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Global modelling of the total OH reactivity: investigations on the "missing" OH sink and its atmospheric implications

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Supplementary material

Tables S1, S2 and S3 below report the values of OH production and loss fluxes in the whole troposphere, in the boundary layer and at the surface respectively, obtained in the UM-UKCA runs described in the current work. The values for the base run are in reasonably good agreement with those reported by Lelieveld and co-workers (Lelieveld et al., 2016).

OH production	**Lelieveld et al., 2016**	**Base run**	**X + OH run**	**CH_3O_2 + OH run 1**	**CH_3O_2 + OH run 2**	**CH_3O_2 + OH run 3**
$O(^1D) + H_2O$	84.0 (33%)	79.1 (39.5%)	79.4 (39.7%)	78.0 (38.6%)	78.1 (38.9%)	78.2 (39.1%)
$NO + HO_2$	76.6 (30%)	55.0 (27.5%)	55.0 (27.5%)	56.7 (28.1%)	56.5 (28.1%)	56.2 (28.1%)
$O_3 + HO_2$	34.4 (14%)	23.5 (11.8%)	23.5 (11.7%)	24.1 (11.9%)	23.9 (11.9%)	23.7 (11.8%)
$H_2O_2 + h\nu$	24.8 (10%)	20.9 (10.5%)	20.9 (10.5%)	22.9 (11.4%)	22.4 (11.2%)	22.0 (11.0%)
OVOCs, ROOH + $h\nu$	31.4 (13%)	21.5 (10.8%)	21.1 (10.6%)	20.2 (10%)	20.0 (10.0%)	19.9 (10.0%)
Total OH production	**251.2**	**200.1**	**199.9**	**201.9**	**200.9**	**199.9**
OH loss						
$OH + HO_y$ [a]	46.2 (18%)	32.5 (16.3%)	31.9 (16.0%)	33.0 (16.4%)	32.6 (16.2%)	32.1 (16.1%)
$OH + NO_x$	4.1 (1.5%)	2.2 (1.1%)	2.1 (1.0%)	2.2 (1.1%)	2.2 (1.1%)	2.2 (1.1%)
$OH + NO_z$ [b]		1.4 (0.7%)	1.4 (0.7%)	1.4 (0.7%)	1.4 (0.7%)	1.4 (0.7%)
$OH + CH_4$	29.8 (12%)	26.7 (13.3%)	26.1 (13.1%)	26.2 (13.0%)	26.0 (13.0%)	25.9 (13.0%)
OH + CO	97.8 (39%)	90.3 (45.1%)	89.7 (44.9%)	90.0 (44.6%)	89.5 (44.6%)	89.1 (44.6%)
$OH + C_1$ VOCs [c]	37.0 (15%)	24.8 (12.4%)	24.0 (12.0%)	22.5 (11.2%)	22.7 (11.3%)	22.8 (11.4%)
$OH + C_{2,3}$ VOCs [d]	34.7 (14%)	3.5 (1.7%)	3.4 (1.7%)	3.5 (1.7%)	3.5 (1.7%)	3.5 (1.7%)
OH + isoprene and ox. prod. [e]		17.0 (8.5%)	16.5 (8.3%)	17.1 (8.5%)	17.0 (8.5%)	17.0 (8.5%)
OH + monoterpenes		0.4 (0.2%)	0.3 (0.2%)	0.4 (0.2%)	0.4 (0.2%)	0.4 (0.2%)
OH + halogenated [f]	1.6 (0.5%)	0.0 (0.0%)	0.0 (0.0%)	0.0 (0.0%)	0.0 (0.0%)	0.0 (0.0%)
OH + sulphur species [g]		1.3 (0.6%)	1.2 (0.6%)	1.3 (0.6%)	1.3 (0.6%)	1.3 (0.6%)
OH + X	-	-	2.9 (1.5%)	-	-	-
$OH + CH_3O_2 \rightarrow HO_2 + CH_3O$	-	-	-	4.4 (2.2%)	3.5 (1.7%)	2.6 (1.3%)
$OH + CH_3O_2 \rightarrow CH_3OH + O_2$	-	-	-	-	0.9 (0.4%)	1.7 (0.9%)
Total OH loss	**251.2**	**200.1**	**199.9**	**201.9**	**200.9**	**199.9**

Notes:

[a] HO_y = H_2, O_3, H_2O_2, OH, HO_2, $O(^3P)$

[b] NO_z = PAN, MPAN, PPAN, HONO, HNO_3, HNO_4, NALD, CH_3ONO_2, NO_3, isoprene nitrate

[c] C_1 VOCs = CH_3OH, formaldehyde, methyl hydroperoxide, formic acid

[d] $C_{2,3}$ VOCs = ethane, propane, acetaldehyde, propionaldehyde, ethyl, *n*-propyl and *i*-propyl hydroperoxides, acetone, acetone hydroperoxide

[e] isoprene oxidation products = isoprene hydroperoxide, MACR, MACROOH, hydroxyacetone, methyl glyoxal, acetic acid, peracetic acid

[f] halogenated species = CH_3Br, ClO, BrO, HBr, HCl, $ClONO_2$, OClO, HOCl

[g] sulphur species = SO_2, H_2S, dimethyl sulphide, COS, CS_2

Table S1: Annual mean tropospheric OH production and loss fluxes in Tmol yr^{-1}.

OH production	**Base run**	**X + OH run**	**CH_3O_2 + OH run 1**	**CH_3O_2 + OH run 2**	**CH_3O_2 + OH run 3**
$O(^1D) + H_2O$	20.2 (38.4 %)	20.1 (39.4 %)	19.9 (37.6%)	19.9 (37.8%)	19.9 (38.0%)
$NO + HO_2$	17.0 (32.4%)	16.4 (32.0%)	17.5 (33.0%)	17.4 (33.0%)	17.3 (33.0%)
$O_3 + HO_2$	4.5 (8.5%)	4.3 (8.4%)	4.5 (8.6%)	4.5 (8.6%)	4.5 (8.5%)
$H_2O_2 + h\nu$	2.9 (5.5%)	2.8 (5.5%)	3.2 (6.0%)	3.1 (5.9%)	3.0 (5.8%)
OVOCs, ROOH + $h\nu$	8.0 (15.3%)	7.5 (14.7%)	7.8 (14.7%)	7.7 (14.7%)	7.7 (14.7%)
Total OH production	**52.5**	**51.1**	**52.8**	**52.6**	**52.5**
OH loss					
$OH + HO_y$ [a]	6.9 (13.2%)	6.5 (12.7%)	7.0 (13.2%)	6.9 (13.1%)	6.8 (13.0%)
$OH + NO_x$	1.2 (2.3%)	1.0 (2.0%)	1.2 (2.2%)	1.2 (2.3%)	1.2 (2.3%)
$OH + NO_z$ [b]	0.6 (1.1%)	0.6 (1.1%)	0.6 (1.1%)	0.6 (1.1%)	0.6 (1.1%)
$OH + CH_4$	6.9 (13.1%)	6.5 (12.7%)	6.7 (12.7%)	6.7 (12.7%)	6.7 (12.7%)
OH + CO	19.1 (36.3%)	18.0 (35.2%)	18.9 (35.8%)	18.9 (35.9%)	18.8 (35.9%)
$OH + C_1$ VOCs [c]	6.5 (12.4%)	6.0 (11.7%)	6.0 (11.4%)	6.0 (11.5%)	6.0 (11.5%)
$OH + C_{2,3}$ VOCs [d]	0.9 (1.8%)	0.9 (1.7%)	0.9 (1.8%)	0.9 (1.8%)	0.9 (1.8%)
OH + isoprene and ox. prod. [e]	9.4 (18.0%)	8.7 (17.1%)	9.4 (17.8%)	9.4 (17.9%)	9.4 (17.9%)
OH + monoterpenes	0.3 (0.6%)	0.3 (0.5%)	0.3 (0.6%)	0.3 (0.6%)	0.3 (0.6%)
OH + halogenated [f]	0.0 (0.0%)	0.0 (0.0%)	0.0 (0.0%)	0.0 (0.0%)	0.0 (0.0%)
OH + sulphur species [g]	0.7 (1.3%)	0.6 (1.2%)	0.7 (1.2%)	0.7 (1.2%)	0.7 (1.2%)
OH + X	-	2.0 (4.0%)	-	-	-
$OH + CH_3O_2 \rightarrow HO_2 + CH_3O$	-	-	1.0 (2%)	0.8 (1.6%)	0.6 (1.2%)
$OH + CH_3O_2 \rightarrow CH_3OH + O_2$	-	-	-	0.2 (0.4%)	0.4 (0.8%)
Total OH loss	**52.5**	**51.1**	**52.8**	**52.6**	**52.5**

Notes:

[a] HO_y = H_2, O_3, H_2O_2, OH, HO_2, $O(^3P)$

[b] NO_z = PAN, MPAN, PPAN, HONO, HNO_3, HNO_4, NALD, CH_3ONO_2, NO_3, isoprene nitrate

[c] C_1 VOCs = CH_3OH, formaldehyde, methyl hydroperoxide, formic acid

[d] $C_{2,3}$ VOCs = ethane, propane, acetaldehyde, propionaldehyde, ethyl, *n*-propyl and *i*-propyl hydroperoxides, acetone, acetone hydroperoxide

[e] isoprene oxidation products = isoprene hydroperoxide, MACR, MACROOH, hydroxyacetone, methyl glyoxal, acetic acid, peracetic acid

[f] halogenated species = CH_3Br, ClO, BrO, HBr, HCl, $ClONO_2$, OClO, HOCl

[g] sulphur species = SO_2, H_2S, dimethyl sulphide, COS, CS_2

Table S2: Annual mean OH production and loss fluxes in the boundary layer in Tmol yr^{-1}.

OH production	**Base run**	**X + OH run**	**CH_3O_2 + OH run 1**	**CH_3O_2 + OH run 2**	**CH_3O_2 + OH run 3**
$O(^1D) + H_2O$	0.78 (30.6%)	0.78 (32.8 %)	0.77 (30.1%)	0.77 (30.2%)	0.78 (30.4%)
$NO + HO_2$	1.00 (39.3%)	0.91 (38.3%)	1.02 (39.9%)	1.02 (39.8%)	1.02 (39.8%)
$O_3 + HO_2$	0.18 (7.0%)	0.17 (6.9%)	0.18 (7.0%)	0.18 (7.0%)	0.18 (7.0%)
$H_2O_2 + h\nu$	0.10 (3.8%)	0.09 (3.9%)	0.11 (4.1%)	0.10 (4.1%)	0.10 (4.0%)
OVOCs, ROOH + $h\nu$	0.50 (19.4%)	0.43 (18.0%)	0.48 (18.9%)	0.48 (18.9%)	0.48 (18.9%)
Total OH production	**2.56**	**2.38**	**2.57**	**2.56**	**2.55**
OH loss					
OH + HO_y [a]	0.27 (10.7%)	0.25 (10.3%)	0.27 (10.7%)	0.27 (10.6%)	0.27 (10.5%)
OH + NO_x	0.08 (3.2%)	0.07 (2.7%)	0.08 (3.2%)	0.08 (3.2%)	0.08 (3.2%)
OH + NO_z [b]	0.03 (1.2%)	0.03 (1.1%)	0.03 (1.2%)	0.03 (1.2%)	0.03 (1.2%)
OH + CH_4	0.29 (11.5%)	0.27 (11.3%)	0.29 (11.2%)	0.29 (11.2%)	0.29 (11.2%)
OH + CO	0.79 (30.9%)	0.71 (29.3%)	0.78 (30.5%)	0.78 (30.5%)	0.78 (30.6%)
OH + C_1 VOCs [c]	0.27 (10.7%)	0.24 (10.0%)	0.26 (10.0%)	0.26 (10.0%)	0.26 (10.0%)
OH + $C_{2,3}$ VOCs [d]	0.05 (1.8%)	0.04 (1.7%)	0.05 (1.8%)	0.05 (1.8%)	0.05 (1.8%)
OH + isoprene and ox. prod. [e]	0.69 (27.1%)	0.59 (24.8%)	0.69 (26.9%)	0.69 (27.0%)	0.69 (27.0%)
OH + monoterpenes	0.04 (1.6%)	0.03 (1.1%)	0.04 (1.6%)	0.04 (1.6%)	0.04 (1.6%)
OH + halogenated [f]	0.00 (0.0%)	0.00 (0.0%)	0.00 (0.0%)	0.00 (0.0%)	0.00 (0.0%)
OH + sulphur species [g]	0.04 (1.4%)	0.03 (1.4%)	0.04 (1.4%)	0.04 (1.4%)	0.04 (1.4%)
OH + X	-	0.14 (6.0%)	-	-	-
OH + $CH_3O_2 \rightarrow HO_2 + CH_3O$	-	-	0.04 (1.6%)	0.03 (1.3%)	0.02 (1.0%)
OH + $CH_3O_2 \rightarrow CH_3OH + O_2$	-	-	-	0.01 (0.3%)	0.02 (0.6%)
Total OH loss	**2.56**	**2.38**	**2.57**	**2.56**	**2.55**

Notes:

[a] HO_y = H_2, O_3, H_2O_2, OH, HO_2, $O(^3P)$

[b] NO_z = PAN, MPAN, PPAN, HONO, HNO_3, HNO_4, NALD, CH_3ONO_2, NO_3, isoprene nitrate

[c] C_1 VOCs = CH_3OH, formaldehyde, methyl hydroperoxide, formic acid

[d] $C_{2,3}$ VOCs = ethane, propane, acetaldehyde, propionaldehyde, ethyl, *n*-propyl and *i*-propyl hydroperoxides, acetone, acetone hydroperoxide

[e] isoprene oxidation products = isoprene hydroperoxide, MACR, MACROOH, hydroxyacetone, methyl glyoxal, acetic acid, peracetic acid

[f] halogenated species = CH_3Br, ClO, BrO, HBr, HCl, $ClONO_2$, OClO, HOCl

[g] sulphur species = SO_2, H_2S, dimethyl sulphide, COS, CS_2

Table S3: Annual mean OH production and loss fluxes at the surface in Tmol yr^{-1}.

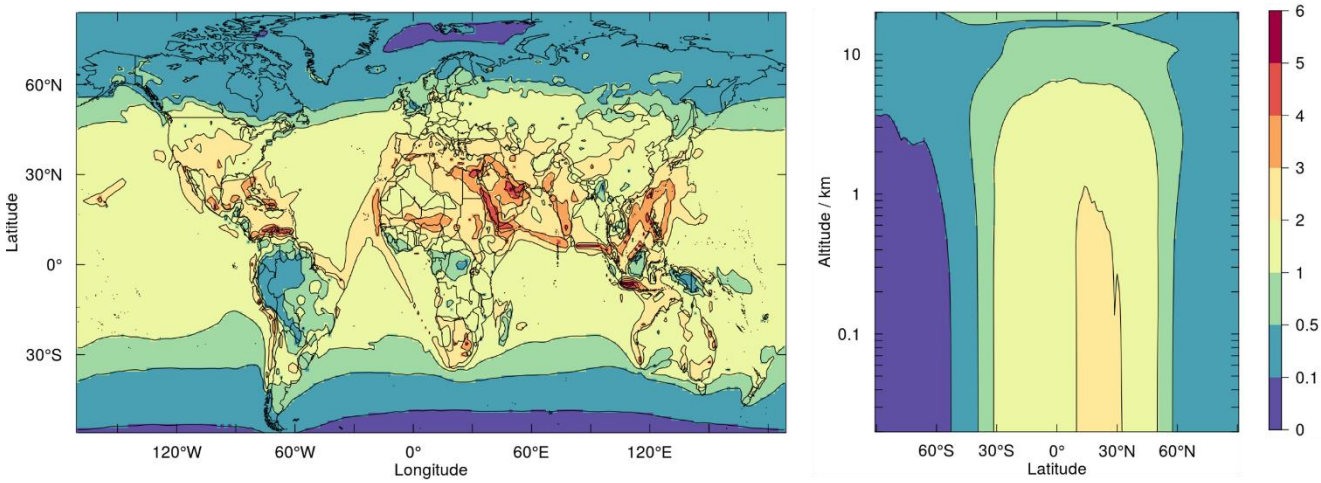

Figure S1: Annual mean OH number density in 10^6 molecules cm^{-3} at the surface (left) and as a zonal mean (right).

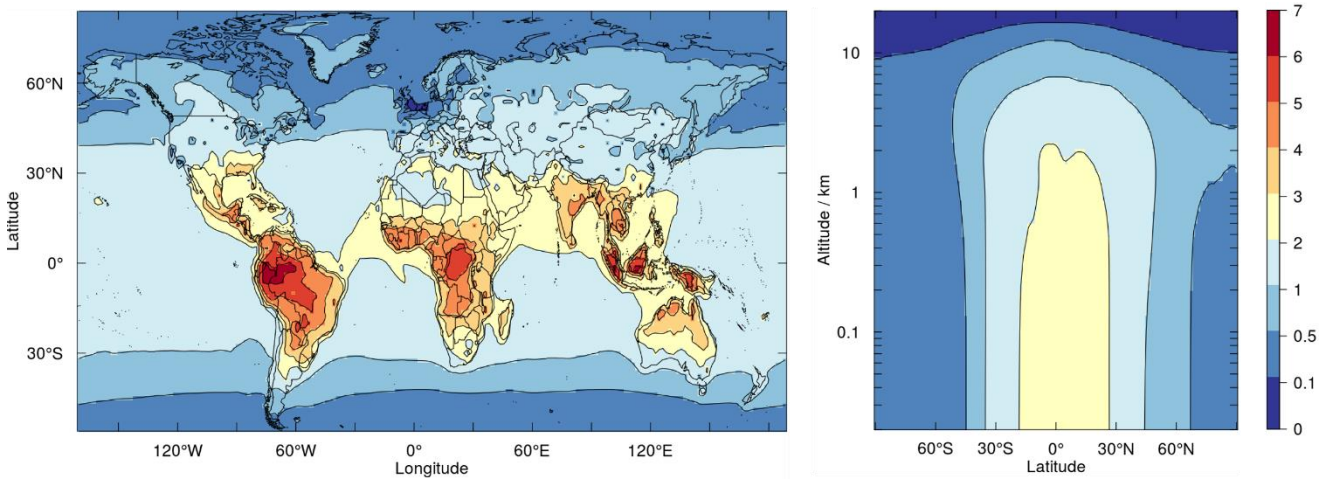

Figure S2: Annual mean HO_2 number density in 10^8 molecules cm^{-3} at the surface (left) and as a zonal mean (right).

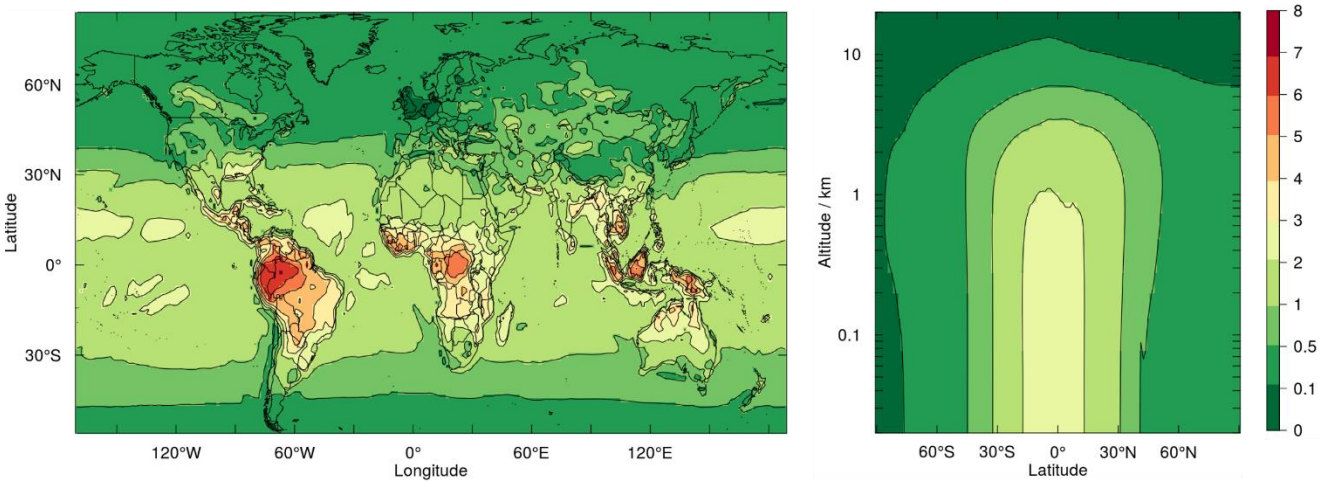

Figure S3: Annual mean RO_2 number density in 10^8 molecules cm^{-3} at the surface (left) and as a zonal mean (right).

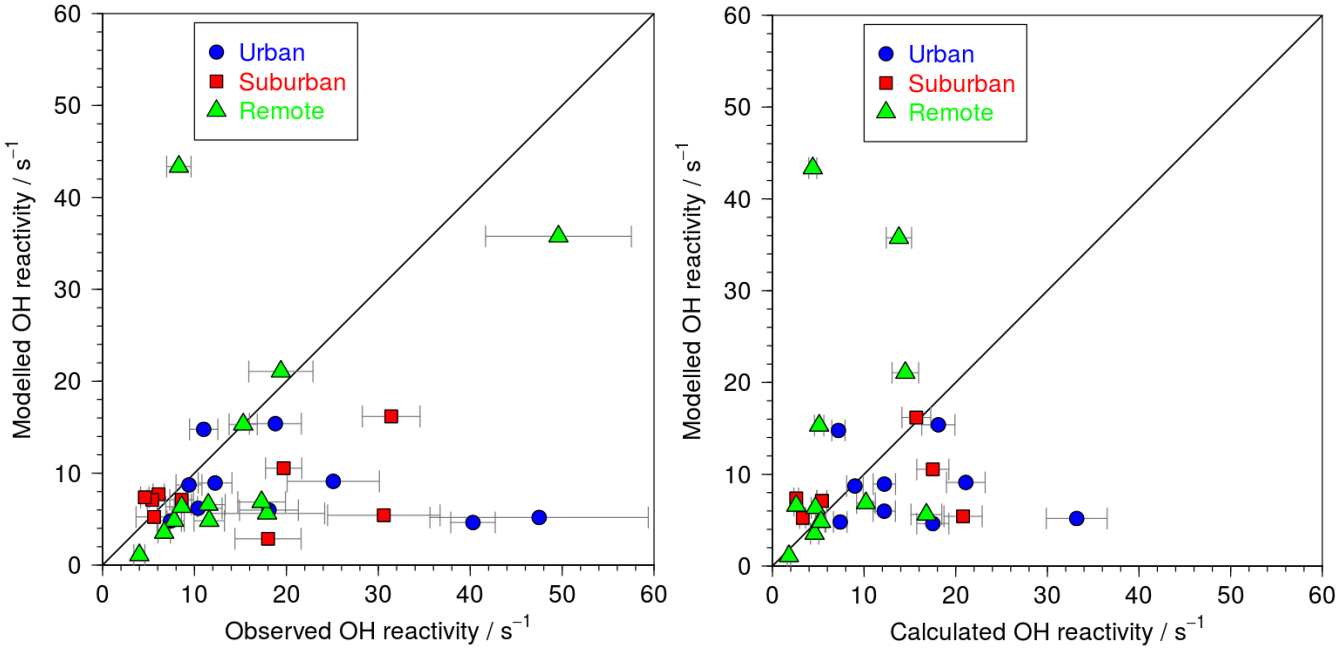

Figure S4: Scatter plots of OH reactivity calculated from modelled sinks against observed total OH reactivity (left) and OH reactivity calculated from individual measured species (right).

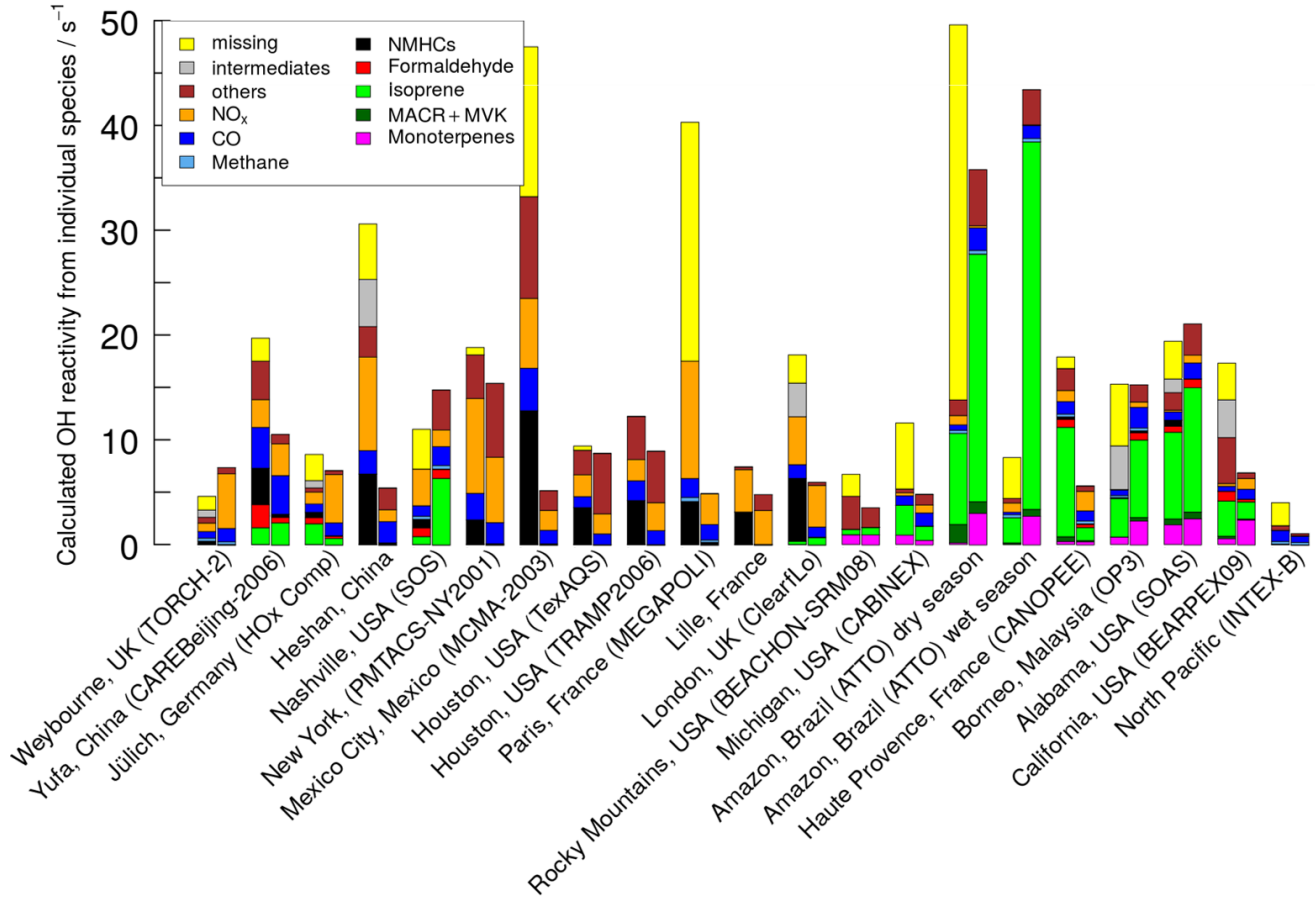

Figure S5: Bar plot comparing the speciated OH reactivity from individual field campaigns. For each pair of bars, the bar on the left represents the reactivities calculated from measured species and the one on the right the reactivities from modelled species.

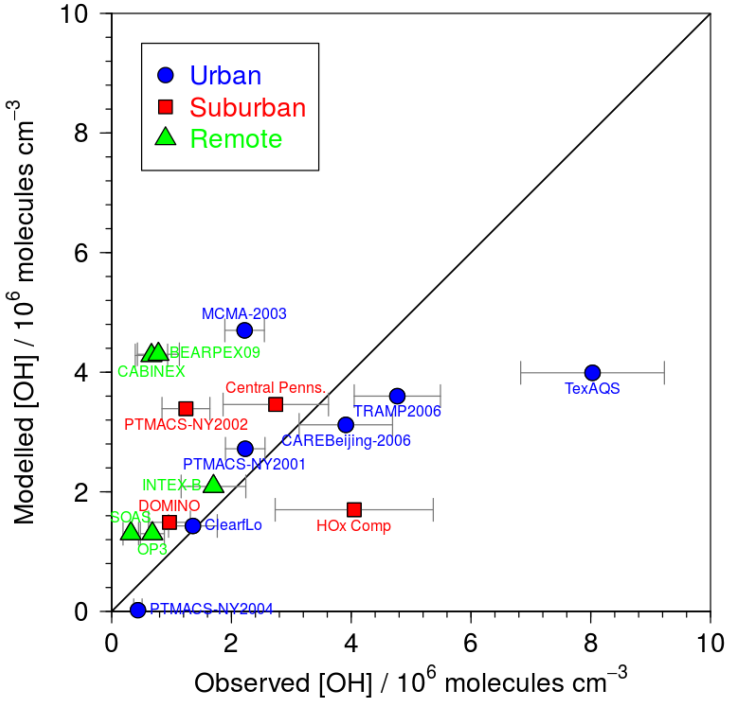

Figure S6: Scatter plot of modelled against observed [OH]. Error bars represent the 2σ uncertainty from each individual measurement.

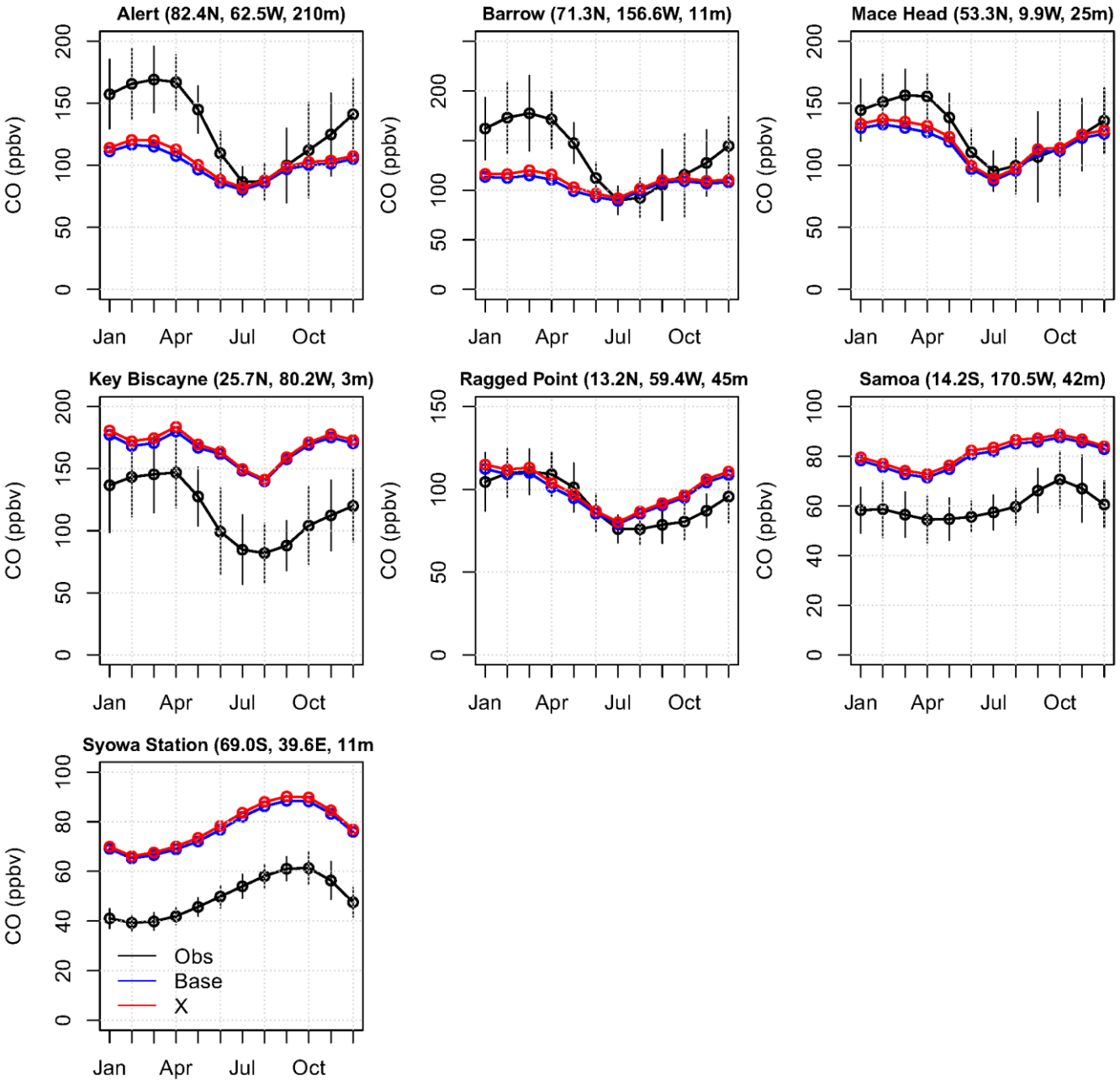

Figure S7: Comparison of CO observations (black data points and lines) with the base run model (blue points and lines) and the model including OH sink X (red points and lines).

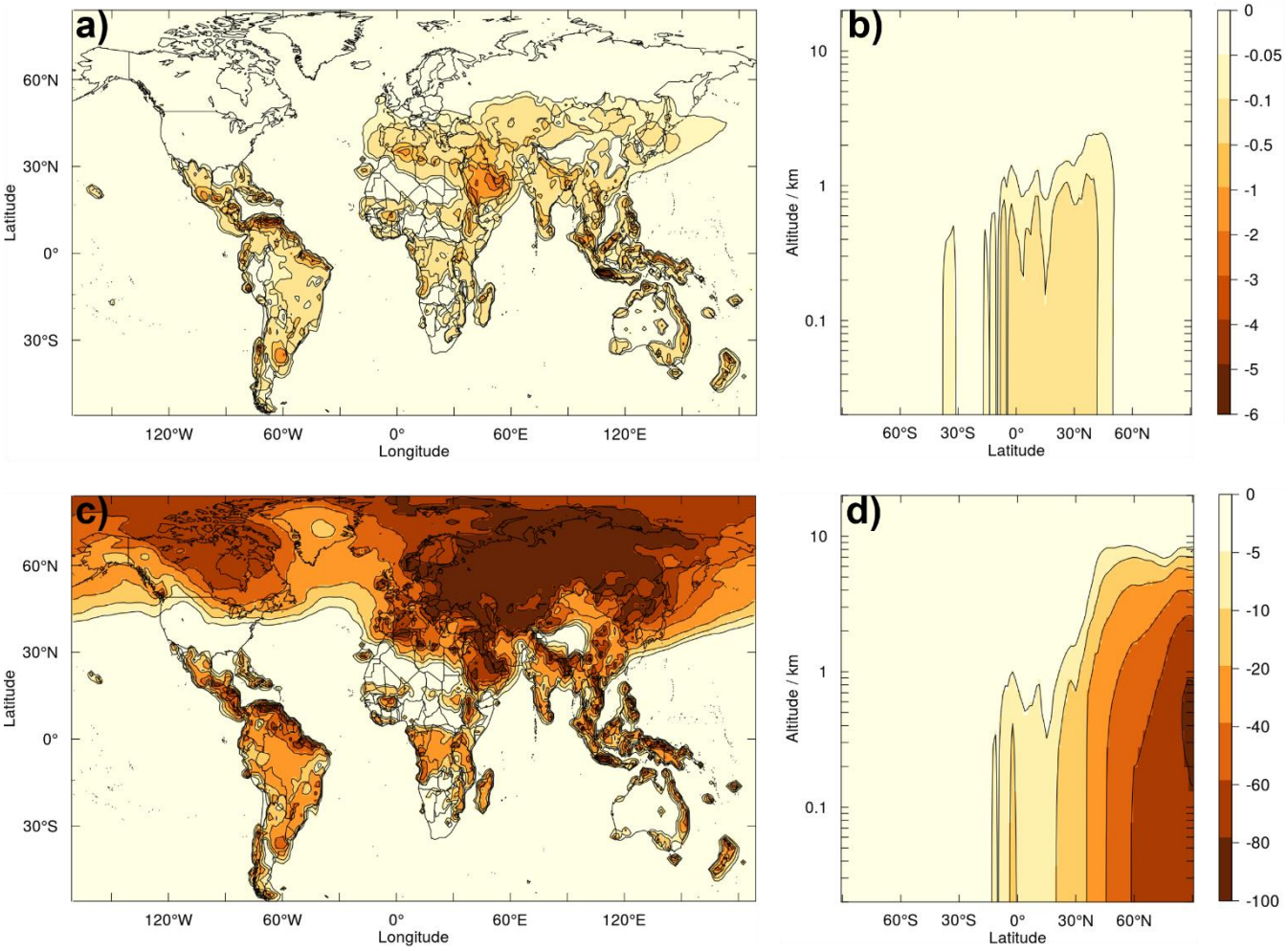

Figure S8: Mean change in OH concentration in the boreal winter (DJF) following inclusion of R3: absolute change in 10^6 molecules cm^{-3} a) at the surface and b) as a zonal mean, and relative percentage change c) at the surface and d) as a zonal mean.

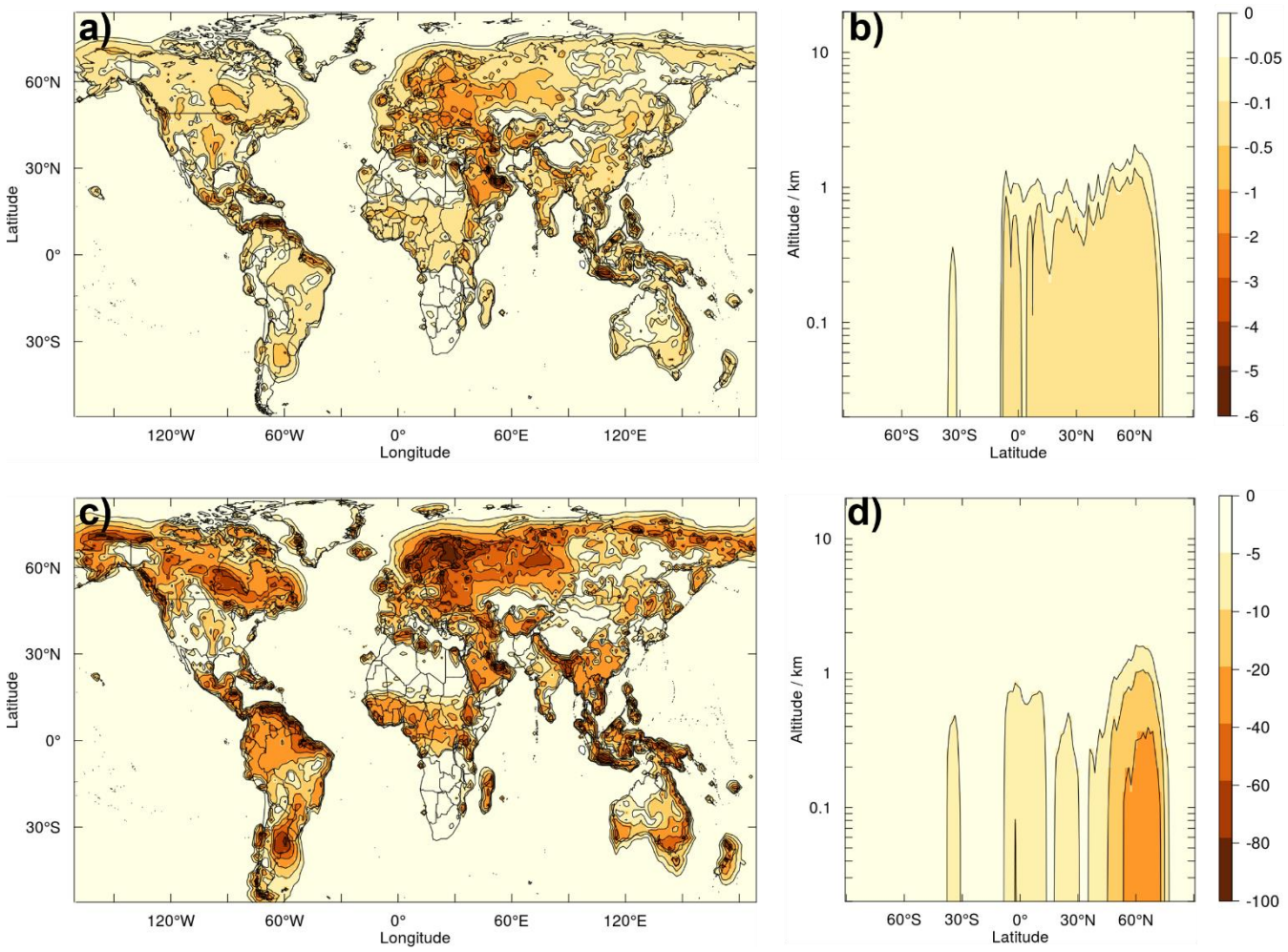

Figure S9: Mean change in OH concentration in the boreal summer (JJA) following inclusion of R3: absolute change in 10^6 molecules cm^{-3} a) at the surface and b) as a zonal mean, and relative percentage change c) at the surface and d) as a zonal mean.

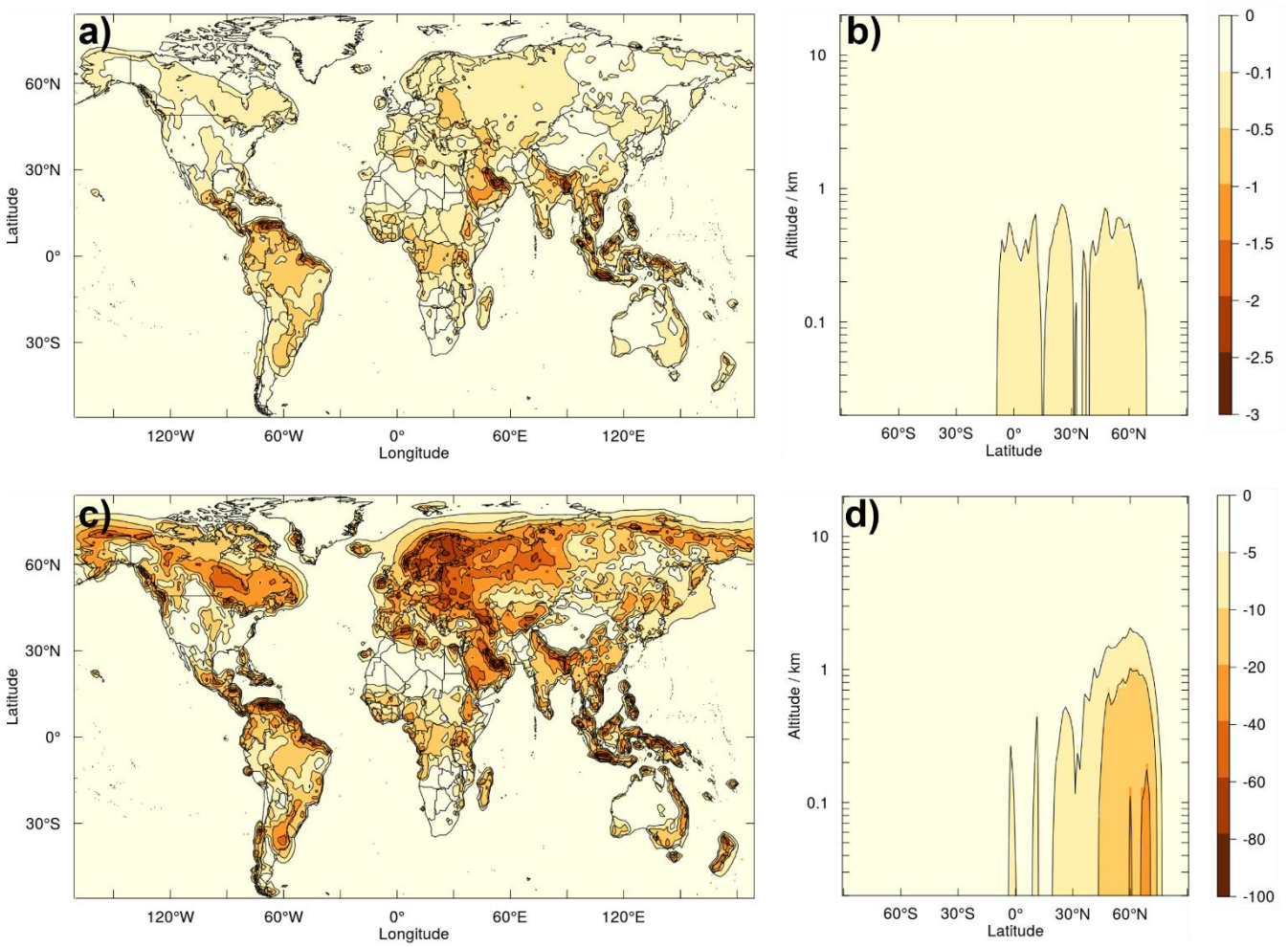

Figure S10: Annual mean change in HO_2 concentration following inclusion of R3: absolute change in 10^8 molecules cm^{-3} a) at the surface and b) as a zonal mean, and relative percentage change c) at the surface and d) as a zonal mean.

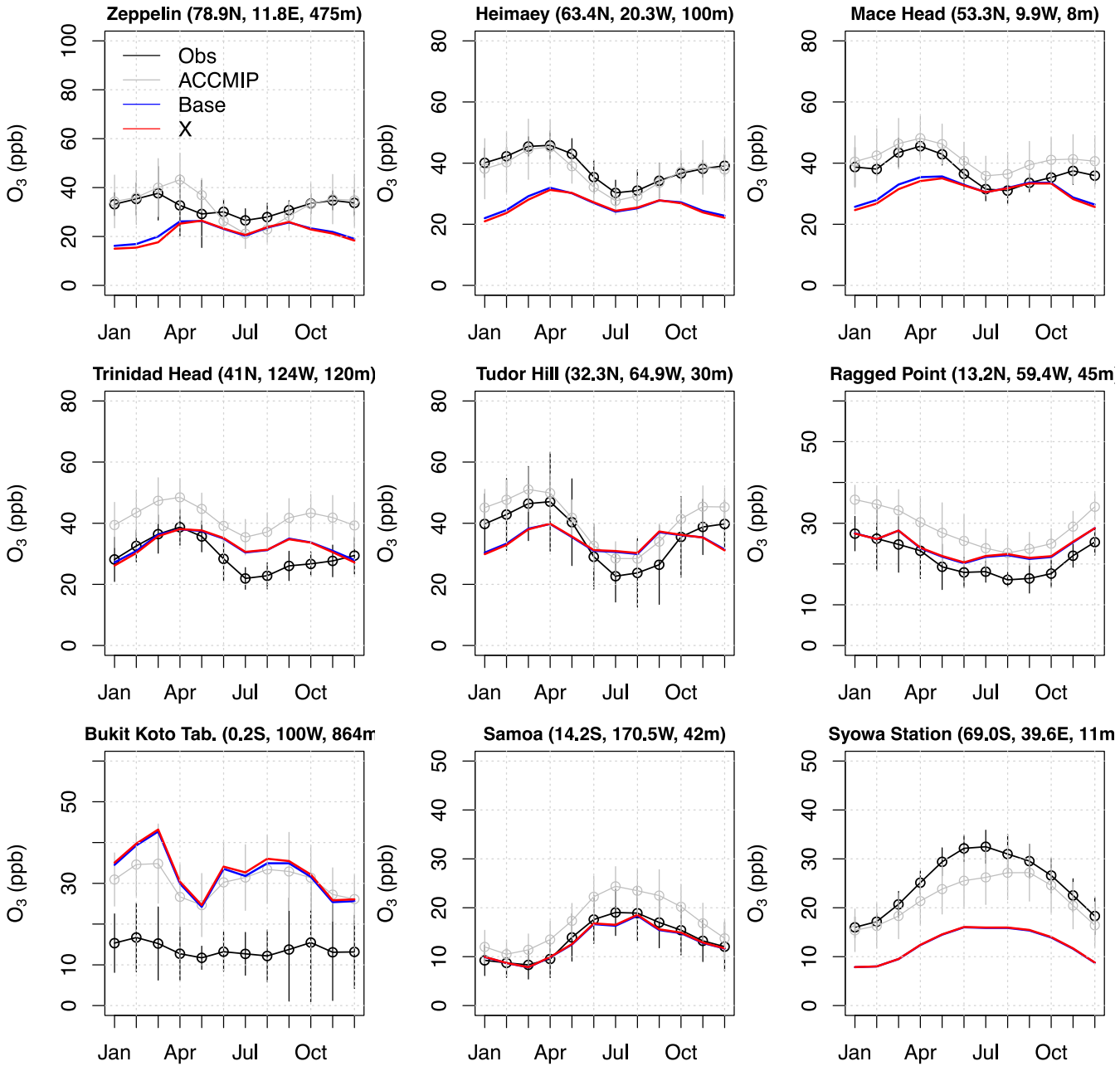

Figure S11: Comparison of ozone observations (black data points and lines) with the base run model (red points and lines) and the model including OH sink X (blue points and lines). The multi-model mean results from the ACCMIP year 2000 simulations are shown in grey with the grey bars indicating one standard deviation of the ACCMIP multi-model ensemble (Young et al., 2013).

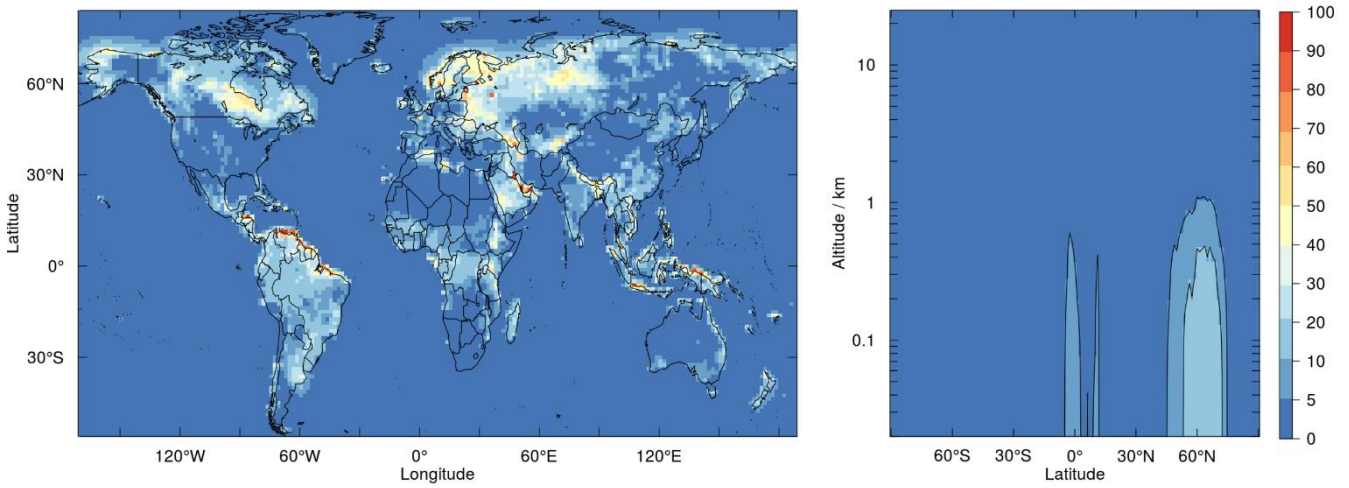

Figure S12: Percentage contribution of the flux through R3 (X + OH) to the total OH loss flux at the surface (left) and as zonal mean (right).

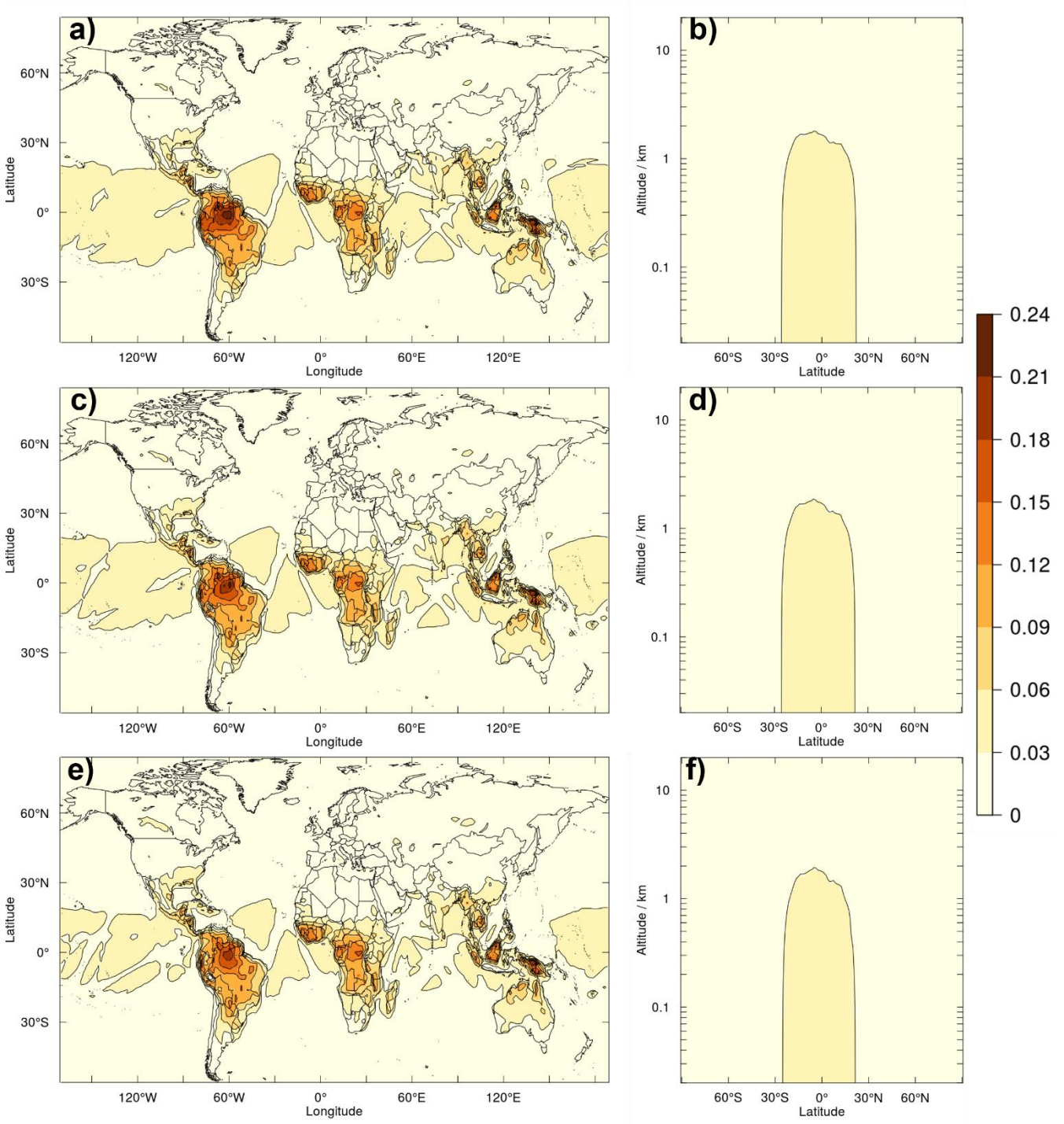

Figure S13: Mean changes in k_{OH} (in s^{-1}) from run 1 (a and b, top row), run 2 (c and d, middle row), and run 3 (e and f, bottom row), relative to the base run. Changes are shown for the surface (left column) and as a zonal mean (right column).

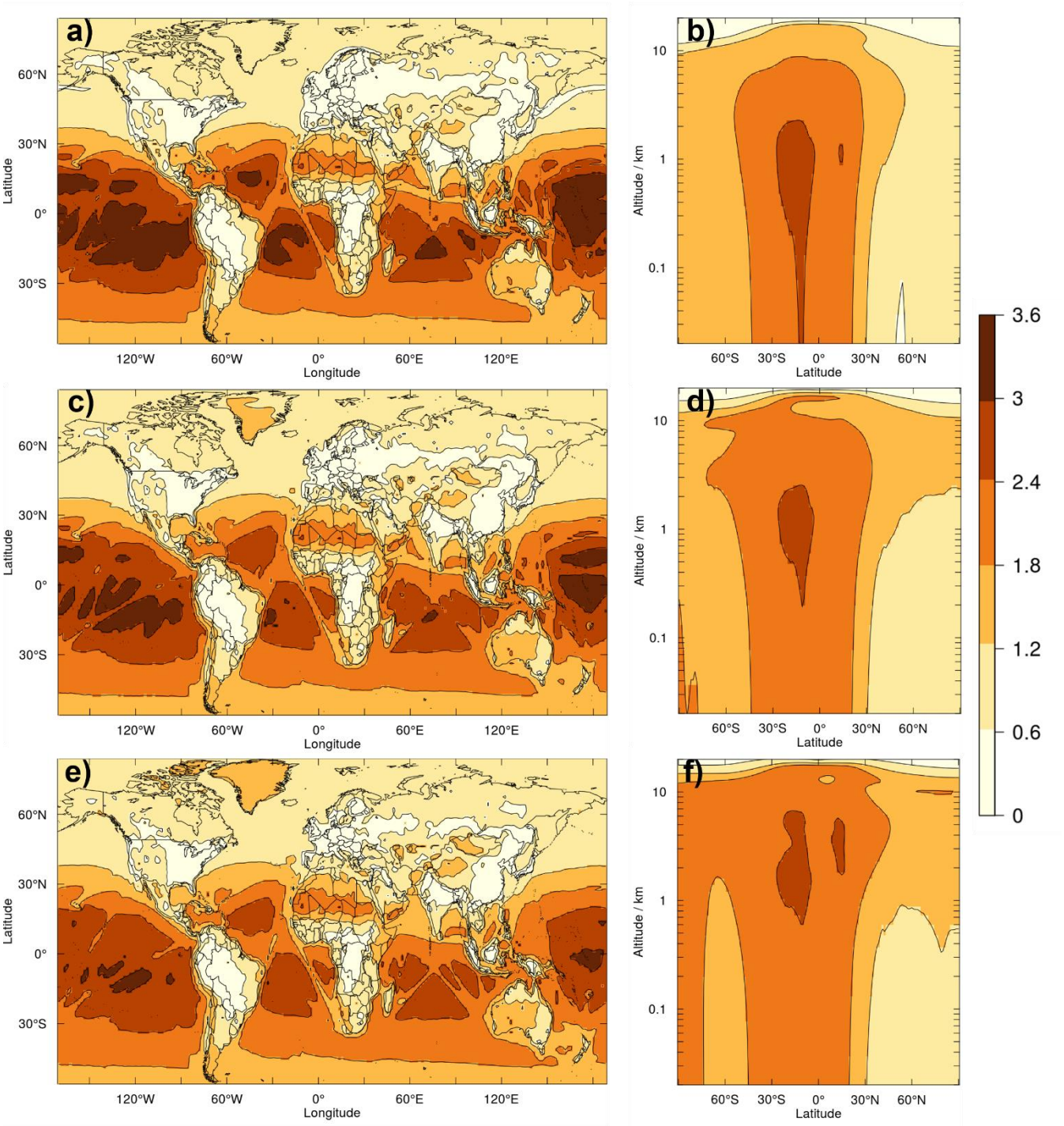

Figure S14: Mean percentage changes in k_{OH} (in %) from run 1 (a and b, top row), run 2 (c and d, middle row), and run 3 (e and f, bottom row), relative to the base run. Changes are shown for the surface (left column) and as a zonal mean (right column).