



Supplement of

Rapid O₃ assimilations – Part 2: Tropospheric O₃ changes accompanied by declining NO_x emissions in the USA and Europe in 2005–2020

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Supplemental Information

Uncertainty Analysis

The uncertainties in the averages of surface and tropospheric column O₃ concentrations are calculated using the bootstrapping method (Efron, 1992):

1. A bootstrap sample is generated by randomly drawing N data points from the full set of N data points. For example, the OMI O₃ data was averaged to produce a dataset with 2866 points (grids with 0.5°×0.625° spatial resolution over the US); N = 2866 data points were then randomly drawn from the dataset; Average of OMI O₃ over the US (for example, 2005) is calculated as the mean value of the randomly drawn N = 2866 data points.
2. Step 1 are repeated by 1000 times. Because the averages are based on the randomly drawn data points, we have different averages for the same year (for example, 2005). The standard deviation of the ensemble of averages represent the uncertainty in the average.
3. The above process is repeated to produce the standard deviations of averages of OMI O₃ over the US for various years (i.e. 2006, 2007, ... 2020).

The trends and uncertainties in the trends of surface and tropospheric column O₃ concentrations are calculated using the linear fitting of averages by using the least squares method.

Reference:

Efron, B.: Bootstrap Methods: Another Look at the Jackknife. In: Kotz, S., Johnson, N.L. (eds) Breakthroughs in Statistics. Springer Series in Statistics. Springer, New York, NY, 1992.

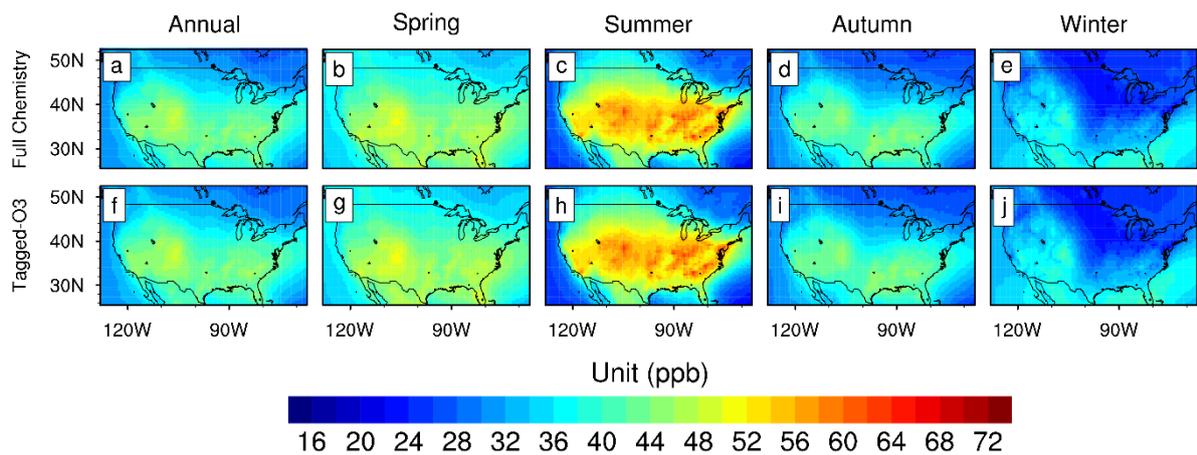


Fig. S1. Surface MDA8 O₃ over the US in 2005-2020 (annual and seasonal averages) simulated by GEOS-Chem model with (a-e) full chemistry mode; (f-j) single O₃ tracer (tagged-O₃) mode.

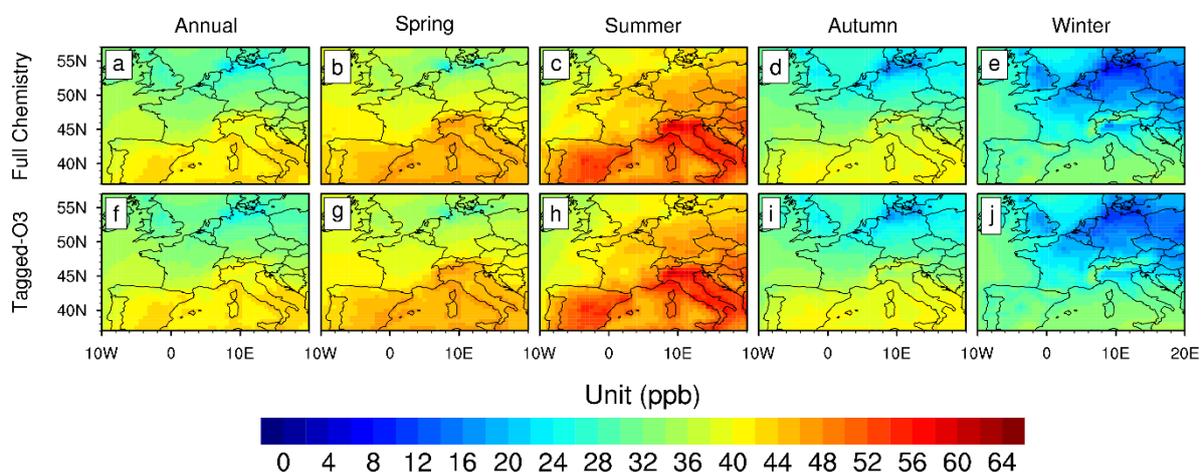


Fig. S2. Surface MDA8 O₃ over Europe in 2005-2020 (annual and seasonal averages) simulated by GEOS-Chem model with (a-e) full chemistry mode; (f-j) single O₃ tracer (tagged-O₃) mode.

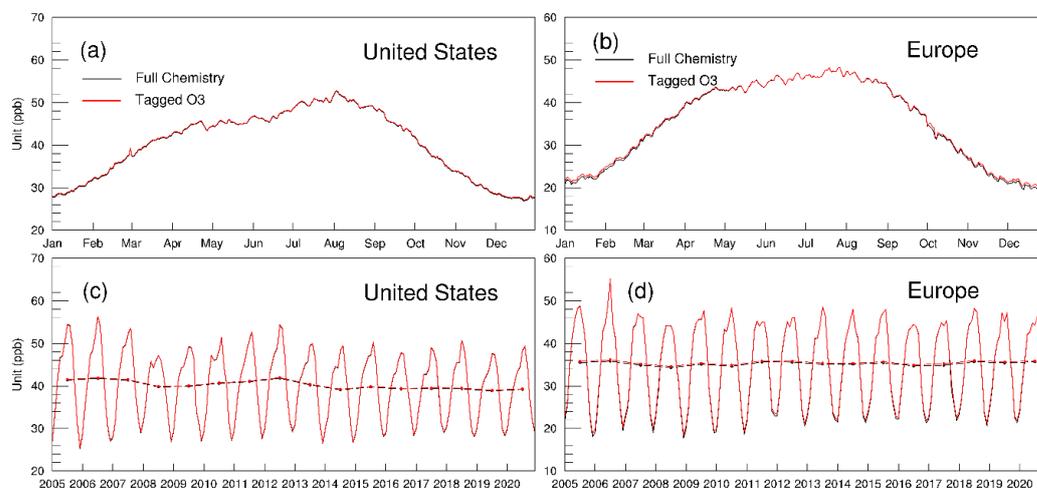


Fig. S3. (a-b) Daily averages of surface MDA8 O₃ over the US and Europe in 2005-2020 from GEOS-Chem full chemistry and single O₃ tracer (tagged-O₃) simulations; (c-d) Monthly averages of MDA8 O₃. The dashed lines in panels c-d are annual averages.

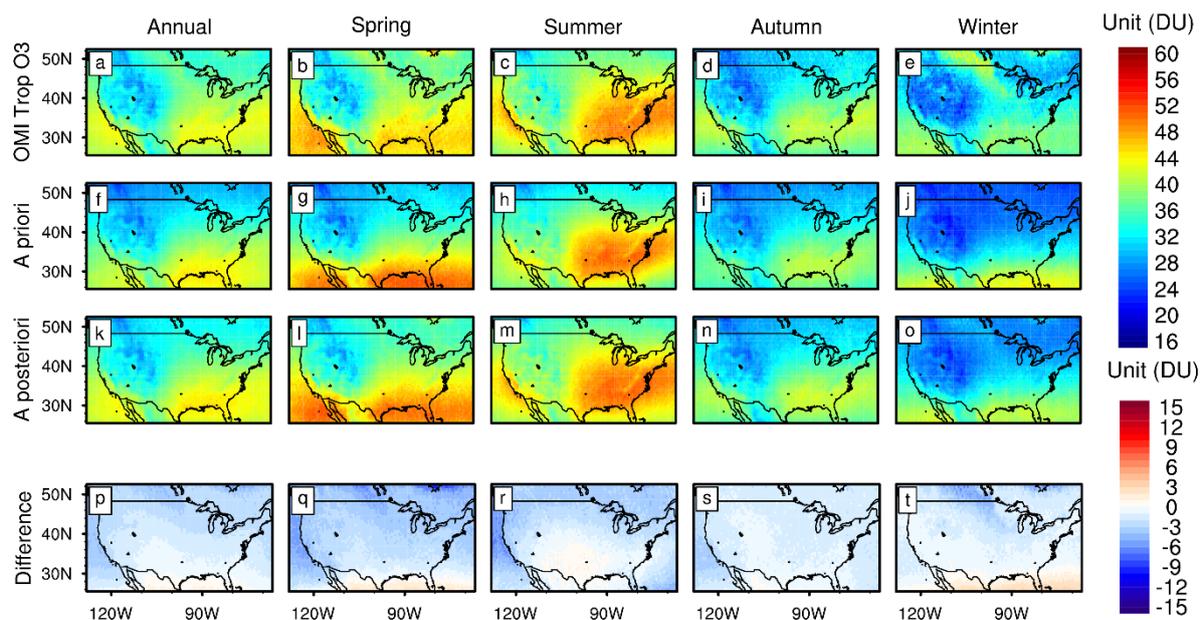


Fig. S4. Tropospheric O₃ columns over the US in 2005-2020 (annual and seasonal averages) from (a-e) OMI observations; (f-j) GEOS-Chem a priori simulation; (k-o) GEOS-Chem a posteriori simulation by assimilating OMI O₃ observations. (p-t) Bias in the a priori simulations calculated by a priori minus a posteriori tropospheric O₃ columns. The output O₃ profiles are convolved with OMI averaging kernels.

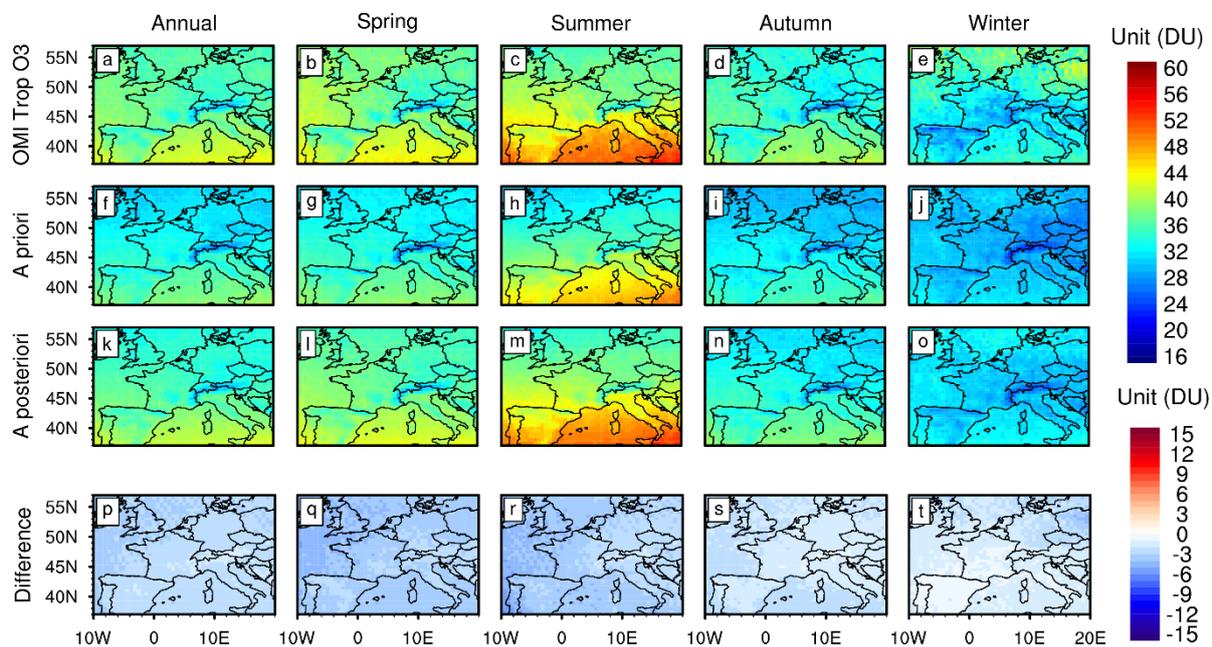


Fig. S5. Tropospheric O₃ columns over Europe in 2005-2020 (annual and seasonal averages) from (a-e) OMI observations; (f-j) GEOS-Chem a priori simulation; (k-o) GEOS-Chem a posteriori simulation by assimilating OMI O₃ observations. (p-t) Bias in the a priori simulations calculated by a priori minus a posteriori tropospheric O₃ columns. The output O₃ profiles are convolved with OMI averaging kernels.

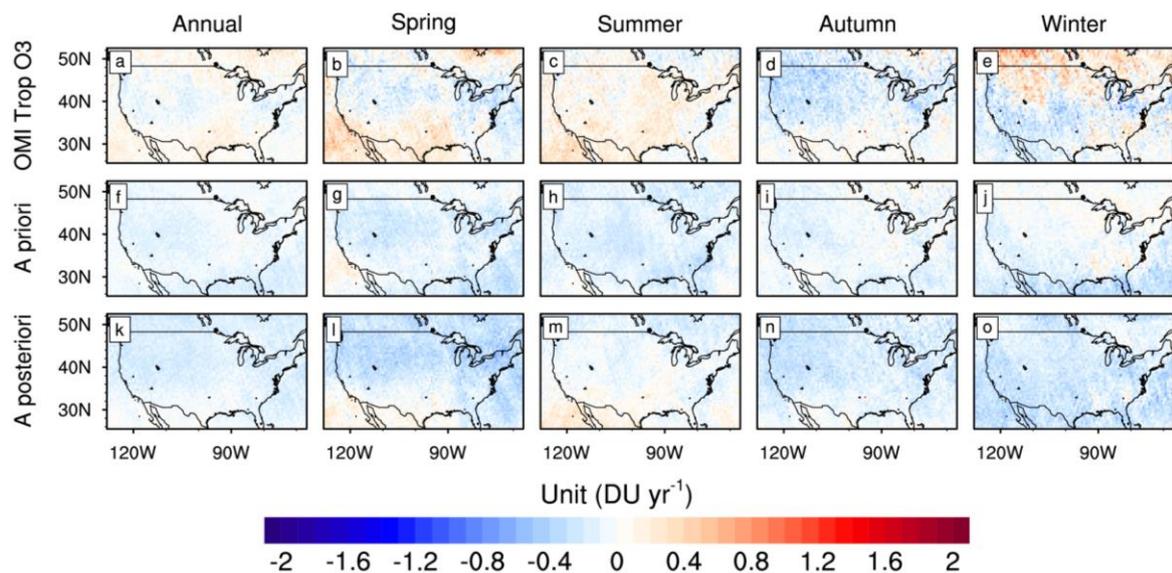


Fig. S6. Trends of tropospheric O₃ columns over the US in 2005-2020 (annual and seasonal averages) from (a-e) OMI observations; (f-j) GEOS-Chem a priori simulation; (k-o) GEOS-Chem a posteriori simulation by assimilating OMI O₃ observations. The output O₃ profiles are convolved with OMI averaging kernels.

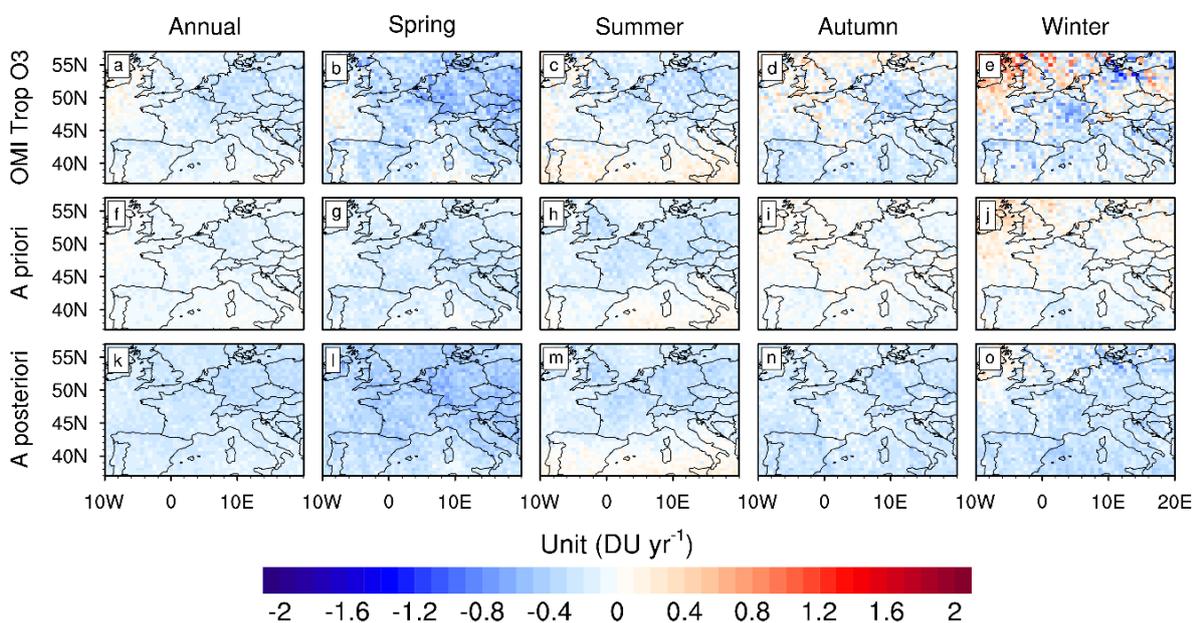


Fig. S7. Trends of tropospheric O₃ columns over Europe in 2005-2020 (annual and seasonal averages) from (a-e) OMI observations; (f-j) GEOS-Chem a priori simulation; (k-o) GEOS-Chem a posteriori simulation by assimilating OMI O₃ observations. The output O₃ profiles are convolved with OMI averaging kernels.

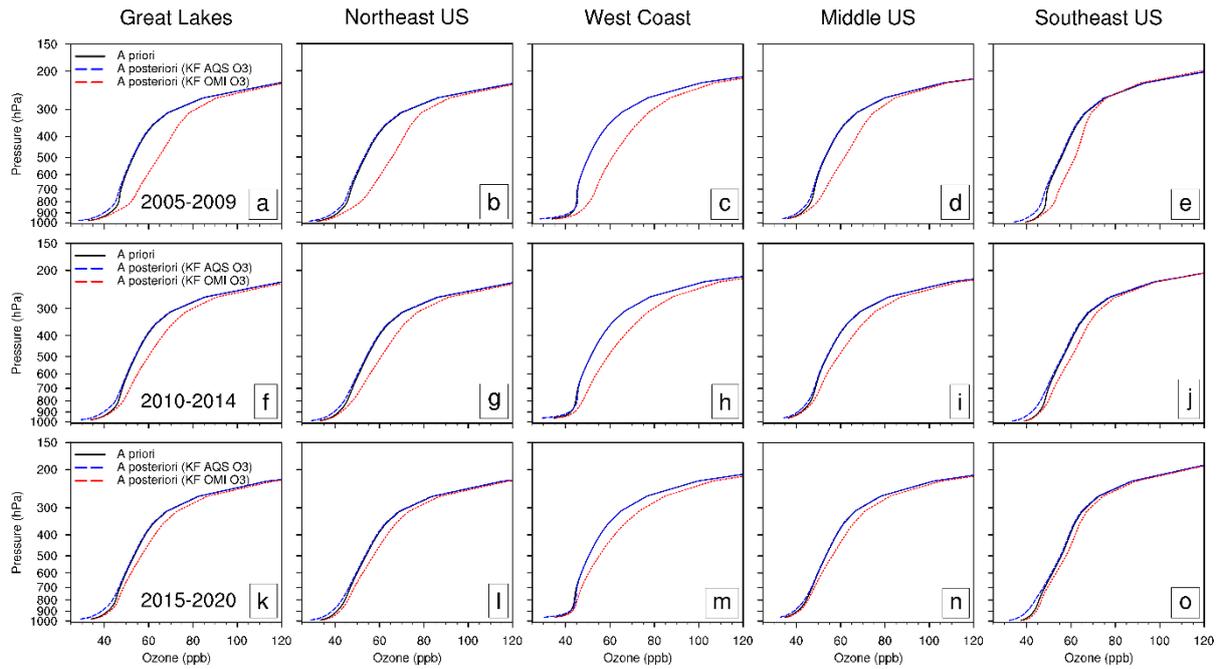


Fig. S8. Averages of O₃ vertical profiles in 2005-2020 in the US from GEOS-Chem a priori (black) and a posteriori simulations by assimilating AQS (blue) and OMI (red) O₃ observations.

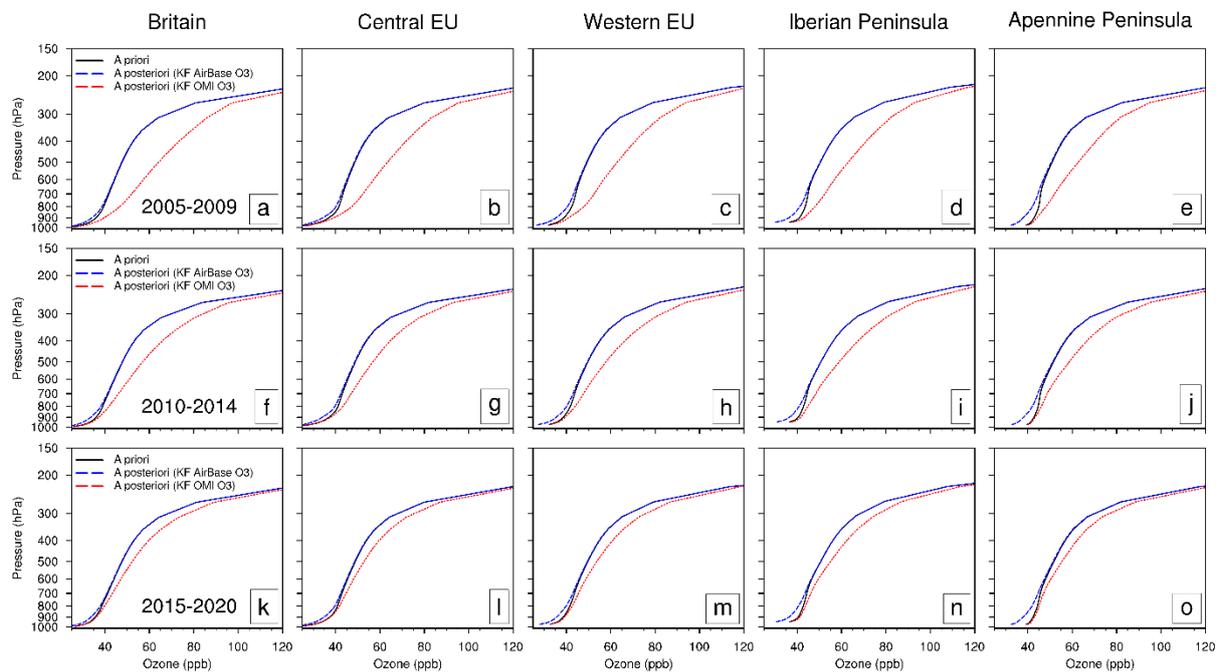


Fig. S9. Averages of O₃ vertical profiles in 2005-2020 in Europe from GEOS-Chem a priori (black) and a posteriori simulations by assimilating AirBase (blue) and OMI (red) O₃ observations.

#1 Great Lakes			Annual		Spring		Summer		Autumn		Winter	
			Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend
TS1.1 surface (sampled)	2005- 2020	AQS	38.2±0.3	-0.11±0.08	44.5±0.2	-0.13±0.09	48.6±0.4	-0.57±0.18	33.8±0.3	-0.24±0.18	26.2±0.7	0.45±0.07
		a priori	40.0±0.3	-0.04±0.06	43.2±0.2	-0.17±0.08	55.7±0.4	-0.45±0.19	37.1±0.4	0.09±0.10	24.1±0.3	0.35±0.06
		KF-AQS	37.7±0.2	-0.07±0.06	43.5±0.2	-0.13±0.08	49.0±0.3	-0.51±0.18	33.1±0.2	-0.03±0.12	25.4±0.2	0.39±0.06
TS1.2 surface	2005- 2020	a priori	40.2±0.2	-0.06±0.06	43.4±0.2	-0.18±0.08	55.7±0.4	-0.47±0.18	37.4±0.3	0.09±0.10	24.8±0.2	0.27±0.07
		KF-AQS	37.9±0.1	-0.09±0.06	43.5±0.1	-0.14±0.08	49.1±0.2	-0.52±0.17	33.5±0.2	-0.04±0.12	25.7±0.2	0.30±0.05
TS1.3 trop. column (convolved)	2005- 2020	OMI	39.3±0.1	-0.09±0.06	40.6±0.1	-0.16±0.11	46.9±0.1	0.12±0.12	36.7±0.2	-0.17±0.05	32.8±0.2	-0.15±0.15
		a priori	36.9±0.2	-0.11±0.05	37.5±0.2	-0.16±0.10	45.9±0.2	-0.13±0.10	35.0±0.2	-0.09±0.04	29.1±0.1	-0.03±0.05
		KF-OMI	38.8±0.1	-0.21±0.04	40.4±0.1	-0.31±0.10	47.7±0.1	-0.03±0.09	36.3±0.1	-0.23±0.03	30.4±0.1	-0.24±0.05
TS1.4 trop. Column	2005- 2020	a priori	37.4±0.2	-0.03±0.05	38.1±0.2	-0.12±0.10	45.9±0.2	-0.13±0.14	35.8±0.2	0.03±0.09	30.1±0.2	0.12±0.08
		KF-AQS	36.8±0.2	-0.03±0.05	37.9±0.2	-0.10±0.09	44.3±0.2	-0.13±0.14	35.0±0.2	0.01±0.09	30.1±0.2	0.12±0.08
		KF-OMI	40.2±0.1	-0.29±0.05	42.5±0.1	-0.44±0.09	48.3±0.2	-0.09±0.12	37.8±0.1	-0.23±0.10	32.2±0.2	-0.39±0.11
	2005- 2009	KF-AQS	36.6±0.2	-0.48±0.16	38.0±0.2	-0.32±0.39	44.1±0.2	-1.57±0.12	34.9±0.2	-0.37±0.72	29.3±0.2	-0.10±0.29
		KF-OMI	41.8±0.1	-0.42±0.29	44.5±0.1	-0.29±0.35	47.6±0.1	-1.30±0.15	39.4±0.1	-0.52±0.82	35.3±0.1	-0.47±0.44
	2010- 2014	KF-AQS	37.1±0.2	-0.63±0.24	38.4±0.2	-1.15±0.74	45.5±0.2	-1.95±0.30	34.7±0.2	-0.11±0.47	30.1±0.2	-0.06±0.88
		KF-OMI	40.2±0.1	-0.97±0.22	43.1±0.1	-1.45±0.73	49.6±0.2	-1.77±0.23	37.0±0.2	-0.41±0.50	30.7±0.2	-0.49±0.76
	2015- 2020	KF-AQS	36.7±0.2	0.05±0.12	37.4±0.1	-0.04±0.29	43.7±0.2	0.38±0.47	35.3±0.1	-0.19±0.43	30.8±0.2	0.11±0.26
KF-OMI		38.9±0.1	-0.15±0.12	40.2±0.1	-0.41±0.34	47.7±0.2	0.16±0.37	37.1±0.1	-0.35±0.37	30.9±0.2	0.02±0.23	

Table. S1. Averages (with units ppb or DU) and trends (with units ppb yr⁻¹ or DU yr⁻¹) of surface and tropospheric column O₃ concentrations in 2005-2020 over Great Lakes (Region #1) from observations (AQS and OMI) and a priori and a posteriori (KF) simulations. Grids with low and medium anthropogenic NO_x emissions are excluded in Great Lakes.

#2 Northeast US			Annual		Spring		Summer		Autumn		Winter	
			Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend
TS2.1 surface (sampled)	2005- 2020	AQS	37.4±0.4	-0.07±0.06	43.9±0.3	-0.11±0.07	47.1±0.6	-0.61±0.13	31.9±0.3	-0.05±0.14	27.2±0.6	0.42±0.08
		a priori	38.1±0.4	0.03±0.04	41.6±0.3	-0.02±0.05	52.2±1.0	-0.41±0.12	34.7±0.4	0.20±0.09	24.1±0.3	0.33±0.06
		KF-AQS	36.4±0.2	-0.04±0.05	42.4±0.2	-0.08±0.06	46.3±0.6	-0.52±0.11	31.0±0.3	0.05±0.12	26.2±0.3	0.36±0.06
TS2.2 surface	2005- 2020	a priori	38.4±0.4	0.01±0.04	41.9±0.3	-0.05±0.05	52.4±0.7	-0.42±0.12	34.9±0.3	0.19±0.09	24.7±0.3	0.28±0.06
		KF-AQS	36.6±0.2	-0.06±0.05	42.5±0.2	-0.11±0.06	46.4±0.7	-0.53±0.11	31.2±0.3	0.03±0.12	26.5±0.2	0.32±0.05
TS2.3 trop. column (convolved)	2005- 2020	OMI	39.3±0.1	-0.12±0.04	40.8±0.2	-0.21±0.09	47.3±0.2	0.02±0.10	37.2±0.2	-0.21±0.06	31.9±0.2	-0.08±0.11
		a priori	37.0±0.2	-0.11±0.04	37.1±0.2	-0.18±0.09	45.8±0.3	-0.13±0.09	35.3±0.2	-0.11±0.04	29.6±0.1	0.00±0.04
		KF-OMI	38.8±0.2	-0.22±0.03	40.1±0.2	-0.36±0.08	48.0±0.3	-0.06±0.08	36.7±0.2	-0.27±0.04	30.5±0.2	-0.17±0.04
TS2.4 trop. Column	2005- 2020	a priori	37.4±0.2	-0.02±0.04	38.0±0.2	-0.07±0.09	45.3±0.3	-0.17±0.12	35.9±0.2	0.04±0.07	30.5±0.2	0.14±0.08
		KF-AQS	36.8±0.2	-0.02±0.04	37.9±0.2	-0.07±0.09	43.7±0.2	-0.18±0.11	35.1±0.2	0.02±0.07	30.6±0.2	0.14±0.08
		KF-OMI	40.3±0.2	-0.31±0.04	42.9±0.1	-0.48±0.09	47.9±0.2	-0.12±0.11	37.8±0.2	-0.21±0.07	32.7±0.2	-0.39±0.12
	2005- 2009	KF-AQS	36.6±0.2	-0.33±0.05	37.7±0.2	0.00±0.16	43.9±0.2	-1.21±0.22	34.9±0.2	-0.67±0.48	30.0±0.2	-0.07±0.46
		KF-OMI	42.0±0.2	-0.33±0.20	45.1±0.1	-0.11±0.23	47.6±0.2	-0.89±0.39	39.1±0.2	-0.74±0.54	36.2±0.2	-0.63±0.47
	2010- 2014	KF-AQS	37.1±0.2	-0.51±0.16	38.6±0.2	-0.91±0.81	44.7±0.3	-1.96±0.13	35.0±0.2	-0.15±0.38	30.2±0.2	-0.03±0.76
		KF-OMI	40.4±0.2	-0.86±0.12	43.9±0.2	-1.16±0.78	48.9±0.3	-1.92±0.11	37.3±0.2	-0.42±0.32	30.6±0.2	-0.46±0.67
	2015- 2020	KF-AQS	36.6±0.2	0.10±0.13	37.4±0.2	-0.07±0.31	42.8±0.2	0.20±0.32	35.4±0.2	-0.13±0.36	31.4±0.2	0.28±0.27
KF-OMI		38.9±0.2	-0.08±0.15	40.2±0.2	-0.46±0.41	47.2±0.2	0.03±0.22	37.2±0.2	-0.23±0.31	31.5±0.2	0.20±0.25	

Table. S2. Averages (with units ppb or DU) and trends (with units ppb yr⁻¹ or DU yr⁻¹) of surface and tropospheric column O₃ concentrations in 2005-2020 over Northeast US (Region #2) from observations (AQS and OMI) and a priori and a posteriori (KF) simulations. Grids with low and medium anthropogenic NO_x emissions are excluded in Northeast US.

#3 West Coast			Annual		Spring		Summer		Autumn		Winter	
			Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend
TS3.1 surface (sampled)	2005- 2020	AQS	41.0±1.5	0.15±0.05	44.5±1.1	-0.07±0.09	50.8±2.5	-0.15±0.12	41.0±1.3	0.39±0.05	28.1±0.8	0.37±0.06
		a priori	41.6±0.7	-0.01±0.03	43.8±0.7	-0.18±0.06	49.2±1.1	-0.09±0.11	40.8±0.6	0.12±0.04	32.9±0.5	0.08±0.03
		KF-AQS	41.2±1.1	0.11±0.04	44.5±1.0	-0.10±0.08	50.6±1.9	-0.13±0.11	41.4±1.1	0.34±0.04	28.8±0.8	0.29±0.05
TS3.2 surface	2005- 2020	a priori	41.6±0.7	-0.01±0.03	43.8±0.6	-0.18±0.06	49.2±1.2	-0.09±0.11	40.8±0.6	0.12±0.04	32.9±0.5	0.08±0.03
		KF-AQS	41.2±1.1	0.11±0.04	44.5±0.9	-0.10±0.08	50.6±2.2	-0.13±0.11	41.4±1.0	0.34±0.04	28.8±0.9	0.29±0.05
TS3.3 trop. column (convolved)	2005- 2020	OMI	39.2±0.4	0.00±0.08	42.1±0.5	0.21±0.12	43.8±0.4	0.13±0.09	37.9±0.4	-0.15±0.10	32.7±0.5	-0.19±0.16
		a priori	37.5±0.4	-0.09±0.04	39.5±0.4	-0.06±0.07	42.7±0.3	-0.12±0.06	35.6±0.3	-0.09±0.06	31.9±0.4	-0.10±0.08
		KF-OMI	39.8±0.3	-0.14±0.06	43.2±0.5	-0.06±0.09	45.2±0.4	0.02±0.08	37.7±0.4	-0.23±0.07	32.7±0.4	-0.28±0.09
TS3.4 trop. Column	2005- 2020	a priori	38.2±0.4	-0.02±0.04	38.9±0.6	-0.09±0.07	42.5±0.4	-0.15±0.09	36.5±0.3	0.00±0.07	34.9±0.6	0.07±0.09
		KF-AQS	38.1±0.4	-0.01±0.04	38.9±0.6	-0.09±0.07	42.6±0.4	-0.16±0.10	36.5±0.4	0.02±0.06	34.6±0.5	0.09±0.09
		KF-OMI	41.9±0.4	-0.26±0.04	44.9±0.4	-0.37±0.11	46.6±0.4	-0.08±0.09	39.5±0.4	-0.29±0.08	36.7±0.5	-0.38±0.10
	2005- 2009	KF-AQS	38.0±0.4	0.07±0.18	38.9±0.4	-0.17±0.28	42.9±0.4	-0.48±0.51	36.6±0.3	-0.08±0.52	34.0±0.6	-0.01±0.66
		KF-OMI	43.2±0.3	0.19±0.19	45.9±0.3	-0.06±0.34	46.1±0.3	-0.17±0.15	41.6±0.3	-0.16±0.74	39.8±0.5	-0.20±0.56
	2010- 2014	KF-AQS	38.3±0.4	-0.11±0.16	39.8±0.7	0.00±0.61	43.1±0.4	-1.21±0.27	36.5±0.4	0.17±0.29	34.5±0.6	0.60±0.50
		KF-OMI	42.3±0.4	-0.72±0.18	46.8±0.5	-1.04±0.67	47.8±0.4	-1.56±0.21	39.1±0.3	-0.11±0.37	35.3±0.5	0.18±0.47
	2015- 2020	KF-AQS	38.0±0.4	-0.07±0.25	38.2±0.4	-0.29±0.23	41.8±0.4	-0.11±0.52	36.5±0.4	0.53±0.23	35.0±0.5	-0.31±0.44
KF-OMI		40.6±0.4	-0.30±0.18	42.6±0.4	-0.72±0.33	46.0±0.4	-0.19±0.29	38.0±0.4	0.15±0.17	35.3±0.6	-0.44±0.40	

Table. S3. Averages (with units ppb or DU) and trends (with units ppb yr⁻¹ or DU yr⁻¹) of surface and tropospheric column O₃ concentrations in 2005-2020 over West Coast (Region #3) from observations (AQS and OMI) and a priori and a posteriori (KF) simulations. Grids with low and medium anthropogenic NO_x emissions are excluded in West Coast.

#4 Middle US			Annual		Spring		Summer		Autumn		Winter	
			Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend
TS4.1 surface (sampled)	2005- 2020	AQS	39.7±0.3	-0.30±0.09	44.3±0.3	-0.29±0.10	46.0±0.5	-0.62±0.21	37.8±0.5	-0.35±0.15	31.0±0.4	-0.06±0.08
		a priori	42.2±0.3	-0.28±0.06	44.8±0.3	-0.40±0.06	55.2±0.4	-0.61±0.18	40.6±0.5	-0.18±0.10	28.3±0.4	0.02±0.07
		KF-AQS	40.4±0.3	-0.29±0.08	44.8±0.2	-0.33±0.09	48.5±0.4	-0.61±0.19	38.4±0.5	-0.30±0.13	30.2±0.4	-0.01±0.07
TS4.2 surface	2005- 2020	a priori	41.3±0.1	-0.23±0.06	44.1±0.1	-0.34±0.06	54.1±0.1	-0.48±0.17	39.3±0.2	-0.18±0.09	27.8±0.2	0.04±0.06
		KF-AQS	40.0±0.1	-0.21±0.06	44.1±0.1	-0.28±0.07	49.8±0.2	-0.46±0.17	37.6±0.2	-0.22±0.10	28.7±0.1	0.05±0.05
TS4.3 trop. column (convolved)	2005- 2020	OMI	39.0±0.1	0.02±0.05	40.8±0.1	0.11±0.11	44.4±0.1	0.16±0.10	37.2±0.1	-0.06±0.07	33.8±0.1	-0.12±0.11
		a priori	38.2±0.2	-0.10±0.04	39.5±0.2	-0.11±0.08	46.3±0.1	-0.17±0.08	36.1±0.1	-0.06±0.05	31.0±0.2	-0.05±0.05
		KF-OMI	39.4±0.1	-0.13±0.05	41.8±0.2	-0.13±0.09	46.4±0.1	0.00±0.08	37.2±0.1	-0.14±0.06	32.1±0.2	-0.24±0.05
TS4.4 trop. Column	2005- 2020	a priori	39.3±0.2	-0.08±0.06	40.8±0.3	-0.19±0.12	46.1±0.1	-0.18±0.12	36.6±0.2	-0.02±0.08	33.9±0.3	0.04±0.11
		KF-AQS	38.9±0.2	-0.07±0.06	40.8±0.3	-0.17±0.12	45.1±0.1	-0.17±0.12	36.2±0.1	-0.02±0.07	33.9±0.3	0.05±0.11
		KF-OMI	41.5±0.2	-0.24±0.05	44.4±0.2	-0.33±0.12	47.7±0.1	-0.09±0.11	38.5±0.1	-0.21±0.08	35.4±0.3	-0.34±0.11
	2005- 2009	KF-AQS	39.0±0.2	-0.43±0.21	41.1±0.2	-0.62±0.78	45.3±0.1	-1.44±0.20	36.1±0.1	-0.15±0.50	33.3±0.3	-0.58±0.53
		KF-OMI	42.6±0.1	-0.30±0.17	45.4±0.2	-0.42±0.75	47.3±0.1	-1.26±0.07	39.8±0.1	-0.14±0.61	37.9±0.3	-0.59±0.49
	2010- 2014	KF-AQS	39.4±0.2	-0.60±0.47	41.6±0.3	-1.20±0.66	45.9±0.1	-1.37±0.76	36.4±0.1	-0.31±0.41	34.0±0.3	-0.49±0.98
		KF-OMI	41.8±0.2	-0.88±0.37	45.7±0.2	-1.47±0.57	48.8±0.1	-1.25±0.75	38.3±0.2	-0.50±0.41	34.2±0.2	-0.81±0.87
	2015- 2020	KF-AQS	38.5±0.2	0.07±0.11	39.7±0.2	0.11±0.50	44.2±0.1	0.28±0.36	36.1±0.1	0.00±0.42	34.3±0.3	-0.02±0.47
KF-OMI		40.3±0.2	-0.12±0.15	42.4±0.2	-0.23±0.52	47.1±0.1	0.19±0.29	37.6±0.1	-0.25±0.38	34.2±0.3	-0.13±0.44	

Table. S4. Averages (with units ppb or DU) and trends (with units ppb yr⁻¹ or DU yr⁻¹) of surface and tropospheric column O₃ concentrations in 2005-2020 over Middle US (Region #4) from observations (AQS and OMI) and a priori and a posteriori (KF) simulations.

#5 Southeast US			Annual		Spring		Summer		Autumn		Winter	
			Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend
TS5.1 surface (sampled)	2005- 2020	AQS	39.9±0.3	-0.47±0.09	46.9±0.2	-0.54±0.13	42.6±0.5	-0.92±0.18	37.6±0.3	-0.44±0.19	32.7±0.3	-0.03±0.06
		a priori	44.4±0.1	-0.38±0.06	46.6±0.1	-0.47±0.07	54.8±0.5	-0.86±0.15	43.3±0.1	-0.24±0.11	33.2±0.3	0.00±0.12
		KF-AQS	40.3±0.1	-0.43±0.08	46.3±0.1	-0.52±0.11	44.7±0.4	-0.87±0.16	37.8±0.2	-0.37±0.15	32.6±0.3	-0.02±0.06
TS5.2 surface	2005- 2020	a priori	43.7±0.1	-0.39±0.06	46.0±0.1	-0.47±0.07	53.1±0.3	-0.84±0.15	42.9±0.1	-0.26±0.10	33.0±0.1	-0.02±0.12
		KF-AQS	40.6±0.0	-0.39±0.07	45.3±0.1	-0.48±0.09	45.3±0.2	-0.78±0.14	38.9±0.1	-0.33±0.13	32.8±0.1	0.00±0.08
TS5.3 trop. column (convolved)	2005- 2020	OMI	42.7±0.1	0.06±0.07	44.5±0.1	0.06±0.12	48.9±0.1	0.09±0.11	40.4±0.1	0.00±0.07	36.9±0.1	0.07±0.12
		a priori	42.8±0.1	-0.11±0.04	45.3±0.1	-0.13±0.10	49.3±0.1	-0.21±0.09	40.1±0.1	-0.08±0.06	36.6±0.2	-0.02±0.06
		KF-OMI	43.6±0.1	-0.10±0.05	46.8±0.1	-0.13±0.10	49.8±0.1	-0.02±0.09	41.0±0.0	-0.13±0.06	36.9±0.1	-0.14±0.06
TS5.4 trop. Column	2005- 2020	a priori	44.3±0.1	-0.19±0.07	47.4±0.2	-0.34±0.15	48.1±0.1	-0.26±0.12	40.0±0.0	-0.07±0.09	42.2±0.2	-0.09±0.15
		KF-AQS	43.5±0.1	-0.18±0.07	47.1±0.2	-0.34±0.15	46.1±0.1	-0.23±0.11	39.2±0.1	-0.08±0.10	42.1±0.2	-0.09±0.15
		KF-OMI	45.8±0.1	-0.28±0.07	49.8±0.2	-0.42±0.15	49.4±0.1	-0.17±0.11	41.6±0.0	-0.20±0.08	42.7±0.1	-0.34±0.17
	2005- 2009	KF-AQS	44.1±0.1	-0.23±0.44	48.0±0.2	-1.06±1.15	46.5±0.1	-0.61±0.32	38.9±0.1	-0.40±0.35	43.3±0.3	-0.32±0.66
		KF-OMI	46.9±0.1	-0.22±0.27	51.0±0.1	-0.98±1.03	49.3±0.1	-0.64±0.30	42.1±0.0	-0.39±0.34	45.9±0.2	-0.15±0.56
	2010- 2014	KF-AQS	44.3±0.1	-1.05±0.30	48.7±0.2	-1.53±0.59	47.3±0.1	-1.83±0.45	40.1±0.1	-0.97±0.54	40.9±0.2	-0.94±0.99
		KF-OMI	46.6±0.1	-1.24±0.22	51.8±0.2	-1.69±0.55	50.8±0.1	-1.74±0.47	42.4±0.1	-1.03±0.49	40.5±0.1	-1.07±1.04
	2015- 2020	KF-AQS	42.4±0.1	0.14±0.25	45.1±0.2	0.14±0.49	44.7±0.1	0.19±0.36	38.6±0.0	-0.15±0.48	42.2±0.2	0.44±0.73
KF-OMI		44.2±0.1	-0.06±0.31	47.2±0.2	-0.13±0.64	48.3±0.1	0.04±0.36	40.6±0.0	-0.39±0.41	41.8±0.2	0.24±0.78	

Table. S5. Averages (with units ppb or DU) and trends (with units ppb yr⁻¹ or DU yr⁻¹) of surface and tropospheric column O₃ concentrations in 2005-2020 over Southeast US (Region #5) from observations (AQS and OMI) and a priori and a posteriori (KF) simulations.

#1 Britain			Annual		Spring		Summer		Autumn		Winter	
			Mean	Trend								
TS6.1 surface (sampled)	2005- 2020	AirBase	24.4±0.6	0.14±0.06	31.8±0.6	0.17±0.10	27.5±0.6	-0.01±0.12	19.7±0.7	0.12±0.06	18.8±0.7	0.35±0.10
		a priori	29.9±0.2	0.10±0.03	36.0±0.1	0.11±0.05	38.7±0.3	-0.10±0.09	25.2±0.2	0.08±0.05	19.9±0.4	0.31±0.09
		KF-AirBase	26.0±0.3	0.09±0.05	32.9±0.3	0.11±0.08	30.7±0.3	-0.07±0.10	21.3±0.4	0.07±0.05	19.1±0.5	0.28±0.09
TS6.2 surface	2005- 2020	a priori	30.0±0.2	0.09±0.03	35.9±0.1	0.10±0.05	38.5±0.3	-0.10±0.08	25.4±0.2	0.07±0.04	20.2±0.4	0.30±0.09
		KF-AirBase	26.7±0.3	0.07±0.04	33.4±0.2	0.09±0.07	31.9±0.4	-0.09±0.09	22.0±0.3	0.06±0.04	19.5±0.4	0.28±0.09
TS6.3 trop. column (convolved)	2005- 2020	OMI	35.9±0.1	-0.05±0.05	37.8±0.2	-0.31±0.15	37.2±0.2	-0.17±0.11	34.1±0.1	0.04±0.10	34.5±0.3	0.30±0.13
		a priori	30.9±0.1	-0.07±0.03	32.3±0.1	-0.16±0.06	32.2±0.1	-0.24±0.06	29.8±0.1	0.03±0.04	29.3±0.1	0.12±0.09
		KF-OMI	33.7±0.1	-0.25±0.03	36.2±0.1	-0.43±0.09	35.9±0.2	-0.29±0.07	31.7±0.1	-0.19±0.06	30.8±0.1	-0.03±0.08
TS6.4 trop. Column	2005- 2020	a priori	30.1±0.1	0.00±0.03	32.9±0.1	0.01±0.06	31.8±0.2	0.02±0.08	27.9±0.1	-0.02±0.06	27.8±0.1	0.01±0.08
		KF-AirBase	29.7±0.1	0.00±0.03	32.6±0.1	0.01±0.06	31.0±0.2	0.02±0.08	27.5±0.1	-0.02±0.05	27.7±0.1	0.01±0.08
		KF-OMI	34.7±0.1	-0.49±0.05	39.8±0.1	-0.60±0.08	36.9±0.2	-0.07±0.09	30.9±0.1	-0.52±0.08	30.6±0.1	-0.71±0.13
	2005- 2009	KF-AirBase	29.7±0.1	-0.29±0.10	32.4±0.1	0.18±0.30	31.1±0.1	-0.85±0.41	27.7±0.1	-0.27±0.34	27.6±0.0	-0.27±0.40
		KF-OMI	37.7±0.1	-0.52±0.31	42.8±0.0	0.03±0.54	37.0±0.1	-0.42±0.52	34.4±0.1	-0.92±0.48	35.9±0.0	-1.35±0.53
	2010- 2014	KF-AirBase	29.5±0.1	-0.16±0.21	32.8±0.1	-0.67±0.39	30.3±0.1	-0.04±0.40	27.3±0.1	0.29±0.40	27.5±0.1	-0.40±0.71
		KF-OMI	34.2±0.1	-0.88±0.31	40.7±0.1	-1.52±0.44	37.0±0.1	-0.41±0.60	29.9±0.1	-0.25±0.41	28.3±0.1	-1.00±0.53
	2015- 2020	KF-AirBase	29.9±0.1	0.07±0.13	32.6±0.1	0.18±0.24	31.4±0.2	0.17±0.41	27.5±0.1	-0.15±0.22	28.1±0.1	-0.12±0.25
KF-OMI		32.5±0.1	-0.35±0.11	36.6±0.1	-0.52±0.12	36.7±0.2	-0.45±0.43	28.7±0.1	-0.42±0.23	28.2±0.1	-0.18±0.24	

Table. S6. Averages (with units ppb or DU) and trends (with units ppb yr⁻¹ or DU yr⁻¹) of surface and tropospheric column O₃ concentrations in 2005-2020 over Britain (Region #1) from observations (AirBase and OMI) and a priori and a posteriori (KF) simulations. Grids with low and medium anthropogenic NO_x emissions are excluded in Britain.

#2 Central EU			Annual		Spring		Summer		Autumn		Winter	
			Mean	Trend								
TS7.1 surface (sampled)	2005- 2020	AirBase	28.0±0.3	0.21±0.06	35.5±0.3	0.14±0.10	38.3±0.4	0.17±0.17	20.5±0.2	0.22±0.08	17.7±0.3	0.33±0.10
		a priori	30.5±0.2	0.14±0.04	36.5±0.2	0.12±0.05	44.7±0.2	0.03±0.11	25.2±0.4	0.14±0.07	15.9±0.3	0.27±0.08
		KF-AirBase	28.2±0.2	0.16±0.06	35.4±0.2	0.13±0.09	39.0±0.3	0.12±0.15	21.3±0.2	0.17±0.07	17.4±0.2	0.26±0.08
TS7.2 surface	2005- 2020	a priori	30.6±0.3	0.14±0.04	36.5±0.2	0.12±0.05	44.8±0.2	0.03±0.11	25.2±0.3	0.14±0.07	15.9±0.3	0.27±0.08
		KF-AirBase	28.3±0.2	0.16±0.06	35.4±0.2	0.12±0.09	39.1±0.3	0.12±0.15	21.3±0.2	0.17±0.07	17.3±0.2	0.25±0.08
TS7.3 trop. column (convolved)	2005- 2020	OMI	35.5±0.1	-0.22±0.07	36.6±0.2	-0.51±0.15	37.8±0.1	-0.25±0.09	33.4±0.1	-0.14±0.12	34.2±0.2	-0.02±0.13
		a priori	31.6±0.1	-0.13±0.04	32.9±0.1	-0.26±0.07	34.4±0.1	-0.23±0.07	30.2±0.1	-0.02±0.03	28.9±0.1	-0.02±0.08
		KF-OMI	34.0±0.1	-0.31±0.05	36.2±0.1	-0.52±0.09	37.5±0.1	-0.29±0.08	31.9±0.1	-0.22±0.06	30.5±0.1	-0.20±0.07
TS7.4 trop. Column	2005- 2020	a priori	31.0±0.0	0.04±0.03	33.4±0.1	0.02±0.07	34.7±0.1	0.09±0.07	28.5±0.0	-0.01±0.06	27.3±0.1	0.06±0.06
		KF-AirBase	30.4±0.0	0.04±0.03	33.0±0.1	0.02±0.07	33.5±0.1	0.10±0.08	27.8±0.0	-0.01±0.06	27.4±0.1	0.06±0.06
		KF-OMI	34.9±0.1	-0.38±0.05	39.3±0.1	-0.50±0.07	38.9±0.0	0.03±0.08	31.1±0.1	-0.44±0.07	30.0±0.1	-0.62±0.12
	2005- 2009	KF-AirBase	30.2±0.0	-0.24±0.13	32.7±0.0	0.07±0.39	33.2±0.0	-0.50±0.40	27.9±0.1	-0.48±0.21	27.0±0.1	-0.28±0.31
		KF-OMI	37.4±0.1	-0.36±0.31	41.6±0.1	-0.12±0.47	38.5±0.1	-0.13±0.47	34.1±0.1	-0.83±0.39	34.8±0.1	-1.25±0.58
	2010- 2014	KF-AirBase	30.2±0.0	-0.11±0.23	33.3±0.0	-0.61±0.53	32.8±0.0	-0.06±0.39	27.7±0.0	0.24±0.42	27.0±0.1	-0.17±0.38
		KF-OMI	34.4±0.1	-0.72±0.14	40.3±0.1	-1.49±0.13	38.8±0.0	-0.30±0.45	30.3±0.1	-0.12±0.37	27.8±0.1	-0.71±0.34
	2015- 2020	KF-AirBase	30.8±0.0	0.11±0.16	33.0±0.1	0.19±0.28	34.5±0.1	0.07±0.41	27.8±0.0	0.13±0.27	27.9±0.1	-0.12±0.27
KF-OMI		33.3±0.0	-0.23±0.14	36.7±0.1	-0.45±0.16	39.3±0.1	-0.31±0.47	29.2±0.0	-0.11±0.29	28.0±0.1	-0.18±0.26	

Table. S7. Averages (with units ppb or DU) and trends (with units ppb yr⁻¹ or DU yr⁻¹) of surface and tropospheric column O₃ concentrations in 2005-2020 over Central EU (Region #2) from observations (AirBase and OMI) and a priori and a posteriori (KF) simulations. Grids with low and medium anthropogenic NO_x emissions are excluded in Central EU.

#3 Western EU			Annual		Spring		Summer		Autumn		Winter	
			Mean	Trend								
TS8.1 surface (sampled)	2005- 2020	AirBase	32.3±0.2	0.11±0.05	38.9±0.2	0.05±0.07	40.1±0.4	-0.06±0.14	27.6±0.3	0.15±0.05	22.6±0.3	0.30±0.06
		a priori	35.9±0.2	0.02±0.03	40.6±0.2	-0.12±0.05	44.9±0.3	-0.10±0.11	32.9±0.3	0.03±0.06	25.2±0.3	0.23±0.06
		KF-AirBase	32.5±0.2	0.05±0.04	38.6±0.2	-0.02±0.07	40.2±0.2	-0.10±0.12	28.4±0.2	0.07±0.05	23.0±0.3	0.25±0.05
TS8.2 surface	2005- 2020	a priori	36.0±0.2	0.00±0.03	40.6±0.2	-0.13±0.05	44.8±0.2	-0.12±0.11	33.2±0.2	0.01±0.06	25.6±0.3	0.22±0.06
		KF-AirBase	32.8±0.2	0.03±0.04	38.6±0.1	-0.05±0.06	40.2±0.2	-0.12±0.12	28.8±0.2	0.05±0.05	23.5±0.2	0.23±0.05
TS8.3 trop. column (convolved)	2005- 2020	OMI	36.3±0.1	-0.16±0.08	37.8±0.1	-0.29±0.18	41.6±0.2	-0.10±0.10	34.7±0.1	-0.07±0.08	31.0±0.1	-0.21±0.16
		a priori	33.3±0.1	-0.10±0.04	34.0±0.1	-0.19±0.07	37.8±0.1	-0.17±0.06	32.0±0.1	-0.01±0.03	29.5±0.1	-0.04±0.06
		KF-OMI	35.7±0.1	-0.26±0.05	37.5±0.1	-0.40±0.10	41.3±0.2	-0.20±0.07	33.7±0.1	-0.20±0.05	30.5±0.1	-0.24±0.07
TS8.4 trop. Column	2005- 2020	a priori	32.5±0.1	0.02±0.03	34.1±0.1	-0.02±0.06	37.0±0.1	0.03±0.06	30.0±0.1	-0.01±0.05	28.7±0.1	0.09±0.06
		KF-AirBase	31.8±0.1	0.03±0.03	33.6±0.1	0.00±0.06	35.9±0.1	0.03±0.06	29.3±0.1	0.00±0.04	28.5±0.1	0.09±0.06
		KF-OMI	36.4±0.1	-0.36±0.04	40.0±0.1	-0.44±0.07	41.4±0.1	-0.02±0.07	32.7±0.1	-0.41±0.06	31.2±0.1	-0.53±0.11
	2005- 2009	KF-AirBase	31.6±0.1	-0.23±0.11	33.3±0.1	-0.15±0.20	35.7±0.1	-0.64±0.30	29.3±0.1	-0.21±0.34	27.9±0.1	-0.06±0.35
		KF-OMI	38.5±0.1	-0.40±0.22	41.8±0.1	-0.53±0.39	41.1±0.1	-0.25±0.34	35.4±0.1	-0.55±0.52	35.4±0.1	-0.81±0.49
	2010- 2014	KF-AirBase	31.7±0.1	-0.25±0.24	33.8±0.1	-0.60±0.51	35.5±0.1	-0.38±0.32	29.2±0.1	-0.14±0.29	28.4±0.1	-0.14±0.49
		KF-OMI	36.2±0.1	-0.79±0.12	41.1±0.1	-1.19±0.33	41.6±0.1	-0.54±0.34	32.1±0.1	-0.47±0.19	29.2±0.1	-0.67±0.25
	2015- 2020	KF-AirBase	32.1±0.1	-0.03±0.08	33.6±0.1	-0.04±0.22	36.5±0.1	-0.20±0.21	29.4±0.1	-0.03±0.17	29.1±0.1	0.02±0.28
KF-OMI		34.8±0.1	-0.32±0.15	37.6±0.1	-0.57±0.23	41.4±0.1	-0.45±0.40	31.1±0.1	-0.35±0.19	29.3±0.1	-0.09±0.28	

Table. S8. Averages (with units ppb or DU) and trends (with units ppb yr⁻¹ or DU yr⁻¹) of surface and tropospheric column O₃ concentrations in 2005-2020 over Western EU (Region #3) from observations (AirBase and OMI) and a priori and a posteriori (KF) simulations.

#4 Iberian Peninsula			Annual		Spring		Summer		Autumn		Winter	
			Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend
TS9.1 surface (sampled)	2005- 2020	AirBase	34.2±0.4	0.06±0.06	39.5±0.4	0.02±0.08	41.1±0.6	-0.15±0.09	31.2±0.4	0.12±0.08	25.3±0.4	0.23±0.08
		a priori	40.3±0.2	-0.06±0.03	43.9±0.1	-0.21±0.05	48.4±0.4	-0.18±0.11	38.1±0.2	-0.01±0.06	30.6±0.1	0.13±0.05
		KF-AirBase	35.2±0.3	-0.02±0.04	40.0±0.3	-0.08±0.07	42.1±0.4	-0.20±0.09	32.5±0.3	0.02±0.07	26.2±0.3	0.17±0.06
TS9.2 surface	2005- 2020	a priori	40.4±0.1	-0.07±0.03	43.9±0.1	-0.22±0.05	48.7±0.3	-0.17±0.12	38.4±0.1	-0.02±0.07	30.9±0.1	0.10±0.05
		KF-AirBase	36.2±0.2	-0.05±0.04	40.6±0.2	-0.13±0.06	43.4±0.3	-0.22±0.09	33.4±0.2	0.00±0.06	27.2±0.2	0.13±0.05
TS9.3 trop. column (convolved)	2005- 2020	OMI	37.8±0.2	-0.12±0.08	39.1±0.2	-0.20±0.18	45.4±0.2	0.05±0.10	35.7±0.2	-0.22±0.07	30.9±0.1	-0.12±0.17
		a priori	35.3±0.1	-0.08±0.04	36.0±0.1	-0.16±0.09	41.5±0.2	-0.06±0.06	33.3±0.1	-0.07±0.04	30.5±0.1	-0.05±0.05
		KF-OMI	37.8±0.1	-0.21±0.05	39.2±0.1	-0.30±0.12	45.3±0.2	-0.03±0.07	35.2±0.1	-0.28±0.05	31.2±0.1	-0.24±0.06
TS9.4 trop. Column	2005- 2020	a priori	34.2±0.1	0.02±0.03	35.0±0.1	-0.06±0.06	40.7±0.2	0.03±0.06	31.3±0.1	0.00±0.05	29.9±0.1	0.09±0.07
		KF-AirBase	33.6±0.1	0.02±0.03	34.5±0.1	-0.04±0.06	39.7±0.2	0.02±0.06	30.6±0.1	0.00±0.05	29.5±0.1	0.10±0.06
		KF-OMI	38.1±0.1	-0.30±0.04	40.8±0.1	-0.39±0.09	45.3±0.2	0.01±0.05	34.2±0.1	-0.36±0.07	32.0±0.1	-0.46±0.10
	2005- 2009	KF-AirBase	33.3±0.1	-0.26±0.05	34.4±0.1	-0.20±0.35	39.5±0.2	-0.69±0.26	30.4±0.1	-0.31±0.38	29.0±0.1	0.02±0.30
		KF-OMI	39.7±0.1	-0.41±0.20	42.1±0.1	-0.33±0.40	44.9±0.2	-0.43±0.29	36.3±0.1	-0.55±0.59	35.6±0.1	-0.50±0.37
	2010- 2014	KF-AirBase	33.6±0.1	-0.38±0.23	34.7±0.1	-0.63±0.28	39.4±0.2	-0.42±0.36	30.7±0.1	-0.68±0.13	29.7±0.1	0.10±0.50
		KF-OMI	38.2±0.1	-0.86±0.27	42.0±0.2	-1.18±0.43	45.6±0.2	-0.44±0.32	34.0±0.1	-0.97±0.24	30.4±0.1	-0.37±0.26
	2015- 2020	KF-AirBase	33.8±0.1	0.03±0.09	34.4±0.1	-0.28±0.23	40.1±0.2	0.05±0.15	30.7±0.1	0.10±0.20	29.9±0.1	0.34±0.34
KF-OMI		36.7±0.1	-0.23±0.19	38.6±0.1	-0.74±0.44	45.4±0.2	-0.07±0.19	32.7±0.1	-0.29±0.24	30.2±0.1	0.20±0.34	

Table. S9. Averages (with units ppb or DU) and trends (with units ppb yr⁻¹ or DU yr⁻¹) of surface and tropospheric column O₃ concentrations in 2005-2020 over Iberian Peninsula (Region #4) from observations (AirBase and OMI) and a priori and a posteriori (KF) simulations.

#5 Apennine Peninsula			Annual		Spring		Summer		Autumn		Winter	
			Mean	Trend								
TS10.1 surface (sampled)	2005- 2020	AirBase	34.0±0.4	0.08±0.07	39.2±0.5	-0.06±0.09	45.5±0.8	-0.03±0.11	29.2±0.5	0.17±0.07	22.0±0.7	0.22±0.06
		a priori	41.8±0.2	-0.05±0.03	45.3±0.1	-0.30±0.05	53.9±0.2	-0.10±0.07	39.0±0.2	0.00±0.05	29.1±0.4	0.20±0.05
		KF-AirBase	35.3±0.3	0.04±0.06	40.1±0.2	-0.09±0.08	46.4±0.4	-0.04±0.11	31.0±0.4	0.11±0.06	23.7±0.6	0.18±0.05
TS10.2 surface	2005- 2020	a priori	41.8±0.1	-0.06±0.02	45.2±0.1	-0.30±0.05	52.8±0.2	-0.10±0.07	39.2±0.1	-0.01±0.05	30.1±0.2	0.17±0.05
		KF-AirBase	36.6±0.2	-0.02±0.05	41.0±0.2	-0.16±0.06	46.1±0.2	-0.11±0.10	33.0±0.2	0.03±0.05	26.5±0.3	0.16±0.04
TS10.3 trop. column (convolved)	2005- 2020	OMI	38.7±0.2	-0.13±0.07	39.5±0.2	-0.25±0.14	46.7±0.2	0.02±0.10	36.5±0.2	-0.20±0.06	31.9±0.1	-0.15±0.11
		a priori	35.4±0.2	-0.08±0.04	35.6±0.1	-0.17±0.07	43.0±0.2	-0.02±0.07	33.4±0.1	-0.07±0.03	29.6±0.1	-0.06±0.06
		KF-OMI	37.7±0.2	-0.21±0.04	38.7±0.2	-0.33±0.09	46.0±0.2	-0.02±0.08	35.2±0.1	-0.25±0.04	30.7±0.1	-0.25±0.06
TS10.4 trop. Column	2005- 2020	a priori	34.9±0.1	0.04±0.04	35.3±0.1	-0.10±0.07	44.1±0.3	0.12±0.07	31.8±0.1	0.02±0.05	28.5±0.1	0.09±0.06
		KF-AirBase	34.0±0.1	0.04±0.04	34.5±0.1	-0.08±0.07	42.7±0.3	0.11±0.08	30.8±0.1	0.03±0.05	28.1±0.1	0.10±0.05
		KF-OMI	38.2±0.1	-0.26±0.04	40.3±0.1	-0.42±0.08	47.6±0.3	0.11±0.08	34.2±0.1	-0.29±0.05	30.8±0.1	-0.46±0.11
	2005- 2009	KF-AirBase	33.5±0.2	-0.24±0.14	34.6±0.1	-0.68±0.13	41.8±0.3	0.17±0.50	30.4±0.1	-0.57±0.34	27.6±0.1	-0.39±0.32
		KF-OMI	39.6±0.2	-0.29±0.18	41.7±0.1	-0.80±0.23	46.5±0.2	0.43±0.44	36.0±0.1	-0.74±0.36	34.5±0.1	-1.06±0.41
	2010- 2014	KF-AirBase	34.1±0.1	-0.28±0.36	34.9±0.1	-0.58±0.55	42.8±0.3	-0.51±0.71	31.0±0.1	-0.02±0.27	27.6±0.1	-0.28±0.17
		KF-OMI	38.3±0.2	-0.81±0.22	41.5±0.1	-1.19±0.50	48.2±0.3	-0.67±0.67	34.0±0.1	-0.45±0.21	28.7±0.1	-0.79±0.33
	2015- 2020	KF-AirBase	34.2±0.1	0.03±0.09	34.2±0.1	-0.24±0.32	43.3±0.2	-0.07±0.16	30.8±0.1	0.25±0.07	28.9±0.1	0.01±0.25
		KF-OMI	37.0±0.2	-0.19±0.07	38.0±0.1	-0.70±0.32	48.0±0.3	-0.16±0.14	32.9±0.1	-0.01±0.09	29.5±0.1	-0.11±0.27

Table. S10. Averages (with units ppb or DU) and trends (with units ppb yr⁻¹ or DU yr⁻¹) of surface and tropospheric column O₃ concentrations in 2005-2020 over Apennine Peninsula (Region #5) from observations (AirBase and OMI) and a priori and a posteriori (KF) simulations.