



Supplement of

Rapid O₃ assimilations – Part 1: Background and local contributions to tropospheric O₃ changes in China in 2015–2020

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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 1868 points (grids with 0.5°×0.625° spatial resolution over China); N = 1868 data points were then randomly drawn from the dataset (individual data points may be drawn multiple times); Average of OMI O₃ over China (for example, 2015) is calculated as the mean value of the randomly drawn N = 1868 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, 2015). 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 China for various years (i.e. 2016, 2017, 2018, 2019 and 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.

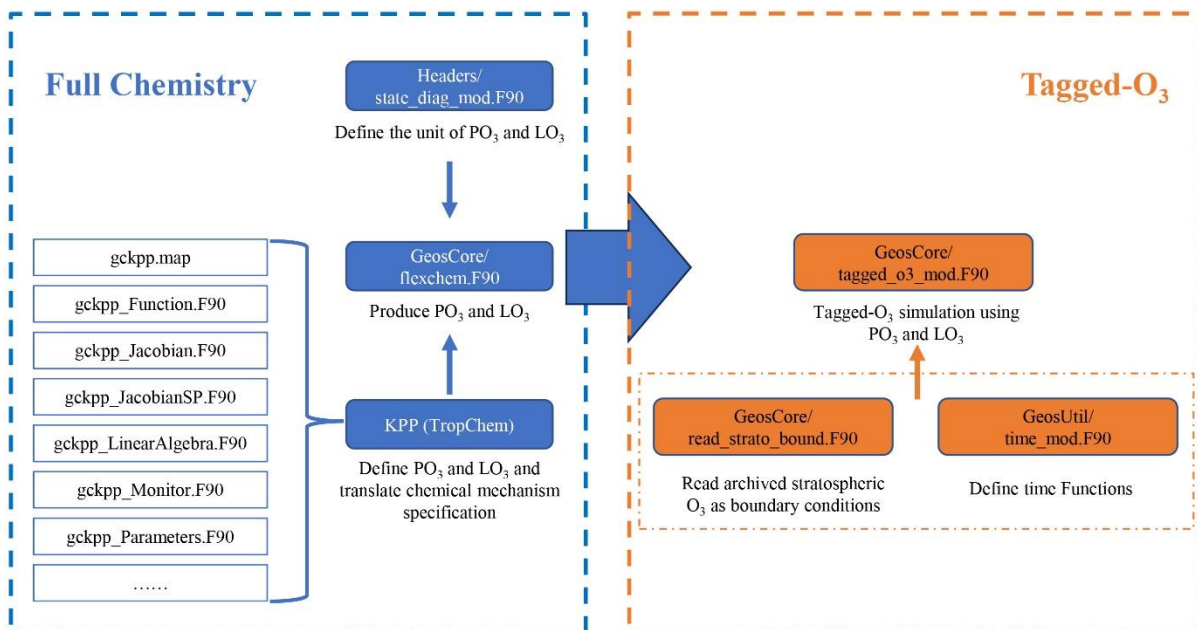


Fig. S1. Framework of the tagged-O₃ mode of GEOS-Chem.

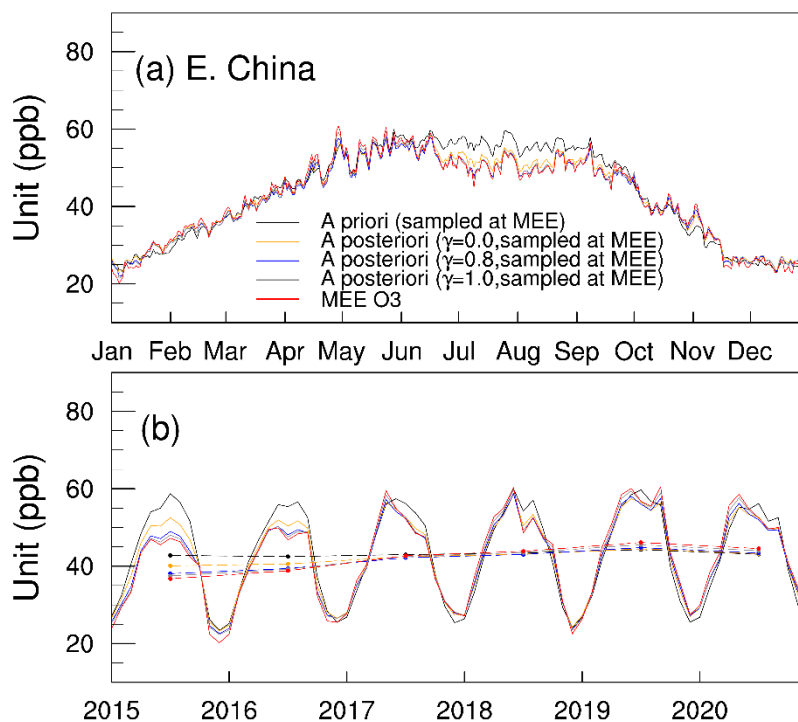


Fig. S2. (a) Daily averages of surface MDA8 O₃ in 2015-2020 from MEE stations, GEOS-Chem a priori and a posteriori simulations by assimilating MEE O₃ observations with different γ parameters (experiments #5-#7 in Table 1). (b) Monthly averages of MDA8 O₃. The dashed lines in panel b are annual averages.

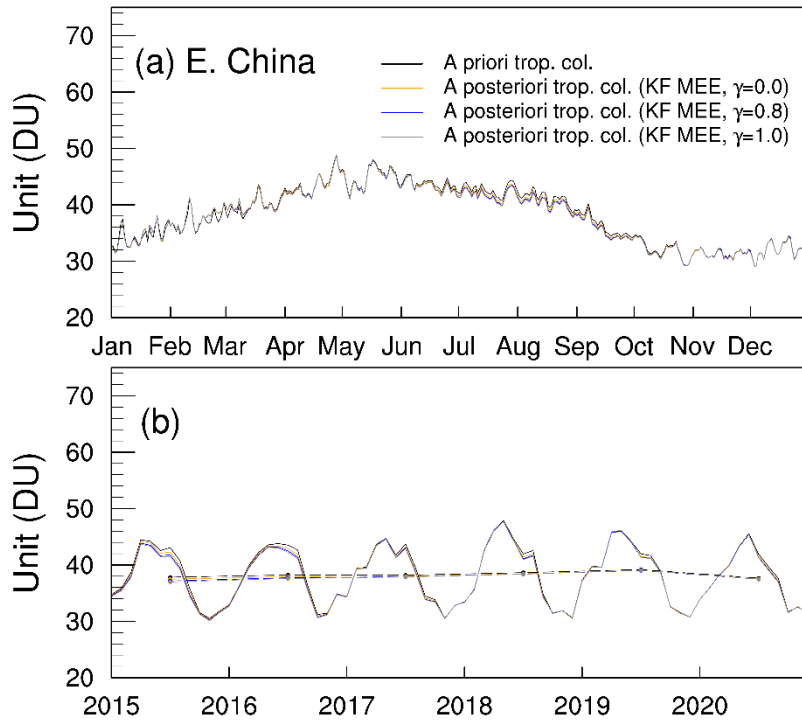


Fig. S3. (a) Daily averages of tropospheric O₃ columns in 2015-2020 from GEOS-Chem a priori simulation and a posteriori simulations by assimilating MEE O₃ observations with different γ parameters (experiments #5-#7 in Table 1). (b) Monthly averages of tropospheric O₃ columns. The dashed lines in panel b are annual averages.

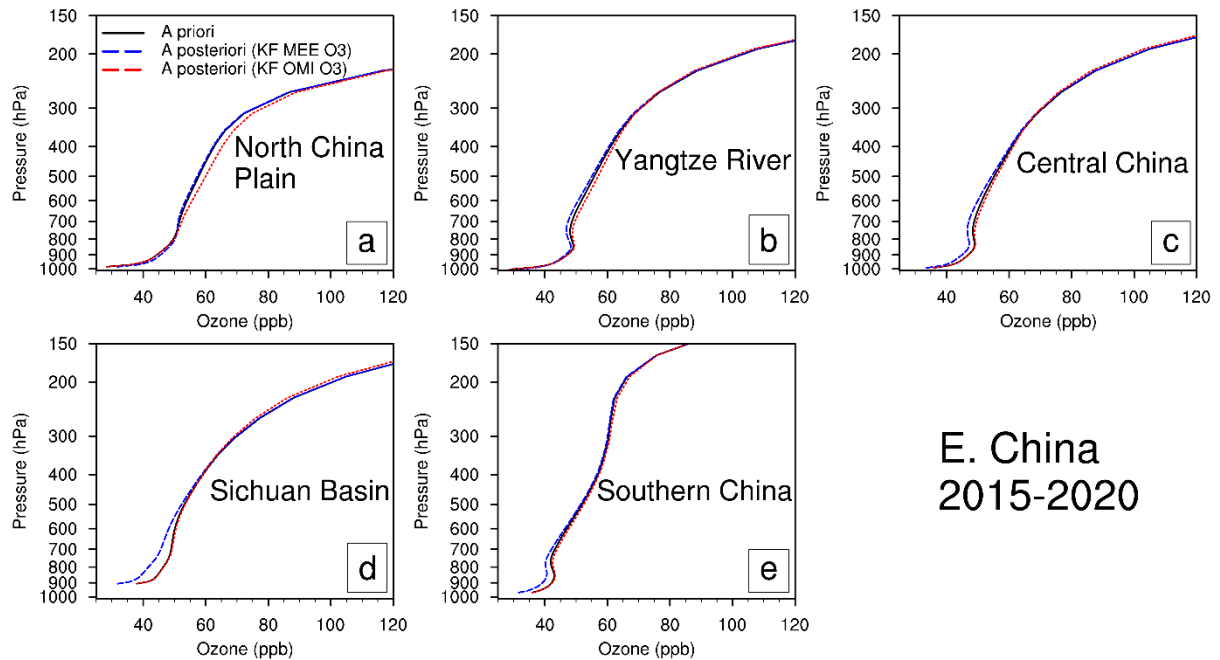


Fig. S4. Averages of O₃ vertical profiles in 2015-2020 from GEOS-Chem a priori (black) and a posteriori simulations by assimilating MEE (blue) and OMI (red) O₃ observations.

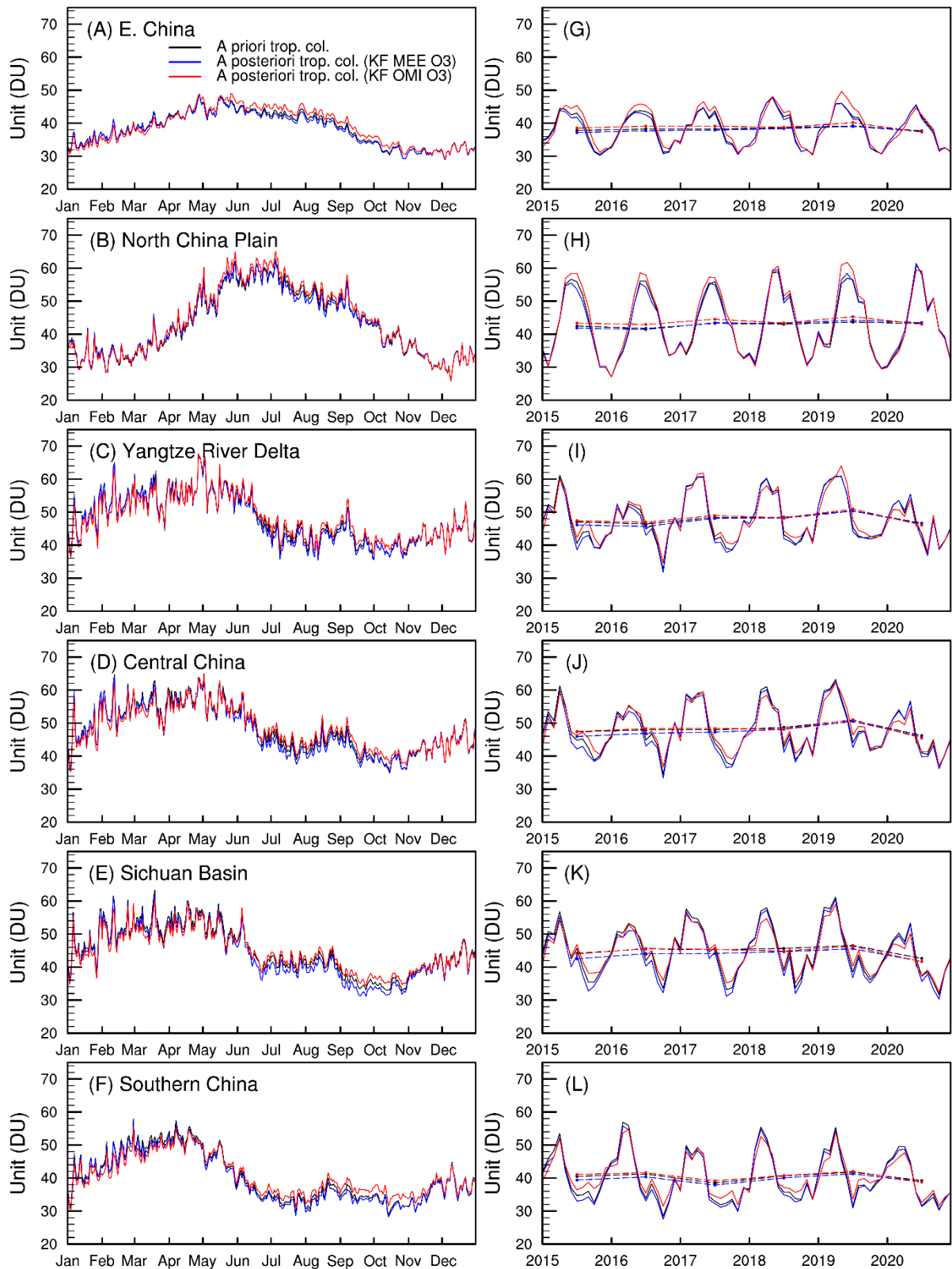


Fig. S5. (A-F) Daily averages of tropospheric O₃ columns in 2015-2020 from GEOS-Chem a priori simulation (black, Exp. #1) and a posteriori simulations by assimilating MEE (blue, Exp. #5) and OMI (red, Exp. #8) O₃ observations. (G-L) Monthly averages of tropospheric O₃ columns. The dashed lines in panels G-L are annual averages. The output O₃ profiles are NOT convolved with OMI averaging kernels.

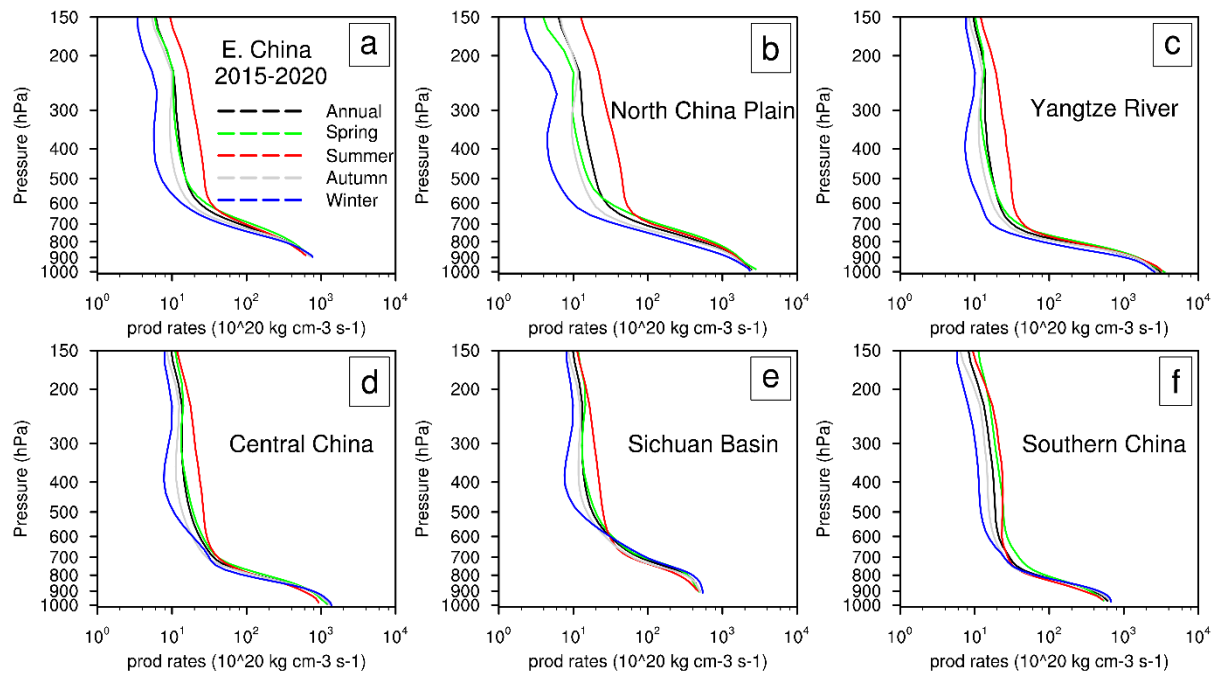


Fig. S6. O₃ production rates in 2015-2020 from GEOS-Chem simulations.

#1 North China Plain (2015-2020)		Annual		Spring		Summer		Autumn		Winter	
		Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend
TS1.1 surface (sampled)	MEE	47.6±0.4	2.34±0.48	55.7±0.4	2.41±0.63	65.2±0.7	3.00±0.88	42.9±0.5	2.29±0.47	27.8±0.4	1.35±0.24
	a priori	42.4±0.2	0.49±0.10	46.0±0.3	0.55±0.11	65.5±0.3	0.68±0.33	36.0±0.3	0.61±0.19	22.4±0.2	0.34±0.18
	KF-MEE	45.6±0.2	1.76±0.35	51.7±0.3	1.94±0.44	63.5±0.5	2.52±0.73	40.8±0.5	1.72±0.31	27.2±0.2	0.81±0.15
TS1.2 surface	a priori	42.7±0.2	0.48±0.10	46.2±0.2	0.53±0.11	66.3±0.3	0.69±0.34	36.0±0.3	0.60±0.22	22.5±0.1	0.34±0.20
	KF-MEE	44.8±0.2	1.56±0.32	50.5±0.3	1.70±0.34	64.1±0.4	2.33±0.70	39.3±0.4	1.53±0.32	25.9±0.2	0.76±0.16
TS1.3 trop. column (convolved)	OMI	45.2±0.3	-0.48±0.22	46.6±0.4	0.08±0.46	59.1±0.3	-1.12±0.46	42.7±0.4	-0.69±0.39	32.0±0.3	-0.69±0.39
	a priori	40.9±0.2	0.04±0.12	41.5±0.4	0.20±0.32	53.8±0.2	-0.30±0.17	38.3±0.2	0.21±0.13	30.0±0.2	0.00±0.19
	KF-OMI	42.7±0.2	-0.20±0.15	43.3±0.4	0.08±0.24	57.0±0.2	-0.72±0.31	40.0±0.3	-0.16±0.22	30.3±0.2	-0.13±0.19
TS1.4 trop. Column	a priori	42.9±0.3	0.27±0.13	44.6±0.5	-0.09±0.73	54.5±0.2	0.12±0.16	39.5±0.2	0.74±0.53	33.4±0.6	0.64±0.71
	KF-MEE	42.9±0.3	0.47±0.17	45.1±0.4	0.14±0.73	53.8±0.2	0.47±0.10	39.6±0.3	0.91±0.48	33.7±0.5	0.70±0.70
	KF-OMI	43.7±0.3	0.12±0.25	45.8±0.3	-0.19±0.75	56.2±0.2	-0.11±0.24	40.3±0.2	0.50±0.49	33.2±0.5	0.52±0.68

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 2015-2020 over North China Plain (Region #1) from observations (MEE and OMI) and a priori and a posteriori (KF) simulations. Grids with low and medium anthropogenic NO_x emissions are excluded in North China Plain.

#2 Yangtze River Delta (2015-2020)		Annual		Spring		Summer		Autumn		Winter	
		Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend
TS2.1 surface (sampled)	MEE	44.9±0.6	2.01±0.43	52.3±0.8	2.84±0.75	55.6±0.8	1.84±1.08	44.6±0.5	2.65±0.81	28.1±0.6	0.53±0.54
	a priori	44.6±0.3	0.65±0.12	49.2±0.4	0.92±0.21	60.8±0.6	0.63±0.57	42.6±0.4	0.94±0.17	26.3±0.4	0.37±0.27
	KF-MEE	45.0±0.5	1.38±0.33	50.7±0.6	2.21±0.58	56.2±0.7	1.34±0.89	44.6±0.4	1.84±0.55	28.9±0.3	0.13±0.41
TS2.2 surface	a priori	44.7±0.2	0.64±0.12	49.4±0.3	0.91±0.20	60.2±0.6	0.54±0.56	42.8±0.4	0.93±0.18	26.9±0.4	0.40±0.26
	KF-MEE	44.9±0.4	1.28±0.31	50.6±0.5	2.07±0.53	55.9±0.7	1.15±0.83	44.5±0.3	1.71±0.51	29.1±0.3	0.19±0.38
TS2.3 trop. column (convolved)	OMI	48.6±0.2	-0.29±0.33	55.0±0.3	0.15±0.54	53.6±0.5	0.23±0.90	45.0±0.2	-1.08±0.57	40.2±0.3	-1.03±0.91
	a priori	45.5±0.1	0.38±0.22	53.4±0.3	0.46±0.37	48.6±0.6	0.52±0.60	40.4±0.2	-0.21±0.30	39.3±0.4	0.34±0.65
	KF-OMI	47.0±0.1	0.10±0.28	53.9±0.2	0.27±0.19	51.9±0.5	0.33±0.86	43.1±0.2	-0.71±0.43	38.8±0.2	0.02±0.70
TS2.4 trop. Column	a priori	47.9±0.2	0.29±0.37	56.4±0.2	0.29±1.02	45.5±0.5	-0.02±0.60	41.2±0.2	0.59±0.26	49.1±0.1	0.63±0.47
	KF-MEE	47.5±0.2	0.45±0.40	56.1±0.2	0.56±1.05	44.3±0.5	0.19±0.64	41.0±0.2	0.74±0.31	49.2±0.2	0.63±0.47
	KF-OMI	48.1±0.2	0.13±0.46	56.0±0.1	0.15±1.15	46.7±0.5	-0.16±0.74	42.3±0.2	0.29±0.31	48.1±0.1	0.48±0.43

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 2015-2020 over Yangtze River Delta (Region #2) from observations (MEE and OMI) and a priori and a posteriori (KF) simulations. Grids with low and medium anthropogenic NO_x emissions are excluded in Yangtze River Delta.

#3 Central China (2015-2020)		Annual		Spring		Summer		Autumn		Winter	
		Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend
TS3.1 surface (sampled)	MEE	43.5±0.4	1.90±0.44	48.8±0.8	2.53±0.56	52.6±0.9	1.89±1.35	45.4±0.4	2.54±1.12	28.0±0.6	0.52±0.78
	a priori	45.1±0.2	0.44±0.29	50.3±0.3	0.61±0.21	59.8±0.5	0.04±0.85	44.3±0.4	0.83±0.40	26.4±0.3	0.49±0.24
	KF-MEE	43.1±0.4	1.55±0.38	47.8±0.4	2.07±0.42	53.0±0.5	1.51±1.20	44.7±0.4	2.09±0.89	27.8±0.4	0.47±0.63
TS3.2 surface	a priori	45.3±0.1	0.45±0.28	50.6±0.1	0.61±0.19	59.6±0.4	0.01±0.82	44.3±0.2	0.83±0.38	27.0±0.2	0.53±0.23
	KF-MEE	43.3±0.2	1.33±0.34	48.0±0.2	1.72±0.34	53.4±0.5	1.16±1.05	44.2±0.2	1.83±0.72	28.2±0.2	0.63±0.42
TS3.3 trop. column (convolved)	OMI	47.4±0.2	-0.60±0.20	54.4±0.2	-0.36±0.31	50.7±0.3	-0.66±0.59	42.8±0.2	-0.94±0.40	40.8±0.2	-0.60±0.40
	a priori	45.2±0.1	-0.08±0.27	53.5±0.3	0.05±0.19	47.7±0.4	-0.20±0.52	39.8±0.2	0.10±0.34	39.4±0.3	-0.19±0.45
	KF-OMI	46.2±0.1	-0.37±0.26	53.6±0.2	-0.20±0.30	50.0±0.4	-0.51±0.55	41.8±0.2	-0.47±0.34	39.1±0.3	-0.33±0.38
TS3.4 trop. Column	a priori	48.2±0.1	0.11±0.41	56.8±0.2	0.33±0.72	46.0±0.3	-0.34±0.56	41.0±0.1	0.36±0.41	49.3±0.1	0.12±0.49
	KF-MEE	47.4±0.1	0.32±0.43	55.9±0.2	0.62±0.72	44.5±0.3	-0.06±0.59	40.5±0.2	0.58±0.49	49.2±0.1	0.15±0.49
	KF-OMI	48.1±0.1	-0.06±0.48	55.7±0.1	0.15±0.85	47.0±0.4	-0.51±0.69	42.1±0.1	0.06±0.42	48.2±0.1	0.00±0.44

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 2015-2020 over Central China (Region #3) from observations (MEE and OMI) and a priori and a posteriori (KF) simulations.

#4 Sichuan Basin (2015-2020)		Annual		Spring		Summer		Autumn		Winter	
		Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend
TS4.1 surface (sampled)	MEE	36.9±0.9	1.13±0.25	45.1±1.2	2.27±0.54	50.7±1.5	0.95±0.67	29.2±0.8	0.50±0.35	23.4±0.7	0.59±0.47
	a priori	45.7±0.5	-0.18±0.13	51.5±0.4	0.06±0.29	60.0±1.0	-0.83±0.43	42.0±0.3	-0.10±0.19	29.5±0.3	0.30±0.12
	KF-MEE	37.5±0.6	0.77±0.20	44.4±1.0	1.69±0.43	50.3±1.2	0.54±0.56	31.7±0.4	0.32±0.25	24.5±0.5	0.45±0.37
TS4.2 surface	a priori	44.6±0.2	-0.16±0.17	50.4±0.2	0.10±0.20	54.9±0.6	-0.75±0.54	41.8±0.1	-0.06±0.19	31.4±0.3	0.25±0.14
	KF-MEE	39.1±0.2	0.38±0.18	44.9±0.3	0.99±0.22	47.3±0.5	-0.04±0.57	35.7±0.3	0.25±0.25	28.9±0.4	0.28±0.27
TS4.3 trop. column (convolved)	OMI	42.6±0.3	-0.39±0.32	47.7±0.4	0.30±0.56	46.3±0.3	-0.59±0.54	37.5±0.3	-0.66±0.40	38.7±0.4	-1.08±0.79
	a priori	42.6±0.3	0.03±0.29	51.3±0.4	0.23±0.60	44.7±0.3	-0.30±0.43	36.0±0.3	0.31±0.30	38.2±0.4	-0.26±0.31
	KF-OMI	42.9±0.3	-0.23±0.31	49.6±0.4	0.05±0.42	46.3±0.3	-0.45±0.61	37.5±0.3	-0.31±0.31	37.7±0.4	-0.51±0.46
TS4.4 trop. Column	a priori	44.9±0.2	-0.12±0.36	53.5±0.3	0.26±0.65	42.6±0.3	-0.67±0.33	36.7±0.2	-0.06±0.30	47.3±0.2	-0.16±0.61
	KF-MEE	43.8±0.2	0.03±0.38	52.3±0.3	0.47±0.66	41.0±0.3	-0.46±0.35	35.4±0.2	0.03±0.32	46.7±0.2	-0.13±0.60
	KF-OMI	44.6±0.2	-0.29±0.42	51.6±0.3	0.04±0.75	43.4±0.3	-0.89±0.43	37.8±0.2	-0.33±0.28	46.0±0.2	-0.23±0.55

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 2015-2020 over Sichuan Basin (Region #4) from observations (MEE and OMI) and a priori and a posteriori (KF) simulations.

#5 Southern China (2015-2020)		Annual		Spring		Summer		Autumn		Winter	
		Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend
TS5.1 surface (sampled)	MEE	38.3±0.5	1.44±0.40	37.8±0.5	1.50±0.84	38.5±0.6	0.79±0.68	44.9±0.7	2.85±1.45	33.2±0.7	1.85±1.39
	a priori	43.2±0.3	-0.02±0.29	47.5±0.3	-0.03±0.34	43.7±0.6	-1.01±0.77	46.1±0.5	0.66±0.58	35.8±0.6	0.83±0.48
	KF-MEE	39.2±0.4	0.97±0.35	39.9±0.4	0.93±0.67	39.5±0.6	0.23±0.66	44.7±0.6	2.21±1.20	33.5±0.6	1.49±1.05
TS5.2 surface	a priori	42.2±0.1	-0.07±0.26	47.0±0.1	-0.06±0.30	42.4±0.3	-0.99±0.77	44.7±0.2	0.63±0.54	34.9±0.3	0.68±0.51
	KF-MEE	39.0±0.2	0.58±0.29	41.4±0.2	0.58±0.50	38.8±0.2	-0.19±0.64	42.9±0.3	1.65±0.94	33.4±0.2	0.95±0.83
TS5.3 trop. column (convolved)	OMI	41.3±0.1	-0.23±0.32	49.1±0.1	0.07±0.44	41.5±0.1	-0.38±0.77	37.4±0.1	-0.09±0.48	36.9±0.2	-0.64±0.42
	a priori	39.7±0.2	0.03±0.33	49.5±0.2	0.01±0.55	35.9±0.2	-0.09±0.66	32.8±0.1	0.18±0.50	40.3±0.1	-0.36±0.32
	KF-OMI	41.3±0.1	-0.19±0.32	49.3±0.1	-0.09±0.43	40.3±0.1	-0.33±0.76	36.9±0.1	-0.14±0.47	38.5±0.1	-0.50±0.32
TS5.4 trop. Column	a priori	40.3±0.2	-0.07±0.33	49.7±0.2	-0.20±0.55	35.2±0.2	-0.18±0.40	34.1±0.1	0.27±0.55	42.0±0.2	-0.46±0.39
	KF-MEE	39.6±0.1	0.06±0.32	48.7±0.2	-0.09±0.57	34.4±0.1	-0.01±0.37	33.6±0.1	0.45±0.60	41.7±0.2	-0.41±0.37
	KF-OMI	40.6±0.1	-0.25±0.33	48.6±0.1	-0.32±0.53	36.7±0.1	-0.35±0.44	36.2±0.1	-0.05±0.59	40.8±0.1	-0.61±0.43

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 2015-2020 over Southern China (Region #5) from observations (MEE and OMI) and a priori and a posteriori (KF) simulations.