

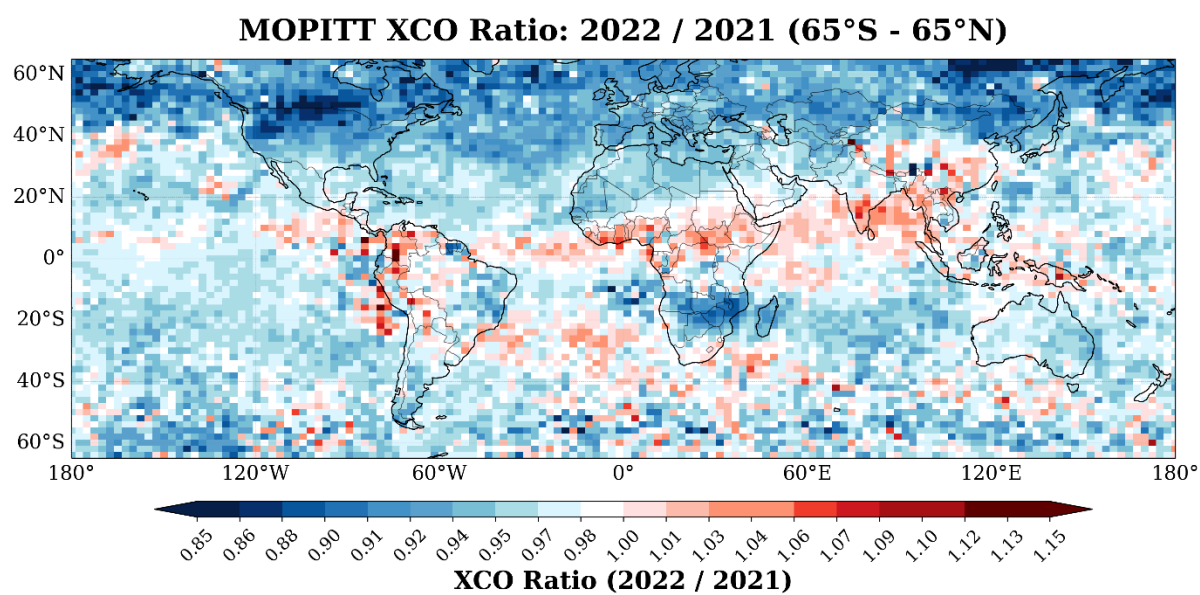


Fig. S1. Ratios between MOPITT CO columns in 2022 and 2021.

Domain	Sources	sites type	L (km)
China	MEE	Urban (Y)	2
		Suburban (N)	4
North America	NAPS	Traffic / Point Source	1
		Urban	2
		Sub-Urban	4
		Non-Urban	10
	EPA	Traffic	1
		Urban	2
		Suburban	4
		Rural	10
		Remote	20
Europe	EEA	Traffic	1
		Urban	2
		Suburban	4
		Rural	10
		Rural-Remote	20

Table. S1. Scale parameter (L) of site representation.

Experiments		Dataset for evaluation	Domain	R	Slope	Bias (ppb)	RMSE (ppb)
S2.1	a priori simulation	Surface	China	0.31	0.22	-331.4	512.8
			NCP	0.47	0.25	-399.8	602.2
			YRD	0.28	0.44	-139.6	411.9
			SCB	0.43	0.28	-257.8	338.7
S2.2	a priori simulation	MOPITT	China	0.73	0.62	6.8	18.8
			NCP	0.11	0.10	-16.6	29.9
			YRD	0.50	0.42	8.4	20.3
			SCB	0.67	0.60	17.6	25.9
S2.3	KF-Surface	Surface	China	0.94	0.80	-80.1	157.0
			NCP	0.96	0.82	-90.6	174.3
			YRD	0.86	0.84	-23.9	125.3
			SCB	0.94	0.81	-40.0	91.2
S2.4	KF-Surface	MOPITT (sampled)	China	0.76	0.98	27.1	33.9
			NCP	0.65	0.86	20.9	28.5
			YRD	0.67	0.72	31.7	35.8
			SCB	0.75	0.91	47.1	50.9
S2.5	KF-MOPITT	MOPITT	China	0.87	0.74	3.9	13.0
			NCP	0.71	0.65	-7.1	15.9
			YRD	0.83	0.74	3.6	11.9
			SCB	0.74	0.67	10.9	20.2

Table. S2. Statistical metrics comparing simulations and assimilations against surface and MOPITT data across different regions of China. Three different experiments are performed, namely the GEOS-Chem a priori simulation (S2.1, S2.2), the Kalman Filter assimilation of surface CO observations (S2.3, S2.4), and the Kalman Filter assimilation of MOPITT CO column observations (S2.5). The evaluation employs three distinct datasets: surface measurements, MOPITT retrievals and a sampled MOPITT dataset matching locations of surface stations.

Experiments		Dataset for evaluation	Domain	R	Slope	Bias (ppb)	RMSE (ppb)
S3.1	a priori simulation	Surface	North America	0.23	0.07	-198.9	282.2
			Northeast	0.46	0.16	-147.8	202.5
			Middle	0.35	0.09	-245.0	286.8
			West Coast	0.17	0.06	-240.8	340.8
S3.2	a priori simulation	MOPITT	North America	0.30	0.14	3.1	13.7
			Northeast	-0.02	-0.01	-0.6	15.4
			Middle	-0.06	-0.03	0.6	13.7
			West Coast	0.45	0.20	2.6	12.6
S3.3	KF-Surface	Surface	North America	0.95	0.72	-68.8	102.3
			Northeast	0.93	0.71	-53.8	81.6
			Middle	0.91	0.66	-104.0	126.5
			West Coast	0.96	0.78	-69.2	103.3
S3.4	KF-Surface	MOPITT (sampled)	North America	0.44	0.27	5.4	14.8
			Northeast	0.05	0.03	3.8	15.8
			Middle	-0.05	-0.02	1.9	14.7
			West Coast	0.58	0.35	4.3	13.6
S3.5	KF-MOPITT	MOPITT	North America	0.80	0.55	1.6	8.6
			Northeast	0.67	0.48	-0.6	9.8
			Middle	0.75	0.51	0.2	8.1
			West Coast	0.76	0.48	1.7	9.3

Table. S3. Same to Table S2 but for statistical metrics comparing simulations and assimilations against surface and MOPITT data across different regions of North America.

Experiments		Dataset for evaluation	Domain	R	Slope	Bias (ppb)	RMSE (ppb)
S4.1	a priori simulation	Surface	Europe	0.29	0.03	-244.7	378.7
			Iberian Peninsula	0.37	0.04	-180.1	250.2
			Apennine Peninsula	0.39	0.07	-342.8	440.8
			Central Europe	0.56	0.12	-145.5	205.9
S4.2	a priori simulation	MOPITT	Europe	0.22	0.08	1.8	11.4
			Iberian Peninsula	0.32	0.11	6.3	11.1
			Apennine Peninsula	0.35	0.17	3.9	11.2
			Central Europe	0.03	0.01	0.4	11.7
S4.3	KF-Surface	Surface	Europe	0.95	0.71	-60.7	128.9
			Iberian Peninsula	0.94	0.65	-52.6	92.7
			Apennine Peninsula	0.93	0.72	-79.0	142.4
			Central Europe	0.93	0.79	-30.7	67.7
S4.4	KF-Surface	MOPITT (sampled)	Europe	0.46	0.33	10.5	14.9
			Iberian Peninsula	0.49	0.26	10.8	13.7
			Apennine Peninsula	0.51	0.42	15.0	18.0
			Central Europe	0.26	0.15	7.1	13.0
S4.5	KF-MOPITT	MOPITT	Europe	0.72	0.45	1.0	8.0
			Iberian Peninsula	0.75	0.45	3.9	7.5
			Apennine Peninsula	0.78	0.52	2.1	7.4
			Central Europe	0.69	0.45	0.1	8.0

Table. S4. Same to Table S2 but for statistical metrics comparing simulations and assimilations against surface and MOPITT data across different regions of Europe.

	Experiment	Domain	Mean	Trend
Surface (ppb or ppb/year)	a priori simulation	China	401.3	-6.84 ± 5.27
		NCP	426.1	-6.52 ± 5.78
		YRD	541.2	-12.01 ± 9.54
		SCB	376.2	-4.34 ± 4.96
	Surface observation	China	735.3	-43.55 ± 7.51
		NCP	827.4	-68.28 ± 14.60
		YRD	681.3	-29.84 ± 5.78
		SCB	643.2	-27.49 ± 7.51
	KF-Surface	China	654.6	-35.21 ± 6.56
		NCP	736.4	-55.35 ± 12.37
		YRD	657.4	-26.79 ± 6.27
		SCB	601.4	-23.40 ± 6.86
Column (Xco ppb or ppb/year)	a priori simulation	China	109.3	-0.28 ± 0.14
		NCP	126.9	0.58 ± 0.98
		YRD	140.0	0.05 ± 0.71
		SCB	134.1	-0.14 ± 0.36
	MOPITT	China	102.4	-0.79 ± 0.40
		NCP	143.2	-2.65 ± 0.64
		YRD	131.8	-1.40 ± 0.79
		SCB	116.8	-0.50 ± 0.50
	KF-Surface	China	120.9	-1.17 ± 0.18
		NCP	165.0	-3.55 ± 1.46
		YRD	165.2	-1.89 ± 0.77
		SCB	158.9	-1.53 ± 0.62
	KF-MOPITT	China	106.4	-0.53 ± 0.24
		NCP	136.2	-1.64 ± 0.65
		YRD	135.2	-0.79 ± 0.56
		SCB	127.5	-0.72 ± 0.43

Table. S5. Averages and trends of surface and column CO concentrations from different experiments and datasets in China in 2015-2024.

	Experiment	Domain	Mean	Trend
Surface (ppb or ppb/year)	a priori simulation	North America	172.1	-0.20 ± 1.42
		Northeast	191.3	-0.23 ± 1.28
		Middle	182.9	-0.98 ± 1.20
		West Coast	163.9	-0.14 ± 3.74
	Surface observation	North America	349.4	-5.29 ± 3.12
		Northeast	316.3	-2.22 ± 2.96
		Middle	431.2	11.61 ± 7.71
		West Coast	370.9	-9.36 ± 4.69
	KF-Surface	North America	287.6	-3.91 ± 2.17
		Northeast	270.7	-1.90 ± 2.45
		Middle	325.8	5.44 ± 4.96
		West Coast	311.5	-7.03 ± 3.72
Column (Xco ppb or ppb/year)	a priori simulation	North America	98.9	-0.10 ± 0.10
		Northeast	104.7	-0.03 ± 0.16
		Middle	101.6	0.02 ± 0.12
		West Coast	97.9	-0.24 ± 0.26
	MOPITT	North America	95.7	-0.11 ± 0.70
		Northeast	105.4	0.13 ± 0.95
		Middle	100.9	0.06 ± 0.86
		West Coast	95.4	-0.47 ± 0.61
	KF-Surface	North America	100.2	-0.15 ± 0.13
		Northeast	107.0	-0.09 ± 0.18
		Middle	103.4	-0.04 ± 0.15
		West Coast	100.0	-0.35 ± 0.28
	KF-MOPITT	North America	97.4	-0.11 ± 0.42
		Northeast	104.8	0.07 ± 0.57
		Middle	101.2	0.05 ± 0.57
		West Coast	97.1	-0.33 ± 0.40

Table. S6. Same to Table S5 but for North America in 2015-2024.

	Experiment	Domain	Mean	Trend
Surface (ppb or ppb/year)	a priori simulation	Europe	140.3	-0.26 ± 0.33
		Iberian Peninsula	124.8	0.15 ± 0.35
		Apennine Peninsula	150.0	-0.11 ± 0.71
		Central Europe	149.9	-0.37 ± 0.54
	Surface observation	Europe	385.0	-3.91 ± 4.01
		Iberian Peninsula	304.8	-2.52 ± 3.90
		Apennine Peninsula	493.1	-11.02 ± 11.28
		Central Europe	294.2	-6.40 ± 1.69
	KF-Surface	Europe	324.3	-3.82 ± 2.59
		Iberian Peninsula	252.2	-1.83 ± 2.15
		Apennine Peninsula	413.9	-8.89 ± 7.97
		Central Europe	263.5	-5.92 ± 1.79
Column (Xco ppb or ppb/year)	a priori simulation	Europe	98.9	-0.02 ± 0.07
		Iberian Peninsula	97.0	-0.08 ± 0.09
		Apennine Peninsula	101.6	-0.03 ± 0.09
		Central Europe	100.6	0.01 ± 0.12
	MOPITT	Europe	97.1	-0.21 ± 0.65
		Iberian Peninsula	90.9	-0.29 ± 0.59
		Apennine Peninsula	97.7	-0.41 ± 0.62
		Central Europe	100.3	-0.12 ± 0.56
	KF-Surface	Europe	103.6	-0.14 ± 0.13
		Iberian Peninsula	101.6	-0.16 ± 0.18
		Apennine Peninsula	111.6	-0.38 ± 0.31
		Central Europe	107.2	-0.29 ± 0.21
	KF-MOPITT	Europe	98.1	-0.10 ± 0.32
		Iberian Peninsula	94.7	-0.10 ± 0.28
		Apennine Peninsula	99.8	-0.17 ± 0.28
		Central Europe	100.3	-0.08 ± 0.29

Table. S7. Same to Table S5 but for Europe in 2015-2024.