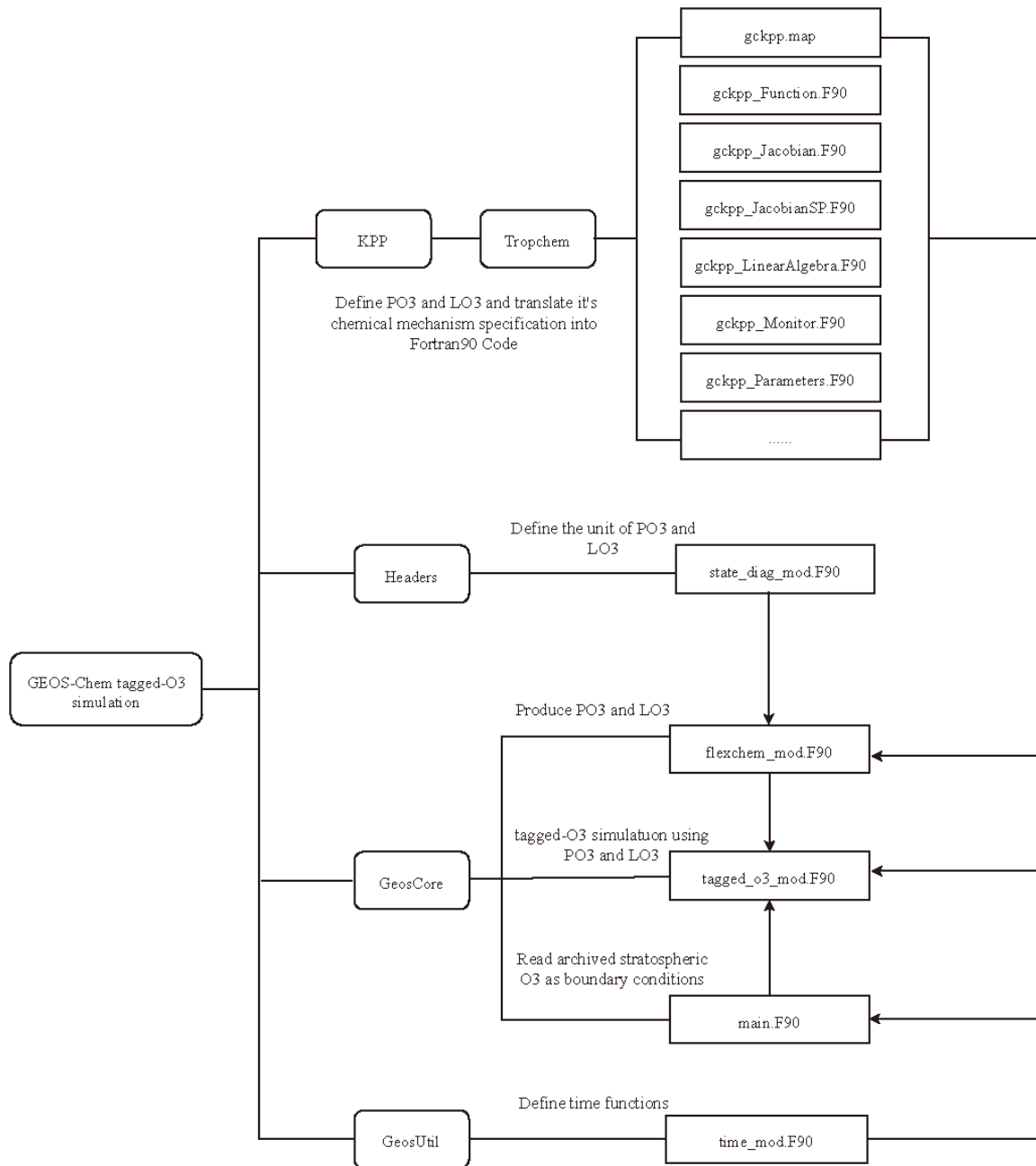
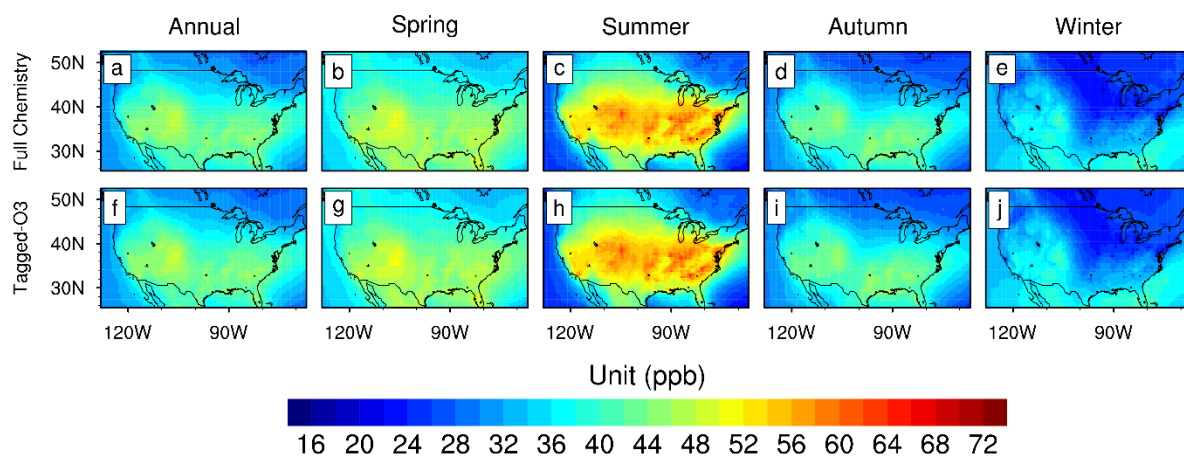


## Supplemental Information

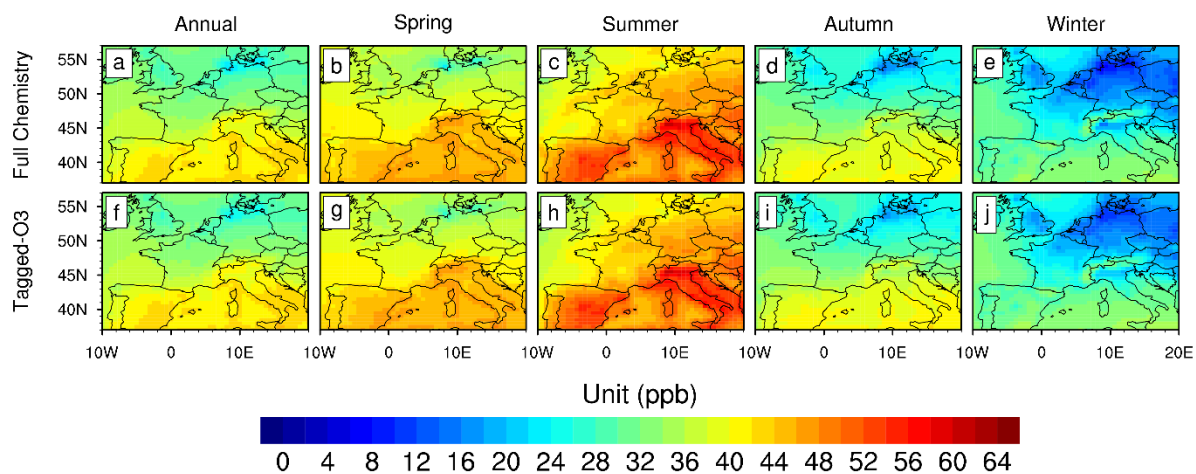


**Fig. S1.** Framework of the tagged-O<sub>3</sub> mode of GEOS-Chem.

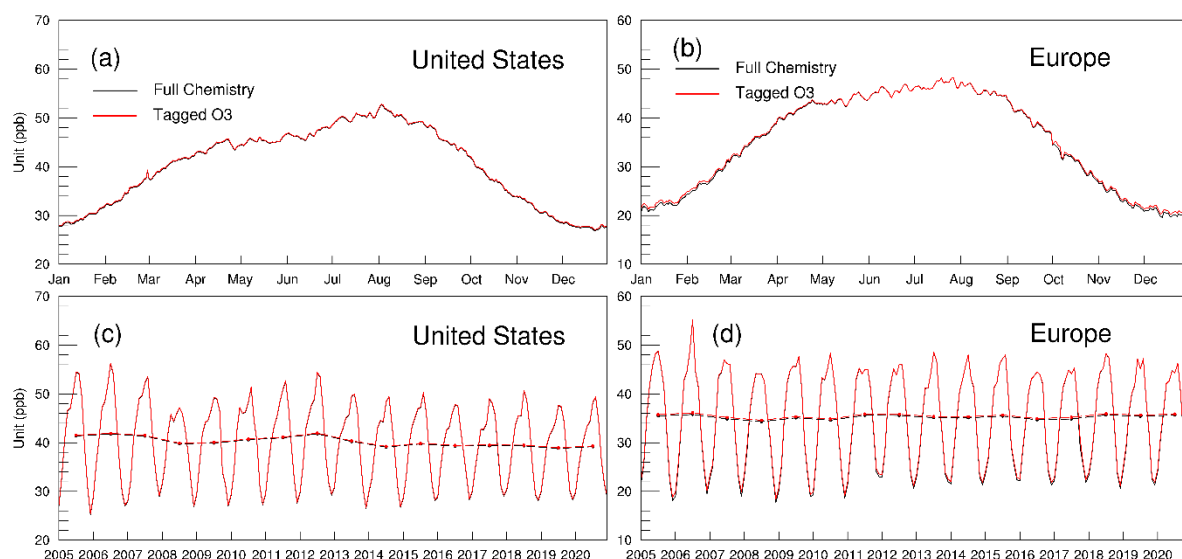
## GEOS-Chem Surface MDA8 Ozone Concentration



## GEOS-Chem Surface MDA8 Ozone Concentration



**Fig. S2.** Surface MDA8 O<sub>3</sub> in 2005-2020 (annual and seasonal averages) simulated by GEOS-Chem model with (A-E) full chemistry mode; (F-J) tagged-O<sub>3</sub> mode.



**Fig. S3.** (A-B) Daily averages of surface MDA8 O<sub>3</sub> over the US and Europe in 2005-2020 from GEOS-Chem full chemistry and tagged-O<sub>3</sub> simulations; (C-D) Monthly averages of MDA8 O<sub>3</sub>. The dashed lines in panels C-D are annual averages.

#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	2.34	55.7	2.41	65.2	3.00	42.9	2.29	27.8	1.35
	a priori	42.3	0.46	45.9	0.53	65.2	0.61	36.0	0.60	22.4	0.34
	KF-MEE	45.6	1.76	51.7	1.94	63.5	2.52	40.8	1.72	27.2	0.81
TS1.2 surface	a priori	42.5	0.46	46.1	0.51	66.0	0.62	35.9	0.59	22.5	0.34
	KF-MEE	44.8	1.56	50.5	1.70	64.1	2.33	39.3	1.53	25.9	0.76
TS1.3 trop. column (convolved)	OMI	45.2	-0.48	46.6	0.08	59.1	-1.12	42.7	-0.69	32.0	-0.69
	a priori	40.9	0.04	41.5	0.20	53.7	-0.32	38.3	0.21	30.0	0.00
	KF-OMI	42.7	-0.20	43.3	0.08	57.0	-0.72	40.0	-0.16	30.3	-0.13
TS1.4 trop. Column	a priori	42.9	0.26	44.6	-0.10	54.5	0.10	39.5	0.74	33.4	0.64
	KF-MEE	41.4	0.66	43.3	0.36	51.3	0.79	38.1	1.08	33.3	0.80
	KF-OMI	43.7	0.12	45.8	-0.19	56.2	-0.11	40.3	0.50	33.2	0.52

**Table. S1.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> 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<sub>x</sub> 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	2.01	52.3	2.84	55.6	1.84	44.6	2.65	28.1	0.53
	a priori	44.5	0.63	49.1	0.88	60.5	0.58	42.5	0.92	26.3	0.37
	KF-MEE	45.0	1.38	50.7	2.21	56.2	1.34	44.6	1.84	28.9	0.13
TS2.2 surface	a priori	44.6	0.61	49.2	0.87	59.9	0.49	42.7	0.91	26.9	0.40
	KF-MEE	44.9	1.28	50.6	2.07	55.9	1.15	44.5	1.71	29.1	0.19
TS2.3 trop. column (convolved)	OMI	48.6	-0.29	55.0	0.15	53.6	0.23	45.0	-1.08	40.2	-1.03
	a priori	45.4	0.38	53.4	0.45	48.5	0.51	40.4	-0.22	39.3	0.34
	KF-OMI	47.0	0.10	53.9	0.27	51.9	0.33	43.1	-0.71	38.8	0.02
TS2.4 trop. Column	a priori	47.9	0.28	56.4	0.28	45.4	-0.04	41.1	0.59	49.1	0.63
	KF-MEE	46.0	0.60	54.2	0.74	42.4	0.41	39.6	0.84	48.6	0.78
	KF-OMI	48.1	0.13	56.0	0.15	46.7	-0.16	42.3	0.29	48.1	0.48

**Table. S2.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> 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<sub>x</sub> 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	1.90	48.8	2.53	52.6	1.89	45.4	2.54	28.0	0.52
	a priori	44.7	0.37	50.0	0.53	59.1	-0.10	44.0	0.78	26.4	0.49
	KF-MEE	43.1	1.55	47.8	2.07	53.0	1.51	44.7	2.09	27.8	0.47
TS3.2 surface	a priori	44.9	0.37	50.2	0.52	58.8	-0.15	44.0	0.77	27.0	0.53
	KF-MEE	43.3	1.33	48.0	1.72	53.4	1.16	44.2	1.83	28.2	0.63
TS3.3 trop. column (convolved)	OMI	47.4	-0.60	54.4	-0.36	50.7	-0.66	42.8	-0.94	40.8	-0.60
	a priori	45.2	-0.09	53.5	0.04	47.7	-0.22	39.7	0.09	39.4	-0.19
	KF-OMI	46.2	-0.37	53.6	-0.20	50.0	-0.51	41.8	-0.47	39.1	-0.33
TS3.4 trop. Column	a priori	48.1	0.09	56.7	0.32	45.8	-0.37	40.9	0.35	49.3	0.12
	KF-MEE	45.9	0.46	54.2	0.79	42.7	0.13	38.8	0.67	48.5	0.33
	KF-OMI	48.1	-0.06	55.7	0.15	47.0	-0.51	42.1	0.06	48.2	0.00

**Table. S3.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> 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	1.13	45.1	2.27	50.7	0.95	29.2	0.50	23.4	0.59
	a priori	45.3	-0.26	51.1	-0.02	59.1	-1.01	41.7	-0.17	29.5	0.30
	KF-MEE	37.5	0.77	44.4	1.69	50.3	0.54	31.7	0.32	24.5	0.45
TS4.2 surface	a priori	43.9	-0.28	49.8	-0.02	53.6	-1.01	41.3	-0.18	31.4	0.25
	KF-MEE	39.1	0.38	44.9	0.99	47.3	-0.04	35.7	0.25	28.9	0.28
TS4.3 trop. column (convolved)	OMI	42.6	-0.39	47.7	0.30	46.3	-0.59	37.5	-0.66	38.7	-1.08
	a priori	42.6	0.02	51.2	0.21	44.5	-0.33	36.0	0.29	38.2	-0.26
	KF-OMI	42.9	-0.23	49.6	0.05	46.3	-0.45	37.5	-0.31	37.7	-0.51
TS4.4 trop. Column	a priori	44.8	-0.14	53.4	0.23	42.4	-0.71	36.6	-0.08	47.3	-0.16
	KF-MEE	42.6	0.17	51.1	0.59	39.4	-0.25	34.3	0.18	46.2	0.02
	KF-OMI	44.6	-0.29	51.6	0.04	43.4	-0.89	37.8	-0.33	46.0	-0.23

**Table. S4.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> 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	1.44	37.8	1.50	38.5	0.79	44.9	2.85	33.2	1.85
	a priori	43.0	-0.05	47.4	-0.04	43.4	-1.06	45.8	0.60	35.8	0.83
	KF-MEE	39.2	0.97	39.9	0.93	39.5	0.23	44.7	2.21	33.5	1.49
TS5.2 surface	a priori	42.0	-0.11	46.9	-0.09	42.0	-1.06	44.3	0.54	34.9	0.67
	KF-MEE	39.0	0.58	41.4	0.58	38.8	-0.19	42.9	1.65	33.4	0.95
TS5.3 trop. column (convolved)	OMI	41.3	-0.23	49.1	0.07	41.5	-0.38	37.4	-0.09	36.9	-0.64
	a priori	39.7	0.03	49.5	0.01	35.9	-0.10	32.7	0.18	40.3	-0.36
	KF-OMI	41.3	-0.19	49.3	-0.09	40.3	-0.33	36.9	-0.14	38.5	-0.50
TS5.4 trop. Column	a priori	40.3	-0.08	49.7	-0.21	35.2	-0.19	34.1	0.26	42.0	-0.46
	KF-MEE	38.8	0.15	47.9	-0.01	33.4	0.13	32.5	0.50	41.2	-0.30
	KF-OMI	40.6	-0.25	48.6	-0.32	36.7	-0.35	36.2	-0.05	40.8	-0.61

**Table. S5.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> concentrations in 2015-2020 over Southern China (Region #5) from observations (MEE and OMI) and a priori and a posteriori (KF) simulations.