



Supplement of

High-resolution regional inversion reveals overestimation of anthropogenic methane emissions in China

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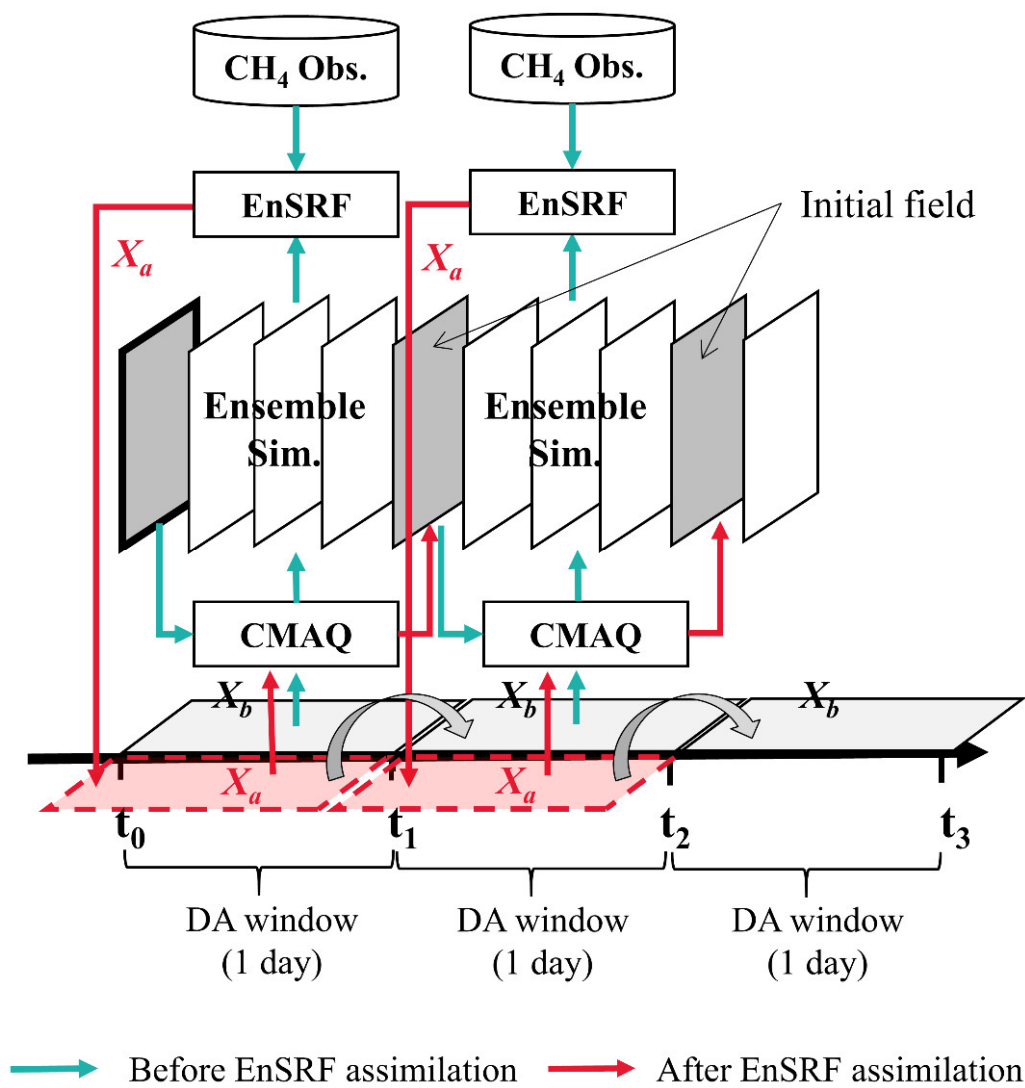


Figure S1 RegGCAS-CH₄ assimilation process

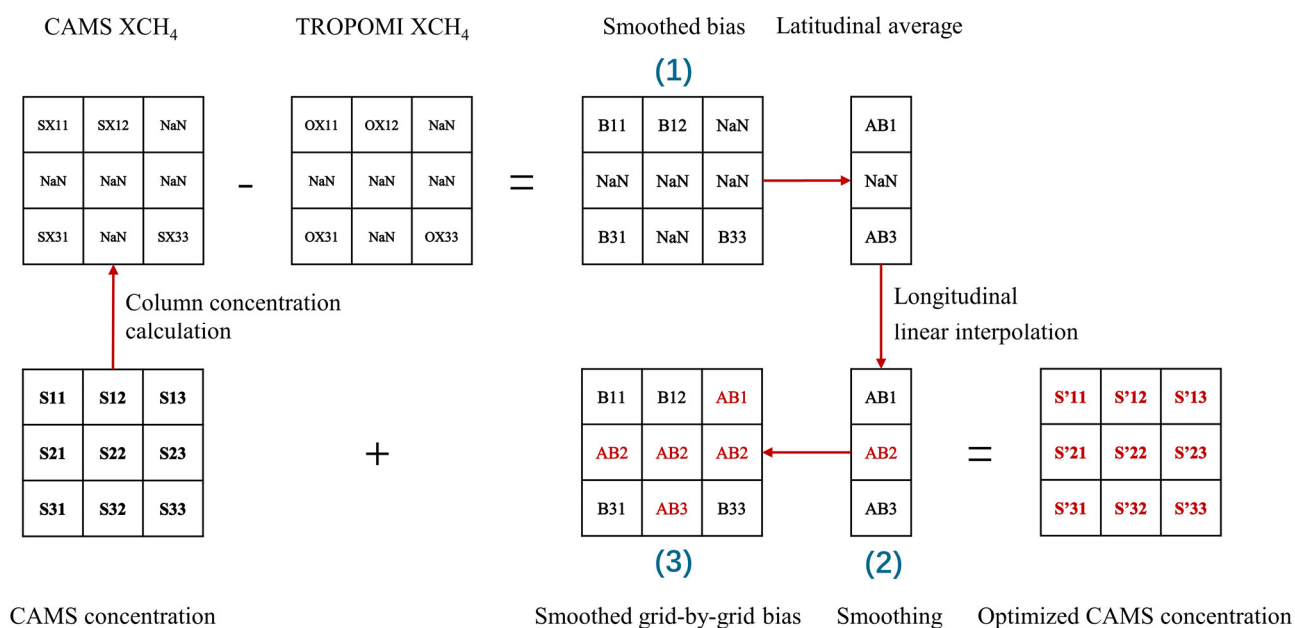


Figure S2 Schematic of constraining the CAMS concentration fields using TROPOMI XCH₄. Blue numbers indicate smoothing steps to remove abrupt variations: (1) smoothing CAMS XCH₄ bias over time, latitude, and longitude; (2) Smoothing after interpolation over time and latitude; and (3) smoothing the final bias over latitude and longitude.

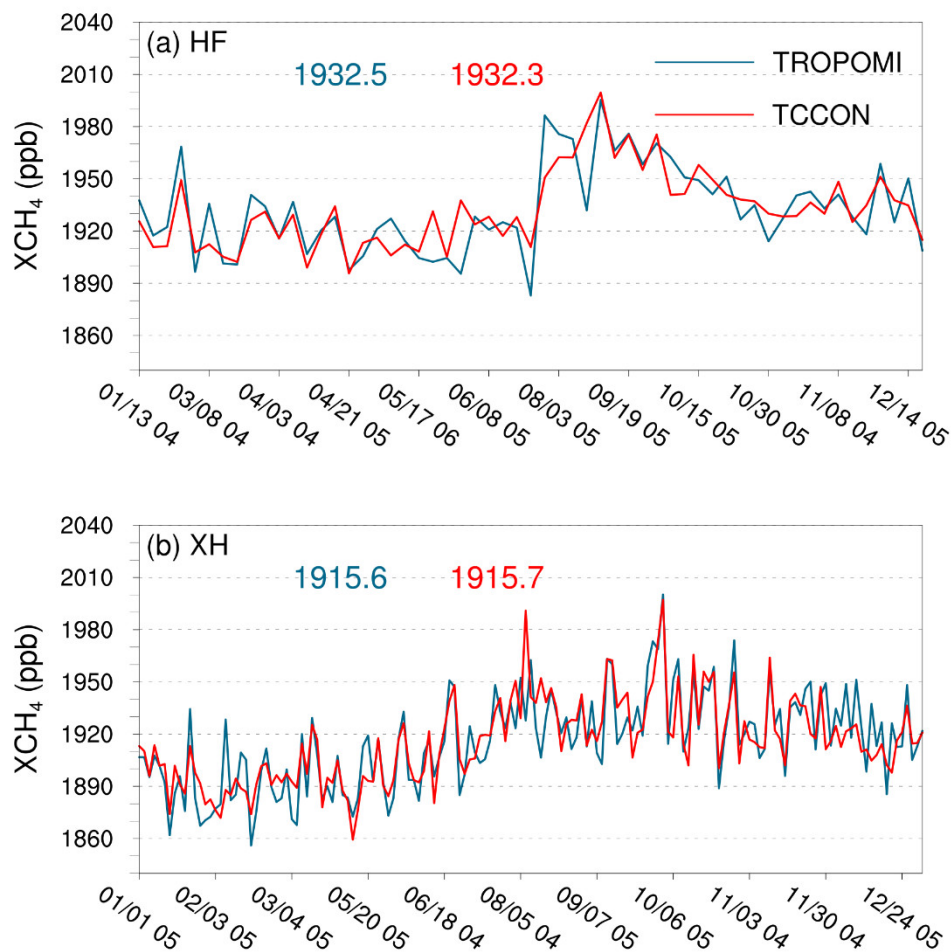


Figure S3 Comparison of time series between TROPOMI XCH₄ product after final quality control and TCCON observations at Hefei and Xianghe stations. For the evaluation, only TROPOMI pixels that are located within a 0.1° radius of the respective TCCON station and have a time difference of less than 1 hour relative to TCCON observational records were selected. Specifically, the number of valid matching pairs was 57 for the Hefei station and 155 for the Xianghe station.

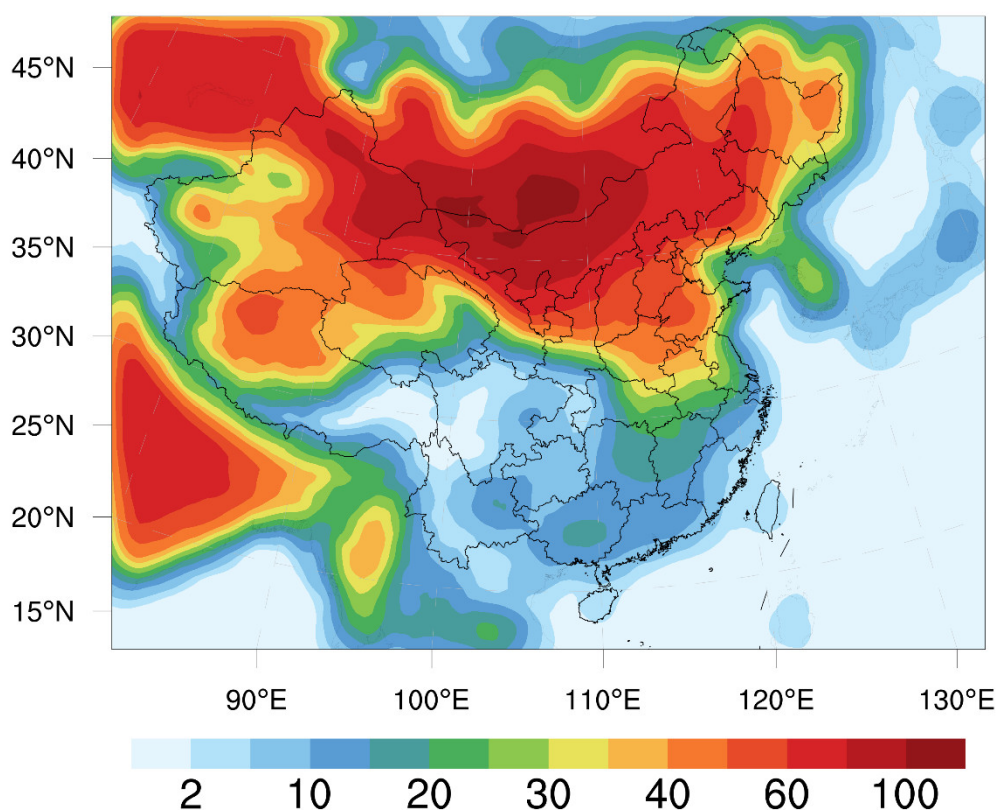


Figure S4 Average number of observations constraining each grid per day

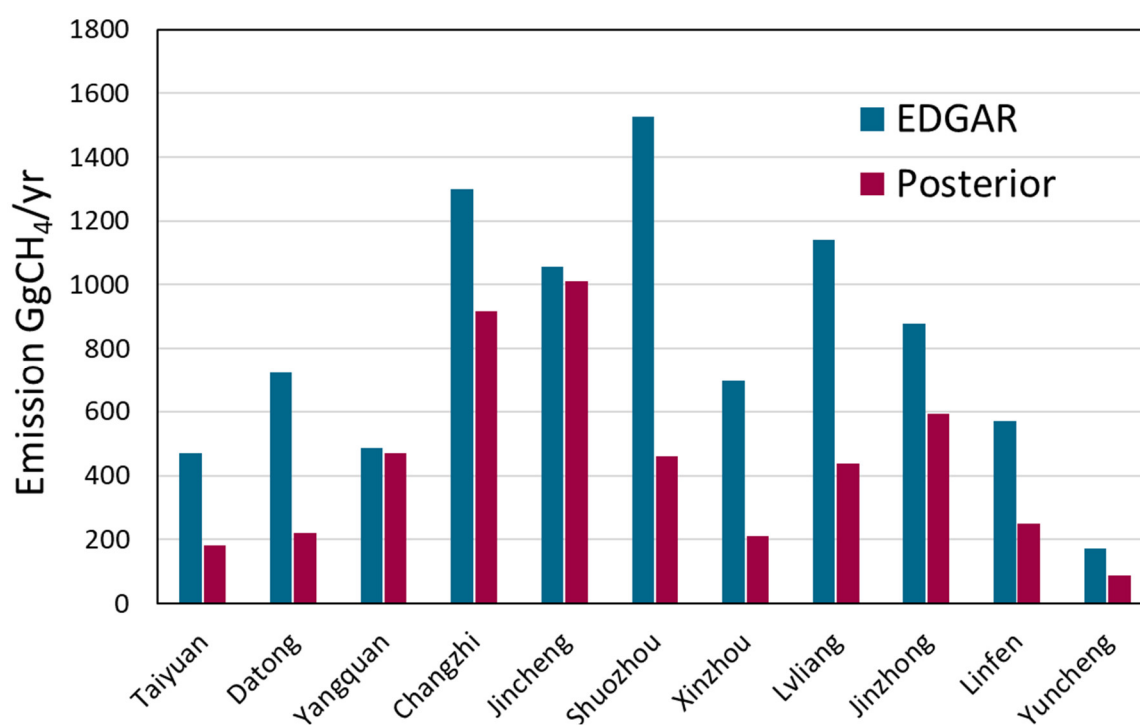


Figure S5 Comparison of prior and posterior emissions (GgCH₄·yr⁻¹) in various cities of Shanxi Province.

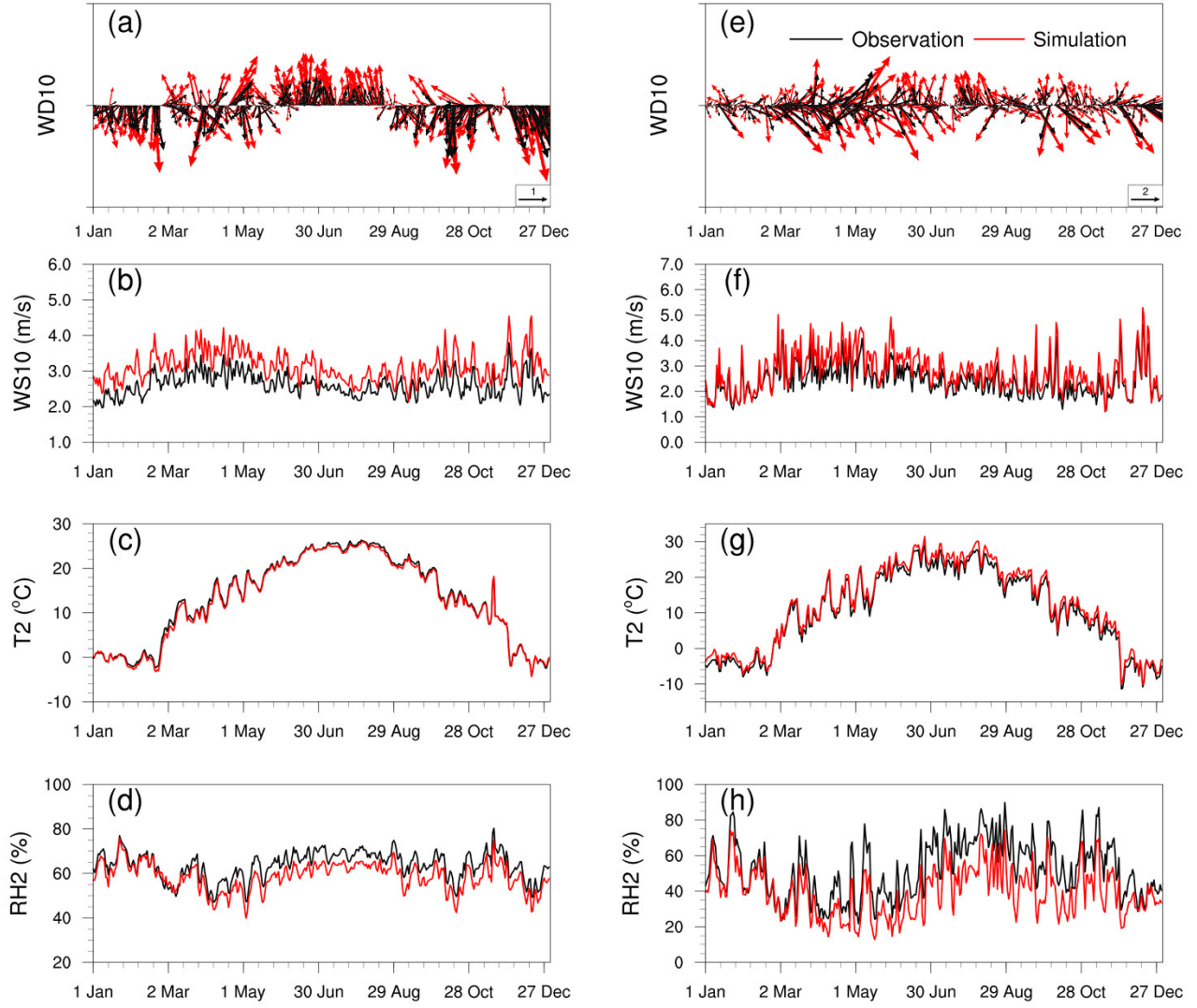


Figure S6 Time series of observed and simulated wind direction at 10 m (WD10), wind speeds at 10 m (WS10, m/s), temperature at 2 m (T2, °C), and relative humidity at 2 m (RH2, %) across (a-d) China and (e-h) Shanxi Province. China and Shanxi Province include 400 and 26 stations, respectively.

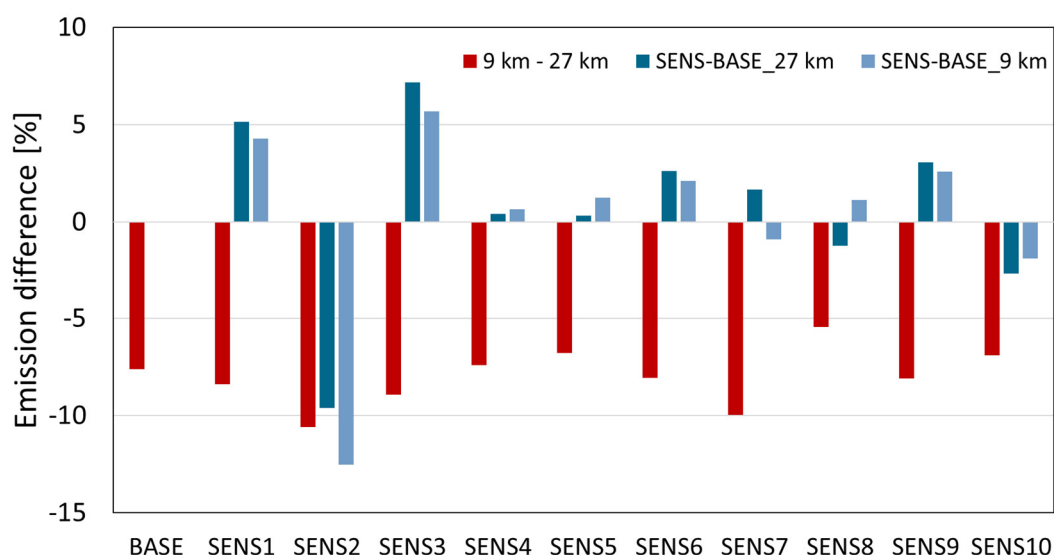


Figure S7 Comparison of emission differences under the D02 domain coverage: one between 9 km and 27 km resolutions within the same BASE or SENS experiment (red), and the other between corresponding SENS and BASE experiments at either 27 km (dark blue) or 9 km (light blue) resolution. Note that only positive differences (i.e., $\text{SENS} > \text{BASE}$ emissions) can indicate unconsidered factors that might lead to the low emission results in our study.

Table S1 Configuration options of WRF/CMAQ.

WRF		CMAQ	
Parameter	Scheme	Parameter	Scheme
Microphysics	WSM6	Horizontal/Vertical advection	yamo/wrf
Longwave	RRTM	Horizontal/Vertical diffusion	multiscale/acm2
Shortwave	Goddard	Deposition	/
Boundary layer	ACM	Chemistry solver	/
Cumulus	Kain-Fritsch	Photolysis	/
Land-surface	Noah	Aerosol module	/
Surface layer	Revised	Cloud module	/
Urban canopy	No	Gas-phase chemistry	/

Table S2 Independent observations of atmospheric CH₄ concentrations used to evaluate the posterior emission estimates.

Site	Latitude (°N)	Longitude (°E)	Altitude (m)	Obs. Number	Frequency	Location
AMY	36.5	126.3	47.	84/7979	Hourly/ Weekly	Coastal (Korea)
DSI	20.7	116.7	3.	49	Weekly	Ocean (Taiwan, China)
GSN	33.3	126.2	72.	7899	Hourly	Ocean (Korea)
LLN	23.5	120.9	2862.	50	Weekly	Mountain (Taiwan, China)
RYO	39.0	141.8	260.	7986	Hourly	Mountain (Japan)
TAP	36.7	126.1	16.	77	Weekly	Coastal (Korea)
ULD	37.5	130.9	220.9	8089	Hourly	Ocean (Korea)
WLG	36.3	100.9	3810.	47	Weekly	Tibetan Plateau (China)
YON	24.5	123.0	30.	8220	Hourly	Ocean (Japan)

Table S3 Monthly emission comparison ($\text{GgCH}_4\cdot\text{mon}^{-1}$) of major emission sectors over Mainland China and Shanxi Province. The numbers in parentheses represent posterior emissions.

	Coal	Gas	Rice	Waste	Livestock	Building	Manure
Mainland China							
Jan	2204.2 (835.9)	120.8 (86)	0 (0)	1316.5 (665.2)	597.5 (300.2)	405.6 (253)	118 (51.3)
Feb	2204.2 (780.1)	120.8 (117.3)	15.7 (13.1)	1316.5 (745.4)	597.5 (319.8)	310.9 (213.2)	118 (55.3)
Mar	2204.2 (869)	120.8 (109.3)	65.6 (52.2)	1316.5 (731.2)	597.5 (313.6)	272.7 (185.7)	118 (53.4)
Apr	2204.2 (1146.7)	120.8 (82.7)	869.3 (618.8)	1316.5 (758.6)	597.5 (319.4)	142.1 (96.3)	118 (53.5)
May	2204.2 (879.9)	120.8 (75.5)	2164.3 (1475.6)	1316.5 (745.9)	597.5 (315.5)	130 (86.5)	118 (52.6)
Jun	2204.2 (865.8)	120.8 (63)	2913.3 (2197.5)	1316.5 (722.4)	597.5 (309)	130 (86.1)	118 (52.7)
Jul	2204.2 (1146.8)	120.8 (71.9)	2771.7 (2783)	1316.5 (1099.8)	597.5 (347.1)	130 (117.7)	118 (61.4)
Aug	2204.2 (1567.9)	120.8 (73.9)	2855.6 (3530.1)	1316.5 (1491.2)	597.5 (401.2)	130 (145)	118 (73.2)
Sep	2204.2 (1018.5)	120.8 (66.6)	1608.4 (1840.5)	1316.5 (1351.7)	597.5 (346.6)	130 (130.5)	118 (62.5)
Oct	2204.2 (855.4)	120.8 (66.5)	400.5 (509.1)	1316.5 (1337.6)	597.5 (338.5)	168.8 (171.6)	118 (61.6)
Nov	2204.2 (904.9)	120.8 (71.9)	0 (0)	1316.5 (858.5)	597.5 (316.9)	288.8 (212.5)	118 (55.1)
Dec	2205.3 (791)	120.9 (71.9)	0 (0)	1317.1 (744.2)	597.8 (309.1)	384.8 (258.9)	118 (53.1)
Shanxi Province							
Jan	706.2 (362.2)	0.9 (0.3)	0 (0)	28.2 (11.7)	5.4 (2.1)	12.2 (5.2)	0.5 (0.2)
Feb	706.2 (230.8)	0.9 (0.3)	0 (0)	28.2 (9.2)	5.4 (1.7)	9.4 (3.1)	0.5 (0.2)
Mar	706.2 (314.6)	0.9 (0.3)	0 (0)	28.2 (10.7)	5.4 (1.9)	8.2 (3.1)	0.5 (0.2)
Apr	706.2 (490.6)	0.9 (0.4)	0.4 (0.2)	28.2 (14.8)	5.4 (2.4)	4.3 (2.1)	0.5 (0.2)
May	706.2 (363.4)	0.9 (0.4)	0.9 (0.5)	28.2 (15.8)	5.4 (2.6)	3.9 (2)	0.5 (0.2)
Jun	706.2 (363.3)	0.9 (0.3)	1.2 (0.6)	28.2 (12.2)	5.4 (2.2)	3.9 (1.8)	0.5 (0.2)
Jul	706.2 (497.5)	0.9 (0.5)	1.2 (0.7)	28.2 (17.7)	5.4 (2.9)	3.9 (2.4)	0.5 (0.3)
Aug	706.2 (617.2)	0.9 (0.5)	1.2 (0.9)	28.2 (24.2)	5.4 (3.5)	3.9 (3.1)	0.5 (0.4)
Sep	706.2 (374.5)	0.9 (0.5)	0.7 (0.4)	28.2 (20.7)	5.4 (3)	3.9 (2.6)	0.5 (0.3)
Oct	706.2 (298.4)	0.9 (0.4)	0.2 (0.1)	28.2 (12.2)	5.4 (2.2)	5.1 (2.2)	0.5 (0.2)
Nov	706.2 (393.6)	0.9 (0.4)	0 (0)	28.2 (12.2)	5.4 (2.2)	8.7 (3.9)	0.5 (0.2)
Dec	706.5 (265.7)	0.9 (0.3)	0 (0)	28.2 (10)	5.4 (1.8)	11.6 (4.1)	0.5 (0.2)

Table S4 Statistics comparing the CH₄ concentrations (ppb) from the simulations with prior (CEP) and posterior (VEP) emissions against independent surface observations, respectively.

Site	Mean Obs.	Mean Sim.		BIAS		RMSE		CORR	
		CEP	VEP	CEP	VEP	CEP	VEP	CEP	VEP
AMY-flask	2038.4	2070.4	2048.6	32.0	10.2	65.0	41.9	0.53	0.67
AMY-insitu	2065.7	2101.0	2069.0	35.3	3.3	98.7	69.5	0.64	0.72
DSI-flask	1983.8	2009.5	1994.0	25.7	10.1	53.6	49.2	0.87	0.80
GSN-insitu	2017.4	2041.1	2019.7	23.7	2.4	59.4	37.9	0.65	0.72
LLN-flask	1942.7	1969.3	1963.0	26.5	20.3	40.8	33.3	0.85	0.85
RYO-insitu	1998.0	2023.0	2018.6	25.0	20.6	37.2	31.7	0.73	0.74
TAP-flask	2032.7	2053.0	2035.0	20.3	2.3	47.7	36.2	0.85	0.85
ULD-insitu	2011.1	2034.4	2018.6	23.3	7.5	45.7	25.8	0.68	0.71
WLG-flask	1983.0	2030.4	2011.3	47.4	28.3	67.8	44.3	0.53	0.54
YON-insitu	1967.1	1994.2	1980.9	27.1	13.8	45.6	29.4	0.87	0.88

* BIAS, mean bias; RMSE, root mean square error; CORR, correlation coefficient

Table S5 Statistics comparing the daily average CH₄ concentrations (ppb) during 12:00–18:00 local time from the simulations with prior (CEP) and posterior (VEP) emissions against six independent surface *in-situ* observation sites in Shanxi Province, respectively. The numbers under the site names represent the number of valid observations.

Site Name	Mean Obs.	Mean Sim.		BIAS		RMSE		CORR	
		CEP	VEP	CEP	VEP	CEP	VEP	CEP	VEP
TY (348)	2356.2	2370.9	2165.5	14.6	-190.8	240.0	289.7	0.60	0.64
DT (284)	2079.3	2252.6	2078.1	173.3	-1.2	282.6	57.1	0.71	0.79
LF (276)	2299.5	2263.8	2134.9	-35.7	-164.7	193.6	240.2	0.44	0.37
SZ (358)	2082.0	2701.4	2218.0	619.4	136.0	885.2	226.9	0.41	0.49
JC (361)	2274.0	2332.0	2194.2	58.0	-79.7	206.3	222.2	0.68	0.64
WTS (364)	2058.3	2158.0	2058.8	99.6	0.5	139.4	58.7	0.53	0.64

* BIAS, mean bias; RMSE, root mean square error; CORR, correlation coefficient

Table S6. The assimilation, sensitivity, and validation experiments conducted in this study.

Exp.Type	Exp. Name	Prior inventory	Boundary conditions	Chemical reactions	Observation	Observation error	Background error	Local scale
Assimilation	BASE	EDGAR v8 (the first DA window), optimized emissions of the previous window (other DA windows)	Corrected CAMS global fields	Deactivated	TROPOMI/SRON	0.7%	40%	300 km
Sensitivity	SENS1	Same as BASE	Raw CAMS global fields	Deactivated	Same as BASE	0.7%	40%	300 km
	SENS2	Same as BASE, but with CAMS-GLOB-ANT v6.2	Same as BASE	Deactivated	Same as BASE	0.7%	40%	300 km
	SENS3	Same as BASE	Same as BASE	Activated	Same as BASE	0.7%	40%	300 km
	SENS4	Same as BASE	Same as BASE	Deactivated	TROPOMI/WFMD	0.7%	40%	300 km
	SENS5-6	Same as BASE	Same as BASE	Deactivated	Same as BASE	0.5%, 0.9%	40%	300 km
	SENS7-8	Same as BASE	Same as BASE	Deactivated	Same as BASE	0.7%	30%, 50%	300 km
	SENS9-10	Same as BASE	Same as BASE	Deactivated	Same as BASE	0.7%	40%	250 km, 350 km
	CEP	EDGAR v8	Same as BASE	Deactivated	/	/	/	/
Validation	VEP	Posterior emissions of BASE	Same as BASE	Deactivated	/	/	/	/