Geophysical Research Letters

Supporting Information for

Intercomparison between Surrogate, Explicit and Full Treatments of VSL Bromine Chemistry within the CAM-Chem Chemistry-Climate Model

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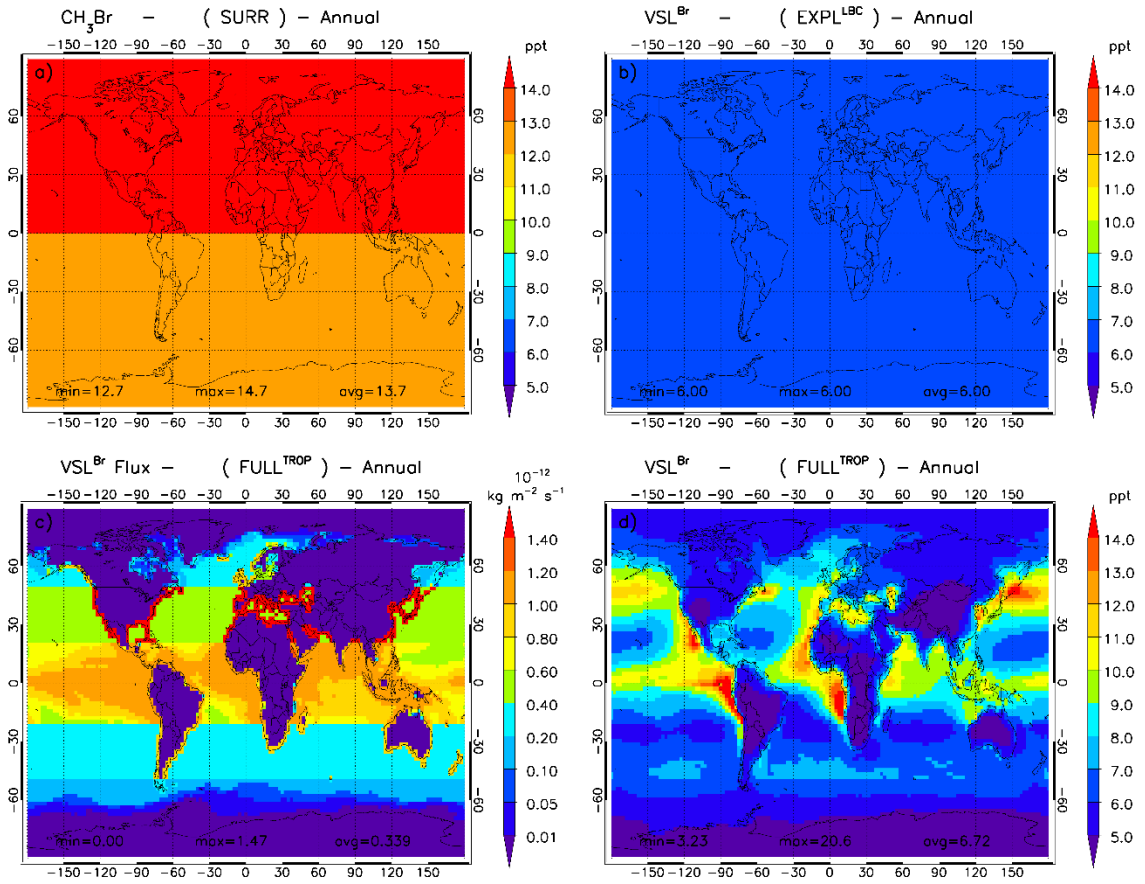

Figure S1. Spatial distribution of CAM-Chem surface LBCs and emissions considered for each sensitivity simulation: **a)** surface LBC for long-lived CH_3Br including the additional contribution of CH_3Br as a surrogate of VSL^{Br} (*SURR*^{LBC}); **b)** surface LBC for the sum of $CHBr_3$ and CH_2Br_2 within the *EXPL*^{LBC} setup; **c)** annual oceanic bromine flux from the six VSL^{Br} included in the Ordóñez et al., (2012) emissions inventory (*FULL*^{TROP}); **d)** annual mean total VSL^{Br} mixing ratio at the model surface for the *FULL*^{TROP} scheme.

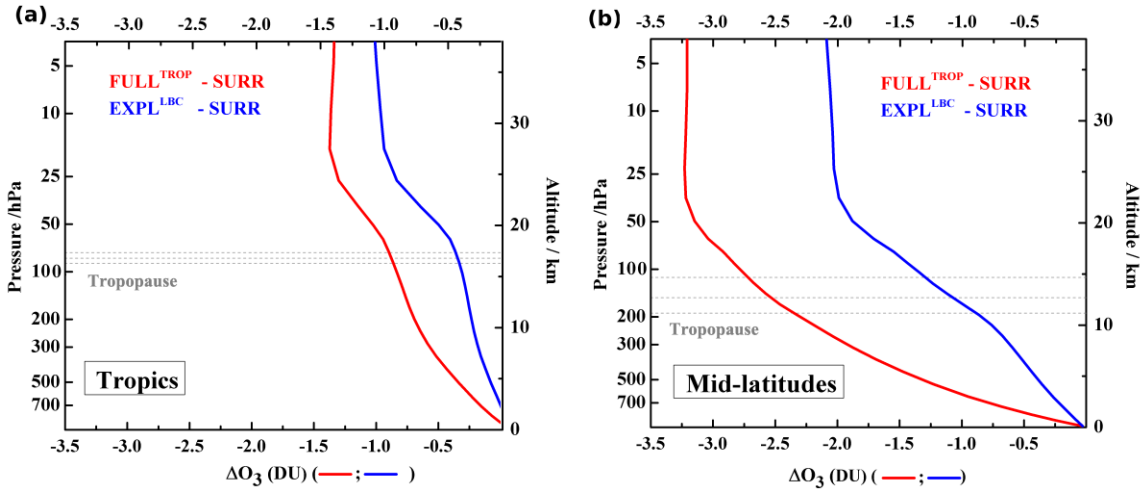

Figure S2. Annual cumulative ozone partial column change (ΣΔpO3) cumulative ozone partial column change between the *FULL*^{TROP} and *EXPL*^{LBC} schemes respect to the *SURR* setup. The vertical integration has been performed from the surface to the model top within the **c)** Tropics (20°N−20°S) and **b)** Mid-latitudes (25°N−50°S N/S).

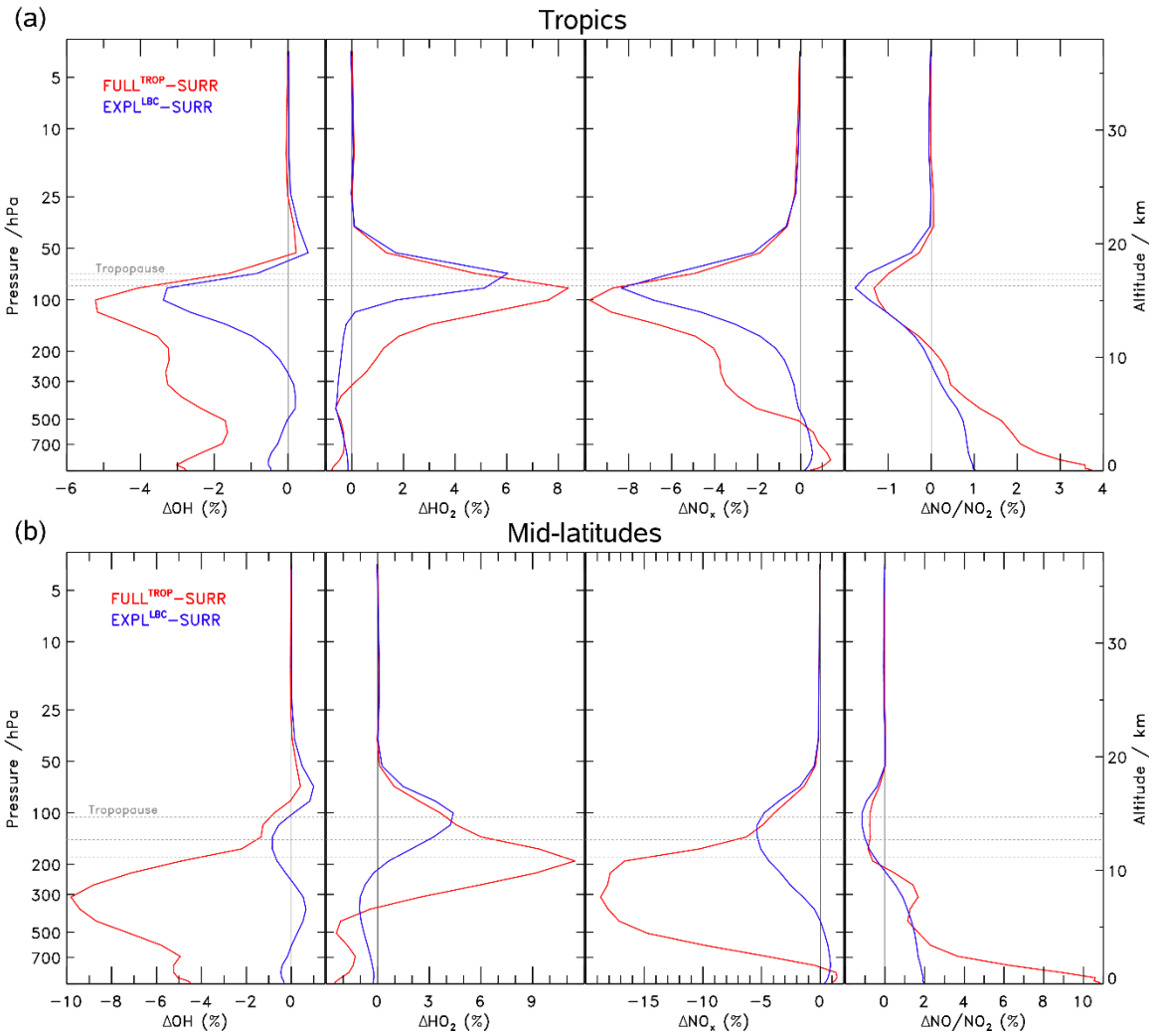

Figure S3. Percent change in mean vertical profiles of the indicated species or ratio for the *FULL*^{TROP} (red) and *EXPL*^{LBC} (blue) schemes, relative to the *SURR* setup. Each panel show the annual mean vertical profile over the **a)** Tropical (20°S–20°N) and **b)** mid-latitude (25°–50° N/S) bands. Species plotted are, from left to right, OH, HO_2, NO_x (=NO+NO_2), and the ratio NO/NO_2.

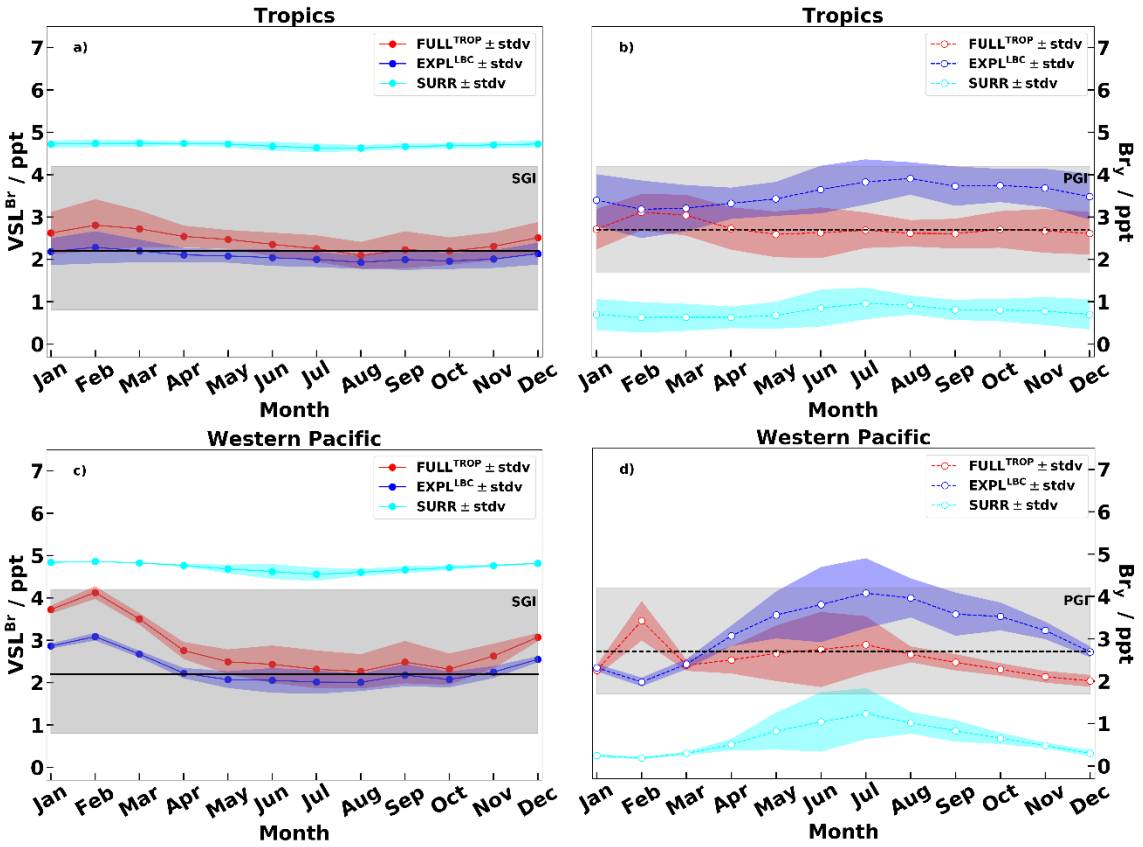

Figure S4. Seasonal variation of SGI (left) and PGI (right) for the Tropical annual mean (top) and Western Pacific (WP) region during February (bottom) for all sensitivity setups. Shaded areas represent the model spatial and monthly variability within the Tropics (20°N-20°S) and the WP (0°N-20°N & 140°E-160°E). The SGI and PGI (mean ± range) assessed tropical mean annual values from WMO (2018) are shown in black.

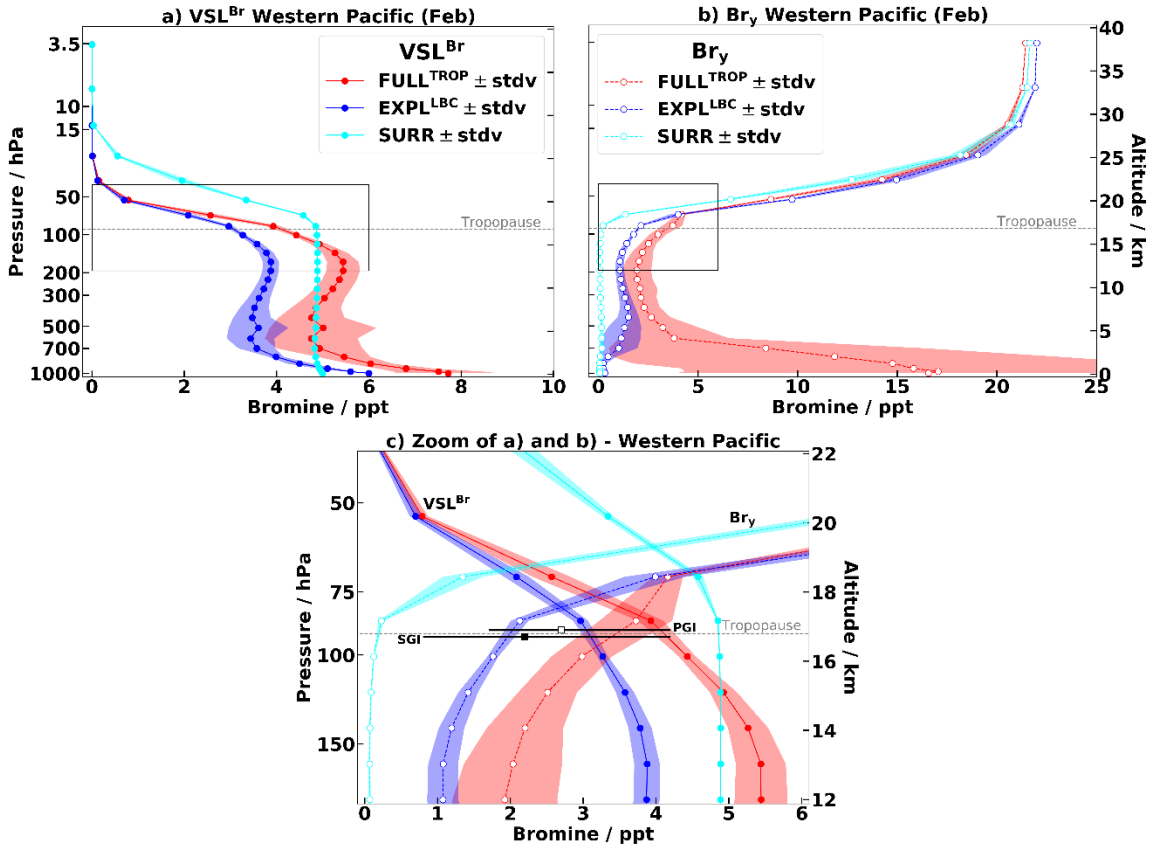

Figure S5. Idem to Fig. 1 in the main text, but for the Western Pacific warm pool region (0°N-20°N & 140°E-160°E) during February.

Table S1. Description of emission sources, surface LBCs and tropospheric processing considered for each chemical scheme.

Sensitivity	Long-lived bromine (CH_3Br + Halons)	Surrogate VSL^{Br} (CH_3Br)	Explicit VSL^{Br} ($CHBr_3$ + CH_2Br_2)	SSA dehalogenation (Br_y)	Washout, ice-uptake & het. recycling (Bry)
SURR	**15.9 ppt Br**	**5 ppt Br**[$]	**–**	**–**	**–**
EXPL^{LBC}	**15.9 ppt Br**	**–**	**1.2 ppt each (6 ppt Br)** [$]	**–**	**–**
FULL^{TROP}	**15.9 ppt Br**	**–**	**3.4×10^{-11} kg m^{-2} s^{-1} (6.72 ppt Br)** [&]	**Yes** [#]	**Yes** [#]

[$] The surrogate and explicit LBCs values are those recommended within the CCMI (Chemistry-Climate Model Initiative) project.

[&] The full approach includes seasonal and geographically varying oceanic fluxes from the Ordóñez et al. (2012) inventory. The corresponding global mean surface VSL^{Br} volume mixing ratio is shown in parenthesis.

[#] SSA dehalogenation and Br_y washout, ice-uptake and recycling within the troposphere are based on Fernandez et al. (2014).

Table S2. Contribution of Bromocarbons and Inorganic Bromine to total Bromine Budget within the Tropics.

Sensitivity[$,#]	VSL^{Br} (ppt)	Br_y (ppt)	$VSL^{Br}+Br_y$ (ppt)	LL^{Br} (ppt)	ΣBr (ppt)	O_3 (ppm)	$\sum pO_3$ (DU)
Tropics-38 km							
***FULL*^{TROP}**	0.0	21.4	21.4	0.0	21.4	8.77	256.6
***EXPL*^{LBC}**	0.0	22.0	22.0	0.0	22.0	8.75	256.9
SURR	0.0	21.7	21.7	0.0	21.7	8.77	258.0
Tropics-22 km							
***FULL*^{TROP}**	0.1	14.8	14.9	6.4	21.3	2.90	86.1
***EXPL*^{LBC}**	0.1	15.4	15.5	6.4	21.9	2.89	86.6
SURR	1.9	13.3	15.2	6.4	21.6	2.91	87.5
Mean Tropical Tropopause[&]-16.8 km							
***FULL*^{TROP}**	2.4	2.7	5.2	15.6	20.8	0.15	27.1
***EXPL*^{LBC}**	2.1	3.5	5.6	15.6	21.3	0.15	27.6
SURR	4.7	0.8	5.5	15.6	21.1	0.15	28.1
Tropics-12 km							
***FULL*^{TROP}**	4.4	0.9	5.4	16.1	21.5	0.04	21.4
***EXPL*^{LBC}**	3.5	1.5	5.0	16.1	21.1	0.04	21.9
SURR	4.9	0.1	5.0	16.1	21.1	0.04	22.3
Tropics-5 km							
***FULL*^{TROP}**	4.2	1.8	6.0	16.0	22.0	0.04	11.9
***EXPL*^{LBC}**	3.3	1.5	4.8	16.0	20.8	0.04	12.3
SURR	4.8	0.2	5.0	16.0	21.0	0.04	12.5
Surface-100 m							
***FULL*^{TROP}**	8.6	2.9	11.5	16.1	27.6	0.02	0.2
***EXPL*^{LBC}**	6.0	0.5	6.5	16.1	22.7	0.02	0.3
SURR	5.0	0.1	5.1	16.1	21.2	0.02	0.3

[$]All values consider the sum of all bromocarbons (VSL^{Br}, LL^{Br}), inorganic (Br_y) and total (ΣBr) bromine species weighted by their respective bromine atomicity. Ozone mixing ratios as well as the cumulative partial ozone column ($\sum pO_3$) vertically integrated from the surface up to each vertical height/pressure is also provided.

[#]For the *SURR* setup, the VSL^{Br} column includes the bromine contribution from the surrogate CH_3Br computed as $VSL^{Br} = LL^{Br}[EXPL^{LBC}] - LL^{Br}[SURR]$.

[&]Values at the mean chemical tropopause were interpolated to fulfill the $O_3 \approx 150$ ppb condition and represent the best estimate of the model bromine SGI and PGI.

Table S3. Contribution of Bromocarbons and Inorganic Bromine to total Bromine Budget within the Mid-Latitudes.

Sensitivity[$,#]	**VSL^{Br} (ppt)**	**Br_y (ppt)**	**$VSL^{Br}+Br_y$ (ppt)**	**LL^{Br} (ppt)**	**ΣBr (ppt)**	**O_3 (ppm)**	**$\sum pO_3$ (DU)**
Mid-latitudes-38 km							
***FULL*^{TROP}**	0.0	21.4	21.4	0.0	21.4	8.13	297.3
***EXPL*^{LBC}**	0.0	22.0	22.0	0.0	22.0	8.12	298.5
SURR	0.0	21.7	21.7	0.0	21.7	8.13	300.6
Mid-latitudes-22 km							
***FULL*^{TROP}**	0.0	18.2	18.3	3.0	21.3	3.46	150.0
***EXPL*^{LBC}**	0.0	18.8	18.9	3.0	21.9	3.45	151.4
SURR	0.8	17.8	18.6	3.0	21.6	3.46	153.4
Mid-latitudes-17 km							
***FULL*^{TROP}**	1.2	7.2	8.4	12.3	20.7	0.77	63.4
***EXPL*^{LBC}**	1.1	8.1	9.1	12.3	21.4	0.77	64.9
SURR	3.7	5.2	8.9	12.3	21.2	0.78	66.4
Mean mid-latitudinal tropopause[&]-12 km							
***FULL*^{TROP}**	2.6	2.1	4.7	15.4	20.1	0.15	33.1
***EXPL*^{LBC}**	2.4	3.5	5.8	15.4	21.2	0.15	34.7
SURR	4.6	0.9	5.5	15.4	20.9	0.16	35.7
Mid-latitudes-5 km							
***FULL*^{TROP}**	3.5	1.3	4.8	15.8	20.6	0.05	15.8
***EXPL*^{LBC}**	3.3	1.6	4.9	15.8	20.8	0.05	17.0
SURR	4.8	0.2	5.0	15.8	20.8	0.05	17.4
Surface-100 m							
***FULL*^{TROP}**	7.0	2.4	9.5	15.9	25.4	0.03	0.3
***EXPL*^{LBC}**	6.0	0.5	6.5	15.9	22.4	0.03	0.4
SURR	5.0	0.1	5.1	15.9	21.0	0.03	0.4

[$]All values consider the sum of all bromocarbons (VSL^{Br}, LL^{Br}), inorganic (Br_y) and total (ΣBr) bromine species weighted by their respective bromine atomicity. Ozone mixing ratios as well as the cumulative partial ozone column ($\sum pO_3$) vertically integrated from the surface up to each vertical height/pressure is also provided.

[#]For the *SURR* setup, the VSL^{Br} column includes the bromine contribution from the surrogate CH_3Br computed as $VSL^{Br} = LL^{Br}[EXPL^{LBC}] - LL^{Br}[SURR]$.

[&]Values at the mean chemical tropopause were interpolated to fulfill the $O_3 \approx 150$ ppb condition and represent the best estimate of the model bromine SGI and PGI.