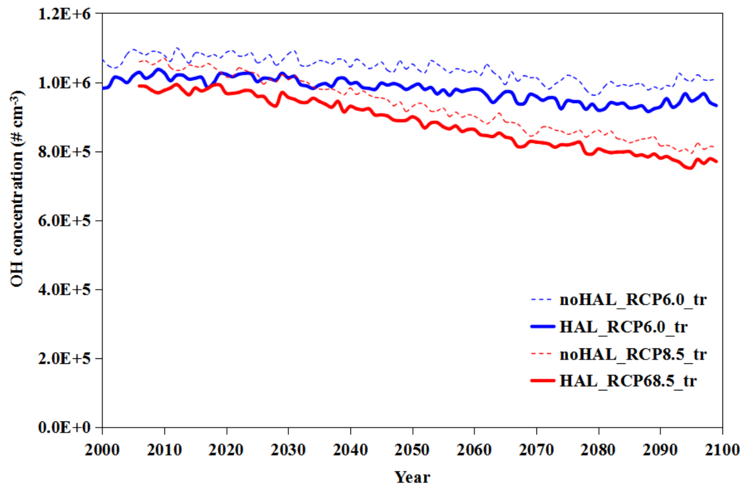

**Supplementary Figure 3 | Predicted OH radical mixing ratios (pptv) in noHAL case and their changes (pptv and %) due to halogens for RCP6.0 (left) and RCP8.5 (right) scenarios in the 21st Century.** Both surface and zonal average results are shown.



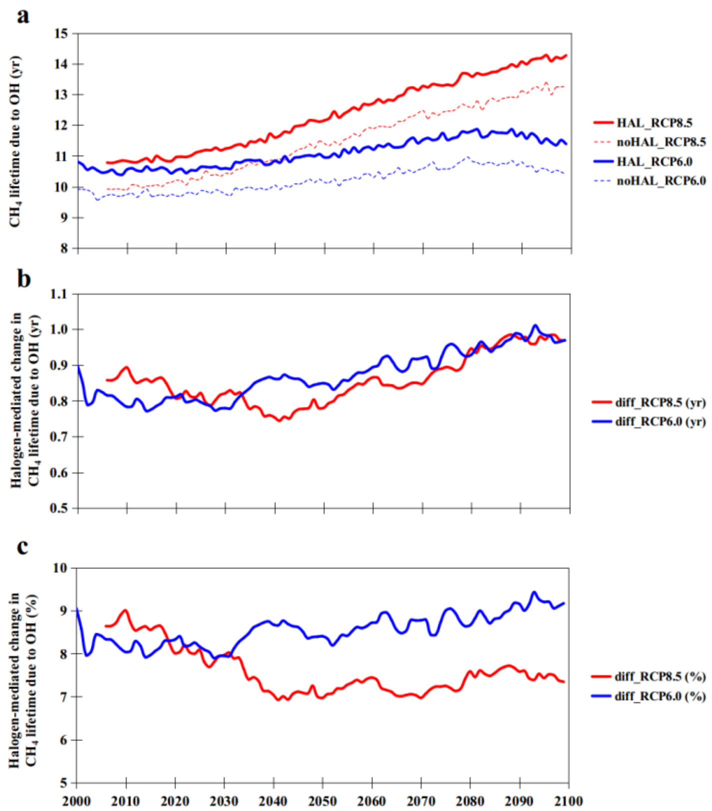

**Supplementary Figure 4 | Predicted Cl atom mixing ratios (ppqv) in noHAL case and their changes (ppqv and %) due to halogens for RCP6.0 and RCP8.5 scenarios in the 21st Century.** Both surface and zonal average results are shown.



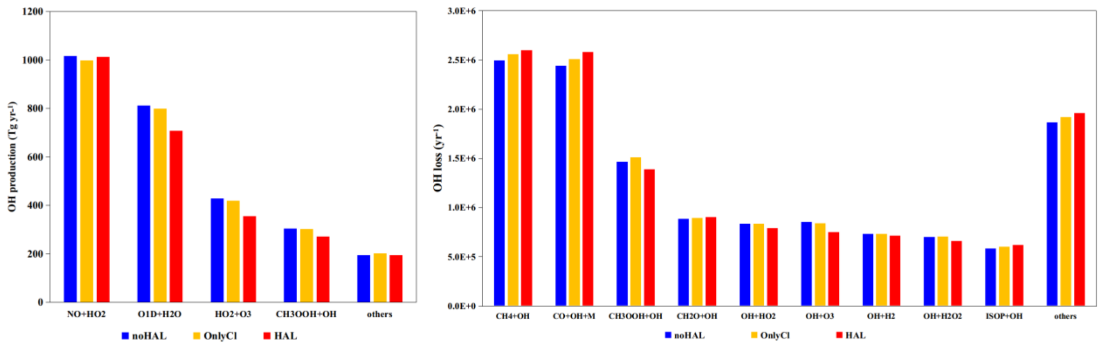

**Supplementary Figure 5 | Spatial patterns of halogen-mediated change in $CH_4$ loss rate averaged during the 21st century for the RCP8.5 scenario.** **a** surface $CH_4$ loss ($yr^{-1}$) without halogens (noHAL); **b** Halogen-mediated changes in $CH_4$ loss ($yr^{-1}$) at the surface; **c** same as **b** but in percentage; **d** Zonal distribution of $CH_4$ loss ($yr^{-1}$) without halogens (noHAL) showing the largest $CH_4$ loss near-surface level in tropics; **e** Zonal distribution of halogen-mediated change in $CH_4$ loss ($yr^{-1}$); **f** same as **e** but in percentage. Results for the RCP6.0 scenario are shown in Figure 2.



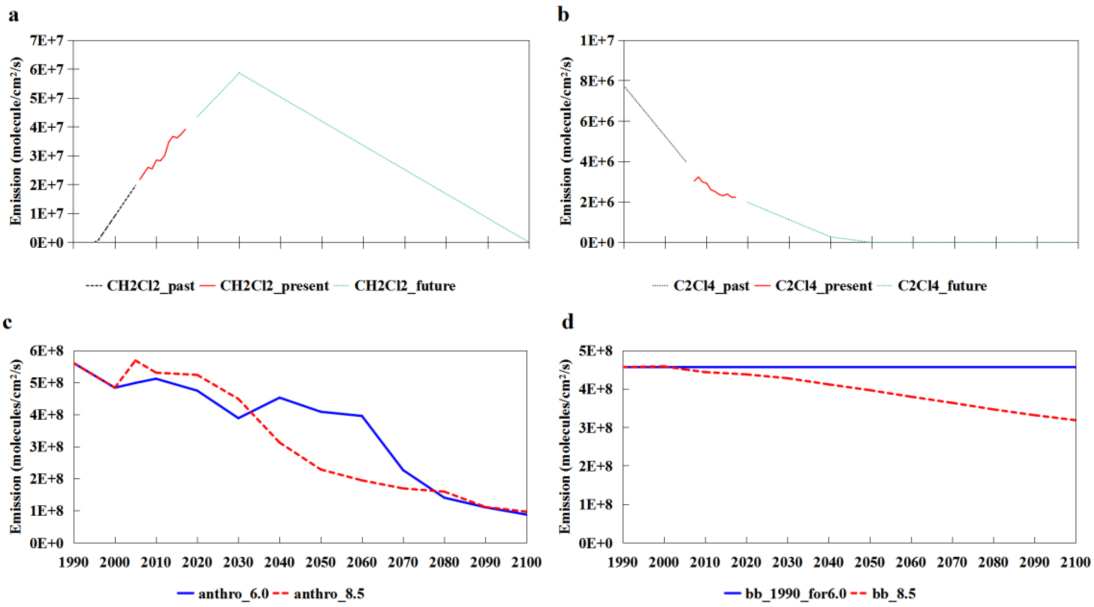

**Supplementary Figure 6 |** The variation of global tropospheric OH concentration (# $cm^{-3}$) in noHAL and HAL cases in both RCP6.0 and RCP8.5 scenarios in the 21st century.



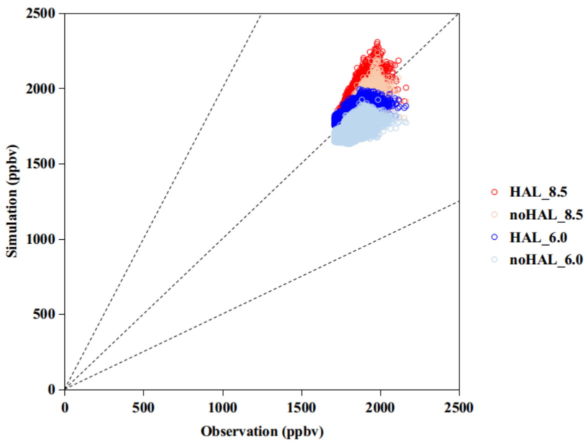

**Supplementary Figure 7 | Halogen-mediated changes in $CH_4$ lifetime with respect to OH in the 21st century for RCP6.0 and RCP8.5 scenarios**. **a** Global $CH_4$ chemical lifetime (yr) with (HAL, solid) and without (noHAL, dashed) halogens; **b** halogen-mediated change in $CH_4$ lifetime in absolute term (yr); **c** the same as **b** but in percentage. Note that the halogen-mediated changes are calculated with a moving average of 10 years (approximately the lifetime of $CH_4$ in the atmosphere). The total $CH_4$ chemical lifetime is shown in Figure 4. Note that the results for RCP8.5 are only shown from 2006 to 2100 and the results before 2006 are identical to those in RCP6.0.



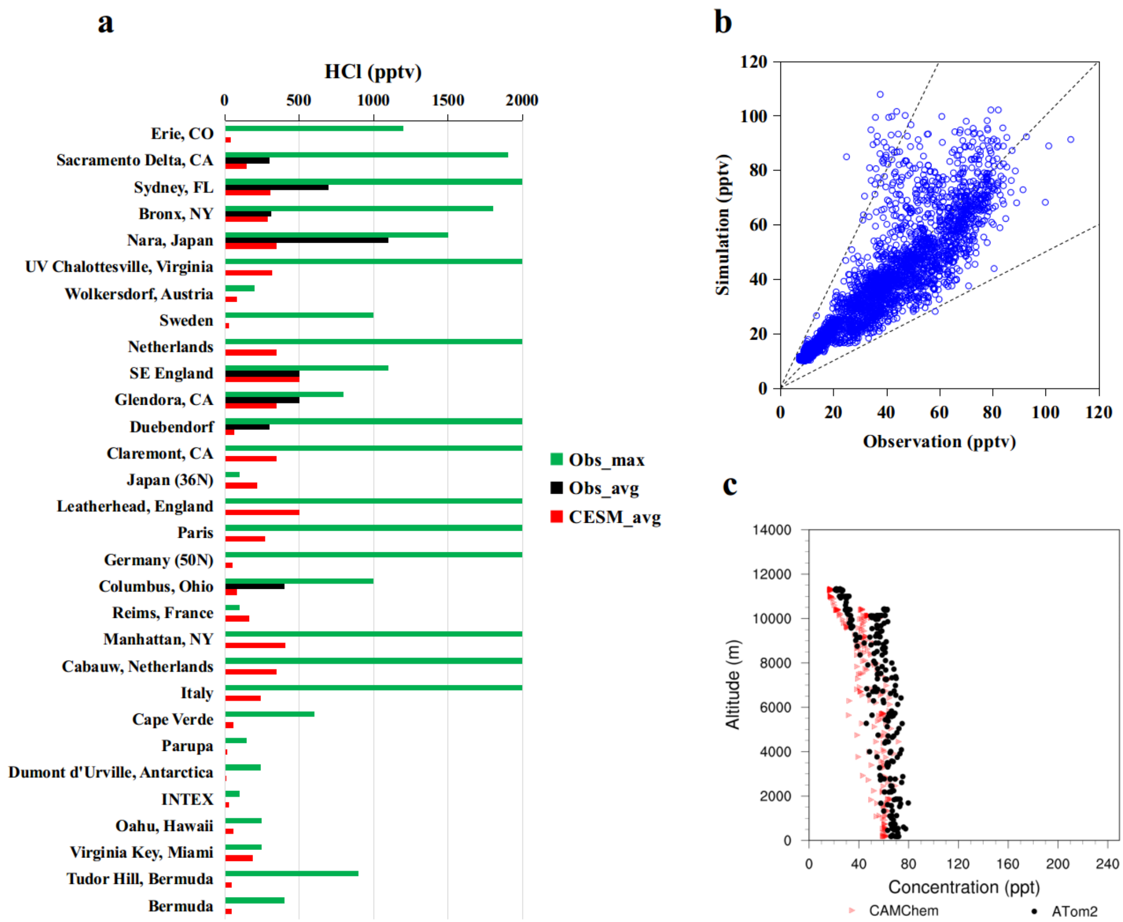

**Supplementary Figure 8** | Budget analysis of global OH production (left, Tg $yr^{-1}$) and loss (right, $yr^{-1}$) in noHAL, OnlyCl, and HAL cases averaged in the entire 21st century. Note that the OH loss is normalized with its burden.




**Supplementary Figure 9** | The emission trend of (**a**) anthropogenic $CH_2Cl_2$, (**b**) anthropogenic $C_2Cl_4$, (**c**) anthropogenic HCl, and (**d**) biomass burning HCl used in the present study.




**Supplementary Figure 10** | Comparison of simulated surface $CH_4$ volume mixing ratio with global surface observations from NOAA. The dotted lines represent 1:2, 1:1, and 2:1 ratios.




**Supplementary Figure 11 |** (a) Comparison of simulated HCl with surface observation, (**b**) comparison of simulated $CH_2Cl_2$ and surface observation (the dotted lines are 1:2, 1:1, and 2:1 ratios), and (**c**) comparison of simulated $CH_2Cl_2$ and flight observation during ATom campaign. Note that the observed maximum of HCl at the following sites are larger than 2000 pptv: Sydney, FL (5600 pptv), UV Chalottesville, Virginia (2800 pptv), Netherlands (3000 pptv), Leatherhead, England (3800 pptv), Paris (5000 pptv), Manhattan, NY (9000 pptv), Italy (2500 pptv).

**Supplementary Table 1** | Design of simulation scenarios

| Scenario | Routine emissions | Natural VSL | Anthropogenic HCl | Biomass burning HCl | $CH_2Cl_2$ | Other organic chlorine |
|---|---|---|---|---|---|---|
| **noHAL_6.0** | RCP6.0 | - | - | - | - | - |
| **HAL_6.0** | RCP6.0 | Iglesias-Suarez et al. (2020) | Scaling with anthropogenic $SO_2$ in RCP6.0 | Fixed from 1960 to 2100 using Keene et al., 1999 (RCEI) | Variation from 1960 to 2030; Decrease afterwards | Variation from 1960 to 2030; Decrease to natural value by 2100. |
| **OnlyCl_6.0** | RCP6.0 | Chlorine species in Iglesias-Suarez et al. (2020) | Scaling with anthropogenic $SO_2$ in RCP6.0 | Fixed from 1960 to 2100 using Keene et al., 1999 (RCEI) | Variation from 1960 to 2030; Decrease afterwards | Variation from 1960 to 2030; Decrease to natural value by 2100. |
| **noHAL_8.5** | RCP8.5 | - | - | - | - | - |
| **HAL_8.5** | RCP8.5 | Iglesias-Suarez et al. (2020) | Scaling with anthropogenic $SO_2$ in RCP8.5 | Scaling with biomass burning CO in RCP8.5 | Variation from 1960 to 2030; Decrease afterwards | Variation from 1960 to 2030; Decrease to natural value by 2100. |

Note: the description of the model design is in Methods.

**Supplementary Table 2 |** Statistics of global $CH_4$ burden, source, and sink at present, in the future, and in the 21[st] century.

| | **Burden (Tg)** | | | | **Source (Tg/yr)** | | | | | | **Sink (Tg/yr)** | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | **RCP6.0** | | **RCP8.5** | | **RCP6.0** | | | **RCP8.5** | | | **RCP6.0** | | | | | | **RCP8.5** | | | | | |
| | | | | | | | | | | | **noHAL** | | | **HAL** | | | **noHAL** | | | **HAL** | | |
| | **noHAL [a]** | **HAL [b]** | **noHAL [c]** | **HAL [d]** | **ANT [e]** | **BB [f]** | **NAT [g]** | **ANT [h]** | **BB [i]** | **NAT [j]** | **OH [k]** | **Cl [l]** | **Str [m]** | **OH [n]** | **Cl [o]** | **Str [p]** | **OH [q]** | **Cl [r]** | **Str [s]** | **OH [t]** | **Cl [u]** | **Str [v]** |
| **Present (2000-2019) [w]** | 4714.7 | 5000.6 | 4994.9 | 5299.2 | 280.8 | 25.7 | 191.0 | 331.8 | 24.7 | 191.0 | 483.5 | 2.7 | 27.1 | 473.9 | 9.6 | 29.0 | 499.4 | 2.7 | 28.1 | 488.8 | 9.9 | 32.3 |
| **Future (2080-2099) [x]** | 5116.0 | 5509.0 | 11771.2 | 12423.6 | 250.3 | 30.1 | 191.0 | 837.0 | 16.4 | 191.0 | 479.7 | 1.9 | 26.3 | 473.1 | 5.7 | 28.5 | 906.3 | 1.8 | 62.6 | 889.7 | 8.0 | 66.7 |
| **21[st] Century [y]** | 4971.9 | 5302.8 | 8381.9 | 8825.9 | 287.4 | 27.4 | 191.0 | 582.3 | 21.0 | 191.0 | 487.3 | 2.3 | 26.8 | 478.6 | 7.9 | 28.8 | 714.3 | 2.2 | 45.0 | 700.0 | 9.3 | 47.8 |

[a] Global $CH_4$ burden (Tg/yr) in the RCP6.0 scenario in the noHAL case;
[b] Global $CH_4$ burden (Tg/yr) in the RCP6.0 scenario in the HAL case;
[c] Global $CH_4$ burden (Tg/yr) in the RCP8.5 scenario in the noHAL case;
[d] Global $CH_4$ burden (Tg/yr) in the RCP8.5 scenario in the HAL case;
[e] Global $CH_4$ source (Tg/yr) in the RCP6.0 scenario from anthropogenic emission;
[f] Global $CH_4$ source (Tg/yr) in the RCP6.0 scenario from biomass burning emission;
[g] Global $CH_4$ source (Tg/yr) in the RCP6.0 scenario from natural emission;
[h] Global $CH_4$ source (Tg/yr) in the RCP8.5 scenario from anthropogenic emission;
[i] Global $CH_4$ source (Tg/yr) in the RCP8.5 scenario from biomass burning emission;
[j] Global $CH_4$ source (Tg/yr) in the RCP8.5 scenario from natural emission;
[k] Global $CH_4$ sink (Tg/yr) in the RCP6.0 scenario in the noHAL case via tropospheric OH;
[l] Global $CH_4$ sink (Tg/yr) in the RCP6.0 scenario in the noHAL case via tropospheric Cl;
[m] Global $CH_4$ sink (Tg/yr) in the RCP6.0 scenario in the noHAL case via stratospheric processes, including stratospheric OH, Cl, and $O^1D$;
[n] Global $CH_4$ sink (Tg/yr) in the RCP6.0 scenario in the HAL case via tropospheric OH;
[o] Global $CH_4$ sink (Tg/yr) in the RCP6.0 scenario in the HAL case via tropospheric Cl;
[p] Global $CH_4$ sink(Tg/yr) in the RCP6.0 scenario in the HAL case via stratospheric processes, including stratospheric OH, Cl, and $O^1D$;
[q] Global $CH_4$ sink (Tg/yr) in the RCP8.5 scenario in the noHAL case via tropospheric OH;
[r] Global $CH_4$ sink (Tg/yr) in the RCP8.5 scenario in the noHAL case via tropospheric Cl;
[s] Global $CH_4$ sink (Tg/yr) in the RCP8.5 scenario in the noHAL case via stratospheric processes, including stratospheric OH, Cl, and $O^1D$;
[t] Global $CH_4$ sink (Tg/yr) in the RCP8.5 scenario in the HAL case via tropospheric OH;
[u] Global $CH_4$ sink (Tg/yr) in the RCP8.5 scenario in the HAL case via tropospheric Cl;
[v] Global $CH_4$ sink (Tg/yr) in the RCP8.5 scenario in the HAL case via stratospheric processes, including stratospheric OH, Cl, and $O^1D$;
[w] Values averaged for the present time (2000 to 2019);
[x] Values averaged for the future (2080 to 2099);
[y] Values averaged for the 21[st] century (2000 to 2099);

**Supplementary Table 3**. Global methane burden (Tg) in the year 2000 in the sensitivity cases with (HAL) and without (noHAL) halogens applying different spin-up periods (10, 20, 30, and 40 years).

| | $CH_4$ burden | difference | | $CH_4$ burden | difference |
|---|---|---|---|---|---|
| noHAL_10 | 4114 | - | HAL_10 | 4244 | - |
| noHAL_20 | 4576 | 11.2% [a] | HAL_20 | 4797 | 13.0% [d] |
| noHAL_30 | 4769 | 4.2% [b] | HAL_30 | 5028 | 4.8% [e] |
| noHAL_40 | 4840 | 1.5% [c] | HAL_40 | 5116 | 1.8% [f] |

[a] The difference is calculated as (nohal_20 - nohal_10)×100/nohal_10.
[b] The difference is calculated as (nohal_30 - nohal_20)×100/nohal_20.
[c] The difference is calculated as (nohal_40 - nohal_30)×100/nohal_30.
[d] The difference is calculated as (hal_20 - hal_10)×100/hal_10.
[e] The difference is calculated as (hal_30 - hal_20)×100/hal_20.
[f] The difference is calculated as (hal_40 - hal_30)×100/hal_30.