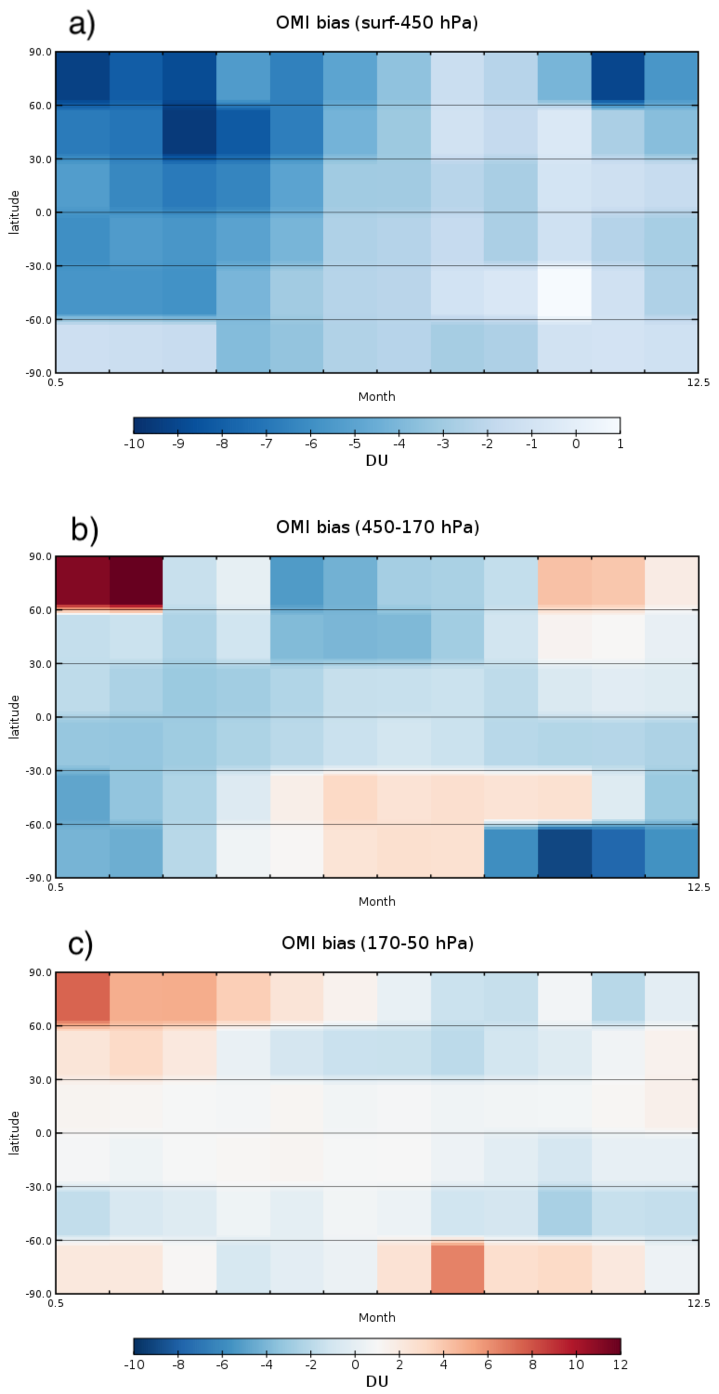
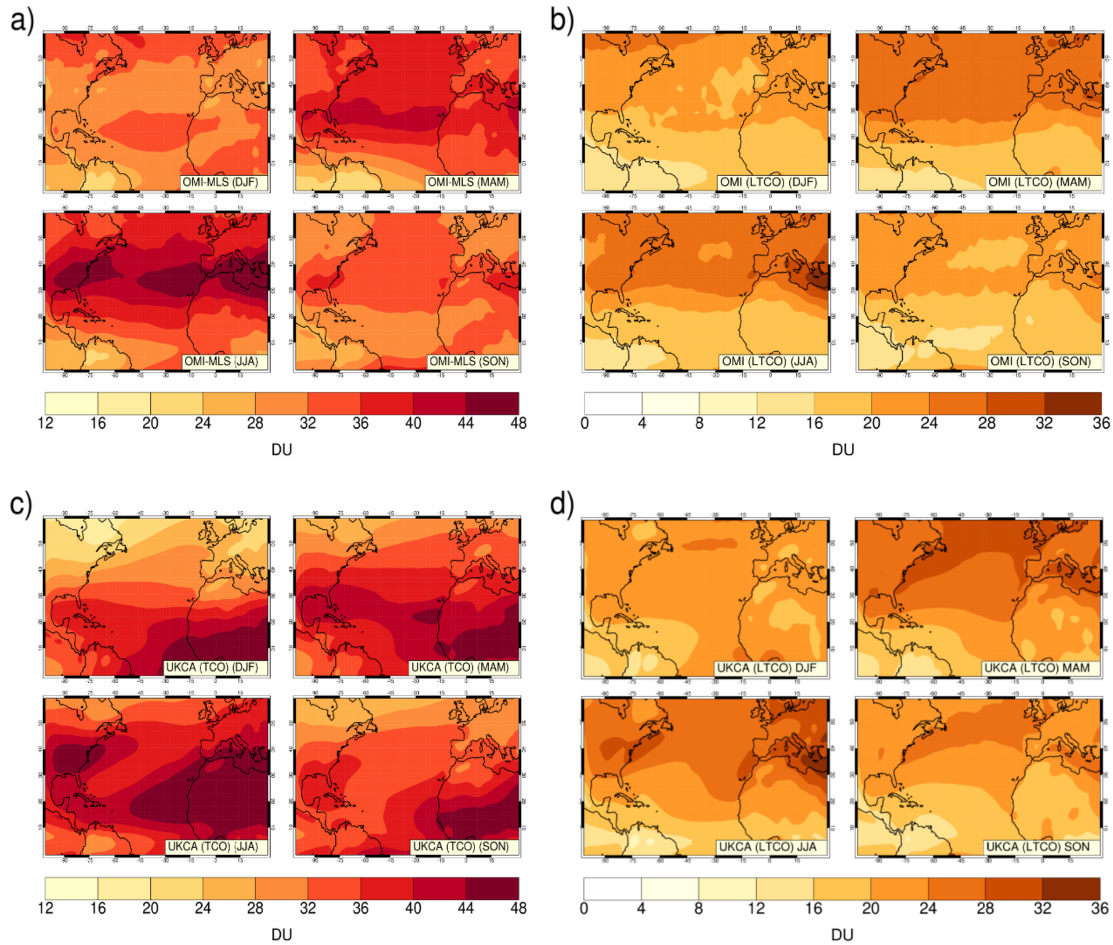
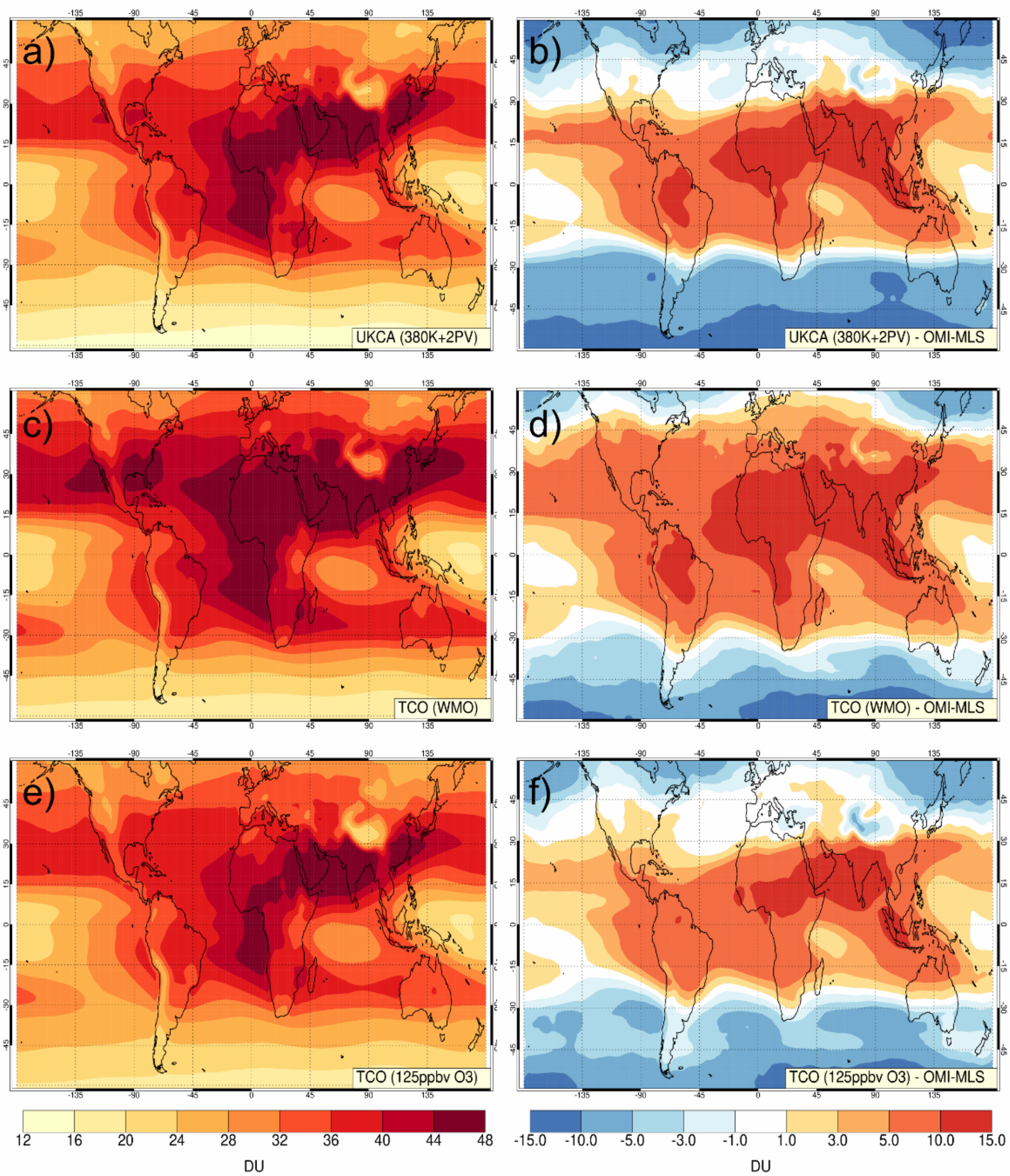


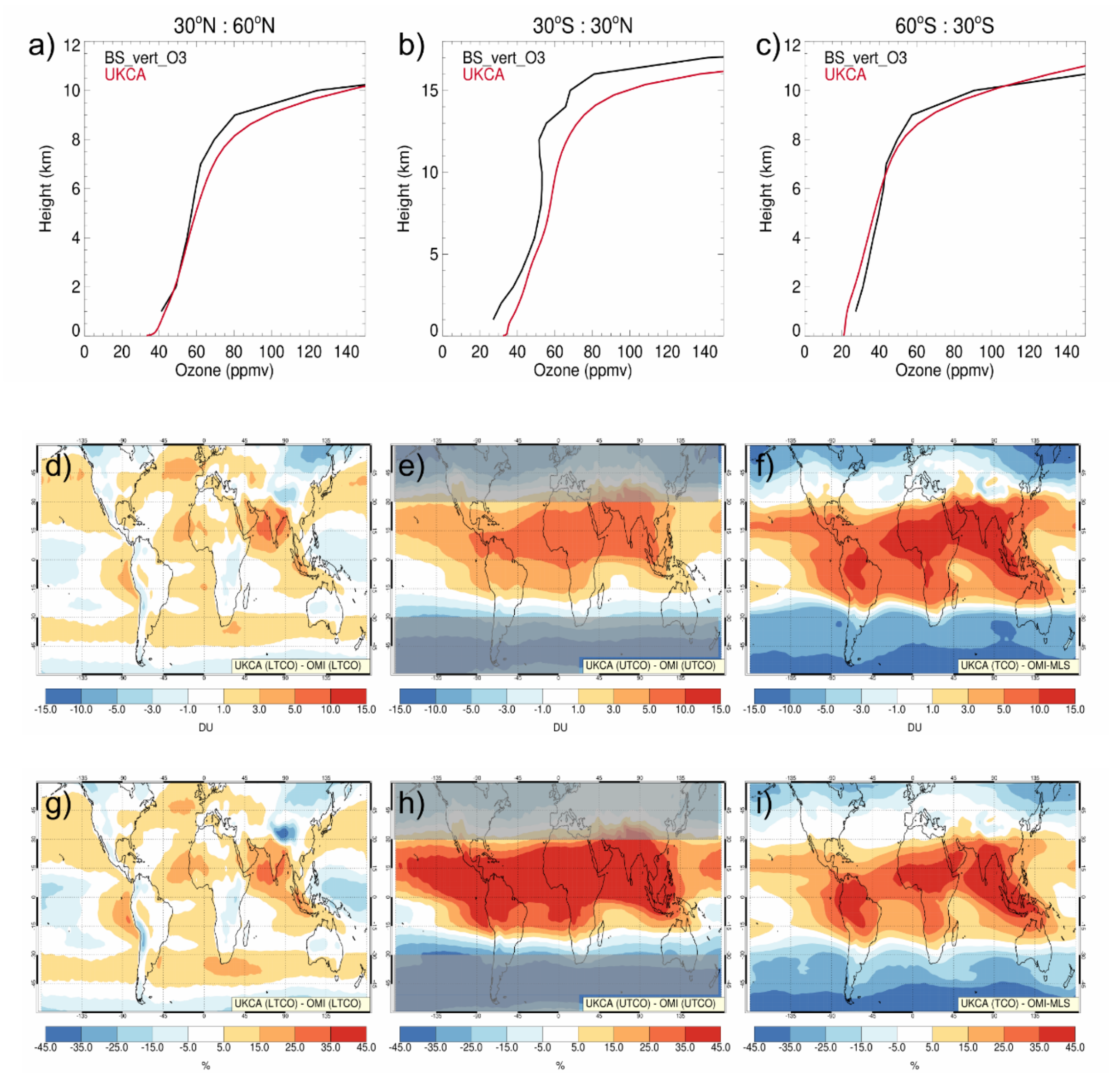
Supplementary Figure 1: OMI bias relative to ozone sonde ensemble as a function of latitude and month for surf-450 hPa column (a), 450-170 hPa column (b), 170-50 hPa column (c).



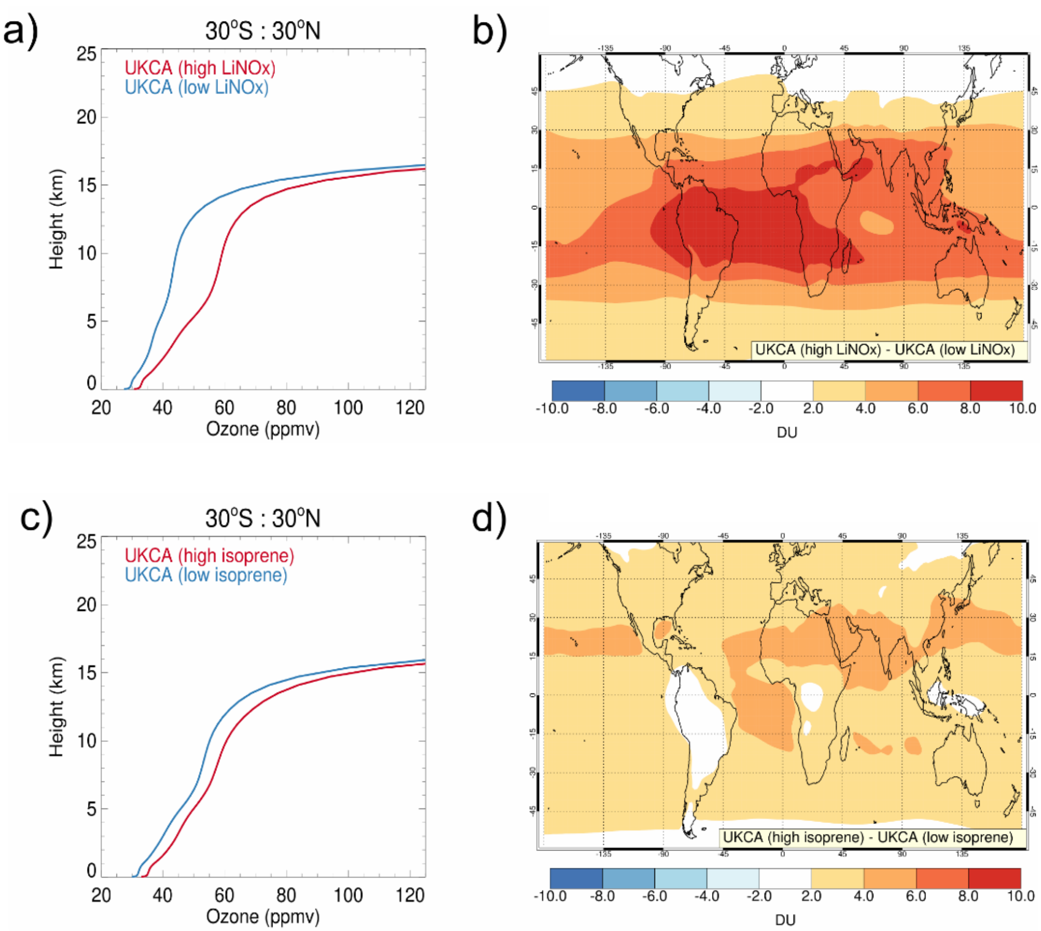
Supplementary Figure 2: TCO and LTCO multi annual seasonal mean maps for the period 2005-2018. OMI-MLS TCO (a), OMI LTCO (b), UKCA TCO (c), and UKCA LTCO (d).



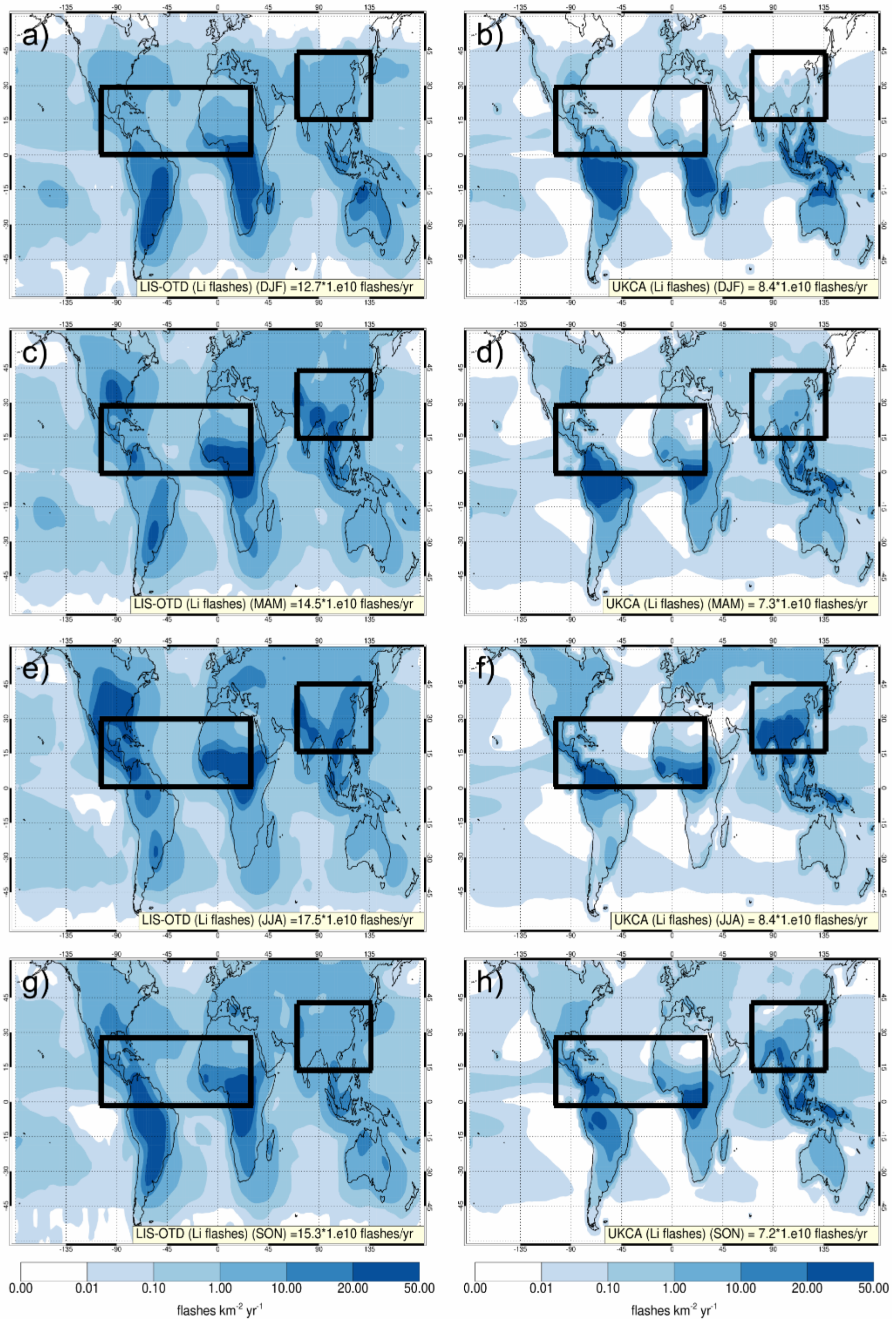
Supplementary Figure 3: Sensitivity of Tropospheric ozone column to tropopause definition for UKCA multiannual mean (2005-2018). Left column shows multiannual means and right column shows difference between modelled TCO and OMI-MLS TCO. The tropopause is defined as follow: a) and b) 380K + 2PV (ozone burden 60º S:60º N = 301 Tg) ; c) and d) WMO tropopause (ozone burden 60º S:60º N = 332 Tg); e) and f) 125ppbv ozoneapause (ozone burden 60º S:60º N =308 Tg). OMI-MLS ozone burden 60º S:60º N = 297 Tg



Supplementary Figure 4: Global Tropospheric ozone. Top row shows zonally averaged, multiannual mean vertical profiles of modelled ozone compared to Bodeker Scientific vertical ozone dataset: a) northern mid latitudes, b) Tropics, c) southern mid latitudes. Second row shows multi annual mean absolute difference maps between modelled and observed ozone in three different vertical columns: d) LTCO, e) UTCO, f) TCO. Third row is the same as second row but for percentage difference. Note that UTCO data is shaded outside the Tropics since this ozone subcolumn, at mid latitudes, samples the lower stratosphere as well as the upper troposphere.



Supplementary Figure 5: First column shows comparison of mean ozone vertical profiles, averaged over the Tropics, for: a) a UKCA simulation with high LiNOx emissions (~7 Tg(N) $^{-1}$yr) and a UKCA simulation with low LiNOx emissions (~2 Tg(N) yr^{-1}); c) a UKCA simulation with high isoprene emissions (~800 Tg yr^{-1}) and a UKCA simulation with low isoprene emissions (~260 Tg yr^{-1}). Second column shows difference between sensitivity experiments (same as first column) but as a difference in TCO. Note that in the control simulation LiNOx emissions are ~ 6 Tg(N) yr^{-1} and isoprene emissions are ~530 Tg yr^{-1}. The difference in global ozone burden between high and low sensitivity experiments is ~50 Tg (for LiNOx) and 30 Tg (for isoprene).



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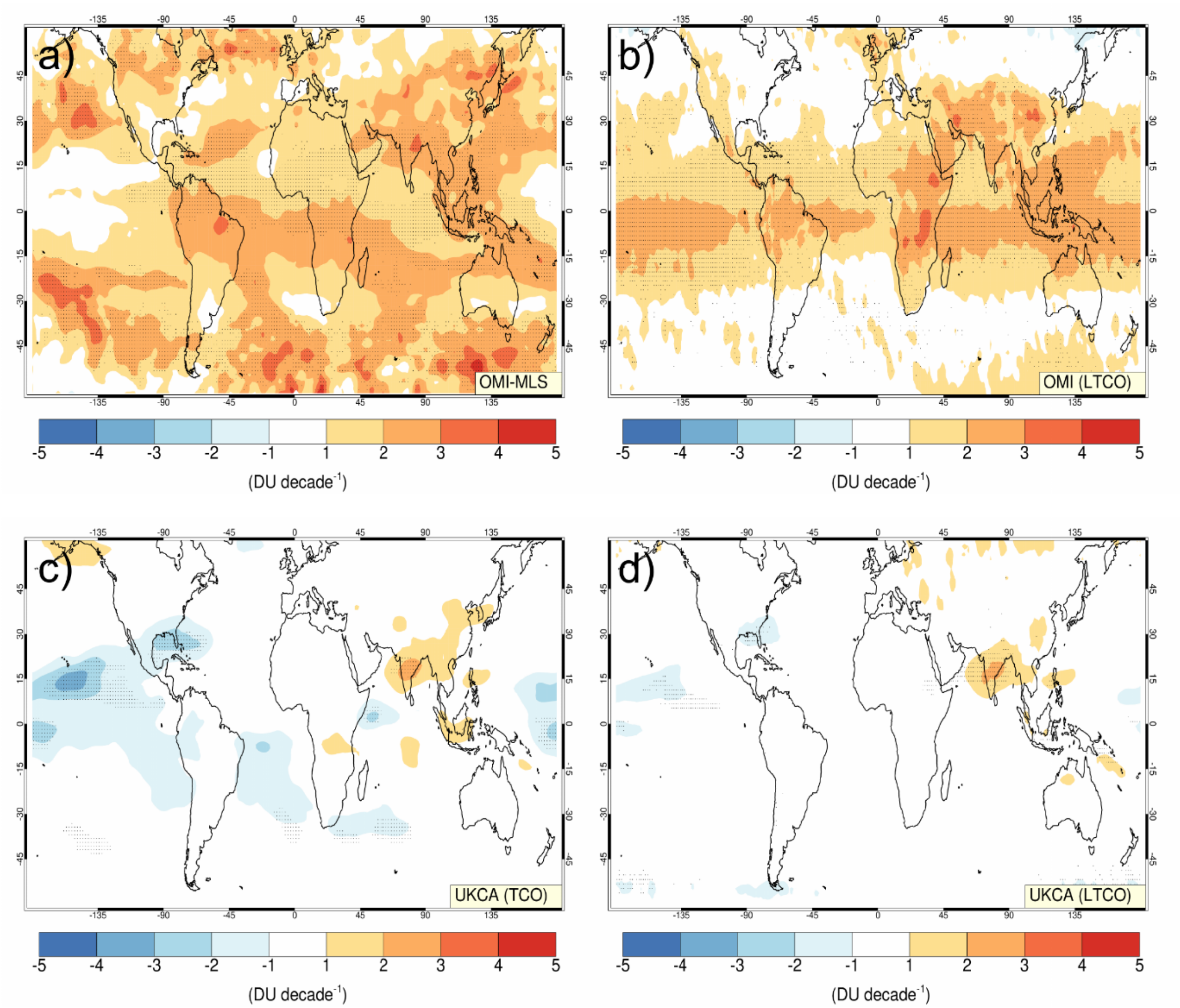
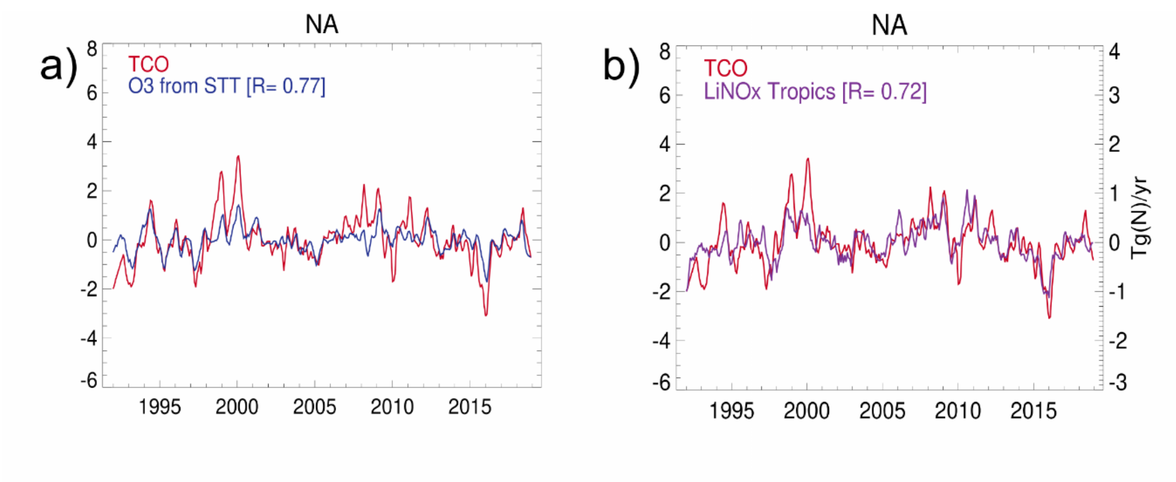
Supplementary Figure 6: Seasonal maps of observed (first column) and modelled (second column) lightning flashes for the period 1996-2013. Top row to bottom row respectively for DJF, MAM, JJA, SON.



Supplementary Figure 7: Tropospheric ozone trends for the period 2005-2018. OMI-MLS TCO trend (a); OMI LTCO trend (b); UKCA TCO trend (c) and UKCA LTCO trend (d). The stippling indicates where trends are significant to the 95 % confidence level, based on the standard error of the residuals.



Supplementary Figure 8: deseasonalised and detrended timeseries anomalies for the period 1992-2018, showing temporal correlation (R) between modelled TCO in the North Atlantic and its key drivers: ozone from STT in the North Atlantic and LiNOx emissions in the Tropics.

CHEMICAL FIELD	1992-2001 TREND			1998-2006 TREND			2005-2018 TREND		
	NA	NH	Tropics	NA	NH	Tropics	NA	NH	Tropics
UKCA TCO DU decade-1	2.3(0.8)	2.1(0.7)	2.8(0.8)	-1.2(0.8)	-0.8(0.7)	-1.3(0.9)	-0.3(0.4)	-0.1(0.4)	-0.5(0.6)
LiNOx ems Tg(N) decade-1	-0.1(0.1)	0.2(0.2)	**0.8(0.3)**	0.0(0.1)	-0.1(0.2)	**0.0(0.3)**	-0.1(0.1)	0.0(0.1)	**-0.1(0.2)**
Isoprene ems Tg decade-1	-7(6)	-3(10)	61(14)	-3.4(6.8)	-4(14)	-13(19)	-8.5(3.2)	-15(5)	- 31(10)
Surf NOx ems Tg(N) decade-1	0.2(0.4)	1.7(0.7)	4.1(1.6)	-1.7(0.5)	4.5(1.3)	3.8(1.4)	-2.5(0.2)	0.2(0.4)	2.3(0.5)
Ozone from STT DU decade-1	**0.9(0.5)**	**0.9(0.4)**	0.7(0.4)	**-0.5(0.4)**	**-0.6(0.3)**	-0.9(0.3)	**0.1(0.3)**	**0.0(0.2)**	-0.2(0.2)

Supplementary Table 1: same as Table 2 but showing absolute trend values and the error of the trend estimate (in brackets).