Supplementary material

Tables S1, S2 and S3 below report the values of OH production and loss fluxes in the whole troposphere, 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,NO_3, 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,NO_3, 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,NO_3, 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}.

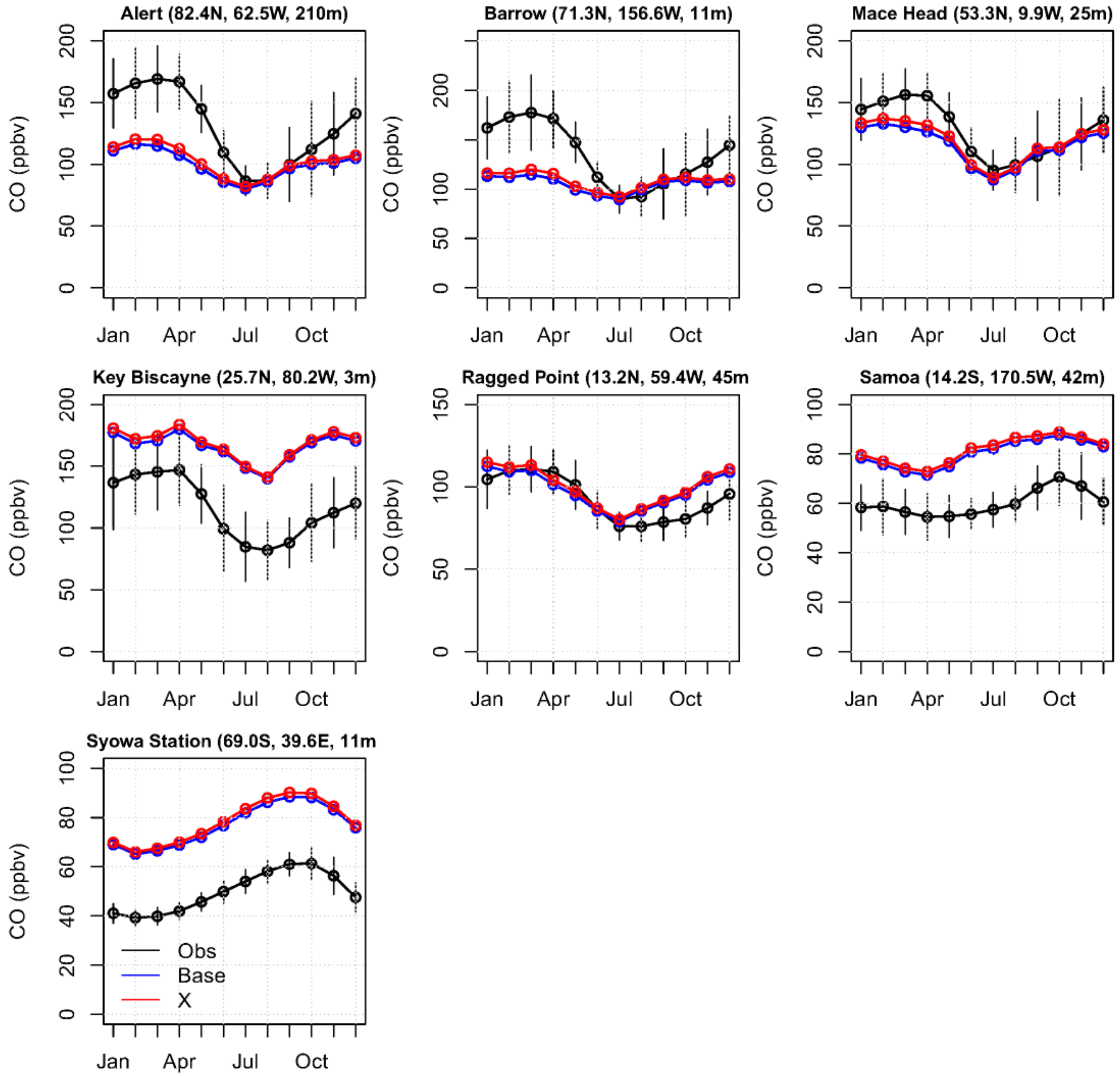

Figure S1: Comparison of CO observations (black data points and lines) with the base run model (blue points and lines) and the model including OH sink X (red points and lines).

5

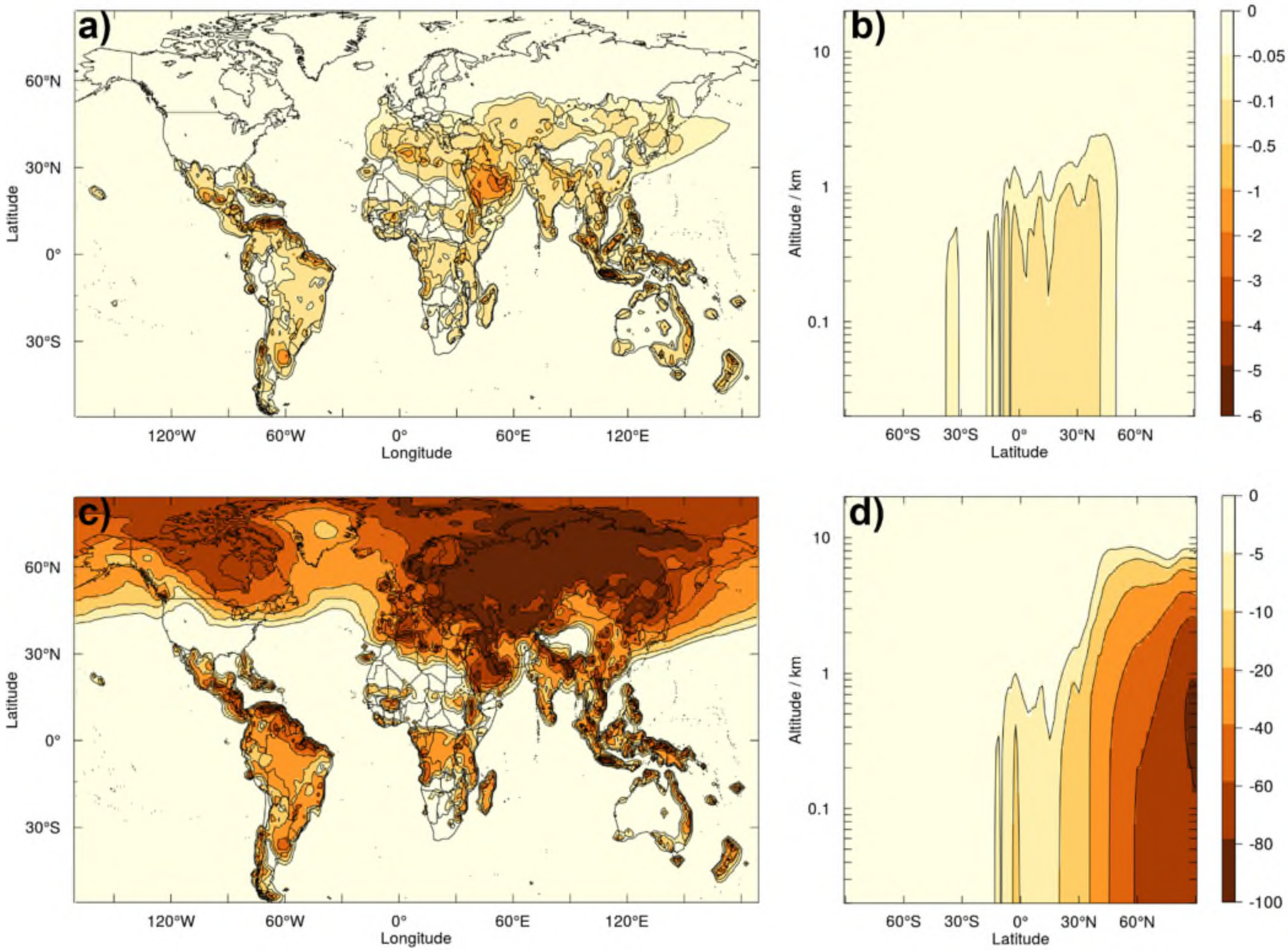

Figure S2: Mean change in OH concentration in the boreal winter (DJF) following inclusion of R3: absolute change in 10^6 molecules cm^{-3} a) at the surface and b) as a zonal mean, and relative percentage change c) at the surface and d) as a zonal mean.

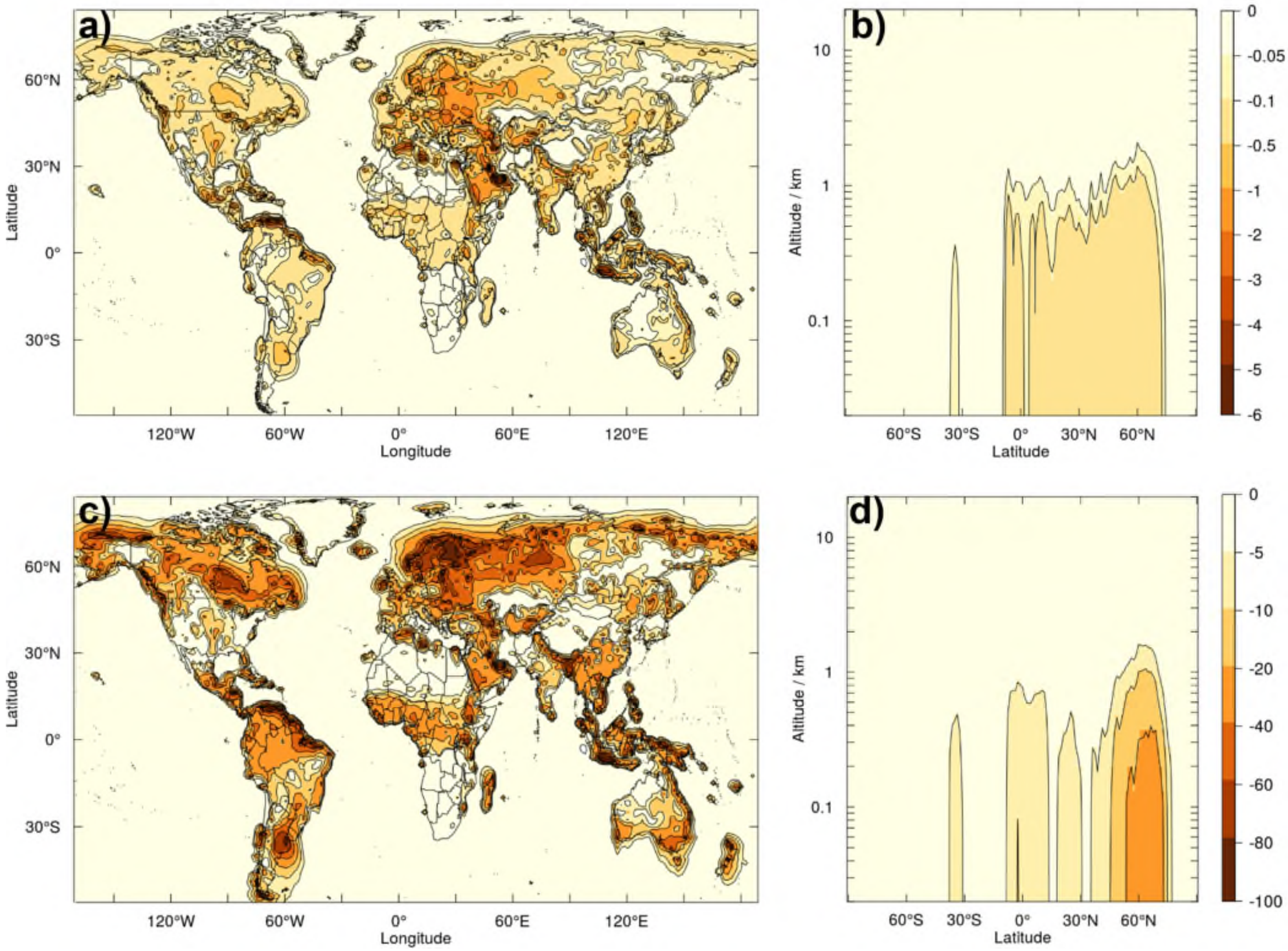

Figure S3: Mean change in OH concentration in the boreal summer (JJA) following inclusion of R3: absolute change in 10^6 molecules cm^{-3} a) at the surface and b) as a zonal mean, and relative percentage change c) at the surface and d) as a zonal mean.

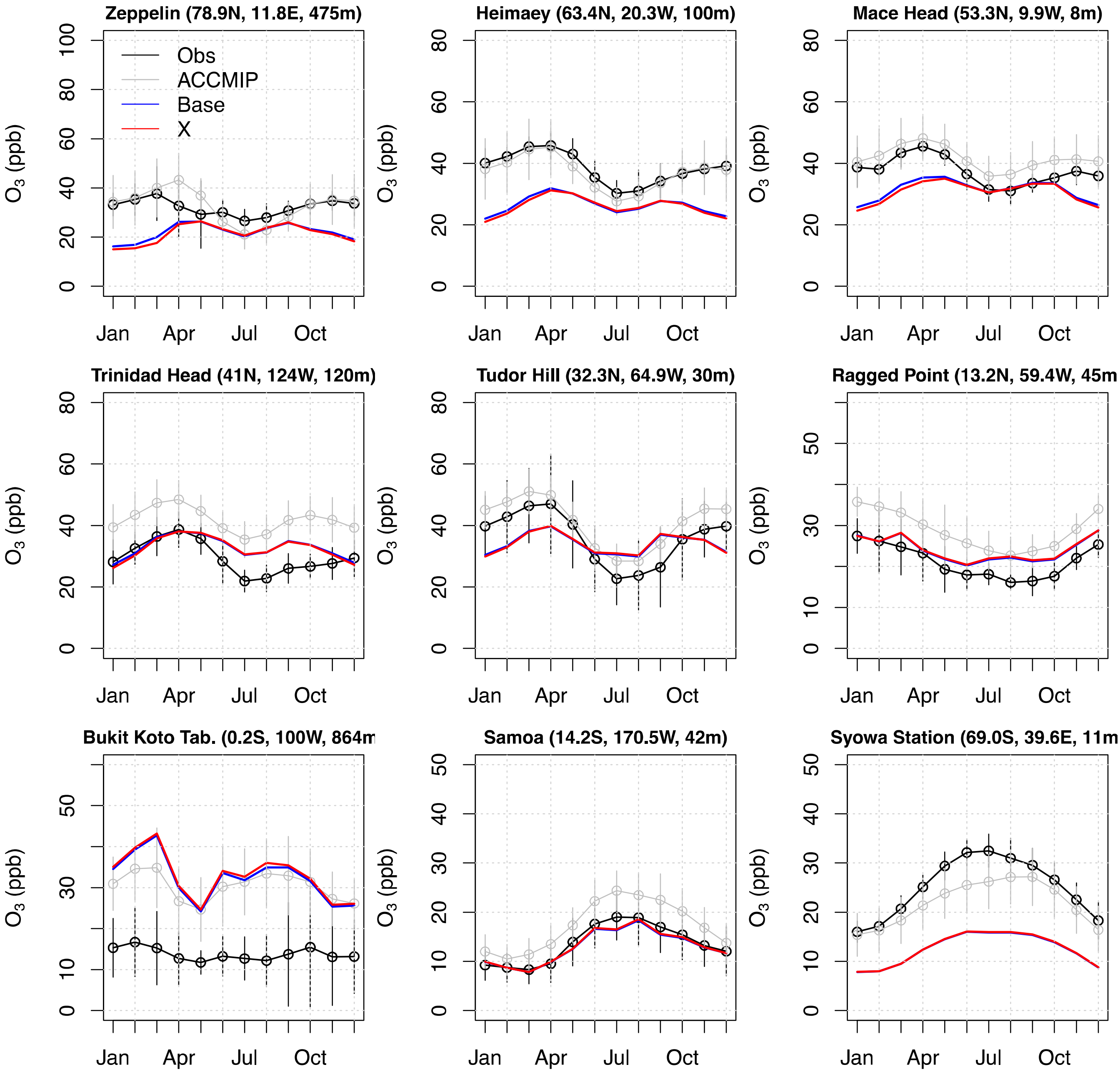

Figure S4: Comparison of ozone observations (black data points and lines) with the base run model (red points and lines) and the model including OH sink X (blue points and lines). The multi model mean results from the ACCMIP year 2000 simulations are shown in grey with the grey bars indicating one standard deviation of the ACCMIP multi model ensemble (Young et al., 2013).

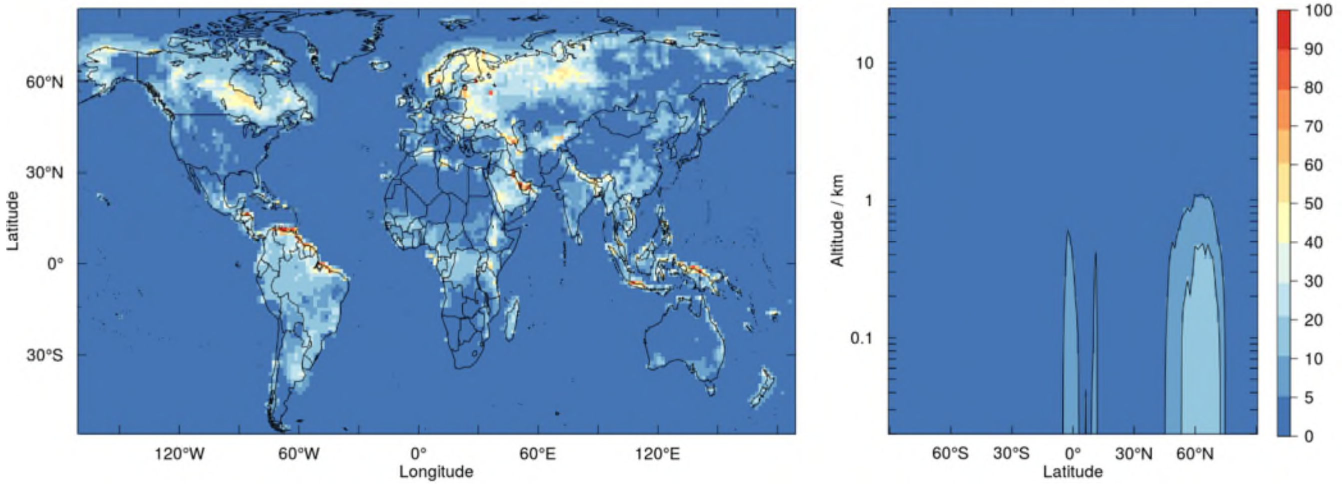

Figure S5: Percentage contribution of the flux through R3 (X + OH) to the total OH loss flux at the surface (left) and as zonal mean (right).